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Importance of XY anisotropy in Sr_2IrO_4 revealed by magnetic critical scattering experiments

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The magnetic critical scattering in Sr_2IrO_4 has been characterized using x-ray resonant magnetic scattering (XRMS) both below and above the three-dimensional antiferromagnetic ordering temperature T_N. The order parameter critical exponent below T_N is found to be $\beta = 0.195(4)$, in the range of the two-dimensional (2D) XYh_4 universality class. Over an extended temperature range above T_N, the amplitude and correlation length of the intrinsic critical fluctuations are well described by the 2D Heisenberg model with XY anisotropy. This contrasts with an earlier study of the critical scattering over a more limited range of temperature, which found agreement with the theory of the isotropic 2D Heisenberg quantum antiferromagnet, developed to describe the critical fluctuations of the conventional Mott insulator La_2CuO_4 and related systems. Our study therefore establishes the importance of XY anisotropy in the low-energy effective Hamiltonian of Sr_2IrO_4, the prototypical spin-orbit Mott insulator.

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The Ruddlesden-Popper series $Sr_{n+1}Ir_nO_{3n+1}$ of perovskite iridates has emerged as a fruitful arena in which to explore the effects of electron correlations in the strong spin-orbit coupling limit. The first two members of this series, single-layer Sr_2IrO_4 ($n = 1$) and bilayer $Sr_3Ir_2O_7$ ($n = 2$), are believed to exemplify a new class of spin-orbit Mott insulators. Of central importance to our understanding of these materials is the emergence of a $j_{\rm eff} = 1/2$ ground state by the combined action of a strong cubic crystal field and spin-orbit interactions on the $5d^5$ electrons of the Ir^{4+} ions [1]. The weakened electron correlations typical of the $5d$ elements then split the $j_{\rm eff} = 1/2$ band, opening a gap, leading to a Mott-like state.

Sr_2IrO_4 in particular has attracted considerable attention because of its striking similarities to La_2CuO_4 in terms of both its structural and magnetic properties. The magnetic structures and excitations of Sr_2IrO_4 have been investigated in a number of x-ray resonant magnetic scattering (XRMS) studies [1–7] which have allowed an effective low-energy Hamiltonian to be proposed and refined. Sr_2IrO_4 forms an antiferromagnetic structure below $T_N \sim 225$ K in which the moments are confined to the $\mathbf{a} - \mathbf{b}$ planes and canted to follow rigidly the correlated rotation of the oxygen octahedra of the $I4_1/acd$ crystal structure [8]. A resonant inelastic x-ray scattering (RIXS) experiment [2] has revealed a dispersion relation somewhat reminiscent of that displayed by La_2CuO_4, albeit with a lower-energy scale and much stronger further neighbor couplings, which can be derived from a smaller ratio of on-site repulsion over hopping amplitude [9]. This result suggests that the low-energy isospin dynamics of the $j_{\rm eff} = 1/2$ states in Sr_2IrO_4 may, to leading order, be mapped onto an effective isotropic two-dimensional Heisenberg Hamiltonian, in agreement with predictions by Jackeli and Khaliullin [10].

Critical scattering studies provide information complementary to that obtainable from the ordered state. For thermally driven transitions in classical systems, issues such as dimensionality, relevant anisotropies, etc., can be addressed by determining the critical exponents both below and above the transition temperature [11]. One question that naturally arises is, in what ways, if any, do the critical fluctuations of a lattice decorated by $j_{\rm eff} = 1/2$ isospins differ from the case of $S = 1/2$ spins? For La_2CuO_4 and related two-dimensional (2D) Cu^{2+} ($S = 1/2$) systems it has been established that the instantaneous magnetic scattering function $S(\mathbf{Q})$ is well described by the critical properties of the 2D quantum $S = 1/2$ Heisenberg antiferromagnet on a square lattice (2DQHAFSL) [12–16]. The 2DQHAFSL model itself has been extensively studied using a range of theoretical and computational techniques, with results that are in broad agreement within the range of applicability of the assumptions used [17–20]. Here, for the sake of brevity, we compare our data to the results obtained by quantum Monte Carlo (QMC) techniques [18].

