Unconventional Nonreciprocal Voltage Transition in Ag_2Te Nanobelts

Peng-Liang Leng(冷鹏亮)[1†], Xiang-Yu Cao(曹翔宇)[1†], Qiang Ma(马强)[1], Lin-Feng Ai(艾临风)[1], Yu-Da Zhang(张钰达)[1], Jing-Lei Zhang(张警蕾)[2], and Fa-Xian Xiu(修发贤)[1*]

[1]*State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China*
[2]*Anhui Province Key Laboratory of Condensed Matter Physics at Extreme Conditions, High Magnetic Field Laboratory of the Chinese Academy of Sciences, Hefei 230031, China*

(Received 11 August 2023; accepted manuscript online 3 November 2023)

Nonreciprocal effects are consistently observed in noncentrosymmetric materials due to the intrinsic symmetry breaking and in high-conductivity systems due to the extrinsic thermoelectric effect. Meanwhile, nonreciprocal charge transport is widely utilized as an effective experimental technique for detecting intrinsic unidirectional electrical contributions. Here, we show an unconventional nonreciprocal voltage transition in topological insulator Ag_2Te nanobelts. The nonreciprocal voltage develops from nearly zero to giant values under the applied current I_{ac} and external magnetic fields, while remaining unchanged under various current I_{dc}. This unidirectional electrical contribution is further evidenced by the differential resistance (dV/dI) measurements. Furthermore, the transition possesses two-dimensional properties under a tilted magnetic field and occurs when the voltage between two electrodes exceeds a certain value. We propose a possible mechanism based on the development of edge channels in Ag_2Te nanobelts to interpret the phenomenon. Our results not only introduce a peculiar nonreciprocal voltage transition in topological materials but also enrich the understanding of the intrinsic mechanism that strongly affects nonreciprocal charge transport.

DOI: 10.1088/0256-307X/40/12/127201

Symmetry breaking in noncentrosymmetric materials gives rise to many exotic physical phenomena, including the nonreciprocal effect. It is a traditional nonlinear response under external magnetic fields and applied electrical fields with nonequal resistances when the current is applied along opposite directions.[1–3] The p–n junction is the most famous nonreciprocal device, characterized by a nonlinear I–V relation resulting from inequivalent charge occupation-induced inversion breaking at the junction interfaces. On the other hand, the rectification effect has been observed as one of the prominent nonreciprocal effects in polar crystals when the applied electrical field is perpendicular to the external magnetic field. The emergent polar current is along the direction of the cross product of the magnetic and electric fields.[2] The intrinsic origins of the nonreciprocal effect include Cooper pair fluctuations in superconductors around the transition temperature,[4–7] anomalous charge and spin-current transformations for the spin-momentum locking of surface states in topological insulators,[3,8–10] high-order responses from Berry curvature dipoles in Weyl semimetals,[11–15] large nonreciprocal resistances from quantum anomalous Hall edge states in magnetic topological insulator thin films,[16,17] and Fermi surface topology.[18] In addition, the thermoelectric effect of the nonuniform current heating in nanostructures could also be an extrinsic origin of the nonreciprocal effect.[19]

Experimentally, the second harmonic method has been used to detect nonreciprocal resistance by applying a current with a frequency of ω but measuring the voltage with a frequency of 2ω in a lock-in amplifier.[2] The measurement configuration is shown in the inset of Fig. 1(a). In conventional Hall bar mesoscopic devices, nontrivial nonreciprocal resistance often emerges between the longitudinal electrodes in noncentrosymmetric materials and scales bilinearly with external magnetic field B and applied electrical field E. It retains its odd function with respect to the magnetic field B in nonlinear planar Hall effects,[3,5,8–10,20–22] superconducting fluctuations,[5] and magnetization dynamics.[16,17,23] In contrast, in Weyl semimetals with finite Berry curvature dipoles,[11] the nonreciprocal resistance appears along the transverse direction because the nonlinear Hall resistance is under time-reversal symmetry and proportional to the high order of the applied current I_{ac}. Although the nonreciprocal resistance from an extrinsic thermoelectric origin[19] can be odd or even with respect to the external magnetic field B owing to a nonuniform temperature gradient from its asymmetric device geometry, it is still linearly proportional to the applied current I_{ac}. At present, the nonreciprocal transport signals are linear with one or a high order of I_{ac} and are odd with B in intrinsic inversion symmetry breaking systems. However, a nonlinear nonreciprocal voltage that has an even function with B has never been reported.

