Effects of spin accumulation in magnetic multilayers

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The diffusion equation for the spin accumulation in magnetic multilayers is extended to capture the timedependent buildup of spin accumulation as well as the influence of an external field and arbitrary magnetization orientations on the spin accumulation. One obtains a consistent formulation for spin accumulation that can explain such important effects as spin injection, spin filtering, and spin transfer on an equal footing. Spin transfer is determined by the amount of spin accumulation at the interfaces whereas spin filtering by the departure from collinear magnetization orientations. The noncollinear magnetization orientations lead to a spatial decay of the spin accumulation in the transverse direction of the multilayer plane. Spin filtering can thus be associated with a new length scale in the transverse direction. Moreover, spin filtering is in correspondence with a biquadratic coupling between layers and changes in the angular dependence of the giant magnetoresistance. What is interesting is that spin filtering is not a prerequisite to spin transfer. Different scenarios for current-induced magnetization reversal are discussed in which, depending largely on the geometry of the magnetic multilayer, either spin injection or spin transfer will be dominant.

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I. INTRODUCTION

Essential to electronic devices are interfaces across which the distribution of individual types of carriers has to change. In magnetic multilayers (MML's) the bulk transport characteristics in alternating magnetic and nonmagnetic layers require the distribution of spin-up and spin-down conduction electrons to change across the interface. In the presence of a current this leads to spin accumulation. The spin degree of freedom of the conduction electrons can be used in a similar fashion as electrons and holes in semiconductors, giving rise to numerous new effects such as spin injection, spin filtering, and spin transfer. To fully exploit MML's in electronic devices, it is thus important to understand the effect of spin accumulation in more detail.

The purpose of this work is then to extend earlier formulations on spin accumulation without adding to their complexity. We will focus on metal-based MML's where transport is considered to be diffusive and spin information conserved over a length scale of the order of the thickness of the layers. Building on a limiting case of combined transport equations for spin and charge in conjunction with coupled Landau-Lifshitz equations derived in a previous work,¹ we will analyze in detail the obtained diffusion equation. In particular, we will study the time development of the buildup of spin accumulation which has, at least to our knowledge, not been addressed so far. The transient behavior of spin accumulation is an important issue since ultrafast switching, required, for example, in magnetic read heads or memory devices, could in principle lead to a number of unexpected effects when one wants to use MML's with currents perpendicular to the plane of the layers (CPP) in future devices. In addition, we will discuss the effects arising from high currents and the noncollinearity of the magnetization in consecutive layers at steady state. These effects are spin injection, spin filtering, and spin transfer.

Spin accumulation is a well-studied phenomena dating back to the seminal works of Johnson and Silsbee.^{2–4} They

also gave a theoretical description based on nonequilibrium thermodynamics⁴ which was formulated independently by van Son, van Kempen, and Wyder in terms of a diffusion equations for spin-dependent chemical potentials,⁵ arriving at the same conclusions. They realized that a current across a single magnetic layer leads to the buildup of nonequilibrium magnetization δM_s —spin accumulation—at the interfaces. This arises from different scattering probabilities for spin-up and spin-down conduction electrons in the ferromagnet.

Much of the impetus to study spin accumulation in more detail was given by the discovery of giant magnetoresistance (GMR) in magnetic multilayers in the CPP geometry by Pratt *et al.*⁶ This led to a number of theoretical works that extended the basic models to describe spin accumulation. More detailed treatments have been given later on the basis of linear response theory and the Boltzmann equation, respectively.^{7,8} It could be shown that the essential features of spin accumulation are captured by the spin-dependent diffusion equation and reasonable quantitative agreement reached when spin-flip scattering at the interfaces was included in the form of boundary conditions.⁸ In general, only collinear orientations of the magnetizations in successive magnetic layers were considered.

Later, Dauguet et al. measured the angular dependence of the CPP-MR effect in MML's.⁹ Their experiments show a small but rather noticeable departure from the predicted $\cos \theta$ dependence of the CPP-MR. The problem of angular dependence of the CPP-MR was first studied by Wang et al. in a numerical band-structure calculation of a superlattice which confirmed that the angular dependence is not solely described by $\cos \theta$.¹⁰ Comparing the linear response calculations of Camblong et al.¹¹ with those of Vedyayev et al.,¹² one notices that the departure of the $\cos \theta$ dependence is due to the band structure. These results have been confirmed by Hernando et al. in a semiclassical calculation.¹³ Since these treatments are rather involved, we would like to revisit this problem and show that in a simple phenomenological formulation based on a generalized diffusion equation the experiments can not only be well explained but one also finds

simple analytical expressions. In particular, our treatment lends itself to the interesting interpretation that the origin for the departure of the $\cos \theta$ behavior is that of spin filtering.

We consider spin filtering as the reorientation of the magnetic moment of the conduction electrons towards the direction of the effective field of the magnetic background. In MML's the effect of spin filtering was first studied by Slonczewski^{14,16} based on a quantum mechanical treatment. The effect of spin filtering reported therein was of the order of the s-d exchange splitting of the conduction electrons in the molecular field of the localized electrons. The spin reorients itself therefore within a few monolayers completely. In the context of MML's this is in contradiction to the measured angular behavior of the CPP-MR where a departure of the $\cos \theta$ dependence is a small correction; there is no complete suppression of the CPP-MR. We will show that spin filtering is indeed a much smaller effect limited by the amount of spin accumulation. It occurs over a length scale comparable to the spin diffusion length. It is only in the limit of half-metallic magnetic layers that spin filtering is bound to a few monolayers at the interface, a case treated by Bazaliy et al.¹⁷

The interesting proposition made in Refs. 14-16 is that spin filtering leads to spin transfer: the dissipative loss of spin angular momentum from one magnetic subsystem to another that can under certain conditions lead to the excitation of spin waves (microwaves). According to Newton's third law, angular momentum must be conserved in a system. As spin filtering is a transverse effect on the magnetic moment of the conduction electrons, the torque thus created in the background magnetization is also transverse.^{14–16,18} The simplicity of the argument adds to its beauty. Yet we find that spin filtering is in general not related to spin transfer between magnetic subsystems. This is due to the fact that the dominant relaxation mechanism for the nonequilibrium magnetization of the conduction electrons is to the lattice, for example, via spin-orbit coupling.^{19,20} Spin filtering is, therefore, likely to be deterrent to spin transfer as it reduces the nonequilibrium magnetization.

Spin transfer only occurs when the energy is conserved in the process of flipping a spin and exciting a spin wave as has been stressed by Berger.²² For the energy to be conserved, a large nonequilibrium magnetizations is required. References 1 and 22 show that spin transfer is governed by this condition. Once the condition is met, spin transfer is a very efficient process to relax the nonequilibrium magnetization. However, it is only this additional contribution of the nonequilibrium magnetization which conserves energy that relaxes within a few monolayers.

Spin filtering, which does not have an energy barrier to overcome, induces a torque on the background magnetization, nevertheless. Because this torque is related to a current-induced biquadratic coupling, it leads, however, to a precession which is conservative and extends into the bulk of the layer. This biquadratic coupling can be regarded as the next order in the expansion of the nonequilibrium exchange interaction (NEXI). In contrast to the bilinear coupling,^{23–25} the biquadratic coupling exists even in symmetric structures. However, for the systems that show current-induced magnetization reversal^{26–28} the dominant contribution comes from

the bilinear term which is discussed here in the diffusive regime.

The NEXI can be regarded as a "figure of merit" for detecting spin injection. We understand the term spin injection as the change in the nonequilibrium magnetization of a system due to the presence of an external source of nonequilibrium magnetization, for instance, another layer. We consider this change a "nonlocal" change as distinct from the "local" change induced by a layer itself. A way to produce such a nonlocal change in the nonequilibrium magnetization is to inject a spin current into a ferromagnetic layer which acts as the "detector" or "analyzer" of the injected spin current. This has been realized experimentally by letting a spin-polarized electron beam produced in a photocathode pass through a magnetic film²⁹ or by using a multilayer structure of at least two magnetic layers separated by a nonmagnetic one.^{26–28,30–35} Whereas in the former setup the photocathode can be regarded as the "polarizer"-the source of spin-polarized electrons-and the magnetic layer takes the function of an "analyzer," the latter scenario is somewhat more complex in that the two magnetic layers both significantly spin-polarize the current. In the limit of a MML with inversion symmetry at steady state, one can no longer distinguish the two magnetic layers and the concept of a polarizer and analyzer breaks down; the bilinear coupling of the NEXI largely vanishes and there is only the, in general, much weaker biquadratic coupling of the NEXI left. This implies that also the spin injection is largely reduced.

One would assume from the works on spin transfer^{14–17,21,22} that the absence of spin injection would necessarily mean that spin transfer cannot occur and spin waves cannot be excited. That this is, however, not the case has been shown in Ref. 1. This also allows one to reconcile the notion of spin transfer with the experiments of Tsoi *et al.*^{30,31} and Myers *et al.*²⁶ where a single magnetic layer was sufficient to observe current-induced excitations. Here, we will discuss how spin transfer and spin injection lead to different current-induced switching behavior. We find that for different geometries of the system studied one or the other mechanism will be prevalent.

The rest of the paper is then structured as follows. In Sec. II we derive phenomenological transport equations for noncollinear structures. In Sec. III we deal with applications of the diffusion equation and discuss the different physical scenarios outlined above. These are (A) the time-dependent buildup of spin accumulation, (B) spin filtering in noncollinear MML's, (C) magnetic field effects due to noncollinearity which turn out to be negligible, (D) the biquadratic coupling of the NEXI which can be regarded as being caused by a spin current-induced quadrupole field, (E) spin transfer and microwave generation, and, finally, (F) a discussion of current-induced switching being either based on spin injection or spin transfer. Conclusions are drawn in Sec. IV.

II. DERIVATIONS

In deriving the transport and continuity equations, we follow a particularly instructive treatment given by Torrey³⁶ (see also Ref. 37). For simplicity, we assume a fixed background of magnetic moments that is constant in time.

The justification for a phenomenological approach lies in the length scales involved in transport across MML's. The transport is governed by two characteristic length scales, the electron mean free path and the spin diffusion length; in general, the thickness of the individual layers exceeds the size of the electron mean free path in metallic structures but not that of the spin diffusion length.⁸ Whereas charge transport is therefore largely diffusive, spin information is conserved on a length scale comparable to the layer thickness. This led to the use of spin-dependent chemical potentials whose spatial dependence is described in terms of a diffusion equation.^{5,8} Although this approach is very intuitive for collinear MML's much of its clarity is lost when considering arbitrary orientations of the magnetization in different layers. Its correspondence to the approach taken in the following is referred to Appendix A.

We will embark on phenomenological arguments based on drift and diffusion currents to derive expressions for both spin and charge current. These have to be combined with the appropriate continuity equations in order to derive the diffusion equation governing the buildup of spin accumulation. For the spin current the corresponding continuity equations turn out to be coupled Landau-Lifshitz equations between the paramagnetic subsystem of conduction electrons and localized magnetic moments. The derivation of the latter is referred to Appendix B.

