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Observation of excess polar magneto-optical Kerr effect signals in asymmetric Co/Tb-multilayers

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ABSTRACT

We study the magneto-optical properties of asymmetric Co/Tb-multilayers that are designed to exhibit considerable Dzyaloshinskii–Moriya interaction (DMI) and low levels of effective perpendicular magnetic anisotropy. Accordingly, non-uniform magnetization states can occur in these films at intermediate magnetic field strengths, which cause a corresponding net magnetization reduction. Simultaneously, we observe an excess polar magneto-optical Kerr effect (MOKE) signal that increases beyond its value at saturation and is fully anticorrelated with the magnetization in its field dependency. We also find a very strong wavelength dependence of this excess polar MOKE signal that is very different from the MOKE behavior at saturation. All experimental data can be quantitatively explained by an additional polar MOKE mechanism that is associated with magnetic non-uniformities, such as, for instance, skyrmions, which have already been demonstrated to generate topological MOKE in certain compounds.

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Magneto-optical effects are an important area of fundamental materials research, and they are also heavily utilized as characterization techniques for magnetic materials due to their contactless and non-destructive nature and their capabilities in terms of spatial and time resolution.^{1–9} Hereby, the linear magneto-optical Kerr effect (MOKE) has especially high utility because it can be used for any type of material and enable full vector magnetometry.^{10–14} Its formal description is based on the linear terms in the series expansion of the dielectric tensor with respect to the Cartesian magnetization components m_i , utilizing a single magneto-optical coupling constant Q . This description turns out to be extremely accurate, with only minor reported deviations.^{15–19} Some of those are related to material symmetry and require a full tensorial description of Q ^{19–21} or the consideration of other magnetic order parameters that can generate effects analogous to linear MOKE in ferromagnets.^{22–25} It has also been documented that non-uniform magnetic structures with non-trivial topologies, such as magnetic skyrmions, can give rise to an unconventional linear magneto-optical effect, the topological MOKE.^{26,27} While this effect was expected to be vanishingly small,²⁸ there have been recent reports of topological MOKE observations for skyrmion lattice states in Gd_2PdSi_3 and CrVGe .^{27,29} So, it is presently unclear if topological

MOKE is a common occurrence in materials that have the potential to exhibit topological non-uniform magnetization structures. Thus, the purpose of this experimental study is to explore, in a quantitatively precise manner, if anomalous linear MOKE signals might also occur in asymmetric magnetic multilayers.

We fabricated a series of asymmetric multilayer samples that exhibit perpendicular magnetic anisotropy by means of DC-sputter deposition, whose layer structure is displayed in Fig. 1(a). The idea of these Pt/Co/Tb/W multilayers is twofold: first, the multilayer should not exhibit inversion symmetry, which is accomplished by making top and bottom non-magnetic interface layers from different materials, namely, Pt and W, and by making the central magnetic layer asymmetric by using Co on the bottom and Tb on top. This approach is designed to generate sizeable Dzyaloshinskii–Moriya interaction (DMI) strength, which has the potential to generate deviations from simple ferromagnetic order.^{30–32} Second, the interface induced perpendicular anisotropy can be tuned by a simple variation of the Co layer thickness t . This allows one to generate magnetic film samples that have weak overall magnetic anisotropy due to the balancing of interface induced perpendicular and magnetostatic in-plane anisotropy.^{33,34} Corresponding anisotropy measurements for our samples exhibit

