In-plane anisotropy of spin excitations in the normal and superconducting states of underdoped YBa$_2$Cu$_3$O$_{6+x}$

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A detailed inelastic neutron scattering study of the in-plane anisotropy of magnetic excitations in twinned YBa$_2$Cu$_3$O$_{6+x}$ ($T_c = 61$ K) reveals that the spin excitation spectra in the superconducting and normal all states are qualitatively different. Below $T_c$, the spectrum consists of upward- and downward-dispersing branches with in-plane anisotropy merging at an energy $E_{res} = 37.5$ meV. In the normal state, the singularity at $E_{res}$ disappears, and the spectrum exhibits a steep dispersion with a strongly anisotropic in-plane geometry. These data have important implications for models based on static or dynamical stripe order of spins and charges.

A variety of different states with unusual spin, charge, or current correlations have been invoked to explain the anomalous normal-state (NS) properties of underdoped copper-oxide superconductors. Prominent examples are "striped" states with spin and charge order extending along one of the principal axes of the CuO$_2$ square lattice. Neutron scattering experiments have detailed information about the microscopic magnetic order and dynamical properties and can thus serve as particularly incisive tests of microscopic models of the copper oxides. Recent neutron scattering work on La$_2$CuO$_4$ and YBa$_2$Cu$_3$O$_{6+x}$ has uncovered tantalizing evidence of a "universal" spin excitation spectrum independent of materials-specific details. The dispersion surface of the spin excitations contains upward- and downward-dispersing excitation branches merging at the wave vector $Q_{AF}$ characterizing antiferromagnetic order in the undoped parent compounds. The full spectrum thus resembles an "hour glass" in energy-momentum space.

Some features of this spectrum agree with calculations for a specific stripe model, according to which nonmagnetic charge stripes separate a set of weakly coupled spin ladders in the copper oxide layers. A key prediction of these models is a pronounced in-plane anisotropy of the magnetic spectrum. This prediction can, in principle, be tested if a crystallographically unique in-plane axis pins the stripe propagation vector. In YBa$_2$Cu$_3$O$_{6+x}$, for instance, the CuO chains along the $b$-axis remain such a direction. In practice, however, crystallographic twinning (that is, the formation of microscopic domains in which $a$- and $b$-axes are interchanged) limits our ability to extract information about the in-plane anisotropy from data on the large specimens typically used for neutron scattering.

In a recent neutron scattering study of arrays of single twinned YBa$_2$Cu$_3$O$_{6+x}$ single crystals with $x = 0.85$ and $0.6$, we have provided detailed information about the in-plane anisotropy of the dynamical spin susceptibility in the superconducting (SC) state at low energies. The results disagree with the predictions of the static stripe model and have stimulated calculations based on fluctuating stripe arrays.

However, whereas the neutron data were taken in the SC state, none of the stripe models consider the presence of superconductivity. In order to provide stringent experimental constraints for theories based on fluctuating stripes, as well as concomitant theories based on anisotropic Fermi liquids and stripe states, we have carried out experiments on the spin dynamics in the normal state of underdoped YBa$_2$Cu$_3$O$_{6+x}$. Previous work on twinned samples was unable to resolve the in-plane anisotropy in the normal state, and the results appeared to indicate that the NS spin excitation spectrum is simply a broadened version of the spectrum in the SC state. Our new data now show that both spectra are in fact qualitatively different. Specifically, the singularity at $E_{res}$, which gives rise to the characteristic "hour glass" shape of the spectrum, disappears in the NS, and the spectrum exhibits an unusually steep dispersion with a marked in-plane anisotropy. The low-energy spectral weight is strongly reduced upon heating above a characteristic temperature $T = 200$ K. These results will be discussed in the light of recent theoretical work.

The experiments were performed on an array of 180 individually detwinne YBa$_2$Cu$_3$O$_{6+x}$ single crystals with superconducting transition temperatures (at midpoint) of $T_c = 61$ K and width $T_c/2$. The crystals were co-aligned on three Al plates with a mosaicity of $< 12$. The volume of the entire array was $450$ mm$^3$, and the twin domain population ratio was 94%. Measurements were performed at the IN8 spectrometer at the Institut Laue Langevin (Grenoble, France) and the 2T spectrometer at the Laboratoire Leon Brillouin (Saclay, France). Scans along $a$ and $b$ were carried out under identical instrumental resolution conditions by working in two different Brillouin zones. No collimators were used in order...
and show a complex dependence upon energy and temperature. A synopsis of the entire data set is shown in Fig. 2 at two temperature plateaux above and below $T_c$. In the following we will discuss the salient features of the spectrum and its temperature evolution by referring to Fig. 2 for overall trends, and to Fig. 1 for more subtle details.

