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MAGNETIC FLUCTUATIONS IN THE COMPOUNDS $YBa_2CU_3O_{6+x}$ ($0< x < 1$)

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Abstract. – We performed neutron scattering experiments using polarization analysis on the high temperature superconductor $YBa_2Cu_3O_{6.9}$ and the related semiconducting compounds $YBa_2Cu_3O_{6+x}$ ($x = 0.11$, 0.37). While we found paramagnetic fluctuations in the non-superconducting samples, we established severe upper bounds for the total magnetic intensity for the superconductor. We conclude that magnetic couplings as a source for superconductivity in $YBa_2Cu_3O_7$ are highly unlikely.

1. Introduction

Since the discovery of the "high-temperature" superconductivity in Cu-oxides, several magnetic coupling mechanisms have been put forward as being responsible for the high transition temperatures (compare Refs. in [1]). All of these models rely on magnetic excitations (or fluctuations) of energy of order kT_C (the superconducting transition temperature). For the $YBa_2Cu_3O_{6+x}$ ($0 < x < 1$) series, the discovery of antiferromagnetic long range order in the non-superconducting, tetragonal samples with $x < 0.5$ [2-5] seemed to support these speculations.

Here, we present results of a neutron scattering study of the magnetic fluctuations and excitations in the $YBa_2Cu_3O_{6+x}$ compounds [1, 6]. Magnetic neutron scattering is the ideal technique for these investigations: Polarization analysis allows the unambiguous distinction of electronic magnetic fluctuations from all other excitations. The energy and wavevector dependence of the magnetic scattering cross-section gives information about magnetic correlations on a microscopic scale.

2. Sample preparation and characterization

According to the procedures described in [7], we prepared three powder samples of $YBa_2Cu_3O_{6+x}$. For all samples, we have examined structure, sample homogeneity and oxygen content on the high-resolution neutron diffractometer D2B at the ILL/Grenoble. The samples were single phase and had the structures described in [8]. The two samples with $x < 0.5$ were tetragonal (space group P4/mmm), while the superconductor with $x \approx 1$ had the orthorhombic structure (space group Pmmm). From a profile-refinement of the powder diffraction patterns, we determined the structural parameters, including the occupancy of the oxygen sites. This lead to the following values for the parameter x : $x = 0.11(2)$, $x = 0.37(6)$ and $x = 0.89(3)$.

3. Principles of the polarization analysis

We have extracted the pure magnetic scattering by means of polarization analysis on the instrument D7 at the high flux reactor of the ILL in Grenoble. Details of the experimental technique were already given in [1, 6, 9]. We repeat only the fundamentals:

to determine the energy-integrated magnetic intensity, the instrument was used in the integral mode without time-of-flight energy analysis. Spin-flip (SF) as well as non-spin-flip (NSF) intensities are recorded simultaneously in 32 detectors for polarization of the incident beam in three orthogonal directions x, y and z. For powder samples, the magnetic scattering rate can then ideally be determined independently from the SF and the NSF intensities:

$$\frac{1}{2}I_{\mathrm{mag}}=I_{\mathrm{SF}}^{(x)}+I_{\mathrm{SF}}^{(y)}-2I_{\mathrm{SF}}^{(z)}$$
$$\frac{1}{2}I_{\mathrm{mag}}=2I_{\mathrm{NSF}}^{(z)}-I_{\mathrm{NSF}}^{(x)}-I_{\mathrm{NSF}}^{(y)}.$$

With this difference technique systematic errors can be minimized at the expense of counting statistics.

For corrections, we measured the empty sample container, a cadmium sample, a vanadium sample and a quartz-sample. We corrected for multiple polarization dependent scattering of the vanadium sample, for the relative detector efficiency, for background and for the finite flipping ratio.

