Optical determination of the Néel vector in a CuMnAs thin-film antiferromagnet

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Recent breakthroughs in the electrical detection and manipulation of antiferromagnets have opened a new avenue in the research of non-volatile spintronic devices1–10. Antiparallel spin sublattices in antiferromagnets, producing zero dipolar fields, lead to insensitivity to magnetic field perturbations, multi-level stability, ultrafast spin dynamics and other favourable characteristics, and may find utility in fields ranging from magnetic memories to optical signal processing. However, the absence of a net magnetic moment and ultrafast magnetization dynamics timescales make antiferromagnets notoriously difficult to study using common magnetometers or magnetic resonance techniques. Here, we demonstrate the experimental determination of the Néel vector in a thin film of antiferromagnetic CuMnAs (refs 9,10), a prominent material used in the first realization of antiferromagnetic memory chips10. We use a table-top femtosecond pump–probe magneto-optical experiment that is considerably more accessible than the traditionally employed large-scale-facility techniques such as neutron diffraction3 and X-ray magnetic dichroism measurements5,6,7,8,9,10.

Well-established optical methods3,7–9 enable the study of magnetic materials with high spatial resolution10 on short timescales11. In particular, Kerr and Faraday magneto-optical (MO) effects, which are linear in magnetization, are frequently used for the characterization of ferromagnets (FMs)12–15. For antiferromagnets (AFs), the use of MO techniques is much more challenging. Several time-resolved studies have been performed on canted AFs16–19, where the Dzyaloshinskii–Moriya interaction induces a canting of the AF spin-sublattices with an angle of about 1°, which leads to a small net magnetization. These canted AFs are much easier to study experimentally, because, despite their antiferromagnetic ordering, it is still possible to influence the spins with relatively weak magnetic fields and, moreover, Kerr and Faraday effects can be used for the characterization of their magnetic ordering. With fully compensated AFs, the signals from oppositely oriented magnetic sublattices cancel for MO effects that are linear (that is, odd) in magnetization, which leaves only MO effects that are quadratic (even) in magnetization15 or transient linear MO effects17 as suitable probes for these materials. Quadratic MO effects have been reported for many magnetic materials14,19,24–26. However, the practical use of quadratic MO effects for the characterization of magnetic materials is much less common than the use of linear MO effects15,27,28. One reason for this is that quadratic MO effects are typically much weaker than linear MO effects15. Moreover, the experimentally measured light polarization change does not alter sign when the direction of magnetization in the sample is reversed (for example, by an external magnetic field in the case of FMs). Experimentally, the second feature, in particular, is rather limiting because it significantly complicates the separation of the magnetic-order-related signal from other sources of the light polarization change (for example, strain- or crystal-structure-related)29.

In this Letter we show that this problem can be circumvented if a two-beam pump–probe detection scheme is used to measure the pump-induced demagnetization-related MO signal in the AF. Our optical technique, based on the Voigt effect, allows an unambiguous direct determination of the Néel vector orientation in thin antiferromagnetic metal films in devices, directly from measured data, without fitting to a theoretical model. Moreover, this optical measurement provides subpicosecond time resolution, which is needed for the study of magnetization dynamics in AFs.

To demonstrate this technique we use a model collinear AF, tetragonal semimetal CuMnAs (ref. 9), grown epitaxially on GaP (001). The spin axis lies in the a–b plane of CuMnAs, which registers with the GaP substrate through a 45° rotation, making CuMnAs (001) parallel to GaP {110} (Fig. 2 in ref. 9). The Néel temperature for a 500-nm-thick film of CuMnAs is 480 ± 5 K (ref. 30).

