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Role of magnetic inertia in damped macrospin dynamics

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Role of magnetic inertia in damped macrospin dynamics

by

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Thesis

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Summary

The goal of this thesis is to introduce the concept of inertia in damped macrospin dynamics. Following the work performed by T. L. Gilbert in this direction, a mass is associated to the macrospin, not related to the displacement of a real mass, but to macrospin's inertia. As a consequence, a generalised form of the Gilbert dynamics equation accounting for macrospin's inertia is derived.

At the scale of a nanostructure ferromagnet, fluctuations are relevant. The macrospin undergoes a Brownian motion in the corresponding configuration space (a sphere of radius M_s). In order to make a description of the macrospin, a simple, rigorous and new theory, that of mesoscopic nonequilibrium thermodynamics (MNET), is used as an alternative to the quite heavy formalism of stochastic processes used by W. F. Brown Jr. The state of the magnetisation is described by a number of degrees of freedom that exert an influence in the overall dynamics of the system: the orientation \mathbf{m} of the magnetisation of angles (θ, ϕ) and the velocity $\mathbf{u} = \frac{d\mathbf{m}}{dt}$, or the angular momentum \mathbf{L} . A distribution function f is introduced related to the probability of finding the magnetisation in a particular state (\mathbf{m}, \mathbf{u}) or (\mathbf{m}, \mathbf{L}) . The combination of the statistical definition of the entropy as a function of the probability, together with the systematic methodology of nonequilibrium thermodynamics, results in a powerful theory describing not only the average dynamics of the macrospin, but also the fluctuations about it.

Hence, a generalised dynamic Gilbert and a generalised stochastic Brown equation are derived within the MNET theory. A relaxation time τ is defined dependent on macrospin's inertia and damping. The magnetisation behaviour determines two regimes: the inertial regime or the short time scale limit $t \ll \tau$ and, the diffusion regime or the long time scale limit. A new phenomenon, *nutation*, is predicted in the inertial regime, whereas the Gilbert dynamic and Brown stochastic equations are retrieved at the long time scale.

Furthermore, the question of extending the application of MNET in the field of spintronics is put in the context of spin-transfer. A two rotating fluid model is presented for the conduction s -electron spins and the d -electron spins of the magnetisation. The model is of interest very near the normal-ferromagnet interface where spin-accumulation is found.

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Chapter 1

Introduction and basic concepts

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1.1 Introduction

We live in the era of nanotechnology, on the continuous chase towards the smallest and the fastest; and the field of magnetism didn't escape it. In the 1990s, IBM introduced a new type of hard-disk drive (HDD) that would revolutionise data storage with higher densities and faster reading speeds. Crucial to this technological revolution was the giant magnetoresistance discovery (GMR) made by the groups of A. Fert [1] and P. Grünberg [2] - for which the two won the 2007 Nobel Prize in Physics. The structure of a GMR sample, the 'new' reading head for HDD, consists of alternate ferromagnetic and non-magnetic thin layers of hundreds of atoms thick. It imposed GMR to be one of the first real applications of nanotechnology, or more precisely of modern magnetic thin film technologies.

A thin ferromagnetic layer, such as the one of a 'GMR sample', is a monodomain magnetic 'particle' of uniform magnetisation, called also a *macrospin*. At equilibrium, the macrospin aligns with the direction of a deterministic magnetic field, which can be a superposition of several types of fields: externally applied, anisotropic or dipolar. However, if a magnetic field is applied with a different direction than that of the macrospin, the latter won't align instantaneously with it. It relaxes towards the new equilibrium direction on a nanosecond timescale. The science studying the equilibrium orientations of a complex magnetic structure bears the name of *magnetostatics*. The way the orientations behave from an equilibrium configuration to another as a response to a magnetic excitation (i.e. an applied magnetic field), is the domain of *dynamics in magnetism*.

This thesis focuses on the damped dynamics of the simplest magnetic structure, the macrospin. The cornerstones of macrospin dynamics are the phenomenological equations of Landau-Lifshitz (LL) [3] from 1935, and that of Gilbert [4] from 1955 (equivalent equations). *One novelty of this thesis consists in introducing an inertial hypothesis in the context of the stochastic behaviour of the magnetisation (macrospin coupled to a reservoir)*. A mass is associated to the macrospin, not related to the inertia of matter, but to that of the magnetisation. It will be shown that this assumption does not contradict the physics related to the dynamics of the macrospin (the LL and Gilbert equations), but instead, reveals a new phenomenon at time scales shorter than the picosecond. Furthermore, thermal fluctuations in macrospin dynamics will be rendered in a mesoscopic nonequilibrium thermodynamics framework (MNET).

1.1.1 Inertia in magnetism

Inertia in domain wall dynamics In 1948, W. Döring introduced for the first time the concept of mass in magnetism [5]. In the context of domain wall dynamics, he pointed out that a moving 180° domain wall exhibits inertia, despite the absence of any mass displacement.

When a magnetic field is applied in the plane of the wall, the wall begins to move. When the field is annulled, the wall keeps on moving with an energy proportional to half of the squared velocity of the wall and the proportionality constant is defined as the wall's virtual mass. The mass or inertia of a domain wall arises because the dynamic wall profile of a moving wall is slightly altered with respect to the static (standing) wall [6] (see Figure 1.1). Note that the domain wall mass should not be confused with a real mass. There is no actual material displaced; only the dynamic wall profile is translated. The mass as introduced by Döring has a macroscopic character, depends on the wall profile and is inversely proportional with the square of the gyromagnetic factor γ [6]. A nice experimental verification for the existence of the domain wall mass was given by de Leeuw and Robertson [7] in 1975 for garnet materials. In their experiment, it was shown that upon changing the equilibrium position of the domain wall, the wall first overshoots then oscillates around its new equilibrium position before coming to rest in the new position [8].

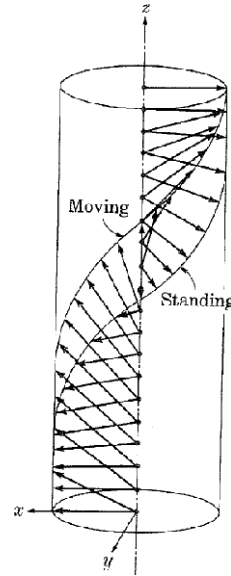


Figure 1.1: Standing versus moving 180° Bloch wall. Figure taken from the book written by S. Chikazumi on “Physics of Ferromagnetism“ [6].

Inertia in ferromagnetic resonance spinwaves The notion of mass is also introduced in the context of uniform ferromagnetic resonance spinwaves. There the inverse of the mass μ^{-1} is proportional to the second derivative of the energy in respect to the angle of precession ϕ of the magnetisation \mathbf{M} of amplitude M_s : $\mu^{-1} \propto \frac{1}{M_s^2 \sin^2 \theta} \frac{\partial^2 E}{\partial \phi^2}$, where θ is the tilting angle of the magnetisation [8].

Inertia in macrospin dynamics In 1955, T. L. Gilbert derived in his PhD dissertation a mathematical equivalent expression for the Landau-Lifschitz equation for the macrospin dynamics, bearing today his name. In his first derivation, he circumvented the problem of defining an inertial tensor and angular variables for spin rotations (without attempting to specify the functional form of the classical kinetic energy term for spin systems) with the help of the implicit assumption that the classical equations of motion could be interpreted as quantum operator equations [4]. However, he noted in the Appendix of his thesis and shown in a 1955 article [9–11] that the same equation can be derived within a Lagrangian formalism. T. L. Gilbert was one of the first to introduce the concept of inertia in the context

of macrospin dynamics. He tried to find an analogy between the macrospin's dynamics and a mechanical object dynamics. More precisely, he searched an expression for the tensor $\overline{\overline{I}}$ of macrospin's inertia that had a physical meaning in classical mechanics.

”One can show that the inertial tensor must have a single nonzero term corresponding to the rotational inertia for rotation about the principal axis. I was unable to conceive of a physical object with an inertial tensor of this kind [4].”

He associated a kinetic energy to the macrospin in the same way as in classical mechanics for an object with principal momenta of inertia $(0, 0, C)$ [9–11].

The **main goal of this thesis** is to reintroduce inertia in the damped dynamics of the macrospin. Following the footsteps performed by Gilbert in this direction, we will find an inertia tensor that makes physical sense and does not contradict the known dynamic behaviour of the magnetisation. Gilbert's equation will be derived in a mesoscopic nonequilibrium thermodynamics framework (MNET) capable to introduce fluctuations in dynamics. A new behaviour will be predicted at time scales shorter than the picosecond to be accounted by additional terms present in a generalised Gilbert equation describing the dynamics of the macrospin.

1.1.2 Thermal fluctuations of a uniform magnetised particle (macrospin)

The Gilbert and the Landau-Lifshitz equations describe only the determinist nature of the macrospin dynamics, and do not consider the effect of thermal fluctuations in the dynamics. It is necessary to point out that at this scale, for magnetic nanostructures such that of a macrospin, thermal fluctuations play an important role.

To introduce fluctuations in the dynamic behaviour of ”sufficiently fine ferromagnetic particles”, W. F. Brown Jr. proposes in his works from 1963 [12] and 1979 [13], a Brownian motion treatment for the macrospin. He supposes that in the presence of thermal agitation, the phenomenological damping field introduced by Gilbert describes the statistical average of rapidly fluctuating random forces. It must be augmented by a random field of statistical average zero and second moment proportional to the temperature. Thus, starting with the Gilbert equation he proceeds with the framework of stochastic processes to derive a rotational Fokker-Planck¹ equation for the probability of having the macrospin oriented in a particular direction (θ, ϕ) for an imposed magnetic field (magnetic potential energy profile) and being subjected to thermal fluctuations. This equation is the achievement of

¹In the field of “Statistical mechanics” [14], the rotational equation derived by Brown is a particular case of a 'real' Fokker-Planck equation and is commonly called the *Smoluchowski equation*. The name of 'Fokker-Planck equation' is generally given to an equation of a probability function f which is not only dependent on one slow variable, the position (our case - the orientation of the magnetisation), but also on a faster degree of freedom, usually the velocity of a Brownian particle.

a continuous-orientation model of the magnetisation, in respect to the discrete-orientation model proposed previously by Néel [15].

Fluctuations within MNET The stochastic processes framework used by Brown to introduce fluctuations in macrospin dynamics involves quite a heavy mathematical formalism, difficult to adapt to more complicated cases. An alternative and simpler approach exists leading to the same type of equations as the one derived by Brown: the *mesoscopic*² *nonequilibrium thermodynamics* framework (MNET) [17–19]. The works of I. Prigogine and P. Mazur from 1953 “Sur l’extension de la thermodynamique aux phénomènes irréversibles liés aux degrés de liberté internes” [20] and S. R. De Groot and P. Mazur from 1962 on “Non-equilibrium thermodynamics” [21] represent milestones for the recent theory of MNET, which leads simply, yet rigorously, to Fokker-Planck type equations. The MNET theory torch passed from P. Mazur [17, 20] to the groups of J. M. Rubi, J. M. G. Vilar and D. Reguera from “Universitat de Barcelona” [18, 19].

The **second goal of this thesis** is to show that fluctuations in macrospin dynamics can be rendered within a thermodynamic framework. More precisely, it is shown that non equilibrium thermodynamics formalism offers a simpler, yet rigorous framework than that proposed by Brown in 1963/1979, that derives not only the average dynamic behaviour of a magnetic particle (Gilbert [4] and Landau-Lifshitz [3] equations), but also the stochastic behaviour of a magnetic particle submitted to thermal fluctuations (Brown’s equation). Even more, with MNET one can go beyond the already known stochastic behaviour of the uniform magnetised particle, proposing a generalisation of Brown’s stochastic equation.

The **third goal of this thesis** is to propose as a perspective, a simple two fluid model based on mesoscopic nonequilibrium thermodynamics which could be applied in the context of the spin transfer [22–28]. A two rotating fluid model is proposed for the spin transfer problem as a perspective. Due to the large separation of time scales the conduction *s*-electron spins and the *d*-electron spins can be modelled as two rotating ‘fluids’ near the normal-ferromagnet interface, where spin-accumulation is found.

1.2 Plan of the thesis

The first chapter is a reminder of the basic concepts used all along the thesis. The application of mesoscopic nonequilibrium thermodynamics to the field of magnetism imposes a

²Here, and throughout the thesis the adjective mesoscopic has the same meaning as that given by van Kampen [16]: “The stochastic description in terms of macroscopic variables will be called mesoscopic. It comprises both the deterministic laws and the fluctuations about them”. Nowadays the term ‘mesoscopic’ is often used to denote the phenomena on the borderline between classical and quantum mechanics.

basic presentation of both. The first part of the chapter deals with the dynamic and stochastic behaviour of a uniform magnetised particle (macrospin), i.e. the Gilbert and the Brown stochastic equations. The original articles will be followed closely. The second part briefly gives a basic understanding of nonequilibrium thermodynamics (NET) and of mesoscopic nonequilibrium thermodynamics (MNET). The concept of local equilibrium is presented and illustrated in the simple case of one dimensional diffusion, and then generalised to the several degrees of freedom space in MNET.

The second chapter applies the nonequilibrium thermodynamics scheme to a statistical ensemble of non-interacting uniform magnetised particles with orientations distributed in the solid angle 4π . This is precisely the idea used by Brown in its 1963 article [12], with a main difference: instead of adding a white noise to the Gilbert dynamic equation, and performing quite heavy calculations characteristic to stochastic processes, we use the NET simple, phenomenological, yet rigorous framework to derive Brown's stochastic equation and Gilbert's equation.

The third chapter has the role of introducing an additional and faster degree of freedom next to that of the orientation $\mathbf{e} = (\theta, \phi)$ of an uniform magnetised particle. The study of magnetisation dynamics submitted to thermal fluctuations 'suggests' the velocity of the magnetisation $\frac{d\mathbf{m}}{dt} = \mathbf{u} = M_s \frac{d\mathbf{e}}{dt}$ to be the natural additional kinetic degree of freedom. Assuming that the magnetisation has inertia (an effective magnetic mass), the MNET framework is applied to a statistical ensemble of magnetic particles having as degrees of freedom: the orientation \mathbf{e} and the velocity \mathbf{u} . A dynamic generalised equation is derived describing the magnetisation behaviour going beyond the one deduced by Gilbert.

The fourth chapter While performing calculations in the third chapter, it was soon realised that the best additional degree of freedom is not the velocity $\frac{d\mathbf{m}}{dt}$, but instead the corresponding angular velocity $\boldsymbol{\omega}$ or the angular momentum $\mathbf{L} = \overline{\overline{\mathbf{I}}} \cdot \boldsymbol{\omega}$, where $\overline{\overline{\mathbf{I}}}$ stands for the tensor of the magnetisation inertia. The main justification for this reasoning was the intent to follow Gilbert's footsteps made in 1955 in his Ph.D. thesis [4]. He searched a mechanic formal analogue of the magnetisation dynamics, i.e. a physical object that has a dynamics similar to that of the magnetisation, or equivalently, an inertia tensor $\overline{\overline{\mathbf{I}}}$ of the magnetisation inertia with a physical meaning in classical mechanics. If in the third chapter the generalised Gilbert equation is derived inductively, in a simple mathematical formalism, in this chapter the magnetisation inertia tensor hypothesis together with the application of the MNET framework in a 'rotational phase-space' (\mathbf{e}, \mathbf{L}), lead deductively not only to the same generalised Gilbert equation, but also to the corresponding rotational Fokker-Planck equation, a generalisation of Brown's work from 1963 [12].

The previous three chapters derive the main dynamic and stochastic traits of the magnetisation in the classic framework of NET and MNET. **The fifth chapter** presents another possible application of MNET in the field of magnetism, in the context of the spin-transfer phenomenon [22–24] and is presented as perspectives for future research. Its final goal is to introduce the spins of the electrons, and treat them on the same footing with that of the magnetisation. The microscopic versus macroscopic, the time scale separation of the two species: spins and macrospin (magnetisation), the cumulative effect of the first on the last, the conservation laws for the energy and angular momentum for the entire system, led to the simple idea of treating the d -electron spins and the s -electron spins as two rotating fluids near the normal-ferromagnet interface, where there is spin-accumulation. This kind of problem imposes the use of the so called two-fluid model scheme [29, 30] used previously with success, in the context of plasma physics [30].

1.3 Macrospin dynamics

1.3.1 Gilbert's equation within a Lagrangian formalism

In this subsection we briefly remind the Lagrangian derivation of Gilbert's equation as he derived it [4, 9–11]. However, it has to be reminded that the first one to point out that the precession can be derived from a Lagrangian was Döring [5, 31].

A Without damping

Suppose the magnetisation \mathbf{M} of a ferromagnetic specimen has the static equilibrium direction described by the azimuth ϕ and the polar angle θ , as in the Fig. 1.2. A Lagrangian function \mathcal{L} can be associated to the magnetisation by considering the macrospin as a classical top with principal moments of inertia $(0, 0, C)$ and Euler angles ϕ, θ and ψ [11]. Here ψ is the angle that describes a rotation of the top about its symmetry axis. The angular velocity of the top about its symmetry axis is then $\dot{\psi}$ plus the component $\dot{\phi} \cos \theta$ (see Fig. 1.2). The kinetic energy T of the top is therefore given by [11, 32]:

$$T = \frac{1}{2}C \left(\dot{\psi} + \dot{\phi} \cos \theta \right)^2 \quad (1.1)$$

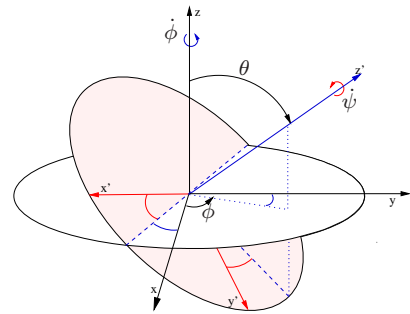


Figure 1.2:

and the Lagrangian function by $\mathcal{L} = T - V(\theta, \phi)$, where V is the potential energy. Now Lagrange's equations of motion have the general form [32]:

$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{q}_\alpha} - \frac{\partial \mathcal{L}}{\partial q_\alpha} = 0 \quad (1.2)$$

where q_α is a coordinate, i.e. either θ , either ϕ , either ψ . The application of the equation to the present problem yields

$$\begin{cases} \frac{d}{dt} [C \cos \theta (\dot{\psi} + \dot{\phi} \cos \theta)] + \frac{\partial V}{\partial \phi} = 0 \\ \frac{d}{dt} (0) + C \sin \theta \dot{\phi} (\dot{\psi} + \dot{\phi} \cos \theta) + \frac{\partial V}{\partial \theta} = 0 \\ \frac{d}{dt} [C (\dot{\psi} + \dot{\phi} \cos \theta)] = 0 \end{cases} \quad (1.3)$$

The last equation of the system affirms that the angular momentum conjugate to the coordinate ψ is conserved.

$$C (\dot{\psi} + \dot{\phi} \cos \theta) = cst. \quad (1.4)$$

In order to return to the magnetic system, it is necessary only to recognise that this angular momentum should be set equal to $\frac{M_s}{\gamma}$ [11], a quantity that is a constant of motion of this approximation. The system of equations becomes:

$$\begin{cases} \frac{M_s}{\gamma} \dot{\theta} \sin \theta = \frac{\partial V}{\partial \phi} \\ -\frac{M_s}{\gamma} \dot{\phi} \sin \theta = \frac{\partial V}{\partial \theta} \end{cases} \Leftrightarrow \boldsymbol{\omega}_\perp = -\dot{\phi} \sin \theta \boldsymbol{\theta} + \dot{\theta} \boldsymbol{\phi} = \frac{\gamma}{M_s} \frac{\partial V}{\partial \mathbf{e}} \quad (1.5)$$

$$\Rightarrow \frac{d\mathbf{M}}{dt} = \boldsymbol{\omega}_\perp \times \mathbf{M} = \gamma \mathbf{M} \times \mathbf{H}_{eff} \quad (1.6)$$

where the angular velocity $\boldsymbol{\omega}$ of the macrospin and the effective magnetic field were introduced: $\mathbf{H}_{eff} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$.

B With damping

Damping may also be introduced within a Lagrangian formulation, following the trends in classical mechanics [31, 32], with the help of the Rayleigh dissipation function \mathcal{F} :

$$\mathcal{F} = \frac{1}{2} \eta \left(\frac{d\mathbf{M}}{dt} \right)^2 = \frac{1}{2} \eta M_s^2 (\dot{\theta}^2 + \dot{\phi}^2 \sin^2 \theta) \quad (1.7)$$

Then, the equations of motion have the general form:

$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{q}_\alpha} - \frac{\partial \mathcal{L}}{\partial q_\alpha} + \frac{\partial \mathcal{F}}{\partial \dot{q}_\alpha} = 0 \quad (1.8)$$

These equations applied to our particular case lead to the following system of equations:

$$\begin{cases} \frac{d}{dt} (0) + C (\dot{\phi} \cos \theta + \dot{\psi}) \dot{\phi} \sin \theta + \eta M_s^2 \dot{\theta} + \frac{\partial V}{\partial \theta} = 0 \\ \frac{d}{dt} [C (\dot{\phi} \cos \theta + \dot{\psi}) \cos \theta] + \eta M_s^2 \dot{\phi} \sin^2 \theta + \frac{\partial V}{\partial \phi} = 0 \\ \frac{d}{dt} [C (\dot{\phi} \cos \theta + \dot{\psi})] = 0 \end{cases} \quad (1.9)$$

With constant of motion, the conjugate angular momentum of the ψ coordinate $\frac{M_s}{\gamma}$, the Gilbert equation [4] is derived.

$$\frac{\partial \mathcal{L}}{\partial \dot{\psi}} = C (\dot{\phi} \cos \theta + \dot{\psi}) = \frac{M_s}{\gamma} \quad (1.10)$$

$$\begin{cases} \dot{\phi} \sin \theta = -\frac{\gamma}{M_s} \frac{\partial V}{\partial \theta} - \gamma \eta M_s \dot{\theta} \\ \dot{\theta} = \frac{\gamma}{M_s \sin \theta} \frac{\partial V}{\partial \phi} + \gamma \eta M_s \dot{\phi} \sin \theta \end{cases} \Leftrightarrow \boldsymbol{\omega}_\perp = -\dot{\phi} \sin \theta \boldsymbol{\theta} + \dot{\theta} \boldsymbol{\phi} = \frac{\gamma}{M_s} \frac{\partial V}{\partial \mathbf{e}} + \gamma \eta \boldsymbol{\omega}_\perp \times \mathbf{M} \quad (1.11)$$

$$\frac{d\mathbf{M}}{dt} = \boldsymbol{\omega}_\perp \times \mathbf{M} = \gamma \mathbf{M} \times \left(-\frac{\partial V}{\partial \mathbf{M}} - \eta \frac{d\mathbf{M}}{dt} \right) \quad (1.12)$$

1.3.2 Thermal fluctuations - Brown's stochastic equation

In this subsection we briefly remind the steps made by W. F. Brown Jr. [12] in his derivation of the rotational Fokker-Planck equation.

Let \mathbf{M} be the magnetisation of a mono-domain ferromagnetic particle, of volume v and energy vV . In average, the dynamics of \mathbf{M} obeys Gilbert's equation:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times [-\partial V / \partial \mathbf{M} - \eta d\mathbf{M} / dt] \quad (1.13)$$

where γ is the gyromagnetic ratio and η is a dissipation constant. The equilibrium configurations of the magnetisation correspond to the annullment of $\frac{d\mathbf{M}}{dt}$ from (1.13), which

translates in finding the energy minima of the particle. One can find more than one energy minima; the valleys of a potential landscape, separated by barriers or hills of the potential landscape.

In the presence of thermal fluctuations, without changing the applied field (energy profile), the passage from one metastable state to another can be accomplished, due to the thermal energy $k_B T$, at long enough time scales. As W. F. Brown Jr. said [13, 33],

”we expect if we put this recording to a shelf that it will stay in the same magnetic state and would be surprised if it suddenly jumped from being a recording of Beethoven to that of Brahms. In principle, the *apparent stability* of the recording is only one of the *many* local minima of the free energy; thermal agitation can cause spontaneous jumps from one such state to another”.

The intention of W. F. Brown Jr. was to include thermal agitation as a factor in the dynamics of the magnetisation. The rotational Fokker-Planck equation accounts of the applied forces and of the thermal agitation. It was first suggested by Néel [15] and then derived by W. F. Brown Jr.

He begins its treatment assuming that in the presence of thermal fluctuations, the dissipative field $-\eta \frac{d\mathbf{M}}{dt}$ from Eq. (1.13) describes the statistical average of rapidly fluctuating forces, and that for an individual particle this expression must be augmented by a term $\mathbf{h}(t)$:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times [-\partial V / \partial \mathbf{M} - \eta d\mathbf{M}/dt + \mathbf{h}(t)] \quad (1.14)$$

He furthermore assumes that the noise field $\mathbf{h}(t)$ is Gaussian (Markovian), and it has the following properties:

$$\begin{cases} \langle h_i(t) \rangle = 0 \\ \langle h_i(t), h_j(t) \rangle = m \delta_{ij} \delta(t) \end{cases} \quad (1.15)$$

where m is a constant.

The exact calculation will be sketched in a second paragraph, however ”we digress to present a simpler, intuitive method of taking account of thermal agitation”.

A Basic concept

W. F. Brown Jr. takes a statistical ensemble of identical particles, that can be represented by a distribution of points over the unit sphere, with surface density $W(\theta, \phi)$. As the particles undergo changes of moment orientation, the representative points move, and there is a net surface-current density \mathbf{J} . The number of points of the unit sphere is conserved and one can write the continuity equation

$$\frac{\partial W}{\partial t} = -\nabla \mathbf{J} \quad (1.16)$$

where, in the absence of thermal agitation, $\mathbf{J} = W\mathbf{v}$ and \mathbf{v} is the velocity of a representative point $\mathbf{v} = \frac{d\mathbf{M}}{dt} \frac{1}{M_s}$. As the expression of $\frac{d\mathbf{M}}{dt}$ is already known Eq. (1.13), one can derive the expression of the surface density current in the absence of thermal agitation.

To the expression of \mathbf{J} , W. F. Brown Jr. adds a diffusion term $-k'\nabla W$ that has the tendency to make the distribution more nearly uniform. "Direct justification of this intuitive procedure would be difficult; but in fact it gives the same result as the Fokker-Planck method of the following paragraph, with considerably less labor" [12]

The intuitive procedure gives:

$$\begin{cases} J^\theta = - \left[\left(h' \frac{\partial V}{\partial \theta} - \frac{g'}{\sin\theta} \frac{\partial V}{\partial \phi} \right) W + k' \frac{\partial W}{\partial \theta} \right] \\ J^\phi = - \left[\left(g' \frac{\partial V}{\partial \theta} + \frac{h'}{\sin\theta} \frac{\partial V}{\partial \phi} \right) W + \frac{k'}{\sin\theta} \frac{\partial W}{\partial \phi} \right] \end{cases} \quad (1.17)$$

and

$$\begin{aligned} \frac{\partial W(\theta, \phi)}{\partial t} = \frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left\{ \sin\theta \left[\left(h' \frac{\partial V}{\partial \theta} - \frac{g'}{\sin\theta} \frac{\partial V}{\partial \phi} \right) W + k' \frac{\partial W}{\partial \theta} \right] \right\} \\ + \frac{1}{\sin\theta} \frac{\partial}{\partial \phi} \left\{ \left(g' \frac{\partial V}{\partial \theta} + \frac{h'}{\sin\theta} \frac{\partial V}{\partial \phi} \right) W + \frac{k'}{\sin\theta} \frac{\partial W}{\partial \phi} \right\} \end{aligned} \quad (1.18)$$

"The Fokker-Planck method will lead directly to the partial differential equation (1.18), without introduction of the current-density components J^θ , J^ϕ " [12]

B The Fokker-Planck method

The method is based on the stochastic general Fokker-Planck equation:

$$\frac{\partial P(x_1, x_2)}{\partial t} = - \sum_{i=1}^2 \frac{\partial}{\partial x_i} (A_i P) + \frac{1}{2} \sum_{i,j=1}^2 \frac{\partial^2}{\partial x_i \partial x_j} (B_{ij} P) \quad (1.19)$$

where, in our case, $x_1 = \theta$, $x_2 = \phi$, A_i and B_{ij} are functions of $x_1 = \theta$, $x_2 = \phi$ defined by:

$$A_i = \lim_{\Delta t \rightarrow 0} \frac{\langle \Delta x_i \rangle}{\Delta t}; \quad B_{ij} = \lim_{\Delta t \rightarrow 0} \frac{\langle \Delta x_i \Delta x_j \rangle}{\Delta t} \quad (1.20)$$

As W. F. Brown Jr. needs to calculate the coefficients A_i and B_{ij} , for deriving the particular Fokker-Planck equation respected by a monodomain magnetic particle under thermal fluctuations, he uses the Landau-Lifshitz form of Gilbert equation (1.14):

$$\frac{d\mathbf{M}}{dt} = g' M_s [\mathbf{M} \times (\mathbf{H} + \mathbf{h})] + h' [\mathbf{M} \times (\mathbf{H} + \mathbf{h})] \times \mathbf{M} \quad (1.21)$$

to derive the equations satisfied by the derivatives of θ and ϕ

$$\begin{cases} \dot{\theta} = h' \left(-\frac{\partial V}{\partial \theta} + P_\theta \right) - \frac{g'}{\sin \theta} \left(-\frac{\partial V}{\partial \phi} + P_\phi \right) \\ \dot{\phi} = \frac{g'}{\sin \theta} \left(-\frac{\partial V}{\partial \theta} + P_\theta \right) - \frac{h'}{\sin^2 \theta} \left(-\frac{\partial V}{\partial \phi} + P_\phi \right) \end{cases} \quad (1.22)$$

where

$$g' = \frac{\gamma}{(1 + \alpha^2)M_s}; \quad h' = \frac{\alpha\gamma}{(1 + \alpha^2)M_s} = \alpha g'; \quad \alpha = \eta\gamma M_s \quad (1.23)$$

and

$$\begin{cases} P_\theta = M_s \{h_1(t) \cos \theta \cos \phi + h_2(t) \cos \theta \sin \phi - h_3(t) \sin \theta\} \\ P_\phi = M_s \{-h_1(t) \sin \theta \sin \phi + h_2(t) \sin \theta \cos \phi\} \end{cases} \quad (1.24)$$

Once the equations respected by $\dot{\theta}$ and $\dot{\phi}$ under thermal fluctuations are derived, $\Delta\theta$ and $\Delta\phi$ are evaluated only to terms of order Δt for A_i , and only to terms of order $(\Delta t)^{\frac{1}{2}}$ for B_{ij} . As it is expected, the evaluations of $\Delta\theta$ and $\Delta\phi$ contain components of the fluctuating field $\mathbf{h}(t)$.

In a second step, W. F. Brown Jr. takes the statistical average of $\Delta\theta$, $\Delta\phi$, $\Delta\theta\Delta\theta$, $\Delta\theta\Delta\phi$ and $\Delta\phi\Delta\phi$, making use of the properties of the fluctuating field $\mathbf{h}(t)$ 1.15. Once these laborious steps are made, the coefficients A_i and B_{ij} are deducted easily.

$$\begin{cases} A_1 = -h' \frac{\partial V}{\partial \theta} + \frac{g'}{\sin \theta} \frac{\partial V}{\partial \phi} + \frac{1}{2} m M_s^2 (h'^2 + g'^2) \cot \theta \\ A_2 = -\frac{g'}{\sin \theta} \frac{\partial V}{\partial \theta} - \frac{h'}{\sin^2 \theta} \frac{\partial V}{\partial \phi} \\ B_{11} = m M_s^2 (h'^2 + g'^2) \\ B_{12} = B_{21} = 0 \\ B_{22} = m M_s^2 (h'^2 + g'^2) \frac{1}{\sin^2 \theta} \end{cases} \quad (1.25)$$

Substitution of (1.25) in eq. (1.19) gives the differential equation followed by P . By the definitions of P and W :

$$P = W \sin \theta \quad (1.26)$$

with some rearranging, the equation satisfied by W can be reduced to the form 1.18, with

$$k' = \frac{1}{2} m M_s^2 (h'^2 + g'^2) \quad (1.27)$$

To relate the constant k' and m to other constants, W. F. Brown Jr. imposes the requirement

that in statistical equilibrium ($\frac{\partial W}{\partial t} = 0$), W must reduce to:

$$W_0 = A_0 e^{-V(\theta, \phi)v/k_B T} \quad (1.28)$$

Substitution of Eq. (1.28) in Eq. (1.18) leads to an identity only if

$$\begin{cases} k' = h' \frac{k_B T}{v} \\ m = 2 \frac{k_B T}{v} \eta \end{cases} \quad (1.29)$$

1.4 Thermodynamics beyond equilibrium

We live in a world that is not in thermodynamic equilibrium. Most of the time we encounter phenomena exhibited by systems not in thermodynamic equilibrium, while equilibrium systems are the exception. Yet, thermodynamics that describes equilibrium states is of great importance and extremely useful. In such cases, the intricate behaviour of large numbers of molecules can completely be characterised by a few variables that describe general average properties and strictly follow the rules of thermodynamics [34, 35].

However, it is possible to extend thermodynamics to situations that are in *local equilibrium*. For almost every macroscopic system we can meaningfully assign a temperature T , and other thermodynamic variables to every "elemental volume" δV . In most situations we may assume that *equilibrium thermodynamic relations are valid for the thermodynamic variables assigned to an elemental volume*. This is the concept of local equilibrium and is the domain of validity of nonequilibrium thermodynamics (NET) [21, 34]. Despite its generality, this theory has notorious limitations: it is applicable only to macroscopic systems, for which fluctuations are not important, and it operates within the linear response domain. Whereas the linear approximation is valid for many transport processes, such as heat conduction and mass diffusion, even in the presence of large gradients it is not appropriate for activated processes in which the system immediately enters the nonlinear domain.

The goal of this section is to present advances aimed at obtaining a simple and comprehensive description of the dynamics of nonequilibrium systems at the mesoscopic scale. These advances have provided not only a deeper understanding of the concept of local equilibrium but also a framework (MNET), reminiscent of nonequilibrium thermodynamics. To set its grounds, the nonequilibrium thermodynamics concepts are discussed first in subsection 1.4.1 and then, the extension to mesoscopic scales is described in subsection 1.4.2.

1.4.1 Nonequilibrium thermodynamics (NET)

While equilibrium thermodynamics deals with large systems at equilibrium, nonequilibrium thermodynamics (NET) [21, 34] extends to situations where the system is *not* in global, but in local equilibrium. It is built on the grounds of two main hypotheses: the local equilibrium hypothesis and the nonnegativity of the entropy production. The theory attributes the deviations from equilibrium to the presence of unbalanced thermodynamic forces, such as electric fields or gradients, which give rise to fluxes, such as electric or heat currents. Forces and fluxes are in a relationship that is cause-effect compatible with the second law of thermodynamics and with the inherent symmetries, either macroscopic or microscopic. The dynamics follows from the local conservation laws for the thermodynamic field quantities, in which the fluxes are linear functions of the forces whose coefficients, the Onsager coefficients, satisfy reciprocity relations.

To illustrate explicitly this method, it will be applied to the simple case of particle diffusion in one dimension.

A Local equilibrium

The local equilibrium hypothesis assumes that the thermodynamic variables defined locally, i.e. in each subsystem of a conveniently partitioned system, admit the same interpretation as in equilibrium and that, the equilibrium thermodynamic relations (the 1st and the 2nd principle of thermodynamics) are also valid for the local variables assigned to each subsystem. Explicitly:

Local variables The extensive variables: the entropy S , the energy E , the number of particles N_k are replaced by the corresponding volume densities $s(\mathbf{x}, t)$, $e(\mathbf{x}, t)$ and $n_k(\mathbf{x}, t)$, while the intensive variables: the temperature $T(\mathbf{x}, t)$, the pressure $p(\mathbf{x}, t)$ and the chemical potential $\mu_k(\mathbf{x}, t)$ become functions of position \mathbf{x} and time t .

Local form of the 1st principle of thermodynamics For any macroscopic system, there is a scalar state function, extensive and conserved called energy E with the following property:

$$dE = dW + dQ + dU_{matter} \quad (1.30)$$

where dW , dQ and dU_{matter} are energies received by the system in forms of work, of heat, respectively from exchanges of matter between different chemical species of the system.

The 1st principle imposes that the energy E is a state function, i.e. that the total differential of the energy dE exists. When introducing the state extensive variables (S, V, N_k, X_j), corresponding intensive variables are defined: the temperature T , the pressure p , the chem-

ical potentials μ_k , and the generalised forces F_j :

$$\begin{aligned} dE &= \frac{\partial E}{\partial S} dS + \frac{\partial E}{\partial V} dV + \sum_k \frac{\partial E}{\partial N_k} dN_k + \sum_j \frac{\partial E}{\partial X_j} dX_j \\ &= TdS - pdV + \sum_k \mu_k dN_k - \sum_j F_j dX_j \end{aligned} \quad (1.31)$$

This explicit expression of the 1st principle posing in balance all forms of energy bears the name of the *Gibbs relation*. Its *local form* is:

$$Tds = de - \sum_k \mu_k dn_k + \sum_j F_j dx_j \quad (1.32)$$

where the volume does not appear, as the entropy density s , the energy density e and the number density n_k are volume densities.

Local form of the 2nd principle of thermodynamics According to the second principle of equilibrium thermodynamics, one can introduce for any macroscopic system a state function S , the entropy of the system with the following properties:

- (a). The variation of the entropy dS may be written as the sum of two terms:

$$dS = d_e S + d_i S \quad (1.33)$$

where $d_e S$ is the entropy supplied to the system by its surroundings, and $d_i S$ is the entropy produced inside the system.

- (b). The supplied entropy $d_e S$ may be positive, negative or zero, depending on the interaction of the system with its surroundings. For example, for an adiabatically insulated system $d_e S = 0$.
- (c). On the other hand, the 2nd principle of thermodynamics states that $d_i S$ must be zero for reversible (or equilibrium) transformations and positive for irreversible transformations of the system:

$$d_i S \geq 0 \quad (1.34)$$

The local form of these two equations (1.33) and (1.34), can be derived if the volume entropy density s , the entropy flux \mathbf{J}^s and the entropy source strength σ or entropy production are introduced.

$$S = \int^V ns dV \quad \frac{d_e S}{dt} = - \int^\Sigma \mathbf{J}^s \cdot d\Sigma \quad \frac{d_i S}{dt} = \int^V \sigma dV \quad (1.35)$$

With the Gauss theorem, the equation for the entropy (1.33) writes as:

$$\int^V \left(\frac{\partial ns}{\partial t} + \frac{\partial \mathbf{J}^s}{\partial \mathbf{x}} - \sigma \right) dV = 0 \quad (1.36)$$

Since (1.33) and (1.34) must hold for any arbitrary volume V , the local form of the entropy balance equation and that of the 2nd law of thermodynamics are obtained:

$$\begin{cases} \frac{\partial ns}{\partial t} = -\frac{\partial \mathbf{J}^s}{\partial \mathbf{x}} + \sigma \\ \sigma \geq 0 \end{cases} \quad (1.37)$$

B Illustration: diffusion of particles in one dimension

The first step is to compute the entropy production σ . We start with the Gibbs relation (1.32). For the sake of simplicity we assume that the process takes place at a constant temperature T .

