Nonreciprocal or even-order nonlinear responses in symmetry-broken systems are powerful probes of emergent properties in quantum materials, including superconductors, magnets, and topological materials. Recently, vortex matter has been recognized as a key ingredient of giant nonlinear responses in superconductors with broken inversion symmetry. However, nonlinear effects have been probed as excess voltage only under broken time-reversal symmetry. In this study, we report second harmonic transport under time-reversal symmetry in the noncentrosymmetric trigonal superconductor PbTaSe₂. The magnitude of anomalous nonlinear transport is two orders of magnitude larger than those in the normal state, and the directional dependence of nonlinear signals are fully consistent with crystal symmetry. The enhanced nonlinearity is semiquantitatively explained by the asymmetric Hall effect of vortex-antivortex string pairs in noncentrosymmetric systems. This study enriches the literature on nonlinear phenomena by elucidating quantum transport in noncentrosymmetric superconductors.
Recently, symmetry breaking in solids has become the focus of research in condensed matter physics. It is also a key strategy for developing novel functionalities. To date, many characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.

Among the various emergent transports originating from symmetry breaking, the second-order nonlinear transport, which includes the intrinsic rectification effect and nonlinear Hall effect, is recognized as a sophisticated probe of symmetry breaking and a potential functionality for rectifying a variety of quantum currents. Recently, however, it has been proposed that second-order nonlinear transport can occur even under time-reversal symmetric conditions. An important example is the nonlinear anomalous Hall effect, which is a new type of Hall effect realized under time-reversal symmetric conditions and has been experimentally observed in few-layer WTe$_2$ and bulk TaIrTe$_4$. Band topology/geometry (i.e., trigonal superconductor and has attracted increased interest as a centrosymmetric crystals, have been reported. For example, the characteristic physical properties, which are unique to noncentrosymmetric crystals, have been reported. For example, the nonlinear optical response such as the second harmonic generation and optical parametric effect are known to occur in noncentrosymmetric crystals. Broken inversion symmetry also affects transport properties via asymmetric scattering, spin–orbit interaction, magnetic structure, and accompanying geometrical/topological characteristics.
respectively. Both $|\rho_{01}^{(2\omega)}|/(\rho_{01}^{(\omega)})$ and $\sigma_{xx}^w$ increased monotonically with a decrease in the temperature, exhibiting similar behavior. In Fig. 2e, we analyze the correlation between these quantities by plotting $|\rho_{01}^{(2\omega)}|/(\rho_{01}^{(\omega)})$ versus $(\sigma_{xx}^w)^2$. A linear dependence on $(\sigma_{xx}^w)^2$, particularly in the high $\sigma_{xx}^w$ (low-temperature) region. Unexpectably, a nonlinear anomalous response was visible even above $T = 100$ K. In general, the second-order nonlinear transverse voltage can be well described by equation $\frac{|\rho_{01}^{(2\omega)}|}{(\rho_{01}^{(\omega)})} = \xi (\sigma_{xx}^w)^2 + \eta$ (where $\xi$ and $\eta$ are phenomenological fitting parameters)\(^{21}\), reflecting the two contributions to the nonlinear transverse response: the first term can be the skew-scattering-like origin, which scales as $\frac{|\rho_{01}^{(2\omega)}|}{(\rho_{01}^{(\omega)})} \propto \tau^2 \propto (\sigma_{xx}^w)^2$, and the second term that satisfies $\frac{|\rho_{01}^{(2\omega)}|}{(\rho_{01}^{(\omega)})} \propto \tau^0 \propto (\sigma_{xx}^w)^0$ corresponds to scattering-time-free mechanisms such as the Berry curvature dipole effect and the side-jump mechanism. In sample 1, fitting parameters $\xi$ and $\eta$ are estimated as $\xi = 2.3 \times 10^{-20}$ m$^3$ V$^{-1}$ Ω$^{-2}$ and $\eta = -3.2 \ \mu$V$^{-1}$, respectively. Unlike a previous study on WTe$_2$\(^{23}\), in which both contributions cannot be neglected, the first term is dominant in PbTaSe$_2$. This might be because the Berry curvature dipole will strictly vanish in trigonal crystals with three mirror planes. Note that a small deviation from the relation $\frac{|\rho_{01}^{(2\omega)}|}{(\rho_{01}^{(\omega)})} \propto \tau^2 \propto (\sigma_{xx}^w)^2$ (black linear dashed line in Fig. 2e) was observed in the low $\sigma_{xx}^w$ (high-temperature) region. This might be attributed to the contribution from the $\sigma_{xx}^w$-linear term $\frac{|\rho_{01}^{(2\omega)}|}{(\rho_{01}^{(\omega)})} \propto \sigma_{xx}^w$ originating from both skew and side-jump scatterings\(^{34}\). The same scaling of $|\rho_{01}^{(2\omega)}|/(\rho_{01}^{(\omega)})$ and $(\sigma_{xx}^w)^2$ was observed in other samples, as depicted for sample 3 in Supplementary Fig. 5.

