Femtosecond magneto-optics of EuO

F. Formisano a,b,⁎, R. Medapalli b,c, Y. Xiaob, H. Renb, E.E. Fullertona, A.V. Kimela

a Institute for Molecules and Materials, Radboud University, 6525 AJ Nijmegen, The Netherlands
b Department of Physics, School of Sciences, National Institute of Technology, Andhra Pradesh 534102, India

c Center for Memory and Recording Research, University of California San Diego, La Jolla, CA 92093, United States

ABSTRACT

We report on the photoinduced dynamics of the magneto-optical Faraday effect and the transmittivity of ferromagnetic semiconducting phase of EuO. Excitation with 8-fs laser pulses launches significantly different dynamics of the Faraday effect compared to magnetic refraction. It is argued that the effects are dynamic probes of the magnetization and the exchange interaction in the material which have distinctly different dynamics at the sub-100 fs time scale.

1. Introduction

The discovery of the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism [1] of the exchange interaction mediated by conduction electrons boosted interest to magnetic semiconductors and naturally led to speculations about optical control of magnetism in these materials by injecting photocarriers [2–4]. Optical control of magnetism became especially appealing after the development of femtosecond laser sources. The seminal discovery of laser-induced demagnetization of metallic Ni on a picosecond time-scale by the team of E. Beaurepaire and J.-Y. Bigot [5] opened up a plethora of intriguing opportunities to study magnetism on a time scale of fundamental interaction between quasiparticles [6]. More recent developments of laser sources for generation of sub-10 fs and even attosecond pulses of light facilitated fundamentally new regimes of coherent spin dynamics in metals driven by light [6–8]. Studies of the femtosecond dynamics in magnetic semiconductors seem to be the key to answer one of the most challenging questions in condensed matter physics - how and how fast does short-range exchange interaction build up a long range magnetic order?

Europium chalcogenides are a particular example of magnetic semiconductors. Although first reports on optical control of magnetism in these materials appeared more than four decades ago, the development of new ultrafast laser sources led to new intriguing observations and keeps boosting interest to the topic [6,9,10]. For instance, several reports claimed that intense laser pulses are able to induce magnetization in EuO [11,12] and EuSe [13,14]. The effect has been explained as a result of the photogeneration of non-equilibrium charge carriers in the conduction band and enhancement of the ferromagnetic d-f exchange interaction [15]. The photo-induced magnetization has been mainly probed with the help of the magneto-optical Faraday effect [16]. Lately, non-linear optical effects, such as Second Harmonic Generation (SHG) [11,17] and Third Harmonic Generation (THG), have succeeded in detecting both the ferromagnetic order parameter (i.e. magnetization) and the exchange interaction [10,12]. Furthermore, the isotropic magnetic refraction effect in Eu-chalcogenides has been also proposed as an eligible probe technique to investigate the d-f exchange energy [18–20]. These features of the europium chalcogenides open up an opportunity to trace the dynamics of spin and spin-spin exchange interaction in one single experiment. It is fundamentally interesting to see if, in such an experiment, the dynamics of the short-range interaction and long-range order can be distinctly different. Obviously, in order to disentangle the dynamics of magneto-optical effects sensitive to spins and spin-spin exchange interaction, respectively, one has to perform the measurements on a time-scale shorter than the period of spin precession in the effective field of the exchange interaction (~100 T) [21]. Aiming to achieve this goal, we performed time-resolved measurements of the magneto-optical Faraday effect and the effect of the magnetic refraction in EuO with sub-10 fs temporal resolution. Here we show that, up to 100 fs, the magneto-optical Faraday effect, which is sensitive to the magnetization dynamics, has distinctly different dynamics from the effect of magnetic refraction, which traces the dynamics of the exchange interaction.

