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## Spin transfer in an open ferromagnetic layer: from negative damping to effective temperature

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### Abstract

Spin transfer is a typical spintronics effect that allows a ferromagnetic layer to be switched by spin injection. All experimental results concerning spin transfer (quasi-static hysteresis loops or AC resonance measurements) are described on the basis of the Landau–Lifshitz–Gilbert equation of the magnetization, in which additional current dependent terms are added, like current dependent effective fields and current dependent damping factors, that can be positive or negative. The origin of these terms can be investigated further by performing stochastic experiments, like one-shot relaxation experiments under spin injection in the activation regime of the magnetization. In this regime, the Néel–Brown activation law is observed which leads to the introduction of a current dependent effective temperature. In order to define these counterintuitive parameters (effective temperature and negative damping), a detailed thermokinetic analysis of the different sub-systems involved is performed. This report presents a thermokinetic description of the different forms of energy exchanged between the electric and the ferromagnetic sub-systems at a normal/ferromagnetic junction.

The derivation of the Fokker–Planck equation in the framework of the thermokinetic theory allows the transport parameters to be defined from the entropy variation and refined with the Onsager reciprocity relations and symmetry properties of the magnetic system. The contribution of the spin polarized current is introduced as an external source term in the conservation laws of the ferromagnetic layer. Due to the relaxation time separation, this contribution can be reduced to an effective damping. The flux of energy transferred between the ferromagnet and the spin polarized current can be positive or negative, depending on the spin accumulation configuration. The effective temperature is deduced in the activation (stationary) regime, provided that the relaxation time that couples the magnetization to the spin polarized current is shorter than the relaxation to the lattice.

(Some figures in this article are in colour only in the electronic version)

In the context of spintronics, the electrical resistances of magnetic nanostructures are tuned with the magnetization states. Giant magnetoresistance (GMR), or anisotropic magnetoresistance (AMR) allows the magnetization states of nanolayers to be measured with great precision. Such magnetoresistances are scalable reading processes and are used for magnetic sensors and random access memory (MRAM) technology. The possibility of controlling the magnetic configuration of a magnetic nanostructure by injecting spins emerged only in recent studies, opening the way to a readily scalable writing process for MRAM application. This approach is also extended to thermally assisted switching, in which the heat flux is exploited in order to help the magnetization reversal. In order to control the magnetic configurations and their stabilities (for reading and writing processes), in such magnetic nanopillars, it is necessary to understand on one hand the processes responsible for the magnetization reversal (in the presence of a magnetic field and heat), and on the other hand, the processes governing spin dependent electronic transport at normal/ferromagnetic interfaces. Taken separately, the two effects are rather well understood today. However, coupling the two processes leads one to consider a large variety of possible mechanisms, called *spin transfer*, that involve an ensemble of non-equilibrium sub-systems in interaction, with different populations of electrons and different populations of spins. The present work tries to clarify this picture with a phenomenological analysis based on non-equilibrium thermodynamics of open systems.

Magnetization reversal provoked by spin injection has been observed in magnetic nanostructures of various morphologies, from spin valve multilayers [1–8] to nanowires [9–11] and point contacts [12–15], and different kinds of magnetic domain walls [16–24]. In order to describe and interpret these observations, physicists were forced to add one or two current dependent terms into the well known dynamical equations that describe a ferromagnetic layer coupled to a heat bath (Fokker–Planck or corresponding Landau–Lifshitz–Gilbert equations). However, the question remains open about the deterministic (e.g. spin torque) or stochastic (e.g. fluctuation and noise) nature of the terms to be added.

It has been observed that for a time window larger than the nanosecond timescale, and in the framework of one-shot measurements (i.e. non-averaged, or irreversible measurements), the magnetization reversal induced by spin injection is an activated process, with two-level fluctuations [25–28] or simple irreversible jumps [26, 29]. In these experiments, governed by stochastic fluctuations and noise, the observed effect is accounted for by a current dependent effective temperature in the Néel–Brown activation law [26]. In contrast, for quasi-static measurements (e.g. magnetoresistance measured as a function of the magnetic field or current with DC systems or a lock-in detection system) and for high frequency measurements, oscillations and resonances indicate, in the frequency domain, the manifestation of quasi-ballistic precession effects [15, 28, 30–33]. In these last experiments, the stochastic nature of the signal is reduced to line shapes of the resonance, and the behaviour is described in terms of current dependent effective fields and damping factors, within a generalized Landau–Lifshitz–Gilbert (LLG) equation. This deterministic formulation is motivated by the pioneering works of Berger [34] and Slonczewski [35] concerning spin torque theory. However, the deterministic approach cannot directly account for the magnetic relaxation measurements performed in the activation regime (as discussed in section 4.1 below). In contrast, the hypothesis used here is that of an open system.

In order to describe the open system composed by the ferromagnet and the spin injection at the interfaces, a detailed analysis of the different sub-systems is performed on the basis of thermokinetic theory [36–49]. The first step (the first section below) is to identify the relevant sub-systems of interest (pointing out the difference between the spin accumulation due to the diffusion of spin dependent conduction electrons at an interface, and the magnetization of a ferromagnetic layer), the coupling between them, and the role of the microscopic

degree of freedom that will be reduced to the action of the environment. In section 2, spin injection and spin dependent transport are described in the framework of the two-spin-channel approximation (a conduction channel that carries spin up and a conduction channel that carries spin down, defined by the conductivities). Giant magnetoresistance, spin accumulation, and corresponding entropy production, or heat transfer, are deduced. Beyond the two-spin-channel approximation, the analysis is extended to four channels with the introduction of two other electronic populations (typically s-like for conduction electrons and d-like for the ferromagnetic order parameter), and the relaxation between them. In the same manner as spin-flip scattering coupled the spin up and spin down channels, this relaxation defines a dissipative coupling between the ferromagnet and the spin dependent electric sub-systems. The third section is devoted to the detailed description of the ferromagnetic order parameter coupled to a heat bath (without spin injection). Both the rotational Fokker–Planck equation and the corresponding LLG equation are derived in the framework of the thermokinetic theory, i.e. with the help of the first two laws of thermodynamics and the Onsager reciprocity relations only. The coupling of the ferromagnetic order parameter to the heat bath is introduced via the chemical potential with a typical Maxwell–Boltzmann diffusion term including the temperature [36, 43]. The Néel–Brown law is deduced in the activation regime.

The last section is devoted to spin transfer, i.e. to the ferromagnetic Brownian motion activated by spin injection. The consequences of the Slonczewski spin torque hypothesis is first analysed as a deterministic term added to the Gilbert equation. A criterion for the validity of the spin torque approach is deduced. In contrast to the deterministic point of view, the open system approach is introduced with the contribution of s–d-like spin accumulation at the interfaces, as a source term in the conservation laws of the magnetization. Explicitly, it is shown that if  $n$  is the density of magnetic moments oriented in a given direction  $(\theta, \phi)$  of the unit sphere, and  $\vec{J}_M$  is the corresponding flux of magnetic moments (this flux is not a displacement in the usual space), the conservation of  $n$  can be written as  $\partial n / \partial t = -\text{div } \vec{J}_M + \int_{\text{N-F}} \dot{\Psi}(z) dz$ , where the divergence is defined on the sphere and  $\dot{\Psi}$  is the relaxation rate, integrated through the normal–ferromagnetic interfaces. This equation defines the irreversible spin transfer occurring in the ferromagnetic layer, taken as an open system. The relaxation rate is related to the spin accumulation  $\Delta\mu$  through an Onsager transport coefficient  $L$ ,  $\dot{\Psi} = L\Delta\mu$  (where  $\Delta\mu$  is proportional to the current).  $L$  is linked to the relaxation times through the charge conservation laws (or electric screening properties).

Due to the large relaxation time separation, the contribution of the source term can be reduced to the effect of an environment that is responsible for an *effective damping* and *effective fluctuations* (or effective temperature). The energy transferred between the ferromagnetic layer and the sub-system defined by the spin accumulation conduction electrons can be positive or negative, depending on the sign of the spin accumulation at the different interfaces. The effective temperature is deduced in the activation (stationary) regime, because the relaxation time that couples the magnetization to the spin polarized current short cuts the relaxation to the lattice.

## 1. Thermokinetic approach

### 1.1. Interacting sub-systems

The general scheme of the thermokinetic approach is described in [37, 39–41, 43]. The method consists in defining the state of the system with a set of the relevant extensive variables, say  $\{s, x_i\}$ , where  $x_i$  is, e.g., the density of particles in the sub-system  $i$ , or equivalently, the density of component  $i$  of a multicomponent fluid, and  $s$  is the total entropy density. The conservation

equations should then be written out, and the two laws of thermodynamics applied. The conservation equation for the component  $i$  can be written as

$$\frac{\partial n_i}{\partial t} = -\text{div}(\vec{J}_i) + \sum_j v_{ij} \dot{\Psi}_j. \quad (1)$$

The divergence of the current  $\vec{J}_i$  describes the conservative part of the process, and the term  $\dot{\Psi}_j$  is a source term that describes the relaxation of  $v_{ij}$  components  $i$  into the component  $j$  ( $v_{ij} \leq 0$ ), or inversely ( $v_{ij} \geq 0$ ).<sup>1</sup> It is proportional to the inverse of the relaxation time  $\dot{\Psi}_j \propto \tau^{-1}$  (see the appendix). Physically, the term  $\dot{\Psi}_j$  describes the relaxation process that changes the internal degree of freedom (e.g. spins, electric charges, internal configuration). In terms of chemical reactions,  $\dot{\Psi}_j$  is the velocity of the reaction, i.e. the generalized flux thermodynamically conjugated to the chemical affinity  $A_j$  (defined below). The summation over all sub-systems, or all components of the fluid, is that of a conserved variable:  $\sum_i \frac{\partial n_i}{\partial t} = -\text{div}(\sum_i \vec{J}_i)$ . The same holds, of course, for the energy  $E$ :  $\frac{\partial E}{\partial t} = -\text{div}(\vec{J}_E)$ , where  $J_E$  is the flux of energy. In contrast, the entropy production of the total system is not conservative in general, due to the irreversible processes (in other words, information is lost). The equation for the entropy production of the whole system takes the canonical form  $\frac{\partial s}{\partial t} = -\text{div}(\vec{J}_s) + \mathcal{I}$ , where  $J_s$  is the flux of entropy, and  $\mathcal{I}$  is the internal entropy production, or *irreversibility*, which is a consequence of the second law of thermodynamics:  $\mathcal{I} \geq 0$  (assuming  $T \geq 0$ ). According to the first law of thermodynamics, the energy  $E$  is a state function that is also scalar, extensive and conserved, so

$$\frac{\partial E(s, \{x_i\})}{\partial t} = \frac{\partial E}{\partial s} \frac{\partial s}{\partial t} + \sum_i \frac{\partial E}{\partial x_i} \cdot \frac{\partial x_i}{\partial t} \quad (2)$$

where  $\partial E/\partial s = T$  is the temperature,  $\partial E/\partial x_i \equiv F_i$  is the generalized force associated with the flux  $\partial x_i/\partial t$ . In the following we will deal exclusively with the chemical potentials  $\mu_i = \partial n_i/\partial E$ , unless otherwise specified (i.e. there is no need to introduce other extensive variables). The following Gibbs relation is obtained as a direct consequence of the first law:  $T \frac{\partial s}{\partial t} = \frac{\partial E}{\partial t} - \sum_i \frac{\partial n_i}{\partial t} \mu_i$ . After having inserted the conservation equations, equation (1), into (2), the following form is obtained<sup>2</sup>:

$$T \frac{\partial s}{\partial t} = -\text{div}(\vec{J}_E) + \sum_i \mu_i \text{div}(\vec{J}_i) - \sum_{ij} \mu_i v_{ij} \dot{\Psi}_j. \quad (3)$$

Using the development  $\text{div}(\mu_i \vec{J}_i) = \mu_i \text{div}(\vec{J}_i) + \vec{J}_i \cdot \vec{\text{grad}}(\mu_i)$ , equation (3) can be rewritten in the canonical form

$$\frac{\partial s}{\partial t} = -\text{div}(\vec{J}_s) + \mathcal{I} \quad (4)$$

where

$$\begin{aligned} \vec{J}_s &= \frac{1}{T} \vec{J}_E - \sum_i \frac{\mu_i}{T} \vec{J}_i \\ \mathcal{I} &= \vec{J}_E \cdot \vec{\text{grad}}\left(\frac{1}{T}\right) - \sum_i \vec{J}_i \cdot \vec{\text{grad}}\left(\frac{\mu_i}{T}\right) - \frac{1}{T} \sum_{ij} \mu_i v_{ij} \dot{\Psi}_j \end{aligned} \quad (5)$$

where the last term on the right-hand side defines the dissipative coupling between the sub-systems. As will be shown in the last section, *this term will be responsible for the irreversible*

<sup>1</sup> The coefficient  $v_{ij}$  is defined by the stoichiometric coefficients. If the chemical reaction  $i$  can be written as  $v'_{iA} A \rightarrow v''_{iA} A$ , with stoichiometric coefficients  $v'_{iA}$  and  $v''_{iA}$ , the coefficient  $v_{iA}$  appearing in the continuity equations is  $v_{iA} = v''_{iA} - v'_{iA}$ .

<sup>2</sup> In what follows, there is no kinetic energy, i.e. no inertial effects. This is why the equations are reduced to the simplest expression.

*spin transfer* effect described in this work. What is unusual in dealing with the second law is manipulation of an inequality ( $\mathcal{I} \geq 0$ ) instead of an equality, and consequently dealing with sufficient conditions instead of equivalences. Here, the condition  $\mathcal{I} \geq 0$  leads to introduction of a positive matrix  $\{\mathcal{L}_{ij}\}_{ij}$  of Onsager–Casimir transport coefficients that are state functions of the variables  $\{s, x_i\}$ , in order to build a positive quadratic form. The condition is fulfilled if the flux  $J_i$  and the relaxation velocity  $\dot{\Psi}_i$  have the form

$$\begin{aligned}\vec{J}_i &= - \sum_j \mathcal{L}_{ij} \vec{\text{grad}}(\mu_j) \\ \dot{\Psi}_i &= \sum_j L_{ij} A_j\end{aligned}\quad (6)$$

where

$$A_j \equiv - \sum_k v_{ik} \mu_k \quad (7)$$

is the chemical affinity of the corresponding reaction  $j$  (and we have  $A_j = -\partial E/\partial \Psi_j$ ) [50]. Furthermore, due to the time reversal symmetry of the microscopic equations, the transport coefficients follow the Onsager–Casimir reciprocity relations [51]. In the case of chemical reaction, the cross-coefficients that couple the flux  $\vec{J}$  to the relaxation process  $\dot{\Psi}$  are assumed to be zero, because, according to the Curie principle, only processes of identical tensorial nature are coupled. Note however that, if the relaxation rates describe a relaxation process like spin-flip relaxation, this may no longer be the case, and cross-coefficients may exist. This problem will be invoked in the last section of this work.

Assuming zero cross-coefficients, and inserting equation (6) into the continuity equation (1), we obtain an equation for the time variation of the density  $\partial n_i/\partial t$  in terms of derivatives of the chemical potentials  $\mu_j$ :

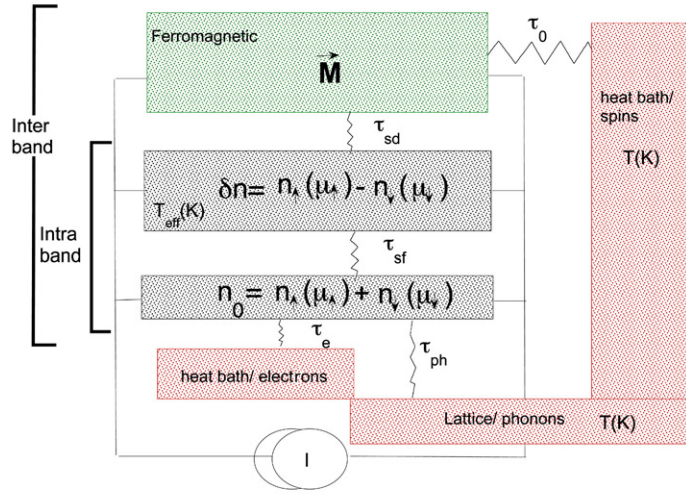
$$\frac{\partial n_i}{\partial t} = \sum_j \mathcal{L}_{ij} \nabla^2 \mu_j + \sum_{jk} v_{ij} L_{ik} A_k. \quad (8)$$

It is then sufficient to know the form of the chemical potential as a function of the density (for pure fluids:  $\mu(n_i) = \mu_0 + kT \ln(n_i)$ ) in order to derive the corresponding differential equation, or Fokker–Planck equation, with diffusion and relaxation terms (see sections 2, 3 and the appendix below).

What we gain in performing this analysis is the clear identification of the conservative and dissipative flux (through the internal entropy production), and the ability to define a dissipative process that couples the sub-systems beyond the usual deterministic coupling (electric field, magnetic field, pressure, etc). This dissipative coupling appears with an additional transport coefficient  $L$ , defined unequivocally via the transport equations. In the case studied below, the matrix  $\mathcal{L}$  is composed by the conductivities  $\sigma_i$  associated with each channel (i.e. associated with a given electronic population), the thermal conductivity, or the corresponding Seebeck (thermoelectric power) and Peltier coefficients [48, 52, 53] and the ferromagnetic transport coefficients: gyromagnetic ratio  $\Gamma$  and the Gilbert damping coefficient  $\eta$ . Beyond this, the flux of entropy or heat allows the *spin transfer* to be understood in an open system in terms of relaxation with a supplementary Onsager coefficient  $L$ . As shown in the last section, this term is responsible for an effective temperature  $T_{\text{eff}}$  and effective (negative) damping  $\alpha_{\text{eff}}$ .

## 1.2. The model

The model is based on the hypothesis that the ferromagnetic order parameter  $\vec{M}$  is well differentiated from the sub-system composed by spin polarized conduction electrons, although



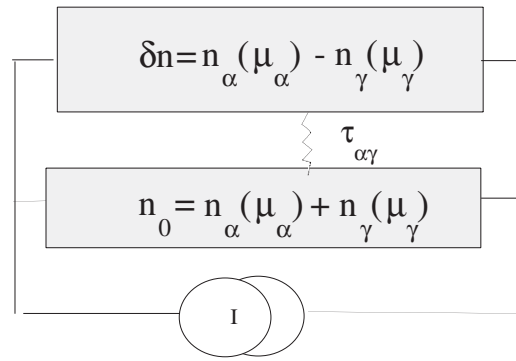
**Figure 1.** Thermokinetic picture of irreversible spin transfer. A ferromagnetic system (with magnetization  $M$ ), and an electric system with spin accumulation density  $\Delta n$  and electronic density at the Fermi level  $n_0$ . The chemical potential  $\mu$  is defined for each spin channel. The three sub-systems are coupled together through the relaxation times  $\tau_{sd}$  (interband s–d-like relaxation) and  $\tau_{sf}$  (intra band spin-flip relaxation). The sub-systems are also coupled to the current generator  $I$ , and to the heat reservoirs, through the corresponding well known relaxation times  $\tau_0$  (Néel–Brown waiting time),  $\tau_e$  and  $\tau_{ph}$ : elastic and inelastic electronic relaxation times.

the two systems exchange charges, spins, and heat through a relaxation mechanism that will be described in terms of internal variables [36, 40, 43]. As shown above, the relaxation of an internal variable (or internal degree of freedom) defines a transport coefficient  $L_{sd}$  related to the corresponding relaxation time  $\tau_{sd}$  ( $L_{sd} \propto \tau_{sd}^{-1}$ ; see the appendix for the relation to the relaxation time, and the electric screening properties).

We hence start with the two sub-systems: the ferromagnet described with the magnetization  $\vec{M}$  and the system of two conducting spin channels of the conduction electrons. The two sub-systems are dynamically coupled through the relaxation time  $\tau_{sd}$ . This relaxation is qualified as interband relaxation, as opposed to the intraband spin-flip relaxation  $\tau_{sf}$  introduced in the usual two-spin-channel approximation. The conducting channels are usually described in terms of the density  $n_{\uparrow}$  of conduction electrons with spin up and the density  $n_{\downarrow}$  of conduction electrons with spin down. The intraband coupling (accounted for by  $L_{sf}$  or  $\tau_{sf}$ ) is responsible for the spin accumulation mechanism for the stationary regime. For convenience, we redefine the two channels with the density of spin polarized electrons  $\Delta n = n_{\uparrow} - n_{\downarrow}$  (‘spin conduction channel’) and the total density of electrons  $n_0 = n_{\uparrow} + n_{\downarrow}$ .

Furthermore, the conduction channels are contacted to a power supply (current generator here). Strictly speaking, the magnetic system is also contacted to the power supply, e.g. through the electron of d character [55]. The conduction electrons thermalize each other through a well known mechanism of elastic scattering  $\tau_e$  (that defines the conduction electron reservoir), at the femtosecond timescale (or below), and are also contacted to the lattice through the Fermi–Dirac distribution, and inelastic scattering  $\tau_{ph}$ . On the other hand, the ferromagnetic order parameter is contacted to the lattice with a well known relaxation time  $\tau_0$  that is measured in ferromagnetic resonance (FMR) experiments, and is typically of the order of the nanosecond (or a few hundreds of picoseconds). This description leads to the model depicted in figure 1.





