

PDF hosted at the Radboud Repository of the Radboud University Nijmegen

The following full text is a publisher's version.

For additional information about this publication click this link.

<https://hdl.handle.net/2066/215060>

Please be advised that this information was generated on 2021-04-15 and may be subject to change.

PAPER • OPEN ACCESS

Terahertz modulation of the Cotton-Mouton effect

To cite this article: S Barsaume *et al* 2019 *J. Phys.: Conf. Ser.* **1389** 012040

View the [article online](#) for updates and enhancements.



IOP | ebooks™

Bringing together innovative digital publishing with leading authors from the global scientific community.

Start exploring the collection—download the first chapter of every title for free.

Terahertz modulation of the Cotton-Mouton effect

S Barsaume¹, E A Mashkovich¹, K A Grishunin^{1,2}, A V Telegin³,
Y P Sukhorukov³, P C M Christianen^{1,4}, Th Rasing¹ and A V Kimel^{1,2}

¹Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, the Netherlands

²MIREA - Russian Technological University, Moscow 119454, Russia

³M.N. Miheev Institute of Metal Physics Ural Branch of Russian Academy of Sciences, 620108, Russia

⁴High Field Magnetic Laboratory (HFML - EFML), Radboud University, 6525 ED Nijmegen, the Netherlands

E-mail: e.mashkovich@science.ru.nl

Abstract. The ultrafast dynamics of the magneto-optical response triggered in the ferromagnetic semiconductor CdCr₂Se₄ by an intense nearly single cycle THz pulse is investigated. The dependence on the strength and the polarity of the external magnetic field, and the polarization of the THz pulse can be explained by a phenomenological expression that attributes the observed dynamics to the Cotton-Mouton effect at THz frequencies.

1. Introduction

Over the past 10 years a plethora of intriguing examples of ultrafast optical control of magnetism were demonstrated. These include ultrafast demagnetization [1], all-optical magnetic recording [2] and high efficient spintronic THz emitters [3]. Most of this work concerned magnetic metals or dielectrics. It is interesting that the idea to control magnetism by light was first suggested for magnetic semiconductors, inspired by the discovery of the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism [4] of the exchange interaction. This naturally led to speculations about optical control of magnetism in magnetic semiconductors by injecting photocarriers [5]. Ultrafast quenching of ferromagnetism induced by an optical laser pulse was demonstrated in the diluted semiconductor InMnAs [6]. Recent experiments also revealed that optical pumping of electronic transitions can excite magnetization dynamics [7,8], but the majority of the experiments have been explained with the help of the three-temperature model [9]. This raises the questions about the role of the electronic structure and the wavelength of light in the optical control of magnetism. To answer this question terahertz (THz) spectroscopy offers an attractive possibility to study solid state systems in a spectral range where light-matter interaction is defined by low-energy electronic transitions and phonons [10]. Inspired by this opportunity, we studied the THz induced dynamics in the ferromagnetic semiconductor CdCr₂Se₄ spinel.

2. Experimental setup

In this paper we investigate the single sublattice ferromagnetic semiconductor CdCr₂Se₄ with a Curie temperature 130 K [11]. The 100-nm thick polycrystalline film was obtained by vacuum deposition on



a borosilicate glass substrate of thickness 140 μm . Here, the ferromagnetic order originates from superexchange interaction of Cr^{3+} ions provided by Cd. CdCr_2Se_4 possesses an in-plane magnetic anisotropy with a relatively small coercive field of the order of tens of Gauss [12]. The bandgap for the cryogenically cooled sample is around 1.2 eV.

