Letter

Spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$

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We use inelastic neutron scattering to study spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$, where Lanthanum doping triggers a Lifshitz transition by pushing the van Hove singularity in the γ band to the Fermi energy. Strong spin fluctuations emerge at an incommensurate wave vector $\mathbf{Q}_{ic} = (0.3, 0.3)$, corresponding to the nesting vector between α and β Fermi sheets. The incommensurate antiferromagnetic fluctuations shift toward (0.25,0.25) with increasing energy up to ~110 meV. By contrast, scatterings near the ferromagnetic wave vectors $\mathbf{Q} = (1, 0)$ and (1,1) remain featureless at all energies. This contradicts the weak-coupling perspective that suggests a sharp enhancement of ferromagnetic susceptibility due to the divergence of density of states in the associated γ band. Our findings imply that ferromagnetic fluctuations in Sr_2RuO_4 and related materials do not fit into the weak-coupling paradigm, but instead are quasilocal fluctuations induced by Hund's coupling. This imposes significant constraints for the pairing mechanism involving spin fluctuations.

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The pairing mechanism of the Sr_2RuO_4 superconductor has been the focus of tremendous research activities [1–4], but as yet remains a mystery. For a long time, Sr_2RuO_4 has stood as a promising candidate for a spin-triplet superconductor with a chiral p-wave order parameter [5–7]. A series of experiments supported this belief. Early on, nuclear magnetic resonance (NMR) [8] and polarized neutron diffraction [9] measurements show an unchanged magnetic susceptibility across the superconducting transition temperature (T_c), suggesting an odd pairing state. In addition, a spontaneous time-reversal symmetry breaking at T_c has been observed by muon spin relaxation [10] and polar Kerr effect [11] studies, which reveals a chiral character of the superconducting state.

However, some experimental data such as the absence of edge currents [12,13], the Pauli-limited upper critical field [14], and the lack of a linear strain dependency of T_c at zero strain limit [15] cannot be readily explained in this scenario. Furthermore, recent NMR [16,17] and polarized neutron diffraction [18] investigations revealed that the spin susceptibility in Sr_2RuO_4 decreases significantly below T_c—contradicting a simple spin-triplet Cooper pairing. The discrepancies between these results have kicked off a flurry of studies to identify its pairing symmetry, and different composite order parameters have been put forward to reconcile the controversies [19–27]. Yet, the debate on this topic remains far from closed.

Superconductivity in Sr_2RuO_4 arises near magnetic instabilities [28–30], as it does in cuprates and iron pnictides. As a result, spin fluctuations are believed to be critical for the Cooper pairing [31–35]. Theoretically, antiferromagnetic and ferromagnetic fluctuations could cause even and odd parity of superconducting order parameters, respectively [32,33]. Previous inelastic neutron scattering (INS) experiments on Sr_2RuO_4 revealed strong two-dimensional incommensurate antiferromagnetic responses around wave

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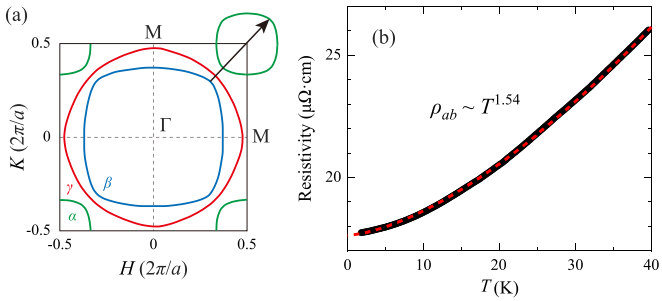

FIG. 1. (a) Schematic of the Fermi surfaces in the first Brillouin zone of $Sr_{2-y}La_yRuO_4$ close to the Lifshitz transition, inferred from ARPES measurements [47]. Green, blue, and red solid curves represent α, β, and γ Fermi sheets, respectively. Black arrow indicates the nesting vector between α and β bands. (b) In-plane resistivity ρ_{ab} versus temperature T in $Sr_{1.8}La_{0.2}RuO_4$. Red dashed curve is a powerlaw fitting $AT^n + \rho_0$ with $n = 1.54 \pm 0.04$.

vector $\mathbf{Q}_{ic} = (0.3,0.3,L)$ and equivalent positions [36–43], which are attributed to the Fermi surface nesting between the quasi-one-dimensional α and β sheets associated with the d_{xz} and d_{yz} orbitals, respectively [see Fig. 1(a)]. Meanwhile, weak magnetic fluctuations appear near the Brillouin zone center [38,41,43], manifesting a ferromagnetic character. The ferromagnetic fluctuations have been attributed to the nesting of γ bands [44,45], but recent dynamical mean-field theory (DMFT) calculations instead suggested that the ferromagnetic fluctuations are quasilocal due to Hund's coupling [46]. Yet, the origin of the ferromagnetic fluctuations is still under debate.

