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Ultrafast spin-photon interaction investigated with coherent magneto-optics

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We investigate the interaction of femtosecond laser pulses with spins including relativistic corrections. The time-ordered magneto-optical signals corresponding to a pump-probe configuration are calculated in the case of one electron submitted to a magnetic field and evolving in eight levels of the fine structure of a hydrogenlike atom. Our simulations explain the origin of the coherent magneto-optical response and ultrafast spin dynamics in ferromagnets excited by intense laser pulses as recently reported in Ni and $CoPt_3$ ferromagnetic thin films. Our detailed analysis allows identifying the respective roles of the coherent spin-photon interaction and spin dynamics, unraveling recent controversies about the laser-induced ultrafast magnetization dynamics.

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Ferromagnetic thin films can be modified on a femtosecond time scale by ultrashort laser pulses.[1–5] This observed phenomenon, as well as the nonthermal optical control of magnetic order with light pulses,[6] is important as it promises major applications in the fields of data storage and time-resolved magnetic imaging, for example.[7] Understanding the underlying physical mechanisms requires a formal description of the laser-induced ultrafast magnetization dynamics which involves the multiple possible interactions between the photons, electrons, the spins, and the lattice, together with the complexity of the electronic band structures of the magnetic materials.

Several models have been proposed to describe the demagnetization process occurring in the subpicosecond time scale. The first one involves three interacting baths at different temperatures corresponding to the charges, the spins, and the lattice which are out of equilibrium.[8] A quantum model, including the effects of exchange and spin-orbit interactions, has accounted for spin flips in terms of their dephasing after they redistribute in the excited states.[9–11] An extension of this model including the combined interaction of the laser and spin orbit successfully described elementary spin flips or so-called lambda processes.[12] Modeling the spins dynamics at later times, a few picoseconds after laser excitation, has reached a consensus. In that case, the spin-phonon interaction prevails and is responsible for the damping of the precession of the magnetization in ferromagnets, the exception being made when approaching the Curie temperature for which long-range fluctuations maintain a nonequilibrium spin bath for a long time.[13,14] A revival of the debate regarding the origin of the ultrafast demagnetization occurred with the assumption that spin flips could be due to a mechanism similar to the Elliott-Yafet scattering of conduction electrons by magnetic impurities.[15,16] Some experiments do not support this model, for example, regarding the effect of magnetic impurities on the spin dynamics in doped ferromagnetic transition metals.[17]

More recently a new controversy has arisen regarding the interpretation of the time-resolved magneto-optical response as a signature of the magnetization dynamics.[18,19] The main origin of this controversy lies in the distinctions that one ought to make between the time-dependent response function (magneto-optical signal) and the system's dynamics (the spin populations). In the present Rapid Communication we show that there is a straightforward manner to clarify the debate by considering separately the coherent and population dynamics in the magneto-optical response. Indeed, it is known that in metals coherent magnetism is important, as shown in a recent detailed study of the charges and spin dynamics performed in Ni and $CoPt_3$ ferromagnetic films.[20]

Let us consider the simplest possible system that consists of eight quantum levels interacting with the laser field such that the interaction takes into account the relativistic corrections to the quantum electron dynamics, including the spin-orbit interaction. We determine the response function from the density matrix formalism including the time-ordered third-order nonlinear terms. This approach allows to understand the main differences between the coherences and spins populations. We consider a one-electron Hamiltonian with an effective Coulomb interaction perturbed by the spin-orbit and kinetic momentum-laser vector potential interactions. Using such a Hamiltonian, applied to the band structure of metals, Argyres has shown that static magneto-optical Kerr and Faraday effects in ferromagnetic materials mainly come from a spin-orbit interaction with the ionic field.[21] For the purpose of understanding the main steps of our approach, which primarily aims at determining the respective roles played by the coherent versus population dynamics in the response function (Faraday rotation), we consider the case of a simple discrete eight-level system representing a reduced hydrogenlike system.

The relativistic contributions to the ultrafast magneto-optical dynamics are considered via the Foldy-Wouthuysen transformation of the Dirac equation which enlightens the various interaction terms between spins and femtosecond laser fields. We add to Argyres' approach the terms of the Foldy-Wouthuysen transformation to second order in $\frac{1}{m}$ for the electron from a hydrogenlike atom submitted to different static fields: the ionic field $\mathbf{E}_i$ and its associated central ionic potential $V_i(\mathbf{r})$ and a strong static homogeneous magnetic field $B_M\mathbf{e}_z$ with potential vector $\mathbf{A}_M = -\frac{1}{2}\mathbf{R} \wedge \mathbf{B}_M$. The electron is interacting with a laser field described in the Coulomb gauge associated with an homogeneous electric field $\mathbf{E}_L$, with its collinear vector potential $\mathbf{A}_L$, and with its related magnetic field $\mathbf{B}_L$.

