Reversible manipulation of the topological Hall effect by hydrogen

Lin Liu, Zhixiang Ye, Ruilin Zhang, Tao Lin, Mingxia Qiu, Shunpu Li, and Hongyu An*
College of New Materials and New Energies, Shenzhen Technology University, Shenzhen 518118, China

(Received 20 March 2023; revised 11 July 2023; accepted 13 July 2023; published 24 July 2023)

We demonstrate that the topological Hall effect (THE) in a Pd/$Tm_3Fe_5O_{12}$ (TmIG) bilayer can be delicately manipulated by H_2 with a maximum 100% tunability in the reversible manner. This phenomenon originates from the variation of the Dzyaloshinskii-Moriya interaction (DMI) at the Pd/TmIG interface, which can be effectively tuned by the absorption/desorption of H_2 in Pd. Furthermore, we show that the THE in a Pd/W/TmIG trilayer barely changes by applying H_2 even the W layer is only 1-nm-thick, which indicates that the DMI generated by the W/TmIG interface is responsible for the THE, and the change in the net spin accumulation at the W/TmIG interface has no effect on the THE. Finally, we show that the Cu/TmIG interface can generate sufficient interfacial DMI to induce THE, even Cu is a light metal with weak spin-orbit coupling. Our study provides a simple approach to delicately manipulate the THE in spintronic devices and paves a practical way for developing more sensitive hydrogen sensors based on the spintronic technology.

DOI: 10.1103/PhysRevB.108.024422

I. INTRODUCTION

Magnetic skyrmions are promising for memory and logic devices due to their topological protection, nanoscale sizes, and low-energy consumption [1–7]. As the signature of the magnetic skyrmions, the topological Hall effect (THE), provides a simple way to electrically detect the skyrmions [8]. Recently, THE has attracted intensive studies in heavy metal (HM)/ferrimagnetic insulator (FMI) bilayers, due to the potential advantages of FMIs [9–13]. The low damping property, long spin transmission length, and absence of the Ohmic loss of FMI ensure high-speed response with ultra-low-energy consumption in the memory and logic devices. Applausive results have been reported in previous studies [8,14–18]. For instance, Shao *et al*. reported the observation of the THE in a Pt/$Tm_3Fe_5O_{12}$ (TmIG) bilayer [8]. Ahmed *et al*. reported a prominent THE in Pt/TmIG and Pt/$Y_3Fe_5O_{12}$ (YIG) bilayers [14]. Lee and co-workers also reported the THE in Pt/TmIG and Pt/YIG bilayers [15,16]. However, despite these applausive achievements, essential problems still remain elusive. First, the source of the interfacial Dzyaloshinskii-Moriya interaction (DMI) responsible for the THE and topological spin texture in the HM/FMI bilayer is still in controversy. In the reports of Lee and co-workers, they interpreted that the DMI responsible for the THE originates from the HM/FMI interface [15,16]. In the report of Caretta *et al*., they interpreted that this DMI stems from the intrinsic spin-orbit coupling (SOC) induced by the rare-earth orbital magnetism near the HM/FMI interface [13]. In the report of Vélez *et al*., they interpreted that the net DMI is a competition between HM/FMI and FMI/substrate interfaces with opposite chirality, and the FMI/substrate interface contributes dominantly [19]. Moreover, to effectively control the THE in a reversible way is essential for the application of THE-based spintronic devices, which is reported here. To solve these two problems is important in both fundamental and practical aspects of spintronics.

In this paper, we demonstrate that the THE in a Pd/TmIG bilayer can be reversibly manipulated by the absorption/desorption of H_2 gas in Pd, and the tunability of the THE resistivity reaches 100%. Our result confirms that the origin of the interfacial DMI is from the HM/FMI interface, which solves the controversy in previous reports. We further fabricated Pd/W/TmIG trilayer and conducted the THE measurement by applying H_2, and interestingly, we find that both the change in the net spin current and the Pd/W interface have no effect on the THE, even the W layer is only 1-nm thick. Finally, we show that the Cu/TmIG interface can generate sufficient interfacial DMI to induce THE, even Cu is a light metal with weak SOC.