A. Transport equations

The axes of the system are chosen to be arbitrarily oriented with respect to the field direction of the layers. The conduction electron spins shall be quantized along the z coordinate axis and n_{σ} is the number of spin-up ($\sigma = \uparrow$) and spin down ($\sigma = \downarrow$) electrons in this direction, respectively. As for electrons and holes in semiconductors, we use here the concepts of drift and diffusion current for spin-up and spin-down electrons. In the presence of both external fields and a carrier density gradient, the carrier current density of the two types can be written as the sum of a term proportional to the drift velocity \vec{v}_{σ} and a term proportional to the density gradient:

$$\vec{J}_{\sigma} = n_{\sigma} \vec{v}_{\sigma} - D \vec{\nabla} n_{\sigma}, \qquad (1)$$

and *D* is the diffusion constant. We assume that the drift velocity is proportional to the force on the conduction electron produced by the action of the field gradients on its electric and magnetic moment, i.e., $K\vec{v}_{\sigma} = \vec{F}_{\sigma}$, where

$$\vec{F}_{\sigma} = -e\vec{\nabla}V \pm \mu_B\vec{\nabla}H_{\rm eff}^z, \qquad (2)$$

and we neglected the cyclotron effect. The positive sign is for spin-up electrons and the negative sign for spin-down electrons. The effective magnetic field $\mathbf{H}_{\text{eff}} = \mathbf{H}_e + \alpha \mathbf{M}_d$ contains the external field and the molecular field coupled to the conduction electrons via an exchange constant α . The molecular field is derived from the energy per unit volume of the *s*-*d*-type exchange interaction between the conduction electrons and the more localized electrons producing the magnetic background,

$$E_{sd} = -\alpha \mathbf{M}_s \cdot \mathbf{M}_d \,. \tag{3}$$

In Eq. (2) only the *z* component is taken into account since the spin is oriented along the *z* axis. When we rewrite Eq. (1) with help of Eq. (2), the *z* component of the magnetization current takes the following form:

$$\vec{J}_M^z = \mu_B(\vec{J}_{\uparrow} - \vec{J}_{\downarrow}) = -\mu_s M_s^z \vec{\nabla} V - D \vec{\nabla} M_s^z + D \chi_s \vec{\nabla} H_{\text{eff}}^z.$$
(4)

We have introduced the conduction electron magnetization $M_s^z = \mu_B(n_{\uparrow} - n_{\downarrow})$, the electron mobility $\mu_s = e/K$ $= eDN(\varepsilon_F)/n$, and the susceptibility $\chi_s = \mu_B^2 n/(DK)$ $= \mu_B^2 N(\varepsilon_F)$, where the expressions after the second equality implies free conduction electrons and $N(\varepsilon_F)$ denotes the density of states at the Fermi level. In principle, for general band structures all transport parameters are spin dependent. However, in a phenomenological approach this dependence can be absorbed in the already existing spin-dependent parameters. The total number of electrons is given by $n=n_{\uparrow}$ $+n_{\downarrow}$ and does not depend on the axis of spin quantization. The *x* and *y* components can be obtained in a similar manner. Realizing that the equilibrium magnetization is given by $\mathbf{M}_s^0 = \chi_s \mathbf{H}_{eff}$, the full magnetization current becomes

$$\vec{\mathbf{J}}_M = -\,\mu_s \mathbf{M}_s \vec{\nabla} V - D \vec{\nabla} \,\delta \mathbf{M}_s \,, \tag{5}$$

where $\delta \mathbf{M}_s = \mathbf{M}_s - \mathbf{M}_s^0$ is the nonequilibrium magnetization, i.e., spin accumulation in the system. The expression $-\mathbf{H}^* = \delta \mathbf{M}_s / \chi_s$ can be regarded as some additional magnetic field, in the presence of which the magnetization would be in equilibrium. Johnson and Silsbee recognized it as a generalized force that drives the magnetization current.⁴ One obtains their result by linearization of Eq. (5), i.e., $\mathbf{M}_s \rightarrow \mathbf{M}_s^0$ and Dindependent of changes in n. In Eq. (5) the flow \mathbf{J}_M is expressed in terms of the forces ∇V and $\nabla \delta \mathbf{M}_s$ as well as their respective kinetic coefficients. An equivalent term has been introduced in Ref. 5 where spin accumulation is understood as the current-induced difference in "chemical potentials" for spin-up and spin-down electrons. The equivalence between the above-described treatment with that of a spinor notation of the chemical potential is given in Appendix A.

By the same thermodynamic principle, one finds an analogous expression for the electric current with different kinetic coefficients. However, employing the line of reasoning that led to Eq. (4) and assuming just for now that the z axis of quantization corresponds to the field direction, we find the following expression for the electric current density:

$$\begin{split} \vec{J}_{e} &= e(\vec{J}_{\uparrow} + \vec{J}_{\downarrow}) \\ &= -\sigma \vec{\nabla} V - eD \vec{\nabla} n + \mu_{s} M_{s}^{z} \vec{\nabla} H_{\text{eff}}^{z} \\ &= -\sigma \vec{\nabla} V - \frac{\mu_{s}}{\chi_{s}} M_{s}^{z} \vec{\nabla} \,\delta M_{s}^{z} - \frac{2eD}{n} \vec{\nabla} (n_{\uparrow} n_{\downarrow}), \end{split}$$
(6)

where $\sigma = e^2 n/K = e^2 DN(\varepsilon_F)$ is the conductivity. The term $\vec{\nabla}(n_{\uparrow}n_{\downarrow})$ is somewhat unexpected from the thermodynamic

reasoning above as it does not appear as a "classical" force on the conduction electrons. From the particular form of this term, one may deduce that this force is due to the electronelectron interactions in the system which are causing a diffusion of the electric charge and can lead to charge accumulation effects. This may be best seen for the case of a nonmagnetic metal where $n_{\uparrow} = n_{\downarrow} = n/2$. The second term in Eq. (6) vanishes, and one simply recovers the spinindependent transport equation in terms of drift and diffusion currents:

$$\vec{J}_{e} = -\sigma \vec{\nabla} V - eD \vec{\nabla} n$$

In the opposite limit of a strong ferromagnet or even halfmetal, where the spin subbands are strongly split $n_{\uparrow} \ge n_{\downarrow}$, the last term in Eq. (6) may be neglected. Assuming for now the latter limit, the conjugate equation to Eq. (5) for arbitrary spin orientation is then

$$\vec{J}_e = -\sigma \vec{\nabla} V - \frac{\mu_s}{\chi_s} \mathbf{M}_s \cdot \vec{\nabla} \,\delta \mathbf{M}_s \,. \tag{7}$$

Since electron and magnetization currents have to vanish in thermal equilibrium, an Einstein relation between diffusion and mobility constants has to hold, i.e., $\mu_s = eD/(k_BT)$. For degenerate conductors one does the following replacement $k_BT = mv_F^2/3$, where *m* is the effective conduction electron mass and v_F the Fermi velocity.

So far we have considered only quasifree conduction electrons. Such a treatment is likely to overestimate the diffusion processes in a ferromagnetic metal. It is, however, possible to define the transport coefficients in a more general form: the conductivity can be written as $\sigma = ne\mu_s = ne^2 \tau_{tr}/m^*$ and the diffusion constant as $D = \kappa \tau_{tr}$, where τ_{tr} is the transport scattering time which has contributions from spinindependent impurity scattering and spin-dependent scattering $\tau_{\rm tr}^{-1} = \tau_i^{-1} + \tau_s^{-1}$; m^* is the renormalized or effective mass of the conduction electrons. The constant $\kappa = \xi v_F^2/3$ is also renormalized. In general, one has to consider quasiparticles close to the Fermi surface, and ξ , which is equal to or smaller than unity, can then be obtained with help of Fermi liquid theory which also allows one to determine the effective mass and a renormalized transport scattering time in order to obtain reasonable agreement with experiments. Here we consider these parameters as phenomenological constants. In particular a reduced value of ξ in the diffusion constant accounts for the delocalized nature of the delectrons in most transition metals.

Such an effectively reduced diffusion constant implies that the magnetization transport is reduced relative to the charge transport. Therefore, later in the text we shall distinguish between the equilibrium polarization $M_s^{0z}/(\mu_B n)$ and a spin-transport asymmetry factor β relating to the nonequilibrium magnetization. Only for simple parabolic conduction bands are they identical. A smaller value for *D* leads also to a more satisfying reasoning why the last term in Eq. (6) is negligible compared to the other two terms. On a more heuristic level this means that in most metallic MML's the spin diffusion length is much larger than the electron mean free path, and one may neglect additional charge accumulation effects. This argument was put forward in the treatment of Valet and Fert to neglect a similar term which arises as a "Boltzmann correction."⁸ It had also been pointed out by Johnson and Silsbee that the use of a renormalized diffusion constant allows for a difference in the spin and charge accumulation length;⁴ i.e., charge and spin are less coupled in realistic MML's than a free-electron description would predict.

B. Continuity equations

Equations (5) and (7) are not yet sufficient to determine the spin accumulation. Out of equilibrium, one requires the additional condition of continuity. For the electric current this is the standard continuity equation

$$e\frac{\partial n}{\partial t} = -\vec{\nabla} \cdot \vec{J}_e \,. \tag{8}$$

For the magnetization current a similar expression is satisfied if we consider a single magnetic layer in the absence of external magnetic fields. In a general MML, however, magnetization is transported across a number of magnetic layers. The conduction electrons, thus, experience a large change in molecular fields from the background magnetization so that we have to describe the continuity of the spin polarization of the conduction electrons instead. The change in magnetic moment is governed by three contributions: the contribution of torque exerted by the magnetic background on the vector moment, the contribution of relaxation processes, and, finally, the rate of change of magnetization through the surface of a unit volume element \mathcal{V}_{0} which is described by the magnetization current. The first two conditions lead to standard Landau-Lifshitz equations which have to be supplemented by the action of the magnetization current. The change of the magnetic moment per volume element due to the magnetization current is

$$-\int \frac{d\mathcal{V}}{\mathcal{V}_0} \left(\frac{\partial \mathbf{M}_s}{\partial t}\right)_{[J_M]} = \int \frac{d\mathcal{S}}{\mathcal{V}_0} \mathbf{n} \cdot \vec{\mathbf{J}}_M = \vec{\nabla} \cdot \vec{\mathbf{J}}_M, \qquad (9)$$

where **n** is the unit vector on the surface S of the volume element. The Landau-Lifshitz equation for a static magnetic background derived in Appendix B reads as

$$\frac{\partial \mathbf{M}_s}{\partial t} - \gamma \mathbf{M}_s \times \mathbf{H}_e + \vec{\nabla} \cdot \vec{\mathbf{J}}_M = -\frac{\delta \mathbf{M}_d}{\tau_{dl}} - \frac{\delta \mathbf{M}_s}{\tau_{sl}}.$$
 (10)

On the left-hand side (LHS) of the equation the additional term $-\alpha(\gamma_s - \gamma_d)(\mathbf{M}_s \times \mathbf{M}_d)$ ought to be included if the system has different gyromagnetic ratios $\gamma_s \neq \gamma_d$ for the *s*- and *d*-electron systems. In Sec. III C, we will show that any of these "precession" terms are negligible for most experimental situations concerning metallic MML's. The terms on the right-hand side are due to relaxation with characteristic times τ_{sl} and τ_{dl} . Both are mainly due to conduction electron spin relaxation to the lattice, for example, via spin-orbit coupling.¹⁹ In general, *s*-*d* scattering conserves angular momentum, so that it does not contribute to relaxation if the

magnetic background \mathbf{M}_d is fixed.³⁸ In such a situation τ_{sl} is the only contribution to the conduction-electron spin relaxation τ_s , also known as T_2 . However, the *s*-*d* scattering can couple conduction electrons to thermal magnons of the background magnetization which is the origin of the first term on the right-hand side (see Appendix B and Refs. 52 and 54 for details). This is a weak effect at low temperatures. The magnitude of $\delta \mathbf{M}_d$ depends approximately on temperature as given by the Bloch law $\delta M_d = M_d(0) - M_d(T) \propto T^{3/2}$. In the following we always consider T=0.

We note that Eq. (10) does not describe the precession of the magnetization due to an oscillating external field, since we assumed that the magnetic background is held fixed. Also in the regime, where spin-wave excitations occur due to high currents,³⁰ Eq. (10) can only describe the onset of spin-wave excitation after which it is no longer valid and one has to retreat to the more general equations given in Ref. 1. In both cases one has to take the dynamics of the magnetic background into account. Nevertheless, Eq. (10) serves our purpose to study the buildup of spin accumulation across a MML before these excitations can occur.

III. APPLICATIONS

A. Time-dependent buildup of spin accumulation

One of the effects not yet studied when considering transport across magnetic multilayers is the time-dependent buildup of nonequilibrium magnetization or spin accumulation. One would like to know how the nonequilibrium magnetization distributes itself in the multilayer before the steady state is reached. The steady state requires *both charge and spin distributions* to be constant over time. Here we study the transient processes of spin accumulation.

We noted before that the length scales for charge and spin transport are very different in MML and, hence, their time scales. Therefore, one can assume that a steady state for charge transport is established instantaneously, i.e., within $\tau_{\rm tr} \approx 10^{-15} - 10^{-14}$ s, whereas the spin accumulation reaches steady state only after $\tau_s \approx \tau_{sl} \approx 10^{-12} - 10^{-11}$ s. In the following we consider times longer than the transport scattering time and shorter than the spin relaxation time.

