Magnetic Dynamics of Superconducting $La_{1.86}Sr_{0.14}CuO_4$

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The general magnetic response function $\chi(Q,\omega)$ of a superconducting ($T_c=33$ K) single crystal of $La_{1.86}Sr_{0.14}CuO_4$ is measured using inelastic neutron scattering. There is no gap in the magnetic fluctuation spectrum of the normal state. Superconductivity reduces $\chi''(Q,\omega)$ in progressively greater proportion as $\hbar\omega$ decreases below 6 meV. The superconducting transition coincides with a sharp maximum in the temperature-dependent $\chi''(Q,\omega)$ for $\hbar\omega=4$ meV and Q fixed at the incommensurate wave vector characterizing the magnetic fluctuations in the normal state.

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Nuclear resonance experiments [1] show that in superconductors the magnetic dynamics change dramatically at the onset of superconductivity. However, because such experiments probe only local susceptibilities at very low frequencies, how the general Q- and ω-dependent response function $\chi(Q,\omega)$ actually changes below T_c is not well understood [2,3]. In the present paper, we describe a direct demonstration that superconductivity actually does affect $\chi(Q,\omega)$ for frequencies larger than those employed in nuclear resonance experiments. Specifically, our inelastic neutron scattering measurements on $La_{1.86}Sr_{0.14}CuO_4$ show that for $\hbar\omega \lesssim 6$ meV, superconductivity suppresses the relatively long-ranged incommensurate fluctuations which develop below 100 K. This new effect is especially apparent at momentum transfers Q near which other workers [2] have not collected data.

The fabrication and characteristics of our single-crystal sample of $La_{1.86}Sr_{0.14}CuO_4$ (volume of 0.7 cm^3) are described elsewhere [4]. Most pertinent for this paper is that dc magnetization measurements [5] in an applied field of 20 G on the entire sample indicate a sharp onset of superconductivity at 33 K. We performed inelastic neutron scattering measurements using the TAS VI cold neutron spectrometer of Risø National Laboratory. Throughout this paper, we label reciprocal space, as probed by the instrument, using notation appropriate for the (nearly) square lattices of the CuO_2 planes. Thus, the nuclear reciprocal-lattice points have coordinates $(n2\pi,m2\pi)$, while the new points introduced by the unit-cell doubling associated with simple antiferromagnetism (AFM) are of the form $((2m+1)\pi,(2n+1)\pi)$. Even though long-range AFM order does not exist in $La_{2-x}Sr_xCuO_4$ for modest x, the magnetic fluctuations remain peaked at reciprocal-lattice points of the latter type [2,6]. For larger x ($\gtrsim 0.075$), however, the magnetic fluctuations acquire the incommensurate character [4] illustrated in the lower right-hand corner of Fig. 1. In particular, $\chi''(q,\omega)$ is peaked at the points $(\pi,\pi)\pm\delta(\pi,0)$ and $(\pi,\pi)\pm\delta(0,\pi)$, where $\delta \cong 2x$. Figure 1 shows a collection of constant-$\hbar\omega$ scans at $T=35$ K$=T_c+2$ K. At $\hbar\omega=3.5$ meV, the lowest energy transfer probed, there are two sharp peaks at $(\pi,\pi)-\delta(\pi,0)$ and $(\pi,\pi)+\delta(0,\pi)$, where $\delta=0.245\pm0.004$. As $\hbar\omega$ is increased, the peaks in the scattering become somewhat broader, which implies shorter correlation lengths for higher-energy fluctuations. The peak intensity is roughly independent of $\hbar\omega$, so that the Q-integrated spectral weight must rise with $\hbar\omega$.

Figure 2 shows how representative constant-$\hbar\omega$ scans change with T. The solid and open circles correspond to data taken along the dashed and solid trajectories, respectively, in the inset of Fig. 1. The spectra evolve considerably in the normal state. Specifically, on cooling from 100 to 35 K, scattering at (π,π) is suppressed and clearer incommensurate peaks emerge for the lower $\hbar\omega$ (3.5 meV). Cooling further, through T_c, attenuates the 6-meV spectra by $\sim 15\%$, but does not visibly broaden the incommensurate peaks. For $\hbar\omega=3.5$ meV, cooling from 35 to 20 K has a more dramatic effect: The incommensurate modulations disappear almost entirely.