The Hamiltonian H_0 corresponding to no interaction with the laser reflects the Zeeman splitting in a strong magnetic

field. At this point the degeneracy of the spin states are already lifted and the effect of spin-orbit interaction is to slightly shift some energy levels:

$$H_0 = \frac{1}{2m}(\mathbf{p} - q\mathbf{A}_M)^2 + qV_i(\mathbf{r}) - \frac{q\hbar^2}{8m^2c^2}\nabla\cdot\mathbf{E}_i - \frac{q}{m}\mathbf{S}\cdot\mathbf{B}_M - \frac{q}{2m^2c^2}\mathbf{S}\cdot[\mathbf{E}_i\wedge(\mathbf{p}-q\mathbf{A}_M)]. \quad (1)$$

Neglecting the second-order terms in the laser vector potential, the interaction Hamiltonian is

$$H_{\text{int}} = -\frac{q}{m}\mathbf{\Pi}\cdot\mathbf{A}_L - \frac{q}{2m^2c^2}[(\mathbf{p}-q\mathbf{A}_M)\wedge\mathbf{S}]\cdot\mathbf{E}_L - \frac{q}{m}\mathbf{S}\cdot\mathbf{B}_L - \frac{\imath q\hbar}{4m^2c^2}\mathbf{S}\cdot(\nabla\wedge\mathbf{E}_L). \quad (2)$$

With the kinetic momentum operator $\mathbf{\Pi} = \frac{m}{\imath\hbar}[\mathbf{R}, H_0]$,

$$\mathbf{\Pi} = \mathbf{p} - q\mathbf{A}_M + \frac{q}{2mc^2}\mathbf{S}\wedge\mathbf{E}_i. \quad (3)$$

We apply the electric dipolar approximation to the interaction Hamiltonian and consider the $2s$ and $3p$ levels of the hydrogen atom; their energy difference without spin-orbit coupling and static magnetic field is associated to the frequency ω_0 as shown in Fig. 1. This energy is close to the typical photon energies used in experiments. We do not consider the magnetic dipolar interaction term, which is off resonant. The two first terms of Eq. (2) imply a variation of the projection of the orbital momentum along the quantization axes $\Delta l = \pm 1$, $\Delta l_z = \pm 1$, $\Delta s_z = 0$, and the terms containing the spin (H_α and H_β) involve spin-flip transitions with $\Delta l = \pm 1$, $\Delta l_z = 0$, $\Delta s_z = \pm 1$. Table I shows the orders of magnitude of each interaction matrix element compared to the predominant interaction term $\langle j|\frac{q}{m}\mathbf{p}\cdot\mathbf{A}_L|i\rangle$ for two states $|i\rangle$ and $\langle j|$ among the eight states sketched in Fig. 1. In the case of our hydrogenlike model, we will see that the contributions $H_\alpha = \frac{q^2}{2m^2c^2}[\mathbf{S}\wedge\mathbf{E}_i]\cdot\mathbf{A}_L$ and $H_\beta = \frac{q}{2m^2c^2}[\mathbf{p}\wedge\mathbf{S}]\cdot\mathbf{E}_L$ can be neglected in the magneto-optical response but not in the spin dynamics. It is not the purpose to discuss here their influence in the case of a ferromagnetic solid, where additional phenomena

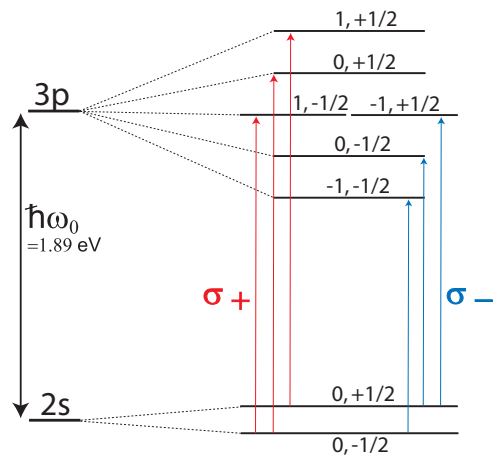

FIG. 1. (Color) Considered transitions in the hydrogenlike atom. For each level we indicate l_z,s_z. $\sigma_\pm$ stands for a circularly polarized field along $\mathbf{e}_x \pm \imath\mathbf{e}_y$.