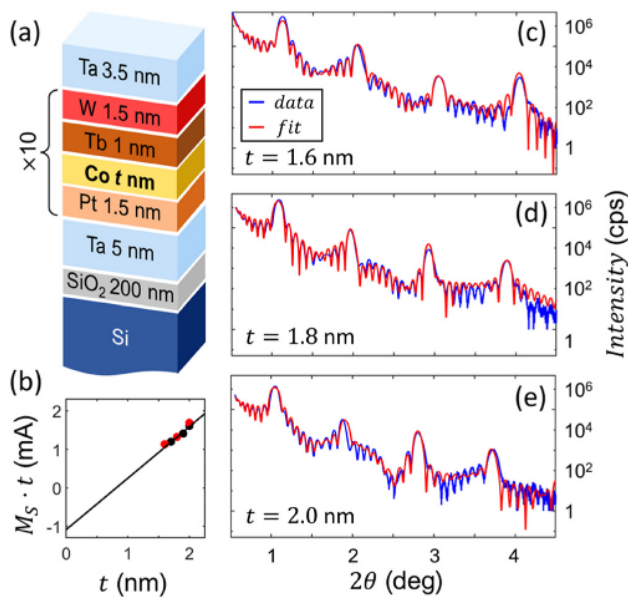


FIG 1 a) Schematic of our sample structure: the multilayer sequence consisting of Pt, Co, Tb, and W layers is repeated 10 times; all layer thicknesses are listed with the exception of the Co layer, whose thickness t was varied; b) Co thickness t evolution of the saturation magnetization M_s of our multilayer films and associated linear fit (the red dots display results for samples whose properties are shown in detail here, and the black dots additional samples); c)–e) grazing angle x-ray reflectivity data of three samples (blue lines), representing $t = 1.6$ c), 1.8 d), and 2.0 nm e) in comparison to least squares fits of the multilayer structures (red lines) [supplementary material].

exactly this dependency (Fig. S3, supplementary material). The Ta layers in our films are for improved adhesion and oxidation protection only.

Overall, the net magnetic anisotropy is modified upon tuning t and, thus, the relevance of DMI and the associated appearance of non-uniform states can be turned on or off in a very simple manner. It is also worth noting that the antiparallel orientation of the Co and Tb magnetizations, which is experimentally confirmed by the negative axis intercept at $t = 0$ in Fig. 1(b), leads to a reduced total magnetization. This shifts the critical Co thickness, at which the anisotropy is compensated, to larger t values, allowing better growth control.^{33,34} The quality of our samples has been confirmed by means of x-ray reflectivity (XRR) measurements, as shown in Figs. 1(c)–1(e) for three samples having different t . Both the total thickness and the multilayer oscillations are very clearly visible, and they match the utilized multilayer model very well (supplementary material).

After fabrication, we have measured the magnetization reversal properties of our samples by means of vibrating sample magnetometry (VSM), using a maximum out-of-plane field of 1 T and a field scan rate of 1 mT per second. In addition, we studied our samples using generalized magneto-optical ellipsometry (GME) and Kerr microscopy to quantify their precise magneto-optical properties and micromagnetic structures. Figures 2(a)–2(c) show the out-of-plane magnetization M vs field H behavior of three samples, exhibiting nominal Co-thicknesses $t = 1.6$, 1.8 , and 2.0 nm. All curves show perpendicular anisotropy behavior with a pinched central hysteresis that is indicative

of perpendicular domain formation at low fields.^{35,36} Our GME measurements in Figs. 2(d)–2(f) for the same samples and perpendicular field orientation utilize an intensity- and polarization-stabilized 5 mW laser of wavelength $\lambda = 635$ nm and corroborate all aspects of our VSM observations. γ is hereby the complex polar MOKE coefficient of the Jones calculus reflection matrix [displayed as an inset in Fig. 2(e)], which is proportional to the perpendicular magnetization [supplementary material]. Given the vector magnetometry capabilities of GME,^{37,38} we also measured the in-plane magnetization components associated with the longitudinal MOKE and transverse MOKE β simultaneously, which are vanishingly small in the entire field range. In Figs. 2(d)–2(f), we specifically display $\text{Re}(\gamma)$, $\text{Re}(\beta)$, and $\text{Re}(\gamma)$ because they allow for the visualization of hysteresis loop behavior. The corresponding imaginary components of γ , β , and γ show the same behavior, but exhibit lower signal-to-noise ratios for our samples. Overall, our GME observations corroborate the occurrence of a perpendicular domain state in the low field region with no net in-plane magnetization. The polar MOKE signal in saturation shows no relevant field dependence, and its amplitude scales with t as displayed in the inset of Fig. 2(f), replicating the magnetization dependence in Fig. 1(b). Hereby, it is important to notice that our GME measurements do not rely on any type of background subtraction, given that none of the optical elements other than the sample itself are exposed to the magnetic field (supplementary material).