II. EXPERIMENTAL METHODS

For the sample fabrication, a 3-nm-thick TmIG film was deposited on (111)-oriented $Gd_3Sc_2Ga_3O_{12}$ single-crystal substrates at 700 °C by magnetron sputtering from a TmIG target. Pure argon gas was applied for the deposition. After cooling down to the room temperature, Pd, W, and Cu layers were deposited on the TmIG surface, respectively, without breaking the vacuum. The film thickness was controlled by the deposition time with a precalibrated deposition rate. The Hall effect was measured by patterning the films into Hall bar devices. For the measurement, the devices were set up in an enclosed cell with inlet and outlet where the mixture of H_2 and N_2 gases was fed through the inlet and exhausted through the outlet. The gas pressure in the cell was kept as same as the outside ambient. The H_2 concentration Q is defined as the amount of H_2 in the mixture of the H_2 and N_2 gases. Before the measurement, pure N_2 was supplied. Then, the mixture of

*anhongyu@sztu.edu.cn

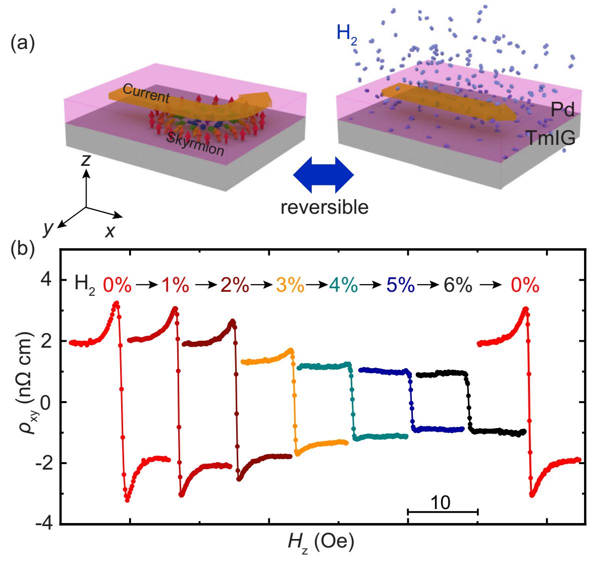

FIG. 1. (a) Schematic of the THE in Pd/TmIG bilayer reversibly manipulated by H_2 absorption and desorption. (b) Variation of the Hall resistivity ρ_{xy} in the Pd (2 nm)/TmIG (3 nm) bilayer by changing the H_2 concentration Q. The OHE are subtracted and the curves are horizontally shifted for clarity. The time interval between each measurement at different Q was about 20 minutes, which is sufficient for the completion of H_2 absorption/desorption due to the ultrathin Pd thickness.

H_2 and N_2 gases were applied for 20 min, which is sufficient for the completion of hydrogen (H) absorption/desorption due to the ultrathin Pd thickness. Therefore, the time interval between each measurement at different Q's is about 20 min.

III. RESULTS AND DISCUSSION

As shown in Fig. 1, when the current flows through the Pd/TmIG interface, the electromagnetic field generated by the skyrmion gives rise to the transverse Hall current. This will induce a peak in the measured Hall resistivity curves. By applying H_2, the skyrmion can be annihilated, leading to the vanishment of the peak in the Hall resistivity curves. The measured Hall resistivity is expressed as $\rho_{xy} = \rho_{OHE} + \rho_{AHE} + \rho_{THE}$, where ρ_{OHE}, ρ_{AHE}, and ρ_{THE} are the ordinary Hall effect (OHE) proportional to H_z, anomalous Hall effect (AHE) proportional to the perpendicular magnetization of TmIG and THE proportional to the density of the topological spin texture, respectively [15]. Figure 1(b) exhibits the Hall resistivity curves of the Pd (2 nm)/TmIG (3 nm) bilayer after subtracting the linear OHE background. When the H_2 concentration Q is 0%, a peak can be clearly observed in the Hall resistivity curve. The saturation at large magnetic field is due to the AHE, and the peak at lower magnetic field is due to the THE. It is noteworthy that the intensity of the peak gradually decreases by increasing Q. When Q is 6%, the peak vanishes, and only the AHE signal can be observed. By changing Q to 0%, the peak recovers again.

