## Terahertz magneto-optics in the ferromagnetic semiconductor HgCdCr<sub>2</sub>Se<sub>4</sub>

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The magneto-optical response of the ferromagnetic semiconductor HgCdCr<sub>2</sub>Se<sub>4</sub> at terahertz (THz) frequencies is studied using polarization sensitive THz time-domain spectroscopy. It is shown that the polarization state of broadband terahertz pulses, with a spectrum spanning from 0.2 THz to 2.2 THz, changes as an even function of the magnetization of the medium. Analysing the ellipticity and the rotation of the polarization of the THz radiation, we show that these effects originate from linear birefringence and dichroism, respectively, induced by the magnetic ordering. These effects are rather strong and reach 10<sup>2</sup> rad/m at an applied field of 1 kG which saturates the magnetization of the sample. Our observation serves as a proof-of-principle showing strong effects of the magnetic order on the response of a medium to electric fields at THz frequencies. These experiments also suggest the feasibility of spin-dependent transport measurements on a sub-picosecond time-scale. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License, [http://dx.doi.org/10.1063/1.4916884]

During the last 50 years, the physics of magnetism has been progressing in a breathtaking fashion and this progress can be characterized by two words: "smaller" and "faster." Exploring the physics of magnetism at ever smaller lengthscales led to the discovery of giant magneto-resistance in magnetic nanostructures, which caused a great advancement of spin-transport electronics (spintronics). 1,2 Simultaneously, the observation of subpicosecond demagnetization by a femtosecond laser pulse<sup>3</sup> triggered the field of ultrafast femtosecond magnetism and the discovery of all-optical switching.<sup>4,5</sup> However, understanding the correlations between transport properties and magnetism at ever faster time-scales has faced a problem since conventional transport measurements, based on the application of an electric voltage to the sample, can hardly achieve sub-100 ps temporal resolution. Recently, it has been demonstrated that terahertz (THz) spectroscopy with the help of freely propagating broadband sub-ps pulses of THz radiation can serve as an ultrafast probe of transport properties of semiconductors and semiconducting nanostructures. It is natural to suggest that a similar technique can be used to perform magneto-transport measurements in the THz spectral range with subpicosecond temporal resolution.

Although research areas as spintronics and femtosecond magnetism would greatly profit from THz magneto-optics, its feasibility is still questionable and studies of magneto-optical phenomena in this spectral range are still scarce. To date, THz magneto-optical phenomena have been mostly studied either in conducting materials without magnetic order or in magnetically ordered non-conducting materials. Moreover, the majority of these works studied the Faraday rotation, while other magneto-optical effects, like magnetization dependent linear birefringence and dichroism, may be equally important for establishing a link between electronic transport properties and THz magneto-optics. Measurements on magnetically ordered and conducting

samples were reported in Refs. 10, 11, and 16. However, their samples consist of ensembles of metallic ferromagnetic particles, in which the collective plasmonic response of the particles rather than the bulk material itself gives rise to spin-dependent terahertz transmission. Up till now, it remained unclear how strong magneto-optical effects at THz frequencies can be in realistic bulk spintronic materials.

Using broadband pulses of THz radiation, we report about magneto-optical studies in a Voigt geometry of the ferromagnetic semiconductor Hg<sub>0.92</sub>Cd<sub>0.08</sub>Cr<sub>2</sub>Se<sub>4</sub> in the range from 0.2 THz to 2.2 THz. Similar samples are known for their unusual high magnetoresistance up to  $80\%^{20,21}$  and large magneto optical effects (e.g., magnetotransmission) for mid-infrared radiation. 22-26 We reveal that the polarization state of a linearly polarized THz pulse substantially changes upon propagation through the sample due to the in-plane magnetization; the polarization becomes elliptical and the main axis of the ellipse rotates. Performing the measurements for different incoming and outgoing THz polarizations, varying applied magnetic field and temperature, we reveal that the ellipticity and rotation are due to rather strong linear birefringence and linear dichroism, respectively, induced by the magnetic order. The effects reach values in the order of 10<sup>2</sup> rad/m at an applied field of 1 kG which saturates the magnetization of the sample.

