

Relating Gilbert damping and ultrafast laser-induced demagnetization

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Based on the breathing Fermi-surface model of Gilbert damping and on the Elliott-Yafet relation for the spin-relaxation time, a relation is established between the conductivitylike contribution to the Gilbert damping α at low temperatures and the demagnetization time τ_M for ultrafast laser-induced demagnetization at low laser fluences. Thereby it is assumed that, respectively, the same types of spin-dependent electron-scattering processes are relevant for α and τ_M . The relation contains information on the properties of single-electron states which are calculated by the *ab initio* electron theory. The predicted value for α/τ_M is in good agreement with the experimental value.

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I. INTRODUCTION: GILBERT DAMPING AND LASER-INDUCED DEMAGNETIZATION

Recently, there has been an extensive research activity on short-time magnetization dynamics for two reasons. First, there is an enormous importance for magnetic devices, and, second, the microscopic mechanisms which determine the dissipative magnetization dynamics are not well understood. The dynamics (e.g., magnetization switching) can be driven by external magnetic fields or by spin-polarized electrical currents on a time scale of nanoseconds to several picoseconds. Since the pioneering paper of Beaupaire *et al.*¹ it is known that the magnetization can be modulated even on a picosecond or subpicosecond time scale when exposing a thin film of Ni, Fe, or Co, e.g., to an optical femtosecond laser pulse. In the following we denote the dynamics on these two scales as fast and ultrafast magnetization dynamics, respectively.

For the theoretical modeling of the fast dynamics of the magnetization \mathbf{M} the Gilbert equation² is commonly used

$$\frac{d\mathbf{M}}{dt} = -\gamma(\mathbf{M} \times \mathbf{H}_{\text{eff}}) + \frac{1}{M} \left(\mathbf{M} \times \alpha \frac{d\mathbf{M}}{dt} \right). \quad (1)$$

Here the first term (γ is the gyromagnetic ratio) describes the precession of \mathbf{M} around the effective field \mathbf{H}_{eff} , and the second term with the damping scalar α represents the damping. It has been shown (for a review, see Ref. 3) that for a general situation the Gilbert scalar α has to be replaced by a damping matrix $\underline{\alpha}$, which depends itself on the magnetization configuration of the whole system but for the present purpose it suffices to consider Eq. (1). For the modeling of the ultrafast dynamics a phenomenological three-temperature model is used¹ which describes the interaction of the electron, spin, and lattice subsystems, or its recently developed microscopic version (see, e.g., Ref. 4). The quantity of interest is the demagnetization time τ_M (which for Ni can be below 100 fs for low laser fluences⁴) describing the rate of magnetization loss of the film after laser excitation.

Both damping of the fast magnetization dynamics, characterized by α , and ultrafast demagnetization after laser excitation, characterized by τ_M , require a transfer of angular momentum from the electronic system to the lattice via electronic spin-flip scattering. Assuming that the dominant mi-

croscopic channels for the angular momentum transfer are the same for both situations, it is desirable to find a relation between α and τ_M .

II. SUMMARY OF A FORMER UNIFIED THEORY

A first unified theory of fast and ultrafast magnetization dynamics has been presented by Koopmans *et al.*⁵ who compared the fast precessional dynamics of a homogeneously magnetized system in the homogeneous effective field $H_{\text{eff}} = H$ (composed of the external field, the anisotropy field, and the demagnetization field) with the ultrafast demagnetization after laser excitation. According to Eq. (1), the precession damps out on the time scale

