Anisotropy in the incommensurate spin fluctuations of Sr_2RuO_4

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(Received 25 July 2003; published 6 May 2004)

It has been proposed that Sr_2RuO_4 exhibits spin-triplet superconductivity mediated by ferromagnetic fluctuations. So far neutron-scattering experiments have failed to detect any clear evidence of ferromagnetic spin fluctuations but, instead, this type of experiments has been successful in confirming the existence of incommensurate spin fluctuations near $\mathbf{q}=(\frac{1}{3}\ \frac{1}{3}\ 0)$. For this reason there have been many efforts to associate the contributions of such incommensurate fluctuations to the mechanism of its superconductivity. Our unpolarized inelastic neutron-scattering measurements revealed that these incommensurate spin fluctuations possess *c*-axis anisotropy with an anisotropic factor $\chi_c''/\chi_{a,b}''$ of ~ 2.8. This result is consistent with theoretical ideas that the incommensurate spin fluctuations with a *c*-axis anisotropy may be the origin of *p*-wave superconductivity of this material.

DOI: 10.1103/PhysRevB.69.174501 PACS number(s): 74.70.Pq, 75.40.Gb, 78.70.Nx

I. INTRODUCTION

Sr_2RuO_4 is the first two-dimensional perovskite oxide material known to exhibit a superconducting transition without containing copper.[1] While Sr_2RuO_4 is isostructural with the high-T_c material $La_{2-x}Sr_xCuO_4$, its normal state shows Fermi-liquid behavior and its superconducting state is not a spin singlet ($S=0$) as observed in the conventional *s*-wave ($l=0$) superconductors or the *d*-wave ($l=2$) high-T_c materials. Its superconducting state is instead a spin triplet ($S=1$) with (most-likely) *p*-wave symmetry ($l=1$) (see Ref. 2 for a recent review).

Muon spin resonance (μSR) and NMR (Knight shift) measurements have provided experimental evidence of the spin-triplet pairing in Sr_2RuO_4. μSR measurements succeeded in confirming the existence of the spontaneous magnetic field below the superconducting transition temperature $T_c \sim 1.5$ K, indicating the time-reversal symmetry-breaking in superconducting state.[3] Knight shift measurements for the oxygen site in the RuO_2 planes revealed that the spin susceptibility remains temperature independent even below T_c.[4]

Since T_c of Sr_2RuO_4 (~ 1.5 K) is strongly suppressed by nonmagnetic impurities,[5] electron-phonon coupling cannot be the origin of pairing mechanism. From the analogy with the superfluid state of ^{3}He and from the fact that the neighbor material $SrRuO_3$ is ferromagnetic, it was speculated that Sr_2RuO_4 exhibits spin-triplet superconductivity mediated by ferromagnetic fluctuations. Up to this date, however, there is no clear experimental evidence of ferromagnetic fluctuations in this material.[6–8]

The electronic structure of Sr_2RuO_4 (*d*-electron system) is much simpler than those of other spin-triplet superconductors.[9,10] This fact has stimulated theoretical efforts on the topics of spin-triplet superconductivity and the symmetry of superconducting order parameters.

Mazin and Singh have calculated the electronic band structure of Sr_2RuO_4 based on the t_{2g} orbital of the Ru^{4+} ($4d^4$) and showed that the Fermi surfaces consist of quasi-one-dimensional α, β planes defined by the d_{yz}, d_{zx} orbital, and of two-dimensional γ planes defined by the d_{xy} orbital.[11] These predictions are consistent with the results of de Haas–van Alphen experiments.[12] Furthermore, the theory predicted that sizable nesting effects in the quasi-one-dimensional bands (α, β planes) may cause the enhancement of the spin susceptibility near the incommensurate propagating vector $\mathbf{q}=(\frac{1}{3}\ \frac{1}{3}\ 0)$.[13] Such an enhancement was indeed confirmed in dynamical spin susceptibility $\chi''(\mathbf{q}_0,\omega)$ at $\mathbf{q}_0=(0.3\ 0.3\ 0)$ by inelastic neutron-scattering (INS) experiments.[6] These results stimulated discussions about the possibility of *p*-wave superconductivity mediated by such incommensurate spin fluctuations.-

Some theoretical works reported that if such incommensurate spin fluctuations possess *c*-axis anisotropy, the spin-triplet superconductivity could be stabilized by such fluctuations.[14–16] It is therefore of great importance to establish if there is any observable anisotropy in $\chi''(\mathbf{q}_0)$ that can be related to the origin of the spin-triplet superconductivity in Sr_2RuO_4.

