Unusual interplay between copper-spin and vortex dynamics in slightly overdoped La$_{1.83}$Sr$_{0.17}$CuO$_4$

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Abstract. – Our inelastic neutron scattering experiments of the spin excitations in the slightly overdoped La$_{1.83}$Sr$_{0.17}$CuO$_4$ compound show that, under the application of a magnetic field of 5 Tesla, the low-temperature susceptibility undergoes a weight redistribution centered at the spin-gap energy. Furthermore, by comparing the temperature dependence of the neutron data with ac-susceptibility and magnetization measurements, we conclude that the filling in of the spin gap tracks the irreversibility/melting temperature rather than $T_{c2}$, which indicates an unusual interplay between the magnetic vortices and the spin excitations even in the slightly overdoped regime of high-temperature superconductors.

Introduction. – Experiments on the high-temperature superconductors (HTSC) have produced a tremendously rich variety of physical behaviours, both from the mesoscopic (physics of vortices [1,2,3]) as well as from the microscopic (fundamental electronic [4] and magnetic excitations [5]) point of views.

From the mesoscopic point of view, the cuprates are type-II superconductors with strong anisotropy giving rise to a complicated magnetic phase diagram [1]: below the lower critical field $H_{c1}$, the magnetic flux is entirely excluded from the material (Meissner phase); above $H_{c1}$ quantized magnetic vortices can penetrate the superconductor (mixed phase) to form an ordered vortex lattice that eventually melts at higher fields and temperatures, giving rise to a vortex fluid phase; finally above the upper critical field $H_{c2}$ the normal state is recovered. In La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) the first order melting transition is almost concomitant to the irreversibility line $H_{irr}$, where reversible magnetization and resistivity appear [6,7]. Surprisingly, very little information exists about the microscopic observation of vortex lattice in LSCO, a compound belonging to the family of the first HTSC that has been discovered. Using small angle neutron scattering (SANS), we have recently reported the first direct evidence of a well ordered vortex lattice in slightly overdoped LSCO (x=0.17), which furthermore undergoes a field-induced hexagonal to square transition at around 0.4 Tesla [8,9].
From the microscopic point of view, the LSCO compounds are characterized by the presence of spin excitations located in the vicinity of the antiferromagnetic (AF) wavevector of the undoped parent compound at the \((\frac{1}{2} \pm \delta, \frac{1}{2})\) and \((\frac{1}{2}, \frac{1}{2} \pm \delta)\) incommensurate positions \([9]\). While such excitations could originate from Fermi surface nesting properties due to coherence effects in the superconducting state \([10, 11]\), it has also been proposed that they could be the signature of either dynamical stripes \([12]\) or SO(5) supersymmetry \([13]\). Since these various models differ in the way the AF state of the undoped compound is related to the superconducting state, the application of a magnetic field provides a clean way to differentiate between them. While conventional vortices emerge from Fermi-liquid like models, the SO(5) model predicts unusual vortices with bound AF-states \([14]\). To our knowledge, no predictions exist for the stripes-model in a magnetic field.

Recently, Lake et al. \([15]\) have reported the magnetic field dependence of the incommensurate spin excitations in optimally doped LSCO. On one side they observe, below \(T=10\) K, the appearance of sub-gap excitations when a field of 7.5 Tesla is applied parallel to the c-axis. On the other side it is shown that the filling in of the gap with increasing temperature seems to track the irreversibility/melting line rather than \(T_{c2}\). Both observations are highly unusual and it is crucial to understand whether or not they share a common origin. In order to answer this question, we have performed field dependent measurements of the incommensurate excitations on a slightly overdoped LSCO crystal.

**Experimental results.** Our experiments were performed on the same high-quality single crystal as the one used for the vortex lattice measurements \([8]\). Details of the sample growth can be found elsewhere \([16]\). The neutron data were taken on the IN22 spectrometer at the high-flux reactor of the Institute Laue Langevin in Grenoble, France. We used a graphite vertically curved monochromator and graphite horizontally curved analyser with a fixed final energy of 13.8 meV. In order to avoid contamination by higher-order reflections a PG-filter in \(k_f\) was installed. The sample, a LSCO single crystal \((x=0.17, T_{c}=37\) K, \(\Delta T_{c}=1.5\) K, \(m=2727\) mg), was mounted in a cryostat with the c-axis oriented perpendicular to the scattering plane. A magnetic field up to 5 Tesla could be applied parallel to the c-axis. The Q-scans were performed by rotating the sample around its c-axis. Within such a scan, one can measure two different incommensurate peaks without changing the analyser and detector positions.

