**Critical Role of La doping for Topological Hall Effect**

**in Epitaxial EuO Films**

Yu Yun1,2†, Yang Ma1,2†, Tang Su1,2, Wenyu Xing1,2, Yangyang Chen1,2, Yunyan Yao1,2, Ranran Cai1,2, Wei Yuan1,2, and Wei Han1,2\*

1 International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, P. R. China

2 Collaborative Innovation Center of Quantum Matter, Beijing 100871, P. R. China

† These authors contributed equally to the work

\* Correspondence to: weihan@pku.edu.cn

**Abstract**

We report the critical role of La doping in the topological Hall effect observed in La*x*Eu1-*x*O thin films (~ 50 nm) grown by molecular beam epitaxy. When the La doping exceeds 0.036, a topological Hall effect is observed close to the Curie temperature, which we attribute to the formation of magnetic skyrmions. Besides, the La doping is found to determine the phases, densities, and sizes of the skyrmions in the La*x*Eu1-*x*O thin films. The maximum region of the skyrmion phase diagram is observed on the La0.1Eu0.9O thin film. As the La doping increases, the skyrmion density increases while the skyrmion size decreases. Our findings demonstrate the important role of La doping for the skyrmions in EuO films, which could be important for theoretical understandings of magnetic skyrmions in 2D Heisenberg ferromagnets.

**I. INTRODUCTION**

Magnetic skyrmion, a topological stable and particle-like spin texture, has attracted a great deal of attention arising from its exotic physical properties and potential application for high density memory devices [1-3]. Dzyaloshinskii–Moriya interaction (DMI), a chiral interaction due to the inversion symmetry breaking, has been demonstrated to be accountable for skyrmions in bulk non-centrosymmetric B20 chiral magnets [2,4,5], including MnSi [6-8], Fe1−xCoxSi [9], FeGe [10,11], etc, and their analog Cu2OSeO3, a multiferroic insulator [12,13]. Besides, interfacial DMI between a ferromagnetic (FM) layer and a heavy metallic (HM) layer with large spin-orbit coupling gives rise to the formation of skyrmions at the FM-HM interface [14-20]. Beyond the DMI, frustrated exchange interaction has also been supposed to be the reason for the formation of the skyrmions in centrosymmetric cubic SrFe1-xCoxO3 systems [21-23]. Very interestingly, a recent observation of the topological Hall effect in EuO1-x thin films provide a strong evidence for the presence of skyrmions in classical 2D Heisenberg ferromagnet [24-26]. The non-monotonic thickness dependence of the skyrmion is reported, which exhibit different behaviors compared to the B20 chiral magnets [10,24]. For EuO thin films, it is well established that the ferromagnetic properties can be tuned by carriers in the 5d band, such as rare earth atoms doping and oxygen vacancies [27-33]. However, the role of the carrier concentration in interacting with the skyrmions in EuO films has remain elusive.

In this paper, we report the critical role of the La doping for the topological Hall effect (THE) observed in La*x*Eu1-*x*O thin films, with *x* systematically varying from 0 to 0.20. The Curie temperature could be enhanced up to ~ 127 K with a La doping of *x* = 0.036, and exhibits a similar trend with the carrier concentration when the La doping changes. The THE emerges as the temperature increases close to the Curie temperature in La*x*Eu1-*x*O thin films, with x ranges from 0.036 to 0.20. Given that the THE stems from the local effective magnetic field experienced by itinerant electrons, the skyrmion density and radius are derived from the topological Hall resistivity. As the La doping increases, the skyrmion density increases, and skyrmion radius decreases. These results present the critical role of La doping in modulating the formation of skyrmion, skyrmion phases, skyrmion density, and skyrmion radius in Heisenberg ferromagnetic La*x*Eu1-*x*O thin films.

**II. EXPERIMENTAL** **DETAILS**

High-quality epitaxial La*x*Eu1-*x*O thin films are grown on (001)-oriented yttrium-stabilized cubic zirconia (YSZ) substrates using molecular beam epitaxy (MBE-Komponenten GmbH; Octoplus 400). Prior to the film growth, the YSZ substrates are annealed by 1 hour at 600 ºC with 3×10-7 mbar oxygen pressure, then 450 ºC is kept in following steps. For EuO thin films, a very thin layer of EuO (~ 6 nm) is grown with thermally evaporating cell of Eu, adjusted to provide a deposition rate of ~ 8 Å/min [34]. Then oxygen is introduced and its partial pressure is maintained at 1.5×10-9 mbar to continue the growth of EuO. For La*x*Eu1-*x*O thin films, La and Eu are co-evaporated from thermal effusion cells with different La deposition rates to control the atomic ratio. During growth, *in situ* reflective high energy electron diffraction (RHEED) is used to monitor and characterize the films’ crystalline quality. At the end, a thin MgO layer (~ 5 nm) is deposited *in situ* via e-beam deposition as a capping layer to avoid degradation during the measurements.

