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In order to explain a number of recent experimental observations of laser-induced femtosecond demagnetization in the large fluence limit, we discuss the consequences of a recently proposed nonlocal approach. A microscopic description of spin flip scattering is implemented in an effective three temperature model, including electronic heat diffusion. Effects of finite film thickness on the demagnetization transients are discussed. Our results show a clear saturation of the ultrafast demagnetization, in excellent agreement with experimental observations. © *2011 American Institute of Physics*. [doi:10.1063/1.3540681]

All optical techniques exploiting femtosecond laser pulses have opened the way toward the exploration of the ultimate limits of magnetization dynamics. It has been shown that it is possible to (partially) quench the magnetic ordering of ferromagnetic thin films within a few hundred femtoseconds after laser excitation [see Refs. 1–5]. One of the outstanding issues is the behavior under very intense laser pulses, raising the temperature to near or above the Curie temperature (T_C). Simple intuition predicts a rapid increase of the demagnetization amplitude as a function of laser fluence while approaching the T_C. In contrast, in experiments the demagnetization seems to level off at values of around a tenth of the saturation magnetization,[6] and it has been speculated whether full demagnetization is possible at all without changing the underlying mechanism.[7] In this paper, we show that such behavior is a natural consequence of the finite optical penetration depth (λ) of the laser light used to investigate the dynamics, and can be quantitatively accounted for by a nonlocal extension of the three-temperature model (3TM), the latter describing the ultrafast equilibration of the electron-, spin-, and lattice systems.

Recently, we have proposed a theory for laser induced demagnetization, based on a finite spin-flip probability upon momentum scattering.[8] Experimental support for such a scenario has been reported earlier on in Refs. 6, 7 and 10. In the present work, we use the microscopic implementation of the 3TM (M3TM), while implementing heat diffusion via conduction electrons to treat the nonhomogeneous case.[9] It will be shown that drastic effects arise even for metallic films with a thickness of only 10 nm to 20 nm, such as for films with a thickness comparable to the extinction depth of the laser light. For the well-studied case of nickel thin films, both the measured demagnetization as a function of laser fluence, as well as the 'saturating' temporal magnetization profiles can be quantitatively described for realistic parameters. We will start by briefly reviewing the M3TM. We will then show how it can be extended to treat the nonhomogeneous heating and large fluence cases. Finally, we will present simulations for a number of exemplary cases, and discuss their correspondence with recent experimental results in the high fluence regime.

Within the 3TM (Ref. 1) heat capacities and temperatures are assigned to the reservoirs of electron charge (e), spin (s), and lattice (l), (C_e, T_e), (C_s, T_s), and (C_l, T_l), respectively. Furthermore, coupling constants are defined as g_{es}, g_{sl}, g_{el}, describing the rate of energy exchange between the systems. Here we make use of a recently introduced microscopic extension to the model that allow us to better describe experiments in the large fluence limit, heating (almost) to the T_C, and in cases where films are not heated homogeneously throughout.[9]

To include the case of nonhomogeneous heating, we restrict ourselves to a one-dimensional model, explicitly making the three temperatures a function of the z-coordinate.[9] For the electron specific heat we make the usual approximation: $C_e(T_e) = \gamma T_e(z)$. We assume the heat diffusion to be dominated by the electrons and to be described by the heat conductivity κ.[11] The ferromagnetic film of thickness d is sandwiched between thermally insulating media, e.g., a vacuum or an oxidic substrate, such as Si/SiO_x. Thus, we derive a set of coupled differential equations for the electron and lattice temperature:

$$\begin{aligned} C_e(T_e(z))\frac{dT_e(z)}{dt} &= \nabla_z(\kappa\nabla_z T_e(z)) + g_{el}(T_l(z) - T_e(z)), \\ C_l\frac{dT_l(z)}{dt} &= g_{el}(T_e(z) - T_l(z)). \end{aligned} \tag{1}$$

We assume instantaneous heating of the electron system by the laser pulse followed by infinitely fast thermalization of the electron gas to a temperature profile $\Delta T_e(z, 0) = \Delta T_{pump} \exp(-z/\lambda)$. For this approximation to hold, it is required that the energy is deposited relatively locally. This condition would not be fulfilled in noble metals such as silver and gold, which have a much longer hot electron scattering length.[12]

