Spin-lattice relaxation of ^{63}Cu in $YBa_2Cu_3O_{7-y}$

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The temperature dependence of the spin-lattice relaxation time of ^{63}Cu nuclei at the frequencies of the nuclear quadrupole resonance of the crystallographically nonequivalent positions of the copper atoms, Cu1 (22 MHz) and Cu2 (31.5 MHz), in the normal state and superconducting state of the compound $YBa_2Cu_3O_{7-y}$ ($y = 0.05$ and 0.24, $T_c = 93$ K and 56 K) is presented.

The temperature dependence of the spin-lattice relaxation time T_1 of ^{63}Cu nuclei in the compound $YBa_2Cu_3O_{7-y}$ ($T_c = 87$ K) was studied experimentally in Ref. 1. The measurements were carried out in the mixed state ($B_0 = 2T$, $\nu = 22.3$ MHz). The exponential dependence of T_1 (T_c/T) was used to find the value $2\Delta/kT_c = 8.0(5)$, where Δ is the energy gap. The states of the copper atoms responsible for such a large value of Δ could not, however, be determined unambiguously.

In the structure of the $YBa_2Cu_3O_{7-y}$ compound the copper atoms occupy two crystallographically nonequivalent positions: Cu1, the Cu–O chains between the planes of Ba atoms, and Cu2, the Cu–O planes which surround the layer of Y atoms. The difference in the electric field gradients of Cu nuclei allows us to selectively study the behavior of the spin-lattice relaxation at the frequencies of the nuclear quadrupole resonance (ν_Q) of the corresponding nonequivalent positions of the copper atoms.

Figure 1 shows the nuclear quadrupole resonance (NQR) spectra of ^{63}Cu and ^{65}Cu in $YBa_2Cu_3O_{7-y}$ samples with various oxygen concentrations in the unit cell [$y = 0.05(1)$, $T_c(\Delta T_c) = 93(3)$ K; $y = 0.24(2)$, $T_c(\Delta T_c) = 56(6)$ K; $y = 1.00(5)$]. The oxygen content was determined from the data on neutron-diffraction analysis and thermogravimetric analysis. The spectra were obtained by using the

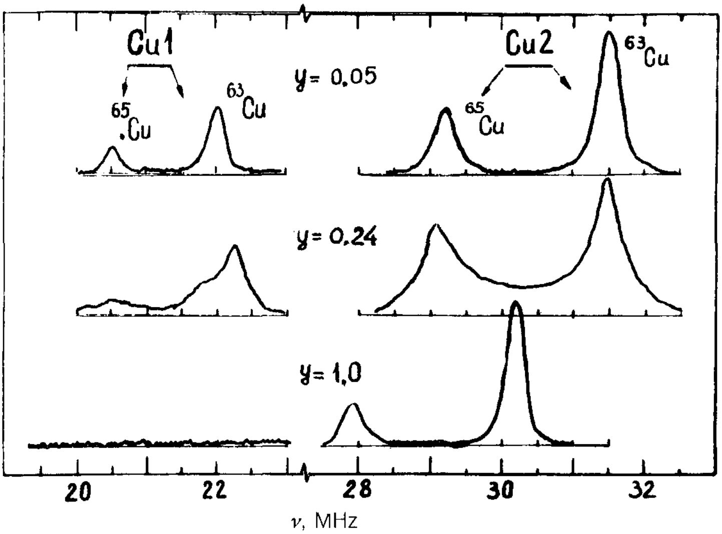

FIG. 1. Nuclear quadrupole resonance spectra of ^{63}Cu and ^{65}Cu at $T = 78$ K in $YBa_2Cu_3O_{7-y}$ samples; $y = 0.05$ ($T_c = 93$ K), 0.24 (56 K), and 1.0.

spin-echo method with an ISSh-3-12 NQR spectrometer. The temperature dependence $\nu_Q(T)$ for $y = 0.05$ is the same as that in Ref. 2. All the lines are broadened in the $YBa_2Cu_3O_{6.76}$ sample, suggesting that the short-range-order disruptions increase in the nearest-neighborhood of the copper atoms. The NQR lines corresponding to the crystallographic positions were identified by comparing the ratio of the resonance-line intensities, with allowance for the frequency dispersion of the transmission coefficient of the spectrometer's receiving circuit, and the difference in the spin-spin relaxation times T_2. The NQR signal of ^{63}Cu($\nu_Q = 22$ MHz) is related to the Cu1 positions and the NQR signal corresponding to $\nu_Q = 31.5$ MHz is related to the Cu2 positions, in agreement with the conclusions of Refs. 2 and 3. An additional confirmation is the change of T_2(^{63}Cu) on switching to $HoBa_2Cu_3O_{7-y}$ ($T_c = 94$ K), where the presence of a magnetic rare-earth ion contributes significantly to the spin-spin relaxation of the copper nuclei with $\nu_Q = 31.5$ MHz. For $y = 1.0$ a second pair of lines (the Cu1 positions) could not be seen in the frequency interval 18–26 MHz.

