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NEURON SCATTERING STUDIES OF THE MAGNETIC FLUCTUATIONS IN $YBa_2Cu_3O_{7-\delta}$

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Abstract—Neutron scattering measurements were made on the spin fluctuations in $YBa_2Cu_3O_{7-\delta}$ for different oxygen doping levels. Incommensurability is clearly observed for oxygen concentrations of 6.6 and 6.7 and is suggested for the 6.93. Measurements of the resonance for the $O_{6.6}$ concentration show that it exists in a broadened and less intense form at temperatures much higher than T_c. © 1998 Elsevier Science Ltd. All rights reserved

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It now seems clear that the magnetic fluctuations play a central role in the behavior of high-T_c superconducting materials. Neutron scattering can determine both the wavevector and energy dependence of the magnetic fluctuations in a straightforward manner. However, the technique is strongly limited by the intensity of available neutron beams, thus, requiring the use of large single crystal samples. $YBa_2Cu_3O_{7-\delta}$ (YBCO) is one of the best characterized of the high-T_c superconductors, but only now are crystals becoming available with sufficient quality and oxygen uniformity that reliable results can be obtained. This is particularly true of the underdoped materials that display interesting pseudogap behavior. In these materials very high oxygen uniformity is necessary, yet this is difficult with the large samples needed. We were successful in developing techniques that insure uniform oxygen concentrations throughout the whole volume of large high quality crystals.

A second problem with the YBCO materials is that the energy scale of interest for the magnetic fluctuations is the same as that for many of the intense phonon excitations. It is often very difficult to separate the magnetic excitations in neutron scattering experiments from the far more intense phonon excitations. We have approached the problem by using polarization analysis when possible to isolate the magnetic signal. In some cases, however, the magnetic scattering is too weak to permit the use of polarization analysis which is far less efficient than conventional techniques. Nevertheless, we have succeeded in characterizing the phonon excitations better through measurements and models so that a more reliable extraction of the magnetic signal is possible.

Another factor that has greatly aided our measurements is the development of the filter integration technique [1]. In this technique use is made of the fact that the magnetic fluctuations are, except for the bilayer coupling, two-dimensional in nature and the scattering is summed over the c^* direction in the reciprocal lattice greatly increasing the magnetic signal relative to other types of scattering. Graphite filters are used to essentially eliminate the elastic scattering so that only the fluctuations are measured. We do not obtain detailed energy selection with this technique, rather sample a band of energies. It is, thus, often desirable to perform standard measurements with triple-axis spectrometry to confirm some of the results if possible. However, the technique is excellent for survey studies and to measure very small signals.

In this paper we will consider results on two of the most interesting aspects of the spin fluctuations in the YBCO materials, the resonance and the incommensurability. Let us first consider measurements for $YBCO_{6.6}$. The magnetic fluctuations are found around the reciprocal lattice position (π,π) of the unit cell which is the position where antiferromagnetism is observed in the highly underdoped ordered compounds. This is the position (0.5, 0.5, L) in reciprocal lattice units. We find the scattering in question stems from coupled bilayers so that L is set for a value to sample acoustic scattering, in our case 1.7. To search for incommensurate fluctuations we scan along the line from (0,0) to $(2\pi, 2\pi)$ or along (h,h) in the reciprocal lattice. In the scan we use a good resolution along the scan direction but must employ poor resolution perpendicular to this direction to obtain sufficient intensity. The scan direction is shown on the top of Fig. 1. along with the position of possible incommensurate magnetic satellites. If the satellites are as shown, a peak in the scattering will be found when a line perpendicular to the scan direction intersects the left and lower satellites at the same time. A similar peak (smaller because of the magnetic form factor) will

