PHYSICAL REVIEW B **92**, 180412(R) (2015)

Electron-magnon scattering in magnetic heterostructures far out of equilibrium

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(Received 1 April 2015; published 23 November 2015)

We present a theory of out-of-equilibrium ultrafast spin dynamics in magnetic heterostructures based on the *s*-*d* model of ferromagnetism. Both in the bulk and across interfaces, the exchange processes between the itinerant *s* and the localized *d* electrons are described by kinetic rate equations for electron-magnon spin-flop scattering. In our treatment, the magnon distribution function remains nonthermalized on the relevant time scales of the demagnetization process, and the relaxation of the out-of-equilibrium spin accumulation among itinerant electrons provides the principal channel for dissipation of spin angular momentum from the combined electronic system.

DOI: 10.1103/PhysRevB.92.180412 PACS number(s): 72.25.Mk, 72.10.Di, 72.20.Pa, 75.40.Gb

Controlling spin flow in magnetic heterostructures at ultrafast time scales using femtosecond laser pulses opens intriguing possibilities for spintronics [1]. These laser-induced perturbations [2,3] stir up the most extreme regime of spin dynamics, which is governed by the highest energy scale associated with magnetic order: the microscopic spin exchange that controls the ordering temperature T_C. In contrast, at microwave frequencies the ferromagnetic dynamics in the bulk are well described by the Landau-Lifshitz-Gilbert (LLG) phenomenology [4], which has been successfully applied to the problem of the ferromagnetic resonance (FMR) [5]. At finite temperatures below T_C, the spin Seebeck and Peltier effects [6,7] describe the coupled spin and heat currents across interfaces in magnetic heterostructures. Despite their different appearances, the microwave, thermal, and ultrafast spin dynamics are all rooted in the exchange interactions between electrons. It is thus natural to try to advance a microscopic understanding of the ultrafast dynamics based on the established phenomena at lower energies.

Although some attempts have been made [8,9] to extend the LLG phenomenology to describe ultrafast demagnetization in bulk ferromagnets, no firm connection exists between the ultrafast spin generation at interfaces and the microwave spin-transfer and spin-pumping effects [10] or the thermal spin Seebeck and Peltier effects. In this Rapid Communication, we unify the energy regimes of microwave, thermal, and ultrafast spin dynamics in magnetic heterostructures from a common microscopic point of view, so that the parameters that control the high and low energy limits of spin relaxation originate from the same electron-magnon interactions. In addition to the unified framework, this Rapid Communication's unique contributions are the history-dependent, nonthermalized magnon distribution function and the crucial role of the out-of-equilibrium spin accumulation among itinerant electrons as the bottleneck that limits the dissipation of spin angular momentum from the combined electronic system.

The first reports on ultrafast demagnetization in Ni [11] challenged the conventional view of low-frequency magnetization dynamics at temperatures well below T_C. A multitude of mechanisms and scenarios have been suggested to explain the observed quenching of the magnetic moment. Some advocate direct coherent spin transfer induced by the irradiating laser light as the source of demagnetization [12]. Alternative theories argue that ultrafast spin dynamics arise indirectly through incoherent heat transfer to the electron system [13,14]. Recent experiments have demonstrated that nonlocal laser irradiation also induces ultrafast demagnetization [15], and atomistic modeling [16] supports the view that heating of magnetic materials is sufficient to induce ultrafast spin dynamics.

Terahertz (THz) magnon excitations in metallic ferromagnets have recently been proposed as an important element of ultrafast demagnetization by several authors [17,18]. The elementary interaction that describes these excitations is the electron-magnon scattering. Our theory is based on kinetic equations for the low-frequency spin and charge transport associated with the microwave magnetization dynamics in heterostructures [19] and with the linear spin-caloritronic response [7,20]. We extend these theories to treat far-from-equilibrium spin dynamics, in which transport is dominated by magnons and hot electrons. Electron-magnon scattering plays a critical role in this regime. We base our understanding of this interaction on the *transverse spin diffusion* [21] in the bulk and the *spin-mixing physics*, e.g., spin transfer and spin pumping [19,22], at the interfaces.

