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# Magnetic-field-induced crossover from the inverse Faraday effect to the optical orientation in EuTe

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A time-resolved optical pump-probe technique has been applied for studying the ultrafast dynamics in the magnetic semiconductor EuTe near the absorption band gap. We show that application of external magnetic field up to 6 T results in crossover from the inverse Faraday effect taking place on the femtosecond time scale to the optical orientation phenomenon with an evolution in the picosecond time domain. We propose a model which includes both these processes, possessing different spectral and temporal properties. The circularly polarized optical pumping induces the electronic transition $4f^7 5d^0 \rightarrow 4f^6 5d^1$ forming the absorption band gap in EuTe. The observed crossover is related to a strong magnetic-field shift of the band gap in EuTe at low temperatures. It was found that manipulation of spin states on intrinsic defect levels takes place on a time scale of 19 ps in the applied magnetic field of 6 T. *Published by AIP Publishing*. https://doi.org/10.1063/1.5027473

## I. INTRODUCTION

Ultrafast phenomena related to the control and manipulation of electronic and spin states are in the focus of current research in physics of magnetism, magnonics, and spintronics.[1–3] This activity is motivated, first of all, by novel experiments on ultrafast magnetic phenomena revealing new fundamentally important mechanisms of electronic, spin and orbital dynamics taking place on femto- and picosecond time scales.[4–9] On the other hand, new results open potential possibilities for constructing high-speed magneto-electronic and magneto-optical devices.

The conservation of angular momentum is one of the most fundamental laws of physics, which plays an important role in various phenomena. For example, the Einstein-de Haas effect is a consequence of this conservation manifesting in the mechanical rotation of a free body, when its magnetic moment is changed.[10] The photon is a particle with an intrinsic angular momentum of one unit of $\hbar$ in the quantum-mechanical description; therefore, the circularly polarized light carries a spin angular momentum. In Ref. 11, it was shown that the torque exerted by circularly polarized light can be transferred to a small electric dipole. An intense circularly polarized light may create a magnetization $\mathbf{M}$ in a medium during the photon-electron interaction due to the inverse Faraday effect

$$\mathbf{M} = -i\chi^{(2)}(0;\omega,-\omega)[\mathbf{E}\times\mathbf{E}^*], \tag{1}$$

where $\chi^{(2)}(0;\omega,-\omega)$ is the second-order nonlinear optical susceptibility describing the two-photon mixing process allowed in any media[12] and $\mathbf{E}$ is an oscillating electric field at angular frequency $\omega$. This phenomenon was theoretically predicted and discussed in Refs. 13–15 and experimentally observed in Ref. 16. Nowadays, the inverse Faraday effect is widely used for experimental and theoretical studies of ultrafast phenomena in magnetic systems.[7,17–20] An inverse transverse magneto-optical Kerr effect related to Eq. (1) was predicted in Ref. 21.

Another physical phenomenon based on the angular momentum transfer from circularly polarized light to a medium is the optical orientation. This phenomenon reflects the exchange of angular momentum between the circularly polarized light and atomic or solid state systems. The principles of optical orientation were established by Kastler for paramagnetic atoms[22] and then were successfully applied for molecules[23] and semiconductors.[24] For example, due to the angular momentum conservation, a circularly polarized photon creates a spin-oriented *s*-electron in GaAs with a rather long life time at room[25] and low temperatures.[26] The phenomenon of optical orientation is a linear optical process taking place during the interaction of circularly polarized light with an absorbing medium.

Here, we report on the magnetic-field control of interplay between the inverse Faraday effect and optical orientation close to the absorption band gap in the magnetic semiconductor EuTe. We show that mechanisms of optical orientation and spin-relaxation in this material are different from those in model band semiconductors $A^{III}B^{V}$ and $A^{II}B^{VI}$ due to a specific electronic structure of EuTe.

## II. EXPERIMENTAL DETAILS AND RESULTS

Magnetic semiconductors Eu$X$ ($X$ = O, S, Se, Te) represent a group of materials possessing unique electronic, magnetic, optical, and magneto-optical properties[27–29] which are determined by strongly localized $4f^7$ electrons of the $Eu^{2+}$ ions with spin $S = 7/2$ and orbital momentum $L = 0$. EuTe is antiferromagnetic with a Néel temperature $T_N = 9.6$ K.