**Figure 2.** Two-channel model, including relaxation that couples the two electronic populations.

The basic idea developed below rests on the fact that the typical timescales of the dynamics of the two sub-systems are greatly separated. There is a slow variable, the magnetization, and fast variables, the degrees of freedom related to the spins of the conduction electrons. It is then possible to reduce the action of the fast variable to the role of an environment as regards to the magnetization, like for spin bath relaxation. The effect of the coupling to the spin dependent electronic sub-system will then be reduced to specific damping and fluctuation terms added to the usual stochastic equations for the magnetization. This will be our line of reasoning followed in the last section, after describing the two sub-systems.

## 2. Spin dependent transport

In order to explain the high resistance and the high thermoelectric power observed in transition metals, Mott introduced the concept of spin polarized current and suggested that s–d interband scattering plays an essential role in the conduction properties [54]. This approach in terms of two conduction bands [55] explained the existence of a spin polarized current in the 3d ferromagnetic materials [56], and was used for the description of anisotropic magnetoresistance (AMR) [57, 58], the description of a spin polarizer [59], and thermoelectric power [60]. With the discovery of giant magnetoresistance (GMR) [61] and related effects [62] (like domain wall scattering [63–66] discussed below), the development of spintronics focused the discussion on spin-flip scattering occurring between spin polarized conducting channels [67–72]. The two-channel model, which describes the conduction electrons with majority and minority spins, is applied with great efficiency to GMR and spin injection effects [47, 73–78], including metal/semiconductor [79] and metal/superconductor interfaces [80]. In this context, it is sufficient to describe the diffusion process in terms of spin-flip scattering without the need to invoke interband s–d scattering.

It is convenient to generalize the two-spin-channel approach to any relevant transport channels, i.e. to any distinguishable electron populations  $\alpha$  and  $\gamma$  defined by an internal degree of freedom (see figure 2). The local out-of-equilibrium state near the junction is then described with a non-vanishing chemical potential difference between these two populations:  $\Delta\mu_{\alpha\gamma} = \mu_{\alpha} - \mu_{\gamma} \neq 0$ . In other words, assuming that the presence of a junction induces a deviation from the local equilibrium, the  $\alpha$  and  $\gamma$  populations can be *defined by the  $\alpha \rightarrow \gamma$  relaxation mechanism* itself, that allows the local equilibrium to be recovered in the bulk material ( $\lim_{z \rightarrow \pm\infty} \Delta\mu(z) = 0$ ) [47]. Such considerations have been presented in some important spintronics studies on the basis of microscopic calculations [54, 57, 58, 67, 69, 77, 78, 81–83].



The thermokinetic approach allows us to deal with interband relaxation on an equal footing with spin-flip relaxation, with the help of the transport coefficients only. For this purpose, the two-spin-channel model is generalized, with the introduction of the corresponding transport coefficients: the conductivities  $\sigma_\alpha$  and  $\sigma_\gamma$  of each channel define the total conductivity  $\sigma_t = \sigma_\alpha + \sigma_\gamma$  and the conductivity asymmetry  $\beta = (\sigma_\alpha - \sigma_\gamma)/\sigma_t$ ; the relaxation between the two channels is described using the parameter  $L$  (or equivalently, the relevant relaxation times  $\tau_{\gamma \leftrightarrow \alpha}$ ).

### 2.1. The generalized two-channel model

In the framework of the two-conducting-channel model, which includes relaxation from one channel to the other, it is easy to follow step by step the method described in the first section. The conservation laws can be written as (assuming a 1D space variable  $z$ )

$$\begin{aligned}\frac{\partial n_\alpha}{\partial t} &= -\frac{\partial J_\alpha}{\partial z} - \dot{\Psi}_{\alpha\gamma} \\ \frac{\partial n_\gamma}{\partial t} &= -\frac{\partial J_\gamma}{\partial z} + \dot{\Psi}_{\alpha\gamma}\end{aligned}\quad (9)$$

where  $n_\alpha$  and  $n_\gamma$  are the densities of particles in the channels  $\{\alpha, \gamma\}$ .

The entropy variation can be written as

$$T\mathcal{I} = -J_\alpha \frac{\partial \mu_\alpha}{\partial z} - J_\gamma \frac{\partial \mu_\gamma}{\partial z} - \dot{\Psi}_{\alpha\gamma}(\mu_\alpha - \mu_\gamma) \quad (10)$$

and the application of the second law of thermodynamics leads to introduction of the Onsager coefficients  $\sigma_\alpha \geq 0$ ,  $\sigma_\gamma \geq 0$ , and  $L \geq 0$  [47], such that

$$\begin{aligned}J_\alpha &= -\frac{\sigma_\alpha}{e} \frac{\partial \mu_\alpha}{\partial z} \\ J_\gamma &= -\frac{\sigma_\gamma}{e} \frac{\partial \mu_\gamma}{\partial z} \\ \dot{\Psi}_{\alpha\gamma} &= L(\mu_\alpha - \mu_\gamma)\end{aligned}\quad (11)$$

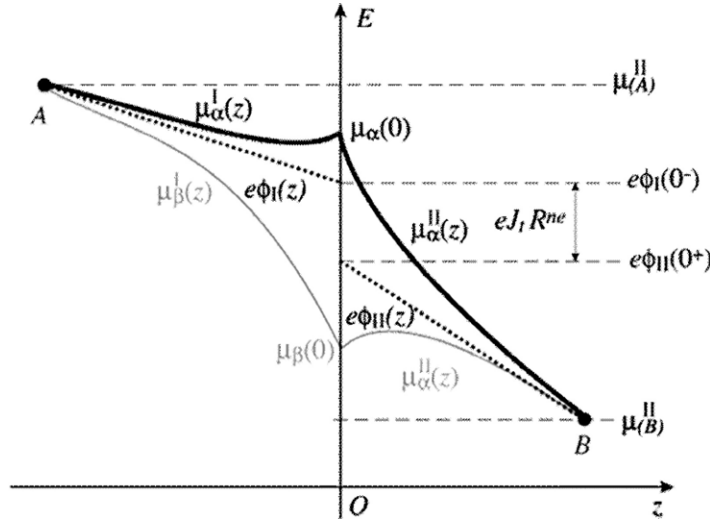
where  $\dot{\Psi}_{\alpha\gamma}$  describes the relaxation from the channel  $\alpha$  to the other channel  $\gamma$  in terms of the velocity of the reaction  $\alpha \rightarrow \gamma$ . It is not necessary, in what follows, to distinguish between the electric part and the pure chemical part of the electrochemical potentials (see [84]). The effects of the electric charge distribution are described in the appendix, with the introduction of the screening length  $l$  and the relation to the relaxation times. As shown in the appendix, the Onsager coefficient  $L$  is inversely proportional to the electronic relaxation times  $\tau_{\alpha \leftrightarrow \gamma}$ :

$$L \propto \left( \frac{g}{\tau_{\alpha \rightarrow \gamma}} + \frac{f}{\tau_{\gamma \rightarrow \alpha}} \right) \quad (12)$$

where  $f$  and  $g$  are two functions close to unity, and related to the electric charge distributions (see the appendix). Note that due to our definition of  $\mu_\alpha$  and  $\mu_\gamma$ , there is no direct coupling between the two channels: there is no transport coefficient that couples the first two equations in (11). This is a consequence of the definition of the electronic populations, or channels, through the relaxation process itself (the populations are stable if  $\dot{\Psi} = 0$ ). In the context presented here, the term for spin mixing, often used in spintronics, should then refer to the relaxation process  $\dot{\Psi}$  (spin-flip or s-d relaxation), and not to the existence of cross-coefficients in equation (11), as introduced in [53].

The total current  $J_t$  is constant:

$$J_t = J_\alpha + J_\gamma = -\frac{1}{e} \frac{\partial}{\partial z} (\sigma_\alpha \mu_\alpha + \sigma_\gamma \mu_\gamma). \quad (13)$$



**Figure 3.** Junction between two layers I and II. The chemical potential profile over the interval  $[A, B]$  in the  $\alpha$  and  $\gamma$  channels. The  $A$  and  $B$  points verify  $\mu_\alpha(A) = \mu_\gamma(A)$  and  $\mu_\alpha(B) = \mu_\gamma(B)$ . The two straight lines represent the  $\Phi$  variation in each region ( $\Phi_I, \Phi_{II}$ ). It can be directly seen that the out-of-equilibrium resistance  $R^{ne}$  is determined by the  $\Phi$  discontinuity at the interface.

However, it is not possible to measure separately the different conduction channels, since any realistic electric contact short cuts the two channels. What is measured is necessarily the usual Ohm's law,  $J_t = -\sigma_t \frac{\partial \Phi}{\partial z}$ , that imposes the reference electric potential  $\Phi$  to be introduced, together with the total conductivity  $\sigma_t = \sigma_\alpha + \sigma_\gamma$ .<sup>3</sup> The potential  $\Phi$  is hence

$$e\Phi = \frac{1}{\sigma_t}(\sigma_\alpha \mu_\alpha + \sigma_\gamma \mu_\gamma). \quad (14)$$

Let us assume that the two channels collapse to a unique conduction channel for a specific configuration, the reference, which is a local equilibrium situation:  $\Delta\mu_{eq} = 0$ . The out-of-equilibrium contribution to the resistance,  $R^{ne}$ , is calculated through the relation

$$J_t e R^{ne} = \int_A^B \frac{\partial}{\partial z}(\mu_\alpha - e\Phi(z)) dz = \int_A^B \frac{\partial}{\partial z}(\mu_\gamma - e\Phi(z)) dz, \quad (15)$$

so

$$R^{ne} = -\frac{1}{J_t e} \int_A^B \frac{\sigma_\alpha - \sigma_\gamma}{2\sigma_t} \frac{\partial \Delta\mu}{\partial z} dz \quad (16)$$

where the measurement points  $A$  and  $B$  are located far enough from the interface (inside the bulk) that  $\Delta\mu(A) = \Delta\mu(B) = 0$  (see figure 3). The integration in equation (15) is performed over the regular part of the function only ( $\Phi$  and  $\sigma_i$  are discontinuous)<sup>4</sup>. Equation (16) allows

<sup>3</sup> In all these calculations, the channel parameters  $\sigma_{\alpha\gamma}, \beta$ , and  $L$  are considered as constant in space. It can be shown that taking into account the gradient only introduces vanishingly small corrections.

<sup>4</sup> The Riemann integral is evaluated in the intervals where the derivative is regular. Note that a global calculation over  $[A, B]$ , within the framework of the distribution theory, yields zero because  $\Delta\mu(A) = \Delta\mu(B) = 0$ . This simply means that the integral of the regular part is opposite to the integral of the Dirac masses located at the interface. Let us note that the out-of-equilibrium resistance is straightforwardly connected to the chemical potential drop between the  $A$  and  $B$  points, diminished by the standard potential drop which would result from the application of Ohm's law. We obtain  $J_t e R^{ne} = [\mu_I(A) - \mu_{II}(B)] - e[\Phi_I(A) - \Phi_I(0^-)] - e[\Phi_{II}(0^+) - \Phi_{II}(B)] = [\mu_I(A) - e\Phi_I(A)] - [\mu(0) - e\Phi_I(0^-)] + [\mu(0) - e\Phi_{II}(0^+)] - [\mu_{II}(B) - e\Phi_{II}(B)] = e[\Phi_I(0^-) - \Phi_{II}(0^+)]$ . It can be seen that the second expression for  $J_t e R^{ne}$  above is nothing but the opposite of equation (6), and is equal to the discontinuity of  $\Phi$  at the interface, which provides a simple physical interpretation as illustrated in figure 3.

the out-of-equilibrium resistance at a simple junction between two layers (composed of the layers I and II) to be easily calculated. If the junction is set at  $z = 0$  and the conductivities are respectively  $\sigma_i^I$  and  $\sigma_i^{II}$  ( $i = \{\alpha, \gamma\}$ ), we have

$$J_{Te} R^{ne} = \int_A^0 \frac{\sigma_\alpha^I - \sigma_\gamma^I}{2\sigma_t} \frac{\partial \Delta\mu^I}{\partial z} dz + \int_0^B \frac{\sigma_\alpha^{II} - \sigma_\gamma^{II}}{2\sigma_t} \frac{\partial \Delta\mu^{II}}{\partial z} dz. \quad (17)$$

The equilibrium is recovered in the bulk, so

$$R^{ne} = \left( \frac{\sigma_\alpha^I - \sigma_\gamma^I}{\sigma_t^I} - \frac{\sigma_\alpha^{II} - \sigma_\gamma^{II}}{\sigma_t^{II}} \right) \frac{\Delta\mu(0)}{2J_{Te}}. \quad (18)$$

The chemical potential difference  $\Delta\mu(z)$ , which accounts for the pumping force opposed to the relaxation  $\alpha \rightarrow \gamma$ , is obtained by solving the diffusion equation deduced from equations (11) and (9), and assuming a stationary regime for each channel,  $\frac{\partial n_\alpha}{\partial t} = \frac{\partial n_\gamma}{\partial t} = 0$  [47, 73–76]:

$$\frac{\partial^2 \Delta\mu(z)}{\partial z^2} = \frac{\Delta\mu(z)}{l_{\text{diff}}^2} \quad (19)$$

where

$$l_{\text{diff}}^{-2} = eL(\sigma_\alpha^{-1} + \sigma_\gamma^{-1}) \quad (20)$$

is the diffusion length related to the  $\alpha \rightarrow \gamma$  relaxation.

At the interface ( $z = 0$ ), the continuity of the currents for each channel can be written as  $J_\alpha^I(0) = J_\alpha^{II}(0)$ , where

$$J_\alpha(0) = -\frac{\sigma_\alpha \sigma_\gamma}{e\sigma_t} \frac{\partial \Delta\mu}{\partial z} + \frac{\sigma_\alpha}{\sigma_t} J_t \quad (21)$$

which leads to the general relation:

$$\Delta\mu(0) = \left( \frac{\sigma_\alpha^I}{\sigma_t^I} - \frac{\sigma_\alpha^{II}}{\sigma_t^{II}} \right) \left( \frac{\sigma_\alpha^I \sigma_\gamma^I}{\sigma_t^I l_{\text{diff}}^I} + \frac{\sigma_\alpha^{II} \sigma_\gamma^{II}}{\sigma_t^{II} l_{\text{diff}}^{II}} \right)^{-1} eJ_t. \quad (22)$$

Inserting equation (22) into (18), we obtain the general expression for the out-of-equilibrium resistance (per unit area) produced by the  $\alpha \rightarrow \gamma$  relaxation mechanism at a junction:

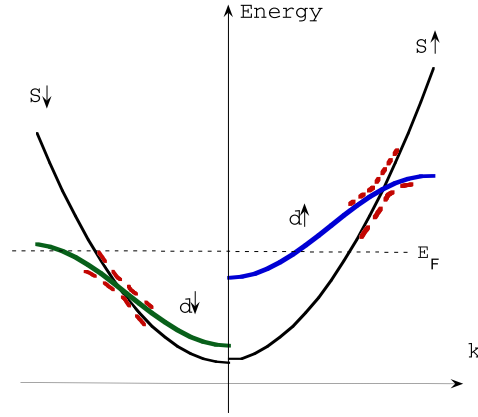
$$R^{ne} = \left( \frac{\sigma_\alpha^I - \sigma_\gamma^I}{2\sigma_t^I} - \frac{\sigma_\alpha^{II} - \sigma_\gamma^{II}}{2\sigma_t^{II}} \right) \left( \frac{\sigma_\alpha^I}{\sigma_t^I} - \frac{\sigma_\alpha^{II}}{\sigma_t^{II}} \right) \left( \sqrt{\frac{\sigma_\alpha^I \sigma_\gamma^I eL^I}{\sigma_t^I}} + \sqrt{\frac{\sigma_\alpha^{II} \sigma_\gamma^{II} eL^{II}}{\sigma_t^{II}}} \right)^{-1} \quad (23)$$

where we have used the relation

$$l_{\text{diff}}^{-1} = 2\sqrt{\frac{eL}{\sigma_t(1-\beta^2)}}. \quad (24)$$

It is convenient to describe the conductivity asymmetry using a parameter  $\beta$  such that  $\sigma_\alpha = \sigma_t(1+\beta)/2$  and  $\sigma_\gamma = \sigma_t(1-\beta)/2$ . The out-of-equilibrium contribution to the resistance then takes the following form:

$$R^{ne} = \frac{1}{2} \frac{(\beta_I - \beta_{II})^2}{\sqrt{eL^I \sigma_t^I (1-\beta_I^2)} + \sqrt{eL^{II} \sigma_t^{II} (1-\beta_{II}^2)}}. \quad (25)$$



**Figure 4.** Generic band structure for a 3d ferromagnet with s and d bands schematized for an arbitrary direction of the wavevector  $k$ . The shift between the two d bands for the two spin carriers up and down is exemplified. The hybridized zone is schematized with dotted lines at the junction between s and d bands. At the Fermi level four different electronic populations can be identified.

In the case of the sub-system described in terms of two *spin* channels, the relaxation  $\dot{\Psi}_{\uparrow\downarrow}$  leads to a *spin accumulation* effect  $\Delta\mu_{\uparrow\downarrow}$  at the interface of two identical ferromagnets with an antiparallel configuration. The corresponding resistance contribution is

$$R_{sa}^{\uparrow\downarrow} = \frac{\beta_s^2}{\sigma_t(1 - \beta_s^2)} l_{sf} = \frac{\beta_s^2}{\sqrt{eL\sigma_t(1 - \beta_s^2)}}. \quad (26)$$

This expression is the well known giant magnetoresistance contribution [47, 73–76, 85, 86].

## 2.2. The four-channel approximation

In the previous subsections, two different electronic relaxation mechanisms have been invoked separately in order to describe giant magnetoresistance or anisotropic magnetoresistance. It is clear however that the two relaxations would take place in parallel, leading to a more complex redistribution of spins within the different channels. In the present subsection, we consider a system in which the two mechanisms coexist, leading to a four-channel model [49].

The generic band structure (energy as a function of wavevector  $\vec{k}$  for a given direction) of a 3d ferromagnet is schematized in figure 4. The band s is parabolic and the exchange splitting is very small. In contrast, the d bands are strongly shifted between up and down spin carriers. The hybridized zone is schematized by the dotted lines at the intersection.

The system is composed of the reservoirs of the injected s electrons and the ferromagnetic layer composed of the d electrons. At the interface, current injection leads to a redistribution of the different electronic populations that are governed by spin polarization and charge conservation laws. Let us assume that the current injected is spin polarized in the down polarization ( $\downarrow$ ). The conservation laws should be written out taking into account the reaction mechanisms between the different populations. At short timescales (electronic scattering) the relaxation channels are assumed to be the following four:

- (I)  $e_{s\downarrow} \rightarrow e_{d\downarrow}$  (spin-conserved s–d scattering);
- (II)  $e_{s\downarrow} \rightarrow e_{s\uparrow}$  (spin-flip scattering for the s population);
- (III)  $e_{s\downarrow} \rightarrow e_{d\uparrow}$  (spin-flip s–d scattering);
- (IV)  $e_{d\downarrow} \rightarrow e_{d\uparrow}$  (spin-flip scattering for the d population).

Process (I) is assumed to be the main mechanism responsible for anisotropic magnetoresistance (AMR). Process (II) leads to the well known spin accumulation effect and was also described in detail in the first subsections. *According to the fact that the majority-spin  $d$  band is full* and lies at a sizable energy below the Fermi level, the current  $J_{d\uparrow}$  is negligible and the channel  $d\uparrow$  is frozen. Processes (III) and (IV) are hence negligible [59]. Consequently, we are dealing with a three-channel model  $\{s\uparrow, s\downarrow, d\downarrow\}$ .