TABLE I. Scale of the considered interaction matrix elements in hydrogen compared to the canonical momentum-laser vector potential interaction matrix element.

	$\langle j\|\frac{q}{m}p\cdot A_L\|i\rangle$	$\simeq$	1
	$\langle j\|\frac{q^2}{m}A_M\cdot A_L\|i\rangle$	$\simeq$	$3\times10^{-5}B_M$
$\langle j\|H_\alpha\|i\rangle =$	$\langle j\|\frac{q^2}{2m^2c^2}S\wedge E_i\cdot A_L\|i\rangle$	$\simeq$	1×10^{-6}
$\langle j\|H_\beta\|i\rangle =$	$\langle j\|\frac{q}{2m^2c^2}p\wedge S\cdot E_L\|i\rangle$	$\simeq$	9×10^{-7}

such as a dynamical anisotropy can be induced by the presence of an external field,[22] for example, the laser field in our case.

In order to model magneto-optical pump-probe experiments, the evolution of the system is calculated in the Liouville formalism to the third order of the laser perturbation.[23] The relaxation time of the coherences (ρ_{nm},$n\neq m$) is T_2 whereas it is T_1 for the population differences ($\rho_{nn}-\rho_{mm}$,$n\in\{3,8\}$;$m\in\{1,2\}$). For simplicity they are assumed to be the same for each transition. The electric field of the laser pulses is given by $\mathbf{E}_L(t) = \frac{1}{2}[\epsilon(t,\tau)e^{-\imath\omega_L t} + \epsilon^*(t,\tau)e^{\imath\omega_L t}]\mathbf{e}_x$, where $\epsilon(t,\tau)$ is a Gaussian centered on the pump-probe delay τ or 0 in the case of the probe or pump fields. The laser frequency ω_L is equal to or close to ω_0. The effective dipolar moment of the system $\mathbf{D}$ gives the first- and third-order polarizations of the atom $\mathbf{P}^{(1)}(t) = \text{Tr}(\rho^{(1)}\mathbf{D})$ and $\mathbf{P}^{(3)}(t,\tau) = \text{Tr}(\rho^{(3)}\mathbf{D})$. The polarization is calculated using the rotating wave approximation and depends on both the time t and delay τ between the pump and probe pulses. As we do not consider any propagation effect in this simple atomistic approach, we set arbitrarily that the radiated electric field at order (n) is $\mathbf{E}^{(n)} \equiv \mathbf{P}^{(n)}$. We consider pump and probe linearly polarized in the plane perpendicular to the quantization axes and calculate the dynamical magneto-optical rotation induced by the sample in the Jones formalism. In order to follow the common experimental procedure for magneto-optical measurements with a polarization bridge, we define $\mathbf{E}'^{(3)}(t,\tau)$ as the rotated $\mathbf{E}^{(3)}(t,\tau)$ due to a half-wave plate tilted by an angle of $\frac{\pi}{8}$ with respect to the ($\mathbf{e}_x$,$\mathbf{e}_y$) axes, and the measured differential intensities $\Delta I_{x,(y)}$ are

$$\Delta I_{x,(y)} = \int_{-\infty}^{+\infty} 2\,\text{Re}\left[\mathbf{E}'^{(3)}_{x,(y)}(t,\tau)\cdot\mathbf{E}'^{*(1)}_{x,(y)}(t)\right]dt. \quad (4)$$

The differential rotation for a positive magnetic field $\Delta\Theta_{+B_M}(\tau) = [\Delta I_x - \Delta I_y]_{+B_M}$ is linked to the first- and third-order field's amplitude and to their instantaneous rotations $\Theta^{(1)}_{+B_M}(t)$ and $\Theta^{(3)}_{+B_M}(t,\tau)$; here we assume the ellipticity to be negligible:

$$\Delta\Theta_{+B_M}(\tau) = \int_{-\infty}^{+\infty} \sin\left[\Theta^{(3)}_{+B_M}(t,\tau) + \Theta^{(1)}_{+B_M}(t)\right] \times 2\left[\left|\mathbf{E}^{(3)}_{x,(y)}(t,\tau)\right|\left|\mathbf{E}^{*(1)}_{x,(y)}(t)\right|\right]dt. \quad (5)$$

