Driving force of ultrafast magnetization dynamics

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New Journal of Physics 13 (2011) 123010 (8pp)
Received 11 June 2011
Published 7 December 2011
Online at http://www.njp.org/
doi:10.1088/1367-2630/13/12/123010

Abstract. Irradiating a ferromagnetic material with an ultrashort laser pulse leads to demagnetization on the femtosecond timescale. We implement Elliott–Yafet-type spin-flip scattering, mediated by electron–electron and electron–phonon collisions, in the framework of a spin-resolved Boltzmann equation. Considering three mutually coupled reservoirs, (i) spin-up electrons, (ii) spin-down electrons and (iii) phonons, we trace non-equilibrium electron distributions during and after laser excitation. We identified the driving force for ultrafast magnetization dynamics as the equilibration of temperatures and chemical potentials between electronic subsystems. This principle can be used to easily predict the maximum quenching of magnetization upon ultrashort laser irradiation in any material, as we show for the case of 3d-ferromagnetic nickel.

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Figure 1. Scheme of the spin-resolved Boltzmann equation: it separates the phononic and the electronic system, the latter subdivided into a majority and a minority part. Due to the spin mixing, both electronic systems are coupled by electron–electron and electron–phonon spin-flips. In essence, the driving force for ultrafast demagnetization is determined by an equilibration of temperatures and chemical potentials between the two electronic systems.

It has been known for more than a decade that the interaction of a femtosecond laser pulse with a ferromagnetic metal causes ultrafast quenching of the magnetization [1]. Typical demagnetization times extracted from experiments on the 3d-ferromagnetic metals Co, Fe and Ni are between 100 and 300 fs, depending on the laser fluence and the specific sample [2–5]. On the theoretical side, intensive efforts are being made to identify the microscopic mechanisms responsible for the observed behavior [6–15]. For a general classification of laser-induced magnetization dynamics, we have to divide into the coherent interaction as pointed out in [6, 7] and into the longer enduring incoherent ultrafast demagnetization. Among the incoherent mechanisms, Elliott–Yafet (EY)-type spin-flip scattering is definitely one of the most implemented methods for the modeling of ultrafast demagnetization. The most popular EY-type spin-flip mechanism is electron–phonon (el–ph) scattering [2, 10, 13–16], while recently electron–electron (el–el) Coulomb (EEC) scattering has also been considered [11, 17]. Based on EY-type el–ph scattering, Koopmans et al [2] developed the so-called microscopic three-temperature model (M3TM), which has been shown to possess great predictive power, explaining not only the demagnetization in ferromagnetic transition metals but also the specific demagnetization characteristics of gadolinium. EEC scattering has been implemented in a dynamical model including momentum- and spin-dependent carrier scattering [11], in the following referred to as the EEC model. Since the band structure is taken as input for the calculations, the EEC model can be successfully applied to understand the demagnetization dynamics in materials with a peculiar band structure, as recently shown for the case of half-metallic Heusler alloys [17].

However, a fundamental question remains unsolved: what is the ultimate ‘driving force’ for ultrafast demagnetization?

To answer this question, we introduce a simplified, yet microscopic kinetic model describing non-equilibrium electron dynamics in laser-excited ferromagnetic metals. It combines the basic ingredients of the M3TM and the EEC model in the framework of a spin-resolved Boltzmann equation. The main idea behind this approach is schematically depicted...
in figure 1. Based on the EEC model, we describe a ferromagnetic metal as consisting of three subsystems: the spin-up (majority) electrons, the spin-down (minority) electrons, and the phonon system. In line with the typical spin-flip mechanisms in semiconductor physics [18–20], we consider el–el spin-flips and, additionally, in the spirit of the M3TM, we include el–ph-mediated spin-flips but without the need of introducing a separate spin subsystem. In order to enable the implementation of complete Boltzmann collision integrals and to ensure a clear interpretation of the results, it is necessary to make simplifying assumptions such as the free electron gas density of states (DOS) for the two spin subsystems. The strength of our approach is that it allows us to assign a respective temperature and chemical potential to up and down electrons, denoted by $T^\uparrow$ and $\mu^\uparrow$. We found that the equilibration of these quantities is the driving force for ultrafast demagnetization. This equilibration condition can be easily applied to real materials with complex DOS to obtain the maximum magnetization quenching following laser excitation, as we exemplarily show for nickel. As a further result, our approach is capable of clarifying the relative role played by el–el and el–ph scattering during ultrafast demagnetization.

