Observation of two-dimensional spin fluctuations in the bilayer ruthenate $Sr_3Ru_2O_7$ by inelastic neutron scattering

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We report an observation of two-dimensional incommensurate magnetic fluctuations in the layered metallic perovskite $Sr_3Ru_2O_7$. The wave vectors where the magnetic fluctuations are strongest are different from those observed in the superconducting single layer ruthenate Sr_2RuO_4 and appear to be determined by Fermi surface nesting. No antiferromagnetic ordering is observed for temperatures down to 1.5 K. For temperatures $T \gtrsim 20$ K, the fluctuations become predominately ferromagnetic. Our inelastic neutron scattering measurements provide concrete evidence of the coexistence of competing interactions in $Sr_3Ru_2O_7$ and of the low-energy scale of the fluctuations.

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The nature of magnetic correlations in layered oxide perovskites such as cuprates, manganites, and ruthenates is at the heart of theoretical and experimental challenges in contemporary solid state physics. In recent years, the discovery of unconventional superconductivity in the single-layer ruthenate Sr_2RuO_4 (Ref. 1) has generated great interest in this and related ruthenates. The observation of low energy incommensurate two-dimensional (2D) spin fluctuations in Sr_2RuO_4 (Refs. 2–4) has raised the question of the relevance of spin fluctuations to *p*-wave pairing in this material. The closest relative of Sr_2RuO_4, the bilayer $Sr_3Ru_2O_7$, is a paramagnet where ferromagnetic and antiferromagnetic correlations may be in competition, and ferromagnetism can be induced by pressure or impurities.[5–10] At low temperatures, high-quality single crystals of $Sr_3Ru_2O_7$ exhibit Fermi liquid behavior, such as a T^2 temperature dependence of the resistivity[6,11] and a linear electronic heat capacity with $\gamma = 110$ mJ/(K^2 mol Ru).[8] However, a moderate magnetic field (5.5–7.7 T, depending on field direction) induces a metamagnetic transition, which is accompanied by a striking deviation from Fermi liquid behavior.[8,12] $Sr_3Ru_2O_7$ appears to be a strong candidate to exhibit a metamagnetic quantum critical end-point, driven by the magnetic field and characterized by the absence of spontaneous symmetry breaking.[12] In this paper we report observations in zero field of low-energy spin fluctuations in $Sr_3Ru_2O_7$ as measured by inelastic neutron scattering.

With respect to the conducting and magnetic properties of $Sr_3Ru_2O_7$, the fundamental building blocks of its crystal structure are the RuO_2 bilayers joined by an SrO layer. These slabs are separated along the crystal ***c*** direction by two rock salt-type layers of SrO which decouple the slabs electronically and magnetically. In contrast to the single layer compound Sr_2RuO_4, the RuO_6 octahedra in $Sr_3Ru_2O_7$ are rotated around the ***c*** axis, by $\sim 7°$. This changes the unit cell from body-centered tetragonal to a $\sqrt{2}\times\sqrt{2}$ larger face-centered cell which is orthorhombic but has *a* and *b* equal within experimental error.[13] The rotation is expected to reduce the in-plane Ru-Ru hopping, and hence increase the density of states at the Fermi level,[14] which may enhance the magnetic fluctuations.

Single crystals of $Sr_3Ru_2O_7$ were grown in a mirror furnace, and were checked for homogeneity and purity by magnetic, resistive, and crystallographic measurements. All crystals used in this study showed the characteristic peak in susceptibility at $T\sim$ 17 K and no ferromagnetism. For the inelastic neutron scattering experiments, three crystals were mounted so that their axes coincided to form a mosaic sample with total mass 0.9 g. This was mounted in a cryostat on the cold neutron three-axis spectrometer IN14 at the ILL. For simplicity, we describe our results using the tetragonal cell of the compound, which has the *a* and *b* lattice parameters equal to the in-plane Ru-O-Ru distance 3.87 Å. The ***c*** axis is perpendicular to the RuO_2 planes and has the magnitude 20.7 Å, which is twice the spacing of the RuO_2 bilayers, reflecting the body-centred stacking of bilayers.[13] Using this unit cell, the main nuclear Bragg peaks of the 3D structure occur at points (h,k,l) in reciprocal space with integer *h*, *k*, and *l* and $(h+k+l)$ even. The less intense ones, arising from the rotations of the octahedra, occur at some of the points where *h* and *k* are half-integral and *l* is an integer.