We would like to motivate the physical situation by studying two simple examples. One could imagine the following situation. A ferromagnet is in contact with a "battery" that allows no spin-flip scattering. Then the current entering the ferromagnet becomes only polarized by the spin-flip scattering in the ferromagnet itself. The buildup of spin accumulation is entirely determined by the relaxation time τ_{sl} of the ferromagnet. Although this scenario seems, at first, likely to be realized in many experiments, it leads to the interesting, somewhat paradoxical situation that the current across a "perfect" ferromagnet, i.e., one without spin-flip processes, stays always unpolarized. This can be seen from the following line of reasoning.

The corresponding boundary condition that requires the battery not to produce a nonequilibrium magnetization is $\delta \mathbf{M}_s(-\infty) = \delta \mathbf{M}_s(\infty) = 0$. At the interface between ferromagnet *F* and nonmagnetic battery *N*, one has therefore from Eq. (5)

$$J_M^F(\pm\infty) = -\mu_s^F M_s^z \vec{\nabla} V, \qquad (11a)$$

$$J_M^N(\pm\infty) = 0, \tag{11b}$$

assuming a current flowing along a fixed axis. In Eq. (11b) the drift part is absent as the battery in nonmagnetic. Since J_M has to be continuous everywhere, it follows that there is no polarization of the current at the interface $J_M^F(\pm\infty) = J_M^N(\pm\infty) = 0$. Therefore, the current across a "perfect" ferromagnet, where Eq. (11a) determines J_M^F everywhere in the ferromagnet, is always unpolarized. The resistance of the ferromagnet is held identical for both spin channels by the boundary conditions.

In general, however, it is more likely that there exist spinflip processes outside the structure under consideration. In other words, it seems reasonable to assume that it is possible to draw different currents for spin-up and -down electrons from the battery. In such a case the battery produces a nonequilibrium magnetization, and one has only $\delta \mathbf{M}_s(\infty)$ $-\delta \mathbf{M}_s(-\infty)=0$. To be more explicit, we consider the buildup of spin accumulation at the interface, x=0, between a ferromagnet and a nonmagnetic metal layer, when a current is flowing in the *x* direction across it. With Eq. (5) the magnetization current in each layer can be approximated by

$$J_M^F \approx -\mu_s^F M_s^z \vec{\nabla} V \approx \frac{\mu_B}{e} \sigma_F^* \beta E, \qquad (12a)$$

$$J_{M}^{N} = -D \frac{\partial}{\partial x} \delta M_{s}^{z} \equiv -D \frac{\partial m(x,t)}{\partial x}.$$
 (12b)

In Eq. (12a) the first approximation is given by the fact that due to metallic conduction the current in the ferromagnet is dominated by the drift part whereas the second approximation is a linearization in the dependence of the background magnetization where β is the spin "asymmetry" factor in the ferromagnet.⁸ The asymmetry factor is a transport parameter which, in general, should be considered as a renormalized or phenomenological quantity, respectively. For simple parabolic conduction bands it can be directly associated with the conduction electron magnetization $\beta = M_s^{0z}/(\mu_B n)$. The average conductivity in the ferromagnet is given by $\sigma_F^* = (1$ $(-\beta^2)/\rho_F^*$ and the electric field defined as $E = -\vec{\nabla}V$. As before, in Eq. (12b) the drift part is absent as there is no background magnetization in the nonmagnetic metal. The function m(x,t) determines the spin accumulation in the nonmagnetic layer.

In order to study its time development, we further assume that due to the dominance of the drift contribution to the electric current for both magnetic and nonmagnetic layers its divergence is zero, i.e., that it has reached steady state. This happens within the transport scattering time τ_{tr} which is of the order of $10^{-15}-10^{-14}$ s. Before the spin accumulation has reached a steady state $t_0 \approx \tau_{tr} < t < \tau_{sl} \approx 10^{-12}-10^{-10}$ s, the form of Eqs. (12) allows one to treat the ferromagnetic layer as a constant source of nonequilibrium magnetization with $m_0 = m(0,t)$ at all times for the above interval. For the transient problem in the nonmagnetic layer x > 0 one has the



FIG. 1. Spin accumulation profile in the nonmagnetic layer developing in the time steps from left to right Dt=0.01, 0.1, 1, 10, 100, 1000 in units of an arbitrary area.

initial condition that $m(x>0,t_0)=0$ and Eq. (10) reduces at zero temperature and in the absence of relaxation to

$$\frac{\partial \widetilde{m}(x,t)}{\partial t} = D \frac{\partial^2 \widetilde{m}(x,t)}{\partial x^2},$$
(13)

where $\tilde{m}(x,t) = m_0 - m(x,t)$. One, however, has to bear in mind that the spin accumulation at the interface depends on the total resistance of the structure. Therefore, there is only a fraction of the rate given by the drift current (12a) which one can actually drive across the interface. This fraction is given by the spin accumulation calculated in the resistor-in-series model, where due to the time scale in consideration spin diffusion can be neglected. The semi-infinite layers have resistivities for spin-up and spin-down channels $\rho_F^{\sigma} = 2\rho_F^*(1 \pm \beta)$ and $\rho_N^{\sigma} = 2\rho_N^*$. For a constant external bias V that is the same for each spin channel and the boundary condition $m(-\infty,t_0) - m(\infty,t_0) = 0$, the initial spin accumulation at the interface is given by

$$m_0 = eV\mu_B N(\varepsilon_F) \frac{2\beta \rho_F^* \rho_N^*}{(\rho_F^* + \rho_N^*)^2 - (\beta \rho_F^*)^2}.$$
 (14)

As pointed out before, at $t=t_0$ there is no spin accumulation in the nonmagnetic layer, so that $m(x,t_0)=0$ or $\tilde{m}(x,t_0)$ $=m_0$. The solution to Eq. (13) can be found via Laplace transformation into an image function given by simple exponentials similar to the steady-state problem. The backtransformation leads to an error function,

$$\widetilde{m}(x,t) = m_0 \operatorname{Erf}\left(\frac{x}{2\sqrt{D_N t}}\right),\tag{15}$$

or the error function complement for m(x,t), respectively, shown normalized to the initial spin accumulation m_0 in Fig. 1. At time $t=\infty$, $m(x,t)=m_0$. This result is only valid assuming that the spin diffusion length is infinite. In reality the accumulation stops growing after $t \approx \tau_{sl}$ because of spin relaxation on this time scale.

The magnetization current in the nonmagnetic layer can be derived together with Eq. (15) from Eq. (12b):

$$J_M^N(x,t) = -D_N \frac{\partial m(x,t)}{\partial x} = m_0 \sqrt{\frac{D_N}{\pi t}} e^{-x^2/4D_N t}.$$
 (16)

The buildup of spin accumulation is connected to the flow of nonequilibrium magnetization across the interface, i.e., at x = 0 from the ferromagnet into the nonmagnetic layer, which is determined by Eq. (16),

$$J_{M}^{N}(x,t)\big|_{x=0^{+}} = m_{0}\sqrt{\frac{D_{N}}{\pi t}}.$$
(17)

This flow tends to decrease over time. The amount of magnetization having entered the nonmagnetic layer after time t,

$$\mathcal{M}(t) = \nu \int_{0}^{t} dt' J_{M}^{N}(x,t') \big|_{x=0^{+}} = 2 \nu m_{0} \sqrt{\frac{D_{N}t}{\pi}}, \quad (18)$$

where ν is the area of the cross section of the interface, allows us to determine the spin accumulation m_0 . This could be done by measuring the magnetic moment \mathcal{M} as a function of time by femtosecond magnetometry (see, for example, Ref. 29 for related experiments). Such a measurement gives a direct way to access the amount of spin accumulation at an interface and would be of great value.

Although the flow across the interface vanishes in Eq. (17) if $t \rightarrow \infty$, it is limited to $t = \tau_{sl}$; therefore one reaches a steady-state condition for the magnetization current. In such a case, we replace the LHS of the diffusion equation (13) by $\tilde{m}(x,t)/\tau_{sl}$ and impose again the boundary condition that both J_M and $\tilde{m}(x,t)$ be conserved at the interface. In addition, the diffusion term in the ferromagnet has to be added to Eq. (12a). We also neglect terms in the nonequilibrium magnetization that are of order β^3 and assume that the average density of states is the same in both materials. Then we can substitute in Eq. (17) πt by τ_{sl} and m_0 by⁸

$$m_0 = e J_e \mu_B N(\varepsilon_F) \frac{\beta \lambda_F \lambda_N \rho_F^* \rho_N^*}{\lambda_F \rho_F^* + \lambda_N \rho_N^*},$$
(19)

where $\lambda_{F(N)} = \sqrt{D_{F(N)} \tau_{sl}^{F(N)}}$ is the spin diffusion length in the respective material. Due to spin diffusion, this result differs from that in Eq. (14).

B. Spin accumulation in noncollinear structures: Spin filtering

As was pointed out in the Introduction, we consider spin filtering as the reorientation of the magnetic moment of the conduction electrons towards the magnetic background. The idea is that there is a higher scattering rate for unaligned moments than for already aligned ones. This effect, which is caused by the anisotropy in spin space, can be described by two different spin diffusion lengths for the conduction electron spin: one for aligned spins (termed parallel) and a second shorter one for spins that are not aligned with the background magnetization (termed transverse or antiparallel). If the nonequilibrium magnetization is, thus, antiparallel to the magnetic background, its decay away from the interface will be fastest. The effect of spin filtering is studied on two identical semi-infinite ferromagnetic layers, whose magnetization vectors \mathbf{M}_L and \mathbf{M}_R are at an arbitrary angle θ in the *zy* interface plane at x=0. We rewrite the electric field in Eq. (5) with help of Eq. (7) in terms of the electric current:

$$\vec{\mathbf{J}}_{M} = -\frac{\mu_{B}}{e} \mathbf{p} \vec{J}_{e} - D[\vec{\nabla} \,\delta \mathbf{M}_{s} - \mathbf{p}(\mathbf{p} \cdot \vec{\nabla} \,\delta \mathbf{M}_{s})], \qquad (20)$$

where we defined a vector of polarization $\mathbf{p} = \mathbf{M}_s^0/(\mu_B n)$ which for parabolic free conduction bands is related to the asymmetry factor in the ferromagnet as $|\mathbf{p}| = \beta$. In the two ferromagnets we have then $\mathbf{p}_L = (0,0,\beta)$, and \mathbf{p}_R $= (0,\beta \sin \theta,\beta \cos \theta)$. The electric current shall be directed along the *x* direction and we neglect spin-dependent scattering at the interfaces so that the magnetization current is conserved at the interface: $\mathbf{J}_M(0^+) - \mathbf{J}_M(0^-) = 0$. To determine the nonequilibrium magnetization, we need to apply also Eq. (10). In the present case we assume that one can neglect effects from the precession term caused by the external field \mathbf{H}_e (see Sec. III C). At steady state Eq. (10) with Eq. (20) reads as

$$D\left[\frac{\partial^2 \delta \mathbf{M}_s}{\partial x^2} - \mathbf{p}\left(\mathbf{p} \cdot \frac{\partial^2 \delta \mathbf{M}_s}{\partial x^2}\right)\right] = \frac{\delta \mathbf{M}_s}{\tau_{sl}}.$$
 (21)

Without the second term in the square parentheses, Eq. (21) would be the standard spin diffusion equation written in vector form⁵ and the nonequilibrium magnetization proportional to β . Including the second term in the square parentheses, which has a prefactor proportional to β^2 , the nonequilibrium magnetization contains then in addition corrections to the order of β^3 .

The term in the square parentheses can be written in matrix form and mixes the different coordinates if the matrix is not diagonal, leading to the following ansatz:

$$\delta \mathbf{M}_{s}(x) = \mathbf{m}_{1\pm} e^{\pm x/\lambda_{\parallel}} + \mathbf{m}_{2\pm} e^{\pm x/\lambda_{\perp}}, \qquad (22)$$

where $\mathbf{m}_{1(2)\pm}$ are vectors in spin space. We introduced a spin diffusion length for aligned spins $\lambda_{\parallel} = \sqrt{D_F \tau_{sl}}$ (termed parallel) and a shorter one for spins which are not aligned $\lambda_{\perp} = \lambda_{\parallel}\sqrt{1-\beta^2}$ (termed transverse or antiparallel). We see that the "transverse effect" gets larger the larger the polarization β of the conduction electron is. The necessity of an additional spin diffusion length has been recognized before in transport across domain walls.³⁹ However, this is the first time that the presence of an additional spin diffusion length leads to an anisotropic behavior in the spin diffusion with respect to spin space. Although our treatment is closely related to that Hernando and co-workers¹³ who express the effects of the noncollinearity via a mixing conductance for spin-up and spin-down electrons, such an anisotropy had not been recognized.