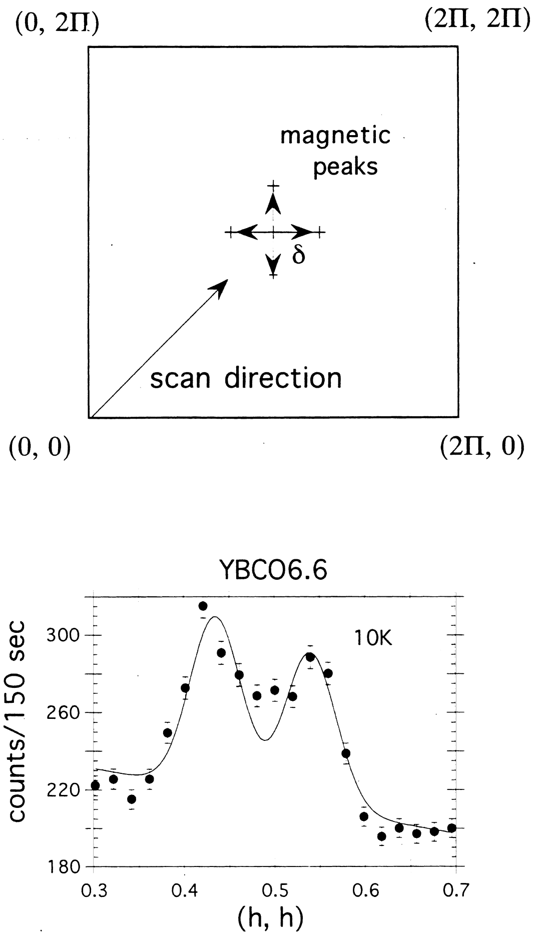

Fig. 1. The top panel shows a possible position of the incommensurate satellites in YBCO. A scan along the arrow would produce two peaks stemming from the very relaxed resolution perpendicular to the scan direction. The bottom graph shows such a scan for $YBCO_{6.6}$.

be found when the perpendicular intersects the upper and right hand satellites simultaneously. Another set of possible positions of the satellites would be along the scan direction in which case three peaks would be observed, two from individual satellites on the scan direction and the third at the (0.5, 0.5) position as the perpendicular intersects peaks above and below the scan direction. The peak at (0.5, 0.5) could be smaller depending on the extent of the resolution perpendicular to the scan. There could of course be other possible arrangements of the magnetic scattering.

The graph in the lower part of Fig. 1. shows a scan made for $YBCO_{6.6}$ using the filter integration technique. Peaks are observed on either side of the (0.5, 0.5) position for (h,h) showing that the magnetic fluctuations are incommensurate. Further measurements with triple axis spectrometry show the fluctuations to be incommensurate [2] at energies below the resonance energy of about 34 meV. The incommensurate position of the peaks from a series of scans was determined to be (0.057, 0.057) with an error of 0.006. Note the actual satellite wavevector would be 2×0.057 if the satellites are actually as pictured in Fig. 1. We are in the process of making sets of scans along different paths to determine the actual satellite wavevector. This is much more difficult than for the 214 superconductors because of the energy scale and the bilayer nature of the scattering.

We move now to measurements of the resonance scattering in $YBCO_{6.6}$. The resonance scattering in the fully doped YBCO materials is a very narrow peak found at 41 meV. The first observation of increased scattering at low temperatures at 41 meV in YBCO was made by Rossat-Mignod *et al.* [3] although the low temperature response was considered a rather broad band of scattering at that time. Mook and coworkers [4] showed using polarized beam techniques that for $YBCO_{6.93}$ the 41 meV scattering was a sharp peak that dominated the low temperature scattering. The measurements suggested that a small peak remained at 100 K. Fong *et al.* [5] determined that for highly doped YBCO the resonance is not found above the superconducting transition temperature. The present results on $YBCO_{6.6}$ show that the resonance remains as a broadened and considerably less intense peak in energy at temperatures as high as a temperature T^* which could be interpreted as the pseudogap temperature.

The resonance energy for $YBCO_{6.6}$ was found to be about 34 meV [6, 7]. We show in Fig. 2 scans in energy through the resonance for six temperatures. These scans were obtained by making a series of scans in momentum q to determine the phonon background. The phonons tend to be reasonably flat in q near (0.5, 1.5, 1.7) where the measurements were taken so they could be subtracted out with a considerable degree of confidence. Our scan in energy was then derived from the scans in momentum. Our success at removing the phonon background is verified by the 293 K data which shows little scattering as expected for the magnetic excitations.

The resonance at 10 K is resolution limited for our experiment. As the temperature is raised through $T_c = 63$ K the resonance broadens and diminishes in intensity, but is still observed as a definite peak in energy. A signature of the resonance remains at 150 K, but appears to vanish by about 200 K with little scattering observed by 293 K. A scan of intensity vs temperature made at the resonance energy of 34 meV shown in Ref. [6] indicates that the character of the scattering changes at T_c. The scattering intensity has a noticeable upturn upon cooling through T_c and increases more rapidly as the temperature is lowered further. A possible way to consider the observed behavior is to characterize the resonance as a property of the superconducting state with a precursor that exists in the normal state at temperatures as high as a pseudogap temperature T^*.

