Coherent magnetism: pushing the limits of spin-photon interaction

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Abstract: We report on magneto-optical coherent dynamics measurements on a 9 fs time scale. We show that the laser field induces an anisotropy, driving the magnetization out of equilibrium. The corresponding mechanism is the spin-orbit interaction.

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1. Introduction

The question of how fast can one modify the magnetic state of magnetic materials is important in the context of controlling the recording process in magnetic media [1]. It has been previously shown that coherent magnetism can result from the strong spin-photon interaction, mediated by the spin-orbit coupling [2]. This finding has been deduced from magneto-optical Faraday measurements in Ni and CoPt ferromagnetic thin films using 50 fs pulse duration. Alternatively, it has been shown that the in-plane magnetization of Garnet films can be controlled optically, resulting in a motion of precession [3]. Its origin has been attributed to a magneto-electric effect that is a coherent mechanism corresponding to an induced anisotropy $\delta H_{\mathbf{a}}$ which modifies the effective field sensed by the material. Here, we explore the limits of this coherent spin-photon interaction by performing time resolved Magneto-Optical Four Wave Mixing (MO-FWM) in a ferrimagnetic Garnet film with 9 fs pulses. We propose a simple model based on the spin-orbit interaction coupled to the laser field which was previously used to explain the coherent Faraday measurements in metals [4].

2. 9 fs magneto optical four wave mixing experiment:

The experiment is sketched in the inset of Fig 1. It consists in a degenerate four wave mixing configuration performed at 800nm using 9 fs pulses with 80 MHz repetition rate. $\mathbf{k}_p$ (respectively $\mathbf{k}_s$) corresponds to the pump (respectively probe) wave vectors. Note that in such configuration both laser fields can have similar amplitudes. The sample is a 7 µm thick bismuth doped iron garnet (BIG) film and has been chosen for its strong spin orbit coupling, giving rise to a large magneto optical response in the visible and near IR. A static magnetic field $\mathbf{H}_0$ is applied perpendicular to the sample plane ($\phi = 0°$ or 180° being the angle between $\mathbf{H}_0$ and the normal to the sample). The magnetic signal is obtained from the difference between the measurements for opposite directions of $\mathbf{H}_0$.

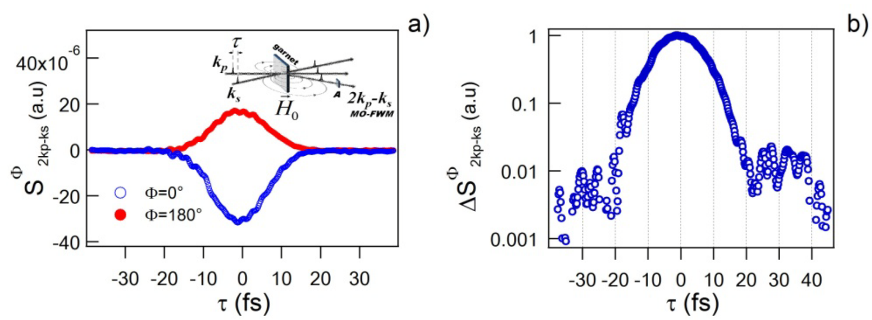

Fig. 1. a) Two beams MO-FWM signals for two opposite directions of $\mathbf{H}_0$ (ϕ = 0°blue opened circles and ϕ = 180° red closed circles). Inset: sketch of the two beams MO-FWM configuration. b) MO-FWM given by $\Delta S^{\phi}_{2kp-ks} = S^{180°}_{2kp-ks} - S^{0°}_{2kp-ks}$ in log scale and relaxation time T_2^{MO}.

The MO-FWM signal S^{ϕ}_{2kp-ks} shown on Fig 1.a) is obtained by analyzing the polarization state of the coherent four wave mixing emission in the $2\mathbf{k}_p$-$\mathbf{k}_s$ direction, using a polarization bridge. Note the opposite rotation for the complementary angles of $\mathbf{H}_0$. The differential signal $\Delta S^{\phi}_{2kp-ks} = S^{180°}_{2kp-ks} - \Delta S^{0°}_{2kp-ks}$ is shown on Fig 2.b). The coherent dynamics of the charges is expected to decay with a characteristic time T_2, corresponding to the dephasing time of charges with respect to the laser field. By analogy, the magnetic field dependent coherent emission (Fig 1.b) corresponds to a magneto optical dephasing timeT_2^{M0}. The asymmetry of the ΔS^{ϕ}_{2kp-ks} allows to retrieve T_2^{M0} = 2.8 fs assuming an inhomogeneously broadened medium, due to the multi-domain structure of Garnet on the spatial scale of the focused beams (~30 μm diameter). This is confirmed by a detailed analysis of the signals emitted in the two directions $2\mathbf{k}_p$-$\mathbf{k}_s$ and $2\mathbf{k}_s$-$\mathbf{k}_p$.

