HAL open science

Magneto-optical four-wave mixing transients in garnets

M. Barthélémy, M. Vomir, M. Sanches Piaia, H. Vonesch, P. Molho, B. Barbara, J.-Y. Bigot

To cite this version:

M. Barthélémy, M. Vomir, M. Sanches Piaia, H. Vonesch, P. Molho, et al.. Magneto-optical four-wave mixing transients in garnets. Optica, 2017, 4 (1), pp.60-63. 10.1364/OPTICA.4.000060 . hal-01765147

HAL Id: hal-01765147

https://hal.science/hal-01765147v1

Submitted on 22 Sep 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

optica

Magneto-optical four-wave mixing transients in garnets

M. Barthelemy,[1] M. Vomir,[1] M. Sanches Piaia,[1] H. Vonesch,[1] P. Molho,[2] B. Barbara,[2] and J.-Y. Bigot[1,*]

[1]Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, Strasbourg 67034, France
[2]Université Joseph Fourier, CNRS, Institut Néel, UPR 2940, Grenoble 38042, France
*Corresponding author: bigot@ipcms.u-strasbg.fr

Received 23 September 2016; revised 30 November 2016; accepted 1 December 2016 (Doc. ID 276213); published 4 January 2017

Femtosecond optical transients are extensively used to study electronic coherence and population relaxation in condensed matter. In this work we show that it is possible to determine the spin coherence and population dynamics in magnetic materials using magneto-optical (MO) four-wave mixing transients. The methodology is applied to Bi-doped garnet films where an ultrashort coherent MO response is observed during the laser pulse and attributed to the spin–orbit interaction. In addition, long-lived spin populations ($T_1 = 1.35$ ps) are shown to depend on the laser frequency detuning with respect to Fe^{3+} electronic resonance. Otherwise, our approach paves the way for realizing diffractive MO systems that can be controlled with external magnetic fields. © 2017 Optical Society of America

OCIS codes: (210.3820) Magneto-optical materials; (300.2570) Four-wave mixing; (300.6240) Spectroscopy, coherent transient; (320.0320) Ultrafast optics; (320.7120) Ultrafast phenomena; (320.7130) Ultrafast processes in condensed matter, including semiconductors.

https://doi.org/10.1364/OPTICA.4.000060

As is well known, four-wave mixing (FWM) in molecular and semiconductor materials allows determining the relaxation times associated to the electronic coherent and population dynamics. Depending on the studied system, the coherent dephasing time (T_2) and the population lifetime (T_1) occur on a broad temporal range from tens of femtoseconds to nanoseconds [1–4]. Concerning the coherent dynamics of electronic spin states, many studies have been performed, for example, near the excitonic resonances of semiconductors [5]. So far little is known about the coherence of spin states in correlated spin phases, even though it is important for understanding how to manipulate and control the magnetization of metals, oxides, or molecular magnets with optical beams. It requires development of adapted measurement methods as shown here.

Regarding spin dynamics, extensive studies of the ultrafast demagnetization in ferromagnets have been performed using femtosecond laser pulses [6–8]. A variety of approaches have been used, such as spin-resolved photo-emission [9], femtoslicing [10,11], table-top x-ray probing [12], terahertz resonant emission [13] or excitation, and ultrafast magneto-acoustics [14]. In those experiments, the demagnetization results from a thermal process associated to the change of magnetization modulus and/or orientation, which can be described phenomenologically by a three temperature model [6,15] or by the time-dependent density functional theory [16,17]. The initial demagnetization is accompanied during the first few hundreds of femtoseconds by the thermalization of spins near the Fermi level [9,10,18,19].

