Topological Kerr Effect

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We study the magneto-optical Kerr effect in SrRuO₃ thin films, at different laser wavelengths and incidence angles, uncovering regimes of temperature and magnetic field where the Kerr rotation displays two-component behavior. One component of the Kerr signal tracks the magnetization, while the second component bears a strong similarity to the topological Hall effect seen in transport experiments and is thus attributable to the presence of skyrmions. A comparison of different substrates suggests that strain plays a key role in this phenomenon. We substantiate this with a Landau theory analysis of magneto-elastic coupling to strain gradients that generates the chiral Dzyaloshinskii-Moriya exchange required to stabilize skyrmions in our intermediate thickness films. A model of electrons coupled to a skyrmion-elastic coupling is found to exhibit a significant Kerr angle at a frequency scale set by the effective Hund’s coupling, lending further support to our proposal. We term this observed optical incarnation of the topological Hall effect as the “topological Kerr effect”.

Skyrmions, particles with topologically twisted magnetic or electric dipole configurations, are of great interest as classical “topological memories” [1–3]. Originally proposed in particle physics [4], skyrmions and anti-skyrmions have been observed in numerous condensed matter settings including quantum Hall devices [5–8], magnetic metals [9–20], and ferroelectric materials [21]. The challenge to stabilize and manipulate small skyrmions for dense information storage has driven an exploration of skyrmions in new platforms such as frustrated magnets [22–27] where a strong spin twist is imposed by antiferromagnetic exchanges, and in oxide heterostructures or ultrathin films [28–36] which feature a strong chiral magnetic exchange at the interface called the interfacial Dzyaloshinskii-Moriya (DM) interaction.

In addition to uncovering new materials which host skyrmions, it is also important to find routes to probe and manipulate these topological entities. Skyrmions have been directly visualized via Lorentz force transmission electron microscopy (TEM) which detects the spatially varying winding of the in-plane component of the skyrmion magnetization [16, 17], and somewhat more indirectly using magnetic force microscopy (MFM) [37, 38]. In metallic magnets, the non-coplanar spin texture of magnetic skyrmions imprints a real-space Berry curvature onto conduction electrons, which acts as an emergent orbital magnetic field [39–41]. This emergent magnetic field induces an additional contribution to the Hall resistivity, first observed in MnSi, termed the topological Hall effect (THE) [11, 12, 42]. The converse phenomenon, where the skyrmions exhibit a transverse response to forcing, termed the skyrmion Hall effect, has also been observed in more recent experiments [43, 44], and it acts as a potential deterrent for proposed “racetrack memories” [20]. In addition to its impact on electrical transport, the Berry curvature of skyrmions also impacts the flow of heat through topological Nernst and thermal Hall effects [44–45].

In our paper, we report the measurement of a magneto-optical Kerr effect (MOKE) on thin-film samples of SrRuO₃, discovering wide regimes of temperature and magnetic fields where the Kerr angle shows signatures strikingly similar to the skyrmion topological Hall effect. The Kerr rotation, like the Hall resistivity, is proportional to the off-diagonal components of the conductivity tensor, but measured at optical frequencies rather than at zero frequency. In contrast to previous work, which found the THE in ultrathin (< 6 unit cells) films of SrRuO₃ [36, 40, 47] as well as SrRuO₃-SrIrO₃ and SrRuO₃-BaTiO₃ bilayers [33, 35, 38], here we observe both the THE and the associated Kerr effect in films of thicknesses ~ 50-120 nm, which are not expected to be dominated by the interfacial DM exchange at the film-substrate interface. We nonetheless show that a symmetry-allowed coupling between the strain-gradient and the magnetic exchange can induce an additional Dzyaloshinskii-Moriya interaction even away from the interface, and it can stabilize skyrmions, providing a theoretical basis for assigning the observed Kerr effect to skyrmions. A simple model of electrons coupled to a skyrmion crystal (SkX) is used to shed light on the origin of this Kerr effect and its dependence on frequency.

To the best of our knowledge, our experiments constitute the first observation of the THE-like Kerr effect arising from skyrmions. In line with the nomenclature used to describe the topological Hall effect of skyrmions, we term this observed effect the “topological Kerr effect”. Our work provides an optical probe of skyrmions, and highlights the future potential to use intense light fields for ultrafast manipulation of skyrmions.
FIG. 1. Comparison of (a) magnetization $M$, (b) Hall resistance $R_{xy}$, and Kerr rotation $\theta_K$ at normal incidence with (c) 594 nm, (d) 633 nm and (e) 810 nm lasers for an 88 nm thick SRO sample on LSAT. The magnetization and 810 nm Kerr rotation data both show a typical hysteresis loop. Both the Hall resistance and the Kerr rotations at 594 and 633 nm, however, show additional bump features. We note that the bump features in the Kerr rotation data differ in that they are of opposite sign when compared to those that appear in the Hall resistance measurements.

Experimental observations. — We have grown SrRuO$_3$ films using pulsed-laser deposition onto both (LaAlO$_3$)$_{0.3}$(Sr$_2$TaAlO$_6$)$_{0.7}$ (LSAT) and SrTiO$_3$ (STO) substrates. In contrast to previous studies which explored ultrathin few-unit-cell films, our SrRuO$_3$ films range in thickness from 30 – 200 nm; see Supplemental Material (SM) for details. SrRuO$_3$ films in this thickness range display ferromagnetic order below $T_c \sim 150$ K. We have measured the Kerr rotation in these films using a low-power (≈170 $\mu$W) continuous-wave laser which was weakly focused onto the sample, with the reflected beam passed through a Wollaston prism into a pair of balanced photodiode detectors. To improve signal-to-noise ratio, a chopper modulated the beam at $\approx 5$ kHz, and the signal was fed through a lock-in amplifier. We used both a typical polar Kerr configuration, in which the beam was normally incident onto the sample, and one with a large angle of incidence (around 70°). In either case, a magnetic field was applied perpendicular to the film and swept from large negative to large positive and back again to produce a hysteresis loop.