The resulting magneto-optical signal is then proportional to $\Delta\Theta(\tau) = \Delta\Theta_{+B_M}(\tau) - \Delta\Theta_{-B_M}(\tau)$. obtained for the two directions $\pm B_M$ of the magnetic field. As done experimentally, we perform a differentiation on the magnetic field and normalize the resulting rotation with the linear magneto-optical

rotation $\Theta_{\pm B_M}$:

$$\frac{\Delta\Theta(\tau)}{\Theta} = \frac{\Delta\Theta_{+B_M}(\tau) - \Delta\Theta_{-B_M}(\tau)}{\Theta_{+B_M} - \Theta_{-B_M}}. \quad (6)$$

Considering the time ordering of a single probe and two pump pulses, one can distinguish three terms corresponding to different coupling between pump, probe, and the polarization of the system as described in the case of charge dynamics by Brito-Cruz *et al.*.[25] First, the time ordering of the fields $\epsilon_{\text{pump}}(t=0)\epsilon^*_{\text{pump}}(t=0)\epsilon_{\text{probe}}(t=\tau)$ describes the "population dynamics" induced by the pump field $\epsilon_{\text{pump}}, \epsilon^*_{\text{pump}}$. It is maximal for positive pump-probe delays and relaxes with the population decay time T_1. The second term, referred to as "pump-polarization coupling" (PPC), is given by the sequence $\epsilon^*_{\text{pump}}(t=0)\epsilon_{\text{probe}}(t=\tau)\epsilon_{\text{pump}}(t=0)$, which corresponds to a convolution of the pump pulse with the exponential decay of the density matrix's coherences within the T_2 time. The third term $\epsilon_{\text{probe}}(t=\tau)\epsilon^*_{\text{pump}}(t=0)\epsilon_{\text{pump}}(t=0)$ is maximum at negative pump-probe delays and corresponds to the coherences generated by the probe which couple to the pump, sometimes also referred to as "pump-perturbed free-induction decay" (PP-FID). These two latter terms, also hereafter referred to as "coherent terms," are strongly related to the dephasing of the polarization as pump and probe can only couple if the density matrix's coherences are nonzero. Figure 2 shows the dynamical differential magneto-optical rotation calculated for each of these three terms using the interaction Hamiltonian in Eq. (2).

In the numerical simulations, we have considered Gaussian pulses of width 10 fs and a probe field ten times less intense than the one of the pump. The dephasing time of the coherences and the lifetime of the differences of population are chosen to be 10 and 100 fs. The static magnetic field is 1 T. The pump and probe laser fields are taken to be linearly polarized along $\mathbf{e}_x$. The unperturbed ground-level populations are 0.9 for $s_z = \mp\frac{1}{2}$ and 0.1 for $s_z = \pm\frac{1}{2}$ for a magnetic field $\pm B_M$. These results clearly show that the coherent terms are important in the magneto-optical signals and that their contributions with respect to the spin population dynamics depend on the laser detuning.

The next issue that we address now is the difference between the pump-probe magneto-optical signal and the spin and orbital momentum dynamics. Toward that purpose we determine two different quantities: the projection of the spin and the orbital momentum operators $\langle S_z^{(2)}\rangle$ and $\langle L_z^{(2)}\rangle$ along the quantification axis $\mathbf{e}_z$. Note that the dynamics is now represented by the trace of the operators multiplied by the second-order nonlinear terms of the density matrix as they are related to the density matrix's populations. The spin dynamics is due to the H_α and H_β spin-flip terms. Figure 3 shows their dynamics for the three time orderings of the pump and probe fields. The population terms [Figs. 3(a) and 3(b)] are represented as a function of time t as they do not depend on the probe field. The two coherent terms are integrated over t and represented as a function of pump-probe delay τ, Figs. 3(c) and 3(d) for PPC and Figs. 3(e) and 3(f) for PP-FID terms, as they explicitly depend on time-ordered sequences involving both the pump

