Emergence of Weyl fermions in an epitaxial ferromagnetic oxide

Kosuke Takiguchi,1,2† Yuki K. Wakabayashi,1,4† Hiroshi Irie,1 Yoshiharu Krockenberger,1 Takuma Otsuka,3 Hiroshi Sawada,3 Masaaki Tanaka,2 Yoshitaka Taniyasu,1 and Hideki Yamamoto†

1NTT Basic Research Laboratories, NTT Corporation, Atsugi, Kanagawa 243-0198, Japan.
2Department of Electrical Engineering and Information Systems & Center for Spintronics Research Network (CSRN), The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan.
3NTT Communication Science Laboratories, NTT Corporation, Soraku-gun, Kyoto 619-0237, Japan.
*email: yuuki.wakabayashi.we@hco.ntt.co.jp
†These authors contributed equally to this work.

Magnetic Weyl fermions, which occur in magnets, have novel transport phenomena related to pairs of Weyl nodes1,2,3,4,5, and they are, of both, scientific and technological interest, with the potential for use in high-performance electronics, spintronics and quantum computing. Although magnetic Weyl fermions have been predicted to exist in various oxides6,7,8, evidence for their existence in oxide materials remains elusive9,10,11. SrRuO3, a 4d ferromagnetic metal often used as an epitaxial conducting layer in oxide heterostructures12,13,14,15, provides a promising opportunity to seek for the existence of magnetic Weyl fermions. State-of-the-art oxide thin film growth technologies, advanced by machine learning techniques, may allow access to such topological matter. Here we show direct quantum transport evidence of magnetic Weyl fermions in an epitaxial ferromagnetic oxide SrRuO3: unsaturated linear positive magnetoresistance (MR)16, chiral-anomaly-induced negative MR16,17, π Berry phase accumulated along cyclotron orbits16, light cyclotron masses18 and high quantum mobility of about 10000 cm2/Vs18,19. We employed machine-learning-assisted molecular beam epitaxy (MBE)20 to synthesize SrRuO3 films whose quality is sufficiently high to probe their intrinsic quantum transport properties. We also clarified the disorder dependence of the transport of the magnetic Weyl fermions, and provided a brand-new diagram for the Weyl transport, which gives a clear guideline for accessing the topologically nontrivial transport phenomena. Our results establish SrRuO3 as a magnetic Weyl semimetal and topological oxide electronics as a new research field.

Weyl semimetals, which host Weyl fermions described by the Weyl Hamiltonian, have intriguing and fascinating transport phenomena based on the chiral anomaly and linear band dispersion with spin-momentum locking1,2,3,4,21, such as chiral-anomaly-induced negative MR and high mobility16,17,18. Compared with space-inversion-symmetry-breaking Weyl semimetals22, time-reversal-symmetry (TRS)-breaking ones are thought to be more suitable for spintronic applications3,23,24. For example, since the distribution of Weyl nodes in magnets is determined by the spin texture1, this distribution is expected to be controlled by the magnetization switching technique25,26. Recent angle-resolved photoemission spectroscopy (ARPES) studies have found experimental evidence for the electronic structure of magnetic Weyl semimetals Co3Sn2S2,3,3 and
Co$_2$MnGa$^4$, such as the presence of bulk Weyl points with linear dispersions and surface Fermi arcs. Demonstrating the relevance of magnetic Weyl fermions to spintronic and electronic applications requires information on quantum oscillations, which allows us to characterize transport properties of individual orbits in a magnetic Weyl semimetal. However, systematic and comprehensive measurements of quantum transport, including the quantum oscillations, have been hampered in magnetic Weyl semimetals because of the difficulty in achieving a quantum lifetime long enough to observe the quantum oscillations in metallic systems. Since specimens in the form of epitaxial films are advantageous for future device applications of magnetic Weyl semimetals, it is urgently required to prepare single-crystalline thin films$^{27}$ of magnetic Weyl semimetals whose quality is sufficiently high to probe quantum transport properties.

Theoretically, the presence of magnetic Weyl fermions has been predicted for SrRuO$_3$, a 4$d$ ferromagnetic material$^7$. SrRuO$_3$ is widely used as an epitaxial conducting layer in oxide electronics and spintronics owing to the unique nature of ferromagnetic metal, compatibility with other perovskite-structured oxides, and chemical stability$^{12,13,14,15,26}$. Theoretical studies predicted that the electronic structure of SrRuO$_3$ includes a large number of Weyl nodes caused by the TRS breaking and spin-orbit coupling (SOC) (Fig. 1a)$^7$, and suggested that the Berry phase from the Weyl nodes gives rise to an anomalous Hall effect (AHE) in it.$^7,10,28$ However, a definitive conclusion of the presence of Weyl fermions near the Fermi energy ($E_F$) cannot be drawn from observations of the AHE alone$^{7,9,10}$, because the AHE reported so far for SrRuO$_3$ can be well reproduced using a function composed of both intrinsic (Karplus-Luttinger (KL) mechanism) and extrinsic (side-jump scattering) terms$^{29,30}$.

In this study, we conducted comprehensive high field magnetotransport measurements (see Methods section ‘Magnetotransport measurements’) including quantum oscillations in the resistivity, i.e., Shubnikov-de Haas (SdH) oscillations, on an extraordinarily high-quality SrRuO$_3$ film (63-nm thick) epitaxially grown on SrTiO$_3$ (Fig. 1b, c and Extended Data Fig. 1). To probe the contribution of the Weyl fermions to the transport properties, it is necessary to identify the following five signatures from the magnetotransport data: (1) unsaturated linear positive MR$^{16,18,31,32,33}$, (2) chiral-anomaly-induced negative MR$^{1,16,17}$, (3) $\pi$ Berry phase accumulated along the cyclotron orbits$^{16,31,32,33}$, (4) light cyclotron mass$^{16,18,31,32,33}$ and (5) high quantum mobility$^{16,18,31,32,33}$. We have clearly observed these five signatures of magnetic Weyl fermions in SrRuO$_3$.