In our model, we calculate the time- and energy-dependent dynamics of the considered subsystems on the basis of a kinetic approach applying complete Boltzmann collision integrals [21]. Due to the use of an electron distribution, holes are simultaneously covered by this approach as unoccupied states below the Fermi energy. The interactions between electrons (el–el), electrons and phonons (el–ph) as well as the absorption of laser energy (absorp) are described as independent collision integrals $\Gamma_{\text{col}}$, respectively. Their sum yields the transient evolutions of the electronic distribution $f(E)$ and the phononic distribution $g(E)$. The particular collision integrals are generally derived by Fermi’s golden rule

$$\Gamma_{\text{col}} = \sum_{\text{all states}} \frac{2\pi}{\hbar} |\langle \varphi_0 | \hat{H}_{\text{col}} | \varphi_1 \rangle|^2 \delta(E_0 - E_1),$$

where $\hat{H}_{\text{col}}$ indicates the Hamiltonian for the considered collision process, $\varphi_0$ the initial state with energy $E_0$ and $\varphi_1$ the final state with energy $E_1$. Such a model was developed in [21] to describe non-equilibrium electron and phonon dynamics during and after ultrafast laser excitation for a non-magnetic material. It is capable of accounting for non-equilibrium distribution functions where no real temperatures are defined.

Firstly, to apply this approach to ferromagnetic metals we include the Stoner model in the existing theory. This leads to a separated description of the two spin systems (up- and down-electrons), with a constant energy shift by the exchange energy $\Delta_{\text{ex}}$ between the corresponding distribution functions.

Secondly, we include spin-mixing in our approach. This allows a coupling between both electronic reservoirs: typically, the orthogonality of the up and down states guarantees that the matrix element $\langle \uparrow | \downarrow \rangle$ vanishes and for a spin diagonal interaction operator, there will be no possibility for an electron to change its spin during a collision process. But due to the spin–orbit coupling in solids, no pure up or down states are defined, but a mixture of both [13, 14]. In this case, an up-electron, for instance, is described by the state

$$|\tilde{\uparrow}\rangle = a |\uparrow\rangle + b |\downarrow\rangle,$$

whereby $|b| \ll |a|$. As the matrix element of the mixed state is larger than zero ($|\langle \tilde{\uparrow} | \tilde{\downarrow} \rangle| > 0$), the spin–orbit coupling allows for spin-flip and thus induces an interaction between the two spin systems in our model [11, 20]. The importance of spin–orbit coupling for the ultrafast demagnetization has been verified experimentally in [22]. The collisions including spin-mixing...
are thus described by

$$\Gamma_{\text{col}} = \sum_{\text{all states}} \frac{2\pi}{\hbar} |\langle \sigma_0 | \hat{H}_{\text{col}} | \sigma_1 \rangle|^2 \delta(E_0 - E_1)$$

$$= |\langle \sigma_0 | \sigma_1 \rangle|^2 \sum_{\text{all states}} \frac{2\pi}{\hbar} |\langle \sigma_0 | \hat{H}_{\text{col}} | \sigma_1 \rangle|^2 \delta(E_0 - E_1)$$

$$\equiv |\langle \sigma_0 | \sigma_1 \rangle|^2 \Gamma_{\text{col}},$$ (3)

whereby $\varphi_i$ is the spatial and $\sigma_i \in \{ \tilde{\uparrow}, \tilde{\downarrow} \}$ the (mixed) spin wave function of the electron.

