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Unifying Ultrafast Magnetization Dynamics

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We present a microscopic model that successfully explains the ultrafast equilibration of magnetic order in ferromagnetic metals at a time scale τ_M of only a few hundred femtoseconds after pulsed laser excitation. It is found that τ_M can be directly related to the so-called Gilbert damping factor α that describes damping of GHz precessional motion of the magnetization vector. Independent of the spin-scattering mechanism, an appealingly simple equation relating the two key parameters via the Curie temperature T_C is derived, $\tau_M \approx c_0 \hbar / k_B T_C \alpha$, with $\hbar$ and k_B the Planck and Boltzmann constants, respectively, and the prefactor $c_0 \sim \frac{1}{4}$. We argue that phonon-mediated spin-flip scattering may contribute significantly to the sub-ps response.

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Among the most challenging and outstanding questions in today's condensed matter physics is the ultrafast quenching and growth of ferromagnetic order at a time scale τ_M of only a few hundred femtoseconds after pulsed laser excitation. An apparently unrelated issue in applied magnetism is the damping of GHz precessional motion of the magnetization vector, described by the Gilbert damping factor α . The latter is of utmost importance for high-frequency switching of magnetic devices and media, but microscopically being poorly understood. Within this Letter we unify the two phenomena, relating them even though their characteristic time scales differ by many orders of magnitude.

In 1996, Beaupaire *et al.* reported pioneering experiments on the magneto-optical (MO) behavior of nickel thin films after pulsed laser (60 fs) irradiation [1]. It was found that roughly half of the magnetic moment was lost well within the first picosecond. Soon, this surprising result got well confirmed by several groups [2,3]. Although it was shown that during the initial strongly nonequilibrium phase utmost care has to be taken with too naïve an interpretation of the MO response [4–6], by now a full consensus about a typical demagnetization time of $\tau_M \sim 100$ –300 fs for elementary ferromagnetic transition metals has been achieved [7]. Apart from all-optical experiments, it has been shown by time-resolved photoemission that the exchange splitting between majority and minority spin bands is affected at a similar time scale [8], while the loss of magnetization was directly detected by microwave radiation [9]. Very recently, the reverse effect of sub-ps generation rather than quenching of ferromagnetism was reported independently by two groups, driving FeRh thin films through an anti-ferromagnetic to ferromagnetic phase transition [10,11].

Surprisingly enough, despite the interest the topic received because of its elementary relevance in ferromagnetism, theoretical efforts to understand the novel phenomenon have been sparse. Phenomenologically, the process can be described within a so-called three tempera-

ture model. Absorbed laser light creates highly energetic “hot” electrons that rapidly thermalize to an equilibrated electron sea at an electron temperature T_e . Electron-phonon (e - p) interaction successively takes care of equilibration with the lattice (temperature T_l). Eventually, interactions between the spin system and the electrons, the lattice, or a combination thereof, cause a final heating of the spin system (T_s). Assigning heat capacities to the three interacting baths (c_e , c_l and c_s , respectively), and fitting coupling constants between them, allows for reproducing the transient reflectivity as well as magnetization dynamics profiles [1], but does not give any insight as to the microscopic origin of the processes involved. Microscopic modeling has been restricted to attempts by Zhang and Hübner [12] who speculated on the combined action of spin-orbit coupling and the laser field to cause a demagnetization within only tens of fs. It was demonstrated elsewhere [4,7], however, that the required conditions are not met in the experiments reported so far. Thus, fs scale magnetization processes in itinerant ferromagnets remain a major theoretical challenge, and even the characteristic time scale and corresponding elementary processes have not been identified yet. Finding the proper time scale is one of the main challenges addressed in this Letter.

For reasons to be clarified later, our analysis starts with considering precessional dynamics of the magnetization vector $\vec{M}$ in an effective field $\vec{H}$, as described by the Landau-Lifshitz-Gilbert (LLG) equation:

$$\frac{d\vec{M}}{dt} = \gamma \mu_0 (\vec{M} \times \vec{H}) + \frac{\alpha}{M} \left(\vec{M} \times \frac{d\vec{M}}{dt} \right), \quad (1)$$

with $\gamma = g \mu_B / \hbar$, the Bohr magneton μ_B , and the Landé factor $g \approx 2$. The first term describes the torque that leads to a precession at frequency $\omega_L = \gamma \mu_0 H$. The second term describes dissipation of energy and a convergence of the magnetic moment to align with $\vec{H}$. In the fully isotropic case, the typical dissipation time is given by

$\tau_{\text{LLG}} = 1/\omega_L \alpha$. Even for fields of a Tesla and a relatively large damping of $\alpha \sim 0.1$ this yields a τ_{LLG} of more than 100 ps, 3 orders of magnitude slower than the laser-induced (de)magnetization phenomena.