The total current  $J_t$  is composed of the three currents for each channel:  $J_t = J_{s\uparrow} + J_{s\downarrow} + J_{d\downarrow}$ . In order to write out the conservation laws, the relaxation rate  $\dot{\Psi}_{sd}$  is introduced to account for s–d spin-conserved scattering, and the relaxation rate  $\dot{\Psi}_s$  is introduced in order to account for spin-flip scattering. *Assuming that all channels are in a steady state* (this condition will relax in the last section, where the magnetic system is coupled to the channels  $d\downarrow$ ),

$$\begin{aligned} \frac{\partial n_t}{\partial t} &= -\frac{\partial J_t}{\partial z} = 0 \\ \frac{\partial n_{s\uparrow}}{\partial t} &= -\frac{\partial J_{s\uparrow}}{\partial z} - \dot{\Psi}_s = 0 \\ \frac{\partial n_{s\downarrow}}{\partial t} &= -\frac{\partial J_{s\downarrow}}{\partial z} - \dot{\Psi}_{sd} + \dot{\Psi}_s = 0 \\ \frac{\partial n_{d\downarrow}}{\partial t} &= -\frac{\partial J_{d\downarrow}}{\partial z} + \dot{\Psi}_{sd} = 0 \end{aligned} \quad (27)$$

where  $n_t, n_{s\uparrow}, n_{s\downarrow}, n_{d\downarrow}$  are respectively the total densities of particles and the density of particles in the  $s\uparrow, s\downarrow, d\downarrow$  channels. The system is described in terms of the number of electrons present in each channel at a given time, that defines the four currents, plus the entropy of the system. The conjugate (intensive) variables are the chemical potentials  $\{\mu_{s\uparrow}, \mu_{s\downarrow}, \mu_{d\uparrow}, \mu_{d\downarrow}\}$ . The application of the first and second laws of thermodynamics [47] allows us to deduce the Onsager relations of the system:

$$\begin{aligned} J_{s\downarrow} &= -\frac{\sigma_{s\downarrow}}{e} \frac{\partial \mu_{s\downarrow}}{\partial z} \\ J_{s\uparrow} &= -\frac{\sigma_{s\uparrow}}{e} \frac{\partial \mu_{s\uparrow}}{\partial z} \\ J_{d\downarrow} &= -\frac{\sigma_{d\downarrow}}{e} \frac{\partial \mu_{d\downarrow}}{\partial z} \\ \dot{\Psi}_{sd} &= L_{sd} (\mu_{s\downarrow} - \mu_{d\downarrow}) \\ \dot{\Psi}_s &= L_s (\mu_{s\uparrow} - \mu_{s\downarrow}) \end{aligned} \quad (28)$$

where the conductivity of each channel  $\{\sigma_{s\uparrow}, \sigma_{s\downarrow}, \sigma_{d\uparrow}, \sigma_{d\downarrow}\}$  has been introduced. The first four equations are nothing but Ohm's law applied to each channel, and the two last equations introduce new Onsager transport coefficients,  $L_{sd\downarrow}$  and  $L_s$ , that respectively describe the s–d relaxation (I) for minority spins under the action of the chemical potential difference  $\Delta\mu_{\downarrow} = \mu_{s\downarrow}/2 - \mu_{d\downarrow}$  and the spin-flip relaxation (II) under spin pumping  $\Delta\mu_s = \mu_{s\uparrow} - \mu_{s\downarrow}/2$ . According to the appendix, the Onsager coefficients are proportional to the corresponding relaxation times.

For convenience, we define the usual charge current  $J_{0s} = J_{s\uparrow} + J_{s\downarrow}$ , the minority-spin current  $J_{0\downarrow} = J_{s\downarrow} + J_{d\downarrow}$ , and the two *polarized currents*  $\delta J_{\downarrow} = J_{s\downarrow} - J_{d\downarrow}$  and  $\delta J_s = J_{s\uparrow} - J_{s\downarrow}$ . We introduce the  $\sigma_s$  and  $\sigma_{\downarrow}$  conductivities  $\{\sigma_s = \sigma_{s\uparrow} + \sigma_{s\downarrow}$  and  $\sigma_{\downarrow} = \sigma_{s\downarrow} + \sigma_{d\downarrow}\}$ . The conductivity imbalances  $\beta_{\downarrow}$  and  $\beta_s$  between respectively the  $s\downarrow$  and  $d\downarrow$  channels and the  $s\uparrow$  and  $s\downarrow$  channels are

$$\begin{aligned} \beta_{\downarrow} &= \frac{\sigma_{s\downarrow} - \sigma_{d\downarrow}}{\sigma_{\downarrow}} \\ \beta_s &= \frac{\sigma_{s\uparrow} - \sigma_{s\downarrow}}{\sigma_s}. \end{aligned} \quad (29)$$

Equation (27) becomes

$$\begin{aligned}
\frac{\partial J_t}{\partial z} &= \frac{\partial J_{d\downarrow}}{\partial z} + \frac{\partial J_s}{\partial z} = 0 \\
\frac{\partial J_{0\downarrow}}{\partial z} &= \dot{\psi}_s \\
\frac{\partial \delta J_{\downarrow}}{\partial z} &= -2\dot{\psi}_{sd} - \dot{\psi}_s \\
\frac{\partial J_{0s}}{\partial z} &= -\dot{\psi}_{sd} \\
\frac{\partial \delta J_s}{\partial z} &= \dot{\psi}_{sd} - 2\dot{\psi}_s
\end{aligned} \tag{30}$$

and, defining the quasi-chemical potentials  $\mu_s = \mu_{s\uparrow} + \mu_{s\downarrow}/2$  and  $\mu_{\downarrow} = \mu_{s\downarrow}/2 + \mu_{d\downarrow}$ , equation (28) becomes

$$\begin{aligned}
J_{0\downarrow} &= -\frac{\sigma_{\downarrow}}{2e} \left( \frac{\partial \mu_{\downarrow}}{\partial z} + \beta_{\downarrow} \frac{\partial \Delta \mu_{\downarrow}}{\partial z} \right) \\
\delta J_{\downarrow} &= -\frac{\sigma_{\downarrow}}{2e} \left( \beta_{\downarrow} \frac{\partial \mu_{\downarrow}}{\partial z} + \frac{\partial \Delta \mu_{\downarrow}}{\partial z} \right) \\
J_{0s} &= -\frac{\sigma_s}{2e} \left( \frac{\partial \mu_s}{\partial z} + \beta_s \frac{\partial \Delta \mu_s}{\partial z} \right) \\
\delta J_s &= -\frac{\sigma_s}{2e} \left( \beta_s \frac{\partial \mu_s}{\partial z} + \frac{\partial \Delta \mu_s}{\partial z} \right) \\
\dot{\Psi}_{sd} &= L_{sd} \Delta \mu_{\downarrow} \\
\dot{\Psi}_s &= L_s \Delta \mu_s.
\end{aligned} \tag{31}$$

The equations of conservation (equation (30)) and the above Onsager equations lead to the two coupled diffusion equations:

$$\begin{aligned}
\frac{\partial^2 \Delta \mu_{\downarrow}}{\partial z^2} &= \frac{1}{l_{sd}^2} \Delta \mu_{\downarrow} - \frac{1}{\lambda_s^2} \Delta \mu_s \\
\frac{\partial^2 \Delta \mu_s}{\partial z^2} &= \frac{1}{\lambda_{sd}^2} \Delta \mu_{\downarrow} - \frac{1}{l_{sf}^2} \Delta \mu_s
\end{aligned} \tag{32}$$

where

$$\begin{aligned}
l_{sd} &\equiv \sqrt{\frac{\sigma_{\downarrow} (1 - \beta_{\downarrow}^2)}{4 e L_{sd}}} \\
\lambda_s &\equiv \sqrt{\frac{\sigma_{\downarrow} (1 + \beta_{\downarrow})}{2 e L_s}} \\
l_{sf} &\equiv \sqrt{\frac{\sigma_s (1 - \beta_s^2)}{4 e L_s}} \\
\lambda_{sd} &\equiv \sqrt{\frac{\sigma_s (1 - \beta_s)}{2 e L_{sd}}}.
\end{aligned} \tag{33}$$

A solution of equations (32) is

$$\begin{aligned}
\Delta \mu_{\downarrow} &= \Delta \mu_1 + \Delta \mu_2 \\
\Delta \mu_s &= \lambda_s^2 \left( \left( \frac{1}{l_{sd}^2} - \frac{1}{\Lambda_+^2} \right) \Delta \mu_1 + \left( \frac{1}{l_{sd}^2} - \frac{1}{\Lambda_-^2} \right) \Delta \mu_2 \right)
\end{aligned} \tag{34}$$

with

$$\begin{aligned}\Delta\mu_1 &= a_1 e^{\frac{z}{\Lambda_+}} + a_2 e^{-\frac{z}{\Lambda_+}} \\ \Delta\mu_2 &= b_1 e^{\frac{z}{\Lambda_-}} + b_2 e^{-\frac{z}{\Lambda_-}}\end{aligned}\quad (35)$$

where

$$\Lambda_{\pm}^{-2} = \frac{1}{2}(l_{sd}^{-2} + l_{sf}^{-2}) \left( 1 \pm \sqrt{1 - 4 \frac{l_{sd}^{-2} l_{sf}^{-2} - \lambda_s^{-2} \lambda_{sd}^{-2}}{(l_{sd}^{-2} + l_{sf}^{-2})^2}} \right).$$

The constants  $a_1$ ,  $a_2$ ,  $b_1$ ,  $b_2$  are defined by the boundary conditions. It can then be seen that the usual spin accumulation corresponding to  $\Delta\mu_s$  also depends on the spin-conserved s–d electronic diffusion which is known to be efficient [59] and, conversely, that spin-conserved diffusion is able to lead to a spin accumulation, or d *spin accumulation* effects. Accordingly, we expect to measure some typical effects related to spin accumulation in single magnetic layers, or if  $\beta_s = 0$ : this point will be illustrated in the new expression for the magnetoresistance (equation (39) below), and in section 4 through the effect of current induced magnetization switching (CIMS). s–d relaxation adds a new contribution to the resistance, which plays the role of an interface resistance arising from the diffusive treatment of the band mismatch [67–69].

The resistance produced by the usual spin accumulation contribution and the contribution of s–d relaxation are defined (see equation (16)) by

$$R_{sa} = \frac{-1}{eJ_t} \int_B^A \frac{\partial}{\partial z} (\mu_i - \Phi(z)) dz \quad (36)$$

where  $\Phi(z)$  is the total electric field and  $\mu_i$  is one of the chemical potentials. Provided that the total current is  $J_t = J_{s\uparrow} + J_{s\downarrow} + J_{d\downarrow}$ , or

$$J_t = -\frac{\sigma_t}{e} \frac{\partial}{\partial z} \left( \frac{\sigma_{d\downarrow}}{\sigma_t} \mu_{d\downarrow} + \frac{\sigma_{s\downarrow}}{\sigma_t} \mu_{s\downarrow} + \frac{\sigma_{s\uparrow}}{\sigma_t} \mu_{s\uparrow} \right), \quad (37)$$

the total electric field can also be written (from equation (28)) as

$$\Phi(z) = \frac{J_t}{\sigma_t} = -\frac{1}{e} \left( \frac{\sigma_{d\downarrow}}{\sigma_t} \frac{\partial \mu_{d\downarrow}}{\partial z} + \frac{\sigma_{s\downarrow}}{\sigma_t} \frac{\Delta \mu_{\downarrow}}{\partial z} + \frac{\sigma_{s\uparrow}}{\sigma_t} \frac{\Delta \mu_s}{\partial z} \right) \quad (38)$$

where  $\sigma_t = \sigma_{s\uparrow} + \sigma_{s\downarrow} + \sigma_{d\downarrow}$ . The resistance is given by

$$R_{sa} = -\frac{1}{eJ_t} \int_A^B \left( \frac{\sigma_{s\downarrow}}{\sigma_t} \frac{\partial \Delta \mu_{\downarrow}}{\partial z} + \frac{\sigma_{s\uparrow}}{\sigma_t} \frac{\partial \Delta \mu_s}{\partial z} \right) dz. \quad (39)$$

This three-channel model brings to light the interplay between band mismatch effects and spin accumulation, in a diffusive approach. It is interesting to note that the local neutrality charge condition which is often used (see for instance equation (4) in [87]) was not included, as described in the appendix. On the contrary, we have imposed the conservation of the current at any point of the conductor. Indeed, electron transfer from one channel to another where the electron mobility is different induces a local variation of the total current.

The resolution of the coupled diffusion equations is discussed elsewhere [49].

### 2.3. Domain wall scattering

In the description given so far, the spin quantification axis that defines up and down spin states was fixed through the whole structure (i.e. through the layers and the interfaces). Provided that the spin quantification axis follows the direction of the magnetization, it could be non-uniform throughout a ferromagnetic layer, or crossing an interface. This is especially the case in the presence of a magnetic domain wall. In a thin enough magnetic domain wall the spin



would not follow the quantification axis adiabatically, leading to spin dependent domain wall scattering (DWS) [63–66]. This effect has been investigated intensively in the last few decades in various structures [72]. The underlying idea is however rather simple, and can be formulated easily with a generalization of the two-spin-channel approach. For the sake of simplicity, this generalization will be performed only for the two electronic populations  $\{\alpha, \gamma\}$ .

Like in [47], we start with the conservation of the particles for the two channels, in a discrete model. The system is described as a layer  $\Sigma_k$  in contact with a left layer  $\Sigma_{k-1}$  and a right layer  $\Sigma_{k+1}$ . The spin-flip scattering introduced in the previous sections is described by the reaction rate  $\dot{\Psi}^k$ . A probability  $(1 - \Delta\epsilon(k))$  of spin-flip alignment along the quantification axis is introduced. In the case of ballistic alignment  $(1 - \Delta\epsilon(k)) = \cos^2(\Delta\theta(k)/2)$  where  $\Delta\theta(k)$  is the angle between the magnetization of two adjacent layers  $\Sigma_{k-1}$  and  $\Sigma_k$ . The conservation of the particles is now described by

$$\begin{aligned} \frac{dN_\alpha}{dt} &= (1 - \Delta\epsilon(k))I_\alpha^{k-1 \rightarrow k} - I_\alpha^{k \rightarrow k+1} + \Delta\epsilon(k)I_\gamma^{k-1 \rightarrow k} - \dot{\Psi}^k \\ \frac{dN_\gamma}{dt} &= (1 - \Delta\epsilon(k))I_\gamma^{k-1 \rightarrow k} - I_\gamma^{k \rightarrow k+1} + \Delta\epsilon(k)I_\alpha^{k-1 \rightarrow k} + \dot{\Psi}^k. \end{aligned} \quad (40)$$

With the notation introduced in the previous sections, the entropy variation can be written in the following way:

$$\begin{aligned} T \frac{dS}{dt} &= P_\Phi^{R' \rightarrow 1} - P_\Phi^{\Omega \rightarrow R'} \\ &+ \sum_{k=2}^{\Omega} \frac{1}{2} (\Delta\mu^{k-1} - \Delta\mu^k + 2(1 - \Delta\epsilon(k))\Delta\mu^k) \delta I_s^{k-1 \rightarrow k} \\ &+ \sum_{k=2}^{\Omega} \frac{1}{2} (\mu^{k-1} - \mu^k) I_0^{k-1 \rightarrow k} + \sum_{k=1}^{\Omega} \Delta\mu^k \dot{\Psi}^k \end{aligned} \quad (41)$$

where we have introduced  $I_0 = I_\alpha + I_\gamma$ ,  $\delta I = I_\alpha - I_\gamma$ ,  $\mu_0 = \mu_\alpha + \mu_\gamma$ , and  $\Delta\mu = \mu_\alpha - \mu_\gamma$ . The terms  $P_\Phi^{R' \rightarrow 1}$  and  $P_\Phi^{\Omega \rightarrow R'}$  stand for heat and chemical transfer from the reservoirs to the system  $\Sigma$ .

After taking the continuum limit, the internal entropy production  $\mathcal{I}$  (or irreversibility) reads

$$T \cdot \mathcal{I} = -\frac{1}{2} \frac{\partial \mu_0}{\partial z} J_0 + \frac{1}{2} \left( -\frac{\partial \Delta\mu}{\partial z} + 2\epsilon \Delta\mu \right) \delta J + \Delta\mu \dot{\Psi}. \quad (42)$$

The first term is the Joule effect, the second is the dissipation related to the spin accumulation process that occurs at the interface, or for a magnetic domain wall, and the third term is the dissipation due to spin-flip (or s-d) electronic relaxation. The expression for the entropy production equation (42) allows the Onsager relations generalizing equation (11) or (28) to be deduced:

$$\begin{aligned} J_0 &= -\frac{\sigma_0}{2e} \left( \frac{\partial \mu_0}{\partial z} + \beta \left( \frac{\partial \Delta\mu}{\partial z} - 2\epsilon \Delta\mu \right) \right) \\ \delta J &= -\frac{\sigma_0}{2e} \left( \beta \frac{\partial \mu_0}{\partial z} + \frac{\partial \Delta\mu}{\partial z} - 2\epsilon \Delta\mu \right) \\ \dot{\Psi} &= L_{\alpha\gamma} \Delta\mu \end{aligned} \quad (43)$$

where  $\epsilon = \lim_{\Delta k \rightarrow 0} \frac{\Delta\epsilon(k)}{\Delta k}$ , and, as already introduced,  $\sigma_0 = \sigma_\alpha + \sigma_\gamma$  and  $\beta = (\sigma_\alpha - \sigma_\gamma)/\sigma_\tau$ .

The diffusion equation for  $\Delta\mu$ , obtained in the stationary regime, is modified accordingly:

$$\frac{\partial^2 \Delta\mu}{\partial z^2} = \left( \frac{1}{l_{\text{diff}}^2} + \frac{1}{l_{\text{DW}}^2} \right) \Delta\mu + \frac{1}{\kappa} \frac{\partial \Delta\mu}{\partial z} \quad (44)$$

where the length  $l_{\text{diff}}$  as been defined in the first section, equation (20):

$$l_{\text{diff}} = \sqrt{\frac{\sigma_0(1 - \beta^2)}{2eL}} \quad (45)$$

and the domain wall diffusion length  $l_{\text{DW}}$  is defined as

$$l_{\text{DW}} = \sqrt{\frac{(1 - \beta^2)}{4\epsilon}} \quad (46)$$

while the length  $\kappa$  is given by

$$\kappa^{-1} = \epsilon \frac{2\beta^2}{1 - \beta^2}. \quad (47)$$

The magnetoresistance is modified with respect to equation (19), due to the new term  $\partial_z \Delta\mu(z)$  in the diffusion equation. It is worth pointing out that a spin accumulation  $\Delta\mu(z) \neq cst$  should be expected in the case of spin polarized current ( $\beta \neq 0$ ) even without the usual spin-flip contribution, i.e. in the ballistic limit.

### 3. Ferromagnetic Brownian motion and magnetization switching

#### 3.1. Thermokinetic derivation of the Fokker–Planck equation

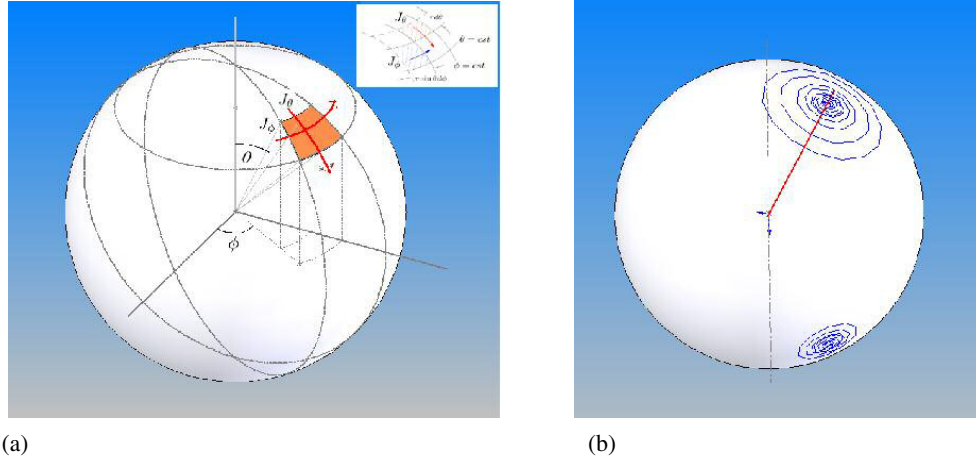
The description given in the previous sections is related to the transport properties of charge carriers in the case of spin polarized current. In spintronics experiments, the electric current is spin polarized through a ferromagnetic layer, but it is not necessary to describe the ferromagnetic order parameter as such. This is of course no longer the case for current induced magnetization switching experiments, where the magnetization is the measured variable.

The magnetization is a fascinating degree of freedom, that has to be described in terms of rotational Brownian motion. The description of the dynamics of ferromagnetic particles coupled to a heat bath is a very active field of investigation [88–93], and the resulting predictions are rather well known and validated experimentally at large [94–96] and short [97–101] timescales. The magnetization relaxation described here is limited to the so-called Néel relaxation that involves only the magnetic moment, in contrast to the Debye inertial relaxation occurring in ferrofluids (in which the ferromagnetic particles rotate in a viscous environment, leading to surprising inertial effects like negative viscosity [46]).

The aim of this subsection is first to show that the rotational Fokker–Planck equation governing the dynamics of the magnetization  $\vec{M}$  of one monodomain particle coupled to the heat bath can also be obtained applying step by step the approach used in the previous sections. The resulting Fokker–Planck equation with the corresponding Onsager transport coefficients, and the hypothesis used, can then be compared term by term to the previous study of spin dependent charge transport.

*3.1.1. Geometrical representation of the statistical ensemble.* Let  $\Sigma$  be a statistical ensemble of  $N$  identical monodomain particles of volume  $v$ , having the same energy per unit volume  $V^{\text{mag}}(\theta, \phi)$ , magnetization  $\vec{M}$  and thermostat temperature  $T$ . The vector  $\vec{M}$  is defined by the angles  $\theta$  and  $\phi$ . The ensemble  $\Sigma$  can be represented by a distribution of representative points over the unit sphere (figure 5) with a density  $n(\theta, \phi)$ .

We divide the ensemble of representative points  $\Sigma$  into sub-ensembles  $\Sigma_{\theta, \phi}$  such that the magnetization is confined within the solid angle  $\delta V_{\theta, \phi} = \sin \theta d\theta d\phi$  (i.e. the representative points lie between two consecutive parallels and meridians over the sphere).



