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Terahertz magneto-optics in the ferromagnetic semiconductor HgCdCr$_2$Se$_4$

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The magneto-optical response of the ferromagnetic semiconductor HgCdCr$_2$Se$_4$ 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 $10^2$ 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 timescale.

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 length-scales led to the discovery of giant magneto-resistance in magnetic nanostructures, which caused a great advancement of spin-transport electronics (spintronics). Simultaneously, the observation of subpicosecond demagnetization by a femtosecond laser pulse triggered the field of ultrafast femtosecond magnetism and the discovery of all-optical switching.

THz radiation can serve as an ultrafast probe of 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 semiconductor 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$_{0.92}$Cd$_{0.08}$Cr$_2$Se$_4$ in the range from 0.2 THz to 2.2 THz. Similar samples are known for their unusual high magnetoresistance up to 80% and large magneto optical effects (e.g., magnetotransmission) for mid-infrared radiation. 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 $10^2$ rad/m at an applied field of 1 kG which saturates the magnetization of the sample.

The sample is a 250 μm thick crystalline plate of Hg$_{0.92}$Cd$_{0.08}$Cr$_2$Se$_4$ grown by the chemical transport reaction technique using CrCl$_3$ as a carrier agent. Its crystal structure is close to cubic and belongs to the spinel symmetry group (space group $Fdar{3}m$). 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.
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 [112] 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 $2 \times 10^5$. 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

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

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 $3^\circ$ for 1 kG bias field. This corresponds to 52 mrad over 250 $\mu$m thickness of the sample, i.e., $2.1 \times 10^5$ rad/m at an applied field of 1 kG which saturates the magnetization of the sample. These effects are huge when compared to typical magneto-optical signals measured in the visible spectrum. 29 This rotation is also large compared to some of the known...
magneto-optical signals measured in the THz spectral range.\textsuperscript{7,18} 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, is odd with respect to the applied field. From this, we can see that the maximum magnetization induced rotation is 6° 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.\textsuperscript{31} 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.

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 ($T_C \approx 120$ K), confirming that the observed effects appear due to the magnetic order of HgCdCr$_2$Se$_4$. Figure 3 also shows a curve of the magnetization which overlaps with the magnetization induced ellipticity. The rotation and ellipticity reach 6° from the expected temperature dependence of the magnetization, which is 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 $4.2 \times 10^5$ rad/m at an applied field of 1 kG which saturates the magnetization of the sample. 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

$$\lambda(\omega) = -\ln \left( \frac{|\mathbf{E}(\omega)|}{|\mathbf{E}_{\text{ref}}(\omega)|} \right).$$

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.
HgCdCr$_2$Se$_4$ 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 $4.2 \times 10^5$ rad/m at an applied field of 1 kG 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. 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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