

Magnetic-field-driven phase switching in the antiferromagnetic Mott insulator $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ Ksenia S. Rabinovich^{1,*}, Tim Priessnitz^{1,*}, Nils Gross^{1,*}, George Jackeli^{1,2}, Maximilian J. Krautloher¹, Pascal Reiss¹, Eberhard J. Goering¹, Jurgen H. Smet¹, Bernhard Keimer¹, and Alexander V. Boris^{1,†}¹*Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany*²*Andronikashvili Institute of Physics, Tbilisi State University, Tamarashvili str. 6, 0177 Tbilisi, Georgia*

(Received 22 June 2026; accepted 10 September 2026; published 30 September 2026)

A bandwidth-controlled antiferromagnetic Mott-insulating phase in $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$ is realized through isovalent substitution of the Ru site. For a dilute substitution with only 1% Ti, the Mott insulator ground state remains nearly degenerate with the ground state of pristine $\text{Ca}_3\text{Ru}_2\text{O}_7$, where the Ru moments are ferromagnetically aligned within the metallic RuO_2 bilayers, which are stacked in an antiferromagnetic fashion. The exceptionally shallow free-energy landscape of this doped compound arises from intertwined electron–electron and electron–lattice interactions. This makes its magnetic and transport properties highly sensitive to external perturbations. We systematically investigated magnetic-field-induced phase switching in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ to explore its magnetic H - T phase diagram. With the field applied along the easy b axis, parallel to the antiferromagnetic moments, the magnetization exhibits a first-order spin-flop transition at ≈ 6 T, indicating reorientation of the Ru moments perpendicular to the field. The transition is accompanied by a decrease in the electrical resistance, but the spin-flop phase remains insulating. Above 10.5 T, all Ru moments align with the b axis, resulting in a forced ferromagnetic metallic phase. In contrast, neither spin-flop nor forced ferromagnetic phases are observed up to 14 T when the field is applied along the a axis. While the electronic kinetic energy and the electron–lattice coupling contribute to the free-energy balance of this system, the resulting H - T phase diagram is remarkably simple and closely resembles that of a canonical anisotropic antiferromagnet, albeit with substantially renormalized critical fields.

DOI: [10.1103/z696-28zn](https://doi.org/10.1103/z696-28zn)**I. INTRODUCTION**

In $4d$ transition-metal layered perovskites, in particular the ruthenates, the energy scales of competing collective instabilities, intra-atomic spin–orbit coupling, and electronic correlation strength are comparable. As a consequence, they host a variety of nearly degenerate ground states ranging from superconductivity and Mott insulating states to alter-magnetism [1–6]. The degeneracy implies that even minor chemical substitution or slight external stimuli—such as pressure, electric field, and illumination—can significantly affect the phase behavior and macroscopic properties [7–11]. A notable example of such electronic phase sensitivity to small perturbations is bilayer perovskite calcium ruthenate $\text{Ca}_3\text{Ru}_2\text{O}_7$. Its ground state is a correlated polar metal with itinerant electrons within the antiferromagnetically (AFM) stacked ferromagnetic (FM) RuO_2 bilayers [2,12–16]. Remarkably, substituting just 1% of Ru with isovalent Ti, thus minimally tuning the effective Ru electronic bandwidth, transforms $\text{Ca}_3\text{Ru}_2\text{O}_7$ into an AFM Mott insulator [11,17–19],

placing it in a regime where competing FM metallic and AFM insulating phases coexist at nearly identical energies.

At room temperature, both $\text{Ca}_3\text{Ru}_2\text{O}_7$ and $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ are paramagnetic metals (PM-M) with an orthorhombic crystal structure, characterized by the polar space group $Bb2_1m$. Upon cooling, pristine $\text{Ca}_3\text{Ru}_2\text{O}_7$ undergoes two consecutive isostructural magnetic transitions: an A-type AFM- a ordering at $T_N = 56$ K, with FM alignment along the a axis within the RuO_2 bilayers, followed by a metamagnetic spin reorientation transition to the AFM- b phase at $T_s = 48$ K, stabilizing a polar metallic state with FM moments aligned along the b axis. In contrast, the 1% Ti-substituted compound $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ undergoes a first-order metal-to-insulator transition (MIT) concomitant with G-type AFM ordering, i.e., AFM alignment of the Ru magnetic moments both within and between the bilayers along the b axis (G-AFM- b), at $T_{\text{MIT}} = T_N = 55$ K. This transition is accompanied by significant disproportionate changes in the crystal lattice parameters, and the itinerant electronic behavior is replaced by charge localization coupled in a robust manner to the spin alignment.

The confluence of electron–electron and electron–lattice interactions renders the material extraordinarily sensitive to external perturbations such as light illumination [11]. An exceptionally shallow free-energy landscape of its G-AFM- b Mott insulator ground state, nearly degenerate with the metallic AFM- a , b state, highlights $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ as a unique model system for building and testing a theory of Mott transition dynamics in the presence of strong

*These authors contributed equally to this work.

†Contact author: A.Boris@fkf.mpg.de

electron–lattice coupling. Although the response to magnetic fields has been investigated extensively in $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$, reported H - T phase diagrams vary widely in complexity depending on the level of Ti substitution and, presumably, preparation details. Disentangling the roles of spin reorientation, lattice distortions, and electronic localization in the evolution and development of the various phases has proven challenging. Moreover, increasing the substitution of Ru ions above 1% reduces the effective Ru electronic bandwidth W so that the kinetic energy no longer competes with the onsite U , stabilizing the Mott-insulating state. The collective instabilities in the electronic, magnetic, and structural properties of these compounds, and their manifestations in charge dynamics, become less sensitive to external perturbations. Exploring the transport and magnetic properties of the $x = 1\%$ compound across an extended range of magnetic fields is therefore highly desirable to clarify the underlying physics.

From a technological perspective, the exceptional light sensitivity of the MIT suggests potential applications in designing nanodevices with quantum-level photosensitivity [11]. Recent studies have also demonstrated that the MIT in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ at heterointerfaces with the relativistic $5d$ Mott insulator Sr_2IrO_4 offers an effective method for tuning magnons, indicating a promising avenue for AFM magnonics [20]. Furthermore, AFM insulators with a preferred magnetization direction, such as $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$, are attractive alternatives to ferromagnets for spintronic applications, since they possess a field-switchable spin-flop transition that combines ultrafast dynamics with low magnetic susceptibility [21–24].

In $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$, the magnetic anisotropy is intrinsically linked to its electronic configuration: the Ru^{4+} ions adopt a low-spin $S = 1$ configuration, where the extended $4d$ orbitals, associated with a strong ligand field, stabilize a state dominated by spin–orbit coupling and crystal field effects. This pronounced anisotropy turns $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ into an ideal platform for field-controlled spin reorientation. While conventional anisotropic AFMs exhibit spin-flop transitions determined by a simple balance between exchange and anisotropy energies, in strongly correlated systems with magnetoelastic coupling, the physics is far richer, as spin reorientation, lattice distortion, and electronic localization are intertwined. This can potentially lead to a significant renormalization of the critical switching fields. This phenomenon is addressed in this work by systematically recording the H - T phase diagram of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ under fields up to 14 T applied along the in-plane crystallographic axes a and b , i.e., the magnetic hard and easy directions, respectively, and by comparing the magnetization and magnetic susceptibility behavior with transport data.

