

Magnetic domains in ultrathin, bulk-like and proximity-coupled europium oxide

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The control of electron spins in materials that are simultaneously ferromagnetic and insulating opens up a wealth of quantum phenomena in spin-based electronics. Thin films of europium oxide (EuO) are ideal for the generation and manipulation of spin-polarized states, but so far, there are no experimental literature reports on the magnetic domain patterns of EuO. However, at these microscopic length scales, magnetic relaxation between the remanent and demagnetized states takes place in any spintronic device. This relaxation process involves the displacement of magnetic domain walls and can therefore be strongly influenced by the film's structure and thickness. Here, we present an investigation of the temperature-dependent behavior of magnetic domains and hysteresis in bulk-like (25 nm) and ultrathin (3 nm) EuO films. Magneto-optical Kerr microscopy is used, a technique that is a valuable tool for exploring microscopic features such as spin dynamics and magnetic domain walls. Significant Kerr rotation in EuO led to high-contrast magnetic domain images in thick films, facilitating the observation of domain dynamics. The Curie temperature (T_C) and coercivity show thickness-dependent variations. The analysis and comparison of hysteresis loops and domain imaging in EuO and EuO/Co reveal proximity effect-induced antiferromagnetic coupling of both layers. To elucidate the magnetization reversal dynamics in EuO, micromagnetic simulations using MuMax³ were performed below and above T_C . This comprehensive approach was designed to comprehend the impact of magnetism and magnetic proximity effects in EuO on the micromagnetic scale, potentially extending its magnetic ordering beyond T_C .

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I. INTRODUCTION

Magnetic insulators are ubiquitous materials in spintronic devices [1–5]. The material class of rare-earth europium chalcogenides (EuX, X = O, S, Se, and Te) features strong optical responses and enormous magneto-optic Kerr effects that facilitate spin-based applications, including nanophotonic devices, for example, spin diodes, spin transistors, spin injectors, spin qubits, and nonvolatile magnetic memory [6,7]. Among them, EuO has attracted the most attention in the past couple of decades due to its Silicon-like band gap of 1.12 eV and a reasonable Curie temperature (T_C) of 69 K. Below T_C , the magnetic moments of Eu^{2+} are exchange-coupled with a very large localized magnetic moment of $7.0 \mu_B$. Because of the significant exchange splitting of the conduction band, EuO also shows a strong spin polarization near 100 % [8]. EuO is a potentially topologically driven material with significant spin-orbit coupling, as evidenced by several observed phenomena, such as the anomalous Hall effect and the presence of magnetic skyrmions [9].

In the wealth of literature available on EuO, most studies on its magnetic properties have been dedicated to its macroscopic characteristics, such as T_C , electronic structure, and temperature-dependent magnetization [10,11]. The practical application of EuO is restricted due to its T_C of 69 K, and experimental efforts are dedicated to enhancing the T_C . The improved exchange coupling by the donor states and/or conduction electrons with electron doping via oxygen vacancies or trivalent rare-earth ions [12–14] were the sources of the rise in T_C . Doping EuO with rare-earth ions such as Lu, Gd, and La has resulted in T_C values of 119, 116, and 120 K, respectively [15–17]. Proximity effects by placing a room temperature ferromagnet close to EuO have been investigated by Pappas *et al.* [7], and because of Co, thin EuS layers showed significantly high spin polarization up to room temperature. Using the proximity-induced effect from Co and Fe, this strategy has been tested for ultrathin EuO (3 nm); very recently, signatures of magnetic ordering up to 80 K have been observed [18]. However, all of these reports are confined to the physical properties in the bulk or nano regime.

Given this, the microscopic and mesoscopic properties, such as magnetic domains, magnetization reversal, and domain dynamics in EuO, remain unknown. There is a dearth of literature on magnetic domains in EuO, and one reason for this could be the oxidation-prone nature of EuO compared with the more stable paramagnetic phase Eu_2O_3 [19]. Despite the fact that EuO has the largest Kerr rotation of $\sim 4^\circ$, there are only a handful of reports on Kerr studies of EuO [20]. Some of these focus on the energy-dependent Kerr rotation of Eu [6,20–22], while the others are related to the second

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harmonic generation in EuO [23]. Recently, Mönkebüscher *et al.* studied EuO/Co bilayers using static and time-resolved magneto-optical Kerr effect measurements and explored the influence of the magnetic proximity of Co on the T_C and spin dynamics in EuO [24].