The RRR, which is defined as the ratio of the longitudinal resistivity $\rho_{xx}$ at 300 K [$\rho_{xx}(300 \text{ K})$] and $T\to0$ K [$\rho_{xx}(T\to0 \text{ K})$] ($T$: temperature), is a good measure to gauge the purity of a metallic system, that is, the quality of single-crystalline SrRuO$_3$ thin films (See Methods section ‘Determination of the RRR in SrRuO$_3$’). In fact, high RRR values are indispensable for exploring intrinsic electronic states. More specifically, RRR values above 40 and 60 have enabled observations of sharp, dispersive quasiparticle peaks near the Fermi level by ARPES$^{34}$ and quantum oscillations via the electrical resistivity$^{35}$, respectively. To form SrRuO$_3$ with quality exceeding current levels, we employed machine-learning-assisted MBE (see Methods section ‘Machine-learning-assisted MBE’), which we developed recently$^{20}$.

The resistivity $\rho_{xx}$ vs $T$ curve of SrRuO$_3$ thin film shows a clear kink at the Curie temperature ($T_c$) of $\sim152$ K (Fig. 1d)$^{12}$, while the magnetization measurement at $T = 10$ K shows a typical ferromagnetic hysteresis loop (Fig. 1d, right inset). With a residual
resistivity \(\rho_{xx}(T\rightarrow 0 \text{ K})\) of 2.23 \(\mu\Omega\cdot\text{cm}\) and a RRR of 84.3, SrRuO\(_3\) thin films grown by machine-learning-assisted MBE are superior to those prepared by any other method\(^{12,20,35,36}\). Below approximately 20 K, the \(T^2\) scattering rate (\(\rho_{xx} \propto T^2\)) expected in a Fermi liquid is observed (Fig. 1d, left inset)\(^{12,34}\), indicating that the intrinsic transport phenomenon is seen below this temperature (hereafter called \(T_F\)).

In this Fermi liquid region \([T < T_F (20 \text{ K})]\), semimetallic behavior is seen in the MR \((\rho_{xx}(B) - \rho_{xx}(0 \text{ T})/\rho_{xx}(0 \text{ T})\) (Fig. 1e) and Hall resistivity \(\rho_{xy}(B)\) (Fig. 1f) curves with the magnetic field \(B\) applied in the out-of-plane \([001]\) direction of the SrTiO\(_3\) substrate. As shown in Fig. 1e, \(\rho_{xx}\) above \(T_F\) shows negative MR due to the suppression of magnetic scattering\(^{12,37,38}\), and the MR changes its sign below \(T_F\). Importantly, the positive MR at 2 K shows no signature of saturation even up to 14 T, which is typical of a semimetal\(^{16,39}\) and also commonly seen in Weyl semimetals\(^{1,16,18,31,32,33}\). Especially in the case of Weyl semimetals, linear energy dispersion of Weyl nodes is considered to be one of the most plausible origins of unsaturated linear positive MR\(^{40,41}\). In addition, as shown in Fig. 1f, the \(\rho_{xx}(B)\) curves below \(T_F\) are nonlinear, indicating the coexistence of multiple types of carriers (electrons and holes). We note that, below \(T_F\), the AHE, which stems from the extrinsic side-jump scattering and the intrinsic KL mechanisms in SrRuO\(_3\),\(^{29,30}\) is well suppressed due to the small residual resistivity of the SrRuO\(_3\) film with the RRR of 84.3, and thus \(\rho_{xy}(B)\) curves below \(T_F\) are dominated by the ordinary Hall effect (see Methods section ‘Temperature dependence of the AHE in the SrRuO\(_3\) film with the RRR = 84.3’). Below 10 K, where the AHE is negligible, both the \(\rho_{xy}(B)\) values and the slopes of \(\rho_{xy}(B)\) change their signs in the high-\(B\) region, signaling the possibility of the coexistence of high-mobility electrons with low-mobility holes\(^{16}\).

Next, we observed the chiral-anomaly-induced negative MR, which is an important signature of Weyl fermions\(^{1,16,17,42}\). To clarify the anisotropic character of the chiral-anomaly-induced negative MR thoroughly and systematically, we measured \(\rho_{xx}(B)\) at \(B\) angles \(\alpha, \beta, \gamma\) in the \(x-y, y-z, z-x\) planes, respectively (Fig. 2a–c). The rotation angles \(\alpha, \beta, \gamma\) are defined in the insets of Fig. 2a–c. When \(B\) is applied perpendicular to the current \(I (B \perp I, \alpha = 90^\circ \text{ or } \beta = 0-90^\circ \text{ or } \gamma = 90^\circ)\), the unsaturated linear positive MR is observed. The unsaturated linear positive MR seen here is expected owing to the presence of the Weyl fermions\(^{40,41}\), and those states are supposedly anisotropic because of the \(\sim 0.5\%\) compressive strain of SrRuO\(_3\) induced by the SrTiO\(_3\) substrates\(^{39}\). This anisotropy is confirmed by varying \(\beta\) (Fig. 2b). In contrast, when \(B\) is rotated parallel to the current \((B//I, \alpha = 0^\circ \text{ or } \gamma = 0^\circ)\), the MR turns negative and becomes linear above 8 T (Fig. 2a, c and Extended Data Fig. 5). Theoretical calculations based on the semiclassical Boltzmann kinetic equation predict that TRS-breaking Weyl semimetals show a negative MR that is linear in \(B^1\), in comparison with the quadratic dependence expected for space-inversion-symmetry-breaking Weyl semimetals\(^{16,17}\). Thus, the observed linear increase of the negative MR is consistent with a chiral anomaly in magnetic Weyl semimetals. The chiral-anomaly-induced negative MR can be understood from the violation of the conservation rules of chiral charges as shown in Fig. 2d\(^{42}\), and it should be maximum when \(B\) is exactly parallel to \(I (\alpha = 0^\circ, 180^\circ \text{ or } \gamma = 0^\circ, 180^\circ)\). As expected, this anisotropic feature of the negative MR is observed at \(\alpha = 0^\circ, 180^\circ \text{ or } \gamma = 0^\circ, 180^\circ\) under 14 T (Fig. 2e). In addition, the peak structures in the angle dependence of the negative MR at \(B//I (\alpha = 0^\circ, 180^\circ \text{ or } \gamma = 0^\circ, 180^\circ)\) are similar to those in previous observations of the chiral anomaly in other Weyl semimetals\(^{1,16,17,43,44}\). These results confirm that this linear negative MR is induced by the chiral anomaly. As a consequence
of the composition of the positive MR and negative MR, $\rho_{xx}(B)$ is lower than the zero field resistivity $\rho_0 = \rho_{xx}(0 \, \text{T})$ when $\alpha$ and $\gamma$ are near 0 or 180°.

