The magneto-optical Barnett effect and spin momentum transfer

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The interaction of polarized light with a spin in the presence of dissipation is shown to be equivalent to a spin transfer process that can cause switching. In plasmas, the spin transfer is dominated by a spin-spin exchange term while at lower energy densities it is dominated by an optical Barnett-like effect. This latter effect is used in conjunction with optical phonons to predict femtosecond magnetization reversal believed to be recently measured in GdCoFe thin films. Conventional approaches based on the Bloch and the Landau-Lifshitz equations do not reproduce this ultrafast switching.

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In 1908, Richardson proposed of what has come to be known as the Einstein-de Haas effect [1]. By conservation of angular momentum, a body which is free to rotate around an axis will indeed start rotating upon magnetization. A few years later, the inverse effect, that is magnetization by mechanical rotation, was proposed and observed by Barnett [2]. Barnett showed that such rotation is equivalent to a magnetic field directed along the axis of rotation and effectively contributes to the internal field in a magnet. The direction of magnetization in a body at rest was shown to differ from that in a rotating body. A similar effect was also recently observed in paramagnets subjected to rotating magnetic fields instead of mechanical rotation [3]. Rotating black holes also emit particles (Hawking radiation) with net polarization due to the angular momentum of the black hole and can also be understood as a manifestation of a gravitational Barnett effect [4].

In this paper, we show that there is an optical version of the Barnett effect that can lead to magnetization reversal in the femtosecond regime. By optical we mean that the laser imparts spin, rather than orbital momentum, to the magnetization. The optical Barnett effect is shown to act in similar fashion to a direct spin momentum transfer between the laser and the spin polarization of a plasma at high power.

We believe that the optical Barnett effect has already been observed in a recent experiment [5]. The experiment demonstrated that it is possible to switch the magnetization of a GdCoFe film with a circularly polarized, 40 fs, 800 nm laser pulse at relatively low power, of the order of 1011 W/cm2. Using a single spin picture, we show rigorously that the optical Barnett effect in combination with coupling to an appropriate bath, can provide a consistent explanation of the observed femtosecond switching result. Since in this case a real understanding of the short time response of the magnetization to the laser is crucial, we refrain from using the phenomenological modified Bloch equations (MB) [2] or the Landau-Lifshitz (LL) [5] equation but instead use a self consistent formulation for the spin and its environment.

First we provide a motivation for the Hamiltonian that will be adopted for the interaction of the laser with the electronic degrees of freedom. Starting from the Dirac equation, Foldy and Wouthuysen [6] used a series of canonical transformations to derive the Pauli equation and other terms of higher order in inverse mass. To order 1/m02, the Hamiltonian is given by

\[
\mathcal{H} = \frac{1}{2m_0} \left( \mathbf{p} - \frac{e}{c} \mathbf{A} \right)^2 - e\Phi + \frac{e\hbar}{2m_0c} \mathbf{B} \cdot \mathbf{\sigma} 
\]

(1)

\[
+ \frac{e\hbar}{8m_0^2c^2} \left\{ \mathbf{p} \cdot (\mathbf{\sigma} \times \mathbf{E}) + (\mathbf{\sigma} \times \mathbf{E}) \cdot \mathbf{p} \right\} 
\]

\[
+ \frac{e^2\hbar}{4m_0^2c^3} \mathbf{E} \times \mathbf{\sigma} + \frac{e\hbar}{8m_0^2c^2} \nabla \cdot \mathbf{E} 
\]

This Hamiltonian can also be derived using the gauge-invariant proper-time method [7]. The terms in this expansion are well known. The third and fourth are the Zeeman and spin-orbit coupling energies. The fifth, less well known, term is a Heisenberg exchange-like interaction between the spin of the laser pulse and that of the electron. This term becomes important at laser powers of 1015 W/cm2 and above.

