Laser-Induced Ultrafast Demagnetization in Ferromagnetic Metals

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The laser-induced femtosecond demagnetization in ferromagnetic metals is investigated theoretically. Different from the conventional nanosecond one, this ultrafast demagnetization is a cooperative effect of the external laser field and the internal spin-orbit coupling. The spin-orbit coupling smears out the original identities of triplets and singlets while the laser field uses it as an avenue to influence demagnetization. Importantly, this demagnetization can be manipulated controllably, an essential point to future applications, such as ultrafast control of magneto-optical gating. Finally, the polarization filter effect on the ultrafast time scale is discussed based upon the present theoretical results.

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Thermally and magnetically driven demagnetization in ferromagnetic metals occurs on a nanosecond time scale [1]. This time scale is basically set by the spin-lattice, magnetic dipole, and Zeeman interactions and has long been considered as a speed limit for magneto-optical technology. However, this limit is now challenged by recent experimental observations [2–4]. Using pump-probe magneto-optics, Beaurepaire *et al.* [2] first demonstrated the laser-induced demagnetization in a ferromagnetic Ni film within 1–2 ps. This finding has greatly motivated intensive experimental [3,4] as well as theoretical [5] investigations. The very latest experiment shows a decay of the magneto-optical signal already within 50 fs [6], 3 orders of magnitude shorter than any conventional demagnetization process. This cannot be easily explained within the existing theory since none of the above interactions plays a major role within such a short time. Indeed, the standard spin-wave theory addresses a quasistatic process activated by the thermal or magnetic fields [7], rather than a demagnetization triggered by the laser field on a femtosecond time scale. We believe that understanding this novel phenomenon is not only of great theoretical importance but also of considerable technological significance.

In this Letter, we will demonstrate that the laser-induced demagnetization is genuine and is a cooperative effect of the external laser field and the internal spin-orbit coupling (SOC). In the absence of spin-orbit coupling, the laser field cannot effectively change the magnetic spin moment on the fs time scale; on the other hand, without the laser field, the spin-orbit coupling can change the magnetic spin moment only by a few percent. Only when both SOC and the laser field act on the system can such a novel demagnetization occur obviously: *a laser-induced demagnetization via spin-orbit coupling*. Importantly, such a demagnetization can be tuned externally, e.g., by the variation of laser intensity or pump-pulse sequence, which is indispensable to applications.

We employ one monolayer of ferromagnetic Ni as an example. A generic set of Ni parameters is used [5]. The Hamiltonian consists of two parts: $H_{\rm sys}$ for the system and $H_{\rm ext}$ for the laser field, $H = H_{\rm sys} + H_{\rm ext}$, where $H_{\rm sys} = \sum_{i,j,k,l,\sigma,\sigma',\sigma'',\sigma'''} U_{i\sigma,j\sigma',l\sigma''',k\sigma''} c_{i\sigma}^{\dagger} c_{j\sigma'}^{\dagger} c_{k\sigma''} c_{l\sigma'''} + \sum_{\nu,\sigma,K} \mathcal{E}_{\nu}(K) n_{\nu\sigma}(K) + H_{\rm SO}$. Here $U_{i\sigma,j\sigma',l\sigma''',k\sigma''}$ is the on-site electron interaction with the orbital indices i, j, k, l and spin indices $\sigma, \sigma', \sigma'', \sigma'''$ [8]. c ($c^{\dagger}$) is the electron annihilation (creation) operator. The last two terms in $H_{\rm sys}$ are the band structure [9] and spin-orbit coupling [5], respectively. $H_{\rm ext} = \tilde{E}(t) \cdot D$ [10], where $\tilde{E}(t)$ is the electric field [11], $\tilde{E}(t) = E_1(t) + E_2(t - \tau)$, D is the dipole operator, and τ is the time delay between two laser pulses. $E_i(t) = A_i e^{-t^2/\Gamma_i^2} \cos(\omega_i t)$, where A_i is the amplitude, Γ_i is the temporal width, and ω_i is the incident laser frequency of pulse i.