The central result of this paper is the change of the topology of the dispersion surface from the SC to the normal state. First, we focus on the spectrum deep in the SC state (Fig. 1, 5 K). Starting from low excitation energies, first decreases with increasing energy, so that the IC peaks merge at $Q_{AF}$ at an energy of $E_{res} = 37.5$ meV. For $E > E_{res}$, increases again, so that the spectrum from the \textquotedblleft hour-glass\textquotedblright{} dispersion (Fig. 2) already familiar from previous work $[3,4,5,22]$ is preserved. In the NS, however, the singularity at $E_{res}$ is no longer present, and the incommensurability is only weakly energy-dependent over a wide energy range including $E_{res}$ (Fig. 2c,d). As a consequence, the NS spectrum no longer shows the \textquotedblleft hour-glass\textquotedblright{} shape that is characteristic of the magnetic spectrum below $T_c$, and that has been the subject of much recent debate. This constitutes a qualitative difference of the spectra in the SC and normal states.

A second major difference between the SC and normal states is manifested in the in-plane anisotropy. In the SC state, the dispersion is found to be steeper along $b^*$, the direction of the CuO chains (Fig. 1). The in-plane spectral weight is moderately anisotropic for $E < E_{res}$, that is, the peak intensity of cuts along $a^*$ is higher than that along $b^*$. As pointed out before [14], the anisotropy decreases with increasing energy approaching $E_{res}$. A new aspect revealed by the data in Fig. 1 is that for $E > E_{res}$ the spectral weight anisotropy disappears within the experimental sensitivity. Apart from the slight difference along $a$ (the two in-plane directions), the spectrum is thus fully two-dimensional at high energies.

In the NS, the in-plane anisotropy increases strongly for $E < E_{res}$ (70 K pro les in Figs. 1 and 2). Upon heating through $T_c$, and the overall extent of the signal in $Q$-space shrinks by about 30% along $b^*$, but only by 10% along $a^*$. The constant-energy pro les become at-topped, and if these pro les are fitted to two peaks displaced from $Q_{AF}$ the resulting a $b$ anisotropy of is nearly 40%, compared to 15% in the SC state. At energies up to $E_{res}$, the geometry of the NS spectrum is thus more anisotropic than the one in the SC state. In contrast, hardly any difference between spectra in the normal and SC states is discernible at excitation energies significantly exceeding $E_{res}$. This distinction is highlighted in Fig. 2e-f, where the difference between the magnetic spectra in the SC and normal states is shown. The difference signal comes from the down ward-dispersing branch below $E_{res}$, which draws its spectral weight from a limited range above and a more extended range below $E_{res}$ (negative signal in Figs. 2e,f). It is significantly less anisotropic than the NS spectrum itself (Fig. 2c,d), with respect to both and the spectral weight distribution. Notably, the difference spectrum is very similar to its analogue in almost optimally doped YBa$_2$Cu$_3$O$_{6.85}$, which was shown to exhibit a nearly circular geometry [14]. This suggests that the main characteristics of the SC state (such as the gap anisotropy) are similar at both doping levels.

The spectral arrangement associated with the formation of the down ward-dispersing branch at $T_c$ results in a sharp upturn of the intensity at points along this branch (Fig. 3a-b), while at $Q_{AF}$ and 30 meV there is only a broad maximum at $T_c$ (Fig. 3c). Upon further heating, however, the spectral weight at energies at and below
Figure 2: Color representation of the magnetic intensity. Panels a-b show the SC regime, c-d the regime just above $T_c$, and e-f the difference between both spectra. The upper and lower rows show scans along $(H, -1.5, -1.7)$ and $(1.5, K, 1.7)$, respectively. In order to obtain a meaningful color representation, the intensity at 250 K was subtracted for $E < 38$ meV and the data was corrected for a $Q$-linear background at all energies. The normalized wave-vector $k_F$ was set to $2.66\alpha$ below 38 meV and to $4.5\alpha$ above. Scans taken at the overlapping energy 38 meV were used to bring both energy ranges to the same scale. The white lines connect the fitted peak positions of the constant-energy cuts. Dotted lines represent upper bounds on the incommensurability.

$E_{\text{res}}$ declines uniformly at all $Q$-values and vanishes (to within the experimental sensitivity) at a characteristic temperature $T_\text{c}$ of 200 K. This is a further manifestation of the qualitative difference of the SC and normal state spectra. $T_\text{c}$ is comparable to the temperature below which various observables exhibit a 'pseudogap' in this doping range. Corresponding constant-energy cuts show that for $T > T_\text{c}$, the low-energy spectral weight is severely depleted over the entire Brillouin zone below a characteristic energy $E < 40$ meV (Fig. 1 e-h, see also Ref. [20]). Within our experimental sensitivity, the magnetic spectral weight for $E < E_{\text{res}}$ is indistinguishable from zero. However, it is unlikely that $E_{\text{res}}$ represents a true gap, because $YBa_2Cu_3O_6.6$ is metallic at room temperature, and significant Korringa relaxation has been observed in NMR experiments. At energies above $E_{\text{res}}$, in contrast, the spectral weight is only moderately reduced upon heating above $T_\text{c}$ (Fig. 1 a-d). We can thus distinguish a third temperature regime above $T_\text{c}$, with a magnetic excitation spectrum differing distinctly from the spectra deep inside the SC state and just above $T_\text{c}$. The intriguing question whether or not the approximate coincidence of $E_{\text{res}}$ and $E_{\text{pseudo}}$ is accidental should be addressed by theory.

