Ultrafast demagnetization of ferromagnetic transition metals:
The role of the Coulomb interaction

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The Elliott-Yafet (EY) mechanism is arguably the most promising candidate to explain the ultrafast demagnetization dynamics in ferromagnetic transition metals on timescales on the order of 100 fs. So far, only electron-phonon scattering has been analyzed as the scattering process needed to account for the demagnetization due to the EY mechanism. We show for the first time that the electron-electron scattering contribution to the EY mechanism has the potential to explain time-resolved magneto-optical Kerr effect measurements on thin magnetic Co and Ni films, without reference to a phononic “spin bath.”

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Current research in femtosecond magnetism is concerned with elucidating the fundamental mechanisms of light-induced spin dynamics as well as searching for potential applications in data processing. Despite of important experimental studies employing various time-dependent magneto-optical techniques, no consensus on a microscopic understanding of ultrafast magnetization dynamics in ferromagnets has emerged. Rather, demagnetization dynamics is typically described in the framework of the phenomenological three-temperature model. In this model, temperatures are assigned to the electron, lattice and spin “subsystems,” and the exchange of energy (and spin) is driven by the temperature differences between the respective subsystems. Although the three-temperature model provides an intuitive picture of demagnetization, its relation to the microscopic dynamics behind the demagnetization is still an active field of research.

The most popular candidate for the microscopic process behind ultrafast demagnetization is a mechanism of the Elliott-Yafet (EY) type. In the EY mechanism, the demagnetization arises because, in the presence of the spin-orbit interaction, spin is not a good quantum number, so that any momentum-dependent scattering mechanism changes the spin admixture when an electron is scattered from state $|k\rangle$ to $|k + q\rangle$. So far, the scattering process responsible for the EY mechanism has been assumed to be electron-phonon and electron-defect scattering in several theoretical and experimental studies. Unlike these papers, we apply, for the first time, an EY mechanism based exclusively on electron-electron Coulomb scattering to the ultrafast demagnetization in ferromagnetic metals. As a proof of principle, we demonstrate quantitative agreement for the demagnetization time and magnetization changes the spin admixture when an electron is scattered from state $|k\rangle$ to $|k + q\rangle$. The Elliott-Yafet (EY) mechanism is arguably the most promising candidate to explain the ultrafast demagnetization dynamics in ferromagnetic transition metals on timescales on the order of 100 fs. So far, only electron-phonon scattering has been analyzed as the scattering process needed to account for the demagnetization due to the EY mechanism. We show for the first time that the electron-electron scattering contribution to the EY mechanism has the potential to explain time-resolved magneto-optical Kerr effect measurements on thin magnetic Co and Ni films, without reference to a phononic “spin bath.”

The role of the Coulomb interaction

For the electron-electron Coulomb scattering we use the Boltzmann equation in the form

$$\frac{\partial n^\mu_k}{\partial t} = \frac{\partial n^\mu_k}{\partial t}\bigg|_{\text{opt}} + \frac{\partial n^\mu_k}{\partial t}\bigg|_{\text{e-e}}.$$  \hspace{1cm} (1)

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$$\frac{\partial n^\mu_k}{\partial t}\bigg|_{\text{e-e}} = \frac{2m}{\hbar} \sum_{lq} \sum_{\mu_1, \mu_2, \mu_3} |V_{\mu_1 \mu_2 \mu_3}^\mu_k(k, \ell, q, \omega)|^2 \left\{ (1 - n^\mu_k)n^\mu_{l+q} - (1 - n^\mu_{l+q})n^\mu_k - \{ (1 - n) \leftrightarrow n \} \right\} (\epsilon^\mu_k - \epsilon^\mu_{l+q} + \epsilon^\mu_{l+q} - \epsilon^\mu_k).$$  \hspace{1cm} (2)
where \( V \) is the dynamically screened Coulomb potential that depends on the initial and final states of the two scattering electrons \(|\mu, k\rangle \rightarrow |\mu_1, k + q\rangle\) and \(|\mu_2, l + q\rangle \rightarrow |\mu_3, l\rangle\), and \(\hbar \omega = \epsilon_k^\mu - \epsilon_{k'}^\mu\). The optical excitation contribution in Eq. (1) is calculated by adiabatic elimination of the optical polarization \([14]\)