This is a typical time-dependent problem and the system evolves according to the Schrödinger equation [12],

$$i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = H|\Psi(t)\rangle, \tag{1}$$

where $\hbar$ is the Planck constant over 2π. Its solution can be formally written as

$$|\Psi(t)\rangle = \overrightarrow{T} e^{(1/i\hbar)\int_{t_0}^{t} H(t')\,dt'} |\Psi(t_0)\rangle, \tag{2}$$

where $|\Psi(t_{(0)})\rangle$ is the electronic state at time $t_{(0)}$ and $\overrightarrow{T}$ is the time-ordering operator [12] acting on the right term. In real calculations, we solve Eq. (1) numerically. In order to quantify our results, we introduce a new quantity, the transient magnetic moment $M(t)$ with the definition as $M(t) = \langle\Psi(t)|S_z|\Psi(t)\rangle = \langle\Psi(t_0)| e^{-(1/i\hbar)\int_{t_0}^{t} H(t')\,dt'} \overleftarrow{T} S_z \times \overrightarrow{T} e^{(1/i\hbar)\int_{t_0}^{t} H(t')\,dt'} |\Psi(t_0)\rangle$ where S_z is the z component of the spin operator. Without going into too much detail at this stage, we already can make some comments on the demagnetization process. From the Hamiltonian, we know that in the absence of SOC the total S_z commutes with the total Hamiltonian, $[S_z, H] = 0$ [10]. Consequently, we have $M(t) = \langle\Psi(t_0)|S_z|\Psi(t_0)\rangle = M(0)$, independent of time. This means physically that, without SOC, there is no clear spin relaxation for our Hamiltonian, irrespective of the presence of a laser field [10]. This observation is certainly true in our case. As a check, we turn off the spin-orbit coupling while keeping the laser field. Doing so, we truly find that even with the laser field [of intensity $I = 0.3$ (a.u.)] [13], $M(t)$ (a.u.) is precisely constant (see the upper curve in Fig. 1).

Complementarily, we next turn off the laser field while keeping SOC. Thus now only the SOC is responsible for the dynamics. The initial excited states are prepared to be a Gaussian distribution of energy. In Fig. 1, we plot $M(t)$ versus time t (see the lower curve). Interestingly, comparing with the upper curve, one can indeed see some variation of the magnetic moment due to the fact that the spin-orbit coupling couples the triplets and singlets, but the relative change is rather small, only about 2%–5%. This tells us that SOC alone is not sufficient to yield a clear reduction of the magnetic moment. As one will see later, without laser field the demagnetization is roughly proportional to the SOC constant λ, which explains why the change of magnetization is so tiny.

The whole scenario changes drastically if both laser field and spin-orbit coupling act on the system. We choose the pulse shape as a Gaussian function with the temporal width $\Gamma = 10$ fs and the photon energy $\hbar\omega = 2$ eV. For the moment, only one laser pulse ($A_1 \neq 0, A_2 = 0$) is employed at $t = 0$ fs. We start with a very weak laser intensity $I = 0.03$ (see the long-dashed line in Fig. 2). One notices that after the laser is turned on the magnetic moment is reduced, with the total variation of about 4%. The present laser field is too weak to greatly alter the system. However, if we enhance the intensity to $I = 0.3$ (see the dot-dashed line), a very large reduction of the magnetic moment, more than 40%, is accomplished *on a time scale shorter than 20 fs*. With a higher intensity, the reduction becomes even much stronger (see the solid line with $I = 1.5$). This is consistent with the experimental observation. In the pump-probe second-harmonic generation (SHG) experiment, Hohlfeld *et al.* [3] clearly showed that the SHG signal is reduced with the increase of the laser intensities. However, from these experiments, it is difficult to know definitely whether the magnetic moment has changed or not since there is no simple and direct connection between the magneto-optical SHG signal and the magnetic moment of the system [14]. In other words, the experiments provide only an indirect evidence. Here we directly prove that this is a genuine change of the magnetic moment. For $I = 1.5$, almost 50% reduction of the magnetic moment is reached. The explicit dependence of the magnetic moment on intensity is illustrated in the inset of Fig. 2. It can be fitted nicely to $M(I) = M_0 + M_1 e^{-aI}$. Here $M_0 = 0.46$, $M_1 = 0.6$, and $a = 12.2$. Because of the bleaching effect, a saturation appears for $I \geq 0.5$.

