I. INTRODUCTION

Critical fluctuations of the order parameter emerge in proximity to the transition temperature $T_c$ of second-order phase transitions. These fluctuations are characterized by a correlation length $\xi$ and a response time $\tau$, which diverge at $T_c$ [1–4]. In the critical regime close to $T_c$, fundamental physical properties of a material, such as the magnetic susceptibility and the heat capacity, adopt critical behavior and can be described by power-laws $\propto |t|^\lambda$, with critical exponents $\lambda$ and $t \equiv (T/T_c - 1)$ [2–6]. Furthermore, the scaling behaviors in the spatial and time domains are related via $\tau \propto \xi^\nu$, containing $\nu$, $\gamma$, and $\xi$ respectively. We find that the critical behaviors of the single-layer compound Ca$_2$RuO$_4$ follow universal scaling laws that are compatible with predictions of the 2D-XY model. The bilayer compound Ca$_3$Ru$_2$O$_7$ is only partly consistent with the 2D-XY theory and best described by the three-dimensional Ising (3D-I) model, which is likely a consequence of the intra-bilayer exchange interactions in combination with an orthorhombic single-ion anisotropy. Hence, our results suggest that layered ruthenates are promising solid-state platforms for research on the 2D-XY model and the effects of 3D interactions and additional spin-space anisotropies on the magnetic fluctuations.

A key experimental technique for the investigation of critical magnetic scattering is neutron triple-axis spectroscopy (TAS), which exploits the proportionality between the magnetic neutron scattering cross section and the dynamic scattering function $S(q, \omega)$, containing $\kappa$ and $\Gamma$ [21–23]. More specifically, $\Gamma$ can be derived from TAS energy scans of the critical magnetic scattering, while $\kappa$ corresponds to the energy-integrated $Q$-width in momentum space. Along these lines, pioneering studies investigated the critical magnetic fluctuations in classical magnetic systems, such as the 3D ferromagnet (FM) EuO [24–26] and the 3D antiferromagnet (AFM) RbMnF$_3$ [27, 28]. Furthermore, TAS studies were carried out on systems with quasi-2D magnetic correlations, including the isotropic square-lattice AFMs Rb$_2$MnF$_4$ [29], Sr$_2$CuO$_2$Cl$_2$, and Sr$_2$Cu$_3$O$_2$Cl$_2$ [30], as well as the AFM parent compounds of the cuprate superconductors [31], which exhibit 2D Heisenberg (2D-H) scaling properties above their Neél temperatures. More recently, critical magnetic fluctuations were investigated in 5d-electron transition metal oxides (TMOS) using X-ray scattering. In single-layer Sr$_3$Ir$_2$O$_7$, which exhibits a Mott-insulating AFM ground state with a $J_{eff} = 1/2$ effective total angular momentum due to strong spin-orbit coupling (SOC) [32, 33], 2D-H scaling with a small easy-plane anisotropy was reported [34, 35]. On the other hand, in bilayer Sr$_3$Ir$_2$O$_7$ the scaling behavior close to the transition is consistent with the 3D Ising (3D-I) universality class, but
significant deviations were found and attributed to disorder [36].

In 4d-electron TMOs, such as single- and bilayer ruthenates, critical fluctuations have remained unexplored to date. Notably, ruthenates show a plethora of electronic ground states [37–41] such as unconventional superconductivity in Sr₂RuO₄ [42] and excitonic AFM order in the Mott insulator Ca₃RuO₄ [43, 44], arising from a delicate competition between the energy scales of SOC, crystal field splitting, Hund’s coupling, and intersite exchange interactions. In the latter compound, spins are arranged in an AFM fashion within square-lattice RuO₂ planes and stacked along the c-axis in a G-type pattern [Fig. 1a] with a Néel temperature $T_N \sim 110$ K [45–47]. The excitonic character is believed to result from excitonic transitions between non-magnetic singlet ($J_{eff}=0$) and magnetic triplet states ($J_{eff}=1$) [43, 44]. The nature of the excitonic magnetism was recently corroborated by resonant inelastic x-ray scattering (RIXS) [48], Raman scattering [49], as well as inelastic neutron scattering (INS), detecting a soft amplitude mode (‘Higgs-mode’) in the spin-wave spectrum [50].

The unquenched orbital angular momentum of the Ru magnetic moments in Ca₂RuO₄ further results in a highly unusual spectrum of transverse magnons in the AFM state [50]. The low-energy magnetic Hamiltonian derived from an analysis of this spectrum is dominated by an XY-type single-ion anisotropy, which is much larger than the nearest-neighbor exchange interaction and an Ising-type single-ion anisotropy resulting from an orthorhombic distortion of the crystal structure. At the same time, the INS experiments did not reveal any dispersion of the magnons perpendicular to the RuO₂ layers, which implies that the interlayer interactions are much weaker than the interactions within the layers. The evidence for an approximate 2D-XY symmetry of the magnetic Hamiltonian derived from the analysis of the magnon dispersions has motivated the present study.

In contrast to the Mott insulator Ca₃RuO₄, the bilayer compound Ca₃Ru₂O₇ is metallic in the paramagnetic state and maintains considerable electrical conductivity below the Néel temperature $T_{N,1} \sim 56$ K [51]. The magnetic structure is A-type AFM (i.e., FM bilayers with alternating orientation along the c-axis) [Fig. 1d] [51, 52]. A second magnetic transition associated with a reorientation of the spins from the a- to the b-axis in the RuO₂ planes [53] and a greater reduction of the electrical conductivity occurs at $T_{N,2} \sim 48$ K [51]. As the crystal structure of Ca₃Ru₂O₇ comprises two closely spaced RuO₂ layers within a unit cell [Fig. 1d], substantial interlayer interactions within a bilayer unit are expected and were indeed identified in INS studies of the magnon dispersions [54, 55]. As exchange interactions between bilayer units are weak, the dimensionality of the exchange-bond network is intermediate between 2D and 3D. The INS data also revealed an anisotropy gap, but were insufficient for a determination of the nature of the dominant anisotropy (Ising versus XY).

In this work, we use TAS to examine the critical scattering in single-layer Ca₂RuO₄ and bilayer Ca₃Ru₂O₇ in vicinity and above $T_N$. We extract the critical static and dynamical exponents to determine the spin dimensionalities and anisotropies, which we compare to the model Hamiltonians employed in previous INS and RIXS studies below $T_N$. For Ca₂RuO₄, we derive the critical exponent $\beta$ of the order parameter from the temperature-dependence of the AFM (100) Bragg intensity. The static critical exponents $\nu$ and $\gamma$ are extracted from the Q-width and amplitude, respectively, of the magnetic diffuse scattering around (100.83) above $T_N$. We find that the temperature-dependence of the order parameter, Q-width, and amplitude are well-captured by a 2D-XY model, as expected based on the spin Hamiltonian extracted from the magnon dispersions [50].

The dynamic critical exponent $z$ is derived from the broadening in energy of the diffuse scattering at (100) for $T > T_N$ and is also in reasonable agreement with the 2D-XY model. For Ca₃Ru₂O₇, we derive $\beta$ from the temperature-dependence of the AFM (001) Bragg intensity below $T_N$, while $\nu$, $\gamma$ and $z$ are extracted from the Q-width, amplitude, and energy-width, respectively, of the diffuse scattering around (001) above $T_N$. From a combined consideration of all extracted exponents, we conclude that the critical behavior of Ca₃Ru₂O₇ is only partly consistent with the 2D-XY model, and is best described by the 3D-I model. We discuss these observations in the context of prior experiments on single-layer and bilayer iridates [34–36].

II. METHODS

High-quality single crystals of Ca₂RuO₄ and Ca₃Ru₂O₇ were grown by the optical floating zone method [47], as described in Ref. [50]. Ca₂RuO₄ exhibits the orthorhombic space group $Pbca$ at 11 K and the lattice parameters $a = 5.39$ Å, $b = 5.63$ Å, and $c = 11.75$ Å [45]. Ca₃Ru₂O₇ crystallizes in an orthorhombic space group $Bb\bar{2}1m$ at 50 K with the lattice parameters $a = 5.36$ Å, $b = 5.53$ Å, and $c = 19.54$ Å [52]. Single crystals that included orthorhombic (a,b)-twins were co-aligned on Si-plates with thicknesses of 0.5 mm and arranged in Al-sample holders. In case of Ca₂RuO₄, approximately 100 single crystals were co-aligned, yielding a total mass of 1.5 g. In case of Ca₃Ru₂O₇, approximately 30 single crystals were used with total mass of 0.8 g. The mosaicity of both sample arrays was 2-3°. Due to the (a,b)-twinning, the scattering planes were (H0L)/(0KL). All values of $Q$ are given in reciprocal lattice units.