**Figure 5.** (a) The figure on the left illustrates the flow of representative points over the unit sphere:  $J^\theta$  and  $J^\phi$ . (b) The figure on 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).

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  $\vec{J}^{\text{mag}}$ ; 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  $dV_{\theta,\phi}$  at a given time  $t$  is  $dP(\theta, \phi, t) = \frac{n(\theta,\phi,t)}{N} dV_{\theta,\phi}$ .

**3.1.2. Conservation laws.** The sub-ensembles of representative points  $\Sigma_{\theta,\phi}$  are described using the following extensive parameters: the entropy  $dS = s(\theta, \phi, t) dV_{\theta,\phi}$ , the number of points  $dN = n(\theta, \phi, t) dV_{\theta,\phi}$  and the energy  $dE = e(\theta, \phi, t) dV_{\theta,\phi}$ , where  $s$  and  $e$  are the entropy and energy densities. The flow (of points, energy, and entropy) is described as the two-dimensional flux  $\vec{J}$  ( $\vec{J}_n$ ,  $\vec{J}_e$  and  $\vec{J}_s$ ):

$$\vec{J} = J^\theta \vec{u}_\theta + J^\phi \vec{u}_\phi \tag{48}$$

and accounts for the flow of the corresponding magnetic moments relaxing or precessing along the coordinates  $\theta, \phi$ , where  $\vec{u}_r, \vec{u}_\theta, \vec{u}_\phi$  are the unit vectors in the spherical coordinate system.

The conservation laws of the number, energy and entropy of the particles contained in the sub-ensemble  $\Sigma_{\theta,\phi}$  can be written as

$$\begin{cases} \frac{\partial n}{\partial t} = -\text{div } \vec{J}_n \\ \frac{\partial e}{\partial t} = -\text{div } \vec{J}_e \\ \frac{\partial s}{\partial t} = -\text{div } \vec{J}_s + \mathcal{I} \end{cases} \Rightarrow \begin{cases} \frac{\partial n}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial\theta} (J_n^\theta \sin\theta) - \frac{1}{\sin\theta} \frac{\partial J_n^\phi}{\partial\phi} \\ \frac{\partial e}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial\theta} (\mathcal{J}_e^\theta \sin\theta) - \frac{1}{\sin\theta} \frac{\partial \mathcal{J}_e^\phi}{\partial\phi} \\ \frac{\partial s}{\partial t} = -\frac{1}{\sin\theta} \frac{\partial}{\partial\theta} (\mathcal{J}_s^\theta \sin\theta) - \frac{1}{\sin\theta} \frac{\partial \mathcal{J}_s^\phi}{\partial\phi} + \mathcal{I} \end{cases} \tag{49}$$

where in contrast to the energy and number of particles, the entropy  $s$  is not a conservative quantity, and an internal entropy production term  $\mathcal{I}$  (or *irreversibility*) is added to the entropy flux  $\vec{J}_s$  (third equation in equation (49)).

The expression for the first law of thermodynamics allows the energy variation to be expressed as a function of the partial derivatives that define the chemical potentials and the

temperature  $\tilde{\mu} \equiv \frac{\partial e}{\partial n}$  and  $T \equiv \frac{\partial e}{\partial s}$ . The intensive variables  $\tilde{\mu}$  and  $T$  are also functions of  $(\theta, \phi, t)$  except if imposed by a reservoir.

$$\frac{\partial s}{\partial t} = \frac{1}{T} \frac{\partial e}{\partial t} - \frac{\tilde{\mu}}{T} \frac{\partial n}{\partial t} \quad (50)$$

where  $\tilde{\mu} \equiv \partial_n e$  contains all contributions to the energy (see below).

The expression for the internal entropy variation can be obtained using the conservation laws

$$\frac{\partial s}{\partial t} = -\frac{1}{T} \operatorname{div} \vec{J}_e + \frac{\tilde{\mu}}{T} \operatorname{div} \vec{J}_n \quad (51)$$

or, in another form,

$$\frac{\partial s}{\partial t} = -\operatorname{div} \left( \frac{1}{T} \vec{J}_e - \frac{\tilde{\mu}}{T} \vec{J}_n \right) + \vec{J}_e \cdot \vec{\operatorname{grad}} \left( \frac{1}{T} \right) - \vec{J}_n \cdot \vec{\operatorname{grad}} \left( \frac{\tilde{\mu}}{T} \right). \quad (52)$$

Comparing this last equation with the third equation from (49), we can deduce the form of the entropy production  $\mathcal{I}$ :

$$\begin{aligned} \vec{J}_s &= \frac{1}{T} \vec{J}_e - \frac{\tilde{\mu}}{T} \vec{J}_n \\ \mathcal{I} &= \vec{J}_e \cdot \vec{\operatorname{grad}} \left( \frac{1}{T} \right) - \vec{J}_n \cdot \vec{\operatorname{grad}} \left( \frac{\tilde{\mu}}{T} \right). \end{aligned} \quad (53)$$

The entropy production  $\mathcal{I}$  is a sum of products of the fluxes  $\vec{J}_k$  and the corresponding conjugate forces  $\vec{F}_k$  [43].

We assume in the following that the temperature  $T(\theta, \phi) = T$  is fixed by a unique thermostat: the first term in the right-hand side of the equation (53) vanishes.

A sufficient condition for imposing the second law of thermodynamics  $\mathcal{I} \geq 0$  is then building a quadratic form. This leads us to define the matrix  $\mathcal{L}$  of Onsager transport coefficients  $L_{ij}(\theta, \phi)$  (that are state functions of dimension  $[\text{energy}]^{-1}[\text{time}]^{-1}$ ) such that  $J_i = \sum_j L_{ij} \partial_j \tilde{\mu}$ , where the symmetrized  $\mathcal{L}$  matrix is positive. The *Onsager reciprocity relations* impose furthermore that  $L_{ij} = \pm L_{ji}$ , where the sign ( $-$ ) is present if  $L_{ij}$  is a function of the magnetic field (there is no angular velocity here) [40].

The following relations are deduced:

$$\vec{J}_n = -\mathcal{L} \vec{\operatorname{grad}} \tilde{\mu} \Rightarrow \begin{cases} J_n^\theta = -L_{\theta\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - L_{\theta\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} \\ J_n^\phi = -L_{\phi\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - L_{\phi\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} \end{cases} \quad (54)$$

where  $L_{\theta\phi} = -L_{\phi\theta}$ . The first of equations (49) can be rewritten as

$$\frac{\partial n_{(\theta, \phi, t)}}{\partial t} = -\operatorname{div} \vec{J}_n = +\operatorname{div} (\mathcal{L} \vec{\operatorname{grad}} \tilde{\mu}) \quad (55)$$

$$\begin{aligned} \Rightarrow \frac{\partial n_{(\theta, \phi, t)}}{\partial t} &= \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left[ \sin \theta \left( L_{\theta\theta} \frac{\partial \tilde{\mu}_{(\theta, \phi, t)}}{\partial \theta} + \frac{1}{\sin \theta} L_{\theta\phi} \frac{\partial \tilde{\mu}_{(\theta, \phi, t)}}{\partial \phi} \right) \right] \\ &\quad + \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \left[ L_{\phi\theta} \frac{\partial \tilde{\mu}_{(\theta, \phi, t)}}{\partial \theta} + \frac{1}{\sin \theta} L_{\phi\phi} \frac{\partial \tilde{\mu}_{(\theta, \phi, t)}}{\partial \phi} \right] \end{aligned} \quad (56)$$

where  $L_{\theta\theta} \geq 0$  and  $L_{\phi\phi} \geq 0$ . This is the general expression for the density variation  $\partial_t n(\theta, \phi, t)$  of particle number from the sub-ensemble  $\Sigma_{\theta, \phi}$ . Note that there is no relaxation term ( $\Psi$ ) in the conservation law (55) of the magnetization: the flow of representative points is conserved on the unit sphere. This assumption will be removed in the case of spin injection performed with electric currents (see the next section).

In the same manner as for equation (12), the Onsager coefficients are related to the relevant relaxation times. In the case of ferromagnetic insulators, the relaxation channels are well defined [102], and the coefficients  $L_{ij}$  are directly related to the relaxation times  $T_1$  and  $T_2$  measured in ferromagnetic resonance experiments.

### 3.2. The rotational Fokker–Planck equation

In thermokinetics, the intensive parameter which controls the number of particles of a sub-ensemble is the chemical potential  $\tilde{\mu}$ . The relevant energy terms are contained in the deterministic potential  $V^{\text{mag}}$ , and the stochastic term is defined by thermal fluctuations due to the coupling to a relevant heat bath. Anticipating the last section, it is worth pointing out that the relevant heat bath is defined by the degrees of freedom of the environment which are those of the lattice or those of the electronic system (as discussed below). The fluctuations are taken into account through a temperature dependent chemical potential that takes the following form (derived in the general case by Mazur in [43]):

$$\tilde{\mu} \equiv k_B T \ln(n) + v V^{\text{mag}}(\theta, \phi) + \mu_0. \quad (57)$$

The first term on the right-hand side of equation (57) accounts for the density of particles at temperature  $T$ , the second term  $v V^{\text{mag}}$  represents the magnetic potential energy that defines the local magnetic field  $\vec{H}_{\text{eff}} = -\vec{\nabla} V^{\text{mag}}$  (where the two-dimensional gradient  $\vec{\nabla}$  is defined on the surface of the unit sphere) and the third term is a constant which is related to the chemical nature of the particles.

The local equilibrium condition  $\partial_i \tilde{\mu} = 0$  defines stationary fluxes (due to both drift and diffusion) that are mutually compensated along the coordinate  $i$ . This point is well illustrated in the work of Guggenheim while introducing the electrochemical potential [38] in order to generalize the description of an electric fluid to ionic solutions.

Inserting the expression for  $\tilde{\mu}$ , and using the reciprocal relation  $L_{\theta\phi} = -L_{\phi\theta}$ , the equations for fluxes and the variation of particles take the form

$$\begin{aligned} J_n^\theta &= -\left(h' \frac{\partial V^{\text{mag}}}{\partial \theta} - \frac{g'}{\sin \theta} \frac{\partial V^{\text{mag}}}{\partial \phi}\right) n - \left(h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} - \frac{g'}{\sin \theta} \frac{k_B T}{v} \frac{\partial n}{\partial \phi}\right) \\ J_n^\phi &= -\left(g' \frac{\partial V^{\text{mag}}}{\partial \theta} + \frac{k'}{\sin \theta} \frac{v}{k_B T} \frac{\partial V^{\text{mag}}}{\partial \phi}\right) n - \left(g' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} + \frac{k'}{\sin \theta} \frac{\partial n}{\partial \phi}\right) \end{aligned} \quad (58)$$

where the magnetic diffusion coefficients have been defined in the following form:

$$h' = \frac{L_{\theta\theta} v}{n}; \quad g' = -\frac{L_{\theta\phi} v}{n} = \frac{L_{\phi\theta} v}{n}; \quad k' = L_{\phi\phi} \frac{k_B T}{n} \geq 0. \quad (59)$$

Assuming that  $g'$  is constant, equation (56) can be rewritten as

$$\begin{aligned} \frac{\partial n(\theta, \phi)}{\partial t} &= \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta \left[ \left( h' \frac{\partial V^{\text{mag}}}{\partial \theta} - \frac{g'}{\sin \theta} \frac{\partial V^{\text{mag}}}{\partial \phi} \right) n + h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \right] \right\} \\ &+ \frac{1}{\sin \theta} \frac{\partial}{\partial \phi} \left\{ \left( g' \frac{\partial V^{\text{mag}}}{\partial \theta} + \frac{k'}{\sin \theta} \frac{v}{k_B T} \frac{\partial V^{\text{mag}}}{\partial \phi} \right) n + \frac{k'}{\sin \theta} \frac{\partial n}{\partial \phi} \right\}. \end{aligned} \quad (60)$$

The expression (60) represents the rotational Fokker–Planck equation obtained by thermokinetic means. It is identical to that obtained by Brown [89] through stochastic calculations.

Furthermore, the Onsager matrix also follows the symmetry of the system, and is invariant on rotation around the anisotropy axis, so  $L_{\phi,\phi} = L_{\theta,\theta}$ . The following relations are obtained for the magnetic transport coefficients:

$$L_{\theta\theta} = L_{\phi\phi} = \frac{h'n}{v} = \frac{k'n}{k_B T} \quad (61)$$

which permits us to write out the equations above in a compact vectorial form:

$$\begin{aligned}\vec{J} &= -g'\vec{u}_r \times \left( n\vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v}\vec{\nabla}n \right) + h'\vec{u}_r \times \left[ \vec{u}_r \times \left( n\vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v}\vec{\nabla}n \right) \right] \\ \frac{\partial n}{\partial t} &= g'\vec{\nabla} \left\{ \vec{u}_r \times \left( n\vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v}\vec{\nabla}n \right) \right\} - h'\vec{\nabla} \left\{ \vec{u}_r \times \left[ \vec{u}_r \times \left( n\vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v}\vec{\nabla}n \right) \right] \right\}\end{aligned}\quad (62)$$

where  $\vec{\nabla}$  is the two-dimensional gradient operator defined on the surface of the unit sphere and  $\vec{u}_r$  is the spherical radial unit vector. It is to be noted that the second equation has drift terms which contain  $\nabla V^{\text{mag}}$ , and diffusion terms which contain  $\vec{\nabla}n$ . The terms  $k' = h'\frac{k_{\text{B}}T}{v}$  and  $g'\frac{k_{\text{B}}T}{v}$  are the rotational diffusion coefficients and the terms  $g'n$  and  $h'n$  represent drift coefficients.

### 3.3. Landau–Lifshitz–Gilbert equation with diffusion

Using the first equation from (62), one can deduce the Landau–Lifshitz–Gilbert equation with diffusion. As the flux is related to the time derivative of the unit vector  $\vec{u}_r$  through the relation

$$\vec{J} = n \frac{d\vec{u}_r}{dt} \quad (63)$$

we arrive at the following equation:

$$\frac{d\vec{u}_r}{dt} = -g'\vec{u}_r \times \left( \vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v} \frac{\vec{\nabla}n}{n} \right) + h'\vec{u}_r \times \left[ \vec{u}_r \times \left( \vec{\nabla}V^{\text{mag}} + \frac{k_{\text{B}}T}{v} \frac{\vec{\nabla}n}{n} \right) \right] \quad (64)$$

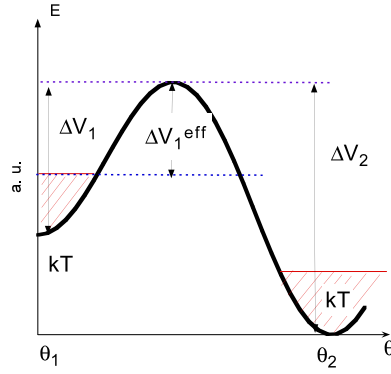
where the first term in the right-hand side is *the precession* term, and the second term in the right-hand side describes the *longitudinal relaxation*. Multiplied by the amplitude of the magnetization  $M_s$ , equation (64) becomes the well known LLG equation:

$$\frac{d\vec{M}}{dt} = g'M_s \left( \vec{M} \times \vec{H}_{\text{eff}} \right) + h' \left( \vec{M} \times \vec{H}_{\text{eff}} \right) \times \vec{M} \quad (65)$$

where the effective field  $\vec{H}_{\text{eff}} = \vec{H}_{\text{eff}} - \frac{k_{\text{B}}T}{nvM_s}\vec{\nabla}n$  includes the diffusion term. Experimentally, the first contribution can be observed through ferromagnetic resonance measurements (FMR) at typical frequencies of tens of GHz (around 100 ps timescales). The thermalization time (proportional to  $1/g'$ ; see the next section) is given by the width of the resonance peaks. Both frequency and time resolved noise experiments have been also performed in order to measure precession and thermal spin waves by using giant magnetoresistance properties [97–101]. In the corresponding measurements, the data are averaged over many shots near an equilibrium position (linear response regime). In contrast, the measurements at large timescales, typically beyond a few nanoseconds, access the magnetization reversal for which *the precession terms can be neglected*. The *one-shot measurements* give direct access to the stochastic nature of the signal [25, 26, 29, 94, 95]: a snapshot is a statistical event, namely the magnetization reversal from one metastable state to the other, that is governed by the random fluctuations, described as a ‘Langevin force’ (not present in the averaged LLG equation).

Equation (64) can be put into the Gilbert form by constructing the cross-product  $\times \vec{M}$  on the left- and right-hand sides of the equation. We obtain the well known Gilbert equation, that defines the Gilbert damping parameter  $\eta$ :

$$\frac{d\vec{M}}{dt} = \Gamma \vec{M} \times \left( \vec{H}_{\text{eff}} - \eta \frac{d\vec{M}}{dt} \right). \quad (66)$$



**Figure 6.** Double-well potential (continuous line) with stochastic fluctuations (dashed area), and the definition of the barrier heights.

The transport coefficients  $h'$ ,  $g'$  and  $k'$  are related to the Gilbert damping coefficient  $\eta$ , the gyromagnetic factor  $\Gamma$  and the magnetization at saturation  $M_s$  through the following relations:

$$g' = \frac{\Gamma}{(1 + (\eta\Gamma M_s)^2) M_s}; \quad h' = \eta\Gamma M_s g'. \quad (67)$$

The Gilbert damping factor is usually expressed through the damping parameter  $\alpha$  without dimension:

$$\alpha = \eta\Gamma M_s. \quad (68)$$

### 3.4. Activation regime and Néel–Brown law

**3.4.1. Neglecting precession.** In the slow relaxation measurements (the so-called magnetic after-effect), relaxation is governed by activation over a potential barrier. At long timescales (beyond tens of nanoseconds to hours), the precessional terms can be neglected. The expression for the surface current fluxes equation (58) becomes

$$\begin{aligned} J_n^\theta &= -h' \frac{\partial V^{\text{mag}}}{\partial \theta} n - h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \\ J_n^\phi &= -g' \frac{\partial V^{\text{mag}}}{\partial \theta} n - g' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \end{aligned} \quad (69)$$

and the corresponding Fokker–Planck equation becomes

$$\frac{\partial n(\theta, \phi)}{\partial t} = \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left\{ \sin \theta \left[ h' \frac{\partial V^{\text{mag}}}{\partial \theta} n + h' \frac{k_B T}{v} \frac{\partial n}{\partial \theta} \right] \right\}. \quad (70)$$

These expressions will be used in the following subsection for deriving the Néel–Brown relaxation time.

**3.4.2. The double-well potential and the relaxation times.** The double-well potential (see figure 6) is the first approximation of the ferromagnetic particle energy  $V^{\text{mag}}(\theta, \phi)$  beyond the harmonic potential, but it is also a realistic magnetic potential in the case of a uniform magnetization with uniaxial anisotropy [26]:

$$V^{\text{mag}} = K \sin^2 \theta - M_s H_{\text{ap}} \cos(\theta - \varphi) \quad (71)$$



where  $\theta$  is the direction of the magnetization,  $\varphi$  is the direction of the applied field  $H_{\text{ap}}$ , and  $K$  is the anisotropy constant in terms of energy per unit volume.