The present work focuses on coercivity behavior and magnetic domain reversal studies of high-quality stoichiometric EuO films with different thicknesses, from the bulk-like (25 nm) to the ultrathin film regime (3 nm). Using magneto-optic Kerr microscopy, we simultaneously observed magnetic domains and recorded magnetic hystereses on EuO films. We also investigated the effects of temperature and film thickness on the coercivity of EuO. Moreover, magneto-optic studies were conducted on a EuO/Co heterostructure to examine the impact of the close proximity of Co on the magnetic domains and saturation behavior of EuO. To picture the magnetization reversal dynamics in EuO and EuO/Co, micromagnetic simulations were performed using MuMax³ and compared with the experimental findings.

II. METHODS

A. Experimental

Using molecular beam epitaxy, EuO thin films and EuO/Co heterostructures were deposited on Nb-doped SrTiO₃ (Nb:STO)(Crystec GmbH). A 25-nm-thick EuO film was deposited as a bulk reference at 400° C at an Eu evaporation rate maintained at 0.13 Å/s for 30 minutes and with an oxygen partial pressure of 10⁻⁸ mbar after 30 minutes of substrate preheating at 500° C. For the ultrathin 3-nm EuO film, a redox growth mechanism was used, and Eu was evaporated without oxygen flow for 5 minutes and 25 seconds. More information about this mechanism can be found elsewhere [25,26]. The Nb-doped STO [100] direction and EuO [110] direction have a lattice mismatch of ≈7%, and EuO is proven to grow epitaxially on STO after a 45° rotation [27]. All samples were subjected to low-energy electron diffraction and *in situ* X-ray photoelectron spectroscopy (XPS) (representative spectrum in Fig. 1S in the supplemental material) for preliminary structural and chemical examination. We also performed a rough estimation of EuO thickness in a 3-nm film using XPS, which can be seen in Fig. 2S in the supplemental material [28]. Then, using e-beam evaporation, a 4-nm-thick Co overlayer was deposited onto the 3-nm EuO film at room temperature. Co was deposited at room temperature on half of the sample area using a shadow mask with a deposition rate of 0.16 Å/s, and thickness was controlled using quartz crystal. Lastly, a 25-nm coating of MgO was deposited onto all films to prevent air oxidation.

A wide-field Kerr microscope from Evico Magnetics, Germany, was used for the magnetic measurements at low temperatures [29]. It employs a Carl Zeiss Axioscope design microscope, equipped with a brilliant white LED light source. Measurements in Longitudinal geometry (longitudinal magneto-optic Kerr effect, or LMOKE) were made using an applied magnetic field that was varied between ±360 mT. Further details about these measurements can be found in the supplemental Material [28].

B. Simulations

Micromagnetic simulations were performed using Mumax³ to obtain qualitative information about the domain reversal mechanisms in EuO [30,31]. We assumed a micromagnetic model to reproduce the magnetization properties of an experimentally observed thin EuO film. For simplicity, the thin film's crystalline texture was neglected in simulations. The field-dependent curves for EuO were obtained along with snapshots of magnetic moments at each value of the applied field. The magnetic parameters were kept close to the theoretical values for EuO to correlate with the respective experimental results by MOKE. The following set of parameters was used in the simulation process: $M_S = 1.77 \times 10^6$ A/m and $A_{\text{exch}} = (1.1658 \times 10^{-12} \pm 3.668 \times 10^{-17})$ J/m (estimated from the critical temperature using structure-specific constants [32]), where M_S and A_{exch} are the saturation magnetization and exchange constant, respectively [33]. For details, refer to the supplemental material [28] and Ref. [34].

III. RESULTS AND DISCUSSION

A. Bulk-like EuO film

First, we present the magnetic hystereses measured for the 25-nm EuO bulk-like film. We recorded magnetic domain pictures in LMOKE geometry [Fig. 1(a)] and measured magnetic hysteresis by systematically varying the sample temperature below and up to T_C . The LMOKE hystereses are displayed in Fig. 1(b). Well-defined, fully saturated hystereses were observed at every measured temperature up to T_C . The coercivity (H_c) of the loops decreases with increasing temperature, as is expected for ferromagnetic behavior [35]. The value of $H_c = 7.5$ mT at 20 K is in good agreement with the values reported in the literature, which are characteristic of high-quality EuO films measured with vibrating sample magnetometry and MOKE [24]. As the temperature approaches the bulk EuO T_C , that is., 69 K, both the H_c and remanent magnetization fall drastically. The magnetic hysteresis could be observed up to 70 K, which corresponds to the bulk T_C of EuO.