Figure 1a presents a schematic demonstrating the Voigt effect, which is closely connected to magnetic linear dichroism (MLD); see Supplementary Section ‘Magneto-optical Voigt effect’1, in a compensated AF. The rotation of light polarization (or the change of its ellipticity) occurs due to the different complex index of refraction for normal incidence light with a polarization plane oriented parallel and perpendicular to the magnetic moments14 (Supplementary Fig. 1a and Supplementary equations (1) to (6)). In common magnetic materials, polarization rotation due to the Voigt effect typically does not exceed the scale of milliradians17,19,25,26. Consequently, it is rather difficult to experimentally separate this MO signal from other (non-magnetic) sources of light polarization change. In Fig. 1b we show a situation where a second—considerably stronger—pump laser pulse illuminates the place where the probe pulse is experiencing the sample MO response. The pump-induced local heating of the sample leads to its partial demagnetization31,32 and, consequently, to a reduction in the static MO signal. The MO signal measured in this pump–probe experiment is given by (Supplementary equations (7) and (8))

$$\text{MO}(\Delta t, \varepsilon) = \frac{2P}{M} \sin 2(\varphi - \varepsilon) \delta M(\Delta t)$$

(1)

where $P$ is the corresponding MO coefficient, which scales quadratically with the sublattice magnetization $M$ projection onto the plane.

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perpendicular to the probe light propagation direction, and $\varphi$ and $\epsilon$ describe the in-plane orientation of magnetic moments and light polarization, respectively (Supplementary Fig. 1b). $\delta M$ is a pump-induced change in the magnetization that depends on time delay $\Delta t$ between pump and probe pulses. In Fig. 1c,d we show the time-resolved pump-induced change in the MO signal and sample transmission, respectively, measured at 15 K in a transmission geometry in our 10 nm thin CuMnAs epilayer by probe pulses with several orientations of the polarization plane $\epsilon$. Figure 1e presents a more detailed polarization dependence of the measured MO signal, which confirms the harmonic dependence predicted by equation (1) for the Voigt-effect-related MO signal. We stress that the key ingredient that enables separation of light polarization rotation due to the Voigt effect from that of a non-magnetic origin (for example, due to strain in the cryostat windows or the crystal structure of the GaP substrate) is the local modification of the magnetic order of the investigated CuMnAs epilayer by pump pulses, which is consequently measured by probe pulses. Any polarization changes experienced by probe pulses during their propagation in the optical set-up that are not modified by the pump pulses are not detected by this technique. Hence, it is sensitive only to changes occurring in the ~100 µm region where

Figure 1 | Experimental observation of uniaxial magnetic anisotropy in a 10 nm film of CuMnAs. a, Schematic illustration of the Voigt effect, which leads to a rotation of the light polarization plane. b, Pump-induced demagnetization reduces the probe polarization rotation. c, Pump-induced change in the MO signal measured for various probe polarization orientations $\epsilon$ as a function of time delay $\Delta t$ between pump and probe pulses at 920 nm in transmission geometry at 15 K. d, Transient transmission measured simultaneously with the data in c. e, Probe-polarization dependence of the MO signal measured at 756 nm in transmission geometry for $\Delta t = 60$ ps at 15 K (black points) and 300 K (red points). $\epsilon = 0^\circ$ corresponds to the crystallographic direction [1–10] in the GaP substrate. Solid lines are fits by equation (1) plus polarization-independent backgrounds (dashed horizontal lines). f, As in e, but in reflection geometry at 920 nm and 15 K. Error bars indicate the uncertainty in determining the displayed value from the experimentally measured data.
the pump and probe beams overlap spatially. When the sample temperature is increased to 300 K, the overall character of the measured polarization dependence does not change significantly; only the signal magnitude is larger at the higher temperature (Fig. 1e and Fig. 4). In Fig. 1f we show the polarization dependence measured in the same sample at 15 K in reflection geometry. The signal shows the same periodicity as the signal measured in transmission geometry, but the sign of the measured MO signal is opposite for these two geometries due to the opposite sign of $P$.

Without knowing the actual sign of $P$ in the studied material it is not possible to deduce the absolute Néel vector orientation from these measurements. Nevertheless, the measured data imply (equation (1)) that the spin axis in the studied 10 nm film of CuMnAs is either along the direction corresponding to $\varepsilon = 0^\circ$ or $\varepsilon = -90^\circ$. For the actual sample orientation in Fig. 1, these directions correspond to the crystallographic directions [1–10] and [110] in the GaP substrate, respectively. Consequently, we can conclude that for the 10 nm film of CuMnAs there is an in-plane uniaxial magnetic anisotropy such that the Néel vector is preferentially oriented along the [100] or [010] crystallographic direction in CuMnAs.