In this Letter, we report the giant nonlinear nonreciprocal voltage in topological insulator Ag_2Te nanobelts.

†These authors contributed equally to this work.
*Corresponding author. Email: Faxian@fudan.edu.cn
© 2023 Chinese Physical Society and IOP Publishing Ltd

The unconventional nonreciprocal voltage grows nonlinearly with increasing current I_{ac} and reads an even function with external magnetic field B. Alternative differential resistance (dV/dI) measurements are carried out to confirm the giant nonlinear nonreciprocal voltage. Furthermore, the transition occurs when the voltage between two electrodes exceeds a certain value and shows two-dimensional properties under a tilted magnetic field. All these unconventional nonreciprocal voltage transitions can be well interpreted by the possible development of edge channels in the Ag_2Te nanobelts.

Ag_2Te has been predicted theoretically and confirmed experimentally to be a new type of topological insulator with a narrow bulk gap.[24–27] The high-quality nanobelts synthesized by the chemical vapor deposition method possess ultra-high mobility[26] and ultra-confined anisotropic acoustic terahertz plasmon polaritons[28] for a special $(\bar{1}01)$ surface plane with the b-axis lying in the plane. The prominent quantum oscillations from the two-dimensional surface states and three-dimensional bulk states can be observed in Ag_2Te nanobelts, which have high mobility and low carrier density.[26]

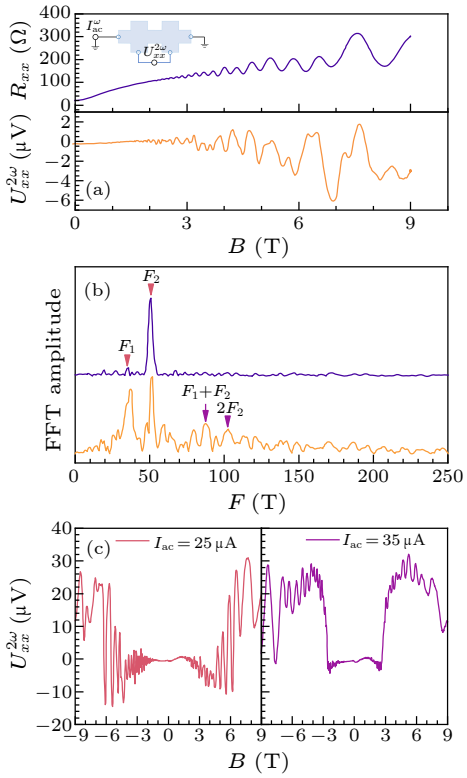

Fig. 1. Giant nonreciprocal response from the thermoelectric effect in Ag_2Te nanobelt #1. (a) The contrast between the longitudinal magnetoresistance R_{xx} and nonreciprocal voltage $U_{xx}^{2\omega}$ under external perpendicular magnetic fields. The applied current is 5 μA. Inset: the measurement configuration. (b) The resultant fast Fourier transform spectrum from resistance and nonreciprocal voltage. (c) The giant quantum oscillations from the thermoelectric effect merge with the transition in nonreciprocal voltage when the applied current increases to $I_{ac} = 25$ μA and 35 μA.

Figure 1 shows quantum oscillations in a typical Ag_2Te nanobelt. High-frequency quantum oscillations in longitudinal resistance R_{xx} originate from the surface states, according to previous research.[26] The nonreciprocal voltage $U_{xx}^{2\omega}$ also shows strong oscillations with nearly zero background, which results from the extrinsic thermoelectric effects inducing periodic changes in the density of states (DOS) when the discrete Landau levels develop under external magnetic fields. The corresponding fast Fourier transform is extracted as shown in Fig. 1(b) to capture the frequencies of the quantum oscillations, which are proportional to the cross section of the Fermi surface intersecting with Fermi energy E_F. The F_1 and F_2 are assigned to the bulk states and surface states,[26] respectively. The high peak in R_{xx} indicates the dominant surface state behavior, while the multiple-frequency response in $U_{xx}^{2\omega}$ shows a competing behavior. Apparently, the thermoelectric origin of nonreciprocal voltage $U_{xx}^{2\omega}$ has the inherent advantage of detecting the details of the Fermi surface more sensitively, capturing more information from the bulk state other than a completely surface-state dominated response. More interestingly, when the applied current I_{ac} is enhanced from 5 μA in Fig. 1(a) to 25 μA and 35 μA in Fig. 1(c), an unconventional transition emerges, accompanied by enhanced extrinsic quantum oscillations. The background of the nonreciprocal voltage $U_{xx}^{2\omega}$ changes from nearly zero at low magnetic fields to giant responses at high magnetic fields, as shown in Fig. 1(c).