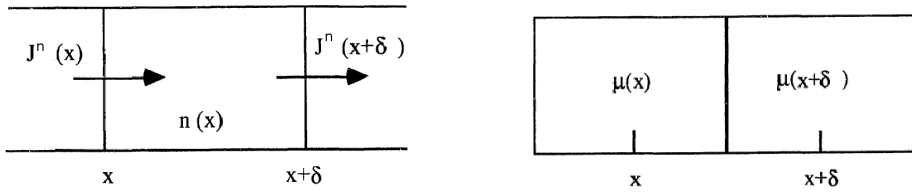
$$T \frac{\partial s}{\partial t} = \frac{\partial e}{\partial t} - \mu \frac{\partial n}{\partial t} \quad (1.38)$$

With the conservation laws (1.39) for the extensive parameters entropy density s , energy density e and number density n (see Fig. 1.3), the expression for the entropy production σ is derived as the product of the flux of particles J^n and its corresponding thermodynamic force $\frac{\partial \mu}{\partial x}$, where J^e and J^s are the energy, respectively the entropy flux.

$$\begin{cases} \frac{\partial n}{\partial t} = -\frac{\partial J^n}{\partial x} \\ \frac{\partial e}{\partial t} = -\frac{\partial J^e}{\partial x} \end{cases} \xrightarrow{\text{with (1.38)}} \begin{cases} \frac{\partial s}{\partial t} = -\frac{\partial}{\partial x} \left(\frac{J^e}{T} - \frac{J^n \mu}{T} \right) - \frac{J^n}{T} \frac{\partial \mu}{\partial x} \\ \frac{\partial s}{\partial t} = -\frac{\partial J^s}{\partial x} + \sigma \end{cases} \Rightarrow \sigma = -J^n \frac{\partial \mu}{\partial x} \quad (1.39)$$

Entropy production σ Generally, the structure of the expression for σ is that of a bilinear form: it consists of a sum of products of two factors. One of these factors in each term is a flow quantity (heat flow, diffusion flow, momentum flows or viscous pressure tensor, chemical reaction rate) already introduced in conservation laws. The other factor in each term is related to a gradient of an intensive state variable (gradients of temperature, chemical potential and velocity) and may contain the external force; it can also be a difference of thermodynamic state variables. These quantities which multiply the fluxes in the expression for the entropy production are called *thermodynamic forces*.

Phenomenological law In the absence of nonlocal effects, the flux J^n is proportional to the thermodynamic force $\frac{\partial \mu}{\partial x}$, whose coefficient, the Onsager coefficient $L = L[x, n(x)]$



(a) The change in the number of particles $\frac{\partial n \delta}{\partial t}$ in a small cell of size δ , at location x , is due to the difference in the flow J^n into and out of the cell $J^n(x) - J^n(x + \delta) = -\frac{\partial J^n}{\partial x} \delta$, translated into a conservation law: $\frac{\partial n}{\partial t} = -\frac{\partial J^n}{\partial x}$

(b) The thermodynamic force, which is the difference in the chemical potential, is given by $\mu(x) - \mu(x + \delta) = -\frac{\partial \mu}{\partial x} \delta$.

Figure 1.3: If in the first figure it is illustrated the flux of particles J^n together with the corresponding conservation law, in the second figure we have its cause: the thermodynamic force $\frac{\partial \mu}{\partial x}$. Similar conservation laws can be written for the energy and entropy, where the entropy is not conserved according to the second law of thermodynamics ($\sigma \geq 0$).

depends on the thermodynamic variables as well as on the x -coordinate.

$$J^n = -L \frac{\partial \mu}{\partial x} \quad (1.40)$$

For a chemical potential that does not depend explicitly on the spatial coordinate, i.e. $\mu = \mu[n(x)]$, the conservation law of particles with the phenomenological law (1.40) can be rewritten as the well-known diffusion equation:

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial n}{\partial x} \right) \quad (1.41)$$

where the diffusion coefficient is $D = L \frac{\partial \mu}{\partial n}$.

C Limitations of nonequilibrium thermodynamics (NET)

Nonequilibrium thermodynamics uses a set of local variables whose global counterparts coincide with those defined at equilibrium. This choice clearly restricts its application domain to the macroscopic level, at typical length scales much larger than any molecular size. In such a situation, the inherent molecular nature of matter can be ignored and one can adopt a continuum description in terms of a few conserved fields. Whereas this approximation has been extremely useful in the characterisation of many irreversible processes, it is no longer valid for systems defined at the mesoscale when the typical time and length scales are such that the presence of fluctuations becomes relevant.

The linear character of the constitutive relations proposed by the theory should in principle be appropriate only when the magnitude of the gradients is small. In practice, linear relations have been proved to work well for many transport processes, even in the presence of large gradients. In contrast, for activated processes, the assumption of linearity fails. A first attempt to overcome such difficulties was pioneered by I. Prigogine and P. Mazur [20] who extended thermodynamic concepts to irreversible phenomena in systems with internal degrees of freedom. Building on these ideas, it is possible to develop a mesoscopic extension of the conventional nonequilibrium thermodynamics, as described in the following section, that is able to overcome such difficulties.

1.4.2 Mesoscopic nonequilibrium thermodynamics (MNET)

“To a chemist the entropy of a system is a *macroscopic* state function, i.e. a function of the thermodynamics variables of the system. In statistical mechanics, entropy is a *mesoscopic*³ quantity, i.e. a functional of the probability distribution function P . It is never a *microscopic* quantity, because on the microscopic level there is no *irreversibility*” (van Kampen [36]). Following closely the works of P. Mazur, J. M Rubi, D. Reguera and J. M. G Vilar [17–19], this section has the role to present such a mesoscopic formalism, the MNET framework.

The reduction of the observational time and length scales of a system usually entails an increase in the number of degrees of freedom that have not yet equilibrated and that therefore exert an influence in the overall dynamics of the system. The nonequilibrated degrees of freedom will be denoted by $\gamma = \{\gamma_i\}$ and may, for example, represent the velocity of a particle, the orientation of a spin, the size of a macromolecule, or any coordinate or order parameter whose values define the state of the system in a phase space. The characterisation at the mesoscopic level of the state of the system follows from $P(\gamma, t)$, the probability density of finding the system at the state $\gamma \in (\gamma, \gamma + d\gamma)$ at time t .

Gibbs entropy postulate The entropy of the system in terms of this probability can be expressed through the Gibbs entropy postulate [36]:

$$S = S_{eq} - k_B \int P(\gamma, t) \ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} d\gamma \quad (1.42)$$

where S_{eq} is the entropy of the system when the degrees of freedom γ are at equilibrium. If they are not, there is a contribution to the entropy that arises from deviations of the probability density $P(\gamma, t)$ from its equilibrium value $P_{eq}(\gamma)$.

³Here, and throughout the thesis the adjective mesoscopic has the same meaning as that given by van Kampen [16]: “The stochastic description in terms of macroscopic variables will be called mesoscopic. It comprises both the deterministic laws and the fluctuations about them”. Nowadays the term ‘mesoscopic’ is often used to denote the phenomena on the borderline between classical and quantum mechanics.

This expression (1.42) represents the starting point for the derivation of the entropy production σ , which gives further access to the dynamics of the mesoscopic degrees of freedom γ . Differentiating it in respect to time, the following expression is obtained:

$$\frac{dS}{dt} = -k_B \int \frac{\partial P(\gamma, t)}{\partial t} \ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} d\gamma \quad (1.43)$$

Conservation law The probability density is governed by the conservation law written for the γ space, where a flux $\mathbf{J}(\gamma, t)$ in the freedom degrees space γ -space has been introduced.

$$\frac{\partial P}{\partial t} = -\frac{\partial \mathbf{J}}{\partial \gamma} \quad (1.44)$$

Entropy production The conservation law (1.44) together with the Gibbs entropy postulate (1.43) lead to the derivation of the entropy production σ :

$$\frac{dS}{dt} = k_B \int \left[\frac{\partial}{\partial \gamma} \left(\mathbf{J}(\gamma, t) \ln \frac{P}{P_{eq}} \right) - \mathbf{J}(\gamma, t) \frac{\partial}{\partial \gamma} \left(\ln \frac{P}{P_{eq}} \right) \right] d\gamma \quad (1.45)$$

$$\Rightarrow \begin{cases} \mathbf{J}^s = -k_B \mathbf{J}(\gamma, t) \ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} \\ \sigma = -k_B \int \mathbf{J}(\gamma, t) \cdot \frac{\partial}{\partial \gamma} \left(\ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} \right) d\gamma \end{cases} \quad (1.46)$$

Note that the entropy production σ is a sum of products of the components of the probability flux \mathbf{J} and of the corresponding ‘‘thermodynamic forces’’ $\frac{\partial}{\partial \gamma} \left(\ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} \right)$. In this scheme, the thermodynamic forces are identified as the gradients in the space of mesoscopic variables of the logarithm of the ratio of the probability density to its equilibrium value.

Phenomenological law We will now assume a linear dependence between fluxes and forces and establish a linear relationship between them, where $L(\gamma, P(\gamma))$ is an Onsager coefficient, which, in general, depends on the state variable $P(\gamma)$ and on the mesoscopic coordinates γ .

$$\mathbf{J}(\gamma, t) = -k_B L(\gamma, P(\gamma)) \frac{\partial}{\partial \gamma} \left(\ln \frac{P(\gamma, t)}{P_{eq}(\gamma)} \right) \quad (1.47)$$

To derive this expression, locality in γ -space has been taken into account.

Fokker-Planck equation The resulting kinetic equation follows by substituting the expression for the flux (1.47) into the conservation law (1.44), where the diffusion coefficient

$D(\gamma)$ was also defined:

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \gamma} \left(DP_{eq} \frac{\partial P(\gamma, t)}{\partial \gamma P_{eq}(\gamma)} \right) \quad D(\gamma) \stackrel{def}{=} \frac{k_B L(\gamma, P)}{P} \quad (1.48)$$

It is important to stress that MNET provides a simple and direct method to determine the dynamics of a system from its equilibrium properties. In particular, by knowing the equilibrium thermodynamic potential of a system in terms of its relevant variables, one could easily derive the general form of the kinetics. The method proposed thus offers a general formalism able to analyse the dynamics of systems away from equilibrium.

In the following chapters we will illustrate its applicability by considering the mesoscopic degree of freedom the velocity \mathbf{u} , next to that of the orientation \mathbf{e} for a system of noninteracting uniform magnetisations \mathbf{m} .

Bibliography

- [1] M. N. BAIBICH, J. M. BROTO, A. FERT, F. N. V. DAU, F. PETROFF, P. ETIENNE, G. CREUZET, A. FRIEDERICH, and J. CHAZELAS, *Phys. Rev. Lett.* **61**, 2472 (1988).
- [2] G. BINASCH, P. GRÜNBERG, F. SAURENBACH, and W. ZINN, *Phys. Rev. B* **39**, 4828 (1989).
- [3] L. LANDAU and E. LIFSHITZ, *Phys. Z. Sowjetunion* **8**, 153 (1935).
- [4] T. L. GILBERT, *IEEE Transactions on Magnetics* **40**, 3443 (2004).
- [5] W. DÖRING, *Zeit. Naturforsch* **3a**, 373 (1948).
- [6] S. CHIKAZUMI, *Physics of Ferromagnetism*, Oxford Univ. Press, 1997.
- [7] F. H. DE LEEUW and J. M. ROBERTSON, *J. Appl. Phys.* **46**, 3182 (1975).
- [8] U. EBELS, L. D. BUDA, K. OUNADJELA, and P. E. WIGEN, Small amplitude dynamics of nonhomogenous magnetization distributions :The excitation spectrum of stripe domains, In *Spin dynamics in confined magnetic structures I* [37], pp. 167–219.
- [9] T. L. GILBERT, *Phys. Rev.* **100**, 1243 (1955).
- [10] W. F. BROWN, *Magnetostatic principles in ferromagnetism*, North-Holland Publishing Co., 1962.
- [11] A. H. MORRISH, *The physical principles of magnetism*, John Wiley and Sons, 1983.
- [12] W. F. BROWN JR., *Physical Review* **130**, 1677 (1963).

- [13] W. F. BROWN JR., *IEEE Trans. Magn.* **15**, 1196 (1979).
- [14] R. ZWANZIG, *Nonequilibrium statistical mechanics*, Oxford University Press, 2001.
- [15] L. NÉEL, *Ann. Geophys.* **5**, 99 (1949).
- [16] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, p. 57, In [38], third edition, 2007.
- [17] P. MAZUR, *Physica A* **274**, 491 (1999).
- [18] J. M. G. VILAR and J. M. RUBI, *Proc. Natl. Acad. Sci. USA* **98**, 11081 (2001).
- [19] D. REGUERA, J. M. RUBI, and J. M. G. VILAR, *J. Phys. Chem. B* **109**, 21502 (2005).
- [20] I. PRIGOGINE and P. MAZUR, *Physica* **19**, 241 (1953).
- [21] S. R. DEGROOT and P. MAZUR, *Non-Equilibrium Thermodynamics*, Dover Publications Inc. New York, second edition, 1984.
- [22] J. C. SLONCZEWSKI, *J. Magn. Magn. Mater.* **159**, L1 (1996).
- [23] L. BERGER, *Phys. Rev. B* **54**, 9353 (1996).
- [24] M. D. STILES and J. MILTAT, Spin transfer torque and dynamics, in *Spin dynamics in confined magnetic structures III*, volume **101**, pp. 225–308, Springer-Verlag Berlin Heidelberg, 2006.
- [25] J. Z. SUN, *IBM J. Res. and Dev.* **50**, 81 (2006).
- [26] D. C. RALPH and M. D. STILES, *J. Magn. Magn. Mater.* **320**, 1190 (2008).
- [27] J.-E. WEGROWE, S. M. SANTOS, M.-C. CIORNEI, H.-J. DROUHIN, and J. M. RUBI, *Physical Review B* **77**, 174408 (2008).
- [28] J.-E. WEGROWE, M.-C. CIORNEI, and H.-J. DROUHIN, *J. Phys.:Condens Matter* **19**, 165213 (2007).
- [29] I. PRIGOGINE and P. MAZUR, *Physica* **17** (1951).
- [30] L. C. WOODS, *The thermodynamics of fluid systems*, Calendron Press, 1975.
- [31] J. MILTAT, G. ALBUQUERQUE, and A. THIAVILLE, An introduction to micromagnetics in the dynamic regime, In *Spin dynamics in confined magnetic structures I* [37], pp. 1–34.

- [32] H. GOLDSTEIN, C. POOLE, and J. SAFKO, *Classical mechanics*, Addison Wesley, third edition, 2000.
- [33] W. T. COFFEY, Y. P. KALMYKOV, and J. T. WALDRON, *The Langevin Equation With Applications to Stochastic Problems in Physics, Chemistry and Electrical Engineering*, World Scientific, second edition, 2004.
- [34] D. KONDEPUDI and I. PRIGOGINE, *Modern Thermodynamics, From Heat Engines to Dissipative Structures*, John Wiley and Sons, 1998.
- [35] H. B. CALLEN, *Thermodynamics and an Introduction to Thermostatistics*, John Wiley and Sons, 1985.
- [36] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, p. 185, In [38], third edition, 2007.
- [37] B. HILLEBRANDS and K. OUNADJELA, *Topics Appl. Phys* **83** (2002).
- [38] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, Elsevier Science and Technology Books, third edition, 2007.

Chapter 2

Gilbert and Brown stochastic equations within NET

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Introduction

The purpose of this first chapter is to prove that the Gilbert equation [1], the Landau-Lifshitz equation [2] and the Brown stochastic equation, or otherwise called the rotational Fokker-Planck equation [3] can all be derived [4, 5] following a common classic framework, that of nonequilibrium thermodynamics (NET) [6, 7].

The chapter is structured as follows: The first section 2.1 concerns with the presentation of the model. It introduces thermodynamic variables associated to a system of noninteracting magnetic monodomain particles. Once the thermodynamic variables are introduced, the non equilibrium thermodynamics framework is applied to derive averaged dynamic and stochastic equations associated to the system of magnetic particles. The explicit application of the method is presented in section 2.2, the “NET framework“. As it is a deductive method, it has several cornerstones: the first and the second principle of thermodynamics, and conservation laws, all to be presented in subsection 2.2.1. With these as a foundation, the entropy production is derived defining the fluxes and their corresponding “thermodynamic forces“. A phenomenological law is imposed by the local equilibrium hypothesis together with the second principle of thermodynamics between the causes (the forces) and the effects (the fluxes). The law leads to the derivation of the Brown stochastic equation, the Landau-Lifshitz equation and the Gilbert equation meeting the purpose of this chapter (see subsections 2.2.3, 2.2.4, respectively 2.2.5).

2.1 Model

To model the dynamics of a monodomain particle subjected to thermal agitation, we study an ensemble of identical monodomain particles of magnetization \mathbf{m} , magnetic energy per unit volume $V(\theta, \phi)$ and thermostat temperature T . Each magnetization vector \mathbf{m} has the same origin O , a constant M_s modulus, an orientation $\mathbf{e} = (\theta, \phi)$ and is represented by its vector tip, a point on the surface of a unit radius sphere. If the magnetization \mathbf{m} is reoriented due to an applied magnetic field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$ or due to thermal fluctuations, its corresponding point moves on the sphere surface, and reciprocally.

Let Σ be such a statistical ensemble of N identical monodomain particles of volume v and let it be represented by points distributed over the unit sphere (fig. 2.1) with a density $n(\theta, \phi)$. The ensemble Σ can be divided in sub-ensembles $\Sigma_{\theta, \phi}$ including the magnetisations \mathbf{m} with orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$, i.e. which are confined within the solid angle $\delta V_{\theta, \phi}^{vol} = \sin\theta d\theta d\phi$. As the particles undergo changes of magnetization orientation, the representative points move on the sphere, and there is a net surface flux of representative points \mathbf{J}^n . The representative points move from one sub-ensemble $\Sigma_{\theta, \phi}$ to another sub-ensemble $\Sigma_{\theta+\Delta\theta, \phi+\Delta\phi}$. The probability of finding a particle with the magnetization orientation within the solid angle $\delta V_{\theta, \phi}^{vol}$ at a given time t is $dP(\theta, \phi, t) = \frac{n(\theta, \phi, t)}{N} \delta V_{\theta, \phi}^{vol}$.

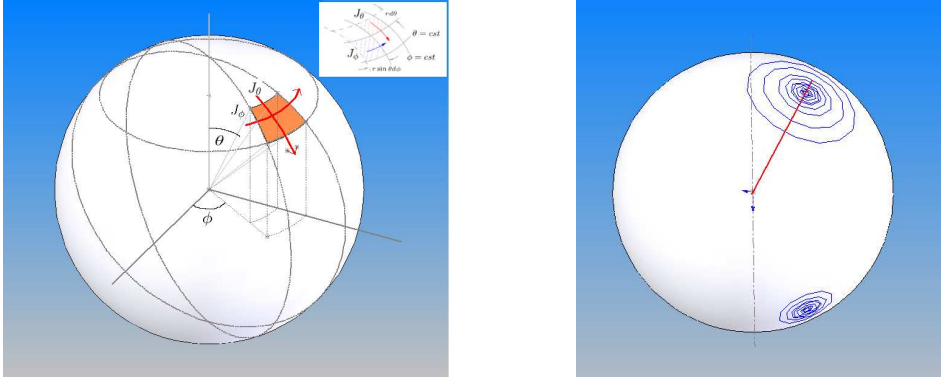


Figure 2.1: a) The figure from the left illustrates the flow of representative points over the unit sphere: J^θ and J^ϕ . b) The figure from the right illustrates a particular case of distribution of points on the sphere: the points are concentrated at two attractors, one with more particles than the other (asymmetric double well potential).

For almost every macroscopic system we can meaningfully assign a temperature T , and other thermodynamic variables to every "elemental volume" δV^{vol} . The entire system of magnetic particles Σ is viewed as a collection of subsystems $\Sigma_{\theta,\phi}$ interacting with each other. The sub-systems of representative points $\Sigma_{\theta,\phi}$ are described by extensive and intensive parameters. The extensive parameters are: the entropy $dS = s(\theta, \phi, t) \delta V_{\theta,\phi}^{vol}$, the number of points $dN = n(\theta, \phi, t) \delta V_{\theta,\phi}^{vol}$ and the energy $dE = e(\theta, \phi, t) \delta V_{\theta,\phi}^{vol}$, where s , e and n are volume densities of entropy, energy, respectively number of points. The flow (of points, energy, and entropy) is described by a two dimensional flux \mathbf{J} (\mathbf{J}^n , \mathbf{J}^e and \mathbf{J}^s): $\mathbf{J} = J^\theta \mathbf{e}_\theta + J^\phi \mathbf{e}_\phi$ and accounts for the flow of the corresponding magnetic moments relaxing or precessing along the coordinates (θ, ϕ) , where \mathbf{e} , $\boldsymbol{\theta}$, $\boldsymbol{\phi}$ are spherical unit vectors. The intensive parameters are the temperature T and the chemical potential $\mu(\theta, \phi)$ 'controlling' the flow of energy, respectively the number of points (particles).

2.2 NET framework

2.2.1 Conservation laws and the Gibbs relation

A Conservation laws

The conservation laws for the number, energy and entropy densities n , e , respectively s , of the particles contained in the sub-ensemble $\Sigma_{\theta,\phi}$ write:

$$\left\{ \begin{array}{l} \frac{\partial n}{\partial t} = -\frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^n \\ \frac{\partial e}{\partial t} = -\frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^e \\ \frac{\partial s}{\partial t} = -\frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^s + \sigma \end{array} \right. \Rightarrow \left\{ \begin{array}{l} \frac{\partial n}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left(J^{n-\theta} \sin\theta \right) - \frac{1}{\sin\theta} \frac{\partial J^{n-\phi}}{\partial \phi} \\ \frac{\partial e}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left(J^{e-\theta} \sin\theta \right) - \frac{1}{\sin\theta} \frac{\partial J^{e-\phi}}{\partial \phi} \\ \frac{\partial s}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial \theta} \left(J^{s-\theta} \sin\theta \right) - \frac{1}{\sin\theta} \frac{\partial J^{s-\phi}}{\partial \phi} + \sigma \end{array} \right. \quad (2.1)$$

where in contrast to the energy and number of particles, the entropy s is not a conserved extensive parameter. It is explicit by the presence of a nonnegative extra term $\sigma \geq 0$ next to the entropy flux \mathbf{J}^s , called the *entropy production*, where the nonnegativity is a consequence of the *first principle* of thermodynamics.

B The Gibbs relation

In most situations we may assume that *equilibrium* thermodynamic relations are valid for the thermodynamic variables assigned to each δV . This is the concept of *local equilibrium*. Then, the Gibbs relation valid for systems in equilibrium $dE = TdS - p\delta V + \sum_k \mu^k dN^k$, translates for systems *not* in equilibrium, *but* in local equilibrium to the following relation:

$$\frac{\partial s}{\partial t} = \frac{1}{T} \frac{\partial e}{\partial t} - \sum_k \frac{\mu^k}{T} \frac{\partial n^k}{\partial t} \quad (2.2)$$

In this equation the volume δV does not appear because s , e and n^k are volume densities.

In our particular case of one 'chemical' specie of particles, the local equilibrium hypothesis imposes the following Gibbs relation:

$$\frac{\partial s}{\partial t} = \frac{1}{T} \frac{\partial e}{\partial t} - \frac{\tilde{\mu}}{T} \frac{\partial n}{\partial t} \quad (2.3)$$

The chemical potential $\tilde{\mu}_{(\theta,\phi)}$ is the intensive parameter "controlling" directly the number of particles n of a sub-ensemble $\Sigma_{\theta,\phi}$. The thermal fluctuations affecting the number of particles n are accounted by a temperature dependent chemical potential $kT \ln n$, while

the magnetic field reorienting the magnetic particles from one subsystem to the other is accounted by a magnetic energy term vV (derived in the general case by S. R. De Groot and P. Mazur in Ref. [6]) :

$$\tilde{\mu} \equiv k_B T \ln(n) + vV(\theta, \phi) + \mu_0 \quad (2.4)$$

The second term vV represents the magnetic potential energy that defines the local magnetic field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$, v is the physical volume of one magnetic particle, and the third term μ_0 is a constant related to the chemical nature of the particles.

2.2.2 Thermokinetic equations

A The entropy production

Rewriting the first principle of thermodynamics (2.3) using the conservation laws (2.1), the entropy production σ is derived:

$$\frac{\partial s}{\partial t} = -\frac{1}{T} \frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^e + \frac{\tilde{\mu}}{T} \frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^n \quad (2.5)$$

\Leftrightarrow

$$\frac{\partial s}{\partial t} = -\frac{\partial}{\partial \mathbf{e}} \cdot \left(\frac{1}{T} \mathbf{J}^e - \frac{\tilde{\mu}}{T} \mathbf{J}^n \right) + \mathbf{J}^e \cdot \frac{\partial}{\partial \mathbf{e}} \left(\frac{1}{T} \right) - \mathbf{J}^n \cdot \frac{\partial}{\partial \mathbf{e}} \left(\frac{\tilde{\mu}}{T} \right) \quad (2.6)$$

The last equation (2.6) represents the conservation law for the entropy (2.1), leading to the expressions of the entropy flux \mathbf{J}^s and entropy production σ as:

$$\begin{cases} \mathbf{J}^s = \frac{1}{T} \mathbf{J}^e - \frac{\tilde{\mu}}{T} \mathbf{J}^n \\ \sigma = \mathbf{J}^e \cdot \frac{\partial}{\partial \mathbf{e}} \left(\frac{1}{T} \right) - \mathbf{J}^n \cdot \frac{\partial}{\partial \mathbf{e}} \left(\frac{\tilde{\mu}}{T} \right) \end{cases} \quad (2.7)$$

The entropy production σ is a sum of products between the fluxes \mathbf{J}^k and their corresponding “thermodynamic forces” \mathbf{F}^k [6]. Assuming, that all the magnetic particles \mathbf{m} are equivalently affected by thermal fluctuations, i.e. the temperature T is uniform all over the sphere, the first term from the right hand side of the entropy production equation (2.7) disappears.

B Phenomenological law

The local equilibrium hypothesis and the second law of thermodynamics $\sigma \geq 0$ are met (sufficient condition) when the entropy production is built as a quadratic form. A phenomenological symmetric positive tensor $\overline{\overline{L}}$ of Onsager transport coefficients $L_{ij}(\theta, \phi)$ is

introduced such that $J^i = \sum_j L_{ij} \frac{\partial \tilde{\mu}}{\partial e_j}$. The symmetry of the tensor $\overline{\overline{L}}$ is a reflection of systems symmetry, and is translated into the *Onsager reciprocity relations*: $L_{ij} = \pm L_{ji}$ [6]. The phenomenological law is then written:

$$\mathbf{J}^n = -\overline{\overline{L}} \cdot \frac{\partial \tilde{\mu}}{\partial \mathbf{e}} \Rightarrow \begin{cases} J^{n-\theta} = -L_{\theta\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - L_{\theta\phi} \frac{1}{\sin\theta} \frac{\partial \tilde{\mu}}{\partial \phi} \\ J^{n-\phi} = -L_{\phi\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - L_{\phi\phi} \frac{1}{\sin\theta} \frac{\partial \tilde{\mu}}{\partial \phi} \end{cases} \quad (2.8)$$

where $L_{\theta\phi} = -L_{\phi\theta}$. Let's introduce a new tensor $\overline{\overline{D}}$ of coefficients h' , g' and k' , which allows the rewriting of the phenomenological law (2.8) in a different form:

$$\mathbf{J}^n = -\overline{\overline{L}} \cdot \frac{\partial \tilde{\mu}}{\partial \mathbf{e}} = -\frac{n}{k_B T} \overline{\overline{D}} \cdot \frac{\partial \tilde{\mu}}{\partial \mathbf{e}} \quad (2.9)$$

where

$$\overline{\overline{D}} = \frac{k_B T}{n} \overline{\overline{L}} = \frac{k_B T}{v} \begin{pmatrix} h' & -g' & 0 \\ g' & \frac{k'v}{k_B T} & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (2.10)$$

$$h' = \frac{L_{\theta\theta}v}{n}; \quad g' = -\frac{L_{\theta\phi}v}{n} = \frac{L_{\phi\theta}v}{n}; \quad k' = L_{\phi\phi} \frac{k_B T}{n} \geq 0 \quad (2.11)$$

With the expression of the chemical potential (2.4), the magnetic analogous of Fick's diffusion law is derived, and the previously defined tensor $\overline{\overline{D}}$ is identified as a *diffusion* tensor:

$$\mathbf{J}^n = -\overline{\overline{D}} \cdot \left(\frac{\partial n}{\partial \mathbf{e}} + \frac{nv}{k_B T} \frac{\partial V}{\partial \mathbf{e}} \right) \quad (2.12)$$

$$\Leftrightarrow \begin{cases} J^{n-\theta} = - \left(h' \frac{\partial V}{\partial \theta} - \frac{g'}{\sin\theta} \frac{\partial V}{\partial \phi} \right) n - \left(h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} - \frac{g'}{\sin\theta} \frac{k_B T}{v} \frac{\partial n}{\partial \phi} \right) \\ J^{n-\phi} = - \left(g' \frac{\partial V}{\partial \theta} + \frac{k'}{\sin\theta} \frac{v}{k_B T} \frac{\partial V}{\partial \phi} \right) n - \left(g' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} + \frac{k'}{\sin\theta} \frac{\partial n}{\partial \phi} \right) \end{cases} \quad (2.13)$$

The flux \mathbf{J}^n represents the number of magnetic particles changing their orientation from $\mathbf{e} = (\theta, \phi)$ to $\mathbf{e} + d\mathbf{e} = (\theta + d\theta, \phi + d\phi)$ per unit of time, and is caused by the application of a conservative magnetic field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$ and by the nonuniform repartition of the magnetisations \mathbf{m} in the 4π solid angle. The magnetic field \mathbf{H} generates a drift flux $-\overline{\overline{D}} \cdot \frac{nv}{k_B T} \frac{\partial V}{\partial \mathbf{e}}$, while the nonuniform repartition generates a diffusion flux $-\overline{\overline{D}} \cdot \frac{\partial n}{\partial \mathbf{e}}$.

2.2.3 The Brown stochastic equation

The conservation law for the number of magnetic particles n (2.1) together with the assumption of a constant g' coefficient leads to the rotational Fokker-Planck equation obtained previously by Brown [3].

$$\frac{\partial n(\theta, \phi, t)}{\partial t} = -\frac{\partial}{\partial \mathbf{e}} \cdot \mathbf{J}^n = \frac{\partial}{\partial \mathbf{e}} \left[\overline{\overline{D}} \cdot \left(\frac{\partial n}{\partial \mathbf{e}} + \frac{n v}{k_B T} \frac{\partial V}{\partial \mathbf{e}} \right) \right] \quad (2.14)$$

\Leftrightarrow

$$\begin{aligned} \frac{\partial n(\theta, \phi)}{\partial t} = \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta \left[\left(h' \frac{\partial V}{\partial \theta} - \frac{g'}{\sin \theta} \frac{\partial V}{\partial \phi} \right) n + h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \right] \right\} \\ + \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \left\{ \left(g' \frac{\partial V}{\partial \theta} + \frac{k'}{\sin \theta} \frac{v}{k_B T} \frac{\partial V}{\partial \phi} \right) n + \frac{k'}{\sin \theta} \frac{\partial n}{\partial \phi} \right\} \quad (2.15) \end{aligned}$$

Whereas Brown derived the equation using a rather complicated formalism characteristic for stochastic processes, here the same equation was derived in a few lines using the non-equilibrium thermodynamics framework (NET). Also, the previously defined coefficients g' , h' and k' are identified to be the LL gyromagnetic coefficient, the LL damping coefficient, respectively the 'stochastic' coefficient introduced by Brown in its article [3].

2.2.4 The Landau-Lifschitz equation with diffusion

It is generally known that a flux \mathbf{J}^n can be written (following its definition), as the product between the density of particles n and their corresponding average velocity $\langle \frac{d\mathbf{e}}{dt} \rangle$. Let \mathbf{M} be the magnetization vector corresponding to the average velocity of the magnetic particles \mathbf{m} of orientation within $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$:

$$\mathbf{J}^n = n \left\langle \frac{d\mathbf{e}}{dt} \right\rangle = \frac{n}{M_s} \frac{d\mathbf{M}}{dt} \quad (2.16)$$

The expression of the flux \mathbf{J}^n together with the assumption that the diagonal coefficients of the diffusion tensor are equal, i.e. $h' = \frac{k' v}{k_B T}$, leads to the derivation of the Landau-Lifschitz equation *with* diffusion:

$$\frac{d\mathbf{M}}{dt} = -\frac{M_s}{n} \overline{\overline{D}} \cdot \left(\frac{\partial n}{\partial \mathbf{e}} + \frac{n v}{k_B T} \frac{\partial V}{\partial \mathbf{e}} \right) \quad (2.17)$$

where the diffusion tensor $\overline{\overline{D}}$ is written as:

$$\overline{\overline{D}} = \frac{k_B T}{v} \begin{pmatrix} h' & -g' & 0 \\ g' & h' & 0 \\ 0 & 0 & 0 \end{pmatrix} = \frac{k_B T}{v} h' (\overline{\overline{U}} - \mathbf{e}\mathbf{e}) + \frac{k_B T}{v} g' \mathbf{e} \times \overline{\overline{U}} \quad (2.18)$$

Knowing that $\overline{\overline{U}}$ is the unit tensor and $\mathbf{e} \times (\mathbf{b} \times \mathbf{e}) = (\overline{\overline{U}} - \mathbf{e}\mathbf{e}) \cdot \mathbf{b}$, the Landau-Lifshitz equation is recovered from the 'diffusion' equation (2.17):

$$\frac{d\mathbf{M}}{dt} = -h' \mathbf{M} \times \left[\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \left(\frac{k_B T}{v} \ln n + V \right) \times \mathbf{M} \right] - g' M_s \mathbf{M} \times \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \left(\frac{k_B T}{v} \ln n + V \right) \quad (2.19)$$

$$\frac{d\mathbf{M}}{dt} = g' M_s \mathbf{M} \times \mathbf{H}_{eff} + h' (\mathbf{M} \times \mathbf{H}_{eff}) \times \mathbf{M} \quad (2.20)$$

where the first and the second term in the right hand side of the equation (2.20) represent *precession*, respectively *longitudinal relaxation* and $\mathbf{H}_{eff} = \mathbf{H} - \frac{k_B T}{nvM_s} \nabla n$ represents an an effective magnetic field including the diffusion term.

2.2.5 The Gilbert equation with diffusion

The equation (2.20) can be also written in the Gilbert form by performing a cross product $\times \mathbf{M}$. The Gilbert equation is derived together with the Gilbert damping parameter η , identified as a function of the LL gyromagnetic and damping coefficients g' , respectively h' :

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left(\mathbf{H}_{eff} - \eta \frac{d\mathbf{M}}{dt} \right) \quad (2.21)$$

$$g' = \frac{\gamma}{(1 + \alpha^2) M_s} \quad h' = \alpha g' \quad \alpha = \gamma \eta M_s \quad (2.22)$$

where the dimensionless damping coefficient α has been introduced.

Conclusions

The purpose of this chapter was to prove that the application of the framework of nonequilibrium thermodynamics (NET) to an ensemble of magnetic particles Σ leads not only to the derivation of the averaged dynamic behaviour of one monodomain magnetic particle, but as well to the derivation of its stochastic behaviour. The Gilbert and the Landau-Lifshitz equations, as well as the Brown's rotational Fokker-Planck equation can be derived within a common classic simple framework: non-equilibrium thermodynamics (NET).

The ensemble Σ consists of monodomain magnetic particles of orientation $\mathbf{e} = (\theta, \phi)$,

changed either by an applied magnetic field \mathbf{H} , either by thermal fluctuations (temperature T). State variables such as: the entropy density s , the energy density e and the number density of particles n are associated to each sub-ensemble $\Sigma_{\theta,\phi}$ of magnetic particles having the orientation within the solid angle $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$. The NET method is applied for each subsystem $\Sigma_{\theta,\phi}$ supposed to be in *local equilibrium*. With cornerstones as: the first and the second principle of thermodynamics, and conservation laws, the NET method is applied. It leads to the derivation of a phenomenological law between the flux of magnetic particles \mathbf{J}^n and its corresponding *thermodynamic force* $\frac{\partial \tilde{\mu}}{\partial \mathbf{e}}$. In this way, the flux \mathbf{J}^n is expressed as the sum of a drift flux depending on the magnetic field \mathbf{H} , and of a diffusion flux caused by the nonuniform repartition of the orientations of the magnetic particles. The Gilbert, the Landau-Lifshitz and as well the Brown stochastic equations are derived starting from this law.

Bibliography

- [1] T. L. GILBERT, *IEEE Transactions on Magnetics* **40**, 3443 (2004).
- [2] L. LANDAU and E. LIFSHITZ, *Phys. Z. Sowjetunion* **8**, 153 (1935).
- [3] W. F. BROWN JR., *Physical Review* **130**, 1677 (1963).
- [4] J.-E. WEGROWE, M.-C. CIORNEI, and H.-J. DROUHIN, *J. Phys.:Condens Matter* **19**, 165213 (2007).
- [5] J.-E. WEGROWE, M.-C. CIORNEI, and H.-J. DROUHIN, *Thermokinetics of Spin-Dependent Transport and Ferromagnetism*, chapter 15, American Scientific, 2009.
- [6] S. DEGROOT and P. MAZUR, *Non-Equilibrium Thermodynamics*, Dover Publications Inc. New York, second edition, 1984.
- [7] D. KONDEPUDI and I. PRIGOGINE, *Modern Thermodynamics, From Heat Engines to Dissipative Structures*, John Wiley and Sons, 1998.

Chapter 3

The generalised Gilbert equation Inductive approach

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Introduction

As proved in the previous chapter and in earlier works [1, 2], associating two degrees of freedom to each uniform magnetisation $\mathbf{m} = M_s \mathbf{e}$ from an ensemble of identical magnetic particles with different orientations $\mathbf{e} = (\theta, \phi)$ is enough to derive the Gilbert [3], the Landau-Lifshitz [4] and the Brown [5] stochastic equations.