Band structure studies of Sr_2RuO_4 reveal a van Hove singularity (vHS) in the γ band closely (∼49 meV) above the Fermi level [44,45,47,48]. Modest external perturbations can therefore significantly influence the Fermi surface topology and electronic properties [15,45,48–52]. For example, applying uniaxial strain on Sr_2RuO_4 may induce a Lifshitz transition by pushing the vHS to the Fermi level and enhance T_c [50,51]. Carrier doping by chemical substitution provides an alternative path to tune the electronic properties. For example, La^{3+} substitution for Sr^{2+} induces electron doping and generates a Lifshitz transition at the critical doping in $Sr_{2-y}La_yRuO_4$ ($y_c = 0.2$), where the vHS crosses the Fermi level [45,47,48]. A non-Fermi-liquid (NFL) behavior has been observed at the Lifshitz transition, manifesting the occurrence of a strong electronic renormalization and quasiparticle scatterings (see Fig. 1(b) and Ref. [45]). In the weak coupling scenario, the divergence of density of states near the vHS would greatly enhance the ferromagnetic spin fluctuations associated with the γ band [45], whereas quasilocal fluctuations are expected to be marginally affected and remain very weak [46]. The origin of ferromagnetic fluctuations in Sr_2RuO_4 and related materials can thus be directly addressed by studying the La^{3+} doping effect.

In this paper, we report an INS study of spin fluctuations in $Sr_{2-y}La_yRuO_4$ at the critical concentration $y_c = 0.2$, where the vHS in the γ band crosses the Fermi level. Strong incommensurate antiferromagnetic fluctuations are observed up to ∼110 meV and disperse from $\mathbf{Q}_{ic} = (0.3,0.3)$ to $(0.25,0.25)$. This differs from undoped Sr_2RuO_4 where the antiferromagnetic fluctuations show little dispersion. On the other hand, no discernible ferromagnetic response has been detected with our experimental sensitivity. These results contradict the weak-coupling scenario that ferromagnetic fluctuations are induced by the nesting of the γ band; instead they support the proposal that magnetic responses near the ferromagnetic wave vectors have a quasilocal character.

High-quality $Sr_{1.8}La_{0.2}RuO_4$ single crystals were synthesized using the floating zone method [53]. Electron probe micro analysis (EPMA) and x-ray diffraction (XRD) measurements are performed to confirm the chemical composition and characterize the quality of our sample. The XRD Rietveld refinement shows that $Sr_{1.8}La_{0.2}RuO_4$ adopts the same space group ($I4/mmm$) as Sr_2RuO_4. No impurity, disorder, or lattice distortion is detected (see the Supplemental Material [54]). Below 40 K, the in-plane resistivity follows a temperature dependence $\rho_{ab} = AT^n + \rho_0$ with $n = 1.54 \pm 0.04$ [Fig. 1(b)], deviating from the Fermi liquid behavior observed in undoped Sr_2RuO_4. This is consistent with previous reports [45]. Our INS experiments were carried out on the MERLIN time-of-flight spectrometer at the Rutherford Appleton Laboratory [56], and the cold triple-axis spectrometer PANDA at the Heinz Maier-Leibnitz Zentrum (FRMII) [57]. 21 pieces of single crystals with a total mass of ∼12 g were coaligned for the INS measurements. The time-of-flight experiment was performed with the incident neutron energies fixed at $E_i =$ 18.8, 40.2, and 135.8 meV. Data were normalized to absolute units using the incoherent elastic scattering from a standard vanadium sample. Triple-axis experiments were performed with the final neutron energy fixed at $E_f = 5.1$ meV. Pyrolytic graphite (002) [PG(002)] was used as a monochromator and analyzer. A Be filter was employed to reduce the contamination from higher-order neutrons. We define the wave vector $\mathbf{Q}$ in the tetragonal unit cell at $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$ as (H, K, L) in reciprocal lattice units (r.l.u.), where $\mathbf{a}^* = 2\pi\hat{\mathbf{a}}/a$, $\mathbf{b}^* = 2\pi\hat{\mathbf{b}}/b$, and $\mathbf{c}^* = 2\pi\hat{\mathbf{c}}/c$ with lattice parameters $a = b = 3.86$ Å and $c = 12.72$ Å.