II. EXPERIMENTAL METHODS

High-quality single-domain crystals of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ were grown using the optical floating-zone technique and selected based on the reflectivity contrast in polarized-light optical microscopy. Careful sample selection avoided artifacts from multiphase behavior and phase separation that have been reported to affect the magnetic and transport properties of $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$

at higher Ti substitution levels [25–27]. High-resolution x-ray diffraction (XRD) and THz spectroscopy confirmed their phase purity and monodomain character [11,19]. Typical crystal dimensions were $2 \times 1 \times 0.2 \text{ mm}^3$. Magnetic phase transitions in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ were identified from anomalies in the temperature-dependent magnetic susceptibility $\chi(T)$ under various magnetic fields and/or in the isothermal magnetization $M(H)$ below $T_N = 55 \text{ K}$. Magnetization was measured up to 7 T with a Quantum Design SQUID magnetometer and up to 14 T with a Quantum Design physical properties measurement system (PPMS) equipped with a vibrating sample magnetometer (VSM) option. Similarly, electronic transitions were detected from anomalies in the resistivity recorded as functions of temperature and magnetic field. The in-plane resistivity was measured up to 14 T and down to 5 K on oriented samples contacted with conductive silver glue. Through sample rotation, the magnetic field was aligned with the crystal axes. To address uncertainties in absolute resistivity arising from anisotropy and possible current inhomogeneity, we employed both two- and four-terminal configurations. Unless stated otherwise, we report the normalized resistivity change $(\rho - \rho_0)/\rho_{\max}$, where ρ_0 and $\rho_{\max}$ are the zero-field and maximum resistivity, respectively. Low-frequency lock-in detection at 38 Hz was used to improve the signal-to-noise ratio and served as a proxy for the DC resistivity. To avoid current-induced phase transformations known from the monolayer member of the same Ruddlesden–Popper series [9,28,29], an AC excitation current as low as 200 nA was used. Samples were appropriately mounted to minimize strain and to avoid affecting the structural transition.

III. RESULTS AND DISCUSSION

A. Magnetic properties

The magnetic susceptibility $\chi(T)$ of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ exhibits a sharp drop with decreasing temperature below T_N , accompanied by pronounced thermal hysteresis. Figure 1 illustrates that the hysteresis loop shifts to lower temperatures with increasing magnetic field. When the magnetic field is aligned along the a axis [Fig. 1(a)], the transition temperature decreases quadratically with increasing field up to 14 T. A linear fit of T_N versus H^2 , shown in Fig. 1(b), yields

$$T_N(H) = T_N(0) - \alpha H^2, \quad (1)$$

with $T_N(0) = 55.4 \pm 0.1 \text{ K}$ and $\alpha = 0.127 \pm 0.004 \text{ K/T}^2$. This quadratic behavior reflects the competition between the Zeeman energy and exchange interactions in a collinear AFM, leading to a characteristic field-dependent suppression of the Néel temperature. Within a molecular-field approximation, the quadratic coefficient α is given by [30,31]

$$\alpha = \frac{3(2S^2 + 2S + 1)}{40(S + 1)^2} \left[\frac{\chi_N}{M_s} \right]^2 T_N(0), \quad (2)$$

where S is the spin quantum number, χ_N is the susceptibility at $T_N(0)$, and M_s is the saturation sublattice magnetization. Using $S = 1$, $\chi_N = 0.065 \text{ emu/Ru mol Oe}$ from Fig. 1(a), $M_s = 0.75 \mu_B/\text{Ru}$ [19,32], and $T_N = 55.4 \text{ K}$, we obtain $\alpha \approx 0.13 \text{ K/T}^2$. This is consistent with the value extracted from

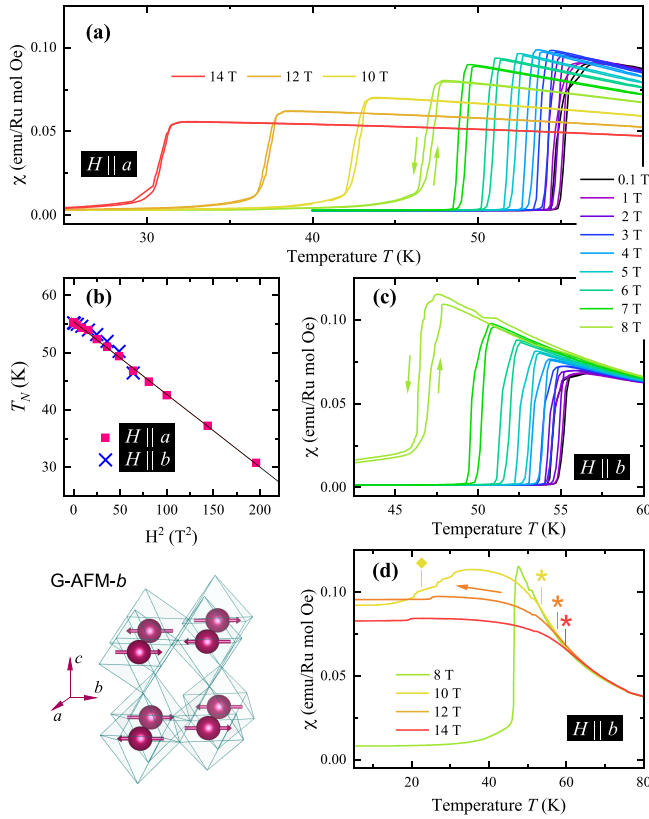


FIG. 1. Temperature dependence of magnetic susceptibility $\chi(T)$ of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ measured at different magnetic fields. (a) $\chi(T)$ measured at magnetic fields up to 14 T applied along the a axis. (b) The G-AFM to PM phase transition temperature from field-cooled (FC) $\chi(T)$ measurements plotted against magnetic field squared, with $H||a$ axis (red squares) and $H||b$ axis (blue crosses). (c), (d) $\chi(T)$ measured with $H||b$ axis at fields below (c) and above (d) 8 T. In (d), the diamond and stars correspond to the positions of the broad maximum and minima in the derivatives of FC $\chi(T)$, respectively. The image of Ru moments (red) surrounded by oxygen octahedra illustrates the magnetically ordered G-AFM- b state at zero field.

the experiment. By extrapolating $T_N(H^2)$ down to zero temperature, we find that the complete suppression of the AFM order requires a magnetic field of ≈ 20.9 T. Below T_N , both zero-field-cooled (ZFC) and field-cooled (FC) $\chi(T)$ curves [Fig. 1(a)] drop to a common minimum value, indicative of AFM ordering. This behavior is maintained up to 14 T for magnetic fields aligned along the a axis, without the emergence of new magnetic phases.

For fields applied along the b axis, the same behavior persists up to 7 T, as shown in Fig. 1(c) and denoted by crossed symbols in Fig. 1(b). At 8 T, however, the $\chi(T)$ develops a finite background below T_N , accompanied by an increasing disparity between the FC and ZFC measurements. The enhanced low-temperature $\chi(T)$ indicates the presence of uncompensated magnetic moments. Applying such a strong field competes with the AFM exchange interaction, promoting spin alignment along the field direction. As the magnetic field is increased further, the susceptibility cusp becomes progressively smeared out. Figure 1(d) thus suggests that the collapse