The two-axis mode experiments on Ca₂RuO₄ were carried out at the thermal neutron spectrometer TRISP [56, 57] at the FRM II neutron source at the Heinz Maier-Leibnitz Zentrum (MLZ), Garching. The instrument was operated with clockwise scattering sense at the monochromator and sample ($SM = -1$, $SS = -1$).
FIG. 1. Crystal structure, critical scattering and magnetic order parameter in Ca$_2$RuO$_4$ (a-c) and Ca$_3$Ru$_2$O$_7$ (d-f). (a) Schematic of the crystallographic unit cell of single-layer Ca$_2$RuO$_4$ (black lines). Oxygen ions are omitted for clarity. Green and purple spins indicate the G-type AFM order. (b) Intensity of the magnetic (100) peak measured as a function of temperature. The red solid line is a power-law (PL) fit $I \propto M^2 \propto |q|^2 \beta$, with $\beta = 0.158(6)$ and $T_N = 112.20(1)$ K, convoluted with a Gaussian-distribution of Néel temperatures $T_N$ with a FWHM of $4.84(1)$ K (grey shaded area). The inset shows the intensity measured at $Q = (100.83)$. The increase of intensity in vicinity to $T_N$ indicates the presence of critical scattering from critical magnetic fluctuations. (c) Same data as in panel (b), but on a double-logarithmic scale. Note that the data and fits are non-linear due to the distribution of $T_N$. (d) Schematic of the unit cell of bilayer Ca$_3$Ru$_2$O$_7$ (black lines) with the A-Type AFM order indicated. (e) Intensity of the magnetic (001) peak. The green and red dashed lines indicate the two AFM transitions. Below $T_{N,2} = 48$ K, the magnetic moments reorient from $m \parallel a$ to $m \parallel b$. The red solid line is a PL fit, which yields $T_{N,1} = 54.16(2)$ K and $\beta = 0.230(6)$. The inset shows the intensity measured at $Q = (001.3)$, with the increase of intensity in vicinity to $T_N \approx 54$ K indicating the presence of critical scattering. (f) Same data for $T < T_{N,1}$ as in panel (e), but on a double-logarithmic scale.

Laue-Langevin (ILL), Grenoble. The instrument was operated (i) in two-axis mode with $SM = 1$, $SS = -1$ and $k_i = 1.3$ Å$^{-1}$ and (ii) in three-axis mode with $SM = 1$, $SS = -1$, $SA = 1$ and $k_f = 1.3$ Å$^{-1}$ with double focusing monochromator and analyzer (energy resolution $\approx 0.08$ meV). A Be-filter was used to suppress higher monochromator orders.

The cross section of magnetic neutron scattering [21, 61] is proportional to the dynamic scattering function $S(q, \omega)$, with $Q = G_m + q = k_i - k_f$ and $\omega = h(k_i^2 - k_f^2)/(2m)$. Here, $G_m$ is a magnetic reciprocal lattice vector, $q$ the relative momentum transfer, and $k_i, f$ the incident and final neutron wave vectors. $S(q, \omega)$ is related to the imaginary part of the generalized magnetic suscep-
bility via

\[ S(q, \omega) = \frac{\chi''(q, \omega)}{1 - \exp(-\hbar \omega/k_B T)}. \]  

The real and imaginary parts of the generalized susceptibility \( \chi(q, \omega) \) are Kramers–Kronig related. A general form of \( \chi''(q, \omega) \) is given by \( \chi''(q, \omega) = \chi'(q) F(\omega) \), where \( \chi'(q) \) is the real part of the static susceptibility and \( F(\omega) \) the spectral weight function, which is an even function of \( \omega \) and satisfies the normalization condition \( \int_{-\infty}^{\infty} F(\omega) d\omega = 1 \). Above the ordering temperature, spin fluctuations at small \( q \) are strongly damped and the spectral-weight function takes on a Lorentzian shape [21]:

\[ F(\omega) = \frac{1}{\pi} \frac{\Gamma}{\omega^2 + \omega_0^2}. \]  

To extract the inverse of the magnetic correlation length \( \kappa \) from \( Q \)-scans, we use the following Lorentzian form for the static susceptibility [4]

\[ \chi'(q) = \frac{\chi'(0)}{1 + q^2/k^2}. \]  

where \( \chi'(0) \equiv \chi_0 \) corresponds to the staggered magnetic susceptibility. The Kramers-Kronig relation connects \( \chi'(q) \) and \( S(q, \omega) \) via

\[ k_B T \chi'(q) = \int_{-\infty}^{\infty} \frac{1 - \exp(-\hbar \omega/k_B T)}{\hbar \omega/k_B T} S(q, \omega) \, d(\hbar \omega) \approx \int_{-\infty}^{\infty} S(q, \omega) \, d(\hbar \omega) = S(q), \]  

where \( \hbar \omega \ll k_B T \) was assumed [21, 61]. Hence, \( S(0) \propto \chi_0 T \) follows for the static case at \( q = 0 \). To determine \( S(q) \), in principle, it would be required to measure the entire \( S(q, \omega) \) function and perform a numerical \( \omega \)-integration, which can be avoided in 2D systems by using an energy integrating TAS configuration, as introduced by Birgeneau et al. [62]. In this configuration the TAS analyzer is removed (two-axis mode) and \( k_f \) is aligned perpendicular to the 2D-layers, corresponding to the ab-plane in Ca\(_2\)RuO\(_4\). The magnitude of \( k_f \) varies with \( \omega \), but the relevant components of \( q \) in the 2D planes are constant and independent of \( \omega \). In consequence, the detector signal corresponds to an energy integration with lower integration limit (energy-gain scattering) given by the thermal energy of the fluctuations, and upper limit (energy-loss scattering) given by the energy \( E_i \) of the incident neutrons:

\[ S(q) = \int_{-\infty}^{\infty} S(q, \omega) \, d(\hbar \omega) \approx \int_{-k_B T}^{E_i} S(q, \omega) \, d(\hbar \omega). \]  

In the case of Ca\(_2\)RuO\(_4\), we achieved this energy-integrating configuration with \( k_f \parallel c \) by choosing \( Q = (1 0 0.83) \) for \( k_i = 1.75 \, \text{Å}^{-1} \). For Ca\(_3\)Ru\(_2\)O\(_7\), due to the 3D character of the AFM order [55], the ideal energy-integration configuration with \( k_f \parallel c \) can in principle not be obtained, as \( Q_L \) cannot be chosen arbitrarily. However, as discussed in App. A, the effect of this imperfect energy-integration on the measured linewidth \( \kappa \) in the two-axis configuration (without analyzer) is insignificant for our determination of the critical exponent \( \nu \). Thus, the two-axis data of Ca\(_3\)Ru\(_2\)O\(_7\) were not corrected for the integration effect.

### III. RESULTS

#### A. Static critical properties of Ca\(_2\)RuO\(_4\)

In Ca\(_2\)RuO\(_4\), the magnetic moments point along the \( b \)-axis of the orthorhombic unit cell [45] [Fig. 1a] with a possible small canting in the \( c \)-direction \( (m_c \approx 0.1 m_b) \) [63]. The magnetic susceptibility in the paramagnetic state indicates quasi-2D spin fluctuations [64], which was recently also found from the magnon dispersion in the ordered phase [50], where the following parameters were derived: \( J = 5.8 \, \text{meV}, J_{XY} = 0.87 \, \text{meV} \) for the Heisenberg and XY-type exchange couplings; and \( E = 25 \, \text{meV}, \epsilon = 4 \, \text{meV} \) for the single-ion terms of the tetragonal and orthorhombic symmetries, respectively. An interlayer coupling \( J' \) was not required to describe the magnon dispersion [50], which is in line with studies on 1\% Ti-doped Ca\(_2\)RuO\(_4\), where a very small \( J' = 0.03 \, \text{meV} \) was reported [65]. Hence, the strong tetragonal term \( E \) and the small \( J' \) signal that Ca\(_2\)RuO\(_4\) can be regarded as a quasi-2D-XY AFM.

