Evidence for incommensurate spin fluctuations in Sr$_2$RuO$_4$.

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We report first inelastic neutron scattering measurements in the normal state of Sr$_2$RuO$_4$ that reveal the existence of incommensurate magnetic spin fluctuations located at $q_0=(\pm 0.6\pi/a, \pm 0.6\pi/a, 0)$. This finding confirms recent band structure calculations that have predicted incommensurate magnetic responses related to dynamical nesting properties of its Fermi surface.

Being the only example of a non-cuprate layered perovskite superconductor, Sr$_2$RuO$_4$ has attracted considerable attention despite its rather low critical temperature, $T_c \sim 1$ K. Its normal state is characterized as an essentially two-dimensional Fermi liquid and the coherent interlayer transport settles in at low temperature only. The susceptibility is most likely dominated by an enhanced Pauli spin susceptibility, $\Delta$. Meanwhile, the Sommerfeld coefficient, $\gamma$, in the specific heat is enhanced by a factor 3.5 with respect to band structure calculations. It yields a Wilson ratio, $R_W (\sim \Delta/\gamma)$, of 1.7. This value indicates that the enhancements in both susceptibility and electronic specific heat can be ascribed to the same origin: most likely correlations among electrons.

Noticing that SrRuO$_3$ is ferromagnetic (FM), it has been conjectured that Sr$_2$RuO$_4$ is close to a FM instability as well. This assertion is corroborated by microscopic calculation of magnetic properties of ruthenates. Since FM fluctuations disfavor both s- and d-wave superconductivity, it has been suggested that superconductivity in Sr$_2$RuO$_4$ should possess p-wave symmetry (triplet pairing). Conventional local density approximation (LDA) calculations give a correct Fermi surface topography, probed by de Haas-van Alphen measurements, as well as the magnetic enhancement due to Stoner exchange enhancement, although the mass renormalization cannot be explained within LDA calculations. In the superconducting state, the $^{101}$Ru nuclear spin lattice relaxation rate, $1/T_1$ exhibits a sharp decrease without a coherence peak (Hebel-Slichter peak) just above $T_c$, supporting the idea that an anisotropic pairing is effectively realized in Sr$_2$RuO$_4$. In addition, the spontaneous appearance of an internal magnetic field below the transition temperature, reported by neutron spin rotation measurements ($\mu$SR), and the absence of Knight shift modifications below $T_c$ point towards the triplet p-wave superconductivity. However, few experiments have really probed the exact nature of the spin fluctuations. Only the observation of a similar temperature dependence for $^{101}$Ru $1/T_1T$ and for $^{17}$O $1/T_1T$ in the NMR experiments by Imai et al. has suggested that spin fluctuations are predominantly FM in origin.

The determination of the antiferromagnetic order in the closely related compound Ca$_2$RuO$_4$ has suggested that the picture of a near-by FM instability in Sr$_2$RuO$_4$ is too simple. Furthermore, recent calculations which take into account the particular topology of the Fermi-surface, have predicted a sizeable magnetic response at the incommensurate wave-vector $(2\pi/3a, 2\pi/3a, 0)$, i.e. far away from the zone-center. The enhanced susceptibility arises from pronounced nesting properties of the almost one-dimensional $d_{z^2}$ bands. Mazin and Singh discuss the possibility of a competition between p-wave and d-wave superconductivity in Sr$_2$RuO$_4$.

In this letter, we report first inelastic neutron scattering (INS) measurements performed on single crystals of Sr$_2$RuO$_4$ in the normal state. Our data reveal dominant magnetic scattering at the incommensurate wave vectors $q_0=(\pm 0.6\pi/a, \pm 0.6\pi/a, 0)$, i.e. very close to the positions predicted by the band-structure calculations. The relevance of these findings for the mechanism of superconductivity in Sr$_2$RuO$_4$ will be discussed.

Most of the INS measurements presented here have been carried out on a single crystal of cylindrical shape (4mm in diameter and 35mm long) grown by a floating zone method. The sample exhibits the superconducting transition at $T_c \sim 0.62$ K. The single crystal was mounted in an aluminum can and attached to the cold finger of a closed cycle helium refrigerator. The INS experiments were performed on the triple axis spectrometers 2T (thermal beam) and 4F2 (cold beam) at the Laboratoire Léon Brillouin, Saclay, France. These spectrometers use neutron optics that focus the beam to the sample, with a resulting gain of neutron flux that proved to be crucial for these experiments. The experimental set up incorporates PG002 monochromator and analyzer and 14.7 meV fixed final energy. A pyrolytic graphite filter was inserted into the scattered beam in order to remove higher order contaminations. Data were taken within the scattering plane spanned by (1,0,0) and (0,1,0) directions. Some additional measurements were performed using several smaller single crystals with higher transition temperatures, $T_c=1.4-1.5$ K; these experiments have revealed similar signals. Throughout this article, the wave vector $Q=(H,K,L)$ is indexed in units of the reciprocal tetragonal lattice vectors $2\pi/a = 2\pi/b = 1.63$ Å$^{-1}$ and...
2π/ε = 0.49 Å⁻¹ (I4/mmm space group).  

