Observation of the Crystal Hall Effect in a Collinear Antiferromagnet

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An electric current passing through a magnetic conductor can generate a dissipationless transversal current of topological Berry curvature origin. This anomalous Hall effect requires the breaking of spin-degeneracy of electronic bands, conventionally arising from a macroscopic moment in ferromagnets, or a non-collinear magnetic order in complex magnets. Here we report the experimental observation of a new anomalous Hall effect mechanism – the crystal Hall effect – in a system with the abundant collinear antiferromagnetic order. We detect a large crystal Hall conductivity of $\sim$330 S cm$^{-1}$, consistent with our density functional theory calculations, by performing Hall measurements up to 50 T on high quality epilayers of RuO$_2$. We demonstrate that this crystal Hall effect is an experimental manifestation of unconventional spin-splitting originating from a complex crystal structure in combination with collinear antiferromagnetism with zero net moment. This opens a previously unexplored chapter, associated with the new spin-splitting physics, of dissipationless transport and other quantum and topological phenomena in condensed matter.
As early as in 1879, Edwin Hall discovered that the electrical current in a silver wire is drawn towards one side of the wire by a magnetic field (1). Two years later, he further found that such a transverse effect in ferromagnetic metals such as nickel and cobalt is almost ten times larger than that in non-magnetic metals (2). The former effect was named the Hall effect, which, to a great extent, enabled the birth of semiconductor electronics, while the latter larger effect in ferromagnetic materials was coined as the anomalous Hall effect (AHE) that does not originate from the Lorentz force and can be non-zero in zero magnetic field.

Over a century, the AHE has been one of the most fascinating phenomena of transport in solid materials, connected to the difficulty to reach a full understand of it and its increasing importance in revealing the topological nature of the material phases. As the topological and Berry phase physics have been utilized to understand the nature of the AHE in the past two decades (3-5), it has been generally believed that both the intrinsic mechanism related to Berry curvature and the extrinsic spin-dependent scattering mechanisms can attribute to the AHE in ferromagnetic spin-orbit coupled materials (6).

Despite the lack of a full microscopic understanding on the AHE, symmetry arguments can be used to determine if the AHE can be allowed to occur in a particular material, irrespective of its microscopic mechanism (7-9). Based on this perspective, the discovery (9,10) of the AHE in antiferromagnets with a non-collinear spin structure, which lowers their original crystal symmetries and leads to a non-vanishing Hall conductivity, has been established. Such a discovery breaks with our previous intuition that the AHE is an intrinsic feature of ferromagnetism alone. On the other hand, collinear antiferromagnets are commonly believed to be prevented from having the AHE (10-12). This is because, when looking at the magnetic ordering alone, time-reversal symmetry combined with another symmetry operation (inversion or half-unit cell translation) $T_{AF}$ connecting the two antiferromagnetic sublattices prohibits the anomalous Hall conductivity. Examples include the simple two-sublattice collinear antiferromagnet Mn$_2$Au shown in Fig. 1A, which exhibits Kramers spin degenerate bands (13) illustrated schematically in the inset. However, a recent theoretical prediction demonstrates that collinear antiferromagnets crystallizing in magnetic symmetry groups allowing for Hall pseudovector can generate a surprisingly strong Hall signal due to its low symmetric crystal structure rather than the magnetic order alone (9). Commonly the magnetization effects in these
antiferromagnets are microscopically of relativistic perturbative origin and are thus inherently small \((14)\). Here we show that in contrast to this common belief, the corresponding Hall conductivity can be large. Microscopically, the effect can be attributed to its strong topological \((15)\) and symmetry breaking origin due to the nonmagnetic atoms and collinear antiferromagnetism \((9)\). The dependence of the Hall effect on the distribution of nonmagnetic atoms represents a new degree of freedom in the design of materials and together with its distinct microscopic origin motivates labeling this phenomenon as the crystal Hall effect.