To directly compare the THE variation with H_2, we further subtracted the AHE contribution using the method reported in previous studies [14,15]. The AHE resistivity is fitted by a $\tanh(H_z/H_0)$ function, where H_0 is a fitting parameter as shown in Figs. 2(a) and 2(c). Figures 2(b) and 2(d) show the ρ_{THE} curves after the ρ_{AHE} subtraction for $Q = 0\%$ and 6%, respectively. When Q is 0%, a ρ_{THE} peak value of 1.3 nΩ cm with a THE field range of about 4 kOe can be observed. Although, the ρ_{THE} peak completely vanishes by increasing Q to 6%. This result indicates that the tunability of the THE in the Pd/TmIG bilayer can reach 100%. Figure 2(e) further exhibits the delicate manipulation of ρ_{THE} by gradually changing Q. We also conducted the measurement by increasing the repeated cycles to test the long-term performance of the THE manipulation, and confirm that the reversible change is relatively stable without obvious decay as shown in Fig. 2(f).

The above result unambiguously demonstrates that the THE in the Pd/TmIG bilayer can be delicately manipulated by H_2 with 100% tunability in the reversible manner. Pd is known as a H_2 sensing material, and the H_2 absorption/desorption in Pd has been widely studied [20,21]. The absorption of hydrogen can expand the Pd lattice. One might doubt that the strain induced by this expansion may affect the THE. In our Pd/TmIG bilayer, by increasing Q from 0% to 6%, the Pd resistance changes about 2%. According to the previous study, the 2% change of the resistance indicates a 2% atomic ratio between the absorbed H and Pd atoms [22]. With this small amount of H atoms, the expansion of Pd lattice can be neglected due to that the H atom is much smaller than Pd atom, which will first occupy the interstitial positions between the Pd atoms. Thus, the strain effect is unlikely to be responsible for the variation of the THE. Another possible origin is that the variation of the net spin accumulation at the Pd/TmIG interface induced by the modified spin-diffusion length in Pd may affect the THE since the change in the Pd resistance induced by the H_2 absorption affects its spin-diffusion length [23]. This will be verified in the latter section, which can also be excluded. The change in the THE can originate from the hydrogen-induced variation of the interfacial DMI. It is well known that the interfacial DMI is responsible for the THE and skyrmion [1]. Previous study on the spin-orbit torque in a Pd/$Ni_{81}Fe_{19}$ bilayer manipulated by H_2 has confirmed that the H_2 absorption/desorption significantly influences the Pd/$Ni_{81}Fe_{19}$ interface especially the spin mixing conductance [24]. In our Pd/TmIG bilayer, the absorbed hydrogen at the Pd/TmIG interface essentially affects the interfacial DMI, which leads to the variation of the THE. Since hydrogen only tunes the Pd/TmIG interface, our result provides a solid evidence that the DMI at the HM/TmIG interface is responsible for the THE and skyrmion in the HM/FMI bilayers.

Our paper demonstrates that H_2 absorption in Pd is a powerful tool to tune the Pd/H_2-unreactive-material interface. Therefore, in the following, we fabricated Pd (1.5-nm)/W (1-nm)/TmIG (3-nm) trilayer to investigate whether the change in the net spin accumulation at the HM/TmIG plays a role on the THE. Figure 3(a) shows the OHE-subtracted Hall resistivity curves by systematically changing Q. The curves were vertically shifted for clarity. By increasing Q from 0% to 100%, it can be seen that although ρ_{xy} gradually increases, the peak due to the THE barely changes. Figure 3(b) shows that the electrical resistance of the device gradually increases due to the H_2 absorption, which confirms the H_2 absorption of Pd, consistent with previous reports [21,24]. We separately plotted ρ_{AHE} and ρ_{THE} in Figs. 3(c) and 3(d) to clearly