Figure 1 shows representative constant-ω scans taken in the (H,K,0)-plane: at ¯ω=6.2 meV and around Q₀=(1.3,0.3,0) along the (0,1,0) direction. The scan at 10.4 K shows a sharp maximum of intensity peaked at Q₀=(1.3,0.3,0) on top of a smooth background. At room temperature, this sharp peak has almost disappeared. The horizontal bar indicates the spectrometer resolution.

The Fermi surface of Sr₂RuO₄ is formed by three sheets [10]: one, related to the 4dₓ²−y² orbitals is quasi-2D, whereas, the two others, related to 4dxz,dyz orbitals are quasi-1D. The 1D-sheets can be schematically described by parallel planes separated by q=±2π/3a, running both in the x and in the y directions. These peculiarities give rise to dynamical nesting effects at the wave vectors k=(q,y), k=(kₓ,q) and in particular at q=(q,q). The nesting effects become dominant when calculating the bare spin susceptibility of a non interacting metal [11], given by the Lindhard-function [12]:

\[ \chi(\vec{q},\omega) = -2\mu_B^2 \sum_k \frac{f_{k+q} - f_k}{\varepsilon_{k+q} - \varepsilon_k - \hbar\omega + i\epsilon} \]  

(2)

where ε →0, f_k is the Fermi distribution function and ε_k the quasiparticle dispersion relation. Our INS are in very good agreement with the predicted four spots of magnetic scattering situated at q=(±2π/3a,±2π/3a) [13]. In the experiment the incommensurate magnetic responses are actually observed slightly away, at q=((±2π/3a,±2π/3a), which is most likely related to details of the band-structure [10].
a Gaussian profile, located at \( q_0 \) with an energy independent q-width, on top of a constant background. In addition, two energy scans have been performed at \( Q=(1.3,0.3,0) \) and at \( Q=(1.3,0.46,0) \), the latter providing a background reference. These measurements allow us to determine the energy dependence of the magnetic response at \( q_0 \) from 1.5 to 12 meV. The analysis could not be extended to higher and lower energies due to the contaminations by phonons and elastic incoherent scattering respectively. Using Eq. [4], the magnetic intensity has been converted to the dynamical spin susceptibility \( \chi'' \) after correction by the thermal population factor and the squared magnetic form factor reported in figure [5]. We have then calibrated \( \chi'' \) in absolute units against the squared magnetic form factor reported in figure 2, slightly increases up to 7 meV and then almost saturates. This energy dependence can be parameterized following linear response theory:

\[
\chi''(q_0, \omega) = \chi'(q_0, 0) \frac{\Gamma \omega}{\omega^2 + \Gamma^2}
\]

where \( \Gamma \) is a damping energy of 9 meV and \( \chi'(q_0, 0) = 180 \mu_B^2 \cdot \text{eV}^{-1} \) corresponds to the static spin susceptibility at \( q_0 \). It is worth emphasizing that \( \chi'(q_0, 0) \) is 6 times larger than that at \( Q=0 \), i.e. the uniform susceptibility: \( \tilde{\chi} = \chi'(Q = 0, 0) = 30 \mu_B^2 \cdot \text{eV}^{-1} \) (\( \approx 10^{-3} \) emu/mole) [17]. In \( \text{La}_{1.86}\text{Sr}_{0.14}\text{O}_4 \), usually referred to as a strongly correlated system, \( \chi''(Q, \omega) \) at incommensurate wave vectors exhibits almost the same magnitude and a similar \( \omega \)-dependence [22].

In \( \text{Sr}_2\text{RuO}_4 \), electronic correlations are incorporated in RPA calculations: the spin susceptibility \( \chi(q, \omega) \) becomes enhanced through the Stoner-factor \( I(q) \) [8,10]:

\[
\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - \frac{I(q)}{2(\mu_B^2) \chi_0(q, \omega)}}
\]

The q-dependence of the Stoner factor, for an individual RuO$_2$ plane, reflects the fact that FM interactions are favored over antiferromagnetic interactions in Sr$_2$RuO$_4$ : in our units, \( I(q) = 0.86(1+0.8(a/\pi)^2 q^2) \text{ eV} \) (q in A$^{-1}$) [17]. INS results point towards a strong enhancement of the spin susceptibility by the Stoner factor (see Eq. [4]), such that the system should be close to a magnetic instability at \( q_0 \). With \( \chi'(q_0, 0) = 180 \mu_B^2 \cdot \text{eV}^{-1} \), one deduces from Eq. [4] that \( \frac{I(q_0)}{2(\mu_B^2) \chi_0(q_0, 0)} \approx 0.099 \), instead of being larger than 1 for a magnetic instability. Thus, incommensurate spin fluctuations are stronger than FM fluctuations in Sr$_2$RuO$_4$, as suggested in ref. [10].