The spin-lattice relaxation time T_1 was measured on the basis of the reconstruction of the amplitude of the quadrupole spin echo $A(t)$ after the saturating series of rf pulses ($H_1 \approx 100$ Oe, t is the time interval between the saturating series of pulses and a pair of pulses that forms the echo). In the normal state we have $A(t) \sim \exp - t/T_1$, with the exception of a sample with $y = 0.24$, where we saw a slight nonexponential dependence which was apparently attributable to the considerable NQR linewidth of this sample. A comparison of T_1 for the ^{63}Cu and ^{65}Cu isotopes in the temperature region $T_c/2 < T < 2T_c$ leads to the conclusion that the spin-lattice relaxation of copper nuclei is determined primarily by the fluctuating part of the magnetic hyperfine interaction of the nucleus with the electronic surroundings: $T_1(^{63}\mathrm{Cu})/T_1(^{63}\mathrm{Cu}) = 1.15(5)$

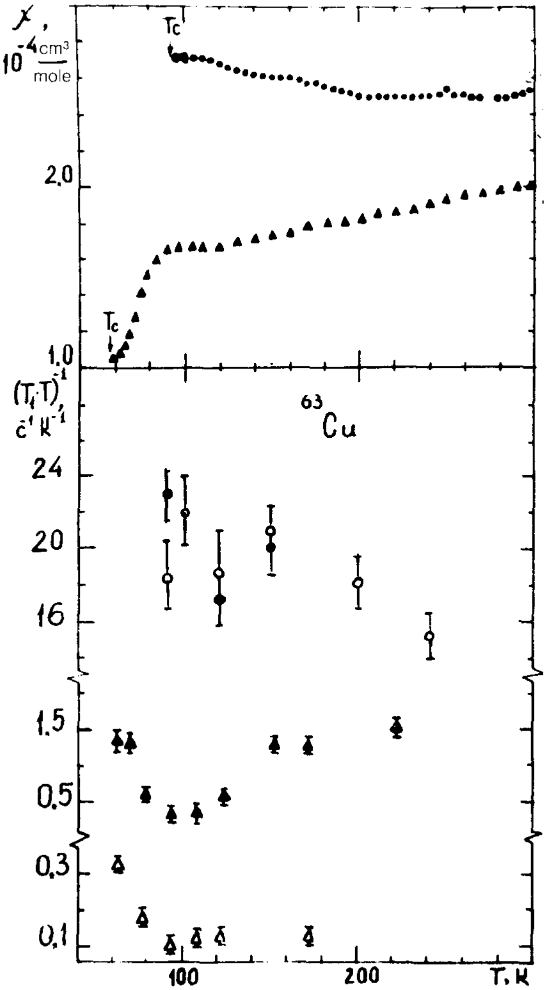

FIG. 2. Temperature dependence of the magnetic susceptibility in the normal state (a) (●—$y = 0.05$, ▲—$y = 0.24$) and of the quantity $(T_1T)^{-1}$ of ^{63}Cu nuclei (b) ($y = 0.05$, ●—22 MHz (Cu1), O—31.5 MHz (Cu2); $y = 0.24$, ▲—22.1 MHz (Cu1), △—31.3 MHz (Cu2).

