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Nonlinear Transverse Transport in a Ferromagnetic Polar Metal

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ABSTRACT

Symmetry-driven nonlinear responses in quantum materials hold key importance to uncover emergent electronic phenomena. While the nonlinear transport has been extensively studied in nonmagnetic materials, its correlation with ferromagnetism remains an open and intriguing question. Here, we explore the nonlinear transverse transport in a ferromagnetic polar metal. Specifically, in non-centrosymmetric SrRuO₃(111) thin films, we observed an abrupt enhancement of the time-reversal-even transverse nonlinear signal during the paramagnetic-ferromagnetic transition. Scaling analysis reveals a sign reversal of both the skew scattering and side jump contributions upon the emergence of ferromagnetism. Combined with theoretical analysis, we correlate this behavior with a sign change of Berry curvature triple on the Fermi surface, driven by the redistribution of Berry curvature hot spots across the magnetic transition. Our findings unveil that magnetic ordering can induce a dramatic nonperturbative change of nonlinear responses, establishing correlated magnetic oxides as a promising platform for exploring tunable magnetic nonlinear transport effects.

1 | Introduction

Polar metals—materials that simultaneously host metallic conduction and inversion symmetry breaking – have emerged as fertile ground for unconventional electronics [1–4]. In these materials, a particularly intriguing phenomenon is nonlinear transverse transport, a second-order response that requires

inversion-symmetry breaking but does not need an external magnetic field [5–9]. This nonlinear effect is fundamentally linked to the momentum-space pattern of Berry curvature, which becomes asymmetric when inversion symmetry is broken. While inversion symmetry breaking is recognized as a prerequisite, the role of broken time-reversal ($\mathcal{T}$) symmetry—a hallmark of ferromagnetic systems—has remained largely unnoticed. Most

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recent works have focused on antiferromagnetic systems such as MnBi_2Te_4 [10–12], where the combined $\mathcal{PT}$ symmetry is preserved and the observed nonlinear signals are predominantly $\mathcal{T}$ -odd [13]. In contrast, ferromagnetic polar metals, which simultaneously host metallicity, polarity, and spontaneous magnetization, provide an ideal yet largely unstudied platform to probe $\mathcal{T}$ -even nonlinear transport upon presence of magnetic order. This limited understanding stems from the scarcity of suitable materials that combine polar displacements, metallicity, and ferromagnetic order, which requires harnessing strong electron-electron correlations and makes it nontrivial to probe their interplay with nonlinear phenomena [14].

Notably, previous studies have shown that the nonlinear transport can vary dramatically during topological or ferroelectric phase transitions, where the dipole pattern of underlying asymmetric Berry curvature landscape changes [15–20]. This raises the compelling possibility that a magnetic phase transition, which lifts the antisymmetric constraint on the Berry curvature distribution hence permits richer patterns of the latter, can greatly modulate the behavior of nonlinear responses. In particular, if the exchange coupling reshapes the hot-spot structures of Berry curvature around the Fermi surface [21–23], the nonlinear response is anticipated to be dramatically altered. Once achieved, this will not only illuminate the so far obscure physics of exchange-driven nonlinear Berry curvature effects, but also unlock new potential for harnessing nonlinear responses to create novel spintronics device functionalities [6, 24].

Complex magnetic oxides offer an ideal platform for exploring the correlation between magnetism and nonlinear transverse transport. Their strong charge-spin-lattice coupling enables the engineering of a wide range of structural, electronic, and magnetic properties, and allows flexible control over both inversion and time-reversal symmetries [1, 25–27]. It was shown that strain engineering in ferromagnetic metallic oxides can induce electrical polarity, stabilizing a magnetic polar metal phase [14, 28–30], which creates an ideal platform to investigate nonlinear transport in ferromagnets. Here, ferromagnetic polar metal refers to a metallic ferromagnetic thin-film state with strain-induced structural inversion-symmetry breaking, rather than an intrinsic bulk polar phase of SrRuO_3 . Interestingly, in thin film format [31, 32], a threefold rotational axis can be maintained along the growth direction, which prohibits the dipole form of Berry curvature distribution but enforces a novel Berry curvature triple (BCT) pattern [33–36]. In such a case, the $\mathcal{T}$ -even nonlinear transport in transverse direction can only arise from disorder induced extrinsic mechanisms such as skew scattering and side jump [37–39], which are intimately linked to BCT [33–36]. As a geometric quantity uniquely characterizing threefold symmetric Berry curvature distributions, BCT remains unexplored in magnetic phases and upon magnetic phase transitions. Therefore, examining how magnetic phase transitions affect $\mathcal{T}$ -even nonlinear transport signals is of significant interest for elucidating exchange-driven variations in BCT.