The sample is a  $250 \,\mu\text{m}$  thick crystalline plate of  $\text{Hg}_{0.92}\text{Cd}_{0.08}\text{Cr}_2\text{Se}_4$  grown by the chemical transport reaction technique using  $\text{CrCl}_3$  as a carrier agent.<sup>27</sup> Its crystal structure is close to cubic and belongs to the spinel symmetry group (space group Fd3m). The sample is cut perpendicular to the [111] crystallographic axis. The Curie temperature of the material, which is a p-type semiconductor, is around 120 K. The THz spectrometer used for measuring the THz transmission of the sample is shown in Fig. 1(a). It employed 50 fs laser pulses generated by a Ti:sapphire amplified laser

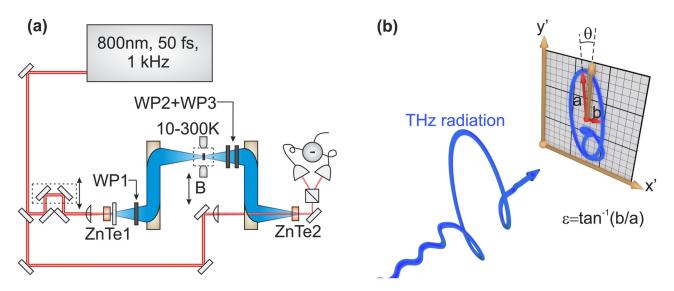


FIG. 1. (a) Setup employed for measuring the THz transmission of the magnetic semiconductor  $HgCdCr_2Se_4$ . A ZnTe crystal is used to convert laser light into THz radiation, the remaining laser light is filtered out. An additional ZnTe crystal is used to detect the THz radiation, in which the THz radiation introduces ellipticity to a laser beam. Using a balanced bridge detection scheme, the THz radiation induced ellipticity is measured. Wiregrid polarizers (WP) provide THz polarization sensitive measurements. (b) The electric field of a THz pulse is projected onto a plane perpendicular to the propagation direction, showing the rotation  $\theta$  and ellipticity  $\varepsilon$ . The axes of the plane are chosen parallel to x' and y', which in the experiment correspond to  $45^\circ$  and  $-45^\circ$  with respect to the direction of the applied field.

system at a central wavelength of 800 nm and with a repetition rate of 1 kHz. Each of these pulses was split in two. One part was incident on a 1 mm thick ZnTe crystal and thus generated THz radiation via the phenomenon of optical rectification. Using a pair of gold coated parabolic mirrors, the THz radiation was focused on the sample inside a cold-finger helium-flow cryostat, capable of setting the temperature between 10 and 300 K. The cryostat was placed in between the coils of a magnet capable of producing fields up to 1 kG. The field was applied in-plane of the sample along its  $[11\overline{2}]$ axis. An additional pair of parabolic mirrors was used to collect the transmitted THz radiation and focus it on the detector ZnTe crystal which was gated by a probe pulse from the laser. The ellipticity acquired by the probe pulse due to the THz electric field was measured using a balanced bridge detector. The output signal of the detector was therefore proportional to the magnitude and the sign of the electric field of the THz pulse at the time of arrival of the probe pulse. In our setup, the lowest spectral limit is around 0.2 THz, as determined by the propagation losses of terahertz radiation arising from the finite apertures of the parabolic mirrors. The highest spectral limit is around 2.2 THz, which is mainly determined by the transmission of the cryostat windows.

For sensitive detection of magneto-optical effects at THz frequencies, we employed wiregrid polarizers (WP), as shown in Fig. 1(a). The wiregrid polarizers have a transmission larger than 95% in the frequency range from 0 to 2 THz and an extinction ratio for an electric field at 1 THz of approximately  $2 \times 10^3$ . The first wiregrid polarizer (WP1 in Fig. 1(a)) was situated between the ZnTe crystal used for generation of the THz radiation and the sample. Directly after the sample, the second and the third wiregrid polarizers (WP2 and WP3 in Fig. 1(a)) were placed. The second polarizer was alternated between  $-45^\circ$  and  $45^\circ$  with respect to the applied magnetic field. The third polarizer was orientated perpendicular to the magnetic field. As described in Ref. 28,

such a setup provides the possibility to determine the rotation and ellipticity of the transmitted THz radiation. Figure 1(b) shows how the rotation  $\theta$  and ellipticity  $\varepsilon$  of the THz radiation appear in the projection of the electric field of a THz pulse on a plane perpendicular to the propagation direction. The axes of this plane are chosen parallel to x' and y', corresponding to the two used orientations of WP2. Applying a bias field in-plane can introduce anisotropic changes to the refractive index and absorption coefficient, resulting in ellipticity and rotation dependent on the magnetization

$$\varepsilon(M) = \varepsilon_{M=0} + \Delta \varepsilon(M),$$
  

$$\theta(M) = \theta_{M=0} + \Delta \theta(M),$$
(1)

where  $\varepsilon_{M=0}$  and  $\theta_{M=0}$  are the magnetization independent ellipticity and rotation, while  $\Delta\varepsilon$  and  $\Delta\theta$  are the magnetization induced ellipticity and rotation, respectively.

