Dynamics of femtosecond magnetization reversal induced by circularly polarized light in the presence of fluctuations and dissipation

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Magnetization reversal by a femtosecond circularly polarized laser pulse has been recently demonstrated in rare-earth doped transition metals (RE-TM). The switching mechanism has been attributed to an inverse Faraday effect and thermal effects. Based on the parameters provided in the experimental work, we show that this claim is unlikely to give rise to femtosecond reversal. Using a hybrid itinerant-localized picture of the RE-TM system, we propose a new mechanism that requires the presence of the rare earth element to reduce the symmetry of the system as well as a strong enhancement of spin-orbit coupling between the d electrons and the f moments in the presence of the laser. Our model does not require the heating close to the Curie temperature of the sample.

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Using perpendicular magnetized $Gd_{22}Fe_{74.6}Co_{3.4}$ thin films, it was shown that a 40 fs circularly polarized laser pulse with a wavelength of 800 nm is capable of switching the magnetization [1]. What is remarkable about this important and unexpected result is that it was all-optical and no external fields were needed. Instead the switching was critically dependent on the chirality and the intensity of the laser pulse. The minimum intensity of the laser needed to accomplish reversal was of the order of $10^{11}$ W/cm$^2$ which is still too low for the direct exchange of the angular momentum between the laser and the magnetization to be significant. The authors of Ref. [1] argued that a magnetic field generated by the inverse Faraday effect, $H_{IFE}$, in combination with the enhanced susceptibility at elevated temperatures is responsible for the switching. They estimated $H_{IFE}$ to be of the order of 10 T and the switching temperature $T_{SW}$ to be close to the Curie point $T_C$. However an equally valid reinterpretation of their data leads to $T_{SW} \approx (T_C - T_{room})/2$ [2]. The dynamics
of the reversal was not investigated in Ref. [1], but it is believed to occur on the femtosecond time scale. This belief is based on the fact that controlled switching is only possible if it is faster than relevant energy and phase relaxation times.

In this paper, we first discuss the ideas put forward in Ref. [1] in a more quantitative way and show that their picture can not explain femtosecond magnetization reversal. After that, we suggest qualitatively a different richer picture of the dynamics in the system and point out the need to go beyond LLG to study magnetization dynamics on the femtosecond time scale. The necessity stems from the fact, that strong magnetic fields generated by the laser are alone not sufficient to drive femtosecond magnetization reversal. Instead it is the simultaneous optical enhancement of dissipation that enables ultrafast switching. This dissipation is made possible by the fast relaxation of the hot electrons which requires the full account of the non-adiabatic dynamics in the laser-spin-electrons system. This latter aspect of the problem is treated in detail in this paper since it is of tremendous importance to any non-equilibrium system and in particular fast spin dynamics in systems driven far from equilibrium by a laser pulse. Consequently, a self-consistent treatment of dissipation and fluctuations is required which is also included here. Our method of solution goes beyond the Born approximation and is non-perturbative which is needed to accurately treat spin dynamics in an interacting environment as discussed by DiVincenzo and Loss [3]. It is based on the two-point effective action [4] and provides a systematic way to obtain solutions more accurate than those of the Bloch equations [5].

Before we discuss our model and method of solution, we first show that the ideas proposed in Ref. [1], which are based on LLG, can not form a basis for the understanding of the fast switching of the magnetization. The LLG equation is a phenomenological equation written for magnetization close to equilibrium and accounts for damping in the simplest possible way

\[
\frac{dM}{dt} = -\gamma M \times H_{\text{eff}} + \alpha M \times \frac{dM}{dt},
\]

(1)

The internal field and the external laser field, \(H_l = H_l(\cos \omega_l t, \sin \omega_l t, 0)\), are part of \(H_{\text{eff}}\). In a frame rotating with the frequency \(\omega_l = 10^{15} \text{Hz}\), \(H_l\) becomes time independent and the lab frame time derivative \(\frac{dM}{dt}\) becomes \(\frac{dM}{dt} - \omega_l z \times M\). Hence in the rotating frame, the effective field is \(H_{\text{eff}} + H_B\), where \(H_B = -\omega_l / \gamma z\) is the equivalent of the Barnett field which is of the order of \(5 \times 10^7\) Oe. This shows that the magnetization in the presence of the laser precesses at almost five orders of magnitude faster than in typical FMR experiments. The
LLG equation has been successful in reproducing most magnetization data with 'reasonable' damping constants $\alpha$ but it does a poor job in systematically accounting for damping in different correlated data sets. An example of such data is the measurement of damping in rare-earth doped transition metals (RE-TM) as a function of the orbital momentum of the impurities [6].

