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Magnetic chirality of the spin triplet in the spin-ladder compound $\mathrm{Sr_{14}Cu_{24}O_{41}}$ as seen via polarized inelastic neutron scattering

J. E. Lorenzo,[1] L. P. Regnault,[2] C. Boullier,[2] N. Martin,[2] S. Vanishri,[3] and C. Marin[3]

[1]*Institut Néel,Centre de la Recherche Scientifique/Université Joseph Fourier, F-38042 Grenoble, Cedex 9, France*

[2]*Service de Physique Statistique, Magnétisme et Supraconductivité, Commissariat a la Énergie Atomique,Institut de Nanotechnologie et Cryogénie/Université Joseph Fourier-Grenoble 1, laboratoire de Magnétisme et Diffraction Neutronique, F-38054 Grenoble Cedex 9, France*

[3]*Service de Physique Statistique, Magnétisme et Supraconductivité, Commissariat a la Énergie Atomique-Institut de Nanotechnologie et Cryogénie/Université Joseph Fourier-Grenoble 1, laboratoire Instrumentation, Matériaux Avancés, Physique des Électrons Corrélés (IMAPEC), F-38054 Grenoble Cedex 9, France*

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We report the direct observation by polarized inelastic neutron scattering experiments at low fields (3.5 T) of the splitting of the spin triplet of magnetic excitations in the response associated with the ladders in the composite cuprate $\mathrm{Sr_{14}Cu_{24}O_{41}}$. By conveniently choosing the magnetic field configuration and by making use of the spin chirality of the excitations, the splitting can be observed at relatively low fields. In this way, resonant excitations can be separated from the remainder of magnetic and phonon excitations at high temperatures. In $\mathrm{Sr_{14}Cu_{24}O_{41}}$, the 32-meV gapped mode vanishes above ~200 K, close to the hole-crystal melting temperature previously observed by resonant x-ray and neutron diffraction.

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Introduction. The study of the excitations in low-dimensional quantum magnetic systems is a subject of great interest, undoubtedly sparked by the discovery of the resonance peak in the high-T_C cuprates and more recently by the realization that a similar response appears in the superconducting pnictides. Despite the abundant literature, many questions remain as yet unanswered.[1] Among these, the degeneracy of the resonance stands as a long-time query. Namely, is the resonance peak a triplet, a doublet, or a spin-wave-like excitation, or something else? Or is the resonance effectively disappearing at T_C? Very oddly, these questions have never been unambiguously solved in the high-T_C cuprates. Certainly the magnetic fields required to split this resonance (in the case of a triplet) beyond the instrument resolution are prohibitively large for commercial split-pair superconducting magnets (delivering at best 17 T).

Here we have undertaken the study of *another* resonance peak issued from the condensation of holes into a charge density wave (CDW) in the ladder subsystem of $\mathrm{Sr_{14}Cu_{24}O_{41}}$(Ref. 2). We have applied a recently developed technique[2] that takes advantage of the *longitudinal* polarized neutron scattering cross section in combination with moderate magnetic fields (3.5 T). We will summarize this approach and show how fields of less than 2 T can be used to monitor the nature of these quantum excitations.

Longitudinal polarization analysis and trivial magnetochirality. Spin waves are odd-parity dynamical objects, characterized by both symmetric and antisymmetric spin-spin correlation functions. Although the symmetric correlation functions are currently measured in a regular, unpolarized, inelastic-neutron-scattering (INS) experiment, little is known about the effect of the antisymmetric part and thus about the dynamical magnetochirality. Antisymmetric correlation functions can be revealed through neutron *longitudinal* polarization analysis studies (LPA), provided that spin domains can be dissymmetrized. In the case of a uniaxial ferromagnetic compound a magnetic field can be used to favor a single domain state. More challenging is the case of antiferromagnetic compounds where both spin states are part of the chiral antisymmetric scattering cross section and thus very difficult to disentangle. A particular case where the antisymmetric spin-spin correlation functions can be extracted in an antiferromagnetic compound is realized in spin-singlet ground-state compounds.[2]