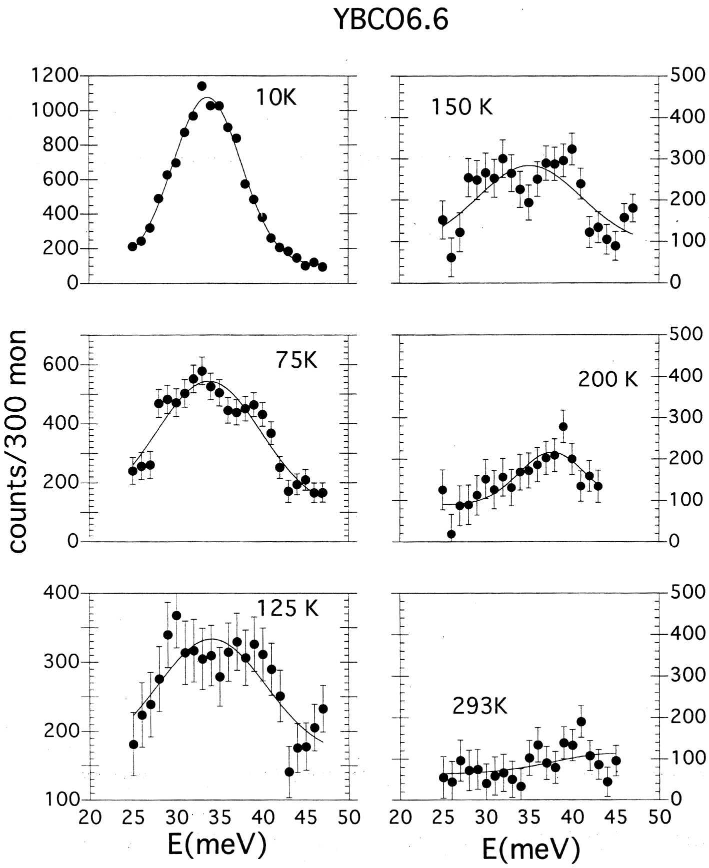

Fig. 2. Triple-axis scans through the resonance energy for $YBCO_{6.6}$. We note the resonance peak survives in a broadened and less intense form at temperatures as high as 150 K.

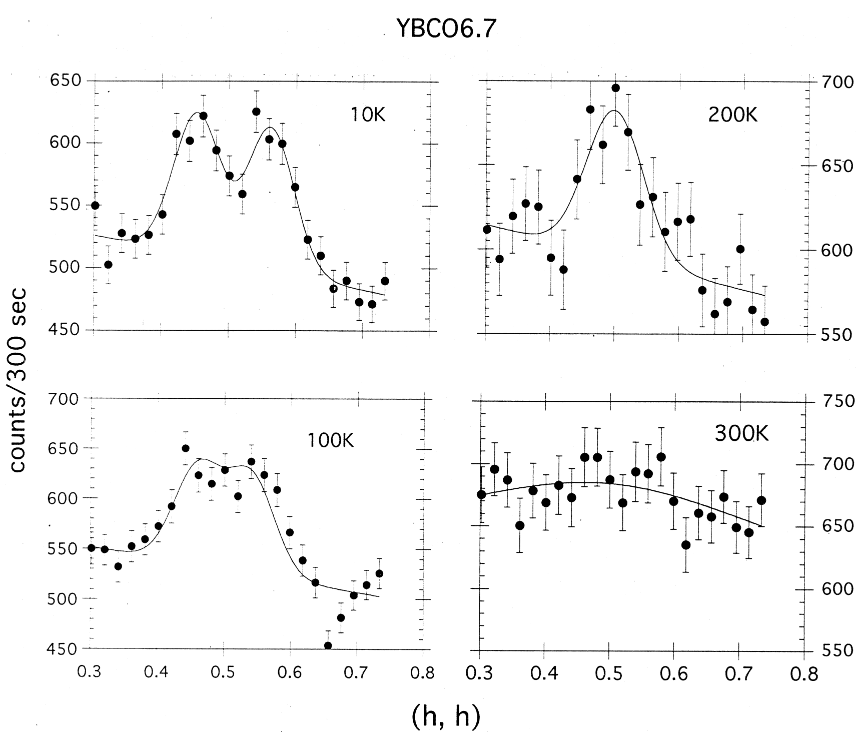

Fig. 3. Scans made along the direction shown in Fig. 1 for $YBCO_{6.7}$. Incommensurability is observed at low temperatures with little magnetic intensity remaining at 300 K.

We now turn to measurements on a sample with an oxygen concentration of 6.7, $YBCO_{6.7}$. We find that T_c is 74 K for this sample while the resonance energy is 38.5 meV. Fig. 3 shows our measurements of the magnetic scattering made for this sample using the filter integration technique. The scan direction is along (π,π) and was done in the same way as the scan shown in Fig. 1. Again we observe the scattering is incommensurate at low temperatures. The incommensurability is still observed as the temperature is raised to 100 K, but is clearly gone by 200 K. Little if any scattering is observed at room temperature showing that the scans are free of phonon contributions. Least squares analysis of low temperature scans show an incommensurability wavevector of (0.061, 0.061) with an error of 0.005. The measured incommensurate wavevector is, thus, larger than for the $YBCO_{6.6}$ sample, but the error in position is sufficiently large that a clear difference is not established. The precise determination of the incommensurability wavevector is hampered by the weakness of the scattering coupled with an energy scale that results in reduced momentum resolution.

