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OBSERVATION OF ANTIFERROMAGNETIC ORDER IN YBa$_2$Cu$_3$O$_{6.15}$ by Cu NQR

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Abstract. – The NQR spectra of YBa$_2$Cu$_3$O$_{y}$ (6.0 < y < 6.25) show two different antiferromagnetic structures with a different stacking sequence. In both structures the magnetic moment is located at the Cu-2 sites perpendicular to the c-axis. The structure with antiferromagnetic coupling between all Cu-2 planes reveals strong temperature dependent magnetic fluctuations with correlation times of about $10^{-6}$ s at 15 K. No fluctuations are found in the second structure which reveals a constant internal field of about 0.2 T.

From muon spin rotation [1] and neutron scattering [2] it became evident that antiferromagnetic order exists in the system YBa$_2$Cu$_3$O$_{y}$ at low oxygen concentration 6.0 < y < 6.4 with the magnetic moments located at the Cu-2 sites (CuO$_2$ planes). Since these magnetic moments induce an hyperfine field at the corresponding Cu nuclei they must have a strong influence on the NQR spectra. In spite of that, the NQR spectra of some semiconducting samples with y ≈ 6.2 and the superconducting ones with y > 6.8 look quite similar [3] exhibiting two pairs of lines in the region between 18 and 36 MHz (Fig. 1 of Ref. [3]). The lines of each pair correspond to the ±3/2, ±1/2 transitions of the $^{63}$Cu and $^{65}$Cu isotopes, respectively. Each pair reflects a Cu site with a well defined electric field gradient (EFG). After some controversy about the assignment of the lines there is now general agreement [3, 4] that the high-frequency lines in superconducting samples (31.5 MHz for $^{63}$Cu) correspond to Cu-2 and the low-frequency lines (22.0 MHz) to Cu-1 (all frequency values hold for 4.2 K). For semiconducting samples we have deduced from the symmetry of the EFG and from the influence of Gd substituted for Y that Cu-1 with only 2 oxygen nearest neighbours leads to the high frequency line (30.1 MHz) and Cu-1 with 3 nn to the line at 24.0 MHz [3]. Correspondingly this line becomes very weak if y approaches 6.0. The lines of the Cu-2 sites have been detected at 89.89(5) MHz for $^{63}$Cu corresponding an hyperfine field of 7.665(5) T [5].

For some semiconducting samples a different NQR spectrum has been found with either a total or a partial splitting of the line at 30.1 MHz by an internal field (Fig. 5 of Ref. [3]). In this communication we present a detailed investigation of this splitting. By applying a small magnetic field the direction of the internal field was determined. Further we discuss the influence of the temperature on the spectra from which we obtain information about magnetic fluctuations in the antiferromagnetic structure.

For the spin-echo measurements powder samples with a random distribution of the crystal orientation have been used. They have been prepared either by quenching from high temperature or by thermal degassing of YBa$_2$Cu$_3$O$_{y}$ samples in ultrahigh vacuum and reloading them with oxygen. In two samples 1 or 0.2 % of the Cu was substituted by $^{57}$Fe. From the 9 samples with y ≤ 6.25 only two (y = 6.0 with 1 % $^{57}$Fe and y = 6.15 with the pure $^{63}$Cu isotope) reveal the full splitting, two (y = 6.0 and 6.05) show a superposition of a strong single line and a weak doublet and five (6.1 ≤ y ≤ 6.25) only a single line. The correlation between the conditions of preparation and the appearance of the doublet is still unclear. For the two Cu isotopes the splitting scales exactly with the γ values thus proving that it is caused by a magnetic field of 0.15 T for y = 6.0 and 0.20 T for y = 6.15, respectively.

The different spectra prove that two different magnetic structures exist in this system: In the structure proposed from neutron scattering (Fig. 1A) the influence of the moments cancels at the Cu-1 sites leading to the "normal" NQR spectrum without splitting. If, however, the stacking sequence changes from + + + to ++ + -- for example (Fig. 1B), the dipolar and the transferred hyperfine fields from nearest Cu-2 neighbours sum up at the Cu-1 sites resulting in an internal field parallel to the direction of the magnetic moments.

Fig. 1. – Spin structure of YBa$_2$Cu$_3$O$_6$; (A) deduced from neutron scattering, no internal field at Cu-1; (B) proposed to explain an internal field at Cu-1.
For any simple antiferromagnetic structure the internal field must be either parallel or perpendicular to the c-axis. Since the symmetry axis of the EFG is parallel to the c-axis both cases can be distinguished by applying a small magnetic field \( B_0 \). By the superposition of an internal field of a fixed direction and an external field of random direction in the powder sample, each line of the doublet is split. For the internal field parallel to the symmetry axis of the EFG, the maximum and minimum values of the frequency shift as \( \pm \gamma/2\pi B_0 \) whereas for the internal field perpendicular to it the initial slope is \( \pm 1.87/2\pi \) [6]. The field-sweep spectra observed at frequencies between 26 and 34 MHz show a slope revealing that the internal field is directed perpendicular to the c-axis of the crystal. This means that in the proposed structure B of figure 1 the magnetic moments are perpendicular to the c-axis as has been deduced for the structure A from neutron scattering.

Any transition between the two structures should lead to a temperature dependence in the shape of the spectrum. For two samples with \( y = 6.15 \) showing the doublet and single line spectrum, respectively, no change has been found up to 50 K. However, the temperature dependence of the echo intensity observed at a delay time of 50 \( \mu \)s between the two rf pulses is different for both spectra (Fig. 2). In the doublet the intensity is exactly proportional to \( 1/T \) as expected if all parameters (relaxation times and number of nuclei contributing to the signal) are constant. For the single line, in contrast, the \( 1/T \) behaviour is found only for \( T > 40 \) K (Fig. 2B). At lower temperature there is a minimum at about 15 K. This temperature dependence proves that the spin-spin relaxation rate \( 1/T_2 \) depends on the temperature.

No exact measurements of \( T_2 \) have been performed. But for the doublet holds \( T_2 > 0.8 \) ms at \( 4.2 < T < 100 \) K. For the single line \( T_2 \) changes from about 0.3 ms at 4.2 K to less than 0.1 ms at 15 K and increases to more than 0.5 ms at 50 K. A similar, even stronger temperature dependence has been reported for the 30 MHz line in a sample with \( y = 6.3 \) [7].

A maximum of the relaxation rate \( 1/T_2 \) can be explained by a modulation of the resonance frequency by a fluctuating magnetic field with a frequency of the same magnitude as the amplitude of the modulation. Since the magnetic field from one nearest Cu-2 neighbour at a Cu-1 site results in a frequency shift of about 1 MHz, a maximum in \( 1/T_2 \) is expected if the neighbouring Cu-2 planes are fluctuating with a rate of about 1 MHz one against the other. Thus, there exists a strong temperature dependence of the fluctuation rate in the structure A with a correlation time of about \( 10^{-6} \) s at 15 K. Since the relaxation rate in the doublet is smaller without any maximum, the coupling between neighbouring Cu-2 planes must be much stronger in the structure B allowing no low frequency fluctuations.

Note added in proof:

Chemical analysis of the sample prepared from the pure \( ^{63} \)Cu isotope showing the spectrum of figure 2A has proved a contamination by 0.8 % Fe (relatively to Cu). Thus the stable ferromagnetic coupling between the CuO\(_2\) planes is most probably induced by a small iron content.

[6] We are indebted to H. Riesemeier, Berlin, for calculating the field-sweep spectra.