# Evidence for Incommensurate Spin Fluctuations in $Sr_2RuO_4$

Y. Sidis,[1] M. Braden,[1,2] P. Bourges,[1] B. Hennion,[1] S. NishiZaki,[3] Y. Maeno,[3,4] and Y. Mori[3]
[1]*Laboratoire Léon Brillouin, C.E.A./C.N.R.S., F-91191 Gif-sur-Yvette CEDEX, France*
[2]*Forschungszentrum Karlsruhe, INFP, Postfach 3640, D-76021 Karlsruhe, Germany*
[3]*Department of Physics, Kyoto University, Kyoto 606-8502, Japan*
[4]*CREST, Japan Science and Technology Corporation, Kawaguchi, Saitama 332-0012, Japan*
(Received 23 April 1999)

We report inelastic neutron scattering measurements in the normal state of $Sr_2RuO_4$ that reveal the existence of significant incommensurate magnetic spin fluctuations located at $\mathbf{q}_0 = (\pm 0.6\pi/a, \pm 0.6\pi/a, 0)$. This finding confirms recent band-structure calculations that have predicted incommensurate magnetic responses related to dynamical nesting properties of its Fermi surface and points towards the possibility of a competition between $p$-wave spin triplet and $d$-wave spin singlet superconductivity in $Sr_2RuO_4$. We present a comparison between low energy spin fluctuations, converted in absolute units, from inelastic neutron scattering measurements and previous nuclear magnetic resonance studies.

PACS numbers: 78.70.Nx, 74.70.–b, 75.40.Gb

Being the only example of a noncuprate layered perovskite superconductor [1], $Sr_2RuO_4$ has attracted considerable attention despite its rather low critical temperature, $T_c \sim 1$ K [1–3]. Indeed, superconductivity in $Sr_2RuO_4$ is referred to as unconventional: (i) Cooper pairs are in a triplet spin state [4,5] and (ii) an anisotropic pairing is realized [6]. These unambiguous experimental evidences corroborate earlier theoretical predictions that superconductivity in $Sr_2RuO_4$ should possess $p$-wave symmetry (triplet pairing), mediated by strong ferromagnetic (FM) fluctuations that disfavor both $s$- and $d$-wave spin singlet superconductivity [7–9]. The fact that the three-dimensional parent compound $SrRuO_3$ is ferromagnetic, in addition to microscopic calculations of magnetic properties of ruthenates seem to support the idea of the predominant role of FM fluctuations in $Sr_2RuO_4$ [7,10]. 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 Ishida *et al.* [11] points towards the same conclusion.

The determination of the antiferromagnetic order in the closely related compound $Ca_2RuO_4$ [12,13] has suggested that the picture of a nearby FM instability in $Sr_2RuO_4$ is too simple. Furthermore, recent calculations which take into account the particular topology of the Fermi surface have predicted a sizable magnetic response at the incommensurate wave vector $(2\pi/3a, 2\pi/3a, 0)$ [14], i.e., far away from the zone center. The enhanced susceptibility arises from pronounced nesting properties of the almost one-dimensional $d_{xz,yz}$ bands. Mazin and Singh discuss the possibility of a competition between $p$-wave and $d$-wave superconductivity in $Sr_2RuO_4$ [14].

In this Letter, we report first inelastic neutron scattering (INS) measurements performed on single crystals of $Sr_2RuO_4$ in the normal state. Our data reveal dominant magnetic scattering at the incommensurate wave vectors $\mathbf{q}_0 = (\pm 0.6\pi/a, \pm 0.6\pi/a, 0)$, i.e., very close to the positions predicted by the band-structure calculations. The relevance of these findings for the mechanism of superconductivity in $Sr_2RuO_4$ will be discussed.

Most of the INS measurements presented here have been carried out on a single crystal of cylindrical shape (4 mm in diameter and 35 mm 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 setup 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 $\mathbf{Q} = (H,K,L)$ is indexed in units of the reciprocal tetragonal lattice vectors $2\pi/a = 2\pi/b = 1.63$ Å$^{-1}$ and $2\pi/c = 0.49$ Å$^{-1}$ ($I4/mmm$ space group) [1].