In order to investigate the anomalous nonreciprocal voltage transition, we conduct second harmonic measurements of samples without quantum oscillations. Different from previous nonreciprocal studies,[2,3] the nonreciprocal voltage in Ag_2Te nanobelts grows nonlinearly and displays a sharp transition from nearly zero to a giant response when sweeping the applied current I_{ac}, but it reduces at higher temperatures, for example, at 100 K, as shown in Fig. 2(a). This nonlinear relation distinguishes this phenomenon from the earlier intrinsic and extrinsic origins of the nonreciprocal effect. In a traditional Hall bar device with eight electrodes, we explore the magnetic-field-dependent nonreciprocal voltage transition by applying current with a frequency ω along the x direction [Fig. 2(b)]. The corresponding resistance is shown in Fig. S1 of the Supplementary Materials. Various methods for connecting two electrodes are employed. The most prominent transition appears between electrons with diagonal connections. The large transitions occur under both positive and negative magnetic fields from three channels along the longitudinal direction. In contrast, three channels along the transverse direction have nearly zero response, proving that the giant nonreciprocal voltage transitions come from the two electrodes along the longitudinal direction. However, there are some confusing features as follows: Firstly, voltages $U_{26}^{2\omega}$ and $U_{84}^{2\omega}$ exhibit high asymmetry with the magnetic field while they are antisymmetric with each other. Secondly, the sum of voltages U_{23} and U_{34} should be equal to the voltage U_{34}, which is an inevitable result in resistance but invalid in nonreciprocal voltage, as shown in

Figs. S2(b)–S2(c). Furthermore, the second harmonic voltages between electrodes in connections 2–6 and 8–4 are greater than those in 2–3, 3–4, and 2–4.

Taking one typical channel with a symmetric transition as an example, we measure the nonreciprocal voltage as a function of the magnetic field at various current I_{ac}, as displayed in Fig. 2(c). The transition follows the rule that the critical magnetic field decreases when the applied current I_{ac} increases from 3 μA to 30 μA. Apart from the changes in the critical field, the initial and final state nonreciprocal voltages at various current I_{ac} remain similar, showing that the two longitudinal electrons acquire a constant nonreciprocal voltage when the external magnetic field exceeds the critical magnetic field. To exclude the extrinsic contribution from thermoelectric effects, we then apply different current I_{dc} while the current $I_{ac} = 7$ μA is fixed, as shown in Fig. 2(d). Surprisingly, the critical magnetic field of the nonreciprocal voltage transition remains unchanged, while the final state transition voltage under a higher magnetic field varies with the applied I_{dc} from −20 μA to 20 μA because of the Joule heating. Other data acquired under different current I_{ac} are shown in Fig. S3 of the Supplementary Materials. It is also noted that the transition is not only related to current I_{ac}, but is also frequency independent, as shown in Fig. S2(a).