Figure 3, which shows a constant-Q scan collected at

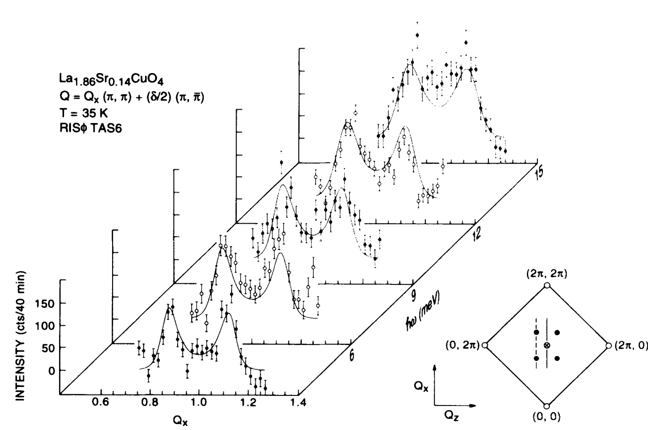

FIG. 1. Magnetic scattering at $T=35$ K$\gtrsim T_c=33$ K. Inset: Reciprocal space with solid circles indicating locations near (π,π) with maximal magnetic response and open circles corresponding to (nuclear) Bragg points for a square lattice. The main figure consists of a series of background-corrected constant-energy scans, collected along reciprocal-space trajectory indicated by the dashed line in the inset. Solid lines are derived from fits described in the text.

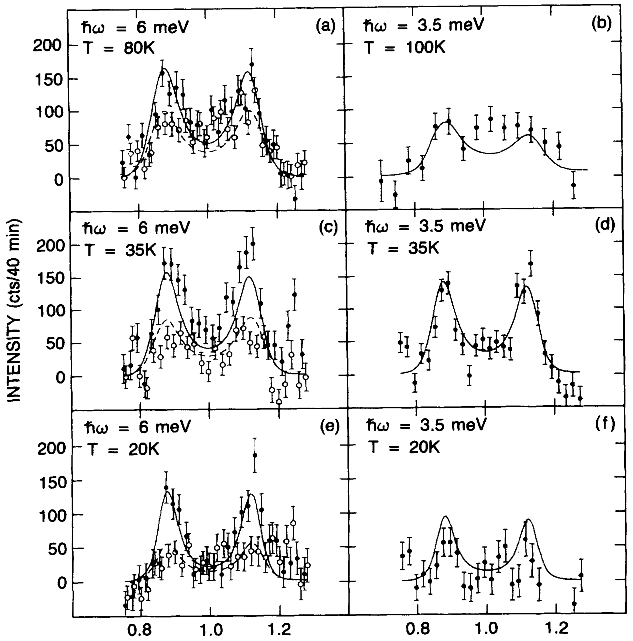

FIG. 2. Temperature dependence of background-corrected constant $\hbar\omega = 6$ and 3.5 meV scans. Solid and open circles are from reciprocal-space trajectories indicated by the dashed and solid lines, respectively, in the inset of Fig. 1. Solid and dashed curves are from fits using Eq. (1) (for $T > T_c$) and Eq. (3) (for $T < T_c$).

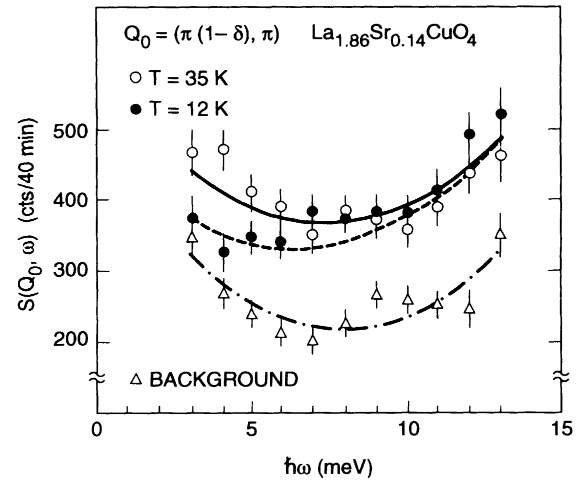

FIG. 3. Constant $Q_0 = (\pi,\pi) - \delta(\pi,0)$ energy scan for $T = 35$ K $\gtrsim T_c = 33$ K and $T = 12$ K $\cong T_c/3$. The data are not corrected for background, identified with signal at $Q = (0.55\pi, 1.15\pi)$, and indicated by open triangles. Solid and dashed curves are from fits using Eqs. (1) and (3), respectively.

