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Author(s)	Fujiwara, Naoki; Fujimaki, Y.; Uchida, S.; Matsubayashi, K.; Matsumoto, T.; Uwatoko, Y.
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NMR and NQR study of pressure-induced superconductivity and the origin of critical-temperature enhancement in the spin-ladder cuprate $Sr_2Ca_{12}Cu_{24}O_{41}$

N. Fujiwara,[1,*] Y. Fujimaki,[2] S. Uchida,[2] K. Matsubayashi,[3] T. Matsumoto,[3,4] and Y. Uwatoko[3]
[1]*Graduate School of Human and Environmental Studies, Kyoto University, Yoshida-Nihonmatsu-cyo, Sakyo-ku, Kyoto 606-8501, Japan*
[2]*Department of Superconductivity, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[3]*Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan*
[4]*National Institute for Materials Science, Tsukuba 305-0047, Japan*

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Pressure-induced superconductivity was studied for a spin-ladder cuprate $Sr_2Ca_{12}Cu_{24}O_{41}$ using nuclear magnetic resonance under pressures up to the optimal pressure 3.8 GPa. Pressure application leads to a transitional change from a spin-gapped state to a Fermi-liquid state at temperatures higher than T_c. The relaxation rate $1/T_1$ shows activated-type behavior at an onset pressure, whereas Korringa-like behavior becomes predominant at the optimal pressure, suggesting that an increase in the density of states at the Fermi energy leads to enhancement of T_c. Nuclear quadrupole resonance spectra suggest that pressure application causes transfer of holes from the chain to the ladder sites. The transfer of holes increases DOS below the optimal pressure. A dome-shaped T_c versus pressure curve arises from naive balance between the transfer of holes and broadening of the band width.

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A spin-ladder system, $Sr_{14-x}Ca_xCu_{24}O_{41}$ ($x=11.5-13.5$), which is known as the only cuprate superconductor with nonsquare lattice Cu-O layers, offers a good model for study of quasi-one-dimensional superconductivity. To date, various theoretical studies have been performed for isolated two-leg ladders or the Trellis lattice.[1–11] Although the system is attractive for comparison with high-T_c cuprates, little experimental effort has been devoted to investigation of superconductivity because of experimental difficulties under high pressure; the onset and optimal pressures are around 3 and 4 GPa, respectively.[12,13] The experiments at high pressures, above 3 GPa, have been performed only for resistivity, ac susceptibility, x-ray diffraction and NMR measurements.[12–21] Appearance of a dome-shaped T_c curve on the P-T phase diagram and a decrease in the spin gap caused by pressure application are reminiscent of hole doping in high-T_c cuprates.[13,18,19] The pressure-induced superconducting state survives even at high fields beyond the Pauli paramagnetic limit, when the field $\boldsymbol{H}$ is applied parallel to the leg direction ($\mathbf{H}\parallel$c), for which orbital suppression is depressed.[15] The NMR measurement at 3.5 GPa shows that the quasiparticle excitation accompanies a full gap; however, the Knight shift shows no appreciable change below T_c.[20]

This system also shows features common to quasi-one-dimensional systems such as organic superconductors. An example is emergence of a charge-density wave (CDW) in a lightly doped regime ($x\leq 0.8$) at ambient pressure.[22,23] The stable superconducting state beyond the Pauli paramagnetic limit and no change in the Knight shift below T_c are also common to $(TMTSF)_2X$ ($X=ClO_4$ or PF_6). These features have often been discussed in relation to the possibility of p-wave superconductivity or singlet-Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase transition.[24–26] Recently, in the cuprate spin-ladder system, the possibility of FFLO state has been investigated theoretically at high field.[8–10] NMR study at optimal pressure is required to investigate the reasons that lead to T_c enhancement after pressure application. Questions that need to be addressed are: (1) what is the origin of the dome-shaped T_c curve and (2) what is the spin susceptibility (χ_{spin}) like around T_c. We performed ^{63}Cu-NMR measurements up to the optimal pressure (∼3.8 GPa) to study these problems.