In order to have bounded solutions at $\pm \infty$, we choose the coefficients in Eq. (22) as follows: $\mathbf{m}_{1-} \neq 0$, $\mathbf{m}_{2-} \neq 0$, and $\mathbf{m}_{1+} = \mathbf{m}_{2+} = \mathbf{0}$ in the right layer, whereas $\mathbf{m}_{1+} \neq 0$, $\mathbf{m}_{2+} \neq 0$, and $\mathbf{m}_{1-} = \mathbf{m}_{2-} = \mathbf{0}$ in the left layer; i.e., the left layer is equivalent to coefficients with a "+" index and the right layer with a "-" one. When we use the continuity condition

for the nonequilibrium magnetization at the boundary, $\delta \mathbf{M}_s(0^+) - \delta \mathbf{M}_s(0^-) = 0$, the solution is still underdetermined as we have 12 instead of 6 coefficients. To determine the remaining coefficients, we make use of the fact that the ansatz (22) has to be a solution of Eq. (21) at x=0.

For our example we find that there is no contribution to the nonequilibrium magnetization along the *x* coordinate so that $m_{1\pm}^x = m_{2\pm}^x = 0$. An intermixing between the coordinates occurs only between the *z* and *y* directions in the left and right ferromagnets. Since for these coefficients the general expression is rather complex, we would like to focus on the case of perpendicular alignment, $\theta = \pi/2$, where in the left layer

$$m_{1\pm}^{z} = -m_{2\pm}^{y} = \zeta \frac{\lambda_{\parallel} - \lambda_{\perp}}{\beta} = \zeta \lambda_{\parallel} \frac{1 - \sqrt{1 - \beta^{2}}}{\beta} \approx \frac{\zeta \lambda_{\parallel}}{2} \beta,$$

and all other terms being zero. The factor $\zeta = e \mu_B N(\varepsilon_F) J_e / \sigma_F^*$ depends on the current. The last expression only holds approximately for $\beta^2 \ll 1$. For $\beta^2 \approx 1$ one has $\zeta \lambda_{\parallel}$ instead. With these coefficients the expression (22) for the spatial distribution of the nonequilibrium magnetization is particularly simple as it keeps only one of the exponentials for each coordinate:

$$\delta \mathbf{M}_{s}(x \leq 0) = \zeta \frac{\lambda_{\parallel} - \lambda_{\perp}}{\beta} \left[\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} e^{+x/\lambda_{\parallel}} - \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} e^{+x/\lambda_{\perp}} \right]$$
$$\delta \mathbf{M}_{s}(x \geq 0) = \zeta \frac{\lambda_{\parallel} - \lambda_{\perp}}{\beta} \left[\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} e^{-x/\lambda_{\perp}} - \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} e^{-x/\lambda_{\parallel}} \right].$$

From the above expression follows that at the interface the nonequilibrium magnetization is for both layers at 45° to their respective background magnetization. Since $\lambda_{\parallel} > \lambda_{\perp}$, the nonequilibrium magnetization in the bulk of the layers sufficiently far away from the interface will be directed along the z direction in the left layer and along the -y direction in the right layer. For a better understanding this result is to be compared with an antiparallel configuration $\theta = \pi$, where $m_{1\pm}^{y} = m_{2\pm}^{y} = m_{1\pm}^{z} = 0$ and $m_{2\pm}^{z} = \zeta \beta \lambda_{\perp} / (1 - \beta^{2})$ as given previously in Ref. 8. Whereas in the antiparallel configuration the decay of the nonequilibrium magnetization away from the interface scales with λ_{\perp} and is confined to the z coordinate, in the perpendicular configuration the decay has both contributions in the y and z coordinates, but scales differently. If it were to scale in the same way, the nonequilibrium magnetization would be always at 45° to the magnetization of the layers; i.e., its direction takes always the mean of \mathbf{p}_L and \mathbf{p}_R , as it is at the interface.

The occurrence of the shorter spin diffusion length λ_{\perp} —transverse spin diffusion length—leads to the effect of *spin filtering*. In the present form, one can think of this effect as an anisotropy in the relaxation of nonequilibrium magnetization. The preferential direction of the nonequilibrium magnetization is parallel to the background magnetization along \mathbf{p}_L or \mathbf{p}_R , respectively. In particular for half-metals,¹⁷



FIG. 2. Shown is the effect of spin filtering. In order to make the figure easier to view, the background magnetization of the right layer is now in the *z* direction and that of the left layer at an angle θ (left and right are exchanged with respect to the calculated example). The spin accumulation profile changes along the *x* direction of a ferromagnetic layer. At the interface (closest to the viewer) the nonequilibrium magnetization is at 45° to the background magnetization of the layer. Since here it is assumed that $\lambda_{\perp} \ll \lambda_{\parallel}$, the nonequilibrium magnetization relaxes to the background magnetization, along the *z* axis, before it finally decays away from the interface.

where $\beta \approx 1$, the effect becomes very large, since $\lambda_{\perp} \rightarrow 0$. The result of spin filtering is a "twisting" and *relaxation* of the nonequilibrium magnetization as shown in Fig. 2. From a microscopic point of view, electrons precess around the local *s*-*d* field^{15,21} given the populations and energies are different for spin-up and spin-down electrons. This process, when averaged over elastic and inelastic scattering events, leads to different speeds for electrons diffusing along the parallel and perpendicular directions to the local exchange field. Since we describe a different to that of Slonczewski who was first to point out a similar effect in a ballistic treatment of the conduction through a tunnel junction.¹⁴

In Slonczewski's treatment, however, the effect was proportional to the exchange splitting of the conduction electron bands, i.e., αM_d , and not just to the nonequilibrium magnetization δM_s as found here. Thus the associated "precession" is several orders of magnitudes faster than for the case treated here.14,21 Also in that example spin filtering was a large effect and localized at the interface. This does not seem to be applicable to the MML's for which one has measured the angular dependence of the CPP-MR; for these one finds only small departures from the $\cos \theta$ dependence, while a strong spin filtering would completely suppress the CPP-MR.⁹ It seems when simply matching wave functions certain aspects of spin transport are omitted. An instructive example is to compare Eqs. (8a) and (8b) of Ref. 24 with Eq. (2) of Ref. 40 which lacks the effects arising from the nonequilibrium magnetization. In Sec. III E we will show that Slonczewski's idea of spin filtering is only applicable beyond a spin accumulation threshold. In our phenomenological approach spin filtering occurs in the bulk of the layer and is only for half metals strictly localized at the interface.

In the following, we would like to show how the effect of spin filtering influences the angular dependence of the CPP-MR, a quantity of experimental interest. The CPP-MR is then the resistance change of the structure with respect to the mutual orientation of the magnetization of the layers, i.e., to the angle θ , which is associated to the voltage drop across the interface,

$$R_{\rm SI} = -\frac{1}{\nu J_e} \int_{-\infty}^{\infty} dx \frac{\partial V}{\partial x}$$
$$= \frac{\mu_B}{e \chi_s \nu J_e} [\mathbf{p}_L \cdot \delta \mathbf{M}_s(x) \big|_{-\infty}^0 + \mathbf{p}_R \cdot \delta \mathbf{M}_s(x) \big|_{0}^{\infty}]$$
$$= \beta^2 \frac{\lambda_{\perp} \rho_F^*}{\nu} (1 - \cos \theta) F(\theta), \qquad (23)$$

where ν is the area of the interface between the two ferromagnetic layers. We introduced a function describing the departure from the $\cos \theta$ behavior of the resistance:

$$F(\theta) = \frac{8\frac{\lambda_{\perp}}{\lambda_{\parallel}} \left[1 + \frac{\lambda_{\perp}}{\lambda_{\parallel}} - \beta^2 \cos^2\left(\frac{\theta}{2}\right)\right]}{8 - \beta^2 [7 + \cos(2\theta)] + \frac{\lambda_{\perp}}{\lambda_{\parallel}} \{8 - \beta^2 [7 - \cos(2\theta)]\}}$$
$$\approx \left[1 - \frac{\beta^2 \cos^2\left(\frac{\theta}{2}\right)}{1 + \sqrt{1 - \beta^2}}\right]. \tag{24}$$

For an antiparallel alignment of the magnetization of the ferromagnets the last term in expression (24) vanishes and we obtain the result that $\nu R_{\rm SI} = 2\beta^2 \lambda_{\perp} \rho_F^{*.8}$ The departure of the strict $\cos \theta$ dependence is largest for close to parallel alignment and is proportional to β^4 as expected from Eq. (21). For Co layers, where $\beta \approx 0.4$, the correction is then up to 8%—a small but noticeable effect.⁹ The form of the fitting function in Ref. 9 is somewhat corresponding to Eq. (23). However, therein it was argued that this effect can only occur for different magnetic layer materials. Here we show that the departure from the $\cos \theta$ behavior is a feature of the polarization effects arising from noncollinearity: spin filtering.

C. Spin accumulation in noncollinear structures: Magnetic field effects

One would expect that another interesting effect present in noncollinear structures is that of the magnetic field \mathbf{H}_{e} on the nonequilibrium magnetization. However, this effect is in fact rather small and becomes only relevant at steady state if its related term in Eq. (10) is comparable to or larger than the inelastic scattering rate, i.e., $\omega \approx 1/\tau_{sl}$. In the following we shall explain this in more detail. A similar treatment of *spin precession* due to the influence of an external magnetic field on the nonequilibrium magnetization in the nonmagnetic spacer between two ferromagnetic electrodes was given in Ref. 13.

For simplicity, we study again two semi-infinite ferromagnets with a steady-state current as before but now ignore the corrections to the nonequilibrium magnetization in β^3 calculated in the previous section. We consider, in addition, that the right layer shall be pinned and that the influence of the magnetic field compared to inelastic scattering is small

 $1/\tau_{sl}^R \gg \omega_R$. However, in the left ferromagnet these quantities are comparable to each other $1/\tau_{sl}^L \approx \omega_L$. We choose its magnetization to be directed along the *z* direction for convenience and let that of the right layer oriented at an arbitrary angle θ in the *zy* plane. Instead of Eq. (21), we need to solve for the left ferromagnet

$$D\frac{\partial^2 \delta \mathbf{M}_s}{\partial x^2} = \frac{\delta \mathbf{M}_s}{\tau_{sl}^L} + \gamma \delta \mathbf{M}_s \times \mathbf{H}_e$$
(25)

and the corresponding equation for the right one that does not contain the last term. In order not to complicate our model, the diffusion constants in both layers are roughly identical as well as the resistivities. These equations have to be supplemented with the usual boundary conditions of continuity at the interface x=0.