With all the described ingredients, we can now write down the spin-resolved Boltzmann equation

$$\frac{\partial f_{\mu}}{\partial t} = \sum_{\sigma, \nu, \lambda} |\langle \mu, \sigma | \nu, \lambda \rangle|^2 \Gamma_{\text{el} - \text{el}} + \sum_{\sigma} |\langle \mu | \sigma \rangle|^2 \Gamma_{\text{el} - \text{ph}} + \sum_{\sigma} |\langle \mu | \sigma \rangle|^2 \Gamma_{\text{absorp}},$$ (4a)

$$\frac{\partial g_{\mu}}{\partial t} = \sum_{\mu, \sigma} |\langle \mu | \sigma \rangle|^2 \Gamma_{\text{ph} - \text{el}},$$ (4b)

with $\mu, \nu, \sigma, \lambda \in \{ \tilde{\uparrow}, \tilde{\downarrow} \}$ indicating the spin states. Equation (4) consists of three equations for $f_{\uparrow}, f_{\downarrow}$ and $g$, respectively. The collision terms $\Gamma_{\text{col}}$ can be found in [21, 23]. Each of the distinct equations is coupled to the others; thus the dynamics of all three subsystems mutually depend on each other. Since the description allows for spin-flips, the number of particles in each electronic reservoir is not conserved. Instead, the sum of the transient number of particles of spin-up ($n_{\uparrow}$) and spin-down ($n_{\downarrow}$) is conserved. The transient magnetization $M$ is calculated through

$$M(t) = \mu_B (n_{\uparrow}(t) - n_{\downarrow}(t)) = \mu_B \Delta n(t),$$ (5)

where $\mu_B$ is the Bohr magneton.

We note that, from a conceptual point of view, the spin-resolved Boltzmann equation is similar to a model proposed earlier by Bigot [24]. In particular, both models consider the conservation of the total number of spin-up and spin-down electrons, and introduce ferromagnetism by the Stoner model. Our description, however, goes a step further by implementing the kinetic details of the electronic and phononic subsystems, thus tracing the non-equilibrium dynamics microscopically. Moreover, by implementing EY-type scattering, we consider that the electrons need a finite time to establish an equilibrium satisfying the conditions of the Stoner model.

We solve the spin-resolved Boltzmann equation (4) for a prototype of a ferromagnetic material with the Fermi energy $E_F = 8$ eV, the effective mass $m^* = 1.45 m_e$ and the DOS of a free electron gas. We assume the exchange energy $\Delta_{\text{ex}} = 2$ eV. The spin-mixing parameter $b^2 \approx 0.03$ in equation (2) is chosen according to \textit{ab initio} calculations for Ni [2, 13, 14]. For the phononic system, we apply the Debye model with $T_D = 428$ K as the Debye temperature. These parameters represent typical values for real metals. Note that none of them is used as a fit parameter, because it is not in the scope of this paper to reproduce the experimental data, but to identify the driving force for ultrafast demagnetization. In this spirit, we chose the DOS of a free electron gas because essential relations are given analytically and the solution of the spin-resolved Boltzmann equation can thus be easily interpreted. Finally, to clearly investigate the processes after optical excitation, we assume a rectangular laser profile in time.
Figure 2. Simulation neglecting the contribution of phonons. (a) The internal energy $U^{\uparrow,\downarrow}$ and the number of particles $n^{\uparrow,\downarrow}$ of each electronic system. (b) Applying equation (8) it is possible to define approximated temperatures $T^{\uparrow,\downarrow}$ and chemical potentials $\mu^{\uparrow,\downarrow}$. The chemical potentials and the corresponding temperatures approach each other asymptotically.

In order to solve the spin-resolved Boltzmann equation (4) we compute a system of approximately 300 strongly coupled integro-differential equations. The solution provides the evolution of the electron distribution in figure 1 of [21], but extended by information on the two spin reservoirs. In particular, a net spin-flip is observed as a slight particle exchange between both electron reservoirs in the solution of equation (4) during and after irradiation.