Figure 1 shows representative constant-$\omega$ scans taken in the $(H,K,0)$ plane: at $\hbar\omega = 6.2$ meV and around $\mathbf{Q}_0 = (1.3, 0.3, 0)$ along the $(0,1,0)$ direction. The scan at 10.4 K shows a sharp maximum of intensity peaked at $\mathbf{Q}_0 = (1.3, 0.3, 0)$ on top of a smooth background. At room temperature, this sharp peak has almost disappeared. The horizontal bar indicates the spectrometer resolution.

At 10.4 K, several constant-$\omega$ scans, with 6.2 meV energy transfer and performed along different directions $[(1,0,0), (0,1,0), (1,1,0), (1,-1,0)]$ have revealed the

 0031-9007/99/83(16)/3320(4)$15.00 

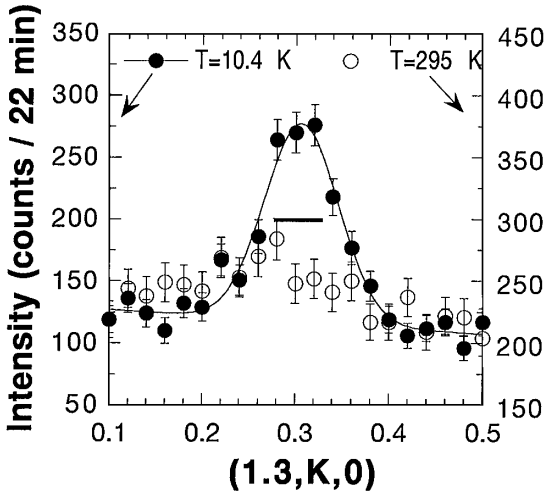


FIG. 1. Constant-$\omega$ scans performed at $\hbar\omega = 6.2$ meV around $\mathbf{Q} = (1.3, 0.3, 0)$ along the $(0, 1, 0)$ direction: $T = 10.4$ K (•), $T = 295$ K (∘).

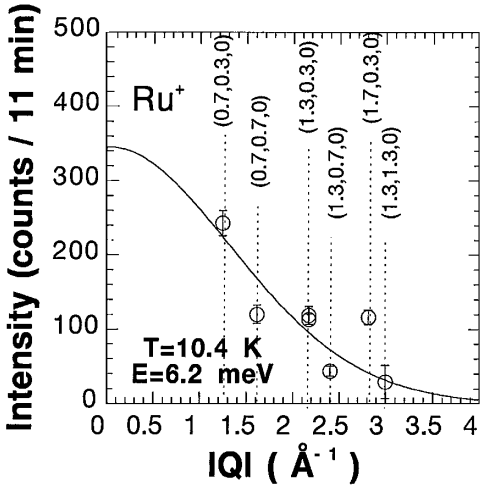


FIG. 2. Magnetic intensity, measured at $T = 10.4$ K and $\hbar\omega = 6.2$ meV as a function of $|Q|$. For each point, the corresponding wave vector, $(H, K, L)$, is also reported. The full line corresponds to the square of the $Ru^+$ magnetic form factor.

existence of comparable peaks at $\mathbf{Q}_0 = \mathbf{q}_0 + \mathbf{G}$, where $\mathbf{q}_0 = (\pm 0.3, \pm 0.3, 0) \equiv (\pm 0.6\pi/a, \pm 0.6\pi/a, 0)$ and $\mathbf{G}$ is a zone center or a $Z$ point (001) in the $(HK0)$ plane. The best fit of the data to a Gaussian profile [15] incorporating experimental resolution function demonstrates that the peak intensity is isotropic with an intrinsic $q$ width (FWHM), $\Delta q = 0.13 \pm 0.06$ Å$^{-1}$.

The interpretation of the scattering at $\mathbf{q}_0$ as magnetic in origin is supported by the large number of points in reciprocal space where it has been observed. Further, the lowest phonon frequencies at $\mathbf{q}_0$ are above 12 meV [16]. In addition, in contrast to a phonon-related scattering that increases at large $|Q|$ or with temperature, the scattering at $\mathbf{q}_0$ decreases both at large wave vector (Fig. 2) and at high temperature (Fig. 1). These different points establish the magnetic origin of the scattering observed around $\mathbf{q}_0$. In contrast, in spite of several attempts, no sizable FM spin fluctuations have been observed.