In the following TAS measurements on Ca\(_2\)RuO\(_4\), we use the energy-integrating two-axis mode (see Methods and Ref. [62]), which can be applied due to the 2D-character of the magnetism, with critical fluctuations that are expected to be independent of \( Q_L \). We perform \( Q_H \)-scans around \( Q = (1 0 0.83) \), which lies on the rod of the 2D magnetic scattering intensity. This corresponds to an energy-integrating configuration with alignment of \( k_f \parallel c \) at \( Q = (1 0 0.83) \) for \( k_i = 1.75 \, \text{Å}^{-1} \). Moreover, the advantage of a momentum \( Q \) that is slightly off from a magnetic Bragg peak position is that the signatures of critical scattering can be particularly pronounced [66]. Accordingly, we observe an enhancement of the scattered intensity at \( Q = (1 0 0.83) \) [inset in Fig. 1b] for temperatures in vicinity to the anticipated \( T_N \) of approximately 110 K [45, 64]. More specifically, we observe that the critical scattering intensity peaks at a temperature slightly higher than 110 K. This behavior is likely related to the fact that \( T_N \) of our Ca\(_2\)RuO\(_4\) sample is not sharply defined, but a distribution of Néel temperatures is present [see gray shaded area in Fig. 1b], in spite of the confirmed excellent crystalline quality (see Methods and Ref. [50]). This variance of \( T_N \) likely results from microstrains within the crystal, which emerge below the concomitant structural and metal-to-insulator transition at 360 K [46] and could be reminiscent of the (pseudo)spin-lattice coupling in Sr\(_2\)IrO\(_4\) [67, 68]. In the following analysis, we take this distribution of \( T_N \) into
account, which allows us to extract the critical properties of Ca$_2$RuO$_4$ similarly to the case of a sharply defined $T_N$.

For an ideal second order phase transition, the order parameter (staggered magnetization $M$) is $I \propto M^2 \propto |t|^2$ and vanishes above $T_N$. Thus, the critical exponent $\beta$ can be determined from the measured nominal magnetic (100) peak intensity $I_{100}$ in Fig. 1b. In general, $\beta$ and the other critical exponents are extracted from the slopes of linear fits in double-logarithmic plots (see Ca$_2$RuO$_4$ below). However, due to the present variance of $T_N$, the data in Fig. 1b cannot be described directly with the power law (PL) scaling function. Note that especially the intensity around 110K does not show the expected sharp drop but is smeared out. This rounding of the intensity evolution cannot be attributed to critical scattering above $T_N$, since the data in the inset of Fig. 1b indicate that the critical contribution is two orders of magnitude smaller. Thus, we fit the (100) data in the range 90 - 120K ($-0.2 < t < 0.1$) with a convolution of the above mentioned PL and a Gaussian distribution of $T_N$ with full width at half maximum (FWHM) $\Delta T_N$ [Fig. 1b]. The resulting fit parameters are $T_N = 112.20(1)$ K, $\Delta T_N = 4.84(1)$ K, and $\beta = 0.158(6)$. The $\beta$-value lies in between the limits of the 2D Ising (2D-I) ($\beta = 0.125$ [4]) and 2D-XY model ($\beta = 0.23$ [69, 70]), as suggested for a XY system with fourfold crystal field anisotropy ($XY_{h4}$) [71]. Figure 1c shows $I_{100}$ and the fit curve on a double-logarithmic scale, illustrating that the PL fit provides an adequate description of the data below $T_N$, and also for a range of temperatures above $T_N$. Note that the strong deviation from a simple PL (straight line in double logarithmic plot) is due to the Gaussian distribution of $T_N$.

As a next step we perform $Q_{\parallel}$-scans around (100.83) to determine the inverse correlation length $\kappa(T)$ [Fig. 2]. Prior to fitting of the scans with Voigt-profiles, we thoroughly determine the background (BG) contributions. Representative scans are shown in Fig. 2a-d. We identify several components of the BG: (i) A temperature-independent component, which is determined at 170K [Fig. 2a], i.e. well above $T_N$. The obtained fit (H-T BG) is employed as BG in the analysis of the data measured at all other temperatures (see dashed-dotted lines in Fig. 2a-d). (ii) The scan at the lowest measured temperature $T = 80$K [Fig. 2b] shows two incommensurate peaks besides the H-T BG. By comparing the temperature-dependent intensity of these resolution limited peaks with the intensities of the magnetic (100) [Fig. 1c] and (101) peaks [45], they can be assigned to the (101) peak of the main domain, and the (011) peak of the twin domain [63]. The (101) peak is likely associated with a 'B-centered' phase with a different propagation vector and transition temperature $T_{N,101} \approx 150$K [45]. Finally, we subtract the H-T BG and the aforementioned two peaks with proper $T$-scaling from the $Q$-scans and obtain the corrected data shown in Fig. 2e. These data are well described by Voigt-profiles, which correspond to the convolution of the intrinsic Lorentzian with half-width-half-maximum (HWHM) $\kappa$ [Eqn. (3)] and the Gaussian instrumental resolution. The constant width of the instrumental Gaussian (FWHM $\approx 0.034$ r.l.u.) was extracted from the 80K scan in agreement with simulations carried out with the RESLIB [72] and TAKIN [73] softwares, respectively.

Prior to the discussion of the inverse correlation length $\kappa$ extracted from Fig. 2e, we address the possible presence of concomitant longitudinal and transverse fluctuations. In general, critical longitudinal (parallel to the static ordering vector) and non-critical transverse fluctuations are expected to be both visible and not separated for all measurement configurations used in this work. However, neu-

![Fig. 2](https://example.com/fig2.jpg)

**FIG. 2.** Selected energy-integrated $Q_{\parallel}$-scans around (100.83) for Ca$_2$RuO$_4$ before (a-d) and after (e) background (BG) subtraction. The data contain a $T$-independent constant BG, which was determined at $T = 170$K and labelled as 'high-temperature' (H-T) BG (a). Panels (b-d) show the H-T BG as a dashed-dotted line. At low-$T$, two small $T$-dependent resolution limited Gaussian-peaks (black dotted lines) appear (b-d), which are attributed to the magnetic (101) peak of the main domain and the (011) peak of the twin domain, respectively. The critical scattering component (Voigt-profile) is shown as a red line. (e) Selected $Q_{\parallel}$-scans after BG subtraction with corresponding fit functions (solid lines). For clarity the data are plotted with a constant offset.
tron spin-echo (NSE) spectroscopy [74–77] is capable to separate the two components [78]. To this end, we carried out high-resolution NSE measurements on Ca$_2$RuO$_4$ at TRISP (see Methods), at $Q = (1\, 0\, 0)$ with $k_i = 2.66\ \text{Å}^{-1}$. Figure 3 shows the resulting spin-echo polarization vs. the spin-echo time $\tau \equiv (m^2 \omega_L L)/(\hbar^2 k_i^2)$ for selected temperatures, with the Larmor-frequency $\omega_L = \gamma_n B_0$ ($\gamma_n = 2.916\ \text{kHz/Oe}$) corresponding to the static magnetic fields $B_0$ in the spin-echo arms with length $L$. A high-temperature BG (170 K) was subtracted from the data. According to Ref. [78], possible transverse fluctuations in Ca$_2$RuO$_4$ (along the c-axis) would lead to a polarized signal for $\tau > 0$, i.e. for parallel magnetic fields $B_0$. Conversely, longitudinal fluctuations (along the b-axis) lead to a polarized signal for $\tau < 0$. Figure 3 shows a significant polarization at $\tau < 0$ that depolarizes as a function of $\tau$ and $T$, respectively, indicative for the $T$-dependent linewidth $\Gamma(T)$ of the critical fluctuations. By contrast, no significant polarization at $\tau > 0$ was measured. Moreover, no oscillations of the polarization at small $\tau$ were observed, which are a hallmark of interference effects between transverse and longitudinal fluctuations [78]. Hence, we conclude that transverse fluctuations along the c-axis are negligible or absent in Ca$_2$RuO$_4$ and we assume in the following that the fluctuations observed in the TAS experiments are also purely longitudinal. A possible explanation for this absence is that these fluctuations are gapped and thus not excited in the $T$-range of our study.