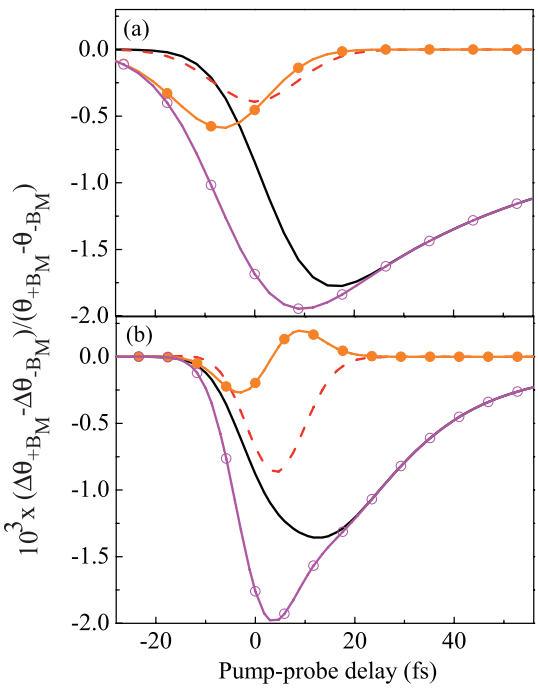

FIG. 2. (Color) Magneto-optical rotation for the population (black straight line), pump-polarization coupling (red dashed line), pump-perturbed free-induction decay (orange closed circle), and total (purple open circle) signals. (a) On resonance ($\hbar\omega_L = \hbar\omega_0 = 1.89$ eV) and (b) off resonance ($\hbar\omega_L = 1.51$ eV).

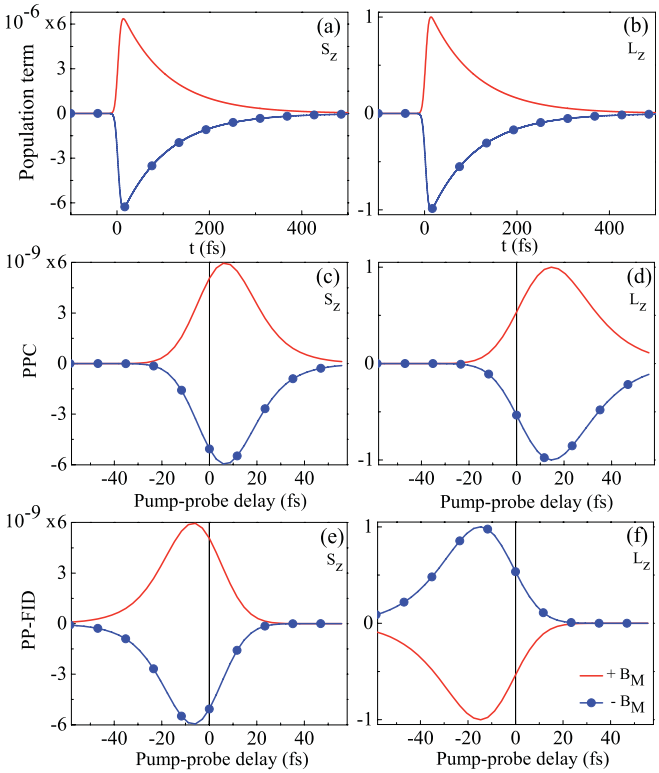

FIG. 3. (Color) Spin and orbital momentum dynamics for the three pump-probe field sequences at $+B_M$ (red straight line) and $-B_M$ (blue closed circle). (a) $\text{Tr}[\rho^{(2)}S_z(t)]$ and (b) $\text{Tr}[\rho^{(2)}L_z(t)]$ for the population term. (c) $\text{Re}\{\int dt\, \text{Tr}[\rho^{(2)}S_z(t,\tau)]\}$ and (d) $\text{Im}\{\int dt\, \text{Tr}[\rho^{(2)}L_z(t,\tau)]\}$ for the pump-polarization coupling term. (e) and (f) The same quantities as (c) and (d) but for the pump-perturbed free-induction decay term. (c)–(f) have the same normalization factor.

and probe pulses. $\langle S_z^{(2)} \rangle$ and $\langle L_z^{(2)} \rangle$ are, respectively, real and imaginary because they are generated by the pulses ϵ^*_{pump} and ϵ_{probe}. In order to calculate real quantities, one should also have considered ϵ_{pump} and $\epsilon^*_{\text{probe}}$, which does not correspond to the experimental configuration chosen here.

In conclusion, we have shown that in an ultrafast magneto-optical experiment the time ordering of the pulses has to be taken into account, especially in order to distinguish the coherent response from the populations dynamics. Out of resonance the magneto-optical coherent signal increases. More importantly, the spin and orbital momentum's dynamics, due to the second order in the perturbation of the density matrix, both participate in the coherent magneto-optical response. Extrapolating the present results to more complex magnetic systems shows that the coherent spin-photon interaction can be used to manipulate the magnetization of spin devices at the femtosecond time scale.

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