We performed extensive measurements with $(h,k,0)$ as the scattering plane and further measurements in the $(h,0,l)$ plane. Unlike Sr_2RuO_4, magnetic fluctuations at our base temperature of 1.5 K were *not* observed to peak along the $(h,h,0)$ direction from a reciprocal lattice point; instead they were detected along $(h,0,0)$. Figure 1 shows representative scans along major symmetry directions at a constant energy

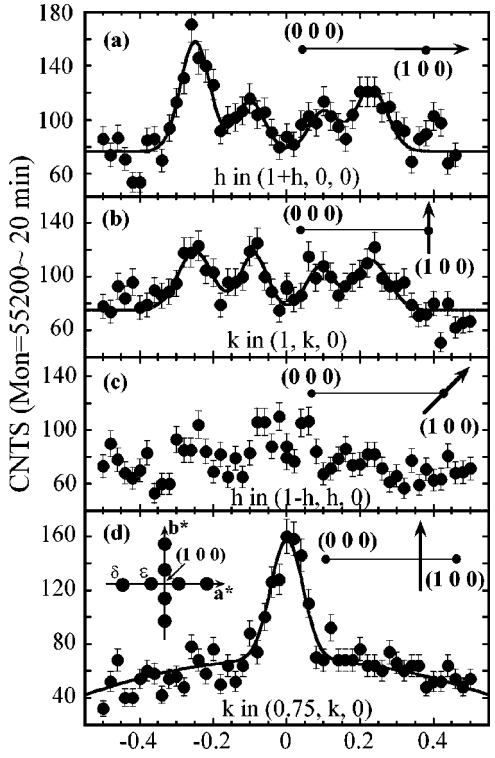

FIG. 1. Inelastic neutron scattering at 1.5 K: intensity versus wavevector along lines in the $(h,k,0)$ plane, at a constant energy transfer of 2 meV. Solid lines are a sum of Gaussians as a guide to the eye. The fwhm Q resolution, calculated for the relaxed collimation of our setup, is comparable to the width of the symbols. The inset to (d) indicates schematically the $\boldsymbol{Q}$ positions where peaks are observed. All measurements in this Letter, except those in Fig. 2, were taken with a varying incident energy and constant final energy of 4.97 meV, using PG (002) monochromator and analyser and a cooled beryllium filter before the analyser to remove higher order contamination.

transfer (from neutrons to the sample) of 2 meV. Figure 1(a) shows a double set of peaks along the $(h,0,0)$ direction at the positions $\boldsymbol{Q}\approx(1\pm0.25,0,0)$ and $(1\pm0.09,0,0)$. The intrinsic nature of such peaks was demonstrated by the observation of a signal of similar intensity around the symmetry-related (0,1,0) point and the presence of four peaks in the ''perpendicular'' scan through (1,0,0) shown in Fig. 1(b). The variation of intensity within each set of peaks is quantitatively consistent with the rapid falloff of the Ru magnetic form factor with the magnitude of $\boldsymbol{Q}$,[15] providing strong evidence for the magnetic nature of the excitations. Furthermore, as shown later, the intensity does not increase with temperature as would be expected if the scattering were due to lattice vibrations. The extent of the magnetic fluctuations in the $(h,k,0)$ plane of reciprocal space was established by the scans shown in Figs. 1(c) and 1(d). These show that the excitations give a broad peak centred on the $(h,0,0)$ axis. The results are summarized in the inset: the excitation intensity peaks at two incommensurate wave vectors of the form $\boldsymbol{q}_\delta\approx\{0.25,0,0\}$ and $\boldsymbol{q}_\epsilon\approx\{0.09,0,0\}$, distributed symmetrically about (1,0,0). It is natural to assume that these arise from peaks in the wave vector–dependent susceptibility at nesting