The magnetic cross section for inelastic neutron scattering is given by

\[
\frac{d^2\sigma}{d\Omega d\omega} \sim \frac{k_f}{k_i} f^2(Q) \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) S^{\alpha\beta}(Q, \omega)
\]

where \(f(Q)\) is the magnetic form factor, \(S^{\alpha\beta}(Q, \omega)\) is the dynamic structure factor, \(\hbar\omega = E_i - E_f\) and \(Q = k_i - k_f\) are the neutron energy and momentum transfers. The imaginary part of the generalized susceptibility is related to the measured dynamic structure factor via the fluctuation-dissipation theorem,

\[
\chi''(Q, \omega) = S(Q, \omega)(1 - e^{-\frac{\hbar\omega}{k_B T}})
\]

We now start with the description of the data obtained at zero-field. Fig.\(\PageIndex{1}\) shows Q-scans through the incommensurate peaks at an energy transfer \(\hbar\omega=4\) meV for temperatures above and below \(T_c\). Fig.\(\PageIndex{2}\) shows a similar scan at \(\hbar\omega=11\) meV and \(T=5\) K. Above \(T_c\) we observe clear peaks at the expected incommensurate positions \(Q_\delta = (\frac{1}{2}, \frac{1}{2} + \delta)\) and \((\frac{1}{2} + \delta, \frac{1}{2})\) with \(\delta=0.13\) \([17]\). Below \(T_c\) we observe that the magnetic intensity at low energy disappears, which is a clear sign for the opening of a spin gap in the superconducting state.
R. Gilardi, A. Hiess, N. Momono, M. Oda, M. Ido and J. Mesot: Unusual interplay between copper-spin and vortex dynamics in slightly overdoped La$_{1.83}$Sr$_{0.17}$CuO$_{4.3}$.

Fig. 1 – Constant energy-scans through two incommensurate peaks at zero field: a) for an energy transfer $\hbar \omega = 4$ meV, $T = 5$ K and 40 K, and b) for $\hbar \omega = 11$ meV, $T = 5$ K.

Fig. 2 – a) Energy dependence of $\chi''(Q_\delta, \omega)$ in zero-field at $T = 5$ K and 40 K. A background measured away from $Q_\delta$ has been subtracted from the raw neutron data. While at high temperature the intensity is roughly constant, at low temperature a gap is present. b) Difference of the high- and low-temperature susceptibility $\chi''(Q_\delta, \omega)$ (shown in Fig. 2a) measured at $H = 0$ T, and of the $H = 5$ T and zero-field susceptibility $\chi''(Q_\delta, \omega)$ (shown in Fig. 3b) and Fig. 2a) measured at $T = 5$ K. Similarly to the temperature effect, the application of a magnetic field induces a transfer of weight from high to low-energies. The field-induced increase of $\chi''$ at 4 meV, obtained independently from the $Q$-scan (see text), has also been included (filled circle).
The presence of the gap is further confirmed by the energy scans performed at \( Q_\delta \), see Fig. 2. For \( T = 5K < T_c \), \( \chi''(Q_\delta, \omega) \) drops sharply below about 6.5 meV, which we identify with the spin gap energy \( \Delta_{SG} \), whereas for \( T = 40K > T_c \), \( \chi''(Q_\delta, \omega) \) doesn’t vary much as a function of energy.