The purpose of this chapter is to go beyond these equations, by considering other degrees of freedom [6, 7], four and not only two: the orientation $\mathbf{e} = (\theta, \phi)$, and the velocity $\mathbf{u} = (u_\theta, u_\phi)$. With two fundamental hypothesis, a model is built in the framework of the theory of mesoscopic nonequilibrium thermodynamics MNET [8] to derive an averaged dynamic equation governing the motion of a magnetic particle submitted to an applied magnetic field \mathbf{H} and to thermal fluctuations. The theory leads to a generalised Gilbert equation characterising the magnetisation dynamics, where a mass is associated to each magnetisation inertia, and not to that of the matter inertia.

The structure of the chapter presents as follows: the first section 3.1 familiarises the reader with the model. In order to characterise the dynamics of a magnetic particle in contact to the thermal bath, the average behaviour of a statistical ensemble of identical magnetic particles of uniform magnetisation \mathbf{m} is studied. The properties of the ensemble will be hence introduced. Once the model established, the following section 3.2 pursues the theoretical scheme of MNET to derive an averaged dynamic equation. A detailed analysis of the scheme's results, leads to the introduction of a relaxation time τ separating two regimes: the diffusion and the inertial regime (see section 3.3). It will be shown that the traits of the diffusion regime ($t \gg \tau$) are the Gilbert and the Brown's stochastic equations, while for the inertial regime ($t \ll \tau$) is a generalised Gilbert equation.

3.1 The model

The ergodic property¹ states: "The time average of a single Brownian particle equals the ensemble average of several identical non-interacting Brownian particles". In order to model the average dynamics of a uniform magnetisation subjected to thermal fluctuations, an ensemble average of identical non-interacting magnetic particles of uniform magnetisations \mathbf{m} and potential energy per unit volume $V(\theta, \phi)$ is studied. Each magnetisation vector \mathbf{m}

¹"One may actually observe a large number of Brownian particles and average the result; that means that one really has a physical realisation of the ensemble (provided the particles do not interact). One might also observe one and the same particle on successive days; the results will be the same if one assumes that sections of the trajectory that lie 24 hours apart are statistically independent. In practice, one simply observes the trajectory of a single particle during a long time. The idea is that irregularly varying function may be cut into a collection of long time intervals and that this collection can serve as the ensemble that defines the stochastic process. The condition for this "self-averaging" to work is that the behaviour of the function during one interval does not affect the behaviour during the next interval. If this is so the time average equals the ensemble average and the process is called *ergodic*", N. G. van Kampen in [9].

has the same origin O , a constant M_s modulus, a physical volume v and is represented by its vector tip, a point on the surface of a M_s radius sphere. If the magnetisation \mathbf{m} is reoriented due to the applied magnetic field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$, its corresponding point moves on the sphere surface, and reciprocally.

One magnetic particle is identified in phase space by its orientation (θ, ϕ) or $\mathbf{m} = M_s \mathbf{e}$ and by its velocity $\mathbf{u} = M_s \dot{\mathbf{e}} = M_s \dot{\theta} \boldsymbol{\theta} + M_s \dot{\phi} \sin \theta \boldsymbol{\phi} = u_\theta \boldsymbol{\theta} + u_\phi \boldsymbol{\phi}$. The number of magnetic particles with the orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ and the velocity $(\mathbf{u}, \mathbf{u} + d\mathbf{u})$ is represented by the distribution function $f(\mathbf{e}, \mathbf{u})$. It is assumed that f vanishes for infinite velocities $\lim_{\mathbf{u} \rightarrow \pm\infty} f(\mathbf{e}, \mathbf{u}) = 0$. The number of magnetic particles with the orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ is represented by the density of particles $n(\mathbf{e})$, where $n(\mathbf{e}) = \int f(\mathbf{e}, \mathbf{u}) d\mathbf{u}$. The average velocity of the magnetisations \mathbf{e} with the orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ is defined as $n\bar{\mathbf{M}} = \int f(\mathbf{e}, \mathbf{u}) \mathbf{u} d\mathbf{u}$. The velocity of one magnetic particle is either changed due to the applied magnetic field \mathbf{H} , either due to the interaction with the heat bath, modelled through a phase space flux $\mathbf{J}^{\mathbf{u}}$, whose value for infinite velocities is zero $\lim_{\mathbf{u} \rightarrow \pm\infty} \mathbf{J}^{\mathbf{u}} = 0$.

Each magnetisation \mathbf{m} has a mass m , *not* related to the inertia of matter, but to that of inertia of the magnetisation. Two hypothesis accompany the model:

- (a). the magnetisation dynamics follows a Newtonian type of law: $\mathbf{H} = m \frac{d\mathbf{u}}{dt}$
- (b). the kinetic energy \mathcal{K} of one magnetic particle is proportional to the square of its velocity \mathbf{u} : $\mathcal{K} = m\mathbf{u}^2/2$.

It is worth mentioning that these two hypothesis implicitly impose an inertia of the magnetisation centered in one point, as if it were its magnetic centre of mass (inertia). If it weren't the case of a magnetic centre of mass, but instead that of a complex magnetic inertia tensor $\bar{\bar{I}}$ associated with the magnetisation \mathbf{m} then, different hypothesis would have been necessary; for example, the associated kinetic energy \mathcal{K} wouldn't have been simply $m\mathbf{u}^2/2$, but instead $\mathbf{L}\mathbf{L} : \bar{\bar{I}}^{-1}/2$, where \mathbf{L} would have been its angular momentum. Precisely this point, of associating a complex magnetic inertia tensor since the beginning of the model, will be treated in the next chapter. In this chapter however, the simpler case of the magnetic centre of mass, furtherly extended to a more complex magnetic inertia $\bar{\bar{I}}$ will be treated. The magnetic centre of mass approach leads to the same main results, but within a simpler mathematical formalism. To separate the magnetic centre of mass formalism from that of a complex magnetic inertia tensor formalism, the terms *point-like*, respectively *body-like* are used for differentiation.

Extensive parameters are associated to the volume elements $(\mathbf{e}, d\mathbf{e})$ and/or $(\mathbf{e}, \mathbf{u}, d\mathbf{e}, d\mathbf{u})$: a) the density of magnetic particles $n(\mathbf{e})$, respectively $f(\mathbf{e}, \mathbf{u})$; b) the density of entropy $s(\mathbf{e})$; c) kinetic and potential energy densities $\int f m \mathbf{u}^2 d\mathbf{u}$, respectively $\int f V(\mathbf{e}) d\mathbf{u} = n(\mathbf{e}) V(\mathbf{e})$, as well as an intensive parameter: the temperature T . We suppose the heat bath affects equivalently all the orientations \mathbf{e} of the magnetic particles, i.e. T is considered uniform all over the sphere.

With these characteristics, we proceed to the derivation of the generalised Gilbert equation. The scheme of MNET is used in order to establish phenomenological laws describing the interaction of the magnetic particles with the thermal bath. The irreversible process of energy dissipation from the magnetic particles towards the bath is represented by dissipative forces and fluxes. To determine their explicit expressions, the entropy production $\sigma(\boldsymbol{\alpha})$, 'establishing' the irreversible processes has to be first determined.

Next section: The purpose of the next section is to derive the expression for the entropy production, the dissipative forces and finally, the magnetisation's dynamic and stochastic equations in terms of the magnetisation \mathbf{M} corresponding to the average velocity $\dot{\mathbf{M}}$.

3.2 MNET framework

Two cornerstones lie at the basis of the MNET framework: the conservation law (3.1) and the Gibbs entropy postulate (3.4) (subsection 3.2.1). Starting with these, the entropy production σ is derived and the dissipative force and flux are determined in subsection "Kinetic equations" 3.2.2. The second law of thermodynamics and the hypothesis of local equilibrium impose a phenomenological law between the cause (the dissipative force) and the effect (the flux). As a consequence, an Onsager coefficient is introduced as a proportionality coefficient between the two. With this law, the magnetisation's dynamic equation is derived in terms of the magnetisation \mathbf{M} in subsection 3.2.3. A cosmetic treatment performed on these results leads to the generalised Gilbert equation to be presented in the next section 3.3.

3.2.1 The Boltzmann equation and the Gibbs entropy postulate

A The Boltzmann equation

The first cornerstone of the MNET framework is the conservation equation for the number of particles $f(\mathbf{e}, \mathbf{u}, t)$ of orientation $(\mathbf{e}, d\mathbf{e})$ and velocity $(\mathbf{u}, d\mathbf{u})$, or otherwise called the Boltzmann equation [10, 11], and is written as:

$$\mathcal{D}f = \frac{\partial f}{\partial t} + \frac{\mathbf{u}}{M_s} \cdot \frac{\partial f}{\partial \mathbf{e}} + \frac{\mathbf{H}}{m} \cdot \frac{\partial f}{\partial \mathbf{u}} = \left(\frac{\partial f}{\partial t} \right)_c = C \quad (3.1)$$

The quantity $C = \left(\frac{\partial f}{\partial t} \right)_c$ represents the rate of change in the velocity-distribution function $f(\mathbf{e}, \mathbf{u}, t)$ for a *fixed* orientation \mathbf{e} due to the magnetic particles interactions with the thermal bath (No interaction between the magnetic particles), referred as the *collision term*. The conservation of the number of magnetic particles at interactions with the bath ('collisions')

allows the collision term C to be written as a divergence of a 'collision flux' $\mathbf{J}^{\mathbf{u}}$ (see Appendix 3.A for more details).

$$\mathcal{D}f = \left(\frac{\partial f}{\partial t} \right)_c = - \frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} \quad (3.2)$$

The conservation equation writes as:

$$\frac{\partial f(\mathbf{u}, \mathbf{e}, t)}{\partial t} = - \frac{\mathbf{u}}{M_s} \cdot \frac{\partial f}{\partial \mathbf{e}} - \frac{\mathbf{H}}{m} \cdot \frac{\partial f}{\partial \mathbf{u}} - \frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} \quad (3.3)$$

B The Gibbs entropy postulate

The second cornerstone of the MNET framework is represented by the Gibbs entropy postulate, which defines the expression for the entropy density ρs . As in kinetic theory and statistical physics, it is expressed as [12–14]:

$$Tns = -k_B T \int f \ln \frac{f}{f^{l.eq.}} d\mathbf{u} + T\rho s^{eq} \quad (3.4)$$

where $f^{l.eq.}$ is the local equilibrium distribution, meaning the local Maxwellian

$$f^{l.eq.}(\mathbf{e}, \mathbf{u}, t) = \exp \left(\frac{\mu_{l.eq.} - mu^2/2 - V(\mathbf{e})}{k_B T/v} \right) \quad (3.5)$$

with $\mu_{l.eq.}$ the chemical potential of particles at local equilibrium. Written differently eq. (3.4), takes the form:

$$Tns = - \int f \left(\frac{k_B T}{v} \ln f + \frac{mu^2}{2} + V(\mathbf{e}) - \mu^{eq} \right) d\mathbf{u} \quad (3.6)$$

3.2.2 Thermokinetic equations

A Conservation laws

A couple of conservation laws will be found useful in a later time, which will be derived here from the conservation law (3.3).

Number of particles Considering the case of a potentially driven field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V(\mathbf{e})}{\partial \mathbf{e}}$ (not a damping type field, but a conservative one), the conservation equation for the density of particles $n(\mathbf{e})$ is derived starting from the conservation law (3.3):

$$\frac{\partial n(\mathbf{e})}{\partial t} = \int \frac{\partial f}{\partial t} d\mathbf{u} = - \frac{1}{M_s} \frac{\partial n \dot{\mathbf{M}}}{\partial \mathbf{e}} \quad (3.7)$$

Momentum $\rho\dot{\mathbf{M}}$ The conservation law for the average magnetic 'momentum' $\rho\dot{\mathbf{M}}$ is derived, starting also from the conservation law (3.3):

$$\begin{aligned}\frac{\partial \rho\dot{\mathbf{M}}}{\partial t} &= \int \frac{\partial f}{\partial t} \mathbf{u} d\mathbf{u} \\ &= -\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \int m f(\mathbf{u} - \dot{\mathbf{M}})(\mathbf{u} - \dot{\mathbf{M}}) d\mathbf{u} - \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \cdot (\rho\dot{\mathbf{M}}\dot{\mathbf{M}}) - n \frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}} + \int m \mathbf{J}^{\mathbf{u}} d\mathbf{u} \\ &\Rightarrow \rho \frac{d\dot{\mathbf{M}}}{dt} = -\frac{1}{M_s} \frac{\partial \overline{P}}{\partial \mathbf{e}} - n \frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}} + \int m \mathbf{J}^{\mathbf{u}} d\mathbf{u}\end{aligned}\quad (3.8)$$

where ρ stands for $\rho = mn$ and P for the pressure of the dilute 'gas' of magnetic particles.

$$\overline{P} \stackrel{def}{=} \int m f(\mathbf{u} - \dot{\mathbf{M}})(\mathbf{u} - \dot{\mathbf{M}}) d\mathbf{u} \quad (3.9)$$

The dynamic equation (3.8) simply states: the average dynamics of the magnetisations is controlled by the magnetic field $\mathbf{H} = -\frac{1}{M_s} \frac{\partial V}{\partial \mathbf{e}}$, by the interactions with the heat bath presented in a form of a damping field $\mathbf{R} = \int m \mathbf{J}^{\mathbf{u}} d\mathbf{u}$ (see Appendix 3.A for more details) and by the pressure of the magnetic 'gas' \overline{P} .

B The entropy production σ

When equation (3.6) is derived in respect to time, reversible and irreversible processes are identified. The first ones contribute to the entropy flux \mathbf{J}^s , while the latter ones contribute to the production of entropy σ . The dissipation in terms of the collision flux $\mathbf{J}^{\mathbf{u}}$ is expected to appear as an irreversible process, hence in the analytical expression for the entropy production σ . (see appendix 3.B for explicit calculations)

$$T \frac{\partial ns}{\partial t} = \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \underbrace{\int f \mathbf{u} \left(\frac{k_B T}{v} \ln f + \frac{mu^2}{2} + V(\mathbf{e}) - \mu^{eq} \right) d\mathbf{u}}_{-\mathbf{J}^s} - \int \mathbf{J}^{\mathbf{u}} \left(\frac{k_B T}{vf} \frac{\partial f}{\partial \mathbf{u}} + m\mathbf{u} \right) d\mathbf{u} \quad (3.10)$$

$$\mathbf{J}^s = - \int f \mathbf{u} \left(\frac{k_B T}{v} \ln f + \mu^{l.eq.} \right) d\mathbf{u} - \mathbf{q} - \frac{\rho\dot{\mathbf{M}}^2}{2} \dot{\mathbf{M}} - nV \dot{\mathbf{M}} - \overline{P} \cdot \dot{\mathbf{M}} - \rho u \dot{\mathbf{M}} \quad (3.11)$$

Fluxes of heat \mathbf{q} , kinetic energy $\frac{\rho\dot{\mathbf{M}}^2}{2} \dot{\mathbf{M}}$, potential energy $nV \dot{\mathbf{M}}$, 'mechanical' energy $\overline{P} \cdot \dot{\mathbf{M}}$, internal energy $\rho u \dot{\mathbf{M}}$ and a purely entropic flux $\int f \mathbf{u} \left(\frac{k_B T}{v} \ln f + \mu^{l.eq.} \right) d\mathbf{u}$ contribute to

the entropy flux \mathbf{J}^s . The heat flux \mathbf{q} and the internal energy density ρu were defined as:

$$\mathbf{q} \stackrel{def}{=} \int (\mathbf{u} - \dot{\mathbf{M}}) \cdot f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} \quad \rho u \stackrel{def}{=} \int f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} \quad (3.12)$$

The remaining expression of the entropy increase in respect to time (3.10) is the entropy production σ . It consists of a product of two factors. One is a flow quantity, the 'collision' flux \mathbf{J}^u , already introduced in the conservation laws (3.3) and (3.8). The other factor is related to a gradient of an intensive parameter expressed as: $\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{u}} + m\mathbf{u}$. These quantities which multiply the fluxes in the expression for the entropy production are called *thermodynamic forces*, and represent the causes for their effects - the fluxes. The presence of the collision flux \mathbf{J}^u was expected in the expression of the entropy production σ , as it is related to the irreversible process of momentum exchange between the ensemble of magnetic particles \mathbf{m} and the heat bath, meaning to the *dissipation* process.

$$T\sigma = - \int \mathbf{J}^u \cdot \left(\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{u}} + m\mathbf{u} \right) d\mathbf{u} \quad (3.13)$$

C Phenomenological law

In conformity to the second law of thermodynamics $\sigma \geq 0$, assuming locality and isotropy in the phase space (\mathbf{e}, \mathbf{u}) , a phenomenological relationship between the collision flux \mathbf{J}^u and its cause, the thermodynamic force $\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{u}} + m\mathbf{u}$ imposes. An Onsager positive coefficient $L_{\mathbf{u}\mathbf{u}}$ is introduced as a proportionality coefficient:

$$\mathbf{J}^u = -L_{\mathbf{u}\mathbf{u}} \left(\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{u}} + m\mathbf{u} \right) \quad (3.14)$$

Defining a damping coefficient η as $\eta = \frac{m^2}{f} L_{\mathbf{u}\mathbf{u}}$, the flux \mathbf{J}^u takes the form:

$$\mathbf{J}^u = -\frac{\eta}{m} \left(f\mathbf{u} + \frac{k_B T}{m v} \frac{\partial f}{\partial \mathbf{u}} \right) \quad (3.15)$$

Couplings: It is worth mentioning that here, it was closely followed the MNET approach presented by J. M. Rubi and A. Perez-Madrid in their article on "Inertial effects in non-equilibrium thermodynamics" [12]. An equivalent MNET approach could have been followed, the one presented by J. M. G. Vilar and J. M. Rubi in their article on "Thermodynamics "beyond" local equilibrium" [15], leading to the same Fokker-Planck equation. However, it has to be annotated that the velocity fluxes introduced in both approaches \mathbf{J}^u are different. They are implicitly defined by the conservation law of the number of particles f . The different definitions lead to the introduction of only one Onsager coefficient in the

first approach (the one that we followed), while extra couplings are necessary in the second approach.

3.2.3 The dynamic equation of the magnetisation \mathbf{M}

A Space-fixed frame

With the collision flux expression (3.15), the dynamic equation (3.8) is rewritten:

$$m \frac{d\dot{\mathbf{M}}}{dt} = \mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \quad (3.16)$$

The up-above equation (3.16) represents the averaged dynamic behaviour of one magnetic particle submitted to a conservative magnetic field \mathbf{H} and to interactions with the thermal bath expressed in terms of:

- (a). an averaged damping field $\frac{1}{n} \mathbf{R} = \frac{1}{n} \int C m \mathbf{u} d\mathbf{u} = \frac{1}{n} \int m \mathbf{J}^u d\mathbf{u} = \eta \dot{\mathbf{M}}$;
- (b). a pressure gradient $\frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}}$ - a 'field' which at long time scale limit (see subsection 3.3.1), redirects the magnetic particles from the highly concentrated areas, big $n(\mathbf{e})$, towards the lowest concentrated areas, low $n(\mathbf{e})$. This term is actually the macroscopic field at the origin of the diffusion term $-k' \nabla n$ introduced by Brown in its stochastic Fokker-Planck equation [5].

It is worth pointing out that the equation (3.16) (without the pressure gradient term) is derivable from the equation (23) of Gilbert's article [3], if the kinetic energy \mathcal{T} introduced there, is explicitly written as: $\mathcal{T} = \frac{m \dot{\mathbf{M}}^2}{2}$.

B Body-fixed frame

In order to derive the Gilbert and the generalised Gilbert equation, a passage from the space-fixed frame to a rotating frame, called the *body-fixed* frame is necessary. Anticipating the next chapter: the passage from one frame to the other is mainly due to the magnetic inertia tensor \bar{I} having constant components only in the frame moving along with the magnetisation vector \mathbf{M} .

As the magnetisation \mathbf{M} rotates, an angular velocity $\boldsymbol{\Omega}$ (see Appendix 3.C.1 for its properties) and a rotating frame $Ox'y'z'$ (body-fixed frame) having as $Oz'//\mathbf{e}$ axis the orientation of the magnetisation are associated to its movement (see Fig. 3.1) . The dynamic equation (3.16) is rewritten in respect to the body frame $Ox'y'z'$, which takes the form of a balance of fields: the applied field \mathbf{H} together with the damping field $-\eta \dot{\mathbf{M}}$ and the pressure gradient field $\frac{\partial \bar{P}}{\partial \mathbf{e}}$ counter a "centrifugal field" $-m \boldsymbol{\Omega} \times \frac{d\mathbf{M}}{dt}$ and an "Euler field"

$$-m \frac{d\Omega}{dt} \times \mathbf{M}.$$

$$0 = \mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} - m \Omega \times \frac{d\mathbf{M}}{dt} - m \frac{d\Omega}{dt} \times \mathbf{M} \quad (3.17)$$

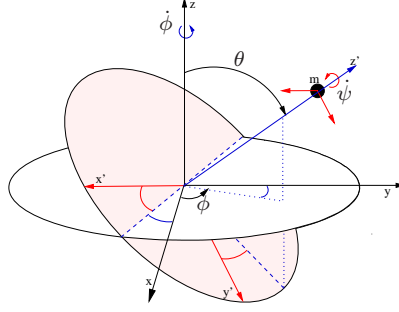


Figure 3.1: The averaged dynamics of one magnetic particle \mathbf{M} is a superposition of three elementary rotations: a) the rotation around the mobile axis Oz' with $\dot{\psi}$; b) the rotation around the fixed Oz axis with $\dot{\phi}$; and c) the rotation around Ox'' (the blue dashed line) with $\dot{\theta}$. The vector corresponding to the average velocity $\dot{\mathbf{M}}$, a magnetisation \mathbf{M} is parallel with the Oz' (\mathbf{e}) axis of the mobile frame.

Angular momentum conservation law A cross product of the last equation (3.17) with \mathbf{M} gives the conservation law of the angular momentum \mathbf{L} (3.21). With the following identity (3.18), its explicit form is derived, where \bar{U} is the unit tensor.

$$\mathbf{a} \times (\mathbf{b} \times \mathbf{a}) = \mathbf{b} \cdot |\mathbf{a}|^2 - \mathbf{a} \cdot (\mathbf{a} \cdot \mathbf{b}) = |\mathbf{a}|^2 \cdot \left[\left(\bar{U} - \frac{\mathbf{a}\mathbf{a}}{|\mathbf{a}|^2} \right) \cdot \mathbf{b} \right] \quad (3.18)$$

$$\underbrace{mM_s^2 (\bar{U} - \mathbf{e}\mathbf{e})}_{\bar{I}} \cdot \frac{d\Omega}{dt} - m \frac{d\mathbf{M}}{dt} (\mathbf{M} \cdot \Omega) = \mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.19)$$

$$\bar{I} \cdot \frac{d\Omega}{dt} + \Omega \times \left[\underbrace{mM_s^2 (\bar{U} - \mathbf{e}\mathbf{e})}_{\bar{I}} \cdot \Omega \right] = \mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.20)$$

$$\left(\frac{d\mathbf{L}}{dt} \right)_s = \left(\frac{d\mathbf{L}}{dt} \right)_b + \Omega \times \mathbf{L} = \mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.21)$$

where s and b indexes refer to time derivatives defined in respect to the space-fixed frame, respectively the body fixed-frame.

The expression $mM_s^2 (\overline{\overline{U}} - \mathbf{ee})$ represents the axial inertia tensor $\overline{\overline{I}}^{pt}$, with symmetry axis the orientation of the magnetic particle $\mathbf{M} = M_s \mathbf{e}$. Written by components in the body-fixed frame (b index), the tensor takes the following shape:

$$\overline{\overline{I}}_b^{pt} = \begin{pmatrix} mM_s^2 & 0 & 0 \\ 0 & mM_s^2 & 0 \\ 0 & 0 & 0 \end{pmatrix} = \begin{pmatrix} I_1^{pt} & 0 & 0 \\ 0 & I_1^{pt} & 0 \\ 0 & 0 & I_3^{pt} \end{pmatrix} = I_1^{pt} (\overline{\overline{U}} - \mathbf{ee}) + I_3^{pt} \mathbf{ee} \quad (3.22)$$

where the principal momenta of inertia $I_1^{pt}, I_2^{pt} = I_1^{pt}$ and I_3^{pt} associated to rotations of the center of mass m around Ox' , Oy' , respectively Oz' axis were introduced. The equality between the two principal momenta of inertia I_1^{pt} and I_2^{pt} was expected due to the axial symmetry of the system around the Oz' axis (or the unit vector \mathbf{e}). The third momenta of inertia $I_3^{pt} = 0$ is equal to zero, as the two hypothesis of the model, the Newtonian type of law and the expression for the kinetic energy (see section 3.1), impose a magnetic centre of mass (point-like).

Body-like magnetic mass (Not point-like) However, a general case would impose a repartition of mass respecting the magnetic properties of the particles. One magnetic particle has a symmetry axis given by the magnetisation orientation \mathbf{e} , imposing an inertial tensor with the expression $\overline{\overline{I}} = I_1 (\overline{\overline{U}} - \mathbf{ee}) + I_3 \mathbf{ee}$, which written by components in the body-fixed frame is given by the formula (3.23).

$$\overline{\overline{I}}_b^{bd} = \begin{pmatrix} I_1 & 0 & 0 \\ 0 & I_1 & 0 \\ 0 & 0 & I_3 \end{pmatrix} \quad (3.23)$$

It is necessary to point out, that the centre of mass for such an inertia tensor, is found on the axis given by the magnetisation vector $\mathbf{M} = M_s \mathbf{e}$. Supposing the angular momentum conservation law (3.21) still holds at changing the inertia momentum tensor expression from the centre of mass inertia tensor $\overline{\overline{I}}^{pt}$ to the body-like inertia tensor $\overline{\overline{I}}^{bd}$, the law written by

components in the body-fixed frame writes as:

$$\begin{cases} I_1 \dot{\Omega}_1 - \Omega_2 \Omega_3 (I_1 - I_3) = -M_s H_2 - \frac{I_1}{\tau} \Omega_1 + \left(\frac{1}{n} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right)_2 \\ I_1 \dot{\Omega}_2 - \Omega_1 \Omega_3 (I_3 - I_1) = +M_s H_1 - \frac{I_1}{\tau} \Omega_2 - \left(\frac{1}{n} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right)_1 \\ I_3 \dot{\Omega}_3 = 0 \end{cases} \quad (3.24)$$

where the relaxation time τ has been introduced.

$$\tau \stackrel{def}{=} \frac{I_1}{\eta M_s^2} \quad (3.25)$$

The conservation of the axial angular momentum $L_3 = \mathbf{L} \cdot \mathbf{e} = I_3 \Omega_3$, as seen in the last line of the system of equations (3.24) is the consequence of an applied torque $\mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right)$ always perpendicular on the magnetisation $\mathbf{M} = M_s \mathbf{e}$.

Angular momentum conservation law in terms of $\boldsymbol{\Omega}$ The same conservation law (3.21) and its equivalent, the system of equations (3.24) can be written also vectorially, in terms of the angular velocity $\boldsymbol{\Omega}$ (see appendix 3.C.2 for further details):

$$\frac{d\boldsymbol{\Omega}}{dt} = -\bar{\beta}_r \cdot \boldsymbol{\Omega} + \frac{1}{I_1} \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.26)$$

where a damping tensor $\bar{\beta}_r$ was introduced as:

$$\bar{\beta}_r = \left[\frac{\eta M_s^2}{I_1} (\bar{\mathbf{U}} - \mathbf{e}\mathbf{e}) - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 \mathbf{e} \times \bar{\mathbf{U}} \right] = \begin{pmatrix} \tau^{-1} & \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & 0 \\ -\left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & \tau^{-1} & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (3.27)$$

Next section: The purpose of the next section is to prove that the Gilbert, the Landau-Lifshitz (LL) and Brown's stochastic equations can be obtained by associating an axial magnetic inertia tensor ($I_1 = I_2 \neq 0$ and $I_3 \neq 0$) to the magnetisation $\mathbf{M} = M_s \mathbf{e}$. It is also proved that a cosmetic treatment performed on the angular momentum conservation law, written either in terms of the angular momenta \mathbf{L} (3.21), either in terms of the angular velocity $\boldsymbol{\Omega}$ (3.26), leads to a generalised Gilbert equation.

3.3 Beyond Gilbert approximation

The conservation law of the angular momentum (3.24), or its equivalent (3.26), introduce a characteristic time scale $\tau = \frac{I_1}{\eta M_s^2}$, which separates the behaviour of the magnetic system of particles in two regimes: the diffusion and the inertial regime (to be treated separately in subsection 3.3.1, respectively 3.3.2). Defining a coefficient γ , such as $\gamma = \frac{M_s}{L_3}$ ($L_3 = cst.$), the Gilbert equation [3] and the stochastic Brown equation [5] are derived in the diffusion regime as long time scale limits of a more complex behaviour. For short time scales $t \ll \tau$, in the inertial regime, a generalised Gilbert equation is derived, characterised by *nutation*, a phenomenon to be detailed in the next chapter.

3.3.1 Diffusion regime $t \gg \tau$

A Brief presentation

For times t much longer than τ , the inertial terms $\dot{\Omega}_1$ and $\dot{\Omega}_2$ from the conservation laws (3.24) or (3.26), are negligible in respect to their damping correspondents $\frac{\Omega_1}{\tau}$, respectively $\frac{\Omega_2}{\tau}$ leading to a regime usually called in thermodynamics or statistical physics as the *diffusion regime*. It is typically characterised by two equations: the Fick's law and the Smoluchowski equation. More, the distribution function f is approximately a Maxwellian with a non vanishing average velocity.

The distribution function In the case of point-like magnetic masses (centre of mass) of orientation \mathbf{e} and velocity \mathbf{u} , the diffusion is related to the variation with time of the density of magnetic particles $n_{(\mathbf{e})}$. Due to thermalisation, after a time $t \gg \tau$, the magnetic particles will follow a distribution function f^{diff} close to a Maxwellian centered on the average angular velocity $\dot{\mathbf{M}}$:

$$f^{diff}(\mathbf{u}, \mathbf{e}, t) \simeq f^{l.eq.}(\mathbf{u}, \mathbf{m}, t) = \exp\left(\frac{\mu_{l.eq.} - m(\mathbf{u} - \dot{\mathbf{M}})^2/2 - V(\mathbf{e})}{k_B T/v}\right) \quad (3.28)$$

With the expression of the distribution function f^{diff} (3.28), the pressure tensor (3.9) reduces to the diagonal form $\overline{\overline{P}}^{diff} = \frac{nk_B T}{v} \overline{\overline{U}}$.

However, when the magnetic inertia tensor $\overline{\overline{I}}^{bd}$ is taken into account, the pressure and the distribution function f , should account for it (the inertia $\overline{\overline{I}}^{bd}$). As it will be pointed out in the next chapter, this leads to the same main results - the Gilbert, Landau-Lifshitz, Brown and generalised Gilbert equations. The important difference is that the demonstration will be done by deduction, and not by induction (not by extending a centre of mass treatment to a body-like treatment).

Either by importing a result from the next chapter, either by extending the result obtained in the case of magnetic centre of mass, the pressure tensor in the case of an axial inertia tensor takes the same value, meaning: $\overline{\overline{P}}^{diff} = \frac{nk_B T}{m v} \overline{\overline{U}}$.

The dynamic equation Neglecting the inertial term $\frac{d\Omega}{dt}$ in respect to the damping term $\overline{\overline{\beta}}_r \cdot \Omega$, the conservation law (3.26) leads to a typical dynamic equation for the diffusion regime. It represents the starting point for the Gilbert, Landau-Lifschitz and Brown equations.

$$0 \simeq -\overline{\overline{\beta}}^* \cdot \Omega + \frac{1}{I_1} \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \cdot \overline{\overline{P}}^{diff} \right) \quad (3.29)$$

where the damping tensor $\overline{\overline{\beta}}^*$ is the previously defined tensor $\overline{\overline{\beta}}^r$ (3.27) together with the definition of a coefficient γ as: $\gamma = \frac{M_s}{L_3} = \frac{M_s}{I_3 \Omega_3}$.

$$\overline{\overline{\beta}}^* = \begin{pmatrix} \tau^{-1} & \left(\frac{I_3}{I_1} - 1\right) \frac{M_s}{\gamma I_3} & 0 \\ -\left(\frac{I_3}{I_1} - 1\right) \frac{M_s}{\gamma I_3} & \tau^{-1} & 0 \\ 0 & 0 & 0 \end{pmatrix} = (\tau \alpha^*)^{-1} \begin{pmatrix} \alpha^* & 1 & 0 \\ -1 & \alpha^* & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (3.30)$$

where

$$\gamma \stackrel{def}{=} \frac{M_s}{L_3} \quad \alpha = \gamma \eta M_s \quad \alpha^* = \gamma^* \eta M_s \quad \gamma^* = \frac{\gamma}{1 - \frac{I_1}{I_3}} \quad (3.31)$$

B Gilbert's approximation

The dynamic equation (3.29) leads to the Gilbert equation (3.32), where the first part of its expression $I_1 \overline{\overline{\beta}}^* \cdot \Omega$ will be written in terms of the magnetisation \mathbf{M} as follows:

$$I_1 \overline{\overline{\beta}}^* \cdot \Omega = I_1 \tau^{-1} \left(\overline{\overline{U}} - \frac{\mathbf{M} \mathbf{M}}{M_s^2} \right) \cdot \Omega + I_1 (\tau \alpha^* M_s)^{-1} \frac{d\mathbf{M}}{dt} = \eta \mathbf{M} \times \frac{d\mathbf{M}}{dt} + \frac{1}{\gamma^*} \frac{d\mathbf{M}}{dt}$$

$$\frac{d\mathbf{M}}{dt} = \gamma^* \mathbf{M} \times \left(\mathbf{H} - \eta \frac{d\mathbf{M}}{dt} - \frac{k_B T}{n v} \frac{1}{M_s} \frac{\partial n}{\partial \mathbf{e}} \right) = \gamma^* \mathbf{M} \times \left(\mathbf{H}_{eff} - \eta \frac{d\mathbf{M}}{dt} \right) \quad (3.32)$$

The derivation of the Gilbert equation allows the physical interpretation of the previously defined coefficients: γ^* - the gyromagnetic factor, α^* - the dimensionless damping coefficient, η - the Gilbert damping coefficient. An effective magnetic field \mathbf{H}_{eff} accounting for the applied magnetic field \mathbf{H} and for the nonuniform distribution of the orientations of the

magnetisations, was also defined: $\mathbf{H}_{eff} = \mathbf{H} - \frac{k_B T}{nv} \frac{1}{M_s} \frac{\partial n}{\partial \mathbf{e}}$.

C Landau-Lifshitz equation (LL)

The same dynamic equation (3.29) also leads to the LL equation [4], where $\overline{\overline{D}}$ is a tensor with the LL gyromagnetic g^* and damping coefficients h^* :

$$\boldsymbol{\Omega} = (I_1 \overline{\overline{\beta}}^*)^{-1} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \quad \Leftrightarrow \quad \boldsymbol{\Omega} = \frac{v \overline{\overline{D}}}{k_B T} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \quad (3.33)$$

$$\overline{\overline{D}} = \frac{k_B T}{v} (I_1 \overline{\overline{\beta}}^*)^{-1} = \frac{k_B T}{v} g^* \begin{pmatrix} \alpha^* & -1 & 0 \\ 1 & \alpha^* & 0 \\ 0 & 0 & 0 \end{pmatrix} = \frac{k_B T}{v} \begin{pmatrix} h^* & -g^* & 0 \\ g^* & h^* & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (3.34)$$

$$g^* = \frac{\gamma^*}{M_s(1 + \alpha^{*2})} \quad h^* = \frac{\alpha^* \gamma^*}{M_s(1 + \alpha^{*2})} = \alpha^* g^* \quad (3.35)$$

The expression of the angular velocity $\boldsymbol{\Omega}$ (3.33) leads to the Landau-Lifshitz equation (3.36):

$$\frac{d\mathbf{M}}{dt} = \underbrace{\left[\frac{v \overline{\overline{D}}}{k_B T} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \right]}_{\boldsymbol{\Omega}} \times \mathbf{M} \quad \Rightarrow \quad \frac{d\mathbf{M}}{dt} = \mathbf{M} \times (g^* M_s \mathbf{H}_{eff} - h^* \mathbf{M} \times \mathbf{H}_{eff}) \quad (3.36)$$

D Brown's stochastic equation

The conservation equation for the density of particles $n_{(\mathbf{e})}$ (3.7) represents the starting point for the magnetic analogous of Smoluchowski's equation, the Brown's stochastic equation [5].

$$\frac{\partial n(\mathbf{e})}{\partial t} = -\frac{1}{M_s} \frac{\partial n \dot{\mathbf{M}}}{\partial \mathbf{e}} = \frac{\partial}{\partial \mathbf{e}} (n \mathbf{e} \times \boldsymbol{\Omega}) \quad \Leftrightarrow \quad \frac{\partial n}{\partial t} = -\frac{\partial \mathbf{j}^e}{\partial \mathbf{e}} \quad (3.37)$$

The expression of the angular velocity $\boldsymbol{\Omega}$ (3.33) gives the analytic form of the flux of particles \mathbf{j}^e . It represents the number of particles changing their magnetic orientation from \mathbf{e} to $\mathbf{e} + d\mathbf{e}$ per unit of time, and is caused by the application of a magnetic field \mathbf{H} and by the nonuniform repartition of the orientations in the 4π solid angle. The magnetic field generates a drift flux, while the nonuniform distribution generates a diffusion flux. The

magnetic analogous of Fick's law is then obtained (3.38):

$$\begin{aligned} \mathbf{j}^e &= n\boldsymbol{\Omega} \times \mathbf{e} = \frac{nv}{k_B T} \left[\overline{\overline{\mathbf{D}}} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \right] \times \mathbf{e} = \frac{nv\overline{\overline{\mathbf{D}}}}{k_B T} \cdot M_s \mathbf{H}_{eff} \Rightarrow \\ \mathbf{j}^e &= -\frac{nv}{k_B T} \overline{\overline{\mathbf{D}}} \cdot \frac{\partial V}{\partial \mathbf{e}} - \overline{\overline{\mathbf{D}}} \cdot \frac{\partial n}{\partial \mathbf{e}} \end{aligned} \quad (3.38)$$

With the conservation law (3.37) and the expression of the diffusion flux \mathbf{j}^e (3.38) the Brown equation derived in the framework of stochastic processes in 1963 [5] is easily derived:

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \cdot \left[\frac{nv\overline{\overline{\mathbf{D}}}}{k_B T} \cdot \frac{\partial}{\partial \mathbf{e}} \left(V + \frac{k_B T}{v} \ln n \right) \right] \Leftrightarrow \quad (3.39)$$

$$\begin{aligned} \frac{\partial n}{\partial t} &= \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta \left[\left(h^* \frac{\partial V}{\partial \theta} - g^* \frac{1}{\sin \theta} \frac{\partial V}{\partial \phi} \right) n + \frac{k_B T}{v} h^* \frac{\partial n}{\partial \theta} \right] \right\} \\ &+ \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \left\{ \left(g^* \frac{\partial V}{\partial \theta} + \frac{1}{\sin \theta} h^* \frac{\partial V}{\partial \phi} \right) n + \frac{k_B T}{v} h^* \frac{1}{\sin \theta} \frac{\partial n}{\partial \phi} \right\} \end{aligned} \quad (3.40)$$

3.3.2 Inertial regime $t \ll \tau$ - The generalised Gilbert equation

For times $t \ll \tau = \frac{I_1}{\eta M_s^2}$, the dynamic equation for the magnetisation is the angular momentum conservation law (3.21) written in terms of the magnetisation vector \mathbf{M} . The conservation law is rewritten:

$$\left(\frac{d\mathbf{L}}{dt} \right)_b + \boldsymbol{\Omega} \times \mathbf{L} = \mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \overline{\overline{P}}}{\partial \mathbf{e}} \right) \quad (3.41)$$

The first, as well as the second term of the left side of the equation (3.41) can be expressed in terms of the magnetisation \mathbf{M} as follows:

$$\begin{cases} \boldsymbol{\Omega} \times \mathbf{L} = (I_3 - I_1) \Omega_3 \boldsymbol{\Omega} \times \mathbf{e} = \frac{1}{\gamma^*} \frac{d\mathbf{M}}{dt} \\ \overline{\overline{\mathbf{I}}} \cdot \frac{d\boldsymbol{\Omega}}{dt} = \overline{\overline{\mathbf{I}}} \cdot \left(\mathbf{e} \times \frac{d^2 \mathbf{e}}{dt^2} + \Omega_3 \frac{d\mathbf{e}}{dt} \right) = \frac{I_1}{M_s^2} \mathbf{M} \times \frac{d^2 \mathbf{M}}{dt^2} + \frac{I_1}{I_3 \gamma} \frac{d\mathbf{M}}{dt} \end{cases} \quad (3.42)$$

Therefore, the generalised Gilbert equation is derived after replacing the two terms $\frac{d\mathbf{L}}{dt} = \overline{\overline{\mathbf{I}}} \cdot \frac{d\boldsymbol{\Omega}}{dt}$ and $\boldsymbol{\Omega} \times \mathbf{L}$ with expressions depending on the magnetisation \mathbf{M} :

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \overline{\overline{P}}}{\partial \mathbf{e}} - \eta \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2 \mathbf{M}}{dt^2} \right) \right] \quad (3.43)$$

Convergence towards the Gilbert equation With the expression of the generalised Gilbert equation (3.43), the existence of two regimes separated by a characteristic time scale τ is clearer. The main unknown of the equation is the principal moment of inertia I_1 . If I_1 is nul, the magnetisation's acceleration $\frac{d^2\mathbf{M}}{dt^2}$ dissapears, the Gilbert equation is retrieved and a classic analogous of the magnetisation dynamics does not exist as Gilbert predicted².

