Comparing Ultrafast Demagnetization Rates Between Competing Models for Finite Temperature Magnetism

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We investigate a recent controversy in ultrafast magnetization dynamics by comparing the demagnetization rates from two frequently used but competing descriptions for finite temperature magnetism, namely a rigid band structure Stoner-like approach and a system of localized spins. The calculations on the localized spin system show a demagnetization rate and time comparable to experimentally obtained values, whereas the rigid band approach yields negligible demagnetization, even when the microscopic spin-flip process is assumed to be instantaneous. This shows that rigid band structure calculations will never be in quantitative agreement with experiments, irrespective of the investigated microscopic scattering mechanism.

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More than a decade ago, it was shown that a femtosecond (fs) laser pulse can quench the magnetization in the transition metal ferromagnets on subpicosecond time scales [1]. Since then, both experimental [2–7] and theoretical [8–17] efforts have been made to identify the origin of this ultrafast demagnetization, but no consensus has been reached yet. One of the most prominent candidates to explain ultrafast demagnetization upon fs laser excitation is scattering of various (quasi-)particles, leading to an ultrafast transfer of angular momentum from and/or to the spin system [4,5,9,13]. Multiple scattering mechanisms have been suggested to accommodate this angular momentum transfer, but if and which one dominates the fs magnetization dynamics is still being debated. More strongly, conflicting results on the demagnetization efficiency of a single scattering mechanism have been obtained using different descriptions for finite temperature magnetism [12,15,16]. In this Letter we identify why these large discrepancies occur, and which approaches are suitable for describing ultrafast magnetization dynamics. We argue that such an analysis is of crucial importance for unraveling the governing microscopic mechanism.

To model ultrafast heating of a ferromagnet from first principles one first needs to be able to fully calculate the equilibrium properties of a ferromagnet at finite temperatures. Unfortunately, describing finite temperature magnetism from first principles is one of the most difficult problems in solid state physics as of today. In itinerant ferromagnets both the delocalized wavelike as the localized particlelike character of the electrons play a crucial role [18]. An *ab initio* framework capturing both these aspects is extremely complicated; hence, using such methods to calculate ultrafast dynamics seems to be a forbidding endeavor. Therefore, all calculations performed in the literature on ultrafast demagnetization resorted to a simplified representation of the spin system or spin excitations, which we can generally divide into two types of models.

The first type of models [10–12,17] is based on the assumption that the magnetic moments are localized in real space, such as in the Weiss or Heisenberg model for ferromagnetism. The atomic magnetic moments are aligned due to the exchange interaction, and thermal fluctuations of the direction of these moments change the average magnetic moment. Although this approach is rather phenomenological, it has been highly successful in describing thermodynamical magnetic properties such as the Curie temperature T_C and the magnetic susceptibility at large temperatures, which are not easily reproduced by band structure models.

On the other hand, ultrafast demagnetization rates have been calculated using a zero temperature band structure [13,15,16,19], where scattering changes the occupation of spin up and spin down states. This approach is similar to the Stoner model, where spin flips of delocalized electrons quench the atomic magnetic moment instead of change the local direction of the magnetization. An advantage of such an approach is that the microscopic properties, such as scattering rates, can be fully calculated from first principles.

Recently it has been shown that the two aforementioned models of a ferromagnet give conflicting results for the calculated demagnetization rates [12,15,16]. This begs the question which of the approaches is suitable for quantitative calculations on ultrafast magnetization dynamics. We will answer this question by performing calculations on the demagnetization of Ni due to a single microscopic scattering mechanism. By only changing the Hamiltonian describing the spin system it is possible to discuss the driving forces for demagnetization and directly compare the results for these two approaches.

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For the presented calculations a model is used which includes all key ingredients to describe ultrafast demagnetization, while remaining as simple as possible to easily compare the two approaches. To this end all calculations are performed in the random-k approximation and all matrix elements are assumed to be constant for all electronic states. The microscopic scattering mechanism investigated is a phonon assisted Elliott-Yafet spin flip, as this is the topic of recent controversy [12,15]. We would like to stress, though, that the final conclusions hold for all microscopic scattering mechanisms.