In particular and for neutron polarization parallel (antiparallel), + (−), to the scattering vector Q, the magnetic components accessible for scattering are those perpendicular to Q, and in addition, the neutron polarization is spin flipped after scattering. The scattering cross section for the spin-flip process parallel to Q reads

$$\sigma_x^{\pm\mp} \propto M_y + M_z \mp M_{ch},$$

where $\sigma_\alpha^{\beta,\gamma}$ is the short form of $(d^2\sigma/d\Omega d\omega)^{\beta,\gamma}(P_0 \parallel \alpha)$ and $M_y = \langle M_{Qy} M_{Qy}^\dagger\rangle_\omega$, $M_z = \langle M_{Qz} M_{Qz}^\dagger\rangle_\omega$, $M_{ch} = i(\langle M_{Qy} M_{Qz}^\dagger\rangle_\omega - \langle M_{Qz} M_{Qy}^\dagger\rangle_\omega)$, where $\langle M_{Q\alpha} M_{Q\alpha}^\dagger\rangle_\omega$ (α = y, z) are the space and time Fourier transforms of the symmetric spin-spin correlation functions, respectively. M_{ch} is the chiral (or antisymmetric) correlation function. Two remarks are worth noting: (i) M_{ch} is an intrinsic feature of the magnetic excitations, which are chiral objects, independent of the interactions that may favor chiral spin arrangements. We have named M_{ch} as *trivial* magnetochirality.[2] (ii) The antisymmetric part of the interference terms and the symmetric counterpart of the chiral correlation function are not accessible by the LPA technique as the polarization of the incident neutrons results rotated after scattering by these terms. In order to access these correlation functions the use of spherical neutron polarimetry based on, e.g., "Cryopad" devices[3,4] is mandatory.

Quantum dimers and INS. Magnetic excitations in antiferromagnetic quantum spin-dimer chain and ladders are very well defined and long-lived at low temperatures; the ground state remains spin singlet all the way down to the lowest temperature. Excitations at low temperature are therefore composite bosons of two-particle states forming a four-state base: a nonmagnetic singlet $|0\rangle = |00\rangle = |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$ and a magnetic triplet $|1\rangle = \{|10\rangle, |11\rangle, |1\bar{1}\rangle\} = \{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle, |\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$. Each component of the triplet is going to split under the application of a magnetic field following the Zeeman energy contribution, $\hbar\omega_{1,\bar{1}} = \hbar\omega_0 \mp g\mu_B H$.

In a previous paper we showed that the correlation functions for spin-singlet to spin-triplet transitions are just textbook calculations.[2] In particular, the *only* components of the spin triplet that are visible in the $P_0 \parallel Q$ (or $\alpha \equiv x$) configuration are $|11\rangle$ and $|1\bar{1}\rangle$, and the scattering cross sections are

$$\sigma_x^{+-}(|11\rangle) \propto 4a_1^2\delta(E - \hbar\omega_1), \quad \sigma_x^{-+}(|11\rangle) \approx 0,$$
$$\sigma_x^{+-}(|1\bar{1}\rangle) \approx 0, \quad \sigma_x^{-+}(|1\bar{1}\rangle) \propto 4a_{\bar{1}}^2\delta(E - \hbar\omega_{\bar{1}}). \qquad (1)$$

Alternatively, one can use the *half*-polarized neutron analysis configuration, with, for instance, the incoming beam unpolarized, $I_+ \propto \sigma_x^{0+}(\equiv \sigma_x^{++} + \sigma_x^{-+})$ and $I_- \propto \sigma_x^{0-}(\equiv \sigma_x^{+-} + \sigma_x^{--})$. It is straightforward to realize that a nuclear term, necessarily not spin flip (σ_x^{++} and σ_x^{--}), should contribute both to I_+ and I_-.

The compound $Sr_{14}Cu_{24}O_{41}$. This compound displays a composite crystallographic structure made up of stacking two distinct low-dimensional Cu-O arrangements. The first subsystem is a one-dimensional (1D) lattice of edge-sharing CuO_2 chains, and the second one is a 2D subsystem of two-leg Cu_2O_3 ladders, with the stacking direction being the b axis. The occurrence of a quantum spin-singlet ground state is revealed in INS experiments through the absence of any elastic or quasielastic magnetic scattering and the appearance of a spin gap in the magnetic excitation spectra at sufficiently low temperatures.[5] In $Sr_{14}Cu_{24}O_{41}$the chain subsystem exhibits a gap at about 11 meV,[6,7] whereas the gap amounts to 32.5 meV for the ladder subsystem.[8] At low temperatures the spin excitations originating form the chain sublattice are concentrated within a narrow band of 1 meV above the gap,[6,7] thus implying that the composite bosons are weakly interacting. The spin dimers of the ladder subsystem are more tightly bounded and strongly interact along the quasi-1D c direction. As shown by INS, the dispersion of excitations extends up to 300 meV.[8–10] The study of the spin gap originating at the ladders below the critical temperature by INS constitutes the goal of this Rapid Communication.