Figure 2 demonstrates how the ambiguity in the Néel vector orientation can be removed on a purely experimental basis. As schematically illustrated in Fig. 2a, tilting of the sample around an axis that is perpendicular to the direction of the magnetic moments leads to a reduction in the moment projection onto the plane perpendicular to the probe light propagation direction. On the other hand, tilting the sample around an axis that is parallel to the direction of the magnetic moment does not change this projection. MO signal measured at 920 nm for $\Delta t = 60$ ps (points) as a function of the sample tilt around the [1–10] substrate direction at 300 K. Solid lines depict the function $\cos^2 \delta$ describing the moment projection reduction expected for the situation shown in a (Supplementary equation (8)). As in c, for a tilt around the [110] substrate direction when the moment projection was not reduced by the sample tilt. Error bars indicate the uncertainty in determining the displayed value from the experimentally measured data.

To check the validity of the MO determination of the Néel vector orientation, we performed an X-ray magnetic linear dichroism
The XMLD was detected using total electron yield, with a probing depth of ∼3 nm. The surface of the uncapped 10 nm CuMnAs epilayer (used for the MO experiments described so far) was oxidized. Consequently, the measured XMLD signal was relatively weak in this sample. Surface oxidation was suppressed by using a 2-nm-thick aluminium capping layer (note that we also see a clear uniaxial anisotropy in the MO signal in the Al-capped sample; Supplementary Fig. 9). Figure 3a presents the XMLD spectrum measured in the capped 10 nm CuMnAs epilayer. The XMLD spectrum was obtained by taking the difference between Mn $L_{2,3}$ X-ray absorption spectra for X-rays at normal incidence with polarization parallel and perpendicular to the [110] direction of the GaP substrate. A clear linear dichroism is observed at the Mn $L_3$ edge, but it is absent at the Cu $L_{2,3}$ edges, which confirms the magnetic origin of the measured XMLD signal. To determine the spin axis orientation from this experiment, we compared the measured XMLD spectrum with that predicted by ab initio calculations. Figure 3b plots the calculated XMLD spectrum, defined as the absorption for AF moments parallel to the X-ray polarization minus the absorption for AF moments perpendicular to the X-ray polarization. The positions of the principal peaks at both the $L_3$ and $L_2$ edges are in good agreement with the experimental data. From the XMLD sign we infer that in the 10 nm CuMnAs film the spin axis is along the crystallographic direction [110] of the GaP substrate, that is, the [010] direction in CuMnAs, in agreement with the conclusions derived from the MO experiments described above. In a self-standing tetragonal crystal of CuMnAs, the directions [100] and [010] should be equivalent. However, it is the surface reconstruction of the zinc-blende GaP substrate, which determines the specific symmetry of bond alignments on the CuMnAs/GaP interface, that breaks the symmetry between these two crystal directions. Note that the present situation in CuMnAs/GaP is fully analogous to the deposition of Fe on a GaAs substrate, for which this surface-induced uniaxial magnetic anisotropy has been investigated in detail. Moreover, this interpretation of the origin of the in-plane uniaxial anisotropy in the 10 nm CuMnAs epilayer on the GaP substrate is also supported by the fact that we do not observe any in-plane uniaxial anisotropy in thicker films (Supplementary Fig. 11), where, instead, the in-plane biaxial anisotropy of bulk CuMnAs dominates.

In Fig. 4 we show the measured temperature dependence of the MO signal, from which the Néel temperature can be determined. Without the pump pulse, the temperature dependence of the quadratic MO signal can be approximated by a power law, $(T_N - T)^{-\beta}$, where $T$ is the sample temperature, $T_N$ is the Néel temperature, and $\beta$ is the critical exponent (Fig. 4a; see Supplementary Information for more details). Absorption of the pump pulse leads to a sample temperature increase $\Delta T$, which results in a change in sample MO activity, which is the signal measured in the pump–probe experiment (Fig. 4b). By fitting the measured MO data for the uncapped CuMnAs epilayer (Fig. 4c) we obtained a Néel temperature of $T_N = 365 \pm 20$ K and a pump-induced temperature increase of $\Delta T = 120 \pm 30$ K. The deduced Néel temperature agrees well with that obtained from the peak position in the temperature derivative of the sample resistivity, which is shown as the red solid line in Fig. 4c. The CuMnAs epilayer capped with aluminium
has a considerably higher Néel temperature of $T_N = 465 \pm 20$ K (Fig. 4d), which is close to the value $T_N = 480 \pm 5$ K reported previously for a 500-nm-thick CuMnAs film\textsuperscript{30}.