The Berry phase has become an important concept in condensed matter physics over the past three decades, since it represents a topological classification of the system and it plays a fundamental role in various phenomena such as electric polarization, orbital magnetism, etc\textsuperscript{45}. However, revealing the $\pi$ Berry phase, originating from a band touching point of the Weyl node\textsuperscript{16,31,32,33}, has been challenging in magnetic Weyl semimetals. Here, we detect the $\pi$ Berry phase accumulation along the cyclotron orbits, for the first time in magnetic Weyl semimetals, by measurement techniques sensitive to the quantized energy levels, \textit{e.g.}, SdH oscillation. The nontrivial $\pi$ Berry phase, which is acquired by a surface integral of the Berry curvature $\Omega$ over a closed surface containing a Weyl point in $k$-space (Fig. 3a)\textsuperscript{16,31,32,33}, causes a phase shift of quantum oscillations. According to the Lifshitz-Kosevich (LK) theory, the magnitude of the SdH oscillation is described as\textsuperscript{16,31,32,33,46}

$$\Delta \sigma_{xx} = \sum_i A_i \frac{X_i}{\sinh X_i} \exp \left( -\frac{2\pi^2 k_B T_{Di}}{\hbar \omega_{ci}} \right) \cos \left[ 2\pi \left( \frac{F_i}{B} - \frac{1}{2} + \beta_{Bi} \right) \right]$$

(1)

where $\Delta \sigma_{xx}$ is the oscillation component of the longitudinal conductivity, $A_i$ are the normalization factors, $X_i = 2\pi^2 k_B T_i / \hbar \omega_{ci}$, $k_B$ is the Boltzmann constant, $\hbar$ is the reduced Planck constant, $\omega_{ci}$ is the cyclotron frequency defined as $eB/m_i^* \omega_{ci}$, $m_i^*$ is the cyclotron mass, $T_{Di}$ is the Dingle temperature, $F_i$ is the frequency of the SdH oscillation, and $2\pi \beta_{Bi}$ is the phase shift caused by the Berry phase as mentioned above. The subscript $i$ indicates the label of an orbit of carriers. To extract the Berry phases in SrRuO$_3$, we used the LK formula [eq. (1)] for two frequencies to fit to the SdH oscillation data. Figure 3b shows the SdH oscillation data at 2 K and the fitting results by eq. (1). (see Methods section ‘Data pretreatment for quantum oscillations’). The oscillation spectrum is considerably complex because of the contribution from several subbands.\textsuperscript{7} To reduce the fitting parameters, we first carried out Fourier transform of the SdH oscillation (Fig. 3c), and extracted the $F_1$ (26 T), $F_2$ (44 T), $m_1^*$ (0.30$m_0$), and $m_2^*$ (0.49$m_0$) ($m_0$; electron rest mass) values from the peak positions and the temperature dependence of $F_1$ and $F_2$ based on the LK theory (Fig. 3c, insets). Small masses are expected for Weyl fermions, which would fulfill the light cyclotron mass signature\textsuperscript{16,18,31,32,33}. From the fitting to the data in 0.07 T $^{-1} < 1/B < 0.2$ T $^{-1}$ shown in Fig. 3b, we obtained $T_{D1} = 0.75$ K, $T_{D2} = 0.40$ K, $\beta_{B1} = 0.27$, and $\beta_{B2} = 0.48$. The $2\pi \beta_{B2}$ value of 0.96$\pi$ states the presence of the nontrivial $\pi$ Berry phase arising from the mass-less dispersion of the Weyl fermions. The $2\pi \beta_{B1}$ of 0.54$\pi$ implies that the energy dispersion of the $F_1$ orbit has both quadratic (trivial) and linear (non-trivial) features\textsuperscript{47,48}. These results cannot be reproduced by fixed zero Berry phases, confirming the existence of the non-zero Berry phase (Extended Data Fig. 7).