for the Cu1 positions and $T_1(^{63}\mathrm{Cu})/T_1(^{65}\mathrm{Cu}) = 1.00(5)$ for the Cu2 positions. This result differs from the results of Ref. 3, where it was concluded for Cu1 positions that the quadrupole relaxation mechanism is the dominant mechanism in the temperature interval indicated above. At $T > T_c$ the value $(T_1T)^{-1}$ with $y = 0.05$ increases with decreasing temperature (Fig. 2), in agreement with the results of Refs. 3 and 4 for $\nu_Q = 31.5$ MHz. With an increase in y, the quantity $(T_1T)^{-1} \sim \langle N^2(E_F)\rangle_T$ decreases sharply apparently because of the decrease of the density of states at the Fermi level, $N(E_F)$. With an increase in the oxygen deficiency, the magnetic susceptibility χ decreases, taking on the value $\chi_0 = 0.80 \times 10^{-4}$ cm^2/mole for $y = 1.0$, where χ_0 is the temperature-independent component used in the data analysis, $\chi(T) = \chi_0 + C/(T - \theta)$. Assuming that the spin component of $YBa_2Cu_3O_{6.0}$ is close to zero, $\chi_{\mathrm{spin}} = 2\mu_B^2\langle N(E_F)\rangle_T$, and that the components which differ from the Pauli compo-

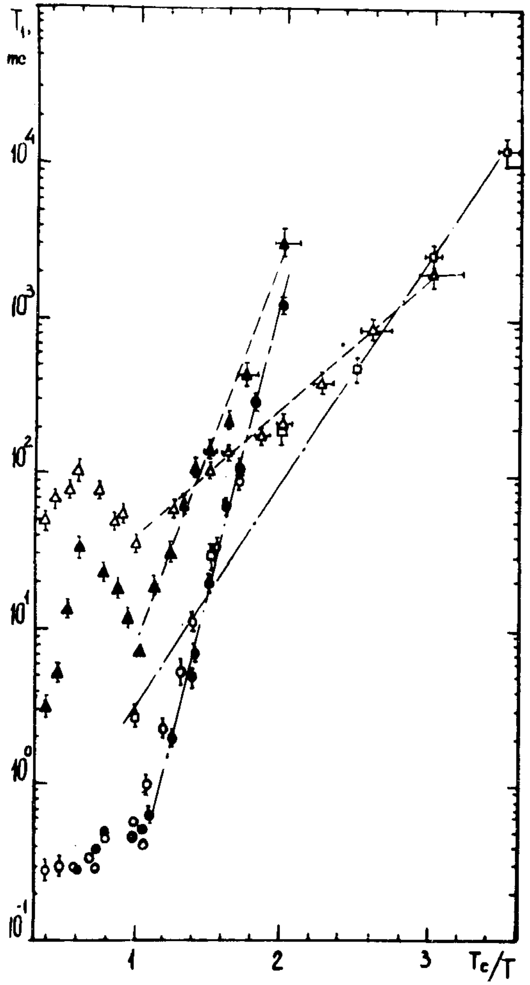

FIG. 3. Evolution of the spin-lattice relaxation time T_1 of ^{63}Cu nuclei in the superconducting state for $YBa_2Cu_3O_{7-y}$; $y = 0.05$ ($T_c = 93$ K) ●—22.05 MHz, ○—31.5 MHz; $y = 0.24$ ($T_c = 56$ K) ▲—22.1 MHz, △—31.3 MHz ($T_c = 87$ K) □—31.5 MHz.

nent do not depend of y, we can postulate that the electron state density at the Fermi level decreases by at least a factor of two as we switch to the $YBa_2Cu_3O_{6.76}$ compound.

We found that when all the samples are in the superconducting state, the curve for the reconstruction of the amplitude of the echo signal, $A(t)$, is not an exponential curve. This behavior is attributable, in our view, to the spatial distribution of Δ from zero to the maximum value as it moves from the surface of the sample into its interior (the average size of powder particles is 5 μm). Figure 3 shows the values of T_1 corresponding to the flattest part of the $A(t)$ curve. In the case of $YBa_2Cu_3O_{6.95}$ the ratio of the width of the energy gap to the critical temperature is approximately the same for the two positions: for the Cu1 position we have $2\Delta/kT_c = 14(2)$ and for the Cu2 position we have $2\Delta/kT_c = 12(2)$, which is higher than the value $2\Delta/kT_c = 7.5(5)$ obtained for the Cu2 positions of a sample with $T_c = 87$ K, which was studied previously.[1] With an increase in y, the slope of the $T_1(T_c/T)$ curve decreases sharply and for the $YBa_2Cu_3O_{6.76}$ sample we have $2\Delta/kT_c = 9(1)$ for the Cu1 position and $2\Delta/kT_c = 3.7(5)$ for the Cu2 position. These results suggest that the pairing mechanism weakens appreciably with increasing oxygen deficiency in the $YBa_2Cu_3O_{7-y}$ compound.

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