In this work, we report the observation of $\mathcal{T}$ -even nonlinear transverse transport response in polar metal SrRuO_3 (111) thin films. The SrRuO_3 (111) films exhibit inversion symmetry breaking by flexoelectric effect [40] but preserve an effective threefold rotational symmetry in the transport plane, allowing

for nonvanishing BCT. During the paramagnetic-ferromagnetic phase transition at 138 K, we observed a large enhancement of nonlinear transverse conductivity. Remarkably, the scaling analysis shows that when the system enters the ferromagnetic phase, both the skew scattering and side jump nonlinear contributions reverse their signs, which are consistent with our first-principles calculations revealing a sign change of BCT across the magnetic phase transition. Our analysis also suggests that this sign reversal can be traced back to the distinct nature of Berry curvature hot spots in the two phases, i.e., spin-orbit local gap in paramagnetic phase versus exchange gap in ferromagnetic phase. This qualitative change is rooted in the emergent strong internal magnetization, going beyond the perturbative picture for nonlinear Hall transport induced by external magnetic field [41–43]. Our observations illuminate nonlinear transport behaviors during magnetic phase transitions and underscore the need for further investigations into the influence of magnetism on $\mathcal{T}$ -even nonlinear transverse transport.

2 | Results

2.1 | Basic Properties of SrRuO_3 (111) Thin Films

SrRuO_3 (111) thin films were grown on SrTiO_3 (111) substrate via pulsed laser deposition technique (See Methods for details). As shown in Figure 1a, standard Hall bar devices were fabricated via photolithography for transport measurements. The first (V^ω) and second order harmonic voltages ($V^{2\omega}$) can be obtained under an ac source current $I_x = I \sin(\omega t)$ using lock-in techniques. Figure 1b shows the longitudinal resistivity ρ_{xx}^ω as function of temperature for the thin film with 8 nm thickness. The film shows typical metallic behavior as the ρ_{xx}^ω monotonically decreases with lowering temperature. A kink at 138 K indicates the paramagnetic-ferromagnetic phase transition. To further verify the transition, we measured anomalous Hall effect at different temperatures. We plot the linear Hall resistivity ρ_{yx}^ω as function of out-of-plane magnetic field for several representative temperatures in Figure 1c. Above 138 K, the anomalous Hall effect is negligible indicating the absence of magnetic order. Below 138 K, the anomalous Hall signal starts to emerge and exhibit hysteresis behavior, consistent with the ferromagnetic metallic properties reported in SrRuO_3 thin films [21, 44]. Note that ρ_{yx}^ω has a very small contribution from the contact misalignment of the Hall bar devices (Figure S1).

Strain gradient due to heteroepitaxy can induce non-centrosymmetric structure that breaks inversion symmetry [45]. In SrRuO_3 (111) thin films, recent reports show that shear strain gradient causes large off-centering displacement of Ru ions along $[\bar{1}\bar{1}0]$ direction, as illustrated in Figure 1e,f [40] (see also Note S1 and Figure S2 for STEM confirmation). The observed displacement is induced by strain gradients rather than intrinsic bulk polar order, reflecting a polar metallic state that arises from structural inversion symmetry breaking instead of true bulk ferroelectricity. Such displacement gives rise to a non-centrosymmetric polar structure. To examine its existence, we performed optical second-harmonic generation (SHG) and the results are shown in Figure 1d. Threefold symmetric SHG anisotropy pattern is observed which aligns well with point group m with three equivalent electric dipoles originated from 120°

