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Two-Dimensional Magnetic Correlations and Partial Long-Range Order in Geometrically Frustrated Sr_2YRuO_6

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Neutron diffraction on the double perovskite Sr_2YRuO_6 with a quasi-face-centered cubic (FCC) lattice of Ru moments reveals planar magnetic correlations that condense into a partial long-range ordered state with coupled alternate antiferromagnetic (AFM) YRuO_4 square layers coexisting with the short-range correlations below $T_{N1} = 32$ K. A second transition to a fully ordered AFM state below $T_{N2} = 24$ K is observed. The reduced dimensionality of the spin correlations is arguably due to a cancellation of the magnetic coupling between consecutive AFM square layers in FCC antiferromagnets, which is the simplest three-dimensional frustrated magnet model system.

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Geometrically frustrated magnets are fascinating materials displaying a rich variety of physical states. In these systems, magnetic moments interact by exchange, but no long-range magnetic structure is able to satisfy all antiferromagnetic (AFM) interactions simultaneously [1]. Long-range magnetic order, if present at all, occurs only for $T < T_N \ll |\Theta_{CW}|$, where Θ_{CW} is the Curie-Weiss temperature, while exotic correlated magnetic states may occur for $T_N < T < |\Theta_{CW}|$. The simplest three-dimensional structure leading to frustrated magnetism and the first one to be investigated is the face-centered cubic (FCC) lattice with antiferromagnetic (AFM) nearest-neighbor (n.n.) interactions [2]. In this case the magnetic order is theoretically predicted to be unstable down to the lowest temperatures as long as both next-nearest-neighbor (n.n.n.) interactions and magnetic anisotropy are negligible [2, 3]. Despite the fundamental interest in the FCC antiferromagnets as one of the examples of frustrated magnetism, the short-range correlations in this lattice have not been investigated in detail, to the best of our knowledge, leaving a gap in the mapping and understanding of the behavior of frustrated magnets.

Sr_2YRuO_6 (SYRO) crystallizes in the ordered double perovskite structure with Ru^{5+} ions defining an FCC magnetic network, which orders in a type-I AFM structure at low- T [4] (see Fig. 1). The magnetic order may be stabilized by weak anisotropy and/or n.n.n. interactions, which are three orders of magnitude weaker than the frustrated n.n. interactions [3]. Specific heat and magnetic susceptibility measurements indicate two phase transitions, at ~ 30 K and ~ 26 K [5], while the physical state of SYRO between these temperatures still remains to be elucidated. Additional interest in this material has been generated by the occurrence of superconductivity when Ru is partially ($< 15\%$) replaced by Cu in both powders [6–12] and single crystals [13, 14], with superconducting volume fractions below 10 %. The origin of the superconductivity in this system is still controversial, and the scenarios so-far considered are (i) a superconducting hole-condensate in the nonmagnetic SrO layers [11] and (ii) the existence of a Cu-based superconducting impurity phase [15–17]. In the present work, we performed a detailed investigation of the crystal and magnetic structure and correlations of the parent compound SYRO by means of high resolution and high intensity neutron powder diffraction. Our results indicate that the short-range magnetic correlations above $T_{N2} = 24$ K are two-dimensional (2D) and the phase between T_{N2} and $T_{N1} = 32$ K is a partially ordered state with alternated AFM ordered and disordered square layers. The 2D nature of the correlations and the possibility of a partially ordered phase should be general trends in FCC frustrated antiferromagnets.