Rather than using a linear combination of exponential as an ansatz, we transform Eq. (25) into circular coordinates so that we have

$$\delta M_s^+(x \le 0) = m_L^+ e^{-i\kappa x + (x/\lambda)},$$

$$\delta M_s^-(x \le 0) = m_L^- e^{+i\kappa x + (x/\lambda)},$$

$$\delta M_s^z(x \le 0) = m_L^z e^{x/\lambda_L},$$

 $\lambda_L = \sqrt{D \tau_{sl}^L}, \qquad 1/\lambda = \cos(\psi_0/2)/\lambda_L,$ where and к $=\sin(\psi_0/2)/\lambda_L$. The latter two quantities describe a decaying and oscillatory behavior of the nonequilibrium magnetization, respectively. Their contribution is determined by the ratio between ω_L and relaxation time, $\tan \psi_0 = \tau_{sl}^L \omega_L$ (assuming that the field is oriented along the z direction so that $\omega_I = |\gamma H_e^z|$). The corresponding treatment for the right ferromagnet is largely simplified due to the relative smallness of the field term: $\delta M_s^{\nu}(x \ge 0) = m_R^{\nu} e^{-x/\lambda_R}$, where λ_R $=\sqrt{D\tau_{sl}^R}$ and $\nu=\{+,-,z\}$. After determining the coefficients $m_{L(R)}^{\nu}$ with help of the continuity conditions at the interface, the nonequilibrium magnetization is a real quantity in Cartesian coordinates:

$$\delta M_s^{x/y}(x \le 0) = \pm \zeta \beta \sin \theta [\lambda_{s/c} e^{x/\lambda} \cos(\kappa x) \mp \lambda_{c/s} \sin(\kappa x)],$$
$$\delta M_s^z(x \le 0) = \zeta \beta \Lambda (1 - \cos \theta) e^{x/\lambda_L},$$

where ζ has been defined in the previous section, Λ $=\lambda_L\lambda_R/(\lambda_L+\lambda_R)$ is a ratio of the spin diffusion lengths left from the and right layers, $\lambda_c = [\lambda_L]$ $+\lambda_R \cos(\psi_0/2)]/[(\lambda_R/\lambda_L)+2\cos(\psi_0/2)+1]],$ and λ, $=\lambda_R \sin(\psi_0/2)/[(\lambda_R/\lambda_L)+2\cos(\psi_0/2)+1]]$. The expression for $\delta M_s^z(x \le 0)$ shows that at large distances from the interface the total nonequilibrium magnetization tends to zero. However, $\delta M_s^{x/y}(x \le 0)$ retains an oscillatory dependence. For this dependence to have a noticeable effect, the period of the oscillations should not be significantly longer than the system size. As a demonstration that this, however, will be generally the case we take $\omega_L \tau_{sl}^L = 1/4$. For an external magnetic field of 1.4 T the relaxation time has then to be τ_{sl}^{L} =1.0 ns which corresponds roughly to a spin diffusion length λ_L of several 100 nm. For these values $\cos(\psi_0/2)$

= 1.0 and $\sin(\psi_0/2) = 0.1$ so that $\lambda = \lambda_L$ and $\kappa = 0.1/\lambda_L$. The latter leads to a period for the oscillations of 6.3 μ m. In order to obtain shorter periods which lead to an observable spatial variation, one needs to apply significantly higher fields or needs extremely long spin lifetimes; both assumptions seem to be unrealistic for metallic MML even for very pure nonmagnetic spacer layers.¹³ It becomes only feasible in semiconductors which show extraordinary long spin lifetimes.⁴¹⁻⁴³ The terms δM_s^x , therefore, can be neglected since, in addition, $\lambda_s \approx 0$. Finally, one can write $\delta M_s^y(x)$ $\leq 0 \approx -\zeta \beta \Lambda \sin \theta e^{x/\lambda_L}$ which is what one would also obtain in the absence of the field term in Eq. (25). The same line of reasoning holds if we would include the term $\alpha(\gamma_s)$ $(\delta \mathbf{M}_s \times \mathbf{M}_d)$ in the LHS of Eq. (25) which is present for different gyromagnetic ratios $\gamma_s \neq \gamma_d$ of the *s*- and *d*-electron systems.

An important aspect of the above calculation is that also for noncollinear orientations of the magnetic moments in different layers the distribution of the nonequilibrium magnetization $\delta \mathbf{M}_s$ is governed by a standard diffusion equation; the effect of local magnetic fields on the nonequilibrium magnetization is rather small. Although calculated in a phenomenological picture, our results also point to the fact that the field due to the *s*-*d* type of coupling of the conduction electrons to the local magnetic moments is not altering the nonequilibrium magnetization $\delta \mathbf{M}_s$ as long as the local background magnetization is stationary. The way the exchange field acts on the nonequilibrium magnetization is only indirect as has been described in the previous section.

D. Spin injection and NEXI: Bilinear, biquadratic, and bicubic coupling

The occurrence of spin accumulation has the important consequence that it can act as an additional magnetic field on the magnetic background. This effect, the bilinear coupling of the nonequilibrium exchange interaction,²³⁻²⁵ is compensated in structures with an inversion symmetry and linear transport. However, if either the transport is nonlinear, as for example across point contacts and tunnel junctions, or the structure is asymmetric, the magnetic layers become bilinearly coupled and certain magnetization configurations become preferable. For sufficiently large currents the coupling forces the magnetization of the layers to reorient.²⁵ The NEXI will be largest if the nonequilibrium magnetization in a ferromagnetic layer is completely determined by an external source, the polarizer. Hence, the NEXI can be regarded as a "figure of merit" for detecting spin injection by a magnetic layer, the analyzer.

A single magnetic layer always creates an excess of nonequilibrium magnetization at one interface and a deficiency on the opposite interface.^{5,44} This distribution of nonequilibrium magnetization depends on the direction of the current so that it is reversed by reversing the current. For a single magnetic layer the nonequilibrium magnetization exists only "locally" (in the sense as determined in the Introduction); as long as the electric current is uniform throughout the system it is fully compensated when integrated over the leads. Two magnetic layers in close proximity will establish a nonequilibrium magnetization profile that depends on the mutual orientation of their local magnetizations. Similar layers will each produce similar profiles, so that the change between different magnetization directions will be large, yet the nonequilibrium magnetization within each layer is almost unchanged. In the limit of a MML with inversion symmetry at steady state, one can no longer distinguish the two magnetic layers and the concept of "polarizer" and "analyzer" breaks down.

In essence, the concept of two magnetic layers acting as a polarizer and analyzer is best fulfilled if one replaces the analyzer by a single spin impurity, so that there is no back effect on the polarizer. Due to the exchange interaction between the conduction electrons and the spin impurity, the local moment orients itself with respect to the direction of the moments of the conduction electrons emitted from the polarizer. The current mediates thus a NEXI between the polarizer and the analyzer that determines the minimum energy configuration.^{23–25} In a multilayer the sign of the NEXI is determined by the direction of the current due to the respective change in the nonequilibrium magnetization profile. In this sense the NEXI can be regarded as the figure of merit for detecting the spin injection with the analyzer.

So far, when calculating the NEXI, only the nonequilibrium magnetization to first order in the polarization β which produced bilinear coupling has been treated.^{23–25} We have seen in Sec. III B that there are, however, higher-order contributions to the nonequilibrium magnetization. These terms proportional to β^3 led to a deviation of the cos θ behavior in the angular dependence of the magnetoresistance. The same model that predicts the resistance also yields the coupling,²⁴ and here we would like to find the associated change in the coupling.

An instructive case is that of two semi-infinite ferromagnets, which shows no bilinear coupling of the NEXI proportional to β^2 . This is also one of the few cases that gives simple algebraic expressions, because usually the expressions for such a calculation become rather involved. The averaged nonequilibrium contribution to the *s*-*d* exchange interaction can be written according to Eq. (3) as

$$E'_{sd} = -\alpha \frac{\nu}{\mathcal{V}_0} \int_{-\infty}^{\infty} dx \,\delta \mathbf{M}_s(x) \cdot \mathbf{M}_d(x).$$
(26)

It is interesting to note that the integrand is similar to the expression for $R_{\rm SI}$ that led to Eq. (23).²⁴ We now insert the solutions to the nonequilibrium magnetization (22) into Eq. (26) and find

$$E'_{\text{NEXI}} = -\alpha_2 [M^4 - (\mathbf{M}_L \cdot \mathbf{M}_R)^2] \left[1 - \frac{\Lambda(\mathbf{M}_L \cdot \mathbf{M}_R)}{M^2} \right]$$
$$= -\alpha_2 M^4 \sin^2 \theta (1 - \Lambda \cos \theta), \qquad (27)$$

where we made the same approximation in the denominator that led to Eq. (24). From the first expression, one can identify bilinear, biquadratic, and bicubic coupling terms. All coupling terms depend linearly on current which produces the feature that they change sign on reversing the current. Therefore, the notion of bilinear, biquadratic, and bicubic coupling makes only sense relative to each other for a given direction of the current. In Eq. (27) we introduced a biquadratic coupling constant α_2 and a scaling factor Λ which is unity for $\beta \approx 1$ and approaches $\beta^2/4$ for $\beta \ll 1$:

$$\alpha_2 = \frac{\alpha c}{(2\mu_B n)^2} \frac{\nu \lambda_\perp}{\mathcal{V}_0},$$
$$\Lambda = \frac{\lambda_{\parallel} - \lambda_\perp}{\lambda_{\parallel} + \lambda_\perp} = \frac{1 - \sqrt{1 - \beta^2}}{1 + \sqrt{1 - \beta^2}}.$$

Therefore, the bilinear and bicubic terms to the coupling can be ignored for most ferromagnets apart from half-metals since they are of the order of β^5 rather than β^4 . The current dependence is given by the dimensionless parameter c $= e J_e \rho_F^* \lambda_F \mu_B N(\varepsilon_F) \beta / M$, where $M = |\mathbf{M}_{L(R)}|$. Here, we focus on the biquadratic term which becomes minimal or maximal, depending on the direction of the current, for perpendicular alignment $\theta = \pi/2$. This is in correspondence to the standard bilinear NEXI term which range is determined by the spin diffusion length λ_{\perp} . However, there is a subtle difference in that the energy discriminates now between a perpendicular configuration of the magnetic moments (minimum in energy at $\theta = \pi/2$) and a collinear one (maximum in energy at $\theta = \pi/2$) and its range is also determined by λ_{\perp} . Therefore, the effect of the nonequilibrium magnetization is that of a quadrupole field distinguishing perpendicular from collinear alignment.

It seems surprising that a current can induce a biquadratic coupling for two identical semi-infinite magnetic layers. Likewise the departure of the $\cos \theta$ dependence of the magnetoresistance was unexpected. Yet when the latter effect was put into the context of spin filtering, it occurred naturally. Spin filtering can also be regarded as the driving force for the biquadratic coupling. As the nonequilibrium magnetization reorients itself in a truncated "corkscrew" towards the background magnetization (see Fig. 2), the field created by the nonequilibrium magnetization changes its direction accordingly. This leads to a perpendicular component in the nonequilibrium magnetization. As the nonequilibrium magnetization takes an intermediate orientation between the background magnetization of the two layers, the perpendicular component has contributions along the axes. Whereas the linear contributions cancel in a symmetric structure, the quadratic ones add. These can be either positive or negative depending on the direction of the current. The distinguishing feature of the biquadratic coupling contribution from the bilinear one is that the biquadratic coupling is always present. When the bilinear term exists, however, the biquadratic coupling is negligible for most structures apart from those based on half-metals.

E. Spin transfer and microwave generation

Berger¹⁵ and Slonczewski^{16,21} pointed out that at an interface between a ferromagnet and a nonmagnetic metal spin waves can be excited. Their theories build on band structure effects at such an interface where an incoming conduction electron has different refractive indices depending on its spin



orientation; the magnetic moment of an incoming conduction electron at an angle with the magnetization of the layer precesses in the local exchange field of the layer (see Sec. III B). Both theories are similar in relying on spin injection but fundamentally different in the way spin waves are excited. Slonczewski's main idea is that spin filtering is associated with the relaxation of angular momentum to the magnetic background. In Secs. III B and III C we have considered a rather general situation in which the nonequilibrium magnetization does not transfer its angular momentum to the background magnetization but to the lattice via τ_{sl} . The relaxation of magnetic moment to the background magnetization-spin transfer-exists only above a dynamic threshold determined by the spin-wave gap of the background magnetization. This is the distinguishing feature of Berger's theory.¹⁵

Experimentally, however, it has been found by Tsoi and co-workers^{30,31} and Myers and co-workers,²⁶ that a spin flip of the conduction electron can excite collective spin waves of distinct frequency above a certain threshold current *even in a single magnetic layer*. In Ref. 1 it was argued that above a dynamic threshold spin transfer is promoted by the scattering rate τ_{sd} which leads to spin-flip processes that excite the local moments to emit microwaves (spin waves). For these processes to occur, the nonequilibrium magnetization has to be sufficiently large at the interfaces of a ferromagnet. Therefore, the excitation processes can, for example, occur even for an initially unpolarized current, but may be absent in an antiparallel configuration of two ferromagnetic layers.