Concerning coherent processes in ferri or ferromagnets, several mechanisms are important. First, the inverse Faraday effect induces a transient magnetic field, which has been used, for example, to control the precession of the magnetization in garnets [20,21]. The magnetic component of an intense terahertz field can also be used to coherently control antiferromagnetic spin waves via time-dependent Zeeman interaction [22]. Second, the strong laser field may induce a magneto-electric anisotropy that modifies the magnetization dynamics. The underlying mechanism is the spin–orbit interaction. In ferromagnetic metals, it can be observed in femtosecond pump–probe magneto-optical (MO) Kerr or Faraday experiments [23]. In bismuth-doped garnets, it is well known that a strong spin–orbit interaction occurs, which manifests by a large MO susceptibility far from the main resonances associated to the octahedral and tetrahedral Fe^{3+} sites [24]. We therefore expect that this material is a good candidate for observing coherent effects associated to MO transient gratings. In this Letter, magnetic-field-dependent FWM signals are measured in Bi-doped garnet film. We use a three-beam configuration [25] so that the coherent and population components can be separately time resolved. The role of the spin–orbit interaction is investigated using a simple model consisting of an eight-level hydrogen-like system coupled to the laser field via a relativistic dipolar interaction, including the generalized electron momentum [26].

The MO-FWM experimental configuration is displayed in Fig. 1. Three linearly polarized frequency degenerate femtosecond pulses propagate in the directions $\mathbf{k_1}$, $\mathbf{k_2}$, and $\mathbf{k_3}$. The FWM signal is observed in the direction $\mathbf{k_1} - \mathbf{k_2} + \mathbf{k_3}$. $S^{\phi}_{k_1-k_2+k_3}(\tau, T)$ is defined as the rotation of the FWM polarization for an angle ϕ of the external magnetic field $\mathbf{H}$ with respect to the normal to the sample plane. $S^{\phi}_{k_1-k_2+k_3}$ is obtained with a polarization bridge consisting of a Wollaston prism and a synchronous detection scheme. $S^{\phi}_{k_1-k_2+k_3}(\tau, T)$ can be measured either as a function of the temporal delay τ between pulses propagating along the directions $\mathbf{k_1}$

and $\mathbf{k_2}$ or as a function of the delay T between pulses along directions $\mathbf{k_1}$ and $\mathbf{k_3}$. The linearly polarized pulses with a duration of 50 fs are obtained from an amplified 10 kHz titanium: sapphire-based optical parametric amplifier laser system. In practice, the static magnetic field $\mathbf{H}$ is set along the two complementary directions $\phi^{-H} = 0°$ and $\phi^{+H} = 180°$, and the magnetic contribution to the FWM emission is obtained as

$$\Delta S(\tau, T) = \left[S^{\phi=0°}_{k_1-k_2+k_3} - S^{\phi=180°}_{k_1-k_2+k_3}\right]/2. \quad \textbf{(1)}$$

Therefore, for a fixed delay $T = T_0$, one can access the coherent contribution of the magnetization as a function of the delay τ by measuring $S^{\phi}_{k_1-k_2+k_3}(\tau, T_0)$. On the other hand, for a fixed delay $\tau = 0$, $S^{\phi}_{k_1-k_2+k_3}(\tau = 0, T)$ carries the information about the spin population dynamics.

Importantly, in the case of a simple two-level system with dephasing time T_2 and lifetime T_1, the two aforementioned signals would allow retrieving both T_2 and T_1 by varying the delays τ and T, respectively. In a first set of experiments, designed to emphasize the coherent magnetic contribution, the central wavelength of the laser pulse is set off-resonance at 800 nm (1.55 eV) with the broadband energy levels of our sample as shown on the absorption spectrum displayed in the inset of Fig. 1. In a second set of experiments, designed to increase the ratio of the spin population component versus the coherence one, the central wavelength is tuned at 600 nm (2.07 eV) and 640 nm (1.94 eV) toward the resonant states associated to the higher energy Fe^{3+} levels at 2.38 and 2.87 eV in the tetrahedral and octahedral sites of our Bi-doped garnet film.

The sample is a 7 μm thick $(GdTmPrBi)_3(FeGa)_5O_{12}$ garnet deposited on a $Gd_3Ga_5O_12$ garnet substrate (0.5 mm thick) using liquid phase epitaxy [27]. In the absorption spectrum of Fig. 1, the fringes correspond to the Fabry–Perot effect on the sample. It has a perpendicular magneto-crystalline anisotropy and a weak ferrimagnetic remanence at room temperature. Let us note that the off-resonance excitation is also tuned so that the lower energy ${}^6A_{1g}({}^6S) \rightarrow {}^4T_{1g}({}^4G)$ transitions [28] (at 1.38 eV) are also avoided, even though they have a much weaker absorption cross section, as seen in the absorption spectrum in Fig. 1.