Figure 1 shows measurements of Kerr rotation $\theta_K$ at normal incidence, Hall resistance $R_{xy}$, and magnetization $M$, at temperatures from 50 K to 130 K for an 88 nm thick SrRuO$_3$ sample on LSAT. While the magnetization (a) has the shape of a typical hysteresis loop, both $R_{xy}$ (b), and the Kerr rotations at $\lambda = 594$ nm (c) and $\lambda = 633$ nm (d), show a large additional bump-like contribution similar to the THE seen in systems hosting magnetic skyrmions. Such an additional contribution was also observed in $R_{xy}$ measurements on few-unit cell ultrathin films and attributed to skyrmion THE. For longer wavelengths such as $\lambda = 810$ nm (e) and 950 nm (seen in Figs. 2 and 3 for 69 nm and 120 nm thick samples, respectively), however, this additional contribution is no longer present in our samples, and the Kerr rotation appears to track the magnetization curve. We note that $R_{xy}$ at saturation magnetization, seen at large positive or negative magnetic fields, changes sign with temperature between $T = 95$ K and $T = 80$ K, as has been reported previously. We observe that the Kerr angles at $\lambda = 594$ nm and $\lambda = 633$ nm exhibits a similar sign change, but occurring between $T = 130$ K and $T = 115$ K, while this sign change is entirely absent at the highest wavelength.

Directly from the raw MOKE data in Figs. 1(c) and 1(d), it is clear that the Kerr angle $\theta_K$ generically exhibits two components: one component tracks the $M$ hysteresis loop (up to a possible sign flip discussed above), while the second component leads to the bump-like feature. This justifies separating $\theta_K$ into a corresponding “normal contribution” $\theta_{K,M}$, and an additional contribution $\theta_{K,T}$, which we call the “topological Kerr angle” and attribute to skyrmions, so that $\theta_K = \theta_{K,M} + \theta_{K,T}$. We illustrate this analysis in Fig. 2 for $\lambda = 633$ nm at $T = 80$ K, where we choose $\theta_{K,M}$ as follows. We assume $\theta_K$ at $\lambda = 950$ nm as a reference signal, since it appears to track $M$ (see SM for a detailed discussion) and thus does not show an additional contribution $\theta_{K,T}$ (as discussed later, we attribute this to the strong frequency dependence of $\theta_{K,T}$). We next rescale this reference signal (see dashed line in Fig. 2(a)), and subtract it from the $\lambda = 633$ nm Kerr signal loop (line with dots), choosing the scaling factor and its sign in order to cancel off the Kerr rotation at saturation magnetization. This defines $\theta_{K,M}$ over the entire loop, and the difference signal $\theta_K - \theta_{K,M}$ (gray shaded area) defines the topological Kerr angle $\theta_{K,T}$. The full field and temperature dependence of $\theta_{K,T}$ quantified in this manner is displayed in Fig. 2(b), where the dots indicate the coercive field $B_c$ defined by the zero crossing of $\theta_K$ at $\lambda = 950$ nm. We observe that the largest $\theta_{K,T}$ occurs roughly around $B_c$, which is consistent with assigning this feature to skyrmions, since skyrmions have been observed, in MFM measurements, to occur in the vicinity of magnetization reversals.

We have carried out similar measurements on films of varying thicknesses, finding a nonzero $\theta_{K,T}$ for thicknesses in the range $\sim 70$-120 nm; see SM for additional data. The fact that $\theta_{K,T}$ vanishes in the 88 nm film for $\lambda \gtrsim 800$ nm, but is nonzero at $\lambda = 594$ nm and 633 nm, signals its strong frequency dependence. We find no sign of $\theta_{K,T}$ in very thick films, where skyrmions are...
FIG. 2. (a) Kerr rotation measured with a 633 nm laser at 80 K for a 69 nm SRO film on LSAT, showing the contribution from the magnetization $\theta_{K,M}$ (inferred from data taken with a 950 nm laser) and the additional contribution $\theta_{K,T}$. (b) Colour plot of the $\theta_{K,T}$ in the temperature - field plane. $B_C$, the field strength where the net magnetization becomes zero, is indicated by the red dots.

expected to be absent. In some thinner films ($\sim 30$ nm), bumps were clearly present in $R_{xy}$, signalling a THE, but such an additional contribution was absent in our MOKE data for the available wavelengths. The most likely explanation for this is that $\theta_{K,T}$ for thinner films becomes much smaller for the wavelengths used in our measurements. Finally, films grown on LSAT which has greater lattice mismatch (1.4%) with SrRuO$_3$ when compared with the STO substrate (lattice mismatch: 0.45%), exhibit a larger, and broader (in field), $\theta_{K,T}$, suggesting that strain plays a crucial role in stabilising skyrmions.

In a previous work on ultrathin SrRuO$_3$ films, it was suggested that the bumps observed in $R_{xy}$ did not, in fact, reflect an intrinsic THE, but instead arose from the temperature dependence of the bulk anomalous Hall effect and $T_c$ inhomogeneities across the sample [49]. However, this alternative model is unable to explain our MOKE results (see SM [48] for details). We thus attribute the observed bumps to an intrinsic effect.

Further evidence in favor of an intrinsic second component in $\theta_K$ comes from Kerr rotation measurements at non-normal incidence. Figure 3 shows one such dataset, for a 70° incidence angle on a 120 nm film grown on LSAT. The s-polarized data (b) is similar to that shown previously, with $\theta_{K,M}$ changing sign just above 95 K. The p-polarized data (a) on the other hand, has $\theta_{K,M}$ which stays fairly constant throughout the temperature range shown. In both cases the additional contribution $\theta_{K,T}$ is clearly visible, as shown in (c) and (d). Despite the dramatic differences in the temperature dependence of $\theta_{K,M}$, the additional contribution $\theta_{K,T}$ remains similar in sign and magnitude between the different polarizations. The fact that polarization affects the two portions of the signal so differently indicates that they are indeed distinct features, again suggesting that the additional signal is intrinsic, and plausibly attributable to skyrmions.