In a paramagnetic state, the magnetic neutron cross section per formula unit can be written in terms of the imaginary part of the dynamical spin susceptibility, $\chi''(\mathbf{Q}, \omega)$, as [17,18]

$$\frac{d^2\sigma}{d\Omega\, d\omega} = \frac{k_f}{k_i} r_0^2 \frac{2F^2(\mathbf{Q})}{\pi(g\mu_B)^2} \frac{\chi''(\mathbf{Q}, \omega)}{1 - \exp(-\hbar\omega/k_B T)}, \quad (1)$$

where $k_i$ and $k_f$ are the incident and final neutron wave vectors, $r_0^2 = 0.292$ barn, $F(\mathbf{Q})$ is the magnetic form factor, and $g \simeq 2$ is the Landé factor. The intensity of the scattering can be reasonably well described by the squared magnetic form factor of the $Ru^+$ ion [19] (note that the magnetic form factor of $Ru^{4+}$ is not available) after correction for geometrical factors related to the unfavorable shape of the sample; see Fig. 2. According to our measurements, the $q$ dependence of $\chi''$ can be parametrized by $\chi''(\mathbf{Q}, \omega) = \chi''(q_0, \omega) \exp[-4\ln(2)(\mathbf{Q} - \mathbf{Q}_0)^2/\Delta q^2]$.

The Fermi surface in $Sr_2RuO_4$ is formed by three sheets [14]: one, related to the $4d_{xy}$ orbitals is quasi-2D, whereas the two others, related to $4d_{xz,yz}$ orbitals, are quasi-1D. The 1D sheets can be schematically described by parallel planes separated by $\bar{q} = \pm 2\pi/3a$, running both in the $x$ and in the $y$ directions. These peculiarities give rise to dynamical nesting effects at the wave vectors $k = (\bar{q}, k_y)$, $k = (k_x, \bar{q})$, and, in particular, at $\bar{\mathbf{q}} = (\bar{q}, \bar{q})$. The nesting effects become dominant when calculating the bare spin susceptibility of a noninteracting metal [14], given by the Lindhard function [17]

$$\chi_0(q, \omega) = -2\mu_B^2 \sum_k \frac{f_{k+q} - f_k}{\varepsilon_{k+q} - \varepsilon_k - \hbar\omega + i\epsilon}, \quad (2)$$

where $\epsilon \to 0$, $f_k$ is the Fermi distribution function, and $\varepsilon_k$ the quasiparticle dispersion relation. Our INS data are in very close agreement with the predicted four spots of magnetic scattering situated at $\bar{\mathbf{q}} = (\pm 2\pi/3a, \pm 2\pi/3a)$ [14]. In the experiment the incommensurate magnetic responses are actually observed slightly away, at $\mathbf{q}_{0\parallel} = (\pm 0.6\pi/a, \pm 0.6\pi/a)$, which is most likely related to details of the band structure [14].

Let us now consider the energy dependence and magnitude of $\chi''(q_0, \omega)$. At $T = 10.4$ K, constant-$\omega$ scans have been measured at $\mathbf{Q} = (1.3, 0.3, 0)$ along the $(0, 1, 0)$ direction for different transferred energies between 2.4 and 12 meV. The magnetic response always displays 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 $\mathbf{Q} = (1.3, 0.3, 0)$ and at $\mathbf{Q} = (1.3, 0.46, 0)$, the latter providing a background reference. These measurements allow us to determine the energy dependence of the magnetic response at $\mathbf{q}_0$ from 1.5 to 12 meV. The analysis could not be extended to higher and lower energies due to the contaminations by phonon [16] and elastic incoherent scattering, respectively. Using Eq. (1), the magnetic intensity has been converted to the dynamical spin

susceptibility $\chi''$ after correction by the detailed balance factor and the squared magnetic form factor reported in Fig. 2. We have then calibrated $\chi''$ in absolute units against acoustic phonons, according to a standard procedure [20]. $\chi''(\omega, Q_0)$, whose energy dependence is reported in absolute units in Fig. 3, slightly increases up to 7 meV and then almost saturates. This energy dependence can be parametrized following linear response theory,