Interestingly, $Sr_{14}Cu_{24}O_{41}$is a self-doped compound with six holes per formula unit (0.25 hole per Cu^{2+} ion) and with an unequal distribution of holes between chains and ladders. The total number of holes and their distribution among the two sublattices vary as a function of the substitution of Sr by La or Y and by Ca, respectively. Today, it is understood that, while a charge ordering (CO) appears in the chain sublattice below ≈150 K, a CDW is established in the ladder subsystem below ≈210 K in pure $Sr_{14}Cu_{24}O_{41}$(Refs. 12 and 13). These characteristic temperatures[11] vary from compound to compound, and the exact repartition of holes between the two sublattices is a matter of very active discussion.[12,13] Therefore, this material provides the opportunity not only to study quantum magnetism in low-dimensional compounds but also to investigate how the carriers interact with such a magnetic environment. The answer to this question is relevant to the field of high-T_C cuprates, of which $Sr_{14}Cu_{24}O_{41}$is a very close system.

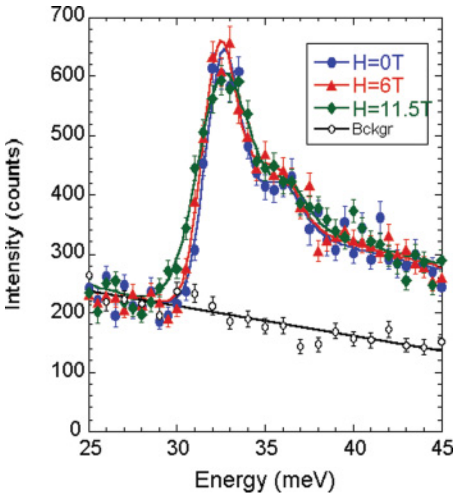

FIG. 1. (Color online) Unpolarized neutron energy scans at $T = 5$ K and constant $Q = (-4.5, 0, Q_L)$, with $Q_L = 0.5$, showing the gap of the spin fluctuations of the ladder subsystem at $E = 32.5$ meV at 0 T (blue circles), 6 T (red triangles), and 11.5 T (green diamonds). The scan at $Q_L = 0.65$ is taken as the background (Bckgr). The solid line is a fit to *ad hoc* functions.

Experiment and results. INS experiments were carried out on three-axis spectrometer (TAS) IN22 at Institut Laue-Langevin (Grenoble, France), and experimental details can

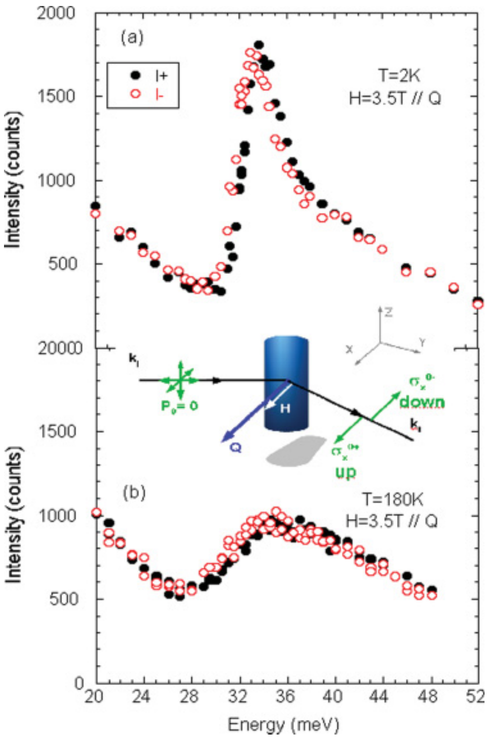

FIG. 2. (Color online) (a) Energy scans taken at $Q = (-4.5, 0, 0.5)$ with *half*-polarized neutrons at $T = 2$ K and under a magnetic field of 3.5 T parallel to Q. Solid and empty circles correspond to two different states of polarization. (b) The same as (a) but at $T = 180$ K.