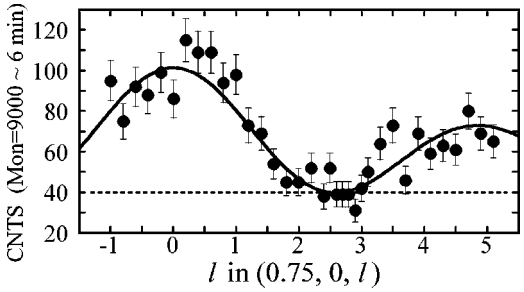

FIG. 2. l dependence of inelastic scattering at 2 meV and $h=0.75$, showing the effects of the bilayers. The solid line is the fit described in the text, plus a constant background. These measurements were performed at a constant incident energy of 14.67 meV, with a PG higher order filter in the incident beam.

vectors of the $Sr_3Ru_2O_7$ Fermi surface. This is not yet known experimentally, although it has been calculated.[14,16] The coupling between the two halves of a bilayer splits each of the three sheets observed in Sr_2RuO_4. This, and the rotation of the octahedra cause hybridization between the bands. It appears from the calculations,[14] that compared with Sr_2RuO_4, much of the nesting at the Fermi level is removed, except between parts of the α sheets (Ru d_{xz} and d_{yz} orbitals). The calculated sheets have nesting vectors along the (tetragonal) $\{1,0,0\}$ directions with values which are comparable with (although not equal to) those we observe. It seems reasonable to conclude that the differences of our results from those on single-layer Sr_2RuO_4 (Refs. 3,4) arise from the effects of bilayers and octahedral rotation on the Fermi surface in our system.

Measurements as a function of l allow us to determine the fundamental fluctuating unit in $Sr_3Ru_2O_7$ in this energy range. Figure 2 shows the variation along $\boldsymbol{c}^*$ of the intensity of the signal at $\boldsymbol{q}_\delta$. The experimental data are well represented by $I\propto f(\boldsymbol{Q})^2\cos^2(2\pi lz/c)$, where $f(\boldsymbol{Q})$ is the Ru form factor and $2z=0.194c$ is the distance between the RuO_2 planes in a bilayer. This function corresponds to the two halves of a bilayer fluctuating *in phase* with each other, but with *no* correlation between bilayers, so that the fluctuations are effectively two dimensional. A similar argument[17] was used to demonstrate 2D fluctuations in YBCO, but with the two halves of the bilayer in *antiphase*. We point out an important consequence of our results: since (1,0,0) *is* a reciprocal lattice point of a RuO_2 bilayer, the values of the $\boldsymbol{q}$ vectors of excitations should be measured from this point, rather than (0,0,0) or (1,0,1), which are the closest reciprocal lattice points of the 3D crystal structure.

We now consider the energy dependence of these excitations. Figure 3 shows four representative $\boldsymbol{Q}$ scans with energy transfers of 1, 2, 3, and 4 meV at $T=1.5$ K. The peaks appear to disperse slightly, and merge at higher energies. We have also performed a $\boldsymbol{Q}$ scan over this region at zero energy transfer, which showed no evidence for static magnetic ordering near $\boldsymbol{q}_\delta$ or $\boldsymbol{q}_\epsilon$. This result is in agreement with those of Refs. 18,19. We conclude that at finite temperature only finite frequency, short-range magnetic correlations exist.

Figure 4 shows the energy dependence of the signal at $\boldsymbol{q}_\delta$. To obtain a measure of the characteristic energy of these

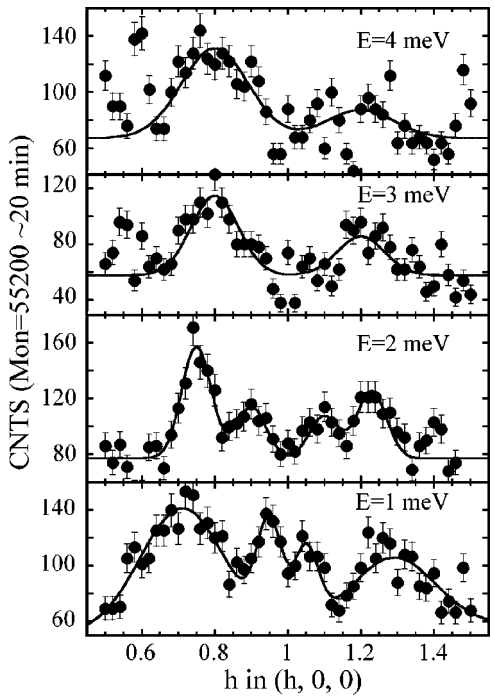

FIG. 3. Energy and $\boldsymbol{Q}$ dependence of scattering at 1.5 K. Scans along $(h,0,0)$ with energy transfers of 1, 2, 3, and 4 meV. We believe that the sharp features at the ends of the scan at 4 meV are due to optic phonons excited by second order contamination of the incoming beam.