The inverse correlation length $\kappa(T)$ resulting from the fits in Fig. 2 is shown in Fig. 4. Notably, a $T$-dependent broadening above 116 K ($T_N + 4\ \text{K}$) can be observed, while the Q-width is approximately constant for $T < 116\ \text{K}$. This is in contrast to the conventional critical scaling theory where $\kappa$ should converge to zero at $T_N$. In the following we will discuss several models to explain this saturation of $\kappa$ at $T < 116\ \text{K}$: (i) An obvious reason for such lower bound of the linewidth are crystallographic defects [79, 80]. One possible type of defect in Ca$_2$RuO$_4$ can be domain walls of the structural twins, which can disrupt the long-range magnetic ordering. However, from the Q-width of the (1\,0\,0) magnetic peak (not shown here) we derive a domain size of $> 300\ \text{Å}$, which is much larger than the extracted correlation length of $20\ \text{Å}$ at $Q = (1\,0\,0)$.35 and $T = 110\ \text{K}$. Thus we exclude domain size effects as the origin of the observed linewidth saturation. (ii) At $T \approx T_N$, i.e. close to the 3D ordering, one expects a crossover of the critical fluctuations from a 2D to a 3D character with an increasing influence of the $Q_L$ component on $\kappa$. This effect was described in Refs. [17, 81] and modelled by an effective $\kappa$ with $\kappa^2_{\text{eff}} = \kappa^2_{3\text{D}} + \kappa^2_{\text{pow}}$, $\kappa^2_{3\text{D}} \equiv Q_L^2 \nu'/J$, and $\kappa^2_{\text{pow}} \equiv \kappa_0\nu^2$. A fit of our experimental $\kappa(T)$ with a convolution of $\kappa_{\text{eff}}$ and the aforementioned Gaussian distribution of $T_N$ results in an exponent $\nu = 1.0(1)$ and $\kappa_{3\text{D}} = 0.035(1)$, and describes the data over the entire measured $T$-range (green dashed-dotted line in Fig. 4). The exponent $\nu$ matches the universal value of the 2D-I model ($\nu_{2\text{D-I}} = 1.4$) [4]. From $\kappa_{3\text{D}} = 0.035(1)$ we obtain the ratio $J'/J = 0.002$. This is in agreement with $J'/J = 0.004$ derived from INS on 1% Ti-doped Ca$_2$RuO$_4$ [65]. Assuming $J = 5.8\ \text{meV}$ [50], this corresponds to an interlayer coupling $J' = 0.01\ \text{meV}$.

The red dashed line in Fig. 4 shows a PL fit $\kappa \propto |t|^\nu$ without a $\kappa$-offset at $T_N$, i.e. without a $Q_L$ dependence due to 3D correlations, convoluted with the variance of $T_N$. Only the data for $T > 116\ \text{K}$, i.e. beyond the saturation region, were included in the fit. The rounded shape of the red line towards $T_N$ results from the $T_N$ variance. The resulting $\nu = 0.42(4)$ is close to the mean-field (MF) value of $\nu_{\text{MF}} = 0.5$, but is at odds with $\nu = 1.0(1)$ obtained in the previous PL fit with the offset in $\kappa$, although both fits give a satisfactory description of the data for $T > 116\ \text{K}$. Furthermore, we note that a fit of $\kappa(T)$ with a 2D quantum Heisenberg model [82–84] with an anisotropy parameter accounting for 3D correlations close to $T_N$ [31] also gives a good agreement with the data, but we exclude this model due to the large easy-plane anisotropy in Ca$_2$RuO$_4$.

We now focus on the 2D-XY model, which was already suggested in the context of the magnon dispersion [50] and describes a topological phase transition accompanied by an unbinding of vortex/antivortex pairs [9–11]. The parameters of this model are the Kosterlitz-Thouless (KT) temperature $T_{\text{KT}}$, a critical exponent $\eta = 0.25$, and a dimensionless non-universal parameter $b$ [11, 85], which was previously determined to be approximately 1.9 [81]. The correlation length in this model is defined as [11]

\[ \xi \propto \exp \left( \frac{b}{\sqrt{t_{\text{KT}}}} \right), \quad \text{with} \quad t_{\text{KT}} \equiv (T/T_{\text{KT}} - 1). \tag{6} \]
For systems with magnetic long-range order the actual KT-transition at \( T_{KT} < T_N \) is usually obscured by the 3D ordering with nonzero interlayer couplings \( J' \), which set in around \( T_N \). The relation between \( T_{KT} \) and \( T_N \) is given by \([69, 81]\)

\[
\frac{T_N - T_{KT}}{T_{KT}} = \frac{4b^2}{[\ln(J/J')]^2}. \tag{7}
\]

Assuming \( J'/J = 0.002 \), as derived from the above PL fit with \( Q_L \) dependence to capture the \( \kappa \)-offset, we obtain \( T_{KT} = 82 \) K, with \( T_{KT} \) denoting the KT temperature derived from Eqn. (7) and \( T_{KT} \) the KT temperature extracted from fits to \( \kappa(T) \) in the following. As expected for a system with a KT temperature below \( T_N \), our data do not show any signatures of a transition around 82 K. Nonetheless, we use this model in the following to describe the scaling above \( T_N \), as it was demonstrated \([86–88]\) and experimentally confirmed \([16, 17, 89]\), that even a XY anisotropy much weaker than in the case of Ca\(_2\)RuO\(_4\) can result in 2D-XY scaling. Hence, as a next step, we fit Eqn. (6) to \( \kappa(T) \) for \( T > 116 \) K. Note that \( T_{KT} \) is much lower than the lower limit of the fitting range, i.e. a possible distribution of \( T_N \) and KT temperatures in the sample will not affect the result of the fit significantly and is therefore not considered here. The resulting fit (solid black line in Fig. 4) with \( T_{KT} = 87(2) \) K \( \approx 0.87 T_N \) provides an excellent description of the data, and is in reasonable agreement with \( T_{KT} = 82 \) K from Eqn. (7).

We note that a fit with \( b \) as a free parameter did not converge, since it couples strongly to \( T_{KT} \). Thus, \( b \) was fixed to 1.9 \([81]\). We also note that \( \eta \) was fixed to 0.25 \([10]\), although \( \eta \) can deviate from this value in specific models on critical scattering, which then would affect the line-shape of \( V(q) \) from the Lorentzian form employed in this work (see Methods). However, we find that in our case the deviation of the line-shape for \( \eta = 0.25 \) is relatively subtle (see App. E) and lies below the detection threshold of the statistics of our data. Therefore, we use the simplest approach for \( V(q) \) in the present work, which is the Lorentzian-function.

In summary, the data in Fig. 4 are consistent with both, the 2D-XY (black solid line) and 2D-I (green dashed-dotted line) scaling behavior for \( T > 116 \) K. Nevertheless, we rule out the latter scaling for the description of \( \kappa(T) \), as the critical peak amplitudes \( S_0(T)/T \) [Fig. 5] with the corresponding critical exponent \( \gamma \) (see below) are not compatible with the 2D-I model, although a crossover to 2D-I scaling close to \( T_N \) is expected due to the orthorhombic anisotropy \( \epsilon \) \([50]\). While this crossover from 2D-XY to 2D-I scaling presumably occurs in a \( T \)-range very close to \( T_N \) and is not resolved in our data, we attribute the observed saturation for \( T < 116 \) K to a crossover to 3D coupling, which eventually drives the magnetic transition.

In addition to \( \kappa(T) \), we analyzed the staggered susceptibility \( \chi_0 \), which also shows critical behavior close to \( T_N \). Related via the Kramers-Kronig relation, \( \chi_0 \) is proportional to \( S(0) \propto \chi_0 T \) (see Methods), i.e. the peak amplitude of the Lorentzian-profile \( S(q) \). In the following, \( S(0) \) will be denoted as \( S_0 \). Figure 5 shows the temperature dependence of the amplitude measured at \((1.00.83)\). First, we fit a \( PL \propto |t|^{-\gamma} \) in the range \( 110 K < T < 140 K \), convoluted with the Gaussian \( T_N \) distribution by assuming \( S_0 = 0 \) for \( T < T_N \). The agreement with the data is not convincing and the extracted critical exponent \( \gamma = 0.47(2) \) does not match universal values \([4]\), especially not the value predicted for the 2D-I model \((\gamma_{2DI} = 1.75)\). Furthermore, since scaling theory predicts PL behavior for temperatures both above and below \( T_N \), we also carried out a fit over the entire \( T \)-range (see App. B). Nevertheless, such fit yields a similar value for \( \gamma \), corroborating that PL scaling is not suitable to capture the temperature dependence of the amplitudes.