Ishida *et al.* have reported the observation of the anisotropic behavior of the spin susceptibility measured by the NMR technique.[17] In NMR measurements, one can observe the **q**-integrated spin susceptibility $\Sigma_{\mathbf{q}}\chi''(\mathbf{q},\omega)/\omega|_{\omega\to 0}$. Judging from the similarities with the INS data reported by Sidis *et al.*,[6] Ishida *et al.* attributed the temperature-dependent part of the **q**-integrated spin susceptibilities (observed by the NMR) to the spin susceptibility at $\mathbf{q}_0$, and reported that $\chi''(\mathbf{q}_0,\omega)$ has a *c*-axis anisotropy with an anisotropic factor $\chi_c''/\chi_{a,b}''$ $[\chi''_{IC,out}(\mathbf{q}_0,\omega)/\chi''_{IC,in}(\mathbf{q}_0,\omega)$ in their notation] of ~ 3.

In order to ascertain the anisotropic nature of the incommensurate spin fluctuations in Sr_2RuO_4, however, it is necessary to measure its **q** dependent spin susceptibility $\chi''(\mathbf{q},\omega)$ using INS. We have performed such measurement and found that the dynamical spin susceptibility of this system at $\mathbf{q}_0$ indeed exhibits *c*-axis anisotropy with an anisotropic factor of ~ 2.8. This value is consistent with the anisotropic factor estimated from the NMR measurements.[17] Our conclusion is different from those of the recent reports by Servant *et al.* and Braden *et al.*,[7,8] this will be discussed at the end of this paper.

II. EXPERIMENT

A. Sample preparation and experimental setup

For this neutron-scattering study, we grew large single crystals of Sr_2RuO_4 by the floating-zone method. The crystals were cut into smaller cylindrical pieces (4 mm in diameter and 30 mm in length). We performed resistivity measurements on these crystals using a Quantum Design PPMS instrument equipped with a ^{3}He option. These measurements revealed that T_c(onset) of all the samples lies between 1.4 and 1.6 K.

The unpolarized INS experiments were performed using the triple-axis spectrometer GPTAS installed at the JRR-3M reactor at the Japan Atomic Energy Research Institute (JAERI) in Tokai, Japan. Neutrons with a fixed final momentum of $k_f = 3.83$ Å^{-1} and a combination of horizontal collimations of 40′-80′-40′-80′ [FWHM (full width space at half maximum) from the monochromator to the detector] were utilized. A pyrolytic graphite filter was placed after the sample position to eliminate higher-order wavelength contaminations. Three sets of crystals were prepared in order to probe three different scattering planes, $(h\ k\ 0)$, $(h\ h\ l)$, and $(0.7h\ 0.3h\ l)$. The total volume of each of these sets was ~ 3 cm^3.[18] The crystals were sealed in aluminum cans (filled with He exchange gas to ensure a uniform temperature) that were attached to the cold head of a closed-cycle He gas refrigerator. Throughout this paper the scattering vector $\mathbf{Q} = (Q_h\ Q_k\ Q_l)$ is indexed in reduced lattice units with tetragonal reciprocal lattice of $a^* = b^* = 1.63$ Å^{-1} and $c^* = 0.49$ Å^{-1}, respectively.