If however, a classic mechanic analogous exists ($I_1 \neq 0$), the Gilbert equation is retrieved for time scales bigger than a characteristic time τ , with a correction on the gyromagnetic coefficient. In order to see the convergence from the generalised Gilbert (3.43) to the Gilbert equation (3.32), two terms of interest will be compared from the generalised expression:

$$\left\{ \begin{array}{l} \mathbf{M} \times \frac{d\mathbf{M}}{dt} = \mathbf{M} \times (\boldsymbol{\Omega} \times \mathbf{M}) \\ \tau \mathbf{M} \times \frac{d^2\mathbf{M}}{dt^2} = \mathbf{M} \times \left(\tau \frac{d\boldsymbol{\Omega}}{dt} \times \mathbf{M} \right) + \tau \mathbf{M} \left(\boldsymbol{\Omega} \times \frac{d\mathbf{M}}{dt} \right) = \mathbf{M} \times \left(\tau \frac{d\boldsymbol{\Omega}}{dt} \times \mathbf{M} \right) - \frac{1}{\eta\gamma} \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt} \end{array} \right. \quad (3.44)$$

For long time scales $t \gg \tau$, the term containing the magnetisation's acceleration $\frac{d^2\mathbf{M}}{dt^2}$ converges to $-\frac{1}{\eta\gamma} \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt}$, leading to the Gilbert equation with a correction performed on the gyromagnetic coefficient $\gamma^* = \frac{\gamma}{1-I_1/I_3}$.

$$\gamma\eta \mathbf{M} \times \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2\mathbf{M}}{dt^2} \right) \xrightarrow{t \gg \tau} \gamma\eta \mathbf{M} \times \frac{d\mathbf{M}}{dt} - \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt} \quad (3.45)$$

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} - \eta \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2\mathbf{M}}{dt^2} \right) \right] \quad (3.46)$$

$$\begin{array}{l} \searrow t \gg \tau \\ \frac{d\mathbf{M}}{dt} = \gamma^* \mathbf{M} \times \left(\mathbf{H} - \frac{k_B T}{n v} \frac{\partial n}{\partial \mathbf{M}} - \eta \frac{d\mathbf{M}}{dt} \right) \end{array} \quad (3.47)$$

As it will be proved in the next chapter, the new dynamics of the magnetisation is the magnetic analogous of a heavy symmetrical top with damping. It will also introduce a new phenomenon, *nutation*.

²“One can show that the inertial tensor must have a single nonzero term corresponding to the rotational inertia for rotation about the principal axis. I was unable to conceive of a physical object with an inertial tensor of this kind.” - Gilbert, 1956 [3]

Conclusions

In respect to what has been done previously, in this chapter is applied the framework of mesoscopic nonequilibrium thermodynamics MNET (and not that of NET) to an ensemble of magnetic particles \mathbf{m} to go beyond the usual Gilbert equation describing the dynamics of one magnetic particle, and Brown's stochastic equation, relating to the probability of having one magnetic particle oriented in a particular direction (θ, ϕ) .

Each uniform magnetisation has associated four degrees of freedom, and not only two as before: the orientation $\mathbf{e} = (\theta, \phi)$ and the velocity $\mathbf{u} = \dot{\mathbf{e}} = (u_\theta, u_\phi)$. The particles change their orientation and velocity either due to an applied magnetic field \mathbf{H} , either due to the existing thermal fluctuations (temperature T). The model has two fundamental hypothesis concerning their kinetics: a Newtonian type of law and an expression for the kinetic energy associated to each magnetic particle. Their expressions impose a magnetic centre of mass m associated to the inertia of the magnetisations, and not to that of matter.

The result of the MNET framework is a dynamic equation for the average velocity of the magnetic particles $\dot{\mathbf{M}}$ of magnetic centre of mass m . This result is then extended to a magnetic inertia tensor $\overset{bd}{\overline{I}}$ respecting the magnetic properties of the particles: an axial symmetry given by the magnetisations orientation \mathbf{e} . The extended dynamic equation introduces a relaxation time τ related to the Gilbert's damping η and to one of the principal momenta of inertia I_1 . This relaxation time τ separates the dynamic behaviour of the magnetisations in two regimes: the diffusion regime or the long time scale limit $t \gg \tau$, and the inertial regime or the short time scale limit $t \ll \tau$.

It is proved that a classic mechanic analogous for the magnetisation dynamics exists, i.e. a physical object can be conceived having an analogous behaviour with that of the magnetisation. It is also proved that the Gilbert, the Landau-Lifshitz (LL) and the Brown stochastic equations are derived for long time scales $t \gg \tau$, while for short time scales $t \ll \tau$ a generalised Gilbert equation is derived, containing the magnetisation acceleration $\frac{d^2\mathbf{M}}{dt^2}$.

Next chapter: The purpose of the next chapter is to prove that not only the same results derived here are recovered by deduction, and not by induction (by extending the case of a magnetic centre of mass to a magnetic inertia tensor), but also that more complex stochastic equations characterise the short time scales regime $t \ll \tau$, i.e. Fokker-Planck type equations.

Appendix

3.A The collision flux $\mathbf{J}^{\mathbf{u}}$

When a magnetic particle $\mathbf{m} = M_s \mathbf{e}$ interacts with the thermal bath, its velocity $\mathbf{u} = M_s \dot{\mathbf{e}}$ and energy changes. The lack of conservation of the velocity and energy of the ensemble of magnetic particles due to the interactions with the bath leads to the appearance of *dissipation*, which appears explicitly in the dynamic and energy equations for the ensemble of particles. The interactions with the bath are analytically expressed in terms of the 'collision flux' $\mathbf{J}^{\mathbf{u}}$, having the following properties:

- (a). The number of magnetic particles is unchanged at the interaction with the bath, allowing the writing of the collision term as a divergence of a flux vanishing for infinite velocities $\lim_{\mathbf{u} \rightarrow \pm\infty} \mathbf{J}^{\mathbf{u}}$:

$$\int C d\mathbf{u} = 0 \quad \xrightarrow{\text{def}} \quad C = -\frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} \quad (3.48)$$

- (b). To deal with the interactions between the magnetic particles and the bath, it is also convenient to define a *damping field* \mathbf{R} :

$$\mathbf{R} = \int C m \mathbf{u} d\mathbf{u} = \int m \mathbf{J}^{\mathbf{u}} d\mathbf{u} \quad (3.49)$$

3.B The entropy production σ

Starting from the cornerstones of the framework, the Gibbs postulate (3.4) and the conservation equation (3.3), the expression for the entropy production σ is derived. Their definitions are recalled:

$$Tns = - \int f \left(\frac{k_B T}{v} \ln f + \frac{m u^2}{2} + V(\mathbf{e}) - \mu^{eq} \right) d\mathbf{u} \quad (3.50)$$

$$\frac{\partial f}{\partial t} = -\mathbf{u} \cdot \frac{1}{M_s} \frac{\partial f}{\partial \mathbf{e}} - \frac{\mathbf{H}}{m} \cdot \frac{\partial f}{\partial \mathbf{u}} - \frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} \quad (3.51)$$

To obtain σ , $T\rho s$ is derived in respect to time:

$$T \frac{\partial(ns)}{\partial t} = -\frac{k_B T}{v} \int \frac{\partial f}{\partial t} (\ln f + 1) d\mathbf{u} - \int \frac{\partial f}{\partial t} \left(\frac{m u^2}{2} + V(\mathbf{e}) - \mu_{eq} \right) d\mathbf{u} \quad (3.52)$$

Each integral is calculated:

(a).

$$\begin{aligned}
\int \frac{\partial f}{\partial t} (\ln f + 1) d\mathbf{u} &= -\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \int f \mathbf{u} (\ln f + 1) d\mathbf{u} \quad + \int f \mathbf{u} \cdot \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} (\ln f + 1) d\mathbf{u} \\
&\quad - \frac{\mathbf{H}}{m} \cdot \int \frac{\partial}{\partial \mathbf{u}} [f (\ln f + 1)] d\mathbf{u} \quad + \frac{\mathbf{H}}{m} \cdot \int f \frac{1}{f} \frac{\partial f}{\partial \mathbf{u}} d\mathbf{u} \\
&\quad - \int \frac{\partial}{\partial \mathbf{u}} [\mathbf{J}^{\mathbf{u}} (\ln f + 1)] d\mathbf{u} \quad + \int \mathbf{J}^{\mathbf{u}} \cdot \frac{1}{f} \frac{\partial f}{\partial \mathbf{u}} d\mathbf{u} \\
&= -\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \int f \mathbf{u} \ln f d\mathbf{u} + \int \mathbf{J}^{\mathbf{u}} \cdot \frac{\partial (\ln f)}{\partial \mathbf{u}} d\mathbf{u} \tag{3.53}
\end{aligned}$$

(b).

$$\begin{aligned}
\int \frac{\partial f}{\partial t} \frac{mu^2}{2} d\mathbf{u} &= -\int \frac{mu^2}{2} \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} (f \mathbf{u}) d\mathbf{u} - \frac{\mathbf{H}}{m} \cdot \int \frac{\partial f}{\partial \mathbf{u}} \frac{mu^2}{2} d\mathbf{u} - \int \frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} \frac{mu^2}{2} d\mathbf{u} \\
&= -\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \int f \mathbf{u} \frac{mu^2}{2} d\mathbf{u} + n \dot{\mathbf{M}} \cdot \mathbf{H} + \int \mathbf{J}^{\mathbf{u}} \cdot \frac{\partial}{\partial \mathbf{u}} \left(\frac{mu^2}{2} \right) d\mathbf{u} \tag{3.54}
\end{aligned}$$

(c).

$$\begin{aligned}
\int \frac{\partial f}{\partial t} (V(\mathbf{e}) - \mu_{eq}) d\mathbf{u} &= -\int \mathbf{u} \cdot \frac{1}{M_s} \frac{\partial f}{\partial \mathbf{e}} (V - \mu_{eq}) d\mathbf{u} - \int \frac{\mathbf{H}}{m} \cdot \frac{\partial f}{\partial \mathbf{u}} (V - \mu_{eq}) d\mathbf{u} \\
&\quad - \int \frac{\partial \mathbf{J}^{\mathbf{u}}}{\partial \mathbf{u}} (V - \mu_{eq}) d\mathbf{u} = -\frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \int f \mathbf{u} (V(\mathbf{e}) - \mu_{eq}) d\mathbf{u} - n \dot{\mathbf{M}} \cdot \mathbf{H} \tag{3.55}
\end{aligned}$$

During the calculations use has been made of the independence of the freedom degrees \mathbf{e} and \mathbf{u} , and of the properties of the distribution function f and that of the velocity flux $\mathbf{J}^{\mathbf{u}}$: $\lim_{\mathbf{u} \rightarrow \pm\infty} f = 0$ and $\lim_{\mathbf{u} \rightarrow \pm\infty} \mathbf{J}^{\mathbf{u}} = 0$. The entropy flux \mathbf{J}^s and the entropy production σ are identified:

$$T \frac{\partial ns}{\partial t} = \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \underbrace{\int f \mathbf{u} \left(\frac{k_B T}{v} \ln f + \frac{mu^2}{2} + V(\mathbf{e}) - \mu^{eq} \right) d\mathbf{u}}_{-\mathbf{J}^s} - \int \mathbf{J}^{\mathbf{u}} \cdot \frac{\partial}{\partial \mathbf{u}} \left(\frac{k_B T}{v} \ln f + \frac{mu^2}{2} \right) d\mathbf{u} \tag{3.56}$$

$$\begin{cases} \mathbf{J}^s = - \int f \mathbf{u} \left(\frac{k_B T}{v} \ln f + \frac{m u^2}{2} + V(\mathbf{e}) - \mu^{eq} \right) d\mathbf{u} \\ \sigma = - \int \mathbf{J}^u \left(\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{u}} + m \mathbf{u} \right) d\mathbf{u} \end{cases} \quad (3.57)$$

The entropy flux \mathbf{J}^s is rewritten in a form with more physical meaning:

$$\begin{aligned} \mathbf{J}^s = & - \int f \mathbf{u} \left(\frac{k_B T}{v} \ln f - \mu^{l.eq.} \right) d\mathbf{u} - \frac{\rho \dot{\mathbf{M}}^2}{2} \dot{\mathbf{M}} - \overline{\overline{\mathbf{P}}} \cdot \dot{\mathbf{M}} - nV \dot{\mathbf{M}} \\ & - \dot{\mathbf{M}} \cdot \int f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} - \int (\mathbf{u} - \dot{\mathbf{M}}) \cdot f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} \end{aligned} \quad (3.58)$$

$$= - \int f \mathbf{u} \left(\frac{k_B T}{v} \ln f - \mu^{l.eq.} \right) d\mathbf{u} - \mathbf{q} - \frac{\rho \dot{\mathbf{M}}^2}{2} \dot{\mathbf{M}} - nV \dot{\mathbf{M}} - \overline{\overline{\mathbf{P}}} \cdot \dot{\mathbf{M}} - \rho u \dot{\mathbf{M}} \quad (3.59)$$

The entropy flux \mathbf{J}^s is a sum of fluxes of heat \mathbf{q} , kinetic energy $\frac{\rho \dot{\mathbf{M}}^2}{2} \dot{\mathbf{M}}$, potential energy $nV \dot{\mathbf{M}}$, mechanical energy $\overline{\overline{\mathbf{P}}} \cdot \dot{\mathbf{M}}$ and 'internal energy' $\rho u \dot{\mathbf{M}}$, where kinetic theory definitions for the internal energy density ρu and for the heat flux \mathbf{q} have been used:

$$\mathbf{q} \stackrel{def}{=} \int (\mathbf{u} - \dot{\mathbf{M}}) \cdot f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} \quad \rho u \stackrel{def}{=} \int f \frac{m(\mathbf{u} - \dot{\mathbf{M}})^2}{2} d\mathbf{u} \quad (3.60)$$

3.C The angular velocity Ω

3.C.1 Properties

$$(i) \quad \Omega \stackrel{def}{=} \left(\dot{\phi} \sin \theta \sin \psi + \dot{\theta} \cos \psi \right) \hat{\mathbf{i}} + \left(\dot{\phi} \sin \theta \cos \psi - \dot{\theta} \sin \psi \right) \hat{\mathbf{j}} + \left(\dot{\psi} + \dot{\phi} \cos \theta \right) \hat{\mathbf{k}} \quad (3.61)$$

$$(ii) \quad \dot{\mathbf{M}} = \Omega \times \mathbf{M} \quad (3.62)$$

$$(iii) \quad \frac{d^2 \mathbf{M}}{dt^2} = \frac{d\Omega}{dt} \times \mathbf{M} + \Omega \times \dot{\mathbf{M}} = \frac{d\Omega}{dt} \times \mathbf{M} + \Omega \times (\Omega \times \mathbf{M}) \quad (3.63)$$

3.C.2 The angular momentum conservation law

The angular momentum conservation law can also be expressed in terms of the angular velocity Ω , as follows:

$$\left(\frac{d\mathbf{L}}{dt}\right)_b = -\Omega \times \mathbf{L} + \mathbf{M} \times \left(\mathbf{H} - \eta \dot{\mathbf{M}} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}}\right) \quad (3.64)$$

$$\bar{I}_b^{bd} \cdot \frac{d\Omega}{dt} = -\Omega \times \left\{ \left[I_1 (\bar{U} - \mathbf{e}\mathbf{e}) + I_3 \mathbf{e}\mathbf{e} \right] \cdot \Omega \right\} - \eta M_s^2 (\bar{U} - \mathbf{e}\mathbf{e}) \cdot \Omega + \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.65)$$

$$\frac{d\Omega}{dt} = - \overbrace{\left[\frac{\eta M_s^2}{I_1} (\bar{U} - \mathbf{e}\mathbf{e}) - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 \mathbf{e} \times \bar{U} \right]}^{\bar{\beta}_r} \cdot \Omega + \frac{1}{I_1} \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.66)$$

$$\frac{d\Omega}{dt} = -\bar{\beta}_r \cdot \Omega + \frac{1}{I_1} \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{1}{M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} \right) \quad (3.67)$$

where

$$\bar{\beta}_r = \begin{pmatrix} \frac{\eta M_s^2}{I_1} & \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & 0 \\ - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & \frac{\eta M_s^2}{I_1} & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (3.68)$$

Bibliography

- [1] J.-E. WEGROWE, M.-C. CIORNEI, and H.-J. DROUHIN, *J. Phys.:Condens Matter* **19**, 165213 (2007).
- [2] J.-E. WEGROWE, M.-C. CIORNEI, and H.-J. DROUHIN, *Thermokinetics of Spin-Dependent Transport and Ferromagnetism*, chapter 15, American Scientific, 2009.
- [3] T. L. GILBERT, *IEEE Transactions on Magnetics* **40**, 3443 (2004).
- [4] L. LANDAU and E. LIFSHITZ, *Phys. Z. Sowjetunion* **8**, 153 (1935).
- [5] W. F. BROWN JR., *Physical Review* **130**, 1677 (1963).
- [6] I. PRIGOGINE and P. MAZUR, *Physica* **19**, 241 (1953).

- [7] S. DEGROOT and P. MAZUR, *Non-Equilibrium Thermodynamics*, Dover Publications Inc. New York, second edition, 1984.
- [8] D. REGUERA, J. RUBI, and J. VILAR, *J. Phys. Chem. B* **109**, 21502 (2005).
- [9] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, pp. 55–56, In [16], third edition, 2007.
- [10] S. CHAPMAN and T. COWLING, *The mathematical theory of non-uniform gases*, chapter 3 - The equations of Boltzmann and Maxwell, Cambridge University Press, third edition, 1970.
- [11] L. C. WOODS, *The thermodynamics of fluid systems*, chapter XIII - Kinetic processes, Calendron Press, 1975.
- [12] J. M. RUBI and A. PEREZ-MADRID, *Physica A* **264**, 492 (1999).
- [13] J. M. RUBI and P. MAZUR, *Physica A* **250**, 253 (1998).
- [14] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, p. 185, In [16], third edition, 2007.
- [15] J. M. G. VILAR and J. M. RUBI, *Proc. Natl. Acad. Sci. USA* **98**, 11081 (2001).
- [16] N. G. VAN KAMPEN, *Stochastic processes in physics and chemistry*, Elsevier Science and Technology Books, third edition, 2007.

Chapter 4

The generalised Gilbert equation Deductive approach

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Introduction

As shown in the first chapter, associating two degrees of freedom (θ, ϕ) to a monodomain magnetic particle \mathbf{m} is enough to derive the Gilbert and the Brown's stochastic equations governing its motion. It was also shown that by adding kinetic type degrees of freedom such as the velocity \mathbf{u} , faster time scales are accessed where the dynamics is ruled by a generalised Gilbert equation. In their derivation, each magnetic particle had associated four degrees of freedom $(\theta, \phi, u_\theta, u_\phi)$ and a mass, not related to the matter inertia but to magnetisation inertia. The mesoscopic nonequilibrium thermodynamics framework (MNET) lead to a dynamic equation written in terms of the angular momentum of the magnetic centre of mass. Supposing the same dynamic equation also held for a more complex magnetic inertia tensor $\overset{\equiv bd}{I}$ associated to the magnetisation inertia, the generalised Gilbert equation was derived by induction. The inertia tensor corresponding to the centre of mass was replaced by an axial inertia tensor having as symmetry axis the magnetisation direction.

The purpose of this chapter is to derive the generalised Gilbert equation from scratch, i.e. by associating an axial inertia tensor to the magnetisation inertia since the beginning of the formalism to confirm by deduction, that the inductive conclusion obtained previously is correct.

We adopt the same structure as before, meaning: the first section 4.1 familiarises the reader with the model; the following section 4.2 pursues the theoretical scheme of MNET to deduce averaged dynamic and stochastic equations; the Gilbert, the Landau-Lifshitz, the Brown equations are derived for times scales bigger than a characteristic time τ , while a generalised Gilbert equation, and a more complex Fokker-Planck equation are derived for time scales shorter than τ in section 4.3. As it is difficult to give predictions only with the analytical results, a numerical solution of the generalised Gilbert equation is performed (see section 4.4). For time scales t shorter than τ a new phenomenon arises: *nutaton*.

4.1 The model

To model the dynamics of a monodomain particle subjected to thermal agitation, we study an ensemble of identical monodomain particles of magnetisation \mathbf{m} and magnetic energy $V(\theta, \phi)$. Each magnetisation vector \mathbf{m} has the same origin O , a constant M_s modulus, a physical volume v and is represented by its vector tip, a point on the surface of a M_s radius sphere. The magnetisation \mathbf{m} is reoriented either by an applied magnetic field, either by thermal fluctuations. One magnetic particle is identified in phase space by its orientation $\boldsymbol{\alpha} = (\theta, \phi, \psi)$ and by its angular momentum \mathbf{L} . The number of magnetic particles with the orientation within $(\boldsymbol{\alpha}, \boldsymbol{\alpha} + d\boldsymbol{\alpha})$ and angular momenta within $(\mathbf{L}, \mathbf{L} + d\mathbf{L})$ is represented by the distribution function $f(\boldsymbol{\alpha}, \mathbf{L})$. It is assumed that f vanishes for infinite angular momenta $\lim_{\mathbf{L} \rightarrow \pm\infty} f(\boldsymbol{\alpha}, \mathbf{L}) = 0$. The number of magnetic particles with the orientation

within $(\boldsymbol{\alpha}, \boldsymbol{\alpha} + d\boldsymbol{\alpha})$ is represented by the density of particles $n(\boldsymbol{\alpha})$, where $n(\boldsymbol{\alpha}) = \int f(\boldsymbol{\alpha}, \mathbf{L}) d\mathbf{L}$. The average angular momenta of the magnetisations with the orientation within $(\boldsymbol{\alpha}, \boldsymbol{\alpha} + d\boldsymbol{\alpha})$ is defined as $n(\boldsymbol{\alpha}) \langle \mathbf{L} \rangle = \int f(\boldsymbol{\alpha}, \mathbf{L}) \mathbf{L} d\mathbf{L}$. The angular momenta \mathbf{L} of one magnetic particle is either changed by an applied magnetic torque \mathbf{N} , either by the interaction with the heat bath, modelled through a phase space flux $\mathbf{J}^{\mathbf{L}}$. It is assumed that $\lim_{\mathbf{L} \rightarrow \pm\infty} \mathbf{J}^{\mathbf{L}} = 0$.

To each magnetisation \mathbf{m} is attributed an inertia tensor $\overline{\overline{I}}$, *not* related to the inertia of matter, but to that of magnetisation dynamics. The inertia tensor is assumed to have a symmetry axis \mathbf{e} given by the magnetic properties of the particle, i.e. by the magnetisation orientation $\mathbf{m} = M_s \mathbf{e}$. Its expression will then be $\overline{\overline{I}} = I_1 (\overline{\overline{U}} - \mathbf{e}\mathbf{e}) + I_3 \mathbf{e}\mathbf{e}$, where $\overline{\overline{U}}$ is the dyadic unit. Also, the kinetic energy \mathcal{T} of one magnetic particle is assumed to be $\mathcal{T} = \mathbf{L}\mathbf{L} : \overline{\overline{I}}^{-1} / 2$.

Each volume element $(\boldsymbol{\alpha}, d\boldsymbol{\alpha})$, $(\boldsymbol{\alpha}, \mathbf{L}, d\boldsymbol{\alpha}, d\mathbf{L})$ is described by extensive parameters such as: a) the density of magnetic particles $n(\boldsymbol{\alpha})$, respectively $f(\boldsymbol{\alpha}, \mathbf{L})$; b) the density of entropy $s(\boldsymbol{\alpha})$; c) kinetic and potential energy densities $\int f \mathbf{L}\mathbf{L} : \overline{\overline{I}}^{-1} / 2 d\mathbf{L}$, respectively $\int f V(\boldsymbol{\alpha}) d\mathbf{L} = n(\boldsymbol{\alpha}) V(\boldsymbol{\alpha})$. We suppose the heat bath affects equivalently all the orientations $\boldsymbol{\alpha}$, i.e. the temperature T is considered uniform all over the sphere.

With these in mind, we proceed to the derivation of the generalised Gilbert equation and its corresponding Fokker-Planck equation. The scheme of MNET is used in order to establish phenomenological laws describing the interaction of the magnetic particles with the thermal bath. The irreversible process of energy dissipation from the magnetic particles towards the bath is represented by dissipative forces and fluxes. To determine their shape, the entropy production $\sigma(\boldsymbol{\alpha})$ has to be first determined.

Next section: The purpose of the next section is to derive the expression for the entropy production, the dissipative forces and finally, the magnetisation's dynamic and stochastic equations in terms of the angular momenta \mathbf{L}_s .

4.2 The MNET framework

There are two cornerstones at the basis of the MNET framework: the conservation law (4.1) and the Gibbs entropy postulate (4.3) (subsection 4.2.1). Starting with these, the entropy production σ is derived and the dissipative force and flux are determined in "Thermokinetic equations" (subsection 4.2.2). The second law of thermodynamics and the hypothesis of local equilibrium impose a phenomenological law between the cause (the dissipative force) and the effect (the flux). As a consequence, an Onsager coefficient is introduced as a proportionality coefficient between the two. With this law, the magnetisation's dynamic and stochastic equations are derived in terms of the average angular momentum $\langle \mathbf{L}_s \rangle$ in "The dynamic and the stochastic equations" (subsection 4.2.3). A cosmetic treatment performed on these

results leads to the generalised Gilbert equation and the Fokker-Planck equation, to be presented in the following section 4.3.

4.2.1 Cornerstones

A The conservation law

In cartesian coordinates, in the space fixed reference-frame, the conservation law for the number of particles $f(\boldsymbol{\alpha}, \mathbf{L}_s)$ writes as [1]:

$$\frac{\partial}{\partial t} f(\boldsymbol{\alpha}, \mathbf{L}_s) = - \left(\frac{\partial f \dot{\boldsymbol{\alpha}}}{\partial \boldsymbol{\alpha}} \right)_{\mathbf{L}_s} - \mathbf{N}_s \cdot \frac{\partial f}{\partial \mathbf{L}_s} - \frac{\partial \mathbf{J}_s^{\mathbf{L}}}{\partial \mathbf{L}_s} \quad (4.1)$$

where the subscript s stands for the space-fixed reference frame, $\boldsymbol{\alpha}$ is a symbolic notation for the three Euler angles, and the derivatives in respect to the angles are made while the cartesian components of the angular momenta \mathbf{L}_s are held constant.

$$\left(\frac{\partial f \dot{\boldsymbol{\alpha}}}{\partial \boldsymbol{\alpha}} \right)_{\mathbf{L}_s} = \frac{1}{\sin \theta} \left\{ \frac{\partial}{\partial \theta} (\dot{\theta} \sin \theta f) \right\}_{\mathbf{L}_s} + \left\{ \frac{\partial}{\partial \phi} (\dot{\phi} f) \right\}_{\mathbf{L}_s} + \left\{ \frac{\partial}{\partial \psi} (\dot{\psi} f) \right\}_{\mathbf{L}_s} \quad (4.2)$$

B The Gibbs entropy postulate

The entropy density $n s$ is defined as in kinetic theory and statistical physics, by the Gibbs entropy postulate:

$$T n s = -k_B T \int f \ln \frac{f}{f^{l.eq.}} d\mathbf{L}_s + T n s^{eq} \quad (4.3)$$

where $f^{l.eq.}$ is the local equilibrium distribution, meaning the local Maxwellian, and $\mu_{l.eq.}$ the chemical potential of particles at local equilibrium.

$$f^{l.eq.}(\boldsymbol{\alpha}, \mathbf{L}_s, t) = \exp \left(\frac{\mu_{l.eq.} - \mathbf{L}_s \mathbf{L}_s : \bar{\mathbf{I}}^{-1} / 2 - V(\boldsymbol{\alpha})}{k_B T / v} \right) \quad (4.4)$$

Written differently eq. (4.3), takes the form:

$$T n s = - \int f \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\mathbf{I}}^{-1}}{2} + V(\boldsymbol{\alpha}) - \mu^{eq} \right) d\mathbf{L}_s \quad (4.5)$$

4.2.2 Thermokinetic equations

A Conservation laws

From the conservation law of particles (4.1), two conservation laws are deduced which serve as starting point for the Brown's stochastic equation, and the generalised Gilbert equation: the conservation law for the number of magnetic particles $n(\boldsymbol{\alpha})$ with orientation within $(\boldsymbol{\alpha}, \boldsymbol{\alpha} + d\boldsymbol{\alpha})$ and respectively, the conservation law for the averaged angular momentum $\langle \mathbf{L}_s \rangle$ of the $n(\boldsymbol{\alpha})$ magnetic particles. In all cases, the interaction with the heat bath $\mathbf{J}_s^{\mathbf{L}}$ has to be found.

The conservation equation for the density of particles $n(\boldsymbol{\alpha})$:

$$\frac{\partial n}{\partial t} = \int \frac{\partial f}{\partial t} d\mathbf{L}_s = -\frac{\partial}{\partial \boldsymbol{\alpha}} \int f \bar{\bar{\Lambda}}(\boldsymbol{\alpha}) \bar{\bar{I}}^{-1}(\boldsymbol{\alpha}) \mathbf{L}_s d\mathbf{L}_s = -\frac{\partial}{\partial \boldsymbol{\alpha}} \left(\bar{\bar{\Lambda}}(\boldsymbol{\alpha}) \bar{\bar{I}}^{-1}(\boldsymbol{\alpha}) \cdot n \langle \mathbf{L}_s \rangle \right) \quad (4.6)$$

The conservation law for the averaged angular momentum $n \langle \mathbf{L}_s \rangle$ (see appendix 4.A.1 for details): :

$$\frac{\partial n \langle \mathbf{L}_s \rangle}{\partial t} = \int \frac{\partial f}{\partial t} \mathbf{L}_s d\mathbf{L}_s = -\frac{\partial}{\partial \boldsymbol{\alpha}} \int \mathbf{L}_s \left(f \bar{\bar{\Lambda}}(\boldsymbol{\alpha}) \bar{\bar{I}}^{-1}(\boldsymbol{\alpha}) \cdot \mathbf{L}_s \right) d\mathbf{L}_s + n \mathbf{N}(\boldsymbol{\alpha}) + \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s \quad (4.7)$$

$$\Rightarrow n \frac{d}{dt} \langle \mathbf{L}_s \rangle = -\frac{\partial}{\partial \boldsymbol{\alpha}} (\bar{\bar{P}} \bar{\bar{\Lambda}}^t) + n \mathbf{N}(\boldsymbol{\alpha}) + \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s \quad (4.8)$$

where t stands for transpose and $\bar{\bar{P}}$ stands for the 'pressure' tensor:

$$\bar{\bar{P}} \stackrel{def}{=} \left[\int f(\mathbf{L}_s - \langle \mathbf{L}_s \rangle) (\mathbf{L}_s - \langle \mathbf{L}_s \rangle) d\mathbf{L}_s \right] \bar{\bar{I}}^{-1} \quad (4.9)$$

The calculations were made for the case of a potentially driven field $\mathbf{H}(\boldsymbol{\alpha}) = -\frac{1}{M_s} \left(\frac{\partial V}{\partial \boldsymbol{\theta}} \hat{\boldsymbol{\theta}} + \frac{\partial V}{\partial \boldsymbol{\phi}} \hat{\boldsymbol{\phi}} \right)$, whose associated torque \mathbf{N} depends only on the degree of freedom $\boldsymbol{\alpha}$. A tensor $\bar{\bar{\Lambda}}$, as defined by Condiff et al. [1, 2], was also introduced to make the connection between the temporal derivative of the angles $\dot{\boldsymbol{\alpha}}$ and the angular velocity $\boldsymbol{\omega}$ as: $\dot{\boldsymbol{\alpha}} = \bar{\bar{\Lambda}}(\boldsymbol{\alpha}) \cdot \boldsymbol{\omega} = \bar{\bar{\Lambda}}(\boldsymbol{\alpha}) \bar{\bar{I}}^{-1}(\boldsymbol{\alpha}) \mathbf{L}_s$.

Axial symmetry For the particular case of axial symmetric bodies, all pertinent quantities are independent of ψ . In this case, Condiff et al. [1, 2] prove that $\boldsymbol{\alpha}$ can be replaced with \mathbf{e} , $\dot{\boldsymbol{\alpha}}$ with $\dot{\mathbf{e}} = \boldsymbol{\omega} \times \mathbf{e}$, $\frac{\partial}{\partial \boldsymbol{\alpha}}$ with $\frac{\partial}{\partial \mathbf{e}} = \boldsymbol{\theta} \frac{\partial}{\partial \boldsymbol{\theta}} + \boldsymbol{\phi} \frac{1}{\sin \theta} \frac{\partial}{\partial \boldsymbol{\phi}}$ and $\bar{\bar{\Lambda}}$ with $-\mathbf{e} \times \bar{\bar{U}}$, where \mathbf{e} is the

axial unit vector and $\overline{\overline{U}}$ is the dyadic unit. The conservation law (4.8) is then written as:

$$\frac{d}{dt} \langle \mathbf{L}_s \rangle = -\mathbf{e} \times \frac{1}{n} \frac{\partial}{\partial \mathbf{e}} \overline{\overline{P}} + \mathbf{N}_{(e)} + \frac{1}{n} \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s \quad (4.10)$$

The conservation law or the dynamic equation (4.10) states that the variation with time of the average angular momentum $\langle \mathbf{L}_s \rangle$ is controlled by the applied torques \mathbf{N} , by the interactions with the heat bath $\int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s$ and by the torque of the "pressure gradient" of the magnetic gas $\overline{\overline{P}}$. The different torques are expected to act as follows: a) the collisions with the heat bath manifested through $\int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s$ are expected to act as a damping like torque; b) the torque of the "pressure gradient" $\mathbf{e} \times \frac{1}{n} \frac{\partial}{\partial \mathbf{e}} \overline{\overline{P}}$ is expected to act, in a certain limit, as a driving torque redirecting the magnetic particles from the highly concentrated areas, to lowly concentrated areas, in order for the distribution of particles $n_{(e)}$ to become uniform.

The purpose of the following subsections is to find out the expression for the flux $\mathbf{J}_s^{\mathbf{L}}$, which models the interaction of the magnetisation with the bath. In this sense, the entropy production has to be derived.