**First and second harmonic resistance in the superconducting state.** Next, we focus on nonlinear transport in the SC state. Figure 3a, b depict the current dependences of $R_{xx}^{(2\omega)}$ (left) and $R_{xx}^{(\omega)}$ (right) in configurations A (sample 1) and B (sample 2), respectively, at $T = 2$ K. With an increase in the current, the superconducting zero-resistance state was broken and a finite resistance state appeared (black dotted curve). Around this transition, a sharp peak of $R_{xx}^{(2\omega)}$ was observed when $I$ was applied parallel to the zigzag (armchair) direction. Note that such anomalies are negligibly small in other directions, in fair agreement with the directional dependence of second-order nonlinear transport in the trigonal systems, as in the case of the normal state (Fig. 2b, c). Figure 3c, d depict the temperature dependences of $R_{xx}^{(2\omega)}$ (left) and $R_{xx}^{(\omega)}$ (right) in configurations A (sample 1, $I = 0.06$ mA) and B (sample 2, $I = 0.3$ mA), respectively. A peak behavior similar to Fig. 3a, b was observed in $R_{xx}^{(2\omega)}$ ($R_{xx}^{(\omega)}$), whereas such a signal was small or absent in the other direction when $I$ was parallel to the zigzag (armchair) direction. Figure 3a–d indicate that both the nonlinear transverse response and the rectification effect were significantly enhanced in the transition region and suppressed in the zero-resistance state. Such nonlinear anomalous transport, which satisfies the directional dependence of trigonal symmetry and is enhanced in the SC fluctuation region, was observed in all the samples we measured (Supplementary Note 5).
Around the superconducting transition, excited vortex–antivortex pairs or vortex loops are known to cause a resistive state in 2D or layered superconductors even under the time-reversal symmetric condition\(^3,^6\). In the present case of layered PbTaSe\(_2\), our simulation revealed that the vortex–antivortex string pair had the lowest energy excitation, as described in Supplementary Note 3. Therefore, the system can be regarded as 2D from the vortex point of view. We propose that this vortex/antivortex dynamics causes the nonlinear transverse response during the superconducting transition, as discussed below, in a manner similar to the vortex rectification effect in trigonal superconductors under an out-of-plane magnetic field\(^15,^18\). Although we note another possible contribution from the amplitude fluctuations above the superconducting transition temperature, we mainly focus on the vortex/antivortex dynamics in this work because it will be dominant below the transition temperature and the following theoretical model can also semiquantitatively explain the results.

In Fig. 3e, we depict a possible mechanism for the observed nonlinear transverse voltage in trigonal superconductors by considering the asymmetric vortex/antivortex Hall effect owing to the trigonal potential. The first clue came from the observation of the excess component in the Hall resistance, which was interpreted as a vortex Hall effect\(^37-44\) (Supplementary Note 2 and Supplementary Figs. 1, 2). The origin of the vortex Hall effect is still being debated. One potential mechanism is the charging of the vortex core due to the difference between the chemical potentials of the normal core and superconducting states. We consider that the vortices and antivortices are excited by a finite temperature or current as string pairs even without magnetic fields (see Supplementary Note 3, Supplementary Fig. 3). When current is applied along the zigzag direction (configuration A), vortices/antivortices are first driven in the armchair direction and then curved in the transverse zigzag direction owing to the vortex/antivortex Hall effect. During this process, vortices/antivortices are rectified, reflecting the trigonal potential; therefore, the vortex Hall effect is asymmetric. This results in the antiparallel motion of vortices and antivortices, which is equivalent to the net flow of vorticity current (purple arrow) in Fig. 3e; the excess voltage appears perpendicular to it, or along the armchair direction, and is observed as the nonlinear transverse voltage. A similar scenario also explains the intrinsic rectification effect (see Supplementary Note 3, Supplementary Fig. 3).