In our approach, we assume that the localized spins that result in the experimentally detectable macroscopic magnetization [23] are distinct from the itinerant electrons at the energy scales of interest. According to the accepted description of relaxation in ferromagnetic metals, the loss of energy and angular momentum from localized *d* electrons is mediated by the exchange interaction to the itinerant *s* electrons. The spin transfer from *d* to *s* states is accompanied by the relaxation of the *s* electron spins to the lattice through an incoherent spin-flip process caused by the spin-orbit coupling. Mitchell formulated such a model several decades ago to describe the longitudinal relaxation of ferromagnetic metals [24]. A similar description was later employed to describe Gilbert damping in ferromagnets at low frequencies [25,26].

In the following, we start by outlining the basic quantum-kinetic formalism for ultrafast spin dynamics in bulk ferromagnetic metals. Later, we show that the ferromagnet

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(F) | normal-metal (N) interfacial spin transport due to electron-magnon interactions follows a similar essential structure, unifying the bulk and interfacial spin dynamics in magnetic heterostructures. The Hamiltonian that describes F is $\hat{H} = \hat{H}_0 + \hat{H}_{sd}$, where $\hat{H}_0$ consists of decoupled s- and d-electron energies, including the kinetic energy of the itinerant electron bath, the d-d exchange energy, dipolar interactions, and the crystalline and Zeeman fields. The s-d interaction is

$$\hat{H}_{sd} = J_{sd} \sum_j \mathbf{S}_j^d \cdot \mathbf{s}(\mathbf{r}_j), \tag{1}$$

where J_{sd} is the exchange energy and $\mathbf{S}_j^d$ $[\mathbf{s}(\mathbf{r}_j)]$ is the d-electron (s-electron) spin vector (spin density) at lattice point j. We express the s-d interaction in terms of bosonic and fermionic creation and annihilation operators:

$$\hat{H}_{sd} = \sum_{qkk'} V_{qkk'} a_q c_{k\uparrow}^\dagger c_{k'\downarrow} + \text{H.c.}, \tag{2}$$

where $a_q^\dagger$ (a_q) is the Holstein-Primakoff creation (annihilation) operator for magnons with wave number q and $c_{k\sigma}^\dagger$ $(c_{k\sigma})$ is the creation (annihilation) operator for s electrons with momentum k and spin σ. $\hat{H}_{sd}$ describes how an electron flips its spin while creating or annihilating a magnon with momentum q and spin $\hbar$. The scattering strength is determined by the matrix element $V_{qkk'}$.

In Eq. (2), we have disregarded terms of the form $\sim a_q^\dagger a_{q'} c_{k\sigma}^\dagger c_{k'\sigma}$, which describe multiple-magnon scattering and do not contribute to a net change in magnetization along the spin-quantization axis. We have also disregarded higher-order terms associated with the Holstein-Primakoff expansion. Fully addressing magnonic correlation effects in the ultrafast regime would require a rigorous approach, e.g., using nonequilibrium Keldysh formalism [27]. However, when the s-d coupling (1) is not the dominant contribution to $\hat{H}$, we follow a mean-field approach and use Fermi's golden rule to compute the spin transfer between the s and d subsystems. We assume that all relevant energy scales are much smaller than the Fermi energy $\epsilon_F \equiv k_B T_F$ of the itinerant s electrons. In this limit, the electronic continuum remains largely degenerate, with electron-hole pairs present predominantly in the vicinity of the Fermi level.

We orient the coordinate system such that the localized spin density points in the negative z direction at equilibrium, with saturation value S (in units of $\hbar$). In the presence of a magnon density n_d, the longitudinal spin density becomes $S_z = n_d - S$. The magnons are assumed to follow a quadratic dispersion relation $\epsilon_q = \hbar\omega_q = \epsilon_0 + Aq^2$, where ϵ_0 is the magnon gap and A parametrizes the stiffness of the ferromagnet. $\langle a_q^\dagger a_{q'} \rangle = n(\epsilon_q)\delta_{qq'}$ defines the magnon distribution function $n(\epsilon_q)$, which is related to the total magnon density through $n_d = \int_{\epsilon_0}^{\epsilon_b} d\epsilon_q \mathcal{D}(\epsilon_q) n(\epsilon_q)$, where $\mathcal{D}(\epsilon_q) = \sqrt{\epsilon_q - \epsilon_0}/(4\pi^2 A^{3/2})$ is the magnon density of states. The integral over $\mathcal{D}(\epsilon_q)$ is cut off at an energy corresponding to the bandwidth, $\epsilon_b \sim k_B T_C$, which is the magnon energy at the edge of the Brillouin zone.