B The entropy production σ

Following the scheme of MNET, equation (4.5) is derived in respect to time in order to solve for the entropy production σ . The resulting vector divergence is then associated with the divergence of the entropy flux \mathbf{J}^s , while the remaining expression is associated with the entropy production σ . For the case of particles with axial symmetry \mathbf{e} , the two physical quantities of interest take the following shape: (see appendix 4.A.2 for explicit calculations)

$$\begin{aligned} T \frac{\partial n s}{\partial t} = & - \frac{\partial}{\partial \mathbf{e}} \left[\mathbf{e} \times \int f \overline{\overline{I}}^{-1} \cdot \mathbf{L}_s \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{I}}^{-1}}{2} + V(\boldsymbol{\alpha}) - \mu^{eq} \right) d\mathbf{L}_s \right] \\ & - \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{\partial}{\partial \mathbf{L}_s} \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{I}}^{-1}}{2} \right) d\mathbf{L}_s \end{aligned} \quad (4.11)$$

As noticed in (4.11), the flux \mathbf{J}^s is the sum of several fluxes: a kinetic energy flux, a potential energy flux and an entropic flux. The remaining expression of the entropy increase in respect to time is the entropy production σ . It is associated to the 'collision' process between the magnetisations \mathbf{m} and the heat bath, or more precisely to the process of irreversible energy loss from the magnetisations to the bath, called also damping. Its

expression is the product between the flux $\mathbf{J}_s^{\mathbf{L}}$ and its corresponding force $\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{L}_s} + \bar{\bar{\mathbf{I}}}^{-1} \cdot \mathbf{L}_s$.

$$\begin{cases} \mathbf{J}^s = \mathbf{e} \times \int f \bar{\bar{\mathbf{I}}}^{-1} \cdot \mathbf{L}_s \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\bar{\mathbf{I}}}^{-1}}{2} + V(\boldsymbol{\alpha}) - \mu^{eq} \right) d\mathbf{L}_s \\ T\sigma = - \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{\partial}{\partial \mathbf{L}_s} \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\bar{\mathbf{I}}}^{-1}}{2} \right) d\mathbf{L}_s \end{cases} \quad (4.12)$$

C Phenomenological law

According to the second law of thermodynamics and to the hypothesis of local equilibrium in the $(\mathbf{e}, \mathbf{L}_s)$ space, a phenomenological relationship between the flux $\mathbf{J}_s^{\mathbf{L}}$ and the force $\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{L}_s} + \bar{\bar{\mathbf{I}}}^{-1} \cdot \mathbf{L}_s$ imposes. A tensor of Onsager coefficients $\bar{\bar{\mathcal{L}}}_{\mathbf{LL}}$ is introduced as a proportionality constant.

$$\mathbf{J}_s^{\mathbf{L}} = - \bar{\bar{\mathcal{L}}}_{\mathbf{LL}} \left(\frac{k_B T}{v f} \frac{\partial f}{\partial \mathbf{L}_s} + \bar{\bar{\mathbf{I}}}^{-1} \cdot \mathbf{L}_s \right) = - \bar{\bar{\beta}} \left(f \mathbf{L}_s + \frac{k_B T}{v} \bar{\bar{\mathbf{I}}} \frac{\partial f}{\partial \mathbf{L}_s} \right) \quad (4.13)$$

Here $\bar{\bar{\beta}}$ is the damping tensor, defined as $\bar{\bar{\beta}} = \frac{1}{f} \bar{\bar{\mathcal{L}}}_{\mathbf{LL}} \bar{\bar{\mathbf{I}}}^{-1}$. As the Onsager coefficients are a reflection of the symmetry of the system [3], the tensor $\bar{\bar{\mathcal{L}}}_{\mathbf{LL}}$, and consequently $\bar{\bar{\beta}}$ mirror the axial symmetry of the system. Under these conditions the damping tensor writes as: $\bar{\bar{\beta}} = \beta_1(\bar{\bar{\mathbf{U}}} - \mathbf{e}\mathbf{e}) + \beta_3\mathbf{e}\mathbf{e}$.

4.2.3 The dynamic and the stochastic equations

A The angular momentum conservation law

Space-fixed frame With the expression of the flux $\mathbf{J}_s^{\mathbf{L}}$ (4.13), the dynamic equation (4.10) is rewritten:

$$\frac{d}{dt} \langle \mathbf{L}_s \rangle = - \bar{\bar{\beta}}_s \cdot \langle \mathbf{L}_s \rangle + \mathbf{N}_s - \mathbf{e} \times \frac{1}{n} \frac{\partial}{\partial \mathbf{e}} \bar{\bar{P}} \quad (4.14)$$

The dynamic equation (4.14) takes the shape of a rotational analogue of the Langevin equation written for the space-fixed reference frame.

Magnetisation (e)-fixed frame To get more insight, the equation will be translated in terms of the average angular velocity $\boldsymbol{\Omega} = \langle \boldsymbol{\omega} \rangle$. As the inertial tensor $\bar{\bar{\mathbf{I}}}_s$ is orientation \mathbf{e} dependent, hence time dependent in the space-fixed reference frame, the dynamic equation

(4.14) will be written in the magnetisation-fixed frame moving with the angular velocity $\mathbf{\Omega}_b$. The magnetisation-fixed frame is defined by the principal axes of the inertial tensor, the axial vector \mathbf{e} of the magnetisation and two perpendicular vectors found in the plane orthogonal to \mathbf{e} . In this frame, the inertial tensor $\overline{\overline{I}}_b$, as well as the damping tensor $\overline{\overline{\beta}}_b$ are diagonal and time independent with (I_1, I_1, I_3) and respectively, $(\beta_1, \beta_1, \beta_3)$ components.

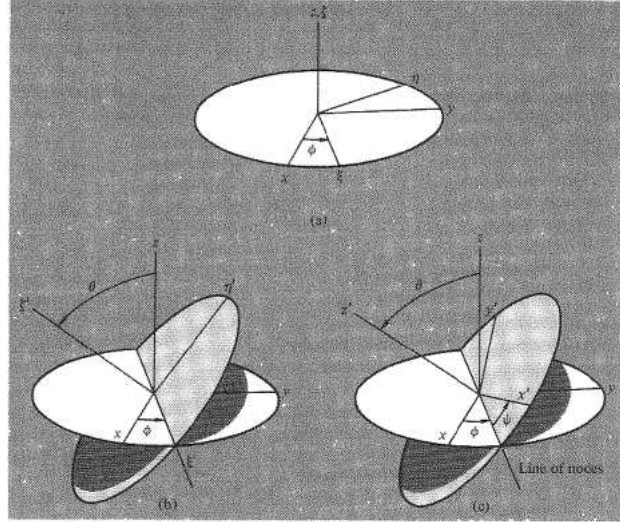


Figure 4.1: Dynamics of the magnetisation as a superposition of three elementary rotations: the rotation around the fixed axis Oz with $\dot{\phi}$ (see a), the rotation around the line of nodes $O\xi'$ with $\dot{\theta}$ (see b) and the rotation around the mobile axis Oz' with $\dot{\psi}$ (see c). The three frames of reference are identified: the fixed-space frame $Oxyz$ (see a) and the two mobile frames: $O\xi'\eta'\zeta'$ (see b) and the $Ox'y'z'$ (see c). The magnetisation vector \mathbf{M} is parallel with the two parallel axes Oz' and $O\xi'$ and with the unit vector \mathbf{e} . (Figure taken for the book of “Classical mechanics“ written by H. Goldstein, C. Poole and J. Safko [4])

Let $\overline{\overline{R}}$ be the rotation matrix which maps vectors from the space-fixed $Oxyz$ to the magnetisation-fixed frame $Ox'y'z'$, as $\mathbf{N}_b = \overline{\overline{R}}\mathbf{N}_s$. Using the property of the rotation matrix: $\frac{d\overline{\overline{R}}}{dt} = -\mathbf{\Omega}_b \times \overline{\overline{R}}$, equation (4.14) transforms then to:

$$\frac{d}{dt} \langle \mathbf{L}_b \rangle = -\mathbf{\Omega}_b \times \langle \mathbf{L}_b \rangle - \overline{\overline{\beta}}_b \cdot \langle \mathbf{L}_b \rangle + \mathbf{N}_b - \mathbf{e} \times \frac{1}{n} \frac{\partial \overline{\overline{P}}}{\partial \mathbf{e}} \quad (4.15)$$

Changing the variable from the average angular momentum $\langle \mathbf{L}_b \rangle$ to the average angular velocity $\mathbf{\Omega}_b$, the angular momentum conservation law written for the magnetisation-fixed

frame is:

$$\frac{d\mathbf{\Omega}_b}{dt} = - \overbrace{\left[\bar{\beta}_b - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 \mathbf{e} \times \bar{\mathbf{U}} \right]}^{\bar{\beta}^{rot}} \cdot \mathbf{\Omega}_b + \bar{I}_b^{-1} \cdot \mathbf{N}_b - \bar{I}_b^{-1} \cdot \left(\mathbf{e} \times \frac{1}{n} \frac{\partial \bar{\mathbf{P}}}{\partial \mathbf{e}} \right) \quad (4.16)$$

where a new antisymmetric tensor $\bar{\beta}^{rot}$ is defined. The antisymmetry of the tensor is a consequence of rotation:

$$\bar{\beta}^{rot} = \bar{\beta}_b - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 \mathbf{e} \times \bar{\mathbf{U}} = \begin{pmatrix} \beta_1 & \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & 0 \\ - \left(\frac{I_3}{I_1} - 1 \right) \Omega_3 & \beta_1 & 0 \\ 0 & 0 & \beta_3 \end{pmatrix} \quad (4.17)$$

The same equation written for each component is:

$$\begin{cases} \dot{\Omega}_1 = -\beta_1 \Omega_1 - \Omega_3 \left(\frac{I_3}{I_1} - 1 \right) \Omega_2 - \frac{1}{I_1} M_s H_2 - \frac{1}{I_1} \left(\frac{1}{n} \frac{\partial \bar{\mathbf{P}}}{\partial \mathbf{e}} \right)_2 \\ \dot{\Omega}_2 = -\beta_1 \Omega_2 + \Omega_3 \left(\frac{I_3}{I_1} - 1 \right) \Omega_1 + \frac{1}{I_1} M_s H_1 + \frac{1}{I_1} \left(\frac{1}{n} \frac{\partial \bar{\mathbf{P}}}{\partial \mathbf{e}} \right)_1 \\ \dot{\Omega}_3 = -\beta_3 \Omega_3 \end{cases} \quad (4.18)$$

The average angular momentum conservation law written for the magnetisation frame (4.15) or its equivalents (4.16) or (4.18) represents a key formula for the derivation of the generalised Gilbert equation.

B The Fokker-Planck equation

Space-fixed frame With the expression of the flux $\mathbf{J}_s^{\mathbf{L}}$ (4.13), the conservation equation (4.1) transforms to:

$$\frac{\partial f(\mathbf{e}, \mathbf{L}_s)}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \cdot \left(f \mathbf{e} \times \bar{I}_s^{-1} \cdot \mathbf{L}_s \right)_{\mathbf{L}_s} - \mathbf{N}_s \cdot \frac{\partial f}{\partial \mathbf{L}_s} + \frac{\partial}{\partial \mathbf{L}_s} \left(f \bar{\beta}_s \cdot \mathbf{L}_s + \frac{k_B T}{v} \bar{\beta}_s \bar{I}_s \cdot \frac{\partial f}{\partial \mathbf{L}_s} \right) \quad (4.19)$$

The equation (4.19) is the Fokker-Planck equation written for the space-fixed frame. It gives the variation with time of the number of magnetic particles f having the orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ and angular momenta $(\mathbf{L}_s, \mathbf{L}_s + d\mathbf{L}_s)$. In deriving it, use has been made of the axial symmetry of the inertial tensor. Having \mathbf{e} as axis of symmetry, all pertinent quantities are independent of ψ , the angle of rotation around \mathbf{e} . In this case, $\partial/\partial \boldsymbol{\alpha}$ is replaced with $\partial/\partial \mathbf{e}$, $\dot{\boldsymbol{\alpha}}$ is replaced with $\boldsymbol{\omega} \times \mathbf{e}$ and $\bar{\boldsymbol{\Lambda}} = -\mathbf{e} \times \bar{\mathbf{U}}$.

Magnetization(e**)-fixed frame** The Fokker-Planck equation written for the magnetisation-fixed reference frame takes the following form (see appendix 4.A.3 for a detailed derivation):

$$\frac{\partial f_{(\mathbf{e}, \mathbf{L}_b)}}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \left(f \mathbf{e} \times \bar{\mathbf{I}}_b^{-1} \cdot \mathbf{L}_b \right)_{\mathbf{L}_b} + \frac{\partial}{\partial \mathbf{L}_b} \left(f \bar{\beta}^{rot} \cdot \mathbf{L}_b - f \mathbf{N}_b + k_B T \bar{\beta}_b \bar{\mathbf{I}}_b \cdot \frac{\partial f}{\partial \mathbf{L}_b} \right) \quad (4.20)$$

It gives the variation with time of the number of magnetic particles $f_{(\mathbf{e}, \mathbf{L}_b)}$ having the orientation $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ and angular momenta $(\mathbf{L}_b, \mathbf{L}_b + d\mathbf{L}_b)$.

4.3 It's a matter of time scales!

In this section it is proved that a cosmetic treatment performed on the derived averaged dynamic (4.16) and stochastic equations (4.20) of the magnetic particles in terms of angular momenta \mathbf{L}_b , lead to the generalised Gilbert equation and to a reformulation of the Fokker-Planck equation (4.20).

Supposing the averaged axial component of the angular momenta is conserved $\langle L_3 \rangle = cst.$ ($\beta_3 = 0$), the averaged dynamic equation (4.15) (or its equivalents (4.16), (4.18)) introduces one characteristic time scale $\tau = \beta_1^{-1}$. It separates the behaviour of the magnetic system of particles in two regimes: the diffusion regime and the inertial regime (to be treated separately in subsection 4.3.1, respectively 4.3.2). The Gilbert equation [5] and the stochastic Brown equation [6] are derived in the diffusion regime as long time scale limits of a more complex behaviour. For shorter time scales, in the inertial regime, a generalised Gilbert equation is derived leading to a new phenomenon in magnetisation dynamics: *nutaton*.

4.3.1 Diffusion regime or Gilbert's and Brown's stochastic equations

A Brief presentation

For times t much longer than $\tau = \beta_1^{-1}$, the inertial terms $\dot{\Omega}_1$ and $\dot{\Omega}_2$ are negligible in respect to their damping correspondents $\beta_1 \Omega_1$, respectively $\beta_1 \Omega_2$ leading to a regime usually called in thermodynamics or statistical physics as the *diffusion regime*. It is typically characterised by two equations: the Fick's law and the Smoluchowski equation. More, the distribution function f is approximately a Maxwellian with a non vanishing average velocity.

The distribution function In our case, of magnetic particles of orientation \mathbf{e} and angular momenta \mathbf{L} , the diffusion is related to the variation with time of the density of magnetic particles $n_{(\mathbf{e})}$. Due to thermalisation, after a time $t \gg \tau$, the magnetic particles follow a distribution function f^{diff} close to a Maxwellian centered on the average angular

momenta \mathbf{L}_s :

$$f_{(\mathbf{e}, \mathbf{L}_s, t)}^{diff} \simeq f_{(\mathbf{e}, \mathbf{L}_s, t)}^{l.eq.} \propto \exp \left\{ \frac{\mu_{l.eq.} - (\mathbf{L}_s - \langle \mathbf{L}_s \rangle)(\mathbf{L}_s - \langle \mathbf{L}_s \rangle) : \bar{\bar{I}}^{-1} / 2 - V_{(\mathbf{e})}}{k_B T / v} \right\} \quad (4.21)$$

With the approximation of the distribution function f (4.21), the pressure tensor (4.9) reduces to the diagonal form $\bar{\bar{P}}^{diff} = n \frac{k_B T}{v} \bar{\bar{U}}$.

The motion equation Neglecting the inertial terms, the averaged dynamic equation (4.16) leads to a typical motion equation for the diffusion regime. This equation (4.22) written in terms of the averaged angular velocity $\mathbf{\Omega}_b$ will later lead to the Gilbert, Landau-Lifschitz and Brown equations.

$$0 \simeq -\bar{\bar{\beta}}_b^* \cdot \mathbf{\Omega}_b + \bar{\bar{I}}_b^{-1} \cdot \mathbf{N}_b - \bar{\bar{I}}_b^{-1} \cdot \left(\mathbf{e} \times \frac{k_B T}{n v} \frac{\partial n}{\partial \mathbf{e}} \right) \quad (4.22)$$

Here we introduced a damping tensor $\bar{\bar{\beta}}_b^*$ based on the previously defined time dependent tensor $\bar{\bar{\beta}}^{rot}$ (4.17). It is a tensor of constant coefficients obtained for the particular case of conserved axial angular momentum $\mathbf{L}_3 = I_3 \mathbf{\Omega}_3$ ($\beta_3 = 0$).

$$\bar{\bar{\beta}}_b^* = \bar{\bar{\beta}}_b - \frac{M_s}{\gamma} \left(\frac{1}{I_1} - \frac{1}{I_3} \right) \mathbf{e} \times \bar{\bar{U}} = \beta_1 \begin{pmatrix} 1 & 1/\alpha^* & 0 \\ -1/\alpha^* & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} = \frac{\bar{\bar{\beta}}_b}{\alpha^*} \begin{pmatrix} \alpha^* & 1 & 0 \\ -1 & \alpha^* & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (4.23)$$

where

$$\gamma \stackrel{def}{=} \frac{M_s}{I_3 \Omega_3} \quad \gamma^* = \frac{\gamma}{1 - I_1/I_3} \quad \alpha^* = \frac{\gamma \beta_1 I_1}{M_s (1 - I_1/I_3)} = \frac{\gamma^* \beta_1 I_1}{M_s} \quad (4.24)$$

B Gilbert's equation

With the properties of the angular velocity $\mathbf{\Omega}_b$ in respect to the magnetisation vector $\mathbf{M} = M_s \mathbf{e}$ (4.25), the Gilbert equation easily follows from the motion equation (4.22):

$$\frac{d\mathbf{M}}{dt} = \mathbf{\Omega}_b \times \mathbf{M} \Rightarrow \mathbf{\Omega}_b = \frac{\mathbf{M}}{M_s^2} \times \frac{d\mathbf{M}}{dt} + \frac{\mathbf{M}}{I_3 \gamma} \Rightarrow \bar{\bar{I}}_b \bar{\bar{\beta}}_b^* \mathbf{\Omega}_b = \frac{\beta_1 I_1}{M_s^2} \mathbf{M} \times \frac{d\mathbf{M}}{dt} + \frac{\beta_1 I_1}{M_s \alpha^*} \frac{d\mathbf{M}}{dt} \quad (4.25)$$

$$\frac{d\mathbf{M}}{dt} = \gamma^* \mathbf{M} \times \left(\mathbf{H} - \eta \frac{d\mathbf{M}}{dt} - \frac{k_B T}{n v} \frac{1}{M_s} \frac{\partial n}{\partial \mathbf{e}} \right) = \gamma^* \mathbf{M} \times \left(\mathbf{H}_{eff} - \eta \frac{d\mathbf{M}}{dt} \right) \quad (4.26)$$

The equation (4.26) is the Gilbert equation, where in the previously defined γ^* and α^* (4.24) the gyromagnetic, respectively the Landau-Lifshitz damping coefficients are identified, while the Gilbert damping coefficient η was introduced as:

$$\eta = \frac{\beta_1 I_1}{M_s^2} \quad (4.27)$$

An effective magnetic field was also defined accounting for the applied magnetic field \mathbf{H} and for the nonuniform distribution of the orientations of the magnetisations: $\mathbf{H}_{eff} = \mathbf{H} - \frac{k_B T}{n v} \frac{1}{M_s} \frac{\partial n}{\partial \mathbf{e}}$.

C Landau-Lifshitz equation (LL)

With the expression of the average angular velocity $\boldsymbol{\Omega}_b$, the LL equation is easily deduced. The same motion equation (4.22), leads to the introduction of the tensor $\overline{\overline{D}}$, the gyromagnetic coefficient g^* and the damping coefficient h^* :

$$\boldsymbol{\Omega}_b = \overline{\overline{\beta}}_b^{*-1} \overline{\overline{I}}^{-1} \cdot \left(\mathbf{N}_b - \mathbf{M} \times \frac{1}{n} \frac{1}{M_s} \frac{\partial}{\partial \mathbf{e}} \cdot \overline{\overline{P}}^{diff} \right) \Leftrightarrow \boldsymbol{\Omega}_b = \frac{v \overline{\overline{D}}}{k_B T} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \quad (4.28)$$

where :

$$\overline{\overline{D}} = \frac{k_B T}{v} \overline{\overline{\beta}}_b^{*-1} \overline{\overline{I}}^{-1} = g^* \frac{k_B T}{v} \begin{pmatrix} \alpha^* & -1 & 0 \\ 1 & \alpha^* & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (4.29)$$

$$g^* = \frac{\gamma^*}{M_s(1 + \alpha^{*2})} \quad h^* = \frac{\alpha^* \gamma^*}{M_s(1 + \alpha^{*2})} = \alpha^* g^* \quad (4.30)$$

The expression of the angular velocity $\boldsymbol{\Omega}_b$ (4.28) leads to the Landau-Lifshitz equation (4.31):

$$\frac{d\mathbf{M}}{dt} = \left[\frac{v \overline{\overline{D}}}{k_B T} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \right] \times \mathbf{M} \Rightarrow \frac{d\mathbf{M}}{dt} = g^* M_s \mathbf{M} \times \mathbf{H}_{eff} + h^* (\mathbf{M} \times \mathbf{H}_{eff}) \times \mathbf{M} \quad (4.31)$$

D Brown's stochastic equation

The conservation equation for the density of particles $n_{(\mathbf{e})}$ (4.6) represents the starting point for the magnetic analogous of Smoluchowski's equation, the Brown's stochastic equation.

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \left(n \mathbf{e} \times \overline{\overline{I}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) = \frac{\partial}{\partial \mathbf{e}} (n \mathbf{e} \times \boldsymbol{\Omega}_b) \Leftrightarrow \frac{\partial n}{\partial t} = - \frac{\partial \mathbf{j}^e}{\partial \mathbf{e}} \quad (4.32)$$

With the expression of the angular velocity $\boldsymbol{\Omega}_b$ (4.28), the flux of particles \mathbf{j}^e is derived. It represents the number of particles changing their magnetic orientation from \mathbf{e} to $\mathbf{e} + d\mathbf{e}$ per unit of time, and is caused by the application of a magnetic field \mathbf{H} and by the nonuniform repartition of the orientations in the 4π solid angle. The magnetic field generates a drift flux, while the nonuniform repartition generates a diffusion flux. The magnetic analogous of Fick's law is then obtained (4.33):

$$\begin{aligned}\mathbf{j}^e &= n\boldsymbol{\Omega}_b \times \mathbf{e} = \frac{nv}{k_B T} \left[\overline{\overline{D}} \cdot (\mathbf{M} \times \mathbf{H}_{eff}) \right] \times \mathbf{e} = \frac{nv\overline{\overline{D}}}{k_B T} \cdot M_s \mathbf{H}_{eff} \Rightarrow \\ \mathbf{j}^e &= -\frac{nv}{k_B T} \overline{\overline{D}} \cdot \frac{\partial V}{\partial \mathbf{e}} - \overline{\overline{D}} \cdot \frac{\partial n}{\partial \mathbf{e}}\end{aligned}\quad (4.33)$$

With the conservation law (4.32) and the expression of the diffusion flux \mathbf{j}^e (4.33), the Brown equation derived in the framework of stochastic processes is easily obtained:

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \cdot \left[\frac{nv\overline{\overline{D}}}{k_B T} \cdot \frac{\partial}{\partial \mathbf{e}} \left(V + \frac{k_B T}{v} \ln n \right) \right] \Leftrightarrow \quad (4.34)$$

$$\begin{aligned}\frac{\partial n}{\partial t} &= \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta \left[\left(h^* \frac{\partial V}{\partial \theta} - g^* \frac{1}{\sin \theta} \frac{\partial V}{\partial \phi} \right) n + h^* \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \right] \right\} \\ &+ \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \left\{ \left(g^* \frac{\partial V}{\partial \theta} + \frac{1}{\sin \theta} h^* \frac{\partial V}{\partial \phi} \right) n + h^* \frac{k_B T}{v} \frac{1}{\sin \theta} \frac{\partial n}{\partial \phi} \right\}\end{aligned}\quad (4.35)$$

4.3.2 The inertial regime or the generalised Gilbert equation

A The generalised Gilbert equation

For time scales smaller than $\tau = \beta_1^{-1}$, the inertial terms $\dot{\Omega}_1$ and $\dot{\Omega}_2$ from the equation (4.16) or better its equivalent (4.18), gain importance in respect to their damping correspondents $\beta_1 \Omega_1$, respectively $\beta_1 \Omega_2$ and the whole equation has to be taken into account. Supposing the axial component of the angular momentum $L_3 = I_3 \Omega_3 = \frac{M_s}{\gamma} (\beta_3 = 0)$ is conserved, the generalised Gilbert equation is retrieved (see the appendix 4.B.1 for explicit calculations):

$$\boxed{\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[\mathbf{H} - \frac{1}{n} \frac{\partial}{\partial \mathbf{M}} \overline{\overline{P}} - \eta \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2 \mathbf{M}}{dt^2} \right) \right]} \quad (4.36)$$

where

$$\alpha = \gamma \eta M_s \quad f = \frac{M_s}{\gamma I_1} = \frac{I_3 \Omega_3}{I_1} \quad \tau = \frac{1}{f\alpha} = \beta_1^{-1} \quad (4.37)$$

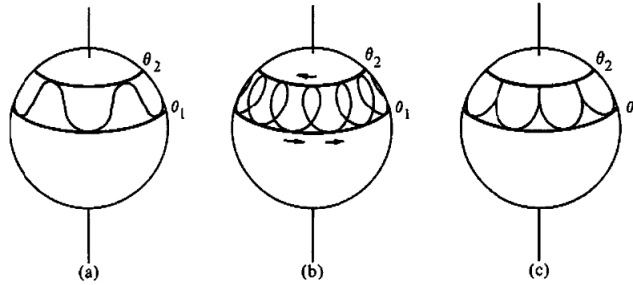


Figure 4.2: The possible locus shapes for a heavy symmetrical top without damping differentiated by the various initial conditions. The top *nutates*: nods up and down between two limit angles θ_1 and θ_2 . The figure c) is representative for a heavy top with no initial velocity, meaning for $\dot{\phi}_{(0)} = 0$ and $\dot{\theta}_{(0)} = 0$ (Figure taken from the book of "Classical mechanics" written by H. Goldstein and C. Poole and J. Safko [4]).

A.1 Expected behaviour As we will soon see, the effect of the acceleration term $\tau \frac{d^2 \mathbf{M}}{dt^2}$ gives a new dynamics trait called *nutations*. The equation (4.36) or its equivalents in terms of angular momenta \mathbf{L}_b (4.15), (4.16), (4.18) with $\beta_3 = 0$ and $L_3 = \frac{M_s}{\gamma}$, describe a movement analogous to that of a heavy symmetrical top with damping. In the line of the mechanical equivalence, one would expect to encounter in the dynamics of the magnetisation some of the features typical for the case of the top. Meaning, without damping, one would expect the magnetisation to nutate and precess (see Figure 4.2). Faster the magnetisation would spin (bigger the Ω_3 , hence bigger the frequency f), smaller the nutation amplitude would be, and bigger the nutation frequency. With damping, for a sufficiently big Ω_3 (or equivalently f), the nutation would be damped out very fast becoming unobservable. The magnetisation would then appear to precess uniformly around the applied magnetic field, as "a *pseudoregular* precession". [4]

A.2 Analytical convergence towards the Gilbert equation With the expression of the generalised Gilbert equation (4.36), the existence of two regimes separated by a characteristic time scale τ is clearer. The main unknown of the equation is the principal moment of inertia I_1 . If I_1 is null, the magnetisation's acceleration $\frac{d^2 \mathbf{M}}{dt^2}$ disappears, the Gilbert equation is retrieved and a mechanical equivalent of the magnetisation dynamics does not exist as Gilbert predicted¹.

If however, a mechanical equivalent exists ($I_1 \neq 0$), the Gilbert equation is retrieved for time scales bigger than a characteristic time τ . In order to see the convergence from

¹"One can show that the inertial tensor must have a single nonzero term corresponding to the rotational inertia for rotation about the principal axis. I was unable to conceive of a physical object with an inertial tensor of this kind." - Gilbert, 1956 [5]

the generalised Gilbert (4.36) to the Gilbert equation (4.26), two terms of interest will be compared from the generalised expression:

$$\left\{ \begin{array}{l} \mathbf{M} \times \frac{d\mathbf{M}}{dt} = \mathbf{M} \times (\boldsymbol{\Omega} \times \mathbf{M}) \\ \tau \mathbf{M} \times \frac{d^2\mathbf{M}}{dt^2} = \mathbf{M} \times \left(\tau \frac{d\boldsymbol{\Omega}}{dt} \times \mathbf{M} \right) + \tau \mathbf{M} \left(\boldsymbol{\Omega} \times \frac{d\mathbf{M}}{dt} \right) = \mathbf{M} \times \left(\tau \frac{d\boldsymbol{\Omega}}{dt} \times \mathbf{M} \right) - \frac{1}{\eta\gamma} \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt} \end{array} \right. \quad (4.38)$$

For long time scales $t \gg \tau$, the term containing the magnetisation's acceleration $\frac{d^2\mathbf{M}}{dt^2}$ converges to $-\frac{1}{\eta\gamma} \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt}$, leading to the Gilbert equation with a correction performed on the gyromagnetic coefficient $\gamma^* = \frac{\gamma}{1 - I_1/I_3}$.

$$\gamma\eta \mathbf{M} \times \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2\mathbf{M}}{dt^2} \right) \xrightarrow{t \gg \tau} \gamma\eta \mathbf{M} \times \frac{d\mathbf{M}}{dt} - \frac{I_1}{I_3} \frac{d\mathbf{M}}{dt} \quad (4.39)$$

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[\mathbf{H} - \frac{1}{n M_s} \frac{\partial \bar{P}}{\partial \mathbf{e}} - \eta \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2\mathbf{M}}{dt^2} \right) \right] \quad (4.40)$$

$$\begin{array}{l} \searrow t \gg \tau \\ \frac{d\mathbf{M}}{dt} = \gamma^* \mathbf{M} \times \left(\mathbf{H} - \frac{k_B T}{nv} \frac{1}{M_s} \frac{\partial n}{\partial \mathbf{e}} - \eta \frac{d\mathbf{M}}{dt} \right) \end{array} \quad (4.41)$$

B The Fokker-Planck equation

The Fokker-Planck equation corresponding to the generalised Gilbert equation is retrieved from the equation (4.20), for the distribution $f(\mathbf{e}, \mathbf{L}_b)$ of magnetic particles having the orientation within $(\mathbf{e}, \mathbf{e} + d\mathbf{e})$ and the angular momentum within $(\mathbf{L}_b, \mathbf{L}_b + d\mathbf{L}_b)$: (see appendix 4.A.3 for detailed calculations)

$$\frac{\partial f(\mathbf{e}, \mathbf{L}_b)}{\partial t} = \frac{\partial}{\partial \mathbf{e}} \cdot \left(f \mathbf{e} \times \bar{I}_b^{-1} \cdot \mathbf{L}_b \right)_{\mathbf{L}_b} + \frac{\partial}{\partial \mathbf{L}_b} \cdot \left[\bar{\beta}_b^* \cdot f \mathbf{L}_b - f \mathbf{N}_b + \frac{k_B T}{v} \bar{\beta}_b \bar{I}_b \cdot \frac{\partial f}{\partial \mathbf{L}_b} \right] \quad (4.42)$$

As $\bar{\beta}_b^{*-1} = \frac{\bar{D}\bar{I}}{k_B T} = \frac{I_1 \bar{D}}{k_B T}$ the Fokker-Planck equation for the magnetisation-fixed frame is rewritten as:

$$\boxed{\frac{\partial f(\mathbf{e}, \mathbf{L}_b)}{\partial t} = \left(\frac{\partial}{\partial \mathbf{e}} \right)_{\mathbf{L}_b} \cdot \left(f \mathbf{e} \times \bar{I}_b^{-1} \cdot \mathbf{L}_b \right) + \frac{\partial}{\partial \mathbf{L}_b} \cdot \left[\bar{\beta}_b^* \cdot \left(f \mathbf{L}_b - \frac{f I_1 \bar{D}}{k_B T} \cdot \mathbf{N}_b + \frac{I_1^2 \bar{D}}{\tau} \cdot \frac{\partial f}{\partial \mathbf{L}_b} \right) \right]} \quad (4.43)$$

4.4 Numerical solution of the generalised Gilbert equation without noise

Having introduced a characteristic relaxation time τ , the nutation time, separating two regimes - the inertial and the diffusion regime, having derived their corresponding dynamic and stochastic equations: the Gilbert and Brown equations, respectively the generalised Gilbert and the Fokker-Planck equations, the next question to be answered is: what are the experimental predictions? To answer it, the solution for the system of equations (4.18) with $\beta_3 = 0$ and $L_3 = \frac{M_s}{\gamma}$ has to be found first. As it is quite impossible to solve the system analytically, the numerical solution is sought.

4.4.1 Analytical simplification

The magnetisation trajectory is given by the system of equations (4.18) with $\beta_3 = 0$ and $L_3 = \frac{M_s}{\gamma}$, which is the equivalent formulation of the generalised Gilbert equation (4.36) in terms of angular momenta. As the distribution function f is not known for time scales smaller than τ , hence the pressure tensor \overline{P} is not known, only the solution without thermal fluctuations will be presented.

$$\begin{cases} \dot{\Omega}_1 = -\beta_1 \Omega_1 + \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_2 - \frac{1}{I_1} M_s H_2 \\ \dot{\Omega}_2 = -\beta_1 \Omega_2 - \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_1 + \frac{1}{I_1} M_s H_1 \\ I_3 \Omega_3 = \frac{M_s}{\gamma} \end{cases} \quad (4.44)$$

where

$$\begin{cases} \Omega_1 = \dot{\phi} \sin \theta \sin \psi + \dot{\theta} \cos \psi \\ \Omega_2 = \dot{\phi} \sin \theta \cos \psi - \dot{\theta} \sin \psi \\ \Omega_3 = \dot{\psi} + \dot{\phi} \cos \theta \end{cases} \quad (4.45)$$

Finding the magnetisation trajectory is equivalent to finding the temporal dependence of the angles defining the orientation of the magnetisation: $\phi(t)$, $\theta(t)$, $\psi(t)$. After some manipulations performed on the system of equations (4.70), a second order differential system is obtained with the variables of interest $\theta(t)$, $\phi(t)$, $\psi(t)$: (see appendix 4.B.2 for the details of the calculation)

$$\begin{cases} \ddot{\phi} \sin \theta = -f\alpha \dot{\phi} \sin \theta - 2 \cos \theta \dot{\phi} \dot{\theta} + f\dot{\theta} + f\gamma H_{\xi'} \\ \ddot{\theta} = -f\alpha \dot{\theta} - f\dot{\phi} \sin \theta + \sin \theta \cos \theta \dot{\phi}^2 - f\gamma H_{\eta'} \\ \dot{\psi} = \Omega_3 - \dot{\phi} \cos \theta \end{cases} \quad (4.46)$$

where

$$\begin{cases} H_{\xi'} = H \sin \phi \sin \beta \\ H_{\eta'} = H \cos \phi \cos \theta \sin \beta + H \sin \theta \cos \beta \\ H_{\zeta'} = -H \cos \phi \sin \theta \sin \beta + H \cos \theta \cos \beta \end{cases} \quad f = \frac{M_s}{\gamma I_1} = \frac{L_3}{I_1} \quad (4.47)$$

Here $H_{\xi'}$, $H_{\eta'}$ and $H_{\zeta'}$ are the components of an applied magnetic field \mathbf{H} in the $\xi'\eta'\zeta'$ frame (see Fig. 4.1), where the field \mathbf{H} forms an angle β with the Oz axis in the yOz plane.

In order to compute the magnetisation trajectory, not only the initial orientation $(\theta_{(0)}, \phi_{(0)}, \psi_{(0)})$ of the magnetisation is necessary, but also the initial values for their first derivatives $(\dot{\theta}_{(0)}, \dot{\phi}_{(0)})$. The trajectory is calculated with the following initial conditions:

$$\begin{cases} \phi_{(0)} = 0 & \theta_{(0)} = -\pi/3 & \psi_{(0)} = 0 \\ \dot{\phi}_{(0)} = 0 & \dot{\theta}_{(0)} = 0 \end{cases} \quad (4.48)$$

The values of the parameters f , α , Ω_3 , γ and H are also needed. Typical values for α , γ and H are found in the literature [7], while for the newly introduced parameters f and Ω_3 no information is given. From the generalised Gilbert equation (4.36), the unknown frequency f is expected to have a strong influence on the dynamics of the magnetisation, while the parameter Ω_3 is not expected to influence it. In order to understand better the importance of the unknown parameter f , several cases will be studied for different values of f .

4.4.2 Nutation

The magnetisation dynamics is studied for times smaller than $\tau = (f\alpha)^{-1}$. It is followed by the observation of the dynamics convergence from the generalised Gilbert (inertial regime) to the usual Gilbert (diffusion regime) behaviour.

As expected, nutation is present for times $t \ll \tau = (f\alpha)^{-1}$. Its amplitude varies strongly with f , while its frequency varies linearly with f (see Fig. 4.3). The amplitude of the nutation is damped out after a time of approx. $\tau = (f\alpha)^{-1}$, and the nutation frequency doesn't vary with the damping α . For sufficiently big values of f , the nutation is barely seen and damped very quickly as expected. For typical values of $\gamma = 10^{10} \left(\frac{\text{A}}{\text{m}}\right)^{-1} \cdot \text{s}^{-1}$, $H = 1 \text{ T}$, $\beta = \pi$, $\Omega_3 = 10^{12} \text{ Hz}$ the nutation is hardly and shortly seen for a frequency of $f = 10^{12} \text{ Hz}$ (for a time $\tau = (f\alpha)^{-1}$ of the order of picoseconds.) (see Fig. 4.4). Also as expected, the trajectory $(\theta_{(t)}, \phi_{(t)})$ is independent of Ω_3 .

With the numerical solution of Gilbert's equation (with the same initial conditions and parameter values), it is possible to plot the Gilbert dynamics and inertial dynamics on the same graph for comparison. It is observed, that a small shift exist between the two, due to the initial amplitude of the nutation. In order for the two behaviours to completely superpose, different initial conditions would impose for the Gilbert equation. Even so, with the same initial conditions, in the diffusion regime, the two trajectories are quasi superposed

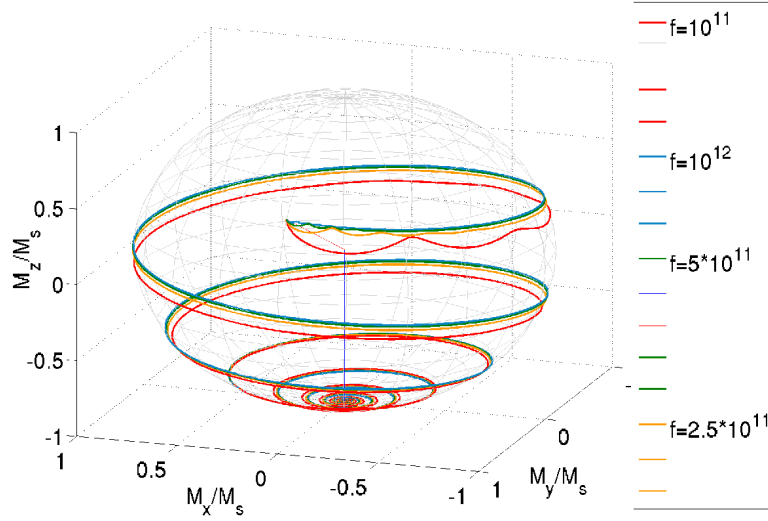
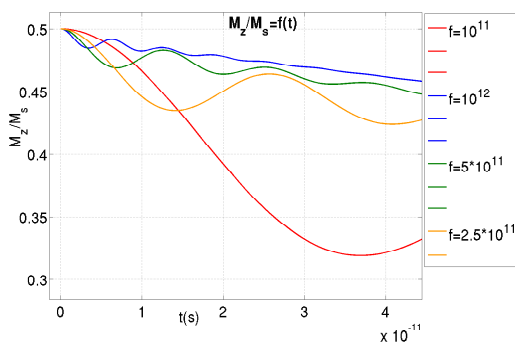
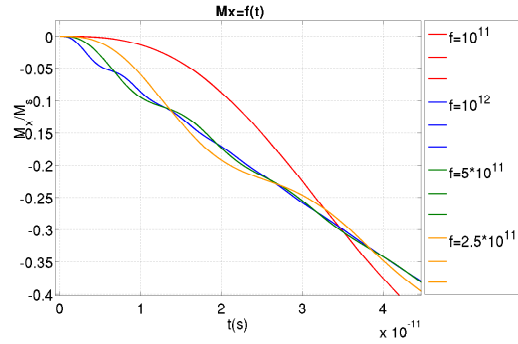
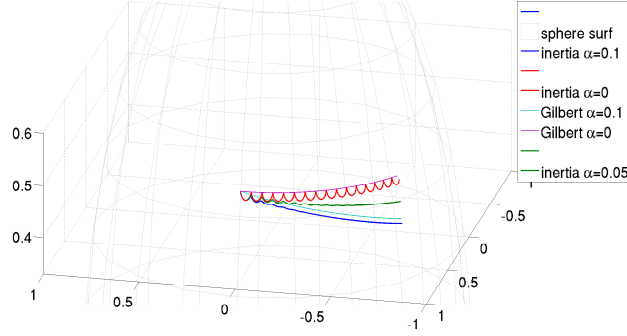
(a) The 3D trajectory of the magnetisation for different frequency values f .(b) The normalised M_z component of the magnetisation as a function of time, during a window time of 40 ps.(c) The normalised M_x component of the magnetisation as a function of time, during a window time of 40 ps.