The 13 g SYRO ceramic sample employed in this work was synthesized by solid-state reaction. Stoichiometric amounts of high-purity Ru, Y_2O_3 and SrCO_3 were ground together thoroughly in an agate mortar, placed in an alumina crucible, and fired first at 900 °C for three days in air, with two intermediate regrindings. The resulting powder was ground again, pressed into pellets and heat treated in air at 1350 °C for three days. High resolution neutron powder diffraction experiments were carried out in the BT-1 powder diffractometer at the NIST Center for Neutron Research (NCNR), using a Ge(311) monochromator with $\lambda = 2.0783$ Å and collimation of 60' before monochromator, 20' before sample and 7' before detectors. High intensity neutron scattering measurements were performed in the BT-7 double focusing triple-axis spectrometer of NCNR operated in two-axis mode using a pyrolytic graphite monochromator with $\lambda = 2.35$ Å and a position-sensitive detector [18]. In both setups, the pellets were placed inside a vanadium can attached to a high cooling power closed-cycle He cryostat with a base T of 2.6 K. Rietveld refinements were performed using the program GSAS+EXPGUI [19] under the monoclinic space group $P2_1/n$ with initial parameters taken from Ref. [4]. Weak peaks due to a minor Y_2O_3 impurity phase (0.14 % weight fraction)

and other unindexed reflections had intensities below 0.5 % of the main reflections, indicating a high purity level of our sample. In this paper, an (hkl) notation is employed for reflection indexing according to the primitive monoclinic unit cell, while $(hkl)'$ indexing refers to the quasi-cubic unit cell (see Fig. 1).

Figure 1 shows the high-resolution neutron powder diffraction pattern of SYRO at 3 K as a function of $Q \equiv (4\pi/\lambda) \sin \theta$. Besides the nuclear Bragg peaks, sharp magnetic reflections were observed below $T_{N1} = 32$ K, with no observable additional broadening with respect to the nuclear peaks. These contributions were initially modeled as a type-I AFM long-range ordering (LRO) of the Ru magnetic moments, in accordance with ref. [4]. A statistically significant improvement in the fitting was obtained by including oxygen moments in the magnetic structure model ($\chi^2 = 1.895$ against $\chi^2 = 1.923$ for the model with pure Ru moments), as suggested by Mazin and Singh based on first principle calculations [20]. The oxygen magnetic structure is such that each oxygen moment is parallel to its nearest Ru moment, consistent with a Ru $4d$ - O $2p$ hybridization mechanism. In addition, our resolution was sufficient to resolve the positions of the (100) and (010) neighboring reflections, revealing contributions solely from (100). The absence of the (010) magnetic reflection allows us to unequivocally determine the moment direction to be along the principal monoclinic axis [010] for all studied temperatures in the ordered phase. Indeed, the intensity ratio between the main magnetic reflections remains unaltered below T_{N1} . The calculated pattern at 3 K after crystal and magnetic structure refinements is given in Fig. 1, showing very good agreement with the observed data. The refined magnetic moment magnitudes are $1.96(2) \mu_B/\text{Ru}$ and $0.056(6) \mu_B/\text{O}$ at 3 K, which are comparable to $1.7 \mu_B/\text{Ru}$ and $0.10 \mu_B/\text{O}$ predicted by first principle calculations [20]. Error bars in parentheses are statistical only and represent one standard deviation. The high sensitivity of our experiment to O moments arises from interference of the scattering by the six O moments per formula unit with the contribution by the Ru moments, leading to measurable effects in the relative Bragg intensities. The similar Ru and Y scattering lengths for both neutrons and x-rays prevent a direct estimation of the Ru/Y anti-site disorder [22]. Nonetheless, the very different refined Ru-O and Y-O average bond distances (1.954 and 2.206 Å, respectively, see [21]) and the reasonably small oxygen Debye-Waller parameters ($U_{iso} \sim 0.005$ Å²) at low- T indicate a negligible degree of Y/Ru chemical disorder, which is favoured by the largely different Shannon radii of Y³⁺ and Ru⁵⁺ ions [22, 23]. No structural phase transition is observed between 3 and 45 K, although small lattice parameter anomalies are noticed below T_{N1} [21], probably due to exchange striction.