NMR and NQR measurements were performed using a single crystal of $Sr_2Ca_{12}Cu_{24}O_{41}$ having a volume of $4\times 2\times 1$ mm^3. The measurements up to 3.5 GPa were carried out using a clamped-type pressure cell. A special variable-temperature insert (VTI) equipped with a 45 ton press was used to achieve a high pressure of 3.8 GPa. Details of the apparatus are described in Fujiwara *et al.*[27] We also determined T_c from resistivity measurements using a cubic anvil cell. A top-loading insert with a 250 ton press was used to control a steady load at low temperatures. Details of the apparatus and calibration method are described in Môri *et al.*[28]

Figure 1 shows $1/T_1$ of ^{63}Cu nuclei measured at 6.2 T for $\boldsymbol{H}\parallel$c up to 3.8 GPa. The rate $1/T_1$ shows activated-type T dependence, which originates from the spin-gap excitation, followed by T-linear dependence;

$$T_1^{-1} = Ae^{-\Delta/T} + BT. \quad (1)$$

The superconducting state appears at low temperatures accompanied by a hump, implying existence of a full gap for the quasiparticle excitation. The coefficient of the T-linear dependence [B in Eq. (1)] is enhanced with increasing pressure up to 3.8 GPa, similar to T_c. The enhancement of T_c is confirmed by a shift of the coherence peak to a higher temperature. The $1/T_1T$ vs T plot normalized by those quantities at T_c fits a single curve. The T-linear dependence comes from the Korringa relation, and thus, the coefficient B is proportional to the square of the density of states (DOS) at the Fermi energy, $D(E_F)$. Therefore, the enhancement of T_c is attributable to an increase in $D(E_F)$. On the other hand, the

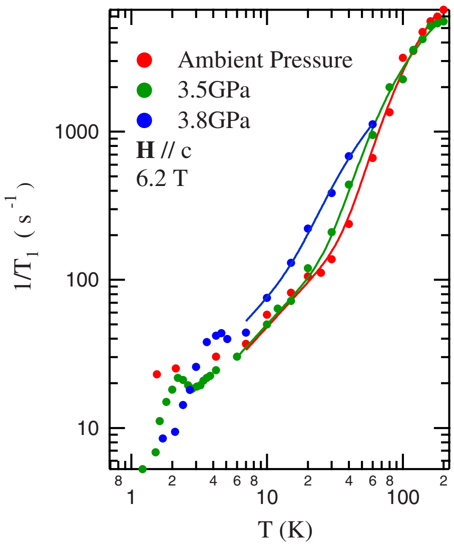

FIG. 1. (Color online) Nuclear magnetic relaxation rate $1/T_1$ of ^{63}Cu nuclei measured at 6.2 T for **H**$\|$c, the leg direction. The data are fitted by Eq. (1).

spin gap Δ and coefficient A in Eq. (1) decrease with increasing pressure. The values of A at ambient pressure, 3.5 and 3.8 GPa are in a ratio of about 6:4:1. The activated-type contribution fades out with increasing pressure, although both mechanisms coexist in the relevant pressure regime. The rate $1/T_1$ under extremely high pressure is expected to become $1/T_1T$=constant. The P dependence of both B and Δ is shown in Fig. 2, together with that of T_c determined from the resistivity measurements. The data determined from the onset and zero resistivity are plotted in Fig. 2. We also plotted the data of T_c determined from the inductance of the NMR probe. Our results show that Δ is almost the same up to the onset pressure, but decreases drastically above it. Corresponding to Δ, B shows a remarkable upturn toward the optimal pressure. The upturn of B (=T independent) means that antiferromagnetic spin fluctuation hardly affects the en-

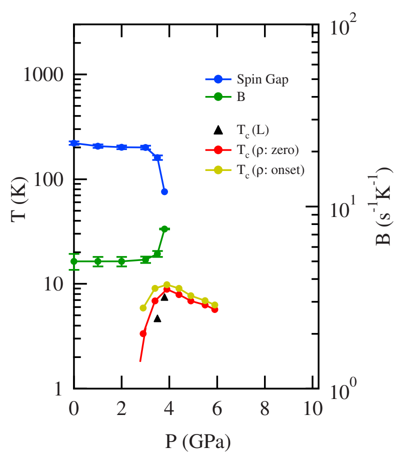

FIG. 2. (Color online) Pressure dependence of spin gap Δ and the coefficient of T-linear term, B in Eq. (1). Closed triangles represent T_c determined from the inductance of the NMR probe. $T_c(\rho\colon \text{onset})$ and $T_c(\rho\colon \text{zero})$ represent values of T_c determined from the onset and zero resistivity, respectively.

