Spin gap and magnetic coherence in a clean high-$T_c$ superconductor

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A notable aspect of high-temperature superconductivity in the copper oxides is the unconventional nature of the underlying paired-electron state. A direct manifestation of the unconventional state is a pairing energy - that is, the energy required to remove one electron from the superconductor - that varies (between zero and a maximum value) as a function of momentum or wavevector \([1,2]\): the pairing energy for conventional superconductors is wavevector-independent \([3,4]\). The wavefunction describing the superconducting state will include not only the pairing of charges, but also of the spins of the paired charges. Each pair is usually in the form of a spin singlet \([5]\), so there will also be a pairing energy associated with transforming the spin singlet into the higher energy spin triplet form without necessarily unbinding the charges. Here we use inelastic neutron scattering to determine the wavevector-dependence of spin pairing in \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\), the simplest high-temperature superconductor. We find that the spin pairing energy (or 'spin gap') is wavevector independent, even though superconductivity significantly alters the wavevector dependence of the spin fluctuations at higher energies.

The experimental technique that we use is inelastic neutron scattering, for which the cross-section is directly proportional to the magnetic excitation spectrum and can be used to probe it as a function of wavevector and energy transfer. In addition, we have selected \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\), the simplest of the high-temperature (high-\(T_c\)) superconductors. The material consists of nearly square \(\text{CuO}_2\) lattices with Cu atoms at the vertices and O atoms on the edges alternating with LaSrO charge reservoir layers. In the absence of Sr doping, the compound is an antiferromagnetic insulator, where the spin on each \(\text{Cu}^{2+}\) ion is antiparallel to those on its four nearest neighbours. Because of the unit cell doubling, magnetic Bragg refections appear at wavevectors such as \((\frac{1}{2},\frac{1}{2})\) (sometimes called \((\pi,\pi)\) in the two-dimensional reciprocal space of the \(\text{CuO}_2\) planes \([6]\)). Doping yields a superconductor without long-range magnetic order but which has low-energy magnetic
excitations peaked at the quartet of wavevectors $Q_\delta = \left( \frac{1}{2}(1 \pm \delta), \frac{1}{2} \right)$ and $\left( \frac{3}{2}, \frac{1}{2}(1 \pm \delta) \right)$, shown in Figure 1a. The recent discovery of nearly identical fluctuations in the high-$T_c$ YBa$_2$Cu$_3$O$_{7-y}$ bilayer materials [1] clearly indicates their relevance to the larger issue of high-$T_c$ superconductivity and validates the continued study of La$_{2-x}$Sr$_x$CuO$_4$ as the cuprate with the least structural and electronic complexity.

The samples are single crystal rods grown in an optical image furnace. The most reliable measure of the quality of bulk superconductors is the specific heat $C$. For our samples, there is a jump of $\Delta C/k_B T_c = 7 \text{ mJ/moleK}^2$ at $T_c = 38.5 \text{ K}$. As $T \to 0$, $C = \gamma_S T$ where $\gamma_S$ is proportional to the electronic density of states at the Fermi level and has the value $\gamma_S < 0.8 \text{ mJ/moleK}^2$. This together with an estimate of 10 $\text{ mJ/moleK}^2$ for the corresponding normal state $\gamma_N$ indicates that the bulk superconducting volume-fraction $1 - \gamma_S/\gamma_N$ of our samples is greater than 0.9. This, as well as the high value of $T_c$ and the narrowness of the transition, is evidence for the very high quality of our large, single crystals. The basic experimental configurations are similar to those employed previously [8,9]. Figure 1a shows the reciprocal space regions probed. A series of scans like those indicated in the figure, performed for a range of energy transfers were used to build up the $Q$-$E$ maps in Figure 1b and c, which show the scattering around the incommensurate peaks in the normal and superconducting states (here $E$ is the energy transfer).

Figure 1b shows that the normal state excitations at 38.5 K are localized near $Q_\delta$ but are entirely delocalized in $E$. In other words the magnetic fluctuations which are favoured are those with a particular spatial period $1/\delta$ corresponding to $Q_\delta$, but no particular temporal period. Cooling below $T_c$ produces a very different image in $Q$-$E$ space. In Figure 1c all low frequency excitations ($E \leq 5 \text{ meV}$) seem to be eliminated and there is an enhancement of the signal above $8 \text{ meV}$ at the incommensurate wavevectors. The signal now has obvious peaks at around $E=11 \text{ meV}$ and $\delta = 0.29 \pm 0.03$ reciprocal lattice units (r.l.u.). We can thus visualize the zero point fluctuations in the superconductor as magnetic density waves undergoing (damped) oscillation with a frequency of 2.75 THz.
In the normal paramagnetic state, the motion of the density waves becomes entirely incoherent.