The magnetic moments of the two sublattices $\mathbf{m}_1$ and $\mathbf{m}_2$ ($|m_1| = |m_2|$) are ordered antiferromagnetically in adjacent (111)-planes. In external magnetic field, EuTe can be ferromagnetically saturated above a critical field of 7.2 T. Most of the early research in 1960s–1970s were performed on bulk single crystals and polycrystalline thin films of Eu*X*. However, during the last decade, high-quality epitaxial thin films of Eu*X* were successfully grown on Si and GaN semiconductor substrates opening new opportunities for applications.[30–32] Eu*X* compounds reveal a new type of nonlinear magneto-optical effects,[33,34] ultrafast spin dynamics,[35–38] and photo-induced spin polarons with a giant magnetic moment.[39,40]

We present results on optical pump-probe studies of epitaxial films of the magnetic semiconductor EuTe. This material exhibits a very strong redshift of the fundamental absorption edge by 130 meV for magnetic fields between 0 and 8 T.[41] Optical effects for photon energies close to the EuTe absorption band gap could be governed by applying external magnetic field. Using a pump-probe technique, we performed experiments on the magnetic-field control of helicity-dependent photo-induced phenomena in EuTe. The pump-probe experiments were done in transmission geometry using an optical parametric oscillator pumped by a Ti:Sapphire laser with 1 ps pulses at 80 MHz repetition rate. We used a degenerate optical scheme for pump and probe beams having a photon energy of 2.19 eV. This energy is slightly below the band gap value of 2.4 eV in EuTe at zero magnetic field. EuTe films were grown by the molecular-beam epitaxy on (111)-oriented $BaF_2$ substrates.[42,43] The 1 $\mu$m thick layers were capped with a 40-nm-thick $BaF_2$ protective layer, and the high sample quality was confirmed by x-ray analyses.

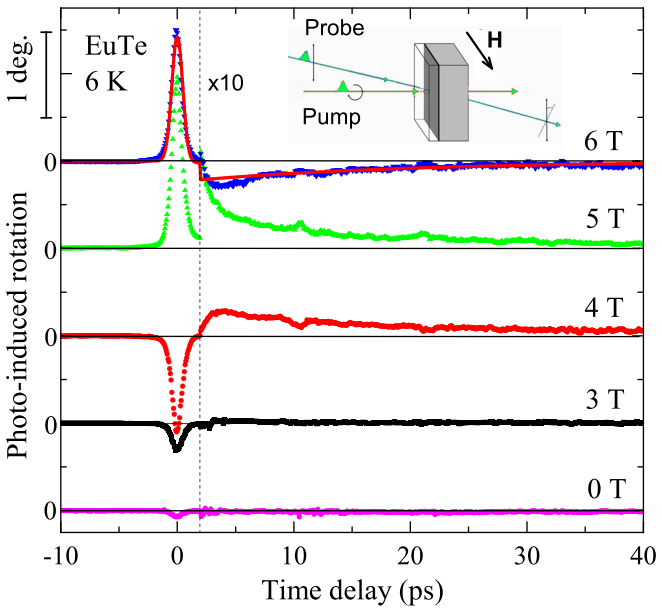


FIG. 1. Temporal behavior of the photo-induced optical rotation in EuTe for different magnetic fields at the photon energy of 2.19 eV. For clarity, the traces are shifted vertically. A solid curve for a magnetic field of 6 T shows the best fit on the basis of Eq. (3) for the following parameters $A = 1.69(1)^\circ$, $B = -0.024(3)^\circ$, $\sigma = 0.67(1)$ ps, and $\tau_s = 19(3)$ ps. There is a scale change of ordinate axis by an enhancement factor of 10 at the time delay of 2 ps (noted as $\times 10$). The inset shows schematically the pump-probe experimental geometry.

Figure 1 shows the probe light polarization rotation induced by the pump beam (photo-induced rotation) in EuTe as a function of the pump-probe time delay for different magnetic fields. Magnetic fields up to 6 T were applied in the Voigt geometry $\mathbf{k} \parallel (111) \perp \mathbf{H}$ (see the inset in Fig. 1). Temporal behavior of the photo-induced rotation is characterized by a narrow Gaussian-shape peak around the zero time delay for magnetic fields of 0–3 T. For magnetic fields above 3 T, a broad tail with a characteristic relaxation time of several picoseconds begins to arise.