**Strain-gradient induced skyrmions** — In ultrathin films, the broken inversion symmetry at the film-substrate interface is well-known to lead to a chiral interfacial-DM exchange which drives skyrmion formation. Here, we show that strain gradients, which are significant away from this ultrathin limit, provide a second microscopic source of inversion breaking. For a cubic crystal, the usual free energy functional for the slowly varying magnetization density in the ordered phase,

$$F_m = \int d^3 r \left[ \frac{J}{2} \sum_{i,j} \nabla_i m_j \nabla_j m_i + u \sum_i m_i^2 + \ldots \right],$$

with $|\vec{m}| = 1$ and $i,j = (x,y,z)$, must be supplemented by symmetry-allowed magneto-elastic couplings between the strain tensor $\epsilon$ and the magnetization density $\vec{m}$,

$$F_{me} = \alpha \int d^3 r \left( \frac{\partial \epsilon_{zz}}{\partial z} \left( m_z \frac{\partial m_z}{\partial x} - m_x \frac{\partial m_z}{\partial y} - m_y \frac{\partial m_z}{\partial y} \right) + \nabla^2 \epsilon_{zz} m_z^2 + \ldots \right) + \gamma \int d^3 r \left( \epsilon_{zz} m_z^2 + \ldots \right),$$

FIG. 3. Kerr rotation at 633 nm for a 120 nm SRO sample on LSAT, taken at oblique incidence. Depending on the choice of polarization, the normal Kerr rotation part of the signal (inferred from data taken with s-polarization at $\lambda=950$ nm, and shown as a dotted line) has a very different temperature dependence. In the s-polarized data (b) it is similar to normal incidence, and changes sign at around 100 K, whereas in the p-polarized data (a) it remains fairly constant with temperature. In contrast, the additional contributions for p- and s-polarized data, shown after subtracting the normal part in (c) and (d) respectively, look almost identical.
with couplings $\alpha, \gamma$. Thus, $D(z) = \alpha \partial_{zz} / \partial z$ acts as a local Rashba-type DM interaction, while $A(z) = \gamma \epsilon_{zz}$ acts as a local uniaxial anisotropy. Details of this symmetry analysis are given in the SM [48].

Fig. 4(a) schematically illustrates three regimes important in such thin films. (i) For ultrathin films with height $w < z_s$, the film conforms to the substrate lattice constant $a \approx 4\AA$ and scaled magnetic field $H$, showing ferromagnetic (FM) phase with moment canted by angle $\varphi$ with respect to the plane of the film and a skyrmion crystal (SKX) phase stabilized by strain gradients [50]. We fix $z_s = 10a$, $z_b = 100a$, and choose $D_s = 0.4J$ and $A_0 = 0.3J$.

To study the phases induced by magneto-elastic couplings, we consider columnar magnetic configurations $\vec{m}(x, y)$ independent of $z$. Integrating the free energy over the film thickness, then leads to an effective 2D model which contains, in addition to the spin stiffness $J = \int_0^w dz J$, a Rashba coupling $D = \int_0^w dz D(z)$, and a uniaxial anisotropy $A = \int_0^w dz A(z)$. A 2D variational computation of collinear, spiral, and periodic SKX within a circular cell ansatz [30, 34], leads to the phase diagram shown in Fig. 4(c); details are in the SM [48]. Here, we have considered a square SKX to conform with the underlying SrRuO$_3$ atomic lattice, but we expect qualitatively similar results for other crystal geometries. Our key finding is the emergence of an island of Néel SKX, within a ferromagnetic phase (FM). As discussed in the SM [48], this SKX is distinct from the interfacial-DM driven phase in the $w < z_s$ ultrathin regime.

**Kerr rotation from skyrmions:** In order to describe the observed Kerr rotation, we couple electrons to a skyrmion texture, assuming a single-orbital square lattice Hamiltonian $H = -t \sum_{(ij)\alpha} \langle c_i^\dagger c_j \rangle + \epsilon_{ij} c_i^\dagger c_j^\dagger \sim J_{\text{H},0} \sum_i \hat{m}_i^\dagger \hat{s}_i^\dagger$. Here, $\hat{m}_i$ is the magnetization arising from skyrmions, with $|\hat{m}_i| = 1$, the electron spin $\hat{s}_i = \frac{1}{2} c_i^\dagger \sigma_{\alpha\beta} c_i$ and the effective Hund’s coupling $J_{\text{H},0}$ originates from underlying multiorbital interactions in the Ru orbitals. This leads to a band structure consisting of a low-energy sector, where the electron spin locally points along $\hat{m}_i$, and a high-energy sector where they point antiparallel. (i) The resulting dispersion has $|C| = 1$ Chern bands, featuring pairs of bands which disperse similarly, with energies differing simply by a constant shift $\sim J_{\text{H},0}$, as shown in Fig. 5(a). (ii) Furthermore, the non-coplanar and spatially varying skyrmion spin texture permits nonzero matrix elements $\langle \vec{k} h | \vec{j} | \vec{k} h \rangle$ of the (spin-conserving) current operator between low and high energy bands ($\ell, h$). These two effects, (i) and (ii), combine to yield a non-trivial frequency-dependent conductivity tensor, leading to Kerr rotation $\sim \text{mrad}$ from skyrmions at a frequency scale set by $J_{\text{H},0}$. Based on our optical observations, this effective scale $J_{\text{H},0} \sim 3eV$. As shown in Fig. 5(b), for a
cut through the phase diagram with \( w = z_b \), we find \( \theta_{K,T} \neq 0 \) in the non-coplanar SkX phase, while it vanishes in the collinear FM phases; see SM [48] for further details. Given the simplicity of our model, which ignores the multiorbital physics, and the complexity of factors which determine the Kerr angle, it is reasonable that we do not quantitatively capture the observed larger value of \( \theta_{K,T} \). The perpendicular magnetization \( M_z \), shown in Fig. 5(c), exhibits small jumps at the phase boundaries.

