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^{63}Cu NQR Study of the Anisotropy in $YBa_2Cu_3O_7$

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We have studied the room temperature anisotropy in the high T_c superconductor $YBa_2Cu_3O_7$ by means of the ^{63}Cu nuclear quadrupole resonance (NQR). For the magnetically oriented powder samples, NQR signals were obtained only when the RF magnetic field is applied perpendicular to the direction of the crystalline c-axis. Significant differences in the spin-lattice relaxation times (T_1) and the lineshapes were observed between the unoriented powder sample and the magnetically oriented sample.

I. INTRODUCTION

Since the discovery of the high T_c superconductors, it has been noticed that the two-dimensionality and anisotropy in these materials play an important role in the high T_c superconductivity mechanism and practical applications. Consequently a number of measurements of the anisotropy in the high T_c superconductors have been made[1]-[5].

Since NQR is a sensitive probe of the electric field gradient (EFG) in the crystals, it is widely used to study the microscopic environments in many solids. Thus NQR has been employed to study the dynamics and local environments of the high T_c superconductors[6]-[9]. Anisotropy of the high T_c superconductors reflected in the distribution of the EFG can also be determined by NQR.

Single crystals of the high T_c superconductors large enough to study the anisotropy are difficult to grow. Powder samples cannot be used to study the anisotropy since they consist of microcrystals with random orientations and thus give the average properties of all the directions. For the YBCO superconductors, the lattice parameters a and b are nearly identical and are much shorter than c, which is the direction of the principal axis of the EFG[2], [13]. It is known that in high magnetic fields the powder samples of the YBCO superconductors are magnetically oriented in the c-direction[1], [3], [5], [10], [11]. These magnetically oriented samples can be used to study the anisotropy instead of the single crystals. It is the purpose of this paper to study the room temperature anisotropy of the $YBa_2Cu_3O_7$ superconductors using the magnetically oriented samples by means of ^{63}Cu(2) NQR.

The Hamiltonian for the interaction of the quadrupole moment of a nucleus with the field gradient at its position due to surrounding charges are[12]

$$H_Q = \frac{e^2QV_{zz}}{4I(2I-1)}\left[3I_z^2 - I(I+1) + \frac{1}{2}\eta(I_+^2 + I_-^2)\right], \qquad (1)$$

where $I_+ = I_x + iI_y$ and $I_- = I_x - iI_y$. Here, η is the anisotropy factor given by

$$\eta = (V_{yy} - V_{xx})/V_{zz}, \qquad (2)$$

where V_{yy}, V_{xx} and V_{zz} are the principal moments of the EFG tensor with

$$V_{yy} \le V_{xx} \le V_{zz}. \qquad (3)$$

For $I = 3/2$, the quadrupole energy levels are

$$E(\pm 3/2) = \frac{3e^2qQ}{4I(2I-1)}\left(1+\frac{\eta^2}{3}\right)^{1/2}, \qquad (4)$$

$$E(\pm 1/2) = -\frac{3e^2qQ}{4I(2I-1)}\left(1+\frac{\eta^2}{3}\right)^{1/2}. \qquad (5)$$

Thus there is only one transition frequency for spin $I = 3/2$,

$$\nu_Q = \frac{E_m - E_{m'}}{h} = \frac{e^2qQ}{2h}\left(1+\frac{\eta^2}{3}\right)^{1/2}. \qquad (6)$$

The two Cu sites in the $YBa_2Cu_3O_7$, so called the Cu(1) chain sites and the Cu(2) plane sites, correspond to NQR lines around 22.0 MHz and 31.21 MHz, respectively[8], [9], [11], [13]. In this work, we performed measurements on the Cu(2) plane sites, where antiferromagnetic correlation is known to exist[7]-[9], [13].

Ⅱ. EXPERIMENTAL

Powder samples of $YBa_2Cu_3O_{7-x}$ were made by the standard solid state reaction and annealed in an O_2 flow for maximum superconductivity, and cheked by the x-ray diffraction and magnetic susceptibility measurements[14]. Then they were mixed with a commercial epoxy (Duro TM-51) in cylindrical sample cells and cured in a strong magnetic field of 4.7 T for 30 min. They were placed in the superconducting magnet with the cylindrical axis either parallel or perpendicular to the magnetic field. The RF magnetic field from the sample coil for the NQR measurements is applied along the cylindrical axis.

The room temperature NQR measurements of the lineshapes and the spin-lattice relaxation times (T_1) for the unoriented and oriented samples were made using a home-built pulsed NQR spectrometer. Because of the broad lineshapes, solid echo sequences had to be used instead of the free-induction decay (FID) signals, using a 90° pulse of about 3.5 μs.

Ⅲ. RESULTS AND DISCUSSION

The ^{63}Cu NQR signals from the Cu(2) plane sites from the $YBa_2Cu_3O_7$ samples were obtained around 31.21 MHz. However, NQR signals were not observed from the samples in which the crystalline c-axis is aligned along the cylindrical axis of the sample cell. This in fact is an expected result since in this case the RF magnetic field is applied along the major principal axis of the EFG tensor, which coincides with the crystalline c-axis[2].

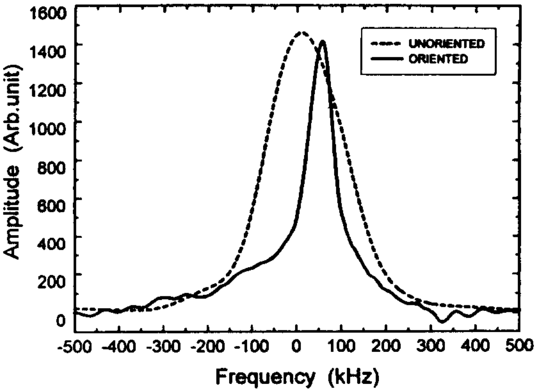

Fig. 1. Room temperture ^{63}Cu NQR $YBa_2Cu_3O_7$ lineshapes for the unoriented powder sample (– – –)and magnetically oriented sample (—) at the RF frequency of 31.21 MHz.