Figure 2 shows the photo-induced optical rotation as a function of magnetic field for two time delays of 0 and 3 ps. These two dependencies display appreciably distinct behavior. The magnetic field dependence for 0 ps time delay has negative values for magnetic fields $0 < H < 4.7$ T; it has a minimum at $H \simeq 4.4$ T, and then it reverses sign for magnetic field of 4.7 T and has a broad maximum for $H \simeq 5.3$ T. Surprisingly, the magnetic field dependence for 3 ps time delay has zero values for magnetic fields $0 < H < 3.2$ T. It has a maximum at $\simeq 4.9$ T and reverses sign for $H \simeq 5.8$ T. Quantitatively, the sign reversal of the photo-induced optical rotation for 0 ps is related to the strong redshift of the absorption band gap in EuTe in external magnetic field[41] when the fundamental absorption edge crosses the probe photon energy of 2.19 eV. Observed different behavior for time delays of 0 and 3 ps can be qualitatively understood by taking into account an influence of excited electronic states on the absorption band gap in EuTe.

## III. DISCUSSION

For explaining optical pump-probe experiments, let us consider the electronic energy diagram of the magnetic semiconductor EuTe in which $Eu^{2+}$ ions in the ground state $4f^7 5d^0$ play the decisive role. The electric dipole transition selection rules imply that the $4f \rightarrow 6s$ transitions are forbidden and one has to take into account the $4f^7 5d^0 \rightarrow 4f^6 5d^1$

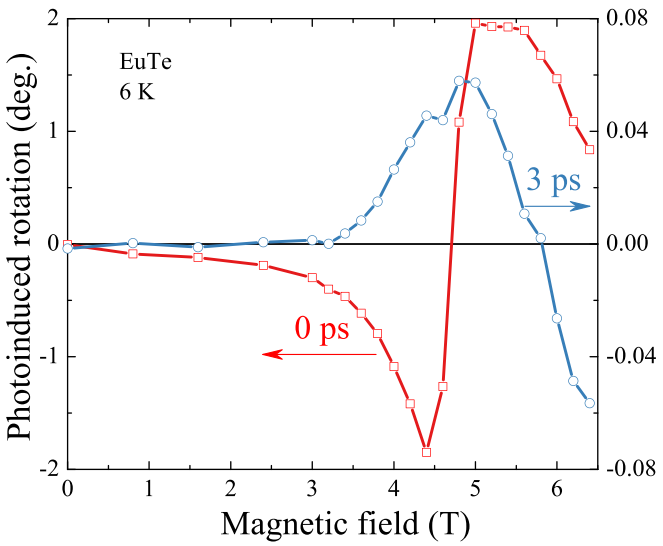


FIG. 2. Magnetic field dependence of the photo-induced rotation in EuTe for two time delays at the photon energy of 2.19 eV.

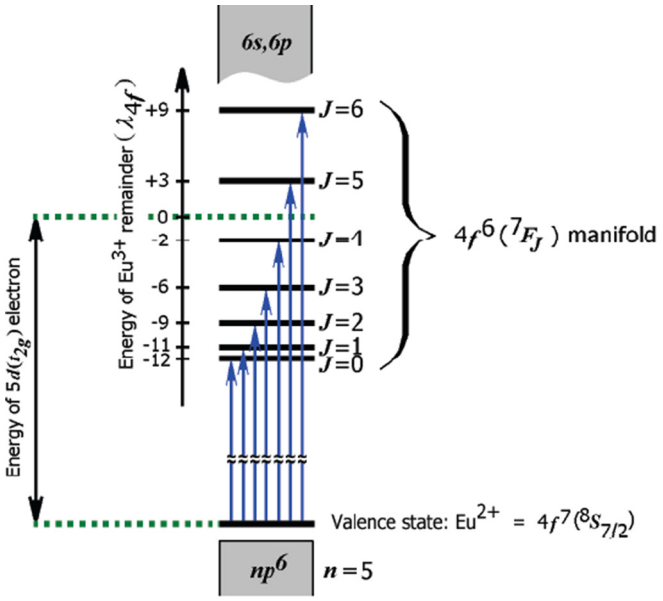


FIG. 3. Electronic energy diagram for unperturbed $4f^7$ and excited $4f^6$ states in EuTe.