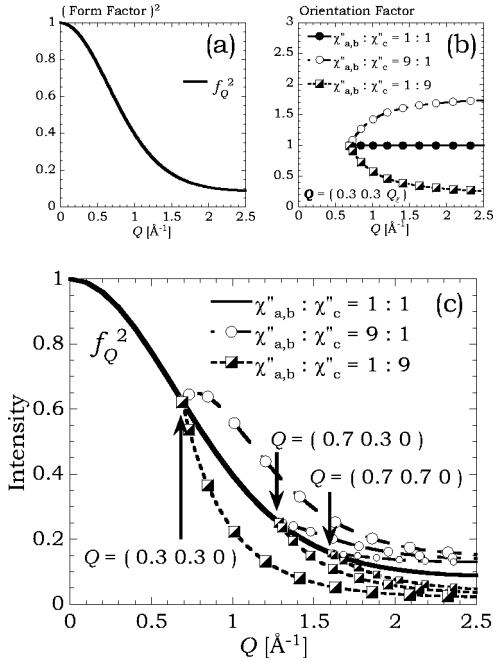

FIG. 1. (a) A schematical Q dependence of the square of magnetic form factor f_Q^2. (b) Q dependence of calculated orientation factors $G(\theta)$ at $\mathbf{Q} = (0.3\ 0.3\ Q_l)$ for different anisotropic factors. Depending on a ratio $\chi''_{a,b} : \chi''_c$, $G(\theta)$ shows different Q dependence. (c) Q dependence of calculated intensities $I[= f_Q^2 \times G(\theta)]$ at $(0.3\ 0.3\ Q_l)$,$(0.7\ 0.3\ Q_l)$, and $(0.7\ 0.7\ Q_l)$ with different anisotropic factors.

B. Magnetic neutron scattering

In this section we describe the method that we used to measure the anisotropic factor of the spin susceptibility $\chi''(q,\omega)$. In a magnetic neutron-scattering experiment,[19] the scattering intensity I is given by

$$I \propto f_Q^2 G, \tag{1}$$

where f_Q is the magnetic form factor, which is the Q component of the Fourier transform of the distribution of unpaired electrons that contribute to the magnetism in the system. If the electronic distribution is isotropic, f_Q shows a monotonic decrease with the absolute value of the scattering vector **Q**, Q as demonstrated in Fig. 1(a). The quantity G in Eq. (1) is an orientation factor related to the fact that neutrons are scattered only by the magnetic components perpendicular to the scattering vector **Q**. In the present study, we assumed that the spin susceptibility within the RuO_2 planes in tetragonal Sr_2RuO_4 is isotropic $(\chi_a'' = \chi_b'' = \chi_{a,b}'')$. The orientation factor G is then given by

$$G(\theta) = (1 + \sin^2\theta)\chi''_{a,b}(\mathbf{q},\omega) + \cos^2\theta\chi''_c(\mathbf{q},\omega), \tag{2}$$

where θ is the angle between the scattering vector **Q** and the ab plane, which changes through the Q_l component of the scattering vector.

In Fig. 1(b) we show a calculation of the Q dependence of $G(\theta)$ for $\mathbf{Q} = (0.3\ 0.3\ Q_l)$ for different anisotropic factors. $G(\theta)$ is constant when the susceptibility is isotropic

$(\chi''_{a,b}=\chi''_c)$ but shows different Q dependence with anisotropy $(\chi''_{a,b}\neq\chi''_c)$; namely, $G(\theta)$ increases (decreases) with Q when $\chi''_c/\chi''_{a,b}<1(>1)$.

The Q dependence of the total intensity I given by Eq. (1) is shown for (0.3 0.3 Q_l), (0.7 0.3 Q_l), and (0.7 0.7 Q_l) in Fig. 1(c). If $\chi''(\mathbf{q},\omega)$ is isotropic, the intensity I is scaled only by f_Q^2 but it decreases slower (faster) than f_Q^2 in the presence of anisotropy $\chi''_c/\chi''_{a,b}<1(>1)$. This illustrates the fact that the anisotropic nature of the spin fluctuations can be directly determined by the comparison of the Q dependence of the intensity I and f_Q^2. We would like to stress that knowing the magnetic form factor accurately is the key to the accurate determination of the spin-susceptibility anisotropy factor. Unfortunately the only magnetic form factor that can be found in the literature for ruthenium is that for Ru^+ $[f_Q(Ru^+)]$.[20] The Ru ions in Sr_2RuO_4 are not Ru^+ but Ru^{4+}(nominally). Furthermore, Sr_2RuO_4 is not an insulator but an itinerant electron system, and the use of $f_Q(Ru^+)$ to characterize the magnetic response of Sr_2RuO_4 is clearly inadequate. For this reason we decided to determine the magnetic form factor for Sr_2RuO_4 $[f_Q(Sr_2RuO_4)]$ experimentally.