Figure 2a-2c shows a series of constant-$E$ cuts through the data in Figure 1. These graphs demonstrate that superconductivity induces a complete loss of signal for $E=2$ meV (2a), a significant intensity-preserving sharpening of the incommensurate peaks for $E=8$ meV (2b), and a large enhancement of the peaks for $E=11$ meV (2c). The peak narrowing in (2b) and (2c) corresponds to a spectacular superconductivity-induced rise in the magnetic coherence lengths (defined as the resolution-corrected inverse half-widths at half-maxima obtained as in [10]) from $20.1 \pm 0.9$ Å to $33.5 \pm 2.0$ Å and $25.5 \pm 0.1$ Å to $34.3 \pm 0.8$ Å, respectively.

Figure 3a-c displays constant-$Q$ spectra both away from $Q_\delta$ (Figure 3a and b) and at $Q_\delta$ (Figure 3c). Superconductivity removes the low-$E$ signal below a threshold energy, while it enhances the higher-$E$ signal close to $Q_\delta$. The threshold for $T < T_c$ appears the same for the three wavevectors shown in Figures 3a-c, with the increase in intensity first visible in all cases at 6 meV. To quantify how superconductivity changes the spectra, we fit the data with the convolution of the instrumental resolution (full-width-half-maximum = 2 meV) and

$$S(Q, E) = \frac{1}{1 - \exp(E/k_B T)} \frac{AE' \Gamma}{\Gamma^2 + E^2}$$

where

$$E' = Re \left\{ [(E - \Delta + i\Gamma_s)(E + \Delta + i\Gamma_s)]^{1/2} \right\}$$

and $A$ is the amplitude, $\Delta$ is the spin gap, $\Gamma$ is the inverse lifetime of spin fluctuations with $E \gg \Delta$ (if $\Delta \ll \Gamma$), $E'$ is an odd function of $E$ which defines the degree to which the spectrum has a gap and $\Gamma_s$ is the inverse lifetime of the fluctuations at the gap edge.

In the normal state, the best fits are obtained for $\Delta = 0$ meV, and the fitted value of $\Gamma$ is essentially $Q$-independent, (Figure 3d). Thus, the lower-amplitude fluctuations with wavevectors different from $Q_\delta$ have lifetimes similar to those at the incommensurate peak.
positions. The \( Q \)-dependence of the signal is entirely accounted for by the \( Q \)-dependence of the real part \( \chi'(Q) \) of the magnetic susceptibility (Figure 3e) which, when \( \Delta = 0 \), is simply the amplitude \( A \). In the superconducting state, \( \Gamma \) (Figure 3d), which characterizes the shape of the spectrum well above the spin gap, becomes strongly \( Q \)-dependent. At the same time, \( \chi'(Q) \) (Figure 3e), related via a Kramers-Kronig relation to the parameters in Equation (1), is suppressed. This explicitly demonstrates that superconductivity reduces the tendency towards static incommensurate magnetic order in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \).

Figure 4 shows the \( Q \) dependence of the spin gap \( \Delta \). As anticipated from inspection of the data in Figure 3, \( \Delta \) is \( Q \)-independent and has the value 6.7 meV. The gap is quite sharp for our sample, with \( \Gamma_s \leq 0.2 \) meV for all \( Q \). Also shown in Figure 4 are the results for \( x=0.15 \) [11,12] and 0.14 [10]. \( \Delta(Q) \) is indistinguishable for the present \( x=0.163 \) and the older \( x=0.14 \) samples; the difference in the low-\( E \) behavior is primarily due to the much larger damping (\( \Gamma_s = 1.2 \) meV for \( x=0.14 \) [13]). In addition, the \( Q \)-independence of \( \Delta(Q) \) is consistent with the \( Q \)-independent but incomplete suppression of the magnetic fluctuations in the \( x=0.14 \) sample [13]. In contrast, the results of [11] and [12] show a large discrepancy with \( x=0.163 \), where the spin gap quoted in these papers is defined as the threshold for visible scattering. Nevertheless the results of [11] are consistent with our work if we use the definition - advocated here and in [13] - of \( \Delta \) given by Equation (2). Fitting the data of [11] to Equation (1) with \( \Delta= 6.7 \) meV yields \( \Gamma_s = 0.5 \) meV, a value intermediate between our findings of 1.2 and 0.1 meV for \( x=0.14 \) and 0.163.