electric dipole transition forming the absorption band gap in EuTe.[44] Figure 3 shows the electronic energy diagram for unperturbed $4f^7$ and excited $4f^6$ states. The absorption edge of EuTe corresponds to the onset of the $4f^7(^8S_{7/2}) \rightarrow 4f^6(^7F_J)5d(t_{2g})$ transition, where the final state is combined of one $5d$-electron and six $4f$-electrons. The $^7F_J$-multiplet ($J = 0, 1, \ldots, 6$) of the six $4f$-electrons with a total spin-orbital splitting is about 0.6 eV. In magnetic fields of 0.5 T, the semiconductor EuTe has two antiferromagnetically ordered sublattices with spins oriented perpendicularly to the magnetic field **H**. Due to spin conservation in the electric-dipole transition $4f^75d^0 \rightarrow 4f^65d^1$ for the circularly polarized photon, immediately after excitation, the electron spin is oriented along the spin of a magnetic sublattice. Moreover, following the Franck-Condon principle, the electronic transition takes place at fixed spatial and spin coordinates of the lattice. Schematically this process, which we call as stage I, is shown in Fig. 4. The width of a single sublevel of the $^7F_J$-multiplet is about 0.1 eV which corresponds

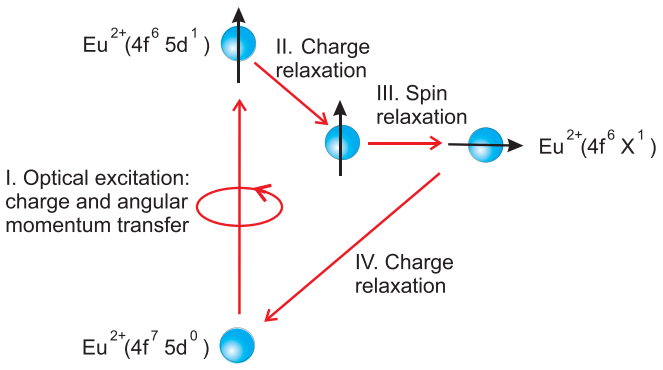


FIG. 4. A scheme of different stages at the electric-dipole transition $4f^75d^0 \rightarrow 4f^65d^1$ in EuTe: I. Optical excitation—charge and angular momentum transfer; II. Charge relaxation; III. Spin relaxation; IV. Charge relaxation.

to the electron lifetime of about 18 fs. After this time interval, some electrons are trapped by long-living intrinsic defect states $4f^6X^1$ (stage II) which are responsible for the luminescence process[41] and magnetic polaron states.[39,40] The life time of electrons at these states is longer than 1 ns depending on an applied magnetic field. During this time interval, the spin of electron at the states $4f^6X^1$ starts to precess (stage III) due to the presence of external magnetic field **H**. This precession can be analysed in terms of the Landau-Lifshitz equation with the Gilbert damping[45–47]

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}^{eff} + \alpha \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}, \tag{2}$$

where **M** is the local magnetization, $\gamma$ is the gyromagnetic ratio, $\mathbf{H}^{eff}$ is the effective magnetic field, which accounts for the external field **H** and exchange field in EuTe, and $\alpha$ is the Gilbert damping constant. In Ref. 47, it was shown that the relativistic extrinsic spin-orbit coupling gives rise to a dominant local spin relaxation mechanism in magnetic solids. In the Voigt geometry $\mathbf{k} \parallel (111) \perp \mathbf{H}$, this spin-relaxation mechanism corresponds to the transverse spin relaxation (see Fig. 4). Finally, trapped electrons at the $4f^6X^1$ states relax to the $4f^7(^8S_{7/2})$ ground state (stage IV).

In order to estimate the characteristic spin relaxation time of electrons at the $4f^6X^1$ states, we propose the following scheme. Assuming that the pump and probe pulses have the Gaussian temporal behavior, the photo-induced rotation $\theta$ can be analyzed by a single-time relaxation model:[25,48]

$$\theta = \frac{A}{\sigma\sqrt{\pi}} \exp\left(-\frac{t^2}{\sigma^2}\right) + \frac{B}{2} \exp\left(\frac{\sigma^2}{4\tau_s^2} - \frac{t}{\tau_s}\right)\left[1 - \mathrm{erf}\left(\frac{\sigma}{2\tau_s} - \frac{t}{\sigma}\right)\right], \tag{3}$$

where $t$ is the pump-probe time delay, $\sigma$ is the width of the Gaussian pulse, and $A$ and $B$ are coefficients related to the instantaneous Gaussian-type and non-instantaneous delayed terms, respectively. The first and second terms in Eq. (3) describe instantaneous contribution due the inverse Faraday effect and non-instantaneous spin-related contribution, respectively. When the laser pulse duration is much shorter than thermal relaxation times, the instantaneous contribution can be analysed in terms of the third-order optical nonlinearity $\chi^{(3)}(\omega; \omega, -\omega, \omega)$ characterising the four-wave mixing process which is relevant to the pump-probe experiment. The photo-induced polarization **P** arising in this process can be written as