The next question is how to understand this ultrafast demagnetization. Let us first look at the many-body level scheme for a ferromagnet, as shown schematically in Fig. 3(a). Without SOC, the transitions are allowed only within the same spin sectors [see Fig. 3(a)], which means no change of the magnetization. With the presence of SOC, triplets mix with singlets. Their original identities are smeared out. The laser field then cooperatively uses this as an avenue to influence the magnetization since the originally forbidden transitions now become possible. For a ferromagnetic material, such as Ni, the ground state is magnetic and most of the large-spin states are close to

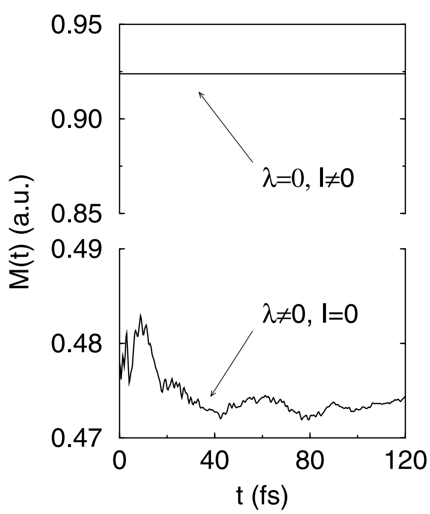

FIG. 1. Time dependence of magnetic moment. Without the spin-orbit coupling ($\lambda = 0$), there is *no* demagnetization, regardless of any existence of laser field (see upper panel) [9]; without the laser field ($I = 0$), the spin-orbit coupling only leads to a tiny effect (see lower panel).

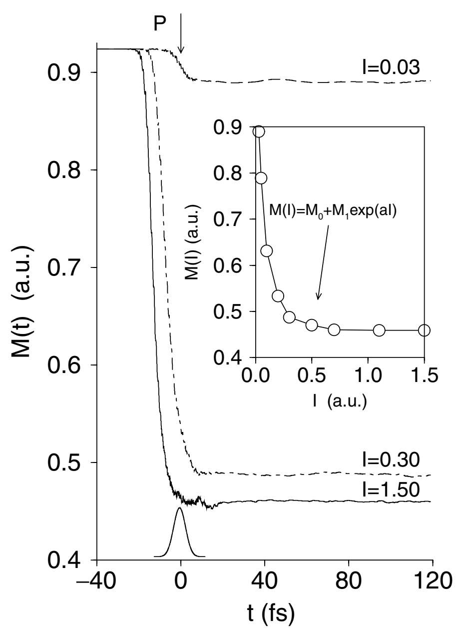

FIG. 2. With the presence of the spin-orbit coupling, the laser field can effectively influence the demagnetization. The intensity I (a.u.) is 0.03 (long-dashed line), 0.3 (dot-dashed line), and 1.50 (solid line). Inset: the exponential dependence of $M(I)$ on the laser intensity I.