The dynamics of the magnetization reversal as obtained from Eq. 1 with $\alpha = 0.2$ and $H_{\text{IFE}} = 10$ T (experimental values of Ref. [1]) is shown in Fig. 1 for various laser fields. The laser intensity $H_l/H_B = 0.001$ corresponds to the experimental power and shows slow reversal within $\approx 20$ ps. The faster reversal curves for $H_l/H_B = 0.01$ and 0.1 is due to the Zeeman interaction with the laser but requires intensities well above the damage threshold. To achieve femtosecond switching at reasonable laser powers, the damping would have to be increased to non-physical values of $\alpha = 80.0$ (cf. inset of figure 1). This calls for a different way to include dissipation in fast spin dynamics. Heating GdFeCo to temperatures some tens of degrees below the Curie point also does not help achieve faster than picosecond reversal. The role of temperature is to increase the fluctuations in the initial values of $M_z$ which will help speed up the reversal but not to the extent of being fast at the femtosecond scale. The Curve with initial magnetization $M_z(0) = -0.5$ shows the effect of increasing temperature on the switching speed.

In the rest of this paper, we give a more careful treatment of the two aspects of time dependent fields and of damping in a simplified model accompanied by a qualitative discussion of the various factors involved in the reversal.

The excitation of GdFeCo by the laser unquenches the orbital angular momentum of the hot electrons which are mainly d electrons provided by Fe. But the d electrons also carry magnetization and hence a strong interplay of orbital magnetization and spin magnetization in this system is expected. The Hamiltonian is

$$\mathcal{H} = \mathcal{H}_d + \mathcal{H}_{\text{ph}} + \mathcal{H}_i,$$

$$\mathcal{H}_i = -eE(t) \cdot \mathbf{r} - H(t) \cdot (\mu_d s + \mu_f \mathbf{S}_f) + J s \cdot S_f + \lambda \mathbf{l} \cdot \mathbf{s} + \lambda_{so} l \cdot \mathbf{S}_f.$$

The Hamiltonian $\mathcal{H}_d$ is that of the d (and s, p) electrons of both Fe and Gd, $\mathcal{H}_{\text{ph}}$ is the Hamiltonian of the phonons and $\mathcal{H}_i$ is the interaction Hamiltonian which includes the dipole interaction, the Zeeman term with both d and f electrons, the spin-orbit coupling term of
FIG. 1: Dynamics of the perpendicular component $M_z$ in an inverse Faraday field $H_{IFE} = 10$ T. The curves are plotted for various laser fields $H_l/H_B = 0.1, 0.01, \text{ and } 0.001$ with initial $M_z(t = 0) = -1$. The curve starting at $M_z(0) = -0.5$ emulates the effect of elevated temperatures on the reversal for $H_l/H_B = 0.001$. The LLG damping is $\alpha = 0.2$. The inset is for $H_l/H_B = 0.001$ and the unphysical damping $\alpha = 80.0$.

The d electrons with coupling constant $\lambda$, the antiferromagnetic coupling between the d and f electrons and finally the effective spin-orbit coupling between the orbit of the itinerant d electrons and the local f moment of Gd that gives rise to the perpendicular anisotropy in GdFeCo. This latter term has many common features to the well known Rashba coupling which gives rise to dissipationless spin currents in time-dependent or inhomogeneous fields [7]. It is the smallest of interactions in the absence of the laser but it lowers the symmetry of the effective field seen by the d electrons and stores the memory of the chirality of the light after the laser is turned off. The enormous complexity of the reversal process in GdFeCo is only partly reflected in this Hamiltonian and since no sufficient real-time data are available to warrant detailed calculations based on this model, we present here a calculation based on a further idealized Hamiltonian that still has the necessary elements needed for a femtosecond reversal process. Our reversal process is based on ideas similar to the (mechanical) Barnett effect [8] which says that an external torque applied to an iron rod affects its magnetization by acquiring a component along the axis of the torque. The torque in our case will be
provided by the electric field of the laser as it acts on the hot d electrons. However the magnetic field of the laser also provides a similar Barnett contribution through the time-dependent Zeeman interaction with the d and f moments. The phonons through the spin-orbit coupling and the dipole terms provide the dissipation required for the reversal process. Polarization in the presence of rotating fields and dissipation has long been recognized in gravitational physics [9], accelerator physics [10] and in semiconductors [11]. Therefore there is no surprise that our model will give rise to polarization induced by a circularly polarized laser. In order to give a clear discussion of the optical Barnett-like effect, we introduce the effective Hamiltonian, \( H^{\text{eff}} \), for the spin degrees of freedom deduced from Eq. [2] and treat the orbital degrees of freedom and the effect of \( E_l \) on them as part of the environment. \( H^{\text{eff}} \) includes energy exchange between three different sub-systems (cf. Fig. [2] inset) and is

\[
H^{\text{eff}} = H_s + H_q + H_{sql} + H_Q
\]

(4)