excitations, we have fitted the response to a simple Lorentzian model for the susceptibility $\chi''(\boldsymbol{Q},\omega)=\chi'(\boldsymbol{Q})\times\omega\Gamma(\boldsymbol{Q})/[\Gamma^2(\boldsymbol{Q})+\omega^2]$. From the fit we obtain a value for the energy width $\hbar\Gamma=2.3\pm0.3$ meV. This is much less than 9 meV obtained by similar methods in the single layer compound Sr_2RuO_4.[3] The presence of dispersion on an energy scale much smaller than ϵ_F and the small energy scale of the fluctuations indicates the strong renormalising effects of electron correlations in our compound. A rough estimate[20] based on our data indicates that the very large observed specific heat γ may be understood in terms of the spin degrees of freedom.

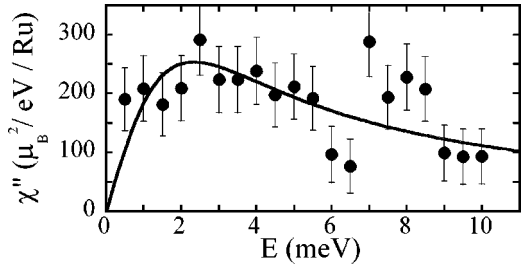

FIG. 4. Energy dependence of magnetic scattering at $\boldsymbol{Q}=(0.75,0,0)$, minus backgrounds taken at (1.48,0,0). The line represents a Lorentzian as described in the text. The ordinate has been corrected by the Bose factor $[n(\omega)+1]$ and the Ru form factor and converted to absolute units (with an accuracy $\sim$20%) by normalization to the intensity of a transverse acoustic phonon, measured at 3.1 meV around (1,1,0) at 100 K.

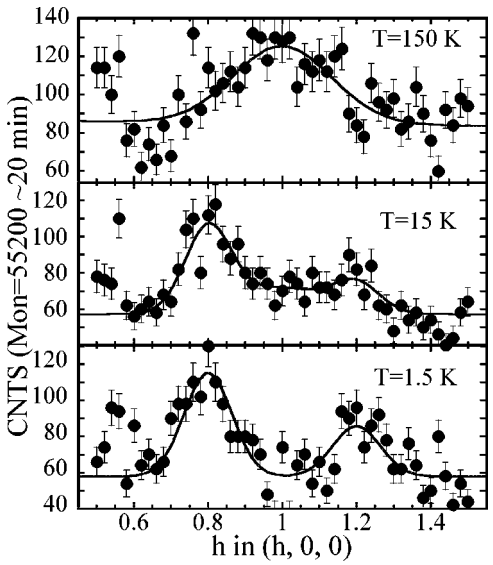

FIG. 5. $\boldsymbol{Q}$ dependence of scattering along $(h,0,0)$ for an energy transfer of 3.1 meV at three different temperatures, 1.5, 15, and 150 K. The peak near $h=0.6$ is believed to be spurious scattering.

We also note that the susceptibility is large, translating to $\chi'(\boldsymbol{Q}_\delta)$ of 1.6×10^{-2} emu/mol Ru. This indicates that $Sr_3Ru_2O_7$ is much closer to magnetic order than its sister compound.