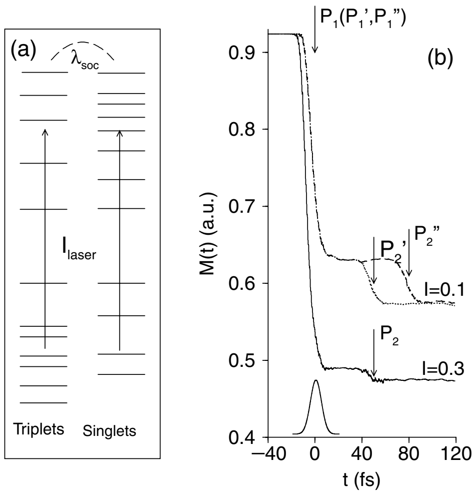

FIG. 3. (a) Many-body level scheme of a ferromagnetic metal. For the sake of clarity, the triplets and singlets are displaced horizontally. (b) Spin manipulation. By intensity variations, one can manipulate the drop of magnetization; by different delays, one can inscribe the information within different time intervals. A combination of them yields a larger flexibility to control spin. Solid line: P_1 and P_2 impinge at 0 and 50 fs, respectively, with intensity $I = 0.3$. Dotted line: P_1' and P_2' at 0 and 50 fs, but $I = 0.1$. Dashed line: P_1'' and P_2'' at 0 and 80 fs with $I = 0.1$.

the ground state while the small-spin states mainly appear in the high-energy window. Excitations from those low-lying states to high-lying ones lead to a pure reduction of magnetic moment while transitions between states of large (small) and large (small) spins roughly keep the original moment, which leads to the demagnetization. Thus, this novel demagnetization is a cooperative effect of SOC and laser field.

More insight into this new mechanism can be gained from a two-level system. The merit of such a system is that one can see directly how SOC and laser field cooperatively affect demagnetization. Assume that these two levels correspond to the ferromagnetic ground state $|\mathrm{gs}\rangle$ and the nonmagnetic excited state $|\mathrm{ex}\rangle$. Because of SOC, the pure triplet $|T\rangle$ admixes with the singlet $|S\rangle$. Thus the ground state and the excited state become $|\mathrm{gs}\rangle = \sqrt{1-\lambda^2}\,|T\rangle - \lambda|S\rangle$ and $|\mathrm{ex}\rangle = \lambda|T\rangle + \sqrt{1-\lambda^2}\,|S\rangle$, respectively, where the spin-orbit coupling λ is taken to be dimensionless. The magnetic moment for the ground state is reduced to $\mathcal{M} = (1-\lambda^2)M_0$, $M_0 = \langle T|S_z|T\rangle$. Upon the external perturbation, the whole system evolves into the state $|\Psi\rangle = \alpha|\mathrm{gs}\rangle + \beta|\mathrm{ex}\rangle$, where $\alpha^2 + \beta^2 = 1$, and β depends on the field strength F and the SOC λ. The magnetic moment of the system can be written as $M(t) = [\alpha^2(1-\lambda^2) + 2\alpha\beta\lambda\sqrt{1-\lambda^2} + \beta^2\lambda^2]M_0$. The relative change is $\delta M(t)/M_0 = -[1 + \cos 2(x+y)]/2$, with $x = \arcsin\alpha$ and $y = \arcsin\lambda$. It is easy to show $\delta M(t)/M_0 \le 0$. This yields a possible demagnetization process and agrees with the above numerical results.

Some special cases are of great interest. (i) Suppose that we switch off λ but keep the laser field, then $\beta = 0$ and $\alpha = 1$, and immediately we have $M(t) = M_0$. This is nothing but another manifestation of the above finding, no λ, no clear spin relaxation [10]. (ii) The field is off but the SOC on. $M(t)/M_0 \propto \lambda\sqrt{1-\lambda^2}\cos(\Delta t)$, $\Delta = (E_{\mathrm{ex}} - E_{\mathrm{gs}})/\hbar$. Thus, $M(t)$ oscillates periodically with time. Importantly, its amplitude is proportional to λ, which explains why, in the absence of laser field, the overall change of the magnetization is so tiny in the above numerical calculations of a real system. (iii) With both SOC and laser field on, the maximal reduction of $M(t)$ is achieved if $\alpha = -\lambda$, and $\beta = \sqrt{1-\lambda^2}$ [15]. Moreover, the large β implies a strong laser field. This can be seen more easily from the first-order perturbation theory (1PT) where β is proportional to $\lambda\sqrt{1-\lambda^2}\,F/\Delta$ and the required F is on the order of Δ/λ. Since usually the gap Δ is finite and λ very small, the field must be very strong. Naturally, a too strong laser strength is beyond the 1PT. Therefore, when we substitute β of the 1PT into $M(t)$, we obtain a linear rather than exponential reduction of the magnetic moment with intensity, which corresponds to the first few points in the inset of Fig. 2. As seen above, an exponential saturation of the magnetic moment appears if the laser intensity is strong enough, which goes beyond the perturbative treatment. Thus, both qualitatively and quantitatively, we can understand this novel ultrafast demagnetization.