Conclusion. — We have measured a nontrivial Kerr signal in thin films of SrRuO\(_3\) over wide ranges of magnetic field and temperature, and different incidence angles, which appears to behave similar to the THE seen in \( R_{xy} \). Our results suggest that this Kerr signal is intrinsic and may be ascribed to skyrmions. We have presented theoretical results showing how strain gradients may stabilize skyrmions away from the previously studied ultrathin film regime, and lead to a topological Kerr effect. This naturally explains why SrRuO\(_3\) on LSAT exhibits a stronger topological Kerr signal, due to its greater lattice mismatch which leads to more significant strain gradients when compared with SrRuO\(_3\) on STO. In future, it would be valuable to experimentally explore the full frequency and strain dependence of this Kerr signal, and use magnetic force microscopy or Lorentz TEM measurements to directly visualize the skyrmions. This could set the stage for future studies of skyrmion-light interactions and its technological applications.

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Supplemental Material

X-Ray Diffraction Data

X-ray diffraction data, plotted in Fig. 6(a), shows the clear changes in the SRO thin-film lattice as the thickness is varied. The out-of-plane lattice parameter shown in Fig. 6(b) is observed to increase as the film thickness is decreased, which can be attributed to the in-plane lattice parameters decreasing towards the LSAT value while the unit cell volume remains constant. In addition, as shown in Fig. 6(c), the peaks are slightly asymmetric. This could be caused by the lattice parameter varying along the out-of-plane direction, as is proposed in our theoretical model. The lattice parameters are directly compared in reciprocal space mapping of SRO thin films with different thicknesses (Fig. S1(d)), also suggesting an increased out-of-plane lattice parameter as well as a decreased in-plane lattice parameter in the thinner film. Moreover, a clear peak broadening in both out-of-plane and in-plane directions is present due to the variation of the lattice parameter along the out-of-plane direction, which provides strong evidence of a gradual strain relaxation throughout the films.

MOKE on Other Samples

The MOKE data was strongly affected by the thickness of the samples, as well as the choice of substrate. Figure 7 shows data for a few different samples. Between the two substrates, the STO samples show much smaller and narrower bump features. Between the different thicknesses shown, the difference was mostly in what the temperature range these bump features occurred, although going to thinner samples (the next being a 51 nm thick sample on LSAT) resulted in these features disappearing entirely. One consistent feature in all of the samples with the topological Kerr signal was the presence of a change in sign of the regular Kerr signal, although it remains unclear as to why this should be the case.

Long-Wavelength Kerr Data vs Magnetization

In order to remove the part of the signal attributed to the ordinary Kerr rotation (i.e. that which is proportional to the magnetization), we have used rescaled long-wavelength data, where no additional bump features were observed. Figure 8 shows comparisons of long-wavelength data with direct magnetization measurements. As shown, the potential issues with using the direct measurements is that slight miscalibrations of the field strength can lead to artificial bump features, and the bulk nature of these measurements may not accurately characterize the small area which is probed by the laser. For these reasons, we have preferred to use the long-wavelength data, as this ensures we are comparing measurements of the same location and with the same field strength.

Alternative Model for Hall Measurements

Recently it was suggested that the Hall effect data on ultrathin SRO films could be explained by inhomogeneity characterized by a distribution of effective temperatures. As a simple model, we can write the signal under increasing field as a step function with a temperature dependent saturation value $A(T)$ and coercive field strength $B_c(T)$

$$S_0(B, T) = A(T)\Theta(B - B_c(T))$$

(3)
We then take the observed signal to be a combination of signals at different temperatures, characterized by a normal distribution with a width $\sigma_T$

$$S(B, T) = \int \text{norm}(\tilde{T}/\sigma_T)S_0(B, T + \tilde{T})d\tilde{T}$$

(4)

Taking the temperature dependence of $A$ and $B_c$ to be approximately linear, and defining parameters $\alpha$ and $\beta$ so that $A(T + \tilde{T}) \approx A(T) + \alpha(T)\tilde{T}/\sigma_T$ and $B_c(T + \tilde{T}) \approx B_c(T) - \beta(T)\tilde{T}/\sigma_T$, where $\beta$ is
FIG. 7. Data at selected temperatures for various samples. A notable difference in the samples on STO is that they have much smaller and narrower bumps compared to those on LSAT.

taken to be positive (as is the case physically), gives a solution

$$S(B, T) = A(T) \cdot \text{erf} \left( \frac{B - B_c(T)}{\beta(T)} \right) + \alpha(T) \cdot \text{norm} \left( \frac{B - B_c(T)}{\beta(T)} \right)$$

(5)

where \(\text{erf}\) and \(\text{norm}\) are the standard error function and normal distributions respectively. The first term produces a typical magnetization loop, and the second produces features that closely resemble the observed bumps. The constraint of this model, however, that the amplitude of these bump features is set by \(\alpha\), as shown in figure 9 (a). In our data (figure 9 (b)-(e)) the saturation value of the signal decreases with temperature for both \(R_{XY}\) and \(\theta_K\), which means that \(\alpha\) must be a negative number. While the bumps in the \(R_{XY}\) data do indeed also have a negative amplitude, those seen in \(\theta_K\) have a positive amplitude and therefore cannot be explained by the model presented above.

**Landau theory with magneto-elastic couplings**

We formulate a Landau free energy with magneto-elastic couplings and show that strain gradients can induce Dzyaloshinsky-Moriya (DM) interactions and stabilize a skyrmion crystal phase. We consider a minimal model of a thin film in the presence of a lattice-parameter relaxation that has a three-part configuration (Fig. 10(a)): (i) tetragonal interface-constrained part, (ii) coherent lattice-parameter relaxation and (iii) strain-free bulk-like section which locally has a cubic symmetry.
In a cubic environment, the free energy describing a ferromagnetic order is given by

$$F_m = \int dr \left( \sum_{i=x,y,z} \frac{J}{2a} (\nabla m_i)^2 + \frac{u}{a^3} \sum_{i=x,y,z} m_i^4 \right) - \frac{H}{a^3} m_z. \quad (6)$$

$m(r)$ is the magnetization field with $|m|^2 = 1$. The first anisotropy term is quartic and plays a role in selecting the magnetization directions. Normalization of the couplings by a length scale $a$ is introduced so that each coupling has an energy unit.

