

Feedback Effect during Ultrafast Demagnetization Dynamics in Ferromagnets

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(Received 30 January 2013; published 16 October 2013)

Motivated by the recent controversy about the importance of spin-flip scattering for ultrafast demagnetization in ferromagnets, we study the spin-dependent electron dynamics based on a dynamical Elliott-Yafet mechanism. The key improvement to earlier approaches is the use of a modified Stoner model with a *dynamic* exchange splitting between majority and minority bands. In the framework of our microscopic model, we find a novel feedback effect between the time-dependent exchange splitting and the spin-flip scattering. This feedback effect allows us to reproduce important properties of the demagnetization dynamics quantitatively. Our results demonstrate that in general Elliott-Yafet spin-flip scattering needs to be taken into account to obtain a microscopic picture of demagnetization dynamics.

DOI: [10.1103/PhysRevLett.111.167204](https://doi.org/10.1103/PhysRevLett.111.167204)

PACS numbers: 75.78.Jp, 05.70.Ln, 75.40.Mg, 75.70.Tj

Sixteen years ago, the first experiments with femtosecond laser pulses on ferromagnetic metals uncovered an ultrafast demagnetization effect [1]. Except for a moderate dependence on the material and experimental conditions, numerous experiments on the transition-metal ferromagnets cobalt, iron, and nickel have unambiguously established typical demagnetization times between 100 and 300 fs after ultrafast optical excitation with linearly polarized light [2–4]. Since the ultrafast change of the magnetization takes place on a time scale much shorter than that of the typical relaxation between electron and lattice temperature, there has been an ongoing theoretical effort to find the fundamental *microscopic* mechanisms responsible for the observed behavior [5–14]. An Elliott-Yafet [15] (EY)-type demagnetization process, based on spin dynamics due to spin-conserving scattering mechanisms in the presence of the spin-orbit interaction, has long been considered the most promising mechanism, with several variants proposed and analyzed over the last few years [2,11–13,16–19]. In particular, electron-phonon scattering was widely considered to be essential for demagnetization, but it has recently been challenged by the authors of Ref. [17], who find that “phonon-mediated EY-like spin-flip scattering cannot be the mechanism of the observed ultrafast demagnetization.” Instead, superdiffusive transport [10] was proposed as a possible main mechanism and advocated [20] as being able to explain all ultrafast laser-induced demagnetization phenomena. We believe that these conclusions, in their generality, are incorrect as they are based on an overreaching interpretation of a calculation using the ground-state band structure and the corresponding matrix elements [17]. It was argued in Ref. [18] that the underestimation of the EY-type scattering arises because the magnetic properties of the material are unchanged in these approaches.

In the present Letter, we show that it is not justified to dismiss EY-type scattering as a potentially decisive contribution to demagnetization of ferromagnetic metals. To this end, we introduce a model that combines EY-type spin-flip scattering with a *dynamic* exchange splitting. The combination of the two mechanisms leads to a feedback effect for the demagnetization during the relaxation dynamics of majority and minority electrons. Our results show that, together with a dynamic exchange splitting, EY-type spin-flip scattering overcomes the limitations imposed by the zero temperature band structure [17,18,20].

Our model, which is schematically depicted in Fig. 1, takes a step into the direction of including dynamical changes of the magnetic properties. We compute electron and hole scattering dynamics within and between majority (spin-up) and minority (spin-down) electron bands that are

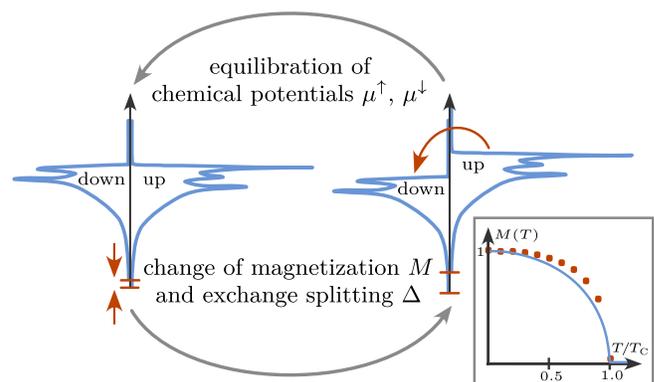


FIG. 1 (color online). Schematic plot of the occupation probability (electron distribution times density of states) in (left) and out of (right) quasiequilibrium. The feedback effect is indicated by gray arrows. Inset: equilibrium magnetization $M(T)$ computed by Eq. (3) for nickel, normalized to the value at $T = 0$ K. The experimental data (red dots) are taken from Ref. [37].