$$\tau_p = \frac{\hbar}{g\mu_B H \alpha} \quad (2)$$

with the Landé factor $g \approx 2$ and the Bohr magneton μ_B . In Ref. 5 the physics behind the drastically different time scales τ_M and τ_p could be explained by a simple hand-waving argument. To do this, the authors of Ref. 5 calculated the transversal spin-relaxation time $\tau_{M,t}$ for laser excited electrons, thereby describing the damped precessional motion of the single electrons again by the Gilbert Eq. (1) with the same damping constant α as for the fast precession but with the effective field $H_{\text{eff}} = H$ replaced by the exchange field H_{ex} , which an individual electron spin feels that is not aligned with the sea of other electrons. Assuming that the longitudinal relaxation time τ_M is equal to the transverse relaxation time $\tau_{M,t}$ even for the extremely fast precession in the Stoner exchange field (preconditions for the validity of this assumption are discussed in Ref. 6) yields^{7,8}

$$\tau_M = \frac{\hbar}{g\mu_B H_{\text{ex}} \alpha}. \quad (3)$$

From Eqs. (2) and (3) it becomes obvious that the reason for the drastically different time scales of fast and ultrafast dynamics is given by the different effective fields. The individual spins of the ultrafast dynamics feel the exchange field, and their precession is extremely fast. In contrast, for the fast dynamics the Stoner exchange field does not appear explicitly (it just functions to guarantee the constant modulus of

the magnetization during the precession), and the precession in the effective field is much slower.

In Ref. 5 the relation in Eq. (3) between τ_M and α has been rederived from quantum-mechanical principles. In both situations an equation of motion is determined which describes the relaxation of the magnetic moment $\mathbf{m}(t)$ of the sample from an initial nonequilibrium situation toward equilibrium. On the microscopic level this relaxation results from an imbalance of spin-up and spin-down electron-lattice scattering events. Thereby it is again assumed that the same types of spin-flip electron scattering (described by a model matrix element) are relevant for τ_M and α . For the femtosecond demagnetization the nonequilibrium situation arises because after the action of the laser pulse (which primarily raises the electronic temperature) the electron, spin, and lattice temperatures of the above-mentioned three-temperature model are different. The increased temperature of the heat bath for the individual spins (provided by the electronic subsystem) causes a repopulation of the spin-up and spin-down levels (defined with respect to the orientation of the exchange field H_{ex}) via spin-flip scattering events which change the energy by $\pm g\mu_B H_{\text{ex}}$. For the calculation of α a homogeneously magnetized sample is considered, where the individual spins are coupled by H_{ex} to a macrospin of fixed quantum number S but variable magnetic quantum number m_S (now defined with respect to the orientation of the effective field \mathbf{H}). The initial nonequilibrium situation where the macrospin is not parallel to \mathbf{H} is again relaxed by spin scattering processes whereby—however—the restriction has to be fulfilled that S is conserved while m_S can be changed. Therefore the change in energy due to an individual spin flip is $\pm g\mu_B H$ rather than $\pm g\mu_B H_{\text{ex}}$. From the equations of motion derived for the respective situation the quantities τ_M and α can be determined, yielding Eq. (3).

It should be recalled that in the quantum-mechanical treatment all the electronic properties of the system are described by just one effective parameter (the spin-flip scattering matrix element). Therefore it cannot be expected that Eq. (3) gives a highly accurate description for all systems, albeit it yielded a correct prediction on the order of magnitude of $\tau_M\alpha$ from a quantum-mechanical treatment. In fact, Eq. (3) could not be confirmed quantitatively when manipulating τ_M and α by transition-metal or rare-earth doping (see, e.g., Ref. 9), either because of the oversimplified treatment of the electronic and scattering properties, or because different relaxation channels are relevant for τ_M and α (in contrast to the basic assumption of the calculation).

III. DESCRIPTION OF THE PRESENT UNIFIED THEORY

In the present paper we derive a relation between α and τ_M by a completely different approach than in Ref. 5. The advantage of the present theory is that it takes into account in a much more detailed manner the specific electronic properties of a material. The disadvantage is that the relation between α and τ_M is much more complicated and does not contain just one parameter [H_{ex} in Eq. (3)] but the properties of all individual electronic states which have to be calculated by the *ab initio* electron theory for a comparison with the

experiment. It will be shown that the value of α/τ_M predicted by the theory agrees well with the corresponding experimental value.