Figure 1(c) shows the representative domain images observed at the marked points during the measurement of hysteresis at 30 K, as indicated in Fig. 1(b). In the positive cycle (field increasing), the light gray contrast shows the emerging areas as stripes, which are aligned in the direction of the applied field, as can be seen at 6.2 mT. These are 180° domains, and they started to appear as branches in the direction of the applied field at different regions in the measured area. This could be attributed to the presence of defects in the film that act as pinning centers for the domain nucleation. The width of these stripes is ≈ 5 μm at 7 mT and was estimated using the scaling feature in ImageJ software, which might contain observational errors [36]. The strip width increased, and they merged as the strength of the applied field increased. A complete reversal of the described behavior can be seen when the applied field was varied from positive to negative direction in Figs. 1(c) and 1(d). The process of magnetic reversal and evolution of magnetic domains can be seen in the multimedia file in Fig. 5S in the supplemental material [28]. A

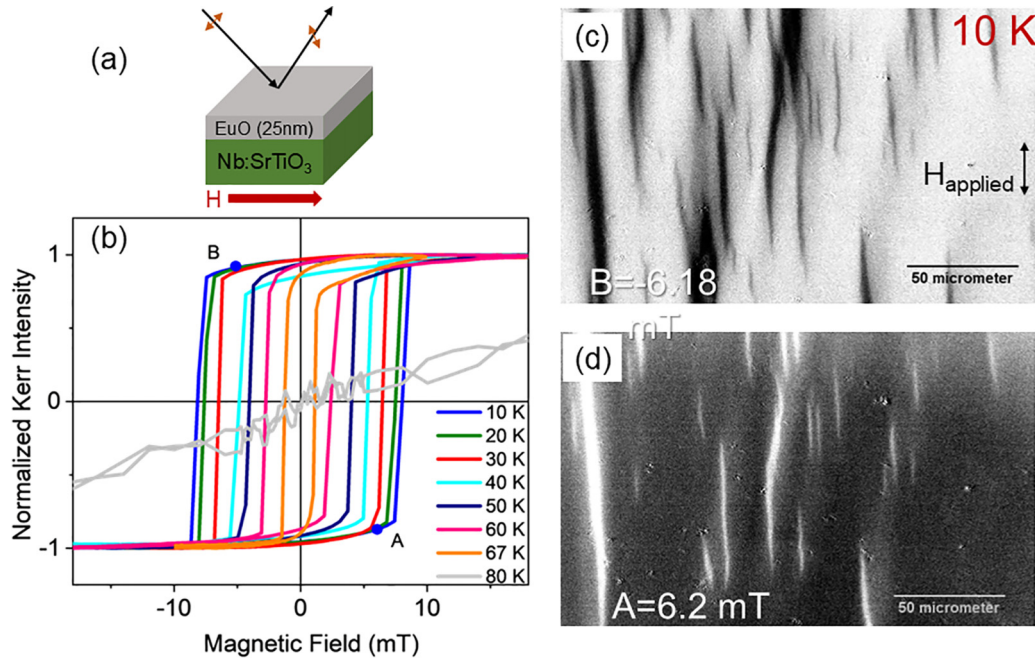


FIG. 1. (a) Schematic showing the sample structure and longitudinal magneto-optic Kerr effect (LMOKE) geometry. (b) Normalized magnetic hystereses obtained on the 25-nm EuO reference film using LMOKE at different temperatures. (c) and (d) The 180° stripe magnetic domains observed at different acquisition points A and B of the hysteresis measured at 30 K shown in (b).

similar kind of domain pattern has been observed at nearly all measurement temperatures, while the Kerr contrast gradually reduced until the signal was lost when T_C was approached, as shown in Fig. 1(b).

