

Anomalous Spectroscopical Effects in an Antiferromagnetic Semiconductor

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(Dated: Nov18, 2024)

Following the recent observation of anomalous Hall effect in antiferromagnetic hexagonal MnTe thin films, related phenomena at finite frequencies have come into focus. Magnetic circular dichroism (MCD) is the key material property here. In the x-ray range, the XMCD has already been demonstrated and used to visualise domains via photoemission electron microscopy (PEEM). Here we report on MCD in optical range and discuss its microscopic mechanism.

PACS numbers: 75.47.-m

I. INTRODUCTION

Several aspects of manganese telluride¹ are worth highlighting: its structure in relation to similar compounds, the semiconducting character and also, the magnetic order it supports. Regarding the last one, the anomalous Hall effect² (AHE) has long been associated only with ferromagnets and often (yet not always^{3,4}) it was assumed to be proportional to magnetisation. Occasional observations which proved these views wrong⁵ went largely unnoticed and their link to weak ferromagnetism was not fully understood¹¹. Even highly cited works⁶ about AHE in antiferromagnets (AFMs) did little to change this state of matters until a prediction appeared concerning Mn₃X systems which are nearly perfectly compensated on one hand and display large AHE on the other hand. The prediction⁷ targeting specifically X=Ir (a non-collinear AFM) motivated experimental effort which soon came to fruition: observation of AHE in this class of materials, with X=Sn, and theoretical discussion is reviewed in Ref. 8. In MnTe¹ which is collinear, the AHE was believed by a large part of the community to be absent until it has recently been rediscovered in thin layers⁹ on InP substrate independently of previous reports⁵ (and confirmed for other substrates¹⁰ and in bulk samples¹¹). Contrary to Mn₃X materials, it is a semiconductor which opens the prospects of changing the material properties by Fermi level manipulation. It is worth stressing that MnTe is a special case within the family of other II-VI materials¹ such as MnO or MnS which are cubic¹² (and do not break symmetries which imply¹³ vanishing AHE) and, on the other hand, its NiAs-structure does not support metallic character as in MnAs which is ferromagnetic moreover.

Effects at zero frequency, such as the AHE, have often their AC counterparts. Measurements of finite frequency magneto-optical (MO) effect in Mn₃X materials¹⁴ and in Mn₃XN systems¹⁵ have once again demonstrated this. Existence of magnetic circular dichroism (MCD) at optical range in non-collinear AFMs has thus been proven (in fact, temperature dependence of MOKE turns out to follow closely that of AHE in Mn₃NiN, for exam-

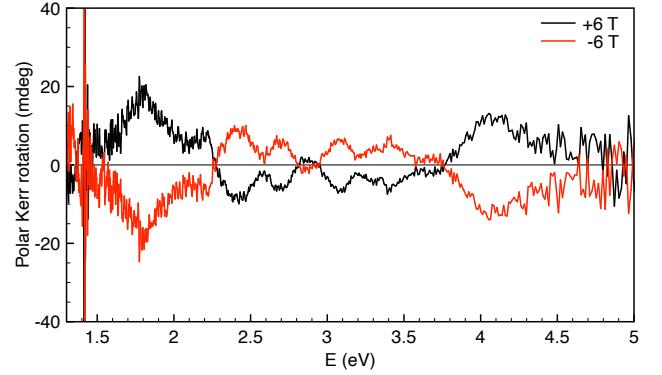


FIG. 1. Magneto-optical spectra at low temperature (40 K, well below T_N) in strong field ($B = \pm 6$ T, perpendicular to MnTe film) which is capable of creating substantial imbalance in population between domains of opposite polarity^{17,21}. Geometry of the measurement setup is described in the text.

ple¹⁵). Spectral shape of MCD in x-ray range (XMCD) in MnTe has been predicted¹⁶ and soon confirmed experimentally¹⁷. (This effect, in combination with XMLD, magnetic linear dichroism, has already been used to visualise antiferromagnetic domains¹⁸ in this material.) However, changing the applied magnetic field is difficult with experiments at synchrotron so we turn our attention to visible-range MO Kerr effect¹⁹ (MOKE), which requires only tabletop experimental setups and allow for continuous sweep of applied magnetic field. These effects express themselves as a differential absorption of left- and right-circularly polarized light in transmission or as a rotation of linear light polarization upon reflection. Moreover, despite the advantages of XMCD (in particular atom sensitivity), its large inherent line width does not allow for detailed description of the electronic structure around the Fermi level. In this case, optical MOKE measurements are more suitable due to their higher energy resolution.