This model is formulated in Supplementary Note 4. In this theoretical description, we consider the vortex/antivortex dynamics, particularly the Hall effect in trigonal potentials (Supplementary Eq. 5). By combining the rectification effect and Hall effect of vortices/antivortices\(^18\), we obtained the expression of \(R_{yx}^{2\omega}\) as \(R_{yx}^{2\omega} = \frac{(\phi_0)^2}{4e^2} n_i^2 \eta^4 \omega^2 \).
Fig. 3 Current and temperature dependences of first/second harmonic signals around superconducting transition and schematic of asymmetric vortex Hall effect as a possible origin for nonlinear transport. 

a. b. Current dependence of $R_{xx}^{(\omega)}$ (left) and $R_{xx}^{(2\omega)}$ (right) at $T = 2 K$ in configuration A (sample 1) and configuration B (sample 2). Red and blue lines indicate longitudinal ($R_{xx}^{(\omega)}$) and transverse ($R_{xx}^{(2\omega)}$) resistance, respectively. 

c. d. Temperature dependences of $R_{xx}^{(\omega)}$ (left) and $R_{xx}^{(2\omega)}$ (right) in configuration A (sample 1) and configuration B (sample 2). The current value is 0.06 mA and 0.3 mA in c, d, respectively. Red and blue lines indicate longitudinal ($R_{xx}^{(\omega)}$) and transverse ($R_{xx}^{(2\omega)}$) resistances, respectively. Error bars indicate the uncertainty of the signals estimated from current dependence of the second harmonic signals at each temperature. e. Schematic of the rectified vortex/antivortex Hall effect in configuration A. Black (green) arrows denote the trajectories of the vortices/antivortices Hall effect when current flows along the zigzag direction. The rectification of vortices/antivortices reflecting the trigonal potential is represented by the thickness differences of arrows. Purple arrow indicates the antiparallel motions of vortices or vorticity current. Nonlinear voltage ($V_f^{(2\omega)}$) appears perpendicular to the vorticity current, in analogy to the inverse spin Hall effect. Regardless of the direction of the current, excess voltage with the same sign appears along the armchair direction, which can be observed as the second harmonic resistance.

$r$ is the Hall angle of vortices, and $\eta_0$ is the friction coefficient. Parameters $\ell_v$, $U$, and $g_2$ are length, energy, and dimensionless function, respectively, which are determined from the detailed shape of the asymmetric pinning potentials by using the Fokker–Planck equation (Supplementary Eq. 9). In this study, we assumed that the dissociation of vortex–antivortex pairs is induced predominantly by the current injected for the observation of the nonlinear transport effect. We employed the realistic phenomenologial parameters: The potential $U$ and the length $\ell_v$ are determined from the experimental current density and magnetic field where the vortices are depinned. $n_v$ is determined from the maximum number of vortices which can be excited in the sample because nonlinear responses are generated by thermal or current-noise fluctuations near the transition point (Supplementary Note 4). Thus, we estimated the value of $R_{xx}^{(\omega)}$ to be approximately 1.4 mΩ near the superconducting transition temperature (Supplementary Fig. 4b), which is in good agreement with the experimental results. Our theoretical model can also explain the temperature dependence of $R_{xx}^{(2\omega)}$ in the superconducting region and the magnitude difference of nonlinear transport between the superconducting and normal states (see Supplementary Note 4). The detailed analysis on the vortex pinning profile and superconducting fluctuation effect will further clarify the nature of nonreciprocal signals.

In the Supplementary Note 6, we also discuss the nonreciprocal transport under a magnetic field10–17 to obtain a comprehensive understanding of the vortex dynamics in this material (Supplementary Fig. 6). The directional dependence of the antisymmetric second-order nonlinear magnetoresistance is rotated by 90° from the case under time-reversal symmetry, which further supports the intrinsic nature of the signals. Significantly, the theoretical estimation of the magnitude of the nonlinear magnetotransport is consistent with the experimental results, as explained in Supplementary Note 4. This result also supports the above scenario, based on the asymmetric vortex Hall effect.