We have performed preliminary experiments to search for incommensurate fluctuations in the optimally doped material $YBCO_{6.93}$. We use the integration technique once again and the results are shown in Fig. 4. The integration technique is not as favorable in this case as the energy window passed by the filter now includes the resonance to some degree. The energy sensitivity of the experiment is given in Ref. [2]. The scan at 10 K shown in the top graph is, thus, expected to show a peak at $(h,h) = 0.5$ that stems from the resonance. We see in addition unresolved structure surrounding the peak at 0.5 that can be interpreted as stemming from incommensurate fluctuations. This structure largely disappears at 100 K with the peak at 0.5 now likely stemming from normal state commensurate excitations. Since now T^* is much closer to T_c for the $O_{6.93}$ material the 100 K spectrum appears similar to the 200 K spectrum for the $O_{6.7}$ sample. The counting rate for the $O_{6.93}$ experiment is considerably higher than for the other concentrations as a much bigger sample was used and the collimation was relaxed to some degree. The bottom panel of Fig. 4 shows the result of taking the difference between the 10 K and 100 K measurements. This result gives an indication of the nature of the incommensurate part of the scattering pattern. The intensity of the magnetic scattering decreases rapidly as higher oxygen doping levels are

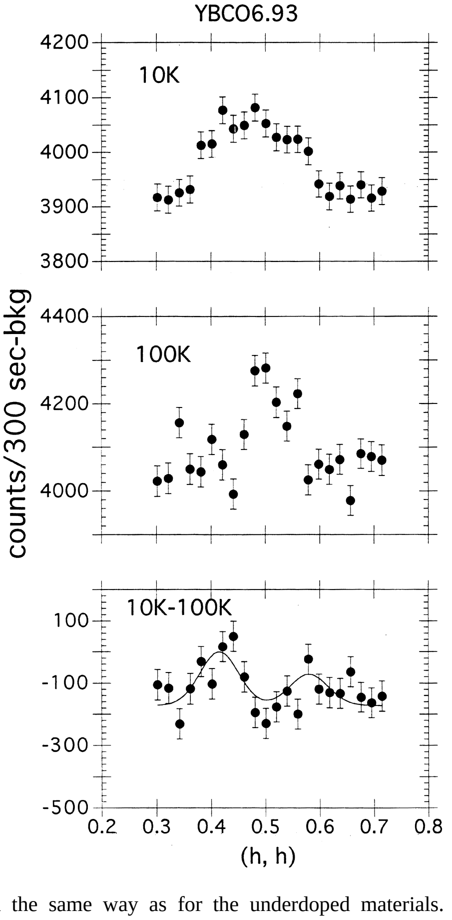

Fig. 4. Scans for $YBCO_{6.93}$ made in the same way as for the underdoped materials. Structure in the 10 K scan suggests incommensurability that is more clearly shown in the difference result shown in the bottom panel.

used so the counting errors, particularly in the difference pattern, are larger than desirable. It can also be misleading to use the differences of patterns taken at different temperatures to determine lineshapes. Nevertheless, the difference pattern serves as an indication of possible incommensurability in optimally doped YBCO. A least squares fit of Gaussian lineshapes to the pattern shown in the figure results in an incommensurability wavevector of (0.08, 0.08). If the satellites are indeed in the position shown in Fig. 1 the incommensurability wavevector would be 2×0.08 or a value near the wavevector for the optimally doped 214 material. However, additional measurements are needed to confirm the incommensurability of $YBCO_{6.93}$ and to determine the momentum space positions of satellite peaks.

We will not discuss in this paper how these results affect various theories of high T_c superconductivity. However, we will make some general observations. It is important that the resonance not only shifts it's position in the underdoped materials, but that its basic character is modified. In the heavily doped material the resonance begins to appear only near T_c while precursor effects with a temperature scale that matches the pseudogap temperature are observed in the underdoped case. We note the resonance is affected by the bulk superconducting transition as a change in slope in the temperature dependence occurs at T_c. Theories to explain the resonance must take these observations into consideration.

The observation of incommensurability is important as it unifies the picture of magnetic fluctuations in the high-T_c materials. The preliminary evidence we have on the doping dependence suggests a close connection between the 214 and 123 materials. Additional measurements are needed to make detailed connections. Obviously any theoretical models that can account for the incommensurability would now appear to be applicable to the high-T_c materials in general.

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