offset by a mean-field Stoner exchange splitting $\Delta(t)$. The Boltzmann equation for the spin- and energy-dependent distribution functions of up and down electrons $f_{\Delta}^{\uparrow,\downarrow}(E, t)$ as well as of phonons $g(E, t)$ reads [11,16]

$$\frac{\partial f_{\Delta}^{\eta}}{\partial t} = \sum_{\sigma, \nu, \lambda} |\langle \eta, \sigma | \nu, \lambda \rangle|^2 \Gamma_{e-e}^{\eta, \sigma, \nu, \lambda}[\Delta] + \sum_{\sigma} |\langle \eta | \sigma \rangle|^2 \Gamma_{e-ph}^{\eta, \sigma}[\Delta] + \sum_{\sigma} |\langle \eta | \sigma \rangle|^2 \Gamma_{\text{exc}}^{\eta, \sigma}[\Delta], \quad (1a)$$

$$\frac{\partial g}{\partial t} = \sum_{\eta, \sigma} |\langle \eta | \sigma \rangle|^2 \Gamma_{\text{ph-e}}^{\eta, \sigma}[\Delta], \quad (1b)$$

with $\eta, \sigma, \nu, \lambda \in \{\uparrow, \downarrow\}$ denoting the spin state. The notations f_{Δ}^{η} and $[\Delta]$ indicate the dependence of the distribution functions and, consequently, the Boltzmann scattering rates on the exchange splitting via the shifted energy dispersions for spin-up and spin-down electrons in a Stoner model. Another important difference with Ref. [16] is that we introduce a realistic density of states and calculate the Boltzmann scattering integrals $\Gamma_{x-y}^{\eta, \sigma}[\Delta]$ between the quasiparticles x and y as in Ref. [21]. As an extension for spin flips, both electron distributions f^{η} and f^{σ} , which are shifted in energy by Δ , are implemented. The excitation of the electrons is dominated by $\Gamma_{\text{exc}}^{\eta, \sigma}[\Delta]$ and is based on inverse bremsstrahlung [21,22]. In Eq. (1) the wave function overlaps $\langle \eta | \sigma \rangle$ and $\langle \eta, \sigma | \nu, \lambda \rangle$ account for the spin mixing between spin-up and spin-down electrons due to the spin-orbit interaction. In principle, our framework allows for energy-dependent spin-flip matrix elements [18,23]. Here, the overlaps are approximated as energy independent to show that a realistic spin mixing may result in a magnetization dynamics, which is in reasonable agreement with the experiment. To that end, the energy-independent overlaps can be pulled out of the collision integrals Γ as in Ref. [11].

As the central point in the present Letter and in agreement with recent experiments [24,25], we take here into account that the exchange splitting changes during the ultrafast demagnetization process. To this end, we use a mean-field approximation and calculate the exchange splitting via a Stoner-like expression [26]. We then assume an instantaneous response of the exchange splitting on the transient magnetization also under nonequilibrium conditions. Therefore, the dynamical exchange splitting is determined by

$$\Delta(t) = U_{\text{eff}} m(t), \quad (2)$$

where U_{eff} is an effective Coulomb interaction. The spin polarization $m(t) = M(t)/(\mu_B N)$ is defined by the Bohr magneton μ_B , the total electron density N , and the magnetization $M(t)$, which is explicitly determined by the transient electron distribution functions $f_{\Delta}^{\uparrow,\downarrow}(t)$. This constitutes a modification of the standard Stoner model [26] in that we use the paramagnetic nickel density of states (DOS) and the effective U_{eff} as a fit parameter, which

allows us to obtain an approximately realistic equilibrium magnetization curve $M(T)$ including Curie temperature, as shown in the inset of Fig. 1.