Using cubic symmetry, we can write down a magneto-elastic free energy $F_{me} = \int dr F_{me}$ as shown below.

$$F_{me} = F_1 + F_2 + \cdots, \quad (7)$$

$$F_1 = \frac{1}{a} \left( \alpha_1 \partial_x \epsilon_{xx} + \alpha_2 \partial_x \epsilon_{yy} + \alpha_3 \partial_x \epsilon_{zz} + \alpha_4 \partial_y \epsilon_{xx} + \alpha_5 \partial_y \epsilon_{yy} \right) (m_z \partial_x m_x - m_x \partial_x m_z) + \cdots \quad (8)$$

$$F_2 = \frac{\gamma}{a^3} \left( \epsilon_{xx} m_x^2 + \epsilon_{yy} m_y^2 + \epsilon_{zz} m_z^2 \right). \quad (9)$$

$\epsilon_{\alpha\beta}$ is the strain field, defined relative to the cubic structure. The dots refer to other magneto-elastic terms, which we do not consider here. One can check that $F_{me}$ is invariant under the generating elements of the octahedral group $O_h$. $F_1$ contains DM-like terms coupled to strain gradients.
and survives in a non-uniform strain field. \( \mathcal{F}_2 \) contains terms which play a similar role to single-ion anisotropies. These are present, for example, in a tetragonal environment, where \( \epsilon_{xx} = \epsilon_{yy} \neq \epsilon_{zz} \). The total free energy can then be expressed as

\[
F[m] = \int_{\text{film}} d\mathbf{r} (\mathcal{F}_m + \mathcal{F}_{me} + \mathcal{F}_I) + \cdots ,
\]

(10)

where the dots account for the free energy of elastic degrees of freedom. Its configuration is fixed to characterize the thin-film model. The new term \( \mathcal{F}_I \) constitutes the interfacial DM term

\[
\mathcal{F}_I = \frac{D_I}{a} \delta(z) [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)].
\]

(11)

Unlike in an ultrathin film where a skyrmion crystal is stabilized by \( \mathcal{F}_I \), our thick samples with the lattice-parameter relaxation accommodate strain-gradient-induced DM terms which are more

FIG. 9. (a) Example plots generated using equation 5 and various \( A \) and \( \alpha \) values, with \( B_c = 0.2 \) and \( \beta = 0.1 \) being held constant. (b) Hall and (c) MOKE data for an 88 nm thick sample on LSAT. Black arrows indicate the saturation values, which are plotted in (d) and (e) as a function of temperature. In both cases, the saturation values are clearly decreasing with temperature, which means \( \alpha < 0 \). In the \( R_{xy} \) data, the bumps have a negative amplitude, matching the prediction of equation 3 but in \( \theta_K \) they are positive, making the data impossible to reproduce with this model.
crucial for stabilizing a skyrmion crystal. This naturally explains the appearance of skyrmions in ultrathin films as mainly driven by the interface, their disappearance in several-layer thin films, and the re-appearance in a thicker sample like in ours thanks to the strain gradient.

To see how DM interactions can arise from a strain-gradient lattice-parameter relaxation, we characterize the film model by the following displacement field.

\[
\mathbf{u} = (u_x, u_y, u_z),
\]

\[
u_x = \begin{cases} 
\epsilon_{xx,1}x & \text{for } 0 < z < z_s, \\
\epsilon_{xx,1} \frac{z-z_b}{z_s-z_b} x & \text{for } z_s < z < z_b, \\
0 & \text{for } z_b < z < w.
\end{cases}
\]

\[
u_y = \begin{cases} 
\epsilon_{yy,1}y & \text{for } 0 < z < z_s, \\
\epsilon_{yy,1} \frac{z-z_b}{z_s-z_b} y & \text{for } z_s < z < z_b, \\
0 & \text{for } z_b < z < w.
\end{cases}
\]

\[
u_z = \begin{cases} 
\frac{z - z_b}{2} (2z - z_s - z_b) & \text{for } 0 < z < z_s, \\
\frac{\epsilon_{zz,1}}{2} (z - z_b)^2 & \text{for } z_s < z < z_b, \\
0 & \text{for } z_b < z < w.
\end{cases}
\]

\(\epsilon_{xx,1}, \epsilon_{yy,1}\) and \(\epsilon_{zz,1}\) are the constant, diagonal strain components characterizing the tetragonal part (i) of the film. \(z\) is the out-of-plane distance from the interface. \(w\) is the film thickness. \(z_s\) and \(z_b\) determine how much proportion of the film belongs to the interface-constrained region, the lattice-parameter relaxation and the strain-free part. Figure 10(b) shows the \(z\)-dependence of the diagonal strain components, which relate to the spatially varying lattice parameters.

In the film model, \(F_1\) vanishes in region (i) and (iii) due to the lack of strain gradient. In the lattice-parameter-relaxation region (ii), the first two terms in \(F_1\) survive. Therefore, \(F_1\) takes a Rashba-DM form:

\[
F_1 = \frac{D(z)}{a^2} [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)],
\]

where \(D(z)\) is shown in Fig 10(c). \(D_s\) is the DM coupling induced by strain gradient. \(F_2\) takes an easy-plane/axis form with a spatially dependent coupling.

\[
F_2 = \frac{A(z)}{a^3} m_z^2,
\]

where the profile of \(A(z)\) is given by Fig 10(d).