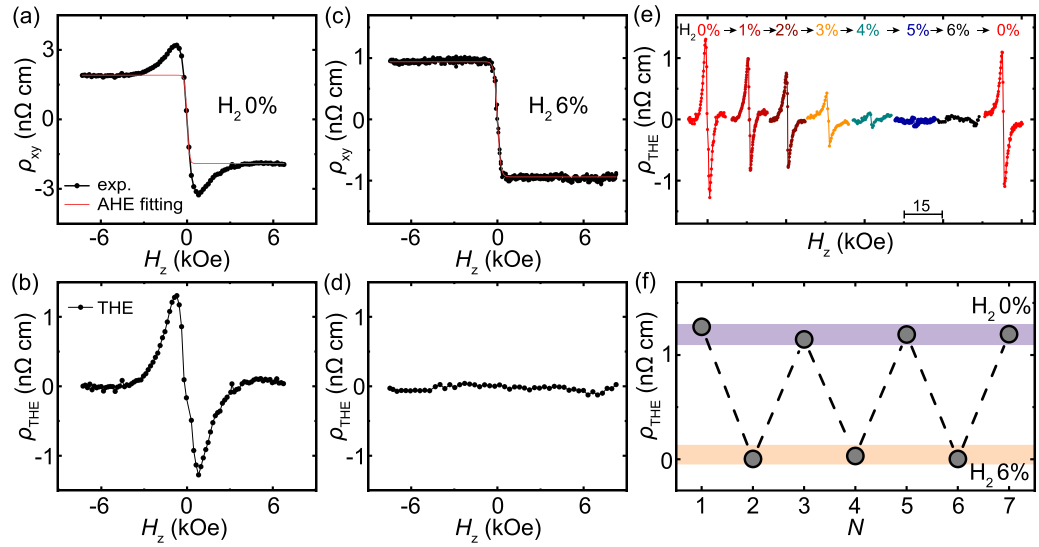

FIG. 2. (a) The Hall resistivity ρ_{xy} and (b) the THE resistivity ρ_{THE} curves of the Pd (2 nm)/TmIG (3 nm) bilayer when the H_2 concentration Q is 0%. (c) The corresponding ρ_{xy} and (d) ρ_{THE} curves when Q is 6%. (e) Variation of the ρ_{THE} curves by changing Q. The curves are horizontally shifted for clarity. (f) Reversible change of ρ_{THE} by alternatively changing Q.

compare their variations with Q. As shown in Fig. 3(c), ρ_{AHE} gradually increases with Q. This is due to the enhancement of the net spin accumulation at the W/TmIG interface. It is known that the sign of the spin Hall angles of Pd and W are opposite [25]. Therefore, in the Pd/W/TmIG trilayer, the spin current generated from the Pd layer injects into the W layer and partially cancels out the spin current generated in the W layer, which weakens the net spin accumulation at the W/TmIG interface. When applying H_2, the absorption of hydrogen at the Pd/W interface reduces the interfacial transparency, and cuts down the spin current injected into the W layer, leading to an enhancement of the net spin accumulation at the W/TmIG interface. This will enhance the AHE resistivity signal due to that the AHE resistivity signal in the Pd/W/TmIG trilayer originates from the spin Hall magnetoresistance [26]. Figure 3(d) exhibits the Q dependence of ρ_{THE}. Interestingly, ρ_{THE} barely changes with Q, even we applied maximum 100% H_2. This result indicates that the change in the net spin accumulation at the W/TmIG interface has no effect on the THE. The average value of the ρ_{THE} is obtained as about 0.28 nΩ cm in the Pd/W/TmIG trilayer.