C. Determination of the magnetic form factor of Sr_2RuO_4

To determine the magnetic form factor of Sr_2RuO_4, $f_Q(Sr_2RuO_4)$, we measured the Q dependence of spin susceptibility at several **Q** positions with $\mathbf{q}_0=(0.3\ 0.3\ 0)$ in the (h k 0) plane ($\theta=0$). The Q dependence of the observed intensities is shown in Fig. 2, the filled and open symbols indicate our data and those reported by Sidis *et al.*,[6] respectively. Note that throughout the present paper all the quoted intensities have been corrected for resolution-volume effects, and that all the quoted experimental errors correspond to 2σ in order to reflect the ambiguities of the scattering technique.

Note that (0.7 0.3 0) and (1.3 0.3 0) are not $\mathbf{q}_0$ positions from the reciprocal zone center, Γ point, but those from the Z point [e.g., (1 0 0)]. These data can be treated equally with other data, because the spin susceptibility at $\mathbf{q}_0$ shows a strong two dimensionality and a rod-type scattering along the c^* axis so that one can observe the signal even on the (h k 0) zone.

Our first observation of Fig. 2 is that it is clear that the data do not scale with $f_Q^2(Ru^+)$, and decrease faster than it. This behavior is consistent with the fact that Sr_2RuO_4 is an itinerant electron system where mobile electrons distribute wider in real space than localized electron system and strongly indicates that one cannot use $f_Q^2(Ru^+)$ to evaluate anisotropic factor of spin susceptibility of Sr_2RuO_4.

The full line in this figure is $f_Q^2(Sr_2RuO_4)$ determined in the present study, then we fitted the observed intensities to the expression

$$f_Q(Sr_2RuO_4)=A\exp[B(Q/4\pi)^2]+C. \tag{3}$$

Here, we assumed that the f_Q in the (h k 0) plane is isotropic, so that the Q dependence of the f_Q is described as a single Q function.[21]

This form factor was used to evaluate the anisotropic factor of the incommensurate spin fluctuations. Note that conductivity and coherence length of Sr_2RuO_4 show anisotropic behavior ($\sigma_{a,b}>\sigma_c$ and $\xi_{a,b}>\xi_c$).[2] Such results indicate that a distribution of unpaired electrons along the c axis is confined and then the decrease of f_Q with Q_l must be slower than that for Q_h or Q_k. It should be stressed here that we assumed an isotropic form factor f_Q in all directions in the present study which causes an *underestimation* of the c-axis anisotropy.

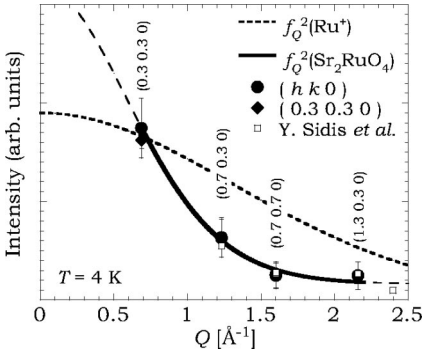

FIG. 2. Q dependence of intensities observed at **Q** with $\mathbf{q}_0=(0.3\ 0.3\ 0)$ in the (h k 0) plane ($\theta=0$). Filled symbols are the present results, in which circle and diamond symbols correspond to the data taken with different sample set with (h k 0) and (h h l) with $l=0$, respectively. Open symbols are taken from Ref. 6 reported by Sidis *et al.* Taking into account ambiguities of scattering experiments, we conservatively adopt 2σ error bars. A broken and a full line correspond to the square of magnetic form factor of Ru^+, $f_Q^2(Ru^+)$, and that of Sr_2RuO_4, $f_Q^2(Sr_2RuO_4)$, respectively. The latter was determined and parametrized with Eq. (3) in the present study. (See the main text.)