**Variational phase diagrams**

We examine the stability of a skyrmion crystal phase in the free energy formulated in the previous section:

\[
F[m] = \int_{\text{film}} d^3 r (F_m + F_{mc} + F_I) + \cdots,
\]

\[
F_m = \frac{J}{2a} \sum_{i=x,y,z} (\nabla m_i)^2 + \frac{u}{a^3} \sum_{i=x,y,z} m_i^4 - \frac{H}{a^3} m_z,
\]

\[
F_{mc} = \frac{A(z)}{a^3} m_z^2 + \frac{D(z)}{a^2} [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)],
\]

\[
F_I = \frac{D_I}{a} \delta(z) [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)].
\]
FIG. 10. (a) Schematic plot of the three-part film model: (i) \( z < z_s \) substrate-constrained part with a local tetragonal symmetry, (ii) \( z_s < z < z_b \) lattice-parameter relaxation and (iii) \( z > z_b \) bulk-like strain-free part which has a local cubic symmetry. \( z \) is the distance from the interface in the normal direction, and \( w \) is the thickness of the film. Its value relative to \( z_s \) and \( z_b \) determines the thin-film structure. (b) Diagonal strain components as a function of \( z \). Strain gradients are present in the lattice-parameter relaxation region. (c) \( z \)-dependence of the strain-gradient-induced DM coupling \( D(z) \) depicting a constant coupling strength \( D_s \) in the relaxation region in our model. (d) \( A(z) \) of the magneto-elastic single-ion-like anisotropy \( A(z)m_z^2 \) as a function of \( z \).

We consider columnar magnetic orders uniform in the out-of-plane direction, namely \( \mathbf{m}(\mathbf{r}) = \mathbf{m}(x, y) \). The free energy is then reduced to an effective free energy defined in the xy-plane, \( \tilde{F}[\mathbf{m}] = \tilde{F}[\mathbf{m}] \),

\[
\tilde{F} = \int d^2r (\tilde{F}_m + \tilde{F}_{me} + \tilde{F}_I) + \cdots , \tag{22}
\]

\[
\tilde{F}_m = \frac{Jw}{2a} \sum_{i=x,y,z} (\nabla' m_i)^2 + \frac{uw}{a^2} \sum_{i=x,y,z} m_i^4 - \frac{Hw}{a^3} m_z , \tag{23}
\]

\[
\tilde{F}_{me} = \frac{\tilde{A}}{a^3} m_z^2 + \frac{\tilde{D}}{a^2} [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)] , \tag{24}
\]

\[
\tilde{F}_I = \frac{\tilde{D}_I}{a^2} [(m_z \partial_x m_x - m_x \partial_x m_z) + (m_z \partial_y m_y - m_y \partial_y m_z)] , \tag{25}
\]

where \( \nabla' \) is the 2D gradient, \( w \) is the film’s thickness, and

\[
\tilde{A} = \int_0^w A(z)dz, \tag{26}
\]

\[
\tilde{D} = \int_0^w D(z)dz , \tag{27}
\]

\[
\tilde{D}_I = aD_I . \tag{28}
\]

We can now view \( \tilde{D} + \tilde{D}_I \) as the total DM coupling. In the rest of the section, we will set \( a = 1 \) and \( z_s = 10a \), where \( z_s \) is defined in Fig. 10(a). Figure 11 shows how \( \tilde{D} + \tilde{D}_I \) changes as a function of the thickness \( w \). In the ultrathin limit \( w \to 0 \), the interfacial contribution dominates, and total DM coupling declines as the thickness grows. In a thicker film, the strain-gradient DM becomes more important, especially when the thickness corresponds to \( z_s < w < z_b \).

The variational states are obtained by minimizing the free energy with respect to the ansatze for the following three magnetic orders: skyrmion crystal, spiral order and uniform magnetization.

(1) Square skyrmion-crystal ansatz is defined by its square unit cell. The unit-cell dimension is given by \( 2R_{skx} \times 2R_{skx} \). The magnetization vector in the unit cell is described by a circularly symmetric function:

\[
\mathbf{m}_{skx} = \left( \frac{x}{r} \sin \Theta(r), \frac{y}{r} \sin \Theta(r), \cos \Theta(r) \right) , \tag{29}
\]
FIG. 11. (a) Effective strain-gradient DM coupling \( \tilde{D} \) as a function of the thickness, featuring an increase in the relaxation region \( z_s < w < z_b \) and becomes less important for \( w \gg z_b \). (b) Total effective DM coupling \( \tilde{D} + \tilde{D}_I \) with a dominant interfacial contribution at small thickness \( w < z_s \), while the strain-gradient contribution becomes important in a thicker film, e.g. for \( w > z_s \). Here, we have set \( D_I = 0.6 J, D_s = 0.4 J, z_s = 10 a \) and \( z_b = 100 a \).

where \( r \) is the radial variable of a polar coordinate centering at the center of each unit cell. \( r^2 = x^2 + y^2 \). \( \Theta(r) \) is defined on a disk with radius \( R_{skx} \) embedded in the square unit cell. Its boundary conditions are \( \Theta(0) = \pi \) and \( \Theta(R_{skx}) = 0 \) for \( \tilde{D} + \tilde{D}_I < 0 \) (or \( \Theta(R_{skx}) = 2\pi \) otherwise.) The region complement to the disk has \( \Theta = 0 \).

(2) Spiral ansatz is defined by a function with a periodicity of \( R_{spi} \) in the x-direction.

\[
\mathbf{m}_{spi} = (\sin \phi(x), 0, \cos \phi(x)).
\]  

(30)

It satisfies boundary conditions: \( \phi(0) = 0 \) and \( \phi(R_{spi}) = -2\pi \) for \( \tilde{D} + \tilde{D}_I < 0 \) (or \( \phi(R_{spi}) = 2\pi \) otherwise.)

(3) The ansatz for the uniform magnetic order allows a canting angle \( \varphi \) away from the film surface.

\[
\mathbf{m}_{fm} = (\cos \varphi, 0, \sin \varphi).
\]  

(31)

It is chosen to lie in the \( xz \)-plane, compatible with the choice \( u < 0 \) in the rest of the text. These ansatze are adapted from Ref.57.