However, one aspect of Fig. 2 is not compatible with this conventional picture. For the $t = 2.0$ nm sample that exhibits weak effective anisotropy as well as DMI, the VSM magnetization shows a noticeable reduction of its magnetization in the field range $\mu_0 H = 70$ –450 mT, even before conventional domain states form, which does not occur for thinner films. Furthermore, this M reduction is not accompanied by a polar MOKE reduction, but instead γ vs H seems to increase slightly above its saturation value upon reducing H in this field range.

To visualize this anomaly better, Fig. 3(a) shows a zoom-in of γ vs H data for the decreasing field hysteresis loop branch of these three samples. Here, we observe conventional behavior for the samples with $t = 1.6$ and 1.8 nm and correspondingly high magnetic anisotropy. Their magneto-optical signal is very stable upon reducing the field, exhibiting only a very minor reduction that appears synchronously with a slight perpendicular magnetization reduction, until sufficiently small field values trigger the transition into the domain state. In contrast, our data show an anomalous increase in the polar MOKE signal upon decreasing the field for the $t = 2.0$ nm sample that occurs simultaneously with the small magnetization reduction in the 70–450 mT field range, which is visualized as a gray line in Fig. 3(a). Thus, in the $t = 2.0$ nm sample and this field range, the conventional linear relationship between magnetization and linear MOKE is not conserved but is instead inverted.

This aspect is displayed in Fig. 3(b), which shows a conventional linear polar MOKE γ vs M correlation for the $t = 1.6$ and 1.8 nm samples in the $\mu_0 H > 70$ mT field range, while the $t = 2.0$ nm sample shows a clear anticorrelation in between γ and M . It is important to emphasize that we do not observe a loss of correlation, but an obvious inversion of its sign, resulting in the anticorrelation of excess polar MOKE γ with magnetization. Second order magneto-optical effects cannot be responsible for this deviation, because they are rigorously removed from the data in the GME analysis scheme.¹² So, an unconventional excess polar MOKE signal occurs in this type of sample in