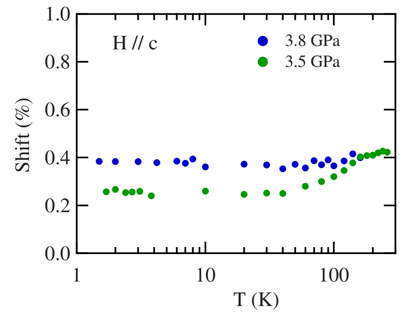

FIG. 3. (Color online) Knight shift of ^{63}Cu nuclei for the ladder sites at low temperatures derived from high fields.

hancement of T_c; when antiferromagnetic fluctuations are predominant such as high-T_c cuprates, T dependence of $1/T_1T$ shows a upturn toward T_c.

An increase in $D(E_F)$ is also observed from the Knight shift for $\boldsymbol{H}\|$c; the spin-gap behavior observed clearly at 3.5 GPa almost vanishes at 3.8 GPa (see Fig. 3). The Knight shift is decomposed into the orbital and spin parts; $K^c = K^c_{orb} + K^c_{spin}$. The orbital part K^c_{orb} is almost unchanged for a pressure of 3.5–3.8 GPa as explained in the following paragraphs. Therefore, the spin part K^c_{spin} or χ_{spin}, which is proportional to $D(E_F)$ in a normal metal, is enhanced with increasing pressure. We estimated K^c_{orb} from the NQR measurements because K^c_{orb} is related to the NQR frequency ν as

$$K^c_{orb} = \alpha\nu + \beta. \tag{2}$$

The relation is derived from Eqs. (3)–(5) in the following paragraphs.

Before presenting the results, we state the NQR results measured at 4.2 K. The NQR spectra at 3.5 GPa and ambient pressure are shown in Fig. 4. We measured the NQR signals up to 40 MHz with a resolution of 0.1 MHz/point. The signals originating from the ladder and chain sites were observed at 10–25 and 30–35 MHz, respectively. At ambient pressure, ^{63}Cu and ^{65}Cu signals are clearly assigned only in case of chain sites, whereas they overlap with each other in case of ladder sites. The overlap is decomposed into two Gaussian functions. The resonance position is almost the same as that measured in an earlier study.[29] At 3.5 GPa, the signals arising from the ladder sites move to high frequencies, whereas those from the chain sites move to low frequencies. The broadening of the line width is caused by

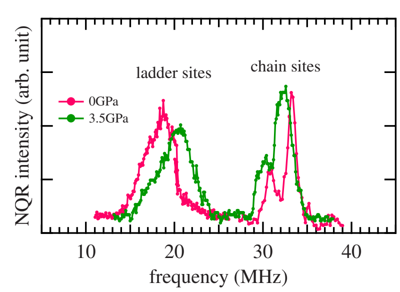

FIG. 4. (Color online) NQR spectra measured at 4.2 K for the ladder (15–25 MHz) and chain sites (30–35 MHz).

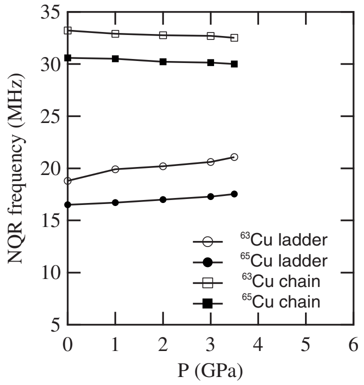

FIG. 5. Pressure dependence of the NQR frequency of ^{63}Cu and ^{65}Cu nuclei.