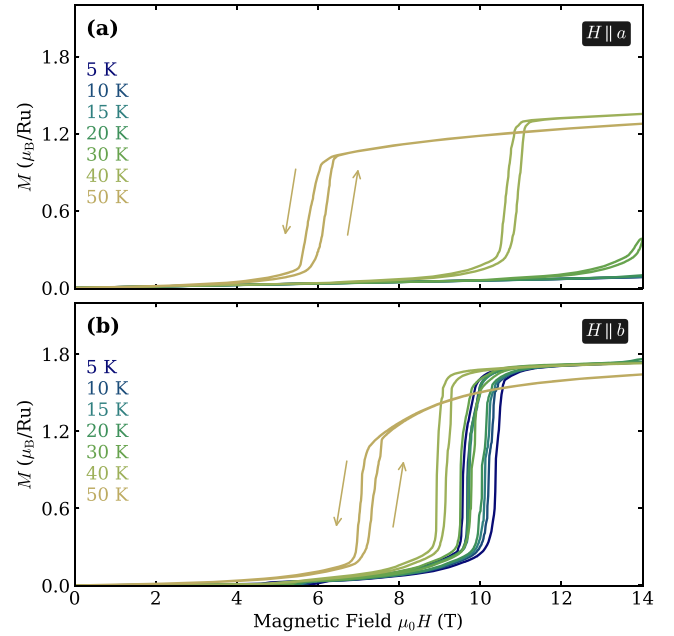


FIG. 2. Magnetization of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ as a function of the magnetic field, measured at temperatures below T_N . Magnetic fields up to 14 T are applied along the a axis (a) and b axis (b). In (b), the magnetic moment at 14 T for temperatures below 40 K reaches about $1.73 \mu_B/\text{Ru}$, indicating a trend towards saturation.

of the AFM order leads to a ferromagnetic-like state at fields above 10 T. This is indicative of the presence of a net magnetic moment, which will be discussed in the context of magnetization measurements in the following section. The crossover point, where thermal fluctuations become strong enough to cause a reduction of the net moment, is determined by extrema in the derivatives of the FC curves. These extrema are marked by star symbols in Fig. 1(d).

To unambiguously identify the nature of the observed magnetic ground states, we now examine the field-dependent magnetization $M(H)$. Figure 2(a) shows that at fields along the a axis and at low temperatures, $M(H)$ remains linear in the field up to 14 T. However, as the temperature approaches $T_N(0)$, a field-driven first-order transition emerges at ≈ 10.5 T and ≈ 6 T at 40 K and 50 K, respectively. After a steep increase of about $1 \mu_B/\text{Ru}$, $M(H)$ does not saturate but continues to rise in a nearly linear fashion. The critical magnetic fields observed during these field sweeps are in good agreement with the G-AFM to PM phase boundary described by the quadratic relationship in Eq. (1) that was derived for $\chi(T)$ in Fig. 1(b). So far, this behavior is fully consistent with the mean-field nature of the phase transition, indicating a conventional quantum critical point at a magnetic field of ≈ 20.9 T.

Conversely, at low temperatures, when the field is increased along the b axis, $M(H)$ exhibits a sharp jump with a clear trend towards saturation at high fields. Figure 2(b) shows that for temperatures below 40 K, the (nearly) saturated magnetic moment corresponds to $1.73 \mu_B$ per Ru at 14 T, which is close to the Ru magnetic moment in pristine $\text{Ca}_3\text{Ru}_2\text{O}_7$ [2,12,16]. Hence, we conclude that above the corresponding critical fields, H_b^{sat} , the G-AFM phase collapses into a forced

FM state, i.e., the fully saturated state of the PM phase. Notably, at 50 K, close to $T_N(0)$, $M(H)$ does not saturate above the critical field of ≈ 7 T, but closely resembles the behavior for fields along the a axis. Accordingly, this transition corresponds to the H - T phase line represented by the cross symbols in Fig. 1(b) along the quadratic field dependence of $T_N(H)$. We note that a similar field-induced transition was reported for $x = 3\%$ Ti-substituted $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$ at fields below H_b^{sat} [27], where incomplete saturation of the magnetic moment was attributed to a canted AFM structure comprising an AFM component along the a -axis and a FM component along the b axis. However, recent neutron diffraction measurements on the one $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ single crystal from the same batch as that used in the present study confirm nearly complete FM alignment of the Ru moments in the forced FM state above H_b^{sat} [32].

In a conventional AFM with uniaxial anisotropy, upon application of a magnetic field along the easy axis, i.e., parallel to the sublattice magnetic moments, the field-driven spin-flip transition from an AFM to a forced FM ground state is typically preceded by a spin-flop transition. This spin-flop transition is characterized by a sudden reorientation of the AFM sublattices perpendicular to the applied field, enabling a progressive canting towards full alignment as the field is increased further. To search for this intermediate spin-flop phase as the magnetic field increases along the easy b axis, before transitioning into the high-field forced FM state, we take a closer look at the low-temperature $M(H)$ for fields below 8 T. First, Fig. 3(a) shows that when the field is applied along the hard a axis, perpendicular to the AFM moments, it causes the Ru moments to cant in the field's direction. This results in a linear dependence of the magnetization, expressed as

$$M_i(H) = \beta_i H, \quad (3)$$

where the index i refers to the crystallographic direction, here the magnetic hard a axis and $\beta_a = (5.75 \pm 0.01) \times 10^{-3} \mu_B/\text{Ru T}^{-1}$ at $T = 5$ K.

In contrast, for the field along the easy b axis (parallel to the AFM moments), the net magnetization at first increases linearly but at a slow pace, with $\beta_b = (1.91 \pm 0.02) \times 10^{-3} \mu_B/\text{Ru T}^{-1}$ at $T = 5$ K, consistent with partial flips of the Ru moments that were initially antiparallel to the field. However, as the field is ramped up further, a hysteretic jump in the magnetization ΔM_b^f occurs above the critical field H_b^f , as shown in Fig. 3(b). At $T = 5$ K and $H_b^f \approx 6$ T, $\Delta M_b^f \approx 0.025 \mu_B/\text{Ru}$. Notably, after the abrupt increase, the magnetization value coincides with that obtained when the same field is applied along the a axis,

$$M_b(H) \approx M_a(H) + \Delta M_b^f \quad (4)$$

at $H \gtrsim H_b^f$, indicating a field-induced reorientation of the spin structure into a canted arrangement perpendicular to the field. This observed spin-flop transition is first-order and accompanied by a hysteresis that narrows progressively with increasing temperature as a result of thermal fluctuations. The phase boundary between the G-AFM- b ground and G-AFM- a spin-flop states can also be identified from the temperature dependencies of $\chi(T)$ measured at magnetic fields applied along the b -axis near but below H_b^f . Figure 3(c) shows that the ZFC and FC $\chi(T)$ curves diverge below the critical tempera-

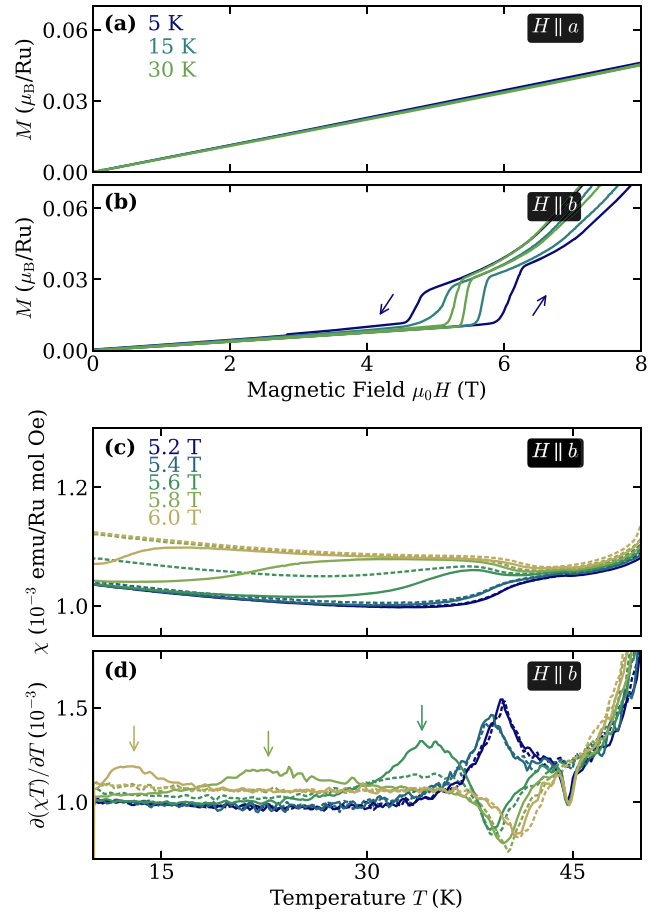


FIG. 3. (a), (b) Detailed view of the field-dependent magnetization of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ from Fig. 2 measured at low temperatures with fields applied along the a axis (a) and b axis (b). In (b), $M(H)$ measured along the easy b axis exhibits a sharp, step-like hysteretic change at the spin-flop transition field H_b^f , indicating the reorientation of the Ru moments from AFM order into a canted spin-flop state, perpendicular to the field. (c) Magnetic susceptibility $\chi(T)$ measured at magnetic fields near H_b^f applied along the b axis. The zero-field-cooled (ZFC, solid line) and field-cooled (FC, dashed lines) $\chi(T)$ curves diverge below the critical temperatures corresponding to the applied fields, as determined by the peaks in the derivatives of ZFC $\chi(T)$, indicated by arrows in (d).