A polarized laser pulse carries angular momentum [8], but more importantly it also carries spin angular momentum. The total angular momentum \( \mathbf{J} \) of the laser beam occupying volume \( V \) is proportional to the momentum \( \mathbf{E} \times \mathbf{B} \) of the wave and given by (cgs)

\[
\mathbf{J} = \frac{1}{4\pi c} \int_V d^3r \quad \mathbf{r} \times (\mathbf{E} \times \mathbf{B}) .
\]

(2)

This vector can be decomposed into two parts according to \( \mathbf{J} = \mathbf{J} + \mathbf{S} \), with \( \mathbf{L} = 1/4\pi c \int_V d^3r \mathbf{E} \cdot (\mathbf{r} \times \nabla) \mathbf{A} \) being the orbital contribution, while the spin contribution is

\[
\mathbf{S} = \frac{1}{4\pi c} \int_V d^3r \quad \mathbf{E} \times \mathbf{A} .
\]

(3)

For circularly polarized light, with polarization vectors \( \mathbf{e}_\pm = 1/\sqrt{2}(\mathbf{x} \pm iy) \), and a vector potential \( \mathbf{A}(t,\mathbf{r}) = \int d^3k/(2\pi)^3 \{\mathbf{e}_{k\pm} a_{\pm}(k) \exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)] + c.c. \}, \) the spin angular momentum can be written as

\[
\mathbf{S} = \frac{1}{2\pi c} \int \frac{d^3k}{(2\pi)^3} \mathbf{k} \left[ |a_+(k)|^2 - |a_-(k)|^2 \right] .
\]

(4)
For a laser with $B_l = (B_l \cos \omega_l t, B_l \sin \omega_l t, 0)$, $a_-(k) = 0$ and $a_+(k) = 0$ correspond to positive and negative frequencies $\omega_l$, respectively. This form clearly shows the chirality of the laser wave and most importantly that $S$ is parallel to the $k$-vector which is perpendicular to the $B$ field. For completeness, we also point out that for beams with a finite cross section, the laser will have an additional component of $B$ in the direction of propagation of the wave which is proportional to the gradient of the transverse components [9]. In the following we will ignore this last contribution since it is important only at the edges of the beam. Moreover the orbital degrees of freedom will be considered as part of the environment.

In order to give a clear discussion of the optical Barnett effect, we introduce an effective Hamiltonian, $H^{\text{eff}}$, for the spin degrees of freedom derived from Eq. (1). $H^{\text{eff}}$ includes energy exchange between three different subsystems (cf. Fig. 1 inset) and is ($\hbar = 1$)

$$H^{\text{eff}} = H_s + H_q + H_{\text{qsl}} + H_Q$$  \hspace{1cm} (5)

The spin Hamiltonian, $H_s = -\gamma B_1 \cdot \sigma - \frac{1}{2} \alpha \sigma^2_z$, includes the interaction with the laser field $B_1$ and an axial anisotropy term. The $\sigma$’s are Pauli spin matrices for spin 1/2. The anisotropy term is taken in the mean field approximation to avoid having it trivial for spin 1/2 fields. It is only relevant after the the laser source is off. The gyromagnetic ratio $\gamma$ is assumed positive. The Hamiltonian, $H_q = \frac{Q^2}{2} + \frac{\omega_q^2}{2} q^2$, represents a single optical phonon mode with energy $\omega_q$ [10]. The Hamiltonian $H_Q$ is that of the macroscopic bath [11]. The interaction Hamiltonian $H_{\text{qsl}} = -\frac{1}{2} \mathbf{z} \cdot \sigma - \lambda(E) \sigma \cdot q - q \cdot \mathbf{Q}$, includes an exchange term and a linear coupling to the optical phonon mode. The effect of the electric field on the mode $q$ is implicit in this interaction. The coupling constant in the exchange term is

$$J = \frac{\pi e^2}{m^2 c^2}.$$  \hspace{1cm} (6)