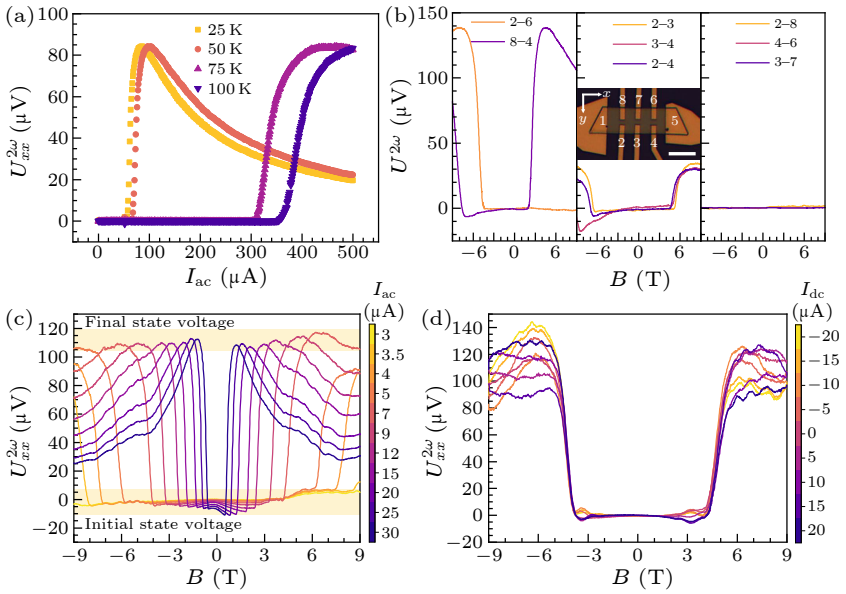

Fig. 2. Unconventional nonreciprocal voltage in Ag_2Te nanobelt #2. (a) The nonreciprocal voltages under different temperatures develop from nearly zero into a giant response when sweeping the applied current. The transition is sharp at a critical current. (b) The nonreciprocal voltages between different electrons under a perpendicular magnetic field. The left panel shows the giant responses from electrons with diagonal connections. The middle panel displays the large nonreciprocal voltages from three channels along the longitudinal direction. The right panel shows three channels along the transverse direction that have no nonreciprocal voltages. Inset is the optical image of the Ag_2Te nanobelt with a traditional Hall bar structure, the electrodes are labeled, and the scale bar is 10 μm. (c) The magnetic-field-dependent nonreciprocal voltages under different current I_{ac} varying from 3 μA to 30 μA. (d) The magnetic-field-dependent nonreciprocal voltages under different current I_{dc} varying from −20 μA to 20 μA when $I_{ac} = 7$ μA.

To validate the unconventional nonreciprocal voltage transition in Ag_2Te nanobelts, we perform differential resistance (dV/dI) measurements by connecting a constant current I_{ac} and a gradually changing current I_{dc} with a small step in series, which is an alternative technique to reflect the local DOS around Fermi energy E_F and is widely employed in superconducting measurements.[29,30] For a conductive topological insulator, the dV/dI reflects the slope of the I–V relation. It should be symmetric with respect to applied positive and negative current I_{dc}, with the asymmetry contribution arising from the unidirectional intrinsic nonreciprocal part,[1] as shown in Fig. 3(a). After deducting the symmetric part from the dV/dI measurements, we can obtain $\Delta dV/dI$ as a function of the current I_{dc}, the magnitude of which represents the intensity of the nonreciprocal effect. In a channel with an asymmetric nonreciprocal voltage transition, as shown in Fig. 3(b), the negative magnetic field can induce the transition. The corresponding $\Delta dV/dI$ curves under various magnetic fields show an antisymmetric behavior with the current I_{dc}, as displayed in Fig. 3(c). At a negative magnetic field, the $\Delta dV/dI$ grows rapidly to a maximum and shrinks linearly with the increased current I_{dc}, as shown by purple curves in Fig. 3(c). With a positive magnetic field, the $\Delta dV/dI$ grows linearly and gradually with the current I_{dc} into infinite and negligible values, as shown by the yellow curves in Fig. 3(c). The asymmetry response of the $\Delta dV/dI$ fits well with the nonreciprocal voltage measurement in Fig. 3(b), proving the unconventional nonreciprocal voltage transition in Ag_2Te nanobelts.