III. EXPERIMENTAL RESULTS

A. Q_l dependence of intensity

In order to study the $Q_l(\theta)$ dependence of the intensity, we performed several series of constant-E scans along (0.3 0.3 Q_l), (0.7 0.7 Q_l), (0.7 0.3 Q_l), and (1.3 0.7 Q_l), and found that, because of low intensity, it is difficult to get accurate Q_l dependence at **Q** positions farther than (0.7 0.7 Q_l). For this reason we report only the results at the (0.3 0.3 Q_l) and (0.7 0.3 Q_l) scans. Furthermore, to collect reliable data, one needs to select a clear window of energy where any spurious peaks including phonon scattering do not appear. For the constant-E scans at (0.3 0.3 Q_l) and (0.7 0.3 Q_l), neutron-transfer energies were selected to be 4 meV and 8 meV, respectively, by measurements of energy dependence of intensity at (0.3 0.3 0) and (0.7 0.3 0) with energy transfer between $\sim -2<E<\sim 20$ meV. The energy dependence of intensity at (0.3 0.3 0) is shown in an inset of Fig. 3(a). The result clearly shows that the intensity at $E=4$ meV is affected by neither incoherent nor forward scattering.

Q_l dependences of integrated intensity at (0.3 0.3 Q_l) and (0.7 0.3 Q_l) are depicted in Fig. 3(a); the integrated intensities were calculated as the product of intensities at (0.3 0.3 Q_l), (0.7 0.3 Q_l) and the width determined by constant-E

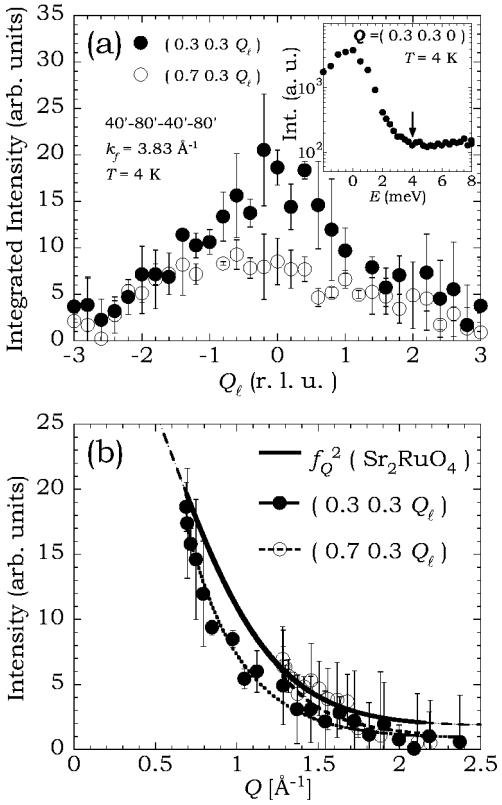

FIG. 3. (a) Q_l dependence of integrated intensity I at (0.3 0.3 Q_l) and (0.7 0.3 Q_l) and at 4 K. Some data points are missing because of uncertainty due to existence of spurious peaks. Inset : Energy dependence of scattering intensity at (0.3 0.3 0). (b) Q dependence of resolution corrected intensity at (0.3 0.3 Q_l) and (0.7 0.3 Q_l). Averaged values between (Q_h Q_k $|Q_l|$) and (Q_h Q_k $-|Q_l|$) are plotted. The full line is the square of magnetic form factor of Sr_2RuO_4, $f_Q^2(Sr_2RuO_4)$, determined in the present study (see Fig. 2). Dashed line on (0.3 0.3 Q_l) is a fitting curve to Eqs. (1) and (2) with $\chi_c''/\chi_{a,b}''=2.8$ and a broken line on (0.7 0.3 Q_l) is a simulation line calculated with the parameters evaluated by data at (0.3 0.3 Q_l). In both plots, 2σ was conservatively adopted as error bars.