In this work, we present spectra of MOKE in terms of polar Kerr rotation (for more details see Appendix A) in a collinear antiferromagnet MnTe for the first time and argue, using comparison to ab initio calculations, that

the origin of the effect is related to mechanism described in Refs. 9 and 11 (AFM with broken PT and tT symmetries¹³) rather than to canting of magnetic moments in external magnetic field that generates magnetisation akin to ferromagnets.

II. MAGNETO-OPTICAL EFFECTS IN A COLLINEAR ANTIFERROMAGNET

MnTe orders antiferromagnetically²⁰ below Néel temperature $T_N = 310$ K and its semiconducting character is discussed in Appendix A. Spectral dependence of polar Kerr rotation at low temperature ($T < T_N$) is shown in Fig. 1. The relation of MOKE to magnetic order is evidenced by its vanishing above T_N , see Fig. 2(a) where spectrum at $T = 360$ K (black) is compared to spectrum at $T = 40$ K (grey). These data can be viewed as the counterpart to XMCD in Fig. 2(a) of Ref. 17 in optical range and magnetic field (direction and strength) was chosen the same for the sake of easier comparison. Spectral amplitude of MOKE scales as an odd function of magnetic field applied in perpendicular direction to the MnTe thin film; we observe no clear saturation up to the measurement shown in Fig. 1.

Low temperature MOKE spectrum exhibits a positive (for $B = +6$ T) polar Kerr rotation at energies below 2.3 eV, followed by negative rotation region between 2.3 and 3.9 eV with several negative spectroscopic structures located near 2.5, 2.7, 3.1 and 3.4 eV. This spectral behavior resembles those reported on other Mn-based compounds, such as $\text{Mn}_3\text{NiN}^{15}$, Ni-Mn-Ga^{27} or $\text{La}_{1/3}\text{Sr}_{2/3}\text{MnO}_3^{28}$ marking Mn $3d$ electrons responsible for optical transitions in the visible region. Spectral features originate from maxima in joint density of states at various points across the Brillouin zone. Concerning the dependence on the Néel vector direction, it has been shown^{9,24,33} that upon its in-plane rotation (measured by angle θ from the direction where AHE⁹ and MOKE are forbidden by symmetry¹³), the spectra in visible range are modulated $\propto \sin 3\theta$. Deviations from this behaviour are minute, see Fig. 4 in Ref. 24, except for situations where $\sin 3\theta \approx 0$ and the absolute magnitude of MOKE is small. In our experiments, the in-plane orientation of the Néel vector should be close²⁰ to $\sin 3\theta \approx 1$ because of magnetocrystalline anisotropy in the Mn plane (in terms of crystallography, these planes are perpendicular to $[0001]$, i.e. the c -axis).

Regarding microscopic mechanism of the observed MOKE, we must first consider canting of the Mn moments induced by the applied magnetic field. At $B = 0$, the moments lie nearly perfectly in-plane (except for the very small canting corresponding likely to arc seconds¹¹) and thin layer samples tend to develop domains¹⁸ (we are unaware of any similar investigation into bulk MnTe). The effect of magnetic field applied perpendicular to Mn planes (i.e. $\vec{B} \parallel c$ as it corresponds to χ_\perp in Ref. 20) is then twofold: magnetic moments cant out of the plane

and domains of one polarity²¹ favoured^{9–11} over those of the other one. The latter effect is crucial as there would be no measurable AHE, for example, when the two polarities are equally populated. On the other hand, the effect of canting has little influence on AHE but large effect on XMCD (compare the bottom two spectra in Fig. 2(a) of Ref. 17). It is therefore important to first understand what the situation is in MOKE. At this point, a word of caution is in order. Our interpretation of MOKE (and also the interpretation of AHE in $\text{MnTe}^{9–11}$) centers on the idea that the effect can be thought of as a sum of two contributions: one which depends on B and another which depends on magnetic order. Specifically, AHE in ferromagnets has often been written as² $R_0B + R_sM$ and regardless whether the dependences are simply linear (described by the Hall constant R_0 and an AHE coefficient R_s) or more complicated, there is no room for any 'crossed term' in this scheme. The latter are nevertheless permitted by Onsager relations.³¹ Based on the present experiments, we cannot exclude the possibility of MOKE being governed by some kind of such product between a variable related to magnetic order and B .