Next, we fit the range \( 110 K < T < 140 K \) with the 2D-XY model by using the scaling relation \( S(0) \propto \xi^{2-\eta} \) \([11]\)

\[
\frac{S(0)}{T} \propto \exp \left( \frac{B}{\sqrt{T_{KT}}} \right), \tag{8}
\]

with \( B \equiv b(2 - \eta) \) and \( T_{KT} = 87 K \) from above. We fixed \( \eta = 0.25 \) as suggested for the 2D-XY model \([11]\). The model gives a good description of the data with only one free parameter in the fit, that is, the proportionality constant in Eqn. (8).

**B. Static critical properties of Ca\(_2\)Ru\(_2\)O\(_7\)**

Ca\(_2\)Ru\(_2\)O\(_7\) exhibits ferromagnetic (FM) bilayers \([Fig. 1d]\), which are stacked in an AFM fashion along
FIG. 5. Peak amplitude $S_0(T)/T$ of Ca$_2$RuO$_4$. The red dotted line is a PL scaling fit ($\gamma = 0.47(2)$) and the black solid line corresponds to the 2D-XY model. The grey data points were not included in the fit. The black vertical line indicates $T_N$ and the grey bar the variance of $T_N$.

the c-axis (A-type AFM) [51, 52]. From the magnon dispersion in the ordered phase [54, 55] the following terms of the Hamiltonian were derived: $J = -3.75$ meV, $J' = -6.5$ meV for the nearest neighbor and intra-bilayer coupling; and $E = 5.5$ meV, $\epsilon = 2.5$ meV for the tetragonal and orthorhombic anisotropy, respectively. Notably, the magnon dispersion along the c-direction and inter-bilayer coupling $J'$ are very small or absent [54, 55]. Hence, Ca$_3$Ru$_2$O$_7$ exhibits an easy-plane anisotropy $E$ and strongly coupled bilayers (large $J_c$) that can possibly act as one magnetic entity [90], suggesting that Ca$_3$Ru$_2$O$_7$ could also be a candidate for quasi-2D-XY critical behavior. This calls for an investigation whether the critical behavior in the bilayer compound Ca$_3$Ru$_2$O$_7$ falls either into the quasi-2D or the 3D limit, or corresponds to an intermediate case.

Figure 1e shows the magnetic (001) peak intensity $I_{001}$ measured upon warming, with a first-order transition at $T_{N,2} \approx 48$ K and a second-order transition at $T_{N,1} \approx 56$ K, in good agreement with Ref. [91]. In contrast to the magnetic peak of Ca$_2$RuO$_4$ [Fig. 1c], $I_{001}$ of Ca$_3$Ru$_2$O$_7$ drops sharply towards $T_{N,1}$, suggesting that possible distribution of Néel temperatures $\Delta T_{N,1}$ is negligible. We explain this observation with the appearance of less pronounced intrinsic crystal strains above the structural transition at $T_{N,2}$. Moreover, in comparison to Ca$_2$RuO$_4$, the crystal-field distortions are expected to be weaker in Ca$_3$Ru$_2$O$_7$ [55]. In order to establish the presence of critical scattering we measure the scattering intensity in distance to the (001) Bragg position at $Q = (001.3)$ as a function of temperature (see inset Fig. 1e). Notably, the critical intensity in the inset in Fig. 1e peaks at $T_{N,1}$ and its magnitude is compatible with the remaining intensity for $T > T_{N,1}$ in Fig. 1e. A PL fit (without a $T_{N,1}$ distribution) in the range 49 K $< T < T_{N,1}$ (0 < $|t|$ < 0.1), i.e. between $T_{N,1}$ and $T_{N,2}$, yields a critical exponent $\beta = 0.23(6)$ and $T_{N,1} = 54.16(2)$ K [Fig. 1e,f]. Figure 1f shows $I_{001}$ and the PL fit on a double-logarithmic scale, suggesting a purely linear evolution of $I_{001}$ in such a plot for the measured temperatures. The obtained value of $\beta$ matches the universal value of the 2D-XY model ($\beta = 0.23$ [69, 70]), although it should be taken with caution as the point density in close vicinity of $T_{N,1}$ is sparse. Moreover, the intrinsic scaling behavior could be obfuscated due to a contribution in the scattering intensity from an overlap with the second transition at $T_{N,2}$.

To extract $\kappa(T)$ and the amplitude of the critical scattering, we carried out $Q_H$-scans around (H01) in the two-axis mode. The ideal energy-integrating configuration used for Ca$_2$RuO$_4$ is not applicable for 3D systems (see Methods). However, a numerical simulation confirmed that the integration according to Eqn. (5) is sufficiently satisfied for our two-axis configuration in Ca$_3$Ru$_2$O$_7$ (see App. A). Furthermore, we note that for Ca$_3$Ru$_2$O$_7$, spin-echo experiments to discern longitudinal and transverse fluctuations have not been carried out. Nonetheless, from the fact that in the present study both, the static and dynamical critical fluctuations in Ca$_3$Ru$_2$O$_7$ are well captured by power-laws (see below), we conclude that the non-critical transverse fluctuations do not contribute significant intensity around $T_{N,1}$ [66]. From the $Q_H$-scans we subtracted two BG components: (i) A sharp peak at $H = 0$, which is clearly visible at high-$T$ (150 K) [Fig. 6a]. In addition, we performed a scan around (H01.25) at 100 K and found that the sharp feature is independent of $Q_L$ and $T$ (see App. C). Thus, we assign it to 2D diffuse nuclear scattering from disorder along the c-axis due to e.g. stacking faults. (ii) The sharp resolution-limited (001) peak with Gaussian-width $\approx 0.01$ r.l.u. [Fig. 6b], which rapidly vanishes above $T_{N,1}$. The (001) peak is intense at $T \leq 54.5$ K and the extraction of critical scattering is not reliable. The BG corrected $Q_H$-scans can be captured by a Voigt-profile (intrinsic Lorentzian critical scattering convoluted with instrumental Gaussian-profile) [Fig. 6e]. A critical scattering intensity can be clearly observed at least up to 70 K. We note that there might be a contribution from critical fluctuations even at 100 K, but since it is very weak we do not consider this temperature in the following analysis.

The resulting Q-width $\kappa(T)$ is plotted in Fig. 7 on double-logarithmic scales. Note that in the following analysis of Ca$_3$Ru$_2$O$_7$, we extract the critical exponents of the PLs from the slopes of linear fits in plots with double logarithmic scaling, whereas plots with linear scaling were employed in the above analysis of Ca$_2$RuO$_4$ [Fig. 4 and Fig. 5], due to the variance of $T_N$ in the latter material. The red dotted line is a linear fit in the range 55 K $\leq T \leq 70$ K with the slope corresponding to the critical exponent in the scaling relation $\kappa \propto t^{\nu}$. The obtained $\nu = 0.550(4)$ lies between the values predicted for the 3D-I ($\nu_{3D-I} = 0.630$, [6]) and the MF model ($\nu_{MF} = 0.5$, [7]...
The MF model, however, is at odds with the modeling of the magnon dispersion of Ca$_3$Ru$_2$O$_7$ in Ref. [55], which used only nearest neighbor couplings and no long-ranged interactions in the spin Hamiltonian [4]. Thus, we assign the critical scaling of $\kappa(T)$ rather to the 3D-I model (green solid line in Fig. 7), which also captures the data well. In analogy to Ca$_2$RuO$_4$, we also fit $\kappa(T)$ of Ca$_3$Ru$_2$O$_7$ with the 2D-XY model, using Eqn. (6), $\eta = 0.25$, and $b = 1.9$. The obtained KT-temperature is $T_{\text{KT}} = 45.42(6)$ K, but the agreement between the fit (black dashed-dotted) and the data is unsatisfactory for most temperatures [Fig. 7]. We also test a fit with $b$ as a free parameter, since lower values of $b$ were reported in some experiments [92, 93] and derived in numerical calculations [85]. Such a fit (not shown here), with $b = 0.44(1)$ and $T_{\text{KT}} = 53.18(7)$, yields a better agreement with the $\kappa(T)$ data of Ca$_3$Ru$_2$O$_7$. However, using the latter values of $b$ and $T_{\text{KT}}$ as an input for the fit of the critical amplitudes (see below) results in a very strong deviation from the data (not shown here) and is therefore disregarded. In summary, we conclude that the 3D-I model is most appropriate to describe the critical behavior of $\kappa(T)$ above $T_N$.