Because of the s-d interaction (1), the itinerant s electrons have a finite spin density at equilibrium (see Fig. 1). One of the key driving forces of the out-of-equilibrium spin dynamics is the spin accumulation $\mu_s \equiv \delta\mu_\uparrow - \delta\mu_\downarrow$. The bands for spin-up and spin-down electrons are split by $\Delta_{xc} \sim J_{sd} S a^3$, where a is the lattice constant of F. By introducing a dynamic exchange splitting, we can write $\mu_s = \delta n_s / D + \delta\Delta_{xc}$ [28], where δn_s is the out-of-equilibrium spin density of the s electrons, $D = 2D_\uparrow D_\downarrow/(D_\uparrow + D_\downarrow)$, and $D_{\uparrow(\downarrow)}$ is the density of states for spin-up (spin-down) electrons at the Fermi level. Because the mean-field band splitting due to the s-d exchange vanishes when the d orbitals are fully depolarized, $\delta\Delta_{xc}/\Delta_{xc} = \pm n_d/S$, where the sign determines whether the s and d orbitals couple ferromagnetically $(-)$ or antiferromagnetically $(+)$.

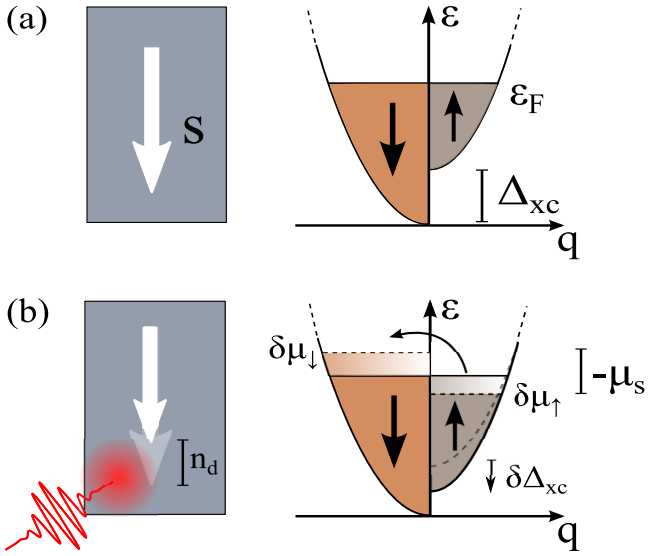

FIG. 1. (Color online) (a) Sketch of the density of s electron states in a ferromagnetic metal with saturation spin density S. At equilibrium, the exchange splitting Δ_{xc} shifts the bands for spin-up and spin-down electrons. (b) A laser pulse heats the s electron bath. The out-of-equilibrium spin accumulation $\mu_s = \delta\mu_\uparrow - \delta\mu_\downarrow$ results from two different mechanisms: (1) electron-magnon scattering induces a spin density among the s electrons, and (2) the mean-field exchange splitting is shifted by $\delta\Delta_{xc}$ by the induced magnon density n_d.

The rate of spin transfer (per unit volume) between the s and d subsystems due to electron-magnon spin-flop processes is determined from Eq. (2) by Fermi's golden rule [22]:

$$I_{sd} = \int_{\epsilon_0}^{\epsilon_b} d\epsilon_q \Gamma(\epsilon_q)(\epsilon_q - \mu_s)\mathcal{D}(\epsilon_q)[n_{\text{BE}}(\epsilon_q - \mu_s) - n(\epsilon_q)], \tag{3}$$

where $\Gamma(\epsilon_q)$ parametrizes the scattering rate at energy ϵ_q. In the derivation of Eq. (3) we have assumed that the kinetic energy of the itinerant electrons and the empty states (holes) thermalize rapidly due to Coulombic scattering and that they are distributed according to Fermi-Dirac statistics. Correspondingly, after standard manipulations [29], it can be shown that the electron-hole pairs follow the Bose-Einstein (BE) distribution function, $n_{\text{BE}}(\epsilon_q - \mu_s) = \{\exp[\beta_s(\epsilon_q - \mu_s)] - 1\}^{-1}$, at the electron temperature $T_s = 1/(k_B\beta_s)$. The number of available scattering states is influenced by the spin accumulation μ_s, as expected.

