Temperature Dependence of the Magnetic Excitations in $La_{1.85}Sr_{0.15}CuO_4$ ($T_c = 33$ K)

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Using neutron-scattering techniques, we have studied the temperature dependence of the magnetic excitations at varied energies in a high-quality single crystal of $La_{1.85}Sr_{0.15}CuO_4$. This crystal exhibits a sharp superconducting transition at 33 K with nearly 100% flux exclusion. For magnetic excitation energies of 9 meV and lower, the intensity drops dramatically with decreasing temperature for temperatures below ~150 K. However, at 12 meV the intensity is independent of temperature between 300 and 5 K.

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It is broadly viewed that magnetism plays an essential role in the superconductivity in the lamellar CuO_2 superconducting materials. Experimentally, one of the most important issues is the nature of the spin excitations in the normal and superconducting states. Over the past two years extensive studies of the magnetism in the CuO_2 materials using neutron-scattering,[1,2] muon-spin-resonance,[3] and nuclear-resonance techniques[4] have been reported. However, a number of basic questions remain unanswered. For example, there is so far very little information on the temperature dependence of the spin excitations at energies comparable to the superconducting gap energy. For such studies the inelastic-neutron-scattering technique plays a unique role. However, such experiments require large single crystals (~1 cm^3 in volume) which are homogeneous and ideally have the gap energy in a convenient energy range (10–20 meV).

In this Letter we report a neutron-scattering study of the magnetic excitations in a large single crystal of $La_{1.85}Sr_{0.15}CuO_4$ which has a sharp superconducting transition at $T_c = 33$ K and at low fields (< 1 G) exhibits nearly 100% flux exclusion. We have observed the spin excitation response function for energies between 3 and 12 meV and for temperatures between 5 and 300 K. For energies above 3 meV the cross section is clearly incommensurate as observed previously.[2,5] For energies below ~12 meV the intensity is ~const between 300 and 150 K, drops precipitously for temperatures down to about 50 K, and is then again ~const down to 5 K. However, at 12 meV the excitation intensity appears to be independent of temperature between 300 and 5 K. This suggests the appearance of a partial or full gap in the spin excitation spectrum well in advance of the superconducting transition.

The single crystal was grown by use of the traveling-solvent-floating-zone method as described in Ref. 6. The sample, which we label KOS-1, was in the form of a cylinder with diameter ~6.5 mm and length 17 mm; the orthorhombic *b* axis (space group *Cmca*) was perpendicular to the cylinder axis. The crystal exhibits a tetragonal-orthorhombic structural transition at $T_0 = 200 \pm 10$ K; from the data compiled in Ref. 7 we conclude that the Sr concentration is 0.15 ± 0.01, consistent with the concentration in the feed material. Further, from the sharpness of the transition we conclude that the Sr and O concentrations are uniform to within ± 0.01 over the entire crystal. Detailed transport and magnetic measurements have been carried out on a thin slice cut from the top of the crystal. The sample shows nearly 100% flux excursion in both field cooling and zero-field cooling at very low fields (0.1 G). Measurements on the entire neutron sample imply a flux exclusion of $80\% \pm 20\%$. Resistivity data within and between the CuO_2 planes are shown in Fig. 1; these are typical for high-quality single crystals of superconducting $La_{2-x}Sr_xCuO_4$. As discussed in Ref. 8, effects of the superconductivity may be seen directly in neutron depolarization measurements.

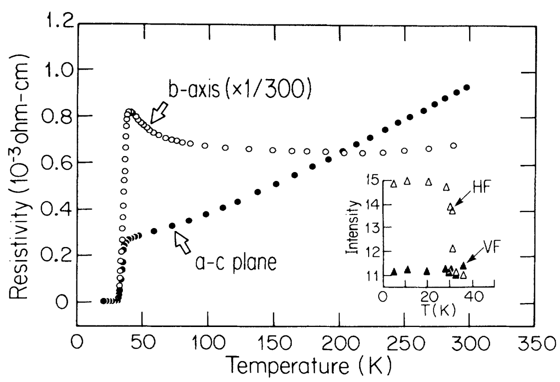

FIG. 1. In-plane and between-plane resistivities in a $La_{1.85}Sr_{0.15}CuO_4$ single crystal cut from KOS-1. Inset: spin-flip neutron intensity with the applied field vertical (▲) and horizontal (△).

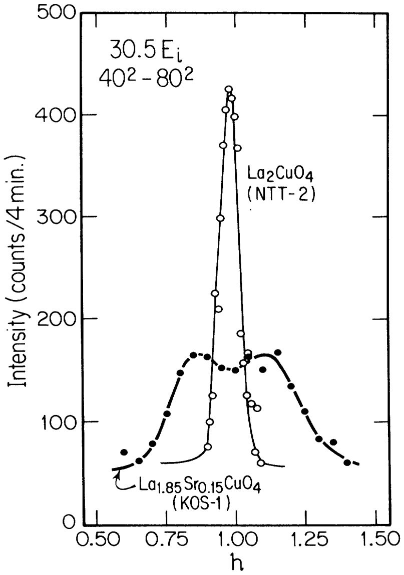

FIG. 2. Inelastic neutron-scattering spectra in La_2CuO_4 (NTT-2) and $La_{1.85}Sr_{0.15}CuO_4$ (KOS-1) at $E=-6$ meV and $T=300$ K. The spectrometer had a fixed incoming neutron energy of 30.5 meV with the collimator sequence 40′-40′-S-80′-80′. The solid lines are guides for the eye.