\[
\frac{\partial n_{\mu k}^\mu}{\partial t} \bigg|_{\text{opt}} = \sum_{\nu \neq \mu} |d(k)_{\mu \nu}| \epsilon_0 |E|^2 (\epsilon_k^\mu - \epsilon_{k'}^\mu) g(\epsilon_k^\mu - \epsilon_{k'}^\mu).
\]  

(3)

where \( E \) is the classical electromagnetic field, and the function \( g(\hbar \omega) \) (peaked around the central frequency of the optical pulse) models the spread of photon energies which can induce electronic transitions via the dipole matrix element \( d_{\mu \nu}(k) \) between states \(|\nu, k\rangle\) to \(|\mu, k\rangle\). Note that the electron-scattering based version of the EY mechanism is contained in Eqs. (1)–(3), if the Coulomb matrix elements include the spin-orbit interaction in the presence of the static lattice, so that scattering transitions change the average spin of the scattered electrons. The lattice effectively acts as a sink for the electronic angular momentum, which is “lost” from the electronic system by the spin-non-conserving scattering processes described by Eq. (2). The important Coulomb and dipole matrix elements can, in principle, be determined from \textit{ab initio} treatments \([7]\), and parameter-free results can be achieved by a dynamical solution of Eqs. (1)–(3). However, due to the numerical complexity of the \(k\)-resolved Boltzmann scattering integral \([2]\), we use a simplified model that contains parameters. We stress that the parameters and simplifications introduced below can be eliminated by using input from \textit{ab initio} methods. We approximate the energy bands as spherically symmetric, \(\epsilon_k^\mu = \epsilon_{|\mu|}^\mu\), and the screened Coulomb interaction as

\[
V_{\mu \mu_1}^{\mu_2 \mu_3}(k, l, q, \omega) = f_\mu f_{\mu_2} f_{\mu_3} \frac{v(q)}{\varepsilon(q, \omega)}
\]  

(4)

where

\[
f_\mu = \begin{cases} 
1 & \text{if } \mu = \mu' \\
\alpha & \text{if } \mu \neq \mu'
\end{cases}
\]  

(5)

The parameter \(\alpha\) is roughly comparable to the \(\alpha\) parameter introduced by Yafet \([15]\) and calculated recently for metallic ferromagnets \([3]\). In Eq. (4), \(v(q)\) is the bare Coulomb potential, and \(\varepsilon^{-1}(q, \omega)\) the dynamical inverse dielectric function. For \(k, l, q\) dependent factors \(f\), this form can be shown to be valid if there are no short-range contributions to the Coulomb interaction \([16]\), and we use this as an approximate explicit expression for the Coulomb matrix element of metals. Important dynamical screening effects are included via the Lindhard dielectric function \(\varepsilon(q, \omega)\). In semiconductors, it has recently been demonstrated that an approach closely related \([13]\) to the one presented here leads to a parameter free agreement for the spin dynamics in theory and experiment \([17]\) because quite accurate wavefunctions can be obtained using \(k \cdot p\) theory. Finally, we assume that optical excitation connects only majority bands and minority bands with each other, respectively, and we approximate the strength of the optical dipole matrix elements by a momentum and band-independent constant \(d\). Although this is a drastic oversimplification, especially in view of the hybridization between \(s\) and \(p\) bands, it is in the same spirit as the approximations introduced for the band structure and the Coulomb interaction: The dependence on the electron vector momentum should either be included in all these quantities, or modeled in a way that introduces the least amount of parameters in the model. In this paper, the electronic excitation after the optical pulse is therefore determined by the band structure, the central photon energy and width of pump pulse, as well as the fluence. These quantities are used as input for the numerical calculations.