The SdH oscillations not only give an insight into the topological nature, but also provide evidence of very high mobility of the Weyl fermions enclosing the Weyl points. We quantitatively determine the mobility of the charge carriers of the $F_1$ and $F_2$ orbits by calculating the quantum mobility, $\mu_q = eB/(2\pi k_B m_i^* T_{Di})$. The obtained $\mu_q$ and $\mu_{q2}$ values are $9.6 \times 10^3$ and $1.1 \times 10^4$ cm$^2$/Vs, respectively. In addition, assuming isotropic Weyl nodes, we can estimate the carrier concentrations $m_i = \frac{1}{6\pi^2} \left( \frac{2eF_i}{\hbar} \right)^{3/2}$ ($n_1 = 3.8 \times 10^{17}$ cm$^{-3}$ and $n_2 = 8.3 \times 10^{17}$ cm$^{-3}$) and the chemical potentials $\mu_i = eF_i/m_i^*$ ($\mu_1 = 10.1$ meV and $\mu_2 = 10.5$ meV).\textsuperscript{49} These results mean that $F_1$ and $F_2$ come from the high-mobility and low-concentration Weyl fermions enclosing the Weyl points.
In addition to the Weyl fermions as evidenced by the five signatures described above, there are trivial Ru 4d bands crossing the $E_F$ in SrRuO$_3$. SdH experiments performed at 0.1 K (Fig. 3d) confirmed some of these trivial Fermi pockets with $F_3 = 300$ T, $F_4 = 500$ T, $F_3 = 3500$ T, and $F_6 = 3850$ T (see Methods section ‘SdH oscillations of trivial orbits’). These four orbits have heavier masses (> 2.8m$_0$) than those of $F_1$ and $F_2$, which is consistent with the reported values for SrRuO$_3$. The Fermi pocket areas of $F_3$ (0.029 Å$^2$) and $F_4$ (0.048 Å$^2$) are close to those of the 364 T (0.035 Å$^2$) orbit reported in earlier de Haas–van Alphen measurements, and the Fermi pocket areas of $F_5$ (0.334 Å$^2$) and $F_6$ (0.368 Å$^2$) correspond to the $\alpha_1$ band (0.33-0.37 Å$^2$) observed by the ARPES and in earlier SdH measurements. Noteworthy is that the Fermi pocket areas of $F_1$ (0.0025 Å$^2$) and $F_2$ (0.0042 Å$^2$) are more than ten times smaller than those of the trivial orbits, indicating that $F_1$ and $F_2$ stem from the small Fermi pockets as a feature of the Weyl fermions.

Observing the intrinsic transport signatures of Weyl fermions requires a high-quality SrRuO$_3$ sample, since it is easily hindered by disorders such as defects and impurities. To show the disorder dependence of the transport phenomena in SrRuO$_3$, we investigated the RRR dependence of $T_C$, $T_F$, and the highest temperature where the linear positive MR, one of the clear features of the transport of the Weyl fermions in SrRuO$_3$, remains (hereafter called $T_w$) (Fig. 4a–c) (see Methods section ‘RRR dependence of the ferromagnetism, Fermi liquid behavior, and Weyl behavior’). As shown in Fig. 4c, ferromagnetism becomes weaker ($T_C < 150$ K) below RRR = 8.93, the Fermi liquid behavior remains in all the films even at low RRR = 6.61, and the positive MR is observed over RRR = 19.4. It is remarkable that the positive MR ratio at 9 T increases with increasing RRR (Fig. 4a, b), indicating that the Weyl fermions become more dominant in the transport. Thus, a high-quality SrRuO$_3$ sample is essential for observing the intrinsic transport of the Weyl fermions, and the diagram presented in Fig. 4c will be an effective guideline for realizing topologically non-trivial transport phenomena of the magnetic Weyl fermions to connect magnetic Weyl semimetals to spintronic devices.

In conclusion, we have observed the emergence of magnetic Weyl fermions in epitaxial SrRuO$_3$ film with the best crystal quality ever reported, which was grown by machine-learning-assisted MBE. The observation of the five important transport signatures of magnetic Weyl fermions—the linear positive MR, chiral-anomaly-induced negative MR, $\pi$ phase shift in a quantum oscillation, light cyclotron mass, and high quantum mobility of about 10000 cm$^2$/Vs—establishes SrRuO$_3$ as a magnetic Weyl semimetal. In addition, the RRR dependence of the ferromagnetism, the Fermi liquid behavior, and the positive MR serves as a road map to merge two emerging fields: topology in condensed matter and oxide electronics. Our results will pave the way for topological oxide electronics.

**Author contributions**

Y.K.W. conceived, designed, and coordinated the study. Y.K.W. and Y.K. designed and built the MBE system. Y.K.W., T.O., and H.S. implemented the Bayesian optimization algorithm for the sample growth. Y.K.W. grew the samples. Y.K.W. and K.T. carried out the sample characterizations. K.T., H.I., and Y.K.W. fabricated the Hall bar structures and carried out transport measurements. K.T. and Y.K.W. analyzed the transport data. K.T. and Y.K.W. wrote the paper. H.I., Y.K., T.O., H.S., M.T., Y.T., and H.Y. contributed to the manuscript and the interpretations of the data.
Data availability
The data that support the plots in this paper and other findings of this study are available from the corresponding author upon request.

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Author information Correspondence and requests for materials should be addressed to Y.K.W. (yuuki.wakabayashi.we@hco.ntt.co.jp).