This constant is very small but since $|S|$ is proportional to the power, $JS$ gives rise to fields $\geq 1$ Tesla for powers $\geq 10^{15}$ W/cm$^2$. While this exchange is negligible for the experimental conditions in Ref. [8], it becomes important in plasmas [12]. The interaction between the spin and the bath is mediated by the spin-orbit coupling. The material-dependent parameter $\lambda(E)$ is proportional to the power of the laser which is responsible for the excitation of the optical modes [10]. As first pointed out in Ref. [12], a strong spin-orbit coupling can provide a fast relaxation channel for the spin of the electrons in the femtosecond regime.

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

$$H' = -\frac{\gamma B_1}{2} M_1 - \lambda \sum_i M_i b_i(t) - \frac{\gamma}{2} (B_0 + H_B) M_3$$  \hspace{1cm} (7)

where $H_B = \omega_l/\gamma z$ will be called the Barnett field and $B_0 = A \langle \sigma_z \rangle$ + $JS_z$. The equations of motion for $\langle M \rangle$, the average of the spin operator, are

$$\langle M_1 \rangle = -\gamma (B_0 + H_B) \langle M_2 \rangle + 2 \lambda (\langle M_2 b_3 \rangle - \langle M_3 b_2 \rangle)$$ \hspace{1cm} (8)

$$\langle M_2 \rangle = \gamma (B_0 + H_B) \langle M_1 \rangle - \gamma B_1 \langle M_3 \rangle + 2 \lambda (\langle M_3 b_1 \rangle - \langle M_1 b_3 \rangle)$$

$$\langle M_3 \rangle = \gamma B_1 \langle M_2 \rangle - 2 \lambda (\langle M_2 b_1 \rangle - \langle M_1 b_2 \rangle),$$

where $b_i$’s are the harmonic mode variables in the rotating frame. The equations of motion for $q$ are

$$\left( \frac{d^2}{dt^2} + \omega_q^2 \right) q_i = \langle Q_i \rangle + \lambda(E) \langle \sigma_i \rangle.$$  \hspace{1cm} (9)

The equations for the average $q_i$ are solved by treating the spin source as a perturbation. The bath $Q$ is assumed Ohmic and is the source of dissipation in the mode $q$ which we take to be $\Gamma = 0.2$ using Shen and Bloembergen notation [10]. 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. [14]. Here we study the more difficult and experimentally relevant case of non-adiabatic magnetization reversal.

![FIG. 1: Dynamics of $M_z$, the component of the magnetization $M$ along the Barnett field $H_B$, induced by circularly polarized light with frequency $\omega_l$. For curves c1, c2, c3 and c4, $\omega_l = 10^{15} Hz$. For c3–, $\omega_l = -10^{15} Hz$. The laser field $B_1 = 10^{-3} H_B$ for c3 and $c3-$, $2 \times 10^{-3} H_B$ for c4, $10^{-5} H_B$ for c1, and $5 \times 10^{-4} H_B$ for $c2$. $M_z$ switches from +1(-1) to -1(-1) for positive (negative) $\omega_l$ as shown by the curves c3, c3–. The curve c3n shows no switching occurs for $\omega_l$ and $M_z(0)$ both positive. The different energy transfer channels involved in the excitation of the magnetization $M$ by the laser $\omega_l$ and its fast relaxation by the macroscopic bath $Q$ via an optical mode $q$ with energy $\omega_q \approx \omega_l$ are shown in the inset. Presented results are for $\omega_0 = 0.8 \omega_l$, $\Gamma = 0.2$, and $A = 10^4$ Oe.](image-url)
In the rotating frame, the instantaneous torque is modified by the Barnett field. For visible light with \( \omega \approx 10^{15} \text{Hz} \), 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, the Barnett 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. Secondly, the magnetization has to be coupled to at least one optical mode \( q \) of energy \( \omega_q \approx \gamma H_B \). 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, \( H_B, B_l, \omega_0 \) and \( \lambda(E) \), but the range of each individual parameter depends on the particular choice of the others.