Figure 12 displays variational phase diagrams as a function of the film thickness \( w \) and the applied field \( H \). Panel (a) illustrates a skyrmion crystal stabilized by the strain-gradient DM, which sits between a canting FM and a polarized FM order. The color plot in the FM phase denotes the canting angle \( \varphi \) relative to the film plane, as shown in the inset. Panel (b) depicts a similar phase diagram with an increased strength of \( D_s \) coupling. This results in a spiral order at low field, which is not observed in the experiment. In panel (c) and (d), we switch on the interfacial \( D_I \) to illustrate its unimportance in a thick film; it does not affect the phase diagrams much in the thick-film limit, e.g. \( w > 5z_s \). The only difference is that a new skyrmion crystal appears in the ultrathin limit \( w < z_s \), which is driven solely by the interfacial DM, consistent with previous studies, e.g. a few layers of \( \text{SrRuO}_3 \) on \( \text{SrIrO}_3 \) substrate[58].
FIG. 12. (a) Variational phase diagram depicting stable phases as a function of the film thickness $w$ and the field strength $H$ for $D_s = 0.4J$ without the interfacial DM. A skyrmion crystal phase arises purely due to the strain-gradient DM (see Fig. 11(a)) in between a canting FM phase and a polarized FM phase. Color gradient denotes the canting angle $\varphi$ defined in the inset. (b) A similar phase diagram with $D_s = 0.5J$. With a larger $D_s$, a spiral order becomes stable in place of the canting FM. (c) and (d) are phase diagrams similar to (a) and (b) but with an additional interfacial DM coupling $D_I = 0.6J$. In the thick-film regime, the phase diagrams are roughly the same as (a) and (b). In the ultrathin limit $w < z_s$ (bottom panel), another skyrmion crystal phase is present and is driven solely by the interfacial term. Here we have set parameters of the free energy to $A_0 = 0.3J, u = -0.02J$ and $z_b = 10z_s$.

Kerr effect in the presence of skyrmions

Model

In this section, we discuss how electrons coupled to a skyrmion crystal can lead to the field dependence of Kerr rotation $\theta_K$, as seen in the experiment, that resembles the topological Hall effect. To see this, we begin by noting multiple contributions to the Kerr rotation observed in the experiment.

$$\theta_K = \theta_{K,M} + \theta_{K,T},$$

(32)

where $\theta_{K,M}$ is the conventional Kerr effect that tracks the magnetization. The topological Kerr component $\theta_{K,T}$ is attributed to electrons coupled to skyrmions, which we will study in detail. The additive form is expected to be valid when the angles are small, e.g. in the mrad range, which is the case in our system. We will show that $\theta_{K,T}$, as a function of the applied field, vanishes everywhere except in a field window where a skyrmion crystal phase is stabilized.

We will focus on the topological component $\theta_{K,T}$ by considering an electron hopping problem without spin-orbit coupling (SOC); SOC is more relevant to the $\theta_{K,M}$ component. In a small angle limit, the polar Kerr angle is determined by the dielectric tensor through the following relation [59]

$$\theta_{K,T}(\omega) = \text{Re} \left( \frac{\varepsilon_{xy}(\omega)}{\left(\varepsilon_{xx}(\omega) - 1\right)\varepsilon_{xx}(\omega)^{1/2}} \right).$$

(33)

The dielectric tensor $\varepsilon_{IJ}$ is related to the conductivity tensor via

$$\varepsilon_{IJ} = \varepsilon_b + i \frac{\sigma_{IJ}}{\omega \varepsilon_0}.$$  

(34)

$\varepsilon_b$ accounts for the background contribution, e.g. from the lattice. $\sigma_{IJ}$ has two contributions: (1) $\sigma^0_{IJ}$ for SRO without SkX, and (2) an extra topological component $\sigma^T_{IJ}$.

$$\sigma_{IJ} = \sigma^0_{IJ} + \sigma^T_{IJ}.$$  

(35)

A phenomenological form of $\sigma^0_{IJ}$ as a function of $\omega$ at 80 K is adapted from Ref. [60] and is used for the computation of $\theta_{K,T}$.

$$\sigma^0_{IJ} \approx \frac{10^6 (\Omega m)^{-1}}{(1 - i \frac{\hbar \omega}{0.017eV})^{0.4}} \delta_{IJ}.$$  

(36)
This bulk value becomes increasingly suitable for the calculation as the thickness increases. The background \( \varepsilon_b \approx 6.2 \) is obtained by imposing that the real part of \( \varepsilon_{xx}(\omega) \) vanishes at \( \hbar \omega \approx 1.3 \text{eV} \) at 40K [61].

\[
\text{Re} \left( \varepsilon_{xx}^0 \right)_{40K} = \text{Re} \left( \varepsilon_b + \frac{i\sigma_{xx}^0}{\omega\varepsilon_0} \right)_{40K} = 0,
\]

where the \( \sigma_{xx}^0(\omega) \) at 40K is also adapted from Ref.60.

To obtain \( \sigma_{IJ}^T \), we consider electrons on a square lattice coupled to a skyrmion crystal.

\[
H_{\text{e-skx}} = -t \sum_{(ij)\sigma} c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.} - J_{\text{H}}^{\text{eff}} \sum_i \mathbf{m}_i \cdot \mathbf{c}_i^\dagger \sigma_{\alpha\beta} \frac{\sigma_{\alpha\beta}}{2} c_{i\beta},
\]

where \( i, j \) are sites of the square lattice. \( J_{\text{H}}^{\text{eff}} \) is viewed as originating from the atomic Hund’s coupling in Ru sites. \( \mathbf{m}_i \) denotes the local moment direction at site \( i \) obtained from the continuum solution to the variational problem discussed in the previous section.

We then compute a relative conductivity \( \sigma_{IJ}^{T,2D} = \sigma_{skx,2D}^{skx,2D} - \sigma_{IJ}^{\text{FM},2D} \), where \( \sigma_{skx,2D} \) is the conductivity of electrons coupled to a skyrmion crystal and \( \sigma_{IJ}^{\text{FM},2D} \) is for electrons coupled to a uniform FM order. The topological \( \sigma_{IJ}^T \) is assumed to be related to the relative conductivity \( \sigma_{IJ}^{T,2D} \) by a factor of \( 1/d \), where \( d \) is a length scale taken to be the out-of-plane lattice parameter, namely \( d \approx 3.9\text{Å} \).

\[
\sigma_{IJ}^T = \frac{\sigma_{IJ}^{T,2D}}{d}.
\]

The reason for taking the relative conductivity is that this additional contribution must vanish when the system is in a FM order.