Figure 2(a) shows the high-intensity neutron scattering profile for $0.35 \text{ Å}^{-1} < Q < 1.4 \text{ Å}^{-1}$ at selected temperatures. At 300 K, only incoherent scattering due to uncorrelated magnetic moments is observed as expected for a paramagnetic phase. At 3 K, Gaussian peaks with widths given by the instrumental resolution are seen at the magnetic Bragg positions consistent with the type-I AFM structure (see above). At intermediate temperatures, both below and above T_{N1} , a broad and asymmetric contribution to the scattering is also observed, peaked at the position of the (001)' Bragg reflection. This is more clearly viewed in the data shown in Fig. 2(b) at 35 K, obtained after a background subtraction procedure [24]. Careful inspection of the data over an extended Q -range (not shown) indicates the diffusive scattering at 35 K is almost totally concentrated at low Q , which is indicative of the magnetic nature of this signal. On the other hand, isotropic type-I AFM correlations should also lead to a strong contribution at the (110)' position at $Q \sim 1.09 \text{ Å}^{-1}$. However, attempts to fit the asymmetric profile shown in Fig. 2(b) with two peaks centered at (001)' and (110)' positions were unsuccessful. In fact, no scattering component centered at the (110)' position is detectable in our experiment at 35 K. This is consistent with magnetic scattering from 2D AFM square layers (see Fig. 1), in which the (10)' reflection is expected at $Q_0 \sim 0.77 \text{ Å}^{-1}$ and the (11)' reflection at $Q \sim 1.09 \text{ Å}^{-1}$ is forbidden. Indeed, the asymmetric shape of the diffusive scattering is characteristic of 2D diffraction [25, 26]. In the limit of very large 2D layers, the powder-averaged diffraction from truncation rods shows a resolution-limited jump from zero to maximum intensity at $Q = Q_0$ and a slow falloff for $Q > Q_0$ [25, 26]. For finite in-layer correlation length L , scattering also occurs in the lower Q side of the peak, due to particle size broadening. The observed intensities in Fig. 2(b) were captured by this model [25, 26] assuming scattering planes with two possible values of L ($L_1 = 23$ Å and $L_2 = 130$ Å at 35 K, see thick solid line in Fig. 2(b)). The scattering component with large L is identified by the relatively sharp contribution in close vicinity of the intensity maximum, while the other component with much shorter L must be invoked to explain the slow intensity falloff for $Q < Q_0$ (see Fig. 2b). Despite the good fit to the data, this model is likely a simplification of the more plausible scenario of planar spin correlations with a broad and continuous distribution of L 's. An estimation of the average L for each temperature could be obtained from fits of the low Q side of the scattering to a Lorentzian lineshape, $I(Q) \propto |f_M(Q)|^2 / [4(Q - Q_0)^2 + w_l^2]$, where $f_M(Q)$ is the Ru magnetic form factor and $w_l = 0.105(5) \text{ Å}^{-1}$ at 35 K, yielding $\bar{L} = 2\pi/w_l = 60(3) \text{ Å}$ (see Fig. 2(b)).

The temperature dependencies of the distinct components of the magnetic scattering are given in Figs. 3(a-f), taken from the high-intensity data. The intensity of the (110)' magnetic Bragg reflection is given in Figs. 3(a) and (b). Besides the onset of the long-range magnetic order at T_{N1} , a kink in the LRO parameter with an enhancement of the intensity below $T_{N2} = 24$ K is noticed. Figures 3(c) and (d) show the T -dependence of the integrated scattering intensity for $0.82 \text{ Å}^{-1} < Q < 0.96 \text{ Å}^{-1}$, which is an interval dominated by scattering from 2D correlations (see Fig.

2(b)). These correlations are observed below ~ 200 K, and enhance on cooling down to T_{N1} . In the ordered phase, the signal from 2D correlations decreases on cooling and nearly disappears below $\sim T_{N2}$. The suppression of the 2D correlations and simultaneous enhancement of the LRO component seem to be related with the phase transition at $\sim T_{N2}$ captured by previous specific heat measurements [5]. Figure 3(e) shows the integrated intensity for $0.35 \text{ \AA}^{-1} < Q < 0.47 \text{ \AA}^{-1}$, representing the development of incoherent magnetic scattering from uncorrelated paramagnetic moments. The scattering weight from the uncorrelated paramagnetism is progressively suppressed on cooling from 300 K, being transferred to the scattering by 2D correlations. Figure 3(f) displays $\bar{L}(T)$, obtained from the Lorentzian fits illustrated in Fig 2(b), showing a maximum at $T \sim T_{N1}$.