Returning to the usual scheme (where magnetic field plays role only indirectly through magnetic order), ab initio models of MOKE spectra (explained in detail in Appendix B) show that in optical regime, canted Mn moments cannot possibly explain the signal measured at $B = 6$ T. As shown in Fig. 2(a), the MOKE spectrum measured above T_N exhibits different spectral behavior than the spectrum below T_N . At energies below 2.5 eV, the spectrum at $T > T_N$ is virtually zero. Above 2.5 eV the spectral dependence is significantly different from the spectrum at $T < T_N$ and more resembles the spectrum in Fig. 2(b) which is calculated for magnetic moments canted by as much as 5 deg. Canting of few degrees at fields up to 10 T can be expected (see p. 4 in Ref. 17), but still leads to Kerr rotation that is significantly smaller than the measured signal at $T < T_N$. This suggests that the observed effect is related to collinear order with domains (of opposite polarities) out of balance. There is an important consequence to this finding: MOKE shown in Fig. 1 should effectively only depend on magnetic order through the domain imbalance.²¹ In other words, as the canting caused by applied magnetic field has only negligible effect, the increase of signal in stronger fields should be attributed to gradual suppression of one polarity of domains¹¹.

We also observe a comparatively large MOKE signal at energies close to the MnTe band gap energy E_g (in Ref. 22, value $E_g = 1.46 \pm 0.10$ eV at 40 K was reported). This feature, shown in Fig. 3 becomes stronger and blue-shifts under applied magnetic field (we observe no similar shift in energy with spectral features far above E_g). While we leave the explanation of this phenomenon for future research, we remark that it could either be related to the large sensitivity of MnTe band gap to rotations of magnetic moments out of the manganese basal plane²³ or to magneto-optical activity of the InP sub-

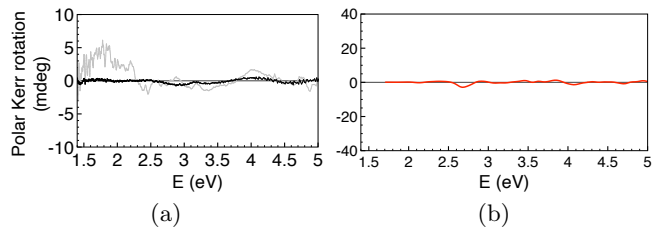


FIG. 2. (a) Experimental MOKE spectra in 1 T field above Néel temperature (black) and at $T = 40$ K (grey). Note the smaller range of vertical axis compared to Fig. 1. (b) Calculated MOKE spectra assuming canted moments.

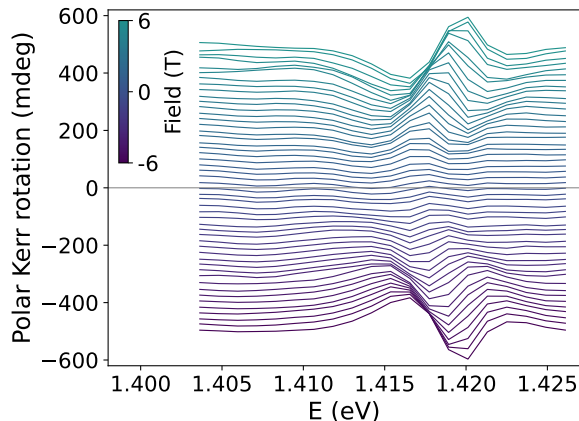


FIG. 3. Stacked plot of low-temperature MOKE at energies close to E_g of MnTe subject to magnetic field $\vec{B} \parallel c$.

strate. Accidentally, band gap of InP is very close to E_g and moreover, excitonic features in optical spectra²⁶ blue-shift with applied magnetic field. Magneto-optical experiments on bulk MnTe¹¹ should easily distinguish between these two mechanisms.

III. CONCLUSIONS

The spectral dependence of magneto-optical polar Kerr rotation was measured at temperature below T_N in a collinear antiferromagnet, which cannot be explained by ferromagnetic moment induced by external magnetic field. This way, we demonstrated that non-collinear order¹⁴ is not essential for the effect to appear, in agreement with expectations²⁵ associated to the class of magnetic systems called altermagnets (a more detailed symmetry analysis of MOKE-active collinear systems has been recently given by Radaelli³⁰). Moreover, non-vanishing linear magneto-optical effect opens possibilities for future use of collinear antiferromagnets in applications where magneto-optical effects are required to be stable under externally applied magnetic fields.