In contrast, let us next consider an individual electron spin that is not aligned with the sea of other electrons. Such an electron will experience an exchange field (H_{ex}) of the order of 10^3 Tesla, and thereby precesses at an extremely high frequency. Let us further conjecture, as originally suggested by Ruigrok [13], that the damping of the resulting single electron precession is governed by a phenomenological damping parameter α similar to its macroscopic counterpart in (1). Then, the precession of the single electron would damp out at an extremely rapid time scale of $\tau = 1/\omega_L \alpha = 1/\gamma \mu_0 H_{\text{ex}} \alpha \sim 100$ fs even for a moderate damping of 0.01. It is the main goal of this Letter to demonstrate the validity of the naïve conjecture from quantum-mechanical principles; i.e., we directly relate (i) the relaxation of (nonequilibrium) elementary spin fluctuations towards equilibrium among the spin, lattice, and electron system, with (ii) the Gilbert damping of the mesoscopic or macroscopic magnetization vector during its alignment with the effective field.

As to the experimental verification of the forthcoming predictions, we choose an all-optical approach in which both demagnetization and precession can be triggered by a single laser pulse and successively probed by recording the MO response after an adjustable delay time. Details of this pump-probe technique have been published elsewhere [14]. A typical result for a nickel thin film (Si/5 nm SiO₂/10 nm Ni) is represented in Fig. 1. Right after laser excitation a drop in the MO contrast is observable associated with a lowering of the magnetic moment. A characteristic time scale $\tau_M = 150$ fs is found. After an optimum in the signal at ~ 300 fs, the signal recovers due to a cooling

FIG. 1 (color online). Main: experimental development of the induced perpendicular component of $\vec{M}$ [$\Delta M_z(t)/M_z(0)$] after laser heating a nickel thin film at $t = 0$, and starting with $\vec{M}$ canted out of plane by applying an external field $\vec{B}_{\text{appl}}$. A sub-ps quenching of M is observed followed by a recovery via e - p relaxation, and finally, at $t > 100$ ps a damped precession of the magnetization. Insets: precession of an individual spin in the exchange field (left) and of $\vec{M}$ in the effective field (right).

of the electronic system when equilibrating with the lattice. From the exponential recovery an e - p energy relaxation time $\tau_E = 0.45$ ps is fitted. At a much longer time scale, an oscillatory signal represents a precession of $\vec{M}$, launched by the sudden perturbation of the magnetic anisotropy by the laser heating and driven by the in-plane component of the canted applied magnetic field. Fitting the damped oscillation and using the Kittel equation, valid for the thin film system, yields $H = 33$ kA/m, $\tau_{\text{LLG}} = 690$ ps, and $\alpha = 0.038$ [15]. Our microscopic theory will relate this value of α with τ_M .

We introduce a Hamiltonian inspired by Elliot-Yafet type of spin-flip scattering by electrons interacting with impurities or phonons [16]. Both scattering processes are facilitated by spin-orbit interactions that transfer angular momentum between the electrons and lattice. In order to derive analytical expressions for the various time scales, we make the following crude approximations. We consider a Fermi sea of spinless electrons with a constant density of states D_F , described by Bloch functions $|\vec{k}\rangle = N^{-1/2} \sum_j \exp(i\vec{k} \cdot \vec{r}_j) |u_j\rangle$ on a lattice of N sites, where $\vec{k}$ is a reciprocal lattice vector, and u_j is a local orbital of site j at position $\vec{r}_j$. A separate spin bath is defined, obeying Boltzmann statistics and described by a total number of $N_s \equiv ND_s$ equivalent two-level systems with an exchange splitting Δ_{ex} that depends in a self-consistent way on the average spin moment $\bar{S}$, i.e., using a mean-field (Weiss) description: $\Delta_{\text{ex}} = J\bar{S}$, where the exchange energy J is related to the Curie temperature via $k_B T_C = J/2$. Throughout this work spin operators are defined in units of $\hbar$, i.e., $S_z = \pm 1/2$.

