

# Anomalies on temperature dependence of nuclear relaxation of $^{63}\text{Cu}$ nuclei in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$

A. V. Bondar', S. M. Ryabchenko, Yu. V. Fedotov, and A. A. Motuz  
*Institute of Physics, Academy of Sciences of the Ukrainian SSR*

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Structural features have been found on the temperature dependence of the spin-spin and spin-lattice relaxation times of  $^{63}\text{Cu}$  nuclei in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  at 35 K. These results, combined with previous results, suggest either a second-order phase transition or a restructuring of the dynamic state of intrinsic defects in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  at 35 K.

The temperature dependence of the spin-lattice relaxation of copper nuclei in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  has been the subject of many studies (e.g., Refs. 1 and 2). The temperature dependence of the spin-spin relaxation in this compound has received far less attention, although studies in this direction might provide important information about the dynamics of the system of magnetic moments associated with copper ions. Such a study seems extremely timely in view of the suggested possibility of a magnetic mechanism for pairing in the high- $T_c$  superconductors.<sup>3</sup>

We have used the nuclear-quadrupole-resonance (NQR) method for a detailed study of the temperature dependence of the spin-spin relaxation time  $T_2$  and the spin-lattice relaxation  $T_1$  of  $^{63}\text{Cu}$  nuclei in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  samples synthesized by a ceramic procedure. The time  $T_2$ , which is a measure of the homogeneous NQR linewidth, is found from the dependence of the spin-echo amplitude on the interval between exciting pulses in the Hahn method. Within the error of our measurements, this dependence is described by a single exponential function. For the test samples,  $T_c$  was 91 K. Bakharev *et al.*<sup>4</sup> studied the temperature dependence  $T_2 = T_2(T)$  over a wide temperature range and reached the conclusion that, after the uniform NQR linewidth of copper nuclei in Cu(2) planes decreases significantly as  $T_c$  is crossed, a further lowering of the temperature causes essentially no change in this width. Our measurements show, however, that the value of  $T_2$  for these nuclei changes significantly in the temperature range 60–10 K and that, furthermore, the dependence  $T_2 = T_2[T]$  is not monotonic in this region (Fig. 1). The nature of the well-defined anomaly on the  $T_2(T)$  curve near 35 K is not clear at this point. This anomaly is apparently associated with changes in certain fundamental characteristics of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  in this temperature range, since it is near 35 K that anomalies have been observed in many other physical properties of this compound. Specifically, anomalies have been observed in the thermal expansion, the elastic modulus,<sup>5</sup> and the attenuation of sound<sup>6</sup>; a sharp change has been observed in a torque.<sup>7</sup> One possible reason for these anomalies might be a second-order phase transition accompanied by a significant change in the correlation time  $\tau_c$  of the fluctuating local fields at copper nuclei. Under conditions such that the decay kinetics of the spin-echo signal is determined spectral diffusion processes, the time  $T_2$  may go through a minimum as the temperature is varied.<sup>8</sup> The position of

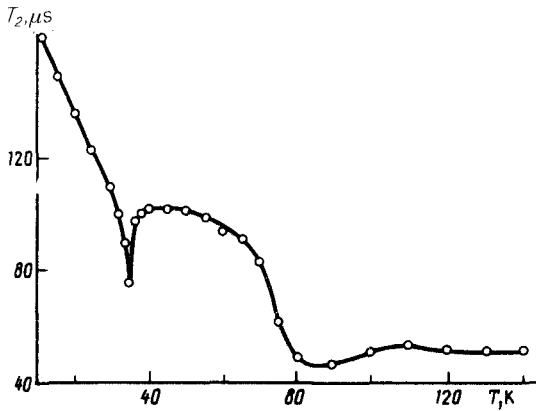


FIG. 1. Temperature dependence  $T_2(T)$  of  $^{63}\text{Cu}$  nuclei in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  planes.

this minimum would be determined by the condition  $1/\tau_C \approx \Delta\omega_0$ , where  $\Delta\omega_0$  is the inhomogeneous NQR linewidth. A sharp change in the nature of the temperature dependence  $T_2(T)$  as the point 35 K is crossed might also be a consequence of a phase transition at this temperature.