**Discussion**

In Fig. 4a, we compare the nonlinear transverse signals in the normal and SC states. The temperature dependence of $\frac{|E^{(2\omega)}|}{|E^{(\omega)}|}$ in both the normal state (blue; $I = 4.3 mA$) and the SC state (red; $I = 100 \mu A$) are plotted as well as $R^{(\omega)}_{xy}$ at $I = 100 \mu A$ (right). Note that superconductivity is destroyed even below $T_c$ when a large current ($I = 4.3 mA$) is applied. The obtained values of $\frac{|E^{(2\omega)}|}{|E^{(\omega)}|}$ below $T_c$ are smoothly connected to the normal state contribution. $\frac{|E^{(2\omega)}|}{|E^{(\omega)}|}$ in the SC state at $I = 100 \mu A$ indicates a remarkable enhancement by orders of magnitude compared to that in the normal state. A similar gigantic enhancement of second-order nonlinear transport is also observed in the nonreciprocal magnetotransport13–15,17, implying that nonlinear transport is universally enhanced in the SC state, regardless of the time-reversal symmetry being preserved or not.

Finally, we compare the nonlinear transverse signals observed in the present system of PbTaSe$_2$ and with those previously reported for few-layer WTe$_2$, TaIrTe$_4$, and Bi$_2$Se$_3$ surface.
In Fig. 4b, the values of $\frac{|E_y|}{I_{ex}}$ are plotted as a function of $(\sigma_{xx}^2)^2$ for all materials. Similar plots of anomalous transverse signal versus longitudinal conductivity are known to be useful for discussing the mechanisms of the linear anomalous Hall effect in itinerant magnets and anomalous thermal Hall effect in insulators. In the mechanisms of the linear anomalous Hall effect in itinerant magnets, the longitudinal conductivity are known to be useful for discussing the mechanisms of the linear anomalous Hall effect in itinerant magnets and anomalous thermal Hall effect in insulators. In the mechanisms of the linear anomalous Hall effect in itinerant magnets, the longitudinal conductivity are known to be useful for discussing the mechanisms of the linear anomalous Hall effect in itinerant magnets and anomalous thermal Hall effect in insulators.

**Methods**

**Device fabrication.** Bulk PbTaSe$_2$ single crystals were grown using a flux method in an evacuated quartz tube. Stoichiometric amounts of Pb, Ta, and Se were sealed in an evacuated quartz tube, and 50 mol% KCl and 50 mol% PbCl$_2$ were mixed. The quartz tube was heated at 900 °C for 24 h and then cooled to room temperature. After crystal growth, the flux was removed by dissolution in water. The obtained PbTaSe$_2$ single crystals were exfoliated into thin flakes using the Scotch-tape method, and the flakes were transferred onto a Si/SiO$_2$ substrate. The thickness of the exfoliated flakes was measured using atomic force microscopy. A Hall bar configuration was fabricated on the flakes with Au (150 nm)/Ti (9 nm) electrodes. The pattern was fabricated using electron beam lithography, and the electrodes were deposited using an evaporator.

In fabricating the Hall bar configuration on the exfoliated flakes, we judged the crystal orientation from the straight edges of the flakes, which can be assumed to be in the zigzag direction. It is known that straight edges in exfoliated transition-metal dichalcogenides are identical to zigzag directions with high probability. Although PbTaSe$_2$ has intercalated Pb layers in TaSe$_2$, we also adopted this criterion to determine the crystal orientation of PbTaSe$_2$. After the transport measurement of
sample 1, it was double checked by the STEM measurement, as discussed in the main text. From the results of the STEM measurement for sample 1, we conclude that the above method of determining the crystal orientation can also be applied to PbTaSe2. Schematic images of PbTaSe2 and 2H-NbSe2 in the main text and Supplementary Notes are drawn by VESTA.

**Transport measurements.** The first and second harmonic resistances were measured using AC lock-in amplifiers (Stanford Research Systems Model SR830 DSP) with a frequency of 13 Hz in a quantum design physical measurement system.

As discussed in previous studies, the voltage in the noncentrosymmetric system can be given as follows:

$$V = R^{(1)} I_0 \sin \omega t + R^{(2)} I_0 \sin^2 \omega t$$

$$= R^{(1)} I_0 \sin \omega t + \frac{1}{3} R^{(2)} I_0 \left[ 1 + \sin \left( 2\omega t - \pi \right) \right].$$

Therefore, by extracting the first and second harmonic resistances, we obtain

$$R^{(1)} = \frac{V^{(1)}}{I_0} = R^{(1)}$$

and

$$R^{(2)} = \frac{V^{(2)}}{I_0} = \frac{1}{2} R^{(2)} I_0.$$.