EuO is a ferromagnetic semiconductor with a band-gap energy (Egap) of 1.2 eV and a rock-salt (fcc) crystal structure with a lattice constant of 5.144 Å. The valence band is made of 4f electron orbitals, while the conduction band is formed mainly of Eu 5d electron orbitals [22,23]. The nominal Curie temperature (TC) of the ferromagnet is TC = 69 K. Ferromagnetism of EuO is explained as a result of two competing exchange mechanisms [24,25]: a) nearest-neighbor ferromagnetic Eu-Eu interaction and b) next-nearest neighbor antiferromagnetic Eu-Eu interaction. The ferromagnetic part of the exchange interaction is due to the virtual excitations of the 4f valence band to the 5d band conduction.
band. The antiferromagnetic part of the exchange interaction is explained as a result of the Kramers-Anderson $ff$ super-exchange mechanism via orbitals of oxygen. In the case of EuO, the ferromagnetic contribution is a dominating one [26].

In Ref. [18], it was shown that in the case of EuTe using the magneto-optical Faraday effect and the effect of Magnetic Refraction, one can trace the dynamics of the magnetization and the $df$ exchange interaction, respectively. This approach for probing the $df$ exchange interaction can also be applied for the case of EuO. In particular, it is known that the bandgap of EuO, $E_{\text{gap}}$, experiences dramatic changes upon crossing the Curie temperature. Such a coupling of optical and magnetic properties is due to the fact that a part of the bandgap energy in this semiconductor is defined by the product of spin-spin correlation function of Eu-ions, which changes upon melting of the magnetic order:

$$E_k = E_k^{(0)} + \sum_{ij} J_{ij} S_i S_j$$  \hspace{1cm} (1)

where $J_{ij}$ is the exchange constant of the $df$ exchange interaction and, $S_i$ and $S_j$ are the spins of the $i$-th and $j$-th neighboring ions.

Probing a change of optical transmittivity $\Delta T$ in a semiconductor we expect that:

$$\Delta T = \frac{\partial T}{\partial E_g} \Delta E_g + \frac{\partial T}{\partial n} \Delta n$$  \hspace{1cm} (2)

where $\Delta E_g$ is the band gap change and $\Delta n$ is the concentration of charge carriers [27].

Therefore, if we can demonstrate that the transmittivity change, $\Delta T$, is due to the change of the bandgap energy $\Delta E_g$, the transmittivity can be seen as an ultrafast probe of the exchange interaction $\sum_{ij} J_{ij} S_i S_j$. In order to reveal the dynamics of spins and the exchange energy, we measure the laser-induced transmittivity changes and differential Faraday rotation using an optical time-resolved two colors pump-probe technique similar to the one described in Ref. [18]. In such a measurement, the Faraday rotation will manifest in a change of the rotation of the detected probe beam, while the magnetic refraction effect will detect a change of the differential transmission [19,28]. In particular, in each set of measurements, the Faraday rotation and the transmission changes of the probe beam were detected contemporary by means of a custom made balanced photo-detector that measured the differential and sum channel signals, respectively. Two types of experiments were performed using long pulses with duration of 200 fs and short pulses with duration of 8 fs, respectively. The sample studied here was a 100-nm-thick EuO film grown by my magnetron sputtering onto a (1 1 0)-oriented MgO substrate. The film was capped with a protective Cr layer of 5 nm. Magneto-optical characterization of the film was performed by measuring the magneto-optical Faraday effect as a function of magnetic field applied at the angle of 90 degrees to the sample surface.

2. Results and discussion

The measurements shown in Fig. 2 confirm the Curie temperature $T_C = 69$ K. The results of the time-resolved measurements of the differential transmission with long laser pulses are shown in Fig. 3 and the Faraday rotation in Fig. 4. The experiments were performed for probe photon energies below ($h\omega_1 = 0.99$ eV in Figs. 3a and 4a) and above the bandgap ($h\omega_2 = 1.24$ eV in Figs. 3b and 4b). A constant magnetic field $H_{\text{ext}} = 0.13$ T was applied perpendicular to the sample surface. The central photon energy of the pump was 1.55 eV. The pump energy was chosen to be above the bandgap of the EuO ($E_{\text{gap}} = 1.2$ eV) to promote interband electron transitions from the Eu $4f^6 (6S_{7/2})$ orbital to the lower J values of the Eu $4f^6 (7F_J) 5d (t_{2g})$ states, with $J = 0, \ldots, 6$ [24,26,30] (see Fig. 1). At this energy, the main peak of the absorption spectrum has been theoretically predicted and observed experimentally [31].