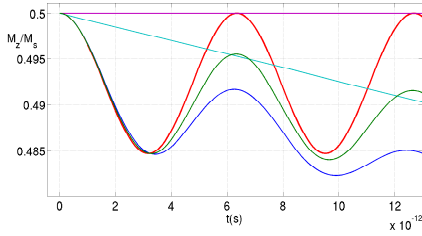
Figure 4.3: Nutation of the magnetisation for typical parameter values of $\alpha = 0.1$, $\gamma = 10^{10} \left(\frac{\text{A}}{\text{m}}\right)^{-1} \cdot \text{s}^{-1}$, $H = 1 \text{ T}$, $\beta = \pi$ and $\Omega_3 = 10^{10} \text{ Hz}$. The amplitude of the nutation varies strongly with f , while its frequency varies linearly with f .

(see Fig. 4.5).

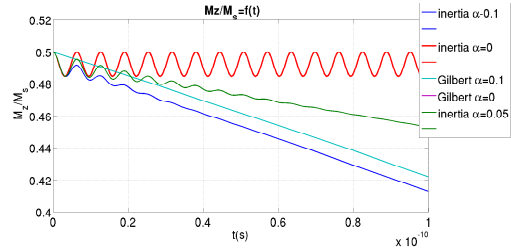
For the particular case of $f = 10^{11} \text{ Hz}$, the nutation amplitude is significant and it would have been observed until now, if the value of f had been equal or smaller than 10^{11} Hz (see Fig. 4.3). It is concluded that if the phenomenon really exists, the values of f have to be higher than $2.5 \cdot 10^{11} \text{ Hz}$, leading to characteristic times τ smaller than 40 ps. In order



(a) The 3D trajectory of the magnetisation for different values of the damping coefficient α .



(b) The normalised M_z component of the magnetisation as a function of time, during a window time of 12 ps.



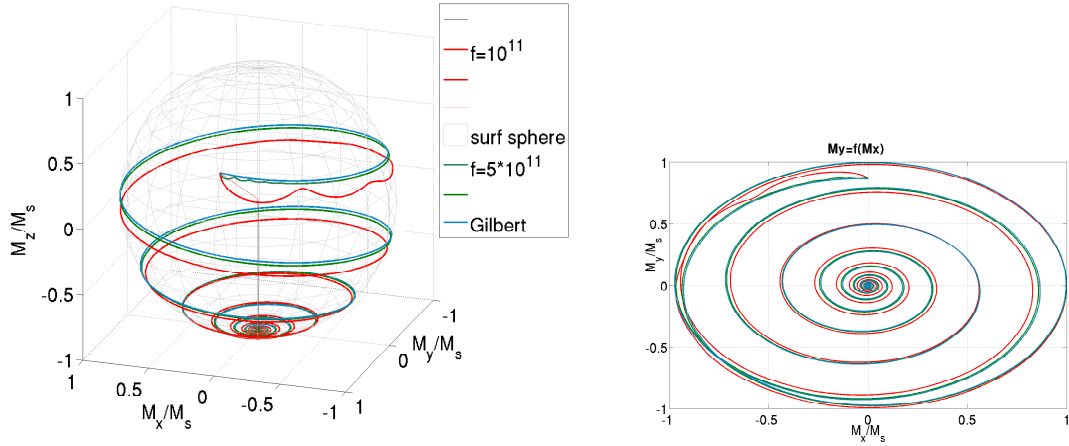
(c) The normalised M_z component of the magnetisation as a function of time.

Figure 4.4: Nutation of the magnetisation for typical parameter values of $\gamma = 10^{10} \left(\frac{\text{\AA}}{m}\right)^{-1} \cdot s^{-1}$, $H = 1 \text{ T}$, $\beta = \pi$, $f = 10^{12} \text{ Hz}$ and $\Omega_3 = 10^{12} \text{ Hz}$. With damping, the nutation is barely seen for a time of approximately 50 ps $[(f\alpha)^{-1}]$, after which the inertial behaviour is quasisuperposed with the Gilbert behaviour. As expected, the frequency of the nutation doesn't change with the damping coefficient α .

for the nutation to be observed, a temporal resolution of more than picoseconds has to be experimentally attained.

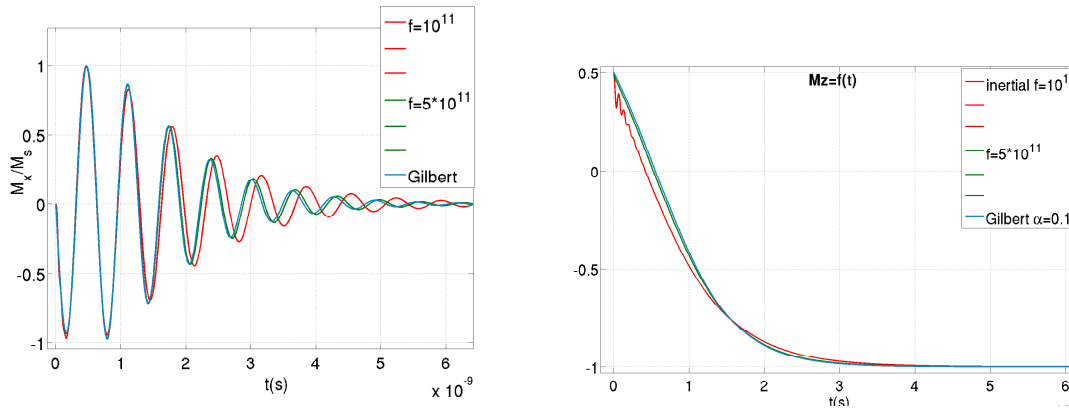
Conclusions

In this chapter we applied the laws of mesoscopic nonequilibrium thermodynamics (MNET) to an ensemble of magnetic particles \mathbf{m} to go beyond the usual Gilbert equation describing the dynamics of one magnetic particle, and Brown's stochastic equation, relating to the probability of having one magnetic particle oriented in a particular direction (θ, ϕ) .



(a) The 3D trajectory of the magnetisation for different values of the frequency f .

(b) Projection of the magnetisation trajectory given in 6 ns on the Oxy plane.



(c) The normalised M_x component of the magnetisation as a function of time, during a window time of 6 ns.

(d) The normalised M_z component of the magnetisation as a function of time, during a window time of 6 ns.

Figure 4.5: Nutation of the magnetisation for typical parameter values of $\gamma = 10^{10} \left(\frac{\text{A}}{\text{m}}\right)^{-1} \cdot \text{s}^{-1}$, $H = 1 \text{ T}$, $\beta = \pi$, $\alpha = 0.1$ and $\Omega_3 = 10^{10} \text{ Hz}$. The frequency of the nutation doesn't change with the damping coefficient α . After a time of the order of $(f\alpha)^{-1}$ the inertial behaviour is quasisuperposed with the Gilbert behaviour. The small shift between the two is due to the nonnegligible nutation amplitude. In order for the two behaviours to completely superpose, different initial conditions would impose for the Gilbert equation.

The ensemble was described by a distribution function f giving the number of particles

having an orientation $\boldsymbol{\alpha} = (\theta, \phi, \psi)$, and an angular momentum \mathbf{L} . Properties of inertia $\overline{\overline{I}}$ (not related to the inertia of matter, but to that of the magnetisation) and kinetic energy $\mathcal{K} = \mathbf{L}\mathbf{L} : \overline{\overline{I}}^{-1} / 2$ were associated to each magnetic particle \mathbf{m} . The average behaviour of the ensemble was derived by applying laws of thermodynamics to the ensemble of particles.

A characteristic time τ (*nutaton time*), related to the Gilbert's damping η and the inertia tensor $\overline{\overline{I}}$ separates two regimes: the *diffusion regime* or the long time scale limit, and the *inertial regime* or the short time scale limit. It was shown that the Gilbert, the Landau-Lifshitz and the Brown equations are representative for the first, while a generalised Gilbert equation (4.36) and a more complex Fokker-Planck are representative for the inertial regime.

The main feature of the generalised Gilbert equation in the inertial regime was shown to be nutation, given by the additional term: $\gamma\eta\tau \mathbf{M} \times \frac{d^2\mathbf{M}}{dt^2}$, where γ is a gyromagnetic coefficient. After solving numerically the equation, it was discovered that for sufficiently big values of the newly introduced frequency parameter $f = (\gamma\eta\tau M_s)^{-1}$ the nutation is barely observable in terms of its amplitude, as well as in terms of its duration. It is concluded that if such a phenomenon exists, the necessary temporal resolution for it to be observable has to be at least that of picoseconds (for typical values of $\gamma = 10^{10} \left(\frac{\text{A}}{\text{m}}\right)^{-1} \cdot \text{s}^{-1}$, $H = 1\text{ T}$ and $\alpha = 0.1$). The exact time scale cannot be predicted as the associated principal moment of inertia I_1 is unknown.

This model could represent a good starting point for other classic models accounting for example, for the spin transfer phenomenon, for different symmetries of the system (reflected by the matrix of Onsager coefficients and by the inertia tensor $\overline{\overline{I}}$), at short and long time scales under the presence of thermal fluctuations.

Next chapter: The purpose of the following chapter is precisely to build a classic framework accounting for the spin-transfer phenomenon having as cornerstone the classic treatment of the magnetisation dynamics presented here.

Appendix

4.A Derivations within the MNET framework

4.A.1 The average angular momentum

$$\begin{aligned}
\frac{\partial n \langle \mathbf{L}_s \rangle}{\partial t} &= \int \frac{\partial f}{\partial t} \mathbf{L}_s d\mathbf{L}_s = -\frac{\partial}{\partial \alpha} \int \mathbf{L}_s \left(f \bar{\Lambda}(\alpha) \bar{\Gamma}^{-1} \cdot \mathbf{L}_s \right) d\mathbf{L}_s + n \mathbf{N}(\alpha) + \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s \\
&= -\frac{\partial}{\partial \alpha} \left\{ \left[\int f(\mathbf{L}_s - \langle \mathbf{L}_s \rangle)(\mathbf{L}_s - \langle \mathbf{L}_s \rangle) d\mathbf{L}_s \right] \bar{\Gamma}^{-1} \bar{\Lambda}^t(\alpha) \right\} \\
&\quad - \frac{\partial}{\partial \alpha} \left\{ (n \langle \mathbf{L}_s \rangle \langle \mathbf{L}_s \rangle) \bar{\Gamma}^{-1} \bar{\Lambda}^t(\alpha) \right\} + n \mathbf{N}(\alpha) + \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s \\
&\Rightarrow n \frac{d}{dt} \langle \mathbf{L}_s \rangle = -\frac{\partial}{\partial \alpha} (\bar{P} \bar{\Lambda}^t) + n \mathbf{N}(\alpha) + \int \mathbf{J}_s^{\mathbf{L}} d\mathbf{L}_s
\end{aligned} \tag{4.49}$$

where \bar{P} stands for

$$\bar{P} \stackrel{def}{=} \left[\int f(\mathbf{L}_s - \langle \mathbf{L}_s \rangle)(\mathbf{L}_s - \langle \mathbf{L}_s \rangle) d\mathbf{L}_s \right] \bar{\Gamma}^{-1} \tag{4.50}$$

4.A.2 The entropy production σ

From the Gibbs postulate (4.3) and the conservation equation in the cartesian form (4.1), the expression of the entropy production $\sigma(\alpha)$ is derived.

The Gibbs postulate (4.5) and the conservation equation (4.1) are recalled:

$$Tns(\alpha) = - \int f \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\Gamma}^{-1}}{2} + V(\alpha) - \mu^{eq} \right) d\mathbf{L}_s \tag{4.51}$$

$$\frac{\partial f}{\partial t} = -\frac{\partial}{\partial \alpha} \left(f \bar{\Lambda} \bar{\Gamma}^{-1} \cdot \mathbf{L}_s \right) - \mathbf{N}(\alpha) \cdot \frac{\partial f}{\partial \mathbf{L}_s} - \frac{\partial \mathbf{J}_s^{\mathbf{L}}}{\partial \mathbf{L}_s} \tag{4.52}$$

To obtain σ , Tns is derived in respect to time:

$$T \frac{\partial (ns)}{\partial t} = -\frac{k_B T}{v} \int \frac{\partial f}{\partial t} (\ln f + 1) d\mathbf{L}_s - \int \frac{\partial f}{\partial t} \left(\frac{\mathbf{L}_s \mathbf{L}_s : \bar{\Gamma}^{-1}}{2} + V(\alpha) - \mu_{eq} \right) d\mathbf{L}_s \tag{4.53}$$

Each integral is calculated:

(a).

$$\begin{aligned}
& \int \frac{\partial f}{\partial t} (\ln f + 1) d\mathbf{L}_s \\
&= -\frac{\partial}{\partial \alpha} \int f (\ln f + 1) \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} d\mathbf{L}_s + \int f (\overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s) \cdot \frac{\partial}{\partial \alpha} (\ln f + 1) d\mathbf{L}_s \\
&\quad - \mathbf{N}(\alpha) \cdot \int \frac{\partial}{\partial \mathbf{L}_s} [f (\ln f + 1)] d\mathbf{L}_s + \mathbf{N}(\alpha) \cdot \int f \frac{1}{f} \frac{\partial f}{\partial \mathbf{L}_s} d\mathbf{L}_s \\
&\quad - \int \frac{\partial}{\partial \mathbf{L}_s} [\mathbf{J}_s^{\mathbf{L}} (\ln f + 1)] d\mathbf{L}_s + \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{1}{f} \frac{\partial f}{\partial \mathbf{L}_s} d\mathbf{L}_s \\
&= -\frac{\partial}{\partial \alpha} \int f \ln f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} d\mathbf{L}_s + \int f \frac{\partial}{\partial \alpha} \left(\overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} \right) \cdot \mathbf{L}_s d\mathbf{L}_s + \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{\partial (\ln f)}{\partial \mathbf{L}_s} d\mathbf{L}_s
\end{aligned} \tag{4.54}$$

(b).

$$\begin{aligned}
& \int \frac{\partial f}{\partial t} \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} d\mathbf{L}_s \\
&= -\int \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} \frac{\partial}{\partial \alpha} \left(f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} \right) d\mathbf{L}_s - \mathbf{N}(\alpha) \int \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} \frac{\partial f}{\partial \mathbf{L}_s} d\mathbf{L}_s - \int \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} \frac{\partial \mathbf{J}_s^{\mathbf{L}}}{\partial \mathbf{L}_s} d\mathbf{L}_s \\
&= -\frac{\partial}{\partial \alpha} \int f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} d\mathbf{L}_s + \int \left[f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} \right] \cdot \frac{1}{2} \frac{\partial}{\partial \alpha} \left(\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1} \right) d\mathbf{L}_s \\
&\quad + n \mathbf{N}(\alpha) \cdot \left(\overline{\overline{T}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) + \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{\partial}{\partial \mathbf{L}_s} \left(\frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} \right) d\mathbf{L}_s
\end{aligned} \tag{4.55}$$

(c).

$$\begin{aligned}
& \int \frac{\partial f}{\partial t} (V(\alpha) - \mu_{eq}) d\mathbf{L}_s \\
&= -\int \frac{\partial}{\partial \alpha} \left(f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} \right) (V - \mu_{eq}) d\mathbf{L}_s - \mathbf{N}(\alpha) \cdot \int (V - \mu_{eq}) \frac{\partial f}{\partial \mathbf{L}_s} d\mathbf{L}_s - \int \frac{\partial \mathbf{J}_s^{\mathbf{L}}}{\partial \mathbf{L}_s} (V - \mu_{eq}) d\mathbf{L}_s \\
&= -\frac{\partial}{\partial \alpha} \int f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \mathbf{L}_s} (V(\alpha) - \mu_{eq}) d\mathbf{L}_s + n \left(\overline{\overline{\Lambda \overline{\overline{T}}^{-1}} \cdot \langle \mathbf{L}_s \rangle \right) \cdot \frac{\partial V(\alpha)}{\partial \alpha}
\end{aligned} \tag{4.56}$$

With the integrals 1., 2. and 3., the variation of the entropy density $n(\alpha)s(\alpha,t)$ in respect

to time is derived:

$$\begin{aligned}
T \frac{\partial(ns)}{\partial t} &= \frac{\partial}{\partial \boldsymbol{\alpha}} \int \underbrace{f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}}} \cdot \mathbf{L}_s \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} + V(\boldsymbol{\alpha}) - \mu^{eq} \right)}_{-\mathbf{J}_s} d\mathbf{L}_s \\
&\quad - \int \mathbf{J}_s^{\mathbf{L}} \cdot \frac{\partial}{\partial \mathbf{L}_s} \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1}}{2} \right) d\mathbf{L}_s \\
&\quad + \int f \frac{\partial}{\partial \boldsymbol{\alpha}} \left(\overline{\overline{\Lambda \overline{\overline{T}}^{-1}}} \right) \cdot \mathbf{L}_s d\mathbf{L}_s + \int \left[f \overline{\overline{\Lambda \overline{\overline{T}}^{-1}}} \cdot \mathbf{L}_s \right] \cdot \frac{1}{2} \frac{\partial}{\partial \boldsymbol{\alpha}} \left(\mathbf{L}_s \mathbf{L}_s : \overline{\overline{T}}^{-1} \right) d\mathbf{L}_s \\
&\quad + n \mathbf{N}(\boldsymbol{\alpha}) \cdot \left(\overline{\overline{T}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) + n \left(\overline{\overline{\Lambda \overline{\overline{T}}^{-1}}} \cdot \langle \mathbf{L}_s \rangle \right) \cdot \frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} \tag{4.57}
\end{aligned}$$

It has to be remembered that the derivatives in respect to $\boldsymbol{\alpha}$ were made while \mathbf{L}_s is kept constant, and the equations derived are written for the fix-spaced reference frame. The properties of the distribution function f and of the velocity flux $\mathbf{J}_s^{\mathbf{L}}$ ($\lim_{\mathbf{L}_s \rightarrow \pm\infty} f = 0$; $\lim_{\mathbf{L}_s \rightarrow \pm\infty} \mathbf{J}_s^{\mathbf{L}} = 0$) were also used.

Axial symmetry For the particular case of axial symmetric bodies, all pertinent quantities are independent of ψ . In this case, Condiff et al. [1, 2] prove that $\boldsymbol{\alpha}$ can be replaced with \mathbf{e} , $\dot{\boldsymbol{\alpha}}$ with $\dot{\mathbf{e}} = \boldsymbol{\omega} \times \mathbf{e}$, $\frac{\partial}{\partial \boldsymbol{\alpha}}$ with $\frac{\partial}{\partial \mathbf{e}} = \boldsymbol{\theta} \frac{\partial}{\partial \boldsymbol{\theta}} + \boldsymbol{\phi} \frac{1}{\sin \theta} \frac{\partial}{\partial \boldsymbol{\phi}}$ and $\overline{\overline{\Lambda}}$ with $-\mathbf{e} \times \overline{\overline{U}}$. Here \mathbf{e} is the axial unit vector and $\overline{\overline{U}}$ is the dyadic unit.

In this situation, the entropy production σ takes a much simpler form, meaning the last two lines of equation (4.57) reduce to zero. To prove it, we will make use of the axial symmetry of the body and write the inertial tensor $\overline{\overline{T}}$ as: $\overline{\overline{T}} = I_1(\overline{\overline{U}} - \mathbf{e}\mathbf{e}) + I_3\mathbf{e}\mathbf{e}$. Each term of the last two lines of the equation (4.57) is recalculated:

•

$$\begin{aligned}
\int f \frac{\partial}{\partial \boldsymbol{\alpha}} \left(\overline{\overline{\Lambda \overline{\overline{T}}^{-1}}} \right) \cdot \mathbf{L}_s d\mathbf{L}_s &= \int f \left[\frac{\partial}{\partial \mathbf{e}} \cdot \left\{ -\mathbf{e} \times \left[I_1^{-1}(\overline{\overline{U}} - \mathbf{e}\mathbf{e}) + I_3^{-1}\mathbf{e}\mathbf{e} \right] \right\} \right] \cdot \mathbf{L}_s d\mathbf{L}_s \\
&= -I_1^{-1} \int f \left[\frac{\partial}{\partial \mathbf{e}} \cdot \left(\mathbf{e} \times \overline{\overline{U}} \right) \right] \cdot \mathbf{L}_s d\mathbf{L}_s = 0 \tag{4.58}
\end{aligned}$$

•

$$\begin{aligned}
\int \left[f \bar{\bar{\Lambda}} \bar{\bar{I}}^{-1} \cdot \mathbf{L}_s \right] \cdot \frac{1}{2} \frac{\partial}{\partial \boldsymbol{\alpha}} \left(\mathbf{L}_s \mathbf{L}_s : \bar{\bar{I}}^{-1} \right) d\mathbf{L}_s &= -I_1^{-1} \int (\mathbf{e} \times \mathbf{L}_s) \cdot \frac{1}{2} \frac{\partial}{\partial \mathbf{e}} \left(\sum_{i,j} L_i L_j (\bar{\bar{I}}^{-1})_{ji} \right) d\mathbf{L}_s \\
&= -I_1^{-1} (I_3^{-1} - I_1^{-1}) \int (\mathbf{e} \times \mathbf{L}_s) \cdot [\mathbf{L}_s \cdot (\mathbf{L}_s \cdot \mathbf{e})] d\mathbf{L}_s = 0
\end{aligned} \tag{4.59}$$

where

$$\left[\frac{\partial}{\partial \mathbf{e}} \sum_{i,j} L_i L_j (\bar{\bar{I}}^{-1})_{ji} \right]_k = (I_3^{-1} - I_1^{-1}) \sum_{i,j} L_i L_j \left(\frac{\partial}{\partial \mathbf{e}} \right)_k (e_j e_i) \tag{4.60}$$

$$= (I_3^{-1} - I_1^{-1}) \sum_{i,j} L_i L_j (e_j \delta_{ik} + e_i \delta_{jk}) = 2(I_3^{-1} - I_1^{-1}) [\mathbf{L}_s \cdot (\mathbf{L}_s \cdot \mathbf{e})]_k \tag{4.61}$$

•

$$\begin{aligned}
\left(\bar{\bar{\Lambda}} \bar{\bar{I}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) \cdot \frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} &= \sum_{i,j} \left(\bar{\bar{\Lambda}} \bar{\bar{I}}^{-1} \right)_{ij} \langle L \rangle_j \left(\frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} \right)_i \\
&= - \sum_{i,j,k} \bar{\bar{\Lambda}}_{ki} \bar{\bar{I}}_{kj}^{-1} \langle L \rangle_j \left(\frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} \right)_i = - \left(\bar{\bar{\Lambda}} \cdot \frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} \right) \cdot \left(\bar{\bar{I}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) \\
&= \left(\mathbf{e} \times \frac{\partial V(\boldsymbol{\alpha})}{\partial \boldsymbol{\alpha}} \right) \cdot \left(\bar{\bar{I}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right) = -\mathbf{N}(\boldsymbol{\alpha}) \cdot \left(\bar{\bar{I}}^{-1} \cdot \langle \mathbf{L}_s \rangle \right)
\end{aligned} \tag{4.62}$$

With these in mind, the entropy flux \mathbf{J}^s and the entropy production $\sigma(\boldsymbol{\alpha})$ of a system of identical axial symmetric particles submitted to thermal noise are:

$$\begin{cases} \mathbf{J}^s = \mathbf{e} \times \int f \bar{\bar{I}}^{-1} \cdot \mathbf{L}_s \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\bar{I}}^{-1}}{2} + V(\boldsymbol{\alpha}) - \mu^{eq} \right) d\mathbf{L}_s \\ T\sigma = - \int \mathbf{J}_s^L \cdot \frac{\partial}{\partial \mathbf{L}_s} \left(\frac{k_B T}{v} \ln f + \frac{\mathbf{L}_s \mathbf{L}_s : \bar{\bar{I}}^{-1}}{2} \right) d\mathbf{L}_s \end{cases} \tag{4.63}$$

4.A.3 The Fokker-Planck equation in the magnetisation-fixed reference frame

In the magnetisation-fixed frame, the conservation law for the number of particles $f(\boldsymbol{\alpha}, \mathbf{L}_b)$ writes as [1]:

$$\frac{\partial f}{\partial t} = - \left(\frac{\partial f \dot{\boldsymbol{\alpha}}}{\partial \boldsymbol{\alpha}} \right)_{\mathbf{L}_b} - (\mathbf{N}_b + \dot{\mathbf{L}}_b) \cdot \frac{\partial f}{\partial \mathbf{L}_b} - \frac{\partial \mathbf{J}_b^{\mathbf{L}}}{\partial \mathbf{L}_b} \quad (4.64)$$

where the subscript b stands for the magnetisation-fixed frame and $\dot{\mathbf{L}}_b$ is a notation for

$$\dot{\mathbf{L}}_b = \left(\frac{1}{I_3} - \frac{1}{I_2} \right) L_{b3} L_{b2} \mathbf{i}' + \left(\frac{1}{I_1} - \frac{1}{I_3} \right) L_{b1} L_{b3} \mathbf{j}' + \left(\frac{1}{I_2} - \frac{1}{I_1} \right) L_{b2} L_{b1} \mathbf{e} \quad (4.65)$$

As we treat the case of axial symmetric particles, the inertial tensor and the damping tensor take the form: $\bar{\mathbf{I}} = I_1(\bar{\mathbf{U}} - \mathbf{e}\mathbf{e}) + I_3\mathbf{e}\mathbf{e}$, respectively $\bar{\boldsymbol{\beta}} = \beta_1(\bar{\mathbf{U}} - \mathbf{e}\mathbf{e}) + \beta_3\mathbf{e}\mathbf{e}$. Also, $\boldsymbol{\alpha}$ can be replaced with \mathbf{e} , $\dot{\boldsymbol{\alpha}}$ with $\dot{\mathbf{e}} = \boldsymbol{\omega} \times \mathbf{e}$, $\frac{\partial}{\partial \boldsymbol{\alpha}}$ with $\frac{\partial}{\partial \mathbf{e}} = \boldsymbol{\theta} \frac{\partial}{\partial \boldsymbol{\theta}} + \boldsymbol{\phi} \frac{1}{\sin \boldsymbol{\theta}} \frac{\partial}{\partial \boldsymbol{\phi}}$ and $\bar{\mathbf{L}}$ with $-\mathbf{e} \times \bar{\mathbf{U}}$, where \mathbf{e} is the axial unit vector and $\bar{\mathbf{U}}$ is the dyadic unit [1, 2].

For this matter, the conservation law (4.64) is rewritten as:

$$\frac{\partial f}{\partial t} = \left(\frac{\partial}{\partial \mathbf{e}} \right)_{\mathbf{L}_b} \cdot \left(f \mathbf{e} \times \bar{\mathbf{I}}_b^{-1} \cdot \mathbf{L}_b \right) - \left[\mathbf{N}_b + L_{b3} \left(\frac{1}{I_1} - \frac{1}{I_3} \right) \mathbf{e} \times \mathbf{L}_b \right] \cdot \frac{\partial f}{\partial \mathbf{L}_b} - \frac{\partial \mathbf{J}_b^{\mathbf{L}}}{\partial \mathbf{L}_b} \quad (4.66)$$

With the expression (4.13) for the flux $\mathbf{J}_b^{\mathbf{L}}$, the conservation law takes the form of a Fokker-Planck equation written for the magnetisation-fixed reference frame:

$$\begin{aligned} \frac{\partial f}{\partial t} = & \left(\frac{\partial}{\partial \mathbf{e}} \right)_{\mathbf{L}_b} \cdot \left(f \mathbf{e} \times \bar{\mathbf{I}}_b^{-1} \cdot \mathbf{L}_b \right) \\ & + \frac{\partial}{\partial \mathbf{L}_b} \cdot \left\{ f \underbrace{\left[\bar{\boldsymbol{\beta}}_b - L_{b3} \left(\frac{1}{I_1} - \frac{1}{I_3} \right) \mathbf{e} \times \bar{\mathbf{U}} \right]}_{\bar{\boldsymbol{\beta}}^{ot}} \cdot \mathbf{L}_b - f \mathbf{N}_b + \frac{k_B T}{v} \bar{\boldsymbol{\beta}}_b \bar{\mathbf{I}}_b \cdot \frac{\partial f}{\partial \mathbf{L}_b} \right\} \end{aligned} \quad (4.67)$$

A Particular case: $\beta_3 = 0$ and $L_{b3} = \frac{M_s}{\gamma}$

When the axial component of the angular momentum L_{b3} is constant ($\beta_3 = 0$), with $L_{b3} = \frac{M_s}{\gamma}$ the time dependent tensor $\bar{\boldsymbol{\beta}}^{ot}$ transforms into a damping tensor with constant

coefficients $\overline{\beta}_b^*$:

$$\overline{\beta}_b^* = \overline{\beta}_b - \frac{M_s}{\gamma} \left(\frac{1}{I_1} - \frac{1}{I_3} \right) \mathbf{e} \times \overline{\mathbf{U}} = \beta_1 \begin{pmatrix} 1 & 1/\alpha^* & 0 \\ -1/\alpha^* & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} = \frac{\overline{\beta}_b}{\alpha^*} \begin{pmatrix} \alpha^* & 1 & 0 \\ -1 & \alpha^* & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad (4.68)$$

where

$$\alpha^* = \gamma^* \eta M_s = \frac{\gamma \beta_1 I_1}{M_s (1 - I_1/I_3)} \quad (4.69)$$

4.B Derivations within the inertial regime

4.B.1 The generalised Gilbert equation

For time scales smaller than $\tau = \beta_1^{-1}$, the inertial terms $\dot{\Omega}_1$ and $\dot{\Omega}_2$ from the equation (4.18) cannot be neglected anymore and the whole equation is taken into account. Supposing the axial angular momentum is conserved with $\langle L_3 \rangle = I_3 \Omega_3 = \frac{M_s}{\gamma}$ ($\beta_3 = 0$), the equation (4.18) is rewritten as:

$$\begin{cases} \dot{\Omega}_1 = -\beta_1 \Omega_1 + \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_2 - \frac{1}{I_1} M_s H_2 \\ \dot{\Omega}_2 = -\beta_1 \Omega_2 - \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_1 + \frac{1}{I_1} M_s H_1 \\ I_3 \Omega_3 = \frac{M_s}{\gamma} \end{cases} \quad (4.70)$$

or more concise:

$$\left(\frac{d\Omega_1}{dt} \mathbf{i}' + \frac{d\Omega_2}{dt} \mathbf{j}' \right) - \frac{M_s}{I_1 \gamma} (1-r) \mathbf{e} \times \boldsymbol{\Omega} = -\beta_1 (\Omega_1 \mathbf{i}' + \Omega_2 \mathbf{j}') + \frac{M_s}{I_1} \mathbf{e} \times \left(\mathbf{H} - \frac{1}{n M_s} \frac{\partial \overline{\mathbf{P}}}{\partial \mathbf{e}} \right) \quad (4.71)$$

where $r = \frac{I_1}{I_3}$.

In order to translate the angular velocity $\boldsymbol{\Omega}$ into an expression depending on $\mathbf{M} = M_s \mathbf{e}$, some of its properties will be used:

(a).

$$\frac{d\mathbf{M}}{dt} = \boldsymbol{\Omega} \times \mathbf{M} \quad \Rightarrow \quad \boldsymbol{\Omega} = \frac{\mathbf{M}}{M_s^2} \times \frac{d\mathbf{M}}{dt} + \frac{\mathbf{M}}{\gamma I_3} \quad (4.72)$$

(b).

$$\begin{aligned}\frac{d\boldsymbol{\Omega}}{dt} &= \frac{d\Omega_1}{dt} \mathbf{i}' + \frac{d\Omega_2}{dt} \mathbf{j}' + \frac{d\Omega_3}{dt} \mathbf{e} + \boldsymbol{\Omega} \times (\Omega_1 \mathbf{i}' + \Omega_2 \mathbf{j}' + \Omega_3 \mathbf{e}) \stackrel{\Omega_3 \equiv \text{cst.}}{=} \frac{d\Omega_1}{dt} \mathbf{i}' + \frac{d\Omega_2}{dt} \mathbf{j}' \\ \Rightarrow \frac{d\boldsymbol{\Omega}}{dt} &= \frac{d\Omega_1}{dt} \mathbf{i}' + \frac{d\Omega_2}{dt} \mathbf{j}' = \frac{\mathbf{M}}{M_s^2} \times \frac{d^2 \mathbf{M}}{dt^2} + \frac{1}{\gamma I_3} \frac{d\mathbf{M}}{dt}\end{aligned}\quad (4.73)$$

With these properties, equation (4.71) takes a more compact form:

$$\frac{1}{\gamma} \frac{d\mathbf{M}}{dt} = \mathbf{M} \times \left(\mathbf{H} - \frac{1}{n} \frac{\partial}{\partial \mathbf{M}} \overline{\overline{P}} \right) - \frac{\beta_1 I_1}{M_s^2} \mathbf{M} \times \left(\frac{d\mathbf{M}}{dt} + \frac{1}{\beta_1} \frac{d^2 \mathbf{M}}{dt^2} \right) \quad (4.74)$$

The generalised Gilbert equation is retrieved:

$$\boxed{\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \left[\mathbf{H} - \frac{1}{n M_s} \frac{\partial}{\partial \mathbf{e}} \overline{\overline{P}} - \eta \left(\frac{d\mathbf{M}}{dt} + \tau \frac{d^2 \mathbf{M}}{dt^2} \right) \right]} \quad (4.75)$$

where

$$\eta = \frac{\beta_1 I_1}{M_s^2} \quad \tau = \beta_1^{-1} = f \alpha \quad f = \frac{M_s}{\gamma I_1} \quad \alpha = \gamma \eta M_s \quad (4.76)$$

4.B.2 Towards the numerical solution of the differential system of equations

Finding the magnetisation dynamics is equivalent to finding the temporal dependence of the angles defining the orientation of the magnetisation: $\phi(t)$, $\theta(t)$, $\psi(t)$. The magnetisation orientation is given by the system of equations (4.18) with $\beta_3 = 0$ and $L_3 = \frac{M_s}{\gamma}$, the equivalent formulation of the generalised Gilbert equation (4.36):

$$\begin{cases} \dot{\Omega}_1 = -\beta_1 \Omega_1 + \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_2 - \frac{1}{I_1} M_s H_2 \\ \dot{\Omega}_2 = -\beta_1 \Omega_2 - \Omega_3 \left(1 - \frac{I_3}{I_1}\right) \Omega_1 + \frac{1}{I_1} M_s H_1 \\ I_3 \Omega_3 = \frac{M_s}{\gamma} \end{cases} \quad (4.77)$$

where

$$\begin{cases} \Omega_1 = \dot{\phi} \sin \theta \sin \psi + \dot{\theta} \cos \psi \\ \Omega_2 = \dot{\phi} \sin \theta \cos \psi - \dot{\theta} \sin \psi \\ \Omega_3 = \dot{\psi} + \dot{\phi} \cos \theta \end{cases} \quad (4.78)$$

As the angular velocity vector $\mathbf{\Omega}$ has simpler components in the $\xi'\eta'\zeta'$ magnetisation frame, the system of equations (4.77) is rewritten for this frame.