Next, we derive the expression for the normalized nonlinear transverse signal

$$\frac{E^{(2)}_{xy}}{E^{(2)}_{yx}},$$

where $E^{(2)}_{xy}$ and $E^{(2)}_{yx}$ are the second-order nonlinear electric fields in the transverse direction and the linear electric field in the longitudinal direction, respectively, when current is applied along the zigzag direction. $E^{(2)}_{xy}$ is written as

$$E^{(2)}_{xy} = \rho^{(2)}_{xy} I_0,$$

where $i_x$ is the current density and $\rho^{(2)}_{xy}$ is the second-order resistivity. By considering $V^{(2)} = W E^{(2)}_{xy}$ and $I_0 = W I_1$, where $V^{(2)}$ is the linear transverse voltage, $W$ is the thickness of the flake, and $I_1$ is the current, it transforms into

$$V^{(2)} = \frac{\rho^{(2)}_{xy}}{\rho^{(2)}_{yx}} I_1 = \frac{E^{(2)}_{xy}}{E^{(2)}_{yx}} I_1.$$.

Therefore, using $\rho^{(2)}_{xy} = \rho^{(1)}_{xy}$, where $\rho^{(1)}_{xy} = \frac{1}{2} \frac{W}{L} \rho^{(1)}_{yx}$ is the linear longitudinal resistivity with channel length $L$, $E^{(2)}_{xy}$ is calculated as

$$E^{(2)}_{xy} = \frac{\rho^{(2)}_{xy}}{\rho^{(2)}_{yx}} I_1$$

$$= \frac{\rho^{(2)}_{xy}}{\rho^{(2)}_{yx}} I_1 = \frac{I_1}{W} \frac{2}{L} \frac{\rho^{(1)}_{yx}}{\rho^{(1)}_{xy}} = \frac{I_1}{W} \frac{2}{L} \frac{\rho^{(1)}_{yx}}{\rho^{(1)}_{xy}} R^{(2)}.$$.

**Selection rules for nonlinear transport under time-reversal symmetric condition in trigonal systems.** Nonlinear current density $f^{(1)}$ in the noncentrosymmetric system is generally written as $f^{(1)} = \beta E$. On the other hand, for the electric field applied along the zigzag direction ($E_x = 0$, $E_y = E$), $f^{(1)}$ leads to

$$f^{(1)} = \begin{pmatrix} -\beta_{11} E_x \\ 0 \\ 0 \end{pmatrix}.$$.

In both cases, $f^{(2)}$ has only the $x$-component. This directional dependence (selection rule) in trigonal systems was confirmed in this study.

**Data availability**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: 18 November 2021; Accepted: 2 March 2022;
Published online: 29 March 2022

References

1. Shen, Y. R. *The Principles of Nonlinear Optics* (Wiley, 1984).

2. Sánchez, J. C. R. et al. Spin-to-charge conversion using Rashba coupling at the interface between non-magnetic materials. Nat. Commun. 4, 2944 (2013).

3. Armitage, N. P., Mele, E. J. & Vishwanath, A. Weyl and Dirac semimetals in three-dimensional solids. Rev. Mod. Phys. 90, 015001 (2018).

4. Rikken, G. L. J. A. & Wyder, P. Electrical magnetochiral anisotropy. Phys. Rev. Lett. 87, 236602 (2001).

5. Olejník, K., Novák, V., Wunderlich, J. & Jungwirth, T. Electrical detection of magnetization reversal without auxiliary magnets. Phys. Rev. B 91, 180402 (2015).

6. Avci, C. O. et al. Unidirectional spin Hall magnetoresistance in ferromagnet/normal metal bilayers. Nat. Phys. 11, 570–575 (2015).

7. Ideue, T. et al. Bulk rectification effect in a polar semiconductor. Nat. Phys. 13, 578–583 (2017).

8. Yasuda, K. et al. Large unidirectional magnetoresistance in a magnetic topological insulator. Phys. Rev. Lett. 117, 127202 (2016).