The exchange splitting $\Delta(t)$ is determined *dynamically* by the transient distributions of minority and majority carriers. For the unexcited system, we determine the magnetization M and the chemical potential μ solving a set of two coupled implicit integral equations for the total carrier density and magnetization condition of the Stoner model

$$N_{\Delta}^{\uparrow}(T, \mu) + N_{\Delta}^{\downarrow}(T, \mu) = N_{\Delta}^{\uparrow}(0, E_F) + N_{\Delta}^{\downarrow}(0, E_F), \quad (3a)$$

$$M = \mu_B [N_{\Delta}^{\uparrow}(T, \mu) - N_{\Delta}^{\downarrow}(T, \mu)], \quad (3b)$$

with the density of spin-up and spin-down electrons

$$N_{\Delta}^{\uparrow,\downarrow}(T, \mu) = \int_0^{\infty} dE D(E) f_{\Delta}^{\uparrow,\downarrow}(E, T, \mu), \quad (4)$$

the shifted Fermi distribution $f_{\Delta}^{\uparrow,\downarrow}(E, T, \mu)$, and density of states $D(E)$. The saturation magnetization M_0 is determined by applying Eq. (3b) for $T = 0$ K. The result of Eq. (3) is the chemical potential of the electrons and an equilibrium magnetization curve $M(T)$ (see inset of Fig. 1). The aforementioned approach can be applied for all itinerant ferromagnets [26].

We solve the Boltzmann equation (1) for nickel with the DOS taken from Ref. [27]. For the effective Coulomb energy parameter we obtain $U_{\text{eff}} = 5.04$ eV, which through Eq. (3) yields a Curie temperature of $T_C = 631$ K [28]. For $T = 0$ K, we obtain an exchange splitting of $\Delta = 0.259$ eV, which is a reasonable value [29]. The Debye temperature $T_D = 390$ K has been extracted from experimental data of the phononic specific heat for nickel [30]. The speed of sound $c_s = 6040$ m/s and the volume of the unit cell $\Omega = 1.094 \times 10^{-29}$ m³ are both taken from Ref. [31]. The spin mixing parameter $b^2 = 0.047$ has been used as a fitting parameter and lies within the same range as the results of density functional theory calculations [2,12]. The particle number and energy conservation in the numerical calculation have been checked.

We compare our results with experimental data of the magnetization dynamics of Roth *et al.* [32]. The data are taken from time-resolved magneto optical Kerr (MOKE) measurements on a 15 nm nickel film irradiated with an 800 nm pump pulse with $\tau = 50$ fs (FWHM). We model the excitation of the pump pulse in the framework of the single-band nickel density of states [21] with a constant reflectivity of $R = 0.98$ and assume a homogeneous heating of the film. Note that the important quantity for the comparison with the experiment is the ratio between different fluences. A more physically relevant quantity is the energy deposited within one unit cell δu after laser excitation. We obtain a δu value that is approximately proportional to the laser fluence, i.e., $\delta u \approx 48.3$ meV/ $\Omega \times (F/F_0)$ with the normalization fluence $F_0 = 2.5$ mJ/cm². In agreement with recent estimations [18], we find for the

lowest ($F = F_0$) and highest excitation ($F = 2F_0$) considered here $\delta u = 48.3 \text{ meV}/\Omega$ and $\delta u = 96.5 \text{ meV}/\Omega$, respectively.

During the excitation process, the energy deposited by the laser pulse leads to different distributions of majority and minority electrons due to their different densities of states (cf. Fig. 1). This results in an imbalance of chemical potentials $\mu^{\uparrow\downarrow}$ as well as temperatures $T^{\uparrow\downarrow}$ of the majority and minority electrons. Consequently, the electrons flip their spin to equilibrate the chemical potentials and electron temperatures. This equilibration process induces the magnetization dynamics [16]. The electron temperature may exceed the Curie temperature for short times but is reduced below T_C due to the cooling by phonons.

Figure 2 shows the transient magnetization obtained by Eq. (1), applying Eq. (2) dynamically (red solid line). To analyze the effect of a dynamic exchange splitting, we compare this result with that obtained by a constant exchange splitting $\Delta_0 = \Delta(t=0)$ (green dashed line). Figure 2 shows that the ferromagnetic material demagnetizes in *both* cases; however, the quenching with a dynamic exchange splitting is much more pronounced than that in the case of a constant Δ_0 . This is due to a feedback effect, sketched in Fig. 1: the equilibration of temperatures and chemical potentials is accompanied by electron spin flips, leading to a change of magnetization. According to Eq. (2), the exchange splitting is modified and the balance between the quasiequilibrium chemical potentials that was just being established is disturbed again, i.e., $\mu^{\uparrow} \neq \mu^{\downarrow}$. This imbalance acts again as a *driving force* for spin-flip scattering between majority and minority electrons and thus leads to an enhancement of demagnetization. The relative demagnetization, resulting from this feedback effect, compares well with the quenching observed experimentally. We thus conclude that a dynamical calculation of the