As a starting point for the calculations the zero-temperature density of states (DOS) of Ni is used, which is depicted in Fig. 1(a). The DOS is filled up to the Fermi level with noninteracting electrons. The lattice is modeled by a set of coupled oscillators (phonons) of a single energy $\hbar\omega_p$ obeying Bose-Einstein statistics, of which the energy is chosen to be 0.03 eV, i.e., close to the Debye energy of Ni. Both electron-electron (e–e) and electron-phonon (e–p) scattering are taken into account by evaluating Fermi's Golden rule.

Without spin-orbit coupling both e–e and e–p scattering are spin-conserving processes; i.e., the total magnetization remains unchanged. However, due to spin-orbit coupling the electronic eigenstates are mixtures of the two spin states. This means that the e–p scattering Hamiltonian connects the spin-mixed up and down states; hence, an e–p scattering event can yield a spin-flip transporting angular momentum from the electrons to the lattice. The probability a_{sf} that an e–p scattering event yields a spin-flip can be calculated from first principles, which yields values for Ni of a_{sf} ranging from 0.04 to 0.25 [12,15]. For simplicity we here assume a constant a_{sf} for all electronic states.

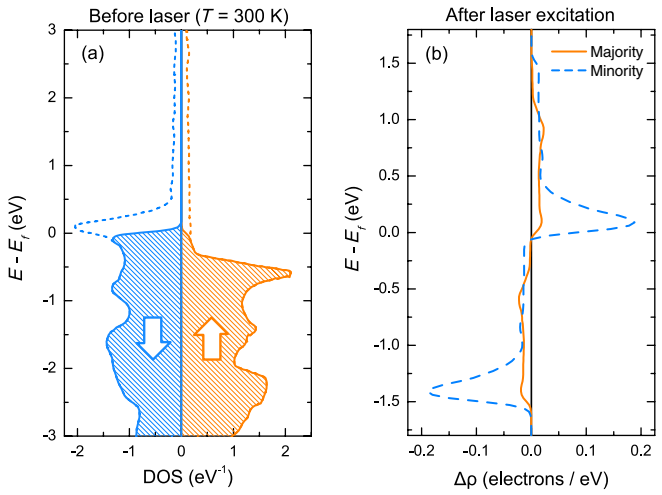

FIG. 1 (color online). (a) DOS and equilibrium occupation of Ni at an ambient temperature of 300 K taken from [15]. (b) Example of redistribution of electrons after a fs laser pulse of 1.5 eV.

In the rigid band structure approach, e–p spin flips are Stoner-like excitations that transport majority electrons to minority states and vice versa, where the spins of the majority (minority) electrons are aligned parallel (antiparallel) to the net average spin in the magnetic material. By evaluating Fermi's golden rule we obtain for example the following rate for an e–p scattering event, where a phonon is absorbed and a spin is flipped from up to down,

$$T_{|\uparrow\rangle\rightarrow|\downarrow\rangle} = a_{\mathrm{sf}} K_{ep} N_p \sum_k \sum_{k'} D_\uparrow(E) D_\downarrow(E+E_p) f_\uparrow(E) \times [1 - f_\downarrow(E+E_p)], \quad (1)$$

where k and k' label are labels for the electronic states, $D_\uparrow$, $D_\downarrow$, $f_\uparrow$, and $f_\downarrow$ the density of states and occupation of the majority and minority electrons, respectively. Furthermore, $K_{ep} = 2\pi D_p \lambda_{ep}^2/\hbar$ in which λ_{ep} is the e–p scattering matrix element for $a_{\mathrm{sf}} = 0$ and D_p is the amount of phonon modes per atom that couple to the electrons, which is in good approximation equal to 1. In all calculations K_{ep} is taken to be 4.9×10^{-3} fs^{-1} eV^{-1}, corresponding to an e–p equilibration time of ≈ 1 ps. Similar expressions to Eq. (1) can be obtained for phonon absorption and/or spin flips from down to up.

Next, we calculate demagnetization in the localized atomic spin approach, for which a simple self-consistent Weiss mean-field model for spin 1/2 is used. This means that the spin system is described by an external spin bath which consists of two energy levels separated by the exchange interaction $J_{\mathrm{ex}} = 2k_{\mathrm{B}}T_{\mathrm{C}}m$, where m is the average normalized magnetization and T_{C} the Curie temperature, which is set to 620 K for Ni. To flip a spin in this mean field approach an energy J_{ex} is required, which is the energy involved for creating a spin excitation, i.e., for breaking ferromagnetic order. This means that an electron gains or loses an energy of $\pm E_p \pm \hbar\omega_{\mathrm{magnon}}$ in an e–p spin flip event. Similar expressions as in Eq. (1) can now be obtained for the Weiss approach. For example, the rate of creating spin excitations due to majority electrons absorbing phonons is given by