For comparison, Fig. 1 shows the lineshape taken at the RF frequency of 31.21 MHz of the unoriented sample and that of the oriented sample in which the RF field is applied perpendicular to the c-axis and along the ab-plane of the microcrystals. Although the bandwidth of the spectrometer is not broad enough to cover the wings of the lineshapes, comparison of the linewidths of the two samples can be safely made. In Fig. 1, the much smaller linewidth of the oriented sample is compared with that of the unoriented sample. This much narrower lineshape and the frequency shift of the magnetically oriented sample, together with the absence of the NQR signals from the samples in which the crystalline c-axis is aligned along the cylindrical axis, are taken as an evidence that the magnetic orientation is nearly perfect in our aligned samples. This indicates that the lineshape of the $YBa_2Cu_3O_7$ superconductor shows anisotropy and that the magnetic field of 4.7 T is high enough for the magnetic orientation in our samples. The much narrower linewidth and noticeable frequency shift for the magnetically oriented sample also indicates that magnetic order in $YBa_2Cu_3O_7$ plays an important role in the ^{63}Cu NQR.

The room temperature spin-lattice relaxation times were measured for the unoriented and oriented samples. The relaxation data are shown in Fig. 2. Single-exponential fit was possible for the spin-lattice relaxations. The room temperature T_1 for the unoriented sample, 280 μs, is in good agreement with previous reports[15]. However, the aligned sample gives a longer T_1 of 380 μs for the spin-lattice relaxation time in the direction of the c-axis. Since the T_1 for the unoriented sample is the average of the relaxation times for random orientations, this means that the relaxation time in the direction of the c-axis is longer than that in the ab-plane. The orientation-dependent spin-lattice relaxation rates $(1/T_1)$ have been reported around and below the superconducting transition temperature (T_c) in $YBa_2Cu_3O_7$[1].

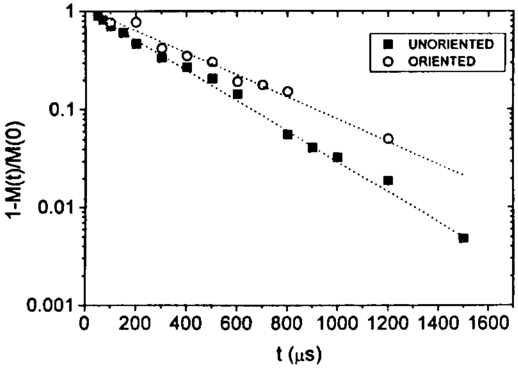

Fig. 2. Room temperature spin-lattice relaxations of the unoriented powder sample (■) and magnetically oriented sample (○) at the RF frequency of 31.21 MHz.

The orientation dependence of the relaxation rate for the Cu(2) is understood as arising from the hyperfine interactions between the electronic spins and the nuclear spins, and from the antiferromagnetic correlations of the electronic spins. The orientation-dependent spin-lattice relaxation in the normal state of $YBa_2Cu_3O_7$ has been explained by Millis, Monien and Pines (MMP)[16]. They derive the spin-lattice relaxation rates in the c-direction and in the ab-plane as

$$W_{/\!/} = \frac{k_B T}{\hbar} \frac{2K^2}{(1+a)^2\triangle} [1.51(1-\alpha)^2\beta(\zeta/a)^2 - 1.51(1-a)\beta\ln(\zeta/a) - (0.15-0.90\alpha+0.13\alpha^2)\beta + 4.71 + 18.9\alpha^2], \quad (7.\ a)$$

$$W_{\perp} = 1/2W_{/\!/} + \frac{k_B T}{\hbar} \frac{2K^2}{(1+a)^2\triangle} [3.02\beta(\zeta/a)^2 - 1.51\beta\ln(\zeta/a) - (0.60\beta + 11.78], \quad (7.\ b)$$

with $\beta = (a/\zeta)^4(\overline{\chi}/\chi)(\Gamma/\overline{\Gamma})$ and $\triangle = \chi_0\hbar\Gamma/\mu_B^2$. Here Γ is the characteristic energy of spin fluctuation in interacting spin system, $\overline{\Gamma}$ is the characteristic energy of spin fluctuation in noninteracting spin system, ζ is the spin correlation length and is the lattice constant. Thus the different spin-lattice relaxation times in the magnetically oriented and unoriented samples of $YBa_2Cu_3O_7$ as observed in our work can be theoretically explained.

Ⅳ. CONCLUSION

We have measured the lineshapes and the spin-lattice relaxation times of the magnetically oriented samples of $YBa_2Cu_3O_7$ and compared them with those of the unoriented powder samples. As a result, room temperature anisotropy due to the magnetic ordering could be observed.

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$YBa_2Cu_3O_7$의 비등방성에 관한 ^{63}Cu 핵사중극공명 연구

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^{63}Cu 핵사중극공명을 이용하여 자기적으로 정렬된 $YBa_2Cu_3O_7$ 고온초전도체 시료의 비등방성을 조사하였다. 정렬된 시료에서는 RF 자기장이 *c*-축에 수직으로 가해질 때만 공명신호가 검출되었다. 자기적으로 정렬된 시료와 정렬되지 않은 시료로부터 측정된 공명차이로부터 $YBa_2Cu_3O_7$의 자기적 비등방성을 관측할 수 있었다.