Now we are in a position to investigate whether one can control such a demagnetization process. As seen above, even with a single laser beam ($A_1 \neq 0, A_2 = 0$), one can controllably reduce the magnetic moment by a suitable intensity. Here by two laser beams ($A_1 \neq 0, A_2 \neq 0$), we can tune the demagnetization not only in its magnitude but also in its temporal sequence. The results are shown in Fig. 3(b), where the arrows denote when the pulses are turned on. Both pump pulses have the same frequency $\omega = 2\ \mathrm{eV}/\hbar$ and the temporal width $\Gamma = 10$ fs. For the solid line in Fig. 3(b), both intensities are $I = 0.3$; the first pulse impinges at 0 fs, and the second one is delayed by $\tau = 50$ fs. One notices that after the first sharp drop of $M(t)$ upon the first pulse P_1, there is an additional reduction of the magnetization due to the second pulse P_2, but owing to the saturation effect, the net change is smaller than the first drop, only a few percent. We can enhance the second reduction by weakening the laser intensity. For instance, we reduce both intensities to $I = 0.1$ (see the dotted line with pulses P_1' and P_2'). Even with the same delay between two pulses, the second reduction becomes much more pronounced, about 20% of the first reduction. This means that one has more freedom to manipulate spins if the saturation is not reached. Actually, one can modify not only the relative reduction of the magnetization, but also the onset of the second reduction in the time domain. The latter can be done with different delays. For example, if one prolongs the delay of P_2'' with respect to P_1'' up to $\tau = 80$ fs, the second reduction starts around 70 fs [see the dashed line in Fig. 3(b)]. This gives us a chance to inscribe information into the medium at a selected time and realize temporal ultrafast writing. Of course, a fascinating thing would be to see whether the laser can also enhance the magnetization [16]. Work along this line is in progress.

Finally, it is also interesting to know whether one can observe a polarization filter effect on the ultrafast time scale. In the static limit, the conventional magneto-optical Kerr rotation is an evidence of such an effect in ferromagnets. For instance, circularly polarized light can be generated from an unpolarized input. In the dynamical regime, this is a new and yet open question, where the electron-electron correlation governs and strong dephasing effects dominate the whole process. Moreover, strong laser fluence leads to highly nonlinear excitations in the media, which may subsequently smear out the selection rules to some extent. Our model calculation implies that if the incident laser fluence is so weak that it still maintains the selection rule, the polarization effect may occur on a time scale longer than the electronic dephasing time. This presents an interesting experimental challenge though some experimental evidences have already indicated the existence of the effect [17].

In conclusion, we have demonstrated and explained the laser-induced ultrafast demagnetization process in ferromagnetic Ni. The underlying mechanism is qualitatively different from the conventional one. The conventional thermally or magnetically driven processes are quasistatic where the spin temperature is well defined, while the present one is on the femtosecond scale where the concept of temperature is questionable. This new demagnetization occurs as a cooperative process between the laser field and the spin-orbit coupling on the femtosecond time scale. Importantly, we have shown that one is able to controllably manipulate such a demagnetization process, which paves the way for future applications, such as ultrafast control of magneto-optical gating.

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[1] M. K. Kryder, J. Appl. Phys. **57**, 3913 (1985); A. Vaterlaus *et al.,* Phys. Rev. Lett. **67**, 3314 (1991).