B. Ultrathin EuO film

We performed similar LMOKE measurements on both the EuO and Co sides of the EuO/Co heterostructure one by one, as shown in Fig. 2(a). However, since the EuO film is only 3 nm in thickness and the corresponding Kerr signal obtained was much lower than for the 25-nm bulk reference, we measured in pure LMOKE mode to enhance the signal-to-noise ratio as described in the supplemental material [28]. On the ultrathin EuO film side, the hystereses obtained as a function of temperature are shown in Fig. 2(b). The hystereses showed lesser magnetic remanence (0.83) compared with the reference film (0.95) at 10 K, as can be seen in Fig. 3S in the supplemental material [28]. At 10 K, H_c is 11.8 mT, which is higher than the H_c value for the reference 25-nm film. The remanent magnetization remained close to 1 as the sample temperature was increased; however, the H_c decreased similarly to that of the reference film. The ultrathin EuO film started to lose its Kerr signal at a lower temperature than 25-nm EuO and exhibited a very weak Kerr signal at and above 60 K. The behavior was found to be consistent with the modeling of the thickness-dependence of T_C in EuO films as shown by Müller *et al.* [22] and in other reports as well [21,37]. The T_C decreases with decreasing film thickness, and comparatively lower T_C values have been observed for the EuO films thinner than 5 nm. We also determined the easy axis of magnetization for EuO to be [110] as observed from hysteresis obtained from measuring the same EuO film

after rotating it by 45° in-plane, as shown in Fig. 4S in the Supplemental Material [28].

The magnetic domain images corresponding to two different values of the applied field measured at 15 K for a 3-nm EuO sample are shown in Fig. 2(c). Similar to Fig. 1(c), the domain evolution for the 3-nm EuO film can be observed, and here the magnetic domains clearly show a weaker Kerr contrast compared with Fig. 1(c). Also, the domains have simultaneously more nucleation sites in the form of strips on the film. To compare, we analyzed $250\mu\text{m}^2$ area for both thicknesses of the film. The number of strips originating in a similar area near the coercive field value is 3–5 for 25-nm film, while for 3-nm film, the number is 9–10. Comparing the domain images of two different thicknesses of EuO, 3 and 25 nm, we observed subtle differences such as: (i) the domains in the 3-nm EuO film were smaller in size, (ii) possess low Kerr intensity contrast, (iii) have more nucleation sites in the form of strips in a similar area near H_c .

These observations can be understood by comparison of thickness-dependent thin film properties such as crystallite size, microstrain, concentration of defects, and so on. [38]. Out of these structural properties, strain in the EuO film of thickness 3 nm on STO could attribute the formation of twin structural domains in the form of stripes to minimize the strain, which in turn affects the magnetic domains and the domain evolution pattern as well [39–41]. These strips may exhibit slight variations in anisotropy fields, preferentially facilitating domain nucleation in alternating strips. Such observations have been made in ferromagnetic and ferroelectric thin films [39,42]. These strips were not observed in the 25-nm reference film as the thickness is high enough that the strained film relaxed gradually. Additionally, another factor that could explain such an observation is exchange