tures corresponding to the applied fields. The phase boundary is then defined by the corresponding peak positions in the temperature-weighted derivatives of the ZFC $\chi(T)$, as indicated by arrows in Fig. 3(d). We point out that, in the case of a conventional anisotropic AFM, the specific heat is directly related to $\partial(T\chi)/\partial T$ up to a proportionality constant that only slowly varies with T [33].

Having identified all magnetic phases from the observed anomalies in $\chi(T)$ and $M(H)$ measurements, we can now compile the H - T phase diagram of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$. For the external field H applied parallel to the a axis, perpendicular to the low- T AFM moments, the phase diagram in Fig. 4(a) contains only two phases: an antiferromagnetic (G-AFM- b) and a paramagnetic (PM). When H is applied along the easy b axis, the phase transitions as a function of T (T_N) and H (H_b^f , H_b^{sat}) produce three distinct regions in Fig. 4(b)

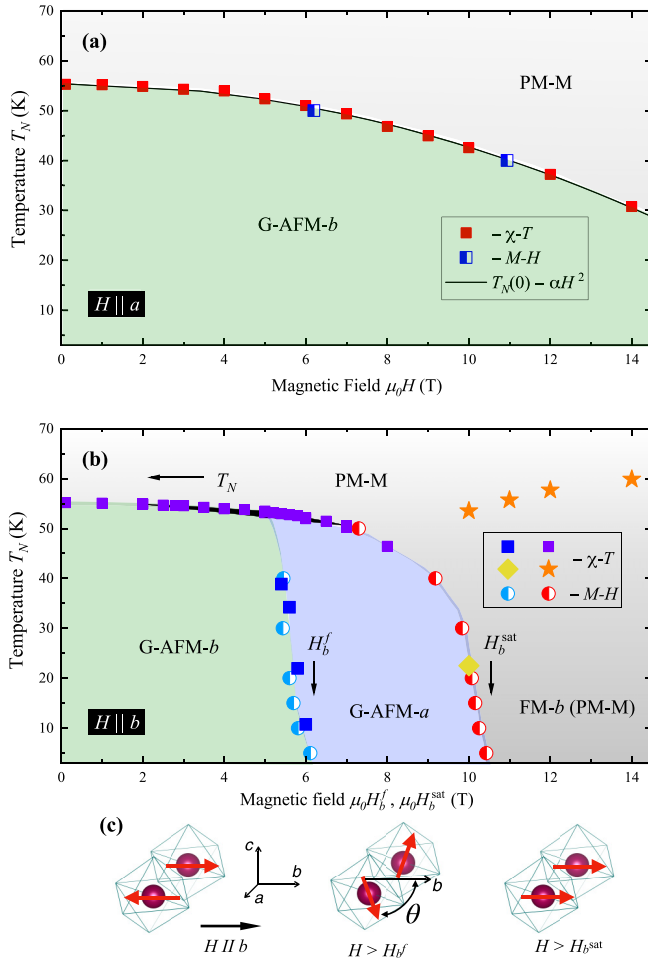


FIG. 4. Magnetic temperature–field (T - H) phase diagrams of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ with the magnetic field applied along the a axis (a) and b axis (b). The phase diagrams are derived from critical temperatures observed in temperature-dependent magnetic susceptibility (χ - T) as shown in Figs. 1 and 3(c), and 3(d) (filled symbols), and critical fields from the field-dependent magnetization (M - H) as shown in Figs. 2 and 3(a), and 3(b) (half-filled symbols). In (a), the critical T_N temperature for AFM ordering decreases quadratically with increasing field along the a axis. The critical field values correspond to temperature-driven transitions from the G-AFM- b to the paramagnetic PM-M state, indicating that the magnetic field along the a axis does not induce a new magnetic order in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$. (b) When the magnetic field is applied along the easy b axis, the low-temperature magnetization exhibits a first-order spin-flop transition at H_b^f , indicating the reorientation of Ru moments perpendicular to the field. Increasing the field above H_b^{sat} aligns all Ru moments with the b axis, resulting in a forced FM phase. With increasing T , the forced FM state is suppressed by thermal fluctuations, restoring PM behavior above the crossover temperatures depicted by star symbols, which correspond to the broad minima in the derivatives of FC $\chi(T)$ marked by stars in Fig. 1(d). The temperature variation of H_b^f and H_b^{sat} forms a typical phase diagram for an anisotropic AFM with the field parallel to the magnetization-easy axis. (c) Schematic illustration of the neighboring Ru magnetic moment alignment corresponding to the G-AFM- b ground state, spin-flop G-AFM- a phase, and field-forced FM- b phase, from left to right, respectively.

separated by phase boundaries: the G-AFM- b phase, the spin-flop G-AFM- a phase, and the forced ferromagnetic FM- b phase that evolves into the PM state at elevated temperatures. At zero field in the G-AFM- b state, magnetizations of the two equivalent Ru-moment sublattices are directed along the b axis, equal in magnitude and antiparallel. Above H_b^f , in the spin-flop G-AFM- a phase, the sublattice moments remain equal and nearly antiparallel but rotate to be nearly perpendicular to $H \parallel b$, forming equal, nonzero angles with H —an arrangement analogous to G-AFM- b for $H \parallel a$ in Fig. 4(a). The star symbols at high fields, derived from the $\chi(T)$ data in Fig. 1(d), mark the temperatures above which thermal fluctuations restore PM behavior in the forced FM- b state.

Magnetic phase boundaries in anisotropic Heisenberg AFMs are typically determined by minimizing the total energy, which reflects the competition among the AFM exchange, anisotropy, and Zeeman terms. Inspired by the experimentally determined phase diagram, reminiscent of that of a conventional anisotropic AFM, we consider a simple spin Hamiltonian, taking into account the bilayer nature of our system with isotropic Heisenberg intersite exchange and uniaxial single-ion anisotropy in a field,

$$\mathcal{H} = J_{\parallel} \sum_{\langle ij \rangle} [\tilde{S}_{1i} \cdot \tilde{S}_{1j} + \tilde{S}_{2i} \cdot \tilde{S}_{2j}] + J_{\perp} \sum_i \tilde{S}_{1i} \cdot \tilde{S}_{2i} - D \sum_i [(S_{1i}^b)^2 + (S_{2i}^b)^2] - \sum_i g\mu_B [\tilde{S}_{1i} + \tilde{S}_{2i}] \cdot \vec{H}, \quad (5)$$

where $\tilde{S}_{1i,2i}$ and $\tilde{S}_{1j,2j}$ are the spin operators for neighboring sites i and j in layers 1 and 2 of the bilayer, $J_{\parallel}$ and $J_{\perp}$ are positive in-plane and interlayer exchange coupling constants, respectively, and D is the orthorhombic single-ion anisotropy term for b -axis distortions to the local cubic crystal symmetry. When the external magnetic field is aligned with the hard a axis, the total magnetic energy per spin is

$$E(\theta) = -(2J_{\parallel} + \frac{1}{2}J_{\perp})S^2 \cos 2\theta - DS^2 \cos^2 \theta - g\mu_B H_a S \sin \theta. \quad (6)$$