The critical exponent $\gamma$ of the staggered susceptibility is obtained by fitting the corresponding peak amplitudes $S_0/T$ with the PL scaling $\chi \propto T^{-\gamma}$ in the range $55 \text{ K} \leq T \leq 70 \text{ K}$. From the slope of the corresponding linear fit (red dotted line) in the double-logarithmic plot [Fig. 8], we extract $\gamma = 1.290(4)$, which is close to the value predicted for the 3D-I model ($\gamma_{3\text{DI}} = 1.238$, [6]), as indicated by the green solid line in Fig. 8. For the comparison with the 2D-XY model, we use Eqn. (8), with $\eta = 0.25$ and $b = 1.9$, as well as $T_{\text{KT}} = 45.42$ K determined from the fit of $\kappa(T)$ above. In spite of a good agreement with the data at $T > T_N + 2K$ (see black dashed-dotted line), the PL fits are more suitable to describe the scaling of the critical amplitudes closer to $T_N$.

C. Dynamic critical properties of Ca$_3$RuO$_4$

Figures 9a–d display selected energy-scans at (100) measured on the cold neutron TAS FLEXX with $k_f = 1.3$ Å$^{-1}$. A $T$-independent BG from elastic incoherent scattering recorded at 170 K was subtracted from the raw data. The BG corrected data are described by a fit with the sum of a resolution-limited Gaussian-peak (FWHM...
\( \approx 0.12 \text{meV} \) and a Voigt-function capturing the critical scattering. The resolution was extracted from the width of the elastic magnetic scattering well-below \( T_N \). The free parameters of the fits are the amplitude of the Gaussian-peak, and the amplitude and width of the Voigt-peak. The amplitudes of the Gaussian and the Voigt match the intensities of the \((100)\) peak [Fig. 1b] and the amplitude of the critical scattering at \((100.83)\) [Fig. 1b, inset] (see App. D). An overview of the energy-scans and the resulting fit curves at selected temperatures is shown in Fig. 9e.

The resulting energy-width \( \Gamma(T) \) of the critical component is plotted in Fig. 10a. Above 115 K the data show a significant broadening, while below no systematic trend was observed. For these data \((T \leq 115 \text{K})\) the intensity of the \((100)\) peak is much stronger than the critical scattering and extraction of \( \Gamma(T) \) in the fit is not reliable. Therefore, we exclude this \( T \)-range in the following scaling analysis (grey data points in Fig. 10a). To determine the dynamic critical exponent \( z \) from \( \Gamma(T) \) above 115 K, in analogy to \( \kappa(T) \), we fit a convolution of the PL \( \Gamma \propto |t|^{z \nu} \) with a Gaussian distribution of \( T_N \) (red dotted line). By assuming \( \Gamma(T) = 0 \) for \( T \leq T_N \), as predicted by dynamic scaling theory [3], the PL fit yields \( z \nu = 1.1(1) \). In spite of the good agreement with the data in Fig. 10a, the error margin is relatively large and only the 3D-H model \((z_{\text{2DH}} = 1.067, [3, 94])\) and the 3D-I model \((z_{\text{2DI}} = 1.26, [3, 6])\) are reasonably close to the obtained exponent. However, 3D-scaling is not expected well above \( T_N \) for this quasi-2D system. Furthermore, \( z \nu = 1.1(1) \) is far away from 2D-I scaling \((z_{\text{2DI}} = 1.75 [4, 95])\), which was discussed in the context of \( \kappa(T) \) of \( \text{Ca}_2\text{RuO}_4 \) (green dashed-dotted line in Fig. 4).

Next, we examine the critical dynamics of \( \text{Ca}_2\text{RuO}_4 \) in terms of the 2D-XY model. For the motion of vortices a dynamic scattering function \( S(q, \omega) \) with a quadratic Lorentzian form (central peak) was derived [85, 96, 97], and also experimentally observed [16, 98, 99]. However, such a central peak is not present in our data [Fig. 9], which can be captured by a simple Lorentzian function. Thus, we use instead the dynamic scaling relation \( \Gamma \propto \kappa z \), suggested to be appropriate for relaxational dynamics in the 2D-XY model [100] to check for 2D-XY scaling above 115 K. The black solid line in Fig. 10a shows the scaling for the critical exponent \( z_{\text{XY}} = 2.0 \), which was postulated for the 2D-XY model [100]. The agreement with the data is not as good as for the PL fit, although at high temperatures the 2D-XY scaling lies essentially within the error bars. The deviation at low temperatures
could be due to the variance of $T_N$ or a crossover to a different scaling behavior. We note that using $z$ as a free fit parameter improves the agreement with the data, but the obtained value of $z_{XY} = 3.04(6)$ (fit not shown here) is at odds with the theory for the universality class.

In addition, we determine $z$ directly from plotting $\Gamma$ vs. $\kappa$ on double logarithmic scales [Fig. 10b]. In such a plot, the slope of a linear fit to the data corresponds to the critical exponent of the scaling relation $\Gamma = \kappa^z$. The resulting dynamical critical exponent $z = 2.9(2)$ (red dashed line) is not compatible with any known universal value. Note, however, that the $\Gamma$ vs. $\kappa$ data points in Fig. 10b were obtained by interpolation, as the $Q$- and energy-scans on Ca$_2$RuO$_4$ were taken at different $Q$-positions and temperatures at two different instruments, which can lead to uncertainties. Specifically, the data points in Fig. 10b were generated by interpolating $\kappa(T)$ to the temperatures at which $\Gamma(T)$ was measured and the same fitting range ($T > 115$ K) was included, where the impact on the $T_N$ distribution is negligible. Nevertheless, while the dynamical critical exponent of the linear fit deviates from a universal value, we find that the 2D-XY model (solid black line) with the exponent $z = 2.0$ [100] captures the $\Gamma$ vs. $\kappa$ data reasonably well [Fig. 10b].

**D. Dynamic critical properties of Ca$_2$Ru$_2$O$_7$**

Figures 11a-d display selected energy-scans at the (001) peak of Ca$_2$Ru$_2$O$_7$. A constant elastic incoherent high-$T$ BG is subtracted from the energy-scans, which was measured at $T = 100$ K, where the contribution of critical scattering is negligible [Fig. 6d]. Subsequently, a single Voigt-function (Gaussian-width $\approx 0.06$ meV) is fitted to the scans. An overview of the energy-scans after BG subtraction with the corresponding fits is plotted in Fig. 11e for selected temperatures.

The resulting energy-width $\Gamma(T)$ of Ca$_2$Ru$_2$O$_7$ is shown in Fig. 12a on double logarithmic scales. The data are well-captured by a linear fit (red dotted line) in the range $55$ K $\leq T \leq 70$ K with a slope $z\nu = 1.186(8)$, corresponding to the critical exponent in the scaling relation $\Gamma \propto \Gamma^z$. This exponent is close to the value predicted for the 3D-I model ($z\nu_{3DI} = 1.260$, [3, 6]) indicated by the green solid line in Fig. 12a, which is consistent with the static critical properties. In analogy to Ca$_2$RuO$_4$, we also carry out a fit with the 2D-XY model, using the dynamic scaling relation $\Gamma \propto \kappa^z$ with $z = 2.0$ [100]. The resulting fit (black dashed-dotted line in Fig. 12a) describes the data reasonably well at high temperatures, but deviates strongly in proximity to $T_N$.

Figure 12b shows a plot of $\Gamma$ vs. $\kappa$ on double logarithmic scales, which allows us to determine $z$ directly. From the slope of a linear fit (red dotted line), we derive $z = 2.14(2)$ as the critical exponent, which is close to $z = 2.0$ proposed for the 3D-I model [3, 101] and consistent with the $\Gamma(T)$ scaling above. Notably, $z = 2.0$ also corresponds to the 2D-XY model [100]. However, since the 3D-I scaling is also compatible with the temperature-dependence of the $Q$-width and critical amplitudes at $T > T_N$ of Ca$_2$Ru$_2$O$_7$, we consider this model as most appropriate.