The crystal Hall symmetry breaking by the low symmetry local magnetization densities is due to the hybridization of the Ru antiferromagnetic orbitals with the nonmagnetic O orbitals. In fact, the \(T_{AF}\) symmetry connecting the antiferromagnetic sublattices is broken by the O octahedra and generates the low symmetry magnetization isosurfaces, as shown in Fig. 1B. Remarkably, even in the calculations without spin-orbit coupling (SOC), the energy bands manifest strong crystal momentum dependent spin splitting \((9,16-18)\). This is in sharp contrast to conventional collinear antiferromagnets \((13)\) (Mn2Au, CuMnAs) where the energy bands are Kramers spin-degenerate. In addition, the spin is a good quantum number in RuO\(_2\) unlike in noncollinear \((19,20)\) or noncoplanar antiferromagnets \((10)\) (Mn3Pt, Mn3Sn), where the bands are spin-split but spin is not a good quantum number. Simultaneously, the octahedra formed around the two Ru atoms in the unit cell are rotated with respect to each other by 90 degrees and this symmetry is related to the perfect spin compensated antiferromagnetic state illustrated on the density of states in Fig. 1D and inset of Fig. 1B (without SOC).

When we further lower the symmetries by adding the SOC in our density functional theory (DFT) calculations (see Methods), the RuO\(_2\) antiferromagnet can exhibit a Hall vector \(\sigma: \mathbf{j}_{CHE} = \sigma \times \mathbf{E}\)

\(\mathbf{j}_{CHE}\) is crystal Hall current, \(\mathbf{E}\) is applied voltage, and \(\sigma = (\sigma_{yz}, \sigma_{zx}, \sigma_{xy})\) whose presence and orientation is sensitive to the antiferromagnetic Néel vector \(\mathbf{N} = (\mathbf{M}_A - \mathbf{M}_B)/2\) orientation \((9)\).

In Fig. 1E we show our first principle calculation of crystal momentum resolved intrinsic Hall conductivity (specifically Berry curvature in crystal momentum \(k_z = 0\) plane) for antiferromagnetic vector oriented along the [110] direction. We observe hotspots of the Berry curvature of magnitude larger than in ferromagnetic Fe \((21)\), or in noncollinear antiferromagnets \((22)\). By integrating the Berry curvature, we obtain the Hall conductivity which show large
values, 100 – 600 S/cm, depending on the Fermi level position as we will discuss later in the manuscript. This indicates the possibility of unexpectedly large Berry curvature effects and particularly crystal Hall conductivity in RuO$_2$ with moments aligned along the [110] crystal direction (13).

Rutile RuO$_2$ is a highly metallic oxide with a room-temperature resistivity of $\rho \sim 35$ $\mu\Omega\cdot$cm (23). Recent neutron scattering (24) and resonant X-ray scattering (25) studies demonstrated the presence of collinear antiferromagnetism in both its thin films and bulk crystals around room temperature, where the spin axis is slightly canted off the $c$-axis. As it has been previously shown (26), that thin-film growth of rutile RuO$_2$ on commonly used oxide single-crystal MgO and SrTiO$_3$ substrates by pulsed laser deposition yields highly-ordered (110)-oriented and (100)-oriented epitaxial films, respectively, which facilitates the investigation of the Hall effect in RuO$_2$ thin films along different crystallographic orientations.

To experimentally verify this new effect, we fabricated high-quality epitaxial thin films of RuO$_2$. To optimize the electrical conductivity, systematic growth of RuO$_2$ onto MgO single-crystal substrates were performed for a wide range of oxygen partial pressure between $10^2$ and $10^6$ Torr and a wide growth temperature range between 500 and 800 °C. The thickness of the RuO$_2$ films was kept at 27 nm. The optimal growth conditions are 550 °C and $10^3$ Torr oxygen pressure, with which a lowest room-temperature resistivity of $\rho \sim 66$ $\mu\Omega\cdot$cm was achieved (Fig. S1). Although it is higher than the bulk resistivity, such an optimized resistivity for rutile RuO$_2$ thin films is one fifth of the resistivity of commonly utilized perovskite metallic oxide SrRuO$_3$ (27).

In addition, a high carrier density $n$ of $\sim3.5\times10^{23}$ cm$^{-3}$ at 300 K and $\sim7.5\times10^{22}$ cm$^{-3}$ at 10 K were obtained by high-field magnetic measurements. Generally, thin films grown below 700 °C are all epitaxial, have a high hole-type carrier density of $\sim10^{23}$ cm$^{-3}$ and exhibit metallic transport behavior (Fig. S1).