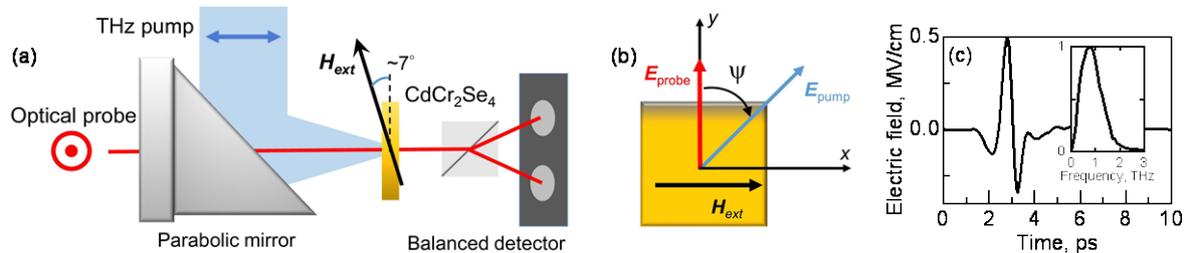


Figure 1. (a) THz pump – optical probe experimental setup. (b) Front view of the sample. The terahertz pump electric field \mathbf{E}_{pump} , the optical probe electric field $\mathbf{E}_{\text{probe}}$ and the external magnetic field \mathbf{H}_{ext} are denoted by arrows. (c) Mapped THz electric field waveform. The corresponding FFT spectrum is shown in the inset.

In order to study dynamical effects induced by intense THz radiation on the magnetism in the semiconductor, we performed time-resolved pump-probe measurements. In the measurements we used a nearly single cycle THz pulse with the electric field up to 500 kV/cm as a pump and a near infrared laser pulse as a probe.

For that we placed a beam splitter (ratio 1:100) right after the 4 mJ pulse energy, 1 kHz repetition rate Ti:sapphire amplifier, to split the output beam into two parts. The schematic of the experimental setup is shown in figure 1a. The stronger pulse (pump) is used to generate THz radiation by means of optical rectification in a LiNbO_3 crystal [13]. Afterwards, the THz pulse was broadened and tightly focused on the sample using a set of parabolic mirrors. The weaker (probe) pulse travels through the hole in the last parabolic mirror, so that both THz and probe pulses are directed collinearly onto the sample (see figure 1a). Both pulses are linearly polarized. To control the polarization direction of the THz pump we imply two wire grid polarizers. The THz polarization angle ψ is shown in figure 1b. The probe beam polarization rotation induced by the THz pulse was measured by an ordinary balanced detection scheme. An external magnetic field \mathbf{H}_{ext} was applied at an angle of approximately 7° with respect to the sample and inducing both an in-plane and an out-of-plane component of the magnetization. All experimental measurements were performed at 6 K in a helium flow cryostat, far below the Curie temperature of the spinel. The measured THz waveform with corresponding Fourier spectrum is shown in figure 1c. The procedure of THz electric field calibration can be found elsewhere [14].

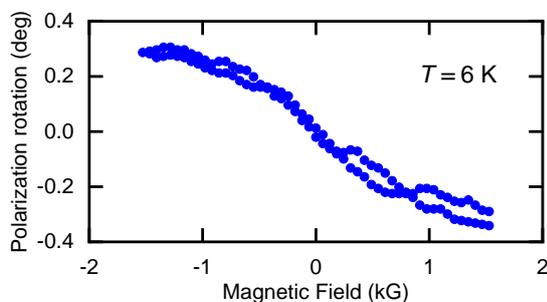


Figure 2. The static magneto-optical Faraday rotation measurement.

In order to characterize the magneto-optical properties of the CdCr_2Se_4 spinel, we performed static optical Faraday rotation measurement at 800 nm wavelength. The external magnetic field was tuned in

the range from -1.5 kG to +1.5 kG. In figure 2 it is seen that at fields above 0.5 kG, the sample rotates the polarization over a fraction of a degree. A saturation of the magnetization is observed at approximately $\pm 0.3^\circ$. This agrees well with previous observations [12].

3. THz induced magneto-optical signal

In order to study the dynamics triggered in the sample with the help of the intense THz pulse, we performed time-resolved measurements of the probe polarization rotation. The typical dependence showing the time-resolved signal as a function of time delay between the THz and probe pulses is shown in figure 3. It is seen that the signal is present only in a short time window comparable with the duration of the THz pulse. The shape of the signal almost follows the THz waveform (see figure 1c). Hence, we can conclude that the THz induced polarization rotation linearly depends on the THz field strength. The same figure shows the curves measured at two values of the external magnetic field. The external field of 0.66 kG corresponds to the value of a nearly saturated magnetization and the value of 2.1 kG is well above the saturation. It is seen that changing the field strength by a factor of 3 does not really change the amplitude of the signal. From that we can conclude that the signal does not depend on the external magnetic field strength.