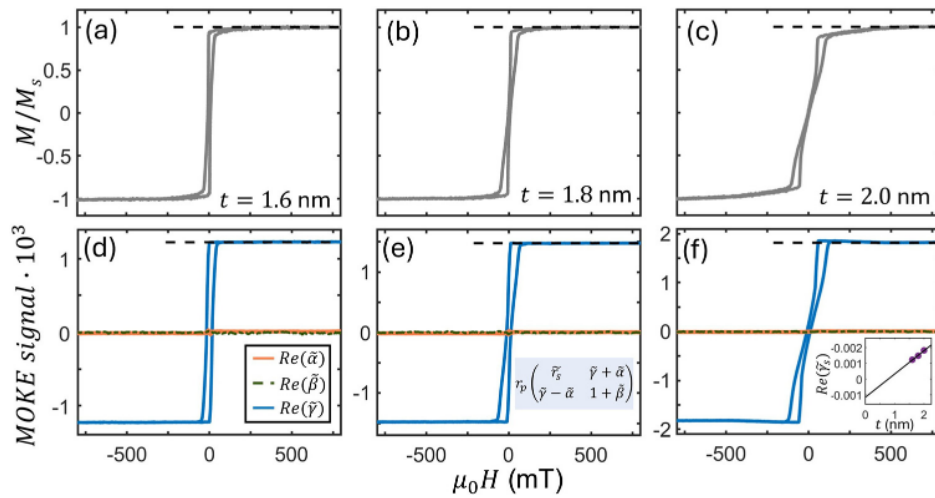


FIG 2 VSM magnetometry M vs H data a) c), compared to GME data d) f) for the same samples using an applied magnetic field H along the surface normal. The plots show data for $t = 1.6$ a) and d), 1.8 b) and e), and 2.0 nm c) and f) samples. The dashed lines in each subplot represent high field saturation values as guides to the eye. The plots d) f) display, in addition to the polar MOKE quantity $\text{Re } \gamma$ (blue line), signals associated with the longitudinal MOKE $\text{Re } \alpha$ (orange line) and transverse MOKE $\text{Re } \beta$ (green dash-dotted line), i.e., quantities that are identified by the Jones calculus reflection matrix $\hat{r}$, which is shown as an inset in e). The inset in f) displays the thickness t evolution of $\text{Re } \gamma_s$ in saturation for our multilayer films (dots) in comparison to a linear fit (line).

addition to the also present conventional linear MOKE that is proportional to M and which can be visualized by directly comparing MOKE with magnetometry data.³⁹ We also find that in this anomalous regime, the excess γ does not coincide with any in-plane MOKE signal [see Fig. 2(f)] and does not exhibit any hysteresis, as can be seen in Fig. 3(c), where the difference signal $\delta\gamma$ between the two hysteresis loop branches is plotted. We observe no difference in behavior in between the field ranges that correspond to high field saturation values and intermediate field strengths (yellow highlighted), where the excess polar MOKE occurs for both branches. This is different from other multilayer observations, in which samples exhibit a breakdown of MOKE vs M correlation due to spin-flop transitions and the different layers contributing differently to the total signal as a function of field.⁴⁰ In our case, the correlation is fully preserved, just its sign is inverted. An anomalous MOKE signal should also be present in the central multi-domain portion of the hysteresis loop. However, meaningful results are difficult to obtain in this case, because the conventional MOKE signal also changes massively in this field range, and a quantitative disentanglement of the different contributions would be problematic. Our results in Fig. 3 are corroborated by measurements on another sample having a thickness of $t = 2.1$ nm, which are shown in the supplementary material (Fig. S4).

To further elucidate the nature of the anomalous polar MOKE signal, we have conducted Kerr microscopy observations for different field strengths.⁶ Exemplary Kerr microscopy images are displayed as insets in Fig. 3(c). Here, the $t = 2.0$ nm sample shows the expected uniform state at high field and a perpendicular multi-domain state at magnetic fields below the domain nucleation threshold.³⁴ In the anomalous MOKE regime, we do not find any indication for static domain or skyrmion generation. Instead, we find seemingly uniform magnetization states on the time and spatial resolution level that we can achieve with our Kerr microscope. While we varied the data acquisition time of our

Kerr microscope by a factor of 16 with the fastest acquisition time being 60 ms, our statistical data analysis did not show any feature that would be indicative of magnetic state fluctuations in our sample. Accordingly, the magnetic non-uniformities that occur in the $t = 2.0$ nm sample and lead to the magnetization reduction must be either smaller than our lateral resolution limit of $0.5 \mu\text{m}$ or they must be fluctuation like states on time scales faster than our fastest Kerr microscopy observations, triggered by the fact that DMI and perpendicular anisotropy are designed to be in near balance. The latter possibility seems to be supported by the lack of detectable hysteresis in the intermediate field range. In addition, it should be mentioned that our observation of having an intermediate field strength occurrence of M and polar MOKE anomalies is consistent with other skyrmion observations.^{27,41}

The simultaneous nature of our M vs H and γ vs H observations can be understood in that below a threshold field of about 450 mT, a field-dependent fraction $f(H)$ of the sample is in a non-uniform magnetization state, such as for instance, a skyrmion or skyrmion fluctuation. Specifically, the fraction $f(H)$ describes the time-averaged total area covered by non-uniform magnetization entities, in which the net magnetization averages to zero. For isolated static skyrmions, this fraction represents the product of their density times the sample surface area, in which each skyrmion generates a vanishing net perpendicular magnetization. Such a non-uniform magnetic state would reduce the total perpendicular magnetization as

$$\frac{M}{M_s}(H) = 1 - f(H) \quad (1)$$

At the same time, the conventional linear polar MOKE γ_{lin} will be reduced proportionally,