$Q = (\pi,\pi) - \delta(\pi,0)$, demonstrates more explicitly how reducing T from 35 K affects the spectrum of magnetic excitations in $La_{1.86}Sr_{0.14}CuO_4$. The data here are raw spectra, and the open triangles represent the background, identified with the signal measured at $Q = (0.55\pi, 1.15\pi)$, a reciprocal-space point far from (π,π). The 12-K data begin to deviate from the 35-K data for $\hbar\omega \cong 6$ meV, and indeed approach the background most closely for the lowest energy transfer, $\hbar\omega = 3$ meV. That this is not due to the decrease of the Bose-Einstein factor, $n(\omega)+1$, is apparent because, for $\hbar\omega = 3$ meV, $n(\omega)+1$ drops by only 33% from $T = 35$ to 12 K.

In Fig. 4(a) we plot the T dependence of the raw scattered intensity (solid circles) for $\hbar\omega = 4$ meV and $Q = Q_0 = (\pi,\pi) - \delta(\pi,0)$. The gradual upward trend of the signal with decreasing T stops abruptly at T_c, below which there is a precipitous decline. That this decline corresponds to a substantial reduction of the magnetic response is obvious from our results for $\chi''(Q_0,\omega)$ (open circles) obtained via the fluctuation-dissipation theorem from the raw data, corrected for the constant background indicated by the dashed line. Figure 4(a) contains two other important qualitative results. First, $\chi''(Q_0,\omega)$ is still appreciable at $T = T_c/3$ for $\hbar\omega = k_BT_c$. This implies, as do also the ω-dependent spectra of Fig. 3, that we are not dealing with an ideal BCS superconductor, where at low T no excitations are expected below the pair condensation energy. The second result is that because $\chi''(Q_0,\omega)$ has a sharp peak rather than merely a break in slope at T_c, superconductivity is not due to the Bose condensation of "preformed" Cooper pairs [7]. The constant-Q data of Fig. 3 also bear out this conclusion: There is no pseudogap in the magnetic excitation spectrum for $T = 35$ K.

To arrive at a more quantitative understanding of the normal-state data, we use the simplest rational function with equivalent extrema at $(\pi,\pi) \pm \delta(\pi,0)$ and $(\pi,\pi) \pm \delta(0,\pi)$ to represent $\chi(Q,\omega)$:

$$\chi(Q,\omega) = \frac{\chi^0 A^2}{i\omega + A^2[\kappa^2 + R(Q)]}, \tag{1}$$

where

$$R(Q) = \frac{[(q_x - q_y)^2 - (\pi\delta)^2]^2 + [(q_x + q_y)^2 - (\pi\delta)^2]^2}{2a_0^2 4\pi^2\delta^2} \tag{2}$$

and $q = (q_x, q_y) = Q - (\pi,\pi)$. Also, $a_0 = 3.8$ Å, the Cu-Cu separation. The inelastic neutron scattering cross section is proportional to $S(Q,\omega) = \chi''(Q,\omega)[n(\omega)+1]$. Forms of the type (1) have been proposed [8] to describe conventional metals close to Fermi surface nesting instabilities. In particular, Noakes *et al.* [9] have shown that Eq. (1) with appropriately chosen $R(Q)$ describes the paramagnetic scattering from the itinerant electron antiferromagnet Cr. For two-dimensional systems such as $La_{1.86}Sr_{0.14}CuO_4$, the local magnetic response, measured at low ω by nuclear resonance spectroscopy, is $\chi_0''(\omega) = \int d^2Q\, \chi''(Q,\omega) \cong \chi^0 4\pi \tan^{-1}(\omega/\omega_0)$ which, if $\hbar\omega_0 = A^2\kappa^2 \sim k_BT$, is recognizable as the form postulated for