Figure 2A shows an X-ray diffraction spectrum of an optimized 27-nm-thick RuO$_2$/MgO heterostructure and reveals the (110) orientation of epitaxial RuO$_2$, which is consistent with the previous growth (26). Magnetometer measurements indicate negligible moments for such a RuO$_2$ thin film (Fig. 2B). To further explore its magnetic order, a soft magnetic layer of Co$_{90}$Fe$_{10}$ with a thickness of 5 nm was deposited on top of it (Fig. 2C) at room temperature in a magnetic field of $\sim20$ mT, yielding an obvious exchange bias at 50 K (Fig. 2D). This agrees with the reported
antiferromagnetic order of rutile RuO$_2$ films revealed by resonant X-ray scattering studies (25). The interfacial exchange coupling between RuO$_2$ and Co$_{90}$Fe$_{10}$ causes a large enhancement of the coercivity field of Co$_{90}$Fe$_{10}$ (Fig. 2E). The blocking temperature is found to be $\sim$200 K (Fig. 2F), which could be further enhanced towards the Néel temperature of RuO$_2$ by post-annealing with a magnetic field (28).

The Hall measurements conducted up to 50 T that was applied along the out-of-plane direction of thin film samples in a pulsed high magnetic field setup demonstrated a clear anomalous Hall effect at all temperatures ranging from 10 to 300 K. The Hall data are shown in Figs. 3A and B and exhibit an anomalous signal, i.e., the departure from ordinary Hall effect linear in magnetic field. We show below in the theory section, that this signal is consistent with the crystal Hall mechanism. Considering the saturation crystal Hall resistivity $\rho_{\text{CHE}}$ and the transverse resistivity $\rho$ (Fig. 3C) of the RuO$_2$ film at different temperatures, the anomalous Hall conductivity estimated from $\sigma_{\text{CHE}} \approx -\rho_{\text{CHE}}/\rho^2$ for a positive magnetic field is plotted as a function of temperature in Fig. 3D. Due to the highly metallic nature of the RuO$_2$ film, $\sigma_{\text{CHE}}$ is rather large at low temperatures. It reaches $\sim$331 S·cm$^{-1}$, which is over three times that of non-collinear antiferromagnet Mn$_3$Sn (29) and even on the same order with the anomalous Hall conductivity of Fe thin films (30). Above 50 K, $\sigma_{\text{CHE}}$ is greatly lowered by increasing temperature and decreases to $\sim$3.2 S·cm$^{-1}$ at room temperature.

Compared with RuO$_2$ films grown on MgO, RuO$_2$ films deposited onto SrTiO$_3$ single-crystal substrates in the same optimized conditions are highly ordered as well (Fig. S2) but (100)-oriented (Fig. 3E). Intriguingly, the Hall effect along the RuO$_2[100]$ direction is predominantly linear up to 50 T for all the temperatures between 10 and 300 K (Fig. 3F). The fitted carrier density $n$ is $\sim$3.28 $\times$ 10$^{23}$ cm$^{-3}$ at 300 K and decreases to $\sim$1.36 $\times$ 10$^{23}$ cm$^{-3}$ at 10 K through a slighter carrier freeze-out effect compared with the (110)-oriented RuO$_2$/MgO heterostructure.

We show next, by first-principle calculations (see Methods), that the experimental observations can be naturally attributed to the crystal Hall effect. The observed positive sign of the measured anomalous Hall conductivity is consistent with the reported DFT calculated sign (9). In Fig. 4A we present our DFT calculated crystal Hall conductivity with Hubbard $U = 1.6$ eV corresponding to the experimental antiferromagnetic moment magnitude (24,25). For hole doping $\sim$0.5 eV,
consistent with hole concentration observed in experiment (see SI), we obtain $\sigma_{H}^{df} > 300 \text{ S/cm}$. The magnitude as well as sign is consistent with the experimental low temperature value $\sigma_{H}^{exp} \approx 330.8 \text{ S/cm}$. In Fig. 4B we show the corresponding out-of-plane and in-plane magnetic anisotropy energies (MAE). We see that the large values of Hall conductivity correspond to an out-of-plane MAE $K_{z} \approx 0.1 - 5 \text{ meV}$ (for the orientation of the crystal axis see inset in Fig. 4B). These values are consistent with antiferromagnetic vector along the [001] axis and a vanishing Hall signal (9) when there is applied no external magnetic field in the experiment.