In contrast to the low-energy treatment in Ref. [22], the derivation of Eq. (3) does not constrict the form of the magnonic distribution $n(\epsilon_q)$ to the thermalized BE distribution

function. When the time scale of the s-d scattering is faster than the typical rates associated with magnon-magnon interactions, magnons are *not* internally equilibrated shortly after rapid heating of the electron bath, as also predicted by atomistic modeling [30]. Consequently, the occupation of the magnon states can deviate significantly from the BE distribution on the time scale of the demagnetization process. Our treatment of this central aspect differs from that of Ref. [31], in which the excited magnons are assumed to be instantly thermalized with an effective spin temperature and zero chemical potential and the thermally activated electron bath is assumed to be unpolarized.

The s-d scattering rate can be phenomenologically expanded as $\Gamma(\epsilon_q) = \Gamma_0 + \chi(\epsilon_q - \epsilon_0)$, where Γ_0 (which vanishes in the simplest Stoner limit [21]) parametrizes the scattering rate of the long-wavelength magnons and $\chi(\epsilon_q - \epsilon_0) \propto q^2$ describes the enhanced scattering of higher-energy magnons due to transverse spin diffusion [21]. In general, one might expect other terms of higher order in q to be present in this expansion as well. We will, however, limit ourselves to extrapolating $\Gamma(\epsilon_q)$ up to the bandwidth ϵ_b, which should be sufficient for qualitative purposes.

Neglecting any direct relaxation of magnons to the static lattice or its vibrations (i.e., phonons), $\partial_t n_d = I_{sd}/\hbar$. The equations of motion for the s-electron spin accumulation and the d-electron magnon distribution function are

$$\partial_t \mu_s = -\frac{\mu_s}{\tau_s} + \frac{\rho}{\hbar} I_{sd}, \tag{4}$$

$$\partial_t n(\epsilon_q) = \frac{\Gamma(\epsilon_q)}{\hbar}(\epsilon_q - \mu_s)[n_{\rm BE}(\epsilon_q - \mu_s) - n(\epsilon_q)], \tag{5}$$

where ρ determines the feedback of the demagnetization on μ_s and τ_s is the spin-orbit relaxation time for the s-electron spin density relaxing to the lattice. τ_s is typically on the order of picoseconds [32] and represents the main channel for the dissipation of angular momentum out of the combined electronic system. In general, τ_s also depends on the kinetic energy of the hot electrons after laser-pulse excitation. However, this discussion is beyond the scope of this Rapid Communication, and we assume that τ_s is independent of energy. $\rho = \rho_D + \rho_\Delta = -1/D \pm \Delta_{xc}/S$ includes effects arising from both the out-of-equilibrium spin density and the dynamic exchange splitting. For ferromagnetic $(-)$ s-d coupling, these effects add up, whereas for antiferromagnetic $(+)$ coupling, they compete.

At low temperatures, low-frequency excitations result in purely transverse spin dynamics. In the classical picture of rigid magnetic precession, the transverse relaxation time τ_2 is determined by the longitudinal relaxation time τ_1 as follows: $1/\tau_2 = 1/(2\tau_1) = \alpha\omega$, where α is the Gilbert damping parameter and ω is the precession frequency. Indeed, in the limit $(q, T_s) \to 0$, Eq. (3) yields

$$\partial_t n_d \to -\frac{\Gamma_0}{\hbar}\epsilon_0 n_d, \tag{6}$$

which is identical to the LLG phenomenology, indicating that $\epsilon_0 = \hbar\omega$ and thus $\Gamma_0 = 2\alpha$. This result establishes the important link between the scattering rate Γ_0 in this treatment and the Gilbert damping parameter that is accessible through FMR experiments.