Fujiyama *et al.* [21] have measured the critical magnetic scattering from Sr_2IrO_4 using XRMS in the interval T_N to $T_N + 25$ K. Their principal results are that the critical fluctuations are 2D over the entire temperature interval investigated, with an evolution of the in-plane correlation length consistent with that expected for the 2DQHAFSL model [17,18]. The latter was interpreted by Fujiyama *et al.* as evidence in favor of the theoretical proposition by Jackeli and Khaliullin [10],

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that the full isospin Hamiltonian (including anisotropies other than that due to Hund's coupling) can be mapped onto an effective, isotropic Heisenberg Hamiltonian. However, the good agreement reported between data and predictions of the 2DQHAFSL model required a value of the nearest-neighbor exchange coupling of $J = 100(10)$ meV which they deduced by fitting their data. A subsequent RIXS study [2] of the one-magnon dispersion in Sr_2IrO_4 provided the value $J = 60$ meV, and a next-nearest-neighbor ferromagnetic coupling $J' = -20$ meV, seemingly difficult to reconcile with the data of Fujiyama *et al.*

Here, we report a comprehensive XRMS study of the critical magnetic fluctuations in Sr_2IrO_4 for temperatures both below and above T_N. Our results provide decisive evidence for the importance of XY anisotropy in the exchange interactions for Sr_2IrO_4. Below T_N, accurate values are obtained for the order parameter β, revealing that Sr_2IrO_4 fits into the 2D XY universality class with h_4 anisotropy. Above T_N we greatly extend the temperature window over which the critical scattering has been measured to $T_N + 73$ K, and carefully apply corrections for the finite instrumental resolution. We establish that, in contrast to an earlier experimental study for Sr_2IrO_4 [21], the intrinsic isospin fluctuations for $T > T_N + 5$ K are dominated by XY type anisotropy.

The critical scattering experiments were performed on beamline I16, Diamond Light Source, UK, and P09, PETRA III, DESY, Germany. These experiments exploited the large enhancement of the x-ray resonant magnetic scattering cross section at the Ir L_3 edge. The single crystals of Sr_2IrO_4 used in this study (dimensions $2 \times 1 \times 0.05$ mm^3) were flux grown from phase-pure polycrystalline Sr_2IrO_4 using techniques described elsewhere [22], and attached to the copper sample mount of a closed-cycle refrigerator. This was in turn mounted on a six-circle diffractometer configured to operate in a vertical scattering geometry. The energy of the incident photon beam was set to 11.218 keV, just below the L_3 edge of iridium, a value found to maximize the intensity of the x-ray resonant magnetic scattering. The incident beam size was 200×20 μm^2, and the polarization of the scattered x rays was determined by using a Au (333) crystal analyzer mounted on the detector arm. The temperature was measured to a precision of ± 0.01 K via a thermocouple secured to the sample mount by Teflon tape. The wave-vector resolution of the instrument, including the effects of sample mosaic, was determined by mapping Bragg peaks in reciprocal space and was found to be typically 1.1×10^{-3} and 3.7×10^{-3} Å^{-1} perpendicular and parallel to $\mathbf{Q}$ in the scattering plane, respectively, and 1.4×10^{-3} Å^{-1} out of the plane.

The first objective was to determine T_N and β from the temperature dependence of the magnetic peak intensity I_M below T_N. The results are summarized in Fig. 1. A fit to the simple power-law form $I_M \propto [(T_N - T)/T_N]^{2\beta}$ is shown in Fig. 1(a), which yields $\beta = 0.195(4)$. This value is consistent with that provided by neutron scattering measurements [23,24]. The value of β deduced by this analysis deviates significantly from theoretical values for both 2D Ising ($\beta = 1/8$) and three-dimensional (3D) systems ($\beta \sim 0.35$), but rather is consistent with value for the 2D XYh_4 universality class in the strong anisotropy limit [25].