Figure 4C demonstrates that for achieving a large crystal Hall conductivity it is sufficient to tilt (not necessary to reorient) the antiferromagnetic moments out of the [001] direction. The value of the spin-flop field, $\sqrt{2K_{z}J}$, for realistic exchange and anisotropy parameters is consistent with the experimental magnetic fields $\lesssim 50 \text{ T}$ required to observe the Hall signal. Typical exchange fields can be estimated from the Néel temperature $J \sim \frac{3}{2}k_{B}T_{N} \sim 300 \text{ K}$ for $T_{N} \sim 300 \text{ K}$. The calculated anisotropy energies shown in Fig. 4D are on the order of $\sim 0.25 \text{ meV} \left(K_{z} \sim 4 \text{ T}\right)$ giving $\sqrt{2K_{z}J} \approx 50 \text{ T}$. Furthermore, we obtain for most of the electron filling a larger Hall conductivity for the antiferromagnetic vector along [110] rather than along the [100] crystal axis. This possibly explains the large anisotropy of the Hall conductivity observed in our experiment, i.e., large value observed for antiferromagnetic vector along [110] and vanishing signal for antiferromagnetic vector along [100]. Also, we observe that besides the tiny region around the charge neutrality point, [110] tends to have the easy axis within the tetragonal plane. The tendency to reorient easier towards [110] rather than [100] direction supports also the experimental observation of the smaller magnitude of the crystal Hall conductivity for [100] rather than [110] layers.

In addition, the calculated dependence of the Hall conductivity on the Néel vector angle saturates at low angles, as shown in Fig. 4C. This explains the saturation of the Hall signal observed in the experiment in Fig. 3A and B. In Fig. 4E and F, we present our calculations of the dependence of the crystal Hall conductivity on the Hubbard correlation parameter $U$ for the antiferromagnetic
vector along the [110] axis. We observe that for small and large Hubbard $U$, the Hall conductivities are negligible. This is due to the vanishing antiferromagnetic moment for small $U$ and opening of an insulating bandgap for large $U$, respectively. The Hubbard parameter $U \approx 1.6 - 2$ eV corresponds to the antiferromagnetic vector magnitude consistent with experiment and previous first principle studies (9,16,24). Decreasing $U$ simulates the quenching of the antiferromagnetic moment close to the Néel temperature, which is consistent with the experimentally observed drop in Hall conductivity with increasing temperature.

We note that the tiny net magnetic moment represents only a very small correction to the crystal Hall conductivity, as shown by an earlier theoretical work (9). We verify that the large experimental magnetic fields have only a small effect on the net magnetization and the electronic structure by calculating them from first principles in a Zeeman field equivalent to a 50 T magnetic field. For the Néel vector along the [110] axis, we obtain a net magnetization of 0.13 $\mu_B$ at 50 T and 0.10 $\mu_B$ at zero field, and 0.07 $\mu_B$ at 50 T and 0.05 $\mu_B$ at zero field for the [100] orientation. We have thus verified that the large experimental magnetic fields have only a small effect on net magnetizations and electronic structure. This indicates that the observed Hall signal originates from the collinear antiferromagnetism (9) and the external magnetic field is required only for the reorientation of the magnetic moments to the directions with a large crystal Hall conductivity.

Finally, we found that (110)-oriented RuO$_2$ films can also be synthesized by using (011)-oriented PMN-PT (0.7PbMg$_{1/3}$Nb$_{2/3}$O$_3$–0.3PbTiO$_3$) single-crystal substrates. However, these films are less ordered as revealed by X-ray diffraction and transmission electron microscopy characterizations (Fig. S3). Although the AHE can be clearly seen at low temperatures (Fig. S4), the anomalous Hall conductivity becomes much smaller than that of highly-ordered RuO$_2$/MgO films at all temperatures (Fig. S5) and decays quickly to zero at ~150 K. This suggests that the crystallinity is a key and therefore an effective tuning factor in determining this new type of anomalous Hall effect.