9. He, P. et al. Nonlinear planar Hall effect. Phys. Rev. Lett. 123, 016801 (2019).

10. Qin, F. et al. Superconductivity in a chiral nanotube. Nat. Commun. 8, 14465 (2017).

11. Watanuki, R. et al. Nonreciprocal charge transport in noncentrosymmetric superconductors. Sci. Adv. 3, e1602390 (2018).

12. Lustikova, I. et al. Vortex rectenna powered by environmental fluctuations. Nat. Commun. 9, 4922 (2018).

13. Yasuda, K. et al. Nonreciprocal charge transport at topological insulator/superconductor interface. Nat. Commun. 10, 2734 (2019).

14. Itahashi, Y. M. et al. Nonreciprocal transport in gate-induced polar superconductor SrTiO3. Sci. Adv. 6, eaay9120 (2020).

15. Itahashi, Y. M., Saito, Y., Ideue, T., Nojima, T. & Iwasa, Y. Quantum and classical ratchet motions of vortices in a two-dimensional trigonal superconductor. Phys. Rev. Res. 2, 023127 (2020).

16. Ando, F. et al. Observation of superconducting diode effect. Nature 584, 373–376 (2020).

17. Ideue, T., Koshikawa, S., Namiki, H., SASAGAWA, T. & IWASA, Y. Giant nonreciprocal magnetotransport in bulk trigonal superconductor PbTaSe2. Phys. Rev. B 101, 024505 (2020).

18. Hoshino, S., Watanuki, R., Hamamoto, K. & Nagaosa, N. Nonreciprocal charge transport in two-dimensional noncentrosymmetric superconductors. Phys. Rev. B 98, 054510 (2018).

19. Tokura, Y. & Nagaosa, N. Nonreciprocal responses from non-centrosymmetric quantum materials. Nat. Commun. 9, 3740 (2018).

20. Ideue, T. & Iwasa, Y. Symmetry breaking and nonlinear electric transport in van der Waals nanostructures. Annu. Rev. Condens. Matter Phys. 12, 201–223 (2021).

21. Sodemann, I. & Fu, L. Quantum nonlinear Hall effect induced by Berry curvature dipole in time-reversal invariant materials. Phys. Rev. Lett. 115, 216806 (2015).

22. Ma, Q. et al. Observation of the nonlinear Hall effect under time-reversal-symmetric conditions. Nature 565, 337–342 (2019).

23. Kang, K., Li, T., Sohn, E., Shan, J. & Mak, K. F. Nonlinear anomalous Hall effect in few-layer WTe2. Nat. Mater. 18, 324–328 (2019).

24. Kumar, D. et al. Room-temperature nonlinear Hall effect and wireless radiofrequency rectification in Weyl semimetal TaIrTe5. Nat. Nanotechnol. 16, 421–425 (2021).