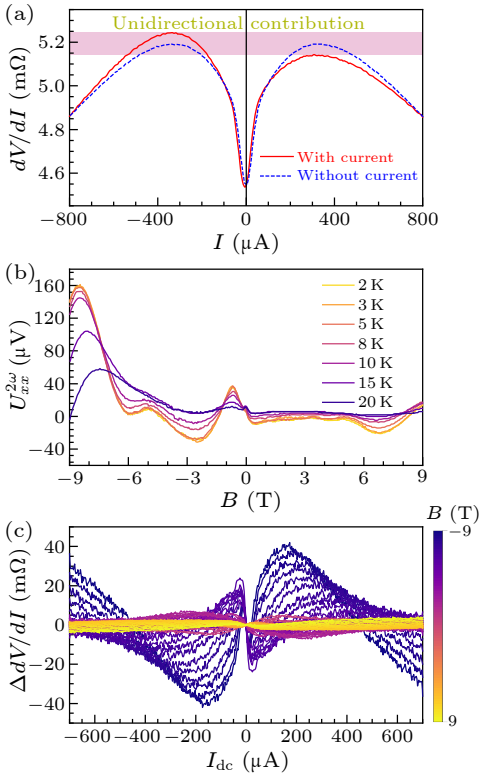

Fig. 3. Unconventional nonreciprocal voltage transition measured through the dV/dI method. (a) Illustration of the unidirectional resistance through dV/dI measurement. The difference between the dV/dI with current and the dV/dI without current corresponds to nonreciprocal voltage. (b) The nonreciprocal voltage results in a giant response under one side when the magnetic field is negative under various temperatures. (c) The $\Delta dV/dI$ as a function of the applied current, showing large signals under one side when the magnetic field is negative.

The second harmonic measurement under the tilted magnetic field can be used to distinguish the dimensionality of the nonreciprocal voltage transition. The nonreciprocal voltage $U_{xx}^{2\omega}$ as a function of the $B\cos\theta$ converges into one curve in Fig. 4(b), where θ is the angle between the magnetic field and the out-of-plane direction. When the magnetic field is tilted, the transition persists but occurs under an enlarged critical magnetic field, as shown in Fig. S5(a). The critical magnetic field can be well fitted to 2D function $1/\cos\theta$, indicating two-dimensional properties.

Having clarified the experimental variable that can tune nonreciprocal transition, we now look for the cause of the anomalous nonreciprocal transition. Taking channel $U_{23}^{2\omega}$ in Fig. 2(b) as an example, we find that when the transition occurs, whether under a positive or negative magnetic field, the voltage between these two electrodes grows to exceed 4.36 mV, as indicated by the dashed lines in Fig. 4(c). We recheck other channels, and this phenomenon is consistently observed without exception, as listed in Table 1. In other words, the nonreciprocal voltage transition from the nearly zero state to a giant response state only occurs when the voltage between the two longitudinal electrodes exceeds a certain threshold. To explain this behavior, we propose the possible development of edge channels within Ag_2Te nanobelts. When an electrical field is applied along the x direction across the nanobelts, these special edge channels form along the two sides of the edge surface, the direction of which is the same as the current direction. Their contribution to the nonreciprocal voltage develops from zero to prominent when the actual voltage exceeds a critical breakdown voltage, as schematized in Fig. 4(a). The direction of the edge channels results in an even function with the magnetic field because the direction of the applied currents remains unchanged when we reverse the direction of the magnetic field. Under an external magnetic field, the topological surface state induces quantum oscillation in the second harmonic voltage. The edge roughness[31] of the edge state could play a crucial role in the unconventional nonreciprocal voltage transition in Ag_2Te nanobelts. The applied current mainly travels within a narrow region near the edge, resulting in a second harmonic voltage transition when the magnetic field or current exceed critical values.

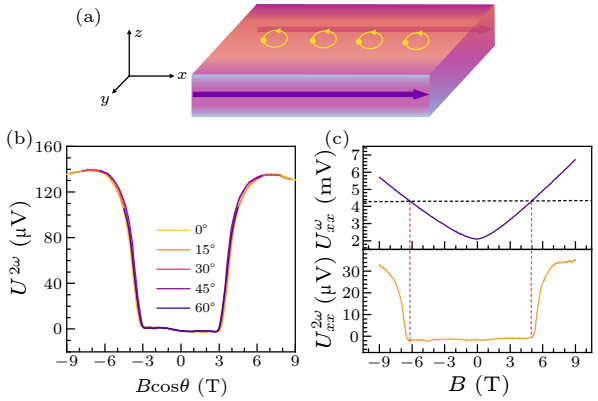

Fig. 4. Nonreciprocal voltage from edge channel in the Ag_2Te nanobelt. (a) Schematic of the special edge channel along the direction of the applied electrical field. (b) The angle-dependent nonreciprocal responses under a tilted magnetic field. All curves converge as a function of the $B\cos\theta$, showing typical two-dimensional properties. (c) The voltage contrast between the voltage difference along two longitudinal electrons and nonreciprocal voltage transition. The dashed lines mark the transition at the critical magnetic field.

Table 1. The voltage between electrodes showing the transitions in Ag_2Te nanobelt #2.