An example of the electric field of transmitted THz pulses projected onto the plane perpendicular to the propagation direction is shown in Fig. 2(a). The black trace is the projected electric field with no bias magnetic field applied, while the red trace is a projection with a field of 1 kG applied. It is seen from this figure that the field induces both changes in rotation and ellipticity to the transmitted THz pulse, providing clear signatures of magneto-optical effects at THz frequencies. From the measured THz electric field, we calculated the ellipticity and rotation using the algorithm explained in Ref. 28. In Figs. 2(b) and 2(c) we show the rotation and ellipticity, respectively, as functions of the bias magnetic field. The observed field induced rotation and ellipticity both reach approximately 3° for 1 kG bias field. This corresponds to 52 mrad over 250  $\mu m$ thickness of the sample, i.e.,  $2.1 \times 10^2$  rad/m at an applied field of 1kG which saturates the magnetization of the sample. These effects are huge when compared to typical magnetooptical signals measured in the visible spectrum.<sup>29</sup> This rotation is also large compared to some of the known

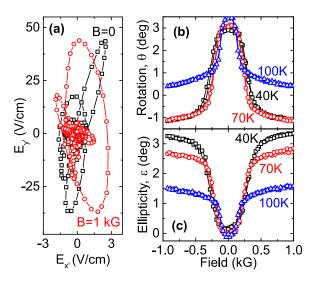


FIG. 2. (a) Traces of the electric field of the THz pulse when projected onto the x' y' plane perpendicular to the propagation direction as indicated in Fig. 1. The black trace shows the transmission for no field applied, while the red trace shows the transmission with a 1 kG applied field. (b) Polarization rotation as a function of the applied field for three different temperatures averaged between 0.4 and 1.5 THz. Here, the rotation is defined with respect to the incident polarization, which is parallel to the y' axis. (c) Ellipticity as a function of the applied field for three different temperatures averaged between 0.4 and 1.5 THz.

magneto-optical signals measured in the THz spectral range. 9,18 Our observations are, however, clearly distinct from previous rotation and ellipticity measurements at terahertz frequencies, as the rotation and ellipticity reported here change as an even function with respect to the applied magnetic field. This fact shows that the Faraday rotation, i.e., a Hall-like conductivity effect odd with respect to the applied field, can be excluded as an origin of the observed behavior.

Our measurements suggest a direct correlation with anisotropic magnetoresistance as measured with conventional electronic methods<sup>30</sup> and which should give rise to magnetooptical signals being an even function of the magnetization. Anisotropic magnetoresistance is the phenomenon in which the conductivity parallel and perpendicular to the applied field change differently with field. The optical manifestation of this effect is magnetic linear dichroism and birefringence. The magnetization dependent linear dichroism and birefringence are expected to give rise to magnetization dependent rotation and ellipticity when the incident THz radiation is polarized differently from parallel or perpendicular to the applied field. Measuring such rotation and ellipticity is preferred compared to measuring phase and amplitude changes, as a polarization change is easily and reliably recognized by measuring the transmission in a cross-polarized orientation of the polarizer before and after the sample. Thus, we chose the experimental geometry in which the incident THz radiation is polarized at −45° with respect to the applied field. We also performed transmission measurements in which the incident THz radiation was polarized perpendicular to the applied field and found no significant effect of magnetic linear birefringence, i.e., we did not observe any phase change in this configuration when changing the applied magnetic field. However, in this configuration, we did observe magnetic linear dichroism, the transmitted electric field decreased approximately 6% when applying a field of 1 kG or -1 kG compared to applying no field at 70 K.

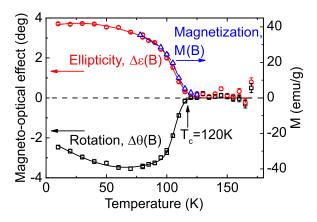


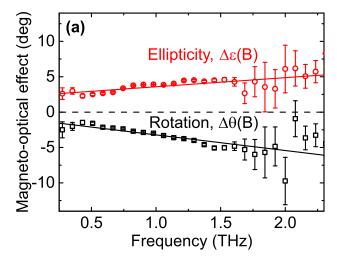
FIG. 3. Magnetization induced rotation and ellipticity averaged between 0.4 and 1.5 THz and at 1 kG applied field as functions of temperature. The solid lines are fits used for guiding the eye. Blue triangles show the magnetization measured at 1 kG applied field.

In order to show that the magneto-optical effects are indeed determined by the magnetic order, we performed temperature dependent measurements. Figure 3 shows the temperature dependence of the field induced rotation and ellipticity. It is seen that the field induced changes disappear as the temperature crosses the Curie point ( $T_c \approx 120\,\mathrm{K}$ ), confirming that the observed effects appear due to the magnetic order of HgCdCr<sub>2</sub>Se<sub>4</sub>. Figure 3 also shows a curve of the magnetization which overlaps with the magnetization induced ellipticity. The temperature dependence of the rotation slightly deviates from the expected temperature dependence of the magnetization, which can be due to the fact that the measurements are affected by a crystallographic linear dichroism independent from magnetization. This effect is also temperature dependent and may result in the observed deviations.