In summary, we have demonstrated the optical determination of the Néel vector orientation and Néel temperature in a fully compensated AF thin film of CuMnAs. We have used the MO Voigt effect combined with the pump–probe experimental technique of ultrafast laser spectroscopy. The research potential of the technique was demonstrated with measurements on CuMnAs, which is a promising material for the development of antiferromagnetic memories. We emphasize that the quadratic MO effects used in our experiments are generic in antiferromagnets, so our approach should be applicable to a broad range of other material candidates for AF spintronics. As a table-top experiment, our approach is considerably more accessible than large-facility measurements, and it also provides a uniquely direct experimental means for detecting Néel vector orientation in thin-film AFs.

Intrinsically, our optical measurements provide high time-resolution matching to the terahertz internal dynamics scale of AFs, which is beyond the reach of the electrical detection tools demonstrated previously.

**Methods**

Methods and any associated references are available in the online version of the paper.

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**References**

1. Shick, A. B., Khmelevskyi, S., Mryasov, O. N., Wunderlich, J. & Jungwirth, T. Spin–orbit coupling induced anisotropy effects in bimetallic antiferromagnets: a route towards antiferromagnetic spintronics. *Phys. Rev. B* **81**, 212409 (2010).

2. MacDonald, A. H. & Tsoi, M. Antiferromagnetic metal spintronics. *Phil. Trans. R. Soc. A* **369**, 3098–3114 (2011).

3. Park, B. G. *et al.* A spin-valve-like magnetoresistance of an antiferromagnet-based tunnel junction. *Nat. Mater.* **10**, 347–351 (2011).

4. Barthem, V. M. T. S., Colin, C. V., Mayaffre, H., Julien, M. H. & Givord, D. Revealing the properties of Mn$_2$Au for antiferromagnetic spintronics. *Nat. Commun.* **4**, 2892 (2013).

5. Marti, X. *et al.* Room-temperature antiferromagnetic memory resistor. *Nat. Mater.* **13**, 367–374 (2014).

6. Krieger, D. *et al.* Multiple-stable anisotropic magnetoresistance memory in antiferromagnetic MnTe. *Nat. Commun.* **7**, 11623 (2016).