Here, θ is the symmetric canting angle of the Ru sublattice moments away from the easy b axis toward $\vec{H} \parallel a$. As H_a increases, this canting yields a gradual increase of the net magnetization described by Eq. (3) with $\beta_a = g\mu_B S/H_a^{\text{sat}}$ up to saturation at

$$g\mu_B H_a^{\text{sat}} = 2S(4J_{\parallel} + J_{\perp} + D). \quad (7)$$

When the field is applied along the easy b axis, the two critical fields are determined by

$$g\mu_B H_b^f = 2S\sqrt{D(4J_{\parallel} + J_{\perp} - D)} \quad (8)$$

and

$$g\mu_B H_b^{\text{sat}} = 2S(4J_{\parallel} + J_{\perp} - D). \quad (9)$$

At low H_b , the AFM state persists up to H_b^f , where it transforms into the spin-flop state with a magnetization jump

$$\Delta M_b^f = g\mu_B S \sqrt{D/(4J_{\parallel} + J_{\perp} - D)}. \quad (10)$$

Further increase of H_b leads to a gradual canting of the Ru moments, until the forced FM state is reached above H_b^{sat} .

An analysis of the magnetic inelastic neutron scattering excitation spectra measured along the high-symmetry directions of the $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ lattice at 5 K yields $J_{\parallel} = 4.5$ meV, $J_{\perp} = 3.0$ meV, and $D = 4.5$ meV [34,35]. Although the applied Heisenberg model with these parameters, $S = 1$, and $g = 2$ reasonably reproduces the low-temperature, weak-field linear magnetization $M_a(H)$ —yielding $\beta_a \approx 4.6 \times 10^{-3} \mu_B/\text{Ru T}^{-1}$ vs the measured $(5.75 \pm 0.01) \times 10^{-3} \mu_B/\text{Ru T}^{-1}$ —it overestimates the critical fields by more than an order of magnitude: 149 T, 285 T, and 440 T compared with the experimentally extracted values of 6 T, 10.5 T, and 20.9 T for H_b^f , H_b^{sat} , and H_a^{sat} , respectively. Such a significant discrepancy indicates a substantial divergence from pure Heisenberg physics in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$. This is supported by the first-order phase boundaries between the PM and ordered AFM states. Unlike canonical AFMs, which exhibit continuous (second-order) PM–AFM transitions [36], $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ undergoes a first-order magnetoelastic phase transition with pronounced hysteresis and phase coexistence. This indicates the presence of strong elastic strain due to disproportionate changes in the lattice parameters at T_N . Strong spin–phonon coupling can substantially weaken long-range Néel order in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$. Cooperative electron–electron and electron–lattice interactions with coupled structural and magnetic order parameters have also been brought into connection with the compound’s exceptional sensitivity to light [11]. In our model Hamiltonian [Eq. (5)], the strong spin–phonon coupling has not been incorporated. This can be accomplished by augmenting the bond-dependent exchange and by including explicit spin–phonon terms [37,38]; however, this is beyond the scope of this experimental work. For the sake of completeness, we mention the prominent example of the manganites, where a strong renormalization of the electronic and magnetic properties has also been reported. There it has been attributed to the combined effect of strong electron–phonon coupling and the double exchange mechanism between itinerant electrons and localized spins. Together, these interactions give rise, for instance, to colossal magnetoresistance and field-driven metal–insulator transitions of enormous magnitude [39]. This warrants a closer look at the magnetotransport properties of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ and a comparison with the rich magnetic phase diagram established above.

In the next section, we show that $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ remains insulating through the spin-flop transition at H_b^f but becomes metallic in the field-induced, saturated FM state above H_b^{sat} . This insulator-to-metal transition indicates that electron itinerancy and low-energy electron–hole excitations—features beyond a minimal Heisenberg spin-only model—also become relevant near the field-driven phase boundary.

B. Magnetoresistance properties

In the absence of a magnetic field, Ti-substituted $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$ undergoes a sharp MIT at T_N upon cooling. The in-plane resistivity increases by three to four orders of magnitude [17,18]. The field dependence of the MIT follows the H – T magnetic phase diagram and gives rise to the colossal

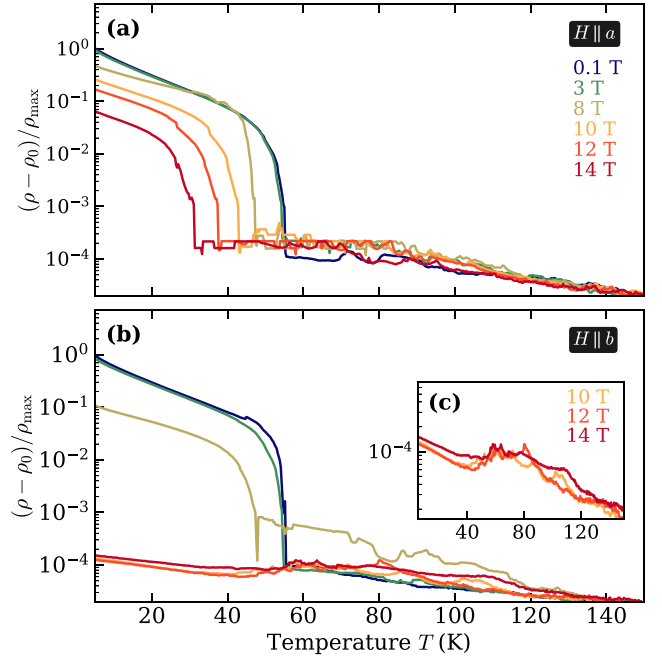


FIG. 5. Temperature dependence of the normalized in-plane, low-frequency (38 Hz) resistivity $(\rho - \rho_0)/\rho_{\text{max}}$ of $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ measured for different magnetic fields. Fields up to 14 T are applied along the a axis (a) and b axis (b). (c) Magnified view for fields ≥ 10 T along the b axis.

magnetoresistance observed at field-induced phase transitions [27], though a reliable interpretation of the emerging phases requires minimal Ti substitution, as higher x can lead to phase separation that complicates the analysis [25].

Therefore, we focus here on $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ as such samples yield a simple H – T phase diagram and show no signs of phase separation. Its Mott-insulating ground state is achieved by finely tuning the one-electron bandwidth W through only 1% substitution on the Ru site. The clean optical gap of 0.45 eV is sharply defined and entirely free of defect-induced excitations, as confirmed across the full spectral range down to 1 meV [11]. The collapse of the Mott state above T_N gives rise to a metallic Drude response with a plasma frequency as large as 2.8 eV, reflecting a substantial itinerant charge carrier density.