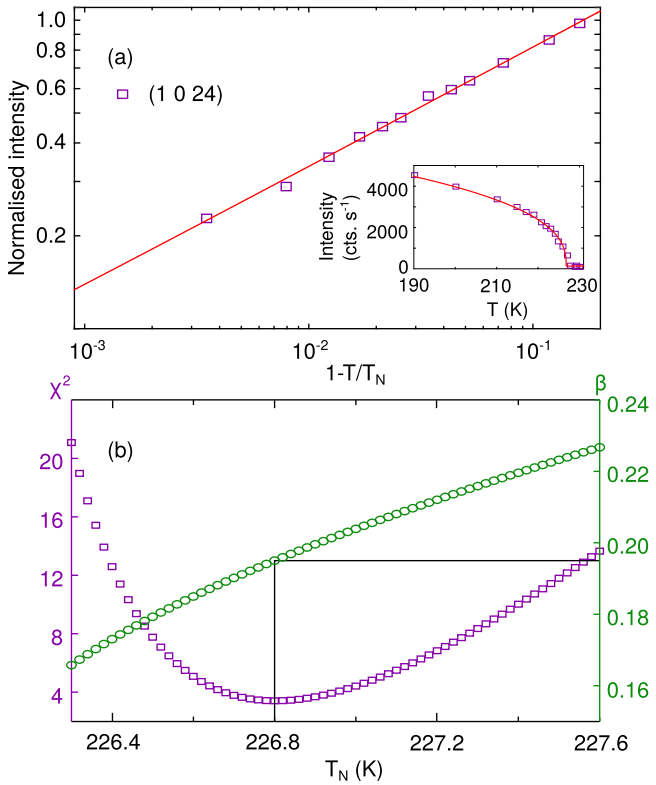

FIG. 1. (Color online) Order parameters of the magnetic critical scattering in Sr_2IrO_4: (a) Integrated intensity and power-law fit (solid line) corresponding to best-fit values of $T_N = 226.80(6)$ K and $\beta = 0.195(4)$. (b) Goodness of fit χ^2 (purple squares) and the resulting exponent β (green circles) as a function of the chosen ordering temperature T_N. The fit in (a) corresponds to the minimum in χ^2.

Next, a detailed investigation of the critical scattering above T_N was undertaken. For these studies the high photon energy and relatively broad energy resolution of x-ray diffractometers ($\gtrsim$1 eV) offer an advantage over neutrons in that they provide an accurate frequency integration to yield the instantaneous magnetic scattering function $S(\mathbf{Q})$. On the other hand, the intrinsic high wave-vector resolution of x-ray techniques presents a challenge in terms of following weak critical magnetic scattering to high temperatures as it broadens and weakens further. X-ray experiments to determine the magnetic critical scattering above T_N have revealed that the critical fluctuations just above T_N have two components: a sharp "central" peak, typically with a Lorentzian squared line shape, believed to be an extrinsic feature due to the presence of defects, and a broader, weaker peak with a Lorentzian line shape arising from intrinsic critical fluctuations [11,26]. The realization that the magnetic critical scattering above T_N can have two such components mirrors earlier results for structural phase transitions [27–29].

Representative scans of the magnetic critical scattering above T_N are shown in Fig. 2. The critical scattering could be followed out to $T_N + 73$ K, roughly trebling the region probed above T_N in Ref. [21], which, as we shall show, places a much tighter constraint on the relevant form of the effective Hamiltonian. The critical scattering above T_N was fitted with