Finally, we include the equilibration of the electronic system with the lattice via a relaxation time approximation

\[
\frac{\partial n_{\mu k}^\mu}{\partial t} \bigg|_{\text{therm}} = \frac{n_{\mu k}^\mu - f_{\mu k}^\mu}{\tau_{\text{phon}}}
\]  

(6)

in Eq. (4). Here, \(f_{\mu k}^\mu\) denotes the Fermi-Dirac distribution of electrons in band \(\mu\) at lattice temperature. Note that in our model the demagnetization solely occurs due to Coulomb scattering, and that the electron-phonon interaction only leads to thermal equilibration, which actually restores the ground-state magnetization. The equilibration times \(\tau_{\text{phon}, \text{Ni}} = 25\, \text{ps}\) and \(\tau_{\text{phon}, \text{Co}} = 5\, \text{ps}\) are extracted from experiment by fitting the remagnetization dynamics. This assumption is based on the experimental observation that the timescale for energy equilibration due to electron-phonon interaction, which includes the effect of heat diffusion in a pragmatic way, is typically longer than the demagnetization time.
FIG. 1: Band structure $\epsilon^F(k)$ of Ni (a) and Co (b) for majority (solid lines) and minority (dashed lines) electrons in $\Gamma$-X direction [19]. The Fermi energy $E_F$ is set to zero. The arrows indicate a typical ultrafast demagnetization scenario: Electrons are excited by an ultrashort laser pulse (vertical light grey arrows), which does not change the total magnetization. They relax via intra- and inter-band scattering (grey arrows). The latter scattering process leads to depolarization of the electrons.

On the experimental side, a variety of techniques are available to excite and detect electron-spin dynamics [18]. Here, we apply an all-optical strategy to trace the spin dynamics on femtosecond timescales. By means of the time-resolved magneto-optical Kerr effect (TR-MOKE) in the longitudinal configuration we excite the ferromagnet by an ultrafast optical pump pulse, and monitor the material response by a delayed and modified optical replica (probe pulse). The femtosecond pulses are generated by a Ti:Sapphire multipass amplifier with 1kHz repetition rate. We use s-polarized 50fs, 800nm pump pulses at normal incidence, and s-polarized 50fs, 400nm probe pulses under 45°. The samples are thin polycrystalline ferromagnetic layers: a 15 nm cobalt film deposited on MgO by dc-sputtering, and a 15 nm Ni film deposited on Si by electron-beam evaporation. The Ni film is capped by a 3 nm Ti layer; another 3 nm Ti layer acts as an adhesion promoter between the Ni film and the substrate.

Qualitatively, the ultrafast demagnetization occurs in our model in the following way. The electronic distributions in the unexcited ferromagnet are assumed to be Fermi-Dirac distributions determined by the lattice temperature and the band structure. The majority and minority energy dispersions are spin split so that a non-zero magnetization exists in equilibrium. The ultrafast optical excitation process creates nonequilibrium electrons in bands accessible by the pump photon energy, and the electrons undergo intraband and interband Coulomb scattering processes. The driving force for the demagnetization are interband scattering processes between the optically excited electrons, which lead to the redistribution of electrons from majority to minority bands as long as the optically excited electrons are spin polarized. Remagnetization occurs due to equilibration at lattice temperature, because the groundstate magnetization is restored when the electrons settle down in the band minima.

For the numerical calculations we use as input for $\epsilon^F(k)$ a KKR-DFT-result [19] for the $\Gamma$-X direction, which is then used for the whole Brillouin zone as if the band structure were spherically symmetric. These dispersions are plotted in Fig. 1. For an experimental and theoretical study of Co and Ni band structures that also discusses the nomenclature of the bands, see Ref. [20]. The exciting laser pulse has a typical full-width at half-maximum of 50fs, photon energy of 1.55 eV and the fluence is numerically adjusted to be in qualitative agreement with an estimate of the absorption of magnetic order in Ni start more than 1 eV above the Fermi energy $E_F$ and mainly take place near the $X$-point.
FIG. 2: Dynamical distribution functions $n_k^\mu$ for four different bands. Electrons are excited from bands $\Delta^\uparrow_2$ and $\Delta^\uparrow_5$ (a) into band $\Delta^\downarrow_1$ (b). Ultrafast demagnetization occurs by scattering into bands $\Delta^\downarrow_2$ (c) and $\Delta^\downarrow_5$ (d). ($k = 8.9 \text{ nm}^{-1} \times$ the index in the figures)