scans along the (h k 0) direction. The obtained widths at (h h Q_l) and (0.7h 0.3h Q_l) were almost constant with Q_l, and we used their averaged values, cf. 0.188 and 0.184 Å^{-1} (in FWHM), respectively. In addition, intensities at (0.5 0.5 Q_l) and (0.7 ± 0.1 0.3 ± 0.04 Q_l), which are almost constant with Q_l, were subtracted as background for calculations of peak intensities at (0.3 0.3 Q_l) and (0.7 0.3 Q_l), respectively. Finally the data at (0.7 0.3 Q_l) with 8 meV were scaled with the data at (0.3 0.3 Q_l) with 4 meV by detailed measurements of energy dependence of signals.

The Q_l dependence of the integrated intensity (0.3 0.3 Q_l) and (0.7 0.3 Q_l) in Fig. 3(a) shows a very broad peak centered at $Q_l=0$, indicating the strong two dimensionality of the spin fluctuations. This result is consistent with the one reported by Servant *et al.*,[7] and allows us to neglect the magnetic correlations along the c axis. Thus we treat data sets with different Q_l independently.

B. Determination of the anisotropic factor

Figure 3(b) shows the Q dependence of the intensities for (0.3 0.3 Q_l) and (0.7 0.3 Q_l), the full line is the magnetic form factor $f_Q^2(Sr_2RuO_4)$ that we measured as described above. This figure clearly shows that the intensities for both (0.3 0.3 Q_l) and (0.7 0.3 Q_l) decrease faster than $f_Q^2(Sr_2RuO_4)$ with increasing Q. Such Q dependence corresponds to the case with $\chi_{a,b}''<\chi_c''$ as demonstrated in Fig. 1(c).

To evaluate the anisotropic factor, Q dependence data for (0.3 0.3 Q_l) was fitted to Eqs. (1) and (2), but the data for (0.7 0.3 Q_l) was not used in this fit because of the poor statistics. From the fitting, we obtained the anisotropic factor of the spin susceptibility $\chi_c''/\chi_{a,b}''\sim2.8\pm0.7$. Note that to evaluate an error in the determination of the anisotropic factor we took into account the error of the magnetic form factor. Furthermore, as explained in the preceding section, we assumed an isotropic form factor and such assumption may cause the underestimation of the c-axis anisotropy. These results let us conclude that the incommensurate antiferromagnetic fluctuations observed at $\mathbf{q}_0=(0.3\ 0.3\ 0)$ exhibit c-axis anisotropy.

IV. DISCUSSION

A. Static and dynamical spin susceptibility observed in Sr_2RuO_4

The magnetic properties in the normal state of Sr_2RuO_4 reported so far are (a) slightly anisotropic uniform susceptibility at $q=0$,[22] (b) anisotropic spin fluctuations with anisotropic factor of $\chi_c''/\chi_{a,b}''\sim3$ at somewhere in $\mathbf{q}$ reported by NMR,[17] (c) spin fluctuations observed at incommensurate $\mathbf{q}$ of (0.3 0.3 0) observed by INS.[6]

The uniform susceptibility of (a) is explained by the Pauli paramagnetism of the conduction electrons in a two-dimensional γ band, and the origin of a slight anisotropy of it ($\chi_c/\chi_{a,b}\sim1.1$) is attributed to the orbital Van Vleck contribution, which is affected by fields parallel to the c axis due to the one-dimensional α and β bands.[23]

On the other hand, the anisotropy in (b) cannot be associated with that in (a), because the anisotropic factor and energy scale of each spin susceptibilities are quite different. Judging from the similarity in temperature dependence of spin fluctuations in (b) and (c), Ishida *et al.* speculated that the anisotropic behavior observed in the NMR measurements has a close relation with spin fluctuations observed at (0.3 0.3 0).[17] Supporting this, our result clearly revealed that the incommensurate spin fluctuation has anisotropy with an anisotropic factor of $\chi_c''/\chi_{a,b}''\sim2.8$. The anisotropic factor reported by NMR measurement is ~3, which is in good agreement with the present result. These results let us conclude that anisotropic behavior observed by the NMR measurements is associated with spin fluctuations at incommensurate $\mathbf{q}_0$ vector of (0.3 0.3 0).