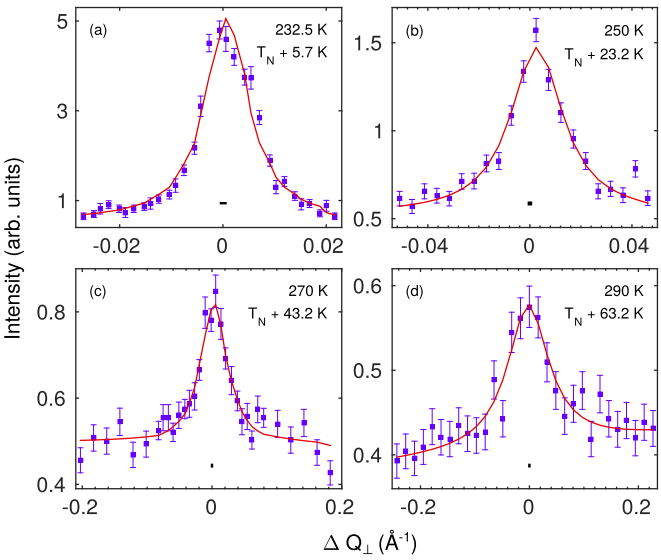

FIG. 2. (Color online) Critical scattering data for $T > T_N$ obtained in scans performed perpendicular to the wave-vector transfer $\mathbf{Q}$ of the appropriate antiferromagnetic Bragg peak. Solid lines: Fits of a Lorentzian line shape convoluted with the experimental resolution. The horizontal bars under the peaks represent the resolution full width at half maximum.

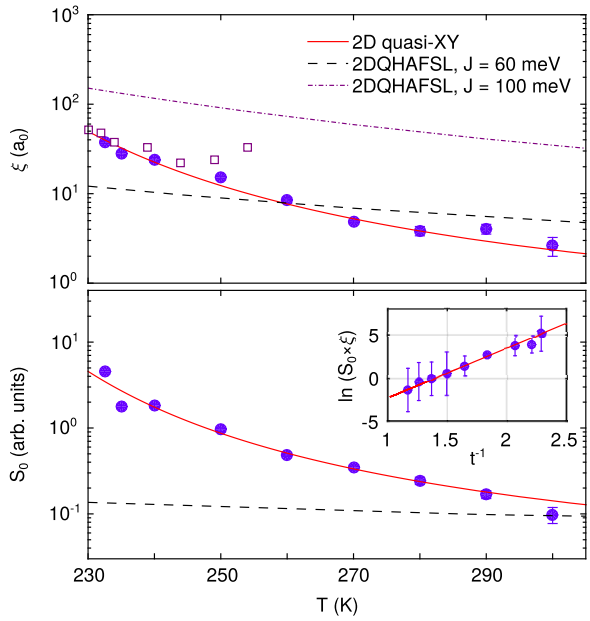

FIG. 3. (Color online) Temperature dependence of the correlation length (top panel) and the amplitude (bottom panel) of the critical scattering for Sr_2IrO_4 as solid circles. Open squares are data taken from Fujiyama [21] divided by a_0. QMC results with $J = 60$ meV (100 meV) as a dashed line (dotted-dashed line) Ref. [18]. Solid line: Fit of a 2D model with XY anisotropy, as discussed in text. Inset: Linear scaling of $\ln(\xi S_0) \sim (3-\eta)bt^{-1}$.

several different line shapes convoluted with the instrumental resolution function. The most satisfactory over most of the temperature range was a Lorentzian, as expected for intrinsic magnetic critical scattering. However, for $T_N < T < T_N + 5.5$ K, a sharper, second component appeared to develop, presumably due to extrinsic, defect mediated scattering. As it proved difficult to obtain unique fits in this temperature interval, we do not further consider the data taken close to T_N.

In Fig. 3 we compare our data with various theories and earlier experimental results [30]. The dashed lines represent QMC results for the 2DQHAFSL model for which $\xi(T) = 0.276\, a_0\, e^{1.25J/k_BT}$ and $S_0 \propto \xi^2 T^2$, where $a_0 = 3.9$ Å is the Ir-Ir nearest-neighbor distance. It is evident that in the 2D regime the values of ξ and S_0 of the intrinsic Lorentzian component have much stronger temperature dependences than the expectations of the 2DQHAFSL. The 2DQHAFSL model cannot therefore describe the critical fluctuations in Sr_2IrO_4, whatever value of J is chosen. This important realization disagrees with the main conclusion of Ref. [21].

