Internal Magnetic Field at Ba Site in Antiferromagnetic $YBa_2Cu_3O_6$ Determined by ^{137}Ba NQR *

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By measuring the beat frequency of the spin-echo amplitude modulation of pulsed ^{137}Ba NQR, the internal magnetic field of 14.8 mT perpendicular to the tetragonal c-axis at the Ba site in the antiferromagnetic $YBa_2Cu_3O_6$ at 80 K was determined. Calculation of the dipole field at the Ba site due to localized antiferromagnetically ordered 0.66 μ_B magnetic moments at Cu(2) sites yields 47 mT.

Key words: Nuclear quadrupole resonance, Antiferromagnetic materials, High-T_c superconductors, Spin-echo.

$YBa_2Cu_3O_6$ is the stoichiometric parent compound of the $YBa_2Cu_3O_{6+x}$ $(x>0.4)$ high-temperature superconducting family, the structural details of which can be found, e.g. in [1]. $YBa_2Cu_3O_6$ has a tetragonal crystal structure and is an antiferromagnetic insulator with Néel temperature $T_N \sim 415$ K. As reported from neutron diffraction measurements [2], the localized magnetic moment of 0.66 μ_B associated with the copper $3d$-hole is at the plane Cu(2) sites, and the magnetic structure consists of antiferromagnetically ordered Cu(2) planes that are themselves antiferromagnetically stacked along the tetragonal c-axis. The staggered magnetization lies parallel to the planes. The antiferromagnetic in-plane coupling is much stronger than the interplane coupling, giving a pronounced 2D antiferromagnetic character to $YBa_2Cu_3O_6$. The antiferromagnetically ordered state and its dynamics were studied, besides by neutron scattering, also by Cu quadrupole-perturbed internal field NMR [3–5]. The resonance experiments were limited to very low temperatures far away from T_N because of extremely short Cu spin-spin and spin-lattice relaxation times. In this contribution we report the first measurements of the internal magnetic field at the Ba site generated by Cu(2) electronic magnetic moments above liquid nitrogen temperature.

The polycrystalline $YBa_2Cu_3O_6$ compound was prepared by deoxidation of uniform large-grain-size

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$YBa_2Cu_3O_7$ material in 0.1 mbar vacuum at 800 °C for one hour. From the measured oxygen weight loss we determined the oxygen content in the unit formula to be 6.05 ± 0.05. The lattice constants $a = 3.856 \pm 0.004$ Å and $c = 11.813 \pm 0.01$ Å, measured by x-ray powder diffraction, confirm this oxygen content. For the sample of 6 mm diameter and 15 mm length we used unoriented $YBa_2Cu_3O_6$ powder.

^{137}Ba has spin 3/2, a natural abundance of 11.3%, a gyromagnetic ratio $\gamma = 2.9729107 \cdot 10^7$ rad $(Ts)^{-1}$, and a quadrupolar moment $Q = 0.28(3) \cdot 10^{-28}$ m². To detect the rather broad natural abundant ^{137}Ba NQR in $YBa_2Cu_3O_6$ we used the pulsed NQR spin-echo technique, sweeping the spectrometer frequency in discrete steps of 50 kHz (Figure 1). To improve the
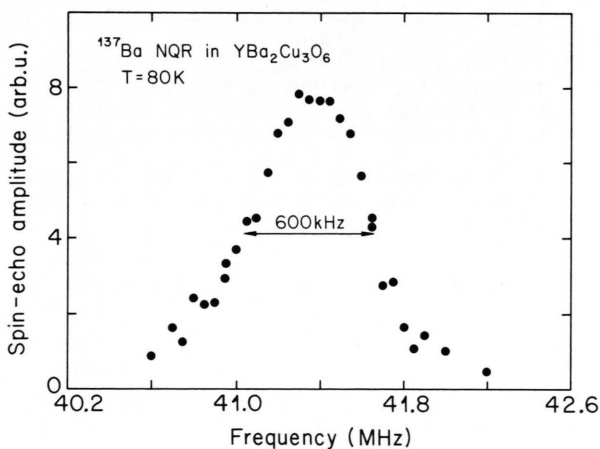

Fig. 1. ^{137}Ba NQR line obtained in a point-wise fashion by sweeping the spectrometer frequency in steps of 50 kHz.

S/N we accumulated the echos employing the following add-subtract phase alternating pulse sequence:

$\pi/2$ pulse (0° phase) $-\tau-\pi$ pulse (0° phase) $-\tau-$ echo, Add,

$\pi/2$ pulse (0° phase) $-\tau-\pi$ pulse (180° phase)$-\tau-$ echo, Add,

$\pi/2$ pulse (180° phase)$-\tau-\pi$ pulse (0° phase) $-\tau-$ echo, Subtract,

$\pi/2$ pulse (180° phase)$-\tau-\pi$ pulse (180° phase)$-\tau-$ echo, Subtract.

From single crystal NQR it is well known that the presence of a small static magnetic field shows up as a slow beat modulation in the echo envelope, which is obtained by recording the peak value of the NQR-echo as a function of the delay time τ between pulses [6, 7]. This also holds true for our polycrystalline YBa$_2$Cu$_3$O$_6$ sample, because in each crystallite the internal magnetic field should have the same orientation with respect to the principal EFG-tensor axes at the Ba site. In this way it was possible to detect the 70 kHz beat frequency of the 600 kHz broad ^{137}Ba NQR line with $v_Q = 41.40$ MHz ($\eta = 0$ due to the symmetry) at $T = 80$ K (Figure 2). For a single sharp value of the internal field an envelope modulation depth of 100% is expected theoretically. Our measurements, however, show a strongly reduced modulation depth, which is attributed to a distribution of field values. This distribution is most probably caused by imperfections of the crystal structure originating from residual oxygen which was not removed during the deoxidation process.
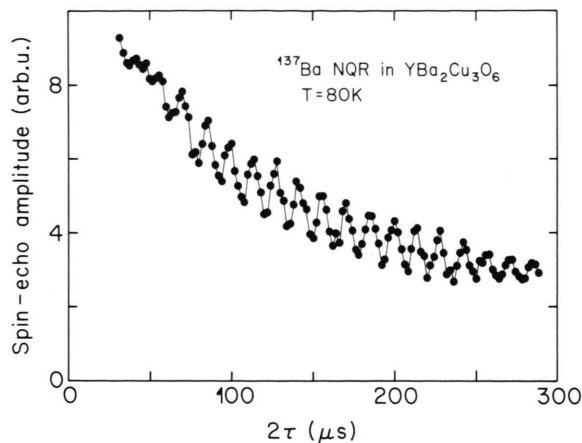

Fig. 2. NQR spin-echo envelope recorded by incrementing τ in steps of 1 μs.

From the beat frequency we determined an internal magnetic field of 14.8 mT perpendicular to the tetragonal c-axis at the Ba site. Calculations of the dipole field by assuming localized 0.66 μ_B moments ordered into the YBa$_2$Cu$_3$O$_6$ magnetic structure gave a much larger magnetic field of 47 mT. Copper-oxygen covalency in effect extending the magnetic moment might be responsible for this obvious discrepancy.

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