We now turn to the magnetic field dependence of the spin excitations. Both \( Q \)- and energy scans are not strongly affected by the application of an external magnetic field of 5 Tesla applied perpendicular to the CuO\(_2\) planes, as shown in Fig. 3. In particular, in our slightly overdoped sample, there is no clear indication of field-induced sub-gap excitations at low temperatures. However one can notice that in the \( Q \)-scan performed at \( H=5 \) T (\( T=5 \) K, Fig. 3a) there is slightly more intensity than in the \( Q \)-scan performed at \( H=0 \) T (\( T=5 \) K, Fig. 1a). By taking the zero-field data at \( T=5 \) K as background and fitting two Gaussians with fixed positions (\( H=\pm \delta \)) and widths, we estimate that the value of \( \chi''(H=5 \) T, \( T=5 \) K) is about 30\%\( \pm \)15\% of \( \chi''(H=0 \) T, \( T=40 \) K). This value is in agreement with \( \chi''(Q_\delta, \omega) \) obtained from energy scan, and is included in Fig. 2b and Fig. 3b (filled circle). In a first step we have analyzed the energy dependence of the susceptibility using the phenomenological function introduced by Lee et al. \[18\]. At low temperature and zero-field, we obtain a spin gap value of \( \Delta_{SG} = 6.5 \pm 0.4 \) meV consistent with earlier results \[18,19\]. At 5 Tesla we observe a decrease of \( \Delta_{SG} \) of the order of 25\% (\( \pm20\%) \). However, due to the large error on the determination of the spin gap value, no definitive conclusion can be reached.

It is very instructive to look on one side at the difference of the high- and low-temperature susceptibility \( \chi''(Q_\delta, \omega) \) in zero-field, and on the other side at the difference between the 5 Tesla and zero-field susceptibility \( \chi''(Q_\delta, \omega) \) at \( T=5 \) K (see Fig. 2b). In both cases we observe a weight transfer from the high-energy to the low-energy region, most likely indicating a conservation of spin as reported earlier \[20\] and also expected in more recent theoretical studies \[21\]. The low-energy field-induced weight transfer at 5 Tesla is about 35\% (\( \pm15\% \))

Fig. 3 – a) Constant energy-scans through two incommensurate peaks in a field of \( H=5 \) T for \( \hbar \omega=4 \) meV, \( T=5 \) K and 40 K. b) Energy dependence of \( \chi''(Q_\delta, \omega) \) at \( T=5 \) K and \( H=5 \) T. The filled circle indicates the value of \( \chi'' \) at 4 meV obtained independently from the \( Q \)-scan measured at \( H=5 \) T, see text.
Unusual interplay between copper-spin and vortex dynamics in slightly overdoped La$_{1.83}$Sr$_{0.17}$CuO$_{4.5}$

Fig. 4 – a) Temperature dependence of the neutron intensity for an energy transfer $\hbar \omega = 2.5$ meV taken in zero-field and in a field of $H=5$ T (measured heating up after field-cooling). b) Irreversibility and superconducting transition lines $H_{irr}$ and $H_{c2}$, respectively. Solid lines are guides to the eye, whereas dashed lines indicate the values of $T_{c2}$ and $T_{irr}$ at $H=0$ T and $H=5$ T. The opening of the spin gap tracks $T_{irr}$ rather than $T_{c2}$ (see text).

Fig. 5 – a) Real and b) imaginary part of the ac-susceptibility $\chi = \chi' + i\chi''$ for $H = H_{ac} \cos(\omega_{ac}t) + H_{dc}$ applied parallel to the c-axis with $H_{ac}=5$ T, $H_{dc}=10$ Oe and $\omega_{ac}=10$ Hz. $T_{irr}$ is given by the peak in $\chi''$ (or maximal slope of $\chi'$). c) Field-cooled magnetization in a dc-field of 5 T applied parallel to the c-axis. $T_{c2}$ is determined as explained in the text (extrapolation method). Notice that even in $M(T)$ there is a sign of the irreversibility line (dip on the right side of the small peak).

of the temperature-induced weight transfer measured between $T=5$ K and $T=40$K through $T_c$. This result is very surprising if one considers that the applied field represents only 10% of $H_{c2}(5K) \sim 50$ Tesla (as determined from resistivity data [22]).

Finally, we present the temperature dependence of the spin fluctuations as a function of magnetic field. Fig. 4a shows the neutron counts (without background subtraction) as a function of temperature at $Q_\delta$ and at an energy transfer of 2.5 meV. The background was found to be dependent on the energy transfer, but independent on temperature ($T < 40$ K) and magnetic field ($H < 5$ T). The sharp decrease of the intensity by decreasing temperature indicates the opening of the spin gap. At zero field the intensity drops around $T_c$, whereas at 5 Tesla the point at which the gap opens is shifted by about 15 K toward lower temperature. Similar results have been obtained for an energy transfer of 4 meV. This indicates an unusually strong effect of the magnetic field on the temperature dependence of the spin fluctuations.