We therefore considered alternative models. In the 2D XY model a phase transition occurs at the Kosterlitz-Thouless temperature T_{KT} [31]. Above T_{KT} the correlation length ξ and scattering amplitude S_0 are described by $\xi \propto \exp(b/\sqrt{t})$ and $S_0 \propto \exp[b(2-\eta)/\sqrt{t}]$, where b is a nonuniversal constant (typically $b \approx 1.9$), $\eta = 1/4$, and $t = (T - T_{KT})/T_{KT}$ the reduced temperature [32]. For real systems, three-dimensional order at a temperature $T_N > T_{KT}$ occurs as a result of weak interlayer coupling J' amplified by the diverging size of correlated regions $\xi(T)^2$. However, attempts to fit the pure 2D XY model to our data yielded unphysical values of $b = 3.0(7)$ and $J'/J = 5 \times 10^{-6}$, and were thus discarded.

Instead, a consistent description of our data was obtained using the 2D anisotropic Heisenberg model (2DAH), where leading Heisenberg interactions are augmented by easy-plane, XY anisotropy [33]. The 2DAH model also has a correlation length that diverges exponentially towards a finite temperature T_{KT}, placing it in the 2D XY universality class. In the limit of long correlation lengths, ξ and S_0 scale the same way as for the pure 2D XY model. However, with increasing temperature there is a crossover towards $\xi(T) = \xi_0 \exp(b/t)$ and $S_0 \propto \exp[b(2-\eta)/t]$ for $\xi \lesssim 100 a_0$ [33]. Our data fall in this regime.

The solid lines in Fig. 3 show the fit to the 2DAH, which is seen to provide an excellent description of the data in the 2D regime. From the fits we obtain $\xi_0 = 0.9(1)$ Å, $T_{KT} = 162(10)$ K, and $b = 2.1(7)$, with the latter in good agreement with theory and experiment for other layered materials [32,34,35]. From the value of the 2D correlation length at T_N it is possible to estimate the ratio of the in-plane and interlayer couplings $J'/J \simeq \xi(T_N)^{-2} = \exp(-2b/T_N) \approx 0.001$. This ratio is an order of magnitude higher than for the cuprates [12,13], which can be understood by comparing the shapes of the ground state orbitals of Sr_2IrO_4 and La_2CuO_4. The $j_{eff} = 1/2$ state is approximately cubic, whereas for La_2CuO_4 the $d_{x^2-y^2}$ orbital lies predominantly in the xy plane. Thus, larger out-of-plane interactions would be expected for Sr_2IrO_4 compared with La_2CuO_4. The description of $\xi(T)$ by inclusion of XY anisotropy is consistent with the extracted value of $\beta = 0.195(4)$, which places the fluctuations below T_N in the 2D XYh_4 universality window, where a fourfold in-plane anisotropy pushes β from 0.231 for 2D XY towards $\beta = 1/8$ for 2D Ising [25] interactions.

In general, the effect of XY anisotropy on the spin-wave dispersion is to lift the degeneracy between the Heisenberg modes. The in-plane fluctuations $\omega_{\parallel}$ maintain a continuous symmetry and are hence gapless (Goldstone mode), whereas the out-of-plane fluctuations $\omega_{\perp}$ are gapped. At the zone center the out-of-plane spin-wave gap is given by $\omega_0 = 4JSZ_c\sqrt{2\Delta_\lambda}$, where $0 \leqslant \Delta_\lambda \leqslant 1$ parametrizes the easy-plane anisotropy [36]. In the data from a recent high-resolution RIXS experiment on Sr_2IrO_4 [5], there appears to be a magnon gap of ∼30 meV at the Brillouin zone center, qualitatively consistent with the existence of XY anisotropy. The degree of XY anisotropy required to produce a gap of this size may be determined by extending the model of Kim *et al.* [2] to include anisotropic exchange. The resulting 2DAH Hamiltonian reads