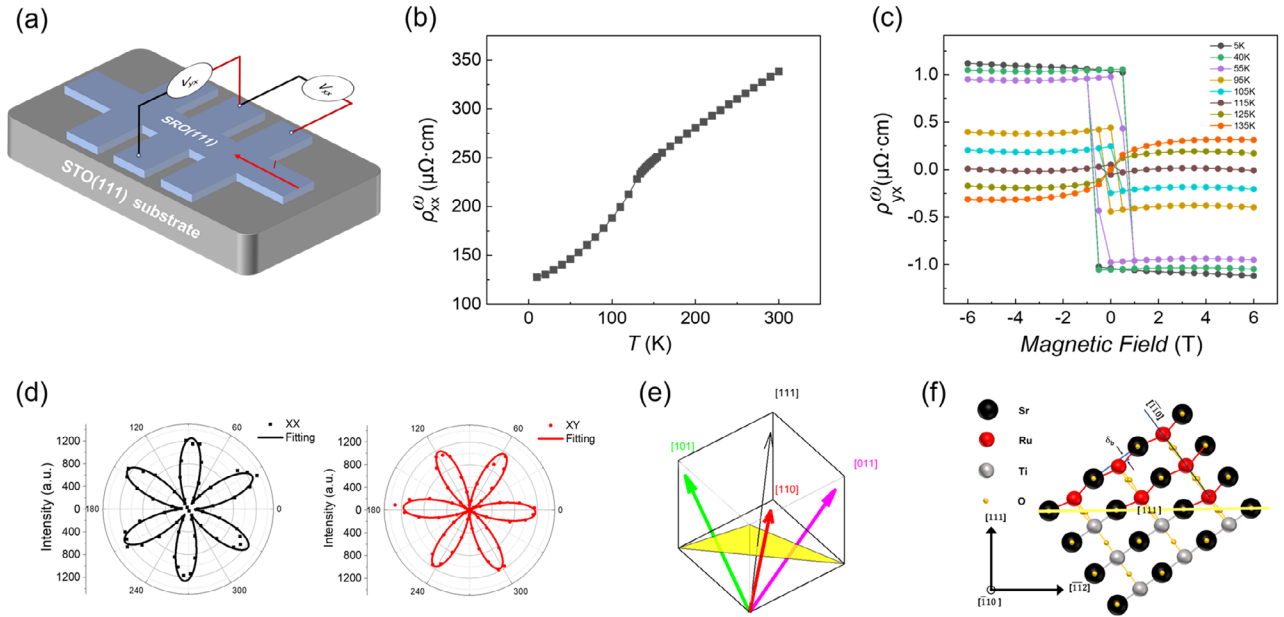


FIGURE 1 | Properties of SrRuO₃(111) thin film. (a) Schematic of Hall bar device of SrRuO₃ thin film. A sinusoidal current I^ω was applied, and the first harmonic V_x^ω and second harmonic $V_y^{2\omega}$ voltages were simultaneously measured along the longitudinal and transverse directions. (b) The longitudinal resistivity ρ_{xx}^ω as a function of temperature. (c) Magnetic field-dependent linear Hall resistivities ($\rho_{yx}^\omega \sim H$) of 8 nm SrRuO₃ thin film. (d) SHG patterns acquired under XX and XY polarization configurations. Here, the excitation and detection beams were linearly polarized, with XX and XY referring to co- and cross-linearly polarized between the two beams, respectively. (e) Schematic of polar symmetry in SrRuO₃ (111) thin film with three electric dipoles along [101], [011] and [110] directions. (f) Schematic showing the off-centering of Ru atoms along $[\bar{1}\bar{1}0]$ induced by a flexoelectric field [40].

rotated domains as illustrated in Figure 1e, in good agreement with previous report [46]. These results confirm the emergence of inversion-symmetry breaking in a ferromagnetic metallic system, offering a rare breed to explore $\mathcal{T}$ -even nonlinear transverse transport under time-reversal symmetry breaking conditions.

2.2 | Nonlinear Transverse Transport Measurement

Figure 2a shows the second-harmonic transverse voltage $V_y^{2\omega}$ measured at zero magnetic field and room temperature. $V_y^{2\omega}$ shows quadratic current dependence $V_y^{2\omega} \propto I^2$, and changes sign when reversing simultaneously the current and corresponding transverse probe directions, validating the nonlinear origin. We further verified that $V_y^{2\omega}$ signal is independent of the ac frequency used (Figure S3), suggesting negligible capacitive contributions. These results unambiguously demonstrate the existence of transverse nonlinear transport in SrRuO₃ (111) thin film and are in line with its inversion-symmetry breaking nature [40, 46].

We then investigated the temperature dependence of nonlinear transport (raw measurements in Figure S4). Figure 2b shows the $V_y^{2\omega} \sim I^2$ curves at different temperatures under zero magnetic field, all of which show good quadratic current dependence. The slope of the $V_y^{2\omega} \sim I^2$ curves is defined as the second order transverse resistance R_{yxx} , which is plotted along with ρ_{xx} in Figure 2c. We achieve R_{yxx} for both upward and downward magnetization (M) at zero magnetic field. The $\mathcal{T}$ -even term is

defined as $R_{yxx}^{T\text{-even}} = \frac{R_{yxx}(+M) + R_{yxx}(-M)}{2}$ while the $\mathcal{T}$ -odd term is defined as $R_{yxx}^{T\text{-odd}} = \frac{R_{yxx}(+M) - R_{yxx}(-M)}{2}$. While $R_{yxx}^{T\text{-even}}$ can exist in both paramagnetic and ferromagnetic phases, $R_{yxx}^{T\text{-odd}}$, which may arise from thermally driven anomalous Nernst effect or $\mathcal{T}$ -odd nonlinear Hall effect [47–49], is only allowed in ferromagnetic phase. As shown in Figure 2c, $R_{yxx}^{T\text{-odd}}$ is indeed vanishingly small in the paramagnetic phase, and remarkably, is still negligible compared to $R_{yxx}^{T\text{-even}}$ in the ferromagnetic phase, showing the dominant $\mathcal{T}$ -even contribution to nonlinear transverse response. The smallness of $R_{yxx}^{T\text{-odd}}$ in ferromagnetic phase is found in all the measured devices (Figure S5), thus solidly validating the $\mathcal{T}$ -even nature of the observed nonlinear transport in both paramagnetic and ferromagnetic phases. Therefore, $R_{yxx} \approx R_{yxx}^{T\text{-even}}$ and in the following analysis we use the $R_{yxx}^{T\text{-even}}$ data for R_{yxx} . As the temperature is lowered through the Curie point, ρ_{xx} exhibits a marked acceleration in its downward trend, whereas the $R_{yxx}^{T\text{-even}}$ undergoes a pronounced enhancement. The starkly contrasting temperature dependence of these two quantities—faster suppression of ρ_{xx} versus the abrupt rise in $R_{yxx}^{T\text{-even}}$ at the magnetic phase transition—strongly implies that, beyond conventional scattering effects, the nonlinear Hall signal is probing additional bandstructure changes (for example, rapid redistribution of Berry curvature or the emergence of new band crossings) in the vicinity of the paramagnetic–ferromagnetic boundary.