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Fig. 1 Sample characteristics and temperature dependence of the magnetoresistance and Hall resistivity of SrRuO$_3$. a, Schematic image of a pair of Weyl nodes with opposite chiralities (L and R). In FM SrRuO$_3$, the TRS breaking lifts the spin degeneracy and leads to linear band crossing at many $k$ points. Then, the SOC opens small gaps with band anti-crossings, except at a pair of points corresponding to the Weyl nodes with opposite chiralities. b, Schematic of sample and crystal structures of the SrRuO$_3$ films (63-nm thick) on a SrTiO$_3$ substrate. In the schematic crystal image, yellow, blue, red, and purple spheres indicate strontium, oxygen, ruthenium, and titanium, respectively. c, Cross-sectional high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image of the SrRuO$_3$ film with the RRR of 71 taken along the [100] axis of the SrTiO$_3$ substrate. The inset in c shows a color overlay of the electron energy loss spectroscopy (EELS)-STEM images for the Ti-L$_{2,3}$- (green) and Ru-M$_{4,5}$-edge (red). Epitaxial growth of the high-quality single-crystalline SrRuO$_3$ film with an abrupt substrate/film interface is seen in the images. d, $\rho_{xx}$-$T$ curves for the SrRuO$_3$ film with the RRR of 84.3. The left inset in d shows the $\rho_{xx}$ versus $T^2$ plot with the linear fitting (black dashed line). We defined the Fermi liquid region as the temperature range where the experimental $\rho_{xx}$ and the fitting line are close enough to each other (< 0.1 $\mu\Omega$ cm). The right inset in d shows the magnetization $M$ versus $B$ curve at 10 K with $B$ applied to the out-of-plane [001] direction of the SrTiO$_3$ substrate. e, f, MR ($\rho_{xx}(B) - \rho_{xx}(0 \text{ T})$)/$\rho_{xx}(0 \text{ T})$ and Hall resistivity $\rho_{xy}(B)$ curves at 2 to 100 K for the SrRuO$_3$ film with the RRR of 84.3 with $B$ applied to the out-of-plane [001] direction of the SrTiO$_3$ substrate. In e and f, the MR and the Hall resistivity at each temperature have been offset by 7% and 0.22 $\mu\Omega \cdot$cm, respectively, for easy viewing.
Fig. 2 Chiral anomaly in the transport of SrRuO$_3$. a-c, MR $\rho_{xx}(B)$ at various $B$ angles $\alpha$ (a), $\beta$ (b), and $\gamma$ (c) at 2 K for the SrRuO$_3$ film with the RRR of 84.3. The angles $\alpha$, $\beta$, and $\gamma$ are defined in the insets of a-c. d, Schematic image of the chiral-anomaly-induced negative MR. Red and blue bands represent Landau levels of a pair of Weyl nodes with opposite chiralities L and R, respectively. Non-orthogonal electric and magnetic fields ($E \cdot B \neq 0$) lead to the chiral charge transfer between the two Weyl nodes with opposite chiralities (L and R). The currents of the chiral charges are observed as a form of negative MR. Here, $\mu_L$ and $\mu_R$ indicate chemical potentials in the Weyl points with opposite chiralities. e, $\alpha$, $\beta$, and $\gamma$-angular dependences of the MR $\rho_{xx}$ with $B = 14$ T at 2 K for the SrRuO$_3$ film with the RRR of 84.3. The black dashed line indicates the $\rho_{xx}(0 \text{ T})$ value. The region below the black dashed line shows negative MR.
Fig. 3 Quantum oscillations of SrRuO$_3$. a, Schematic image of energy dispersion near a Weyl point. Blue and red arrows represent the Berry curvature $\Omega$ and the Berry connection $A$, respectively. The $\pi$ Berry phase is accumulated along the cyclotron orbits (blue circle). b, SdH oscillation measured at 2K with $B$ (5 T $< B < 14$ T) applied to the out-of-plane [001] direction of the SrTiO$_3$ substrate for the SrRuO$_3$ film with the RRR of 84.3. Black curve is the fitting curve of eq. (1). The fitting was carried out by a non-linear least squares method with the fitting parameters $A_1$, $\beta_1$, $T_{D1}$, $A_2$, $\beta_2$, and $T_{D2}$. The inset shows fan diagrams of two oscillation components of $F_1 (= 26$ T) and $F_2 (= 44$ T), which are shown as orange and green symbols, respectively. Here, the circles and triangles indicate integer and half-integer indexes of the oscillation components. c, Fourier transform spectra of the SdH oscillations at 2 to 8 K. Insets in c show the mass estimations of the $F_1$ and $F_2$ orbits according to the LK theory. Black dashed curves are the fitting curves. d, SdH oscillation observed at 0.1 K for the SrRuO$_3$ film with the RRR of 84.3. The inset in d shows a close-up at around 0.075 T$^{-1}$. The oscillation holds four kinds of other trivial orbits with higher frequencies.
Fig. 4 RRR dependence of the transport phenomena in SrRuO$_3$. a, RRR dependence of the MR ($\rho_{xx}(B) - \rho_{xx}(0 \text{T})$)/$\rho_{xx}(0 \text{T})$ measured at 2 or 2.3 K. Above RRR = 19.4, the positive MR, which is one piece of experimental evidence of Weyl fermions in SrRuO$_3$, clearly emerges. b, RRR dependence of the MR ($\rho_{xx}(B) - \rho_{xx}(0 \text{T})$)/$\rho_{xx}(0 \text{T})$ at $B = 9 \text{T}$ and $T = 2$ or 2.3 K. c, Diagram of the RRR dependence of the $T_C$, $T_F$, and $T_W$, which are shown as green stars, blue circles, and red triangles, respectively.
METHODS
Magnetotransport measurements.
We first deposited the Ag electrodes on a SrRuO$_3$ surface. Then, we patterned the samples into 200 × 350 μm$^2$ Hall bar structures by photolithography and Ar ion milling. Resistivity was measured using the four-probe method at 100 μA in a Physical Property Measurement System (PPMS) Dynacool sample chamber equipped with a rotating sample stage. Low-noise measurements below 1 K were performed by an AC analog lock-in technique, and the sample temperature was cooled down in a $^3$He-$^4$He dilution refrigerator.

Determination of the RRR in SrRuO$_3$
The RRR was determined as the ratio of the longitudinal resistivity $\rho_{xx}$ at 300 K [$\rho_{xx}(300 \text{ K})$] and $T\to0$ [$\rho_{xx}(T\to0 \text{ K})$]. SrRuO$_3$ exhibits Fermi liquid behavior at a low temperature, which is characterized by a linear relationship between $\rho_{xx}$ and $T^2$. Based on this relationship, we estimated $\rho_{xx}(T\to0 \text{ K})$ by extrapolating the $\rho_{xx}$ vs $T^2$ fitting line below 10 K (Extended Data Fig. 2).

Machine-learning-assisted MBE
We grew the high-quality SrRuO$_3$ films (63-nm thick) on (001) SrTiO$_3$ substrates in a custom-designed MBE setup equipped with multiple e-beam evaporators for Sr and Ru (Extended Data Fig. 3a). We precisely controlled the elemental fluxes, even those of elements with high melting points, e.g., Ru (2250°C), by monitoring the flux rates with an electron-impact-emission-spectroscopy sensor and feeding the results back to the power supplies for the e-beam evaporators. The oxidation during growth was carried out with ozone (O$_3$) gas. O$_3$ gas (~15% O$_3$ + 85% O$_2$) was introduced through an alumina nozzle pointed at the substrate. Further information about the MBE setup and preparation of the substrates are described elsewhere.$^{51}$ The surface morphology of our SrRuO$_3$ films is composed of atomically flat terraces and steps, observed by atomic force microscopy.$^{20}$ Together with Laue fringes in the θ-2θ X-ray diffraction patterns,$^{10}$ this indicates high crystalline order and a large coherent volume of our SrRuO$_3$ films.