For an estimation of the total magnetic cross section, an integration over the first Brillouin zone and over the frequency spectrum of the magnetic fluctuations must be carried out [1]. The integration over reciprocal space is easily achieved for powder samples, while the experimental energy integration in the integral mode includes several energy-dependent factors, like the transmission function of the supermirror-analyzers [1]. These reduce the contribution of high energy fluctuations with respect to elastic scattering. Therefore, we can only give an estimation of the total scattering cross section within limited energy intervals.

4. Experimental results

4.1 SEMICONDUCTING SAMPLES. – For the $x = 0.11$ sample, we verified the magnetic nature of the Bragg peaks (1/2, 1/2, 1), (1/2, 1/2, 2) and (1/2, 1/2, 3) at room temperature. From the temperature dependence of the magnetic Bragg intensities, we estimate the Néel-temperature for this sample to be T_N=410(20)K. For the $x = 0.37$–sample, we found no clear indication for magnetic long range order at room temperature.

In a search for magnetic fluctuations above T_N, we acquired data at $T = 450$ K for the $x = 0.11$ sample and at $T = 293$ K for the $x = 0.37$ sample. Within statistics, no pronounced short range order modulation could be seen, although we observed enhanced intensities close to the (1/2, 1/2, 1) and (1/2, 1/2, 2) positions. Whether this phenomenon is due to short range order scattering with a large correlation length or to a remaining long range order due to composition fluctuations can be decided unambiguously only with single crystal measurements. The total paramagnetic scattering cross section per formula unit within the energy interval [−25 meV, 25 meV] is estimated to be 0.36 barn for the $x = 0.11$ sample and 0.43 barn for the $x = 0.37$ sample. These values correspond to roughly 10 % of the Cu-atoms carrying a $S = 1/2$–moment. This rather small value can either be due to an important fraction of high energy ($E > 20$ meV) fluctuations or to a small fraction of Cu-atoms carrying a magnetic moment. It is also possible that itinerant magnetism has to be invoked for the understanding of the magnetic properties.

4.2 SUPERCONDUCTING SAMPLE. – In a search for magnetic fluctuations in the superconducting $x =$ 0.89–sample, we took data at $T = 70$ K, 110 K, 150 K, 200 K and 290 K. Figure 1 shows the magnetic differential cross section for the $T = 110$ K measurement. At all temperatures, the estimated values of the total magnetic scattering cross section were zero within the standard deviations. For the $T = 290$ K measurement, where the energy integration is between ±25 meV, the total magnetic cross section is 0.023(19) barn per formula unit. From this value we can estimate an upper

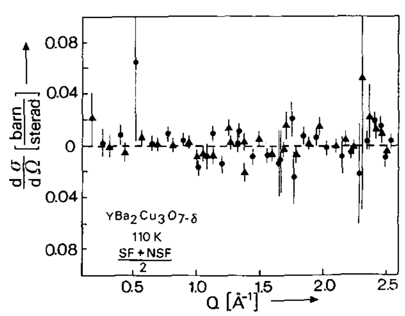

Fig. 1. – The magnetic differential cross-section $d\sigma/d\Omega$ [b/sr] per formula unit $YBa_2Cu_3O_{6.89}$ as a function of the momentum transfer Q at $T = 110$ K.

limit of Cu-atoms with spin 1/2 of 2.5 % . For the 110 K measurement, we estimate this value to be less than 1 % . This last number refers to an energy integration between ±12 meV.

5. Conclusions

To summarize our results, we gave first evidence for the existence of paramagnetic fluctuations in tetragonal, nonsuperconducting $YBa_2Cu_3O_{6+x}$ ($x = 0.11$, 0.37)-samples, while we established within severe upper bounds a vanishingly small total magnetic intensity for the superconducting $x = 0.89$ sample. We conclude from our observations that it is highly unlikely, that magnetic coupling mechanisms can explain the superconductivity in $YBa_2Cu_3O_7$. Since the magnetic intensity vanishes in the superconducting phase, the appearance of magnetism and paramagnetic fluctuations in the non-superconducting samples should not be taken to indicate magnetic mechanisms driving the superconducting phase transition. However the magnetism may be in competition with other many body effects like superconductivity.

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