2.3 | Scaling Analysis

To gain more insight into the impact of paramagnetic–ferromagnetic phase transition, we evaluated the nonlinear

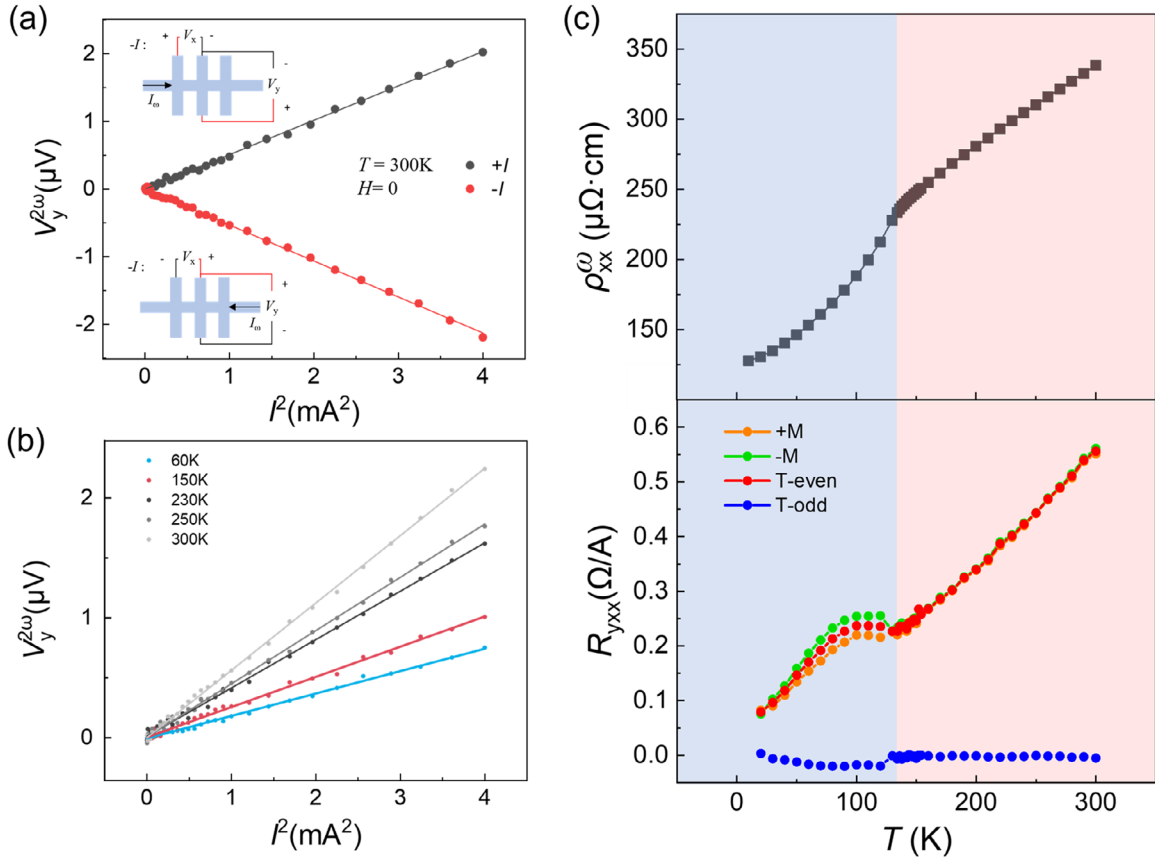


FIGURE 2 | Nonlinear transverse response across magnetic phase transition. (a) The second-harmonic transverse voltage $V_y^{2\omega}$ versus I^2 for the current along the $[1\bar{1}0]$ axis. The solid lines are linear fits to the data. The insets show schematics of current and voltage probe connections under opposite current directions of $+I$ and $-I$. The data were collected at $T = 300$ K without external magnetic field. (b) $V_y^{2\omega}$ versus I^2 at different temperatures, and the slope of which is nonlinear transverse resistance R_{yxx} . (c) The longitudinal resistivity ρ_{xx}^ω (black), the nonlinear transverse resistance R_{yxx} for upward magnetization (orange) and downward magnetization (green), the $\mathcal{T}$ -even term (red) and the $\mathcal{T}$ -odd term (blue) of R_{yxx} versus temperature plotted along. Red and blue regions denote the high temperature paramagnetic and low temperature ferromagnetic phase, respectively.