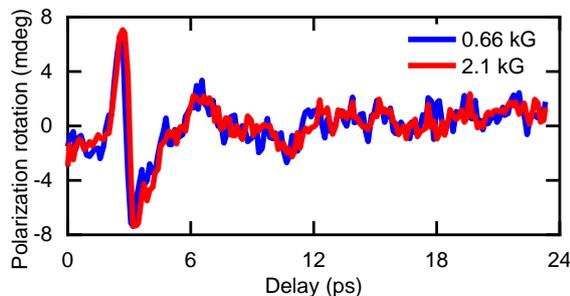


Figure 3. Transient polarization rotation at the external magnetic field strength $H_{\text{ext}} = 0.66$ kG (blue curve) and 2.1 kG (red curve).

However, this result does not exclude that the signal may depend on the magnetization of the medium. With the purpose to understand if the signal depends on the magnetization, we repeated the measurements for two polarities of the magnetic field. The results are shown in figure 4. It is seen that changing the polarity of the magnetic field changes the signal's phase (see figure 4a) over 180 degrees. To better see this effect of polarity, we calculated the sum and the difference of the signals measured at two opposite polarities of the external magnetic field. In particular, the sum was calculated as $\Delta\theta_{\text{sum}} = \theta(H_+) + \theta(H_-)$, where $\theta(H_+)$ corresponds to the probe polarization rotation with the positive external magnetic field polarity and $\theta(H_-)$ is the rotation in the case of the opposite polarity. Accordingly, the difference was calculated as $\Delta\theta_{\text{diff}} = \theta(H_+) - \theta(H_-)$. As a result, we revealed in figure 4b that the THz induced signal is odd with respect to the magnetization.

As the next step, we reveal how the THz induced signal depends on the polarization orientation of the THz pump. The polarization of the THz beam ψ was rotated from -90° to $+90^\circ$. The results of the measurements are shown in figure 5a. It is seen that the orientation of the THz polarization greatly affects the dynamics of the magnetization. To better show this polarization dependence, we plot the peak amplitude at two specific points in time P_1 and P_2 as shown in figure 5a. The P_1 and P_2 amplitudes as a function of the pump polarization angle are shown in figure 5b. It is seen that changing the polarization in the range from -90° to $+90^\circ$ changes the polarity of the induced effect. Note, that the maximum amplitude is observed when the in-plane component of the external magnetic field is perpendicular to the magnetic field of the THz pulse. And opposite, that at 0° , when the in-plane component of the external magnetic field is parallel to the magnetic field of the THz pulse, the signal is null.

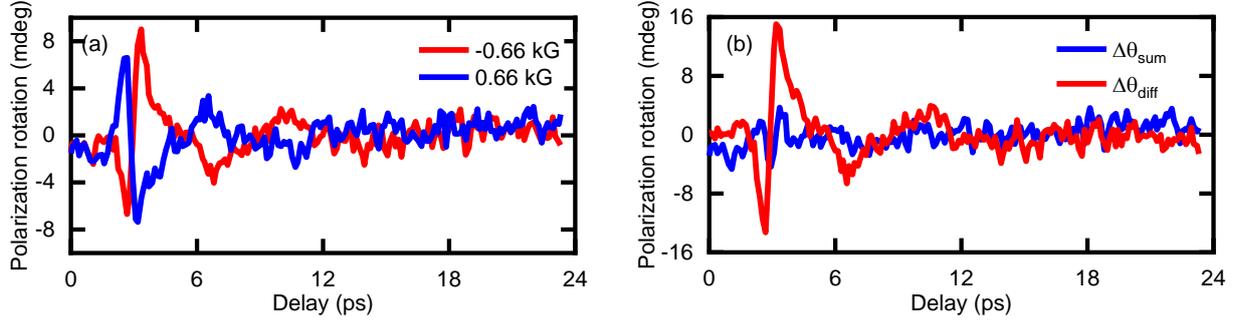


Figure 4. (a) Transient polarization rotation in magnetic fields of opposite polarity. (b) Odd (red curve) and even (blue curve) dynamics with respect to the external magnetic field.