In order to apply Brown's method, we consider a potential which has minima at  $\theta_1$  and  $\theta_2 = \pi - \theta_1$ , and a maximum at  $\theta_m$ . Following Kramers' transition theory, Brown [89] assumes that most of the representative points on the unit sphere are concentrated at the energy minima of  $V^{\text{mag}}(\theta)$  where they are in thermal equilibrium, so locally  $n$  takes the form of the Maxwell-Boltzmann distribution. Thus only a minute fraction of the representative points are outside the energy minima allowing a small diffusion current between them, so manifesting the non-equilibrium conditions;

$$n(\theta, \phi) = \begin{cases} n(\theta_1) e^{-\frac{v}{k_B T} [V(\theta) - V(\theta_1)]}, & \text{for } \theta \in (\theta_1 - \epsilon, \theta_1 + \epsilon) \\ n(\theta_2) e^{-\frac{v}{k_B T} [V(\theta) - V(\theta_2)]}, & \text{for } \theta \in (\theta_2 - \epsilon, \theta_2 + \epsilon). \end{cases} \quad (72)$$

The numbers of particles  $N_1$  or  $N_2$  from the first and the second wells, respectively, are

$$\begin{aligned} N_1 &= 2\pi n(\theta_1) e^{\frac{v}{k_B T} V(\theta_1)} I_1, & \text{where } I_1 &= \int_{\theta_1 - \epsilon}^{\theta_1 + \epsilon} e^{-\frac{v}{k_B T} V(\theta)} \sin \theta \, d\theta \\ N_2 &= 2\pi n(\theta_2) e^{\frac{v}{k_B T} V(\theta_2)} I_2, & \text{where } I_2 &= \int_{\theta_2 - \epsilon}^{\theta_2 + \epsilon} e^{-\frac{v}{k_B T} V(\theta)} \sin \theta \, d\theta. \end{aligned} \quad (73)$$

Assuming that the flow between the two minima  $\theta_1$  and  $\theta_2$  is *quasi-stationary*, approximated by a divergenceless current [89], the total current of particles over the potential barrier can be written as

$$I = 2\pi \sin \theta J_n^\theta. \quad (74)$$

Rewriting the first equation from (69), one obtains

$$\frac{\partial n}{\partial \theta} + \frac{v}{kT} \frac{\partial V^{\text{mag}}}{\partial \theta} n = -\frac{I v}{2\pi h' k_B T \sin \theta} \quad (75)$$

which defines the *activation regime*. Introducing  $I_m$  as

$$I_m = \int_{\theta_1 + \Delta\epsilon}^{\theta_2 - \Delta\epsilon} \frac{e^{\frac{v}{k_B T} V(\theta)}}{\sin \theta} \, d\theta, \quad (76)$$

equation (75) yields

$$I = -\dot{N}_1 = \dot{N}_2 = -\frac{h' k_B T}{v I_m} \left( \frac{N_2}{I_2} - \frac{N_1}{I_1} \right) \quad (77)$$

which has the form

$$\dot{N}_1 = -\dot{N}_2 = \frac{N_2}{\tau_2} - \frac{N_1}{\tau_1} \quad (78)$$

with

$$\begin{aligned} \tau_1 &= \frac{I_1 I_m v}{h' k_B T} \\ \tau_2 &= \frac{I_2 I_m v}{h' k_B T}. \end{aligned} \quad (79)$$

Because of the rapid decrease of the exponential factor with distance from the minima of  $V^{\text{mag}}$ , we may in  $I_1$ ,  $I_2$ ,  $I_m$  replace  $V^{\text{mag}}(\theta)$  by its Taylor's series about  $\theta_1$ ,  $\theta_2$ ,  $\theta_m$ , respectively, truncated at the  $\theta_2$  term, and replace the upper limit of the integrals by  $\infty$ . With these approximations, we find

$$\begin{aligned} \tau_1 &= \tau_{01} e^{\frac{v(V(\theta_m) - V(\theta_1))}{k_B T}} \\ \tau_2 &= \tau_{02} e^{\frac{v(V(\theta_m) - V(\theta_2))}{k_B T}} \end{aligned} \quad (80)$$

where the waiting times are given by the expressions

$$\begin{aligned}\tau_{01} &= \frac{2\pi}{h'} [-V''(\theta_1)V''(\theta_m)]^{-\frac{1}{2}} \frac{\sin \theta_1}{\sin \theta_m} \\ \tau_{02} &= \frac{2\pi}{h'} [-V''(\theta_2)V''(\theta_m)]^{-\frac{1}{2}} \frac{\sin \theta_2}{\sin \theta_m}.\end{aligned}\quad (81)$$

Equation (80) is a formula for a *symmetric bistable* potential which has minima at  $\theta_1$ ,  $\theta_2 = \pi - \theta_1$  and a maximum at  $\theta_m = \pi/2$ .

For  $\varphi \neq 0$ , the potential  $V^{\text{mag}}(\theta, 0)$  has an asymmetric bistable form, and all arguments leading to equation (80) also apply for an arbitrary  $\varphi$  [91]. The general equations are very similar to equation (80); the only difference in the analytic expression is that instead of the symmetric angles,  $\theta_1, \theta_2$  are now asymmetric.

It has to be emphasized that  $\tau_1$  and  $\tau_2$  are the relaxation times related to the first and second potential barriers (starting from the first or the second minima), and that equation (80) constitutes the Néel–Brown law for the particular case of the *symmetric bistable* well. Interestingly, in all cases [91], the Gilbert damping is reduced to the prefactor of the exponential, and consequently plays only a negligible role in the activation process.

In many cases, the potential is highly asymmetric, and the Néel–Brown law is written in the following asymptotic form:

$$\tau = \tau_0 \exp\left(\frac{\Delta V_0(1 - H/H_{\text{sw}}^0)^\alpha}{kT}\right) \quad (82)$$

with three phenomenological parameters  $\alpha \approx 3/2$ ,  $\Delta V_0$  and  $H_{\text{sw}}^0$  [94, 95]. The laws (80) and (82) are well established experimentally in usual magnetic sub-microstructures. More surprisingly, they have been also observed in time resolved one-shot measurements under spin injection with high effective temperature  $T_{\text{eff}}$  (2000–20 000 K) instead of 300–340 K [2, 25–29] (see the next section).

**3.4.3. Hysteresis loops and one-shot measurements.** Let us start from the LLG equation (65). The first term in the right-hand side describes the precession, relevant at the nanosecond timescale and below, and the second term in the right-hand side describes the alignment relaxation. The quasi-static measurements, i.e. the hysteresis loops, are described in a first approach by considering the alignment term only, without activation processes (i.e. at zero kelvins). This corresponds to the Stoner–Wohlfarth model if the uniform magnetization switching mode is assumed [89, 103]. The hysteresis loop is hence described by the condition

$$\vec{H}_{\text{eff}} \times \vec{M} = \vec{0} \quad (83)$$

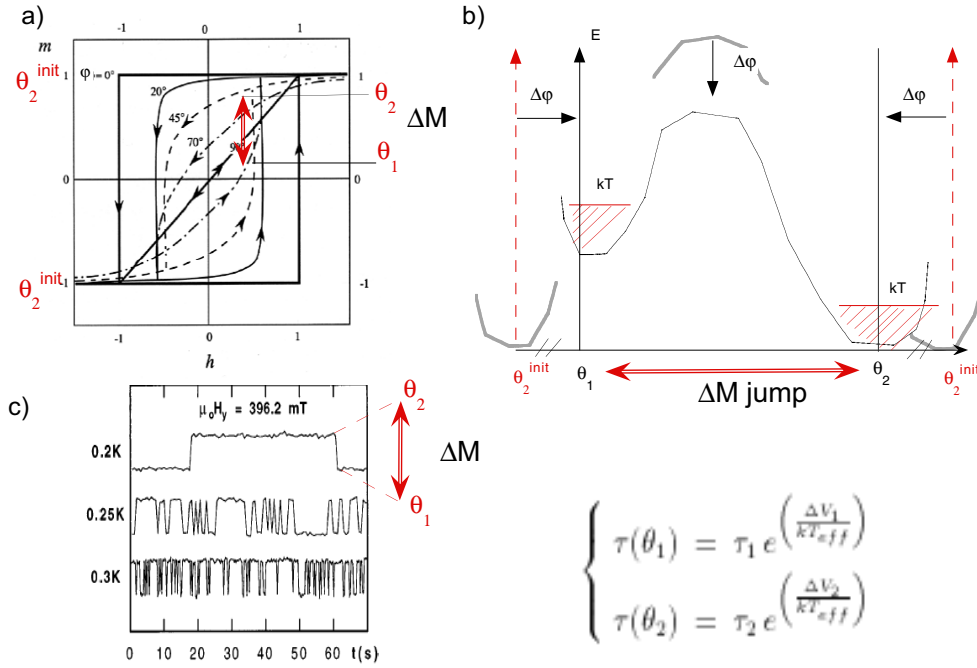
or, in other words, the projection of  $\vec{H}_{\text{eff}}$  over the vectors perpendicular to  $\vec{M}$  is zero:

$$\begin{aligned}\vec{H}_{\text{eff}} \cdot \vec{u}_\theta(\theta, \phi) &= 0 \\ \vec{H}_{\text{eff}} \cdot \vec{u}_\phi(\theta, \phi) &= 0.\end{aligned}\quad (84)$$

In the case of the uniaxial magnetic anisotropy, there is only one variable  $\theta$ , and the quasi-static conditions reduce to  $\vec{H}_{\text{eff}} \cdot \vec{u}_\theta = 0$ . This equation defines the quasi-static, or reversible, branches of the hysteresis. The irreversible jump between two branches is given *independently* by a supplementary condition [94, 104]. *In the case of the Stoner–Wohlfarth model* (uniform rotation at zero kelvins) the condition can be written as  $d/d\theta(\vec{H}_{\text{eff}} \cdot \vec{u}_\theta) = 0$ , and the hysteresis loop, plotted in figure 7(a), is defined by the equations

$$\begin{aligned}\vec{H}_{\text{eff}} \cdot \vec{u}_\theta &= 0 \\ \frac{d}{d\theta} (\vec{H}_{\text{eff}} \cdot \vec{u}_\theta) &= 0\end{aligned}\quad (85)$$

with different values of the applied field.



**Figure 7.** Hysteresis loop and one-shot measurements in the uniaxial anisotropic single magnetic domain. (a) Hysteresis loop at different angles  $\varphi$  of the applied magnetic field. The saturation (i.e. initial states without excitation in the relaxation protocol:  $H_y = 0$ ) corresponds to the magnetization  $\theta_1^{init}$  or  $\theta_2^{init}$ . (b) Double-well potential after the application of the excitation  $\Delta\varphi$ : the whole potential landscape is modified (the barrier height and the two equilibrium configurations). (c) Two-level fluctuations measured between the two configurations (the magnetization jumps from the potential well at  $\theta_1$  to the potential well at  $\theta_2$ ). Right: the Néel–Brown law is verified after taking statistics over a significant number of jumps, in order to access to the mean relaxation times  $\tau(\theta_1)$  and  $\tau(\theta_2)$ . Reprinted with permission from [95]. Copyright ©American Physical Society.

The above conditions are equivalently expressed in terms of the first and second derivatives of the potential energy  $V^{mag}$  (equation (71)), instead of the effective field:

$$\begin{aligned} \frac{\partial V^{mag}}{\partial \theta} &= 0 \\ \frac{\partial^2 V^{mag}}{\partial \theta^2} &= 0. \end{aligned} \tag{86}$$

The advantage of the formulation proposed in equation (85) is that it is not necessary to know the total energy of the system. For example in the case of a ferromagnetic layer close to another magnetic layer (this is typically the case for a pillar spin valve structure in which the second layer plays the role of a spin polarizer: see the next section), a local field due to multipolar fields is present at the level of the ferromagnet studied [5]. The system of the coupled layers is very complicated (e.g. the motion is even chaotic under an oscillatory excitation), but knowledge of the total energy of the coupled system is unnecessary for describing the minor loops due to the switching layer. Only the local expression for the multipolar fields is necessary.

On the other hand, the second condition that gives the position of the irreversible jump (e.g. the second equation in (86) in the case of uniform modes) is no longer valid at non-zero temperature: the hysteresis loop should then be described taking into account the activation

process (see figures 7(b) and (c)). In a single-domain ferromagnet, only the position of the irreversible jump (i.e. the switching field  $H_{sw}$ ) is modified by the activation process (i.e. by the temperature, by the width of the observation time window, or equivalently by the velocity of the sweeping field used to measure the hysteresis loop [105]), but not the rest of the hysteresis loop. The fluctuations do not perturb the quasi-static configurations (e.g. the reversible part of the hysteresis), except at the two critical points where the potential barrier is of the same order as the thermal energy. The reversible part of the hysteresis is not modified by the activation (for a fixed  $M_s$ ) as long as anharmonic effects are negligible.

Of course, this analysis cannot be generalized to non-single-domain samples, i.e. beyond nanoscopic dimensions, because the layer is then an ensemble of sub-systems with a distribution of anisotropies, defects, etc [106], and the hysteresis loops cannot be separated between reversible quasi-static states and irreversible jumps [96, 107].

In contrast to the quasi-static experiments, the time resolved one-shot experiments allow the barrier height—or the amplitude of the thermal fluctuations—to be measured. It is then possible to complete our knowledge of the potential landscape by exploring the barriers between two valleys. If the barrier height is above the energy of the lattice  $kT$ , the irreversible jumps occur within a typical time window that can be tuned with the amplitude of the magnetic field or the temperature. In order to access the mean value of the relaxation times, statistics should be taken over a significant number of shots. If the exponential relaxation is verified, the relevant relaxation times can then be extracted. The ratio of the relaxation times  $\tau(\theta_1)/\tau(\theta_2)$  gives the asymmetry of the double well, and each relaxation time gives the corresponding barrier height. The Néel–Brown law is tested by varying the temperature and the external magnetic field (figure 7). The whole potential can be rebuilt from these measurements.

Figure 7 shows (a) the hysteresis loop (quasi-static states) obtained from the conditions of equation (85), and (b), (c) how to measure the ferromagnetic potential landscape with the help of the quasi-static and slow magnetic relaxation measurements (the so-called magnetic after-effect measurements), *performed on a single-magnetic-domain nanostructure* [26]. In the example shown in (c), the angle and amplitude of the magnetic field is set in order to obtain the two-level fluctuation effect (measurements reported in Wernsdorfer *et al* [95]). The principle of the measurements is sketched in figure 7 for a sample with uniaxial anisotropy: the hysteresis loop describes the succession of equilibrium magnetization configurations as a function of the magnetic field (the applied field is normalized to the anisotropy field) for different angles  $\varphi$  of the applied field. The angle  $\theta$  describes the direction of the magnetization. For a given angle of the field  $\varphi$ , the hysteresis is composed of the reversible configurations and two symmetric irreversible jumps over the potential barrier. The jump occurs from the last equilibrium (metastable) state given by the angle  $\theta_1$  to the other branch of the hysteresis, given by the angle  $\theta_2$  (figure 7(b)). The two angles are defined equivalently by the conditions of equation (85) or (86). The amplitude of the jump is easily measured (it is given by the difference  $M_s(\cos \theta_2 - \cos \theta_1)$ ), and gives direct access, from the hysteresis loop, to the quasi-static configurations (i.e. to the position of the potential wells). Changing the field (amplitude and angle) necessarily changes the equilibrium position and the angles  $\theta_1$  and  $\theta_2$ . Note that the case of uniaxial symmetry with  $\varphi = 0$  is the unique pathological case, where the change in the *amplitude* of the field does not change the equilibrium magnetic configurations. However, since this pathological case is unfortunately that used in most calculations for the sake of simplicity (this was also the case here for the derivation of the Néel–Brown’s law), the fact that the initial and final quasi-static states are necessarily modified is not pointed out in the usual descriptions of the effect of spin transfer torque [108]. Inversely, a change observed in the two equilibrium configurations implies a change of the potential landscape, i.e. a change of the effective field.

**3.4.4. Quasi-ballistic reversal and non-linear resonance.** Finally, it is worth mentioning the situations in which the equilibrium condition  $\vec{H}_{\text{eff}} \times \vec{M} = \vec{0}$  is not sufficient to account for the magnetization reversal. This is the case for a reversal provoked by a pulsed field measured on the timescale of the nanosecond (i.e. in the non-stationary regime), and in the case of a stationary process in which the reversal is helped by a high frequency AC field excitation (anharmonic effects).

In the first case, the relevant time window is shorter than the longitudinal relaxation timescale  $\tau_0$ , the precession now governs the quasi-ballistic dynamics, and the magnetization is not thermalized with the lattice. The collective modes measured are those observed with ferromagnetic resonance; e.g. dynamics of thermal spin waves are observed in GMR structures. Sub-nanosecond time resolved protocols (averaged measurements) allow very rapid magnetization reversal to be measured, that involves few rotations of the magnetization around the effective field before reversal [97–101]. However, we focus our discussion on activation processes, and the quasi-ballistic dynamics is beyond the scope of the present report.

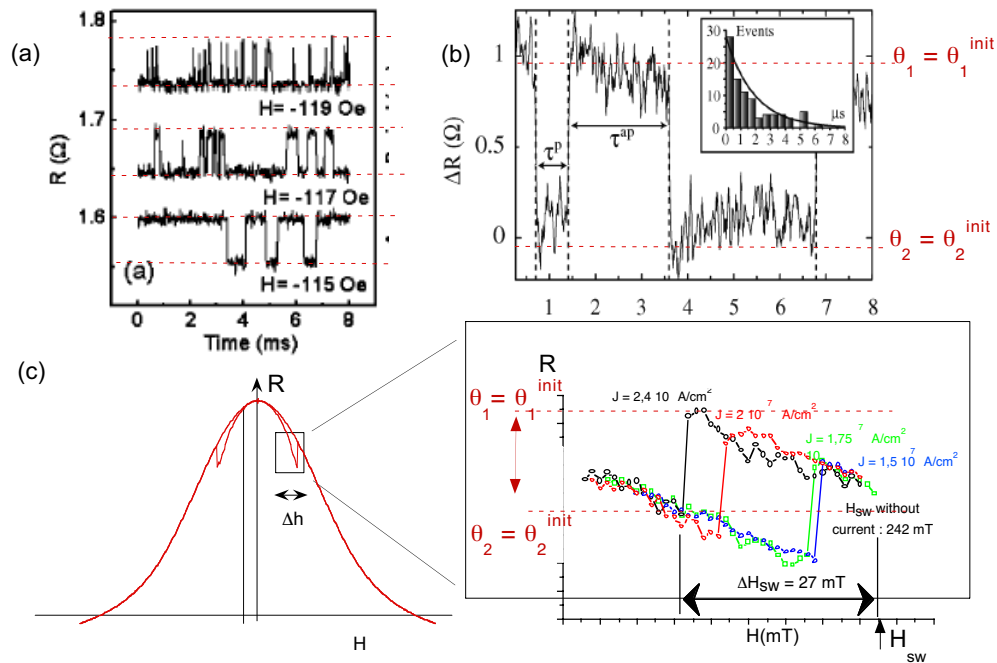
In the second case, a high frequency AC magnetic field is applied in addition to the static field: important variations of the switching field are measured [126]. In that case, the experiments show that the magnetization reversal is affected if the external AC field is in resonance with the precession (frequency of some few GHz). Provided that the resonance is reached, the position of the irreversible jump (the switching field) is driven by the AC field, because the energy of the external wave is absorbed by the magnetic system in the form of kinetic energy. The condition of reversal are then described by the nanolinear stationary condition  $g' M_s (\vec{M} \times \vec{H}_{\text{eff}}) - h' \vec{M} \times (\vec{M} \times \vec{H}_{\text{eff}}) = \vec{0}$ . If the resonance is not reached, the jump is not modified, and the hysteresis loop is not affected. This effect is due to a non-linear resonance process: the precession angle is large so the trajectories of the magnetization are no longer confined within the bottom of the potential wells (the harmonic part of the potential), and at the limit, the trajectories reach the top of the potential barrier: the magnetization reversal occurs deterministically (quasi-ballistic reversal), without the help of the activation process.

#### 4. Spin transfer

Let us now consider a system composed of a ferromagnet coupled to a heat reservoir, in which spin injection (with high current density) is performed. In such spin transfer experiments, the magnetization configurations are measured while injecting the current. Usually the magnetic configurations are measured with magnetoresistance (GMR, AMR or Hall) properties for convenience, but micromagnetometry or magneto-optics measurements are also possible [109].

A lot of experimental results have now been reported. The most common ones are obtained with the measurements of the hysteresis loop as a function of the current amplitude, for a fixed magnetic field [1–8], or the hysteresis loop as a function of the external field, for different currents [9–11]. Many other experiments have been performed in the frequency range with AC currents [12–15, 28, 30–33], with resonance frequencies (beyond GHz) measured as a function of the current amplitude and the applied field. However, since we are interested in the difference between stochastic and deterministic behaviour, we will focus our attention in the following on the one-shot measurements that preserve the stochastic part of the signal [25–29].

Figure 8 shows single-shot measurements with two-level fluctuations in time intervals of milliseconds (a) [25] and microseconds (b) [27], obtained by different groups with GMR measurements on trilayer nanopillars. Figure 8(c) shows the anisotropic magnetoresistance of a single contacted Ni nanowire. A zoom is performed on the irreversible jumps measured in the hysteresis loop [29]. In the last case, each point in the zoom (right part of figure 8(c)) is a 6  $\mu\text{s}$  pulse. The hysteresis is obtained with current varying from  $2.4 \times 10^7$  to  $1.5 \times 10^7$  A cm<sup>-2</sup> (the



**Figure 8.** One-shot measurements under spin injection. (a) Two-level fluctuations measured with GMR in a trilayer system (permalloy/Cu/permalloy, from [25]). (b) Two-level fluctuations measured with GMR in a trilayer structure (Co/Cu/Co, from [27]). (c) Hysteresis loop measured with AMR (left) and zoom around the irreversible jump measured under spin injection (6  $\mu$ s pulse per point) (details reported in [29]). The hysteresis measured with AMR shows the succession of the equilibrium configurations. In all cases, the initial and final states (the equilibrium states) are those measured without current injection ( $\theta = \theta^{init}$ ). Equilibrium configurations are not modified by the spin injection. Reprinted with permission from [25] and [27]. Copyright ©American Physical Society.

position of the jump without current injected is shown by the arrow at  $H_{sw}$ ). In all cases, the metastable states (described with the angles  $\theta_1$  and  $\theta_2$  defined in the previous section) coincide with  $\theta_1^{init}$  and  $\theta_2^{init}$  measured before the application of the current. Everything happens as if the potential landscape were not modified! In contrast, the action of the current is huge (some fraction of an eV to few eV, i.e. from 25% to more than 100% of the anisotropy energy of the ferromagnet), and is observed only for the irreversible jump. By acting on the jump and not on the potential landscape, the effect of the current seems to mimic the action of a temperature.

This is one of the most striking differences observed when comparing to the corresponding measurements without spin injection. The point discussed in the following section is to establish whether this observation can be described in the framework of a current dependent deterministic term added to the LLG equation. The following subsection investigates the general case of a non-deterministic contribution in terms of open systems.