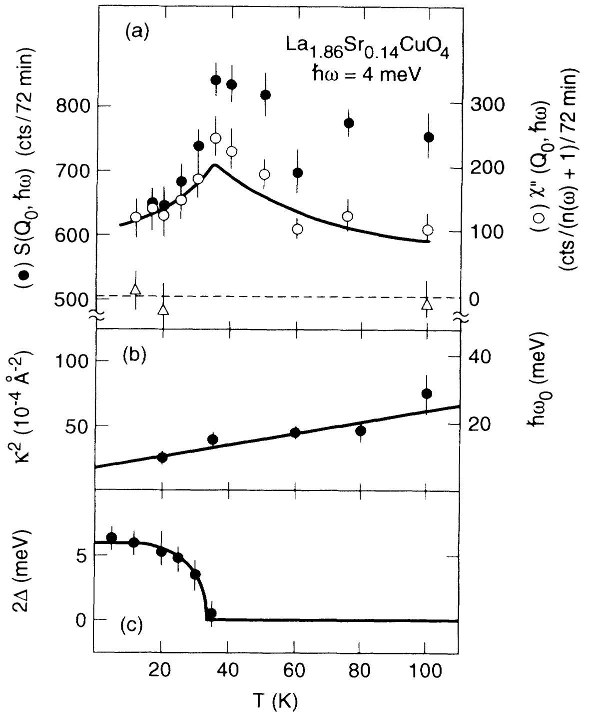

FIG. 4. (a) Temperature dependence of raw intensity for $\hbar\omega=4$ meV, with $Q=Q_0=(\pi,\pi)-\delta(\pi,0)$ (solid circles) and $Q=(0.55\pi,1.15\pi)$ (open triangles). Open circles represent $\chi''(Q_0,\omega)$, obtained by application of fluctuation-dissipation theorem to data indicated by solid circles, where the background is identified with the dashed line through the open triangles. The solid line is computed from Eqs. (1) and (3), assuming fixed amplitude χ^0 and $A^2=3858$ meV Å^2 as well as T-dependent κ^2 and 2Δ given by solid lines in frames (b) and (c). (b) Value of κ^2 and $\hbar\omega_0=A^2\kappa^2$ obtained from fitting Eq. (1) ($T\gtrsim 35$ K) and Eq. (3) ($T\lesssim 35$ K) to data. A^2 and χ^0 were fixed as in (a). (c) Value of 2Δ obtained from fitting Eq. (3) to data. A^2 and χ^0 were fixed as in (a). Except where (35 and 20 K) sufficient data were available to determine it simultaneously, T-dependent κ was fixed at the value given by the solid line in (b). The solid line here corresponds to two-fluid form described in text.

the response of a marginal Fermi liquid [6,10]. Another important feature of Eq. (1) is that, in the extreme quantum limit ($\hbar\omega_0 \gg k_BT$), the equal-time correlation function is

$$S(Q)=\int S(Q,\omega)d\omega=A^2\chi^0\ln(\bar{\omega}/\{A^2[\kappa^2+R(q)]\}),$$

where $\bar{\omega}$ is an upper cutoff frequency. Thus, it is obvious that $S(Q)$ has very weak (i.e., logarithmic) maxima at $(\pi,\pi)\pm\delta(\pi,0)$ and $(\pi,\pi)\pm\delta(0,\pi)$.

To obtain the parameters κ and A^2, we have fitted our data using (1) and (2), corrected for instrumental effects. The solid lines in Fig. 1 correspond to the outcome at 35 K, where $A^2=3858\pm210$ meV Å^2, which is similar to what was found [9] for Cr above its Néel temperature, and $\kappa=0.063\pm0.002$ Å^{-1}. Thus, the incommensurate magnetic fluctuations in $La_{1.86}Sr_{0.14}CuO_4$ are characterized by an $\omega\to 0$ structure factor with an inverse half width at half maximum of $\xi=(0.64\kappa)^{-1}=6.7a_0=25$ Å [the prefactor 0.64 is due to the non-Lorentzian nature of $\chi''(Q,\omega)$ at fixed ω] and $\hbar\omega_0=15$ meV. Since $A^2 \cong \hbar^2/2m$, where m is the mass of a free electron, it appears that quasiparticles with conventional masses are responsible for the incommensurate fluctuations [11]. The length ξ is larger than lengths cited previously [2,4] for $La_{2-x}Sr_xCuO_4$ samples with $x\approx 0.14$ because in many cases the corresponding experiments revealed no incommensurate peaks, and, even where they did [4], they yielded integrals of $\chi''(Q,\omega)$ from $\omega=0$ to cutoffs of the order of 10 meV rather than energy-resolved spectra of the type shown in Fig. 1 of this paper.