The 2D magnetic correlations in a three-dimensional lattice is a consequence of the inherent geometric frustration associated with the FCC Ru network. In fact, the type-I AFM structure of SYRO may be visualized as a stacking of AFM layers in the *ABAB* sequence (see Fig. 1). Within a given AFM layer all the nearest-neighbor (n.n.) Ru-Ru interactions are satisfied, while between consecutive layers only half of the n.n. interactions are satisfied and the other half are frustrated. This leads to a cancellation of the coupling energy between consecutive *A* and *B* AFM layers that is not restricted to n.n. interactions, remaining valid for bilinear Ru(*A*)-Ru(*B*) interactions of any range. Therefore, the observed 2D correlations in SYRO are not entirely surprising. In fact, 2D magnetic fluctuations were theoretically predicted for the FCC lattice [27].

SYRO enters into an interesting state between T_{N1} and T_{N2} . In fact, as suggested by the solid line in Fig. 3(b), the $(110)'$ intensity may be decomposed as the sum of two components with identical weights, one showing a transition at T_{N1} and the other at T_{N2} . On the other hand, the intensity ratio between the main magnetic reflections remains constant and the high-resolution diffraction data could be fit well by a simple type-I AFM structure for all temperatures below T_{N1} , which at first sight might argue against a LRO magnetic transition at T_{N2} . However, these observations can be reconciliated by an intermediate AFM phase for $T_{N2} < T < T_{N1}$, where only alternate AFM square layers are long-range ordered. This phase shows the same magnetic Bragg reflections of the fully ordered type-I AFM phase, however with half of the intensity for each reflection, consistent with Fig. 3(b). Additional evidence of the partially ordered phase is the persisting signal from short-range correlations between T_{N1} and T_{N2} , presumably arising from layers that do not participate in the LRO. A similar partially ordered state has been previously proposed for the FCC antiferromagnet GdInCu₄ [28], indicating this may be a general trend for this geometry, possibly favored by n.n.n. interactions that may couple alternate but not consecutive layers (see Fig. 1).

Our results in stoichiometric SYRO call for a rediscussion of some earlier data obtained in Sr₂YRu_{1-x}Cu_xO₆. Neutron diffraction studies for $x = 0.15$ indicated a magnetic signal up to high- T , which was attributed to Cu ordering with $T_N(\text{Cu}) \sim 86$ K in addition to $T_N(\text{Ru}) \sim 30$ K [11]. It is evident, though, that the persisting magnetic scattering above 30 K may be due to correlated Ru moments (see Fig. 2(a)), and it may not be possible to establish unambiguously whether Cu spins indeed contribute to the ordered magnetism of the doped samples. Also, a relatively small ordered moment of $1.13 \mu_B/\text{formula unit}$ was observed in Sr₂YRu_{0.85}Cu_{0.15}O₆ [11], suggesting that the partially ordered phase revealed here for pure SYRO might be also present in the Cu-doped samples even at low- T . Further studies in the Cu-doped samples are necessary to confirm or dismiss this hypothesis.

Although most ingredients discussed here for SYRO should be present for any FCC antiferromagnet, some signatures of the exotic $4d/5d$ magnetism are found in this compound. Particularly, the Ru-Ru magnetic correlations are observed up to ~ 200 K, even though the n.n. Ru-Ru distance is quite large, $d(\text{Ru-Ru}) \sim 5.77 \text{ \AA}$. This is due to the extended character of the Ru($4d$) electrons, leading to a strong Ru-O-O-Ru superexchange coupling [3] and a significant oxygen magnetic moment (see above). In fact, we anticipate that double perovskites with $4d$ or $5d$ magnetic ions alternated with a non-magnetic cations in the center of oxygen octahedra are excellent candidates to present frustrated magnetism in its simplest and most fundamental geometry in three dimensions, namely the FCC lattice. This is because n.n.n. interactions are very small in such “diluted” magnetic materials, underpinning the interesting phenomena arising from the frustration of n.n. interactions in this lattice.