$$\chi''(q_0, \omega) = \chi'(q_0, 0) \frac{\Gamma \omega}{\omega^2 + \Gamma^2}, \qquad (3)$$

where $\Gamma$ is a damping energy of 9 meV and $\chi'(q_0, 0) = 180 \mu_B^2$ eV$^{-1}$ corresponds to the static spin susceptibility at $\mathbf{q}_0$ (solid line in Fig. 3). It is worth emphasizing that $\chi'(q_0, 0)$ is 6 times larger than that at $Q = 0$, i.e., the uniform susceptibility $\tilde{\chi} = \chi'(Q = 0, 0) = 30 \mu_B^2$ eV$^{-1}$ ($\simeq 10^{-3}$ emu/mole) [1–3]. As a comparison, in the high-$T_c$ cuprate system, $La_{1.86}Sr_{0.14}CuO_4$, the spin susceptibility at the incommensurate wave vectors exhibits almost the same magnitude and a similar $\omega$-dependence [21]. This is surprising as in cuprates, in contrast to $Sr_2RuO_4$, conventional band theory fails to account for the unusual normal state properties. Interestingly, spin fluctuations in both systems are strong and could eventually play a key role in the mechanism of superconductivity.

In $Sr_2RuO_4$, electronic correlations are incorporated in RPA calculations: the spin susceptibility $\chi(q, \omega)$ becomes enhanced through the Stoner factor $I(q)$ [9,14]:

$$\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - [I(q)/2(\mu_B)^2]\chi_0(q, \omega)}. \qquad (4)$$

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 $Sr_2RuO_4$: in our units, $I(q) = 0.43/[1 + 0.08(a/\pi)^2 q^2]$ eV ($q$ in Å$^{-1}$) [9,14]. 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 $\mathbf{q}_0$. With $\chi'(q_0, 0) = 180 \mu_B^2$ eV$^{-1}$, one deduces from Eq. (4) that $\frac{I(q_0)}{2(\mu_B)^2}\chi_0(q_0, 0) \simeq 0.97$, instead of being larger than 1 for a magnetic instability. Incommensurate spin fluctuations are stronger than FM fluctuations in $Sr_2RuO_4$, as suggested in Ref. [14].

The temperature dependence of both $\chi''(\mathbf{Q}_0, 6.2$ meV) and the intrinsic $q$ width are reported in Fig. 4, as deduced from constant-$\omega$ scans performed at 6.2 meV around $\mathbf{Q} = (1.3, 0.3, 0)$ along the $(0, 1, 0)$ direction at different temperatures. $\chi''(\mathbf{q}_0, 6.2$ meV) exhibits a sharp decrease upon temperature increase and simultaneously the magnetic response weakly broadens (the width of the signal can be reliably determined up to only 200 K). In spite of the fact that the system is close to a magnetic instability at $\mathbf{q}_0$, the dynamical magnetic susceptibility does not diverge and the correlation length remains finite down to the lowest temperature. The $T$-dependence of $\chi''$ observed in INS measurements may be described by the outsmearing 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 $\mathbf{q}_0$ and its broadening in $q$ space. The $T$-dependence of the magnetic response at $\mathbf{q}_0$ can indeed be qualitatively

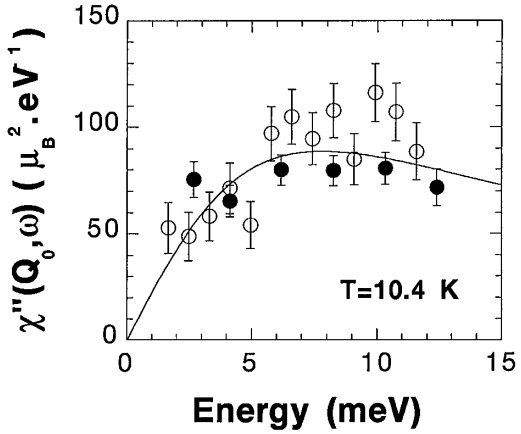


FIG. 3. Energy dependence of the imaginary part of the dynamical magnetic susceptibility at $\mathbf{Q}_0 = (1.3, 0.3, 0)$ as obtained from energy scans (∘) and constant energy scans around $\mathbf{Q}_0$ along the $(0, 1, 0)$ direction (•) (see text).