In the following we are interested in a setup similar to the experiments of Refs. 26–28: two Co layers, of which one is considered to be fixed and thicker than the spin diffusion length λ_F , separated by a Cu layer of negligible resistance and thickness. As previously, we take the magnetization to being constant throughout each layer and we choose the current along the *x* direction. In the present approximation, one can write the orientation of the nonequilibrium magnetization. Without the corrections in β^3 , Eq. (21) takes the form

FIG. 3. Normalized spin accumulation profile along the x axis in a Co/Cu/Co/Cu MML for (a) parallel, (b) antiparallel, and perpendicular orientations. The latter has a profile for the (c) y and (d) z components of the nonequilibrium magnetization, respectively. The profiles are normalized to the spin accumulation at the first Co/Cu interface at x=0. The first Cu layer has zero thickness and resistance. The second Co layer is x=5 nm thick. The spin diffusion lengths were taken 44 nm in Co and 150 nm in the Cu. The resistivity of the second Cu layer is about an order of magnitude less than in the Co.

$$D\frac{\partial^2 \delta \mathbf{M}_s}{\partial x^2} = \frac{\delta \mathbf{M}_s}{\tau_{sl}},\tag{28}$$

and can be solved for each layer seperately by making the following ansatz:¹

$$\delta \mathbf{M}_{s}^{n}(x) = \mathbf{m}_{1}^{n} e^{-x/\lambda_{sl}^{n}} + \mathbf{m}_{2}^{n} e^{x/\lambda_{sl}^{n}}, \qquad (29)$$

where $\mathbf{m}_{1(2)}^n$ are constant vectors for layer *n*, where *n* is either the left or right ferromagnetic layer or nonmagnetic layer. These constants need to be determined by appropriate boundary conditions along the *x* direction. At each interface both nonequilibrium magnetization $\delta \mathbf{M}_s$ and spin current \mathbf{J}_m are continuous. The nonequilibrium magnetization for our full model system turns out to be a superposition of the local moment vectors of the left (fixed) and right (free) magnetic layer. We write then $\delta \mathbf{M}_s(x) = \mathbf{m}_L(x) + \mathbf{m}_R(x)$, where $\mathbf{m}_{L(R)} \| \mathbf{M}_{s,L(R)}^0 \| \mathbf{M}_{d,L(R)} \equiv \mathbf{M}_{L(R)}$. The nonequilibrium magnetization profile is given in Fig. 3 for different orientations of the magnetic moment vectors.

When we introduced the effects of the NEXI in the equation of motion for the background magnetization in Ref. 25, we neglected the effects of relaxation between conduction electrons and background magnetization; i.e., we assumed that $\tau_{sd}^{-1} \approx \tau_{ds}^{-1} \approx 0$. Therefore, the equation of motion in Ref. 25 needs to be supplemented by these additional relaxation terms derived in the Appendix [Eq. (B11b)]:

$$\frac{\partial \mathbf{M}_{R}}{\partial t} = \gamma \mathbf{M}_{R} \times \mathbf{H}_{R} + \mathbf{R}_{R},$$

$$= \lambda_{sd} \mathbf{M}_{R} \times [(\mathbf{m}_{L} + \mathbf{m}_{R}) \times \mathbf{H}_{R}] - \lambda_{d} \mathbf{M}_{R} \times (\mathbf{M}_{R} \times \mathbf{H}_{R}),$$
(30)

where $\lambda_{d(sd)}^{-1} = \tau_{d(sd)} \mathbf{H}_R \cdot \mathbf{M}_R$. The nonlocal effects of the magnetization current are included in the effective field of the background magnetization. For the right layer, we have $\mathbf{H}_R = \mathbf{H}_{\text{eff},R}^0 + \alpha_{\text{eff}} \mathbf{M}_L + \alpha(\mathbf{M}_{s,R}^0 + \mathbf{m}_R)$. Instead of $\alpha_{\text{eff}} \mathbf{M}_L$ we

 $\mathbf{R}_{R} =$

introduced the effective nonequilibrium coupling field $\mathbf{H}_{R}^{\text{NEXI}} = \alpha_{\text{eff}} \mathbf{M}_{L}$ between the layers in Ref. 25.

The expression (30) is identical with the one given in Ref. 1 and a generalization of the damping term given in Ref. 25 in that it includes not only damping terms produced by \mathbf{H}_R but also the spin accumulation effects of each individual layer that enter as spin-transfer terms and are proportional to $\lambda_{sd} \propto \tau_{sd}^{-1}$. Spin transfer sets in when $\lambda_{sd}(\mathbf{m}_L + \mathbf{m}_R)$ overcomes $\lambda_d \mathbf{M}_R$ on the RHS of Eq. (30). For an infinitely thin plate magnetized in the direction perpendicular to the plate so that approximately $H_R^z \approx H_e - 4 \pi M_R \equiv -\omega_H / \gamma$ one obtains the condition¹

$$\hbar \omega_H + \Delta \mu(x_R) \left(\frac{\tau_{dl}}{\tau_{dl} + \tau_{ds}} \right) < 0, \tag{31}$$

where $\Delta \mu(x) = -\hbar \gamma [m_L(x) + m_R(x)]/\chi_s$ is the difference in "chemical potential" for spin-up and spin-down electrons.^{5,8} The presence of a second ferromagnetic layer is not necessary to predict current-induced excitations. As in Ref. 1, we could have introduced an effective field due to spin transfer instead, $\delta \mathbf{H} = (\mathbf{m}_L + \mathbf{m}_R) \tau_d / (\tau_{sd}\chi_d)$, where χ_d / τ_d is given by Eq. (B10). The modulus of this field shows a linear dependence on the current:

$$\delta H = J_e \Lambda_{\rm tr} \,. \tag{32}$$

The parameter Λ_{tr} is a length due to spin transfer. As for \mathbf{H}_{NEXI} , $\delta \mathbf{H}$ changes sign when the direction of the current is reversed.

We study now our example trilayer in more detail where the analysis is reduced to consider the free (right) layer only. Making use of the ansatz (29) and connecting the left ferromagnetic layer, the right ferromagnetic layer, and the nonmagnetic layer by virtue of the above-mentioned boundary conditions, one obtains explicit expressions for the nonequilibrium magnetization in each layer. Since the nonequilibrium magnetization does not change too significantly across the free layer, one can integrate over it. The injected nonequilibrium magnetizations in the right layer take the following form:

$$\mathbf{m}_{R} = -c \frac{\lambda_{F}}{2l_{R}} \frac{(\rho_{F}\lambda_{F} - \rho_{N}\lambda_{N})f_{1}}{\rho_{F}\lambda_{F} + \rho_{N}\lambda_{N}} \mathbf{M}_{R}, \qquad (33a)$$

$$\mathbf{m}_{L} = -c \frac{\lambda_{F}}{2l_{R}} \frac{\rho_{F} \lambda_{F} f_{1} + \rho_{N} \lambda_{N} f_{2}}{\rho_{F} \lambda_{F} + \rho_{N} \lambda_{N}} \mathbf{M}_{L}, \qquad (33b)$$

where $\lambda_{F(N)} = \sqrt{D_{F(N)} \tau_{sl}^{F(N)}}$. The functions $f_1 = (1 - e^{-l_R/\lambda_F})^2$ and $f_2 = 1 - e^{-2l_R/\lambda_F}$ depend on the thickness of the layer. For $l_R \ll \lambda_F$ one can retain only linear terms, so that $f_1 \approx 0$ and $f_2 \approx 2l_R/\lambda_F$. For Co/Cu, $\rho_F \lambda_F \gg \rho_N \lambda_N$ and in this limit the free layer can be considered as an "analyzer" of the injected spin current as the contribution of \mathbf{m}_R vanishes (refer to Fig. 3). This is also the case of maximal NEXI which can be seen from the effective coupling constant,

$$\frac{\alpha_{\text{eff}}}{\alpha} = c \,\nu \lambda_F \frac{(\rho_F \lambda_F - \rho_N \lambda_N) f_3}{\rho_F \lambda_F + \rho_N \lambda_N},\tag{34}$$

given for this particular case of a Co/Cu/Co/Cu structure. The above expression has been obtained from Eq. (26) neglecting the self-energy contributions. The function $f_3 = e^{-l_R/\lambda_F}(1 - e^{-l_R/\lambda_F})$ also depends on the thickness of the right layer so that for $l_R \ll \lambda_F$ one has approximately $f_3 \approx l_R/\lambda_F$. In other words, for this particular configuration the possibility for spin transfer of electrons incident from the left (fixed) layer is largest if NEXI is largest.

If the condition (31) is met and spin transfer sets in, the expressions (33) for \mathbf{m}_L and \mathbf{m}_R are no longer valid to describe the nonequilibrium magnetization, if we were to increase the current further. In such a case one needs to solve the dynamic problem of coupled Landau-Lifshitz equations as pointed out in Ref. 1. However, one can obtain a rough estimate of the *additional* contributions. When spin transfer occurs, the spin diffusion length in the ferromagnet is changed by the additional spin-transfer relaxation channel determined by τ_{sd} , $\lambda_F = \sqrt{D_F \tau_s}$ and $\tau_s^{-1} = \tau_{sd}^{-1} + \tau_{sl}^{-1}$.²⁷

In order to obtain the scattering time τ_{sd} , we can use the "detailed balance" relation between susceptibilities and scattering times of the *s*- and *d*-electron systems given in Eq. (6) of Ref. 1: $\chi_d \tau_{sd} = \chi_s \tau_{ds}$. This is helpful since the scattering time $\tau_{ds} \sim 10^{-9} - 10^{-10}$ has been estimated before.⁴⁵ The static susceptibility of the *d*-electron system is in the case of an infinitely thin plate approximately:

$$\chi_d^0 = \frac{\partial \mathbf{M}_d}{\partial \mathbf{H}_e} \approx \frac{\partial \mathbf{M}_d^0}{\partial \mathbf{H}_{\text{eff} d}^0} \frac{\partial \mathbf{H}_{\text{eff} d}^0}{\partial \mathbf{H}_e} = \frac{1}{4\pi}.$$

Thus, with a Pauli susceptibility $\chi_s \sim 10^{-6}$ we obtain for $\tau_{sd} \sim 10^{-14} - 10^{-15}$ s or a spin diffusion length $\lambda_F \sim 1-10$ nm which is of the order of the electron mean free path so that the concept of spin diffusion is to replaced by that of a simple decay in the nonequilibrium magnetization at the interface. This is in fact comparable to the length scale for which the nonequilibrium magnetization decays in Berger's theory.¹⁵ In other words, Berger's theory provides the spin-transfer time τ_{sd} which is an unknown parameter in our phenomenological approach. The reasoning is similar to Slonczewski's^{16,21} and goes as follows. The *s*-*d* scattering rate equals roughly the precession rate of the nonequilibrium magnetization. Using for example Eq. (B8a) and assuming that there is no external magnetic field, one obtains

$$\gamma \delta \mathbf{M}_{s} \times \alpha \mathbf{M}_{d} = \frac{J\rho}{\hbar |\mathbf{M}_{d}|} \, \delta \mathbf{M}_{s} \times \mathbf{M}_{d} = \frac{\delta \mathbf{M}_{s}}{\tau_{sd}}$$
(35)

or $\tau_{sd}^{-1} = J\rho/\hbar$, where *J* is the *s*-*d* coupling constant and ρ the number of spins per unit volume.²⁴ Typical values for $J\rho$ are 0.1–0.5 eV so that one has again $\tau_{sd} \sim 10^{-14} - 10^{-15}$ s. It is important to understand that expression (35) is a "snapshot" of the magnetization dynamics above threshold (31) and not a condition that determines the threshold.

We can now calculate the contributions to the nonequilibrium magnetization which occur *in addition* to the expressions (33) for \mathbf{m}_L and \mathbf{m}_R . Since the spin diffusion length is now at least an order of magnitude shorter, we use the limit where $l_R > \lambda_F$ so that $f_1 \approx 1$ and $f_2 \approx 1$:

$$\widetilde{\mathbf{m}}_{L(R)} \approx \mp (c - c_{\operatorname{crit}}) \frac{\lambda_F}{2l_R} \mathbf{M}_{L(R)}, \qquad (36)$$

where c_{crit} is the parameter c for the critical current determined from the condition (31). The total nonequilibrium magnetization in the right (free) layer is now $\delta \mathbf{M}_s = \mathbf{m}_L$ $+\mathbf{m}_{R}+\mathbf{\widetilde{m}}_{L}+\mathbf{\widetilde{m}}_{R}$. The last two terms are the above threshold contribution to the nonequilibrium magnetization $\delta \tilde{\mathbf{M}}_{s}$. Whereas the above threshold contribution of the NEXI vanishes $(f_3 \approx 0)$ due to the relaxation of the conduction electron spins to the local moments, spin transfer depends strongly on the mutual orientation of the layers. It vanishes for a parallel configuration of the layers and yields $\delta \tilde{M}_s$ $= c \lambda_F M / l_R$ for an antiparallel configuration. The treatments in Refs. 15,16 and 21 have, in essence, determined the nonequilibrium magnetization by only taking the contribution $\tilde{\mathbf{m}}_L$ for $c_{\text{crit}} = 0$ into account, therefore neglecting the NEXI, the dependence on the mutual orientation of the magnetization of the layers, and the possibility to have spin transfer for an initially unpolarized current.