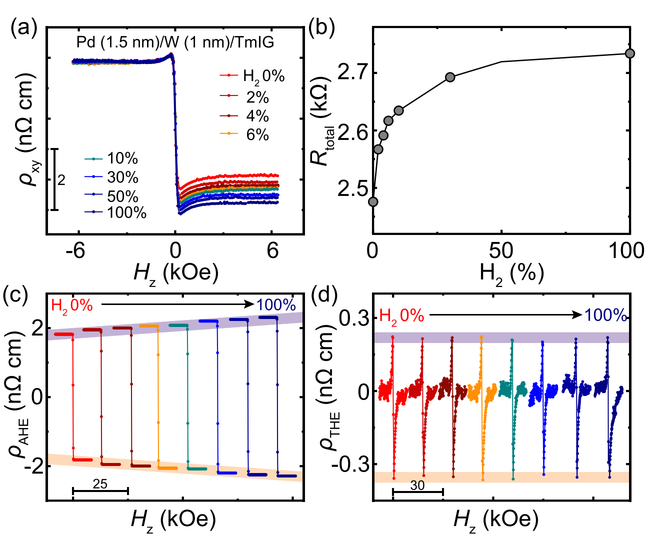

FIG. 3. (a) The Hall resistivity ρ_{xy} curves of the Pd (1.5 nm)/W (1 nm)/TmIG (3 nm) trilayer by changing Q. The curves are vertically shifted for clarity. Variation of the (b) electrical resistance R_{total}, (c) AHE resistivity ρ_{AHE} and (d) THE resistivity ρ_{THE} curves with Q. the curves in (c) and (d) are horizontally shifted for clarity.

The above paper indicates that the net spin accumulation is responsible for the AHE, and the DMI at the metal/TmIG interface is responsible for the THE, which gives us a practical approach to study whether a light metal/TmIG interface can induce sufficient interfacial DMI to generate THE. In the following, we fabricated a Pd (1.5-nm)/Cu (1-nm)/TmIG (3-nm) trilayer and conducted the measurement. Cu is known to have weak SOC with small spin Hall angle (∼0.0032) and long spin-diffusion length (∼500 nm) [27]. Therefore, in the Pd/Cu/TmIG trilayer, the spin current generated from the Pd layer transports through the Cu layer and reaches the Cu/TmIG interface, which is responsible for the AHE signal. If the Cu/TmIG interface has sufficient interfacial DMI, then, the THE signal can be observed. For comparison, we first fabricated a Cu (2-nm)/TmIG (3-nm) bilayer and conducted the measurement. As shown in Fig. 4(a), no signal can be observed in the Cu/TmIG bilayer, consistent with the weak SOC of Cu. Figure 4(b) demonstrates the Hall resistivity curves of the Pd/Cu/TmIG trilayer by changing Q. The THE can be clearly observed in the curves. ρ_{AHE} and ρ_{THE} were separately plotted in Figs. 4(c) and 4(d). By increasing Q from 0% to 100%, ρ_{AHE} gradually decreases. This result is completely opposite to that of the Pd/W/TmIG trilayer and well consistent with our expectation. In the Pd/W/TmIG trilayer, the net spin accumulation at the W/TmIG interface is determined by the competition between Pd and W. Although, in the Pd/Cu/TmIG trilayer, the net spin accumulation at the Cu/TmIG interface is determined by the spin generation

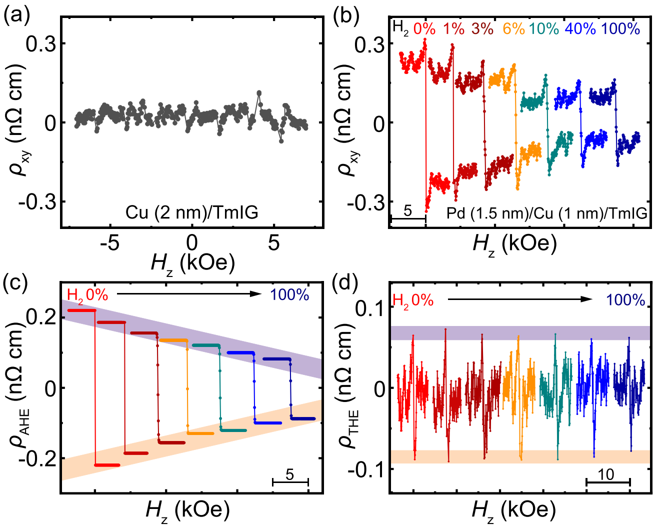

FIG. 4. (a) The Hall resistivity ρ_{xy} curves of the Cu (2 nm)/TmIG (3 nm) bilayer. A 1-nm-thick SiO_2 layer was capped to protect Cu from oxidation. (b) The Hall resistivity ρ_{xy} curves of the Pd (1.5 nm)/Cu (1 nm)/TmIG (3 nm) trilayer by changing Q. (c) AHE resistivity ρ_{AHE} and (d) THE resistivity ρ_{THE} curves with Q. the curves are horizontally shifted for clarity.