Fine-tuning of growth conditions is essential but challenging for high-RRR SrRuO$_3$ growth. Therefore, only a few papers have reported SrRuO$_3$ films with RRRs over 50.$^{20,35,36}$ While a conventional trial-and-error approach may be a way to optimize the growth conditions, this is time-consuming as well as costly, and the optimization efficiency largely depends on the skill and experience of individual researchers. To avoid such time-consuming way and reduce experimental time and cost, we employed machine-learning-assisted MBE, which we developed in previous research.$^{20}$ Here, three important growth parameters [Ru flux rate, growth temperature, and nozzle-to-substrate distance (Extended Data Fig. 3a)] were optimized by a Bayesian optimization (BO) algorithm, which is a sample-efficient approach for global optimization.$^{52}$ This algorithm sequentially produces the three parameter values where a high RRR value is predicted given past trials.

Extended Data Fig. 3b shows the procedure for machine-learning-assisted MBE growth of the SrRuO$_3$ thin films based on the BO algorithm. Here, we optimized each parameter in turn using BO: we first chose one of the growth parameters to update and fixed the rest, ran the BO algorithm to search the growth parameter, and then switched to another growth parameter. This is because BO can be inefficient in large dimensions due
to the difficulty of predicting the outcome value for unseen parameters. Here, \( RRR = S(x) \) is the target function specific to our SrRuO\(_3\) films, and \( x \) is the growth parameter (Ru flux rate, growth temperature, or nozzle-to-substrate distance). BO constructs a model to predict the value of \( S(x) \) for unseen \( x \) using the result of past \( M \) trials \( \{x_m, RRR_m\}_{m=1}^M \), where \( x_m \) are the \( m \)th growth parameters and \( RRR_m \) are the corresponding RRR values. Specifically, we use the Gaussian process regression (GPR) as a prediction model\(^{20,53}\). GPR predicts \( S(x) \) as a Gaussian random variable following \( N(\mu, \sigma^2) \). This means \( \mu \) and \( \sigma^2 \) are calculated from \( x \) and the \( M \) data points. Subsequently, we choose the growth parameter \( x \) in the next run such that the expected improvement (EI)\(^{54}\) is maximized. EI balances the exploitation and exploration by using the predicted \( \mu \) and \( \sigma^2 \) at \( x \). This measures the expectation of improvement over the best experimental RRR so far. This routine is iterated until further improvement is no longer expected. In practice, we terminate the iteration when the number of trials reaches the predetermined budget. Here, we stopped the routine at 11 samples per parameter. After completing 11 samplings for a certain parameter, we chose the value that gave the highest RRR and started the optimization of another parameter. In this optimization procedure, we used the RRR\((T = 4 \text{ K})\) instead of the RRR\((T \rightarrow 0 \text{ K})\) for easy estimation. Further details about the implementation of machine-learning-assisted MBE are described elsewhere\(^{20}\).

In our previous study\(^{20}\), we carried out the optimization of the Ru flux rate while keeping the other parameters unaltered. Subsequently, we tuned the growth temperature and the nozzle-to-substrate distance. As a result, we obtained a high-RRR \((51.8)\) SrRuO\(_3\) film in only 24 MBE growth runs (Extended Data Fig 3c). Since it was still lower than the highest value ever reported \((\sim 80)\) in the literature\(^{12,36}\), we further carried out the re-optimization of the Ru flux rate and growth temperature with previously optimized parameters \((\text{the Ru flux} = 4.2 \text{ Å/s, growth temperature} = 721 \text{ ºC}, \text{and nozzle-to-substrate distance} = 15 \text{ mm})\) as a starting point to find the global-best point in the three-dimensional parameter space. Extended Data Fig. 3c shows the highest experimental RRR values plotted as a function of the total number of MBE growth runs. With the re-optimization of the Ru flux rate and growth temperature, the highest RRR\((T = 4 \text{ K})\) value increased and reached 81 in only 44 MBE growth runs. The highest experimental RRR\((T \rightarrow 0 \text{ K})\), 84.3, was achieved at the Ru flux = 3.65 Å/s, growth temperature = 781 ºC, and nozzle-to-substrate distance = 15 mm. The availability of such high-quality film allowed us to probe the intrinsic transport properties of SrRuO\(_3\).

**Temperature dependence of the AHE in the SrRuO\(_3\) film with the RRR = 84.3**

It is known that the AHE in SrRuO\(_3\) is caused by extrinsic factor (side-jump scattering) in addition to an intrinsic factor (KL mechanism)\(^{12,29,30}\). To determine the contributions of the intrinsic and extrinsic factors to the AHE, we investigated the temperature-dependent scaling of the AHE in the SrRuO\(_3\) film with the RRR = 84.3.

The Hall resistivity \( \rho_{xy}(B) \) in SrRuO\(_3\) is described as the summation of the ordinary \( \rho_{xy}^{OHE}(B) \) and anomalous \( \rho_{xy}^{AHE}(B) \) components of Hall effects\(^{12,29,30}\):

\[
\rho_{xy}(B) = \rho_{xy}^{OHE}(B) + \rho_{xy}^{AHE}(B) \quad (2)
\]

\( \rho_{xy}^{AHE}(B) \) is proportional to the perpendicular magnetization component \( M_{\perp} \): \( \rho_{xy}^{AHE}(B) = c \rho_s M_{\perp}(c; \text{constant}) \). Here, the proportional coefficient \( \rho_s \) differs depending on the origin of the AHE: AHE can have intrinsic (KL mechanism) and extrinsic (side jump scattering) origins\(^{12,29,30}\). As shown in Extended Data Fig. 4a, above \( T_F \), clear AHE, which is
proportional to the magnetization hysteresis curve (Fig. 1d, right inset), is observed, and it dominates Hall resistivity near zero magnetic field. On the other hand, below $T_F$, the AHE components are negligibly small.