Our experiments show that superconductivity produces strongly momentum-dependent changes in the magnetic excitations with energies above a momentum-independent spin gap. The data in their entirety do not resemble the predictions [15–20] for any superconductors, be they \( s \)-wave or \( d \)-wave. Most notably, all \( d \)-wave theories anticipate dispersion in the spin gap which would have been observed over the wavevector range and for the energy resolution of the present experiment. At the same time, \( s \)-wave theory cannot account for the value of the spin gap. We are unaware of calculations which
yield the dramatic incommensurate peak sharpening and enhancements seen above the spin gap, while at the same time showing a large reduction in the real part of the magnetic susceptibility.

There are other difficulties with the conventional weak-coupling $d$-wave approach which posits nodes and therefore a smaller relative superconductivity-induced reduction in scattering between rather than at the incommensurate peaks. Figure 2b shows the opposite - just above the gap energy, the incommensurate peak intensities are preserved while the scattering between the peaks is suppressed. Furthermore, the peak sharpening in momentum space for $\hbar \omega > \Delta$ finds a precedence only in quantum systems, such as $S=1$ antiferromagnetic (Haldane) spin chains and rotons in superfluid helium, which have well-defined gaps with non-zero minima. Thus, while our statistics and resolution cannot exclude a small population of spin-carrying subgap quasiparticles, the systematics of the signal found near the gap energy make such quasiparticles improbable. As for any other spectroscopic experiment, we can only place an upper bound on the signal below the dispersionless gap. Inspection of Figure 3 shows that in between the incommensurate peaks at $Q = \left( \frac{1}{2}(1 + \delta), \frac{1}{2}(1 - \delta) \right)$, where ordinary weak-coupling $d$-wave theories generally anticipate nodes in the spin gap, the intensity for 2 meV at 5 K is less than 14 % of what was seen at $T_c$ and below 5 % of that observed for the incommensurate peaks at 5 K.

Given the overwhelming evidence for $d$-wave superconductivity in the hole-doped high-$T_c$ superconductors [1][2][21][22], we see our data not as evidence against $d$-wave superconductivity but as proof that the spin excitations in the superconducting state do not parallel the charge excitations in the fashion assumed for ordinary $d$- and $s$-wave superconductors. Our measurements, which are sensitive exclusively to the spin sector, taken together with the evidence for $d$-wave superconductivity in the charge sector suggest that the high-$T_c$ superconductors are actually Luther-Emery liquids, namely materials with gapped (triplet) spin excitations and gapless spin zero charge excitations [23][24]. Luther-Emery liquids arise in one-dimensional interacting Fermi systems, which formally resemble
two-dimensional \( d \)-wave superconductors - the dimensionality (zero) of the nodal points where the gap vanishes in the two-dimensional copper oxide is the same as that of the Fermi surface of a one-dimensional metal. There are other arguments for the applicability of the concept of Luther-Emery liquids. The first is that theory indicates that such liquids are the ground states of ladder compounds, one-dimensional strips of finite width cut from CuO\(_2\) planes \[25\,28\]. The second involves the break-down of spin-charge separation when the spin gap collapses to zero, which can be brought about by a magnetic field whose Zeeman energy matches the spin gap energy. The 6.7 meV spin gap which we measure is much closer to the Zeeman energy of the upper critical field measured \[28\] for samples similar to ours than to an ordinary Bardeen-Cooper-Schrieffer pairing energy \( \geq 3.5k_B T_c = 11.6 \) meV.