Electrodes connection	2–3	8–4	8–3	7–4	3–6	3–4	2–4
U^{ω} (mV)	4.36	4.12	4.29	4.25	4.44	4.30	4.30

Based on the assumption of edge channels, we can explain the asymmetry of the nonreciprocal voltage transition behavior between two electrodes connected diagonally since the voltage between these two electrodes shows strong asymmetry with the mixture of magnetoresistance and Hall resistance. On the other hand, the transverse channels have zero nonreciprocal voltage because the two edge channels along the two sides of the nanobelts cancel each other, leading to a giant nonreciprocal voltage be-

tween the longitudinal channels. As a consequence, the voltage along one longitudinal side satisfies the principle of linear superposition but the nonreciprocal voltage is violated because the edge channels contribute giant but infinite nonreciprocal voltage across any two electrodes, regardless of the magnitude of the applied current $I_{\rm dc}$. Moreover, the second harmonic voltages in electrode connections 2–6 and 8–4 include the contribution from the longitudinal edge state and the drain electrode. For the topological chiral edge state, the voltage drop between the source and drain electrodes would be the applied current times quantum resistance h/e^2. However, for the possible edge state in Ag_2Te nanobelts, a considerable voltage drop still occurs, providing the corresponding second harmonic voltage from the thermoelectrical and quantum transport contributions. Therefore, the second harmonic voltages in electrode connections 2–6 and 8–4 are greater than those in other channels along the longitudinal direction. However, further understanding of the physical mechanism of edge channels calls for more theoretical and experimental investigations.

A possible structural phase transition can be excluded since the critical electrical field should be the same across the entire nanobelt. In other words, two longitudinal channels with different lengths should result in different voltages. However, in our experiment, the critical voltage between any channels remains the same.

In summary, we show that in topological insulator Ag_2Te nanobelts, a giant nonreciprocal voltage can be induced by applying a large current $I_{\rm dc}$ and an external magnetic field between two electrodes containing a longitudinal contribution. The transition occurs under certain conditions whereby the actual voltage between any electrodes exceeds a critical breakdown voltage and then contributes a giant but infinite nonreciprocal voltage, which is further proved by the dV/dI measurement. The assumption of edge channels as the underlying mechanism for the giant nonreciprocal transport in Ag_2Te nanobelts provides a possible intrinsic mechanism and experimental viewpoint from which we can generate giant unidirectional intrinsic nonreciprocal contributions. This insight could enhance our comprehension of the nonreciprocal effect.

Acknowledgments. This work was supported by the National Natural Science Foundation of China (Grant Nos. 52225207, 11934005, and 52350001), the Shanghai Pilot Program for Basic Research - FuDan University 21TQ1400100 (Grant No. 21TQ006), and the Shanghai Municipal Science and Technology Major Project (Grant No. 2019SHZDZX01).