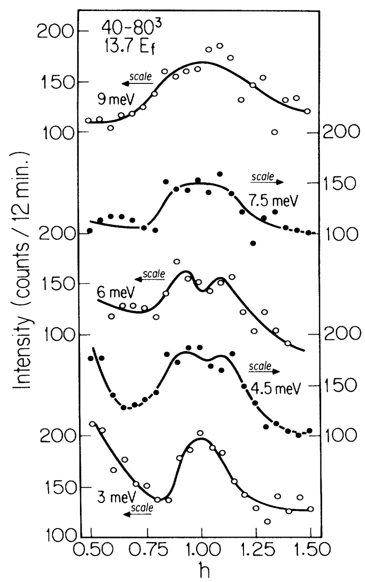

FIG. 3. Inelastic neutron-scattering spectra for varied energies at $T=150$ K in $La_{1.85}Sr_{0.15}CuO_4$ (KOS-1). The spectrometer had a fixed outgoing neutron energy of 13.7 meV with the collimator sequence 40′-80′-S-80′-80′. The solid lines are guides for the eye.

Results for KOS-1 are shown in the inset of Fig. 1. As is evident from the figure both the resistivity data from the thin slice and the neutron depolarization measurements from the entire crystal reveal a sharp superconducting transition at 33 ± 1 K.

The neutron experiments were carried out on the triple-axis spectrometers H-7 and H-4M at the Brookhaven high-flux beam reactor. The sample was mounted with the orthorhombic a axis [tetragonal (110)] vertical. The experimental techniques were identical to those discussed by Thurston *et al.*[2] In general, a variety of triple-axis-spectrometer configurations was used in order to optimize the visibility of the magnetic scattering above the background. We also exploited the fact that the magnetic scattering is two dimensional in character in order to achieve focusing and to minimize contamination from double-scattering and phonon processes. We remark that the inelastic magnetic signal is typically 6 orders of magnitude weaker than the nuclear Bragg scattering so that these experiments are technically quite difficult.

Shown in Fig. 2 are scans at an energy transfer of 6 meV at room temperature in KOS-1 and in a sample of pure La_2CuO_4, labeled NTT-2, which has previously been studied in detail by Endoh *et al.*[1] Two features are immediately evident. First, KOS-1 exhibits a double-peaked incommensurate response as in other metallic and superconducting samples of $La_{2-x}Sr_xCuO_4$.[2,5] Second, allowing for the differences in sample volumes, the integrated intensities are identical to within the errors in the two crystals. That is consistent with previous results which demonstrate that the Cu^{2+} moments are preserved in the superconducting materials.[7]

A series of scans at $T=150$ K at various energies are shown in Fig. 3. As in samples with lower Sr^{2+} concentrations, the integrated intensity of the magnetic scattering depends weakly, at best, on energy.[1,7] In this figure the solid lines are all simply guides for the eye. The response at all energies except possibly 3 meV is flat topped; only with very careful masking and with a clean background is it possible to resolve the two split peaks clearly. Comparison with corresponding data in the sample NTT-35 ($T_c=10$ K) which has $x\simeq0.11$ shows that at high temperatures the dynamical spin response is closely similar in the two samples in spite of the differences in the superconducting transition temperatures.[2] There is, however, a fundamental difference in the temperature dependences. In NTT-35 the intensity of the 6-meV spin scattering is independent of temperature between 300 and 5 K while there is a suggestion of some temperature dependence at 4.5 meV. Representative scans at varied temperatures for $E=6$ and 12 meV in KOS-1 are shown in Fig. 4. It is evident that at 6 meV the intensity decreases significantly between 150 and 12 K. The 12-meV scans exhibit some phonon contamination for $h>1.1$. However, the intensity for $h<1.1$, which arises from magnetic scattering alone, appears to be independent of temperature.