Figure 5 shows FC temperature dependencies of the normalized in-plane resistivity $(\rho - \rho_0)/\rho_{\text{max}}$ for different representative magnetic fields. The data set illustrates that an applied magnetic field lowers the temperature at which the metal–insulator transition, signaled by a sharp resistivity increase, occurs. The temperature change of the MIT follows the quadratic relation of Eq. (1) describing the field-induced suppression of the Néel temperature. The steep MIT-related resistivity upturn is present for all fields up to 14 T when the field is applied along the a axis [Fig. 5(a)]. When the field is aligned with the easy b axis, the MIT follows the same trend for low magnetic fields, but it disappears for fields above 10 T [Figs. 5(b) and 5(c)]. Hence, $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ remains in the conductive PM phase at all temperatures when the field is applied along the easy b -axis and exceeds the critical field value H_b^{sat} . In this high-field regime, the temperature-driven

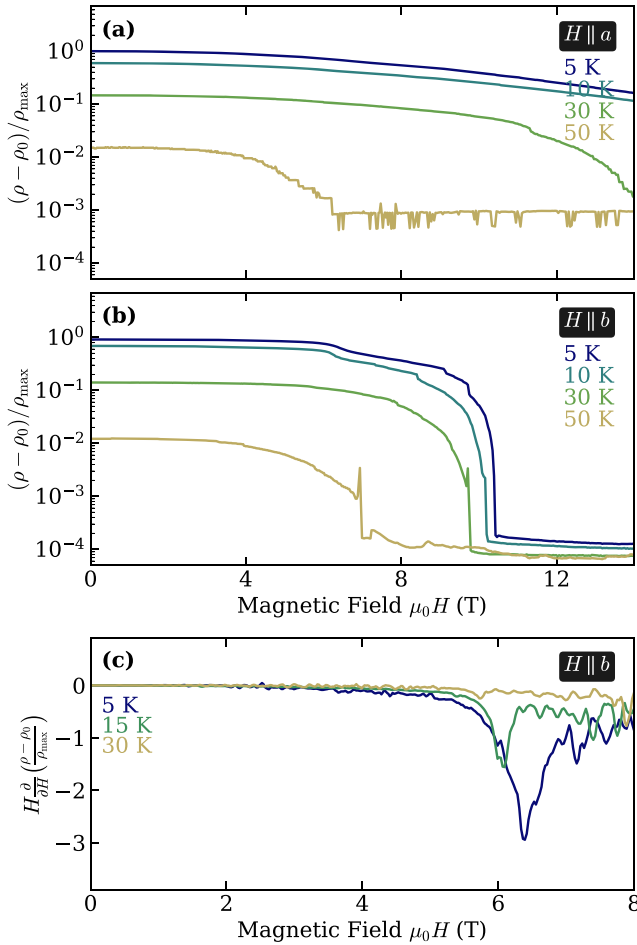


FIG. 6. Magnetic-field dependence of the normalized in-plane, low-frequency (38 Hz) resistivity $(\rho - \rho_0)/\rho_{\max}$ measured at temperatures below T_N with fields applied along the a axis (a) and b axis (b). (c) Field-weighted derivative $H \partial((\rho - \rho_0)/\rho_{\max})/\partial H$ for the field applied along the b axis. The minimum in the derivative corresponds to a slight drop in in-plane resistivity as a canted FM moment develops along the b axis at the field-induced spin-flop transition above H_b^f .

crossover from the field-forced FM state to the magnetically disordered state is accompanied by fluctuations in the resistivity. We attribute these to the formation and growth of ferromagnetic domains as domain walls are bound to alter the microscopic spatial current distribution.

We also examined the isothermal field dependence of the in-plane resistivity to continue mapping the electronic MIT onto the H - T magnetic phase diagram in Fig. 4. Figure 6 shows ZFC curves of the normalized in-plane resistivity $(\rho - \rho_0)/\rho_{\max}$ versus magnetic field at selected temperatures below T_N . The negative magnetoresistance across the field-driven transition from the G-AFM to PM state closely tracks the field-dependent magnetization in Fig. 2. For fields up to 14 T along the a axis in Fig. 6(a), the highly conductive regime appears only near T_N . In contrast, for fields along the easy b axis, the normalized resistivity in Fig. 6(b) drops by two to four orders of magnitude in the forced FM state above H_b^{sat} , while the spin-flop phase for $(H_b^f < H < H_b^{\text{sat}})$ remains

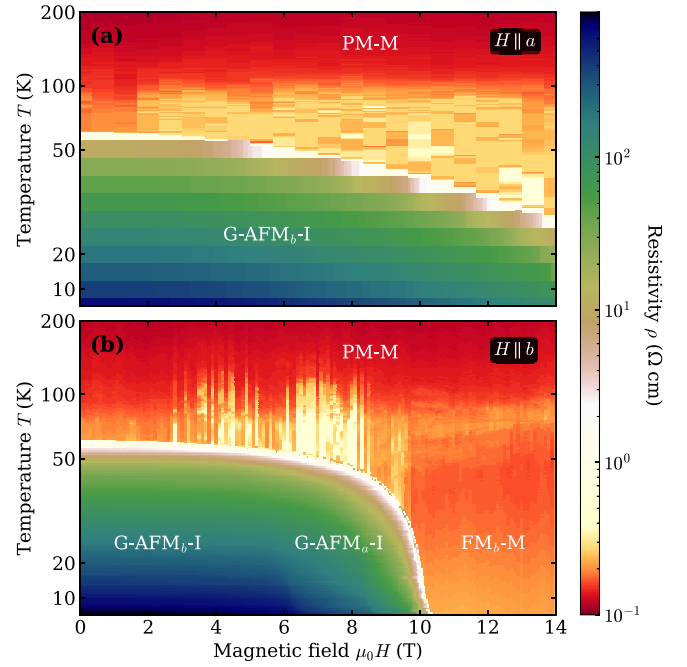


FIG. 7. Color maps of the in-plane resistivity ρ as functions of temperature T and magnetic field H , with H applied along the a axis (a) and b axis (b). Low-frequency (38 Hz) resistivity data are stitched from measurement protocols optimized for the insulating (green) and metallic (orange) regions: in the insulating regime we use $\rho(H)$ field sweeps at fixed T ; in the metallic regime we use $\rho(T)$ sweeps at fixed H , emphasizing phase-specific features.

insulating. Across the spin-flop transition we find only small yet well-reproducible step-like resistivity drops, which correspond to minima in the derivative of the field-dependent normalized resistivity at H_b^f [Fig. 6(c)].

In order to compile a more detailed phase diagram, the in-plane resistivity was measured as a function of both magnetic field and temperature at fine intervals and compiled into the detailed color map in Figs. 7(a) and 7(b). Sharply delineating the MIT, the resistivity color plot traces the phase boundary between the G-AFM and PM phases in the H - T magnetic phase diagrams constructed from the $\chi(T)$ and $M(H)$ data in Figs. 4(a) and 4(b), respectively. Unlike the magnetization and magnetic susceptibility, the magnetoresistance is not a purely thermodynamic quantity and is sensitive to microscopic details and local features. Its behavior can, for instance, depend on the magnetic domain structure, the domain boundaries, and defect-related pinning. Therefore, the magnetoresistance can reveal complementary information that is absent in the magnetic data. In particular, in the metallic phase, as the temperature decreases at constant field, the resistivity increases and pronounced irregularities emerge upon approaching the MIT. Such electrical instabilities become most pronounced near the critical point around 8 T along the b axis, where multiple phase boundary lines converge. These instabilities can be associated with short-range AFM correlations persisting above T_N in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ across the broad T - H phase region near the critical point.

Another notable feature appears in Fig. 7(b) for fields above the critical H_b^{sat} . A distinct boundary on the resistivity

color plot marks the crossover between the field-forced FM state and the high-temperature PM behavior restored by thermal fluctuations, coinciding with the line traced by the star symbols in Fig. 4(b). As illustrated in Fig. 5(c), this phase region also exhibits electrical instabilities, here associated with the dynamics of FM domains. Counterintuitively, the saturated FM state is more resistive than the high-temperature magnetically disordered state, consistent with the overall decrease of resistivity with increasing temperature seen in Fig. 5(c). A similar trend is observed in pristine $\text{Ca}_3\text{Ru}_2\text{O}_7$, where the field-forced FM state shows higher resistivity than the PM state, attributed to the suppression of interbilayer hopping governed by orbital degrees of freedom in the fully polarized FM configuration [40]. This effect stems from the bilayer ruthenate structure, where spin-orbit coupling causes FM alignment to hinder electron hopping more effectively than PM disorder.