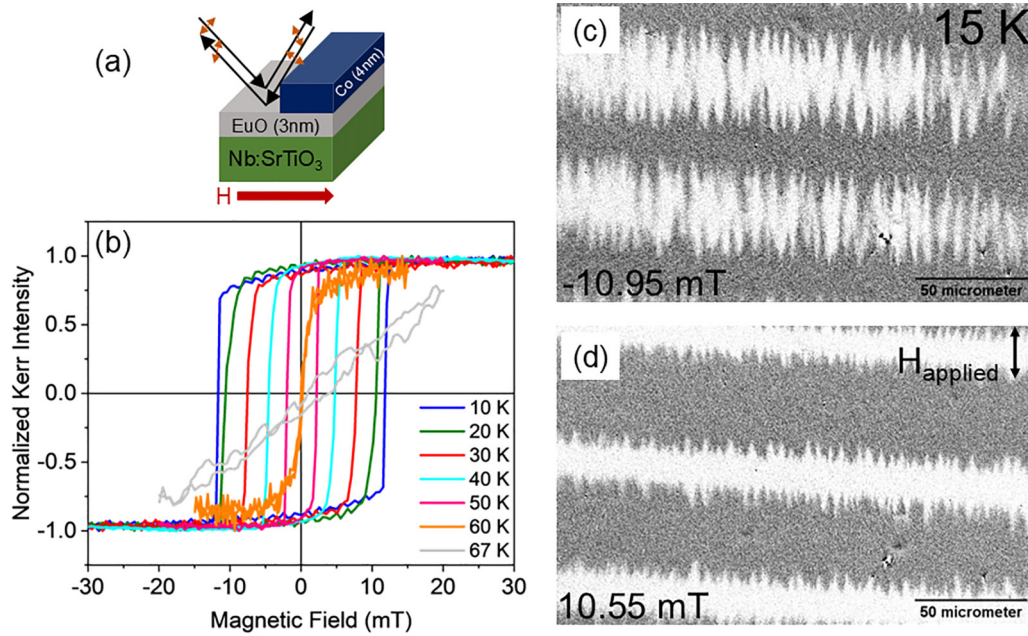


FIG. 2. (a) Schematic showing the two-sided EuO and EuO/Co heterostructure measured in pure longitudinal magneto-optic Kerr effect (LMOKE) geometry. (b) Normalized magnetic hystereses were obtained on a 3-nm EuO film using pure LMOKE at different temperatures. (c) and (d) The magnetic domain observed at different values of the applied field measured at 15 K for a 3-nm EuO film. The light and dark gray colors represent the regions where the magnetization is oppositely aligned.

magnetostriction. The volume of EuO shrinks as it is cooled below T_C , and this follows the magnetization increase below T_C [43]. The strong anisotropic magnetostriction of EuO also has large direction-dependent values as calculated by Argyle *et al.* [44]. This implies that EuO self-organizes below its T_C , giving rise to bands of different strains, yielding regions of different anisotropy as striped domains. The EuO layer may undergo self-organization of bands of different strains to minimize the total energy (domain walls + long-range elastic strain), as found for quasi-two-dimensional strained systems [45]. Since we observe such domains in the ultrathin EuO film, it could be speculated that the domain strips are of magnetostrictive origin. A short video of the domain evolution and magnetic reversal can be seen in the multimedia file in Fig. 6S in the supplemental material [28].

C. EuO/Co heterostructure

The temperature-dependent hystereses obtained are shown in Fig. 3. At 30 K and below, the hysteresis does not show a sudden jump to magnetic saturation, as was observed on the EuO side of the sample. On the Co side, the curves show a different saturation and possess asymmetric shapes as well. Additionally, the Kerr contrast was significantly lower on the Co side, making it difficult to record the domain images. The loops were also reversed in direction. The hystereses also show a small exchange bias field ($\sim 4 - 6$ mT) at 10 and 20 K. The H_c reduced similarly as the temperature increased, as observed for the reference film and 3-nm EuO. Such presence of exchange bias field in EuO films has been generally attributed to the lack of stoichiometry [12,13]. However, our *in situ* x-ray photoelectron spectroscopic investigations debar this as the

EuO film is in the stoichiometric phase, as can be seen in Fig. 1S in the supplemental material [28].

A possible explanation could be the presence of Co in the vicinity of EuO, which leads to proximity coupling at the interface as observed recently in similar heterostructures [18,24]. This proximity-effect-induced coupling could affect the shape of the hysteresis and is found to be more pronounced below 30 K in our case, where the Kerr rotation in EuO is the highest. This implies that the heterostructure now has different Kerr intensity contributions coming from the top FM Co layer, the AFM Co/EuO coupled region, and the remaining FM EuO layer (in case the whole EuO layer is not coupled). The sum of all these contributions could be in an asymmetric loop with a small exchange bias stemming from the antiferromagnetic-coupled layer. Such antiferromagnetic coupling observations have been made in ferromagnetic Fe/MnAs layers; however, their system does not show exchange bias [46,47]. Therefore, up to this point, the presence of exchange bias can not be explained satisfactorily based on the observed data. We would need a systematic thickness-dependent study of (EuO(x nm)/Co(4 nm) coupled layer in the future, where $3 \text{ nm} \leq x \leq 25 \text{ nm}$.

The reversal of loops observed can be attributed to the Kerr rotation in EuO ($\approx 2^\circ$) and Co ($\approx -0.3^\circ$ for blue light); the majority of the Kerr signal coming from the top Co layer of the heterostructure defines the dominant Kerr rotation, which reverses the hystereses. As can be seen in the images in Figs. 3(c) and 3(d), the presence of stripes underneath the top Co layer signifies that the Kerr signal is arising from the whole heterostructure. It seems that, in our case, for the EuO/Co system, Co dominates over EuO in the heterostructure as observed by Mönkebücher *et al.* [24]. Above 30 K, the hystereses exhibited a more symmetric shape while H_c