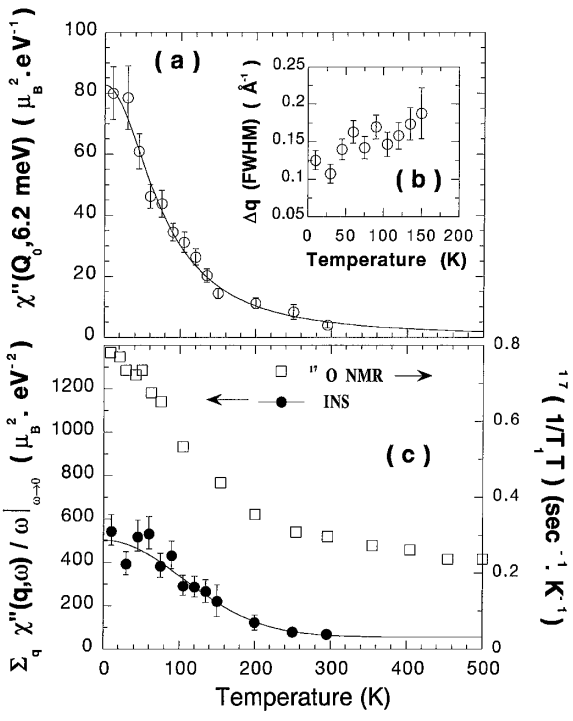


FIG. 4. Results from fits to a Gaussian profile of 6.2 meV constant-$\omega$ scans at $\mathbf{Q}_0 = (1.3, 0.3, 0)$ along the $(0, 1, 0)$: temperature dependences of (a) $\chi''(\mathbf{Q}_0, 6.2$ meV ) and (b) the intrinsic $q$ width of the magnetic signal, $\Delta q$ (FWHM). (c) Comparison between $^{17}(1/T_1T)$ observed by $^{17}$O NMR by Imai *et al.* [4] (□) and the incommensurate contribution calculated from our INS measurements (•). Assuming $\Lambda = 33$ kOe$/\mu_B$ [25], the two scales in this figure are identical. Solid lines are guides to the eye only.

reproduced [22] using Eqs. (2)–(4) and a description of the local density approximation band structure by three mutually nonhybridizing tight-binding bands [9].

INS measurements point out the existence of strong magnetic response at $\mathbf{q}_0$, but do not reveal any sizable FM fluctuations. In contrast, the uniform spin susceptibility [1–3] and the Knight shift measurements [4,23] provide evidence of strong FM correlation in $Sr_2RuO_4$. 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 [4,23]. 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 $(1/T_1T)$, $^{\mathrm{INS}}(1/T_1T)$. In general, $(1/T_1T)$ probes the $q$ summation of the imaginary part of the susceptibility divided by the frequency in the limit $\omega \to 0$ (i.e., its initial slope), $\sum_q \frac{\chi''(q,\omega)}{\omega}|_{\omega\to 0}$; its temperature dependency is shown in Fig. 4(c) (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 $\mathbf{q}_0$, one may approximate $A(q) = A(q_0)$ and get [24]

$$^{\mathrm{INS}}(1/T_1T) \simeq \frac{k_B\gamma_n^2}{(g\mu_B)^2}|A(q_0)|^2 \sum_q \frac{\chi''(q,\omega)}{\omega}\bigg|_{\omega\to 0} \qquad (5)$$

with $|A(q_0)|^2 = \Lambda^2\{1 + 1/2[\cos(2\pi 0.3) + \cos(2\pi 0.3)]\}$ ($\Lambda = 33$ kOe/$\mu_B$ [25]) for $^{17}O$ and $A(q_0) = -299$ kOe/$\mu_B$ [4,11] for $^{101}Ru$. Using these values, we directly compare $^{\mathrm{INS}}(1/T_1T)$ with the measured $^{17}O$ $(1/T_1T)$ in Fig. 4c (right scale). Clearly, the spin fluctuations at $\mathbf{q}_0$ significantly contribute to $^{17}(1/T_1T)$, and can explain a large part of the reported $T$-dependence [4,23]. 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 [22], 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 INS data.