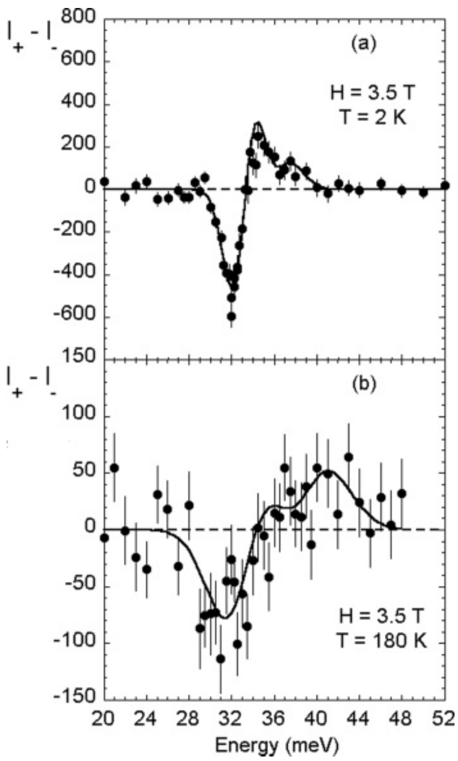

FIG. 3. (a and b) Plots of $I_+ - I_-$ from the data in Figs. 2(a) and 2(b). The solid line is the result of the fit with two triplets under the conditions described in the text.

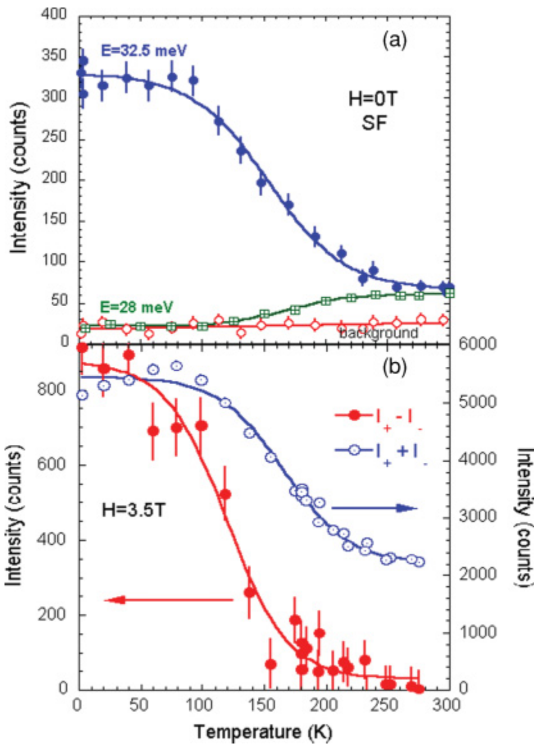

FIG. 4. (Color online) (a) Temperature evolution of the coherent peak intensity at 32.5 meV from polarized-neutron-scattering experiments.[14] The temperature dependence at 28.5 meV (open squares with crosses sign inside) and the temperature dependence of the background (open circles) are also shown. (b) Temperature dependence of $I_+ - I_-$ (solid circles) and $I_+ + I_-$ (open circles).

be found in previous papers.[2,14] Unpolarized INS experiments were carried out with a vertical-field split-pair superconducting magnet, enabling us to reach fields of 11.5 T. Polarized and half-polarized INS experiments were carried out with a horizontal field of up to 3.5 T parallel to Q that uniquely defines the direction of the neutron polarization.

Zero-magnetic-field polarized and unpolarized INS experiments on the ladder excitations in $Sr_{14}Cu_{24}O_{41}$ have revealed the occurrence of a narrow peak (both energy and Q_L unresolved) at 32.5 meV, the so-called resonance or *coherence peak*. The temperature dependence of the intensity scales with the intensity of the CDW that develops below ≈200 K (Ref. 2). Several questions remained unsolved in Ref. 2. One of them was to prove, experimentally, that the sharp resonance at 32.5 meV is really a triplet. Another question relates to the actual temperature where triplets are already coherently formed and the temperature evolution of the resonance peak. Indeed, the thermal contribution of the triplets forming the *coherence peak* can be separated from the remainder of the magnetic excitations, as we expect that excitations progressively broaden as the temperature is increased. Regarding the first question, unpolarized INS data under magnetic field (Fig. 1) show that the resonance-peak width becomes larger and larger as the magnetic field is increased; the full width at half maximum (FWHM) increases from 2.4 meV at $H = 0$ T to 3.8 meV at $H = 11.5$ T, but no clear splitting is observed. Our results in $Sr_{14}Cu_{24}O_{41}$ are in agreement with similar experiments in the high-T_C compound $YBa_2Cu_3O_{6+x}$,[15] but they do not allow us to draw conclusions on the triplet nature of this excitation.