$$\mathcal{H}_{ab} = \sum_{\langle i,j\rangle} \tilde{J}\left[S_i^x S_j^x + S_i^y S_j^y + (1-\Delta_\lambda) S_i^z S_j^z\right] + \sum_{\langle\langle i,j\rangle\rangle} J_2 \vec{S}_i \vec{S}_j + \sum_{\langle\langle\langle i,j\rangle\rangle\rangle} J_3 \vec{S}_i \vec{S}_j. \quad (1)$$

Here, $\tilde{J} = J_{\rm iso}/(1-\Delta_\lambda)$ is the effective nearest-neighbor (NN) exchange parameter, $J_{\rm iso}$ is the isotropic Heisenberg NN exchange, and J_2, J_3 symbolizes the exchange between next-nearest and third-nearest neighbors, respectively. Out-of-plane exchange coupling has been neglected since, as demonstrated above, it is a factor of $\sim 10^3$ weaker than the in-plane terms, and thus not resolvable with RIXS at present.

We fitted the theoretical dispersion of this 2DAH model to the experimental data from Ref. [5] using a linear spin-wave model derived from Eq. (1), with the results shown in Fig. 4. There is good agreement between the experimental dispersion and that calculated from the anisotropic model, with the fitted exchange constants essentially identical to those obtained from the earlier low-resolution study within the error bars [2]. The agreement for the spectral weight is perhaps less compelling but can be said to be qualitatively reasonable, in particular, the model reproduces the nonzero spectral weight at the zone center. Moreover, it proved difficult to extract the spectral weight from the RIXS data [5] as there appears to be a significant variation of line shape as a function of momentum transfer, which may be evidence for an additional excitation mode between the single magnon and bimagnon peaks (the latter was accounted for in the fitting procedure). Fitting the dispersion and intensities to the localized spin model proposed by Igarashi [37] provided similar results, corroborating their prediction of an out-of-plane gap for Sr_2IrO_4. The easy-plane anisotropy parameter $\Delta_\lambda = 0.08(1)$ determined here should be considered as an upper bound to the true anisotropy, since the finite momentum and energy resolution means that the excitation energy at the zone center cannot be fully resolved. Nevertheless, this is significantly larger than that observed for La_2CuO_4 [$\Delta_\lambda = 2.0(5) \times 10^{-4}$] [38], which further illustrates the relative importance of XY anisotropy for Sr_2IrO_4.

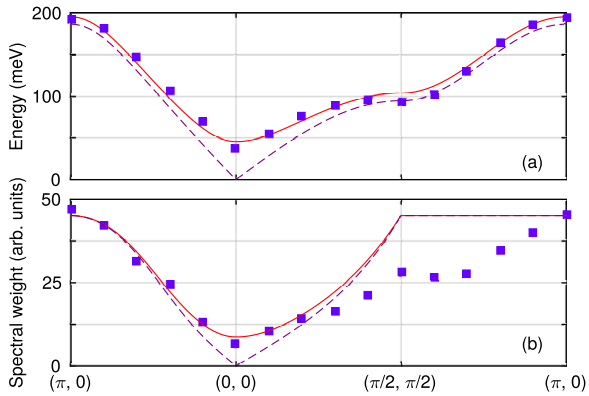

FIG. 4. (Color online) Fit of the anisotropic 2D Heisenberg model to the experimental single magnon dispersion. Squares: (a) Energy and (b) spectral weight of the single magnon peak obtained from fitting RIXS data provided in Ref. [5]. Error bars are smaller than the displayed symbols. Solid and dashed lines: Best fit of gapped and gapless modes to experimental data with $\tilde{J} = 57(7)$ meV, $J_2 = -18(3)$ meV, $J_3 = 14(2)$ meV, and $\Delta_\lambda = 0.08(1)$.

