Observation of Two-dimensional Spin Fluctuations in the Bilayer Ruthenate \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) by Inelastic Neutron Scattering

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We report the first observation of two-dimensional incommensurate magnetic fluctuations in the layered metallic perovskite \( \text{Sr}_3\text{Ru}_2\text{O}_7 \). The wavevectors where the magnetic fluctuations are strongest are different from those observed in the superconducting single layer ruthenate \( \text{Sr}_2\text{RuO}_4 \) 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 predominantly ferromagnetic. Our inelastic neutron scattering measurements provide concrete evidence of the coexistence of competing interactions in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) and of the low energy scale of the fluctuations.

78.70.Nx, 75.40.Gb, 74.20.Mn, 75.30.Kz

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 \( \text{Sr}_2\text{RuO}_4 \) \(^1\) has generated significant interest in this and related ruthenates. The experimental observation of low energy incommensurate 2D spin fluctuations in \( \text{Sr}_3\text{Ru}_2\text{O}_4 \) \(^2\) has raised the question of the relevance of spin fluctuations to the mechanism producing \( p \)-wave pairing in this material. The closest relative of \( \text{Sr}_2\text{RuO}_4 \), the bilayer \( \text{Sr}_3\text{Ru}_2\text{O}_7 \), is a paramagnet where ferromagnetic and antiferromagnetic correlations may be in competition, and ferromagnetism can be induced by pressure or impurities \(^3\). In high-quality single crystals of \( \text{Sr}_3\text{Ru}_2\text{O}_7 \), the resistivity exhibits a Fermi-liquid \( T^2 \) temperature dependence below 10 K \(^4\). However, a moderate magnetic field induces a metamagnetic transition, which is accompanied by a striking deviation from Fermi liquid behaviour \(^5\). \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) 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 \(^6\). In this letter we report the first observations of low-energy spin fluctuations in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) as measured by inelastic neutron scattering.

With respect to the conducting and magnetic properties of \( \text{Sr}_3\text{Ru}_2\text{O}_7 \), the fundamental building blocks of its crystal structure are the \( \text{RuO}_2 \) bilayers joined by an \( \text{SrO} \) layer. These slabs are separated along the crystal \( c \)-direction by two rock salt-type layers of \( \text{SrO} \) which decouple the slabs electronically and magnetically. At the same time, and in contrast to the single layer compound \( \text{Sr}_2\text{RuO}_4 \), the \( \text{RuO}_6 \) octahedra in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) are rotated around the \( c \)-axis, by \( \sim 7^\circ \), changing the unit cell from body-centred tetragonal to a \( \sqrt{2} \times \sqrt{2} \) larger face-centred orthorhombic cell \(^7\). The rotation is expected to reduce the in-plane Ru-Ru hopping, and hence increase the density of states at the Fermi level \(^7\), which may enhance the magnetic fluctuations.

Single crystals of \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) were grown using 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 a temperature of 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. The sample was mounted in a cryostat on the cold neutron three-axis spectrometer IN14 at the ILL. For simplicity, we describe our results using the undistorted 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 \( \text{RuO}_2 \) planes and has the magnitude 20.7 Å, which is twice the spacing of the \( \text{RuO}_2 \) bilayers, reflecting the body-centred stacking of bilayers \(^8\). Using this unit cell, the main nuclear Bragg peaks of the 3-D 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 \( \text{Sr}_2\text{RuO}_4 \), 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 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...
\(Q \approx (1 \pm 0.25, 0, 0)\) and \((1 \pm 0.09, 0, 0)\). 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 Figure 1(b). In addition, 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 \(Q\) [15], providing strong evidence for the magnetic nature of the excitations.

![Image 55x266 to 298x617](Image 55x266 to 298x617)

**FIG. 1.** Inelastic neutron scattering at 1.5K: 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 \(Q\) 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, with a cooled beryllium filter before the analyser to remove higher order contamination.