in Pd and the spin transport through Cu. The hydrogen incorporation at the Pd/Cu interface reduces the spin mixing conductance, which cuts down the spins injected into the Cu layer, leading to the reduction of the spin accumulation at the Cu/TmIG interface. As shown in Fig. 4(d), ρ_{THE} barely changes with Q, which confirms that the THE is determined by the Cu/TmIG interfacial DMI, consistent with the THE result in the Pd/W/TmIG trilayer. The average value of the ρ_{THE} is obtained as about 0.077 nΩ cm in the Pd/Cu/TmIG trilayer, which is smaller than that in the Pd/W/TmIG trilayer due to the weak SOC of Cu. Our paper confirms that even a light metal with weak SOC attached with TmIG can still generate sufficient interfacial DMI to induce THE, which may stimulate the study of the DMI and the skyrmion in light metal/magnet heterostructures in the future.

IV. CONCLUSIONS

To summarize, we demonstrate that the THE in a Pd/TmIG bilayer can be delicately manipulated by H_2 with a maximum 100% tunability in the reversible manner. This phenomenon originates from the variation of the DMI at the Pd/TmIG interface, which can be effectively tuned by the absorption and desorption of H_2. Furthermore, we show that the THE in a Pd/W/TmIG trilayer barely changes by applying H_2 even the W layer is only 1-nm thick, which indicates that the DMI induced at the W/TmIG interface is responsible for the THE, and the change in the net spin accumulation at the W/TmIG interface has no effect on the THE. Finally, we show that the Cu/TmIG interface can generate sufficient interfacial DMI to induce THE, even Cu is a light metal with weak SOC. Our paper provides a simple approach to delicately manipulate the THE and skyrmion in spintronic devices, and paves a practical way for developing more sensitive hydrogen sensors based on the spintronic technology.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Grant No. 52001215), Guangdong Basic and Applied Basic Research Foundation (Grant No. 2021A1515012055), the industrial research and development Project of SZTU (Grant No. KY2022QJKCZ005), the Natural Science Foundation of Top Talent of SZTU (Grant No. 2019210).

[1] A. Fert, V. Cros, and J. Sampaio, Nat. Nanotechnol. **8**, 152 (2013).
[2] C. Moreau-Luchaire, C. Moutafis, N. Reyren, J. Sampaio, C. Vaz, N. Van Horne, K. Bouzehouane, K. Garcia, C. Deranlot, P. Warnicke, P. Wohlhuter, J. George, M. Weigand, J. Raabe, V. Cros, and A. Fert, Nat. Nanotechnol. **11**, 444 (2016).
[3] S. Woo, K. Litzius, B. Krüger, M.-Y. Im, L. Caretta, K. Richter, M. Mann, A. Krone, R. M. Reeve, M. Weigand, P. Agrawal, I. Lemesh, M. Mawass, P. Fischer, M. Klaui, and G. Beach, Nature Mater. **15**, 501 (2016).
[4] 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. te Velthuis, and A. Hoffmann, Science **349**, 283 (2015).
[5] S. Seki, X. Yu, S. Ishiwata, and Y. Tokura, Science **336**, 198 (2012).
[6] N. Nagaosa and Y. Tokura, Nat. Nanotechnol. **8**, 899 (2013).
[7] G. Yu, P. Upadhyaya, Q. Shao, H. Wu, G. Yin, X. Li, C. He, W. Jiang, X. Han, P. K. Amiri, and K. Wang, Nano Lett. **17**, 261 (2017).
[8] Q. Shao, Y. Liu, G. Yu, S. K. Kim, X. Che, C. Tang, Q. L. He, Y. Tserkovnyak, J. Shi, and K. L. Wang, Nat. Electron. **2**, 182 (2019).
[9] C. O. Avci, A. Quindeau, C.-F. Pai, M. Mann, L. Caretta, A. S. Tang, M. C. Onbasli, C. A. Ross, and G. S. Beach, Nature Mater. **16**, 309 (2017).
[10] C. O. Avci, E. Rosenberg, M. Baumgartner, L. Beran, A. Quindeau, P. Gambardella, C. A. Ross, and G. S. Beach, Appl. Phys. Lett. **111**, 072406 (2017).
[11] Q. Shao, C. Tang, G. Yu, A. Navabi, H. Wu, C. He, J. Li, P. Upadhyaya, P. Zhang, S. A. Razavi, Q. L. He, Y. Liu, P. Yang, S. K. Kim, C. Zheng, Y. Liu, L. Pan, R. K. Lake, X. Han, Y. Tserkovnyak *et al.*, Nat. Commun. **9**, 3612 (2018).
[12] C. Guo, C. Wan, M. Zhao, H. Wu, C. Fang, Z. Yan, J. Feng, H. Liu, and X. Han, Appl. Phys. Lett. **114**, 192409 (2019).
[13] L. Caretta, E. Rosenberg, F. Büttner, T. Fakhrul, P. Gargiani, M. Valvidares, Z. Chen, P. Reddy, D. A. Muller, C. A. Ross, and G. S. Beach, Nat. Commun. **11**, 1090 (2020).
[14] A. S. Ahmed, A. J. Lee, N. Bagués, B. A. McCullian, A. M. Thabt, A. Perrine, P.-K. Wu, J. R. Rowland, M. Randeria, P. C.