References

[1] Tokura Y and Nagaosa N 2018 *Nat Commun.* **9** 3740
[2] Ideue T, Hamamoto K, Koshikawa S, Ezawa M, Shimizu S, Kaneko Y, Nagaosa N, and Iwasa Y 2017 *Nat. Phys.* **13** 578
[3] He P, Zhang S S L, Zhu D, Liu Y, Wang Y, Vignale G, and Yang H 2018 *Nat. Phys.* **14** 495
[4] Wakatsuki R, Saito Y, Hoshino S, Itahashi Y M, Ideue T, Iwasa Y, and Nagaosa N 2017 *Sci. Adv.* **3** e1602390
[5] Itahashi Y M, Ideue T, Saito Y, Shimizu S, Nojima T, and Iwasa Y 2020 *Sci. Adv.* **6** eaay9120
[6] Qin F, Shi W, Ideue T, Yoshida M, Zak A, Tenne R, Kikitsu T, Hashizume D, and Iwasa Y 2017 *Nat. Commun.* **8** 14465
[7] Zhang E Z, Xu X, Zou Y C, Ai L, Dong X, Huang C, Leng P, Liu S, Zhang Y, Jia Z, Peng X, Zhao M, Yang Y, Li Z, Guo H, Haigh S J, Shen J, and Xiu F 2020 *Nat. Commun.* **11** 5634
[8] de Martino A, Egger R, and Tsvelik A M 2006 *Phys. Rev. Lett.* **97** 076402
[9] Yasuda K, Tsukazaki A, Yoshimi R, Kawasaki M, and Tokura Y 2016 *Phys. Rev. Lett.* **117** 127202
[10] Legg H F, Rößler M, Münning F, Fan D, Breunig O, Bliesener A, Lippertz G, Uday A, Taskin A A, Klinovaja J, and Ando Y 2022 *Nat. Nanotechnol.* **17** 696
[11] Ma Q, Xu S Y, Shen H, MacNeill D, Fatemi V, Chang T R, Mier V A M, Wu S, Du Z, Hsu C H, Fang S, Gibson Q D, Watanabe K, Taniguchi T, Cava R J, Kaxiras E, Lu H Z, Lin H, Gedik N, and Jarillo-Herrero P 2018 *Nature* **565** 337
[12] Kang K F, Li T X, Shan J, and Mak K F 2019 *Nat. Mater.* **18** 324
[13] Kumar D, Hsu C H, Sharma R, Chang T R, Yu P, Wang J, Liang G, and Yang H 2021 *Nat. Nanotechnol.* **16** 421
[14] Lai S, Liu H, Zhang Z, Zhao J, Feng X, Wang N, Tang C, Liu Y, Yang S A, and Gao W B 2021 *Nat. Nanotechnol.* **16** 869
[15] Du Z Z, Wang C M, Lu H Z, and Xie X C 2018 *Phys. Rev. Lett.* **121** 266601
[16] Zhao W J, Fei Z Y, Song T C, Choi H K, Palomaki T, Sun B S, Malinowski P, McGuire M A, Xu X D, and Cobden D H 2020 *Nat. Mater.* **19** 503
[17] Yasuda K, Morimoto T, Yoshimi R, Mogi M, Tsukazaki A, Kawamura M, Takahashi K S, Nagaosa N, and Tokura Y 2020 *Nat. Nanotechnol.* **15** 831
[18] He P, Hsu C H, Shi S, Cai K, Wang J, Wang Q, Eda G, Pereira V M, and Yang H 2019 *Nat. Commun.* **10** 1290
[19] Zhang C, Yuan X, Zhang J, Leng P, Mou Y, Ni Z, Zhang H, Yang Y, and Xiu F 2021 *Phys. Rev. Appl.* **15** 034084
[20] Murakawa H, Bahramy M S, Tokunaga M, Kohama Y, Bell C, Kaneko Y, Hwang H Y, and Tokura Y 2013 *Science* **342** 1490
[21] Chen G, Zholud A, and Urazhdin S 2020 *Phys. Rev. X* **10** 011064
[22] Wakatsuki R and Nagaosa N 2018 *Phys. Rev. Lett.* **121** 026601
[23] Avci C O, Garello K, Ghosh A, Alvarado S F, and Gambardella P 2015 *Nat. Phys.* **11** 570
[24] Zhang W, Yu R, Feng W, Yao Y, Dai X, and Fang Z 2011 *Phys. Rev. Lett.* **106** 156808
[25] Lee S, In J, Yoo Y, Jo Y, Park Y C, Kim H, Koo H C, Kim B, and Wang K L 2012 *Nano Lett.* **12** 4194
[26] Leng P L, Chen F T, Cao X Y, Wang Y X, Huang C, Sun X D, Yang Y Z, Zhou J C, Xie X Y, Li Z H, Zhang E Z, Yang Y K, and Xiu F X 2020 *Nano Lett.* **20** 7004
[27] Sulaev A, Teo K L, and Wang L 2015 *Sci. Rep.* **5** 8062
[28] Chen S, Leng P L, Konečná A, Modin E, Gutierrez-Amigo M, Vicentini E, Martín-García B, Barra-Burillo M, Niehues I, Maciel E C, Xie X Y, Hueso L E, Artacho E, Aizpurua J, Errea I, Vergniory M G, Xiu F X, and Hillenbrand R 2023 *Nat. Mater.* **22** 860
[29] Huang C, Zhou B T, Zhang H, Yang B, Liu R, Wang H, Wan Y, Huang K, Liao Z, Zhang E, Liu S, Deng Q, Chen Y, Han X, Zou J, Lin X, Han Z, Law K T, and Xiu F 2019 *Nat. Commun.* **10** 2217
[30] Huang C, Narayan A, Zhang E, Xie X, Ai L, Liu S, Yi C, Sanvito S, and Xiu F 2020 *Natl. Sci. Rev.* **7** 1468
[31] Vodolazov D Y and Peeters F M 2005 *Phys. Rev. B* **72** 172508