We show in Fig. 5 the integrated intensities as functions of temperature for scans at 6 and 12 meV. The intensity at 12 meV is independent of temperature, albeit with rather large uncertainties. This is analogous to the

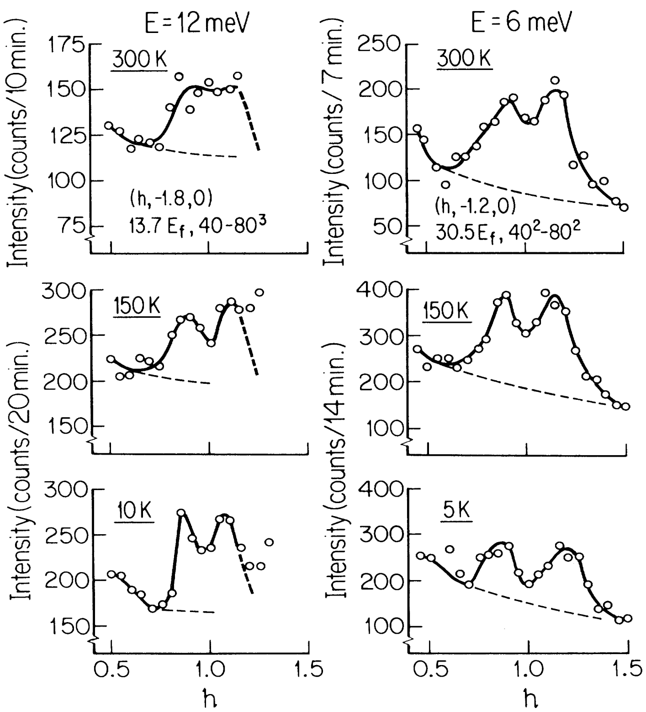

FIG. 4. Inelastic neutron-scattering spectra for $La_{1.85}Sr_{0.15}CuO_4$ (KOS-1) at $E=6$ and 12 meV. For the $E=12$ meV data the spectrometer had a fixed outgoing neutron energy of 13.7 meV with the collimator sequence 40′-80′-S-80′-80′. For the $E=6$ meV data the spectrometer had a fixed outgoing neutron energy of 30.5 meV with the collimator sequence 40′-40′-S-80′-80′. The solid lines are guides for the eye.

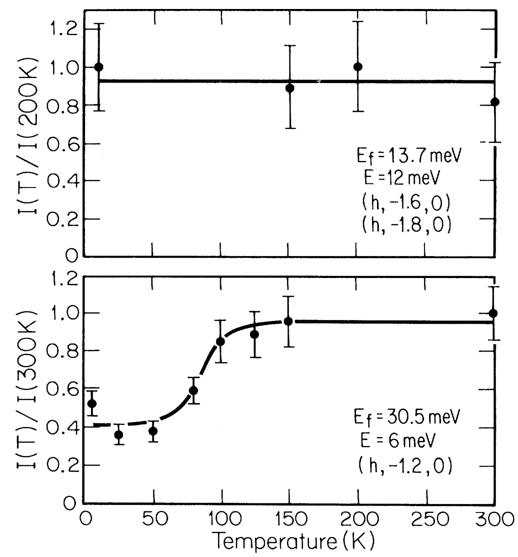

FIG. 5. Normalized integrated intensities of the magnetic response function at energies of 6 and 12 meV. The solid lines are guides for the eye.

behavior at 6 meV in NTT-35 ($T_c=10$ K). However, in KOS-1, as shown in Fig. 5, at 6 meV there is a dramatic diminution in the intensity between ~150 and 50 K. We observe similar behavior at 3 and 9 meV although for the latter, energy contamination from phonon scattering prevents us from making an unambiguous statement.

For $T_c \simeq 10$ K (NTT-35) and $T_c=33$ K (KOS-1) the BCS gaps, $2\Delta=3.5k_BT_c$, are ~3 and 10 meV, respectively. Although weak-coupling BCS theory may well not apply here, the predicted gap energy nevertheless should be of the right order of magnitude. The energy 6 meV is thus above BCS value for the superconducting gap in NTT-35 and below it in KOS-1. It is therefore certainly strongly suggestive that the differences in the temperature dependences of the excitation intensities in the two samples is connected with the energy scales for the superconductivity. The data in KOS-1, in particular, are suggestive of the opening of a gap in the Cu^{2+}-hole spin excitation spectrum with an energy comparable to the BCS gap value. Two features are, however, rather surprising. First, the apparent gap develops well above T_c suggesting that the existence of the gap in the spin excitation spectrum is a prerequisite for the superconductivity. We note that analogous precursor effects are observed in NMR[4] and optical studies[9] of the $YBa_2Cu_3O_{7-x}$ system. Second, there is some residual intensity for the spin fluctuations with energies below the BCS gap value in the superconducting state. There is a variety of possible explanations for this latter effect. These include smearing of the spin gap due to the microscopic structural disorder and microscopic electronic inhomogeneities causing part (say 30%) of the sample to stay in the normal state. The latter would be inconsistent with our flux exclusion measurements on the whole sample. Only experiments on additional materials can resolve this issue.

In summary, these experiments have directly demonstrated the existence of low-lying incommensurate spin excitations in a high-quality single crystal of $La_{1.85}Sr_{0.15}CuO_4$ with $T_c=33$ K and a large Meissner fraction. For energies below the putative BCS gap energy 2Δ, the excitation intensity decreases markedly between 150 and 50 K and is then independent of temperature down to 5 K. Manifestly, this behavior must be predicted by any theory of the superconductivity in the lamellar CuO_2 materials.

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