#### 4.1. Generalized Landau–Lifshitz–Gilbert equation

The system described above has been investigated theoretically on the basis of some different postulates: the one most commonly used is Slonczewski’s spin torque

expression [34, 35, 110–119], but other concepts are also being introduced, like that of radiation damping [120], and other hypotheses related to spin relaxation (see e.g. [78, 121, 122]). However, whatever the mechanism invoked, the result can be described in a very general way, by adding current dependent terms into the LLG equation. The most general case of a *deterministic contribution* is investigated in the present subsection. The case of Slonczewski's spin torque is studied in this context.

Let us imagine a physical system composed of a ferromagnet coupled to an external source of energy. A deterministic coupling is generally due to different forms of external fields, so the energy transferred is in general not reduced to a simple potential energy term (e.g., in the case of electromagnetic fields, a potential vector should be introduced). The question to be addressed is the following: what phenomenological equation describes the dynamics of a ferromagnetic single domain coupled to any kind of field, or in other words, in any kind of deterministic perturbation? The answer is found very simply, by following the basic idea used by Landau and Lifshitz while defining the LL equation. Provided the modulus of the magnetization is conserved, the time variation of the magnetization  $d\vec{M}/dt$  is written in the orthonormal basis composed of the two vectors  $(\vec{M} \times \vec{H})$  and  $(\vec{M} \times (\vec{M} \times \vec{H}))$ . In order to generalize this equation to any deterministic contribution (while maintaining the uniformity of the magnetization), it is sufficient to add a vector  $\vec{X}$  to the Gilbert equation:

$$\frac{d\vec{M}}{dt} = \Gamma \vec{M} \times \left( \vec{H} - \eta \frac{d\vec{M}}{dt} \right) + \vec{X}. \quad (87)$$

In order to obtain the generalized Landau–Lifshitz form, the time variation of the magnetization  $d\vec{M}/dt$  should be expressed in the orthogonal basis  $\{\{\vec{M}\}, \{\vec{M} \times \vec{H}\}, \{\vec{M} \times (\vec{M} \times \vec{H})\}\}$ . After cross-multiplying vectorially by  $\vec{M}$  on both sides of equation (87), and using the double-vector-product formula  $(\vec{A} \times (\vec{B} \times \vec{C})) = (\vec{A} \cdot \vec{C}) \vec{B} - (\vec{A} \cdot \vec{B}) \vec{C}$ , the generalized LL equation is obtained:

$$\frac{d\vec{M}}{dt} = g' M_s \vec{M} \times \left( \vec{H} + \eta \vec{X} \right) - h' \vec{M} \times \left[ \vec{M} \times \left( \vec{H} - \frac{\eta}{\alpha^2} \vec{X} \right) \right] - \frac{g'}{\Gamma M_s} (\vec{M} \cdot \vec{X}) \vec{M} \quad (88)$$

where  $g'$ ,  $h'$  and  $\alpha$  are defined in equations (67) and (68). The major change that is observed, with respect to the usual case without an external contribution, is of course that the last term does not conserve the modulus of the magnetization. However, beyond this trivial observation, the striking result is that the effective field does not have the same intensity and sign for the precession term and for the longitudinal relaxation term. It is then necessary to introduce the collinear effective field experienced during the precession process and the effective field experienced during the alignment process. The two corrections to  $H$  are simply related by the coefficient  $-\alpha^2$ . This is due to the fact that the kinetic energy provided by the external system is injected into the precession term. Of course, as expected in this open system, the effective field cannot be derived from a single potential energy term.

#### 4.2. Slonczewski's spin torque

Let us now be less general, and restrict the interaction with the external source to a contribution that preserves the modulus of the magnetization. Instead of  $\vec{X}$  we introduce now a contribution of the form  $c(I) \vec{M} \times (\vec{M} \times \vec{p})/M_s^2$ , where  $\vec{p}$  is a unit vector. Note that this form is that proposed by Slonczewski in 1996 [35], on the basis of considerations concerning microscopic spin torque conservation at the interface of a spin valve during spin injection. The vector  $\vec{p}$  represents then the polarization of the current. These considerations gave rise to a new and fascinating investigation field concerning the different microscopic mechanisms occurring at the interface.



From the phenomenological point of view, the consequence of the double-torque form of  $\vec{X}$  is a reduction of the general Gilbert equation to the (still rather general) form

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \left( \vec{H} - \eta \frac{d\vec{M}}{dt} \right) + \frac{c(I)}{M_s^2} \vec{M} \times (\vec{M} \times \vec{p}) \quad (89)$$

where  $c(I)$  is proportional to the current  $I$ , and  $\vec{X}$  is the magnetization of the polarization layer (in a three-layer-configuration device).

In the Landau–Lifshitz form, the above equation is equivalent to

$$\frac{d\vec{M}}{dt} = g' \vec{M} \times \left( \vec{H} + c(I)\eta \vec{p} \right) - h' \vec{M} \times \left[ \vec{M} \times \left( \vec{H} - \eta \frac{c(I)}{\alpha^2} \vec{p} \right) \right] \quad (90)$$

so we are left with two effective fields:

$$\begin{aligned} \vec{H}_{\text{eff}}^{\text{pre}}(I) &= \vec{H} + \eta c(I) \vec{p} \\ \vec{H}_{\text{eff}}^{\text{al}}(I) &= \vec{H} - \frac{\eta c(I)}{\alpha^2} \vec{p} \end{aligned} \quad (91)$$

where  $\vec{H}_{\text{eff}}^{\text{pre}}$  is the field experienced during the precession (associated with the kinetic energy) and  $\vec{H}_{\text{eff}}^{\text{al}}$  is the field experienced during the alignment process. The following relation that links the two effective fields is the criterion for the validity of the spin torque hypothesis:

$$\vec{H}_{\text{eff}}^{\text{al}}(I) - H = -\alpha^2 \left( \vec{H}_{\text{eff}}^{\text{pre}}(I) - H \right). \quad (92)$$

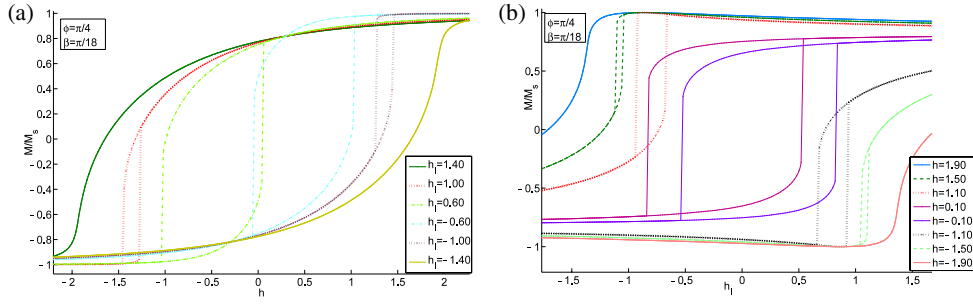
This criterion (92) is one of the basic predictions to be fulfilled in order to show experimentally the validity of the hypothesis of a deterministic contribution of the form  $\vec{X} = c(I) \cdot \vec{M} \times (\vec{M} \times \vec{p}) / M_s^2$ , added to the Gilbert equation. In the case of linear resonance (the so-called ‘reversible’ part in the  $dV/dI$  hysteresis loop), the resonance field is an increasing function of the current amplitude, while in the case of irreversible jumps, the switching field is in contrast a decreasing function of the current.

However, it is necessary to measure separately the precession modes at GHz frequency (stationary dynamical states) and the quasi-static behaviour under current injection, in order to test the criterion (92). If explored close enough to the equilibrium states or more precisely near quasi-static states described with the hysteresis loop (i.e. in the potential valleys), the resonance is the ferromagnetic resonance (FMR), and it is well described by the first term of the LLG equation. If the precession angle is more important, with the result that the non-harmonic part of the potential well is explored, the resulting stationary states involve both terms of the LLG equation, and correspond to non-linear resonance experiments [126].

**4.2.1. Quasi-static spin torque.** For measurement time greatly beyond the nanosecond timescale, the precession is neglected (see the discussion below concerning the case of non-linear resonance), and the quasi-static states are defined by the static equilibrium (first equation of (85)):

$$\left( \vec{H} - \frac{\eta c(I)}{\alpha^2} \vec{p} \right) \cdot \vec{u}_\theta = 0. \quad (93)$$

The system being an open system, the potential energy and kinetic energy of the ferromagnet are no longer defined by a unique energy, a function of the state variables: the energy flux and energy density should be defined with a source term (see below). However, the hysteresis loop, i.e. the succession of quasi-static states for different values of the applied magnetic field and current  $I$ , is still given by the condition of equations (85).



**Figure 9.** Hysteresis loop obtained with the spin torque expression (93) of the text. The field is normalized to the anisotropy field ( $h = \frac{H M_s}{2K}$ ) and applied at an angle of  $\varphi = 45^\circ$ . The polarization  $\vec{p}$  of the current is inside the plane of the layer at an angle of  $\beta = 10^\circ$  and the corresponding normalized field is  $h_I = \frac{nc(I)}{\sigma^2} \frac{M_s}{2K}$ . (a) Hysteresis plotted as function of the applied field for different values of the current. (b) Hysteresis plotted as a function of the current for different values of the applied field.

The effect of the current on the hysteresis loop would then be analogous to that of a local field, like a dipole field. The effect of the spin torque is shown in figure 9 for a vector  $\vec{p}$  in the plane of the layer with an angle of  $10^\circ$  with respect to the anisotropy axis of the ferromagnet (no qualitative differences with other angles): the reversible part of the hysteresis is strongly modified while changing the current. The hysteresis as a function of the current is shown in figure 9(b) for different values of the applied field.

Once again, the reversible effects due to the current on the quasi-static states have not been evidenced experimentally, in contradiction with the prediction from the hypothesis of spin torque.

**4.2.2. Precessional spin torque.** The arguments developed above are based on an analysis performed neglecting the precession terms. If we assume that the precession is maintained in a stationary regime during the one-shot measurements, the magnetization is then driven by the precession [108], like for non-linear resonance, but *without any HF excitations* (in contrast to the experiments in which the current is injected with GHz frequencies). The trace in figure 6 should then be interpreted as deterministic trajectories observed in the phase space composed of two attractors (the two potential valleys, or equilibrium states) and the barrier. In this context, the TLF signal indicates that the magnetization is stuck very close to the attractors, and there is no intermediate trajectory (i.e. large precession angles, averaged over the time, do not exist). The precession angle is always small, except during a very small fraction of time during the jump (the time resolution in reference [28, 29] is of the order of a few nanoseconds). Furthermore, and as already mentioned, the equilibrium states (the attractors) are not modified by the current. Within this context, the precession induces *intermittency*, i.e. a highly specific chaotic behaviour in which the time spent out of the attractors is negligible. Such behaviour is discussed in detail elsewhere, in terms of spin torque hypothesis [124, 125]. This interpretation is however avoided here because the measured traces (figure 6) mimic exactly the full stochastic process composed of the two-level fluctuation (that follows the Néel–Brown activation law), and *the same white noise for the two quasi-static states* [123]. In the case of precession, the precession angle would depend on the current, so the two stationary states should also depend on the current. This is not observed in the TLF signal, or in the hysteresis loops measured as a function of the current amplitude. Furthermore the observation of identical noise in the

two states is in contradiction with published simulations performed with the hypothesis of spin transfer torque [124, 125].

According to the above analysis of the consequences of the spin torque hypothesis, we conclude that another hypothesis should be invoked in order to account for the observed signal, in the activation regime. In the following, we focus our attention on an alternative approach of the effect of the current, namely the *dissipative, or irreversible, spin transfer* effect, for which the spin relaxation is a source term for the magnetization.

### 4.3. Open ferromagnetic systems

According to the description of electronic relaxations given in section 2, it is natural to account for the presence of current dependent terms in the LLG equation exclusively due to this relaxation, i.e. without the need to introduce a deterministic term  $\vec{X}$  into the dynamical equation.

In order to describe the ferromagnetic layer coupled to the spin dependent electric subsystem in terms of open systems, we rewrite the conservation laws of the ferromagnet (equation (55)) adding the source term  $\dot{\Psi}$  due to spin transfer (see figure 10):

$$\begin{aligned}\frac{\partial n^{\text{ferro}}}{\partial t} &= -\text{div}(\vec{J}_n) + \dot{\Psi}(J_e) \\ \frac{\partial n^{\text{elec}}}{\partial t} &= -\text{div}(\delta\vec{J}) - \dot{\Psi}(J_e).\end{aligned}\quad (94)$$

According to the first law of thermodynamics, the contribution due to the electronic relaxation  $\dot{\Psi}$  should be taken into account in the entropy production of the ferromagnet. Inserting equation (94) into (51), the entropy production is

$$\begin{aligned}\vec{J}_s &= \frac{1}{T} \vec{J}_e - \frac{\tilde{\mu}}{T} \vec{J}_n \\ \mathcal{I} &= \vec{J}_e \cdot \text{grad}\left(\frac{1}{T}\right) - \vec{J}_n \cdot \text{grad}\left(\frac{\tilde{\mu}}{T}\right) + \frac{\Delta\mu(J_e)}{T} \dot{\Psi}(J_e)\end{aligned}\quad (95)$$

where  $\Delta\mu(J_e) = \mu_{\text{elec}} - \tilde{\mu}$  (e.g.  $\Delta\mu = \mu_{s\downarrow} - \mu_{d\downarrow}$ ). The Onsager transport equations are modified accordingly. The modification due to the contribution of the electronic relaxation is taken into account by a third relaxation term (we still assume  $T$  constant):

$$\begin{aligned}J_n^\theta &= -\mathcal{L}_{\theta\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - \mathcal{L}_{\theta\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} + L_{\theta\text{ch}} \Delta\mu \\ J_n^\phi &= -\mathcal{L}_{\phi\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - \mathcal{L}_{\phi\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} + L_{\phi\text{ch}} \Delta\mu \\ \dot{\Psi}(J_e) &= L_{\text{ch}\theta} \frac{\partial \tilde{\mu}}{\partial \theta} + L_{\text{ch}\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} + L \Delta\mu\end{aligned}\quad (96)$$

where  $\Delta\mu$  (a function of the current density  $J_e$  described in the first part of this report), is defined as the chemical affinity of the corresponding reaction, or equivalently, is defined as the pumping force associated with the flux  $\dot{\Psi}$  of spins transferred between the spin polarized electric system and the ferromagnet. In other words,  $\Delta\mu$  is the force responsible for irreversible spin transfer. The Onsager cross-coefficients  $L_{\theta\text{ch}}$  (and  $L_{\text{ch}\theta}$ ) and  $L_{\phi\text{ch}}$  (and  $L_{\text{ch}\phi}$ ) have been introduced for the sake of generality in order to account for the dependence of the relaxation mechanisms (e.g. spin accumulation) on the magnetic states or inversely. These coupling terms in the equations (96) are responsible for second-order effects, that will not be discussed in the

present report: the cross-terms proportional to the transport coefficients  $L_{\theta\text{ch}}$ ,  $L_{\text{ch}\theta}$ ,  $L_{\phi\text{ch}}$  and  $L_{\text{ch}\phi}$  will be neglected in the following;

$$\begin{aligned} J_n^\theta &= -\mathcal{L}_{\theta\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - \mathcal{L}_{\theta\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} \\ J_n^\phi &= -\mathcal{L}_{\phi\theta} \frac{\partial \tilde{\mu}}{\partial \theta} - \mathcal{L}_{\phi\phi} \frac{1}{\sin \theta} \frac{\partial \tilde{\mu}}{\partial \phi} \\ \dot{\Psi}(J_e) &= L \quad \Delta\mu(J_e). \end{aligned} \quad (97)$$

Inserting in the continuity equation and integrating over the whole ferromagnetic layer,

$$\frac{\partial n_{(\theta,\phi,t)}}{\partial t} = -\text{div } \vec{J}_n + \int_A^B \dot{\Psi}(z) dz. \quad (98)$$

The Fokker–Planck equation, equation (56), can be rewritten as

$$\frac{\partial n^{\text{ferro}}}{\partial t} = \frac{\partial n_0}{\partial t} + \int_A^B L \Delta\mu(z) dz = 0 \quad (99)$$

where the first term in the right-hand side  $\frac{\partial n_0}{\partial t}$  leads to the standard rotational Fokker–Planck equation (defined by the second equation in equations (62)), and the second term is the contribution coming from the electronic relaxation. *This equation is the main result defining the irreversible spin transfer effect for an open system.* Equation (99) will not be solved here. The aim of the following developments is to define negative damping and effective temperature.

The dynamic equation is obtained in the same way as in the previous section, by writing the flux of representative points on the unit sphere. The relation  $\vec{J} = n d\vec{u}_r/dt$ , equation (63), is not modified by adding the source term (the magnetic flux is not modified because the cross-coefficients are neglected in equation (97)). However, the density  $n(I)$  and diffusion  $\nabla n(I)$  are now current dependent. This can be described in terms of the fluctuating field  $\vec{H}_{\text{eff}}(I)$ , like in equations (64) and (66). As a consequence, the form of the LLG equation is not modified with spin transfer.

Since the effect of the environment conserves the modulus of the magnetization, i.e.  $\vec{u} \cdot d\vec{u}/dt = 0$  [132], the contribution of  $\frac{d\vec{u}}{dt}^{\text{elec}}$  reduces to the two damping factors, parallel to  $\vec{u} \times \nabla V^{\text{mag}}$  and parallel to  $\vec{u} \times (\vec{u} \times \nabla V^{\text{mag}})$ . After averaging, this necessarily leads to the introduction of two parameters  $\alpha_1$  and  $\alpha_2$  such that

$$\frac{d\vec{u}}{dt} = -\alpha_1 \vec{u} \times (\nabla V^{\text{mag}}) + \alpha_2 \vec{u} \times [\vec{u} \times (\nabla V^{\text{mag}})] \quad (100)$$

where the new coefficients  $\alpha_1$  and  $\alpha_2$  account for the modification of the magnetic transport coefficients  $g'$  and  $h'$  due to spin transfer  $\int_A^B L \cdot \Delta\mu(z) dz$ , and can be thought of as *negative damping* or *positive damping*, depending on whether the spin transfer integrated over the whole layer with the two junctions is transferred from the electric system to the magnetic system (negative damping) or inversely (positive damping). In other words, it depends on the balance of spin accumulation at the two interfaces of the ferromagnetic layer. Note that as long as the damping coefficients are not explicitly defined, the validity of the argument used above (Callen’s argument [132]) is not restricted to the relaxation and spin accumulation mechanisms described in the first sections of this work, but is much more general. Beyond Callen’s argument proposed above, the proper derivation of the reduction from equation (99) to (100) (see e.g. [46]) is still to be performed on the basis of a projection operator formalism [128–131].

#### 4.4. Effective thermostat

The Néel–Brown activation laws describe out-of-equilibrium spin systems ( $\partial \tilde{\mu} \neq 0$ ), and are valid for high enough potential barriers  $kT \ll vV^{\text{mag}}$ , or long timescales  $\Delta t/\tau_0 \gg 1$ , where  $\Delta t$

is the measurement time window, and  $\tau_0$  is the relaxation timescale that describes the coupling to the lattice. If the volume  $v$  tends to zero, the energy barrier decreases down to a value such that  $vV^{\text{mag}} \leq kT \ln(\Delta t/\tau_0)$ , and the magnetization is at ‘equilibrium’ with the lattice for the measurement time window  $\Delta t$ . The system is not now metastable but *superparamagnetic*. The equilibrium imposes the condition  $\partial\tilde{\mu} = 0$ , and the statistical distribution is the Maxwell–Boltzmann distribution  $n = N_t \exp(-vV^{\text{mag}}/kT)$ . The magnetization behaves like a paramagnetic spin, with the ferromagnetic order parameter  $\vec{M}$  instead of the spin  $\vec{s}$ . In the case of 3d metallic ferromagnetic nanostructures (Co, Ni, Fe . . .) of sizes typically around 5–10 nm radius, the system is superparamagnetic at room temperature for usual timescales of magnetometric measurements (above  $10^{-4}$  s). The system is nevertheless ferromagnetic and follows the Néel–Brown laws if the measurements are performed in a shorter time window  $\Delta t$  (from microseconds to nanoseconds in the case of  $10 \text{ nm}^3$  particles invoked above).