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 Fig. 1(c) and (d). These show that the excitations give a broad peak centred on the \((h, 0, 0)\) axis. The results are summarised in the inset: the excitation intensity peaks at two incommensurate wavevectors of the form \(q_0 \approx \{0.25, 0, 0\}\) and \(q_0 \approx \{0.09, 0, 0\}\), distributed symmetrically about \((1, 0, 0)\). It is natural to assume that these arise from peaks in the wavevector-dependent susceptibility at nesting vectors of the \(\text{Sr}_2\text{Ru}_2\text{O}_7\) Fermi surface. This is not yet known experimentally, although it has been calculated [15]. The coupling between the two halves of a bilayer splits each of the three sheets observed in \(\text{Sr}_2\text{Ru}_2\text{O}_4\). This, and the rotation of the octahedra cause hybridisation between the bands. It appears from the calculations [17], that compared with \(\text{Sr}_2\text{Ru}_2\text{O}_4\), much of the nesting at the Fermi level is removed, except between parts of the \(\alpha\) 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 \(\text{Sr}_2\text{Ru}_2\text{O}_4\) [16] arise from the effects of bilayers and octahedral rotation on the Fermi surface in our system.

![Image 324x368 to 555x497](Image 324x368 to 555x497)

**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.

Measurements as a function of \(l\) allow us to determine the fundamental fluctuating unit in \(\text{Sr}_3\text{Ru}_2\text{O}_7\) in this energy range. Fig. 2 shows the variation along \(c^*\) of the intensity of the signal at \(q_0\). The experimental data are well-represented by \(I \propto f(Q)^2 \cos^2(2\pi l z/c)\), where \(f(Q)\) 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 [19] 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 \(q\)-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. Fig. 3 shows four representative \(Q\)-scans with energy transfers of 1, 2, 3 and 4 meV at \(T = 1.5\) K.
FIG. 3. Energy and $Q$-dependence of scattering at 1.5 K. Scans along $(h,0,0)$ with energy transfers of 1, 2, 3 and 4 meV.

The peaks appear to disperse slightly, and merge at higher energies. We have also performed a $Q$-scan over this region at zero energy transfer, which showed no evidence for static magnetic ordering near $q_\delta$ or $q_\xi$. This result is in agreement with those of [14,18]. We conclude that at finite temperature only finite frequency, short-range magnetic correlations exist.

Fig. 4 shows the energy dependence of the signal at $q_\delta$. We have fitted the response to a simple Lorentzian model for the susceptibility: $\chi''(Q,\omega) = \chi'(Q) \times \omega \Gamma(Q)/(\Gamma^2(Q) + \omega^2)$. We find that the characteristic energy $\hbar \Gamma = 2.3 \pm 0.3$ meV, which is much less than 9 meV reported in the single layer compound Sr$_2$RuO$_4$ [3]. The presence of dispersion on an energy scale much smaller than $\epsilon_F$ and the small energy scale of the fluctuations indicates the strong renormalising effects of electron correlations in our compound. We also note that the susceptibility is large, translating to $\chi'(Q_\delta)$ of $1.6 \times 10^{-2}$ emu/mol Ru. This indicates that Sr$_3$Ru$_2$O$_7$ is much closer to magnetic order than its sister compound.

We have also followed the fluctuations as a function of temperature at an energy transfer of 3.1 meV around $Q_\delta = (0.75,0,0)$ (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 $c^*$ 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 behaviour with antiferromagnetic finite frequency 2D excitations.
In conclusion, we have observed strong 2D spin fluctuations in the bilayers of Sr$_3$Ru$_2$O$_7$. 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 small characteristic energy of these fluctuations 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 [23] and heavy fermion systems [24]. Thus the behaviour of Sr$_3$Ru$_2$O$_7$ may ultimately be related to its proximity to a quantum critical point [12].

FIG. 6. Temperature-dependence of magnetic response from macroscopic and microscopic measurements: (a) Static susceptibility from [11]. (b) Susceptibility, $\chi''(Q, \omega)$ (units: $\mu^2$/eV/Ru) from neutron scattering at $Q = (0.95, 0, 0)$ and an energy transfer of 2 meV, minus a background at (0.55, 0, 0). (c) As for (b) at $Q = (0.75, 0, 0)$. (d) Fractional magnetoresistance, $(\Delta \rho(2T) - \rho(0))/\rho(0)$, measured with current parallel to the magnetic field in the basal plane [12]. The lines serve as guides to the eye.

In Fig. 6, we show that the change with temperature in the nature of magnetic fluctuations is reflected in macroscopic properties. At a temperature $\approx 20$ 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) [11]. 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 [11]. It is of interest that in the compound Ca$_{2-x}$Sr$_x$RuO$_4$ [21] [22], 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$_3$Ru$_2$O$_7$ the competing interactions coexist in the same high-quality stoichiometric samples.

In conclusion, we have observed strong 2D spin fluctuations of the bilayers in Sr$_3$Ru$_2$O$_7$. 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