To describe how the spin system adapts to the local electron and lattice temperature, we rely on our microscopic model, M3TM, introduced in Refs. 5 and 9. There, spin

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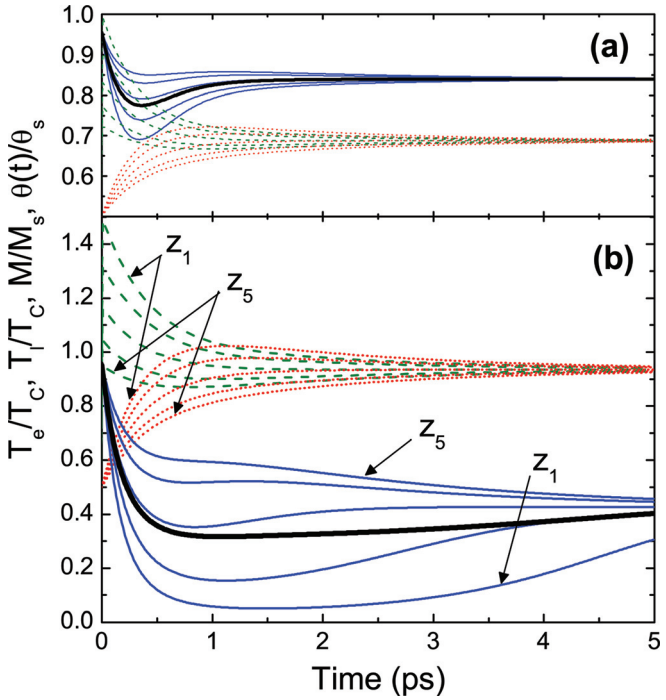

FIG. 1. (Color online) Dependence of electron temperature (dashed), phonon temperature (dotted), and magnetization [M/M_s, (solid)] as a function of delay time after pulsed laser heating at $t=0$, and for five depths in the thin isolated Ni film of 15 nm as indicated. $\mathbf{z}_1$ and $\mathbf{z}_5$ correspond to the first and the fifth slab starting from the interface, respectively. The (thick solid) curves show transient MO signals, $\theta(t)/\theta_S$. Parameters are representative for nickel (see text). (a) low fluence, (b) large fluence.

relaxation is mediated by Elliot-Yafet like processes such as spin-flip scattering upon momentum scattering events with a probability a_{sf}. We derived a compact differential equation that relates the (local) rate of spin change to the (local) electron and phonon temperature. For temperatures near or above the Debye temperature, this equation reads

$$\frac{\mathrm{d}m(z)}{\mathrm{d}t} = Rm(z)\frac{T_l(z)}{T_C}\left(1 - m(z)\coth\left(\frac{mT_C}{T_e(z)}\right)\right), \qquad (2)$$

where $m = \frac{M}{M_s}$, the magnetization relative to the value at $T=0$. The prefactor R can conveniently be written as $R = (8a_{sf}g_{el}k_BT_c^2V_{at}\mu_B)/(\mu_{at}E_D^2)$,[9] with μ_{at} the atomic magnetic moment in units of Bohr magneton μ_B, V_{at} the atomic volume, and E_D is the Debye energy. The temperature dependence of the magnetization is assumed according to the Weiss model. The final task is just to solve the three coupled differential equations [Eqs. (1) and (2)], after applying an initial (nonhomogeneous) perturbation to $T_e(z)$. We calculated the total magneto-optical (MO) signal by $\theta(t) \propto \int m(z,t)\exp(-z/\lambda)dz$, i.e., again accounting for the finite penetration depth.