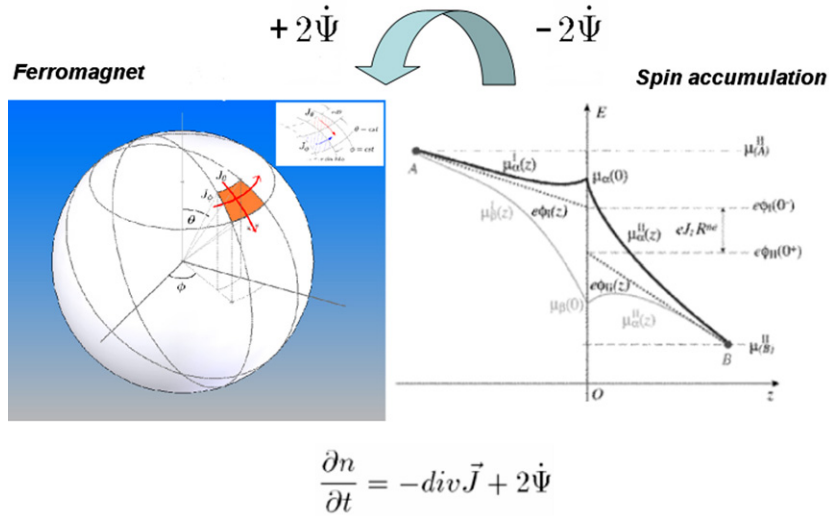
As mentioned in the previous section, if the relevant time window is shorter than the typical ferromagnetic relaxation timescale  $\tau_0$  ( $\Delta t \leq \tau_0$ ), the precession now governs the quasi-ballistic dynamics, which is qualitatively different (because it is not driven by the fluctuations). In the linear regime, the collective modes measured are that observed with ferromagnetic resonance; e.g. dynamics of thermal spin waves are observed in GMR structures [97–101]. It is no longer activated, whatever the potential barrier and the volume  $v$ , and the thermalization process vanishes at short timescales (the ‘quasi-ballistic magnetization reversal’ regime [100]). The temperature of the system (if any [133]) is not necessarily the temperature of the lattice  $T_{\text{eff}} \neq T$ : the system is decoupled from the heat bath. A similar situation justified the introduction of the concept of spin temperature  $T_s$  in the early 1950s with the first spin resonance experiments [134]. Note that if the system is also isolated from other sub-systems at comparable timescales, has an upper bound, and if the populations (up and down) can be inverted (like in nuclear spin systems), the spin temperature of the system can even be negative [135, 136] (but the spin temperature is usually positive and higher than the lattice temperature [136]).

In the situation of interest, with spin polarized currents in 3d metallic nanostructures, the ferromagnetic order parameter is coupled to the lattice through the polarization of the electronic degrees of freedom [127, 137, 138]. Without being coupled to the magnetization (e.g. in the non-magnetic side of a junction), the spin accumulation sub-system relaxes toward equilibrium with a relaxation time  $\tau_{\text{sf}}$  of some picoseconds (as described in the first section with the two-channel model). This relaxation time is shorter than the thermalization of the magnetization with the lattice  $\tau_0$  (nanoseconds). On the other hand, the coupling with the ferromagnetic sub-systems corresponds to a relaxation time  $\tau_{\text{sd}}$  (the period between the electronic relaxation time  $\tau_e$  and  $\tau_{\text{sf}}$ ) shorter than  $\tau_0$ : as a consequence, this relaxation ‘thermalizes’ the ferromagnetic order parameter with the spin accumulation sub-system, which takes the role of the heat bath in place of the lattice. This picture is that schematized in figure 1 in the first section.

In the activation regime, it is possible to assume that the spin accumulation sub-system is a reservoir of energy, and that the ferromagnetic order parameter is thermalized with it (see figure 1). Because the spin accumulation sub-system is not thermalized with the lattice, *the zeroth law of thermodynamics is not valid* [133], and it is possible to identify it as a thermostat at temperature  $T_{\text{eff}}$  in equilibrium with the ferromagnetic system.

The equilibrium condition imposes that  $\mathcal{I} = 0$  [45]. The entropy production  $\mathcal{I}$  of the spin dependent electric sub-system was calculated in section 2. With the temperature  $T_{\text{eff}}$  corresponding to the effective equilibrium temperature, the chemical potential can be written as [43]  $\Delta\tilde{\mu}^{\text{eff}} = \Delta\mu^{\text{eff}} + kT_{\text{eff}} \ln(n_\alpha/n_\gamma)$ .

$$T_{\text{eff}} \cdot \mathcal{I} = \left( -\frac{\partial \Delta\tilde{\mu}^{\text{eff}}}{\partial z} + 2\epsilon \Delta\tilde{\mu}^{\text{eff}} \right) \delta J + \Delta\tilde{\mu}^{\text{eff}} \dot{\Psi} = 0 \quad (101)$$



**Figure 10.** Under spin injection, the ferromagnetic layer is an open system coupled to the spin accumulation reservoir. The variation of the density of magnetic moments (oriented at a given direction in the unit sphere) is given by the divergence of the magnetic flux added to the correction due to the relaxation rate  $\dot{\Psi}$  of spins relaxing from one system to the other at the interfaces.

where the Joule heating contribution  $-\frac{\partial \mu_0}{\partial z} J_0$  has been removed because it does not contribute to the magnetic system and is coupled to the lattice (the whole analysis should also include the Peltier effects: energy can also be extracted from the lattice to the magnetic system).

The entropy production vanishes for the following sufficient condition:

$$\Delta \tilde{\mu}^{\text{eff}} = 0. \tag{102}$$

The condition of equation (102) leads to the expression for the equilibrium temperature  $T_{\text{eff}}$ :

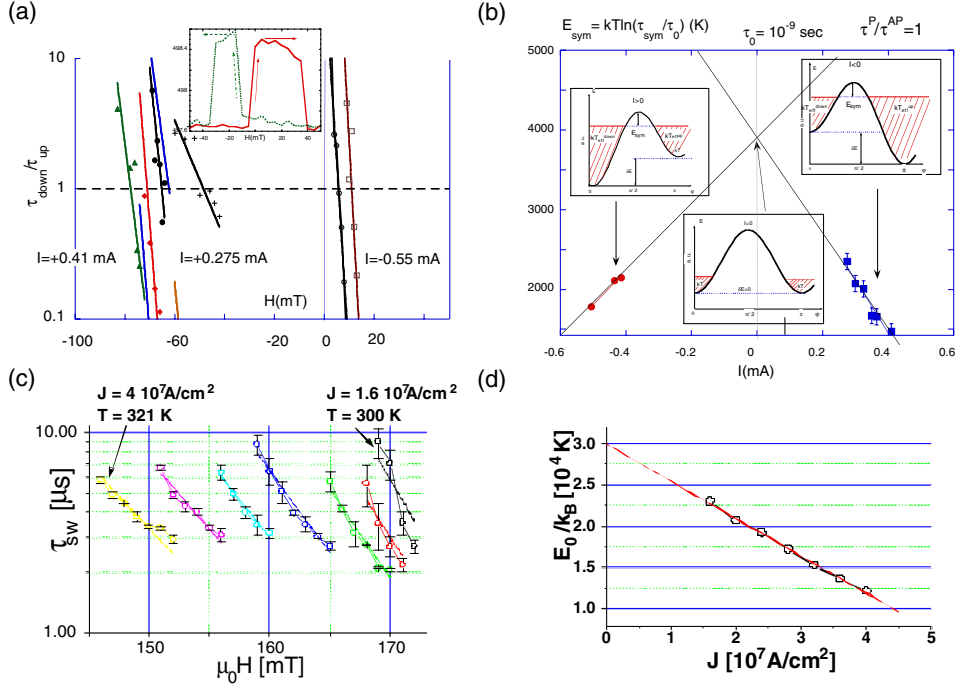
$$kT_{\text{eff}} = -\frac{\Delta \mu}{\ln(n_\alpha/n_\gamma)} \approx -2\Delta \mu \frac{n_0}{\delta n} \tag{103}$$

where the inversion of population implies that  $\delta n \leq 0$ . This equation is simply the equilibrium Curie–Weiss law that accounts for the paramagnetic behaviour of the spin accumulation  $g\mu_B \delta n$ , i.e. the first-order approximation of the averaging over the Boltzmann distribution at temperature  $T_{\text{eff}}$  ( $\delta n$  is the s–d spin accumulation that would be measured with a lattice temperature  $T_{\text{eff}}$ ). The evaluation of  $\delta n$  would necessitate the non-equilibrium distribution at temperature  $T$  being calculated. This task is beyond the scope of the present review. However, according to the evaluation performed below, an energy  $kT_{\text{eff}} \approx 1$  eV can be expected from calculating the ferromagnetic energy for a current of 1 mA due to the spin transfer in the internal field of 1 T for the ferromagnet.

A fundamental consequence of the existence of the effective temperature is that the solution of the stochastic equation for the magnetization is known, and is given by the standard activation equation (75) with the effective temperature  $T_{\text{eff}}$  instead of the lattice temperature  $T$ :

$$\frac{\partial N}{\partial \theta} + \frac{1}{kT_{\text{eff}}} \frac{\partial V}{\partial \theta} N = \frac{I_{\text{eff}}}{2k'_{\text{eff}} \pi \sin(\theta)} \tag{104}$$

where  $I_{\text{eff}}$  is calculated with the Boltzmann distribution with the ferromagnetic energy  $\exp(V(\theta)/kT_{\text{eff}})$ . Assuming a constant effective temperature  $T_{\text{eff}}(\theta) \approx T_{\text{eff}}$ , the equation is



**Figure 11.** Observation of the Néel–Brown activation due to current injection in a Co/Cu/Co trilayer ((a), (b)) and in a Ni nanowire ((c), (d)). (a) Ratio of the two relaxation times (TLF) and (c) relaxation time as a function of the applied field for different currents fitted with the Néel–Brown formula and the effective barrier height as the fitting parameter. ((b), (d)) Variation of the effective barrier height in kelvins. In the case of the two-level fluctuations, the barrier is measured for the symmetric relaxation times (b) (i.e. for different applied fields). The effects of the fluctuations are sketched in the insets (dashed lines). Reprinted with permission from [26] and Ph Guittienne *et al* [29]. Copyright ©American Physical Society.

formally identical to equation (75), so the Néel–Brown activation formula is recovered with  $T_{\text{eff}}$  instead of  $T$ :

$$\begin{aligned}\tau(\theta_1) &= \tau_{01} e^{\left(\frac{\Delta V_1}{kT_{\text{eff}}}\right)} \\ \tau(\theta_2) &= \tau_{02} e^{\left(\frac{\Delta V_2}{kT_{\text{eff}}}\right)}\end{aligned}\quad (105)$$

where  $\tau_{01}$  and  $\tau_{02}$  contain the  $k'_{\text{eff}}$  dependence. This behaviour is experimentally observed [2, 25–28]. Figure 11 shows the typical Néel–Brown activation observed with the sample shown in figures 8(b) and (c). The fit of the mean relaxation time as a function of the applied field and the current amplitude injected in the device is performed with the Néel–Brown formula with the effective barrier height as the fitting parameter (figures 11(a) and (c)). The effective barrier height as a function of the current is presented in figures 11(c) and (d).

Assuming that the mechanism responsible for  $T_{\text{eff}}$  is the spin accumulation occurring at the interface composed of anti-aligned ferromagnets, the voltage drop due to spin accumulation is approximately equal to  $\Delta\mu$  (see equation (18)):

$$kT_{\text{eff}} \approx -\frac{\Delta R^{\text{sa}} I}{\ln(n_\alpha/n_\gamma)} \propto \frac{\Delta R^{\text{sa}} I}{2}\quad (106)$$



with  $-\ln(n_\alpha/n_\gamma) \leq 1$ . The proportionality of  $kT_{\text{eff}}$  and  $R^{\text{sa}}$  was observed by different groups in recent experimental investigations [140, 141] (with DC measurements, the parameter is the ‘critical current’  $I_c \propto T_{\text{eff}}^{-1}$  as shown in figure 11(d)).

How do we estimate the magnetic energy of the spin accumulation system in usual experimental situations, where a current density of some few mA is injected in the nanostructure [1, 2, 4, 9, 25–29]? This current corresponds to some  $10^{16}$  spin  $\text{s}^{-1}$  flowing through the interface. If one assumes that 20% (polarization of the current) of the spins are maintained out of equilibrium within a typical relaxation time  $\tau_{\text{sf}}$  of  $10^{-11}$  s, we are left with about  $2 \times 10^4$  spins that define the magnetization of the spin accumulation sub-system in the volume defined by the corresponding diffusion length. An effect of the electric spin relaxation on the ferromagnetic order parameter should consequently be expected for a nanostructured ferromagnetic system that is only ten to a hundred times larger. In an internal field of  $H_{\text{int}} = 1$  T, this energy  $E = 10^4 \mu_B H_{\text{int}}$  is of the order of 1 eV (beyond the Curie temperature) in accordance with activation experiments performed on various systems [2, 25–29]. Without current injection, the magnetic order parameter is at room temperature and, consequently, the hot sub-system is the spin accumulation system.

## 5. Conclusion

A unified thermokinetics approach involving both spin dependent charge carriers and ferromagnetic Brownian motion has been presented in the context of open systems. The spin dependent electronic relaxation is then introduced as a source term in the conservation equation for the magnetic moment. This leads to the description of the effect of spin injection induced magnetization switching, or irreversible spin transfer in an open ferromagnetic layer.

The description of the spin accumulated charge carriers is based on the two-conduction-channel approximation, generalized to both intraband and interband relaxation. The application of the first and second laws of thermodynamics, together with the conservation laws, leads to the spin dependent transport equations. The relevant Onsager transport coefficients are introduced and related to the typical electronic relaxation times. The effect of charge conservation and screening is also taken into account.

On the other hand, the ferromagnetic order parameter is described on an equal footing by introducing the conservation laws and the relevant chemical potential, with deterministic terms accounting for the effective field, and dissipative terms accounting for the coupling to a relevant heat bath. The corresponding Onsager transport coefficients are defined with the second law of thermodynamics, and refined with the help of the Onsager reciprocal relations. The rotational Fokker–Planck equation and the Landau–Lifshitz–Gilbert (LLG) equation are then derived within the thermokinetic theory. The Onsager coefficients are related to the typical timescales of the ferromagnetic relaxation ( $\tau_0$ ). In the activation regime, the Néel–Brown activation law is deduced.

In the framework of this description, the generalization of both the Fokker–Planck equation and the LLG equation together with the contribution of spin accumulation is straightforward in terms of the flux of representative points in the magnetization sphere. The negative damping appears naturally for describing the exchange of spins from the electric sub-system to the magnetic sub-system, described as a coupling to an environment.

Furthermore, the discussion concerning the different relaxation times shows that the spin polarized current is not thermalized to the lattice in the stationary regime, but is thermalized with the spin accumulation sub-system. The argument is that on one hand the relaxation toward equilibrium of the spin accumulation system (described with  $\tau_{\text{sf}}$ : some tens to hundreds of picoseconds) is shorter than the thermalization of the ferromagnetic system (described with  $\tau_0$ :

nanoseconds), and on the other hand, the coupling between the ferromagnetic order parameter and the spin accumulation sub-system ( $\tau_{sd}$ ) is smaller than or equal to  $\tau_{sf}$ .

An effective temperature is then derived in the activation regime through the entropy production, and this leads to the derivation of an effective Néel–Brown relaxation process due to current injection, that is experimentally observed.

## Acknowledgments

We are indebted to Professor Peter Levy for constant interest throughout this work and for a critical reading of the manuscript.

## Appendix. Microscopic approach and thermokinetic coefficients

### A.1. Relation between $L$ and the electronic relaxation times

Let us consider a simple interface between two metals. Far from the interface, Ohm's law is recovered: the chemical potentials of the channels are identical and the electric distribution is that of equilibrium  $n_{\alpha 0}$  and  $n_{\gamma 0}$ . In the following, we assume that the charge transfer between the two channels can be described with the following relation:

$$f \Delta n_{\alpha}(x) + g \Delta n_{\gamma}(x) = 0. \quad (\text{A.1})$$

The case  $f = g = 1$  describes the local electrical neutrality. We have

$$\begin{aligned} \Delta n_{\alpha}(x) &= (\mu_{\text{ch},\alpha} - \mu^0) N_{\alpha}(E_{\text{F}}) \\ \Delta n_{\gamma}(x) &= (\mu_{\text{ch},\gamma} - \mu^0) N_{\gamma}(E_{\text{F}}) \end{aligned} \quad (\text{A.2})$$

where  $\mu^0$  is the chemical potential in the absence of charge transfer and  $\mu_{\text{ch},\alpha}$  and  $\mu_{\text{ch},\gamma}$  are the purely chemical potentials of the channels (without transfer  $\mu_{\text{ch},\alpha} = \mu_{\text{ch},\gamma} = \mu^0$ );  $N_{\alpha,\gamma}(E_{\text{F}})$  is the density of states at the Fermi level. The separation between the electric potential and the purely chemical potential can be written as

$$\begin{aligned} \mu_{\alpha} &= \mu_{\text{ch},\alpha} + eV \\ \mu_{\gamma} &= \mu_{\text{ch},\gamma} + eV \end{aligned} \quad (\text{A.3})$$

where  $V$  is the local electric potential.

Relation (A.1) gives

$$(\mu_{\text{ch},\alpha} - \mu^0) f N_{\alpha}(E_{\text{F}}) + (\mu_{\text{ch},\gamma} - \mu^0) g N_{\gamma}(E_{\text{F}}) = 0. \quad (\text{A.4})$$

Note that the expressions for  $\Delta n_{\alpha,\gamma}$  come from the fact that at zero kelvins,

$$\begin{aligned} n_{\alpha}(\mu_{\alpha}) &= \int N_{\alpha}(E) f(E) dE = \int_{-\infty}^{\mu_{\text{ch},\alpha}} N_{\alpha}(E) f(E) dE \\ n_{\gamma}(\mu_{\gamma}) &= \int N_{\gamma}(E) f(E) dE = \int_{-\infty}^{\mu_{\text{ch},\gamma}} N_{\gamma}(E) f(E) dE. \end{aligned} \quad (\text{A.5})$$

From these relations we deduce

$$\begin{cases} \Delta n_\alpha = N_\alpha(E_F) \delta\mu_{\text{ch},\alpha} \\ \Delta n_\gamma = N_\gamma(E_F) \delta\mu_{\text{ch},\gamma} = -\frac{f}{g} \Delta n_\alpha \end{cases} \Rightarrow \begin{cases} \Delta n_\alpha = \frac{g N_\alpha(E_F) N_\gamma(E_F)}{f N_\alpha(E_F) + g N_\gamma(E_F)} \delta\mu_{\text{ch},\alpha} \\ \Delta n_\gamma = -\frac{f N_\alpha(E_F) N_\gamma(E_F)}{f N_\alpha(E_F) + g N_\gamma(E_F)} \delta\mu_{\text{ch},\alpha} \end{cases} \quad (\text{A.6})$$

where  $\delta\mu_{\alpha\gamma} = \mu_{\text{ch},\alpha\gamma} - \mu_0$ .