The crystalline structure is also studied using high-resolution X-ray diffraction (Bruker D8 Discover XRD system). The ferromagnetic properties are characterized using Magnetic Properties Measurement System (MPMS; Quantum Design). The topological Hall effect and electron transport properties are measured using either Van der Pauw technique or standard Hall bar configuration in an Oxford Spectromag system.

**III. RESULTS AND DISCUSSION**

The RHEED patterns of the YSZ (001) substrates and the epitaxial La*x*Eu1-*x*O films (~ 50 nm) viewed from both (100) and (110) directions are shown in Fig. 1(a-h). These sharp RHEED patterns indicate the good crystalline quality of the La*x*Eu1-*x*O films with *x* from 0 up to 0.20. Fig. 2(a) shows the XRD results for three typical La*x*Eu1-*x*O films. Clear Laue fringes around the (002) peak of EuO further confirm the epitaxial quality, and from which we estimate the film thickness around 50nm. A slight shift of the (002) peak of the epitaxial La*x*Eu1-*x*O films is observed as the La doping increases. The c-axis lattice constant derived from this peak shift is found to exhibit a tiny increase (0.56%) with La doping, as exemplified in Figure 2(b).

The magnetic moments of the La*x*Eu1-*x*O films are measured as a function of the temperature with an in-plane magnetic field of 1000 Oe. The onset of the magnetic moment could be used to identify the Curie temperature (TC), plotted in Fig. 2(c). As the La doping increases, long range ferromagnetic order strengthens and a maximum TC of ~ 127 K is achieved on the La0.036Eu0.964O film. The double-dome feature of temperature-dependent magnetization curves for La*x*Eu1-*x*O (*x* > 0) films is attributed to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions in the presence of conducting carriers [33]. As the La doping further increases, the TC decreases. The La doping dependence of TC is summarized in Fig. 2(d). To understand this phenomenon, the carrier densities of the films are measured using Hall effect in a perpendicular magnetic field from -5 T to 5 T (See supplementary Fig. S1). The carrier density at 2K also exhibits a peak at the La doping of *x* = 0.036, which is exactly the same doping level for the film with highest TC. The similar La doping dependence of the TC and the carrier density are consistent with the previous study on Gd doped EuO films [27].

A typical magnetic field dependence of the Hall resistivity is shown in Fig. 3(a), measured on the 50 nm La0.1Eu0.9O thin film at 70 K. Apart from the linear Hall region where the magnetic field is larger than 3 T, a strong non-linear Hall effect is observed, which is associated with the magnetic properties of the film. After subtracting linear ordinary Hall contribution, the anomalous Hall resistivity curve is shown in Fig. 3(b). The saturation of the anomalous Hall resistivity at high field is consistent with full alignment of the magnetization. The temperature dependence of the anomalous Hall resistivity (AH) and the longitudinal resistivity of the films are shown in supplementary Fig. S2. Besides the conventional anomalous Hall signal, the topological Hall resistivity is also observed, which is attributed to the formation of the skyrmions under such magnetic fields.

To probe the underlying mechanism for the formation of skyrmions in these ~ 50 nm La*x*Eu1-*x*O thin films, the THE is studied as a function of the temperature and out-of-plane magnetic fields for a series of La*x*Eu1-*x*O films, with *x* systematically increasing from 0 to 0.20. When the La doping is relatively low (*x* = 0.005, 0.009), an unconventional Hall resistance is observed at very low temperatures, as shown in supplementary Fig.S3. These unconventional Hall resistivity is different from the THE due to skyrmions, which usually arises at the temperatures slightly below TC [1,2,6-11]. The mechanisms of these unconventional AHE in low doped La*x*Eu1-*x*O thin films needs further studies. When the La doping exceeds 0.036, a topological Hall effect is observed on the ~50 nm La*x*Eu1-*x*O thin films with *x* = 0.036, 0.10, 0.15, 0.20, as shown in Fig. 4(a-d). For the La*x*Eu1-*x*O thin films with *x* = 0.036, skyrmions are found at the temperatures above ~50 K (Fig. 4(a)). With the increase of La dopant concentration (La0.10Eu0.90O), the skyrmions stability extends to a much wider temperature scope, as shown in Fig. 4(b). As the La doping further increases, the skyrmion phase is suppressed, indicated in Fig. 4(c-d) for the La0.15Eu0.85O and La0.20Eu0.80O thin films respectively. As the thickness of the La*x*Eu1-*x*O films decreases, a smaller skyrmion phase is observed (see supplementary Fig. S4), in line with previous reports [14]. We note that the maximum region for the presence of skyrmions is observed on the ~ 50 nm La0.10Eu0.90O thin film. The topological Hall resistivity as a function of the magnetic field at different temperatures is shown in Fig. 5, indicating the formation of skyrmions down to the temperature of 2 K.