$$\mathbf{P} = \epsilon_0 \boldsymbol{\chi}^{(3)}(\omega; \omega, -\omega, \omega) : \mathbf{E}_{pump}\mathbf{E}^*_{pump}\mathbf{E}_{probe}, \tag{4}$$

where $\mathbf{E}_{pump}$ and $\mathbf{E}_{probe}$ are the optical electric fields of pump and probe beams, respectively. Equation (4) describes a nonlinear optical process enabling an angular momentum transfer from the incoming circularly polarized pump beam to the linearly polarized probe beam. The photo-induced polarization **P** is a source for the outgoing probe light. This process is related to the orbital movement of electrons; therefore, it is called the instantaneous orbital contribution.[6] Moreover,

the spin-selective optical Stark effect for circularly polarized light[49] can contribute to the third-order optical nonlinearity $\chi^{(3)}(\omega;\omega,-\omega,\omega)$. In the case of EuTe, this nonlinearity can be rather strong for the electric-dipole transition $4f^7 5d^0 \rightarrow 4f^6 5d^1$. This gives rise to the photo-induced rotation observed in our experiments which is due to the inverse Faraday effect.

The non-instantaneous contribution appears for applied magnetic field values higher than 3 T. The absorption band gap of EuTe is about 2.4 eV at low temperatures and this value is strongly influenced by an applied field.[41] At a magnetic field of 5 T, the band gap is about 2.2 eV. Thus, the non-instantaneous contribution is related to ultrafast optical orientation for the electric-dipole transition $4f^7 5d^0 \rightarrow 4f^6 5d^1$ with the subsequent charge and spin relaxation with a time constant $\tau_s$ (see Fig. 4). This relaxation corresponds for the Voigt geometry to the transverse component decay of the photo-induced magnetization vector **M** towards its equilibrium orientation parallel to the applied field **H**. Applying the fitting procedure on the basis of Eq. (3) to the experimental data, we found the transverse spin relaxation time $\tau_s = 19$ ps at the $4f^6X^1$ states. We note that this value is about two times shorter than the precession period and about ten times shorter than the decay time of precession oscillations at the $4f^7d^0$ states in the applied magnetic field of 6 T.[37]

Next we would like to discuss the magnetic-field dependence of photo-induced rotation in EuTe (see Fig. 2), which is rather remarkable. For 0 ps time delay for magnetic fields $0 < H < 4.7$ T, the photo-induced rotation is negative, and it has positive values for higher magnetic fields. For 3 ps time delay for magnetic fields $3.2 < H < 5.8$ T, the photo-induced rotation is positive, and for higher fields, it is negative. Besides the strong redshift of the absorption band gap in external magnetic field in EuTe,[41] a change of photo-induced population density with respect to the bottom of the conduction band can modify the exchange interaction and thus magneto-optical constants determining the sign and amplitude of the photo-induced rotation. Such a mechanism with a tuning-parameter dependence via the carrier density was disclosed for Gd-doped EuO.[38] Furthermore, an optical modification of the exchange coupling was discussed in Ref. 50. However, the detailed understanding of physical origin of the observed phenomena taking place at the optical excitation of electronic transition $4f^7 5d^0 \rightarrow 4f^6 5d^1$ and a respective relaxation process would need a microscopic theory that is beyond the present study.

## IV. CONCLUSIONS

In conclusion, we observed a strong optical response in the magnetic semiconductor EuTe for the circularly polarized pump in transmission geometry applying magnetic field in the Voigt geometry $\mathbf{k} \parallel (111) \perp \mathbf{H}$. Observed signals can be attributed to the strong optical nonlinearity of the third order accounting for the inverse Faraday effect and the optical orientation phenomenon at the electronic transition from the localized $4f^7$ states of $Eu^{2+}$ ions on the top of the valence band into $5d$ orbitals forming the conduction band. By applying magnetic field in the range of 0–6 T, one can control the interplay between the inverse Faraday effect and the optical orientation phenomenon in EuTe. We note that such a cross-over mechanism can be important for different classes of intrinsic and diluted magnetic semiconductors in which the band gap can be strongly influenced by the external magnetic field.

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