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PHASE DIAGRAM OF MAGNETIC ORDER AND SUPERCONDUCTIVITY IN HIGH-\(T_c\) \(\text{YBa}_2\text{Cu}_3\text{O}_x\)

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Abstract. – Nuclear quadrupole resonance (NQR) experiments on tetragonal \(\text{YBa}_2\text{Cu}_3\text{O}_x\) with \(x = 6.3\) and 6.4 reveal a successive magnetic transition associated with the chain site at 20 K well below \(T_N \sim 400\) K, while such transition has not been observed down to 1.3 K for \(x = 6.1\). The second transition appears in the vicinity of the superconductivity.

Since the discovery of high-\(T_c\) superconducting oxides, it was recognized that superconductivity contacts with antiferromagnetism (AF). In La-system, a three-dimensional (3D) AF order of undoped \(\text{La}_2\text{CuO}_4\) with \(T_N = 240\) K is markedly destroyed by 1% -Ba or Sr doping, while the magnetic phase with low-\(T_N\) less than 10 K continues gradually just before superconductivity appears around \(x = 0.025\) [1]. The characteristic magnetic phase existing between 3D-AF and superconductivity is of incoherent type along the successive \(\text{CuO}_2\) plane, whereas there exists long-range antiferromagnetic coherency within the \(\text{CuO}_2\) layers. No anomaly of the specific heat appears at \(T_N\) [2], whereas the nuclear relaxation rate \(1 / T_1\) diverges at \(T_N\) [3]. The obtained magnetic phase diagram has been well interpreted in terms of a model proposed by Aharony et al. [4], where a local ferromagnetic frustration among \(\text{Cu}\) spins is induced by \(\mu\) holes introduced on the oxygen sites.

In \(\text{YBa}_2\text{Cu}_3\text{O}_x\), \(\mu\)SR study found firstly that the oxygen-deficient compounds with \(x \sim 6.1\) undergo AF transition above room temperature [5]. Subsequently, neutron diffraction experiment on polycrystal sample with \(x = 6\) and 6.1 confirmed that AF order with \(T_N = 400\) K \(\sim 500\) K possesses the same type of spin structure as that of \(\text{La}_2\text{CuO}_4\) with almost same magnetic moment of 0.6 \(\mu_B\) [6]. From the \(\text{Cu}\) NQR study for \(x = 6.6\), it was found that an AF ordering takes place at 20 K [7]. This type of magnetic order with low-\(T_N\) has also been observed by NQR experiment [8]. On the other hand, recent neutron scattering experiments on single-crystals with \(x = 6.38\) [9] and 6.35 [10] verified that the compounds order antiferromagnetically with the higher \(T_N = 200\) K and 405 K, respectively. This inconsistency between both experiments may be, however, resolved by taking account of the following experimental result. Namely, the neutron scattering study on \(x = 6.35\) revealed a second transition at 40 K successively to the first one with \(T_N = 405\) K [10]. This second magnetic transition may be connected just with AF order with lower \(T_N\) found by NQR and \(\mu\)SR experiments. At present, it is highly desired to investigate separately the magnetic property of the chain and plane sites by NMR technique.

Recently we have observed Cu-NMR signals associated with \(\text{CuO}_2\) plane sites in addition to the NQR signal around 30 MHz [11]. So we can now conclude that NQR signal around 30 MHz observable from \(y = 0.6\) to 1 results from the \(\text{CuO}\) chain sites [7, 12, 13]. Accordingly, the magnetic transition at 20 K for \(x = 6.3\) mentioned above is decisively associated with the \(\text{CuO}\) chain sites.

In this paper, we present the magnetic phase diagram determined from the temperature dependences of NQR line-width and \(1 / T_1\) for the chain sites. \(\text{YBa}_2\text{Cu}_3\text{O}_x\) compounds with \(x = 6.4\) and 6.3 were prepared by quenching the sintered pellets from 900 °C and 950 °C, respectively and the compound with \(x = 6.1\) by annealing in argon atmosphere of 10^{-4} torr at 700 °C for 24 hours and by cooling rapidly to room temperature in the furnace.

In figure 1, we show the NQR spectra for \(x = 6.4\), 6.3 and 6.1. For \(x = 6.4\) and 6.3, we have observed two different types of signals around 23 and 30 MHz with each pair of the isotopes of \(^{63}\text{Cu}\) and \(^{65}\text{Cu}\), while NQR is observable only around 30 MHz for \(x = 6.1\). Since both neighboring chain oxygens are almost vacant for \(x \sim 6.0\), NQR signal observed around 30 MHz for \(x = 6.1\) corresponds to the \(\text{Cu}\) chain site with two oxygen neighbors. On the other hand, the NQR of \(^{63}\text{Cu}\) at 24 MHz for \(x = 6.4\) and 6.3 may be assigned to the site with three oxygen neighbors because the chain site with four oxygens in orthorhombic phase were observed at 22.04 MHz [3].

As was already reported [7], the broad line-width of \(^{63}\text{Cu}\) NQR spectrum for \(x = 6.3\) decreases from 2 MHz to 0.8 MHz with increasing temperature and \(1 / T_1\) exhibits a divergence at 20 K. The same behavior has also been found around 20 K for \(x = 6.4\). Furthermore, the distinct line-splitting with a separation of 2 MHz was

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reported for \( x = 6.15 \) [13]. However, we have observed no line-broadening of the spectrum and no anomaly of \( 1/T_1 \) for \( x = 6.1 \) down to 1.3 K. Accordingly the successive magnetic transition seems to appear in the vicinity of superconductivity. Figure 2 shows the magnetic phase diagram for \( \text{YBa}_2\text{Cu}_3\text{O}_x \), determined by \( \mu \text{SR} \) (triangles) [8], neutron diffraction (circles) [9, 10] and NQR (rectangles). As seen in the figure, the value of \( T_N \) is scattered, suggesting that the serious problem on the identification of oxygen content still remains. Nevertheless, it is evident that the second magnetic transition takes place in \( \text{YBa}_2\text{Cu}_3\text{O}_x \) with \( x > 6.2 \). In characterising the magnetic character of second transition, it is worthwhile that the divergent behavior of \( 1/T_1 \) for \( \text{Y-system} \) is quite analogous to that found at the temperature where a spin-glass like phase appears between 3D-AF and superconducting state for \( \text{La-system} \) [1, 2]. If the concentration of holes increases effectively with increasing oxygen content and holes are almost localized at lower temperature, the strong exchange between hole and Cu spins yields a ferromagnetic frustration among Cu spins as pointed out by Aharony et al. [4]. Hence we may speculate that the successive transition corresponds to a sort of re-entrant spin glass phase. The canting of Cu magnetic moments among the successive CuO₂ layers induced by a frustration applies the inhomogeneous dipole field on the chain sites and then causes the line-broadening of the NQR spectrum. To elucidate the further detailed magnetic nature of second transition, the experiment on CuO₂ plane is now stimulating.