Spin-glass behaviour in non-superconducting YBCO6.31 with planar hole doping p = 0.045 less than the critical doping for superconductivity, p_c = 0.052

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Despite numerous experimental and theoretical studies since the discovery of high temperature superconductivity (HTSC) in cuprates in 1986, there is still no consensus on the mechanism responsible for superconductivity in these materials. A qualitatively common phase diagram as a function of doping and temperature (with some differences in details), however, has emerged from the intensive experimental studies (benefited tremendously from advances in growth of high quality crystals in the past decade) on different HTSC families. For undoped and low doped materials, an insulating and antiferromagnetically (AF) long-ranged ordered state is observed at low temperatures due to the localized spins of Cu^{2+} in the CuO_2 planes common to all HTSC cuprates. Introduction of holes into the CuO_2 planes by doping results in a rapid destruction of the AF ordered state. Increasing doping beyond a critical doping (p_c) eventually leads to the disappearance of the long-ranged AF order and simultaneously the appearance of superconductivity. Further doping enhances superconductivity and a maximum T_c is observed at the optimal doping level beyond which T_c is lowered. Even though long-range AF order does not coexist with superconductivity strong spin fluctuations nonetheless survive well into the superconducting (SC) phase. It is widely believed that only when novel properties of HTSC cuprates are fully elucidated across the whole phase diagram (as a function of doping and temperature), will a complete understanding of these materials be gained. The search for new phases of matter, especially in the underdoped region as the AF phase is approached, continues to be a stimulus for both experiment and theory.

The single-layer $La_{2-x}Sr_xCuO_4$ (LSCO) cuprate family has been extensively studied by neutron scattering [1 - 6]. It is well established that a new spin-glass phase with static short-range AF static correlations coexists with the SC phase for doping levels close to p_c. The spin-glass phase extends to lower doping than p_c until it is eventually substituted by the long-range AF phase. The static magnetic peaks associated with the spin-glass phase appear at incommensurate positions for both superconducting and non-superconducting parts of the phase diagram. However, in the SC phase the static incommensurate peaks are observed along [100] direction whereas in the non-superconducting region they are rotated by 45° appearing along [110] direction.

One of the most well characterized cuprates is the bilayer $YBa_2Cu_3O_{6+x}$ (YBCO6+x) system with a maximum T_c ~ 94 K. Despite the extensive amount of attention this system has received, there have been no detailed studies of the magnetic properties for hole doping near the lower edge of superconducting phase. We have carried out [7 - 11] such detailed studies using unpolarized and polarized thermal neutrons and cold neutrons of the elastic and inelastic spectrum in YBCO6.35 (T_c = 20 K, p = 0.06) and YBCO6.33 (T_c = 8.5 K, p = 0.055) with doping near p_c = 0.052. In these low doped but superconducting samples, we have found that the spins fluctuate on two energy scales: one a damped spin response with a ~ 2 - 4 meV relaxation rate and the other a central mode (not seen in higher doped samples) with a relaxation rate that slows to less than 0.08 meV at low temperatures. The quasistatic mode is centred on commensurate AF positions (0.5 0.5 L) with L = integer showing the development of spin correlations between cells. Our studies have indicated that spatial correlation lengths of the central mode remain finite in the superconducting phase although they increase as doping decreases from YBCO6.35 to YBCO6.33. The central mode intensity smoothly increases on cooling indicating no sign of a critical transition to a Néel state. Lowering doping also results in an increased temperature scale for the growth of the central mode.

In new experiments we have searched for Néel AF order in non-superconducting YBCO6.31 (p = 0.045 < p_c). Surprisingly, none was found [11], indicating that the AF and SC phases are separated by an intervening phase. The spin behaviour evolves smoothly from that of the low-doped, weakly superconducting systems. The correlation lengths in YBCO6.31 non-superconducting system are much larger than ones observed earlier in YBCO6.35 and YBCO6.33, although they remain finite.

A particular feature of interest is a clear decrease in intensity of the central mode that we have observed for YBCO6.31 at low temperatures (below 20 K in figure 1). Such behaviour is seen in conventional spin-glass insulating systems. In addition, for all samples, the temperature scale of the central mode decreases with tighter energy resolution (longer timescales), again reminiscent of a spin-glass. Hence we have demonstrated that a normal (non-superconducting) phase with characteristics similar to those of a spin-glass separates AF and SC phases in the YBCO phase diagram. This spin-glass behaviour extends into the superconducting phase as we showed in earlier studies [7 - 10]. The magnetic spin-glass behaviour is therefore continuous across the superconducting phase transition. Our results show that as doping changes the spinons carrying spin evolve differently than the holons that carry charge. This phase evolution with doping is similar to the LSCO cuprates, except that the static short-ranged magnetic peaks of YBCO are commensurate at all doping levels.