Instead of going to the rotating frame, it is also possible to go to a frame which is rotating around the axis of the effective field \( H_{\text{eff}} = B_l + (H_B + B_0) \hat{z} \). In this case the transformation is explicitly dependent on the Barnett field and is given by

\[
U = U_y(\theta) U_z(\omega_s) U_y^{-1}(\theta) U_z(\omega_l),
\]

with \( U_\alpha(\epsilon) = \exp(-i\epsilon \sigma_\alpha / 2) \), \( \omega_s = \gamma H_{\text{eff}} \) and \( \theta = \tan^{-1}\left(\frac{B_l}{B_0 + H_B}\right) \). It can be shown that this transformation \( U \) is a spin gauge transformation of the full \( U(1) \times SU(2) \) symmetry of the Hamiltonian in Eq. \( 10 \).

To solve equations \( 8 \) and \( 9 \) we need to calculate the average of the product of two operators \( \langle \sigma_i(t) \sigma_j(t') \rangle \). For this we need the density matrix of the whole system or we may use instead the more powerful functional formalism (for a detailed discussion of this method and its application to \( sd \) exchange in metals see \( 17 \) and references therein). The generating functional is

\[
Z[J_1, J_2] = \left\langle \int D\eta Dp Dq \exp \left( -i \int dt \left( \mathcal{H}_s(\eta, t) \right) \right) \right. \\
\left. + \mathcal{H}_q(p, q) + \mathcal{H}_{\text{sd}}(J_1, q - J_2, \sigma) \right\rangle_Q
\]

where the spin variables are written in terms of Grassmann variables, \( \sigma = -\frac{i}{2} \eta \times \eta \) which allows the use of Wick’s theorem in the path integral expansion and \( J_i \) are two virtual external sources. The average values are found in the usual way \( 17, 18 \)

\[
\frac{\delta^2 \ln Z}{\delta J_1 \delta J_2} \bigg|_{J_1 = J_2 = 0} = -\left\langle q^i \sigma^j \right\rangle + \left\langle q^i \right\rangle \left\langle \sigma^j \right\rangle.
\]

Before switching on the laser, the phonon is assumed to be in equilibrium and is not coupled to the spins. 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 \( 17 \), 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. \( 8 \) and \( 9 \) 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. \( 4 \) focus on the power dependence of the magnetic response for \( \omega_l = 10^{15} \text{Hz} \), \( \omega_0 = 0.8 \omega_l \), \( \Gamma = 0.2 \), and \( A = 10^4 \text{Oe} \). While the response to laser fields \( B_l \leq 10^{-5} H_B \) is negligible on a femtosecond time scale (c1), it becomes significant for \( 5 \times 10^{-4} H_B \) (c2) and ultrafast reversal is found at higher fields of \( 10^{-3} H_B \) and \( 2 \times 10^{-3} H_B \) (c3 and c4, respectively). 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 \( \lambda(E) \) of the spins to the mode \( q \) which is proportional to the power. The curves c3- and c3n show the strong dependence of the reversal on the chirality of the laser.

**FIG. 2:** Switching paths for our theory (solid curve), the corresponding modified Bloch equation MB, MB2 for \( 1/|\omega_l|T = 0.06 \), and the Landau-Lifshitz (LL) equation for \( \alpha = 0.25 \). Zero anisotropy has been assumed for MB, MB2 and LL and all other parameters are the same as in Fig. 4c3. Without anisotropy, the MB equations lead to the trivial solution \( M = 0 \). Dynamics of the transverse x-component of the magnetization as predicted by our theory (dashed line), MB2, and LL are shown in the inset.