In summary, neutron powder diffraction experiments in SYRO with quasi-FCC crystal structure reveal planar AFM correlations above ~ 24 K and a partially ordered state between 24 and 32 K. The reduced dimensionality of the spin correlations is argued to be a direct consequence of the inherent geometrical frustration of antiferromagnetism in this lattice, and therefore should be a general trend in FCC antiferromagnets.

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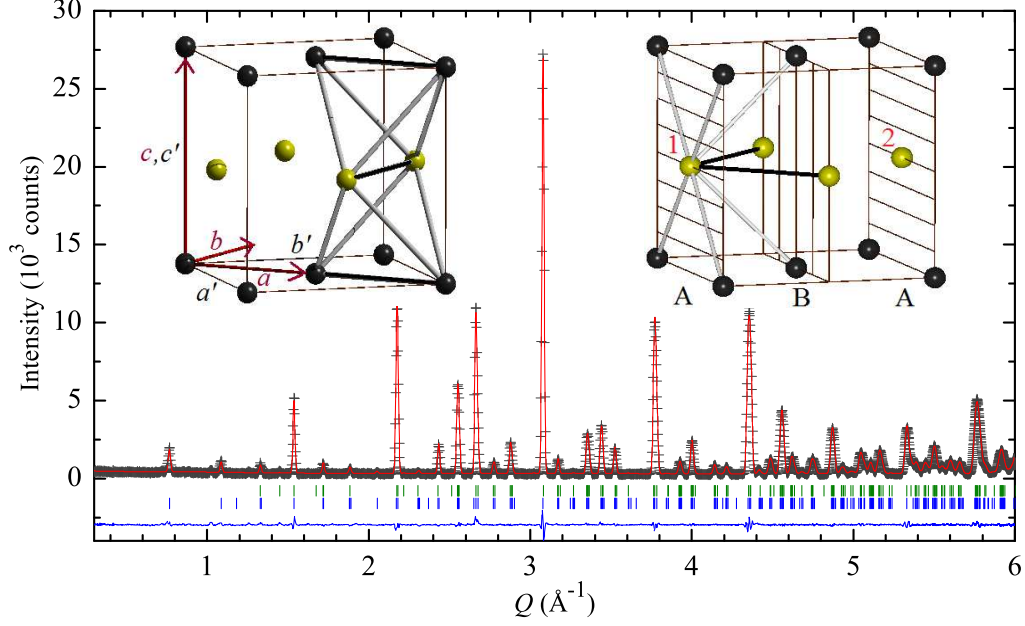


FIG. 1: (Color online) High resolution neutron powder diffraction pattern of Sr_2YRuO_6 at 3 K. The cross symbols and solid lines represent observed and calculated patterns, respectively. The difference curve is shown at the bottom. Vertical bars indicate the expected Bragg peak positions according to the nuclear (upper) and magnetic (lower) structure models described in the text and refined parameters given in [21]. The fitting residuals are $\chi^2 = 1.895$, $R_{wp} = 4.35\%$, and $R_p = 3.45\%$. The insets illustrate the observed type-I antiferromagnetic (AFM) structure of Sr_2YRuO_6 , where only the magnetic Ru ions are displayed and different shades (colors) represent opposite magnetic moments. Two representative edge-shared Ru tetrahedra, which are the basic units leading to geometric frustration in this system, are illustrated in the left, also showing the monoclinic axes with lengths a , b , and c and quasi-cubic unit cell ($a' \sim b' \sim c'$). Satisfied (frustrated) nearest-neighbor AFM interactions are represented as gray (black) bonds. In the right inset, the same magnetic structure is visualized in terms of consecutive AFM square layers in the $ABAB$ sequence (shaded). The net magnetic coupling between consecutive AB layers is null, while alternate AA layers are coupled by weak next-nearest-neighbor exchange interactions such as those between atoms 1 and 2 (see text).