We now discuss the implications of our observations for stripe models of the cuprates. On a qualitative level, the large additional low-energy spectral weight below $T_\text{c}$ and the substantial in-plane NS anisotropy appear compatible with incipient stripe order. Moreover, recent STM experiments in the NS of underdoped $Bi_2Sr_2CaCu_2O_8$ have revealed low-energy charge excitations with a 'vertical' dispersion akin to corresponding features observed in our neutron experiment [24]. A related correspondence has recently been pointed out for the SC state of $La_2-xSr_xCuO_4$, Ref. [23].

However, the calculations of the spin excitation spectra of striped phases reported thus far do not provide a satisfactory description of our data. In static stripe models, $E_{\text{res}}$ is determined by the interaction of spins in adjacent stripes and represents a saddle point separat-
FIG. 3: Temperature dependence of the peak intensity at different positions in energy-momentum space indicated in the legend, and sketched in reference to the "hourglass" dispersion in the SC state.

A low-energy regime of anisotropic two-dimensional (2D) excitations from a high-energy regime with purely 1D excitations [7,8,9]. At $E > E_{\text{gap}}$, the neutron cross section is expected to take the form of streaks in the wave vector direction perpendicular to the stripes, and only one of the two orthogonal in-plane scans is expected to show FC peaks, in contradiction to our data in Fig. 1a(d). This definitely rules out static stripe scenarios for YBa$_2$Cu$_3$O$_{6.8}$, in agreement with previous conclusions based on low-energy excitations in untwinned crystals [12] and high-energy excitations in partially twinned crystals [13] in the SC state.

The spin excitation spectra of dynamical stripes have recently been computed numerically [15]. For slowly fluctuating stripes, the constant-energy cuts exhibit a quasi-2D intensity distribution, as experimentally observed, but the spectrum retains its overall "hourglass" shape. However, since the influence of superconductivity has thus far not been considered, predictions of these models should be compared to our new NS-data, where the "hourglass" shape is no longer present. If both SC and normal state spectra were found to be amenable to an interpretation based on fluctuating stripes, our data imply that the stripe fluctuation rate would have to change dramatically upon crossing $T_c$. To our knowledge, such a scenario has not been predicted. Similar considerations apply for other recently proposed models according to which the "hourglass" shape of the magnetic spectrum is a consequence of spiral spin correlations [15] or other types of modulation [23].

A natural explanation of the qualitative difference between the magnetic excitation spectra in the SC and normal states is provided by models that regard the downward-dispersing branch as an excitonic mode in the spin-triplet channel below the SC energy gap [28]. If the downward dispersion is a manifestation of the d-wave symmetry of the gap, its disappearance in the normal state is not surprising. Quantitative calculations of the spin excitation spectrum associated with the triplet excitation have been carried out mostly in the framework of weak-coupling schemes. It is questionable whether such models are capable of describing the strong temperature evolution we have found for $T_c < T < T_c$. Even in the absence of detailed calculations, however, the qualitative difference between the spectra above and below $T_c$ appears inconsistent with the proposal [28] that the low-energy spin excitations observed for $T_c < T < T$ should be regarded as an incoherent precursor of the triplet excitation below $T_c$.

A microscopic explanation of the unusual dispersion and the strong in-plane anisotropy we have observed in the normal state, as well as its strong temperature evolution for $T_c < T < T_c$, remains an important challenge for theoretical work. An interesting analogy is offered by the spin-1 model compound Y$_1$Ca$_6$BaN$_6$, which exhibits a Haldane gap for $x = 0$. For nonzero density of mobile charge carriers, $n$, additional spin excitations with a "vertical" dispersion develop below the Haldane gap [23]. The similarity with the temperature-driven development of spin correlations below the characteristic energy $E$ in YBa$_2$Cu$_3$O$_{6.6}$ provides hope that the doped Haldane chain could serve as a simple model system for a theoretical description of this behavior.

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tive vectors $a$, $b$ and $c$ where $a = 2 = a = 3.82\text{Å}$, $b = 3.87\text{Å}$ and $c = 11.7\text{Å}$. We choose its out-of-plane component $L_0 = 17 (2n + 1)$, $n$ integer, to probe magnetic excitations that are odd under the exchange of two layers within a bilayer unit. As even excitations show a gap of $54 \text{meV}$ and are much less $T$-dependent, they are presented elsewhere (S. Paihès et al., cond-mat/0512634).

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