We start with the simplest case of spin-flip due to impurity scattering, but will later show that the final result is more generally valid. Then, our Hamiltonian reads:

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_s + \mathcal{H}_{ee} + \mathcal{H}_{si}, \quad (2)$$

$$\mathcal{H}_{si} = \frac{\lambda_{si}}{N} \sum_k \sum_{k'} \sum_j c_k^\dagger c_{k'} (s_{j,+} + s_{j,-}), \quad (3)$$

where $\mathcal{H}_e$ and $\mathcal{H}_s$ represent the electron and spin system, respectively, and $\mathcal{H}_{ee}$ represents the (screened) Coulomb term that is assumed efficient enough to cause an almost instantaneous thermalization of optically excited carriers towards a Fermi-Dirac distribution $\rho(E, T_e)$ at $T = T_e$. In practice, this thermalization takes approximately $\tau_T \sim 50$ – 100 fs for the ferromagnetic transition metals such as nickel. Within the spin-flip term $\mathcal{H}_{si}$, $c_k^\dagger$ and c_k describe creation and annihilation, respectively, of electrons in state $\vec{k}$, whereas $s_{j,+}$ ($s_{j,-}$) denotes a spin-up (-down) flip of spin j . Note that the prefactor λ_{si} scales with impurity concentration.

In order to derive for this $\mathcal{H}$ the dynamical response described by $T_e(t)$ and $T_s(t)$, we start in a full equilibrium situation at ambient temperature T_0 , i.e., $T_e = T_0$ and $T_s = T_0$ (and $\bar{S} = S_0$). Sudden laser excitation leads to

an enhanced electron temperature, $T_e(0) = T_0 + \Delta T_e(0)$, while the spin system remains initially unaffected. The nonequilibrium situation causes an imbalance of spin-up and spin-down scattering. Using (3) and Fermi's golden rule, we can thus calculate the rate at which the average spin changes right after excitation, $\dot{\vec{S}}$, from which the spin temperature rate $\dot{T}_s$ can be derived. The required integrals over all electronic states can be performed analytically, and an estimate of τ_M^{-1} is obtained by normalizing $\dot{T}_s$ by the difference in equilibrium spin value before and after laser heating, $\tau_M^{-1} = \dot{T}_s(0)/\Delta T_s(\infty)$, as illustrated in Fig. 2(c). Assuming $c_s \ll c_e$ throughout our Letter, a closed expression for τ_M in the small pump-fluence limit is derived

$$\tau_M = \frac{c_0 F(T/T_C)}{\pi D_F^2 \lambda_{si}^2} \frac{\hbar}{k_B T_C}, \quad (4)$$

where $c_0 = 1/4$, and where we introduced a function $F(T/T_C)$ that solely depends on T/T_C , with $F(0) = 1$ and deviating from unity by less than 20% up to halfway T_C as plotted in Fig. 2(b).

Next, for the same model Hamiltonian, we derive a value for the Gilbert damping. In practice this means that rather than describing thermal fluctuations of the spin system, we concentrate on the dynamics of the net spin moment. Therefore, in calculating the dynamics of the macro spin $\vec{S}(t)$, we only consider the high-spin state with total spin $S = N_s/2$ (as would be the case for $\Delta_{ex} \gg k_B T$). It is easily shown that in an applied field, the Zeeman splitting between the sublevels causes a precession of this macro spin identical to the prediction of (1). In order to derive the dissipative part of the dynamics, we calculate the impurity-term induced matrix element A for scattering up/down from state $|N_s, m\rangle$ to $|N_s, m \pm 1\rangle$, where m counts the

number of spin-up states. For large N_s , some algebra yields:

$$A_{N_s, m} = \sqrt{m(N_s - m)} \lambda_{si} / N = \sin \theta \lambda_{si} / 2N, \quad (5)$$

where we introduced θ , the angle between $\vec{S}$ and $\vec{H}$. Again using the golden rule and assuming $\gamma \mu_0 \hbar H \ll k_B T_e$, we derived that the alignment of $\vec{S}$ with $\vec{H}$ is described by $\dot{S}_z(t)/S = \pi D_F^2 \lambda_{si}^2 \gamma \mu_0 H \sin^2 \theta(t)$. It can easily be verified that the solution of the macroscopic LLG equation yields $\dot{M}_z(t)/M = \alpha \gamma \mu_0 H \sin^2 \theta(t)$, and, thereby, that the two solutions are identical provided that

$$\alpha = \pi D_F^2 \lambda_{si}^2. \quad (6)$$

We thus derived microscopically the LLG equation and an expression for α for the case of the impurity-induced spin scattering. In passing, we note that our approach is quite similar to the spin-flip scattering treated by Kamberský [17], though does not include ordinary scattering between spin-dependent band levels [17,18].

A comparison of (4) with (6) allows for the reformulation of τ_M in terms of α we searched for:

$$\tau_M = c_0 F(T/T_C) \frac{\hbar}{k_B T_C} \frac{1}{\alpha} \approx c_0 \frac{\hbar}{k_B T_C} \frac{1}{\alpha}, \quad (7)$$

where the last approximation is valid for T well enough below T_C . The final relation between τ_M and α confirms our naïve conjecture at a quantum-mechanical level. Although (7) is expected to yield only a very rough estimate of the magnetization dynamics, since all details of the spin-resolved electronic band structure and spin-scattering processes involved in LLG were neglected, it sets the relevant time scale with surprising accuracy. E.g., the data of Fig. 1 yield $\alpha = 0.038$. Using $T_C = 630$ K, and $F(T) \approx 1$ at room temperature readily predicts $\tau_M = 100$ fs, within a factor of 2 of the measured value.