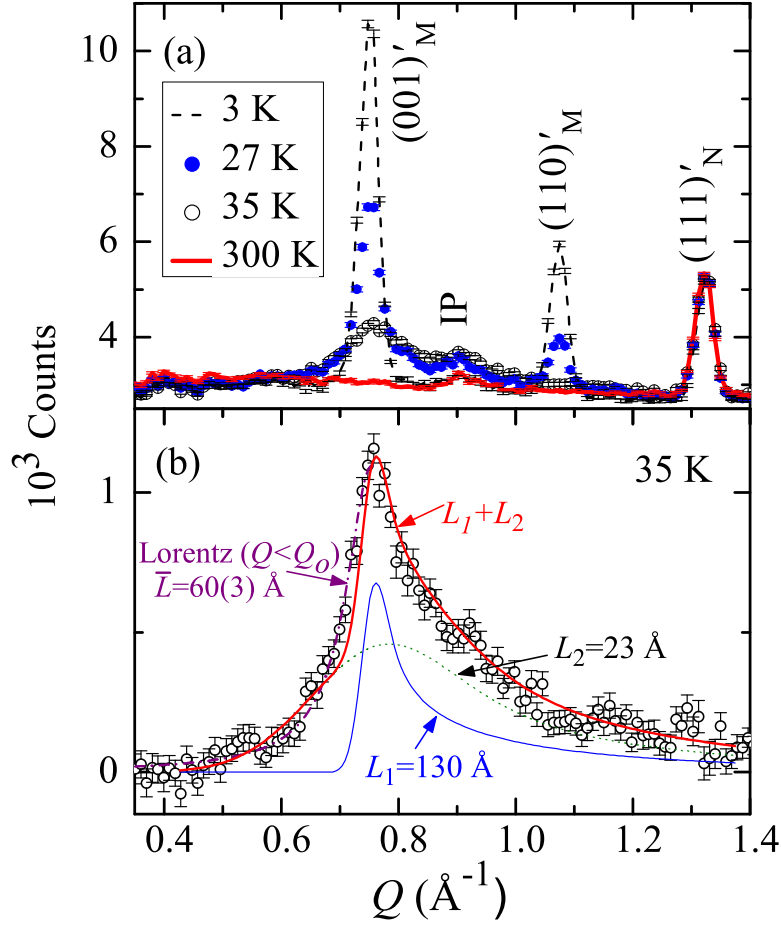


FIG. 2: (Color online) (a) High-intensity diffraction pattern of Sr_2YRuO_6 at selected temperatures. The indexing of magnetic and nuclear Bragg reflections according to the quasi-cubic unit cell of Fig. 1 is indicated, as well as a weak peak due to a minor unidentified impurity phase (IP). (b) Magnetic diffuse scattering at 35 K (symbols) after subtraction of the non-magnetic background [24]. The thin solid and dotted lines are simulations using a two dimensional scattering model with lateral widths $L_1 = 130$ and $L_2 = 23 \text{ \AA}$, respectively, and the thick solid line is the sum of these contributions. The dashed line is a fit of the low- Q side of the scattering to a Lorentzian lineshape, yielding an average in-plane correlation length $\bar{L} = 60(3) \text{ \AA}$.