transverse conductivity $\sigma_{yxx}^{(2)} = -\sigma_{xx} \frac{V_y^{2\omega}}{(V_x^\omega)^2} \frac{L^2}{W}$, where L and W are the length and width of the Hall bar channel. We focused on the temperature dependence of the nonlinear transverse response in the vicinity of the paramagnetic-ferromagnetic transition, where the evolution of magnetic order provides the most direct insight into the role of time-reversal symmetry breaking. The results are shown in Figure 3a. While $\sigma_{yxx}^{(2)}$ varies slowly above the magnetic transition temperature T_c , a sharp increase with decreasing temperature is clearly observed in the ferromagnetic phase. We then analyzed the scaling behavior to help identify the involved physical mechanisms [7, 33, 37]. Using the temperature-dependent data, $\sigma_{yxx}^{(2)}/\sigma_{xx}^2$ is plotted as a function of σ_{xx}^2 as shown in Figure 3b. Two regimes are clearly observed corresponding to the paramagnetic ($\sigma_{xx}^2 < 16 \times 10^6$ (S/cm)²) and ferromagnetic ($\sigma_{xx}^2 > 16 \times 10^6$ (S/cm)²) phases from the transport data. In both regimes we found that the data well satisfy the scaling

$$\sigma_{yxx}^{(2)} = \xi \sigma_{xx}^3 + \eta \sigma_{xx} \quad (1)$$

Here, the cubic term corresponds to the contribution from skew scattering [7, 35], whereas the linear term may originate from Berry curvature dipole [5, 6] or side jump [7, 33]. Notably, SrRuO₃(111) remains a good metal with moderate disorder,

satisfying the key assumptions underlying the scaling analysis in Equation (1). Considering that the system has a threefold rotational symmetry evidenced by the results in Figure 1, the contribution of Berry curvature dipole is excluded. We extracted the values of ξ and η from the fitting. An interesting finding is that both ξ and η experience a sign change during the paramagnetic-ferromagnetic transition. Such sign-changing scaling behaviors were cross-checked in all other samples (Figures S6 and S7). In particular, thick samples (25 nm) also display the sign change of both ξ and η , validating that such behaviors do not originate from surface or interface, but rather reflects intrinsic nonlinear physics of bulk states during the magnetic phase transition. Moreover, in temperature-dependent SHG measurements (Note S2 and Figure S8) [50], we do not observe any structural change or non-collinear magnetic inhomogeneity during the magnetic phase transition, pointing toward an electronic band origin of our observation. Here, we mention that Equation (1) has been employed by a recent work on nonlinear transverse transport in Sb-doped topological insulator MnBi₄Te₇ [51], where a sign change of ξ was also claimed. However, it is unknown whether the signal measured there is $\mathcal{T}$ -even or $\mathcal{T}$ -odd. If it is $\mathcal{T}$ -odd, then the cubic term has totally different mechanisms from skew scattering [52]. More critically, the linear longitudinal resistivity therein shows an insulating feature, thus the validity of Equation (1) is unclear

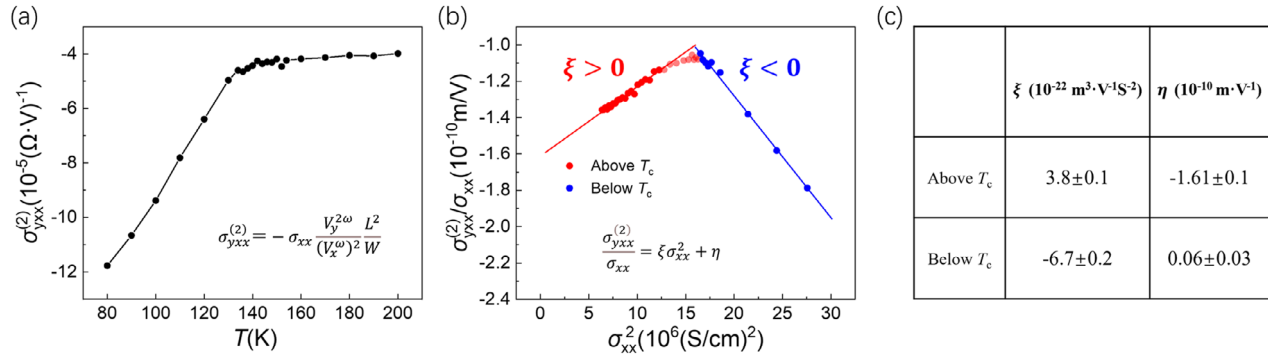


FIGURE 3 | Scaling law of nonlinear transverse conductivity near T_c . (a) The nonlinear transverse conductivity $\sigma_{yxx}^{(2)}$ as a function of temperature. (b) Scaling law analysis by plotting $\sigma_{yxx}^{(2)}/\sigma_{xx}$ versus σ_{xx}^2 . The dots are the experimental data. The solid lines are linear fits to the experimental data in paramagnetic (red) and ferromagnetic (blue) phases. (c) The slopes ξ and intercepts η of linear fitting for paramagnetic (red) and ferromagnetic (blue) phases are shown.