Note that the NQR spectrum does not exhibit the substantial changes near 35 K which would usually accompany phase transitions. Furthermore, there is no sharply defined anomaly in the temperature dependence of the spin-lattice relaxation time,  $T_1(T)$ . One might nevertheless note that certain changes do occur in the nature of this dependence near 35 K for Cu(2) nuclei (nuclei in planes). Previous research<sup>1,2</sup> has shown that in a certain temperature interval whose upper limit is  $T_C$  these nuclei exhibit a  $1/T_1 \sim T^n$  behavior with  $n \approx 4$ . A behavior of this type is characteristic of heavy-fermion compounds<sup>9</sup> and can be explained in terms of the existence of a point or line of zero values of the energy gap for pairing carriers. Figure 2 shows our results on  $T_1 = T_1(T)$  in full logarithmic scale. We see that a power-law approximation works well down to specifically 35 K and that the behavior changes below this temperature.

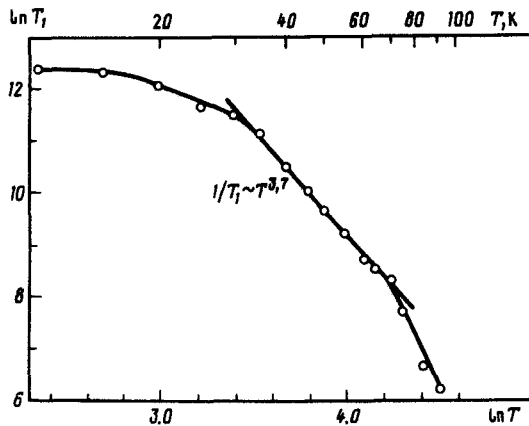


FIG. 2. Temperature dependence  $T_1(T)$  of  $^{63}\text{Cu}$  nuclei in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  planes.

The results found on both  $T_1(T)$  and  $T_2(T)$  are being reproduced with samples synthesized in various laboratories. Furthermore, a careful examination of the published data on  $T_2(T)$  suggests that the anomaly which we have observed has also been present in results found by other investigators, and at the same 35 K, but it has not been identified because of the wide spacing of experimental points along the temperature scale.

In summary, the features of the temperature dependence of the relaxation times [primarily  $T_2(T)$  but also  $T_1(T)$  to some extent] described above suggest that  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  undergoes definite changes at 35 K, possibly even a second-order phase transition. An alternative to this phase transition as a cause of the narrow peak in  $T_2(T)$  might be a temperature dependence of the correlation time for mobile (reorienting) defects which modulate the NQR frequency of neighboring nuclei, similar to that which has been observed in bismuth silicate and germanate crystals on the basis of NQR data.<sup>10</sup> In this case one would have to assume that the defects responsible for the anomaly in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  are intrinsic defects.

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<sup>2</sup>T. Imai *et al.*, J. Phys. Soc. Jpn. **57**, 1771 (1988).

<sup>3</sup>I. E. Dzyaloshinskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 518 (1989) [JETP Lett. **49**, 599 (1989)].

<sup>4</sup>O. N. Bakharev *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **47**, 383 (1988) [JETP Lett. **47**, 458 (1988)].

<sup>5</sup>N. V. Anshukova *et al.*, Problems of High- $T_c$  Superconductivity, Vol. 2, Sverdlovsk, 1987, p. 182.

<sup>6</sup>I. M. Golev, Fiz. Tverd. Tela (Leningrad) **31**, 220 (1989) [Sov. Phys. Solid State **31**, 120 (1989)].

<sup>7</sup>S. M. Ashimov *et al.*, Sverkhprovodimost' **2**, No. 4, 49 (1989).

<sup>8</sup>K. M. Salikhov *et al.*, Electron Spin Echo and Its Applications, Nauka, SO, Novosibirsk, 1976.

<sup>9</sup>N. E. Alekseevskii and D. I. Khomskii, Usp. Fiz. Nauk **147**, 767 (1985) [Sov. Phys. Usp. **28**, 1136 (1985)].

<sup>10</sup>A. Yu. Kudzin *et al.*, Fiz. Tverd. Tela (Leningrad) **24**, 2682 (1982) [Sov. Phys. Solid State **24**, 1520 (1982)].

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