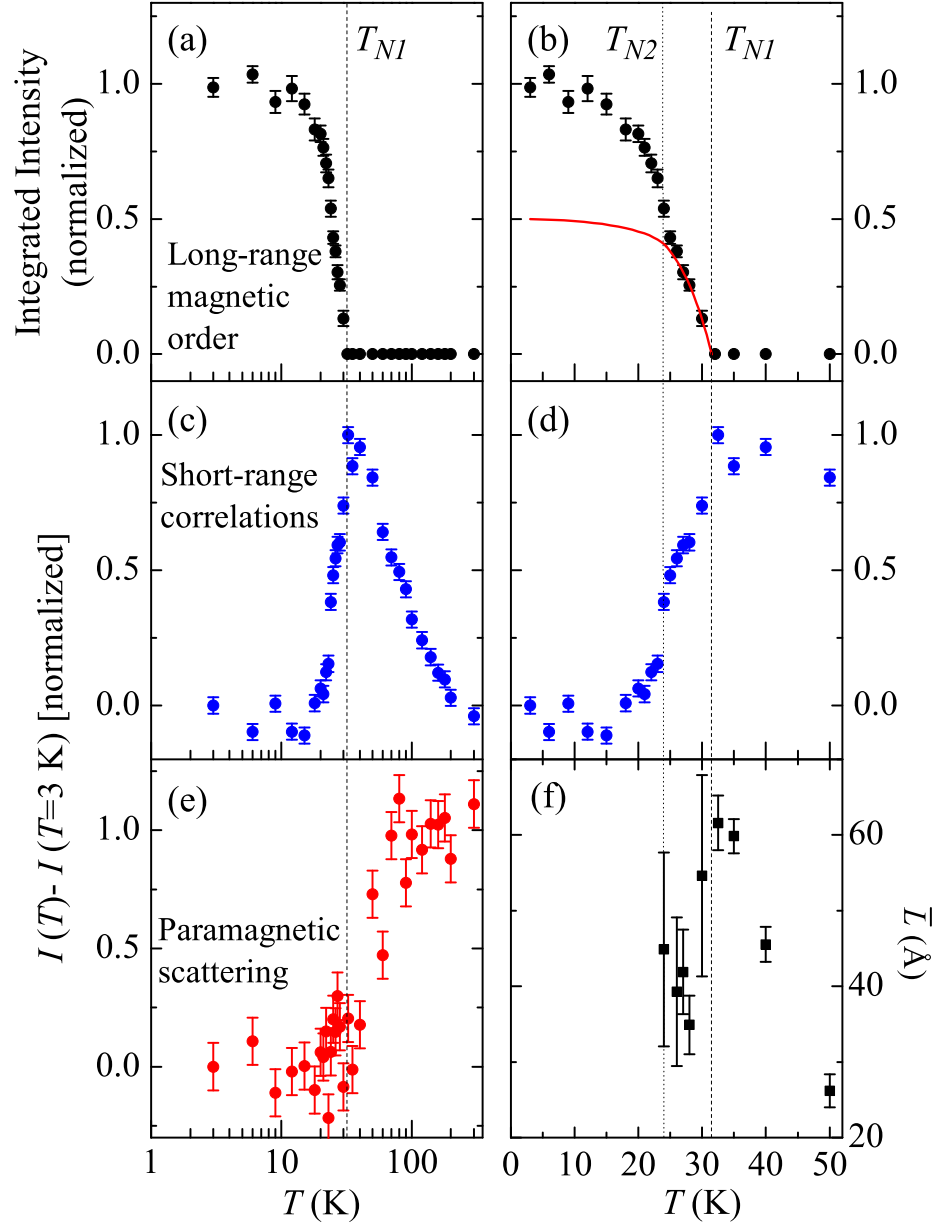


FIG. 3: (Color online) (a,b) Integrated intensity of the $(110)'$ magnetic reflection on both log and linear temperature scales, highlighting the long-range order parameter. The data in (b) clearly indicate there are two components to the scattering with distinct transition temperatures, and the solid line is a guide to the eye; (c,d) integrated scattering for $0.82 \text{ \AA}^{-1} < Q < 0.96 \text{ \AA}^{-1}$, capturing signal from short-range magnetic correlations; (e) integrated scattering for $0.35 \text{ \AA}^{-1} < Q < 0.47 \text{ \AA}^{-1}$, showing the development of weakly correlated spins in the paramagnetic state at high temperatures; (f) average in-plane antiferromagnetic correlation length \bar{L} obtained from half-Lorentzian fits to the diffuse scattering component for $Q < 0.77 \text{ \AA}^{-1}$ (see text and Fig. 2(b)).