To conclude, our INS measurements demonstrate the existence of incommensurate spin fluctuations related to dynamical nesting properties of the $Sr_2RuO_4$ Fermi surface. Our data suggest that the system is close to a magnetic instability at $\mathbf{q}_{0\|} = (\pm 0.6\pi/a, \pm 0.6\pi/a)$. 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 $\mathbf{q}_0$. All these results cast some doubt on the exclusive role of FM spin fluctuations in the mechanism of superconductivity in $Sr_2RuO_4$.

We acknowledge P. Pfeuty, J. Bobroff, and Ph. Mendels for helpful discussions.

[1] Y. Maeno *et al.,* Nature (London) **372**, 532 (1994).
[2] Y. Maeno *et al.,* J. Phys. Soc. Jpn. **66**, 1405 (1997).
[3] J. J. Neumeier *et al.,* Phys. Rev. B **50**, 17 910 (1994).
[4] T. Imai *et al.,* Phys. Rev. Lett. **81**, 3006 (1998).
[5] K. Ishida *et al.,* Nature (London) **396**, 658 (1998).
[6] G. M. Luke *et al.,* Nature (London) **394**, 558 (1998).
[7] T. M. Rice *et al.,* J. Phys. Condens. Matter **7**, L643 (1995).
[8] K. Machida *et al.,* J. Phys. Soc. Jpn. **65**, 3720 (1996).
[9] I. I. Mazin *et al.,* Phys. Rev. Lett. **79**, 733 (1997).
[10] I. I. Mazin *et al.,* Phys. Rev. B **56**, 2556 (1997).
[11] K. Ishida *et al.,* Phys. Rev. B **56**, R505 (1997).
[12] M. Braden *et al.,* Phys. Rev. B **58**, 847 (1998).
[13] S. Nakatsuji *et al.,* J. Phys. Soc. Jpn. **66**, 1868 (1997).
[14] I. I. Mazin and D. J. Singh, Phys. Rev. Lett. **82**, 4324 (1999).
[15] A Lorentzian profile may also account for the $q$-dependence of peak intensity, without significant modification of the overall conclusions of this Letter.
[16] M. Braden *et al.,* Phys. Rev. B **57**, 1236 (1998).
[17] S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Clarendon, Oxford, 1984), Vol. 2.
[18] The spin susceptibility is associated with fluctuations of a single spin component: $\chi(\mathbf{Q},\omega) = \chi^{\alpha\beta} = -(g\mu_B)^2\frac{i}{\hbar}\int_0^\infty dt \exp(i\omega t)\langle[S_i^\alpha(t), S_j^\beta]\rangle$, where $\alpha, \beta$ are Cartesian coordinates. $\chi$ is half of the transverse spin susceptibility $\chi^{+-}$.
[19] *International Tables of Crystallography*, edited by A. J. C. Wilson (Kluwer Academic, Dordrecht, 1995), Vol. C.
[20] We have used acoustic phonons in scans $(2 + \xi, 0, 0)$ and $(2, \xi, 0)$ for $\hbar\omega = 6.75$ and 8.26 meV (sound velocity $\simeq 20.8$ and 43.9 meV Å, respectively) [16].
[21] G. Aeppli *et al.,* Science **278**, 1432 (1997).
[22] P. Pfeuty (private communication).
[23] H. Mukuda *et al.,* J. Phys. Soc. Jpn. **67**, 3945 (1998).
[24] C. Berthier *et al.,* J. Phys. I (France) **6**, 2205 (1996); R. E. Walstedt *et al.,* Phys. Rev. Lett. **72**, 3610 (1994).
[25] For a magnetic field applied along the $c$ axis, $\Lambda^2 = |A_\||^2 + |A_\perp|^2$, with $A_\| = -18.5$ kOe/$\mu_B$ and $A_\perp = -26.8$ kOe/$\mu_B$ [23].