F. Current-induced switching: Spin transfer vs spin injection

Both spin injection-the change in the nonequilibrium magnetization of a system due to the presence of an external source of nonequilibrium magnetization-and spin transfer-the dissipative loss of spin angular momentum from one magnetic subsystem to another-can, in principle, lead to current induced switching. This happens for sufficiently strong currents when \mathbf{H}_{NEXI} and $\delta \mathbf{H}$, respectively, drive the system into resonance at its eigenmodes. Since \mathbf{H}_{NEXI} can be treated as an effective Zeeman term, it usually excites many different modes instantaneously, so that the reversal process sets in immediately. On the contrary, $\delta \mathbf{H}$ is usually delayed by driving the low-frequency modes first. In this sense there is some similarity between spin transfer and an external pumping field. This analogy becomes useful to understand how spin transfer can actually trigger a magnetization reversal since low-frequency modes, in particular, the uniform mode, are as such not sufficient to drive a reversal process; only nonlinear spin waves do. In the case of an external pumping field, nonlinear spin waves are parametrically excited when the field amplitude is large. This picture of the switching dynamics corresponds then to a nonlinear multimode process similar to what has been studied by Safonov and Bertram for small grains in the absence of Gilbert damping⁴⁶ where after a period of uniform oscillations nonlinear excitations set in which can be thought of as "an overheating of the magnetic system" carrying out the reversal process.

In terms of spin-transfer switching requires that the system is constantly pumped or stimulated as in a LASER cavity. (The SWASER concept by Berger needs to be supplemented by a cavity.²¹) Hence, there are two constraints which have to be taken into account: relaxation of the excited modes and the geometry, i.e., the ratio between the region of spin transfer V_{tr} and the volume of the layer V_F . Via condition (31), V_{tr} depends critically on current; as has been

shown in the previous section \mathcal{V}_{tr} is, in general, restricted to the region at the interface and grows only slowly into the bulk of the layer with increasing current as also higher-frequency modes are excited.³¹

We would like to consider a simple example to determine the possible delay time. To turn over a local moment of S = 1, one needs two spin-1/2 conduction electrons to be flipped. Assuming that the damping of the modes occurs on much longer time scales as the spin transfer given condition (31) is satisfied, i.e., $\tau_d \gg \tau_{sd}$, one can calculate a delay time due to the spin transfer by setting the magnetization of the background magnetization equal to the transferred nonequilibrium magnetization:

$$t_{sw} = \frac{\tau_{sd}\tau_d}{\tau_d - \tau_{sd}} \frac{\mathcal{V}_F}{\mathcal{V}_{tr}} \frac{M}{\delta M_s} \approx \tau_{sd} \frac{\mathcal{V}_F}{\mathcal{V}_{tr}} \frac{M}{\delta M_s}.$$
 (37)

For small Co/Cu/Co pillar devices,²⁷ the ratio $\mathcal{V}_F/\mathcal{V}_{tr}$ might vary between 1 and 10. Due to the lateral confinement of the pillar, the spin waves are reflected at the boundaries keeping the energy in the layer. Assuming $\tau_{sd} \approx 10^{-14}$ s, the time delay to the switching would then be of the order of $t_{sw} = 1$ -10 ns for a ratio of $M/\delta M \sim 10^4 - 10^5$. These times are considerably longer than what one obtains from magnetic field switching but is in agreement of switching as a nonlinear multimode process where there is an initial delay time of pumped oscillations before the reversal process can set in.⁴⁶

The pillar structures show, however, different behavior for different mutual alignment of the magnetic layers as can be deduced from the data on critical switching current versus external magnetic field.²⁷ Whereas for switching from an antiparallel to a parallel configuration there seems to be reasonable agreement with the spin-transfer picture, the reverse switching operation requires much shorter relaxation times au_d to fit the data. From our discussion in the previous section there could be in principle three reasons: Because for the parallel configuration the nonequilibrium magnetization is much smaller at the interfaces, switching is more in line with what one would expect from switching due to spin injection.²⁵ On the other hand, one could argue that in the parallel configuration the contribution of spin transfer above threshold (31) is very small since the contributions $\tilde{\mathbf{m}}_{L}$ and $\tilde{\mathbf{m}}_{R}$ cancel each other so that it becomes more difficult to excite higher-frequency modes. Finally the switching is driven by the spin accumulation on the reverse side of the free magnetic layer. Clearer evidence that switching can be caused by spin transfer is given by Wegrowe and co-workers who observe switching only from an antiparallel to a parallel configuration in ferromagnetic nanowires but never the other way around.33

From the ratio $\mathcal{V}_F/\mathcal{V}_{tr}$, one notices, however, that spin transfer cannot lead to switching in experiments with point contacts; the contact area is small compared to the volume of the film and spin waves will be carried away from the contact region, allowing them to decay more easily. In fact, the point contact spectra d^2I/dV^2 were even featureless²⁸ or spin-wave excitations at least occurred for currents well above the switching of the domains at the point contact region.²⁶ Whereas in the former there existed a double-

domain structure in the contact region of which only one could be reversed by the current, so that a double-plateau structure was observed in the GMR, the latter showed clean switching behavior and can be explained by spin injection, i.e., the NEXI.²⁵

IV. CONCLUSIONS

For the development of electronic devices based on MML's where a current is driven across the interfaces it is important to employ a general but relatively simple picture that allows one to capture the main features of spin accumulation. In the present work we derived a simple diffusion equation [Eq. (10) where \mathbf{J}_M is given by Eq. (20)] describing the time and spatial dependence of the conduction electron nonequilibrium magnetization, i.e., the spin accumulation. This diffusion equation is a generalization of the one given in Ref. 5, allowing us to treat arbitrary configurations of the background magnetization within a single magnetic layer or that of a MML on which we focused in this work. Based on this equation, we could develop a coherent framework for treating apart from spin accumulation the phenomena of spin filtering, spin injection, and spin transfer.

The transient behavior of the spin accumulation at an interface between a ferromagnet and nonmagnetic layer could be pictured in terms of a diffusion from a constant source into a semi-infinite body. For metallic systems a steady state is reached within picoseconds and, therefore, does not present a technological obstacle to high-speed switching applications. We could show that the time-dependent process is, nevertheless, of interest as it provides a direct way to measure the amount of spin accumulation at an interface. As for comparison, in semiconductor structures the spin lifetimes can be several orders of magnitudes larger than in metals,⁴¹⁻⁴³ so that the time-dependent buildup could take a few nanoseconds, provided spins can be injected efficiently from a ferromagnet into the semiconductor. For transitionmetal semiconductor junctions this is a well-known problem⁴⁷ since the amount of spin accumulation m_0 at the interface is negligible. All quantities of experimental interest, such as the NEXI (not presented here for a semiconductor spacer layer), scale as ρ_F^*/ρ_N^* which tends to zero due to the large resistance of the semiconductor, as can be seen from Eq. (14), for example. It has been pointed out, however, by Grundler that for ballistic conduction across a band-matched MML, one could conceive an observable nonequilibrium magnetization in the semiconductor since the "resistance" in the semiconductor is effectively reduced for ballistic transport.⁴⁸ Promising candidates to measure the spin accumulation at an interface would be multilayers based on magnetic semiconductors^{49,50} or half-metals as CrO₂.⁵¹

By studying the effects of noncollinearity, we arrived at a new understanding of spin filtering being caused by two different spin diffusion lengths: a transverse one and parallel one. These gave a basis for the intuitive notion that conduction electrons whose moments are not aligned with the background magnetization ought to be more strongly scattered than those whose moments are aligned. In transition metals this gives a small, albeit, noticeable effect which manifests itself in the angular behavior of the CPP-MR.⁹ Yet when studying transport in MML's composed of half-metals, spin filtering can be very large and should not be neglected. Also we pointed out that a concomitant to spin filtering is a biquadratic coupling of the NEXI. All these effects arise from corrections to the spin accumulation when considering noncollinear geometries for MML's.

Further, it was shown how the nonequilibrium magnetization changes as soon as spin-transfer sets in above threshold. The threshold is determined by the amount of spin accumulation at the interfaces whereby the latter depends strongly on the mutual orientation of the magnetization of the layers. The dynamic picture developed by Slonczewski^{16,21} is only applicable in the above threshold regime; i.e., the spintransfer term [first term in Eq. (30)] dominates spin injection and cannot be used to determine the threshold itself. What is important to realize that both spin injection²⁵ and spin transfer¹ can drive a magnetization reversal. Which process will be dominant depends largely on the geometry of the MML under consideration.

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APPENDIX A: RELATION TO SPINOR REPRESENTATION OF CHEMICAL POTENTIAL

Instead of using the notion of separate currents for charge and spin, often another approach is used in terms of spinor currents. The latter occurs more naturally from microscopic calculations, where the wave functions of the system are written as Pauli spinors. For discussing the magnetic properties, however, it is more convenient to use the approach used in the present work. In the following, a few relations are stated that show equivalent expressions in the two different representations.

The electric current and spin current are defined in terms of spinor currents as

$$\vec{J}_e = \text{Tr}\{\vec{J}_{\sigma\sigma'}\},\tag{A1}$$

$$\vec{\mathbf{J}}_{M} = \frac{\mu_{B}}{e} \operatorname{Tr} \{ \boldsymbol{\sigma}_{\sigma\sigma'} \vec{J}_{\sigma'\sigma} \},$$
(A2)

where $\hat{\sigma}$ is the vector of Pauli matrices. This can be used to rewrite the spinor current,

$$\mathbf{\hat{J}} = \begin{pmatrix} \vec{J}_{\uparrow\uparrow} & \vec{J}_{\uparrow\downarrow} \\ \vec{J}_{\downarrow\uparrow} & \vec{J}_{\downarrow\downarrow} \end{pmatrix} = \begin{pmatrix} \vec{J}_e + \frac{e}{\mu_B} \vec{J}_M^z & \frac{e}{\mu_B} \vec{J}_M^+ \\ \frac{e}{\mu_B} \vec{J}_M^- & \vec{J}_e - \frac{e}{\mu_B} \vec{J}_M^z \end{pmatrix}, \quad (A3)$$

where $\vec{J}_M^{\pm} = \vec{J}_M^x \pm i \vec{J}_M^y$. In analogy, it is possible to give an expression of the chemical potential in spinor representation,

$$\hat{\mu} = \frac{1}{2} \begin{pmatrix} \bar{\mu} + \frac{\hbar \gamma}{\chi_s} \delta M^z & \frac{\hbar \gamma}{\chi_s} \delta M^+ \\ \frac{\hbar \gamma}{\chi_s} \delta M^- & \bar{\mu} - \frac{\hbar \gamma}{\chi_s} \delta M^z \end{pmatrix}, \quad (A4)$$

where $\delta M^{\pm} = \delta M^x \pm \delta M^y$ and $\overline{\mu} = 2$ eV is proportional to the applied bias. It is also interesting to write down Ohm's law which can be derived by applying the above relations to Eqs. (5) and (7). After a couple of rather involved transformations, one arrives at

$$\vec{\nabla}\hat{\mu} = -\frac{e}{\sigma^*} \left(\hat{\mathbf{J}} + \frac{1}{1-\beta^2} \hat{p} \hat{\mathbf{J}} \hat{p} \right), \quad \hat{p} = \begin{pmatrix} 1-p^z & -p^+ \\ -p^- & 1+p^z \end{pmatrix},$$
(A5)

where $p^{\pm} = p^x \pm i p^y$. This matrix can also be expressed as $p_{\sigma\sigma'} = \delta_{\sigma\sigma'} - \sigma_{\sigma\sigma'} \cdot \mathbf{p}$, where **p** is the polarization vector introduced before. For collinear alignment, this expression reduced to the standard result⁸

$$\vec{\nabla}\mu_{\sigma} = -e\rho_F^{\sigma}\vec{J}_{\sigma}, \qquad (A6)$$

where the second spin index has been suppressed.