We will discuss a number of elucidating examples for a thin film of nickel because it is by far the most studied elementary material in the field. Parameters used are $\gamma = 5.435 \cdot 10^3$ J/(m^{3}K^2), $C_l = 2.33 \cdot 10^6$ J/(m^{3}K), $g_{el} = 4.05 \cdot 10^{18}$ J/(m^{3}sK), and $E_D = 0.036$ eV. This set of parameters reproduces experimental T_e and T_l transients well. In particular it yields an electron-phonon energy equilibration time $\tau_E \approx 0.5$ ps. Furthermore, $\kappa = 90.7$ J/smK, $\mu_{at} = 0.62\mu_B$, $T_C = 627$ K, $\lambda = 15$ nm, and we use a spin-flip probability $a_{sf} = 0.185$ according to previous results.[9] All calculations were done for an ambient temperature $T = 0.5\ T_C \approx 310$ K.

Using the model and parameters previously discussed, we simulated temperature profiles and demagnetization traces for low and high fluences, as presented in Fig. 1(a) and (b), resp. Simulations are performed for a 15 nm nickel film, i.e., equal to λ. In this case, we did not consider any underlying layer and we will refer to this structure as the isolated layer. To emphasize the nonhomogeneous temperatures, we plotted curves representative for five positions throughout the film.

In Fig. 1(a) we reproduce a transient MO signal typically observed experimentally at low fluences:[3,4] A sharp drop in magnetization followed by a fast recovery, proceeding almost completely within a 1ps to 2 ps. Interestingly, the high fluence case [Fig. 1(b)] yields a completely different behavior. In particular, the recovery is much slower, and the dip in the signal is now much less pronounced compared with the final demagnetized state, again in agreement with experiments.[6] We traced back this different behavior to a superposition of two effects: (i) near the T_C, the magnetization dynamics driven by the average exchange field slows down, and (ii) the temperature of deeper regions in the film is recovering far less rapidly, because of a continuing heat flow from higher up in the film. In passing, we note that a similar slowing down of magnetization recovery when approaching the T_C has been predicted based on atomistic Landau-Lifshitz-Bloch and Landau-Lifshitz-Gilbert approaches.[13,14]

We then investigated the maximum demagnetization, defined as $\Delta M/M_0$, where M_0 is the magnetization at $T = 310$ K, as a function of laser fluence. For very thin isolated films (Fig. 2, filled symbols), $\Delta M/M_0$ increases rapidly with increasing fluence, and the film is completely demagnetized abruptly around $\Delta T_{pump} = 1.0\ T_C$. Such a behavior can be well understood intuitively because of the rapid decrease of M near T_C.

Repeating the calculation for identical parameters, but using a thickness of 30 nm [taken equal to the experiments in Cheskis *et al.* (2005)[7]], yields a very contrasting behavior. It now needs very high fluences to completely quench the magnetization, i.e., $\Delta M/M_0 = 1$. The fluence needed to fully quench the magnetic signal ($\Delta T_{pump} \approx 3.0\ T_C$) is approximately twice as a high as the fluence at which $\Delta M/M_0$ reaches 80% ($\Delta T_{pump} \approx 1.5\ T_C$).

Such a saturation has been seen in experiments and reported more often over the past years. It was the basis of claims that full saturation might be limited by unknown bottlenecks, and that this "anomalous behavior" needs a specific microscopic origin.[7] In contrast, our modeling shows that it is a natural consequence of nonlocal effects accompanying the finite penetration depth of the light, which can intuitively be explained. Although a limited laser fluence is needed to heat up the top part of the film to the T_C, it needs much more power to drive the deepest region of the film above the T_C in cases of films that are much thicker than the penetration depth.

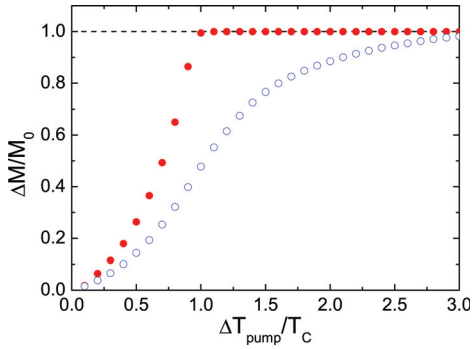

FIG. 2. (Color online) Maximum demagnetization ($\Delta M/M_0$) versus laser fluence (defined in terms of ΔT_{pump}, see text) for isolated nickel thin films of different thicknesses. Filled symbols: Optically thin film of 5 nm. Open symbols: Film of 30 nm, as used in Cheskis *et al*. (Ref. 7).