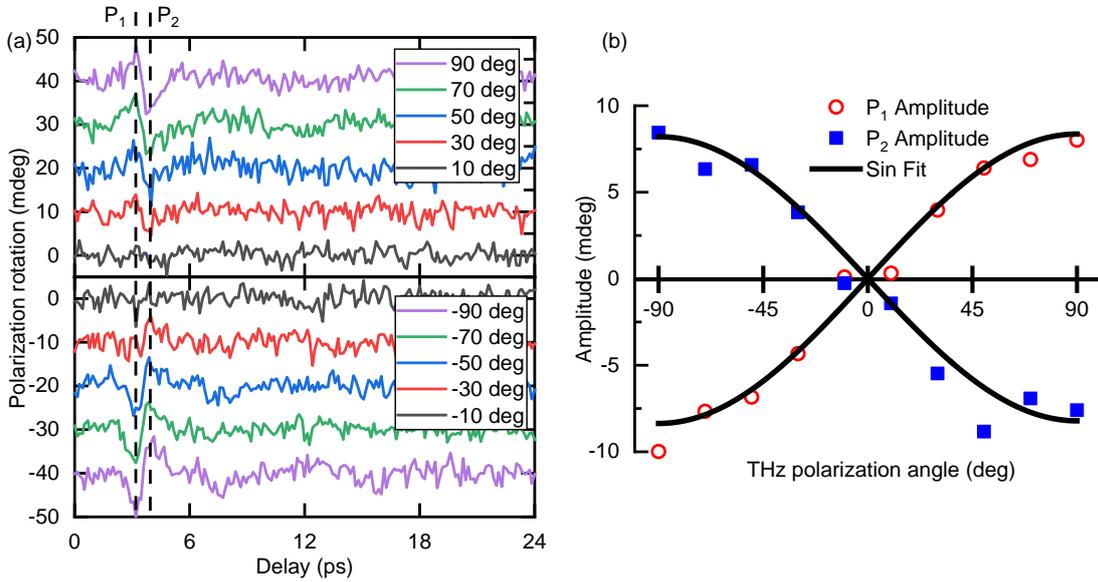


Figure 5. (a) Transient polarization rotation upon changing the angle of the THz polarization ψ from -90° to $+90^\circ$. (b) The amplitudes P_1 and P_2 as a function of the THz polarization angle. The black curves are sinusoidal fits.

Thus we can construct a phenomenological expression for the observed effect. Taking into account that the medium is centrosymmetric and bringing together all the findings listed above, one can conclude that the THz induced change of the symmetric part of the optical susceptibility in the lowest order with respect to the THz fields must be written as

$$\varepsilon_{ij}^s(\omega_{\text{THz}}) = \chi_{ijkl} H_k(\omega_{\text{THz}}) M_l(0), \quad (1)$$

where χ_{ijkl} is a phenomenological tensor and $H_k(\omega_{\text{THz}})$ is the magnetic field of the THz pulse. As you can see, the contribution to the symmetric part of the optical susceptibility is odd with respect to the magnetization $M_l(0)$. Also this contribution does change sign upon changing the polarity of the THz magnetic field. Note that one can neglect terms linear with respect to the electric field of the THz pulse $E_k(\omega_{\text{THz}})$, because the studied medium is centrosymmetric.

Finally, the probe polarization rotation can be seen as a result of different absorption of two linearly polarized waves with polarizations along the x - and the y -axes, respectively. Strong linear dichroism and birefringence at THz frequencies in similar $\text{HgCdCr}_2\text{Se}_4$ ferromagnetic semiconductor reported in [15] agree well with this. Hence, the probe polarization rotation can originate from χ_{xyxy} . This

phenomenon is very similar to the Cotton-Mouton effect with the only difference that now the magnetic field oscillates at the THz frequency. This is why we call it a THz Cotton-Mouton effect. It is also interesting to note that in contrast to the time-resolved studies with the pump in the visible and near infrared spectral ranges, here we did not observe any demagnetization of the material. This is quite surprising, because the semiconductor has a rather large concentration of holes. One would expect that THz radiation can efficiently heat the holes and cause demagnetization, similarly to the cases of ferromagnetic metals [16]. This observation can be explained by the fact that the effective mass of holes is quite large. This means that the THz photon energy is not sufficient to accelerate the heavy carriers and they are barely THz sensitive.