The spin Hamiltonian, \( H_s = -\gamma H_l \cdot S - \frac{1}{2} A S_z^2 \), includes the interaction with \( H_l \) and an axial anisotropy term. The \( S \)'s represent the effective spin degrees of freedom in the GdFeCo system. The anisotropy, which is most relevant after the laser is turned off, is taken in the mean field approximation. The gyromagnetic ratio \( \gamma \) is assumed positive. The Hamiltonian, \( H_q = \frac{p^2}{2} + \omega_0^2 q^2 \), represents a single optical electronic mode with frequency \( \omega_0 \) close to the laser frequency \( \omega_l \) [12]. The Hamiltonian \( H_Q \) is that of the macroscopic bath as defined in [13]. The interaction Hamiltonian \( H_{sql} = -\lambda S \cdot q - q \cdot Q \), includes an effective linear coupling of the spin to the optical mode through spin-orbit coupling, which is taken isotropic for simplicity, and a linear coupling of the optical mode to the phonons \( Q \). The material dependent parameter \( \lambda = \lambda_0 + \lambda_1 E_l^2 / E_c^2 \) is an effective spin-orbit contribution with \( \lambda_0 \approx 0.001 \) eV, \( \lambda_1 \approx 0.1 \) eV and \( E_c \) is the critical field for switching which is taken from experiment. In the presence of the laser, an electron acquires additional orbital angular momentum \( e^2 E_l^2 / m \omega_l^3 \) and hence the power dependence of \( \lambda \). As first pointed out in Ref. [14], a strong spin-orbit coupling in NiO is needed to provide a fast relaxation channel for the spin of the electrons in the femtosecond regime.

This effective Hamiltonian is the simplest model that describes well the femtosecond reversal and reproduces LLG close to equilibrium if \( \omega_0 \) is much larger than the frequency of the spin [13]. In the present case, \( \omega_0 \) is very close to that of the laser-driven spin and consequently one of the main assumptions underlying LLG, the separation of energy scales
FIG. 2: (a): Dynamics of the perpendicular component of the magnetization $M_z$ induced by circularly polarized light $(E_l, H_l)$ with positive frequency $\omega_l = 10^{15}$ Hz and various intensities $H_l/H_B = 0.00001$, 0.0005, 0.001, and 0.002. The initial magnetization is $M_z(0) = -1$. (b): The dependence of switching on the chirality and initial conditions $M_z = \pm 1$ for $H_l/H_B = 0.001$. All results are for $\omega_0 = 0.8 \omega_l$, $\Gamma = 0.2$, and $A = 10^4$ Oe. $H_l = 10^{-3} H_B$ corresponds to a power of $\approx 5 \cdot 10^{11}$ W/cm$^2$.

between the spin and the environment, is broken[16]. Hence we expect non-adiabatic behavior to be important in this system. We will show that a careful treatment of the dynamics give non-exponential switching different than the one expected from the Bloch equations[5] or the LLG equation. This is the physics we would like to address in our analysis of the switching since similar conditions are also present in other important problems such as those in quantum computation where it may be necessary to go beyond the Born approximation to study coherence[3].

The coupled spin-laser-bath system is better studied in a frame $(x_1, x_2, x_3 = z)$ rotating around the $z$–axis with frequency $\omega_l$. In this frame, the Larmor torque is time-independent and the spin Hamiltonian becomes

$$
\mathcal{H}' = -\frac{\gamma H_l}{2} M_1 - \lambda \sum_i M_i b_i(t) - \frac{\gamma}{2} (B_0 + H_B) M_3
$$

where $B_0 = A \langle S_z \rangle$. The equations of motion for $\langle \mathbf{M} \rangle$, the average of the spin operator, are

$$
\langle \dot{M}_1 \rangle = -\gamma (B_0 + H_B) \langle M_2 \rangle + 2\lambda (\langle M_2 b_3 \rangle - \langle M_3 b_2 \rangle)
$$

$$
\langle \dot{M}_2 \rangle = \gamma (B_0 + H_B) \langle M_1 \rangle - \gamma H_l \langle M_3 \rangle + 2\lambda (\langle M_3 b_1 \rangle - \langle M_1 b_3 \rangle)
$$

$$
\langle \dot{M}_3 \rangle = \gamma H_l \langle M_2 \rangle - 2\lambda (\langle M_2 b_1 \rangle - \langle M_1 b_2 \rangle),
$$
where \( b_i \)'s are the harmonic mode variables in the rotating frame. The equations of motion for \( q \) are

\[
\left( \frac{d^2}{dt^2} + \omega_0^2 \right) \langle q_i \rangle = \langle Q_i \rangle + \lambda \langle \sigma_i \rangle ,
\]

(7)

The bath \( Q \) is assumed Ohmic and is the source of dissipation in the mode \( q \) which we take to be \( \Gamma = 0.2 \). In the adiabatic limit and in the absence of anisotropy, the system spin plus bath, \( H_s + H_Q \), has been treated earlier, see e.g. Ref. [17]. Here we study the more complex and experimentally relevant case of non-adiabatic magnetization reversal where the inclusion of memory effects is necessary.