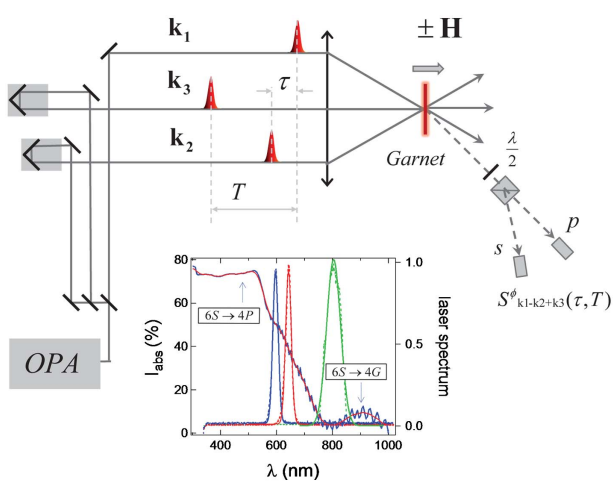

Fig. 1. Three-pulse MO-FWM configuration. **H**, static applied magnetic field; s and p, analyzed polarization components. Inset: intensity absorbed by the Bi-doped garnet film and laser pulse spectra.

Figure 2(a) displays the FWM rotation signals at 1.55 eV $S^{\phi}_{k_1-k_2+k_3}(\tau, T = 300\text{ fs})$ for $\phi = 0°$ and 180°. The difference $\Delta S(\tau)$ (MO-FWM) between these two signals is shown in the inset. This signal corresponds to the coherent contribution of the MO response. Since in the present case the frequency of the laser is detuned from the resonances associated to the crystal field split energy levels Fe^{3+}, the coherence time is shorter than the duration τ_p of the laser. Figure 2(b) shows the magnetic contribution $\Delta S(\tau = 0, T)$ up to $T = 5$ ps at 1.55 eV (closed circles). This signal corresponds to the spin population dynamics associated to the weak remaining absorption at 1.55 eV. An exponential fit to the relaxation leads to a lifetime of ~1.35 ps. During the first 50 fs, when the three pulses are overlapping in time, the coherent contribution dominates again as the three pulses interact coherently. The coordinate scale is interrupted for convenience to better show the weak population contribution for $T > 50$ fs. In Fig. 2(b) we also display the $\Delta S(\tau = 0, T)$ signals for laser photon energies 2.07 and 1.94 eV. The ratio between population contribution and coherence increases when approaching the Fe^{3+} energy levels (transitions ${}^6A_{1g}({}^6S) \rightarrow {}^4T_{1g}({}^4P)$).

We attribute the large MO-FWM emission displayed in the inset of Fig. 2(a) ($\Delta S_{k_1-k_2+k_3}(\tau, T)$) to the coherent coupling between the photons and the spins. This is sustained by the magnetic field dependence of the FWM emission. The microscopic origin of this coherent coupling can be modeled by a dipolar interaction between the laser field and the electrons that are coupled to the spins via the spin–orbit interaction. Such an approach has