magnetic properties, here described by $\Delta(t)$, is essential to explain a realistic quenching with reasonable laser excitation strengths. If no electron-electron collisions are included (see the blue dotted line in Fig. 2), the electron systems only thermalize by electron-phonon collisions, which is a slow process taking several picoseconds. In this case, the quenching of the magnetization is reduced [17,18] and shows an atypical behavior. In Fig. 2, we also show the result of Essert *et al.* [18] for EY-type demagnetization exclusively due to electron-phonon scattering with *ab initio* matrix elements and a constant exchange splitting Δ_0 (brown double-dashed line). This simulation gives qualitatively similar results for the quenching as in our case, when we neglect electron-electron collisions and only electron-phonon spin-flips are applied (blue dotted line).

On the basis of the idea of the EY-type spin-flip mechanism, the microscopic three-temperature model (M3TM) has been developed assuming that the systems of spins, spinless electrons, and phonons are separately in quasiequilibrium [2]. Our method allows us in a straightforward way to verify the assumption of the M3TM that electrons are in equilibrium after an ultrashort laser irradiation. To that end, we performed a simulation in which the electrons thermalize instantaneously after laser excitation. The resulting magnetization dynamics, depicted in Fig. 2 (black dash-dotted line), show that the nonequilibrium *within* the electron subsystems plays a minor role. However, the nonequilibrium *between* both majority and minority electrons is essential for the demagnetization process [16].

In Fig. 3, we plot the dependence of the demagnetization dynamics on the excitation strength and compare it with experimental data [32]. With increasing fluence, the quenching of magnetization is increased due to the higher deposited energy. In accordance with experiment, we find

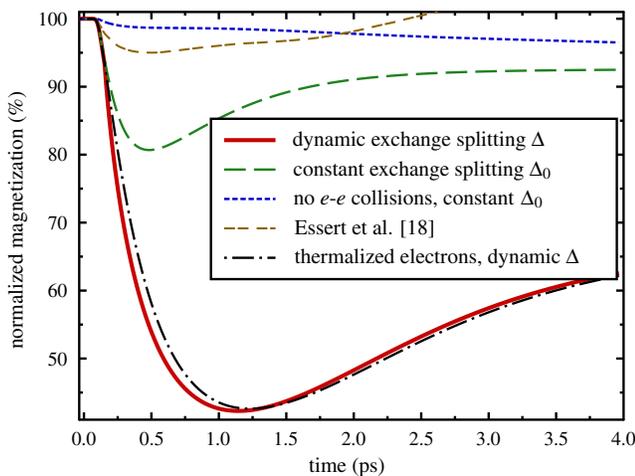


FIG. 2 (color online). Transient magnetization dynamics after ultrashort laser irradiation ($F = 1.4F_0$) for different cases: the role of thermalized electrons and secondary electrons as well as of a transient exchange splitting.

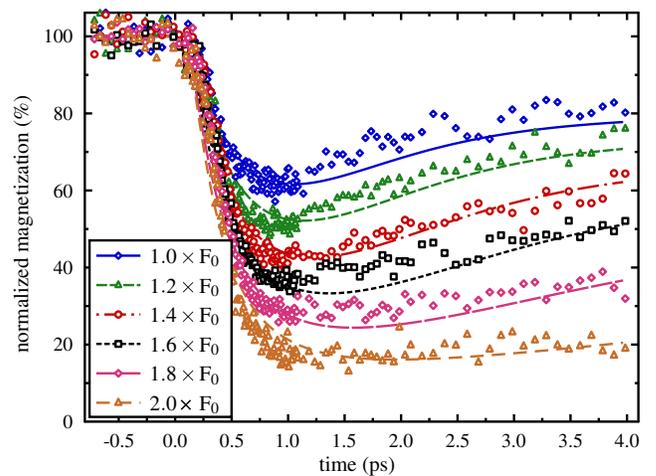


FIG. 3 (color online). Transient magnetization dynamics after ultrashort laser irradiation for different laser fluences. Experimental results, obtained by time-resolved MOKE measurements, are taken from Ref. [32].