On warming, it is very likely that the high-frequency ($\hbar\omega \gg k_BT$) properties defined by A^2 and the scale factor χ^0 remain unchanged. The only parameter which then controls the T dependence of $S(Q,\omega)$ is κ. Our fits to higher T (≤ 100 K) data bear out this hypothesis. Figure 4(b) shows the corresponding results for κ^2. The linear relation $\kappa^2=\kappa_0^2+A_0^{-2}k_BT/\hbar$, with $A_0^2=2000$ meV Å$^2 \cong \frac{1}{2}A^2$ and $\kappa_0=0.043$ Å^{-1}, gives an adequate description of the T dependence of κ^2 shown in Fig. 4(b).

We turn now to the analysis of the data for the superconducting state. In principle, we should compare the data to the appropriate Lindhard function [12] for a superconductor with the Fermi surface of our sample. However, because of the complexity of such a calculation and the probable need to include disorder effects for a random alloy such as $La_{1.86}Sr_{0.14}CuO_4$, we take a more phenomenological approach here. In particular, knowing that $\chi(Q,\omega)$ ultimately originates from electron-hole pairs, which in a clean BCS state exist only for energies in excess of 2Δ (Δ is the gap energy), suggests that the self-energy of the pairs, taken as $i\hbar\Gamma(Q)=i\hbar[\kappa^2+R(Q)]$ in Eqs. (1) and (2), should acquire a real part $\epsilon(Q)\gtrsim 2\Delta$. In a rough sense, $\epsilon(Q)$ represents the energy difference between singlet and triplet states for pairs of carriers with relative momentum Q. Therefore, we replace Eq. (1) by

$$\chi(Q,\omega)=\frac{1}{2}\sum_{\pm}\frac{\chi_0A^2}{i[\omega\pm\epsilon(Q)]+A^2[\kappa^2+R(Q)]}. \qquad (3)$$

We expect $\epsilon(Q)$, like $\Gamma(Q)$, to reach its minimum at the nesting vectors $(\pi,\pi)\pm\delta(0,\pi)$ and $(\pi,\pi)\pm\delta(\pi,0)$, which implies that to lowest order in Q near (π,π), $\epsilon(Q)=2\Delta+A_s^2R(Q)$, where Eq. (2) gives $R(Q)$. The form (3) has obvious shortcomings (notably that its imaginary part never vanishes for $\omega\neq 0$) as a description of what occurs in a good conventional superconductor, but has the virtues of simplicity and analyticity. Using Eq. (3) and assuming that A^2 remains fixed at its normal-state value, we obtain reasonable fits (solid lines in Figs. 2 and 3) to our data. Superconductivity does not seem to arrest the reduction in the inverse length scale κ [see Fig. 4(b)], while the characteristic energy 2Δ rises dramatically [see

Fig. 4(c)] below 35 K to a value depending somewhat on the choice made for A_s^2. We note first that if we neglect the dispersion of the quasiparticles in the superconducting state and set $A_s^2=0$, the fits are generally unsatisfactory. Considerably better results are obtained for $A_s^2=A^2$, in which case $2\Delta(T=12\text{ K})=4.5\pm0.3$ meV. Finally, we have used an ansatz, $A_s^2=2\Delta/\kappa^2$, which reduces the number of fitting parameters and also meets the requirement that Eq. (3), for which $A_s=0$, is recovered as $T\rightarrow T_c$. Among the schemes which we have tried, this provides the best fits to the data, and yields the parameters κ and 2Δ shown in Fig. 4. The two-fluid form, $2\Delta(T)=2\Delta_0[1-(T/T_c)^4]^{1/2}$, with $2\Delta_0=6$ meV and $T_c=33$ K, gives a good description of the T dependence of the pairing energy.

In summary, our neutron scattering measurements on superconducting $La_{1.86}Sr_{0.14}CuO_4$ reveal Cr-like paramagnetic scattering with sharp incommensurate features which are suppressed, but not broadened, below T_c. There is no pseudogap in the normal-state response. The pairing energy deduced from the data in the superconducting state is somewhat lower than $3.5k_BT_c$, but of the same order as the energy characterizing the incommensurate fluctuations.

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