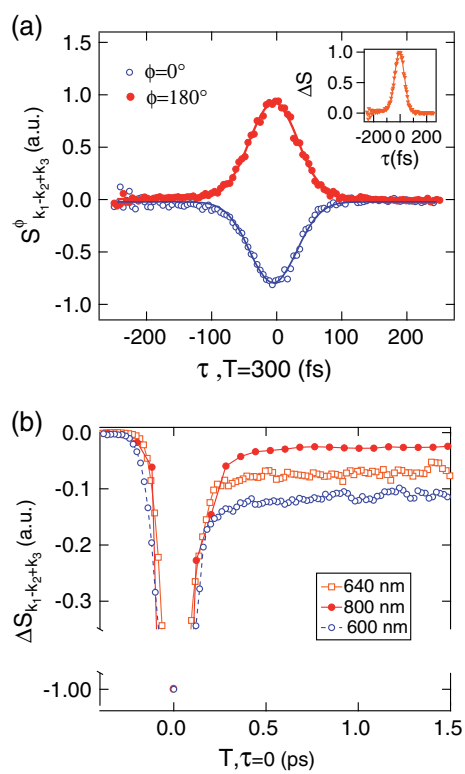

Fig. 2. MO-FWM signals. (a) FWM rotation signals $S^{\phi}_{k_1-k_2+k_3}(\tau, T = 300\text{ fs})$ at 1.5 eV as a function of τ for the magnetic field angles $\phi = 0°$ and 180°. The inset shows the difference (MO-FWM) $\Delta S(\tau, T = 300\text{ fs})$ that corresponds to the spin coherence dynamics. (b) Spin population dynamics $\Delta S_{k_1-k_2+k_3}(\tau = 0, T)$ as a function of T for the three wavelengths (600, 640, and 800 nm).

been proposed to describe MO Faraday experiments performed in ferromagnetic metallic thin films [23]. In the present case of a ferrimagnetic garnet, it is better suited to consider a multilevel system since the description of the MO response in the visible spectrum is usually accounted for by considering the discrete ${}^6A_{1g}({}^6S) \rightarrow {}^4T_{1g}({}^4P)$ transitions associated to the Fe^{3+} sites.

We adopt a Hamiltonian similar to the one previously used to describe the general relativistic electrodynamic interaction of a laser with discrete levels [26]. The corresponding eight levels considered in this model are inspired from a hydrogen-like system with $2s \rightarrow 3p$ transitions to better identify the cases of strong ($\{n, L, J, J_z\}$) and weak ($\{n, L, L_z, S_z\}$) spin–orbit couplings due to the magnetic field. We consider the following interaction Hamiltonian, which takes into account the photon–spin interaction, including spin–flip processes via the coupling of the laser field with the orbital and spin angular momenta:

$$H_{\text{int}} = -\frac{q}{m}\Pi \cdot \mathbf{A}_L - \frac{q}{2m^2c^2}[\mathbf{p}\wedge\mathbf{S}] \cdot \mathbf{E}_L, \tag{2}$$

with the generalized momentum $\Pi = \mathbf{p} - q\mathbf{A}_M + \frac{q}{2mc^2}\mathbf{S}\wedge\mathbf{E}_i$, where $\mathbf{A_M}$ and $\mathbf{A_L}$ are the magnetic and electric potential vectors ($\mathbf{E}_L$: electric field), $\mathbf{S}$ the spin vector, $\mathbf{E_i}$ the field of the ions, and $\mathbf{p}$ the momentum of the electrons. The spin–orbit interaction can be treated consistently in either the weak or the strong magnetic field interactions ($\{n, L, J, J_z\}$ or $\{n, L, L_z, S_z\}$ basis). Even though this level structure ($\Delta E_{2s_{1/2}\rightarrow 3p_{1/2}} = 1.89$ eV) is different than the ones of bismuth, it gives the essential behavior of the MO-FWM signals as we are mainly focusing on off-resonance laser excitation.