$$\frac{\gamma_{lin}}{\gamma_s}(H) = 1 - f(H) \quad (2)$$

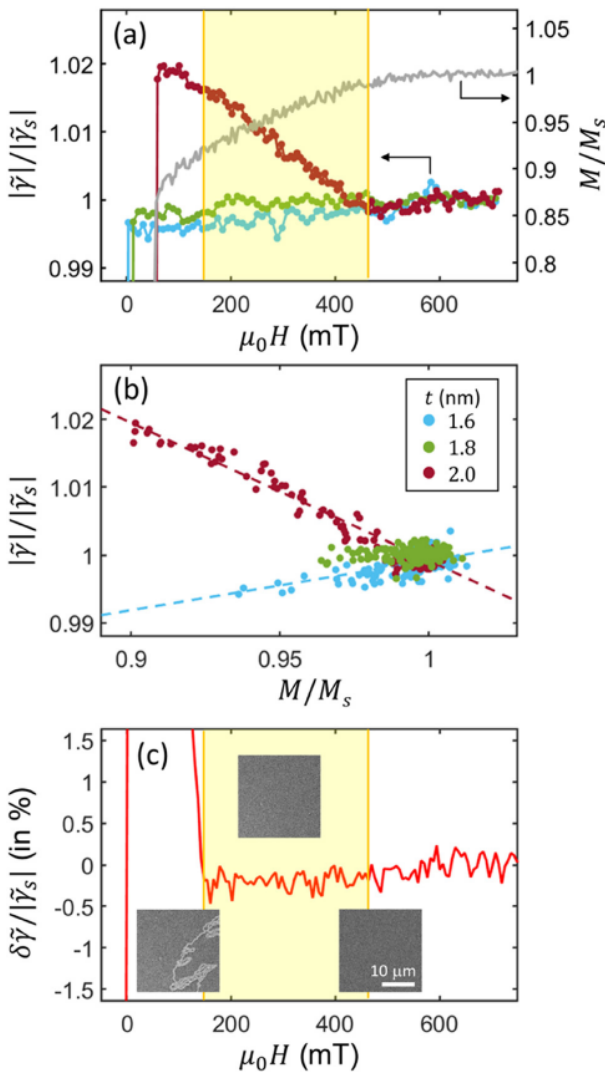


FIG 3 Saturation behavior of the polar MOKE signal γ in our samples; a) displays γ vs H for decreasing field amplitude for the $t = 1.6$ nm (blue dots), 1.8 nm (green dots), and 2.0 nm (red dots) samples; the thin colored lines are guides to the eye. For comparison, the M vs H data for decreasing field strength for the $t = 2.0$ nm sample are displayed as a gray line (the respective axes assignment is indicated by the black arrows); b) γ vs M dependence, measured for decreasing field amplitude for the $t = 1.6$ nm (blue dots), 1.8 nm (green dots), and 2.0 nm (red dots) samples; the dashed colored lines represent linear fits to the data; c) polar MOKE signal difference $\delta\gamma$ between the decreasing and the increasing field branch measurements vs H for the $t = 2.0$ nm sample; insets in c) are MOKE microscopy images taken at $\mu_0H = 60, 75$, and 360 mT, respectively, demonstrating the absence of detectable domain pattern for all fields $\mu_0H > 70$ mT on the decreasing field branch; the vertical yellow lines in a) and c) indicate the field range, where anomalous MOKE signals occur in both branches.