The microscopic origin of the magneto-optical effects may be revealed by their spectral dependence as shown in Fig. 4(a). Interestingly, it seems that the observed magneto-optical effects become stronger with increasing the frequency in the operational spectral range of our spectrometer. We can see that the maximum magnetization induced rotation and ellipticity reach 6°, corresponding to  $4.2 \times 10^2$  rad/m at an applied field of 1 kG which saturates the magnetization of the sample. Three mechanisms may be responsible for magneto-optical effects at terahertz frequencies: spin-dependent transport of free electrons, the ferromagnetic resonance, and other optical resonances affected by the magnetic ordering. Magneto-optical effects arising from the ferromagnetic resonance are expected to be strongest in the vicinity of the ferromagnetic resonance, which at an applied field of 1 kG has a frequency in the order of 1 to 10 GHz.<sup>31</sup> Since the observed magneto-optical effects increase with frequency in the range from 0.2 to 2 THz, the ferromagnetic resonance is not dominantly determining the observed magneto-optical effects.

The spectra of the magneto-optical effects are interesting to compare with the spectrum of the loss function. This function is defined such that it peaks in the presence of absorption

$$\lambda(\omega) = -\ln\left(\frac{|\tilde{E}(\omega)|}{|\tilde{E}_{ref}(\omega)|}\right),\tag{2}$$



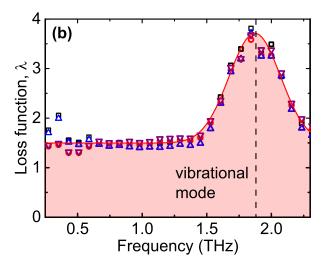


FIG. 4. (a) Spectra of the magnetization induced rotation and ellipticity of the transmitted THz radiation at 40 K and 1 kG applied field. The solid lines are linear fits used for guiding the eye. (b) The loss function (Eq. (2)) as function of frequency at 40 K for THz radiation. The different data sets correspond to the detected electric field measured parallel and perpendicular to the applied field, both measured with 0 and 1 kG applied field. The peak around 1.8 THz corresponds to a vibrational mode reported in Ref. 32. The solid line is a Gaussian fitting, serving as a guide of the eye.

where  $\tilde{E}(\omega)$  and  $\tilde{E}_{ref}(\omega)$  are the complex spectra of the transmitted electric field of the sample and a reference measurement, respectively. A broad resonance is present around 1.8 THz, corresponding to a vibrational mode observed before, using infrared transmission and reflection.<sup>32</sup> The different data sets shown in Fig. 4(b) are for the detected electric field measured parallel and perpendicular to the applied field, both measured with 0 and 1 kG applied field. This spectrum shows that the vibrational mode does not affect the magnetooptical spectra within the experimental accuracy. Despite that the spectra in Fig. 4 reveal that the ferromagnetic resonance and observed vibrational mode are not dominantly responsible for the measured magneto-optical effects, the present data cannot unambiguously indicate the microscopic origin of the magneto-optical effects. Note that if the magneto-optical effects are due to the free-electron response, the magnetization induced ellipticity and rotation are expected to show anomalies near the plasma frequency. For  $HgCdCr_2Se_4$ , the plasma frequency is expected to be in the order of 10 THz,  $^{33,34}$  which is far beyond the ZnTe crystal response used for generation and detection of the THz radiation<sup>35</sup> and thus will be the subject of future studies.

To summarise, we have demonstrated that the p-type HgCdCr<sub>2</sub>Se<sub>4</sub> ferromagnetic semiconductor shows strong linear birefringence and dichroism at THz frequencies depending on its magnetization. We observed a magnetization induced rotation and ellipticity of up to  $4.2 \times 10^2$  rad/m at an applied field of 1kG which saturates the magnetization of the sample. The origin of the observed magneto-optical effects is analogous to the anisotropic magnetoresistance at THz frequencies according to which conductivities parallel and perpendicular to the magnetization are not equal due to the complex electronic structure of the spinel.<sup>20</sup> This inequality also induces linear birefringence and linear dichroism for freely propagating THz waves. Interestingly, our measurements seem to indicate that at higher frequencies the magneto-optical effects are stronger. We believe that our observations provide new prospective to study magnetic semiconductors on picosecond time-scales and strengthen the link between carrier dynamics and THz magneto optics. Our observations may lead to a better understanding of high frequency spin-dependent transport properties and practical implementations.

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