In conclusion, we have presented a detailed study of the critical magnetic properties of the single-layer perovskite iridate Sr_2IrO_4. Our results establish the importance of XY anisotropy in its effective low-energy Hamiltonian. Although in their seminal paper Jackeli and Khaliullin [10] proposed that the interactions in Sr_2IrO_4 are isotropic (Heisenberg), they note that the leading anisotropy term should have an XY character. Here, we have shown that the XY anisotropy is sufficiently strong to push the critical properties of Sr_2IrO_4 significantly away from the isotropic 2DQHAFSL model. Thus, while Sr_2IrO_4 and La_2CuO_4 share some similarities, as far as their effective magnetic interactions and critical properties are concerned, they are quite distinct.

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[1] B. J. Kim, H. Ohsumi, T. Komesu, S. Sakai, T. Morita, H. Takagi, and T. Arima, Science **323**, 1329 (2009).

[2] J. Kim, D. Casa, M. H. Upton, T. Gog, Y.-J. Kim, J. F. Mitchell, M. van Veenendaal, M. Daghofer, J. van den Brink, G. Khaliullin, and B. J. Kim, Phys. Rev. Lett. **108**, 177003 (2012).

[3] S. Boseggia, R. Springell, H. C. Walker, H. M. Rønnow, C. Rüegg, H. Okabe, M. Isobe, R. S. Perry, S. P. Collins, and D. F. McMorrow, Phys. Rev. Lett. **110**, 117207 (2013).

[4] M. Moretti Sala, S. Boseggia, D. F. McMorrow, and G. Monaco, Phys. Rev. Lett. **112**, 026403 (2014).