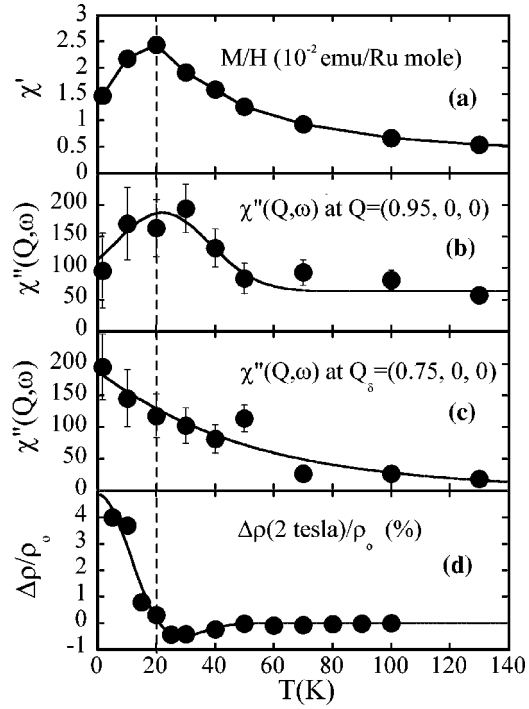

FIG. 6. Temperature-dependence of magnetic response from macroscopic and microscopic measurements (a) Static susceptibility from Ref. 6. (b) Susceptibility, $\chi''(\boldsymbol{Q},\omega)$ (units: μ_B^2/eV/Ru) from neutron scattering at $\boldsymbol{Q}=(0.95,0,0)$ and an energy transfer of 2 meV, minus a background at (0.55,0,0). (c) As for (b) at $\boldsymbol{Q}_\delta=(0.75,0,0)$. (d) Fractional magnetoresistance $[\rho(2T)-\rho(0)]/\rho(0)$ measured with current parallel to the magnetic field in the basal plane (Refs. 8,21). The lines serve as guides to the eye.

We have also followed the fluctuations as a function of temperature at an energy transfer of 3.1 meV around $\boldsymbol{Q}_\delta = (0.75,0,0)$ (Fig. 5). At base temperature, the two peaks associated with the incommensurate spin fluctuations are well defined and intense. However, as the temperature is increased, the intensity of the incommensurate peaks falls off, and is replaced by a broad peak of similar intensity around the 2D reciprocal lattice point (1,0,0). This position is not a Bragg peak of either the tetragonal or the orthorhombic cell, so does not give rise to a low-energy acoustic phonon. Hence the peak at (1,0,0) is most likely of magnetic origin. We have confirmed by measurements along $\boldsymbol{c}^*$ that this signal also arises from fluctuations of a bilayer unit. Our findings point to a crossover in the nature of the low-energy magnetic correlations in this material. At high temperatures, 2D ferromagnetic fluctuations dominate the correlations; as the temperature is lowered, instead of converging to a long-lived ferromagnetic state, the system is sidetracked to a different behavior with antiferromagnetic finite frequency 2D excitations.

In Fig. 6, we show that the change with temperature in the nature of magnetic fluctuations is reflected in macroscopic properties. At a temperature ≈ 20 K, there is a peak in the magnetic susceptibility (a), and also in the susceptibility close to a ferromagnetic position measured by neutron scattering (b). The antiferromagnetic fluctuations (c) fall away rapidly with increasing temperature and this is reflected in the change in sign of the longitudinal magnetoresistance (d).[7] It is not clear what causes this dramatic change in magnetic correlations, but it may be related to a loss in *c*-axis electronic coherence, which reveals itself as a steep rise in the c-axis resistivity in this temperature region.[6] It is of interest that in the compound $Ca_{2-x}Sr_xRuO_4$,[22,23] doping with Sr drives the system from an insulating antiferromagnetic state, through a phase with a ferromagnetic instability to a metallic superconducting one. In contrast, in $Sr_3Ru_2O_7$ the competing interactions coexist in the same high-quality stoichiometric samples. In manganite materials the nature of the magnetic fluctuations can also change with temperature;[24] however, the cause in this case is a structural/magnetic transition.

In conclusion, we have observed strong 2D spin fluctuations of the bilayers in $Sr_3Ru_2O_7$ in zero field. At high temperatures these fluctuations are predominantly ferromagnetic in nature, and cross over to incommensurate ones at low temperatures, with wavevectors close to those expected for nesting vectors of the Fermi surface. The characteristic energy of these fluctuations is small (compared with the sister compound Sr_2RuO_4), and their ambivalent nature suggests that they are implicated in and related to the metamagnetic transition observed at low temperatures. We note that a strong temperature dependence of the electronic properties and magnetic excitations is also observed in high-T_c superconductors[25] and heavy fermion systems.[26] Thus the behavior of $Sr_3Ru_2O_7$ may ultimately be related to its proximity to a quantum critical point.[12]

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