stress anisotropy, which appears when pressure mediation liquid freezes during the pressurizing process. The P dependence of the NQR frequencies is shown in Fig. 5. The ^{63}Cu signals move by 2.27 and −0.69 MHz for the ladder and chain sites, respectively, and those of ^{65}Cu move by 1.05 and −0.60 MHz for the ladder and chain sites, respectively. The ν in Eq. (2) is determined by two factors; the electric-field gradient (EFG) arising from the hole number of the $3d_{x^2-y^2}$ orbital, n_{3d} and that arising from the surrounding $4p$ orbitals. In high-T_c cuprates, the latter contribution is not so sensitive to doping level; therefore, ν is determined mainly by n_{3d},

$$\nu = n_{3d}\nu_{3d} + \nu_{4p} \tag{3}$$

where $\nu_{4p} \simeq -65$ MHz and $\nu_{3d} \simeq -117$ MHz are estimated.[30] Equation (3) can be applied to both ladder and chain sites of $Sr_{14-x}Ca_xCu_{24}O_{41}$ as well as high-T_c cuprates. The increase in the number of holes at the ladder Cu sites comes from the chain sites. We estimated n_{3d} from the resonance frequencies of ^{63}Cu as 0.716 and 0.736 at ambient pressure and 3.5 GPa, respectively.

The hole number is related to the orbital susceptibility χ_{orb}, namely, K^c_{orb}. The orbital susceptibility for $\boldsymbol{H}\|$c is expressed in the same way with high-T_c cuprates[31] as

$$\frac{\chi_{orb}}{N} = n_{3d}\frac{2\mu_B^2}{E_{xy} - E_{x^2-y^2}} \tag{4}$$

where N is the Avogadoro number and μ_B is the Bohr magnetron. E_{xy} and $E_{x^2-y^2}$ represent energy levels of the $3d_{xy}$ and $3d_{x^2-y^2}$ orbitals, respectively. The energy difference between them is estimated to be 4.8 eV from the cluster model.[32] K^c_{orb} is linked with χ_{orb} as

$$K^c_{orb} = A^{orb}\chi_{orb}/N\mu_B \tag{5}$$

where $A^{orb}(\equiv 2\mu_B\langle 1/r^3\rangle)$, $\langle 1/r^3\rangle$ being a radius average for a Cu ion, is a hyperfine field due to the orbital moment induced by the applied field. A^{orb} is estimated as 1028 (kOe/μ_B) using $\langle 1/r^3\rangle = 8.252$ a.u., a value for a free Cu^{2+} ion.[33] From Eqs. (4) and (5), $K^c_{orb} = 0.248 n_{3d}(\%)$ is obtained. A K^c_{orb} of 0.18% is estimated in a pressure of 3.5–3.8 GPa. The estimate shows that K^c_{spin} is enhanced at the optimum pressure.

The increase in T_c or $D(E_F)$ is determined by two factors; the hole number and the band width. The hole number seems to increase beyond the optimal pressure, as is expected from Fig. 5, whereas the band width tends to broaden with increasing pressure. Therefore, the dome-shaped T_c curve arises from naive balance between the two factors: the transfer of holes to the ladder sites increases DOS below the optimal pressure, whereas the broadening of the band width would cause a decrease in DOS over the optimal pressure.

Anomalous behavior, namely, that K^c_{spin} is finite below T_c, is actually observed at rather high fields. If it is related to quasione dimensionality, the singlet-FFLO phase transition is a candidate that can explain the anomaly. In this scenario, K^c_{spin} or χ_{spin} at a low field would decrease with decreasing temperature below T_c. Although NMR measurements at a low field is important, it is difficult to curry out because the overlapped NQR signals in Fig. 4 hardly split at a low field.

In summary, we have measured $1/T_1$ at an optimal pressure of 3.8 GPa, and observed transitional features from the spin-gapped state to the Fermi-liquid state; the activated-type contribution fades out and approaches the Korringa relation with increasing pressure. This implies that antiferromagnetic spin fluctuation is not important unlike high-T_c cuprates. The enhancement of T_c or an increase in DOS at the Fermi energy originates from an increase in the number of holes transferred from the chain sites.

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*naokif@mbox.kudpc.kyoto-u.ac.jp

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