However, the experimentally observed polar MOKE γ_{exp} does not reduce, so that the non-uniform magnetization entities must generate an unconventional excess polar MOKE γ_E . Following optical property calculations of random skyrmion arrays, one would

expect the excess polar MOKE γ_E to be proportional to $f(H)$.⁴² Thus, in total

$$\begin{aligned}\gamma_{\text{exp}}(H) &= \gamma_s [1 - f(H)] + \gamma_E f(H) = \gamma_s + \gamma_E f(H) \\ &= \gamma_s + \gamma_E \left[1 - \frac{M}{M_s}(H) \right],\end{aligned}\quad (3)$$

with $\gamma_E = \gamma_{\text{exp}} - \gamma_s$. Accordingly, the γ_{exp} vs M anticorrelation in Fig. 3(b) is clearly possible if an excess MOKE mechanism exists that is larger per unit area than the conventional linear polar MOKE γ_{lin} . Our experimental data in Fig. 3(b) are very well described by Eq. (3), which supports our formal description.

To better characterize the excess MOKE and elucidate its origin, we have conducted spectroscopic MOKE measurements. Due to its different formal origins, a topological MOKE, for instance, should exhibit a wavelength dependence that is substantially different from the conventional linear MOKE.^{26,27,42} Figure 4(a) shows that there is indeed a very significant wavelength dependence of our excess polar MOKE γ_E/γ_s , with the anomaly becoming stronger for longer wavelengths and basically disappearing at $\lambda = 545$ nm. We also see that the field value, at which the anomaly sets in, is the same for all λ , consistent with the occurrence of magnetic non-uniformities, such as skyrmions or skyrmion-like entities.²⁷ Thus, all data in Fig. 4(a) are described by Eq. (3), making the assumption of a unique and λ -independent $M(H)$ and a λ -dependent γ_E . The solid lines in Fig. 4(a) are fits to Eq. (3) under the additional assumption that $M(H) = M_s$ for $\mu_0H > 450$ mT, which describes all data very well. Figure 4(b) shows the resulting γ_E/γ_s values vs λ in comparison to the λ dependence of the saturation MOKE signal γ_s . While γ_E/γ_s essentially vanishes for $\lambda = 545$ nm, it increases strongly for larger wavelengths and leads to significant excess polar MOKE amplitudes in our $t = 2.0$ nm Pt/Co/Tb/W multilayer sample. At the same, the simultaneously measured saturation signal γ_s shows only a very weak wavelength dependence, demonstrating the different origin of the two MOKE signals. This is also in agreement with calculations of the topological MOKE in skyrmion arrays, showing a very different Berry-phase induced λ dependence of the signal than the spin-orbit generated conventional MOKE.⁴² It should also be mentioned that the quantitative γ_E/γ_s -values, shown in Fig. 4(b), represent a lower estimate, given that they are based on VSM determined $M(H)$ -data, which might differ somewhat from $M(H)$ in the laser spot area that is utilized for our MOKE measurements.⁴³

In conclusion, we have studied the magnetic and magneto-optical properties of asymmetric multilayers that are designed to exhibit considerable DMI and low effective magnetic anisotropy. We observe a net magnetization reduction at intermediate field strengths that is associated with the occurrence of non-uniform magnetization states in a field-dependent fraction of the sample. Simultaneously, we find a polar MOKE signal in the visible spectrum that increases beyond the saturation level, exhibiting a very precise anticorrelation of magnetization and excess polar MOKE signal. All observations can be quantitatively explained by a field-dependent density of non-uniform magnetization states in such samples, which generate an excess polar MOKE signal, such as, for instance, the topological MOKE that a skyrmion state can produce. The λ dependence of the observed excess polar MOKE is very different from the saturation state polar MOKE, which demonstrates that its origin must be different from conventional MOKE,