Remarkably, the demagnetization is almost completely dominated by the two transitions mentioned above, with the nonequilibrium scattering dynamics taking place over more than 100 fs, cf. Figs. 2(c) and (d). Electrons that are not scattered out of band $\Delta^\uparrow_1$ at high energies above the Fermi level accumulate in states close to the Fermi-level because energy- and momentum conservation requirements make out-scattering processes inefficient. For completeness, we mention that band $\Delta^\downarrow_1$ does not play an important role in the demagnetization dynamics of Ni. An analysis of the electronic occupation in the different bands for Co along the same lines leads to the scenario as depicted in Fig. 1(b).

In Figure 3, we plot both the signal obtained from TR-MOKE measurements and the calculated signal for Ni and Co. The magnetization quenching for Ni is significantly stronger than for Co. In our model this is explained by the band structure in combination with the optical excitation process, which yields a more efficient carrier excitation with 800 nm photons: in Ni mainly majority electrons are optically excited (see Fig. 1) so that all interband scattering processes above $E_F$ lead to demagnetization. To obtain quantitative agreement between theory and experiment, we assume the same laser fluence for both materials, and use the Elliott-Yafet factor $\alpha$, introduced in Eq. (4), as a single fit parameter. We obtain $\alpha_{\text{Co}} = 0.15$ and $\alpha_{\text{Ni}} = 0.3$ together with the demagnetization times of $\tau_{\text{Co}} = 215 \text{ fs}$ and $\tau_{\text{Ni}} = 200 \text{ fs}$. The general trend $\alpha_{\text{Co}} < \alpha_{\text{Ni}}$ and the order of magnitude compare well with recent ab initio results for the $\alpha$ parameter [7]. The results in Ref. [2] provide only a qualitative check for our fit parameters, because the ab initio results depend on the band-structure region, over which the wave-function coefficients are averaged. Last, but not least, we stress that the band structure properties influence the microscopic dynamics sufficiently strongly to make it impossible to fit the Co measurements in Fig. 3 using the Ni band-structure and, vice versa.

In conclusion, we applied an Elliott-Yafet mechanism based on electron-electron Coulomb scattering for the explanation of the optically induced ultrafast demagnetization in the ferromagnetic transition metals Ni and Co. The electronic demagnetization in our model occurs through the Coulomb interaction, which is spin-diagonal for free electrons, in the presence of the spin-orbit interaction. Modeling the optical excitation process and the scattering dynamics by Boltzmann scattering integrals for the momentum-dependent dynamical distributions functions in the
various bands, we described a model that can make parameter-free predictions from input based \textit{ab initio} band structure results. In this paper, we made simplifying assumptions for the Coulomb and dipole matrix elements, which led to the introduction of two parameters, the EY-parameter $\alpha$ and the fluence, which are fixed by comparison with experiment. Even though the origin of ferromagnetism is an interaction effect of many electrons, the good agreement between theory and experiment presents evidence that the ultrafast demagnetization dynamics in ferromagnets can be understood in a single-particle picture including electron-electron scattering and the spin-orbit interaction. Unlike other EY based models, we do not take into account the electron-phonon interaction for the demagnetization dynamics. Our goal was to present a proof-of-principle that ultrafast demagnetization can occur due to an EY mechanism based on electron-electron scattering. The question whether the electron-electron or electron-phonon interaction predominates should be investigated further. Theoretically this can be achieved using the tools developed in this paper with the help of \textit{ab initio} band structure calculations. Experimental strategies to separate the two contributions should be developed concurrently.

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