7. Gomonay, E. V. & Loktev, V. M. Spintronics of antiferromagnetic systems. *Low Temp. Phys.* **40**, 17–35 (2014).
8. Jungwirth, T., Marti, X., Wadley, P. & Wunderlich, J. Antiferromagnetic spintronics. Nat. Nanotech. 11, 231–241 (2016).
9. Wadley, P. et al. Tetragonal phase of epitaxial room-temperature antiferromagnet CuMnAs. Nat. Commun. 4, 2322 (2013).
10. Wadley, P. et al. Electrical switching of an antiferromagnet. Science 351, 587–590 (2016).
11. Schreyer, A. et al. Neutron scattering on magnetic thin films: pushing the limits. J. Appl. Phys. 87, 5443–5448 (2000).
12. Alders, D. et al. Temperature and thickness dependence of magnetic moments in NiO epitaxial films. Phys. Rev. B 57, 11623 (1998).
13. Kuiper, P., Searle, B. G., Rudolf, P., Tjeng, L. H. & Chen, C. T. X-ray magnetic dichroism of antiferromagnet Fe₃O₅: the orientation of magnetic moments observed by Fe L₂,3 x-ray absorption spectroscopy. Phys. Rev. Lett. 70, 1549–1552 (1993).
14. Mertins, H.-C. H. et al. Observation of the X-ray magneto-optical Voigt effect. Phys. Rev. Lett. 87, 047401 (2001).
15. Mertins, H.-C. et al. Magneto-optical polarization spectroscopy with soft X-rays. Appl. Phys. A 80, 1011–1020 (2005).
16. Valencia, S. et al. Quadratic X-ray magneto-optical effect upon reflection in a near-normal-incidence configuration at the M edges of 3d-transition metals. Phys. Rev. Lett. 104, 187401 (2010).
17. Zweidin, A. K. & Kotov, V. A. Modern Magnetooptics and Magnetooptical Materials (Institute of Physics, 1997).
18. Mc Cord, J. Progress in magnetic domain observation by advanced magneto-optical microscopy. J. Phys. D 48, 333001 (2015).
19. Kirilyuk, A., Kimel, A. V. & Rasing, T. Ultrafast optical manipulation of magnetic order. Rev. Mod. Phys. 82, 2731–2784 (2010).
20. Kimel, A. V., Kirilyuk, A., Tvetkov, A., Pisarev, R. V. & Rasing, T. Laser-induced ultrafast spin reorientation in the antiferromagnet TmFeO₃. Nature 429, 850–853 (2004).
21. Kimel, A. V. et al. Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses. Nature 435, 655–657 (2005).
22. Kimel, A. V. et al. Inertia-driven spin switching in antiferromagnets. Nat. Phys. 5, 727–731 (2009).
23. Kampfrath, T. et al. Coherent terahertz control of antiferromagnetic spin waves. Nat. Photon. 5, 31–34 (2011).
24. Ferre, J. & Gehring, G. A. Linear optical birefringence of magnetic crystals. Rep. Prog. Phys. 47, 513–611 (1984).
25. Tesarova, N. et al. Systematic study of magnetic linear dichroism and birefringence in (Ga,Mn)As. Phys. Rev. B 89, 085203 (2014).
26. Bossini, D. et al. Macrospin dynamics in antiferromagnets triggered by sub-20 femtosecond injection of nanomagnons. Nat. Commun. 7, 10645 (2016).
27. Tesarova, N. et al. Direct measurement of the three-dimensional magnetization vector trajectory in GaMnAs by a magneto-optical pump-and-probe method. Appl. Phys. Lett. 100, 102403 (2012).
28. Tesarova, N. et al. Experimental observation of the optical spin–orbit torque. Nat. Photon. 7, 492–498 (2013).
29. Tesarova, N. et al. High precision magnetic linear dichroism measurements in (Ga,Mn)As. Rev. Sci. Instrum. 83, 123108 (2012).
30. Wadley, P. et al. Antiferromagnetic structure in tetragonal CuMnAs thin films. Sci. Rep. 5, 17079 (2015).
31. Beurrepaire, E., Merle, J.-C., Daunois, A. & Bigot, J.-Y. Ultrafast spin dynamics in ferromagnetic nickel. Phys. Rev. Lett. 76, 4250–4253 (1996).
32. Bigot, J.-Y., Vomir, M. & Beurrepaire, E. Coherent ultrafast magnetism induced by femtosecond laser pulses. Nat. Phys. 5, 515–520 (2009).
33. Kunes, J. & Oppeneer, P. M. Anisotropic X-ray magnetic linear dichroism at the L₂,3 edges of cubic Fe, Co, and Ni: ab initio calculations and model theory. Phys. Rev. B 67, 024431 (2003).
34. Kneessler, E. M. et al. Influence of substrate surface reconstruction on the growth and magnetic properties of Fe on GaAs(001). Phys. Rev. B 56, 8163–8168 (1997).
35. Moosbühler, R., Bensch, F., Damm, M. & Bayreuther, G. Epitaxial Fe films on GaAs(001): does the substrate surface reconstruction affect the uniaxial magnetic anisotropy? J. Appl. Phys. 91, 8757–8759 (2002).
36. Hills, V. et al. Paramagnetic to antiferromagnetic transition in epitaxial tetragonal CuMnAs. J. Appl. Phys. 117, 172608 (2015).

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Author contributions
R.P.C., V.H. and V.N. prepared the samples. P.N., P.W., B.L.G., P.M. and T.J. planned the experiments. V.S. and F.T. performed the MO experiments. V.H. performed the electrical measurements. P.W., K.W.E., F.M. and S.S.D. performed the XMLD experiment. J.K. and J.Z. performed the XMLD calculations. P.N., V.S. and T.J. wrote the manuscript with contributions from all authors.