The temperature dependence of the AHE in SrRuO$_3$ has been well reproduced by a model where both the intrinsic KL mechanism and the extrinsic side-jump scattering terms are taken into account$^{30}$. In this model, the relationship between $\rho_s$ and $\rho_{xx}$ is described as:

$$\rho_s = \frac{a_1}{\Delta^2 + a_2 \rho_{xx}^2} \rho_{xx}^2 + a_3 \rho_{xx}$$  \hspace{1cm} (3)

where, $a_1$, $a_2$, $a_3$, and $\Delta$ are the fitting parameters, which are associated with the band structure$^{30}$. The first term provides a contribution from the off-diagonal matrix elements of the velocity operators, called the KL term in the model$^{30}$, and the second term means a contribution from the side-jump scattering. Extended Data Fig. 4b shows the $\rho_s$ vs $\rho_{xx}$ plot of the SrRuO$_3$ film with the RRR = 84.3 and the fitting result of eq. (3). The fitting curve reproduces the AHE sufficiently, indicating that the AHE in the SrRuO$_3$ film arises from the intrinsic KL mechanisms along with the extrinsic side-jump scattering. The important point in eq. (3) is that the AHE is asymptotic to zero when $\rho_{xx} \rightarrow 0$. Accordingly, for the SrRuO$_3$ film with such a very high RRR (84.3), equivalently with a very small residual $\rho_{xx}$, AHE becomes negligibly small at low temperatures. Therefore, $\rho_{xy}(B)$ curves below $T_F$ in our data are dominated by the ordinary Hall effect.

Data pretreatment for quantum oscillations

Pretreatments of the SdH oscillation data are crucial for deconvoluting quantum oscillation spectra since magnetoconductivity data generally contain not only oscillation components but also other magneto-resistive components as background signals$^{35}$. In particular, SdH oscillations in SrRuO$_3$ are subject to being masked by large non-saturated positive MR (Extended Data Fig. 6a)$^{35}$. Here, we subtracted the background using a polynomial function up to the fifth order and extracted the oscillation components as shown in Extended Data Fig. 6b. Then, we carried out the well-established pretreatment procedure for Fourier transform of quantum oscillations$^{35,56}$. First, we interpolated the background-subtracted data in order to prepare an equally spaced data set as a function of $1/B$. Then, we multiplied the Hanning window function to obtain the periodicity of the experimental data. Finally, we performed fast Fourier transform on the treated data set.

SdH oscillations of trivial orbits

Together with SdH oscillations from the non-trivial orbits having low frequencies ($F_1$ and $F_2$) (Fig. 3b, c), we observed SdH oscillations from the trivial orbits having high frequencies ($F_3$, $F_4$, $F_5$, and $F_6$) at 0.07 K $< T < 0.75$ K and 12.5 T $< B < 14$ T (Fig. 3d and Extended Data Fig. 8a). Since the carriers in the trivial orbits in SrRuO$_3$ are expected to have larger effective masses than those in the $F_1$ and $F_2$ orbits$^{12,35,50}$, measurements of the former oscillations should be carried out in relatively low-$T$ and high-$B$ regions. We estimated the cyclotron masses of the carriers in $F_3$, $F_4$, $F_5$, and $F_6$ orbits from the temperature dependences of the respective peaks based on the LK theory (Extended Data Fig. 8b-e). In the LK theory for the mass estimation, $B$ is determined as the interval value in the magnetic field range. Extended Data Table 1 shows the estimated cyclotron masses for $F_1$, $F_2$, $F_3$, $F_4$, $F_5$, and $F_6$. The cyclotron masses in the $F_3$, $F_4$, $F_5$, and $F_6$ orbits are relatively high (> 2.8m$_0$), reflecting the trivial band structure (energy dispersions) as their
RRR dependence of the ferromagnetism, Fermi liquid behavior, and Weyl behavior

In Fig. 4a-c, we investigated the RRR dependence of the ferromagnetism, Fermi liquid behavior, and Weyl behavior in SrRuO$_3$. For the ferromagnetism, $T_C$ values are estimated as the position of the kinks in $\rho_{xx}$ vs $T$ curves as shown in Extended Data Fig. 9a. For the Fermi liquid behavior, we defined the Fermi liquid region ($T < T_F$) as the temperature range where the experimental $\rho_{xx}$ and the linear fitting line in $\rho_{xx}$ vs $T^2$ are close enough to each other ($< 0.1 \, \mu\Omega \, \text{cm}$) as shown in Extended Data Fig. 9b. The upper limit temperature for measuring Weyl behavior in SrRuO$_3$ ($T_W$) is defined as the highest temperature at which the resistivity at zero field is lower than that at 9 T ($\rho_{xx}(0 \, \text{T}) < \rho_{xx}(9 \, \text{T})$).
Extended Data Fig. 1 HAADF-STEM and EELS-STEM images of SrRuO$_3$. (From left to right) HAADF-STEM image of the SrRuO$_3$ film with the RRR of 71 taken along the [100] axis of the SrTiO$_3$ substrate. EELS-STEM images for the Ru-$M_{4,5}$- (red), Ti-$L_{2,3}$- (green), Sr-$M_3$- (blue), O-$K$-edge (orange), and a color overlay of the EELS-STEM images for Sr, Ru and Ti.
Extended Data Fig. 2 Fermi liquid behavior in SrRuO$_3$ and the definition of $\rho_{xx}(T \to 0 \text{ K})$. $\rho_{xx}(T)$ versus $T^2$ curve (blue line) for the SrRuO$_3$ film with the RRR of 84.3. The black dashed line is the linear fitting result. Close-up near 0 K$^2$ is shown in the inset. The $\rho_{xx}(T \to 0 \text{ K})$ value is estimated from the extrapolation of the fitting line to the 0 K$^2$ axis.
**Extended Data Fig. 3 Machine-learning-assisted MBE.** a, Schematic illustration of our multi-source oxide MBE system. EIES: Electron Impact Emission Spectroscopy. b, Flowchart of machine-learning-assisted MBE growth based on the BO algorithm. c, Highest experimental RRR values plotted as a function of the total number of MBE growth runs. In c, open circles are data deduced from Ref. 20. Here, the Ru flux rate, growth temperature, and nozzle-to-substrate distance were varied in ranges between 0.18 and 0.61 Å/s, 565 and 815°C, and 1 and 31 mm, in correspondence with the search ranges in BO.
Extended Data Fig. 4 Temperature dependence of the Hall resistivity and scaling of the AHE in SrRuO$_3$. 