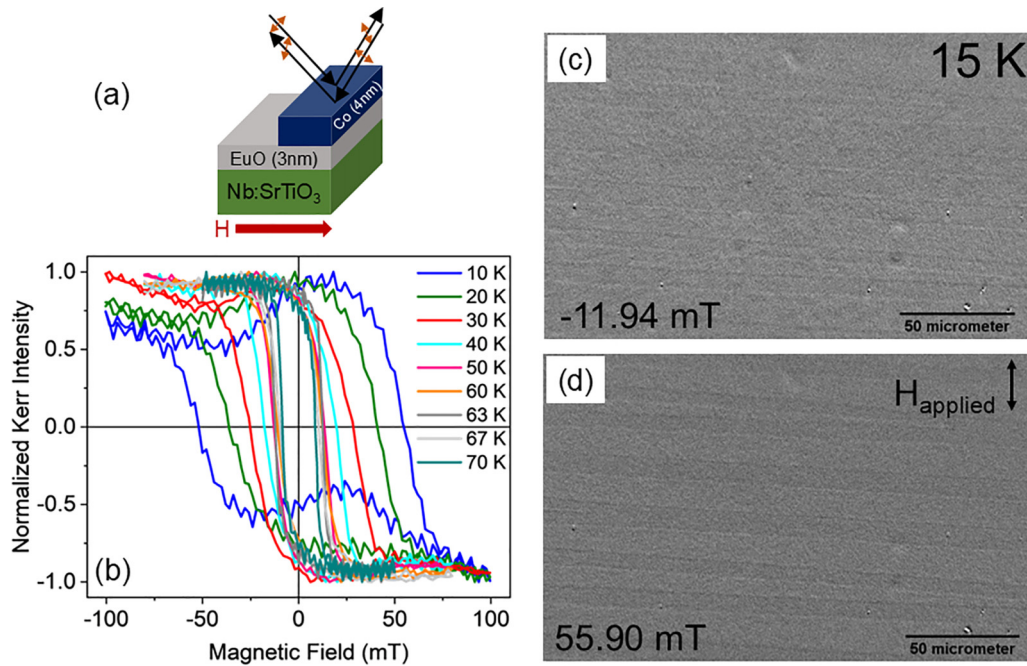


FIG. 3. (a) Schematic showing the EuO/Co heterostructure measured in pure longitudinal magneto-optic Kerr effect (LMOKE) geometry. (b) Normalized magnetic hystereses obtained Co-covered EuO film using pure LMOKE at different temperatures. The magnetic domain observed at different values of the applied field measured at 15 K are shown in (c) and (d).

continues to decrease, and similar behavior continues until 60 K. The hysteresis curve becomes more symmetric afterward while H_c continues to reduce as the temperature is increased, which again hints that the coupling might either be temperature-dependent or the fact that temperature-dependent Kerr rotation of EuO is reducing as T_C approaches [48].

Figures 3(c) and 3(d) shows the domain images observed at different values and directions of the applied magnetic field. A small variation in the Kerr contrast and the presence of stripes can be observed. The multimedia file in Fig. 7S in the supplemental material shows contrast change in the Co overlayer; however, the EuO remains the same [28]. Clear domain images could not be observed on the Co side of the sample, despite optical adjustments made in the microscope. This could be due to very low Kerr contrast on the Co side of the sample, and the antiferromagnetic coupling diminished domain sizes to a length or time scale, which is beyond the scope of the measurement setup.

D. Temperature-dependent H_c behavior

Figure 4 presents the H_c values obtained for the 25-nm EuO, 3-nm EuO, and EuO/Co as a function of temperature. For the bulk-like 25-nm EuO film, a monotonous decrease was observed as temperature is increased. In the whole temperature range until the T_C is reached, the behavior of the decrement of H_c with temperature shows a typical dome shape. The trend of H_c variation can be understood by correlating H_c with the Kerr rotation. The dependence of the Kerr rotation on temperature for EuO films shows a similar behavior as that reported by Swartz *et al.* [48]. However, in the Kerr microscope, we could not directly measure the Kerr rotation but only sense it indirectly through changes in the

image intensity (which is Kerr intensity) [29]. Since the H_c values here have been derived from LMOKE loops obtained by plotting Kerr intensity with the applied field, a similar dependence of Kerr rotation with temperature could be expected.