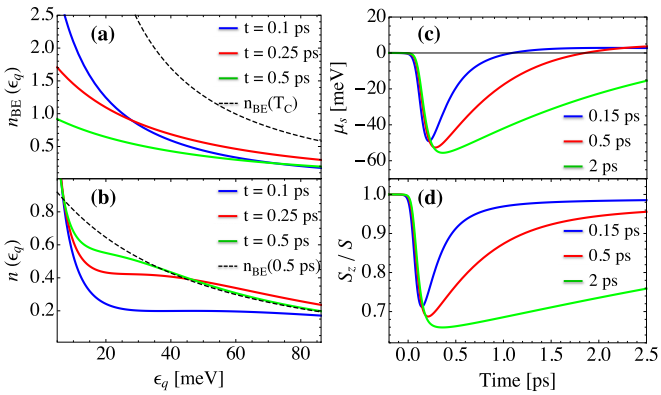

FIG. 2. (Color online) Numerical solutions of Eqs. (4) and (5) after T_s is increased from 10^2 to 10^3 K (T_C) within 50 fs with a decay time of 2 ps. $\epsilon_0 = 5$ meV, $A = 0.6$ meV nm^2, $\rho = 6$ meV nm^3, $\tau_s = 2$ ps, and $\alpha^* = 10\alpha = 0.1$. (a) The itinerant electron-hole pair distribution $n_{\rm BE}(\epsilon - \mu_s)$ is rapidly depleted by the spin accumulation μ_s that is built up via electron-magnon scattering. (b) In the magnon distribution $n(\epsilon_q)$ the high-energy magnon states are rapidly populated, whereas the low-energy states remain unaffected on short time scales. (c) Time evolution of the spin accumulation $\mu_s(t)$ and (d) the longitudinal spin density $-S_z(t)$ with different decay times of T_s: 0.15, 0.5, and 2 ps.

In the opposite high-frequency limit, pertinent to ultrafast demagnetization experiments, we consider F to be in a low-temperature equilibrium state before being excited by a THz laser pulse at $t = 0$, upon which the effective temperature of the itinerant electron bath instantly increases such that $T_s \gtrsim T_C$. This regime is clearly beyond the validity of the LLG phenomenology, which is designed to address the low-energy extremum of magnetization dynamics. Dissipation in the LLG equation, including relaxation terms based on the stochastic Landau-Lifshitz-Bloch treatment [14,33], is subject to a simple Markovian environment without any feedback or internal dynamics. This perspective must be refined for high frequencies when no subsystem can be viewed as a featureless reservoir for energy and angular momentum.

To appreciate the nonthermalized nature of the excited magnons, we consider the limit in which μ_s is small compared with ϵ_0 and no magnons are excited $[n(\epsilon_q) = 0]$ for $t < 0$. After rapid heating of the itinerant electrons at $t = 0$, the time evolution of the magnonic distribution follows

$$n(\epsilon_q, t) \approx n_{\rm BE}(\epsilon_q, t)[1 - e^{-\Gamma(\epsilon_q)\epsilon_q t/\hbar}]. \tag{7}$$

This result implies that, initially, the high-energy states are populated much faster than low-energy states. When μ_s becomes sizable, the coupled equations (4) and (5) must be solved subject to a suitable $T_s(t)$. Figures 2(a) and 2(b) present numerical solutions of (4) and (5) when T_s is increased from 10^2 to 10^3 K within 50 fs with a decay time of 2 ps. By comparison, internal magnon-magnon interactions equilibrate the distribution function on the time scale $\tau_{\rm eq}^{-1} \sim \hbar^{-1}\epsilon_m[\epsilon_m/(k_B T_C)]^3$ [22], where ϵ_m is a characteristic energy of the thermal magnon cloud. For short times, I_{sd} [Eq. (3)] dominates the magnon dynamics, and we expect the magnon population to significantly differ from the thermalized BE distribution.