Figure 2 displays constant-energy plots of the scattering intensity in the (H, K) plane at 5 K. At low energies [Figs. 2(a)–2(c)], clear scatterings are observed near the incommensurate wave vector $\mathbf{Q}_{ic} = (0.3,0.3)$ and equivalent positions. This is similar to the dominant antiferromagnetic spin fluctuations in Sr_2RuO_4 [36,37,39]. It has been shown that La doping mostly influences the γ band, while the nesting condition between the α and β Fermi sheets is weakly affected [47]. The scattering significantly weakens at the equivalent $\mathbf{Q} = (0.7, 0.3)$ [Figs. 2 and 4(j)] due to the reduced Ru^{4+} magnetic form factor with increasing $|\mathbf{Q}|$, corroborating its magnetic origin. With increasing energy, the incommensurate peak positions move toward the lower $\mathbf{Q}$ [Figs. 2(d)–2(i)], revealing a notable dispersion.

The dispersion of the incommensurate spin fluctuations can be better visualized in the contour plot of the E-K plane. As illustrated in Fig. 3, strong spin fluctuations stem from $\mathbf{Q}_{ic} = (0.3,0.3)$ and disperse to $(0.25,0.25)$ around 110 meV. This behavior differs from Sr_2RuO_4 where the peak position of spin fluctuations is essentially fixed at $\mathbf{Q}_{ic}$ at all energies [39]. This difference indicates that La doping may induce a