To explain the observed dynamics, we note that the optical properties of the semiconductor can be changed by modulation of the band-gap energy $\Delta E_g$ or concentration of charge carriers $\Delta n$ (see Eq. (2)). In the case, the first contribution must have a very strong dependence on the energy of probing photons in the vicinity of the

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Fig. 1. Scheme of the energy band structure of EuO at $H = 0$ and photon energy of the pump and probes beam. The inset shows the geometry of the of the experiments.

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Fig. 2. a) Temperature dependence of the Faraday rotation in EuO defined as the height of the hysteresis loop shown in panel (b). The dependence is fitted (solid line) by $\Delta \theta = a(1 - T/T_C)^z$ [29], where $a$, $b$ and $z$ are fit parameters, here the Curie temperature $T_C = 69$ K is determined. In the inset: the geometry of the measurement, the laser source was set at normal incidence respect to the sample surface and with the magnetic field applied at an angle of 90 degrees to the sample surface. b) Hysteresis loops corresponding to field dependencies of the Faraday rotation in EuO for different temperatures.
bandgap energy. From the reflectance spectrum \([32]\) of EuO, it is seen that upon crossing the band gap energy the derivative \(\Delta \rho / \Delta \omega\) and thus the bandgap dependent contribution to the transmissivity is expected to change sign. The expected sign change upon changing the probe photon energy is also seen in the experimentally measured dependencies of the differential transmittivity \(\Delta T/T\) for photon energies below and above the bandgap \(E_\text{g}\) = 1.2 eV. Therefore, we can conclude that the observed dynamics of the differential transmittivity traces the dynamics of the bandgap energy. Moreover, the dynamics drops substantially in amplitude upon crossing the Curie temperature demonstrating that a substantial part of the transmittivity signal has a magnetic origin. Note that although above the Curie temperature, the long-range order is destroyed, short-range order can still be present and, the spin-spin correlation function is non-zero even above \(T_c\). Therefore, even above the Curie temperature, the dynamics of the differential transmittivity can be seen as a probe of randomly magnetized magnetic polarons \([16,33,34]\).

Using the same temporally long and thus spectrally narrow pulses and under the same experimental conditions, we also measured the dynamics of the magneto-optical Faraday effect \((\Delta \theta)\). The results of the measurements performed for probe photon energies below and above the bandgap are shown in Fig. 4. The data are in good agreement with those reported earlier, especially with the one in Ref. \([12]\) for Gd-doped EuO samples. We note that Cr capping and impurities at the interface of those reported earlier, we recognize the process of ultrafast laser-induced demagnetization of type II \([35]\). This type of demagnetization is present when coupling between the electron and spin system is weak. In this case, the first part of the drop in magnetization has a constant time decay which is comparable with non-magnetic one, in which the spin system tries to reach thermal equilibrium with heated electron gas after the laser excitation. This means that the fast initial quenching of the magnetization is followed by a slower demagnetization. More particularly, after an ultrafast, but incomplete demagnetization within the first \(\sim 80\) fs, one observes a slower demagnetization on a time-scale of \(\sim 1.5\) ns. Increasing temperature and reaching the \(T_c\), the demagnetization process slows down and vanishes for temperatures \(T > 50\) K. After crossing \(T_c\), the \(\Delta \theta\) shows the counterpart of the photo-enhanced magnetization effect and, it keeps showing it over the whole temperature scan, in agreement with other works \([16]\). Such a big \(\Delta \theta\) is proof of strong coupling between localized spins and charge carriers, typical for the formation of bounded magnetic polarons \([36]\). This is also an explanation of the two-step process of the \(\Delta T/T\) signal at low temperature \([37]\). In Fig. 4(b) the behavior of the \(\Delta \theta\) presents a similar two-step demagnetization process at low temperatures but less pronounced in the slow demagnetization phase. The slightly different behavior is due to the different position of the probe photon energy with respect to the band of EuO.