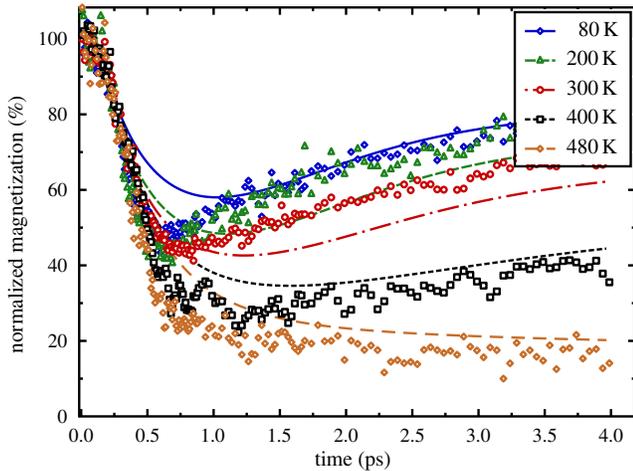


FIG. 4 (color online). Magnetization dynamics after ultrashort laser irradiation at a constant fluence ($F = 1.4F_0$) and various ambient temperatures $T = 80\text{--}480$ K.

that the maximum quenching is almost proportional to the fluence.

In Fig. 4, we investigate the influence of the ambient temperature on the demagnetization dynamics. Both the calculated and the experimental results [32] are shown. The calculations were performed for temperatures between 80 and 480 K with a laser fluence $F = 1.4F_0$.

Note that the experimental magnetization curves in Figs. 3 and 4 are measured under different conditions. In the latter case, a cryostat is used, which cools the sample during the measurement. This additional cooling to the phonon system increases the electron-phonon coupling strength, and the magnetization dynamics is slightly modified. Here, to avoid any further fit parameters in our model, we neglect the (unknown) cooling strength of the cryostat and apply the same fit parameters (R and b^2) for Figs. 3 and 4. Nevertheless, the experimental and theoretical curves show a good agreement. In Fig. 4, we observe larger drops for higher ambient temperatures. We explain this by the shape of the equilibrium magnetization curve $M(T)$ (inset of Fig. 1): For temperatures near Curie temperature, less energy is required for the same quenching compared to room temperature.

The recovery of the magnetization slows down for higher fluences (Fig. 3) as well as for higher ambient temperatures (Fig. 4). This behavior has been also obtained by the Landau-Lifshitz-Bloch model [33]. In both cases, the quasiequilibrium temperatures, reached after the laser pulse, approach Curie temperature. In this temperature range, slight temperature deviations lead to strong changes in the exchange splitting, and a large fraction of the electrons need to flip their spin in order to reach the equilibrium magnetization. Hence, the electronic specific heat blows up [30] and leads to a slowing down of the remagnetization dynamics, as also observed in the experiment.

In summary, we presented a dynamical model for Elliott-Yafet-type spin-flip scattering in ferromagnets that combines Boltzmann scattering dynamics (including a material specific density of states) with a Stoner model for itinerant electrons. By including the time dependence of the Stoner exchange splitting, the magnetization quenching after ultrafast excitation is strongly amplified by an important feedback effect: the spin-flip scattering, which is driven by the difference of the chemical potentials of majority and minority electrons, results in a change of the Stoner exchange splitting that, in turn, leads again to a nonequilibrium in the chemical potentials. We find that such a dynamic exchange splitting is essential to reproduce a quenching of the magnetization for excitation conditions that are comparable with experiment. This model therefore remedies a problem of EY-type spin-flip scattering in a fixed ground-state band structure.

In the same manner as superdiffusive transport, i.e., quantitatively but depending on a fit parameter, our model is in agreement with several key experimental facts. We expect the effects described by both models to be of varying importance, depending on the geometric and alloy structure under study. In general, ultrafast magnetization dynamics of multilayer systems [34,35] has been shown to be strongly dominated by superdiffusive spin transport. On the other hand, we believe that the demagnetization of thin magnetic films deposited on insulators [2,19,32,36] is mostly governed by the Elliott-Yafet mechanism. The question of which microscopic process is more important for a given structure can ultimately only be answered by more detailed experimental and theoretical studies.

Financial support of the Deutsche Forschungsgemeinschaft through the Emmy Noether project RE 1141/11-1 ‘‘Ultrafast Dynamics of Laser-excited Solids’’ is gratefully acknowledged. We thank Bert Koopmans, Tobias Roth, and Daniel Steil for discussions.

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