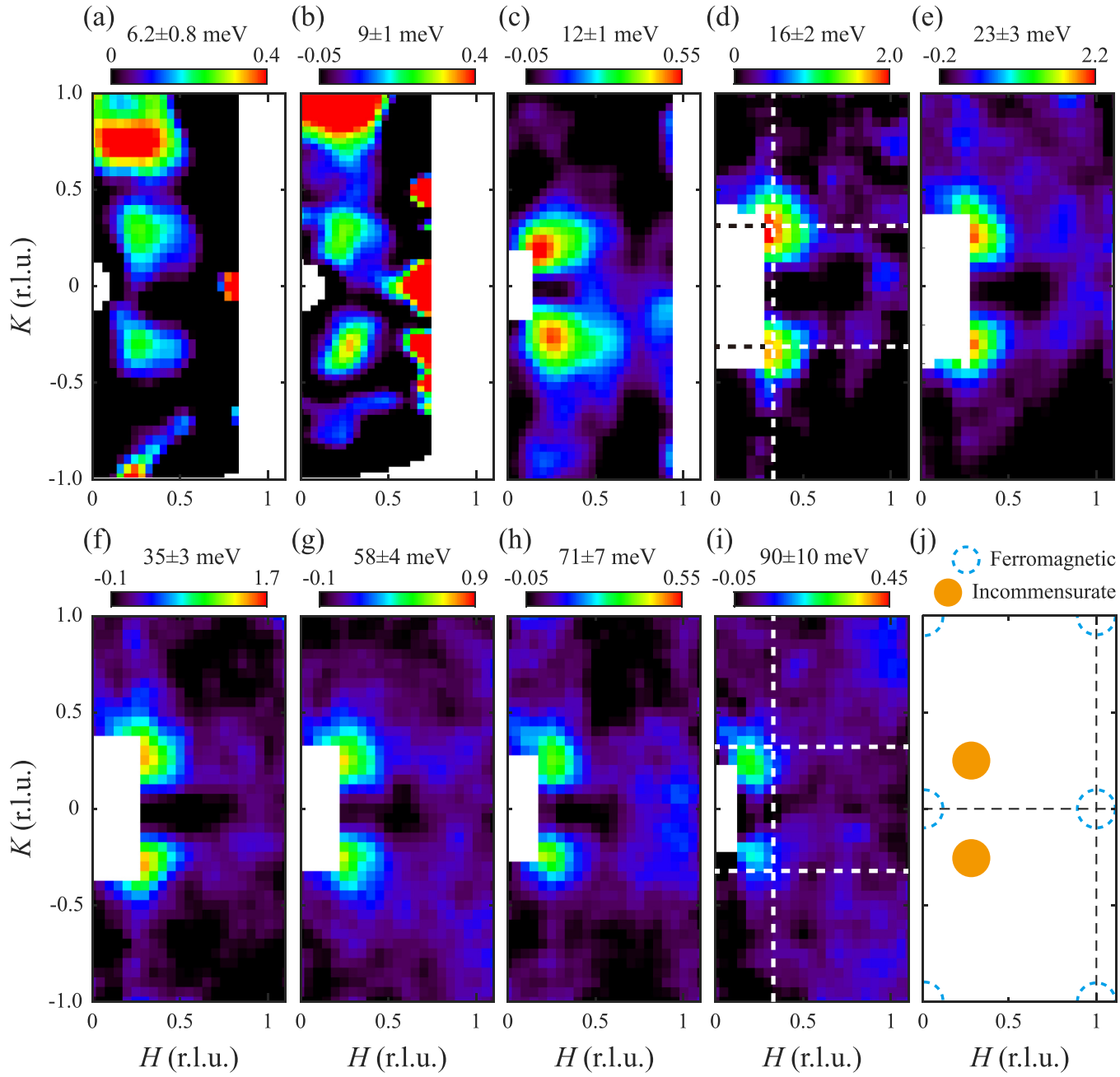

FIG. 2. Momentum dependence of the spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$ at 5 K. Constant-energy images at indicated energies measured with incident neutron energies of $E_i = 18.8$ (a) and (b), 40.2 (c), and 135.8 meV (d)–(i). Contour maps in (a) and (b) are rotated clockwise by 90° according to the C_4 crystal symmetry for direct comparisons with data collected with higher incident energies. Data are symmetrized with respect to the K axis to enhance statistical accuracy and the $|\mathbf{Q}|$-dependent background is subtracted following the method introduced in Ref. [55]. Color bars indicate intensity in unit of mbarn sr^{-1} meV^{-1} $f.u.^{-1}$. Dashed lines in (d) and (i) mark the momenta with $H = 0.3$ and $K = \pm 0.3$. The antiferromagnetic signals at 90 meV locate clearly at smaller wave vectors. (j) Schematic representation of the incommensurate antiferromagnetic and ferromagnetic wave vectors in the (H, K) plane.

non-negligible change to the low-energy dispersion of the α and β bands that slightly departs from the rigid-band shift assumption [45]. Note that since the energy transfer is coupled to L in the time-of-flight scattering geometry, the E-K contour plot yields no observable L modulation of the spin fluctuations.

We made constant-energy cuts through $\mathbf{Q} = (0.3,0.3)$ and (1,0) to quantify the spin fluctuations. As shown in Figs. 4(a)–4(j), the intensity of antiferromagnetic fluctuations displays a maximum at around 16 meV and gradually vanishes above 110 meV. The peak position shifts to a lower $\mathbf{Q}$ with increasing energy in a nearly linear fashion [Figs. 3 and 4(j)]. A careful survey of the scattering intensity has been done around ferromagnetic wave vectors (1,0) and (1,1). However, scans in the $(1, K)$ direction covering these wave vectors are featureless at all energies [Fig. 4(k)], implying that ferromagnetic