[2] E. Beaurepaire *et al.,* Phys. Rev. Lett. **76**, 4250 (1996).

[3] J. Hohlfeld *et al.,* Phys. Rev. Lett. **78**, 4861 (1997); **79**, 960(E) (1997).

[4] M. Aeschlimann *et al.,* Phys. Rev. Lett. **79**, 5158 (1997); A. Scholl *et al.,* Phys. Rev. Lett. **79**, 5146 (1997); G. Ju *et al.,* Phys. Rev. Lett. **82**, 3705 (1999).

[5] W. Hübner and G. P. Zhang, Phys. Rev. B **58**, R5920 (1998); G. P. Zhang and W. Hübner, Appl. Phys. B **68**, 495 (1999); J. Appl. Phys. **85**, 5657 (1999); W. Hübner and G. P. Zhang, J. Magn. Magn. Mater. **189**, 101 (1998). It is important to realize that, due to the translational invariance in solids and the SOC, the angular momentum is not a good quantum number and these original orbital multiplets are not well defined, where those multiplets are replaced by bands. The direct consequence is that, even within the same band, the optical transition is allowed.

[6] J. Güdde *et al.,* Phys. Rev. B **59**, R6608 (1999). Although their time scale is already much shorter than those previously reported, these authors find that the magnetization breakdown is still limited by their pulse width (see the caption of their Fig. 4).

[7] Zhen-Zhong Li, *Solid State Theory* (Advanced Education Press, Beijing, 1985).

[8] We have considered up to three different configurations: $3d^{10}4s^0$, $3d^94s^1$, and $3d^84s^2$.

[9] Note that in Ni the optical absorption spectrum exhibits a big hump and no clear dominant transition can be seen [*Handbook of Optical Constants of Solids,* edited by E. D. Palik (Academic Press, Orlando, 1985)].

[10] Here we do not include the magnetic field since it plays a significant role only on a much longer time scale than that we consider here. For the dynamics on a longer time scale, there have already been some investigations [see W. Hübner and K. H. Bennemann, Phys. Rev. B **53**, 3422 (1996)].

[11] On the ultrafast time scale, the electric field polarization plays a minor effect as observed experimentally [see E. Beaurepaire *et al.,* Phys. Rev. B **58**, 12 134 (1998)]. The role of SOC is blurred by the memory effect caused by the many-body interactions.

[12] G. Baym, *Lectures on Quantum Mechanism* (W. A. Benjamin, Inc., New York, 1969).

[13] The absolute unit for intensity is $\mathrm{mJ/cm^2}$ with a factor of 0.16. The unit for magnetic moment is μ_B with a factor of 0.95.

[14] There is a controversy among experimentalists on whether the change of the susceptibility tensor elements in the pump-probe SHG experiments reflects the change of the magnetization [see Ref. [3]; H. Regensburger, R. Vollmer, and J. Kirschner, Phys. Rev. B **61**, 14 716 (2000); B. Koopmans *et al.,* Phys. Rev. Lett. **85**, 844 (2000)].

[15] Note that α is not exactly zero because the spin-orbit coupling already reduces the magnetic moment for the ground state as compared with the pure triplet $|T\rangle$.

[16] H. Petek *et al.* [Phys. Rev. Lett. **83**, 832 (1999)] recently reported a very important observation. It is certainly of great interest to see whether one could induce the magnetization in noble metals. Our mechanism is a good starting point to study it theoretically since it contains some basic features of $3d$ metals. In particular, in a different topic one recent experiment [Bonn *et al.,* Phys. Rev. B **61**, 1101 (2000)] demonstrates a larger flexibility to investigate the electron-phonon coupling and hot electron transport, which clearly supports our theoretical strategy.

[17] G. Ju *et al.,* Phys. Rev. B **57**, R700 (1998); P. F. Bennett *et al.,* Opt. Lett. **24**, 1373 (1999).