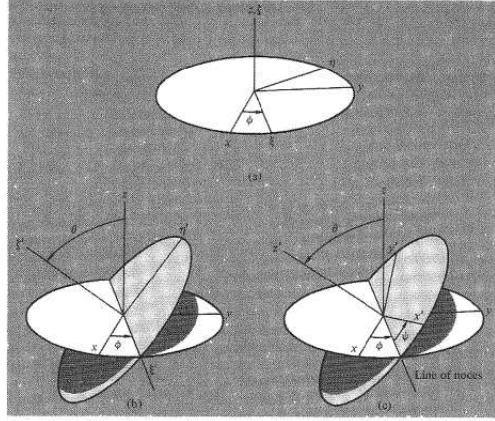


Figure 4.6: Dynamics of the magnetisation as a superposition of three elementary rotations: the rotation around the fixed axis Oz with $\dot{\phi}$ (see a), the rotation around the line of nodes $O\xi'$ with $\dot{\theta}$ (see b) and the rotation around the mobile axis Oz' with $\dot{\psi}$ (see c). The magnetisation vector \mathbf{M} is parallel with the axis Oz' or the unit vector \mathbf{e} .

$$\overline{\overline{R}}_{x'y'z' \rightarrow \xi'\eta'\zeta'} = \begin{pmatrix} \cos \psi & -\sin \psi & 0 \\ \sin \psi & \cos \psi & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad \overline{\overline{R}}_{xyz \rightarrow \xi'\eta'\zeta'} = \begin{pmatrix} \cos \phi & \sin \phi & 0 \\ -\sin \phi \cos \theta & \cos \phi \cos \theta & \sin \theta \\ \sin \phi \sin \theta & -\cos \phi \sin \theta & \cos \theta \end{pmatrix} \quad (4.79)$$

Knowing that the rotation matrix mapping a vector from the $Ox'y'z'$ frame to the $O\xi'\eta'\zeta'$ is $\overline{\overline{R}}_{x'y'z' \rightarrow \xi'\eta'\zeta'}$ and that the rotation matrix mapping a vector from the $Oxyz$ frame to the $O\xi'\eta'\zeta'$ frame is $\overline{\overline{R}}_{xyz \rightarrow \xi'\eta'\zeta'}$, the components of the angular velocity and that of the

magnetic field \mathbf{H} in the $O\xi'\eta'\zeta'$ are:

$$\begin{cases} \Omega_{\xi'} = \dot{\theta} = \Omega_1 \cos \psi - \Omega_2 \sin \psi \\ \Omega_{\eta'} = \dot{\phi} \sin \theta = \Omega_1 \sin \psi + \Omega_2 \cos \psi \\ \Omega_{\zeta'} = \Omega_3 = \dot{\psi} + \dot{\phi} \cos \theta = \frac{M_s}{I_3 \gamma} \end{cases} \quad (4.80)$$

$$\Rightarrow \begin{cases} \dot{\Omega}_{\xi'} = \dot{\Omega}_1 \cos \psi - \dot{\Omega}_2 \sin \psi - \Omega_{\eta'} \dot{\psi} \\ \dot{\Omega}_{\eta'} = \dot{\Omega}_1 \sin \psi + \dot{\Omega}_2 \cos \psi + \Omega_{\zeta'} \dot{\psi} \\ \dot{\Omega}_{\zeta'} = 0 \end{cases} \quad (4.81)$$

$$\begin{cases} H_{\xi'} = H \sin \phi \sin \beta \\ H_{\eta'} = H \cos \phi \cos \theta \sin \beta + H \sin \theta \cos \beta \\ H_{\zeta'} = -H \cos \phi \sin \theta \sin \beta + H \cos \theta \cos \beta \end{cases} \quad (4.82)$$

Here the field \mathbf{H} forms an angle β with the Oz axis in the yOz plane, meaning $H_x = 0$, $H_y = H \sin \beta$ and $H_z = H \cos \beta$.

The simplest way to transform the system is to multiply the first and the second equation of the system (4.77) by $\sin \psi$, respectively $\cos \psi$ and then to add them. In this way, the second equation of the system (4.83) is obtained. In a similar way is proceeded in order to obtain the first equation of the system (4.83), only that now the first equation of (4.77) is multiplied by $\cos \psi$ and the second by $-\sin \psi$. The transformed system is then:

$$\begin{cases} r\dot{\Omega}_{\xi'} - \Omega_{\eta'} (-\Omega_3 + r\dot{\phi} \cos \theta) = -\gamma\Omega_3 H_{\eta'} - \gamma\eta M_s \Omega_3 \Omega_{\xi'} \\ r\dot{\Omega}_{\eta'} + \Omega_{\xi'} (-\Omega_3 + r\dot{\phi} \cos \theta) = \gamma\Omega_3 H_{\xi'} - \gamma\eta M_s \Omega_3 \Omega_{\eta'} \\ \Omega_3 = \dot{\psi} + \dot{\phi} \cos \theta = \frac{M_s}{\gamma I_3} = \text{const.} \end{cases} \quad (4.83)$$

where

$$r = \frac{I_1}{I_3} \quad f = \frac{I_3 \Omega_3}{I_1} = \frac{M_s}{\gamma I_1} \quad \alpha = \gamma\eta M_s. \quad (4.84)$$

With (4.81), the system (4.83) changes to a second order differential system of variables θ , ϕ , ψ which gives the orientation of the magnetisation \mathbf{M} :

$$\begin{cases} \ddot{\phi} \sin \theta = -f\alpha \dot{\phi} \sin \theta - 2 \cos \theta \dot{\phi} \dot{\theta} + f\dot{\theta} + f\gamma H_{\xi'} \\ \ddot{\theta} = -f\alpha \dot{\theta} - f\dot{\phi} \sin \theta + \sin \theta \cos \theta \dot{\phi}^2 - f\gamma H_{\eta'} \\ \dot{\psi} = \Omega_3 - \dot{\phi} \cos \theta \end{cases} \quad (4.85)$$

Bibliography

- [1] D. W. CONDIFF and J. S. DAHLER, *J. Chem. Phys.* **44**, 3988 (1966).
- [2] J. S. DAHLER and N. F. SATHER, *J. Chem. Phys.* **38**, 2363 (1963).
- [3] S. R. DEGROOT and P. MAZUR, *Non-Equilibrium Thermodynamics*, Dover Publications Inc. New York, second edition, 1984.
- [4] H. GOLDSTEIN, C. POOLE, and J. SAFKO, *Classical mechanics*, Addison Wesley, third edition, 2000.
- [5] T. L. GILBERT, *IEEE Transactions on Magnetics* **40**, 3443 (2004).
- [6] W. F. BROWN JR., *Physical Review* **130**, 1677 (1963).
- [7] S. CHIKAZUMI, *Physics of Ferromagnetism*, Oxford Univ. Press, 1997.

Chapter 5

Perspectives: A two rotating fluid model for spin-transfer

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Introduction

The purpose of the previous chapters was to provide a classic model for the macrospin dynamics. Not only the dynamic Gilbert and the stochastic Brown equations were derived, but even additional terms (new behaviour) were predicted for time scales smaller than a characteristic nutation time τ . The role of this chapter is to question how this kind of treatment could be extended for more complex cases, in order to extract more information. Ideas for a first draft thermodynamic model attempting to explain some of the features of the spin-transfer problem [1–5] are presented.

It is well known that a metallic ferromagnet influences the electronic transport properties of the system. The spins of the conducting electrons rotate to follow the local magnetisation. The magnetisation of the ferromagnet changes the flow of spin-angular momentum by exerting a torque on the flowing spins to reorient them, and therefore the flowing spins are expected to exert an equal and opposite torque on the ferromagnet, in order to conserve the total energy and angular momentum of the system. The torque applied by the conduction electron spins on the ferromagnet is what is called *spin transfer torque*. The transfer is microscopic in nature, and is a result of the cumulative effect of the spins.

Two types of magnetic particles are involved in the phenomenon: the spins of the conduction electrons and the macrospin (the uniformed magnetised particle). The spins of the conduction electrons are microscopic and have a response time (dynamics) faster than or of the order of several tens of picoseconds. The macrospin is macroscopic and has a response time usually greater than 100 picoseconds. The time scale separation between these two dynamics allows the physicist to generally treat the two magnetic 'species' separately: when treating the conduction electron spins dynamics the macrospin dynamics is assumed stationary in time, while when treating the macrospin, the conduction electron spins is assumed instantaneous.

The time scale separation between the dynamics of two species that influence one another is an old problem in physics. For example, a fully ionised plasma has potentially two fluids, an electron fluid and an ion fluid occupying the same space, so there will be at least three time scales and length scales to consider. The three collisional relaxation times of importance are those for establishing equilibrium velocity distributions in: (1) the electron gas alone (τ_1), (2) the ion gas alone (τ_2), and (3) the electron-ion composite gas (τ_ϵ). The electron and ion fluids comprising the plasma may also be at different temperatures, but when the temperatures T_1 and T_2 are of comparable magnitude it is approximately the case that there are at least two orders of magnitude separating these times [6].

$$\tau_2 : \tau_1 : \tau_\epsilon = 1 : (m_1/m_2)^{1/2} : (m_1/m_2) \quad \text{that is} \quad \tau_1 \ll \tau_2 \ll \tau_\epsilon \quad (5.1)$$

Due to the large difference of mass $m_1/m_2 \ll 1$, when an ion collides an electron, its momentum undergoes only a relatively small change, while the electron's momentum changes its direction greatly in a collision, but its magnitude only slightly. Hence, the momentum exchange between particles of different components is very small compared to the momentum exchange between particles of the same specie. *Such processes are too rapid for the two species to be in mutual equilibrium.* Each fluid can persist in a stationary state due to the collisions within it which influences its momentum much more than the collisions with the other fluid: in a manner of speaking, the behaviour of one fluid is somehow independent of the other.

A similar case is the one of the liquid Helium II, of a 'normal' fluid at a temperature $T < 2.17K$ and a superfluid at a temperature $T = 0K$, with the important point of difference that mass exchange is possible between the two Helium fluids, but of course impossible for the electric fluids of the plasma: the ions and the electrons.

The idea In a similar way, the spin transfer problem could be approached by treating the conduction s -electron spins and the d -electron spins of the macrospin as two rotating fluids characterised by a large separation of time scales, which can exchange particles, angular momentum, energy, and possibly, having different temperatures. A continuous orientation model could be adopted for the spins of the electrons, and not a discrete one, as spin-up, spin-down. Nonequilibrium thermodynamics is particularly useful with systems like these, in which coupling between the fluids is important, and the model accounting for it is called the *two fluid model* [7, 8].

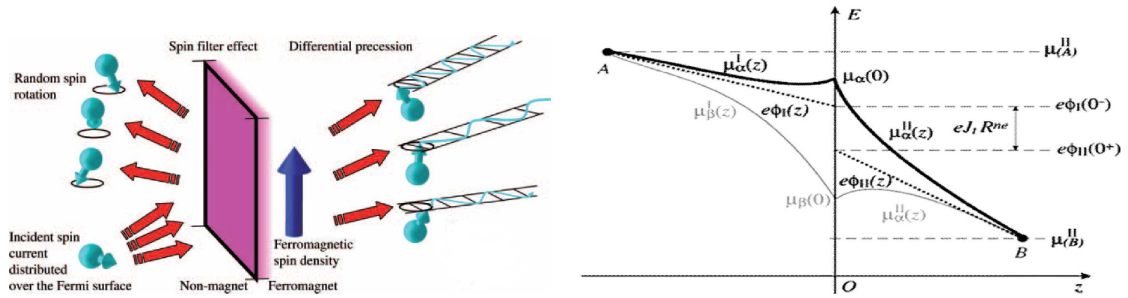
With this idea, this chapter is an attempt in building a two-fluid thermodynamic model for the spin-transfer problem. The attention will be focused on the rotation of the two fluids: the s -electron spins and the macrospin. We restrict in a first-model draft on the study of the cumulative rotational effect of the spins on the macrospin.

5.1 A two rotating fluid model for the spin transfer problem?!

It is known [1, 3] that the electron spins precess as they penetrate the ferromagnet. Very near¹ the normal-ferromagnet interface, at a fixed position in the ferromagnet, the overall conduction electron spins precession and reversal is not zero. There, the spins of the

¹Electron spins precess as they penetrate the ferromagnet. Electrons on different parts of the Fermi surface precess at different rates (in free-electron models, the majority and minority electrons have associated different spherical Fermi surfaces). So, while the dephasing was not complete just after transmission into the ferromagnet, the differential (overall) precession increases the cancellation the greater the distance into the ferromagnet. For transition-metal interfaces it (the transverse spin current) is reduced to 10% of its incident value after a distance of roughly 1 nm [3, 9].

conduction electrons do not precess and reverse randomly as they do far away from the interface in the ferromagnet, leading to an average velocity of rotation (precession and reversal) for the spins different from zero. It is a phenomenon which generally appears under the name of *spin-accumulation*², and is either *transverse*, when speaking about precession, either *longitudinal*, when speaking about reversal (spin-flip) (see Figure 5.1).



(a) Electrons incident from the non-magnet with three different incident directions, have the same spin state, transverse to the ferromagnetic spin density (blue arrow). The reflected electron spins have random spin rotation, leading to no overall precession. The transmitted electrons precess as they go into the ferromagnet. While the transmitted electrons are not immediately dephased as the reflected electrons, the overall precession cancels out the distance 'travelled' in the ferromagnet. The overall precession or the commonly named, differential precession, is a consequence of the *transverse spin accumulation*. (Figure taken from the review article on "Spin transfer torque and dynamics" by M. D. Stiles and J. Miltat [3]).

(b) Profiles for two chemical potentials μ_α and μ_γ defined for two discrete conduction electrons spin populations $\alpha(\uparrow)$ and $\gamma(\downarrow)$ at the normal-ferromagnet interface. The inequality between the two chemical potentials at the interface leads to the appearance of the *longitudinal spin accumulation*. A stationary velocity of reaction $\dot{\psi} = L(\mu_\alpha - \mu_\gamma)$, describes the relaxation from one channel to the other. Hence, an overall reversal of the spins different from zero is present at the interface. (Figure taken from "Spin transfer in an open ferromagnetic layer: from negative damping to effective temperature" by J-E Wegrowe, M C Ciornei and H-J Drouhin [11]).

Figure 5.1: Figures representing two types of spin-accumulation: transverse and longitudinal. The average velocity of rotation of the conduction electrons spins at the normal-ferromagnet interface, precession and reversal (spin-flip) is different from zero.

The model Close to the interface, the conduction *s*-electron spins could be treated classically as a rotating fluid with an average velocity of rotation different from zero. A second

²"The phenomenon of this accumulation can be understood to be similar to charge accumulation. When a current flows in an inhomogeneous conducting medium, continuity of the current requires a larger effective electric field in the regions where the conductivity is smaller. This leads to charge accumulation at the interface between regions of high and low conductivity. Similarly, spin accumulation originates from the spatially varying magnetisation that induces variations in the spin polarisation of the current." (P. M. Levy [10])

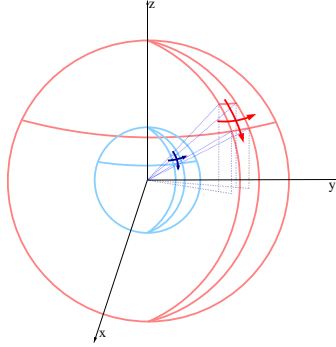


Figure 5.2: The tips of the macrospins move on the surface of an M_s radius (light red sphere), while the tips of the s -electrons spins move on the surface of an s_m radius (light blue sphere). The arrows indicate two components corresponding to precession and reversal for the average velocities of the spins $\dot{\mathbf{S}}$ and the macrospins $\dot{\mathbf{M}}$, or of their corresponding fluxes $n_1\dot{\mathbf{S}}$, respectively $n_2\dot{\mathbf{M}}$.

fluid could be represented either by the microscopic d -electron spins, either by a statistical ensemble of macrospins of much slower dynamics than that of the s -electrons spins. The model would be that of two rotating fluids influencing one another through the exchange of angular momentum. In a first model draft, we will concentrate on the effect of the rotating s -electron spins on the macrospin (d -electron spins).

To describe this situation in a thermodynamic framework the compound fluid could be treated as being a mixture of distinct fluids, having different average fluid velocities $\dot{\mathbf{S}}$, respectively $\dot{\mathbf{M}}$, and, in some situations, involving very short time scales, even having different temperatures T_1 , respectively T_2 . Each fluid would be represented by a distribution function $f_i(\mathbf{u}, \mathbf{e})$ representing the number of magnetic particles having the velocity within the $(\mathbf{u}, d\mathbf{u})$ and orientation within $(\mathbf{e}, d\mathbf{e})$, which could persist as stationary functions close to Maxwellian distributions centered around their average velocities $\dot{\mathbf{S}}$ and $\dot{\mathbf{M}}$ (not as transitory functions towards distributions centered around a common velocity - the processes are too rapid for the two species to be in mutual equilibrium). The number of particles with orientation within $(\mathbf{e}, d\mathbf{e})$ would be represented by the density of particles $n_i(\mathbf{e})$, where $n_i(\mathbf{e}) = \int f_i(\mathbf{u}, \mathbf{e}) d\mathbf{u}$. The average velocity of the spins $\mathbf{s} = s_m \mathbf{e}$ with the orientation within $(\mathbf{e}, d\mathbf{e})$ would be defined as $n_1 \dot{\mathbf{S}} = \int f_1(\mathbf{u}, \mathbf{e}) \mathbf{u} d\mathbf{u}$, while the average velocity for the magnetisations $\mathbf{m} = M_s \mathbf{e}$ would be defined as $n_2 \dot{\mathbf{M}} = \int f_2(\mathbf{u}, \mathbf{e}) \mathbf{u} d\mathbf{u}$.

The velocity \mathbf{u} of one spin would be either changed by an applied magnetic field dependent on the uniform magnetisations \mathbf{m} , $\mathbf{H}_1(\mathbf{m}) = -\frac{1}{s_m} \frac{V_1(\mathbf{e})}{\partial \mathbf{e}}$, either by a transfer of (angular) momenta to other spins, or by a transfer of (angular) momenta to the macrospins \mathbf{m} . The velocity \mathbf{u} of one macrospin would be either changed by an applied magnetic field

$\mathbf{H}_2 = -\frac{1}{M_s} \frac{V_2(\mathbf{e})}{\partial \mathbf{e}}$, either by a transfer of (angular) momentum to the spins. When two magnetic particles of the same kind i transfer (angular) momentum, the (angular) momentum and energy of the fluid i would be conserved, while when s -electron spins and macrospins transfer angular momentum, the (angular) momentum and energy of the fluid i (and also j) is changed. The lack of conservation of the (angular) momentum and energy of the fluid i due to the transfers to particles of a different kind leads to the appearance of *dissipation*. It will explicitly appear in the dynamic and energy equation for each fluid. In spite of it, when writing the dynamic and energy equation for the entire system no dissipative terms are expected as the energy and (angular) momentum of one fluid is transferred to the other.

The *main hypothesis* of the model would be that of treating the s -electron spins on the same footing with the macrospin. Meaning, the model will stand on the magnetic inertia hypothesis: masses m_1 and m_2 will be associated to the s -electron spins and the macrospins related to their magnetic dynamics, and not to the displacement of matter. The ideas presented in chapter 3 will be closely followed in a first-draft model. In a second time, we could pass to the introduction of the angular momentum \mathbf{L} as a kinetic degree of freedom, instead of that of the velocity \mathbf{u} .

The 'independent' nature of the two fluids consists in being able to define partial pressures \overline{P}_i , partial densities of entropy s_i and even different temperatures T_i . Each volume element $(\mathbf{e}, d\mathbf{e})$, $(\mathbf{u}, \mathbf{e}, d\mathbf{u}, d\mathbf{e})$ would be described by extensive parameters such as: a) the density of magnetic particles $n_i(\mathbf{e})$, respectively $f_i(\mathbf{u}, \mathbf{e})$; b) the density of entropy $s_i(\mathbf{e})$; c) kinetic and potential energy densities $\int f_i m_i \mathbf{u}^2 / 2, d\mathbf{u}$, respectively $\int f V_i(\mathbf{r}) d\mathbf{u} = n_i(\mathbf{e}) V_i(\mathbf{e})$.

Following the two fluid thermodynamic scheme as presented by Woods in his book [12], a dynamic equation is expected for the macrospins accounting for the rotating s -spin fluid.

5.2 Dynamic equations

The dynamic equations shapes to be expected for a two fluid system with equal temperatures $T_1 = T_2 = T$ are the following:

$$\begin{cases} \rho_1 \frac{D_1 \dot{\mathbf{S}}}{Dt} = -Div \overline{\overline{P}}_1^1 + n_1 \mathbf{H}_1(\mathbf{m}) + \frac{1}{\tau_{12}} \frac{\rho_1 \rho_2}{\rho} (\dot{\mathbf{M}} - \dot{\mathbf{S}}) \\ \rho_2 \frac{D_2 \dot{\mathbf{M}}}{Dt} = -Div \overline{\overline{P}}_2^2 + n_2 \mathbf{H}_2 - \frac{1}{\tau_{12}} \frac{\rho_1 \rho_2}{\rho} (\dot{\mathbf{M}} - \dot{\mathbf{S}}) \end{cases} \quad (5.2)$$

where $\rho_1 = m_1 n_1$, $\rho_2 = m_2 n_2$, $\rho = \rho_1 + \rho_2$, the 1, 2 indices refer to systems of reference following the flow of the first, respectively the second fluid and τ_{12} is a relaxation time (a detailed derivation is presented in Appendix 5.A). Due to the transfer of (angular) momenta

('collisions') between the spins and the magnetisations, a dissipative field

$$\mathbf{R}_{12} = -\mathbf{R}_{21} = \frac{1}{\tau_{12}} \frac{\rho_1 \rho_2}{\rho} (\dot{\mathbf{M}} - \dot{\mathbf{S}}) \quad (5.3)$$

appears in each motion equation. As the spin bath moves with the velocity $\dot{\mathbf{S}} - \dot{\mathbf{M}}$ relative to the 'fluid' of magnetisations, it exerts a field $\mathbf{R}_{21} = -\mathbf{R}_{12}$ on the magnetisations and the positive work done, namely $\mathbf{R}_{21} \cdot (\dot{\mathbf{S}} - \dot{\mathbf{M}})$ is dissipated in both fluids. The way the dissipative energy divides between the fluids depends on the ratio of the masses m_1/m_2 . As it is supposed that $m_2 \gg m_1$, then almost all the energy is dissipated in the s -electron spin fluid.

The second equation of the system is precisely the key equation from chapter 3 (3.16) which lead to the generalised Gilbert equation. Here it has an extra term whose presence is a consequence of the rotating bath. When the bath is in equilibrium, meaning, the average precession and reversal of the bath is null, represented by an average velocity $\dot{\mathbf{S}} = 0$, the generalised form of the Gilbert equation as presented in chapter 3 is retrieved. However, when the average velocity of the bath is different from zero an extra term is present as a consequence of the transfer of (angular) momentum between the spins and the macrospins. For this matter, the Gilbert damping coefficient η is identified as $\eta = \frac{m_2}{\tau_{12}} \frac{\rho_1}{\rho}$, and the two equations can be rewritten as:

$$\begin{cases} m_1 \frac{D_1 \dot{\mathbf{S}}}{Dt} = -\frac{1}{n_1} \text{Div} \overline{\overline{P_1^1}} + \mathbf{H}_{1(m)} + \eta \frac{n_2}{n_1} (\dot{\mathbf{M}} - \dot{\mathbf{S}}) \\ m_2 \frac{D_2 \dot{\mathbf{M}}}{Dt} = -\frac{1}{n_2} \text{Div} \overline{\overline{P_2^2}} + \mathbf{H}_2 - \eta (\dot{\mathbf{M}} - \dot{\mathbf{S}}) \end{cases} \quad (5.4)$$

Following the same type of cosmetic treatment as the one presented in chapter 3, the two dynamic equations are expected to lead to the following expressions:

$$\begin{cases} \dot{\mathbf{S}} = g_e \mathbf{S} \times \left[-\frac{1}{n_1} \text{Div} \overline{\overline{P_1^1}} + \mathbf{H}_{1(m)} - \eta \frac{n_2}{n_1} \left(\dot{\mathbf{S}} + \tau_1 \frac{D_1 \dot{\mathbf{S}}}{Dt} - \dot{\mathbf{M}} \right) \right] \\ \dot{\mathbf{M}} = \gamma \mathbf{M} \times \left[-\frac{1}{n_2} \text{Div} \overline{\overline{P_2^2}} + \mathbf{H}_2 - \eta \left(\dot{\mathbf{M}} + \tau_2 \frac{D_2 \dot{\mathbf{M}}}{Dt} - \dot{\mathbf{S}} \right) \right] \end{cases} \quad (5.5)$$

If the hypothesis of associating a mass to the inertia of s -electron spins is true (treating the spins on the same footing with the macrospins), then the first equation of the system implies nutation for the spins. Even if it is unexpected, it has to be remembered that the amplitude of the nutation decreases strongly with the 'spinning' frequency, as showed in the Figure 4.3 for the macrospin nutation from the previous chapter. Also, the duration of the nutation is inversely proportional with the spinning frequency. The faster dynamics

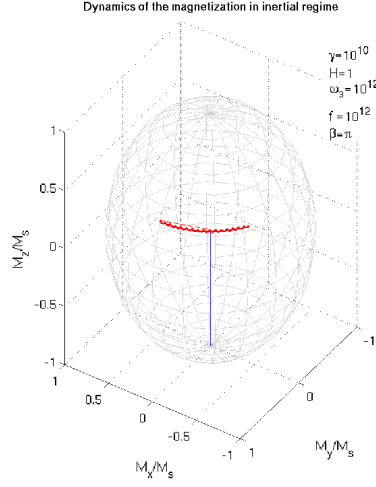


Figure 5.3: Nutation of the macrospin in the absence of damping. It is barely seen for values of the frequency $f = 10^{12} \text{ s}^{-1}$, where $f = \frac{I_3}{I_1} \omega_3$ and ω_3 is the associated spinning frequency of the magnetisation. The faster dynamics of the spins in respect to that of the magnetisation implies faster associated frequencies than those of the magnetisation leading to a barely observable nutation (in terms of its amplitude) for the spins; and even if we could access it experimentally, the measurement would average out over many spins the potential spins nutation.

of the spins (10^{-12} s) in respect to that of the magnetisation (10^{-9} s) of whom nutation is not observed at the accessible measurement times ($t > 10^{-12} \text{ s}$) imply that neither the spins nutation won't be observable; and even if we would have the experimental 'tools' to access it, the measurement would average out over many s -electron spins the potential spin nutation.

Hence, for accessible measuring times ($t > 10^{-12} \text{ s}$), the following type of equations are to be expected (the inertial terms imposing nutation were dropped out):

$$\begin{cases} \frac{d\mathbf{S}}{dt} = g_e^* \mathbf{S} \times \left[-\frac{1}{n_1} \text{Div} \overline{\overline{P_1^1}} + \mathbf{H}_{1(m)} - \eta \frac{n_2}{n_1} \left(\frac{d\mathbf{S}}{dt} - \frac{d\mathbf{M}}{dt} \right) \right] \\ \frac{d\mathbf{M}}{dt} = \gamma^* \mathbf{M} \times \left[-\frac{1}{n_2} \text{Div} \overline{\overline{P_2^2}} + \mathbf{H}_2 - \eta \left(\frac{d\mathbf{M}}{dt} - \frac{d\mathbf{S}}{dt} \right) \right] \end{cases} \quad (5.6)$$

For the case of a dilute 'fluid' of macrospins in a fluid of s -electron spins, meaning when the density of the s -electron spins is much higher than the density of uniform magnetised particles $n_1 \gg n_2$, the role of the macrospins on the spins is expected to be very small: $\eta \frac{n_2}{n_1} \left(\frac{d\mathbf{M}}{dt} - \frac{d\mathbf{S}}{dt} \right) \approx 0$.

Appendix

5.A The two fluid model

It is desired to see the effect of a steady flow of light particles of mass m_1 on the Brownian particles dynamic equation and corresponding Fokker-Planck equation. The Brownian particles of mass m_2 consist in a dilute fluid whom interactions are very rare compared to that of the light fluid. Being interested in time scales t with $\tau_1 < t < \tau_2$, the easiest way is to treat both fluids on the same footing, meaning kinetically (see Table ??). Unless it is wished to remain entirely at a long-time scale phenomenological description, it is necessary to treat the various components as separate fluids and then to approach the long time scales asymptotically. Additive terms are expected in the dynamic and Fokker-Planck equation for the Brownian particles accounting for the cumulative effect of the light fluid.

5.A.1 The model

To each fluid is associated a distribution function $f_i(\mathbf{u}, \mathbf{r})$ representing the number of particles having the velocity in the $(\mathbf{u}, \mathbf{r}, d\mathbf{u}, d\mathbf{r})$ phase space volume element. It is assumed that each distribution function f_i vanishes for infinite velocities $\lim_{\mathbf{u} \rightarrow \pm\infty} f_i(\mathbf{u}, \mathbf{r}) = 0$. The number of particles within $(\mathbf{r}, d\mathbf{r})$ is represented by the density of particles $n_i(\mathbf{r})$, where $n_i(\mathbf{r}) = \int f_i(\mathbf{u}, \mathbf{r}) d\mathbf{u}$. The average velocity \mathbf{v}_i of the particles with the orientation within $(\mathbf{u}, \mathbf{r}, d\mathbf{u}, d\mathbf{r})$ is defined as $n_i(\mathbf{r}) \mathbf{v}_i(\mathbf{r}) = \int f_i(\mathbf{u}, \mathbf{r}) \mathbf{u} d\mathbf{u}$. The velocity \mathbf{u}_i of one particle is either changed by an applied force \mathbf{F}_i , either by the collisions with particles of the same specie i or of the different specie j . The collisions are modelled with a phase space flux $\mathbf{J}_i^{\mathbf{u}}$ accounting for both type of collisions. It is assumed that $\lim_{\mathbf{u} \rightarrow \pm\infty} \mathbf{J}_i^{\mathbf{u}} = 0$.

The 'independent' nature of the two fluids consists in being able to define partial pressures \overline{P}_i , partial densities of entropy s_i and even different temperatures T_i . Each volume element $(\mathbf{r}, d\mathbf{r})$, $(\mathbf{u}, \mathbf{r}, d\mathbf{u}, d\mathbf{r})$ is described by extensive parameters such as: a) the density of magnetic particles $n_i(\mathbf{r})$, respectively $f_i(\mathbf{u}, \mathbf{r})$; b) the density of entropy $s_i(\mathbf{r})$; c) kinetic and potential energy densities $\int f_i m_i \mathbf{u}^2 / 2, d\mathbf{u}$, respectively $\int f V_i(\mathbf{r}) d\mathbf{u} = n_i(\mathbf{r}) V_i(\mathbf{r})$.

With these in mind, we proceed to the derivation of the more general dynamic equation and Fokker-Planck equation for the Brownian particles of mass m_2 accounting for the presence of the other fluid of particles of mass m_1 . The scheme of MNET is used in order to establish simple phenomenological laws accounting for the interactions between the two fluids. The newly introduced phenomenological coefficients would listen to global constraints such as: the conservation of energy and linear momenta. In order to establish the phenomenological laws, the entropy production $\sigma(\mathbf{r})$ has to be determined.

5.A.2 The MNET framework

The derivation of the entropy production σ for the ensemble of the two fluids has two important cornerstones: the Gibbs postulate and the conservation law for the number of particles $f_i(\mathbf{u}, \mathbf{r})$ or otherwise called the Boltzmann equation. With these the entropy production is determined, followed by the establishment of the phenomenological laws. The phenomenological coefficients account for the collisions between fluids in form of dissipative coefficients listening to conservation laws for the energy and momenta of the two fluids ensemble.

A The conservation law and the Gibbs entropy postulate

A.1 The fixed frame

The Gibbs postulate for the fixed frame

The 'independent' nature of the two fluids consists in writing a Gibbs equation for each of the two fluids. It corresponds well to the statistical idea that the distribution functions f_i are close to equilibrium distribution functions f_i^{eq} centered around the average velocities \mathbf{v}_i of each fluid or equivalently, to a small transfer of momenta at the encounter of a heavy Brownian particle m_2 with a light particle m_1 [7]. In the first attempt of writing the model, the mathematical analysis is simplified by considering identical temperatures $T = T_1 = T_2$ for the two fluids. The Gibbs postulate written for each of the two fluids in the fixed frame is:

$$T\rho_i(\mathbf{r})s_i(\mathbf{r}) = -kT \int f_i \ln \frac{f_i}{f_i^{eq}} d\mathbf{u} \quad (5.7)$$

where f_i^{eq} is the local equilibrium distribution, meaning the local Maxwellian with μ_i^{eq} the chemical potential of particles at local equilibrium.

$$f_i^{eq}(\mathbf{u}, \mathbf{r}, t) = \exp\left(\frac{\mu_i^{eq} - m_i u^2/2 - V_i(\mathbf{r})}{k_B T}\right) \quad (5.8)$$

The entropy for the whole system writes then as:

$$T\rho s = - \sum_{i=1,2} \int f_i \left(kT \ln f_i + \frac{m_i u^2}{2} + V_i(\mathbf{r}) - \mu_i^{eq} \right) d\mathbf{u} \quad (5.9)$$

The conservation law for the fixed frame

In the fixed reference frame the conservation equation for the number of particles $f_i(\mathbf{u}, \mathbf{r}, t)$ of coordinate $(\mathbf{r}, d\mathbf{r})$ and velocity $(\mathbf{u}, d\mathbf{u})$ or the Boltzmann equation is written as:

$$\mathcal{D}f_i = \frac{\partial f_i}{\partial t} + \mathbf{u} \cdot \frac{\partial f_i}{\partial \mathbf{r}} + \frac{\mathbf{F}_i}{m_i} \cdot \frac{\partial f_i}{\partial \mathbf{u}} = \left(\frac{\partial f_i}{\partial t} \right)_c = \sum_{j=1,2} C_{ij} \quad (5.10)$$

The quantity $\left(\frac{\partial f_i}{\partial t}\right)_c$ defined above is equal to the rate of change owing to encounters, in the velocity-distribution function f_i at a *fixed* point. C_{ij} is the contribution to the change of f_i due to collisions between particles of i th and j th fluid. This term will be referred as the *collision term*. As a consequence of the conservation of the number of particles at each collision, the sum of the collision terms $\sum_j C_{ij}$ can be written as a divergence of a 'collision flux' \mathbf{J}_i^u (see Appendix 5.B.3 for more details).

$$\mathcal{D}f_i = \left(\frac{\partial f_i}{\partial t}\right)_c = \sum_{j=1,2} C_{ij} = -\frac{\partial \mathbf{J}_i^u}{\partial \mathbf{u}} \quad (5.11)$$

The conservation equation takes then the following shape:

$$\frac{\partial f_i(\mathbf{u}, \mathbf{r}, t)}{\partial t} = -\mathbf{u} \cdot \frac{\partial f_i}{\partial \mathbf{r}} - \frac{\mathbf{F}_i}{m_i} \cdot \frac{\partial f_i}{\partial \mathbf{u}} - \frac{\partial \mathbf{J}_i^u}{\partial \mathbf{u}} \quad (5.12)$$

A.2 Mobile frame following the flow of the whole system - $\frac{D_0}{Dt}$

Before proceeding with the derivation of the entropy production σ , it is necessary to pay attention in which frame the flows of the two fluids will be studied. The entropy density s of the whole system has to be derived in function of time. As we are interested on time scales when the two fluids move independently with average velocities $\mathbf{v}_i(\mathbf{r})$, two different expressions exist for the total temporal derivative: $\frac{D_i}{Dt} = \frac{\partial}{\partial t} + \mathbf{v}_i(\mathbf{r}) \cdot \frac{\partial}{\partial \mathbf{r}}$. In order to gain mathematical simplicity, it is necessary to write all the equations in a common reference frame, and define a unique temporal derivative $\frac{D_0}{Dt}$.

The best reference frame is the mobile frame of centre of mass following the motion of the whole system with the velocity $\mathbf{v}_0 = \frac{\rho_1}{\rho} \mathbf{v}_1 + \frac{\rho_2}{\rho} \mathbf{v}_2$ whom *mobile operator* $\frac{D_0}{Dt}$ is:

$$\frac{D_0}{Dt} = \frac{\partial}{\partial t} + \mathbf{v}_0(\mathbf{r}) \cdot \frac{\partial}{\partial \mathbf{r}} = \frac{D_i}{Dt} - [\mathbf{v}_i(\mathbf{r}) - \mathbf{v}_0(\mathbf{r})] \cdot \frac{\partial}{\partial \mathbf{r}} \quad (5.13)$$

Changing the frame from the fixed frame to the mobile frame following the flow of the system, implies changing the velocities \mathbf{u} with *peculiar velocities* $\mathbf{u}' = \mathbf{u} - \mathbf{v}_0(\mathbf{r})$. The change leads to different expressions for the equilibrium chemical potential μ_i^{eq} , for the total Gibbs equation (5.9) and the conservation law (5.12).

Gibbs postulate in the frame following the flow of the system - $\frac{D_0}{Dt}$

In this case, the variation of the entropy in respect to time is:

$$T \frac{D_0}{Dt} (\rho s) = -\frac{D_0}{Dt} \sum_i \int f_i \left[k_B T \ln f_i + \frac{m_i u'^2}{2} + V_i(\mathbf{r}) - \mu_i^{eq} \right] d\mathbf{u}' \quad (5.14)$$

The conservation law in the frame following the flow of the system - $\frac{D_0}{Dt}$

Using $\mathbf{u}' = \mathbf{u} - \mathbf{v}_0(\mathbf{r}, t)$ as an independent variable instead of \mathbf{u} , changes the meanings of $\frac{\partial f_i}{\partial t}$ and $\frac{\partial f_i}{\partial \mathbf{r}}$ from the conservation law (5.12). The distribution functions f_i are implicitly dependent on t and \mathbf{r} through the dependence of \mathbf{u}' . Hence, $\frac{\partial f_i}{\partial t}$ and $\frac{\partial f_i}{\partial \mathbf{r}}$ are to be replaced by $\frac{\partial f_i}{\partial t} - \frac{\partial \mathbf{v}_0}{\partial t} \cdot \frac{\partial f_i}{\partial \mathbf{u}'}$, respectively $\frac{\partial f_i}{\partial \mathbf{r}} - \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} \cdot \frac{\partial f_i}{\partial \mathbf{u}'}$ [14].

$$\frac{\partial f_i}{\partial t} - \frac{\partial \mathbf{v}_0}{\partial t} \cdot \frac{\partial f_i}{\partial \mathbf{u}'} = -(\mathbf{u}' + \mathbf{v}_0) \cdot \left(\frac{\partial f_i}{\partial \mathbf{r}} - \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} \cdot \frac{\partial f_i}{\partial \mathbf{u}'} \right) - \frac{\partial}{\partial \mathbf{u}'} \cdot \left(\frac{f_i \mathbf{F}_i}{m_i} + \mathbf{J}_i^{\mathbf{u}'} \right) \quad (5.15)$$

$$\Rightarrow \boxed{\frac{Df_i(\mathbf{u}', \mathbf{r}, t)}{Dt} = -\mathbf{u}' \cdot \frac{\partial f_i}{\partial \mathbf{r}} - \frac{\partial}{\partial \mathbf{u}'} \cdot \left(\frac{f_i \mathbf{F}_i}{m_i} - f_i \frac{D\mathbf{v}_0}{Dt} + \mathbf{J}_i^{\mathbf{u}'} \right) + \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\frac{\partial f_i}{\partial \mathbf{u}'} \mathbf{u}' \right)} \quad (5.16)$$

B Thermokinetic equations

B.1 Conservation laws From the conservation law of particles (5.16), several other conservation laws are obtained which will be found handfull at a later time. As in the time frame of interest, the two fluids move independently, the conservation laws will be either written in the common frame with mobile operator $\frac{D_0}{Dt}$, either in their own frames with mobile operators $\frac{D_i}{Dt}$ (see Appendix 5.B.1 for calculation details).