In order to understand this shift we performed ac-susceptibility ($\chi = \chi' + i\chi''$) and magnetization ($M$) measurements on a portion of the crystal used for the neutron experiments [1].

[1] These measurements have been performed with a PPMS at the Paul Scherrer Institute, Switzerland. The whole data will be published elsewhere.
The experimental irreversibility lines are obtained from the loss peak of the imaginary part $\chi''$ of the ac-susceptibility [23], which is directly related to the maximum slope of its real part $\chi'$. In Fig.5a+b we show the ac-susceptibility data for a dc-field of 5 Tesla applied parallel to the c-axis. The small width of the loss peak indicates the high quality of our single crystal. The $H_{c2}(T)$ line is more difficult to measure, since it is only a crossover rather than a true phase transition. Magnetization measurements have been used extensively to determine the upper critical field. The simplest approach is to use the linear extrapolation method based on the linear Abrikosov formula for magnetizations at high fields near $H_{c2}$ [24]. The transition temperature $T_{c2}$ is derived from the intersection of the linear fit with the normal-state horizontal line, as shown in Fig.5c. It was argued that this procedure is not totally correct for HTSC, where the Abrikosov linear region is limited to a small temperature range, because of the rounding of the $M(T)$ curves close to $T_{c2}$ (due to either diamagnetic fluctuations or sample inhomogeneities) [25, 26]. However, in our case, the $H_{c2}(T)$ line determined by extrapolation is in quite good agreement with the line obtained by scaling procedures [26]. By applying dc magnetic fields up to 8 T parallel to the c-axis we determined $T_{irr}$ and $T_{c2}$ and obtained the magnetic phase diagram shown in Fig.4b. Notice that between zero and 5 Tesla, $T_{c2}$ changes by only 4 K. Therefore, one would expect that the application of such a magnetic field would only marginally affect the temperature at which the spin gap opens. As can be seen in Fig.5b, this is clearly not the case since in a field of 5 Tesla the point at which the gap starts to fill in is shifted toward a much lower temperature than the measured $T_{c2}$, and agrees well with the measured $T_{irr}$. Interestingly, in our SANS experiments [7], we observed that the vortex lattice intensity vanishes exactly at the irreversibility line, whereas no indication of vortices can be observed above $T_{irr}$.

The suppression of the spin gap in the vortex fluid phase above $T_{irr}$ can be understood qualitatively by assuming that in this region of the phase diagram, the time scale characterizing the dynamics of the vortices becomes comparable to that of the Cu-spin excitations [15]. Such a coupling between the electronic and vortex-fluid degrees of freedom has been inferred earlier, on one hand from Hall measurements where a sharp change of the conductivity is observed at the melting transition [27], and on the other hand from the observation of an unusually large increase of the specific heat when passing from the solid- to the fluid-vortex state [28]. All these experiments, including our own, point toward a more complex nature of the vortex fluid state than anticipated so far. It remains a theoretical challenge to understand, at a quantitative level, the connection existing between these anomalous observations.

Conclusions. – In conclusion, although our measurements of the spin excitations in a slightly overdoped LSCO do not reveal clear evidence for magnetic field induced sub-gap excitations, we do observe an unusually large spectral weight redistribution centered at the spin gap energy when a field of 5 Tesla is applied. It is interesting to put our results in perspective with those obtained by Lake et al. [15,29]: in the underdoped regime the magnetic field induces static ($\hbar\omega = 0$) moments [29], while at optimal doping excitations at finite energy but within the spin gap ($0 < \hbar\omega < \Delta_{SG}$) are created [15]. Our own results show that upon further doping, weight transfer centered at the the spin gap energy ($\hbar\omega \sim \Delta_{SG}$) occurs. This suggests the existence, beside the superconducting and spin gap energies, of an additional doping dependent magnetic energy scale.

Finally, by comparing the temperature dependence of the neutron data with macroscopic measurements, we conclude that the filling in of the spin gap tracks the irreversibility/melting temperature, rather than $T_{c2}$. This indicates that even in the overdoped regime of HTSC there exists an unusual coupling between the copper-spin and vortex degrees of freedom.
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