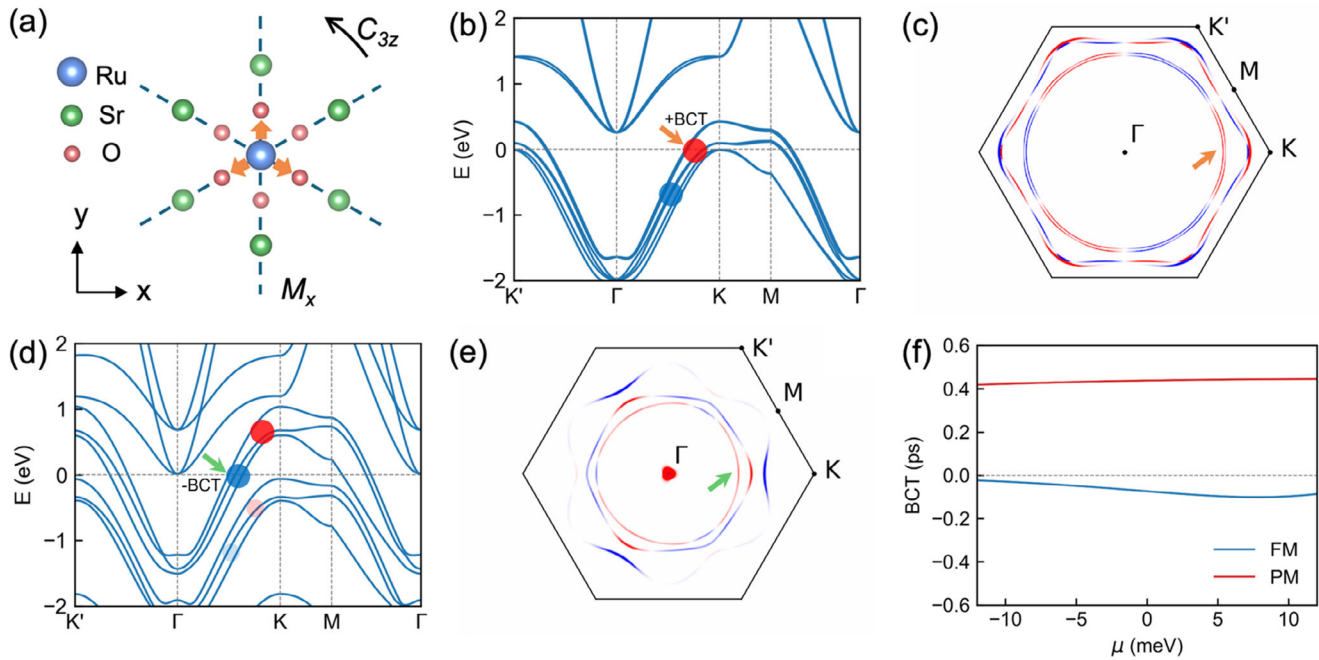


FIGURE 4 | Calculated band structures and Berry curvature distribution. (a) Top view of SrRuO_3 (111). The three vectors represent the displacement of Ru atoms along $[\bar{1}\bar{1}0]$, $[\bar{1}0\bar{1}]$ and $[0\bar{1}\bar{1}]$ for the three polar domains. (b,c) Calculated band structure and Berry curvature distribution at Fermi level for the paramagnetic phase, respectively. The dashed line denotes $E = 0$ eV, corresponding to the Fermi level. (d,e) Same as b), c) but for the ferromagnetic phase. The spin-orbit gap in the paramagnetic phase and exchange gap in the ferromagnetic phase are indicated by orange arrows and green arrows, respectively. Berry curvature hotspots at the Fermi level induced by the two gaps are indicated by red circles (+BCT) and blue circles (-BCT). (f) Calculated BCTs for paramagnetic (PM) and ferromagnetic (FM) phase as function of chemical potential.

[23, 37, 53]. On the other hand, in our case the resistivity shows metallic behavior (Figure 2c), justifying the use of Equation (1).

As both the skew scattering and side jump are related to BCT under a threefold rotational symmetry [33–36], the sign change of ξ and η can be traced back to that of BCT, which we explored from symmetry characteristics and first-principles calculations. The BCT is defined as a convolution of Berry curvature $\Omega_z(\mathbf{k})$ with an angular form factor characteristic of the threefold rotational symmetry: [33–36] $\text{BCT}(\mu) = 2\pi\hbar \int \frac{d\mathbf{k}}{(2\pi)^2} \delta(\epsilon_{\mathbf{k}} - \mu) \Omega_z(\mathbf{k}) \cos 3\theta_{\mathbf{k}}$, where the integration is over the (111) plane and $\theta_{\mathbf{k}}$ is the angle between $\mathbf{k}$ and the Γ -K direction. The structure of the