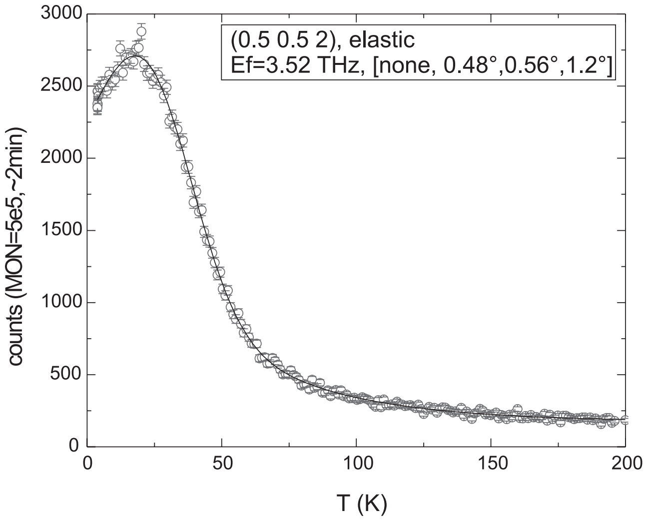

Fig 1. *Temperature dependence of the elastic scattering observed at AF position (0.5 0.5 2). The solid line is a guide to the eye.*

To be able to determine a more precise value for the lower limit of the correlation lengths, we first studied elastic properties of YBCO6.31 at the C5 spectrometer with a tight collimation of [none, 0.273°, 0.477°, 0.44°] and with the other spectrometer parameters the same as Ref. [11]. In addition to be able to fully compare the magnetic properties of YBCO6.31 with that of higher doped materials, we explored the inelastic spectrum observed at AF position and investigated the magnetic field dependence of both elastic and inelastic scattering. The inelastic and field dependence studies were performed with a looser collimation setting of [none, 0.48°, 0.55°, 1.2°]. The sample (described in Ref. [11]) was aligned in the (HHL) plane and mounted in a closed-cycle refrigerator for the zero field measurements and in the horizontal field magnet M2 for the field dependence study. The 335° (for 2-inch beam) horizontal access of the magnet enabled us to perform elastic and inelastic measurements over a large range of wave vectors and energy transfers while applying the magnetic field parallel to the c-axis.

The results of our elastic measurements with the tight collimation setting are shown in figures 2(a) and (b) for (HH2) and (0.5 0.5 L) scans, respectively. In these figures we have also shown similar scans with no filter in the beam so as to reveal the spectrometer and mosaic resolution. This direct experimental comparison of the observed scattering with the resolution shows that for both in-plane and out-of-plane correlations, the observed scattering is broader than the resolution indicating that the correlations are finite. From resolution-convoluted fits to a Lorentzian profile we obtain a lower limit of 350 Å and 100 Å for the correlation lengths in the ab-plane and along the c-direction, respectively. This confirms our lower-resolution results [11] that even though correlations are much longer in non-superconducting YBCO6.31 than higher doped superconducting samples, they remain finite.

Similarly to YBCO6.33, we found that the energy spectrum of YBCO6.31 also consists of two modes. One is the central mode discussed above and the other is a broad component peaked at about 1 THz (~ 4 meV). Figure 3(a) shows the background subtracted spectrum at 3 K observed (corrected for the form factor) at the AF position (0.5 0.5 2) and (0.5 0.5 5.1). Our study indicates that the inelastic scattering follows the coupled bilayer model [12] as shown by the line in figure 3(b), a behaviour again similar to higher doped materials. The agreement of the intensity at L = 5.1 and L = 2, when only the anisotropic Cu^{2+} form factor is extracted, shows that the spins fluctuate isotropically in spin orientation, $\chi_{xx} = \chi_{yy} = \chi_{zz}$. This differs from the low doped LSCO where the spin orientation is anisotropic and confined to the ab-plane.