To certify the La doping effect in these La*x*Eu1-*x*O films, the skyrmion density and radius are estimated from the THE resistivity. As the topological Hall resistivity can be expressed by [7,11,35-37]:

(1)

where *P* is the spin polarization, is the ordinary Hall coefficient, is the skyrmions density, and is one flux quantum. The skyrmion radius is estimated using the length scale of a single skyrmion and *P* = 80% [28], which decreases as the La doping increases, as shown in Fig. 6(a). The skyrmion density is found to monotonically increase as the La doping increases, as shown in Fig. 6(b). These results demonstrate the critical role of the La doping for skyrmions observed in the La*x*Eu1-*x*O thin films. Regarding the origin of the skyrmions observed in the La*x*Eu1-*x*O thin films, one possible reason is related to the La3+ impurities. As the La3+ impurities generate local electric fields around them, which could give rise to local DMI due to the inversion symmetry breaking. Thus, a higher doping of the La3+ impurities generates a higher density of skyrmions.

To unveil the origins of skyrmions formation in Heisenberg ferromagnetic La*x*Eu1-*x*O thin films, future theoretical studies are definitely needed. Besides, direct observation of skyrmions in these films using Lorentz transmission electron microscopy, nitrogen-vacancy quantum sensing, spin-polarized low-energy electron microscopy, or X-ray microscopy would be fruitful to uncover plethora of characteristics for skyrmions [9,38-40].

**IV. CONCLUSION**

In summary, the important role of the La doping for the topological Hall effect in La*x*Eu1-*x*O thin films is reported. The TC and carrier concentration exhibit similar trends as a function of the La doping. THE is observed in La*x*Eu1-*x*O thin films with *x* varies from 0.036 to 0.20, and the maximum range of the skyrmion phase is found in the La0.10Eu0.90O film, where the THE is observed down to a temperature of 2 K. Skyrmion radius shrinks continuously with increasing La doping, and skyrmion density increases with higher La concentration. These experimental results provide helpful clues for future theoretical understandings and experimental searches of magnetic skyrmions in Heisenberg ferromagnets.

**ACKNOWLEDGEMENTS**

We acknowledge the fruitful discussion with Jia Li and Ryuichi Shindou. We also acknowledge the financial support from National Basic Research Programs of China (973 program Grant Nos. 2015CB921104 and 2014CB920902) and National Natural Science Foundation of China (NSFC Grant No. 11574006 and No. 11704011).

**References:**

[1] N. Nagaosa and Y. Tokura, Nat. Nanotech. **8**, 899 (2013).

[2] A. Fert, V. Cros, and J. Sampaio, Nat. Nanotech. **8**, 152 (2013).

[3] W. Jiang, G. Chen, K. Liu, J. Zang, S. G. E. te Velthuis, and A. Hoffmann, Physics Reports **704**, 1 (2017).

[4] I. Dzyaloshinsky, J. Phys. Chem. Sol. **4**, 241 (1958).

[5] T. Moriya, Phys. Rev. **120**, 91 (1960).

[6] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Science **323**, 915 (2009).

[7] A. Neubauer, C. Pfleiderer, B. Binz, A. Rosch, R. Ritz, P. G. Niklowitz, and P. Boni, Phys. Rev. Lett. **102**, 186602 (2009).

[8] C. Pappas, E. Lelièvre-Berna, P. Falus, P. M. Bentley, E. Moskvin, S. Grigoriev, P. Fouquet, and B. Farago, Phys. Rev. Lett. **102**, 197202 (2009).

[9] X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Nature **465**, 901 (2010).

[10] X. Z. Yu, N. Kanazawa, Y. Onose, K. Kimoto, W. Z. Zhang, S. Ishiwata, Y. Matsui, and Y. Tokura, Nat Mater **10**, 106 (2011).

[11] S. X. Huang and C. L. Chien, Phys. Rev. Lett. **108**, 267201 (2012).

[12] S. Seki, X. Z. Yu, S. Ishiwata, and Y. Tokura, Science **336**, 198 (2012).