**a**, Hall resistivity $\rho_{xy}(B)$ curves at 2 to 100 K for the SrRuO$_3$ film with the RRR of 84.3 with $-0.3 \, \text{T} < B < 0.3 \, \text{T}$ applied to the out-of-plane [001] direction of the SrTiO$_3$ substrate. In **a**, the Hall resistivity at each temperature has been offset by 0.15 $\mu\Omega\cdot\text{cm}$ for easy viewing. 

**b**, $\rho_s$ versus $\rho_{xx}$ plot (red circles) and fitting results by eq. (3) (black dashed curve) up to 130 K. In **b**, $\rho_s$ at each temperature was obtained by dividing $\rho_{xy}(0 \, \text{T})$ in Fig. 1f by $M_\perp$ with 100 Oe obtained by the SQUID measurements. Here, the magnitude of $\rho_{xy}(0 \, \text{T})$ is defined as the averaged absolute value of $\rho_{xy}$ at $\pm 0.2 \, \text{T}$. $\rho_{xx}$ at each temperature is the same data as in Fig. 1d.
Extended Data Fig. 5 Chiral-anomaly-induced linear and negative MR in SrRuO$_3$. MR $\rho_{xx}(B)$ at $\alpha = 0$ (brown solid curve) for the SrRuO$_3$ film with the RRR of 84.3 and the linear fitting line (black dashed line) to $\rho_{xx}(B)$ in the negative MR region ($8 \text{T} < B < 14 \text{T}$). The fitting line completely reproduces the negative and linear MR region. The purple dashed line corresponds to the value of the zero field resistivity $\rho_{xx}(0 \text{T})$. 
Extended Data Fig. 6 Pretreatment of the SdH oscillation data. a, Raw $\sigma_{xx}(B)$ data measured at 2 K and $\beta = \gamma = 90^\circ$ for the SrRuO$_3$ film with the RRR of 84.3. The raw conductivity data is obtained by using $\rho_{xx}(B)$ and $\rho_{xy}(B)$ data as $\sigma_{xx}(B) = \rho_{xx}(B)/((\rho_{xx}(B))^2 + (\rho_{xy}(B))^2)$. b, SdH oscillation data $\Delta \sigma_{xx}(B)$ obtained by subtracting a polynomial function up to the fifth order from $\sigma_{xx}(B)$ in a.
Extended Data Fig. 7 LK theory fitting to the SdH oscillations with the fixed zero Berry phases. The SdH oscillation data at 2 K for the SrRuO$_3$ film with the RRR of 84.3 and the fitting results by eq. (1) with the zero Berry phases ($\beta_1 = \beta_2 = 0$). The SdH oscillation is the same data as in Fig. 3b. The fitting was carried out by a non-linear least squares method with the fitting parameters $A_1$, $T_{D1}$, $A_2$, and $T_{D2}$. The fitting curve cannot reproduce the experimental data well, confirming the existence of the non-zero Berry phase.
Extended Data Fig. 8 SdH oscillations and mass estimations of the trivial orbits having high frequencies. **a**, Fourier transform spectra of the SdH oscillations from 70 to 750 mK for the SrRuO$_3$ film with the RRR of 84.3. The spectrum is obtained by fast Fourier transform for the oscillation data $\Delta \rho_{xx}(B)$ from 12.5 T to 14 T. $F_3$, $F_4$, $F_5$, and $F_6$ peaks correspond to 300, 500, 3500, and 3850 T, respectively. **b-e**, Mass estimations for the $F_3$, $F_4$, $F_5$, and $F_6$ orbits according to the LK theory. Black dashed curves are fitting curves.
Extended Data Fig. 9 Estimation of $T_C$ and $T_F$ values in different RRR samples. a, $\rho_{xx}(T)/\rho_{xx}(T\rightarrow 0 \text{ K})$ versus $T$ of the different RRR samples. The kinks around 150 K correspond to $T_C$ of the sample. b, $\rho_{xx}(T)/\rho_{xx}(T\rightarrow 0 \text{ K})$ versus $T^2$ curves with the linear fittings (black lines) for the different RRR samples. The black dashed lines correspond to $T_F^2$ where the experimental $\rho_{xx}$ and the fitting line are close enough to each other ($< 0.1 \mu\Omega \text{ cm}$).
Extended Data Table 1 Frequencies and effective cyclotron masses estimated from SdH oscillations in the SrRuO₃ film with RRR = 84.3. Frequencies and effective cyclotron masses estimated in Fig. 3c and Extended Data Fig. 8b-d. $m_0$ represents the electron rest mass.

|       | $F_1$ | $F_2$ | $F_3$ | $F_4$ | $F_5$ | $F_6$ |
|-------|-------|-------|-------|-------|-------|-------|
| $F$ (T) | 26    | 44    | 300   | 500   | 3500  | 3850  |
| $m^*/m_0$ | 0.30  | 0.49  | 2.9   | 3.1   | 5.0   | 5.8   |

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