SrRuO_3 (111) plane is shown in Figure 4a. It exhibits inversion symmetry along with threefold rotation and mirror symmetries, as indicated by the dashed lines. While individual domains locally break threefold symmetry, the macroscopic transport effectively averages over these domains, thereby restoring the threefold symmetry required for the Berry Curvature Triple analysis. The combined inversion and time-reversal ($\mathcal{PT}$) symmetry enforces double degeneracy in the band structure, leading to vanishing Berry curvature and BCT. As mentioned, the SrTiO_3 (111) substrate imposes shear strain on SrRuO_3 , and the Ru atoms undergo an off-center shift along $[\bar{1}\bar{1}0]$, $[\bar{1}0\bar{1}]$ or $[0\bar{1}\bar{1}]$ direction by 0.1 Å on average in polar domains [40]. Possible effects of domain

boundaries or local strain inhomogeneity would mainly cause quantitative broadening of the Berry curvature distribution, but are negligible for the observed sign reversal of the Berry curvature triple. The strain dependence of the nonlinear signal further supports the key role of strain in driving this Ru off-centering (see Note S3 and Figure S9). As a result, the overall system retains C_{3z} symmetry while the inversion symmetry is broken, enabling nonzero BCT. It should be noted that the electrons on Fermi surfaces are mainly derived from Ru atoms. As such, the shift of Ru atoms shall significantly alter the transport properties of SrRuO₃ thin films.

2.4 | First-Principles Calculations

To perform first-principles calculations, we directly shift the Ru atoms along the [111] direction in the crystal model to simulate the average effect, instead of the realistic displacements as given in Figure 4a. The resulting structure breaks inversion symmetry but preserves the threefold axis C_{3z} , which is in line with our SHG result. We set the displacement to be 0.1 Å, which was selected based on the previous report [40]. We first examined the paramagnetic phase above the critical temperature. Due to PT symmetry breaking, there is a band splitting of approximately 40 meV on the Fermi surface, as shown in Figure 4b. Such a small splitting originates from the spin-orbit coupling (SOC). It enables sizable Berry curvature distributions as shown in Figure 4c, which respects C_{3z} . In contrast, in the ferromagnetic phase below the critical temperature, the band splitting is significantly larger than the paramagnetic case, as it originates from the exchange interaction associated with Ru atoms. The resulting exchange-driven band separation is clearly visible on the Fermi surface (Figure 4e), where the Berry curvature pattern exhibits a distinct redistribution compared with the paramagnetic state. To further clarify the respective roles of SOC and exchange interactions, we calculated the band structures with and without SOC (see Note S4 and Figure S10). These results confirm that in the paramagnetic phase the Berry curvature is primarily induced by the SOC-related spin-orbit gap, whereas in the ferromagnetic phase it is dominated by the exchange gap. We calculated the BCT in both paramagnetic and ferromagnetic phases. As shown in Figure 4f, around the chemical potential, the BCT exhibits opposite signs for the two phases. This sign change feature is consistent with our observed sign reversal of the skew scattering and side jump contributions across the magnetic phase transition.

Moreover, our first-principles results suggest that this sign reversal originates from the distinct mechanisms generating Berry curvature in the two phases (spin-orbit splitting vs exchange splitting), as the spin-orbit gap in the paramagnetic state and the exchange-driven gap in the ferromagnetic state are highlighted by orange and green arrows, respectively in Figure 4b,d. These two gap openings contribute oppositely to the BCT components (see also Note S5 and Figure S11 for a qualitative illustration of Berry curvature redistribution). Finally, we note that while scaling analysis reveals a stronger skew scattering and side jump contribution in the ferromagnetic phase, the calculated BCT appears to have a larger magnitude for the paramagnetic phase. This is possible, because the whole band structure as well as Fermi surface undergo a dramatic change across phase transition

and the scattering related processes should also be strongly altered.

3 | Conclusions

In summary, our study explored nonlinear transverse transport in a ferromagnetic polar metal and uncovered fundamental relationship between time-reversal symmetry breaking and nonlinear transverse effect. We reported an abrupt change of $\mathcal{T}$ -even nonlinear transverse signal and an unexpected sign reversal of skew scattering driven by a ferromagnetic phase transition in a polar metal. The observations match well with the theoretical picture of exchange driven change of BCT. Our results not only open the door to explore nonlinear physics enabled by BCT in ferromagnetic systems, but also uncover a large number of opportunities to explore the impact of magnetic phase transition on various novel $\mathcal{T}$ -even charge [54–58]/spin [59, 60] nonlinear transport effects and the engineering of them for oxide-spintronic devices [61].