**IV. DISCUSSION AND CONCLUSION**

For Ca$_2$RuO$_4$, we revealed that the description of the static critical properties by PL scaling gave only partially satisfactory results, whereas application of the 2D-XY model provided a conclusive picture. In more detail, the obtained critical exponent $\beta = 0.158(6)$ is consis-
FIG. 11. Selected energy scans of Ca$_3$Ru$_2$O$_7$ before (a-d) and after (e) BG subtraction. (a-d) In contrast to Ca$_2$RuO$_4$, the Ca$_3$Ru$_2$O$_7$ data could be well fitted by a single Voigt-profile and a constant elastic high-T BG measured at 100 K. (d) Selected energy scans after BG subtraction with corresponding fit functions (solid lines). For clarity the curves are plotted with a constant offset.

FIG. 12. Energy-width $\Gamma$ of Ca$_3$Ru$_2$O$_7$ vs. temperature (a) and the inverse correlation length (b) on double-logarithmic scales. (a) The red dotted line is a linear fit with the slope corresponding to the critical exponent $z \nu = 1.186(8)$, according to the scaling relation $\Gamma \propto T^{\frac{z \nu}{2}}$. The green solid line indicates 3D-I scaling with $z \nu_{3D} = 1.260$ [3, 6]. The black dashed-dotted line corresponds to the 2D-XY model with the exponent $z = 2.0$ [100]. (b) The red dotted line is a linear fit with the slope corresponding to the critical exponent $z = 2.14(2)$, according to the scaling relation $\Gamma \propto \kappa^z$. The green solid line corresponds to 3D-I scaling and the 2D-XY model, which both exhibit the exponent $z = 2.0$ [3, 100, 101]. Note that only the 3D-I model is also consistent with the static critical properties of Ca$_3$Ru$_2$O$_7$. The grey data points were not included in the fits.

tent with a 2D-XY model with fourfold crystal anisotropy (XY$_4$) [71]. The inverse correlation length $\kappa$, in principle, could be fitted with different PLs and the 2D-XY model. Specifically, the observed saturation of $\kappa$ in the range between $T_N$ and $T_N + 4$ K was captured by a PL with $Q_L$-dependence, likely indicating the presence of 3D fluctuations close to $T_N$. Nevertheless, the employed PL fits, with and without $\kappa$ offset, were not consistent with the 2D nature of Ca$_2$RuO$_4$ far above $T_N$ and results from fits of the amplitudes of the critical scattering. Instead, the 2D-XY model captured both, $\kappa$ and the amplitude adequately. The resulting Kosterlitz-Thouless temperature $T_{KT} = 87(2)$ K was consistent with the ratio $J'/J = 0.002$ derived from the offset in $\kappa(T)$. Our attempt to describe the amplitude of the critical scattering by PL scaling showed that the functional form does not capture the data appropriately and the extracted critical exponent $\gamma = 0.47(2)$ is not compatible with universal values. Yet, 2D-XY scaling captured the critical amplitudes in a broad T-range around $T_N$, in agreement with the Hamiltonian extracted from the magnon dispersion in the ordered phase [50]. The energy-width $\Gamma(T)$ is best captured by a PL fit with $z \nu = 1.1(1)$, which is compatible with 3D AFM Heisenberg scaling ($z \nu_{3DH} = 1.067$, [3, 94]). However, such a model for the dynamical critical scaling would be in stark contrast to the static critical behaviors, the magnon dispersion [50], and the quasi-2D character of Ca$_2$RuO$_4$. Consequently, we fitted the data with the 2D-XY model and $z_{XY} = 2.0$ [100], which also
captures the data reasonably well.

In general, a crossover in the critical behavior from 2D-XY to Ising scaling close to $T_N$ can be expected in Ca$_2$RuO$_4$ due to the orthorhombic terms in the spin Hamiltonian \[ H \]. The observed saturation of $\kappa(T)$ close to $T_N$ might be indicative for a dimensionality crossover to 3D scaling, whereas signatures of such a crossover were less clear in the analysis of the other critical exponents. This apparent absence can be due to the limited instrumental resolution of our TAS measurements and calls for a complementary high-resolution NSE study, focusing on the temperature range in close vicinity to $T_N$. Notably, the enhanced energy resolution of NSE previously helped to resolve controversies about the scaling behavior of heavy Fermion superconductors \[ 102 \] and revealed a crossover from Heisenberg to Ising scaling close to $T_N$ in the classical 3D AFM MnF$_2$ due to uniaxial anisotropy \[ 78 \]. Moreover, subtle signatures of additional phases, which were proposed to exist in Ca$_2$RuO$_4$ above the AFM order, such as orbital order \[ 103, 104 \] and a Jahn-Teller driven spin-nematic phase \[ 105 \], might be detectable in the critical scaling behavior measured with high-resolution NSE. Apart from that, it will be interesting to probe the existence of possible vortex/antivortex-pairs with cryogenic microscopy techniques, such as Lorentz transmission electron microscopy \[ 106 \], which might be particularly pronounced in thin films of Ca$_2$RuO$_4$ \[ 107 \].

Overall, the distribution in $T_N$ in our sample introduces some uncertainty in our analysis of the critical scattering in Ca$_2$RuO$_4$. Nonetheless, we find that our determination of the critical exponents of the $Q$-width, critical amplitudes, and energy-width is relatively insensitive to the details of the variance of $T_N$. Along these lines, we performed fits (not shown here) assuming a difference of $\pm 0.5$ K to our above value of $\Delta T_N = 4.84(1)$ K. The resulting values for $\nu$ and $z$ are closely similar to the above ones (difference smaller than the error bars). This is plausible since we fit the data only for $T > T_N + 4$ K, where the impact of the $T_N$ distribution is relatively small. For the critical amplitudes, where the impact of the $T_N$ distribution on the critical exponent is expected to be strongest, as we fit in the range 110-140 K, we find a deviation of only four percent. Also in the case of the critical amplitudes, the change of the critical exponent is smaller than the error bars. This result suggests that even a putative uncertainty in our value of $\Delta T_N$ would not critically affect our determination of the critical exponents, corroborating the robustness of our analysis. A definitive determination of the critical behavior close to $T_N$ will require the synthesis of large, monolithic Ca$_2$RuO$_4$ single crystals, which appears to be out of reach of the methodologies currently at hand.

For the bilayer compound Ca$_3$Ru$_2$O$_7$, which exhibits strong intra-bilayer couplings, the critical scaling was only partly compatible with the 2D-XY model. Although $\beta = 0.230(6)$ extracted from the temperature-dependence of the magnetic (001) peak matches the expected value for realistic 2D-XY systems \[ 69, 70 \], the result should be taken with caution, due to a relatively low point density around $T_{N,1}$ and a possible overlap with the signal from the transition at $T_{N,2}$. While the extracted $\beta$ is seemingly far from the corresponding value of the 3D-I model ($\beta_{3DI} = 0.327$, \[ 6 \]), previous works in the context of Sr$_2$Ir$_2$O$_7$ pointed out that a significant underestimation of $\beta$ can arise when the power law analysis is not narrowly focused around $T_N$ \[ 36 \], which provides a possible reconciliation between our small $\beta$ and the proposed 3D-I scaling. The critical scaling of the $Q$-width, amplitude, and energy-width above $T_{N,1}$ showed deviations from the 2D-XY theory (especially at low temperatures), whereas the 3D-I model captured the data comprehensively. The Ising character of the magnetic correlations likely results from the orthorhombic anisotropy, which eventually drives the magnetic transition at $T_{N,1}$. We remark, however, that in the $\Gamma$ vs. $\kappa$ plot, 3D-I scaling and the 2D-XY model were indistinguishable, due to identical dynamical critical exponents of the universality classes ($z = 2$). Consequently, the 3D-I model provides the most conclusive description of the critical behaviors in Ca$_3$Ru$_2$O$_7$, although a partial 2D-XY character can not be excluded. This ambiguity likely reflects the geometry of the exchange bonds in the bilayer structure of Ca$_3$Ru$_2$O$_7$, which is intermediate between 2D and 3D. The theoretical description of the resulting crossover phenomena and detailed comparison with the experimental data are important challenges for future research.

In conclusion, our study of the critical magnetic correlations has confirmed Ca$_2$RuO$_4$ as a realization of the 2D-XY AFM on a square lattice. Along with Sr$_2$Ir$_2$O$_7$, which hosts a nearly ideal 2D Heisenberg AFM \[ 34, 35 \], this demonstration illustrates the power of 4$d$- and 5$d$-electron materials with strongly spin-orbit-entangled magnetic moments as a platform for fundamental research on quantum magnetism.