In summary, by combining high-quality epitaxial growth of RuO$_2$ thin films, pulsed high-field Hall measurements and density functional theory calculations, we demonstrate a new type of AHE in a collinear antiferromagnet. This new type of AHE is induced by the symmetry lowering
by the non-magnetic atoms in combination with collinear antiferromagnetism. The identification of the crystal Hall effect in collinear antiferromagnets removes the requirement of a net moment or a complex magnetic structure for the observation of the AHE. The collinear antiferromagnetism can be, in contrast to complex magnetism, more conveniently controlled and together with the crystal Hall effect sensitivity to the nonmagnetic atomic positions represents new degrees of freedom in (quantum) Hall materials design. From a broader perspective, the realization of the new crystal spin-splitting (9,16-18), with which the crystal Hall effect is associated, represents an alternative to the spin-splitting requiring either relativistic high atomic number elements (17,18) (e.g. Rashba effect), ferromagnetism, or complicated noncollinear magnetism. Our work thus opens new research directions and material design of topological insulators, room temperature quantum anomalous Hall effect, magneto-optical effects, giant spin-splitting, or magneto-electrics in the abundant class of collinear antiferromagnets.

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**Author contributions**

Z.F. & Xiaorong Z. performed sample growth, electrical, structural and magnetic measurements with assistance from L.W., Z.Z., H.G., X.W., H.Y., P.Q., Xin.Z., H.W., H.C., & C.J. Theoretical calculations and analysis were performed by L.S., R.G.H., J.S. & T.J. The manuscript was written by Z.L., Z.F. Xiaorong Z., M.C., L.S., J.S. & T.J. All authors commented on manuscript. This project was conceived and led by Z.L.
**Additional information**

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**Data availability:** The data and simulations codes that support the findings of this study are available from the corresponding author upon request.
Fig. 1. Asymmetric collinear antiferromagnetism and crystal Hall Berry curvature in RuO$_2$ from first principles. (A) The effective time-reversal symmetry $T_{AF}$ connects the highly symmetric magnetization isosurfaces in conventional collinear antiferromagnets as Mn$_2$Au and prohibits any spontaneous Hall signal. The inset shows schematics of Kramers spin degenerate Fermi surface. (B) Collinear antiferromagnets with low symmetric magnetization isosurfaces modulated by the hybridization of antiferromagnetic Ru and nonmagnetic O orbitals in the lattice can break $T_{AF}$. In turn, antiferromagnetic RuO$_2$ with moment along [110] crystal axis exhibits spontaneous Hall vector along [110] and crystal Hall voltage can be measured along [1-10]. In the inset, we show spin-split Fermi surface schematics. (C) Strong momentum dependent spin splitting and (D) perfectly compensated density of states in RuO$_2$ antiferromagnet calculated without spin-orbit coupling. (E) Strong crystal momentum resolved Berry curvature $-\Omega_{AF}$ in RuO$_2$ calculated with spin-orbit coupling and moments along [110] corresponding to intrinsic crystal Hall conductivity vector along [110].
Fig. 2. Antiferromagnetic order in a RuO$_2$ thin film. (A) X-ray diffraction spectrum of an optimized RuO$_2$/MgO heterostructure, indicating a highly ordered (110) orientation of the RuO$_2$ film. (B) Magnetic moment versus magnetic field of the RuO$_2$/MgO heterostructure at 50 and 300 K. (C) Schematic of a RuO$_2$/MO heterostructure capped by a 5-nm-thick Co$_{90}$Fe$_{10}$ (CoFe) layer and a 2-nm-thick Pt top layer. (D) Normalized magnetization of the Pt/CoFe/RuO$_2$/MgO stack as a function of magnetic field at 50 K. (E) Comparison of the coercivity field of CoFe in the Pt/CoFe/RuO$_2$/MgO heterostructure with that in a Pt/CoFe/MgO heterostructure. (F) Exchange bias field of the Pt/CoFe/RuO$_2$/MgO heterostructure versus temperature.
Figure 3