In line with the differences observed in the hystereses and domain images of different thicknesses of EuO film, the H_c versus temperature dependence is different for 3-nm EuO. As can be seen in Fig. 4, H_c rapidly decreases with the increase in the temperature in a similar way. From 50 K until the point where the Kerr signal could be obtained, that is 63 K, the

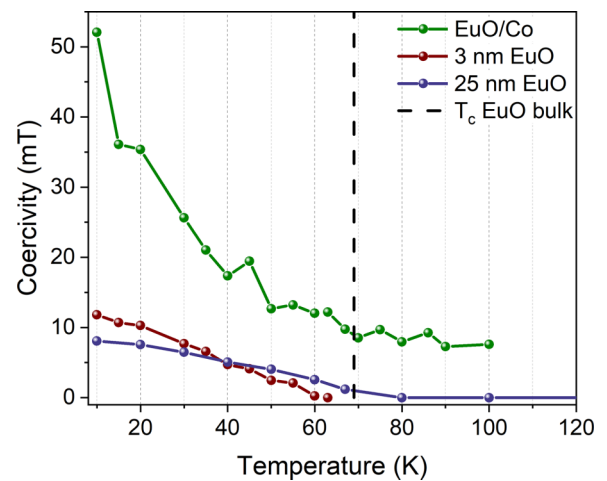


FIG. 4. The coercivity (H_c) dependence of temperature for the reference 25-nm EuO film, 3-nm EuO, and (c) Co(4 nm)/EuO(3 nm) heterostructure. The dashed vertical line is a guide to the eye for T_C of bulk EuO, that is, 69 K. The error bars are less than the size of the symbols in the plot.

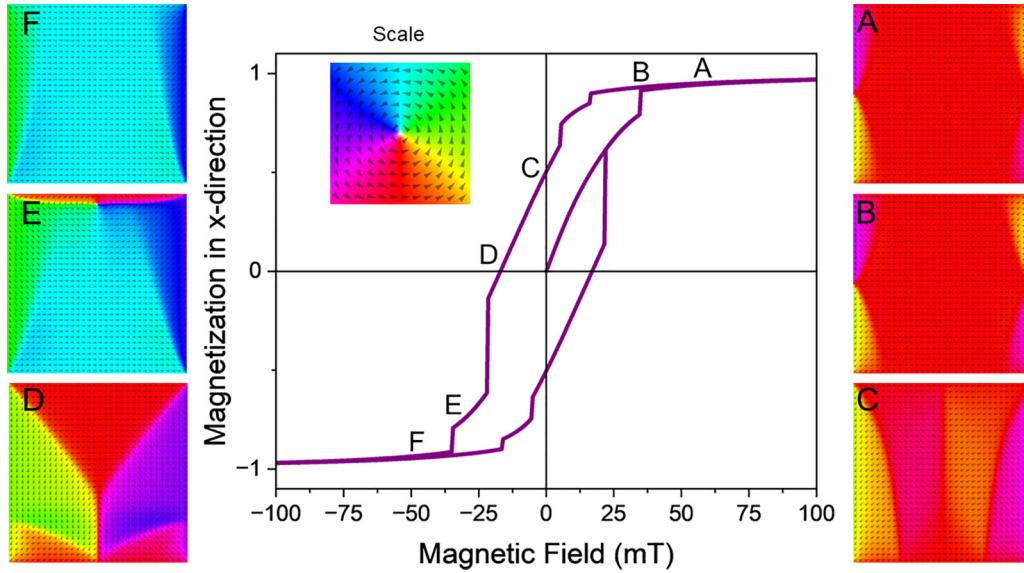


FIG. 5. Simulated magnetic hysteresis and different snapshots of magnetization in x -direction taken at various values of applied magnetic field (A to F = 50, 10, -8.6 , -18.5 , -35 , and -50 mT, respectively) for 3-nm ultrathin EuO at 15 K. The corresponding color scale is shown as the inset.

decrease in H_c is very small. The temperature-dependent H_c behavior of EuO with Co has also been compared in Fig. 4. Here, until 40 K and the decrease in H_c with temperature is rapid as H_c falls from 54 to 18 mT. This region has the maximum effect from the antiferromagnetic coupling between Co and EuO. After 40 K, the variation of H_c up to 60 K is less. In the critical region near T_C of EuO, the shape of the hysteresis does not change anymore, giving the interpretation that in this temperature range, the signal mostly originates from the Co layer. The variations in H_c were very small beyond T_C of EuO. A distinct determination of the T_C of EuO could not be seen as the Kerr signal from the Co layer starts to dominate over EuO beyond its T_C . Overall, the very high H_c of EuO/Co heterostructure in our case could be attributed to the antiferromagnetic interactions between EuO and Co. In these measurements, the hystereses presented could not provide much information about the extent of the proximity effect with temperature, based on the changes in H_c as magneto-optical Kerr measurements lack element-specificity and practically limited Kerr signal resolution between the two layers.