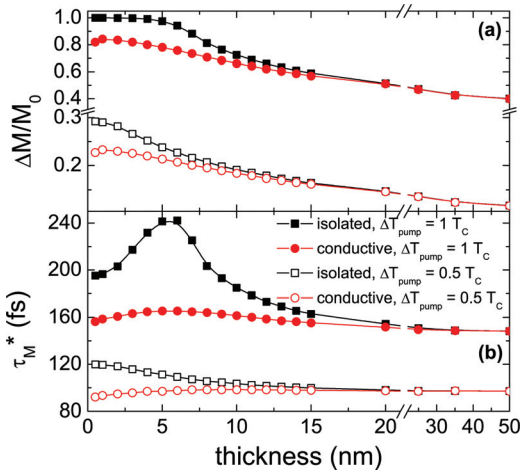

FIG. 3. (Color online) (a) Maximum demagnetization ($\Delta M/M_0$) and (b) effective demagnetization time (τ_M^*) versus the sample thickness for two laser fluences (defined in terms of ΔT_{pump}, see text) for isolated nickel thin film and a nonisolated nickel film (conductive structure, see text). Here, calculations were performed for an ambient temperature of 300 K.

Earlier in this paper, we saw that extrinsic parameters, such as fluence and sample thickness, can influence the demagnetization process enormously. We now want to show that the demagnetization process, largely characterized by $\Delta M/M_0$ and the effective demagnetization time (τ_M^*), is also strongly affected by the sample structure as it influences the heat dissipation. To prove this statement, two different sample structures were simulated. The first one corresponds to an isolated Ni layer as used previously with variable thickness d. The second structure corresponds to a thin film with a constant total thickness of 50 nm. However, this structure consists of two parts: the top part of the film with thickness d is Ni, and thus magnetic, in contrast to the remaining $50-d$ nm. This part is a nonmagnetic metal for which we assume the same thermal and optical parameters as Ni. This structure is referred to as the conductive structure.

In Fig. 3, the results of the simulations performed on the isolated layer and conductive structure are shown for two different fluences. In general, we see in Fig. 3(a) that $\Delta M/M_0$ decreases for increasing thickness and in Fig. 3(b) that τ_M^* is larger for a larger fluence.[9] These observations are in line with Figs. 2 and 1, respectively. For a film thickness larger than the laser penetration depth (15 nm) and equal fluence, both $\Delta M/M_0$ as well as τ_M^* tend to be the same constant value for both structures.

Comparing the isolated structure with the conductive structure , we see that for a structure thinner than the penetration depth, $\Delta M/M_0$ [Fig. 3(a)] is larger for the isolated layer compared with the conductive structure. This can be explained by a much slower heat dissipation in the isolated structure compared with the conductive structure, because the temperature gradient, and therefore the heat diffusion, are faster in the z-direction. The nonhomogeneous temperature and heat diffusion in the isolated structure are also directly reflected in the demagnetization time leading to a larger τ_M^* [Fig. 3(b)]. The effect is obvious for high laser fluence ($\Delta T_{pump} = 1.0\ T_C$) for which a demagnetization time as large as 240 fs is observed for a thickness of 5 nm and decreases to 150 fs for larger thicknesses. The variation is much less pronounced in the case of the conductive structure giving rise to a maximum demagnetization time of 165 fs for a thickness of 5 nm, i.e., 30% smaller than for the isolated structure. Similar trends, though less pronounced, are observed for lower laser fluence.

Summarizing, we implemented a nonlocal (3TM), using a microscopic implementation of the demagnetization process, and allowing for electronic heat conduction. Based on our explicit simulations for nickel thin films, we conclude that no anomalous behavior occurs in the high fluence regime, and the results can be well described by existing theories once nonlocal effects are properly included. Moreover, we have shown that the sample structure, and its thickness, influence the observed demagnetization time. The latter effect should be carefully considered when comparing reported values for the demagnetization time, based on experiments, with a different sample layout.

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