4. Conclusions

To conclude, we have studied the ultrafast dynamics of the magneto-optical response triggered in the ferromagnetic semiconductor CdCr_2Se_4 by an intense nearly single cycle THz pulse. It is shown that the magneto-optical signal can be substantially changed under the action of the THz pulse. Step by step analysis of the sensitivity of the THz induced dynamics to the strength of the external magnetic field, the polarity of the field and orientation of the magnetic field of the THz pulse, reveals a phenomenological expression that can explain the effect. From the expression it is seen that the observed dynamics can be explained as a Cotton-Mouton effect at THz frequencies. We also would like to note that we did not reveal any demagnetization induced by the THz pulse. Regarding the fact that the semiconductor in its ground state has already free charge carriers (holes), one would expect that the THz pulse must heat the free carriers. In accordance with the conventionally accepted three-temperature model such a heating must result in demagnetization of the ferromagnet. As no demagnetization is observed, it indicates that the conventional model is not applicable to this material. Most likely, this is related to a large effective mass of the holes. As a result of it, the THz field is unable to efficiently accelerate and thus increases the temperature of the carriers. Our recent findings show that the same value THz field can efficiently demagnetize and trigger magnetization dynamics in another magnetic semiconductor InMnAs . Such a qualitative difference in the behaviour of the THz induced dynamics is explained by the fact that the effective mass of holes in InMnAs is at least 10 times smaller than in the spinel.

Acknowledgments

The authors thank P. Albers, S. Semin, Ch. Berkhout and F. Janssen for technical support. This research was partially supported by de Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), de Stichting voor Fundamenteel Onderzoek der Materie (FOM), the EU FP7 program Grant Agreement No339813 (EXCHANGE), the Russian Foundation for Basic Research within projects 18-02-40027 and 18-52-53030 and the state assignment of Minobrnauki of Russia (theme “Spin” No.AAAA-A18-118020290104-2).

References

- [1] Beaurepaire E, Merle J-C, Daunois A and Bigot J-Y 1996 *Phys. Rev. Lett.* **76** 4250-53
- [2] Stanciu C D, Hansteen F, Kimel A V, Kirilyuk A, Tsukamoto A, Itoh A and Rasing T 2007 *Phys. Rev. Lett.* **99** 047601
- [3] Seifert T *et al.* 2016 *Nat. Photonics* **10** 483-488
- [4] Ruderman M A and Kittel C 1954 *Phys. Rev.* **96** 99-102
- [5] Pearton S J *et al.* 2003 *J. Appl. Phys.* **93** 1-13
- [6] Wang J, Sun C, Kono J, Oiwa A, Munekata H, Cywiński Ł and Sham L J 2005 *Phys. Rev. Lett.* **95** 167401
- [7] Baierl S, Hohenleutner M, Kampfrath T, Zvezdin A K, Kimel A V, Huber R and Mikhaylovskiy R V 2016 *Nat. Photonics* **10** 715-718
- [8] Stupakiewicz A, Szerenos K, Afanasiev D, Kirilyuk A and Kimel A V 2017 *Nature* **542** 71-74
- [9] Agranat M B, Ashitkov S I, Granovskii A B and Rukman G I 1984 *Zh. Eksp. Teor. Fiz.* **86**

1376-79

- [10] Zhang X-C and Xu J 2010 *Introduction to THz Wave Photonics* (Boston, MA: Springer US)
- [11] Lehmann H W and Harbeke G 1967 *J. Appl. Phys.* **38** 946
- [12] Pogrebna A, Barsaume S, Subkhangulov R R, Telegin A V, Sukhorukov Y P, Chzhan A V, Rasing T and Kimel A V 2018 *Phys. Rev. B* **98** 214427
- [13] Hebling J, Almasi G, Kozma I and Kuhl J 2002 *Opt. Express* **10** 1161
- [14] Sajadi M, Wolf M and Kampfrath T 2015 *Opt. Express* **23** 28985
- [15] Huisman T J, Mikhaylovskiy R V, Telegin A V, Sukhorukov Y P, Granovsky A B, Naumov S V, Rasing T and Kimel A V. 2015 *Appl. Phys. Lett.* **106** 132411
- [16] Bonetti S, Hoffmann M C, Sher M-J, Chen Z, Yang S-H, Samant M G, Parkin S S P and Dürr H A 2016 *Phys. Rev. Lett.* **117** 087205