APPENDIX B: LANDAU-LIFSHITZ EQUATIONS

Although one could derive the full Landau-Lifshitz equations from microscopic theories,^{52,53} we follow the spirit of our phenomenological approach and turn to thermodynamic methods for irreversible processes as outlined in Refs. 37, 54, and 55. This allows us to describe the magnetization dynamics with rather general assumptions and treat the effects of strong interatomic exchange interactions and weak *s*-*d* coupling, simultaneously.

The equilibrium state of the magnetic system will be disturbed when a magnetic field \mathbf{H}_{eff} changes quickly. Its temperature and magnetization \mathbf{M} change so that due to relaxation processes they do no longer satisfy the equation of state. Rather the equation of state now defines a field $\mathbf{\tilde{H}}$ which differs from \mathbf{H}_{eff} . The difference $\mathbf{H}^* = \mathbf{H}_{eff} - \mathbf{\tilde{H}}$ can be regarded as an additional magnetic field as in the case of the magnetization current. By definition, one always has $\mathbf{\tilde{H}} \times \mathbf{M}$ = 0. Since relaxation processes are present, the magnetic subsystem generates entropy:

$$TdS = dU - \mathbf{H}_{\text{eff}} \cdot d\mathbf{M} + \mathbf{H}^* \cdot d\mathbf{M}, \tag{B1}$$

where U is the internal energy of the magnetic subsystem. The first two terms determine the equilibrium contribution and the last term the nonequilibrium contribution. One therefore finds for the rate of entropy production of the magnetic subsystem,

$$T\frac{d\Delta S}{dt} = \mathbf{H}^* \cdot \frac{d\mathbf{M}}{dt}.$$
 (B2)

By means of irreversible thermodynamics the components of dM_i/dt are linear functions of the components of the additional field,

$$\frac{dM_i}{dt} = \sum_{j=1}^3 \mathcal{L}^{ij} H_j^*, \qquad (B3)$$

where \mathcal{L}^{ij} is a tensor of kinetic coefficients whose components are functions of the magnetization, i, j = x, y, z. The Onsager theorem states that $\mathcal{L}^{ij}(\mathbf{M}) = \mathcal{L}^{ji}(-\mathbf{M})$, so that, when writing $\mathcal{L}^{ij} = \mathcal{L}_s^{ij} + \mathcal{L}_a^{ij}$ in terms of a symmetric and antisymmetric tensor, one finds $\mathcal{L}_s^{ij}(\mathbf{M}) = \mathcal{L}_s^{ji}(-\mathbf{M})$ and $\mathcal{L}_a^{ij}(\mathbf{M}) = -\mathcal{L}_a^{ji}(-\mathbf{M})$. The components of the antisymmetric tensor form in linear order an axial vector which is an odd function of magnetization, $\mathcal{L}_a^{ij} = \gamma \Sigma_{k=1}^3 \varepsilon_{ijk} M_k$, where γ will have the meaning of the magnetomechanical ratio. Assuming the presence of an axial anisotropy in an isotropic ferromagnet, the symmetric tensor is taken diagonal in linear order \mathcal{L}_s^{ij} $= \lambda_i \delta_{ij}$ and will be responsible for the relaxation processes in the system. Now, we can write Eq. (B3) as

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \lambda \mathbf{H}^*, \qquad (B4)$$

where we assumed for simplicity $\lambda_i = \lambda_j$. At temperatures far below the Curie point, an external field produces only a change in the direction of the magnetization not in its magnitude. If we take the scalar product of Eq. (B4) with **M** and require **M**² being constant, the motion of **M** can be considered as being confined to the surface of a sphere. By virtue of $\tilde{\mathbf{H}} \times \mathbf{M} = 0$, one finds the standard Landau-Lifshitz equations

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}}).$$
(B5)

In order to describe the magnetic properties of the combined spin systems of the paramagnetic conduction electrons (referred to as *s* electrons) and the localized magnetic moments of the core electrons (referred to as *d* electrons), we must, however, consider two coupled equations of motion for the magnetization \mathbf{M}_s and \mathbf{M}_d , respectively. Therefore, we write similar to Eq. (B3) a system of equations that contain in addition terms that connect both electron subsystems with each other:

$$\frac{dM_{s}^{i}}{dt} = \sum_{j=1}^{3} \mathcal{L}_{ss}^{ij} H_{s}^{*j} + \sum_{j=1}^{3} \mathcal{L}_{sd}^{ij} H_{d}^{*j}, \qquad (B6a)$$

$$\frac{dM_d^i}{dt} = \sum_{j=1}^3 \mathcal{L}_{dd}^{ij} H_d^{*j} + \sum_{j=1}^3 \mathcal{L}_{ds}^{ij} H_s^{*j}, \qquad (B6b)$$

where $\mathbf{H}_{s(d)}^* = \mathbf{H}_{\text{eff},s(d)} - \widetilde{\mathbf{H}}_{s(d)}$. The effective fields are different for *s*- and *d*-electron systems,

$$\mathbf{H}_{\text{eff},s} = \mathbf{H}_{e} + \alpha \mathbf{M}_{d} ,$$
$$\mathbf{H}_{\text{eff},d} = \mathbf{H}_{\text{eff},d}^{0} + \alpha \mathbf{M}_{s} + \mathbf{H}_{dd} ,$$

in that apart from the different *s*-*d* exchange contributions the *d*-electron system includes the effect of direct exchange, anisotropy, dipole-dipole interactions, and the Oerstedt field of the conduction-electron current: $\mathbf{H}_{eff,d}^0 = \mathbf{H}_e + \mathbf{H}_{dip} + \mathbf{H}_{an}$ $+ \mathbf{H}_{dd} + \mathbf{H}_{ind}$. The fields $\mathbf{\tilde{H}}_{s(d)}$ are connected to the magnetizations by the relationships $\mathbf{M}_{s(d)} = \chi_{s(d)} \mathbf{\tilde{H}}_{s(d)}$. In the ferromagnetic subsystem the susceptibility depends on the history of the applied field and has only a formal meaning. It has to be replaced by requiring constancy of the magnitude of the magnetization \mathbf{M}_d . Conversely, since we can assume the system of conduction electrons being paramagnetic, instead of the magnitude of the magnetization being fixed, its susceptibility is constant and given approximately by χ_s $= (\hbar \gamma)^2 N(\varepsilon_F)/4$. We express the kinetic coefficients as

$$\mathcal{L}_{ss}^{ij} = \frac{\chi_s}{\tau_s} \delta_{ij} + \gamma_s \sum_{k=1}^{3} \varepsilon_{ijk} M_s^k, \quad \mathcal{L}_{sd}^{ij} = -\frac{\chi_d}{\tau_{ds}} \delta_{ij},$$
(B7a)

$$\mathcal{L}_{dd}^{ij} = \frac{\chi_d}{\tau_d} \delta_{ij} + \gamma_d \sum_{k=1}^{3} \varepsilon_{ijk} M_d^k, \quad \mathcal{L}_{ds}^{ij} = -\frac{\chi_s}{\tau_{sd}} \delta_{ij},$$
(B7b)

3

where $\tau_s^{-1} = \tau_{sd}^{-1} + \tau_{sl}^{-1}$ and $\tau_d^{-1} = \tau_{ds}^{-1} + \tau_{dl}^{-1}$. We introduced different magnetomechanical ratios for *s* and *d* spin systems and relaxation times that either couple the spin systems, τ_{sd} and τ_{ds} , or couple to some heat bath such as the lattice, τ_{sl} and τ_{dl} . The equations of motion are not yet complete, since we have not taken into account the presence of a magnetization current in Eq. (B6a). The total derivative with respect to time is replaced by $\partial \mathbf{M}_s / \partial t + \vec{\nabla} \cdot \vec{\mathbf{J}}_M$ with the help of relation (9), so that the equations of motion have formally the same form as in Ref. 52:

$$\frac{\partial \mathbf{M}_s}{\partial t} - \gamma_s \mathbf{M}_s \times \mathbf{H}_{\text{eff},s} + \vec{\nabla} \cdot \vec{\mathbf{J}}_M = \frac{\partial \mathbf{M}_d}{\tau_{ds}} - \frac{\partial \mathbf{M}_s}{\tau_s}, \quad (B8a)$$

$$\frac{\partial \mathbf{M}_d}{\partial t} - \gamma_d \mathbf{M}_d \times \mathbf{H}_{\text{eff},d} = \frac{\delta \mathbf{M}_s}{\tau_{sd}} - \frac{\delta \mathbf{M}_d}{\tau_d}.$$
 (B8b)

Since the system relaxes to its instantaneous or local equilibrium value of magnetization, the nonequilibrium magnetization takes the form

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$$\delta \mathbf{M}_{s(d)} = \mathbf{M}_{s(d)} - \chi_{s(d)} \mathbf{H}_{\text{eff},s(d)} \,. \tag{B9}$$

There are a couple of points we would like to discuss. (1) One can see that the scattering rates between *s*- and *d*-electron systems are exactly balanced to their lowest order; a scattering "out" term in one subsystem has always an equivalent scattering "in" term in the other one. They conserve the total spin angular momentum $\mathbf{M}=\mathbf{M}_s+\mathbf{M}_d$ in the system. (2) If we assume for a moment that we are interested in combined solutions of Eqs. (B8a) and (B8b) where (B8b) is independent of time and assume that $\mathbf{M}_d \| \mathbf{H}_{\text{eff},d}^0$ and γ_s $= \gamma_d$, we obtain the equation of motion (10). (3) In order to point out the difference between a fully paramagnetic system treated in Ref. 52 and the system under study, we shall make use of the condition that $|\mathbf{M}_d|$ is constant. Multiplying Eq. (B8b) by \mathbf{M}_d , one can obtain the following relation:

$$\frac{\chi_d}{\tau_d} = \frac{1}{\mathbf{H}_{\text{eff},d} \cdot \mathbf{M}_d} \left(\frac{|\mathbf{M}_d|^2}{\tau_d} - \frac{\delta \mathbf{M}_s \cdot \mathbf{M}_d}{\tau_{sd}} \right), \quad (B10)$$

which, when inserted into Eqs. (B8a) and (B8b), allows one to rewrite the RHS of these equations as

$$\frac{\delta \mathbf{M}_{d}}{\tau_{ds}} - \frac{\delta \mathbf{M}_{s}}{\tau_{s}} = \lambda_{ds} \mathbf{M}_{d} \times (\mathbf{M}_{d} \times \mathbf{H}_{\text{eff},d}) - \frac{\delta \mathbf{M}_{s}}{\tau_{s'}} - \lambda_{s''} \mathbf{M}_{d} \times (\delta \mathbf{M}_{s} \times \mathbf{H}_{\text{eff},d}), \quad (B11a)$$

$$\frac{\delta \mathbf{M}_{s}}{\tau_{sd}} - \frac{\delta \mathbf{M}_{d}}{\tau_{d}} = \lambda_{sd} \mathbf{M}_{d} \times (\delta \mathbf{M}_{s} \times \mathbf{H}_{\text{eff},d}) -\lambda_{d} \mathbf{M}_{d} \times (\mathbf{M}_{d} \times \mathbf{H}_{\text{eff},d}), \quad (B11b)$$

where we introduced $\lambda_{\alpha}^{-1} = \tau_{\alpha} \mathbf{H}_{\text{eff},d} \cdot \mathbf{M}_d$ and the relaxation times $\tau_{s'}^{-1} = \tau_{sl}^{-1} + \tau_{sd}^{-1} (\tau_{dl} + \tau_{ds})^{-1} \tau_{ds}$, and $\tau_{s''}^{-1} = \tau_{sd}^{-1} (\tau_{dl} + \tau_{ds})^{-1} \tau_{dl}$, which couple in Eq. (B11a) the nonequilibrium magnetization $\delta \mathbf{M}_s$ explicitly to the relaxation processes of the magnetic background. These damping terms explicitly conserve the magnitude of the magnetization of the *d*-electron system which is not the case for the *s*-electron system. (4) One can see from Eq. (B8b) or (B11b) that the scattering "in" term can lead to excitations in the *d*-electron system via the nonequilibrium magnetization $\delta \mathbf{M}_s$.

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