$$T_{|\uparrow\rangle\rightarrow|\downarrow\rangle} = \frac{a_{\mathrm{sf}}}{D_{\mathrm{S}}} K_{ep} N_p \sum_k \sum_{k'} D_\uparrow(E) D_\uparrow(E+E_p-J_{\mathrm{ex}}) \times \frac{D_{\mathrm{S}}}{2}(1+m) f_\uparrow(E)[1 - f_\uparrow(E+E_p-J_{\mathrm{ex}})]. \quad (2)$$

The factor $D_{\mathrm{S}}(1+m)/2$ that appears into the rate equation corresponds to the atomic spin up density. Note that in this spin flip event not the atomic magnetic moment is changed, but the local orientation of the quantization axis, which involves a different energy scale. Finally, we conjecture that Eq. (2) correctly describes phonon induced spin flips in a localized spin system, even though they are not identical to the single-electron band excitations originally considered by Elliott and Yafet for nonmagnetic metals.

To simulate the effect of absorption of a 1.5 eV fs laser pulse, the following three approximations are made: (i) the dipole matrix elements are identical for all electronic states, (ii) no spin flips are allowed in the optical excitation process, and (iii) the laser pulse is infinitesimally short, i.e., optical excitation takes place at $t = 0$. An example of a typical absorption profile is depicted in Fig. 1(b), where the change in the amount of electrons per energy $\Delta\rho$ is given.

Before examining the dynamic properties of the two models the equilibrium magnetization as a function of temperature is calculated and plotted in Figs. 2(a) and 2(b) for the Weiss and rigid band approach, respectively. Whereas the Weiss approach yields the typical curves for Ni, the magnetization in the rigid band approach is virtually independent of the ambient temperature. This is in line with the fact that the Stoner model for ferromagnetism predicts Curie temperatures for the transition metal ferromagnetism that are typically five times larger than the experimental observations; i.e., Stoner excitations do not account for the finite temperature properties of ferromagnets.

In Figs. 2(c) and 2(d), calculated demagnetization traces for the Weiss and rigid band structure approach are depicted, where the magnetization divided by the magnetization

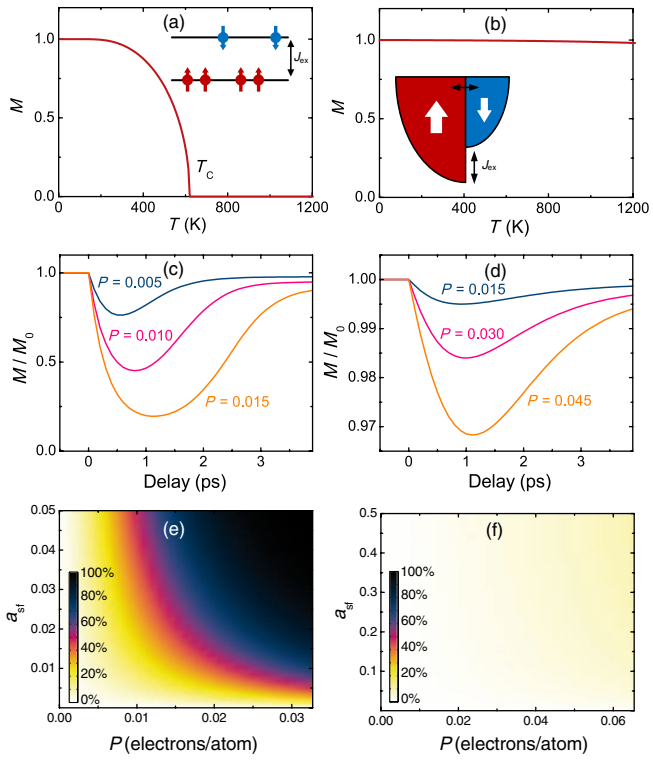

FIG. 2 (color online). Equilibrium magnetization in the Weiss (a) and rigid band structure (b) approach for constant electron and phonon temperatures. Examples of room temperature demagnetization traces of Ni for the Weiss (c) and rigid band structure (d) approach. The different lines correspond to various laser fluences. Maximum quenching as a function of a_{sf} and P for the Weiss (e) and rigid band structure (f) approach, respectively.