Following the analysis of the polarized neutron cross sections carried out above and the application of a moderate magnetic field along Q, we expect that the *half*-polarized configuration yields information on each one of the two Zeeman split components of the triplet. Figure 2(a) shows an energy scan at $T = 2$ K at the position $Q = (-4.5,0,0.5)$ in the two polarization channels, I_+ and I_-, with a magnetic field of 3.5 T applied parallel to Q. The resonance is sitting on top of a continuum of undefined phonon excitations, as can be seen in the tail at low energies. Each point in energy was counted for both polarization channels so that the spectrometer does not move. In this way, the spectrometer positioning errors are minimized, thus yielding smaller error bars. Typical counting times for each point are of the order of 30 min in order to achieve the required statistics. The difference between both polarization channels is a shift of ±0.45 meV $\approx \pm g\mu_B H$ around 32.5 meV, and that can be seen by the naked eye in Fig. 2(a). This should be compared to the scans taken with unpolarized neutrons at larger magnetic fields (Fig. 1). Figure 2(b) displays the same scan as in Fig. 2(a), but at $T = 180$ K. Despite lower counting rates and larger error bars, a general trend is still seen between I_+ and I_- at such high temperatures and relatively low magnetic field.

Figure 3(a) shows the difference $I_+ - I_-$ where the antisymmetric function around 32.5 meV indicates that the response is a triplet. Some features are worth pointing out. (i) The distance between peaks is now an indication of the

FWHM of the resonance peak, 2.6 meV, which is basically the instrument resolution (this can be qualitatively understood by assuming Gaussian line shapes). (ii) The spectral weight in the minus side [left in Fig. 3(a)] is larger than that of the plus side. This is the consequence of an asymmetric line shape of the resonance peak, due to the dispersion. (iii) A second feature appears at 36 meV, which is smaller in intensity but which can be fitted. We know from previous experiments that this is a second triplet appearing at higher energies that remains barely visible at low temperatures. With this method the occurrence of a second triplet is revealed even at low temperatures. (iv) The nuclear component and the tail of magnetic excitations above 38 meV remain unchanged under an applied magnetic field, and they cancel out in the $I_+ - I_-$ plot. Figure 3(b) displays $I_+ - I_-$ at $T = 180$ K, following the data shown in Fig. 2(b). The antisymmetric function is still visible, the FWHM of the resonance peak is of the order of 6 meV, and the positions of the first and second triplets are shifted to 33.5 and 38.5 meV, respectively.

The temperature dependence of the antisymmetric scattering $(I_+ - I_-)$ has been monitored at 32.25 meV, and it is shown in Fig. 4. At this energy, (i) the difference between the scattered intensity of both polarization channels is the largest [see Figs. 3(a) and 3(b)], and (ii) this intensity, within the simple Gaussian line-shape approximation, is just proportional to $I^{\text{peak}}_{\text{triplet}} H/\text{FWHM}_{\text{triplet}}$. As a comparison, we have plotted in Fig. 4 the total scattering, $I_+ + I_-$, at this energy and the temperature dependence of the peak intensity at 32.5 meV carried out with polarized neutrons.[14] Clearly, some inelastic magnetic excitations persist at temperatures well above 200 K, but the the coherent triplet mode disappears at 210 K, i.e., exactly the temperature at which the CDW disappears,[16–18] which points to a hole-induced resonant peak. The similitude with the resonance feature in the high T_C cuprates is remarkable and more than fortuitous. So far, it remains unclear how this triplet relates to the triplet of excitations expected for the ladder arrangement.[19]

Conclusions. By combining small magnetic fields and longitudinal neutron-polarization analysis we have revealed the chirality of the magnetic excitations in the ladder sublattice of $Sr_{14}Cu_{24}O_{41}$. Under appropriate conditions, the magnetic chirality enters the scattering cross section by enhancing or suppressing the scattering intensity and thus can be used to separate the various magnetic components of the spin triplet. Interestingly, only the Zeeman-split modes, $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, are visible in the $P_0 \parallel Q$ configuration, and each one of them appears in a specific polarization channel, I_+ or I_-. By performing the difference $I_+ - I_-$ at $H \neq 0$, a rather large antisymmetric signal appears that accounts for the triplet of excitations alone. This technique can be used to further understand the magnetic excitations in other unconventional magnetic systems, e.g., the resonant mode in the high-T_C cuprates.

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