Hammel, D. W. McComb, and F. Yang, Nano Lett. **19**, 5683 (2019).

[15] A. J. Lee, A. S. Ahmed, J. Flores, S. Guo, B. Wang, N. Bagués, D. W. McComb, and F. Yang, Phys. Rev. Lett. **124**, 107201 (2020).

[16] A. J. Lee, S. Guo, J. Flores, B. Wang, N. Bagués, D. W. McComb, and F. Yang, Nano Lett. **20**, 4667 (2020).

[17] Z. Xu, Q. Liu, Y. Ji, X. Li, J. Li, J. Wang, and L. Chen, ACS Appl. Mater. Interfaces **14**, 16791 (2022).

[18] T. N. Nunley, S. Guo, L.-J. Chang, D. Lujan, J. Choe, S.-F. Lee, F. Yang, and X. Li, Phys. Rev. B **106**, 014415 (2022).

[19] S. Vélez, J. Schaab, M. S. Wörnle, M. Müller, E. Gradauskaite, P. Welter, C. Gutgsell, C. Nistor, C. L. Degen, M. Trassin, M. Fiebig, and P. Gambardella, Nat. Commun. **10**, 4750 (2019).

[20] L. L. Jewell and B. H. Davis, Appl. Catal. A: Gen. **310**, 1 (2006).

[21] A. Mirzaei, H. R. Yousefi, F. Falsafi, M. Bonyani, J.-H. Lee, J.-H. Kim, H. W. Kim, and S. S. Kim, Int. J. Hydrogen Energy **44**, 20552 (2019).

[22] M. Nicolas, L. Dumoulin, and J. P. Burger, J. Appl. Phys. **60**, 3125 (1986).

[23] S. Watt and M. Kostylev, Phys. Rev. B **101**, 174422 (2020).

[24] H. An, S. Haku, Y. Kageyama, A. Musha, Y. Tazaki, and K. Ando, Adv. Funct. Mater. **30**, 2002897 (2020).

[25] S. Karube, N. Tezuka, M. Kohda, and J. Nitta, Phys. Rev. Appl. **13**, 024009 (2020).

[26] H. Nakayama, M. Althammer, Y.-T. Chen, K. Uchida, Y. Kajiwara, D. Kikuchi, T. Ohtani, S. Geprägs, M. Opel, S. Takahashi, R. Gross, G. E. W. Bauer, S. T. B. Goennenwein, and E. Saitoh, Phys. Rev. Lett. **110**, 206601 (2013).

[27] H. L. Wang, C. H. Du, Y. Pu, R. Adur, P. C. Hammel, and F. Y. Yang, Phys. Rev. Lett. **112**, 197201 (2014).