^{63}Cu-NMR Study in High-T_c Superconductor $HgBa_2Ca_2Cu_3O_{8+\delta}$ *

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The ^{63}Cu nuclear transverse relaxation rates have been measured in the normal state on both square and pyramidal CuO_2 planes in $HgBa_2Ca_2Cu_3O_{8+\delta}$ with $T_c = 133$ K. The Gaussian component of the spin-echo decay rate, $^{63}(1/T_{2G})$, for both sites increases with decreasing temperature, followed by a peak around $T^* \sim 150$ K, indicating that the spin correlation becomes stronger with decreasing temperature. Also, it is found that the magnitude of $^{63}(1/T_{2G})$ for a square site is larger than that for a pyramidal one, suggesting that the spin correlation in the square plane is stronger than that in the pyramidal one.

Key words: High-T_c cuprate, $HgBa_2Ca_2Cu_3O_{8+\delta}$, ^{63}Cu-NMR, T_{2G}, Spin correlation.

Introduction

The mechanism of high-T_c superconductivity is still not understood. In this respect it may be helpful to elucidate why T_c in the new mercury-based compound is so high. We used the NMR method, which can give information on the microscopic properties of the CuO_2 plane in high-T_c cuprates. Especially, the ^{63}Cu nuclear transverse relaxation rate, $^{63}(1/T_{2G})$, in the CuO_2 plane provides information on the electron spin-spin correlation through the indirect nuclear spin-spin coupling [1].

At present $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Hg-1223) has the highest known superconduction transition temperature ($T_c = 133$ K) [2, 3]. In this compound there are two crystallographic copper sites, one of which has a pyramidal (5-*fold*) oxygen surrounding, while the other has a square (4-*fold*) one. This provides a good opportunity to reveal the relation between T_c and the number and/or the type of CuO_2 planes.

In [4] we reported the T-dependent Knight shift, ^{63}K, and the nuclear-spin lattice relaxation rate, $^{63}(1/T_1T)$, which exhibits a Curie-Weiss like behavior, of ^{63}Cu in a magnetic field of ~ 11 T, using a well-aligned powder. The full-width at half-maximum (FWHM) of the ^{63}Cu-NMR spectra, which is almost T-independent in the normal state, was very narrow (~ 80 and ~ 130 Oe for the 4- and 5-*fold* site, respectively), assuring that the sample was of high quality.

In the present paper we report results of $^{63}(1/T_{2G})$ measurements for both sites in the normal state in a magnetic field of ~ 11 T.

1. Experimental

The Hg-1223 phase sample was prepared with the high-pressure synthesis technique described in [3]. By X-ray diffraction it was shown that the sample consisted of almost a single-phase [3]. T_c was found to be 133 K from the temperature where the diamagnetic signal appeared in the ac-susceptibility. The powder sample was aligned along the c-axis with an external magnetic field of 11 T and fixed with stycast 1266 epoxy.

The NMR measurements were performed in a conventional phase-coherent standard pulsed spectrometer, using a superconducting magnet (12 T at 4.2 K) to improve the signal to noise ratio. $^{63}(1/T_{2G})$ measurements were made by NMR for the central tansition $I_z(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ at 125.1 MHz in the T-range $130 \sim 300$ K under a magnetic field of ~ 11 T parallel to the c-axis.

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2. Results and Discussion

Figure 1 shows the ^{63}Cu nuclear spin-echo decay at 140 K for the pyramidal site plotted against t, where t is the time interval between the $\pi/2$ pulse and the spin-echo. The spin-echo decay (the solid line) curve was fitted to $M(t) = M_0 \exp\left[-\frac{1}{2}\left(\frac{t}{T_{2G}}\right)^2 - \frac{t}{T_{2L}}\right]$, where T_{2G}, a fitting parameter, is a Gaussian component due to the T_2 process, and T_{2L} is a Lorentzian component due to the T_1 process. $1/T_{2L}$ was determined by the next relation $1/T_{2L} = 3(1/T_1)_c + (1/T_1)_{ab}$ for NMR, where $(1/T_1)_\alpha$ is the spin-lattice relaxation rate for the external field along the α-direction.

In order to determine $1/T_{2G}$ accurately, the linewidth of the NMR spectrum must be small compared with the rf exciting field, because the nuclear spins are uniformly flipped by the exciting pulse. If the spin-excitation is incomplete, the spin-echo decay time becomes longer. The strength of the rf exciting pulse, H_1, was about ~ 80 and ~ 130 Oe for the 4- and 5-*fold* site, respectively, estimated from the width of rf pulse, which is the same as each FWHM of the ^{63}Cu-NMR spectra, and the fitting to the experimental points appears satisfactory, as seen in Figure 1. So, most spins may be flipped. In practice, we measured the H_1-dependence of $^{63}(1/T_{2G})$ in the normal state, and confirmed saturation for both sites.

Figure 2 shows the T-dependence of the Gaussian decay rates, $^{63}(1/T_{2G})$, for square (o) and pyramidal (•) copper sites, respectively. A remarkable feature is that $^{63}(1/T_{2G})$ for both sites increases with decreasing temperature, followed by a peak around $T^* \sim 150$ K, which is almost the same temperature where $^{63}(1/T_1T)$ shows a broad peak. This behavior is similar to that in $Tl_2Ba_2Ca_2Cu_3O_{10}$ [5], whose crystal in similar to that of Hg-1223 but different from that of lightly-doped $YBa_2Cu_3O_{6.63}$ [6] and $YBa_2Cu_4O_8$ [7], showing a spin-gap like behavior, where $^{63}(1/T_{2G})$ continuously increases until just above T_c, while $^{63}(1/T_1T)$ shows a broad peak near 150 K.