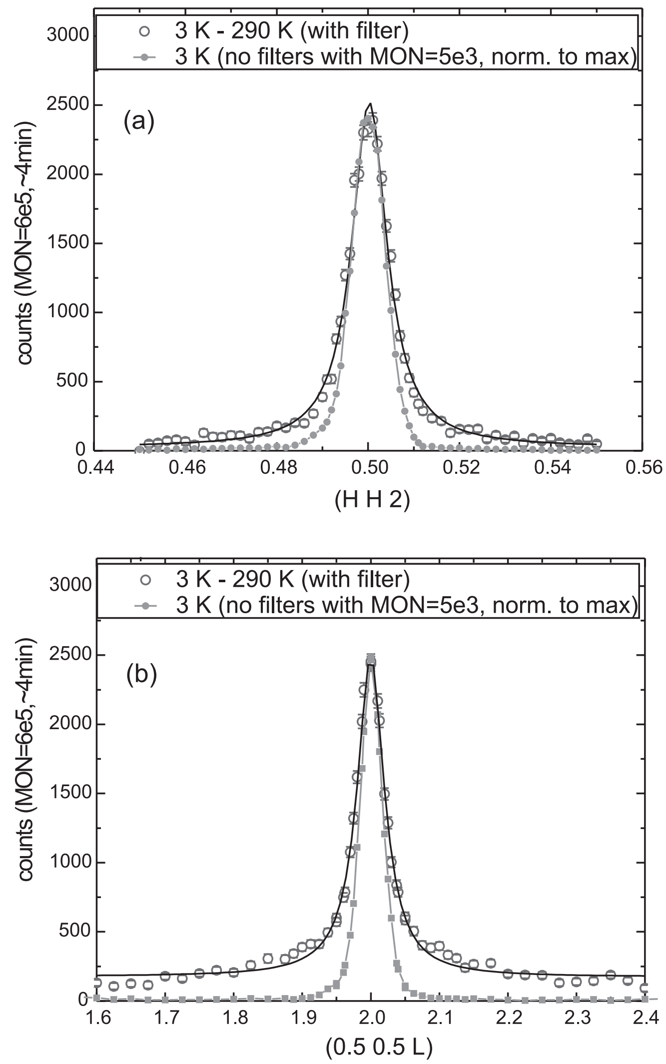

Fig 2. *Background subtracted elastic scans around (0.5 0.5 2) AF position (empty squares). A direct comparison is shown with the resolution data collected with no filter. Data is obtained with the tight collimation setting [none, 0.273°, 0.477°, 0.44°].*

We also studied the effect of a magnetic field of 2.6 T || c-axis on both elastic and inelastic properties of YBCO6.31 using the horizontal magnet M2. As for higher doped YBCO samples, we find that the elastic AF (central mode) intensity and its dependence on temperature do not change with the application of the field, see figure 4(a). For a conventional long-range ordered AF one would expect a reduction in the intensity of the ordered AF moments with the application of a magnetic field. Moreover any spin re-orientation due to the applied field would generally change the magnetic intensity through the Lorentz factor. The field independent intensity shows that the spins remain frozen in the isotropic random orientations suggested above, and as found from earlier polarized neutron experiments on YBCO6.33 [10]. This differs from the easy-plane anisotropy of LSCO [13]. In the non-superconducting region of LSCO, a magnetic field of 4.4 T || c-axis is sufficient to change the magnetic structure and to weaken the incommensurate correlations [13]. Furthermore we find that no change occurs in the intensity of the inelastic scattering in a magnetic field of 2.6 T || c-axis as shown in figure 4(b).