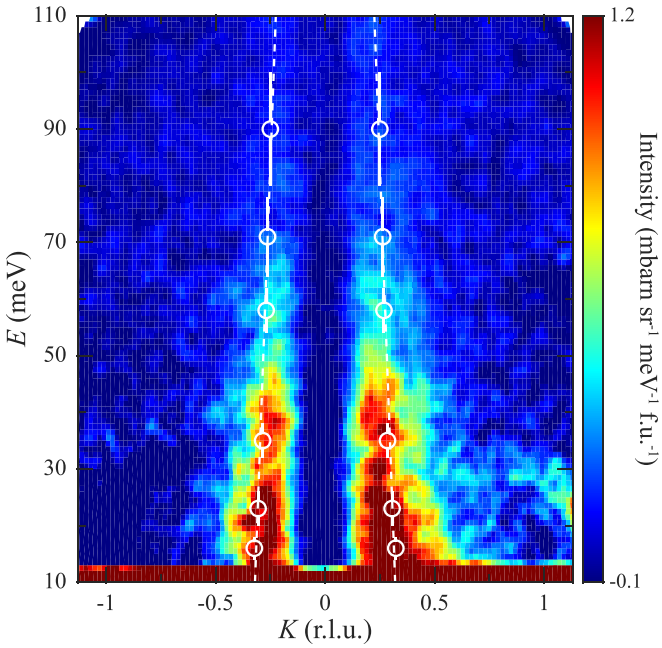

FIG. 3. Dispersion of the incommensurate spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$ at 5 K measured with $E_i = 135.8$ meV. Data are two-fold symmetrized with respect to the K axis and $|\mathbf{Q}|$-dependent backgrounds have been subtracted. The intensity is integrated from $H = 0.1$ to 0.5. Open circles denote peak positions extracted from Gaussian fits of constant-energy scans in Figs. 4(d)–4(i). Vertical bars indicate the energy integration range and the white dashed lines are linear fits to the peak positions.

fluctuations, if they exist, are still below the detection limit of the current INS instrument. This is further confirmed by our triple-axis measurements (see Fig. S3. in the Supplemental Material [54]).

Previous polarized neutron scattering measurements revealed relatively weak ferromagnetic fluctuations in Sr_2RuO_4 [38,41,43]. These studies, however, were not able to distinguish whether the ferromagnetic fluctuations originate from itinerant electrons or local moments. Tight-binding calculations predict a divergence of bare band susceptibility χ_0 and an even more pronounced enhancement of the renormalized susceptibility χ, when La doping pushes the vHS to the Fermi level and induces a divergence of the density of states in the γ band [44,45]. Indications of such enhanced electronic density of states were indeed observed by transport, specific heat and de Haas-van Alphen oscillations measurements [45,48]. Our data, however, reveal no discernible signal around the ferromagnetic wave vectors $\mathbf{Q} = (1, 0)$ and (1,1) at all energies measured, contradicting this weak-coupling scenario. Alternatively, the magnetic response around Γ point with a broad momentum distribution has been interpreted as quasilocal fluctuations driven by Hund's coupling in the DMFT calculation [46]. The absence of a sharp enhancement of the ferromagnetic fluctuations in $Sr_{1.8}La_{0.2}RuO_4$ seems in favor of such a proposal. Interestingly, a recent angle-resolved photoemission measurement on the monolayer $SrRuO_3$ film also suggested that the vHS in the γ band does not lead to ferromagnetism but results in a strongly correlated metal state [58]. This is in line with current neutron scattering results in bulk $Sr_{1.8}La_{0.2}RuO_4$.

The prevalent paradigm of a p-wave triplet pairing state involves the exchange of bosonic ferromagnetic modes, while antiferromagnetic fluctuations favor a singlet pairing. The lack of conventional dispersive ferromagnetic fluctuations in Sr_2RuO_4 seems to make it less likely to host a simple p-wave pairing than other candidates, where substantial well-defined ferromagnetic fluctuations have been observed, such as the heavy Fermion superconductor UCoGe [59] and iron-based compound YFe_2Ge_2 [60]. Recent theoretical and experimental works have suggested composite order parameters in Sr_2RuO_4 [19–23,25], which may involve multiple spin fluctuation pairing channels. Sophisticated theoretical calculations considering both antiferromagnetic and quasilocal ferromagnetic fluctuations on the Cooper pairing are highly desirable.

In summary, we used INS experiments to investigate spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$ covering a broad range of energy-momentum space. Strong antiferromagnetic spin fluctuations emanate from the incommensurate position (0.3,0.3). The excitation extends to above 110 meV and progressively shifts to (0.25,0.25). On the other hand, there is no clearly increased scattering near ferromagnetic wave vectors. This contradicts the weak-coupling hypothesis, which predicts a divergence of ferromagnetic susceptibility as the vHS approaches Fermi level at this La doping. Instead, the inertia of weak ferromagnetic fluctuations against La doping is in line with the description of quasilocal fluctuations driven by Hund's coupling [46]. These results are crucial for a complete understanding of the magnetism in this system, and they set significant restrictions on the mechanism of superconductivity in Sr_2RuO_4.