As shown by Thelen and Pines [8], in a strong correlated limit $^{63}(1/T_{2G})$ is expressed as

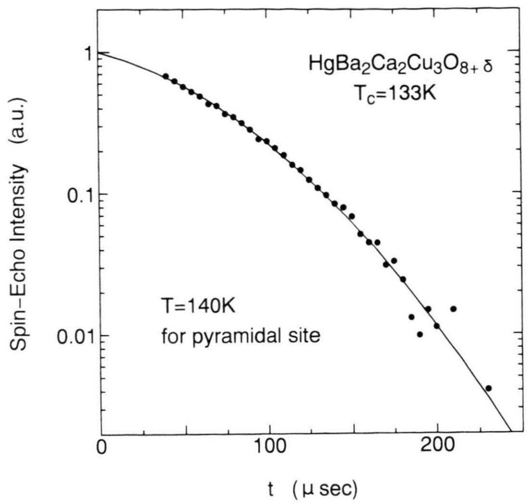

Fig. 1. ^{63}Cu nuclear spin-echo decay at 140 K for the pyramidal site, plotted against t, where t is the time interval between the $\pi/2$ pulse and the spin-echo. The solid line is the best fitting to $M(t) = M_0 \exp\left[-\frac{1}{2}\left(\frac{t}{T_{2G}}\right)^2 - \frac{t}{T_{2L}}\right]$.

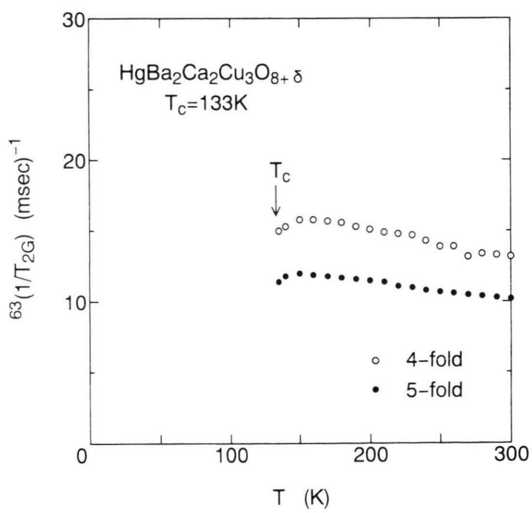

Fig. 2. T-dependence of $^{63}(1/T_{2G})$ in $HgBa_2Ca_2Cu_3O_{8+\delta}$ with $T_c = 133$ K for the square (o) and pyramidal (•) site.

$$^{63}(1/T_{2G}) \propto [\Sigma F(q)^4 \chi'(q,\omega=0)^2]^{1/2} \simeq F(Q)^2 \chi_Q/(\xi/a) = (A_c - 4B)^2 \beta^{1/2} \chi_s(\xi/a), \quad (1)$$

where $F(q)(= [A_c + 2B(\cos q_x a + \cos q_y a)])$ is a hyperfine form factor, $\chi'(q,\omega=0)$ the q-dependent real part of the electron spin susceptibility, and ξ an antiferromagnetic coherence length. Also, χ_Q is the static spin susceptibility at the antiferromagnetic wave vector $Q = (\pi/a, \pi/a)$, which is related to the static spin susceptibility at $q = 0$, $\chi_0(=\chi_s)$, by $\chi_Q = \chi_s(\xi/a)^2 \beta^{1/2}$, where β is assumed to be T-independent.

As seen in (1), the ξ's of both sites increase with decreasing temperature down to T^*, because the χ_s's of both sites decrease with decreasing temperature. It is suggested that the spin correlation becomes stronger with decreasing temperature. Also, it is found that $^{63}(1/T_{2G})$ for the 4-*fold* site is larger than that for the 5-*fold* one, suggesting that the spin correlation in the square plane is stronger than that in the pyramidal one, because the hyperfine field and χ_s for the 4-*fold* site is larger than that for the 5-*fold* one, estimated from the analysis of the Knight shift [4]. So it is expected that the magnetic property in the square CuO_2 plane is very important for the high T_c. This is in contrast to the electron-doped high-T_c compounds, such as the Nd-system, having only a square CuO_2 plane and showing a less high T_c.

3. Summary

We have measured the Gaussian component of the spin-echo decay rate, $^{63}(1/T_{2G})$, on both 4- and 5-*fold* CuO_2 sheets in $HgBa_2Ca_2Cu_3O_{8+\delta}$ with $T_c = 133$ K. In the normal state, $^{63}(1/T_{2G})$ for both sites increases with decreasing temperature followed by a peak around $T^* \sim 150$ K, demonstrating that the spin correlation becomes stronger with decreasing temperature. From a quantitative point of view, it is also found that the magnitude of $^{63}(1/T_{2G})$ for the 4-*fold* site is larger than that of the 5-*fold* one, suggesting that the spin correlation in the square plane is stronger than that in the pyramidal one, and that the magnetic property in 4-*fold* site may be a key for higher T_c.

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