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FIG. 1. The reciprocal space regions over which measurements were made and the resulting data as a function of wavevector and energy transfer. a is a reciprocal space diagram of the CuO$_2$ planes in La$_{2-x}$Sr$_x$CuO$_4$ where the black circles represent the incommensurate peaks surrounding $(\pi,\pi)$. The coloured strips give the data collected at an energy transfer of 9 meV and temperature of 5 K and show the areas probed in this experiment. The lower strip passes through two of the peaks and provides the signal, whereas the upper strip lies far from the peaks and is used as the background. b, is a $Q$-$E$ map measured at the transition temperature of 38.5 K, for the wavevectors shown in a and energy transfers from 2 to 16 meV, the parameter $h$ defines the two-dimensional wavevector $Q=[0.56h,0.44h]$. The data displayed are background subtracted and the thermal population factor $(\exp(-E/k_BT)-1)^{-1}$ has been divided out to give the quantity $S(Q,\omega)$. The colouring of the squares indicates the intensity observed in units of counts per 15 minutes. c, A similar map to b but for the superconducting phase at 5 K. The sample used for these measurements consisted of five crystals grown by the travelling solvent floating zone method; each was approximately 4 mm in diameter and 20 mm long. In order to maximise signal the crystals were mounted on a single holder so that their axes were parallel to within 0.8 degrees. The measurements were performed on the new RITA spectrometer at Risø National Laboratory [30]. RITA differs from its predecessor, TAS6, by use of a velocity selector for better filtering of the incident beam, superior beam optics between the reactor and the sample and a large position sensitive detector. In this experiment pixels at different vertical heights on the detector were binned separately so that a single instrumental scan produced a two-dimensional plot in reciprocal space as seen in a.
FIG. 2. Constant-$E$ scans at various energies through the incommensurate peaks at $T_c$ and in the superconducting phase at 5 K. a, The data for 2 meV well below the spin gap energy of 6.7 meV. The incommensurate peaks are clearly visible at $T_c$ but are completely suppressed in the superconducting phase. b, scattering at 8 meV, somewhat above the gap energy. The peak amplitudes are approximately equal at the two temperatures while their lineshapes are different with scattering at 5 K narrower than at 38.5 K. c The scattering at 11 meV well above the spin gap. The scattering amplitude at 5 K is significantly greater than at 38.5 K. The solid lines through the data were obtained by following the analysis procedures of [10].

FIG. 3. Spectra at various wavevectors, and the $Q$-dependence of the inverse lifetime and susceptibility extracted by fitting such profiles. (The wavevectors range from between the incommensurate peaks up to the peak maxima). Panel a shows the constant-$Q$ spectrum at $h=1.000$ (a wavevector exactly in between the two peaks), b shows the spectrum at $h=1.095$ (a position on the inner slope of the peak) and c gives the spectrum at $h=1.135$ (the peak maximum). The spin gap is present at all three wavevectors in the superconducting phase with the scattering eliminated below 6 meV. As $h$ is changed from 1.000 to 1.135 there is a clear maximum in the response at energies around 11 meV in the superconducting phase. d and e show the values of the energy scale $\Gamma$ and the real part of the magnetic susceptibility $\chi'$ (in units of counts per 15 minutes) as functions of wavevector for both 38.5 K and 5 K. These quantities were extracted by fitting to the data the lineshape given in Equation (1) convolved with the instrumental resolution. $\Gamma$ is approximately constant at $T_c$ but strongly $Q$-dependent at 5 K and $\chi'$ is smaller at 5 K than at 38.5 K although it has a similar wavevector-dependent profile at the two temperatures.
FIG. 4. The wavevector dependence of the spin gap in the superconducting state at 5 K. The gap was extracted by fitting Equation (1), convolved with the instrumental resolution, to constant-$Q$ spectra such as those shown in Figure 3a-c. The gap values are given by the open circles and are independent of wavevector. The solid line gives the fitted wavevector-independent value of the gap which is $\Delta=6.7$ meV. The solid symbols show the spin gaps determined previously for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. The filled square gives the gap for an $x=0.14$ sample, the gap was extracted by the same method as used in this paper and found to be of a similar size. The filled triangle and filled diamond give the gap for two different $x=0.15$ samples; in both cases the gap was defined as the threshold for visible scattering without considering resolution broadening or damping at the gap edge. Using this definition the gap was found to have a much smaller value. In all except the present work, the spin gap was established at only a single wavevector.
Intensity ((counts-background)/15 mins)

- T=38.5K
- T=5K

2 meV
8 meV
11 meV

h in Q=[0.56h,0.44h]
present data, $x=0.163$

Mason, T.E. et al., ref. [13], $x=0.14$

Petit, S. et al., ref. [12], $x=0.15$

Yamada, K. et al., ref. [11], $x=0.15$

spin gap

$\Delta$ (meV)

$h$ in $Q=[0.56h,0.44h]$