In contrast to the significant resistance change observed for the crossover between the field-forced FM state and PM state restored by thermal fluctuations at higher temperatures, only a slight drop in resistivity is observed across the spin-flop transition [Figs. 6(b) and 6(c)], where a canted FM moment emerges above H_b^f [Fig. 3(b)]. The net FM component along the b axis partially aligns neighboring Ru spins across bilayers, reducing spin scattering. Virtual interlayer hopping enhances Ru $4d$ electron itinerancy via a double-exchange-like mechanism, without being subject to the orbital selection rules that suppress hopping in the fully polarized FM state. The normalized resistivity decreases by less than 5%, a change barely discernible in the resistivity color map [green region, Fig. 7(b)]. Both the G-AFM- b ground state and the spin-flop G-AFM- a state remain Mott insulating. The extreme renormalization of the critical field H_b^f relative to the prediction of the minimal Heisenberg spin-only model [Eq. (5)] is therefore primarily attributed to strong spin-phonon coupling. Unlike the G-AFM-I to PM-M transition, the role of electron itinerancy and low-energy electron-hole excitations remains negligible.

IV. CONCLUSIONS

We have systematically investigated the magnetic and electronic phase transitions in bilayer perovskite

$\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ in the presence of in-plane magnetic fields up to 14 T. Using anomalies in $\chi(T)$ under various fields, isothermal $M(H)$ traces below $T_N = 55$ K, as well as temperature- and field-dependent resistivity data, we have constructed the H - T phase diagram of this compound. With the field along the easy b axis, $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ exhibits a first-order spin-flop transition at $H_b^f \approx 6$ T, while retaining its Mott insulating state across the transition. Subsequently, a transition to a forced FM metallic phase occurs above $H_b^{\text{sat}} \approx 10.5$ T. When the field is applied along the hard a axis, neither the spin-flop nor the forced FM phase emerges below 14 T. In the presence of strong electron-lattice coupling, $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ undergoes the first-order magnetoelastic G-AFM-I to PM-M transitions, yet it exhibits the magnetic H - T phase diagram characteristic of canonical anisotropic AFMs. The critical fields H_b^f and H_b^{sat} are hence suppressed by more than an order of magnitude relative to the predictions of a minimal Heisenberg spin-only model that accounts for the exchange interactions in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$. We anticipate that these results will stimulate further theoretical studies to quantitatively describe the magnetic phase diagram of Mott AFMs in the presence of strong electron-lattice coupling. On the experimental side, direct probes are necessary to detail the impact of magnetic fields on structural distortions in $\text{Ca}_3(\text{Ru}_{0.99}\text{Ti}_{0.01})_2\text{O}_7$ and precisely quantify magnetoelastic effects at the phase boundaries.

ACKNOWLEDGMENTS

We gratefully acknowledge A. N. Yaresko, H. Gretarsson, A. Kirilyuk, G. Khaliullin, and L. Wang for fruitful discussions. This work was supported by the European Research Council under Advanced Grant No. 101141844 (SpecTera).

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

-
- [1] G. Cao, C. Alexander, S. McCall, J. Crow, and R. Guertin, From antiferromagnetic insulator to ferromagnetic metal: A brief review of the layered ruthenates, *Mater. Sci. Eng. B* **63**, 76 (1999).
 - [2] G. Cao, S. McCall, J. E. Crow, and R. P. Guertin, Observation of a metallic antiferromagnetic phase and metal to nonmetal transition in $\text{Ca}_3\text{Ru}_2\text{O}_7$, *Phys. Rev. Lett.* **78**, 1751 (1997).
 - [3] S.-I. Ikeda, Y. Maeno, S. Nakatsuji, M. Kosaka, and Y. Uwatoko, Ground state in $\text{Sr}_3\text{Ru}_2\text{O}_7$: Fermi liquid close to a ferromagnetic instability, *Phys. Rev. B* **62**, R6089 (2000).
 - [4] Y. Maeno, T. M. Rice, and M. Sigrist, The intriguing superconductivity of strontium ruthenate, *Phys. Today* **54**(1), 42 (2001).
 - [5] G. Cuono, R. M. Sattigeri, J. Skolimowski, and C. Autieri, Orbital-selective altermagnetism and correlation-enhanced spin-splitting in strongly-correlated transition metal oxides, *J. Magn. Magn. Mater.* **586**, 171163 (2023).
 - [6] A. León, C. Autieri, T. Brumme, and J. W. González, Hybrid d/p-wave altermagnetism in $\text{Ca}_3\text{Ru}_2\text{O}_7$ and strain-controlled spin splitting, *npj Quantum Mater.* **10**, 98 (2025).
 - [7] C. S. Snow, S. L. Cooper, G. Cao, J. E. Crow, H. Fukazawa, S. Nakatsuji, and Y. Maeno, Pressure-tuned collapse of the Mott-like state in $\text{Ca}_{n+1}\text{Ru}_n\text{O}_{3n+1}$ ($n = 1, 2$): Raman spectroscopic studies, *Phys. Rev. Lett.* **89**, 226401 (2002).
 - [8] J. Bertinshaw, N. Gurung, P. Jorba, H. Liu, M. Schmid, D. T. Mantadakis, M. Daghofer, M. Krautloher, A. Jain, G. H. Ryu, O. Fabelo, P. Hansmann, G. Khaliullin, C. Pfleiderer, B. Keimer, and B. J. Kim, Unique crystal structure of Ca_2RuO_4 in the current stabilized semimetallic state, *Phys. Rev. Lett.* **123**, 137204 (2019).