When $T_s > T_C$, the thermally excited electron-hole pairs are populated in accordance with the classical Rayleigh-Jeans distribution, $n_{\mathrm{BE}}(\epsilon_q - \mu_s) \to k_B T_s/(\epsilon_q - \mu_s)$. Assuming, for simplicity, that the expansion for $\Gamma(\epsilon_q)$ is valid throughout the Brillouin zone, Eq. (3) yields $\partial_t n_d|_{t\to 0} = I_{sd}(0)/\hbar = [\Gamma_0 + 3\chi(\epsilon_b - \epsilon_0)/5]k_B T_s S/\hbar$. Thus, the demagnetization rate is initially proportional to the temperature of the electron bath but is reduced by the lack of available scattering states for high-energy magnons within the time scale of the demagnetization process. This finding conflicts with the results obtained from a Langevin treatment of the LLG equation [34], in which the magnetization relaxation rate is proportional to the temperature difference at *all times*. Figures 2(c) and 2(d) illustrate the time evolution of the out-of-equilibrium spin accumulation $\mu_s(t)$ and the longitudinal spin density $-S_z(t)$ for different decay times of T_s.

In the ultrafast regime, the electron-magnon spin-flop scattering is governed by the *effective* Gilbert damping parameter $\alpha^* \equiv \chi(\epsilon_b - \epsilon_0)$. Experimental investigations of the magnon relaxation rates on Co and Fe surfaces confirm that high-q magnons have significantly shorter lifetimes than low-q magnons [18]. It is reasonable to assume that the same effects are also present in the bulk. The initial relaxation time scale in the ultrafast regime is $\tau_i \sim (\alpha^* \hbar^{-1} k_B T_s)^{-1}$. This generalizes the result of Koopmans *et al.* [8] for the ultrafast relaxation of the longitudinal magnetization to arbitrary α^* based on the transverse spin diffusion [21]. The notion of magnons becomes questionable when the intrinsic linewidth approaches the magnon energy, which corresponds to $\alpha^* \sim 1$. Staying well below this limit and consistent with Refs. [18,21], we use $\alpha^* = 0.1$. For $T_C = 10^3$ K the initial relaxation time scale $\tau_i \sim 10^2 (T_C/T_s)$ fs, which is generally consistent with the demagnetization rates observed for ultrafast demagnetization in Fe [35].

We now show that the interfacial scattering follows a structure similar to that of the bulk scattering in a unified description based on the electron-magnon interaction. Figure 3 presents a schematic illustration of an F|N interface. In magnetic heterostructures and for stand-alone ferromagnets on a conducting substrate, the demagnetization dynamics of F are also affected by the spin accumulation in N $\mu_{\mathrm{N}}(x)$, which can impact how nonlocal laser irradiation (e.g., the heating of N alone) induces ultrafast demagnetization of F [15]. By adding terms of the form $\sim \sum_{qkk'} U_{qkk'} a_q \tilde{c}^\dagger_{k\uparrow} \tilde{c}_{k'\downarrow}$ to $\hat{H}_{sd}$, where $\tilde{c}^\dagger_{k\uparrow}$ ($\tilde{c}_{k'\downarrow}$) describes the creation (annihilation) of an electron with spin up (down) at the F|N interface, the *interfacial* spin transfer (per unit area) due to electron-magnon spin-flop scattering is [22]

$$I_i = \int_{\epsilon_0}^{\epsilon_b} d\epsilon_q \Gamma^i(\epsilon_q)\big(\epsilon_q - \mu_{\mathrm{N}}^0\big)\mathcal{D}(\epsilon_q)\big[n_{\mathrm{BE}}\big(\epsilon_q - \mu_{\mathrm{N}}^0\big) - n(\epsilon_q)\big], \tag{8}$$

where $\mu_{\mathrm{N}}^0 \equiv \mu_{\mathrm{N}}(0)$ is the spin accumulation at the interface and $\Gamma^i(\epsilon_q)$ parametrizes the interfacial scattering rate.