It is well known (see, e.g., Refs. 10–15) that there are often two contributions to α , one which is proportional to the conductivity of the material and which dominates at low temperatures, and one which is proportional to the resistivity. We want to derive a relation between the low-temperature damping parameter and the demagnetization time τ_M after laser excitation at low temperatures and such low laser fluences that the electron, spin, and lattice temperatures rise only slightly.

It has been shown (see, e.g., Ref. 13) that the Gilbert damping in metallic ferromagnets results predominantly from the fact that the magnetization dynamics itself generates pairs of excited electrons and holes which then experience spin-dependent scattering at the lattice, thereby transferring angular momentum from the electronic system to the lattice. We can distinguish between pairs for which the excited electrons and holes appear in the, respectively, same band, and pairs which are generated by exciting the electrons to other bands than those for which the holes appear. The relaxation of these two types of electron-hole pairs leads to the above discussed two contributions to α (see Refs. 14 and 15).

The intraband pairs are generated because the spin-orbit energy changes when the orientation $\mathbf{e}(t)$ of the, for example, homogeneous magnetization $\mathbf{M}(t) = M\mathbf{e}(t)$ changes with time t , i.e., the single-electron energies $\varepsilon_{j\mathbf{k}}$ (j and \mathbf{k} denote the band index and the electronic wave vector) change with time. Some states which are just below the Fermi surface for one orientation \mathbf{e} get pushed above the Fermi surface for an orientation \mathbf{e} at another time whereas other states which were originally above are pushed below. This means that excited electrons and holes are generated in the same band when we consider the respective preceding orientation as reference. This means, that, e.g., for a precessional dynamics of $\mathbf{M}(t)$ the Fermi surface “breathes.” The relaxation of the intraband electrons and holes leads to the conductivitylike “breathing Fermi surface” contribution^{13–16} to α .

The interband pairs are generated because the system of electrons feels a time-dependent perturbation due to the changing spin-orbit interaction (see, e.g., Ref. 14), and this leads to electronic transitions between states $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}}$. These excitations are pictured as¹⁴ “bubbling” of individual electrons at the Fermi surface. The relaxation of these interband electrons and holes leads¹⁴ to the resistivitylike “bubbling Fermi surface” contribution to α .

It has been shown in Ref. 14 that the breathing and the bubbling Fermi-surface contributions are incorporated in Kamberský’s¹² torque correlation model. For the conductivitylike contribution itself another type of theory yielded the breathing Fermi-surface model.^{3,16} Because we concentrate on the low-temperature damping, we will consider the breathing Fermi-surface model.

In the theory of Ref. 5 the fast dynamics of the system is described by a statistical approach on the macrospin level, and the macrospin relaxation is driven by the fact that transitions $|S, m_S\rangle \rightarrow |S, m_S + 1\rangle$ lower the energy by $g\mu_B H$. Such transitions are realized by the spin-dependent electron-lattice

scattering events which are characterized by the effective scattering matrix element (see above) but the detailed dynamics on the level of single-electron states does not enter explicitly the statistical approach for the macrospin. In contrast, in the breathing Fermi-surface model the system is described by a statistical approach on the level of single-electron states. As described above, intraband electron-hole pairs are generated, for instance, by a precessional dynamics of $\mathbf{M}(t)$ due to a breathing Fermi surface. The electron-hole pairs generated by the precession survive for some lifetime τ before they relax by electron-lattice scattering, thereby transferring angular momentum to the lattice. Because of the finite lifetime the real occupation numbers $n_{j\mathbf{k}}(t)$ deviate from the equilibrium Fermi-Dirac occupation numbers $f_{j\mathbf{k}}[\varepsilon_{j\mathbf{k}}(t)]$, and the differences between these two occupation numbers represent the driving forces for a statistical treatment of the relaxation on the level of single-electron states. Altogether, the breathing Fermi-surface model yields¹⁷