Mobile frame following the flow of the system - D_0/Dt

Conservation of the density of particles n_i

$$\frac{D_0 n_i}{Dt} = \int \frac{D_0 f_i}{Dt} d\mathbf{u}' = -\frac{\partial}{\partial \mathbf{r}} \cdot [n_i(\mathbf{v}_i - \mathbf{v}_0)] - n_i \frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \quad (5.17)$$

Conservation of linear momentum - Motion equations

The motion equation for the fluid i is obtained from the same conservation law (5.16):

$$\frac{D_0}{Dt} [\rho_i(\mathbf{v}_i - \mathbf{v}_0)] = \int m_i \frac{Df_i}{Dt} \mathbf{u}' d\mathbf{u}' \Rightarrow \quad (5.18)$$

$$\begin{aligned} \frac{D_0}{Dt} [\rho_i(\mathbf{v}_i - \mathbf{v}_0)] = & -Div \overline{\overline{P_i^0}} + \frac{\rho_i}{m_i} \left(\mathbf{F}_i - m_i \frac{D\mathbf{v}_0}{Dt} \right) + \int m_i \mathbf{J}_i^{\mathbf{u}'} d\mathbf{u}' \\ & - \rho_i(\mathbf{v}_i - \mathbf{v}_0) Grad \mathbf{v}_0 - div \mathbf{v}_0 \rho_i(\mathbf{v}_i - \mathbf{v}_0) \end{aligned} \quad (5.19)$$

where the partial pressure tensor $\overline{\overline{P_i^0}}$ of the gas i in the frame following the flow of the entire

system is defined as:

$$\overline{\overline{P}}_i \stackrel{def}{=} \int m_i f_i \mathbf{u}' \mathbf{u}' d\mathbf{u}' = \int m_i f_i (\mathbf{u} - \mathbf{v}_0)(\mathbf{u} - \mathbf{v}_0) d\mathbf{u} \quad (5.20)$$

The equation of motion for the entire system is obtained by summing up the motion equations for each fluid written in the frame of the whole system (5.19):

$$\rho \frac{D_0 \mathbf{v}_0}{Dt} = -Div \left(\overline{\overline{P}}_1 + \overline{\overline{P}}_2 \right) + \frac{\rho_1}{m_1} \mathbf{F}_1 + \frac{\rho_2}{m_2} \mathbf{F}_2 + \int (m_1 \mathbf{J}_1^{\mathbf{u}} + m_2 \mathbf{J}_2^{\mathbf{u}}) d\mathbf{u}' \quad (5.21)$$

Mobile frame following the flow of each gas i - D_i/Dt

Conservation of the density of particles n_i

$$\frac{D_i n_i}{Dt} = \frac{D_0 n_i}{Dt} - [\mathbf{v}_0(\mathbf{r}) - \mathbf{v}_i(\mathbf{r})] \cdot \frac{\partial n_i}{\partial \mathbf{r}} = -n_i \frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_i \quad (5.22)$$

Conservation of linear momentum - Motion equations

The equation (5.19) represents the motion equation written in the frame following the flow of the whole system. When this same equation is written in the frame following the flow of the gas i of average velocity \mathbf{v}_i , the motion equation takes a simpler form (see Appendix 5.B.1 for details).

$$\rho_i \frac{D_i \mathbf{v}_i}{Dt} = -Div \overline{\overline{P}}_i + \frac{\rho_i}{m_i} \mathbf{F}_i + \int m_i \mathbf{J}_i^{\mathbf{u}} d\mathbf{u}' \quad (5.23)$$

where the pressure tensor $\overline{\overline{P}}_i$ corresponds to the pressure of the gas i in the mobile frame following the motion of the gas. It has to be emphasised that even if the pressure tensors $\overline{\overline{P}}_i$ and $\overline{\overline{P}}_i^0$ relate to the pressure of the same gas i , their values depend on the frame. When changing the frame, they differ by a 'dynamic' term related to the relative velocity of the frames $\rho_i (\mathbf{v}_i - \mathbf{v}_0)(\mathbf{v}_i - \mathbf{v}_0)$.

$$\overline{\overline{P}}_i \stackrel{def}{=} \int m_i f_i (\mathbf{u} - \mathbf{v}_i)(\mathbf{u} - \mathbf{v}_i) d\mathbf{u} = \overline{\overline{P}}_i^0 - \rho_i (\mathbf{v}_i - \mathbf{v}_0)(\mathbf{v}_i - \mathbf{v}_0) \quad (5.24)$$

B.2 The entropy production σ in the frame of the system - $\frac{D_0}{Dt}$

The expression for the entropy production for the whole system $\sigma(\mathbf{r})$ is deduced from the 'total' Gibbs equation (5.14). To obtain its expression, is necessary to identify the entropy flux associated to reversible processes after the derivation of the entropy density s with time:

$$T \frac{D_0(\rho s)}{Dt} = -\frac{D_0}{Dt} \sum_i \int f_i \left(k_B T \ln f_i + \frac{m_i u'^2}{2} + V_i(\mathbf{r}) - \mu_i^{eq} \right) d\mathbf{u} \quad (5.25)$$

After some calculations, the expressions for the flux and the entropy production are obtained.(see Appendix 5.B.2 for details)

$$\begin{aligned}
T \frac{D_0(\rho s)}{Dt} = & \frac{\partial}{\partial \mathbf{r}} \cdot \overbrace{\int \mathbf{u}' \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}'}^{-\mathbf{J}^s} \\
& + \left(\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \right) \underbrace{\int \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}'}_{-\rho s} \\
& + \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\overline{\overline{P_1}} + \overline{\overline{P_2}} \right) - \sum_i \int \mathbf{J}_i^u \cdot \frac{\partial}{\partial \mathbf{u}'} \left(kT \ln f_i + \frac{m_i u'^2}{2} \right) d\mathbf{u}'
\end{aligned} \tag{5.26}$$

As noticed in (5.26), the flux \mathbf{J}^s is the diffusive entropy flux and is a sum of several fluxes: kinetic energy fluxes $\sum_i \int \mathbf{u}' f_i m_i u'^2 / 2 d\mathbf{u}'$, potential energy fluxes $\sum_i \int \mathbf{u}' f_i V_i d\mathbf{u}'$ and entropic fluxes $\sum_i \int \mathbf{u}' kT f_i \ln f_i d\mathbf{u}'$. The remaining expression of the entropy increase with time is represented by the convective entropy flux $\rho s \frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0$ and the entropy production σ . The latter is associated to dissipative processes, in this case 'collisions' between particles.

The entropy production has the structure of a bilinear form: it consists of a sum of products of two factors. One of these factors in each term is a flow quantity ('collision' fluxes \mathbf{J}_i^u , momentum flows or pressure tensors $\overline{\overline{P_1}} + \overline{\overline{P_2}}$) already introduced in the conservation laws (5.19) and (5.21). The other factor in each term is related to a gradient of an intensive parameter. These quantities which multiply the fluxes in the expression of the entropy production are called *thermodynamic forces*, causes for their effects - the fluxes.

$$\left\{ \begin{aligned} \mathbf{J}^s(\mathbf{r}) &= -\frac{1}{T} \int \mathbf{u}' \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}' \\ T\sigma(\mathbf{r}) &= -\sum_i \int \mathbf{J}_i^u \cdot \frac{\partial}{\partial \mathbf{u}'} \left(kT \ln f_i + \frac{m_i u'^2}{2} \right) d\mathbf{u}' + \left(\overline{\overline{P_1}} + \overline{\overline{P_2}} \right) : \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} \end{aligned} \right. \tag{5.27}$$

B.3 Phenomenological laws With the expression for the entropy production for the entire system (5.27), phenomenological equations between fluxes and forces are written assuming locality in the phase space. The Curie symmetry principle is respected imposing no coupling between fluxes and thermodynamic forces of different tensorial character in an

isotropic medium. The Onsager coefficients L_{ij} are introduced as proportionality factors.

$$\begin{cases} \mathbf{J}_1^{\mathbf{u}} = -L_{11} \left(\frac{kT}{f_1} \frac{\partial f_1}{\partial \mathbf{u}'} + m_1 \mathbf{u}' \right) - L_{12} \left(\frac{kT}{f_2} \frac{\partial f_2}{\partial \mathbf{u}'} + m_2 \mathbf{u}' \right) \\ \mathbf{J}_2^{\mathbf{u}} = -L_{21} \left(\frac{kT}{f_1} \frac{\partial f_1}{\partial \mathbf{u}'} + m_1 \mathbf{u}' \right) - L_{22} \left(\frac{kT}{f_2} \frac{\partial f_2}{\partial \mathbf{u}'} + m_2 \mathbf{u}' \right) \end{cases} \quad (5.28)$$

The same laws can also be written in terms of damping coefficients $\eta_{ij} \stackrel{def}{=} \frac{m_j L_{ij}}{f_j}$:

$$\Leftrightarrow \begin{cases} \mathbf{J}_1^{\mathbf{u}} = -\eta_{11} \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) - \eta_{12} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \\ \mathbf{J}_2^{\mathbf{u}} = -\eta_{21} \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) - \eta_{22} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \end{cases} \quad (5.29)$$

With the help of the kinetic theory [12], i.e. with conservation laws of momentum and energy which apply at each collision, it can be shown that the newly introduced damping coefficients η_{ij} are not linearly independent. As it is shown in the Appendix 5.B.3, only one coefficient is independent. Let η_{11} be that one, and let's call it simply η .

$$\eta \stackrel{def}{=} \eta_{11} = -\frac{m_2}{m_1} \eta_{21} = -\frac{m_1}{m_2} \frac{n_2}{n_1} \eta_{12} = \frac{n_2}{n_1} \eta_{22} \quad (5.30)$$

With these dependencies, the phenomenological laws write:

$$\begin{cases} \mathbf{J}_1^{\mathbf{u}} = -\eta \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) + \eta \frac{m_2}{m_1} \frac{n_1}{n_2} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \\ \mathbf{J}_2^{\mathbf{u}} = \eta \frac{m_1}{m_2} \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) - \eta \frac{n_1}{n_2} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \end{cases} \quad (5.31)$$

C Dynamic equations

Having an explicit expression for the 'collision fluxes' $\mathbf{J}_i^{\mathbf{u}}$ (5.31), the motion equations (5.21) and (5.23) can now be written as:

$$\begin{cases} \rho_1 \frac{D_1 \mathbf{v}_1}{Dt} = -Div \overline{\overline{P_1}} + n_1 \mathbf{F}_1 - \eta \frac{n}{n_2} \frac{\rho_1 \rho_2}{\rho} (\mathbf{v}_1 - \mathbf{v}_2) \\ \rho_2 \frac{D_2 \mathbf{v}_2}{Dt} = -Div \overline{\overline{P_2}} + n_2 \mathbf{F}_2 + \eta \frac{n}{n_2} \frac{\rho_1 \rho_2}{\rho} (\mathbf{v}_1 - \mathbf{v}_2) \\ \rho \frac{D_0 \mathbf{v}_0}{Dt} = -Div \left(\overline{\overline{P_1}} + \overline{\overline{P_2}} \right) + n_1 \mathbf{F}_1 + n_2 \mathbf{F}_2 \end{cases} \quad (5.32)$$

The friction or the dragging force $\mathbf{R}_{12} = -\eta \frac{n}{n_2} \frac{\rho_1 \rho_2}{\rho} (\mathbf{v}_1 - \mathbf{v}_2)$, as well as the damping coefficient η can be related to collisions either formally and explicitly as done in the Appendix 5.B.3, either in a simple intuitive way as presented by S. Chapman and T.G. Cowling in their book [15]. It can be assumed that there are N_{12} collisions between unlike particles per unit volume and time, and that these reduce the average velocities \mathbf{v}_1 and \mathbf{v}_2 of the colliding molecules to a common velocity \mathbf{v}_0 . By conservation of momentum then $m_1 \mathbf{v}_1 + m_2 \mathbf{v}_2 = (m_1 + m_2) \mathbf{v}_0$. Hence, the average momentum change of one particle m_1 at a collision is $m_1(\mathbf{v}_0 - \mathbf{v}_1) = -m_1 m_2 (\mathbf{v}_1 - \mathbf{v}_2) / (m_1 + m_2)$. Then, the drag force on the first gas is N_{12} times this change of momentum. From this equality, a relaxation time τ_{12} can be defined as:

$$\tau_{12} = \frac{1}{\eta} \frac{n_2}{n} = \frac{n_1 n_2 (m_1 + m_2)}{\rho N_{12}} = \frac{\rho_2}{\rho} \frac{n_1}{N_{12}} + \frac{\rho_1}{\rho} \frac{n_2}{N_{12}} \quad (5.33)$$

Following the line of reasoning of S. Chapman and T.G. Cowling, $\frac{n_1}{N_{12}}$ and $\frac{n_2}{N_{12}}$ represent *collision intervals* for particles m_1 and m_2 in mutual collisions, and τ_{12} is their weighted mean with weighting factors ρ_2/ρ , respectively ρ_1/ρ . The collision intervals for each fluid τ_1 and τ_2 are depending on both collisions within the same fluid, and also within different fluids.

$$\tau_1 = \frac{n_1}{N_{11} + N_{12}} \quad \tau_2 = \frac{n_2}{N_{12} + N_{22}} \quad \tau_{12} = \frac{\rho_2}{\rho} \frac{n_1}{N_{12}} + \frac{\rho_1}{\rho} \frac{n_2}{N_{12}} \quad (5.34)$$

As due to collisions between unlike particles, the force \mathbf{R}_{12} is dissipative. As the fluid 1 moves with the velocity $\mathbf{v}_1 - \mathbf{v}_2$ relative to the fluid 2, it exerts a force $\mathbf{R}_{21} = -\mathbf{R}_{12}$ on the fluid 2 and the positive work done, namely $\mathbf{R}_{21}(\mathbf{v}_1 - \mathbf{v}_2)$ is dissipated in both fluids. The way this energy divides between the fluids depends on the ratio of the masses. Let ξ_{12} be the energy dissipated in the fluid 1 due to the work done by the friction force, then their ratio is (see [6, 12, 16] for further justification):

$$\xi_{12} + \xi_{21} = \mathbf{R}_{21}(\mathbf{v}_1 - \mathbf{v}_2) \quad \frac{\xi_{12}}{\xi_{21}} = \frac{m_2}{m_1} \quad (5.35)$$

To get a better understanding of the physical significance of the existing terms from the dynamic equations above (5.32), the well known case of mutual diffusion will be treated first.

5.B Calculation details

5.B.1 Conservation laws

From the conservation law of particles (5.16), several conservation laws can be obtained which will be found handful on a later time.

The density of particles n_i The conservation equation for the density of particles n_i is:

$$\begin{aligned} \frac{D_0 n_i}{Dt} &= \int \frac{D_0 f_i}{Dt} d\mathbf{u}' = -\frac{\partial}{\partial \mathbf{r}} \int f_i \mathbf{u}' d\mathbf{u}' + \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \int \frac{\partial f_i}{\partial \mathbf{u}'} \mathbf{u}' d\mathbf{u}' \\ &= -\text{div} (n_i \langle \mathbf{u}'_i \rangle) - n_i \text{div} \mathbf{v}_0 = -\text{div} [n_i(\mathbf{v}_i - \mathbf{v}_0)] - n_i \text{div} \mathbf{v}_0 \end{aligned} \quad (5.36)$$

The average velocity \mathbf{v}_i - the motion equation

Mobile frame following the flow of the whole system The motion equation is obtained from the same number of particles conservation law (5.16):

$$\begin{aligned} \frac{D_0}{Dt} [\rho_i(\mathbf{v}_i - \mathbf{v}_0)] &= \int m_i \frac{Df_i}{Dt} \mathbf{u}' d\mathbf{u}' \\ &= - \int m_i \mathbf{u}' \cdot \frac{\partial f_i \mathbf{u}'}{\partial \mathbf{r}} d\mathbf{u}' - \int \left[\frac{\partial}{\partial \mathbf{u}'} \cdot \left(f_i \mathbf{F}_i - m_i f_i \frac{D\mathbf{v}_0}{Dt} + m_i \mathbf{J}_i^{\mathbf{u}} \right) \right] \cdot \mathbf{u}' d\mathbf{u}' \\ &\quad + \int m_i \left[\frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\frac{\partial f_i}{\partial \mathbf{u}'} \mathbf{u}' \right) \right] \cdot \mathbf{u}' d\mathbf{u}' \end{aligned}$$

$$\boxed{\begin{aligned} \frac{D_0}{Dt} [\rho_i(\mathbf{v}_i - \mathbf{v}_0)] &= -\text{Div} \overline{\overline{P}}_i + \frac{\rho_i}{m_i} \left(\mathbf{F}_i - m_i \frac{D\mathbf{v}_0}{Dt} \right) + \int m_i \mathbf{J}_i^{\mathbf{u}} d\mathbf{u}' \\ &\quad - \rho_i(\mathbf{v}_i - \mathbf{v}_0) \text{Grad} \mathbf{v}_0 - \text{div} \mathbf{v}_0 \rho_i(\mathbf{v}_i - \mathbf{v}_0) \end{aligned}} \quad (5.37)$$

where the partial pressure tensor $\overline{\overline{P}}_i^0$ of the gas i in the frame following the flow of the entire system is defined as:

$$\overline{\overline{P}}_i^0 \stackrel{\text{def}}{=} \int m_i f_i \mathbf{u}' \mathbf{u}' d\mathbf{u}' = \int m_i f_i (\mathbf{u} - \mathbf{v}_0)(\mathbf{u} - \mathbf{v}_0) d\mathbf{u} \quad (5.38)$$

Summing up the motion equations for each fluid written for the frame following the motion of the whole system (5.37), the equation of motion for the entire system writes:

$$\rho \frac{D_0 \mathbf{v}_0}{Dt} = -\text{Div} \left(\overline{\overline{P}}_1 + \overline{\overline{P}}_2 \right) + \frac{\rho_1}{m_1} \mathbf{F}_1 + \frac{\rho_2}{m_2} \mathbf{F}_2 + \int (m_1 \mathbf{J}_1^{\mathbf{u}} + m_2 \mathbf{J}_2^{\mathbf{u}}) d\mathbf{u}' \quad (5.39)$$

Mobile frame following the flow of each gas i The equation (5.37) represents the motion equation written for the frame following the flow of the whole system. An equivalent

form of this motion equation is:

$$\rho_i \frac{D_0}{Dt} (\mathbf{v}_i - \mathbf{v}_0) = -Div \left[\overline{\overline{P}}_i^0 - \rho_i (\mathbf{v}_i - \mathbf{v}_0) (\mathbf{v}_i - \mathbf{v}_0) \right] + \frac{\rho_i}{m_i} \left(\mathbf{F}_i - m_i \frac{D\mathbf{v}_0}{Dt} \right) + \int m_i \mathbf{J}_i^{\mathbf{u}} d\mathbf{u}' - \rho_i (\mathbf{v}_i - \mathbf{v}_0) Grad \mathbf{v}_0 \quad (5.40)$$

When this same equation is written in the mobile frame following the flow of the gas i of average velocity \mathbf{v}_i , the motion equation takes a simpler form. Starting from the equation (5.40), the identity writes as:

$$\boxed{\rho_i \frac{D_i \mathbf{v}_i}{Dt} = -Div \overline{\overline{P}}_i^i + \frac{\rho_i}{m_i} \mathbf{F}_i + \int m_i \mathbf{J}_i^{\mathbf{u}'} d\mathbf{u}'} \quad (5.41)$$

where the pressure tensor $\overline{\overline{P}}_i^i$ corresponds to the pressure of the gas i in the mobile frame following the motion of the gas and its corresponding mobile operator $\frac{D_i}{Dt}$ is:

$$\overline{\overline{P}}_i^i \stackrel{def}{=} \int m_i f_i (\mathbf{u} - \mathbf{v}_i) (\mathbf{u} - \mathbf{v}_i) d\mathbf{u} = \overline{\overline{P}}_i^0 - \rho_i (\mathbf{v}_i - \mathbf{v}_0) (\mathbf{v}_i - \mathbf{v}_0) \quad (5.42)$$

5.B.2 The entropy production σ in the frame following the flow of the system - $\frac{D_0}{Dt}$

The expression for the entropy production for the whole system $\sigma(\mathbf{r})$ is deduced from the 'total' Gibbs equation (5.14). To obtain its expression, is necessary to identify the entropy flux associated to reversible processes after the derivation of the entropy density s with time:

$$T \frac{D_0(\rho s)}{Dt} = -\frac{D_0}{Dt} \sum_i \int f_i \left(k_B T \ln f_i + \frac{m_i u'^2}{2} + V_i(\mathbf{r}) - \mu_i^{eq} \right) d\mathbf{u} \quad (5.43)$$

$$= -\sum_i \int \frac{D_0 f_i}{Dt} \left[k_B T (\ln f_i + 1) + \frac{m_i u'^2}{2} + V_i(\mathbf{r}) - \mu_i^{eq} \right] d\mathbf{u} \quad (5.44)$$

The calculation is divided in three steps:

(a).

$$\begin{aligned}
& \int \frac{D_0 f_i}{Dt} [k_B T (\ln f_i + 1) - \mu_i^{eq}] d\mathbf{u}' \\
&= - \int \mathbf{u}' \cdot \frac{\partial f_i}{\partial \mathbf{r}} [k_B T (\ln f_i + 1) - \mu_i^{eq}] d\mathbf{u}' \\
&\quad - \int \frac{\partial}{\partial \mathbf{u}'} \cdot \left(\frac{f_i \mathbf{F}_i}{m_i} - f_i \frac{D_0 \mathbf{v}_0}{Dt} + \mathbf{J}_i^{\mathbf{u}} \right) [k_B T (\ln f_i + 1) - \mu_i^{eq}] d\mathbf{u}' \\
&\quad + \int \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\frac{\partial f_i}{\partial \mathbf{u}'} \mathbf{u}' \right) [k_B T (\ln f_i + 1) - \mu_i^{eq}] d\mathbf{u}' \\
&= - \frac{\partial}{\partial \mathbf{r}} \cdot \int f_i \mathbf{u}' (k_B T \ln f_i - \mu_i^{eq}) d\mathbf{u}' - \left(\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \right) \int f_i (k_B T \ln f_i - \mu_i^{eq}) d\mathbf{u}' \\
&\quad + \int \mathbf{J}_i^{\mathbf{u}} \cdot \frac{\partial}{\partial \mathbf{r}} (k_B T \ln f_i) d\mathbf{u}'
\end{aligned}$$

(b).

$$\begin{aligned}
& \int \frac{D_0 f_i m_i u'^2}{Dt} \frac{1}{2} d\mathbf{u}' \\
&= - \int \mathbf{u}' \cdot \frac{\partial f_i m_i u'^2}{\partial \mathbf{r}} \frac{1}{2} d\mathbf{u}' - \int \frac{\partial}{\partial \mathbf{u}'} \cdot \left(\frac{f_i \mathbf{F}_i}{m_i} - f_i \frac{D_0 \mathbf{v}_0}{Dt} + \mathbf{J}_i^{\mathbf{u}} \right) \frac{m_i u'^2}{2} d\mathbf{u}' \\
&\quad + \int \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\frac{\partial f_i}{\partial \mathbf{u}'} \mathbf{u}' \right) \frac{m_i u'^2}{2} d\mathbf{u}' \tag{5.45} \\
&= - \frac{\partial}{\partial \mathbf{r}} \cdot \int f_i \mathbf{u}' \frac{m_i u'^2}{2} d\mathbf{u}' - \left(\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \right) \int f_i \frac{m_i u'^2}{2} d\mathbf{u}' \\
&\quad + \left(\frac{\mathbf{F}_i}{m_i} - \frac{D_0 \mathbf{v}_0}{Dt} \right) \frac{\rho_i}{m_i} (\mathbf{v}_i - \mathbf{v}_0) + \int \mathbf{J}_i^{\mathbf{u}} m_i \mathbf{u}' d\mathbf{u}' - \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \overline{\overline{P_i^0}}
\end{aligned}$$

where $\overline{\overline{P_i^0}}$ is the partial pressure of the fluid i in the barycenter frame:

$$\overline{\overline{P_i^0}} = \int m_i f_i \mathbf{u}' \mathbf{u}' d\mathbf{u}' \tag{5.46}$$

(c).

$$\int \frac{D_0 f_i}{Dt} V_i d\mathbf{u}' = - \frac{\partial}{\partial \mathbf{r}} \cdot \int f_i \mathbf{u}' V_i d\mathbf{u}' - \left(\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \right) \int f_i V_i d\mathbf{u}' - \frac{\rho_i}{m_i} \mathbf{F}_i \cdot (\mathbf{v}_i - \mathbf{v}_0) \tag{5.47}$$

The expression for the entropy flux and the production of entropy in the barycenter frame is obtained after summing up the above calculated integrals:

$$T \frac{D\rho s}{Dt} = \frac{\partial}{\partial \mathbf{r}} \cdot \int \mathbf{u}' \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}' + \left(\frac{\partial}{\partial \mathbf{r}} \cdot \mathbf{v}_0 \right) \cdot \underbrace{\int \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}'}_{-\rho s} \quad (5.48)$$

$$+ \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\overline{P_i^0} + \overline{P_i^0} \right) - \sum_i \int \mathbf{J}_i^{\mathbf{u}} \cdot \frac{\partial}{\partial \mathbf{u}'} \left(kT \ln f_i + \frac{m_i u'^2}{2} \right) d\mathbf{u}' \Rightarrow \begin{cases} \mathbf{J}^s = -\frac{1}{T} \int \mathbf{u}' \sum_i f_i \left(kT \ln f_i - \mu_i^{eq} + \frac{m_i u'^2}{2} + V_i \right) d\mathbf{u}' \\ T \sigma(\mathbf{r}) = - \sum_i \int \mathbf{J}_i^{\mathbf{u}} \cdot \frac{\partial}{\partial \mathbf{u}'} \left(kT \ln f_i + \frac{m_i u'^2}{2} \right) d\mathbf{u}' + \frac{\partial \mathbf{v}_0}{\partial \mathbf{r}} : \left(\overline{P_i^0} + \overline{P_i^0} \right) \end{cases} \quad (5.49)$$

5.B.3 Conservation laws at each collision - Constraints

When two particles of the same kind i collide, the momentum and energy of the fluid i is conserved, while when particles of different kind collide, the momentum and energy of the fluid i (and also j) is changed. The lack of conservation of the momentum and energy of the fluid i due to the encounter with particles of a different kind leads to the appearance of the dissipation. It explicitly appear in the motion and energy equation for the fluid i . In spite of it, when writing the motion and energy equation for the entire system no dissipative terms are expected as the energy and momentum of one fluid is transferred to the other.

As we stated at the beginning of this section the fluxes $\mathbf{J}_1^{\mathbf{u}}$ and $\mathbf{J}_2^{\mathbf{u}}$ are a reflection of the collisions of the particles of the fluid i either with particles of the same kind, either with particles of a different kind (fluid) j ($j \neq i$). These considerations lead to constraints which have to be satisfied by the collision fluxes $\mathbf{J}_i^{\mathbf{u}}$.

Conservation of the number of particles

The number of particles is unchanged at each collision, leading to the first constraint.

$$\int C_{ij} d\mathbf{u}' = 0 \quad \forall i, j \quad \Rightarrow \quad \sum_j \int C_{ij} d\mathbf{u}' = - \int \frac{\partial \mathbf{J}_i^{\mathbf{u}}}{\partial \mathbf{u}'} d\mathbf{u}' = 0 \quad (5.50)$$

As we immediately see, the first constraint confirms the hypothesis of writing the collision term as a divergence of a flux that vanishes at infinity.

Conservation of momentum and energy

Momentum and energy transferred between particles of the same fluid conserve the momentum and the energy of that fluid in any reference frame [12] (see the first equations from (5.51) and (5.52)). To deal with collisions between unlike particles it is convenient to define *friction forces* \mathbf{R}_{ij} between the i th and the j th fluid, and write the conservation laws in a different manner (see the second equations from (5.51) and (5.52)):

$$\text{Momentum : } \begin{cases} \int C_{ii} m_i \mathbf{u}' d\mathbf{u}' = 0 \\ \int C_{12} m_1 \mathbf{u}' d\mathbf{u}' + \int C_{21} m_2 \mathbf{u}' d\mathbf{u}' = 0 \end{cases} \Leftrightarrow \mathbf{R}_{12} = -\mathbf{R}_{21} \quad (5.51)$$

$$\text{Energy : } \begin{cases} \int C_{ii} \frac{m_i \mathbf{u}'^2}{2} d\mathbf{u}' = 0 \\ \int C_{12} \frac{m_1 \mathbf{u}'^2}{2} d\mathbf{u}' + \int C_{21} \frac{m_2 \mathbf{u}'^2}{2} d\mathbf{u}' = 0 \end{cases} \quad (5.52)$$

where the friction forces \mathbf{R}_{ij} were defined as:

$$\mathbf{R}_{ij} = \int C_{ij} m_i \mathbf{u}' d\mathbf{u}' \quad (5.53)$$

To see the implications of the constraints presented above on the damping coefficients introduced earlier η_{ij} , it is necessary first to identify the explicit expressions for the collision terms C_{ij} . With the expressions for the 'collision fluxes' $\frac{\partial \mathbf{J}_i^{\mathbf{u}}}{\partial \mathbf{u}} : \frac{\partial \mathbf{J}_i^{\mathbf{u}}}{\partial \mathbf{u}} = -\sum_{j=1,2} C_{ij}$ and (5.31), and taking in consideration the constraints, the expressions that make most physical sense³ for the collision terms C_{ij} are the following:

$$\begin{cases} C_{ii} = \eta_{i1} \frac{kT}{m_1} \frac{\partial^2 f_1}{\partial \mathbf{u}'^2} + \eta_{i2} \frac{kT}{m_2} \frac{\partial^2 f_2}{\partial \mathbf{u}'^2} & \forall i \\ C_{12} = \eta_{11} \frac{\partial}{\partial \mathbf{u}'}(f_1 \mathbf{u}') + \eta_{12} \frac{\partial}{\partial \mathbf{u}'}(f_2 \mathbf{u}') \\ C_{21} = \eta_{21} \frac{\partial}{\partial \mathbf{u}'}(f_1 \mathbf{u}') + \eta_{22} \frac{\partial}{\partial \mathbf{u}'}(f_2 \mathbf{u}') \end{cases} \quad (5.54)$$

Replacing these expressions in the conservation laws above, a system of equations is retrieved which has to be satisfied by the damping coefficients η_{ij} . It is concluded that only one

³If no coupling is considered between the fluids, i.e. $\eta_{12} = \eta_{21} = 0$, then the conservation laws written above, (5.51) and (5.52) impose $\eta_{11} = \eta_{22} = 0$, meaning no damping neither for the Brownian particles, nor for the second fluid, the bath. However, if coupling is considered (four damping coefficients), the knowledge gained in the previous chapter 5.3 leads to one solution satisfying the constraints presented (5.51) and (5.52).

damping coefficient is independent, and we denominate it η .

$$\begin{cases} \eta_{11} - \eta_{22} - \eta_{12} \frac{m_1}{m_2} + \eta_{21} \frac{m_2}{m_1} = 0 \\ \eta_{11} n_1 + \eta_{12} \frac{m_1}{m_2} n_2 = 0 \\ \eta_{21} \frac{m_2}{m_1} n_1 + \eta_{22} n_2 = 0 \end{cases} \quad (5.55)$$

$$\Rightarrow \eta \stackrel{def}{=} \eta_{11} = -\frac{m_2}{m_1} \eta_{21} = -\frac{m_1}{m_2} \frac{n_2}{n_1} \eta_{12} = \frac{n_2}{n_1} \eta_{22} \quad (5.56)$$

With these the expressions for the 'collision fluxes' $\mathbf{J}_i^{\mathbf{u}}$ and the friction forces \mathbf{R}_{ij} are:

$$\begin{cases} \mathbf{J}_1^{\mathbf{u}} = -\eta \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) + \eta \frac{m_2}{m_1} \frac{n_1}{n_2} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \\ \mathbf{J}_2^{\mathbf{u}} = \eta \frac{m_1}{m_2} \left(\frac{kT}{m_1} \frac{\partial f_1}{\partial \mathbf{u}'} + f_1 \mathbf{u}' \right) - \eta \frac{n_1}{n_2} \left(\frac{kT}{m_2} \frac{\partial f_2}{\partial \mathbf{u}'} + f_2 \mathbf{u}' \right) \end{cases} \quad (5.57)$$

$$\mathbf{R}_{12} = -\mathbf{R}_{21} = -\eta \frac{n}{n_2} \frac{\rho_1 \rho_2}{\rho} (\mathbf{v}_1 - \mathbf{v}_2) \quad (5.58)$$

Bibliography

- [1] J. C. SLONCZEWSKI, *J. Magn. Magn. Mater.* **159**, L1 (1996).
- [2] L. BERGER, *Phys. Rev. B* **54**, 9353 (1996).
- [3] M. D. STILES and J. MILTAT, Spin transfer torque and dynamics, in *Spin dynamics in confined magnetic structures III*, volume **101**, pp. 225–308, Springer-Verlag Berlin Heidelberg, 2006.
- [4] D. C. RALPH and M. D. STILES, *Journal of Magnetism and Magnetic Materials* **320**, 1190 (2008).
- [5] J.-E. WEGROWE, S. M. SANTOS, M.-C. CIORNEI, H.-J. DROUHIN, and J. M. RUBI, *Physical Review B* **77**, 174408 (2008).
- [6] L. C. WOODS, *Magnetoplasmas and superfluids*, chapter X, In [8], 1975.
- [7] I. PRIGOGINE and P. MAZUR, *Physica* **17** (1951).
- [8] L. C. WOODS, *The thermodynamics of fluid systems*, Calendron Press, 1975.
- [9] M. D. STILES and A. ZANGWILL, *Phys. Rev.* **66**, 14407 (2002).

- [10] S. ZHANG and P. M. LEVY, *Phys. Rev. B* **65**, 052409 (2002).
- [11] J. E. WEGROWE, M. C. CIORNEI, and H. J. DROUHIN, *J. Phys.:Condens Matter* **19**, 165213 (2007).
- [12] L. C. WOODS, *Kinetic processes*, chapter XIII, In [8], 1975.
- [13] J. Z. SUN, *IBM J. Res. and Dev.* **50**, 81 (2006).
- [14] S. CHAPMAN and T. G. COWLING, *The mathematical theory of non-uniform gases*, Cambridge University Press, third edition, 1970.
- [15] S. CHAPMAN and T. G. COWLING, *The mathematical theory of non-uniform gases*, pp. 106–109, 138–142, In [14], third edition, 1970.
- [16] L. C. WOODS, *Fluid mixtures*, chapter IX, In [8], 1975.

Chapter 6

Conclusions

The goal of this thesis was to see the contribution of inertia in damped macrospin dynamics. A macrospin mass (tensor of inertia) was introduced not related to a real mass displacement, but to magnetisation inertia. Following the work performed initially by Gilbert in his search of a mechanical analogue for the macrospin dynamics, a generalised Gilbert equation was derived within the recent theory of mesoscopic nonequilibrium thermodynamics (MNET), accounting for macrospin's inertia. A new relaxation time τ depending on inertia and damping separated the behaviour of macrospin's dynamics into two regimes: the long time scale regime $t \gg \tau$, where the equation of Gilbert was found as a long time scale limit, and the short time scale regime $t \ll \tau$ where a new phenomenon was predicted, *nutaton*. The exact time scale of the new phenomenon couldn't be foretold as the values of the introduced inertia tensor are unknown. However, if the nutation phenomenon exists in macrospin dynamics, it will be observed for time scales shorter than the picosecond scale, for small values of the damping coefficient.

The magnetic moment \mathcal{M} of an electron is related to the value of its angular momentum \mathcal{L} by $\mathcal{M} = \gamma\mathcal{L}$, while the torque exerted on it by a magnetic field \mathbf{H} is $\mathcal{M} \times \mathbf{H}$. The magnetic moment precession is then given by: $\frac{d\mathcal{M}}{dt} = \gamma\mathcal{M} \times \mathbf{H}$. An analogue relation holds for the magnetisation \mathbf{M} in the absence of dissipation. While dealing with macrospin dynamics, T. L. Gilbert generalised this reasoning by replacing the field \mathbf{H} by an effective field including damping. The magnetisation dynamics would then account for damping through a reduced torque, but the definition of the angular momentum \mathbf{L} would still be the same: $\mathbf{M} = \gamma\mathbf{L}$.

It was shown that the introduction of inertia (an inertia tensor) in macrospin dynamics, gave a new definition for the angular momentum \mathbf{L} , that would account not only for damping, but also for the new phenomenon of nutation: $\mathbf{L} = \frac{\mathbf{M}}{\gamma} + \eta\tau\mathbf{M} \times \frac{d\mathbf{M}}{dt}$.

Still, at the scale of a nanostructure ferromagnet, thermal fluctuations play an important role. The macrospin undergoes a Brownian motion in the corresponding configuration space (sphere of radius M_s). The deterministic dynamic behaviour is described by the Gilbert equation, while fluctuations about the average are described by the Brown stochastic equation.

Within the nonequilibrium thermodynamics framework (NET), it was shown that the Brown stochastic equation can be derived in a rigorous scheme, with respect to the quite mathematical heavy scheme of stochastic processes. Meaning, the NET theory was applied to a statistical ensemble of macrospins $\mathbf{m} = M_s \mathbf{e}$ of constant modulus M_s and different orientations $\mathbf{e} = (\theta, \phi)$, described by a distribution function $n(\mathbf{e})$. The conservation law of this function n , together with a phenomenological expression for the probability (particle) flux lead to the desired Brown stochastic equation.

Furthermore, within the MNET theory applied to the case of macrospin dynamics, a true¹ Fokker-Planck equation was derived, a generalisation of Brown's stochastic equation. The generalised form accounts on two degrees of freedom: a faster kinetic degree, the angular momentum of the magnetisation \mathbf{L} and the slower degree, the orientation \mathbf{e} of the macrospins. A function $f(\mathbf{e}, \mathbf{L})$ describing the distribution of macrospins in the (\mathbf{e}, \mathbf{L}) phase space was introduced. The conservation law of this function written for the magnetisation-fixed reference frame (the rotating frame having as Oz axis the orientation of the magnetisation \mathbf{e}), together with the phenomenological expression of the 'collision' flux in the (\mathbf{e}, \mathbf{L}) space, lead to a 'true' Fokker-Planck equation for the magnetisation. It is shown that the Brown form dependent only on the orientation $\mathbf{e} = (\theta, \phi)$ of the macrospin is derived for long time scales $t \gg \tau$.

In the view of these results, of the generalised forms of the Gilbert and Brown equations derived within the simple, yet rigorous theory of MNET, the application of thermodynamics in spintronics arises as a question. A two rotating fluid model is proposed for the spin transfer problem as a perspective. Due to the large separation of time scales the conduction s -electron spins and the d -electron spins can be modelled as two rotating 'fluids' near the normal-ferromagnet interface, where spin-accumulation is found.

¹It is reminded that, in the field of "Statistical mechanics", the rotational Fokker-Planck equation derived by Brown is a particular case of a true Fokker-Planck equation and is commonly called the *Smoluchowski equation*. The name of 'Fokker-Planck equation' is generally given to an equation of a probability function f which is not only dependent on one slow variable, the position (our case - the orientation of the magnetization), but also on a faster degree of freedom, usually the velocity of a Brownian particle.