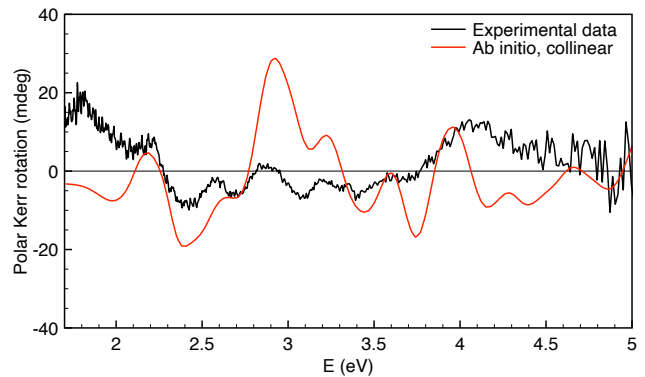


FIG. 4. Comparison of data from Fig. 1 to ab initio calculations in the collinear state as described in Appendix B.

ACKNOWLEDGMENTS

Our work benefited from discussions with T. Jungwirth, D. Kriegner and J. Kolorenč and we express our gratitude to them as well as to funding sources from GA CR (under contract 22-21974S), Austrian Science Funds, grant No. AI0656811 and University of Linz, grant No. LI113221001.

Part of the experiments were carried out at MGML (mgml.eu), which is supported by the Ministry of Education, Youth and Sports, Czech Republic within the program of the Czech Research Infrastructures (project no. LM2023065).

Appendix A: Experimental setup and sample properties

We used rotating analyzer technique to precisely measure the polar Kerr rotation of the sample. The applied magnetic field pointed in out-of-plane direction with respect to the sample surface and the angle of the light incidence was nearly normal. Our experimental technique comprised a wide spectral laser driven light source (Energetiq EQ-99X-FC) a set of parabolic mirrors to collimate the light beam from the source and focus it on to the sample in the Quantum Design PPMS cryostat with superconducting coil. The light passes two polarizers in the experimental setup. First polarizer defines the linear polarization of light prior to the reflection on the sample and the second polarizer after the sample (called analyzer) is rotating several degrees around the crossed position with the first polarizer. The light is finally detected by CCD fiber spectrometer (Ocean Insight QE Pro). From the intensity dependence on the analyzer angle, one can calculate the value of the MOKE.

Our sample was a thin layer (35 nm) of MnTe grown by MBE (molecular beam epitaxy) on InP, described elsewhere.²² In that work, optical absorption edge was confirmed to be close to E_g and hole density inferred from Hall measurements was of the order of 10^{19} per cu-

bic cm which renders Fermi energy shift very small¹¹ so that its non-zero value likely has no measurable effect on MOKE.

Appendix B: Ab initio calculations

Measured data shown in Fig. 1 can be compared to models based on ab initio calculations of electronic structure; the two methods of choice here are the augmented plane wave (APW) and projector augmented-wave (PAW) ones.

Using Kubo formula,¹⁹ conductivity tensor $\sigma_{ij}(\omega)$ is first calculated and then converted to ϵ_{ij} in a standard way²⁴.

Spectral dependence of polar Kerr rotation was calculated from the spectral dependence of permittivity tensor obtained from ab initio methods. Since the thick-

ness of the layer was only 35 nm we had to consider a model structure consisting the MnTe layer on InP substrate. For this purpose we used 4x4 Yeh's formalism for anisotropic multilayers²⁹.

Unlike in the case of canted moments, we now argue that measured Kerr rotation in optical range can be accounted for by density functional theory (DFT) calculations assuming perfectly collinear order. In Fig. 4, the calculated MOKE spectrum was scaled down by a factor of 2. This would correspond to domain imbalance of 75 % (and this parameter should of course depend²¹ on applied magnetic field).

Collinear DFT calculations have been performed using linearised APW method (implemented in `wien2k`) and data in Fig. 2(b) were generated from non-collinear PAW calculation (in VASP) with GGA and Hubbard $U = 4$ eV (Dudarev scheme³²; energy cut-off 400 eV). Lattice constants²⁰ $c = 0.671$ and $a = 0.414$ nm were assumed.

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