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**Appendix A: Energy integration for Ca$_3$Ru$_2$O$_7$**

Due to the 3D character of the AFM order in Ca$_3$Ru$_2$O$_7$ \[ 55 \], the ideal energy-integration configuration with $k_f \parallel c$ in general cannot be obtained, as $Q_L$ cannot be chosen arbitrarily. To estimate the effects of
inelasticity on the experimental $Q$-width we performed a numerical simulation, assuming an intrinsic $\kappa_{in}$ and calculate $\kappa_{out}$, which nominally corresponds to the width in the experimental $Q_H$-scans. For the simulation we assume a Lorentzian $S(q) = 1/|1 + (q/\kappa_{in}(T))|^2$ with a $T$-dependent $Q$-width $\kappa_{in}(T) = \kappa_0 t^{\nu}$, with $\nu = 0.5$ and $\kappa_0 = 0.2 \text{Å}^{-1}$. Note that this choice of parameters is close to experimental values extracted from PL-fitting in the main text. Further, we assume a Lorentzian $S(\omega) = \Gamma_g/(\Gamma^2 + \omega^2)$ with a $q$-dependent energy width $\Gamma_g = \Gamma(T)[1 + (q/\kappa_{in}(T))^2]$ [27, 108, 109]. Here, the $T$-dependent energy-width is $\Gamma(T) = \Gamma_0 t^{\nu}$, with $\Gamma_0 = 1 \text{meV}$ and $z\nu = 1$. We then calculate the integral [Eqn. (5)] in the limits between $-k_B T$ and $E_i$ for each $q_i$ in the $Q_H$-scan and fit the resulting intensity with a Lorentzian with HWHM $\kappa_{out}$. The results of this simulation are shown in Fig. 13. Notably, for the two-axis mode, the reduction of the measured $\kappa_{out}$ compared to the intrinsic $\kappa_{in}$ is only of the order of a few percent. Such a change in the scaling behavior lies within our statistical error of the critical exponent $\nu$. Hence, we did not correct the two-axis data of Ca$_3$Ru$_2$O$_7$ for the integration effect. For comparison, we conducted the same simulation also for the triple-axis case [Fig. 13], where we added a Gaussian-distribution with the energy-resolution ($\pm 0.04 \text{meV}$ for $k_i = 1.3 \text{ Å}^{-1}$) as FWHM. As expected, for the latter case the difference between $\kappa_{in}$ and $\kappa_{out}$ is substantially larger.

![Figure 13](image1.png)

**FIG. 13.** Simulated ratio of $\kappa_{out}/\kappa_{in}$ vs. $T$ in the two- and three-axis mode, respectively (see details in text).

Appendix B: Critical scattering amplitude of Ca$_3$Ru$_2$O$_7$ below $T_N$

Scaling theory predicts for the magnetic susceptiblity PL behavior on both sides of $T_N$ for an ideal second-order phase transition. Specifically, $\chi = A_-|t|^{-\gamma'}$ for $T < T_N$ and $\chi = A_+|t|^{-\gamma}$ with $\gamma = \gamma'$. The resulting critical exponent $\gamma = 0.426(7)$ is slightly reduced compared to the exponent extracted from $A_+|t|^{-\gamma}$ in the range $110 - 140 \text{K}$ ($\gamma = 0.47(2)$), and still far from any universality class. This further supports the notion that the PL fit is not a proper choice for the description of the critical behavior in Ca$_3$Ru$_2$O$_7$.

![Figure 14](image2.png)

**FIG. 14.** Peak amplitude $S_0(T)/T$ of Ca$_3$Ru$_2$O$_7$. The orange line corresponds to a PL fit, with $A_-|t|^{-\gamma}$ and $A_+|t|^{-\gamma}$ for $T < T_N$ and $T > T_N$, respectively. $A_-$ and $A_+$ are the universal amplitudes. The $T_N$ distribution was also taken into account. The extracted critical exponent $\gamma = 0.426(7)$ is close to the value obtained in the main text, where only $A_+|t|^{-\gamma}$ was considered. The black vertical line indicates $T_N$ and the grey bar the variance of $T_N$.

Appendix C: Sharp background peak of Ca$_3$Ru$_2$O$_7$

Figure. 15 shows that a sharp peak at $H = 0$ is present in the scans around (H0Q$_L$). This sharp peak is independent of $T$ and Q$_L$, and thus we assign it to 2D diffuse nuclear scattering from disorder along the c-axis direction in Ca$_3$Ru$_2$O$_7$.

Appendix D: Energy-scan intensities of Ca$_3$Ru$_2$O$_4$

Figure 16 displays the integrated intensities of the fit components deduced from the energy-scans of Ca$_3$Ru$_2$O$_4$ at FLEXX. Upon heating, the intense sharp resolution limited peak decreases significantly while above $\sim 116 \text{K}$ the broad critical component is gradually taking over almost all the spectral weight besides the elastic component. For comparison we added the (1 0 0) peak intensity measured with $k = 1.5 \text{Å}^{-1}$ and normalized on the 80 K point of the entire integrated intensity. The scaling of the normalized (1 0 0) peak resembles that of the sharp component, and thus we assign the latter to elastic magnetic scattering from the (1 0 0) still present above $T_N$ due to the broad transition.
FIG. 15. (H0QL) scans in the two-axis mode at QL = 1 and 1.25 for T = 100 and 150 K. The corresponding TAS angles are: A4 (A3) = 13.8° (83.1°) and 17.3° (81.3°).

FIG. 16. Integrated intensities of the fit components in the E-scans at Q = (1 0 0) vs. T, including the overall integrated intensity (black), the Voigt-profile of the critical scattering (blue), and the resolution-limited Gaussian profile (red). For comparison, we also show the (100) peak intensity (green), measured with kf = 1.5 Å⁻¹ and normalized on the 80 K point of the overall integrated intensity (black). The horizontal gray line indicates the intensity level of the high-temperature (H-T) BG at 170 K.

Appendix E: Peak shape of Q-scans

As described in the Methods section, for the analysis of the Q-scans, we fitted the data with a Voigt-profile, i.e. a convolution of a Gaussian (instrumental resolution) and a simple Lorentzian function [Eqn. (3)]. Beyond this approximation, which is commonly used to describe critical scattering [20, 35, 36], subtle deviations from the Lorentzian form were proposed, accounting for the critical exponent η [1, 4]:

\[
\chi'(q) = \frac{\chi'(0)}{[1 + q^2(1 - \eta/2)^{-1}/\kappa^2]^{1-\eta/2}},
\]

with η ≈ 0 and η = 0.25 predicted for 3D and 2D universality classes [4], respectively. Hence, the peak-shape of the Q-scans also includes information on the critical behavior of the system. For bilayer Ca3Ru2O7, the 3D character of the critical fluctuations above TK suggests η ≈ 0 consistent with the well-matching Voigt-fits [Fig. 6]. For single-layer Ca2RuO4, one would generally expect deviations from a simple Voigt-function due to the 2D character of the fluctuations. However, we find that in our case fits with η = 0 and η = 0.25 yield peak shapes that are essentially the same within the experimental error of our data [insets in Fig. 17]. Nevertheless, we carried out the analysis of the full set of Q-scans not only for η = 0 (Ornstein-Zernike form, Eqn. (E1)), but also for η = 0.25 (2D-XY model, [11]). As can be seen in Fig. 17, the effect of η on our Q-widths is small, and is negligible with respect to the corresponding scaling behavior of κ(T). Specifically, we obtain from 2D-XY fits [Eqn. (6)] on the non-Lorentzian data (η = 0.25) for the Kosterlitz-Thouless temperature TK ≈ 88(2) K for Ca2RuO4, which coincides within the statistical errors with the value discussed in the main text. Furthermore, we remark that in general a variety of other factors such as a non-Gaussian instrumental resolution and surface effects [110, 111] can also influence the peak-shape, which generally make the determination of η challenging.

FIG. 17. Comparison of the Q-widths κ for a fitting of the Q-scans of Ca2RuO4 with η = 0 (blue points) and η = 0.25 (red points) [Eqn. (E1)]. The grey points were not included in the fits. The black vertical line indicates TK and the grey bar the variance of TK. The inset shows the corresponding fits at T = 125 K.
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