The temperature dependence of both \( \chi''(Q_0, 6.2 \text{meV}) \) and the intrinsic q-width are reported in Fig. 4 as deduced from constant-\( \omega \) scans performed at 6.2 meV around \( Q=(1.3,0.3,0) \) along the (0,1,0) direction at different temperatures. \( \chi''(Q_0, 6.2 \text{meV}) \) exhibits a sharp decrease upon temperature increase and simultaneously the magnetic response broadens (the width of the signal can be reliably determined only up to 200 K). The T-dependence of \( \chi'' \) observed in INS measurements may be described by the out-smearing of the Fermi surface due to thermal hopping of electrons into unoccupied states.
(see the numerator in Eq. (2)), yielding a lowering of the dynamical susceptibility at $q_0$, and its broadening in q-space. The T-dependence of the magnetic response at $q_0$ can indeed be qualitatively reproduced [33] using Eqs. (2)-(4) and a description of the LDA band structure by three mutually non-hybridizing tight-binding bands [33].

INS measurements point out the existence of strong magnetic response at $q_0$, but do not reveal any sizeable FM fluctuations. In contrast, the uniform spin susceptibility [13] and the Knight shift measurements [14][24] provide evidence of strong FM correlation in Sr$_2$RuO$_4$. However, the delicate balance between FM and incommensurate spin fluctuations should become visible in the spin-lattice relaxation rate $T_1$ measured by both $^{17}$O and $^{101}$Ru NMR experiments [14][24]. These NMR-techniques probe the low energy spin fluctuations ($\omega \to 0$ with respect to INS measurements); furthermore, they integrate the fluctuations in q-space. Since the INS studies have determined the incommensurate fluctuations on an absolute scale we may estimate their contribution to $(1/T_1T)$, $INS(1/T_1T)$, in general $(1/T_1T)$ probes the q-summation of the the imaginary part of the susceptibility divided by the frequency in the limit $\omega \to 0$ (i.e. its initial slope),

$$\sum_q \chi''(q, \omega)/\omega \bigg|_{\omega \to 0}$$

its temperature dependency is shown in Fig. 4c (left scale).

What renders the quantitative comparison between the INS and NMR-results more difficult is the estimate of the hyperfine field whose q-dependent Fourier transform, $A(q)$, weights the susceptibility in NMR-studies. Considering that INS magnetic fluctuations are sharply peaked around $q_0$, one may approximate $A(q) = A(q_0)$, and gets [23]

$$INS(1/T_1T) \simeq \frac{k_B \gamma^2}{(g\mu_B)^2} |A(q_0)|^2 \sum_q \frac{\chi''(q, \omega)}{\omega} \bigg|_{\omega \to 0}$$

with $|A(q_0)|^2 = \Lambda^2[1 + 1/2(\cos(2\pi q_0) + \cos(2\pi a/3))]$ (\(\Lambda = 33 \text{ kOe/} \mu_B [23]\)) for $^{17}$O and $A(q_0) = -299$ kOe/\(\mu_B[11][14]\) for $^{101}$Ru. Using these values, we directly compare $INS(1/T_1T)$ with the measured $^{17}$O $(1/T_1T)$ in Fig. 4c (right scale). Clearly, the spin fluctuations at $q_0$ significantly contribute to $^{17}(1/T_1T)$, and can explain a large part of the reported T-dependence [14][24]. Similar calculation for the $^{101}$Ru $(1/T_1T)$ (not shown) yields even a stronger contribution. The remaining parts in $^{17,101}(1/T_1T)$, likely associated with FM excitations, should exhibit a less pronounced T-dependence similar to that of the uniform static spin susceptibility. Furthermore, assuming a weak q-dependence for these FM excitations [23], the comparison of NMR and INS measurements allows us to estimate the ferromagnetic characteristic energy to be of the order of 50 meV. This rather elevated value actually provides a satisfactory explanation for the absence of FM fluctuations in our INS measurements.

To conclude, our INS measurements demonstrate the existence of incommensurate spin fluctuations related to dynamical nesting properties of the Sr$_2$RuO$_4$ Fermi surface. Our data suggest that the system is close to a magnetic instability at $q_{0f}/=(\pm 0.6\pi/\alpha,\pm 0.6\pi/\alpha)$. The comparison of INS and $^{17}(1/T_1T)$ measurements suggests that the FM fluctuations are transferred to higher energy with respect to the spin fluctuations at $q_0$. All these results cast some doubt on the predominant role of FM spin fluctuations in the mechanism of superconductivity in Sr$_2$RuO$_4$.

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[21] We have used acoustic phonons in scans (2+\(\xi_0,0,0\)) and (2,\(\xi_0,0,0\)) for $\hbar \omega= 6.75$ and 8.26 meV (sound velocity $\approx 20.8$ and 43.9 meV, respectively) [4].
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[26] For a magnetic field applied along the c axis, $\Lambda^2 = |A_{1f}|^2 + |A_{1l}|^2$, with $A_{1f}= -18.5$ kOe/\(\mu_B\) and $A_{1l}= -26.8$ kOe/\(\mu_B\) [23].