Additional information
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Competing financial interests
The authors declare no competing financial interests.
Methods

MO experiment. A Ti:sapphire oscillator (Mai Tai, Spectra Physics) was used as the light source. Laser pulses, with a time width of 150 fs and repetition rate of 80 MHz, were divided into pump pulses (with a fluence of \(~3 \text{ mJ cm}^{-2}\)) and probe pulses (with at least 50 times weaker fluence), which were focused to the same spot on the sample (with a full-width at half-maximum (FWHM) of \(~30 \mu\text{m}\)). Unless explicitly mentioned (that is, except for Fig. 2), the experiments were performed close to the normal incidence geometry (with an angle between pump and probe beams of \(6^\circ\)). The polarization of the pump pulses was either circular or linear (see Supplementary Fig. 5, where the independence of the measured MO signal on pump polarization is demonstrated), and the probe pulses were linearly polarized with the polarization plane orientation controlled by a half-wave plate and described by angle \(\varepsilon\) (see Supplementary Fig. 1b for a definition of the coordinate system). The dynamics of the polarization plane rotation for the transmitted or reflected probe pulses was obtained by taking a difference of signals measured by detectors in an optical bridge detection system \(^{37}\). Alternatively, the probe ellipticity was measured. The same dynamics of the pump-induced change of rotation and ellipticity for \(\Delta t \geq 20 \text{ ps}\) (Supplementary Fig. 4) confirms the magnetic origin of the measured MO signals \(^{38}\) for \(\Delta t \approx 60 \text{ ps}\), which were used in the above analysis. Simultaneously with the MO signal we also measured the sum of signals from the detectors \(^{37}\), which corresponded to a probe intensity change due to the pump-induced modification of the sample transmission (or reflectivity). As a measure of the transmission changes we used the differential transmission \(\Delta T/T = (T_E - T)/T\), where \(T_E\) (\(T\)) is transmission with (without) the pump pulse \(^{39}\). The very similar dynamics measured for the MO signal and \(\Delta T/T\) (Fig. 1c,d) is a strong indication that both these signals are due to the pump-induced temperature increase \(^{40}\). We also verified that the measured probe-polarization dependence of the MO signal is connected to the orientation of the sample (Supplementary Fig. 2) and that the MO signal measured in the bare GaP substrate is considerably weaker (Supplementary Fig. 3) than those measured in samples with CuMnAs epilayers. The sample was mounted on a cold finger of a closed-cycle helium cryostat (ARS), where the temperature can be changed from 15 to 800 K. The experiments were performed without an external magnetic field applied, because control experiments showed that an external magnetic field up to 500 mT (generated by an electromagnet in the sample plane) has no effect on the measured data (Supplementary Fig. 6). The majority of pump–probe experiments were performed for the same wavelength of pump and probe pulses (see Supplementary Fig. 7 for the measured spectral dependence of the MO signal). However, for the Al-capped sample, experiments using pump pulses at 810 nm and probe pulses at 405 nm (second harmonic generated in a beta barium borate crystal) were also performed.

XMLD experiment. The XMLD study was performed on Beamline I06 at Diamond Light Source. The variable polarization undulator of the beamline allows the X-ray linear polarization vector to be rotated by 90°, allowing XMLD measurements without moving the sample. Mn and Cu \(L_{2,3}\) X-ray absorption edge spectra were obtained using the total electron yield method, with X-rays at normal incidence. The experiment was performed at a temperature of \(T = 250 \text{ K}\).

Data availability. The data that support the plots within this Letter and other findings of this study are available from the corresponding author upon reasonable request.

References

37. Rozkotova, E. et al. Coherent control of magnetization precession in ferromagnetic semiconductor (Ga,Mn)As. Appl. Phys. Lett. 93, 232505 (2008).
38. Koopmans, B., Kampen, M., Kohlhepp, J. T. & Jonge, W. J. M Ultrafast magneto-optics in nickel: magnetism or optics? Phys. Rev. Lett. 85, 844–847 (2000).
39. Horodyska, P. et al. Exciton spin dynamics in spherical CdS quantum dots. Phys. Rev. B 81, 045301 (2010).
40. Carpene, E. et al. Dynamics of electron–magnon interaction and ultrafast demagnetization in thin iron films. Phys. Rev. B 78, 174422 (2008).