before laser excitation M/M_0 is depicted as a function of time. For the calculations K_{ee} is set to 27 fs^{-1} and a_{sf} to 0.05, comparable to recent *ab initio* calculations [15]. At first sight, the traces for the two models seem quite similar; a fast demagnetization is followed by a slower remagnetization, which is frequently observed in experiments on Ni. However, the demagnetization rates in the Weiss model are almost two orders of magnitude larger compared to the rigid band structure for identical laser fluences and microscopic parameters. This large difference is further exemplified by Figs. 2(e) and 2(f), where the maximum quenching $(M_0 - M_{min})/M_0$ is given for a large range of fluences and spin flip parameters. For the Weiss model almost complete demagnetization is observed for relatively small values of a_{sf} and P, whereas the rigid band structure model yields negligible demagnetization rates, even for a spin flip parameter as large as 0.5 and a fluence that brings the whole system to a temperature well above T_C.

The question now remains why there is such an extreme difference between the two approaches. It is important to realize that scattering always tends to equilibrate the various subsystems. In the case of the Weiss model scattering equilibrates the spin temperature to the electron temperature. Because the electron temperature is increased to above T_C by absorption of the laser pulse, and since the equilibrium magnetic moment depends strongly on the temperature in the Weiss model, a large demagnetization is naturally reproduced. The case of a rigid band structure is more complicated, since the equilibrium magnetization is virtually independent of the ambient temperature; see Fig. 2(b). The origin of demagnetization in this approach is equilibration of the spin up and spin down occupation densities for a certain energy level, i.e., $f_\uparrow(E) = f_\downarrow(E)$. This is illustrated by the data depicted in Fig. 3.

In Fig. 3(a) a typical demagnetization trace is depicted, calculated in the rigid band structure approach for $a_{sf} = 0.1$. Again a rapid demagnetization is followed by a slower remagnetization, similar to Fig. 2(d). To examine the origin of this demagnetization in more detail, $D_\uparrow D_\downarrow (f_\uparrow - f_\downarrow)$ is plotted as a function of E for various time delays in Fig. 3(b). From Eq. (1) it can be easily seen that this occupation difference is directly related to the spin flip rate. At $t = 0$ a large positive peak below the Fermi level indicates that there are more majority than minority electrons, which is caused by the excitation of minority electrons by the laser, in agreement with Fig. 1(b). This means that spin flips of majority electrons will cause a demagnetization at $t = 0$.

Now due to e–e scattering the peak will quickly shift to higher energies at larger delays, and eventually changes sign around 0.9 ps due to cooling of the electron system by the phonons. This corresponds to a change from demagnetization to remagnetization, which is in correspondence with the trace in Fig. 3(a). In Fig. 3(c) the maximum quenching connected to the nonequilibrium electron distribution is

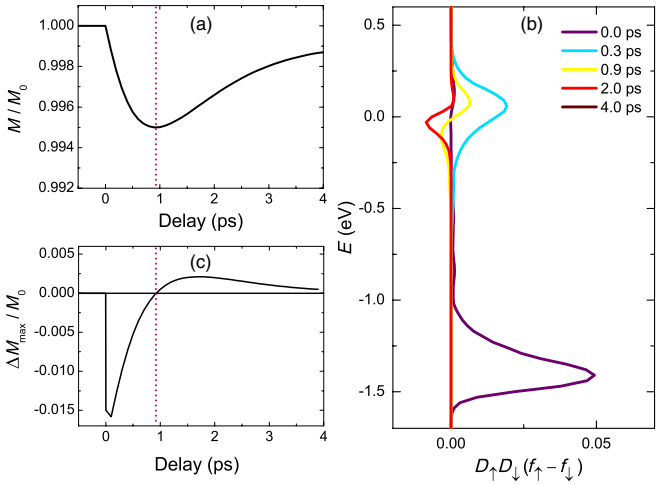

FIG. 3 (color online). Example of an ultrafast demagnetization trace calculated in the rigid band structure model. The dotted line indicates the turning point for de- to remagnetization. (b) Difference of electronic occupation as a function of energy for various time delays. A positive peak indicates demagnetization, whereas a negative one causes remagnetization. (c) Theoretical maximum quenching if demagnetization was instantaneous. The dotted line indicates the intersection with zero, which corresponds to a change from de- to remagnetization.