[5] J. Kim, M. Daghofer, A. H. Said, T. Gog, J. van den Brink, G. Khaliullin, and B. J. Kim, Nat. Commun. **5**, 4453 (2014).
[6] X. Liu, M. P. M. Dean, J. Liu, S. G. Chiuzbăian, N. Jaouen, A. Nicolaou, W. G. Yin, C. R. Serrao, R. Ramesh, H. Ding, and J. P. Hill, J. Phys.: Condens. Matter **27**, 202202 (2015).
[7] P. Olalde-Velasco, Y. Huang, V. Bisogni, J. Pelliciari, M. Dantz, S. Fatale, J. G. Vale, J. Miyawaki, Y. Harada, A. Nicolaou, S. Chiuzbăian, D. F. McMorrow, D. Prabhakaran, A. T. Boothroyd, J. Chang, H. M. Rønnow, V. Strokov, and T. Schmitt (unpublished).
[8] S. Boseggia, H. C. Walker, J. Vale, R. Springell, Z. Feng, R. S. Perry, M. M. Sala, H. M. Rønnow, S. P. Collins, and D. F. McMorrow, J. Phys.: Condens. Matter **25**, 422202 (2013).
[9] B. Dalla Piazza, M. Mourigal, M. Guarise, H. Berger, T. Schmitt, K. J. Zhou, M. Grioni, and H. M. Rønnow, Phys. Rev. B **85**, 100508 (2012).
[10] G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009).
[11] M. F. Collins, *Magnetic Critical Scattering* (Oxford University Press, New York, 1989).
[12] E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).
[13] B. Keimer, N. Belk, R. J. Birgeneau, A. Cassanho, C. Y. Chen, M. Greven, M. A. Kastner, A. Aharony, Y. Endoh, R. W. Erwin, and G. Shirane, Phys. Rev. B **46**, 14034 (1992).
[14] H. M. Rønnow, D. F. McMorrow, and A. Harrison, Phys. Rev. Lett. **82**, 3152 (1999).
[15] H. M. Rønnow, D. F. McMorrow, A. Harrison, I. D. Youngson, R. Coldea, T. G. Perring, G. Aeppli, and O. Syljuåsen, J. Magn. Magn. Mater. **236**, 4 (2001).
[16] P. Carretta, T. Ciabattoni, A. Cuccoli, E. Mognaschi, A. Rigamonti, V. Tognetti, and P. Verrucchi, Appl. Magn. Reson. **19**, 391 (2000).
[17] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988).
[18] M. S. Makivić and H.-Q. Ding, Phys. Rev. B **43**, 3562 (1991).
[19] P. Hasenfratz and F. Niedermayer, Phys. Lett. B **268**, 231 (1991).
[20] A. Cuccoli, V. Tognetti, P. Verrucchi, and R. Vaia, Phys. Rev. B **58**, 14151 (1998).
[21] S. Fujiyama, H. Ohsumi, T. Komesu, J. Matsuno, B. J. Kim, M. Takata, T. Arima, and H. Takagi, Phys. Rev. Lett. **108**, 247212 (2012).
[22] Q. Li, G. Cao, S. Okamoto, J. Yi, W. Lin, B. Sales, J. Yan, R. Arita, J. Kuneš, A. Kozhevnikov, A. G. Eguiluz, M. Imada, Z. Gai, M. Pan, and D. G. Mandrus, Sci. Rep. **3**, 3073 (2013).
[23] C. Dhital, T. Hogan, Z. Yamani, C. de la Cruz, X. Chen, S. Khadka, Z. Ren, and S. D. Wilson, Phys. Rev. B **87**, 144405 (2013).
[24] F. Ye, S. Chi, B. C. Chakoumakos, J. A. Fernandez-Baca, T. Qi, and G. Cao, Phys. Rev. B **87**, 140406 (2013).
[25] A. Taroni, S. T. Bramwell, and P. C. W. Holdsworth, J. Phys.: Condens. Matter **20**, 275233 (2008).
[26] T. R. Thurston, G. Helgesen, J. P. Hill, D. Gibbs, B. D. Gaulin, and P. J. Simpson, Phys. Rev. B **49**, 15730 (1994).
[27] S. R. Andrews, J. Phys. C: Solid State Phys. **19**, 3721 (1986).
[28] D. F. McMorrow, N. Hamaya, S. Shimomura, Y. Fujii, S. Kishimoto, and H. Iwasaki, Solid State Commun. **76**, 443 (1990).
[29] H. Hünnefeld, T. Niemöller, J. R. Schneider, U. Rütt, S. Rodewald, J. Fleig, and G. Shirane, Phys. Rev. B **66**, 014113 (2002).
[30] Furthermore, we believe that the values previously reported for the in-plane correlation length are too large by a factor of a_0. We base this on comparisons of our raw experimental data with those presented in Ref. [21]; the half widths at half maximum are comparable, showing that the two data sets are consistent.
[31] J. M. Kosterlitz and D. J. Thouless, J. Phys. C: Solid State Phys. **6**, 1181 (1973).
[32] S. T. Bramwell and P. C. W. Holdsworth, J. Phys.: Condens. Matter **5**, L53 (1993).
[33] A. Cuccoli, T. Roscilde, R. Vaia, and P. Verrucchi, Phys. Rev. Lett. **90**, 167205 (2003); A. Cuccoli, T. Roscilde, V. Tognetti, R. Vaia, and P. Verrucchi, Phys. Rev. B **67**, 104414 (2003).
[34] J. Als-Nielsen, S. T. Bramwell, M. T. Hutchings, G. J. McIntyre, and D. Visser, J. Phys.: Condens. Matter **5**, 7871 (1993).
[35] H. M. Rønnow, A. R. Wildes, and S. T. Bramwell, Physica B **276**, 676 (2000).
[36] M. Greven, R. Birgeneau, Y. Endoh, M. Kastner, M. Matsuda, and G. Shirane, Z. Phys. B **96**, 465 (1995).
[37] J. I Igarashi and T. Nagao, Phys. Rev. B **89**, 064410 (2014).
[38] B. Keimer, R. Birgeneau, A. Cassanho, Y. Endoh, M. Greven, M. Kastner, and G. Shirane, Z. Phys. B **91**, 373 (1993).