$$\alpha = \frac{\gamma\tau}{M} F_{el}, \quad (4)$$

where the quantity

$$F_{el} = - \sum_{j\mathbf{k}} \frac{\partial f_{j\mathbf{k}}}{\partial \varepsilon_{j\mathbf{k}}} \left(\frac{\partial \varepsilon_{j\mathbf{k}}}{\partial \mathbf{e}} \right)^2 \quad (5)$$

contains the derivatives of the single-electron energies with respect to the orientation \mathbf{e} of the magnetization $\mathbf{M} = M\mathbf{e}$. The quantities $\partial \varepsilon_{j\mathbf{k}} / \partial \mathbf{e}$ can be calculated in the *ab initio* electron theory from the single-electron energies $\varepsilon_{j\mathbf{k}}$ calculated for two close orientations \mathbf{e} of the homogeneous magnetization which are stabilized by the action of constraining fields.³

We now describe the calculation of the demagnetization time τ_M after laser excitations at low temperatures and low laser fluences. For low fluences the laser excitation drives the system only slightly out of thermal equilibrium. In Ref. 5 the quantity τ_M has been calculated within the microscopic three-temperature model described above where the relaxation is driven by the different temperatures of the electron, spin, and lattice subsystems. For low fluences we can use the theory of Yafet^{6,18} in which a weak nonequilibrium situation for the electronic spin states is modeled by prescribing initially two different chemical potentials for electrons with two spin characters, and this difference is the driving force for the proceeding relaxation which is achieved by spin-dependent electron-lattice scattering. Whereas in Ref. 5 the spin-dependent scattering is described from the very beginning by one effective matrix element, the theory of Yafet contains the real matrix elements for the scattering between different electronic states $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}'}$. The key point of the theory is the fact that in a system with spin-orbit coupling the wave functions $\Psi_{j\mathbf{k}}$ are always mixtures of the two spin states $|\uparrow\rangle$ and $|\downarrow\rangle$ with probability $p_{j\mathbf{k}s}$ to find an electron in the spin state s . The degree of spin mixing is described by the parameter

$$b_{j\mathbf{k}}^2 = \min(p_{j\mathbf{k}\uparrow}, p_{j\mathbf{k}\downarrow}), \quad (6)$$

whereby for most states $b_{j\mathbf{k}}^2$ is much smaller than one, i.e., most states have a dominant spin character. In a simplified

version of Yafet's theory the spin-flip matrix elements are not calculated explicitly but estimated by simple physical arguments. Within this simplified version the so-called Elliot-Yafet relation^{6,19} for τ_M is derived

$$\tau_M = \frac{1}{pb^2} \tau_c, \quad (7)$$

where b^2 is an average of $b_{j\mathbf{k}}^2$ over all states involved in the relaxation, p is a material-specific parameter which should be close to 4 (and which should not be mixed up with the above defined probability $p_{j\mathbf{k}s}$ to find a single electron in the spin state s), and the quantity τ_c is the relaxation time entering Drude's theory of electrical conductivity.

Because in the breathing Fermi-surface model the lifetime τ is generally assumed to be identical to the Drude relaxation time τ_c , we can derive from Eqs. (4) and (7) the relation

$$\tau_M = \frac{M}{\gamma F_{el} p b^2} \alpha, \quad (8)$$

which is the central result of the present paper.

Please note two fundamental differences between Eq. (3), which is the central result of Ref. 5 and Eq. (8). First, in Eq. (3) τ_M is proportional to $1/\alpha$ whereas it is proportional to α in Eq. (8). The proportionality to α is related to the fact that we considered the conductivitylike contribution which dominates the damping at long lifetimes τ (respectively, low temperatures). The resistivitylike contribution depends also on the lifetime τ , however, in a more complicated manner. It increases monotonically with increasing τ^{-1} , and for small τ^{-1} (where the conductivitylike contribution dominates) it is proportional to τ^{-1} , $\alpha = \tilde{F}_{el} / \tau$. Thereby, \tilde{F}_{el} is again a quantity which is determined by the properties of the electronic states but it is different from the quantity F_{el} appearing in Eqs. (4) and (5). Whereas F_{el} can be expressed by matrix elements which are formed with, respectively, the same single-electron wave functions $\Psi_{j\mathbf{k}}$, the quantity \tilde{F}_{el} contains matrix elements formed by two different wave functions $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}'}$, respectively, see, e.g., Ref. 14. This procedure yields the relation

