Artificial Topological Hall Effect Induced by Intrinsic Thickness Non-uniformity in Ultrathin SrRuO₃ Films

Liang Wu¹* and Yujun Zhang²

¹Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08854, USA.
²Institute for Solid State Physics, University of Tokyo, Chiba 277-8581, Japan.

*e-mail: lw590@physics.rutgers.edu

Topological Hall effect (THE) originates from the Berry phase that an electron gains when its spin follows the spatially varying non-trivial magnetization texture, such as skyrmions. Such topologically protected magnetization textures can provide great potential for information storage and processing¹. Directly imaging the skyrmions is more challenging than conducting electrical transport measurements. Hence, many researchers studied THE to indirectly attest the presence of skyrmions²-⁶. Recently Wang et al.³ demonstrated electric-field manipulation of THE-like Hall signal in ultrathin SrRuO₃ (SRO) films by ferroelectric BaTiO₃ (BTO). However, since the Anomalous Hall effect (AHE) of SRO thin films has not been fully understood yet⁷, risks remain in obtaining counterfeit artefacts that can mimic THE⁸. Here, by revisiting the data from Wang et al., we propose that the observed THE-like signal in ultrathin SRO films at low temperature is most likely dominated by the effect of intrinsic thickness non-uniformity rather than the advent of skyrmions.

As shown in Figure 1a, thickness non-uniformity is inevitable since the film surface terrace edges do not ideally copy that of the substrate⁹. Even for high quality epitaxial thin film samples, the intrinsic thickness fluctuation is at least 1 unit-cell (uc). That is, except for the major N-uc regions, (N-1) and (N+1)-uc regions always exist in a nominal N-uc film. As the thickness increases, the magnetization of SRO increases (ref. 3, Supplementary Figure 8). Accordingly, the thickness non-uniformity can be detected by magnetic force microscopy (MFM). Wang et al. have observed narrow-ribbon-like regions with different MFM contrast near the step edges in nominal 5-uc SRO films even at a fully saturated state (5 T magnetic field, ref. 3, Supplementary Figure 14). Since the surface quality of the film is nearly perfect (ref. 3, Supplementary Figure 3), the MFM signal can barely be affected by the surface topography. Therefore, the narrow-ribbon-like regions with different MFM contrast should be interpreted as regions with different thicknesses. For ultrathin films, the sample properties are strongly thickness-dependent. The averaged macroscopic properties of the whole sample can be largely affected by this intrinsic thickness non-uniformity, especially for the Hall measurements of ultrathin SRO films as discussed below.
The low-temperature AHE of ultrathin SRO films switch its sign from negative to positive\textsuperscript{2,3,6} as the thickness is diminished to 4 uc, which can become an origin of THE-like signal. The effect of film non-uniformity to the Hall signals has been discussed in ref. 8. The THE-like Hall signals can be disentangled into two AHE components originating from regions with different coercive fields and AHE signs. This mechanism is inspiring but the microscopic origin of the non-uniformity has not been revealed. Moreover, the mechanism in ref. 8 cannot explain the fact that maximal THE-like Hall signal appears at low temperature as observed by Wang \textit{et al.}, rather than at the AHE sign reversal temperature. One may consider that skyrmion-induced THE (Figure 1b, bottom left) can dominate in their samples, but based on the thickness non-uniformity and thickness-dependent AHE in ultrathin SRO, here we are proposing another possible explanation. Since the nominal 5-uc sample is consisting of 4-uc regions (positive AHE) as well as 5- and 6-uc regions (negative AHE), a mismatch of coercive fields of these regions will naturally produce the artificial THE (Figure 1b, bottom right).