Introducing the global transfer rate  $T_{\alpha\rightarrow\gamma}$  ( $T_{\gamma\rightarrow\alpha}$ ) of the channel  $\alpha$  to  $\gamma$  ( $\gamma$  to  $\alpha$ ), the charge conservation between the two channels can be written as ( $e < 0$ )

$$\begin{aligned} \frac{\partial n_\alpha(x)}{\partial t} &= -\frac{1}{e} \frac{\partial J_\alpha(x)}{\partial x} - T_{\alpha\rightarrow\gamma}(n_\alpha(x), n_\gamma(x)) + T_{\gamma\rightarrow\alpha}(n_\alpha(x), n_\gamma(x)) \\ \frac{\partial n_\gamma(x)}{\partial t} &= -\frac{1}{e} \frac{\partial J_\gamma(x)}{\partial x} + T_{\alpha\rightarrow\gamma}(n_\alpha(x), n_\gamma(x)) - T_{\gamma\rightarrow\alpha}(n_\alpha(x), n_\gamma(x)) \end{aligned} \quad (\text{A.7})$$

which leads, in the stationary regime  $\frac{\partial n_{\alpha,\gamma}(x)}{\partial t} = 0$ , to the following relations:

$$\begin{aligned} \frac{\partial J_\alpha(x)}{\partial x} &= -e T_{\alpha\rightarrow\gamma}(n_\alpha(x), n_\gamma(x)) + e T_{\gamma\rightarrow\alpha}(n_\alpha(x), n_\gamma(x)) \\ \frac{\partial J_\gamma(x)}{\partial x} &= +e T_{\alpha\rightarrow\gamma}(n_\alpha(x), n_\gamma(x)) - e T_{\gamma\rightarrow\alpha}(n_\alpha(x), n_\gamma(x)). \end{aligned} \quad (\text{A.8})$$

The Taylor expansion to leading order of the transfer rates  $T_{\alpha\rightarrow\gamma}$  and  $T_{\gamma\rightarrow\alpha}$ , around equilibrium gives

$$\begin{aligned} \frac{\partial J_\alpha(x)}{\partial x} &= -e T_{\alpha\rightarrow\gamma}(n_\alpha^0, n_\gamma^0) - e \frac{\partial T_{\alpha\rightarrow\gamma}}{\partial n_\alpha} \Delta n_\alpha - e \frac{\partial T_{\alpha\rightarrow\gamma}}{\partial n_\gamma} \Delta n_\gamma \\ &\quad + e T_{\gamma\rightarrow\alpha}(n_\alpha^0, n_\gamma^0) + e \frac{\partial T_{\gamma\rightarrow\alpha}}{\partial n_\alpha} \Delta n_\alpha + e \frac{\partial T_{\gamma\rightarrow\alpha}}{\partial n_\gamma} \Delta n_\gamma \end{aligned} \quad (\text{A.9})$$

$$\frac{\partial J_\gamma(x)}{\partial x} = -\frac{\partial J_\alpha(x)}{\partial x}.$$

At equilibrium, the current of each channel is conserved, so

$$-e T_{\alpha\rightarrow\gamma}(n_\alpha^0, n_\gamma^0) + e T_{\gamma\rightarrow\alpha}(n_\alpha^0, n_\gamma^0) = 0. \quad (\text{A.10})$$

Defining the electronic relaxation times  $\tau_{\alpha\rightarrow\gamma}$ ,  $\tau_{\gamma\rightarrow\alpha}$ , such that

$$\begin{aligned} \frac{1}{\tau_{\alpha\rightarrow\gamma}} &= \frac{\partial(T_{\alpha\rightarrow\gamma} - T_{\gamma\rightarrow\alpha})}{\partial n_\alpha} \Big|_{(n_\alpha^0, n_\gamma^0)} \\ \frac{1}{\tau_{\gamma\rightarrow\alpha}} &= \frac{\partial(T_{\gamma\rightarrow\alpha} - T_{\alpha\rightarrow\gamma})}{\partial n_\gamma} \Big|_{(n_\alpha^0, n_\gamma^0)} \end{aligned} \quad (\text{A.11})$$

we have

$$\begin{cases} \frac{\partial J_\alpha(x)}{\partial x} = -e \frac{\Delta n_\alpha}{\tau_{\alpha\rightarrow\gamma}} + e \frac{\Delta n_\gamma}{\tau_{\gamma\rightarrow\alpha}} \\ \frac{\partial J_\gamma(x)}{\partial x} = -\frac{\partial J_\alpha(x)}{\partial x} \end{cases} \quad (\text{A.12})$$

$$\Rightarrow \begin{cases} \frac{\partial J_\alpha(x)}{\partial x} = -e \frac{N_\alpha(E_F) N_\gamma(E_F)}{f N_\alpha(E_F) + g N_\gamma(E_F)} \left( \frac{g}{\tau_{\alpha\rightarrow\gamma}} + \frac{f}{\tau_{\gamma\rightarrow\alpha}} \right) (\mu_{\text{ch},\alpha} - \mu_{\text{ch},\gamma}) \\ \frac{\partial J_\gamma(x)}{\partial x} = -\frac{\partial J_\alpha(x)}{\partial x}. \end{cases} \quad (\text{A.13})$$

The above equations can be rewritten in the following form:

$$\begin{aligned} \frac{\partial J_\alpha(x)}{\partial x} &= -L (\mu_{\text{ch},\alpha} - \mu_{\text{ch},\gamma}) = -L (\mu_\alpha - \mu_\gamma) \\ \frac{\partial J_\gamma(x)}{\partial x} &= +L (\mu_{\text{ch},\alpha} - \mu_{\text{ch},\gamma}) = +L (\mu_\alpha - \mu_\gamma) \end{aligned} \quad (\text{A.14})$$

where the Onsager transport coefficient  $L$  is related to the electronic relaxation times by the following relation:

$$L = e \frac{N_\alpha(E_F)N_\gamma(E_F)}{fN_\alpha(E_F) + gN_\gamma(E_F)} \left( \frac{g}{\tau_{\alpha \rightarrow \gamma}} + \frac{f}{\tau_{\gamma \rightarrow \alpha}} \right)$$

The form of equation (A.14) is that of (9) deduced from the thermokinetic approach in section 1, where the coefficient  $L$  is the Onsager coefficient defined in the third of equations (11).

### A.2. Determination of $f$ and $g$

We have, for the stationary regime,

$$\frac{\partial(J_\alpha(x) - J_\gamma(x))}{\partial x} = -2L(\mu_\alpha - \mu_\gamma). \quad (\text{A.15})$$

Furthermore, the local Ohm's law applied to each channel leads to the following equations:

$$\begin{cases} J_\alpha = -\frac{\sigma_\alpha}{e} \frac{\partial \mu_\alpha(x)}{\partial x} \\ J_\gamma = -\frac{\sigma_\gamma}{e} \frac{\partial \mu_\gamma(x)}{\partial x} \end{cases} \Rightarrow \begin{cases} \frac{\partial J_\alpha(x)}{\partial x} = -\frac{\sigma_\alpha}{e} \frac{\partial^2 \mu_\alpha(x)}{\partial x^2} \\ \frac{\partial J_\gamma(x)}{\partial x} = -\frac{\sigma_\gamma}{e} \frac{\partial^2 \mu_\gamma(x)}{\partial x^2} \end{cases} \quad (\text{A.16})$$

where we assume that the conductivities are constant.

From (A.14) and (A.16), we get

$$\begin{cases} \frac{\partial^2 \mu_\alpha(x)}{\partial x^2} = \frac{eL}{\sigma_\alpha} (\mu_\alpha - \mu_\gamma) \\ \frac{\partial^2 \mu_\gamma(x)}{\partial x^2} = -\frac{eL}{\sigma_\gamma} (\mu_\alpha - \mu_\gamma) \end{cases} \Rightarrow \frac{\partial^2 (\mu_\alpha - \mu_\gamma)}{\partial x^2} = eL \left( \frac{1}{\sigma_\alpha} + \frac{1}{\sigma_\gamma} \right) (\mu_\alpha - \mu_\gamma) \quad (\text{A.17})$$

which leads to the well known diffusion equation for the chemical potential that describes the *spin accumulation* process:

$$\frac{\partial^2 \Delta \mu}{\partial x^2} = eL \left( \frac{1}{\sigma_\alpha} + \frac{1}{\sigma_\gamma} \right) \Delta \mu \quad (\text{A.18})$$

from which the spin diffusion length  $l_{\text{sf}}$  is deduced:

$$\frac{1}{l_{\text{sf}}^2} = eL \left( \frac{1}{\sigma_\alpha} + \frac{1}{\sigma_\gamma} \right). \quad (\text{A.19})$$

From equation (A.17) we have the differential equations

$$\begin{aligned} \frac{\partial^2 \mu_\alpha(x)}{\partial x^2} &= \frac{eL}{\sigma_\alpha} \Delta \mu = \frac{\sigma_\gamma}{\sigma_\alpha} \frac{\partial^2 \Delta \mu}{\partial x^2} \\ \frac{\partial^2 \mu_\gamma(x)}{\partial x^2} &= -\frac{eL}{\sigma_\gamma} \Delta \mu = -\frac{\sigma_\alpha}{\sigma_\gamma} \frac{\partial^2 \Delta \mu}{\partial x^2}. \end{aligned} \quad (\text{A.20})$$

### A.3. Charge distribution and screening

Separating the electric contribution from the chemical contribution, the electrochemical potential can be written as

$$\mu_{\alpha,\gamma} = \mu_{\text{ch},\alpha,\gamma} + eV, \quad (\text{A.21})$$

so

$$\left\{ \begin{array}{l} \frac{\partial^2 \mu_\alpha(x)}{\partial x^2} = \frac{\partial^2 \mu_{\text{ch},\alpha}(x)}{\partial x^2} + e \frac{\partial^2 V(x)}{\partial x^2} \\ \frac{\partial^2 \mu_\gamma(x)}{\partial x^2} = \frac{\partial^2 \mu_{\text{ch},\gamma}(x)}{\partial x^2} + e \frac{\partial^2 V(x)}{\partial x^2} \end{array} \right\} \Rightarrow \left\{ \begin{array}{l} \frac{\partial^2 \mu_{\text{ch},\alpha}(x)}{\partial x^2} = \frac{\sigma_\gamma}{\sigma_t} \frac{\partial^2 \Delta\mu}{\partial x^2} + e^2 \frac{\Delta n_\alpha + \Delta n_\gamma}{\epsilon} \\ \frac{\partial^2 \mu_{\text{ch},\gamma}(x)}{\partial x^2} = -\frac{\sigma_\alpha}{\sigma_t} \frac{\partial^2 \Delta\mu}{\partial x^2} + e^2 \frac{\Delta n_\alpha + \Delta n_\gamma}{\epsilon} \end{array} \right. \quad (\text{A.22})$$

where the Poisson equation has been introduced:

$$-\frac{\partial^2 V(x)}{\partial x^2} = e \frac{\Delta n_\alpha + \Delta n_\gamma}{\epsilon}. \quad (\text{A.23})$$

The equations can be rewritten as

$$\begin{aligned} \frac{\partial^2 \delta \mu_{\text{ch},\alpha}(x)}{\partial x^2} - e^2 \frac{N_\alpha \delta \mu_{\text{ch},\alpha} + N_\gamma \delta \mu_{\text{ch},\gamma}}{\epsilon} &= \frac{\sigma_\gamma}{\sigma_t} \frac{\Delta\mu}{l_{\text{sf}}^2} \\ \frac{\partial^2 \delta \mu_{\text{ch},\gamma}(x)}{\partial x^2} - e^2 \frac{N_\alpha \delta \mu_{\text{ch},\alpha} + N_\gamma \delta \mu_{\text{ch},\gamma}}{\epsilon} &= -\frac{\sigma_\alpha}{\sigma_t} \frac{\Delta\mu}{l_{\text{sf}}^2}. \end{aligned} \quad (\text{A.24})$$

These relations with

$$\mu_\alpha - \mu_\gamma = \mu_{\text{ch},\alpha} - \mu_{\text{ch},\gamma} = \delta \mu_{\text{ch},\alpha} - \delta \mu_{\text{ch},\gamma} \quad (\text{A.25})$$

lead to

$$\begin{aligned} \frac{\partial^2 \Delta \mu_{\text{ch},\alpha}(x)}{\partial x^2} - \frac{\Delta \mu_{\text{ch},\alpha}}{l^2} &= \Delta \mu \left( \frac{\sigma_\gamma}{\sigma_t l_{\text{sf}}^2} - e^2 \frac{N_\gamma}{\epsilon} \right) \\ \frac{\partial^2 \Delta \mu_{\text{ch},\gamma}(x)}{\partial x^2} - \frac{\Delta \mu_{\text{ch},\gamma}}{l^2} &= \Delta \mu \left( -\frac{\sigma_\alpha}{\sigma_t l_{\text{sf}}^2} + e^2 \frac{N_\alpha}{\epsilon} \right) \end{aligned} \quad (\text{A.26})$$

where we have introduced the screening length:

$$\frac{1}{l^2} = e^2 \frac{N_\alpha + N_\gamma}{\epsilon}. \quad (\text{A.27})$$

The solution of the equation is composed of a solution of the equation with zero right-hand side (homogeneous solution) and a particular solution.

(1) *Solution for  $\Delta \mu_{\text{ch},\alpha}$ :*

*homogeneous solution:*

$$\Delta \mu_{\text{ch},\alpha}^{\text{hmg}} = A \exp\left(\frac{x}{l}\right) + B \exp\left(-\frac{x}{l}\right); \quad (\text{A.28})$$

*particular solution:*

$$\begin{aligned} \Delta \mu_{\text{ch},\alpha}^{\text{part}} &= p_\alpha \Delta \mu \\ \Rightarrow p_\alpha \frac{\Delta \mu}{l_{\text{sf}}^2} - \frac{\Delta \mu}{l^2} &= \left( \frac{\sigma_\gamma}{\sigma_t l_{\text{sf}}^2} - e^2 \frac{N_\gamma}{\epsilon} \right) \Delta \mu \end{aligned} \quad (\text{A.29})$$

$$\Rightarrow p_\alpha = \frac{\frac{\sigma_\gamma}{\sigma_t l_{\text{sf}}^2} - \frac{e^2 N_\gamma}{\epsilon}}{\frac{1}{l_{\text{sf}}^2} - \frac{1}{l^2}}. \quad (\text{A.30})$$

(2) *Solution for  $\Delta \mu_{\text{ch},\gamma}$ :*

*homogeneous solution:*

$$\Delta \mu_{\text{ch},\gamma}^{\text{hmg}} = A' \exp\left(\frac{x}{l}\right) + B' \exp\left(-\frac{x}{l}\right); \quad (\text{A.31})$$

particular solution:

$$\begin{aligned}\Delta\mu_{\text{ch},\gamma}^{\text{part}} &= p_\gamma \Delta\mu \\ \Rightarrow p_\gamma \frac{\Delta\mu}{l_{\text{sf}}^2} - \frac{\Delta\mu}{l^2} &= \left( \frac{-\sigma_\alpha}{\sigma_{\text{t}} l_{\text{sf}}^2} + e^2 \frac{N_\alpha}{\epsilon} \right) \Delta\mu\end{aligned}\quad (\text{A.32})$$

$$\Rightarrow p_\gamma = \frac{-\frac{\sigma_\alpha}{\sigma_{\text{t}} l_{\text{sf}}^2} + \frac{e^2 N_\alpha}{\epsilon}}{\frac{1}{l_{\text{sf}}^2} - \frac{1}{l^2}}.\quad (\text{A.33})$$

The general solutions satisfying the condition

$$\Delta\mu_{\text{ch},\alpha} - \Delta\mu_{\text{ch},\gamma} = \Delta\mu\quad (\text{A.34})$$

correspond to

$$A = A' \quad B = B'.\quad (\text{A.35})$$

Inserted in the expression for the charge conservation

$$f \Delta n_\alpha + g \Delta n_\gamma = 0\quad (\text{A.36})$$

we have

$$\begin{aligned}f N_\alpha \left( A \exp\left(\frac{x}{l}\right) + B \exp\left(-\frac{x}{l}\right) + p_\alpha \Delta\mu \right) \\ + g N_\gamma \left( A \exp\left(\frac{x}{l}\right) + B \exp\left(-\frac{x}{l}\right) + p_\gamma \Delta\mu \right) = 0\end{aligned}\quad (\text{A.37})$$

$$\begin{aligned}\Rightarrow (f N_\alpha + g N_\gamma) \left( A \exp\left(\frac{x}{l}\right) + B \exp\left(-\frac{x}{l}\right) \right) + (f N_\alpha p_\alpha + g N_\gamma p_\gamma) \Delta\mu = 0, \\ \text{for all } x\end{aligned}\quad (\text{A.38})$$

$$\Rightarrow f N_\alpha p_\alpha + g N_\gamma p_\gamma = 0 \quad \text{and} \quad A = B = 0\quad (\text{A.39})$$

$$\Rightarrow f N_\alpha \left( \frac{\sigma_\gamma}{\sigma_{\text{t}} l_{\text{sf}}^2} - \frac{e^2 N_\gamma}{\epsilon} \right) + g N_\gamma \left( -\frac{\sigma_\alpha}{\sigma_{\text{t}} l_{\text{sf}}^2} + \frac{e^2 N_\alpha}{\epsilon} \right) = 0.\quad (\text{A.40})$$

A solution can be written as

$$\begin{aligned}f &= N_\gamma \left( \frac{\sigma_\alpha}{\sigma_{\text{t}} l_{\text{sf}}^2} - \frac{e^2 N_\alpha}{\epsilon} \right) \\ g &= N_\alpha \left( \frac{\sigma_\gamma}{\sigma_{\text{t}} l_{\text{sf}}^2} - \frac{e^2 N_\gamma}{\epsilon} \right).\end{aligned}\quad (\text{A.41})$$

#### A.4. $l_{\text{sf}}$ as a function of $l_\alpha$ , $l_\gamma$ and $l$

The relation between  $L$  and the electronic relaxation times has been found to be

$$L = e \frac{N_\alpha(E_{\text{F}}) N_\gamma(E_{\text{F}})}{f N_\alpha(E_{\text{F}}) + g N_\gamma(E_{\text{F}})} \left( \frac{g}{\tau_{\alpha \rightarrow \gamma}} + \frac{f}{\tau_{\gamma \rightarrow \alpha}} \right).\quad (\text{A.42})$$

Inserting the expression for  $f$  and  $g$  obtained in the previous subsection,  $L$  becomes

$$L = e \frac{\frac{1}{\sigma_{\text{t}} l_{\text{sf}}^2} \left( \frac{\sigma_\alpha N_\gamma}{\tau_{\gamma \rightarrow \alpha}} + \frac{\sigma_\gamma N_\alpha}{\tau_{\alpha \rightarrow \gamma}} \right) - e^2 \frac{N_\alpha N_\gamma}{\epsilon} \left( \frac{1}{\tau_{\alpha \rightarrow \gamma}} + \frac{1}{\tau_{\gamma \rightarrow \alpha}} \right)}{\frac{1}{l_{\text{sf}}^2} - \frac{1}{l^2}}.\quad (\text{A.43})$$

Furthermore, according to equation (A.19), the coefficient  $L$  can also be written in the following form:

$$L = \frac{\sigma_\alpha \sigma_\gamma}{e \sigma_{\text{t}} l_{\text{sf}}^2}.\quad (\text{A.44})$$

The two results lead to the equation

$$e^2 \frac{\frac{1}{\sigma_t l_{sf}^2} \left( \frac{\sigma_\alpha N_\gamma}{\tau_{\gamma \rightarrow \alpha}} + \frac{\sigma_\gamma N_\alpha}{\tau_{\alpha \rightarrow \gamma}} \right) - e^2 \frac{N_\alpha N_\gamma}{\epsilon} \left( \frac{1}{\tau_{\alpha \rightarrow \gamma}} + \frac{1}{\tau_{\gamma \rightarrow \alpha}} \right)}{\frac{1}{l_{sf}^2} - \frac{1}{l^2}} = \frac{\sigma_\alpha \sigma_\gamma}{\sigma_t l_{sf}^2}. \quad (\text{A.45})$$

Let us define the typical diffusion lengths per channel  $l_\alpha$  and  $l_\gamma$  such that

$$\begin{cases} l_\alpha^2 = \frac{\sigma_\alpha}{e^2 N_\alpha} \tau_{\alpha \rightarrow \gamma} \\ l_\gamma^2 = \frac{\sigma_\gamma}{e^2 N_\gamma} \tau_{\gamma \rightarrow \alpha} \end{cases} \Rightarrow \begin{cases} \frac{1}{\tau_{\alpha \rightarrow \gamma}} = \frac{\sigma_\alpha}{e^2 N_\alpha l_\alpha^2} \\ \frac{1}{\tau_{\gamma \rightarrow \alpha}} = \frac{\sigma_\gamma}{e^2 N_\gamma l_\gamma^2} \end{cases} \Rightarrow \begin{cases} \frac{\sigma_\alpha N_\gamma}{\tau_{\gamma \rightarrow \alpha}} = \frac{\sigma_\gamma \sigma_\alpha}{e^2 l_\gamma^2} \\ \frac{\sigma_\gamma N_\alpha}{\tau_{\alpha \rightarrow \gamma}} = \frac{\sigma_\gamma \sigma_\alpha}{e^2 l_\alpha^2} \end{cases}. \quad (\text{A.46})$$

Equation (A.45) can be rewritten as

$$\frac{\frac{\sigma_\alpha \sigma_\gamma}{e^2 \sigma_t l_{sf}^2} \left( \frac{1}{l_\gamma^2} + \frac{1}{l_\alpha^2} \right) - \frac{N_\alpha N_\gamma}{\epsilon} \left( \frac{\sigma_\alpha}{N_\alpha l_\alpha^2} + \frac{\sigma_\gamma}{N_\gamma l_\gamma^2} \right)}{\frac{1}{l_{sf}^2} - \frac{1}{l^2}} = \frac{\sigma_\alpha \sigma_\gamma}{e^2 \sigma_t l_{sf}^2} \quad (\text{A.47})$$

$$\Rightarrow \frac{\sigma_t}{N_\alpha + N_\gamma} \left( \frac{N_\gamma}{\sigma_\gamma l_\alpha^2} + \frac{N_\alpha}{\sigma_\alpha l_\gamma^2} \right) l_{sf}^4 - \left[ l^2 \left( \frac{1}{l_\gamma^2} + \frac{1}{l_\alpha^2} \right) + 1 \right] l_{sf}^2 + l^2 = 0. \quad (\text{A.48})$$

(a) *Limits*: For metals, the screening length is much smaller than the diffusion lengths of both channels:

(1)  $l/l_{\alpha\gamma} \ll 0$ :

$$\begin{aligned} & \frac{\sigma_t}{N_\alpha + N_\gamma} \left( \frac{N_\gamma}{\sigma_\gamma l_\alpha^2} + \frac{N_\alpha}{\sigma_\alpha l_\gamma^2} \right) l_{sf}^2 - 1 = 0 \\ \Rightarrow & \boxed{\frac{1}{l_{sf}^2} = \frac{N_\gamma / (N_\alpha + N_\gamma)}{\sigma_\gamma / (\sigma_\alpha + \sigma_\gamma)} \frac{1}{l_\alpha^2} + \frac{N_\alpha / (N_\alpha + N_\gamma)}{\sigma_\alpha / (\sigma_\alpha + \sigma_\gamma)} \frac{1}{l_\gamma^2}}. \end{aligned} \quad (\text{A.49})$$

The other limit gives:

(2)  $l/l_{\alpha\gamma} \Rightarrow \infty$ :

$$l^2 \left[ 1 - \left( \frac{1}{l_\alpha^2} + \frac{1}{l_\gamma^2} \right) l_{sf}^2 \right] = 0 \Rightarrow \frac{1}{l_{sf}^2} = \frac{1}{l_\alpha^2} + \frac{1}{l_\gamma^2}. \quad (\text{A.50})$$

But this second limit is not expected for usual materials.

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