In the rotating frame, the instantaneous torque is modified by the Barnett field. For visible light with \( \omega_l \approx 10^{15} \text{ Hz} \), the optical Barnett field is as large as \( \approx 10^7 \text{ Oe} \). This is larger than most exchange fields and hence a single spin picture should be adequate even for the treatment of the interaction of a laser with a ferromagnet. Despite its tremendous magnitude, we find that this effective field does not induce femtosecond magnetization reversal unless three key requirements are met. First, the Barnett field has to be much larger than the laser field \( H_l \). Secondly, the magnetization has to be coupled through the electric field \( E_l \) to at least one optical mode \( q \) of energy \( \omega_0 \approx \gamma H_B \). Third, the damping of this mode due to its interaction with the macroscopic bath \( Q \) has to enable efficient energy transfer from the spins to the mode \( q \). All three requirements can be met by various combinations of the parameters, \( H_B, H_l, E_l, \omega_0 \) and \( \lambda \), but the range of each individual parameter depends on the particular choice of the others.

To solve equations 6 and 7 we need to calculate the average of the product of two operators \( \langle \sigma_i(t)q_j(t') \rangle \). For this we need the density matrix of the whole system or we may use the more useful functional formalism instead (for a detailed discussion of this method and its application to sd exchange in metals see [18] and references therein). The generating functional is

\[
Z [J_1, J_2] = \left\langle \int D\eta DpDq \exp \left( -i \int dt \left( H_s (\eta, t) + H_q (p, q) + H_{sqq} - J_1 \cdot q - J_2 \cdot \sigma) \right) \right) \right\rangle_Q
\]

(8)

where the spin variables are for simplicity assumed \( 1/2 \) and are expressed in terms of Grassmann variables, \( \sigma = -\frac{i}{2} \eta \times \eta \) [19]. This allows the use of Wick’s theorem in the path integral expansion and \( J_i \) are two virtual external sources. The problem then becomes very similar to that of a Fermi gas which was discussed with the aid of two point functions [4]. The
Before switching on the laser, the phonons and the magnetization are in equilibrium and are weakly coupled since the electrons are in their ground state with a quenched orbital angular momentum. After that, the spin is driven out of equilibrium by the laser and \( q \) is treated as a perturbation which responds linearly to any changes in the spin. As in the \( sd \) exchange problem [18], the equations of motion are non-local in time and integrating out the optical mode \( q \) gives rise to dissipation and fluctuations even at zero temperature.

Using a Runge-Kutta scheme, Eqs. 6 and 7 are solved self-consistently. In line with the discussion above, we find that switching occurs only for a limited range of mutually dependent parameters. The results in fig. 2 focus on the power dependence of the magnetic response for \( \omega_l = 10^{15} \text{ Hz}, \omega_0 = 0.8\omega_l, \Gamma = 0.2, \text{ and } A = 10^4 \text{ Oe.} \) While the response to laser fields \( H_l/H_B \leq 10^{-5} \) is negligible on a femtosecond time scale, it becomes significant for \( H_l/H_B = 5 \cdot 10^{-4} \) and ultrafast reversal is found at higher fields of \( 10^{-3}H_B \) and \( 2 \cdot 10^{-3}H_B \). At the highest fields, \( M_z \) shows an almost instantaneous reversal accompanied by strong oscillations. The period of these oscillations is governed mostly by \( H_B \) while their decay depends on \( \Gamma \). The reversal slows down with time and the approach to equilibrium depends on the coupling \( \lambda \) of the spins to the mode \( q \) which is proportional to the power. The curves in the inset show the strong dependence of the reversal on the chirality of the laser. It is also important to realize that the simple exponential behavior expected from the Bloch equation or the LLG equation for the decay of the \( z \)-component is no longer true in our model. Fig. 2 shows that exponential behavior is only recovered after the laser has been on for several periods. The initial response of the spin system is therefore highly non-adiabatic due to the simultaneous action of the laser and of the dissipation and fluctuations, which are also enhanced by the laser, due to the coupling of the hot electrons to the magnetization.

In summary, we have shown that circularly polarized light can induce femtosecond magnetization reversal, when optical electron-phonon modes with frequencies comparable to those of the light are made available by the electric field of the laser through spin-orbit coupling. We predict a fast non-exponential reversal that cannot be recovered from the modified Bloch equations or the LLG equations and should be observable in future time-resolved experiments. The switching does not require high temperatures and is insensitive to the anisotropy.
which makes the proposed mechanism very attractive to high density magnetic recording.

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