3. Microscopic origin of the Magneto-Optical Coherent Emission

To understand the origin of the coherent magneto optical dynamics, and distinguish it from effects associated to the population dynamics, like thermal equilibrium and magnetization precession, it is necessary to separate the transverse from longitudinal magneto-optical processes. It is best made using the density matrix formalism [5] and considering a simplified Hamiltonian. For the case of a single electron with charge *e* mass *m* momentum **p** and spin ***s*** excited in hydrogenic quantum states and interacting with the laser and magnetic fields, also taking into account the spin-orbit interaction, one has:

$$H_{\text{int}} = \frac{e}{m}\vec{\pi}_{\pm}\vec{A}_L \qquad \text{with} \quad \vec{\pi}_{\pm} = \left(\vec{p} + \frac{e}{c}\vec{A}_M + \vec{s}\times\vec{\nabla}V_{ion}\right)_{\pm}$$

Where $\mathbf{A}_L$ and $\mathbf{A}_M$ are the vector potentials of the laser electromagnetic field and of the magnetic field respectively; V_{ion} is the scalar potential of the ion. The spin-orbit coupling term $\vec{s}\times\vec{\nabla}V_{ion}$ plays a fundamental role in the optical control of the magnetic states. It is a relativistic term present in the quantum electrodynamics of the spin-photon interaction. It can be simply incorporated in the time dependent third order nonlinear response of a 8-level hydrogenoid model system. The corresponding third-order non linear polarization can be expressed in terms of the rotation θ and the ellipticity ε responses either in the Faraday or in the Four Wave Mixing configurations. It allows distinguishing clearly between the population and coherent contributions to the magneto optical signals as usually obtained for the charge dynamics. This is exemplified in Fig. 2 which shows the Faraday rotation obtained with laser pulses of 10 fs, resonant with the main transition. It can be decomposed into two coherent contributions, the pump polarization coupling and the pump-perturbed free induction decay having an electronic dephasing time T_2 = 10fs and a population contribution with a lifetime T_1 = 100 fs . While the coherent contribution has its counter part in the MO-FWM signal, the population is absent for the two-beam configuration. We will also discuss the role played by the coherent interaction terms on the induced electric field anisotropy that is observed in the precession of magnetization vector.

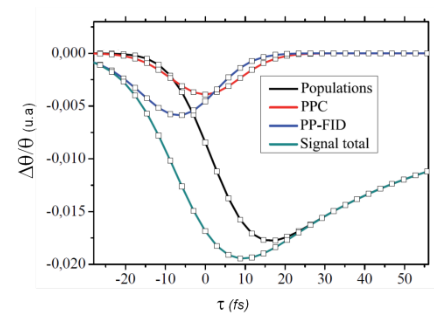

Fig. 2. Magneto-optical rotation. Population dynamics (black). Coherent dynamics: Pump polarization coupling (red) and pump perturbed free induction decay (blue).

In conclusion, the coherent MO-signals observed in transparent ferrimagnetic media reveal the importance of the spin-orbit interaction both for the dephasing dynamics of the charges and spins states at the femtosecond time scale and for the control of the precession at the time scale of tens of picoseconds.

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4. References

[1] J.-Y. Bigot, M. Vomir, *"Ultrafast magnetization dynamics of nanostructures"*, Ann. Phys. 1–29 (2012).
[2] J.-Y. Bigot, M. Vomir & E. Beaurepaire, "*Coherent ultrafast magnetism induced by femtosecond laser pulses*", Nat. Phys. **5**, 515–520 (2009).
[3] F. Hansteen, A. Kimel, A. Kirilyuk, Th. Rasing, "*Nonthermal ultrafast optical control of the magnetization in garnet films"* Phys. Rev. B, **73**, 014421 (2006).
[4] H. Vonesh and J.-Y. Bigot, "*Ultrafast spin-photon interaction investigated with coherent magneto-optics"*, Phys. Rev. B, **85**, 180407(R), (2013).
[5] S. Mukamel, "*Principles of Nonlinear Optical Spectroscopy*", Oxford Series on Optical and Imaging Sciences, Oxford University Press (1999).
[6] J.-Y. Bigot "*Femtosecond magneto-optical processes in metals*", Compte rendu de l'Académie des sciences, **2**, Issue 10, 1483–1504, (2001).