Fig. 3. Hall effect along the out-of-plane direction of RuO₂ thin films. (A)-(B), Hall effect up to 50 T measured for the (110)-oriented RuO₂/MgO heterostructure at 10, 80, 150 and 300 K, respectively. (C) Resistivity versus temperature for the RuO₂/MgO heterostructure. (D) The crystal Hall conductivity (CHC) σ_{CHE} as a function of temperature for the RuO₂/MgO heterostructure. (E) X-ray diffraction spectrum of a RuO₂/SrTiO₃ heterostructure, indicating a highly ordered (100) orientation of the RuO₂ film. (F) Hall effect of the (100)-oriented RuO₂/SrTiO₃ heterostructure up to 50 T at different temperatures ranging from 10 to 300 K.
Figure 4

Fig. 4. Crystal Hall conductivity in RuO₂ calculated from first principles. (A) Crystal Hall conductivity vs energy calculated for the antiferromagnetic vector along [110] and [100] crystal axis. (B) Corresponding out-of-plane (E[N]|110]-E[N]|001]) and in-plane (E[N]|110]-E[N]|100]) magnetic anisotropy energy (MAE) vs Fermi energy. The inset shows the parallel orientation of the applied magnetic field, Hall vector, spin polarization, and antiferromagnetic vector. Calculated dependence of the (C) crystal Hall conductivity components and (D) MAE on the antiferromagnetic vector rotation from [100], to [001] and [110] to [001] crystal axis, respectively. (E) Crystal Hall conductivity and (F) antiferromagnetic vector magnitude dependence on the electronic correlation parameter, i.e. Hubbard U.
Supplementary Materials for
Observation of the Crystal Hall Effect in a Collinear Antiferromagnet

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Materials and Methods

Growth: RuO$_2$ thin films were first grown onto (001)-oriented MgO single-crystal substrates at different growth oxygen pressures ranging from $10^{-2}$ to $10^6$ Torr and different growth temperatures between 500 and 800 °C by pulsed laser deposition with a based pressure of 1.5×10$^3$ Torr. The target-substrate distance was 60 mm. The laser fluence was ~1.6 J/cm$^2$ and the repetition rate was kept at 10 Hz during deposition. The ramp rate was 20 °C/min for heating and 10 °C/min for cooling. The growth of RuO$_2$ thin films onto (001)-oriented SrTiO$_3$ and (011)-oriented 0.7PbMg$_{1/3}$Nb$_{2/3}$O$_3$–0.3PbTiO$_3$ (PMN-PT) single-crystal substrates was performed at 550 °C and 10$^{-3}$ Torr with the same laser fluence of ~1.6 J/cm$^2$ and repetition rate of 10 Hz. Room-temperature growth of the ferromagnetic Co$_{90}$Fe$_{10}$ and the capping Pt thin films was carried out by a d.c. sputtering system with a base pressure of 7.5×10$^{-9}$ Torr. For the Co$_{90}$Fe$_{10}$ deposition, the d.c. sputtering power was 90 W and the Ar pressure was 3 mTorr. The growth rate was ~0.11 Å/s. For Pt sputtering, the power was by 30 W and the Ar pressure was 3 mTorr. The growth rate was determined to be 0.5 Å/s.

X-ray diffraction: X-ray spectra were collected in a XRD-SmartLab diffractometer. The Cu-Ka X-ray wavelength 1.54188 Å.

Transmission electron microscopy: The focused ion beam technique was used to fabricate cross-section samples. Afterwards, the transmission electron microscopy characterization was conducted in a FEI Talos F200X setup under 200 kV.

Electrical measurements: The linear four-probe geometry was utilized for resistivity measurements of RuO$_2$ thin films, which were carried out in a Quantum Design physical property measurement system with a measuring current of 1 mA.

Pulsed high magnetic field Hall measurements: The conventional Hall geometry was established by Cu wires at Wuhan National High Magnetic Field Center, Huazhong University of Science and Technology, Wuhan, China. The magnetic field was applied along the out-of-plane direction of thin film samples. The amplitude of the a.c. current was 3 mA and the frequency was 100 kHz. The Hall voltage collection was performed by a National Instruments PXIe 5105 oscilloscope at a sampling frequency of 4 MHz.