Data shown in Figs. 3 and 4 confirm our hypothesis that the magneto-optical Faraday effect and the effect of magnetic refraction can be employed as dynamic probes of the magnetization and the exchange interaction, respectively. In our final experiment, we perform similar measurements but using spectrally broad 8-fs laser pulses. Such short pulses were produced via a compression technique based on spectral broadening in a hollow-core dielectric waveguide (in the case, Kalexoscope™ from Spectra-Physics Vienna) filled with Ne \([38]\). The original 35 fs pulse, before the compression phase, is produced by a Ti:Sapphire regenerative amplifier working at a repetition rate of 1 kHz. Fig. 5, reports time-resolved dynamics of the differential transmission \(\Delta T/T\) and the magneto-optical Faraday effect \((\Delta \theta)\). Note the observed
Fig. 4. Laser-induced dynamics in EuO a) temporal profiles of the probe Faraday rotation with probe energy, $E_1 = 0.99$ eV, below band-gap. b) temporal profiles of the probe Faraday rotation measured with probe energy, $E_2 = 1.24$ eV, above band-gap. Note that in both case, a constant magnetic field ($H_{\text{ext}}$) of 0.13 T was applied.

Fig. 5. Transient differential transmission $\Delta T/T$ (dashed line) and the Faraday rotation $\Delta \theta$ (solid line) in EuO measured using pump–probe technique and sub-10 fs laser pulses. Panels (a) and (b) show sub-10 ps and sub-100 fs dynamics, respectively. The measurements were performed at constant magnetic field $H_{\text{ext}} = 0.13$ T.
dependencies are different from those in Figs. 3 and 4 because the pulses have now a much broader spectrum, with photon energies from 1.3 eV to 1.9 eV. In particular, due to the broad spectrum of the probe pulses, the photon energies spread over the peaks in the reflectivity spectrum [32]; hence the measurements taken at different temperatures show positive ∆T/T and no change of sign in the ∆T/T trace crossing Tc.

Fig. 5 shows that at low temperatures the differential transmission is systematically faster than the Faraday rotation. As explained above, the observed fast increase of transmissivity within the first femtoseconds can be explained as a result of an ultrafast change of the exchange energy in the medium. Surprisingly, the Faraday rotation has a substantially different dynamics. It is delayed with respect to the differential transmission and shows what, in this work, we will call latency time. From Fig. 5 we estimate the average latency time tlat ≈ 80 fs and note that it has a singularity at T = 30 K, where the latency time is almost doubled. The reason for this deviation could be due to presence of the metal-insulator phase transition, which is expected to start at around 50 K [39]. Assuming that the dynamics of the Faraday effect is caused by demagnetization of EuO, we can interpret the latency as a result of an interplay of two competing processes – laser-induced enhancement of the exchange interaction leading to a strengthening of the ferromagnetic order and laser-induced generation of hot charge carriers that promotes melting of the ordered state. During the latency period, the processes compensate one another and the Faraday rotation does not change. Upon thermalization of electrons, that occurs on a time scale of 100 fs, their effective temperature increases, demagnetization acquires a larger impetus and, eventually dominates the dynamics of the Faraday effect. It is clear from the data that once the long range order is gone, crossing the Tc, the latency time disappeared and, the two dynamics of ∆T/T and ∆θ start at the same time delay.

3. Conclusion

In conclusion, using a pump-probe technique with 8 fs temporal resolution, we studied photoinduced dynamics of the magneto-optical Faraday effect and the transmissivity in magnetic semiconductor EuO. It is shown that the magneto-optical Faraday effect is delayed with respect to the transmissivity with a latency around 80 fs. This latency disappears above the Curie temperature. The observations are consistent with our assumption that the magneto-optical Faraday effect and the differential transmissivity are sensitive to the magnetization dynamics and the dynamics of the exchange interaction, respectively. Therefore, we argue that our experiment has revealed distinct dynamics of the magnetization and the exchange interaction at the sub-100 fs time scale in epitaxial EuO thin film.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jmmm.2020.166479.
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