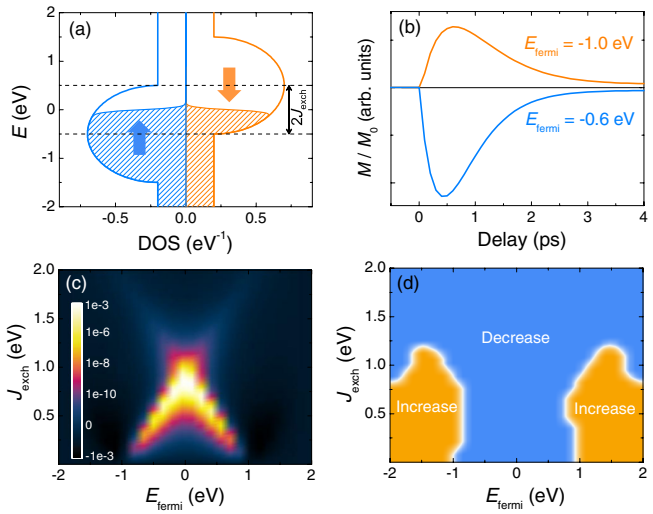

FIG. 4 (color online). (a) Model DOS of a ferromagnet with spin split d bands. (b) Examples of calculated ultrafast magnetization dynamics using the DOS in (a) for two different positions of the Fermi level. (c) Calculated demagnetization rates at $t = 0$ for a wide range of positions of the Fermi level and exchange splittings. (d) Phase diagram showing for which parameters the magnetization of the ferromagnet is increased or decreased by a fs laser pulse.

calculated as a function of delay time, for the hypothetical case that the spin flip scattering mechanism is instantaneous by assuming $a_{\mathrm{sf}} = \infty$. Using Eq. (1) and some simple algebra shows that this maximum quenching is given by $\sum_k D_\uparrow(f_\uparrow - f_{\mathrm{equi}})$. What can be surprisingly concluded from the data, is that even if e–p spin-flip scattering would be instantaneous the maximum quenching would never exceed 1.5% of the total magnetization, although the rise in electron temperature is several hundreds of Kelvin. This unambiguously demonstrates that it is not the strength or effectiveness of the scattering mechanism that limits the demagnetization rate, but rather the fact that in a rigid band structure the magnetic moment is not strongly influenced by the electronic occupation; i.e., there is simply no driving force for demagnetization.

To generalize our calculations to an arbitrary band ferromagnet, the demagnetization efficiency is evaluated for the following fictitious DOS: $D_{\uparrow(\downarrow)}(E) = D_s + D_d\sqrt{1 - [(E \mp J_{\mathrm{ex}})/\Delta E_d]^2}$. This DOS consists of two exchange split d bands with a width ΔE_d and a maximum DOS D_d. Futhermore, there is an energy independent contribution D_s due to s electrons. An example of such a DOS is given in Fig. 4(a). For all calculations $D_s = 0.2$ eV^{-1}, $D_d = 0.5$ eV^{-1} and $\Delta E_d = 1$ eV. In Fig. 4(b) two calculated demagnetization traces using the rigid band structure approach are shown for $J_{\mathrm{ex}} = 0.5$ eV and two different positions of the Fermi level. Surprisingly, depending on the position of E_{fermi}, the magnetization can both increase or decrease due to absorption of the laser pulse. To substantiate this, demagnetization rates at $t = 0$ are calculated for various strengths of J_{ex} and positions of the Fermi level. The results are shown in Fig. 4(c). Two regions where there is an increase in the demagnetization can be identified, which is more clearly shown in the phase diagram in Fig. 4(d). This is in strong contrast with the Weiss approach, where laser pulse heating always yields a decrease of the magnetization, irrespective of the position of the Fermi level.

In conclusion, we have investigated the effect of a fs laser pulse on the ultrafast magnetization dynamics of Ni using two different descriptions of the spin system. It is demonstrated that a rigid band structure approach yields an almost zero demagnetization, whereas for the same parameters a significant demagnetization is obtained in the Weiss model. In that case, using the microscopic materials parameters from *ab initio* rigid band structure calculations reproduces experimentally observed demagnetization and demagnetization times accurately. Finally, we conclude that calculations on ultrafast demagnetization in the rigid band structure approach will never reproduce the experimentally observed demagnetization, irrespective of the type or strength of the investigated scattering mechanism.

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