25. He, P. et al. Quantum frequency doubling in the topological insulator Bi2Se3. Nat. Commun. 12, 698 (2021).
26. Ali, M. N., Gibson, Q. D., Klimczuk, T. & Cava, R. J. Noncentrosymmetric superconductor with a bulk three-dimensional Drac cone gapped by strong spin-orbit coupling. Phys. Rev. B 89, 020503 (2014).
27. Namiki, H. & Sagawa, T. Anisotropic superconducting properties of noncentrosymmetric PbTaSe2 as a candidate exotic superconductor. Sci. Adv. Mater. 8, 2097–2102 (2016).
28. Sankar, R. et al. Anisotropic superconducting property studies of single crystal PbSe: J. Phys. Condens. Matter 29, 095601 (2017).
29. Wang, M. X. et al. Nodeless superconducting gaps in noncentrosymmetric superconductor PbTaSe2 with topological bulk nodal lines. Phys. Rev. B 93, 020503 (2016).
30. Bian, G. et al. Topological nodal-line fermions in spin-orbit metal PbTaSe2. Nat. Commun. 7, 10556 (2016).
31. Guan, S. Y. et al. Superconducting topological surface states in the noncentrosymmetric bulk superconductor PbTaSe2. Sci. Adv. 2, e1600894 (2016).
32. Guo, Y. et al. Distinctive in-plane cleavage behaviors of two-dimensional layered materials. ACS Nano 10, 8980–8988 (2016).
33. Isobe, H., Xu, S. Y. & Fu, L. High-frequency rectification via chiral Bloch electrons. Sci. Adv. 6, eaay2497 (2020).
34. Du, Z. Z., Wang, C. M., Li, S., Lu, H. Z. & Xie, X. C. Disorder-induced nonlinear Hall effect with time-reversal symmetry. Nat. Commun. 10, 3047 (2019).
35. Kosterlitz, J. M. & Thouless, D. J. Ordering, metastability and phase transitions in two-dimensional systems. J. Phys. C: Solid State Phys. 6, 1181–1203 (1973).
36. Matsuda, Y. et al. Thickness dependence of the Kosterlitz-Thouless transition in ultrathin YBa2Cu3Oy−δ films. Phys. Rev. B 48, 10498 (1993).
37. Hagen, S. J., Lobb, C. J., Greene, R. L., Forrester, M. G. & Kang, J. H. Anomalous Hall effect in superconductors near their critical temperatures. Phys. Rev. B 41, 11630–11633 (1990).
38. Luo, J., Orlando, T. P., Graybeal, J. M., Wu, X. D. & Muenchausen, R. Scaling of the longitudinal and Hall resistivities from vortex motion in YBa2Cu3O7. Phys. Rev. Lett. 68, 690–693 (1992).
39. Hagen, S. J. et al. Anomalous flux-flow Hall effect: Nd1.625Ce0.375CuO4−δ and evidence for vortex dynamics. Phys. Rev. B 47, 1068–1068 (1993).
40. Khomski, D. I. & Freimuth, A. Charged vortices in high temperature superconductors. Phys. Rev. Lett. 75, 1384–1386 (1995).
41. Kopnin, N. B. & Lopatin, A. V. Flux-flow Hall effect in clean type-II superconductors. Phys. Rev. B 51, 15291–15303 (1995).
42. Nagaoka, T. et al. Hall anomaly in the superconducting state of high-Tc cuprates: universality in doping dependence. Phys. Rev. Lett. 80, 3594–3597 (1998).
43. Kumagai, K. I., Nozaki, K. & Matsuda, Y. Charged vortices in high-temperature superconductors probed by NMR. Phys. Rev. B 63, 144502 (2001).
44. Auerbach, A. & Arovos, D. P. Hall anomaly and moving vortex charge in layered superconductors. SciPost Phys. 8, 061 (2020).
45. Onoda, S., Sugimoto, N. & Nagaosa, N. Intrinsic versus extrinsic anomalous hall effect in ferromagnets. Phys. Rev. Lett. 97, 126602 (2006).
46. Ideue, T., Kurumaji, T., Ishiwata, S. & Tokura, Y. Giant thermal Hall effect in multiferroics. Nat. Mater. 16, 797–802 (2017).
47. Momma, K. & Izumi, F. VESTA3 for three-dimensional visualization of crystal, volumetric and morphology data. J. Appl. Crystallogr. 44, 1272–1276 (2011).
48. Sturman, B. I. & Fridkin, V. M. The Photovoltaic and Photorefractive Effects in Noncentrosymmetric Materials. (Gordon and Breach Science Publishers, 1992).

**Acknowledgements**

We thank T. Nojima, S. Koshikawa, S. Okazaki, H. Isobe, and N. Nagaosa for fruitful discussions. Y.M.I. was supported by the Advanced Leading Graduate Course for Photon Science (ALPS). T.I. was supported by JSPS KAKENHI grant numbers JP19H01819, grant from Yazaki Memorial Foundation for Science and Technology, JST PRESTO (grant no. JPMJPR19QL), T.S. was supported by JST CREST (grant no. JPMJCR16F2). This work was supported by JSPS KAKENHI grant number JP19H05602 and the A3 Foresight Program.

**Author contributions**

Y.M.I., T.I. and Y.I. conceived the research project. H.N., and T.S. synthesized the bulk material. Y.M.I., and C.G. fabricated the microdevices, performed the experiments, and analyzed the data. S.H. performed the theoretical calculations. Y.M.I., T.I., S.H., and Y.I. wrote the manuscript. All authors have led the physical discussions.

**Competing interests**

The authors declare no competing interests.

**Additional information**

**Supplementary information** The online version contains supplementary material available at https://doi.org/10.1038/s41467-022-29314-4.

**Correspondence** and requests for materials should be addressed to Toshiya Ideue.

**Peer review information** Nature Communications thanks the anonymous reviewers for their contribution to the peer review of this work. Peer reviewer reports are available.

**Reprints and permission information** is available at http://www.nature.com/reprints

**Publisher’s note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

© The Author(s) 2022