However, we introduced the impurity-induced spin-flip mechanism mainly for illustrative purpose, and what remains is to show that the relation between τ_M and α is more general. In a recent paper [19] we numerically explored the case of *phonon*-mediated spin-flip scattering, though merely for the demagnetization process. In the present Letter we aim for analytical expressions, including the case of Gilbert damping. The phonon system is described by identical harmonic oscillators (Einstein model) with phonon energy E_p approximately equal to the Debye energy, a density of oscillators D_p per site, an *e-p* matrix element λ_{ep} , and a probability $0 \leq a \leq 1$ that the *e-p* scattering is accompanied by a spin flip. We again derived (7), only differing (slightly) by the value of c_0 . In this case, τ_M depends on details of the scenario, as illustrated in Fig. 2. For $\tau_M \gg \tau_E$ we find $c_0 = 1/4$ as before, whereas for $\tau_M \ll \tau_E$ an even faster response results, $c_0 = 1/8$. Thereby, also the phonon-assisted model successfully predicts sub-ps values of τ_M for realistic α .

Although the main quest of this Letter has been fulfilled at this point, it is of interest exploiting the fact that within

FIG. 2 (color online). Temperature dependence of the magnetization within the Weiss model (a), and the function $F(T)$ that scales with τ_M as expressed by (4). (c)–(e) Evolution of electron (blue dashed line), lattice (green dotted line), and spin (red line) temperature for the impurity model (c), and the phonon-mediated model in two limiting cases $\tau_M \ll \tau_E$ (d) and $\tau_M \gg \tau_E$ (e), showing the construction of τ_M in the respective cases.

the phonon-mediated scheme analytical expressions for both τ_M and τ_E can be derived. More particularly, in the limit of $T \rightarrow 0$ (i.e., $F(T) \approx 1$), a ratio

$$\frac{\tau_M}{\tau_E} = \frac{3c_0 D_s E_p^2}{\pi^2 a D_F k_B^3 T^2 T_C} \quad (8)$$

is found. Thus, it can be verified whether a realistic value of a can reproduce the ratio τ_M/τ_E obtained from experiment (e.g., Fig. 1). To get an idea of the order of magnitude, we plug in a set of reasonable parameters for Ni: $D_F = 5 \text{ eV}^{-1}$, $E_p = k_B T = k_B T_C/2 = 25 \text{ meV}$, $D_s = 0.6$, $c_0 = 1/8$. This set reproduces the observed $\tau_E \sim 0.45 \text{ ps}$, whereas it requires $a \sim 0.1$ to end up with $\tau_M \sim \tau_E$.

First, we conclude that a demagnetization that is faster than the e - p energy equilibration can be obtained for a value of a that is realistic in the sense that it does not require a spin-flip probability exceeding 1. Second, we note that values of a have been tabulated before for some nonmagnetic metals [20], but not for ferromagnetic transition metals. It was found that a scales roughly with Z^4 . From a comparison with copper, for nickel this dependency would yield a value of a few thousandths at most. However, it was also found that band structure effects—in particular band degeneracies near the Fermi level—can increase a by 2 orders of magnitude [21]. Realizing that such band degeneracies are common for the transition metal ferromagnets, phonon-mediated spin-flip scattering in the spirit of Elliot and Yafet may indeed provide a significant contribution to the sub-ps magnetic response.

In conclusion, we have demonstrated that two formerly unrelated fast dynamic processes in ferromagnets can be related, independent of the details of the spin-flip terms in the Hamiltonian. Values for the demagnetization time in the sub-ps regime are readily derived for a typical Gilbert damping of 0.005–0.05. Clearly, we do not claim any quantitative predictability of the model, knowing that details of the band structure were completely neglected [18], and realizing that Gilbert damping is often dominated by nonintrinsic effects such as a nonlinear spin-wave generation in micromagnetic structures [22], not included in our simplified model at all. Nevertheless, we stress that for the first time the proper time scale could be derived from quantum-mechanical principles. Having established this crucial insight, a wide range of future investigations can be envisioned. Apart from theoretical efforts aiming at implementing a more realistic electronic band structure and spin excitation spectrum, it will be of importance to study experimentally the temperature dependence predicted by the simple model, or trying to confirm the relation between α and τ_M more directly by exploiting especially engineered samples.

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