E. Micromagnetic simulations

A set of simulations was performed to investigate the possible transient domain configurations for the magnetic moments with the applied magnetic field in the x -direction. The normalized magnetization in the x -, y -, and z -directions was recorded and plotted. It was found that with the given assumptions, magnetization is sensitive to the x -direction only, and a representative hysteresis is shown in Fig. 5. The H_c value for this loop is found to be 13.5 mT, which is slightly higher than the experimental value for the 3-nm EuO film at 15 K. However, the snapshots of the magnetic spin configuration of the simulated loop showed similarities with the experimental results in Sec. III C. As the applied field was varied from +100 mT to -100 mT, the snapshots taken at different field

values (A–F) are also shown in Fig. 5. At A, nearly all the spins are aligned with the applied magnetic field. The reversal of magnetization starts from the boundaries, which serve as defects, and they appear as perpendicular domains from both sides. The domains formed at the boundaries also comply with the flux closure domains, which are formed to reduce the demagnetizing energy in a thin magnetic film, giving rise to continuous magnetic field lines. This reversal can be seen in the multimedia file in Fig. S8 in the supplemental material.

Such a simple twin structure might form by alternating domains that touch each other through quasiequidistant parallel plane 90° domain walls and can be correlated to the strip formation due to strain in the film. The twin structures as perpendicular domains observed in the simulations are shown in Fig. 5, C. These walls do not originate due to magnetocrystalline anisotropy as per the assumptions made in the micromagnetic simulations. However, as the sample is a thin film, it leads to high dipolar fields due to more interactions in the film plane than in the out-of-plane direction and high saturation magnetization of EuO. This aligns the moments along the in-plane direction. To model this, we used shape anisotropy, which is computed via the demagnetizing energy in the simulation itself, and excluded the cubic anisotropy, which is significantly smaller than the former.

As can be seen in Fig. 5, D, the moments pointing along the y -axis transition within the plane to switch from one field direction to the other. Further, with the applied field, there is an increase in the area of the unaligned magnetic moments, which finally flip 90° to switch toward the field direction, as can be seen in Fig. 5, E. From B to E, one can understand the magnetization reversal mechanism in an ideal EuO film with the applied field. To affirm the origin of these perpendicular domains, we checked the domain formation with a magnetic field applied along the y -axis, and since the ground state is stabilized by the demagnetizing energy now, both the x - and y -directions show similar perpendicular domains were

appearing at the edges for flux closure, which broadens into larger domains resulting in flipping of the moments along the field direction. At F , nearly all the moments have aligned with the applied field, and uniform magnetization could be observed.

IV. CONCLUSION

In summary, we present the first magnetic domain imaging of bulk-like, ultrathin, and proximity-coupled EuO films utilizing magneto-optical Kerr microscopy. The focus was on the evolution of magnetic domains below and up to T_C . Our observations revealed that the Kerr contrast and domain size in EuO are strongly dependent on the film thickness. Specifically, we observed well-defined, high-contrast magnetic domains in a 25-nm-thick EuO film down to the ultrathin film limit (3 nm). As the thickness of the EuO film decreased, the size of magnetic domains was found to diminish; they possess strip-like features due to uncompensated strain. Simultaneous domain imaging and hysteresis measurements enabled us to analyze the temperature-dependent behavior of H_C . We discovered that variations in H_C with temperature are influenced by film thickness as well. The presence of a Co overlayer on EuO in the heterostructure resulted in asymmetric hysteresis, characterized by relatively high H_C . Additionally, the antiferromagnetic proximity coupling

between EuO and Co significantly influenced both H_C and magnetic domains. Micromagnetic simulations conducted for a 3-nm EuO film exhibited consistency with experimental observations obtained through pure LMOKE measurements. Our findings underscore the potential of designing and conducting fundamental studies on EuO and three-dimensional transition metal-based heterostructures and could lead to the realization of sustained magnetic ordering in EuO beyond its critical temperature.

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DATA AVAILABILITY

The datasets used and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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