Suppose thickness non-uniformity is the only origin of the THE-like signal in BTO-capped nominal 5-uc SRO sample (ref. 3, Figure 2), the 4-uc regions should possess a coercive field of \~{}1.5 T, and that of the 5- and 6-uc regions is \~{}2.6 T. By applying a -2.5 T field after a 5 T saturation, the 4-uc regions are supposed to be switched anti-parallel to 5- and 6-uc regions. Hence, 4-uc regions should appear an opposite MFM signal respect to 5- and 6-uc regions. We found the distribution of MFM contrast in Supplementary Figure 14a\textsubscript{8} of ref. 3 (5 T) is very similar but weaker compared with that of Supplementary Figure 14b\textsubscript{3} (-2.5 T). This indicates that narrow-ribbon-like regions in ref. 3, Supplementary Figure 14 are most possibly 4-uc regions, which is estimated about 23.0 \% in the nominal 5-uc sample. Such large proportion of thickness non-uniformity area cannot be neglected when analyzing the sample properties such as the Hall effect.

In addition, our thickness non-uniformity scenario provides several pivotal advantages towards the understanding of the thickness-dependent Hall signal in ultrathin SRO films. First, it is worth noting that the BTO capping layer plays a role to enlarge the mismatch of coercive fields, which can make the artificial THE more notable. This also explained the reason why THE-like signal vanishes in bare SRO samples since the nominal 4- and 5-uc bare SRO samples have comparable coercive fields at low temperature (ref. 3, Supplementary Figure 10). Second, the THE-like behavior of BTO-capped nominal 4-uc SRO sample can be understood as a major contribution of 4-uc regions with less contribution of 5-uc regions, as 3-uc SRO is much more insulating and has negligible contribution to Hall signal (ref. 3, Supplementary Figure 7). Moreover, the abrupt suppression or even extinction of THE-like signal in SRO films with nominal thickness more than 5 uc\textsuperscript{2,3,6} cannot be reasonably explained by the skyrmion formation induced by interface or surface effect. Nevertheless, this can be naturally interpreted by thickness non-uniformity.
The positive AHE contribution of 4-uc regions will be significantly suppressed as the nominal thickness exceeds 5 uc. Thus, nearly no THE-like signals can be observed in samples with nominal thickness above 5 uc at low temperature. In addition, we also noticed that previous reports about THE-like behaviors in SRO thin films with nominal 4- and 5-uc thickness are quite inconsistent with each other\textsuperscript{2,3,6}. This is possibly due to the fact that it is nearly impossible to obtain the same proportion of 4-uc regions in a nominal 5-uc sample. Within this scenario we proposed, the electric-field control of THE-like signals could be understood as electric-field control of magnetism in a ferromagnetic film with non-uniform thickness.

In conclusion, with consideration of thickness non-uniformity experimentally observed in ultrathin SRO films, THE-like signal can be artificially produced in Hall measurements. The thickness non-uniformity scenario has capabilities to provide more reasonable explanations for previously reported phenomena. We underline that thickness non-uniformity should be more seriously taken into account when analyzing the properties of ultrathin films. And properly harnessing this thickness non-uniformity can also be a promising route to create novel functionalities in thin films and heterostructures.

Reference
1. Fert, A. et al. Nat. Rev. Mater. \textbf{2}, 17031(2017).
2. Matsuno, J. et al. Sci. Adv. \textbf{2}, e1600304 (2016).
3. Wang, L. et al. Nat. Mater. \textbf{17}, 1087–1094 (2018).
4. Neubauer, A. et al. Phys. Rev. Lett. \textbf{102}, 186602 (2009).
5. Ohuchi, Y. et al. Phys. Rev. B \textbf{91}, 245115 (2015).
6. Sohn, B. et al. arXiv 1810.01615 (2018).
7. Nagaosa, N. et al. Rev. Mod. Phys. \textbf{82}, 1539-1592 (2010).
8. Kan, D., et al. Phys. Rev. B \textbf{98}, 180408 (2018).
9. Koster, G. et al. Rev. Mod. Phys. \textbf{84}, 253-298 (2012).
Figure 1. **a**, Schematic of the thickness non-uniformity in a nominal 5-uc SRO on STO(001). **b**, Schematic of thickness non-uniformity induced artificial THE versus skyrmions induced real THE.