[13] T. Adams, A. Chacon, M. Wagner, A. Bauer, G. Brandl, B. Pedersen, H. Berger, P. Lemmens, and C. Pfleiderer, Phys. Rev. Lett. **108** (2012).

[14] S. Heinze, K. von Bergmann, M. Menzel, J. Brede, A. Kubetzka, R. Wiesendanger, G. Bihlmayer, and S. Blügel, Nat. Phys. **7**, 713 (2011).

[15] N. Romming, C. Hanneken, M. Menzel, J. E. Bickel, B. Wolter, K. von Bergmann, A. Kubetzka, and R. Wiesendanger, Science **341**, 636 (2013).

[16] R. Wiesendanger, Nat. Rev. Mater. **1**, 16044 (2016).

[17] W. Jiang, P. Upadhyaya, W. Zhang, G. Yu, M. B. Jungfleisch, F. Y. Fradin, J. E. Pearson, Y. Tserkovnyak, K. L. Wang, O. Heinonen, S. G. E. te Velthuis, and A. Hoffmann, Science **349**, 283 (2015).

[18] S. Woo, K. Litzius, B. Kruger, M.-Y. Im, L. Caretta, K. Richter, M. Mann, A. Krone, R. M. Reeve, M. Weigand, P. Agrawal, I. Lemesh, M.-A. Mawass, P. Fischer, M. Klaui, and G. S. D. Beach, Nat. Mater. **15**, 501 (2016).

[19] O. Boulle, J. Vogel, H. Yang, S. Pizzini, D. de Souza Chaves, A. Locatelli, T. O. Menteş, A. Sala, L. D. Buda-Prejbeanu, O. Klein, M. Belmeguenai, Y. Roussigné, A. Stashkevich, S. M. Chérif, L. Aballe, M. Foerster, M. Chshiev, S. Auffret, I. M. Miron, and G. Gaudin, Nat. Nanotech. **11**, 449 (2016).

[20] C. Moreau-Luchaire, C. Moutaﬁs, N. Reyren, J. Sampaio, C. A. F. Vaz, N. Van Horne, K. Bouzehouane, K. Garcia, C. Deranlot, P. Warnicke, P. Wohlhüter, J. M. George, M. Weigand, J. Raabe, V. Cros, and A. Fert, Nat. Nanotech. **11**, 444 (2016).

[21] S. Ishiwata, M. Tokunaga, Y. Kaneko, D. Okuyama, Y. Tokunaga, S. Wakimoto, K. Kakurai, T. Arima, Y. Taguchi, and Y. Tokura, Phys. Rev. B **84**, 054427 (2011).

[22] T. Okubo, S. Chung, and H. Kawamura, Phys. Rev. Lett. **108**, 017206 (2012).

[23] S. Chakraverty, T. Matsuda, H. Wadati, J. Okamoto, Y. Yamasaki, H. Nakao, Y. Murakami, S. Ishiwata, M. Kawasaki, Y. Taguchi, Y. Tokura, and H. Y. Hwang, Phys. Rev. B **88**, 220405 (2013).

[24] Y. Ohuchi, Y. Kozuka, M. Uchida, K. Ueno, A. Tsukazaki, and M. Kawasaki, Phys. Rev. B **91**, 245115 (2015).

[25] A. A. Belavin and A. M. Polyakov, JETP Lett. **22**, 245 (1975).

[26] A. Abanov and V. L. Pokrovsky, Phys. Rev. B **58**, R8889 (1998).

[27] T. Mairoser, A. Schmehl, A. Melville, T. Heeg, L. Canella, P. Böni, W. Zander, J. Schubert, D. E. Shai, E. J. Monkman, K. M. Shen, D. G. Schlom, and J. Mannhart, Phys. Rev. Lett. **105**, 257206 (2010).

[28] A. Schmehl, V. Vaithyanathan, A. Herrnberger, S. Thiel, C. Richter, M. Liberati, T. Heeg, M. Rockerath, L. F. Kourkoutis, S. Muhlbauer, P. Boni, D. A. Muller, Y. Barash, J. Schubert, Y. Idzerda, J. Mannhart, and D. G. Schlom, Nat. Mater. **6**, 882 (2007).

[29] A. Melville, T. Mairoser, A. Schmehl, D. E. Shai, E. J. Monkman, J. W. Harter, T. Heeg, B. Holländer, J. Schubert, K. M. Shen, J. Mannhart, and D. G. Schlom, Appl. Phys. Lett. **100**, 222101 (2012).