The scattering of coherent long-wavelength magnons at the F|N interface can be described in the language of spin pumping/spin Seebeck effects [22], parametrized by the spin-mixing conductance $g_{\uparrow\downarrow}$ (per unit area) [19]. Motivated by $\Gamma(\epsilon_q)$ in the bulk, we write for the interfacial scattering rate $\Gamma^i(\epsilon_q) = g^*_{\uparrow\downarrow}(\epsilon_q)/(\pi S)$, where $g^*_{\uparrow\downarrow}$ reduces to $g_{\uparrow\downarrow}$ for low-energy scattering, $\epsilon_q \to \epsilon_0$. The interface scattering [Eq. (8)] dominates the microwave spin relaxation in thin ferromagnetic layers of thickness $d_{\mathrm{F}} \lesssim 10$ nm [19,37]. This trend should continue for higher frequencies and is relevant for ultrafast spin dynamics in thin magnetic layers in heterostructures [1]. We expect the energy dependence of the effective spin-mixing conductance to be relatively weak compared to that of the bulk scattering $\Gamma(\epsilon_q)$, which can be severely constrained at low energies due to momentum conservation [21]. For a finite temperature bias δT across the interface and for magnons thermalized at the temperature $T < T_C$, the connection to the thermal spin Seebeck and Peltier effects is made by identifying $\mathcal{S} = \partial_T I_i$ and $\Pi = T\mathcal{S}/\hbar$ [20] as the Seebeck and Peltier coefficients, respectively.

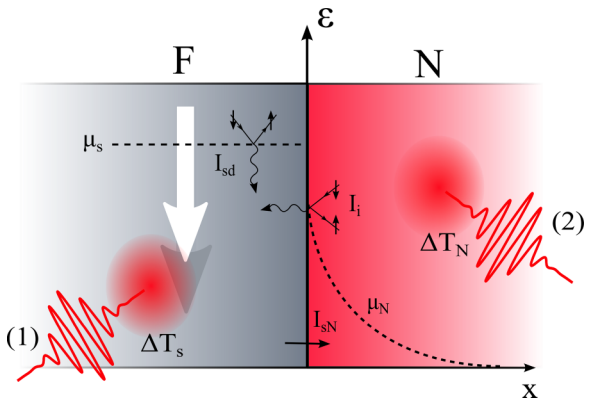

FIG. 3. (Color online) Sketch of a metallic ferromagnet (F) coupled to a normal metal (N). In the ultrafast regime, both the rapid heating of s electrons in F by ΔT_s [labeled (1)] and the heating of N by ΔT_{N} [labeled (2)] can demagnetize F. I_{sd} [Eq. (3)] induces the spin accumulation μ_s in F, whereas I_i [Eq. (8)] induces the spin accumulation μ_{N}^0 at the F|N interface. Subsequently, $\mu_{\mathrm{N}}(x)$ diffuses into N until it vanishes due to spin-flip dissipation to the lattice. The additional interfacial spin current $I_{s\mathrm{N}}$, due to the thermodynamic biases $\delta\mu = \mu_s - \mu_{\mathrm{N}}^0$ and $\delta T = T_s - T_{\mathrm{N}}$, can be described by conventional thermoelectric parameters for longitudinal spin-dependent transport [36].

In conclusion, we have extended the concepts of transverse spin diffusion in bulk ferromagnets and the interfacial spin-mixing physics to address the ultrafast spin dynamics observed in rapidly heated magnetic heterostructures. In the ultrafast regime, the relative importance of the bulk scattering, parametrized by α^*, and the interfacial scattering, parametrized by $g^*_{\uparrow\downarrow}$, can be extracted from measurements of demagnetization strength and spin currents in magnetic heterostructures. For metallic ferromagnets in the bulk, our analysis shows that treating the magnonic subsystems as quasiequilibrated and parametrized by an effective temperature is insufficient to describe the far-from-equilibrium spin dynamics induced by pulsed laser heating. The magnon distribution function remains nonthermalized on the relevant time scale of the demagnetization process, in which the relaxation of the out-of-equilibrium spin accumulation μ_s limits the dissipation of spin angular momentum from the combined electronic system.

The authors thank A. V. Kimel, G. E. W. Bauer, J. Barker, S. Bender, H. Skarsvåg, and E. Fjærbu for valuable discussions.

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