Figures 2 and 3 were calculated for a 100 fs laser pulse with wavelength $\lambda = 630$ nm and a total absorbed fluence of $F = 0.13 \text{ mJ cm}^{-2}$. In figure 3, which will be discussed later in detail, curve (a) shows the transient magnetization (normalized to the initial magnetization) obtained with the spin-resolved Boltzmann equation (4) and applying equation (5). We obtain a demagnetization time of $\tau_M \approx 100$ fs after laser irradiation. Remagnetization occurs on the picosecond timescale.

Generally, when a system is excited out of its equilibrium state, it tends to a new equilibrium. An example is the relaxation between electrons and phonons after ultrafast laser excitation. Here, the initially heated electrons transfer energy to the lattice in a way that both end at a new, higher temperature. The nonequilibrium of temperatures between both systems drives this energy exchange. However, what is the driving force of particle exchange between spin-up and spin-down electrons, which, according to equation (5), is the basis for magnetization dynamics? For such a system, allowing for energy and particle exchange, the equilibrium condition refers to temperature and chemical potential and reads

$$T^{\uparrow} = T^{\downarrow} \quad \text{and} \quad \mu^{\uparrow} = \mu^{\downarrow} + \Delta \mu_{\text{ex}}.$$  \hspace{1cm} (6)

Exploiting the simplifications possible for a free electron gas, the chemical potentials can be explicitly expressed by the Sommerfeld expansion:

$$\mu^{\uparrow,\downarrow}(T^{\uparrow,\downarrow}) = E_F(n^{\uparrow,\downarrow}) \left[1 - \frac{\pi^2}{12} \left( \frac{k_B T^{\uparrow,\downarrow}}{E_F(n^{\uparrow,\downarrow})} \right)^2 + \cdots \right].$$  \hspace{1cm} (7)
After optical excitation, the temperatures \( T^\uparrow \) and \( T^\downarrow \) will be increased. Let us assume that \( T^\uparrow \) and \( T^\downarrow \) have already been equilibrated. Then equation (7) clarifies that in order to equalize also the chemical potentials \( \mu^\uparrow \) and \( \mu^\downarrow \), a change of \( E_F(n^\uparrow) \), mediated by a change of \( n^\uparrow \) and \( n^\downarrow \), is required. Thus, it is the equilibration of chemical potentials of the two electronic subsystems that leads to a particle exchange between both reservoirs and thus provides the ultimate driving force for ultrafast magnetization dynamics.

Our model allows us to verify the conclusion of the driving force behind the demagnetization process because we can selectively suppress different scattering channels in equation (4). In the following, we discard the phononic influence by setting \( \Gamma_{el-ph} = \Gamma_{ph-el} = 0 \).

The transient spin-resolved density \( n^\uparrow \downarrow \) and internal energy \( U^\uparrow \downarrow \) are depicted in figure 2(a) together with the difference \( \Delta n \), being proportional to the magnetization. Due to the higher electron density of majority electrons, the amount of energy absorbed by the spin-up electrons is larger than that for the spin-down electrons. For each moment in time and both spin directions, we may define a temperature \( T^\uparrow \downarrow \) and a chemical potential \( \mu^\uparrow \downarrow \) of the current distribution function. Both quantities are found through the implicit integral equations of the corresponding Fermi distribution \( f_F \):

\[
\begin{align*}
n^\uparrow \downarrow(t) &= \frac{1}{2} \int f_F^\uparrow \downarrow(E, T^\uparrow \downarrow, \mu^\uparrow \downarrow) \text{DOS}(E) \, dE, \tag{8a} \\
U^\uparrow \downarrow(t) &= \frac{1}{2} \int f_F^\uparrow \downarrow(E, T^\uparrow \downarrow, \mu^\uparrow \downarrow) \text{DOS}(E) \, E \, dE. \tag{8b}
\end{align*}
\]