Next we compare our results to those of the modified Bloch equations (MB, MB2)

\[
\frac{dM}{dt} = \gamma M \times (H + H_B) - \frac{1}{T} (M - \chi (H + H_B)),
\]

where \( \chi \) is the magnetic susceptibility.
and the (modified) Landau-Lifshitz equations (LL)

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times (\mathbf{H} + \mathbf{H}_B) - \alpha \gamma \mathbf{M} \times (\mathbf{M} \times (\mathbf{H} + \mathbf{H}_B)).$$

(14)

All equations are written in the rotating frame and will be solved without taking account of the anisotropy. The anisotropy is important only after the reversal of the magnetization and the laser is off. The common modified Bloch equations (MB) do not include the Barnett field in the relaxation term and have been successfully used to interpret low frequency free-induction decay experiments in paramagnets. However, as shown in Fig. 2, MB does not predict switching when the Barnett field is much larger than the laser intensity. In contrast, the modified Bloch equations with the Barnett field accounted for in the relaxation, MB2, agree better with our results.

The LL equations with the Barnett field in the relaxation also give rise to fast reversal (Fig. 2, discontinuous line). The choice of parameters are made such that all solutions cross the point S at the same time. In the lab frame, the LL equations, unlike MB, also give reversal but for much longer times. For $\alpha = 0.25$, a switching time of about 40 ps is found in the lab frame (not shown). It can be shown, with little effort, that the Larmor equation in the lab frame has a solution with a non-zero static $z$-component such that $M_z = \cos \theta$ (cf. Eq. 10).

The inset in figure 2 shows the relaxation of the transverse components of the magnetization. At short times both MB2 and LL equations fail to capture the real dynamics of the reversal process. The failure of LL equations to adequately describe the dynamics in our system clearly invalidates the assumption of the universality of the LL damping term as was claimed by Koopmans et al [10]. Note, that the LL relaxation term is also invalid at short times when the relaxation is caused by momentum relaxation of the conduction electrons [17].

Laser powers of the order of $10^{15} \text{ W/cm}^2$ are relevant only to plasmas since most solids are ionized by such intense radiation [20]. In this case, we have to abandon the identification of the mode $q$ as that of an optical phonon mode and simply assume that it is a term due to collisions between the plasma particles. At these intensities, the spin exchange term $S_2 \sigma_z \sigma_z$ starts to become comparable to the Barnett field. For powers in the range $10^{17} - 10^{20} \text{ W/cm}^2$, full reversal of the polarization of the magnetized plasma is not possible since the Zeeman energy term grows much faster than the exchange term. At powers higher than $10^{20} \text{ W/cm}^2$, the exchange term dominates all other terms and full polarization of the plasma along the $z$-axis becomes again possible.

Figure 3 shows that in the laser configuration studied here ($k \parallel z$), the Barnett effect behaves similar to the spin momentum exchange term. The optical Barnett effect is therefore a spin momentum transfer effect and is not orbital in character as in the original experiment of Barnett since it does not require charged particles. This can also be easily seen if we write the energy, $\mathcal{H}_l = B_l^2/4\pi$, of the laser wave as a Zeeman term, $\mathcal{H}_l = 1/\gamma V \mathbf{H}_B \cdot \mathbf{J}$. The energy of the laser is not modified in the system studied here. Hence, the laser plays the role of a second bath that is strongly coupled to the spins. The spin momentum is transferred from the laser to the spin through the Barnett field as can be seen from the last term in Eq. 13.

In summary, we have shown that circularly polarized light can induce femtosecond magnetization reversal, via spin momentum transfer, when optical phonon modes with frequencies comparable to those of the light are present. This fast reversal can be recovered qualitatively from MB and LL equations only if their relaxation terms are modified to account for the Barnett field. The switching is found to be insensitive to the anisotropy which makes the proposed mechanism very attractive to high density magnetic recording. In high energy plasmas, an additional spin exchange transfer term between the laser and the polarization becomes dominant and can be used to control the induced magnetization.
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