B. The origin of the anisotropic behavior

A short comment about the origin of the anisotropy of the incommensurate spin fluctuations observed at $\mathbf{q}$

$=(0.3\ 0.3\ 0)$. Theoretical calculations within the random-phase approximation using a three-band Hubbard Hamiltonian predict that spin-orbit coupling plays an important role and that, due to strong coupling, the out-of-plane component of the spin susceptibility is about two times larger than the in-plane one at low temperature.[24] The calculated anisotropy and our result are quantitatively consistent. Magnetic properties of (a), (b) and (c) are associated with the orbital of *d* electrons in RuO_2 planes. These facts strongly indicate that the spin-orbit interaction is important to discuss the magnetic properties of this system.

C. Relation between incommensurate spin fluctuations and the superconducting mechanism

As described in the Introduction, some theoretical groups reported that incommensurate spin fluctuations with a *c*-axis anisotropy, $\chi_c'' > \chi_{a,b}''$, may introduce a spin-triplet superconducting state and that the *d* vector turns to a direction of larger antiferromagnetic fluctuations.[14–16] Our results show that the incommensurate spin fluctuations observed in Sr_2RuO_4 satisfy this requirement, namely, $\chi_c'' > \chi_{ab}''$, which makes a direction of *d* vector to be parallel to the *c*-axis consistent with the experimental observations.[3,4,25]

Then the question here is whether these spin fluctuations are really driving forces of the superconductivity of this material or not. Basically, the superconductivity of Sr_2RuO_4 is believed to originate in the quasi-two-dimensional γ main band. On the other hand, the incommensurate antiferromagnetic fluctuations of Sr_2RuO_4 is caused due to the nesting property of the one-dimensional α and β bands. Furthermore, in the $Sr_2Ru_{1-x}Ti_xO_4$ (in which superconductivity is quickly suppressed and the antiferromagnetic fluctuations observed at $x = 0$ develop into a static order with increasing x), the x dependence of T_c seems to be explained only by a doping effect and no enhancement of T_c by the spin fluctuations was observed.[26–28] These results imply that the incommensurate spin fluctuations may not contribute to its superconducting mechanism.[28] In order to further clarify the mechanism of the superconductivity in Sr_2RuO_4, especially of relations between spin-triplet superconductivity and antiferromagnetic fluctuations, information of $\chi''(\mathbf{q}_0, \omega)$ behavior below T_c would be of great help.

D. Discrepancies with unpolarized INS results by other groups

In the present study, we measured data at $(0.3\ 0.3\ Q_l)$ and $(0.7\ 0.3\ Q_l)$ including $(0.3\ 0.3\ 0)$ ($Q \sim 0.70$ Å^{-1}) and estimated the anisotropic factor $\chi_c''/\chi_{a,b}''$ to be ~ 2.8 by evaluating a difference between the I and $f_Q^2(Sr_2RuO_4)$. These results, however, are at odds with other unpolarized neutron-scattering data reported by Servant *et al.*[7] and by Braden *et al.*[8] We attribute these discrepancies to (a) the narrower Q range in these groups' measurements, and (b) a lack of determination of $f_Q^2(Sr_2RuO_4)$ by the other groups. For example, Servant *et al.* measured data at $(0.3\ 0.3\ Q_l)$ with only larger Q part ($Q > 0.80$ Å^{-1}) and $(0.7\ 0.7\ Q_l)$, and concluded an isotropic behavior of spin fluctuations based on the fact that a small number of data points observed along $(0.3\ 0.3\ Q_l)$ and $(0.7\ 0.7\ Q_l)$ scaled at a very narrow Q range near ~ 1.6 Å^{-1}. One can see in Fig. 3(b) of our paper that the accuracy of the data in that Q range (near ~ 1.6 Å^{-1}) is not very good. We also observed data at $(0.7\ 0.7\ Q_l)$ and found that the data scaled with those along $(0.3\ 0.3\ Q_l)$ in this Q range within the huge error bars. Furthermore, they did not get the proper magnetic form factor for Sr_2RuO_4; this prevented them from making a reliable comparison of their data with the magnetic form factor in the small Q region. On the other hand, Braden *et al.* observed Q dependence of I at $(0.3\ 0.3\ Q_l)$ within a very narrow Q range of 1.2–2.5 Å^{-1} and showed that the data decrease slower than that of $f_Q^2(Ru^+)$.[8] This behavior is clearly opposite to our data shown in this paper and to the data of Servant *et al.*[7] At this time we do not understand the source of this discrepancy.