[30] T. Mairoser, F. Loder, A. Melville, D. G. Schlom, and A. Schmehl, Phys. Rev. B **87**, 014416 (2013).

[31] S. G. Altendorf, A. Reisner, C. F. Chang, N. Hollmann, A. D. Rata, and L. H. Tjeng, Appl. Phys. Lett. **104**, 052403 (2014).

[32] T. Yamasaki, K. Ueno, A. Tsukazaki, T. Fukumura, and M. Kawasaki, Appl. Phys. Lett. **98**, 082116 (2011).

[33] P. M. S. Monteiro, P. J. Baker, A. Ionescu, C. H. W. Barnes, Z. Salman, A. Suter, T. Prokscha, and S. Langridge, Phys. Rev. Lett. **110**, 217208 (2013).

[34] R. Sutarto, S. G. Altendorf, B. Coloru, M. Moretti Sala, T. Haupricht, C. F. Chang, Z. Hu, C. Schüßler-Langeheine, N. Hollmann, H. Kierspel, H. H. Hsieh, H. J. Lin, C. T. Chen, and L. H. Tjeng, Phys. Rev. B **79** (2009).

[35] P. Bruno, V. K. Dugaev, and M. Taillefumier, Phys. Rev. Lett. **93**, 096806 (2004).

[36] J. Matsuno, N. Ogawa, K. Yasuda, F. Kagawa, W. Koshibae, N. Nagaosa, Y. Tokura, and M. Kawasaki, Sci. Adv. **2**, e1600304 (2016).

[37] C. Liu, Y. Zang, W. Ruan, Y. Gong, K. He, X. Ma, Q.-K. Xue, and Y. Wang, arXiv:1709.08161 (2017).

[38] M. Pelliccione, A. Jenkins, P. Ovartchaiyapong, C. Reetz, E. Emmanouilidou, N. Ni, and A. C. Bleszynski Jayich, Nat. Nanotech. **11**, 700 (2016).

[39] G. Chen, A. Mascaraque, A. T. N'Diaye, and A. K. Schmid, Appl. Phys. Lett. **106**, 242404 (2015).

[40] S. Woo, K. M. Song, H.-S. Han, M.-S. Jung, M.-Y. Im, K.-S. Lee, K. S. Song, P. Fischer, J.-I. Hong, J. W. Choi, B.-C. Min, H. C. Koo, and J. Chang, Nat. Commun. **8**, 15573 (2017).

**Figure Captions**

FIG. 1. *In situ* RHEED characterization for the YSZ substrates and La*x*Eu1-*x*O thin films. (a-d) RHEED patternsof the YSZ substrates and EuO thin films viewed along the [100] and [110] directions. (e-h) RHEED patternsof the doped La*x*Eu1-*x*O thin films with *x* = 0.10, and 0.20, respectively.

FIG. 2. The crystal structures and ferromagnetic properties of the La*x*Eu1-*x*O thin films as a function of the La doping. (a) XRD results of the La*x*Eu1-*x*O thin films (~ 50 nm). Laue fringe peaks are observed around the (002) main peak of EuO. (b) The *c*-axis lattice constant as a function of La doping. (c) The temperature dependence of the normalized magnetization measured on these La*x*Eu1-*x*O thin films with an in-plane magnetic field of 0.1 T. The onset of the magnetization is used to identify the Curie temperature, as indicated by the arrows. (d) The TC of the La*x*Eu1-*x*O thin films as a function of the La doping.

FIG. 3. The ~~anomalous~~ Hall measurement on a typical film (~ 50 nm La0.1Eu0.9O). (a) Magnetic field dependence of Hall resistivity at 70 K. (b) The anomalous Hall resistivity curve () as a function of the out-of-plane magnetic field after subtraction of the linear Hall contribution. The blue curve is the magnetic field dependence of the magnetization.

FIG. 4. The phase diagrams of topological Hall resistivity in the La*x*Eu1-*x*O thin films as a function of magnetic field and temperature. (a-d) The topological Hall resistivity as a function of magnetic field and temperatures measured on the La0.036Eu0.964O, La0.10Eu0.90O, La0.15Eu0.85O, and La0.20Eu0.80O thin films, respectively. The dashed white lines are guide lines for the saturation magnetic field as a function of the temperature. The skyrmions regions are marked with “SkX”.

FIG. 5. The temperature dependent topological Hall effect on the La0.10Eu0.90O film. The topological Hall resistivity (ρTH) as a function of the magnetic field measured at different temperatures.

FIG. 6. The important role of La doping for the skyrmion density and radius. (a) The estimated skyrmion radius as a function of La doping *x*. (b) La concentration dependence of skyrmion density.