Using the density matrix formalism [29], we have solved the Liouville equation in the weak field basis, with the appropriate temporal sequence of pulses which, for a given orientation ϕ of the external magnetic field ($B = 0.05T$), gives rise to the FWM rotation signal $S^{\phi}_{k_1-k_2+k_3}(\tau, T)$ in the direction $\mathbf{k_1} - \mathbf{k_2} + \mathbf{k_3}$. These signals are then computed for opposite directions (ϕ and $\pi - \phi$) of the static field $\mathbf{B}/\mu_0$, leading to the MO-FWM signal $\Delta S(\tau, T)$. Importantly, the third-order perturbation theory requires consideration of the time ordering of the fields to which corresponds either the coherent contribution when $T \gg \tau$, for a fixed T and $\tau \leq \max\{T_2, \tau_p\}$, where τ_p is the pulse duration and T_2 the dephasing of the spin states, or the population contribution when τ is fixed and $T \leq \tau$ and $T > \max\{T_2, \tau_p\}$. Figure 3 shows the coherent and population contributions of the three-pulse MO-FWM, using the above hydrogen-like system. The parameters used are $\mathbf{E}_L = 4 \cdot 10^6$ Vm^{-1} at a photon energy $\hbar\omega_L = 1.55$ eV, detuned 18% off the main transition, for each of the three pulses. $T_1 = 1.35$ ps, $T_2 = 10$ fs, and $\tau_p = 50$ fs. T_2 is assumed to be shorter than τ_p to simulate the experimental result of Fig. 2(a). Also, for simplification, a single population lifetime is considered and assumed to be equal to the experimental case of Fig. 2(b). The modeling of the FWM signals using the interaction Hamiltonian (Eq. 2) show that they are dependent on the magnetization direction, and that it is possible to distinguish between the coherence and population dynamics. The origin of this behavior is due to the spin–orbit coupling term $\mathbf{S}\wedge\mathbf{E}_i$ appearing in the generalized electron momentum Π. It is, therefore, a manifestation of the relativistic electro-dynamics coupling between the laser field and the spins.

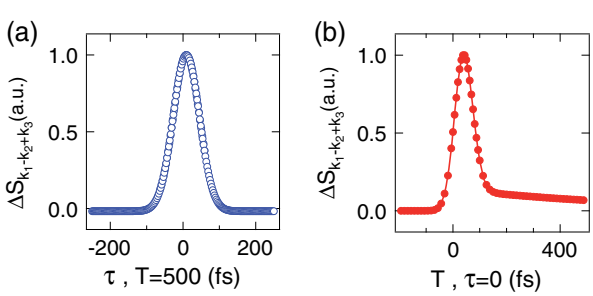

Fig. 3. Calculated MO-FWM signal for a three-pulse configuration. (a) Coherent response as a function of τ for a delay $T = 500$ fs. (b) Spin population dynamics as a function of T for a delay $\tau = 0$.

In conclusion, the coherent emission from Bi-doped garnet film obtained in a MO-FWM configuration is shown to strongly depend on the orientation of an external magnetic field. The three-beam MO configuration adopted allows investigating the spin coherence and population dynamics in ferro- and ferrimagnetic systems. In the case of Bi-doped garnets excited in their transparency region at 800 nm (with a slight overlap with the $6A1g(6S)$ $4T1g(4G)$ transitions), the strong spin–orbit interaction leads to large MO signals. Using femtosecond pulses in a three beam geometry, in the presence of a weak magnetic field, we are able to distinguish between the dephasing and population dynamics of spin states. The dephasing of the spins states (<50 fs) occurs in a time scale much shorter than the relaxation of the populations (1.35 ps). By tuning the laser wavelength toward the Fe^{3+} transitions of the tetrahedral and octahedral sites, we are able to modify the ratio of the coherent versus population components in the MO-FWM emission. Our observations are well described by a model including the spin–orbit Hamiltonian applied to a simple eight-level hydrogen-like system, with phenomenological dephasing time T_2 and lifetime T_1 of the spins states. Apart from understanding the initial coherent photon–spin interaction in ultrafast magnetism, the present work shows the high potential of coherent magneto-optics for realizing femtosecond diffractive optical devices controlled by magnetic fields.

Funding. European Research Council (ERC) (ERC-2009-AdG-20090325); Agence Nationale de la Recherche (ANR) (ANR-10-EQPX-52).