The resulting temperatures \( T^\uparrow \downarrow \) and chemical potentials \( \mu^\uparrow \downarrow \) are depicted in figure 2(b). Due to laser excitation, both temperatures increase, while the chemical potentials drop. When the laser is turned off (at \( t = 100 \text{ fs} \)) the two spin systems differ in temperature and chemical potential. Since this difference is small for the assumed case of free-electron-like DOS (the second-order term in equation (7)), the quenching of magnetization is also considerably lower than that observed in real materials. Subsequently, the reservoirs equilibrate until the same temperature and chemical potential \( \mu \) are reached. According to equation (6), both constraints have to be satisfied to reach equilibrium between the electronic systems. Figure 2(b) verifies that the temperatures and chemical potentials equilibrate simultaneously. Comparing with figure 2(a), we also found that the transient magnetization, being proportional to \( \Delta n \), reaches its asymptotical value on the same timescale. In essence, the particle exchange affected by the difference in chemical potential provides the driving force for ultrafast magnetization.

In order to estimate the effect for more realistic systems, we choose nickel and solve the equilibrium condition (6) applying the implicit equations (8) with the real DOS for Ni [25]. Note that this can be done without explicitly solving the spin-resolved Boltzmann equation. In the case of starting at room temperature and ending up at an electron temperature of \( T_e \equiv T^\uparrow = T^\downarrow = 3000 \text{ K} \), a maximum quenching of 30\% is found. Such transient electron temperatures are typical of ultrafast laser excitation of metals below the melting threshold [21, 26]. Thus, applying the main result of our simplified model to realistic systems leads to quantitative agreement with experiments, demonstrating the validity of our findings.

To show the great potential of the spin-resolved Boltzmann equation, we now exemplarily compare the relative effects of \( el-el \) spin-flips and \( el-ph \) spin-flips on the demagnetization process. As already mentioned in our calculations, holes are also implicitly considered. We again restrict ourselves to the case of a free electron gas.
Figure 3 presents the calculated magnetization dynamics obtained with the spin-resolved Boltzmann equation (4) when different collision processes are regarded separately. The red solid line (d) corresponds to the red solid line in figure 2(a), determining the magnetization dynamics when the influence of the phonon bath is completely discarded. Including the cooling by phonons but disregarding el–ph spin-flips (green dotted line (c)), we see a slight recovery of magnetization. Thus, phonon cooling is essential for the remagnetization process. However, phonons also strengthen the magnetization dynamics: if we additionally allow for phonon-mediated spin-flips, we find the black dash-dotted curve (a). The demagnetization occurs faster than in the case where only electron collisions mediate spin-flips, while the remagnetization is strongly accelerated by phonon-mediated spin-flips. Hence, phonons can act as a catalyst for the ultrafast magnetization process. The blue dashed line (b) is calculated excluding el–el-collision-mediated spin-flips. In this case, the quenching of magnetization will be less. Comparing the simulations shown in figure 3, we conclude that for our system, electron-mediated spin-flips are important mainly during the demagnetization phase, while phonon-mediated spin-flips dominate the remagnetization phase and increase the effectiveness of the demagnetization.

In conclusion, we have described the ultrafast demagnetization with the spin-resolved Boltzmann equation. We identified the driving force for the demagnetization process in the equilibration of the temperatures and chemical potentials of the up- and down-electrons. As shown for the example of Ni this equilibrium condition provides a possibility of easily estimating the maximum quenching for any ferromagnetic material. Our approach is also capable of investigating the role of different scattering processes in ultrafast demagnetization dynamics.
Acknowledgment

We acknowledge fruitful discussions with H C Schneider. Financial support from the Deutsche Forschungsgemeinschaft through the GRK 792 ‘Nonlinear optics and ultrafast dynamics’ and from the Emmy Noether project RE 1141/11-1 ‘Ultrafast dynamics of laser-excited solids’ is gratefully acknowledged.

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