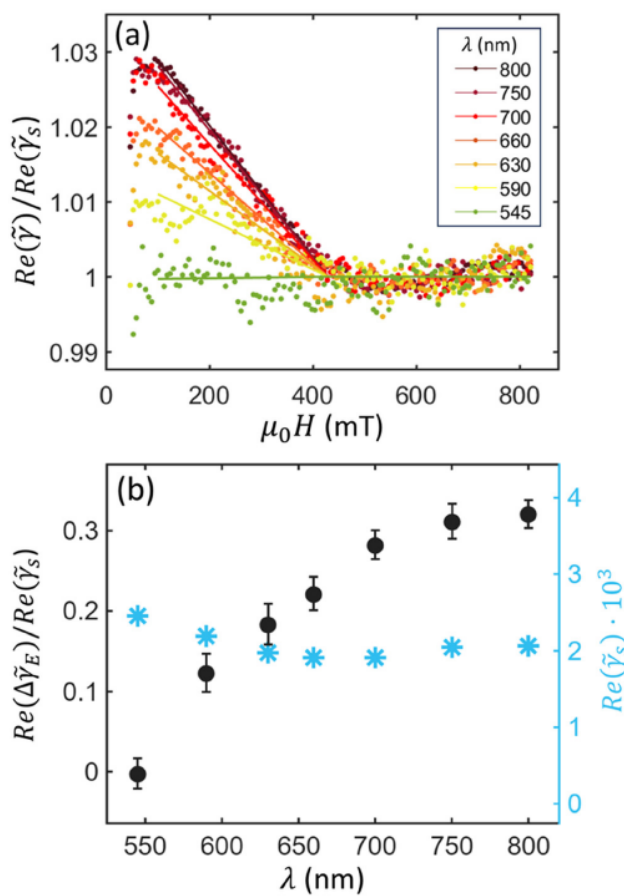


FIG 4 a) Saturation behavior of the polar MOKE signal $Re(\tilde{\gamma})$ vs H for the $t = 2.0$ nm sample and different wavelengths λ ; the experimental data are shown as points, whereas the lines represent least squares fits of the data to Eq. 3; the different wavelength data are represented by different colors as defined in the legend; b) extracted excess polar MOKE values $Re(\Delta\tilde{\gamma}_E)/Re(\tilde{\gamma}_S)$ vs λ according to Eq. 3, shown as black dots using the left-side scale, in comparison to the λ dependence of the saturation polar MOKE signal $Re(\tilde{\gamma}_S)$, shown as blue stars using the right-hand scale. Error bars for $Re(\tilde{\gamma}_S)$ are consistently smaller than the symbol size and thus not shown.

consistent with a topological MOKE interpretation of our data,^{26,27} even if our attempts to visualize the non-uniform magnetic entities were unsuccessful.

See the [supplementary material](#) for GME measurements on another sample exhibiting an anomalous polar MOKE signal and for more details on experimental methods and sample characterization results.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Carmen Martín Valderrama: Conceptualization (lead); Data curation (equal); Formal analysis (lead); Funding acquisition (equal); Investigation (lead); Methodology (equal); Project administration (equal); Resources (equal); Software (lead); Supervision (lead); Validation (equal); Visualization (lead); Writing – original draft (equal); Writing – review editing (equal). **Mikel Quintana:** Data curation (supporting); Formal analysis (supporting); Funding acquisition (equal); Investigation (supporting); Methodology (supporting); Software (lead); Validation (equal); Visualization (lead); Writing – original draft (equal); Writing – review editing (equal). **Kazuhiko Tokunaga:** Data curation (supporting); Formal analysis (supporting); Funding acquisition (equal); Investigation (supporting); Methodology (supporting); Writing – original draft (supporting). **Yoav Urbina:** Funding acquisition (supporting); Investigation (supporting); Methodology (supporting); Validation (supporting); Writing – original draft (supporting). **Yuichiro Kurokawa:** Funding acquisition (supporting); Investigation (supporting); Methodology (supporting); Supervision (supporting); Validation (supporting); Writing – original draft (supporting). **Paolo Vavassori:** Funding acquisition (equal); Investigation (supporting); Methodology (supporting); Project administration (equal); Resources (equal); Supervision (supporting); Validation (supporting); Writing – original draft (supporting). **Hiromi Yuasa:** Conceptualization (supporting); Funding acquisition (equal); Methodology (supporting); Project administration (supporting); Supervision (supporting); Writing – original draft (supporting). **Andreas Berger:** Conceptualization (lead); Funding acquisition (lead); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (lead); Writing – original draft (lead); Writing – review editing (lead).

DATA AVAILABILITY

The data that supports the findings of this study are available within the article and its [supplementary material](#).

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