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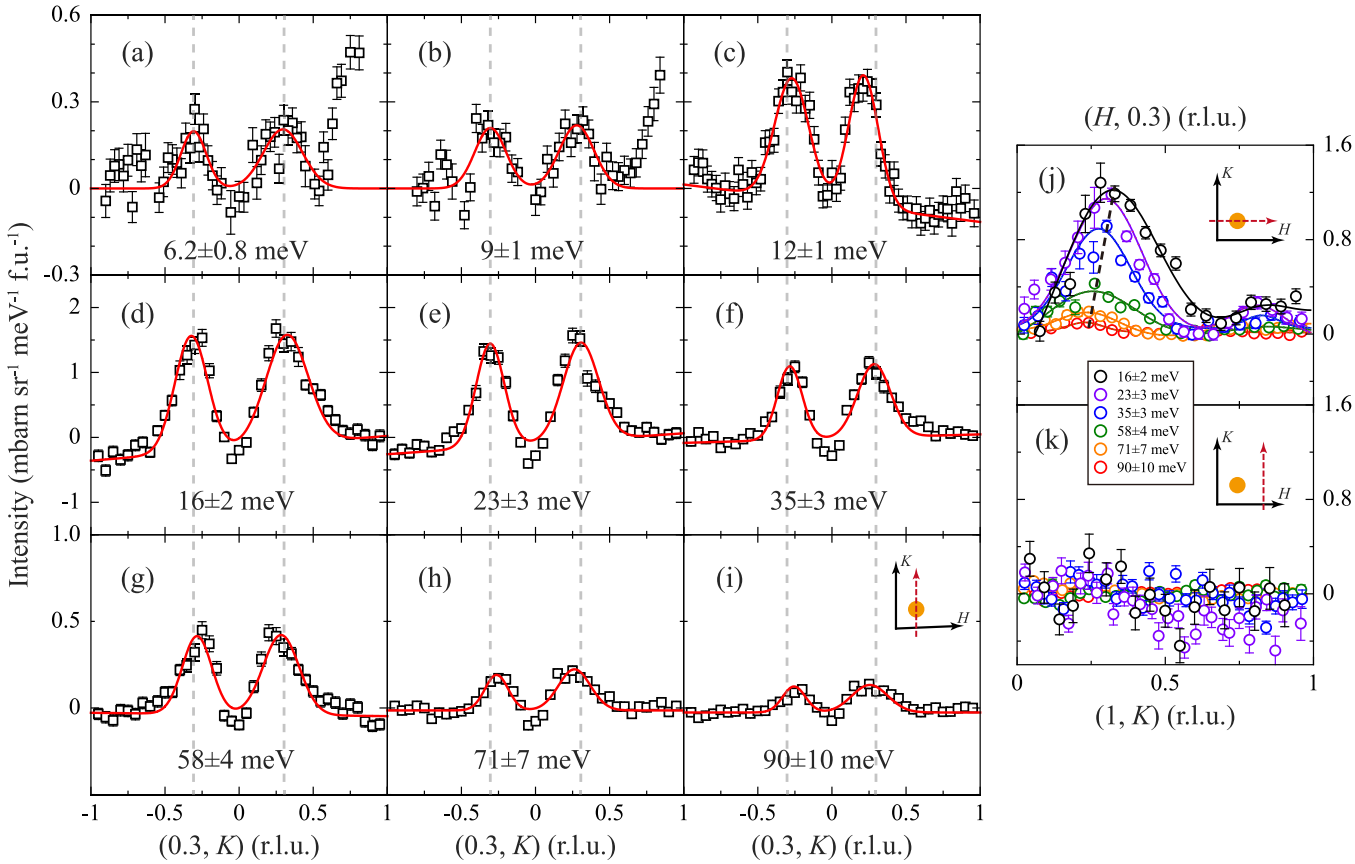

FIG. 4. Constant-energy scans of the spin fluctuations in $Sr_{1.8}La_{0.2}RuO_4$ at 5 K. (a) and (b), (c), and (d)–(k) were collected on MERLIN with $E_i = 18.8$, 40.2, and 135.8 meV, respectively. (a)–(i) Constant-energy scans along $(0.3, K)$ direction at indicated energies. H is integrated from 0.05 to 0.55. Red solid curves are fits with two Gaussian profiles and a linear background. The fitted peak positions are plotted in Fig. 3. Gray dashed lines indicates $K = \pm 0.3$. (j) Constant-energy scans along $(H, 0.3)$ direction with K integrated from 0.10 to 0.50. Solid lines are fits with Gaussian profiles. (k) Constant-energy scans along $(1, K)$ direction with H integrated from 0.95 to 1.05. Data in (j) and (k) are symmetrized with respect to the K, H axes and the (1,1) direction to enhance statistics. Backgrounds have been subtracted as described in Ref. [55]. The black dashed line connects the fitted peak positions at 16 and 90 meV. Arrows in the insets indicate the scan directions. Error bars represent one standard deviation.

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