$$\tau_M = \frac{\tilde{F}_{el}}{pb^2} \frac{1}{\alpha} \quad (9)$$

between the demagnetization time and the resistivitylike contribution to α , and this relation has the same form as Eq. (3) given by Ref. 5. Altogether, it becomes clear that Eq. (3) is not valid for all situations, it is certainly not valid for very long lifetimes (respectively, low temperatures) where the conductivitylike contribution to α dominates. This may be a further reason why relation (3) could not be confirmed quantitatively in the experiments. (Please note that for Fe, Co, and Ni probably both contributions to α are relevant at room temperature.^{10,13})

The second essential difference is that in Eq. (3) just one material parameter (H_{ex}) appears which has nothing to do with spin-orbit coupling, whereas the quantities F_{el} and \tilde{F}_{el} of Eqs. (8) and (9) are determined by the sensitivity of the electronic states on changes in the spin-orbit coupling. On

the first sight it therefore looks as if Eqs. (3) and (9) were, in principle, not compatible with each other. Therefore we must conclude that $\alpha\tau_M = \tilde{F}_{el}/pb^2$ should depend only weakly on the strength ξ of the spin-orbit coupling $\xi(\mathbf{L}\cdot\mathbf{S})$ between the spin angular momentum \mathbf{S} and the orbital angular momentum \mathbf{L} of an electron. Indeed, it has been shown¹⁴ that $\tilde{F}_{el} \sim \xi^2$, and because in first-order perturbation theory b^2 is also proportional to ξ^2 , the quantity $\alpha\tau_M$ does not depend on ξ .

Finally, we want to test our relation (8) against experimental results. We take the case of Ni because in this material the damping is definitely dominated by the conductivity-like contribution at low temperatures, and Ref. 10 gives a value of $\lambda = \gamma M\alpha = 1.07 \times 10^8/\text{s}$ for that contribution at room temperature, where λ is the Landau-Lifshitz damping parameter. Furthermore, the value of τ_M as fitted from $M(t)/M(t=0)$ at low fluences is approximately 100 fs [Fig. 3d of Ref. 4]. This yields the experimental value of $\alpha/\tau_M = 1.2 \times 10^{11}/\text{s}$. To calculate the theoretical value of α/τ_M by Eq. (8), we take $p=4$ and $b^2=0.025$. This value of b^2 has been calculated by the *ab initio* electron theory²⁰ under the assumption that the dominant contribution to the de-

magnetization arises from thermally excited electrons and holes.⁴ The value of F_{el} calculated by the *ab initio* electron theory is taken from Fig. 2 of Ref. 21 (for a precession around [111]). Altogether, Eq. (8) then yields $\alpha/\tau_M = 0.6 \times 10^{11}/\text{s}$, a value which agrees astonishingly well with the experimental value.

To conclude, we have calculated by a purely microscopic approach a relation between the Gilbert damping parameter α at low temperatures and the demagnetization time τ_M for ultrafast laser-induced demagnetization at low fluences. The predicted value for α/τ_M is in good agreement with the experimental value. The theory therefore provides a link between the magnetization dynamics on the fast (nanoseconds to several picoseconds) and the ultrafast (approximately 100 fs) time scale.

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⁸It should be noted that an equation of the form of Eq. (3) relating the single-spin dephasing time $\tau_{M,t}$ to α has been derived by completely different methods also by H. J. Skadsem, Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, *Phys. Rev. B* **75**, 094416 (2007); and by L. Berger, *ibid.* **80**, 144427 (2009).

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