4 | Methods

4.1 | Sample Fabrication and Characterization

SrRuO₃ thin films were grown on SrTiO₃(111) substrates using a pulsed-laser deposition system with a KrF excimer laser (248 nm). Before deposition, SrTiO₃ substrates (miscut <0.1°) were etched with a buffered hydrofluoric acid solution and then annealed in air at 1050°C for 1 h to produce an atomically flat surface with a unit-cell step terrace structure. During deposition, the temperature of the substrate was maintained at 650°C and SrRuO₃ films were grown under an oxygen pressure of 80 mTorr with a laser fluence of 2 J cm⁻² and a frequency of 5 Hz. A high-pressure reflection high-energy electron diffraction system was used to monitor the growth. A smooth surface is observed as shown in Figure S12, which is further confirmed by Atomic force microscopy (Park NX10 with Al-coated tips). X-ray diffraction Φ scan was performed using a Bruker D8 Discover system.

4.2 | Device Fabrication

Conventional photolithography and ion beam etching were used to pattern the SrRuO₃ films into the Hall bar geometry. The channel size was minimized to 50 × 187.5 μm². After patterning, the samples were ex-situ annealed at 300°C in ambient oxygen flow for 3 h to minimize the oxygen deficiency induced during growth and ion-milling. Cr (5 nm) and Au (60 nm) films were sputtered onto the Hall bar as contact electrodes. After the entire fabrication process, the SRO conducting channels still exhibit atomically-flat topography (Figure S12).

4.3 | Electric Transport Measurements

The transport data were measured using a physical properties measurement system (PPMS, Quantum Design). We performed low-frequency ac electric harmonic measurements using Keithley

current source (6221) and Stanford Research lock-in amplifiers (SR830). During the measurements, a sinusoidal current with a frequency $f = \omega/2\pi = 17.3$ Hz is applied to the devices, and the in-phase (0°) first harmonic voltage V^ω and out-of-phase (90°) second harmonic voltage $V^{2\omega}$ along the longitudinal and transverse voltages were measured simultaneously by four lock-in amplifiers. The $\pm\mathbf{M}$ states of the ferromagnetic SrRuO_3 were prepared by cooling the sample to 10 K and applying an out-of-plane magnetic field of +3 T and −3 T, respectively. After establishing these remanent magnetization states, the nonlinear transport measurements were performed at zero field while warming from 10 K, ensuring that the $\pm\mathbf{M}$ datasets correspond to equal-magnitude but opposite-orientation magnetization.

4.4 | Optical Second Harmonic Generation Measurements

The nonlinear SHG measurements were conducted in a variable-temperature optical cryostat housed inside a superconducting magnet with a room-temperature bore. The excitation beam (120 fs, 80 MHz, 820 nm) from a Ti:sapphire oscillator (MaiTai HP, Spectra Physics) was focused onto the sample at normal incidence using a $50\times$ microscopic objective ($\text{NA} = 0.55$). The reflected nonlinear optical signal was collected by the same objective and detected by either a photomultiplier tube in photon-counting mode or a spectrograph equipped with a liquid-nitrogen-cooled charge coupled device. To measure the polarization-resolved intensity of nonlinear optical signal, the polarization of the excitation and signal beams was rotated with respect to the sample simultaneously using a half-wave plate. The polarization state of the signal beam was analyzed by a linear polarizer placed in front of detectors. For the measurements of temperature dependence and magnetic hysteresis loops, the excitation beam was circularly polarized using a quarter-wave plate.

4.5 | Computational Details

The first-principles calculations were performed using the Vienna Ab initio Simulation Package [62–64]. The projector-augmented wave method [65] was employed to describe the interactions between ions and electrons. Exchange-correlation effects were treated self-consistently using the Perdew-Burke-Ernzerhof functional [66]. The cutoff energy was set to be 500 eV. The Brillouin zone was sampled by a k -point grid of $9 \times 9 \times 9$. The convergence threshold for energy and force were set to 10^{-6} eV and 0.01 eV/Å, respectively. The spin-orbit coupling was considered in all calculations. The ab initio tight-binding model was constructed using the Wannier90 package [67], which was used for calculating the Berry curvature and the BCT. The Berry curvature is defined as

$$\Omega_z = -2\text{Im} \sum_{n \neq n'} \frac{\langle \varphi_{nk} | \hat{v}_x | \varphi_{n'k} \rangle \langle \varphi_{n'k} | \hat{v}_y | \varphi_{nk} \rangle}{(\varepsilon_n - \varepsilon_{n'})^2}$$

Here, φ_{nk} is the Bloch wave function with eigenvalue ε_n and the $\hat{v}_{x/y}$ is the velocity operator along the x/y direction.

Author Contributions

Xuyang Sha, Xuejin Zhang, Hao Liu, and Jin Cao contributed equally to this work. H.W.G. and J.S. conceived and supervised the project. X.Y.S. grew the films, fabricated the devices, and performed the transport measurements with the help from H.L. and J.F.Z. R.H.C. and S.W. performed the second harmonic generation measurements. Z.W. performed the STEM measurements. X.J.Z., J.C., C.X., and S.A.Y. performed the theoretical calculation and analysis. All authors discussed the results. X.Y.S., J.Z., R.C., C.X., S.A.Y., P.H., H.G. and J.S. wrote the manuscript with input from all authors.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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