V. SUMMARY

We have performed unpolarized inelastic neutron-scattering measurements on Sr_2RuO_4 to probe the anisotropic behavior of the spin susceptibility observed at the incommensurate wave vector of $\mathbf{q}=(0.3\ 0.3\ 0)$. Our measurements indeed support that the susceptibility exhibits a *c*-axis anisotropy, i.e., $\chi_c''/\chi_{a,b}'' \sim 2.8 \pm 0.7$. This anisotropy ratio is in good agreement with the result obtained by the NMR measurements (~ 3).[17]

Note added in proof. Recently, we became aware that neutron polarization analysis experiments have been performed on Sr_2RuO_4 by two independent groups. These groups succeeded in confirming the *c*-axis anisotropy with anisotropic factor of 2–2.5 (Ref. 29) and 2.0 ± 0.4 (Ref. 30), respectively, being consistent with our unpolarized neutron results presented here. It can be argued that the best way to perform this type of measurements is using the polarized neutron-scattering technique because this technique allows the separation of magnetic components to the scattering from any other nonmagnetic components including phonon and spurious peaks. We would like to stress here, however, that this is not the only reliable way to measure magnetic components. It is true that the unpolarized neutron technique is intrinsically more ambiguous when it comes to measure magnetic components. But being conscious of this fact, we paid the greatest care to reduce such errors, and we made many consistency checks with different scattering zones and even checked background from the cryostat and judiciously chose the best conditions for the experiment. Every experimental result presented in our paper has been examined with great caution and our results are reliable.

ACKNOWLEDGMENTS

We would like to acknowledge Dr. J. A. Fernandez-Baca for valuable discussions and for a critical reading of the manuscript. T.N., H.F., and H.Y. were supported by a Grant-In-Aid from the Ministry of Education, Culture, Sports, Science and Technology, Japan. P. Dai is supported by US NSF DMR-0139882 and DOE under contract No. DE-AC-00OR22725 with UT/Battelle, LLC.

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[18] In order to keep effective sample volume constant, we tried to assemble single crystals symmetrically. Then, to check a reliability, we have measured a ratio of intensities between (0.3 0.3 0) and (0.7 0.7 0) with different sample sets with different scattering planes ($h\ k\ 0$) and ($h\ h\ l$), and found that the ratio is same within experimental accuracy. We also checked absorption factors of Sr, Ru, and O atoms in a text book. They are negligibly small. From these facts, we concluded that geometrical corrections are not necessary in the present experiments, and we just corrected the observed intensity by the instrumental resolution.
[19] As a text, see *Neutron Scattering with a Triple-Axis Spectrometer (Basic Techniques)*, edited by G. Shirane, S.M. Shapiro, and J.M. Tranquada (Cambridge University Press, Cambridge, 2002).
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[21] If there is a remarkable anisotropy in the in-plane susceptibility, data at (0.3 0.3 0) ($Q=0.69$ Å^{-1}), (0.7 0.7 0) ($Q=1.61$ Å^{-1}), and that of (0.7 0.3 0) ($Q=1.25$ Å^{-1}) draw different lines. Within experimental accuracy, such a distinct behavior is not observed, indicating that the sizable anisotropy of magnetic form factor in the RuO_2 plane was not detected.
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