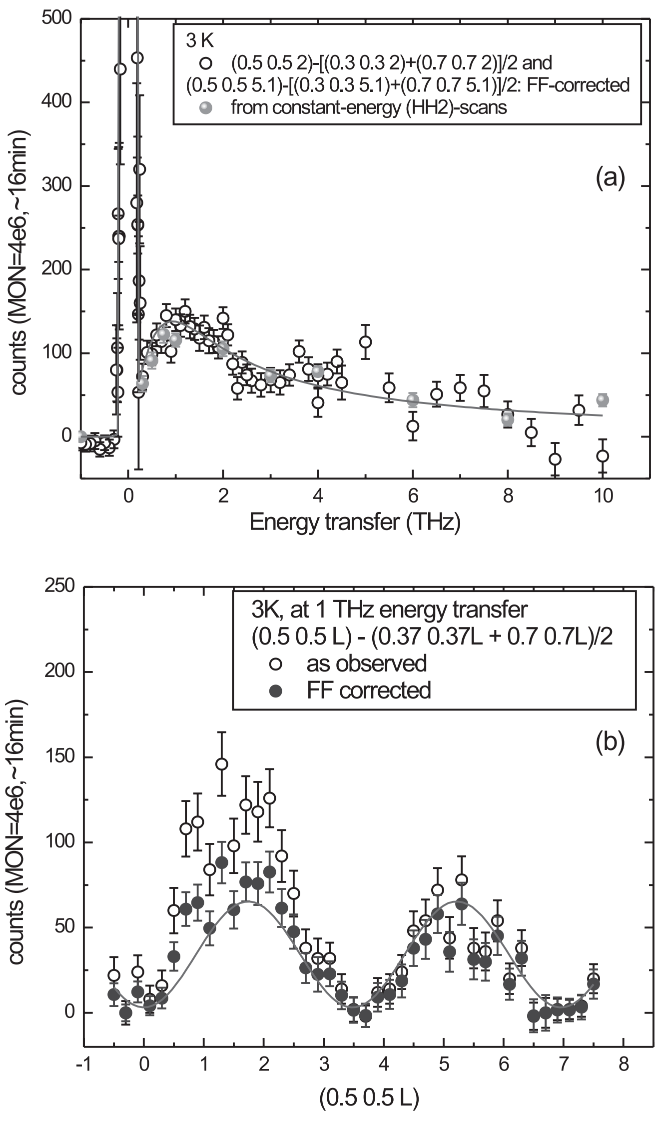

Fig 3. *(a) Inelastic spectrum observed at AF position at 3 K (empty circles: from background subtracted constant-q energy scans and solid circles: from constant energy (HH2)-scans). (b) Inelastic (0.5 0.5 L)-scan at 1 THz and 3 K (empty circles). The form factor corrected data is shown with full circles. The solid line is a fit to the form factor corrected coupled-bilayer model for randomly isotropic spin orientations.*

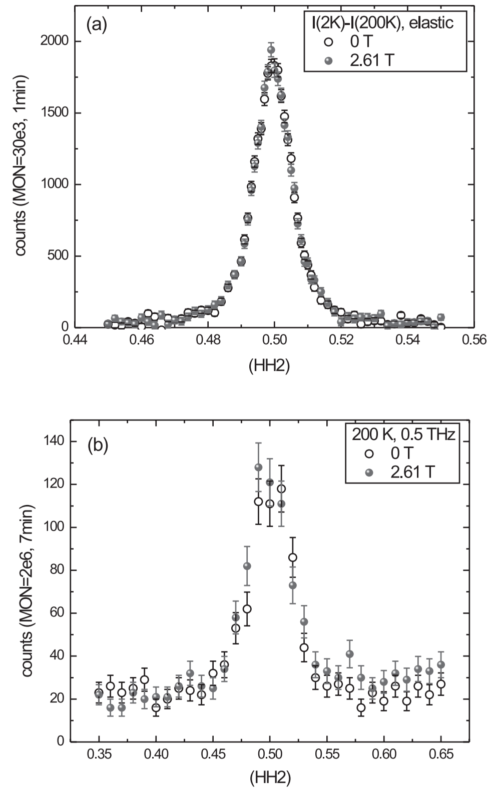

Fig 4. *Magnetic properties of YBCO6.31 do not change in an applied field of 2.6 T || c-axis. (a) elastic scattering and (b) inelastic scattering in zero field compared with 2.6 T.*

In summary, we have shown that no Bragg ordered Néel phase occurs adjacent to the superconducting phase of YBCO system. The correlations remain finite in non-superconducting YBCO6.31 material. Instead the lower doping leads to correlations that are significantly longer especially along the c-axis. The very long range in-plane correlations show that structure or chemical disorder plays a very small role in these high-quality crystals grown at UBC. This is not surprising since in YBCO6+x doping occurs by changing the oxygen content in the CuO chains, which are located away from the CuO_2 planes where both superconductivity and antiferromagnetism reside. Our study also indicates that the spin dynamics in this non-superconducting sample is qualitatively similar to the superconducting samples. All these indicate that the magnetic properties gradually change across p_c, the critical doping for superconductivity and that the phase separating the long range AF ordered and superconducting phases has spin-glass characteristics.

References

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