REFERENCES

1. E. Brown, Q. Zhang, and M. Dantus, J. Chem. Phys. **110**, 5772 (1999).
2. E. J. Brown, I. Pastirk, B. I. Grimberg, V. V. Lozovoy, and M. Dantus, J. Chem. Phys. **111**, 3779 (1999).
3. P. C. Becker, H. L. Fragnito, C. H. B. Cruz, R. L. Fork, J. E. Cunningham, J. E. Henry, and C. V. Shank, Phys. Rev. Lett. **61**, 1647 (1988).
4. J.-Y. Bigot, M. T. Portella, R. W. Schoenlein, J. E. Cunningham, and C. V. Shank, Phys. Rev. Lett. **67**, 636 (1991).
5. D. Chemla and J.-Y. Bigot, Chem. Phys. **210**, 135 (1996).
6. E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
7. J. Hohlfeld, E. Matthias, R. Knorren, and K. H. Bennemann, Phys. Rev. Lett. **78**, 4861 (1997).
8. M. Aeschlimann, M. Bauer, S. Pawlik, W. Weber, R. Burgermeister, D. Oberli, and H. C. Siegmann, Phys. Rev. Lett. **79**, 5158 (1997).
9. A. B. Schmidt, M. Pickel, M. Wiemhöfer, M. Donath, and M. Weinelt, Phys. Rev. Lett. **95**, 107402 (2005).
10. I. Radu, G. Woltersdorf, M. Kiessling, A. Melnikov, U. Bovensiepen, J.-U. Thiele, and C. H. Back, Phys. Rev. Lett. **102**, 117201 (2009).
11. C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruk, H. A. Durr, and W. Eberhardt, Nat. Mater. **6**, 740 (2007).

12. C. La-O-Vorakiat, M. Siemens, M. M. Murnane, H. C. Kapteyn, S. Mathias, M. Aeschlimann, P. Grychtol, R. Adam, C. M. Schneider, J. M. Shaw, H. Nembach, and T. J. Silva, Phys. Rev. Lett. **103**, 257402 (2009).
13. E. Beaurepaire, G. M. Turner, S. M. Harrel, M. C. Beard, J.-Y. Bigot, and C. A. Schmuttenmaer, Appl. Phys. Lett. **84**, 3465 (2004).
14. J. Kim, M. Vomir, and J.-Y. Bigot, Phys. Rev. Lett. **109**, 166601 (2012).
15. J.-Y. Bigot, C. R. Acad. Sci. Ser. IV **2**, 1483 (2001).
16. K. Carva, M. Battiato, D. Legut, and P. M. Oppeneer, Phys. Rev. B **87**, 184425 (2013).
17. K. Krieger, J. K. Dewhurst, P. Elliott, S. Sharma, and E. K. U. Gross, J. Chem. Theory Comput. **11**, 4870 (2015).
18. L. Guidoni, E. Beaurepaire, and J.-Y. Bigot, Phys. Rev. Lett. **89**, 017401 (2002).
19. C. Boeglin, E. Beaurepaire, V. Halté, V. Lopez-Flores, C. Stamm, N. Pontius, H. A. Duerr, and J.-Y. Bigot, Nature **465**, 458 (2010).
20. A. V. Kimel, A. Kirilyuk, P. A. Usachev, R. V. Pisarev, A. M. Balbashov, and T. Rasing, Nature **435**, 655 (2005).
21. M. Deb, M. Vomir, J.-L. Rehspringer, and J.-Y. Bigot, Appl. Phys. Lett. **107**, 252404 (2015).
22. T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mahrlein, M. Dekorsy, T. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, Nat. Photonics **5**, 31 (2011).
23. J.-Y. Bigot, M. Vomir, and E. Beaurepaire, Nat. Phys. **5**, 515 (2009).
24. G. F. Dionne and G. A. Allen, J. Appl. Phys. **75**, 6372 (1994).
25. A. M. Weiner, S. D. Silvestri, and E. P. Ippen, J. Opt. Soc. Am. B **2**, 654 (1985).
26. H. Vonesch and J.-Y. Bigot, Phys. Rev. B **85**, 180407 (2012).
27. P. Molho and M. de Albuquerque, J. Magn. Magn. Mater. **226**, 1388 (2001).
28. G. B. Scott, Phys. Status Solidi B **79**, 203 (1977).
29. S. Mukamel, *Principles of Nonlinear Optical Spectroscopy* (Oxford University, 1995), Chap. 3.

79.81.179.130 {ts '2021-05-22 04:55:37'}