- [9] J. Zhang, A. S. McLeod, Q. Han, X. Chen, H. A. Bechtel, Z. Yao, S. N. G. Corder, T. Ciavatti, T. H. Tao, M. Aronson, G. L. Carr, M. C. Martin, C. Sow, S. Yonezawa, F. Nakamura, I. Terasaki, D. N. Basov, A. J. Millis, Y. Maeno, and M. Liu, Nano-resolved current-induced insulator-metal transition in the Mott insulator Ca_2RuO_4 , *Phys. Rev. X* **9**, 011032 (2019).
- [10] C. T. Suen, I. Marković, M. Zonno, N. Heinsdorf, S. Zhdanovich, N. H. Jo, M. Schmid, P. Hansmann, P. Puphal, K. Fürsich, *et al.*, Electronic response of a Mott insulator at a current-induced insulator-to-metal transition, *Nat. Phys.* **20**, 1757 (2024).
- [11] K. S. Rabinovich, A. N. Yaresko, R. D. Dawson, M. J. Krautloher, T. Priessnitz, Y. Mathis, A. Kirilyuk, B. Keimer, and A. V. Boris, Optically driven dynamics of a Mott insulator-to-metal transition, *Adv. Funct. Mater.* **35**, 2416597 (2025).
- [12] W. Bao, Z. Q. Mao, Z. Qu, and J. W. Lynn, Spin valve effect and magnetoresistivity in single crystalline $\text{Ca}_3\text{Ru}_2\text{O}_7$, *Phys. Rev. Lett.* **100**, 247203 (2008).
- [13] B. Bohnenbuck, I. Zegkinoglou, J. Strempler, C. Schüller-Langeheine, C. S. Nelson, P. Leininger, H.-H. Wu, E. Schierle, J. C. Lang, G. Srajer, S. I. Ikeda, Y. Yoshida, K. Iwata, S. Katano, N. Kikugawa, and B. Keimer, Magnetic structure and orbital state of $\text{Ca}_3\text{Ru}_2\text{O}_7$ investigated by resonant x-ray diffraction, *Phys. Rev. B* **77**, 224412 (2008).
- [14] D. Sokolov, N. Kikugawa, T. Helm, H. Borrmann, U. Burkhardt, R. Cubitt, J. White, E. Ressouche, M. Bleuel, K. Kummer, *et al.*, Metamagnetic texture in a polar antiferromagnet, *Nat. Phys.* **15**, 671 (2019).
- [15] I. Marković, M. D. Watson, O. J. Clark, F. Mazzola, E. A. Morales, C. A. Hooley, H. Rosner, C. M. Polley, T. Balasubramanian, S. Mukherjee, *et al.*, Electronically driven spin-reorientation transition of the correlated polar metal $\text{Ca}_3\text{Ru}_2\text{O}_7$, *Proc. Natl. Acad. Sci. USA* **117**, 15524 (2020).
- [16] Q. Faure, C. D. Dashwood, C. V. Colin, R. D. Johnson, E. Ressouche, G. B. G. Stenning, J. Spratt, D. F. McMorro, and R. S. Perry, Magnetic structure and field dependence of the cycloid phase mediating the spin reorientation transition in $\text{Ca}_3\text{Ru}_2\text{O}_7$, *Phys. Rev. Res.* **5**, 013040 (2023).
- [17] X. Ke, J. Peng, D. J. Singh, T. Hong, W. Tian, C. R. Dela Cruz, and Z. Q. Mao, Emergent electronic and magnetic state in $\text{Ca}_3\text{Ru}_2\text{O}_7$ induced by Ti doping, *Phys. Rev. B* **84**, 201102(R) (2011).
- [18] S. Tsuda, N. Kikugawa, K. Sugii, S. Uji, S. Ueda, M. Nishio, and Y. Maeno, Mott transition extremely sensitive to impurities in $\text{Ca}_3\text{Ru}_2\text{O}_7$ revealed by hard x-ray photoemission studies, *Phys. Rev. B* **87**, 241107(R) (2013).
- [19] M. Krautloher, Neutron scattering studies on layered ruthenates, Ph.D. thesis, Universität Stuttgart, 2018.
- [20] S. Shrestha, M. Souiri, C. J. Dietl, E. M. Pärskke, M. Krautloher, G. A. C. Ortiz, M. Minola, X. Shi, A. V. Boris, J. Hwang, *et al.*, Tunable magnons of an antiferromagnetic Mott insulator via interfacial metal-insulator transitions, *Nat. Commun.* **16**, 3592 (2025).
- [21] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, Antiferromagnetic spintronics, *Rev. Mod. Phys.* **90**, 015005 (2018).
- [22] O. Gomonay, V. Baltz, A. Brataas, and Y. Tserkovnyak, Antiferromagnetic spin textures and dynamics, *Nat. Phys.* **14**, 213 (2018).
- [23] N. O. Antropov, E. A. Kravtsov, M. V. Makarova, V. V. Proglyado, T. Keller, I. A. Subbotin, E. M. Pashaev, G. V. Prutskov, A. L. Vasiliev, Y. M. Chesnokov, N. G. Bebenin, M. A. Milyaev, V. V. Ustinov, B. Keimer, and Y. N. Khaydukov, Tunable spin-flop transition in artificial ferrimagnets, *Phys. Rev. B* **104**, 054414 (2021).
- [24] H. Chen, L. Liu, X. Zhou, Z. Meng, X. Wang, Z. Duan, G. Zhao, H. Yan, P. Qin, and Z. Liu, Emerging antiferromagnets for spintronics, *Adv. Mater.* **36**, 2310379 (2024).
- [25] J. Peng, J. Y. Liu, X. Gu, G. Zhou, W. Wang, J. Hu, F. M. Zhang, and X. S. Wu, Extremely large anisotropic transport caused by electronic phase separation in Ti-doped $\text{Ca}_3\text{Ru}_2\text{O}_7$, *J. Phys. D: Appl. Phys.* **49**, 245004 (2016).
- [26] J. Peng, J. Y. Liu, J. Hu, Z. Q. Mao, F. M. Zhang, and X. S. Wu, Magnetic phase separation in double layer ruthenates $\text{Ca}_3(\text{Ru}_{1-x}\text{Ti}_x)_2\text{O}_7$, *Sci. Rep.* **6**, 19462 (2016).
- [27] M. Zhu, J. Peng, T. Zou, K. Prokes, S. D. Mahanti, T. Hong, Z. Q. Mao, G. Q. Liu, and X. Ke, Colossal magnetoresistance in a Mott insulator via magnetic field-driven insulator-metal transition, *Phys. Rev. Lett.* **116**, 216401 (2016).
- [28] F. Nakamura, M. Sakaki, Y. Yamanaka, S. Tamaru, T. Suzuki, and Y. Maeno, Electric-field-induced metal maintained by current of the Mott insulator Ca_2RuO_4 , *Sci. Rep.* **3**, 2536 (2013).
- [29] R. Okazaki, Y. Nishina, Y. Yasui, F. Nakamura, T. Suzuki, and I. Terasaki, Current-induced gap suppression in the Mott insulator Ca_2RuO_4 , *J. Phys. Soc. Jpn.* **82**, 103702 (2013).
- [30] P. Heller, Nuclear-magnetic-resonance studies of critical phenomena in MnF_2 . I. Time-average properties, *Phys. Rev.* **146**, 403 (1966).
- [31] Y. Shapira and S. Foner, Magnetic phase diagram of MnF_2 from ultrasonic and differential magnetization measurements, *Phys. Rev. B* **1**, 3083 (1970).
- [32] J. Bertinshaw, M. L. Krautloher, L. Wang, H. Trepka, J. Porras, M. Hepting, S. Francoual, E. Ressouche, B. J. Kim, and B. Keimer, Extreme sensitivity to perturbation in Ti-doped $\text{Ca}_3\text{Ru}_2\text{O}_7$ (unpublished).
- [33] M. E. Fisher, Relation between the specific heat and susceptibility of an antiferromagnet, *Philos. Mag.* **7**, 1731 (1962).
- [34] M. J. Krautloher, J. Bertinshaw, J. Porras, D. G. Joshi, H. Trepka, M. Hepting, D. Adroja, H. Walker, A. Ivanov, M. Enderle, *et al.*, Impurity induced localization of low-energy spin excitations in bilayer ruthenate $\text{Ca}_3\text{Ru}_2\text{O}_7$ (unpublished).
- [35] J. Bertinshaw, H. Suzuki, H. Takahashi, M. Krautloher, K. S. Rabinovich, G. Khaliullin, A. V. Boris, H. Gretarsson, and B. Keimer, The emergence of spin-orbit excitations in Ti-doped $\text{Ca}_3\text{Ru}_2\text{O}_7$ (unpublished).
- [36] A. R. King and H. Rohrer, Spin-flop bicritical point in MnF_2 , *Phys. Rev. B* **19**, 5864 (1979).
- [37] K. Aoyama and H. Kawamura, Spin-lattice-coupled order in Heisenberg antiferromagnets on the pyrochlore lattice, *Phys. Rev. Lett.* **116**, 257201 (2016).
- [38] J. S. and D. Bansal, Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO , *Phys. Rev. B* **111**, 104306 (2025).
- [39] Y. Tokura, Critical features of colossal magnetoresistive manganites, *Rep. Prog. Phys.* **69**, 797 (2006).
- [40] X. N. Lin, Z. X. Zhou, V. Durairaj, P. Schlottmann, and G. Cao, Colossal magnetoresistance by avoiding a ferromagnetic state in the Mott system $\text{Ca}_3\text{Ru}_2\text{O}_7$, *Phys. Rev. Lett.* **95**, 017203 (2005).