To compute the conductivity tensor, we use the following Kubo formula [62–64]

\[
\sigma_{IJ}^{2D} = \frac{i2\pi e^2}{L^2} \hbar \sum_{k,l,m} n_F(E_{km}) - n_F(E_{kl}) \left[ \frac{(v_l)_{ml}(v_I)_{lm}}{\hbar \omega + i\eta + (E_{km} - E_{kl})} \right],
\]

where \( (v_I)_{ml} \equiv \langle \mathbf{k} m | \frac{\partial H(k)}{\partial k_I} | \mathbf{k} l \rangle \), and \( | \mathbf{k} m \rangle \) is a Bloch state. We have set \( t = 200\text{meV} \), \( J_{\text{H}}^{\text{eff}} = 2\text{eV} \), \( \eta = 10\text{meV} \), and the electron density = 0.8.

**Results**

Figure 13 shows a frequency dependence of \( \theta_{K,T} \) in a skyrmion crystal phase. The low-energy peak may be understood from the largeness of the off-diagonal \( \sigma_{xy}^T \), which is caused by the low-energy transitions between Chern bands (see Fig. 15). We will discuss this in more detailed in future work. We focus on the behavior of \( \theta_{K,T} \) in the frequency window \( J_{\text{H}}^{\text{eff}} \), which appears to be more relevant to the wavelengths used in our experiment. In this window, \( \theta_{K,T} \) is of the order of mrad. Below this frequency window, \( \theta_{K,T} \) diminishes. This may explains the experimental observation that the bumps in \( \theta_K \) fade away in the lower-frequency lasers and are present in higher-frequency lasers. For instance, the bumps are absent in 810nm measurement, yet they are pronounced in 633nm and 594nm wavelengths (see Fig.1(c)-(e) in the main text.)

To understand this feature, we turn to Fig.14, which shows the conductivity \( \sigma_{IJ}^{T,2D} \) as a function of the optical frequency. Near \( \hbar \omega = J_{H}^{\text{eff}} \), \( \text{Re} \sigma_{xx}^{T,2D} \) exhibits a resonant-like behavior. We trace this back to a large joint density of state \( \delta(h\omega - E_{km} + E_{kn}) \), caused by a large number of pairs of bands that disperse similarly. Their energies differ by a single energy scale of \( J_{H}^{\text{eff}} \), which can be seen in Fig.15. Since \( \text{Re} \sigma_{xx}^{T,2D} \) is proportional to the joint density of state, the resonant-like feature is attributed to
FIG. 13. Topological Kerr component $\theta_{K,T}$ as a function of optical frequency $\omega$ in a skyrmion crystal phase, featuring a pronounced value in the $\hbar\omega \sim J_{H}^{\text{eff}}$ window. In this range, $\theta_{K,T}$ is large enough to produce bumps in the field scan, as observed in our Kerr measurements. It also predicts a significant $\theta_{K,T}$ contribution at low frequencies $\hbar\omega \approx 0.005J_{H}^{\text{eff}}$. Elsewhere, $\theta_{K,T}$ exists but may be insufficient to produce any bumps.

FIG. 14. Relative conductivity $\sigma_{T,2D} = \sigma_{skx,2D} - \sigma_{FM,2D}$ as a function of optical frequency in (a) the low-frequency limit and (b) the $J_{H}^{\text{eff}}$ window. The rich $\omega$-dependence in (a) originates from low-energy transitions between Chern bands. The negative longitudinal relative conductivity implies an increase in resistivity in the presence of skyrmions. The nontrivial frequency dependence in (b) is attributed to the higher-energy optical transitions around $\hbar\omega \sim J_{H}^{\text{eff}}$ with a large joint density of state $\delta(\hbar\omega - E_{k,m} + E_{k,l})$ caused by bands that similarly disperse, as depicted in Fig. 15. This leads to peaks in $\theta_{K,T}$ in Fig. 13 around $\hbar\omega \sim J_{H}^{\text{eff}}$. 
FIG. 15. Band structure of electrons coupled to a skyrmion crystal, displaying similarly dispersing Chern bands whose energies differ by $J^\text{eff}_H$. This leads to a large joint density of state $\delta(\hbar\omega - E_{k,m} + E_{k,l})$ around $\hbar\omega \approx J^\text{eff}_H$ and the rich frequency dependence of the conductivity tensor in Fig. 14(b).

This property of the band structure. One can also show that $\text{Im} \sigma^{T,2D}_{xy}$ is proportional to the joint density of state, thereby exhibiting the same behavior. The frequency dependence of $\text{Im} \sigma^{T,2D}_{xx}$ and $\text{Re} \sigma^{T,2D}_{xy}$ can be understood based on the Kramers-Kronig relations. The behaviour of $\theta_{K,T}$ in the $J^\text{eff}_H$ window can then be associated with the rich frequency dependence of $\sigma^T_{IJ}$ caused by skyrmions. Near and outside this frequency window, the joint density of state is no longer pronounced; the off-diagonal components of $\sigma^T_{IJ}(\omega)$ become small, and so is the $\theta_{K,T}$.

Here, skyrmion crystal plays an important role in producing nonzero matrix elements of the current operator $\langle k|m|j_I|kn \rangle$, despite the fact that $j_I$ is diagonal in spin. Its noncoplanarity is essential for generating an emergent magnetic field [65], leading to a nonvanishing $\sigma^T_{xy}(\omega)$. These two facts lead to $\theta_{K,T}$, which becomes very significant in the optical frequency range around $J^\text{eff}_H$. This leads us to attribute the bumps observed in our Kerr experiment to skyrmions, which we term “topological Kerr effect”.