Magnetic measurements: Magnetic moments were measured in a Quantum Design VersaLab vibrating sample magnetometer with a sensitivity of 10$^{-7}$ emu.

Density functional theory calculations: We performed the density functional theory (DFT) calculations employing the projector augmented plane wave method (SI) implemented in VASP code and we used the spherically symmetric Dudarev DFT+U (S2). We set the energy cut-off of the plane-wave basis to 520 eV, the PBE exchange-correlation functional (S3), and the crystal momentum grid 16x16x24. We used DFT relaxed lattice parameters $a = b = 4.5337$, $c = 3.124Å$ and we set the antiferromagnetic moments along the [110] axis. We constructed the maximally localized Wannier functions in the Wannier90 code (S4) and we calculated the intrinsic Hall conductivity by employing the Berry curvature formula (S5,S6). We used the fine-mesh of 321x321x321 Brillouin zone sampling points. The influence of Zeeman field on the electronic structure was studied by calculations in full potential ELK code with the same parameters as in VASP.
Fig. S1. Conductivity optimization and characterization of RuO$_2$ thin films on (001)-oriented MgO. (A) Phase diagram of electrical conductivity of RuO$_2$ thin films fabricated by pulsed laser deposition at different oxygen pressures and deposition temperatures. (B) Corresponding phase diagram of carrier density of RuO$_2$ thin films. (C) Cross-section transmission electron microscopy image of an optimized RuO$_2$/MgO heterostructure fabricated at 550 °C and $10^{-3}$ Torr oxygen partial pressure. (D) Temperature-dependent sheet resistance contour mapping of the RuO$_2$ films fabricated in the growth temperature range between 500 and 650 °C.
Fig. S2. Cross-section transmission electron microscopy image of a RuO$_2$/SrTiO$_3$ heterostructure fabricated at 550 °C and 10$^{-5}$ Torr oxygen partial pressure.
Fig. S3. RuO$_2$ film grown on a (011)-oriented PMN-PT (0.7PbMg$_{1/3}$Nb$_{2/3}$O$_3$–0.3PbTiO$_3$) single-crystal substrate deposited at 550 °C and 10$^{-3}$ Torr oxygen partial pressure. (A), X-ray diffraction spectrum of the RuO$_2$/PMN-PT heterostructure, indicating the (110) orientation of the RuO$_2$ film. (B), Cross-section transmission electron microscopy image of the RuO$_2$/PMN-PT heterostructure. (C), Temperature-dependent resistivity of the RuO$_2$/PMN-PT heterostructure.
Fig. S4. Hall effect up to 50 T along the out-of-plane direction of the (110)-oriented RuO$_2$/PMN-PT heterostructure at different temperatures.
Fig. S5. Comparison of temperature-dependent crystal Hall conductivity along the out-of-plane direction of (110)-oriented RuO$_2$ films grown on MgO and PMN-PT with different degree of structural ordering.
Table S1. Physical parameters on the RuO$_2$/MgO heterostructure.

| Temperature (K) | $\rho_{xx}$ ($\mu$Ω·cm) | Anomalous Hall Resistivity $\rho_{xy}$ ($\mu$Ω·cm) | $\sigma_{\text{AHE}}$ (S·cm$^{-1}$) | Ordinary Hall Coefficient (cm$^3$/C) | Carrier Density (p-type) (holes/cm$^3$) |
|----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 10 K           | 28.61           | 0.218           | 330.83          | $8.32\times10^{-5}$ | $0.75\times10^{23}$ |
| 80 K           | 31.14           | 0.081           | 103.76          | $3.25\times10^{-5}$ | $1.92\times10^{23}$ |
| 150 K          | 40.26           | 0.054           | 41.23           | $2.66\times10^{-5}$ | $2.35\times10^{23}$ |
| 300 K          | 71.00           | 0.013           | 3.20            | $1.77\times10^{-5}$ | $3.53\times10^{23}$ |
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