Antiferromagnetic phase transition in four-layered high-$T_c$ superconductors
Ba$_2$Ca$_3$Cu$_4$O$_8$(F$_y$O$_{1-y}$)$_2$ with $T_c = 55 - 102$ K: $^{63}$Cu- and $^{19}$F-NMR studies

Sunao SHIMIZU$^1$*, Hidekazu MUKUDA$^1$, Yoshio KITAOKA$^3$,
Hijiri KITO$^2$, Yasuharu KODAMA$^2$, Parasharam M. SHIRAGE$^2$, and Akira IYO$^2$

$^1$Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan
$^2$National Institute of Advanced Industrial Science and Technology (AIST), Umezono, Tsukuba 305-8568, Japan

We report on magnetic characteristics in four-layered high-$T_c$ superconductors Ba$_2$Ca$_3$Cu$_4$O$_8$(F$_y$O$_{1-y}$)$_2$ with apical fluorine through $^{63}$Cu- and $^{19}$F-NMR measurements. The substitution of oxygen for fluorine at the apical site increases the carrier density ($N_h$) and $T_c$ from 55 K up to 102 K. The NMR measurements reveal that antiferromagnetic order, which can uniformly coexist with superconductivity, exists up to $N_h \approx 0.17$ being the quantum critical point (QCP) for five-layered compounds. The fact that the QCP for the four-layered compounds moves to a region of lower carrier density than for five-layered ones ensures that the decrease in the number of CuO$_2$ layers makes an interlayer magnetic coupling weaker.

KEYWORDS: high-$T_c$ superconductivity, copper-oxide, antiferromagnetism, NMR, phase diagram, apical-fluorine

1. Introduction

Remarkably high superconducting transition temperature ($T_c$) in the copper oxides has been realized in a multilayered structure of CuO$_2$ planes. $T_c$ depends on the number of CuO$_2$ layers ($n$) in multilayered compounds with a maximum at $n = 3$. In particular, the highest $T_c$ was observed around 133 K in a Hg-based three-layered ($n=3$) copper oxide HgBa$_2$Ca$_2$Cu$_3$O$_y$ (Hg-1223). Copper oxides with more than three layers comprise inequivalent types of CuO$_2$ layers, an outer CuO$_2$ plane (OP) in a five-fold pyramidal coordination, and an inner plane (IP) in a four-fold square coordination (see Fig.1 as example). Site-selective $^{63}$Cu-NMR studies have revealed that the local carrier density ($N_h$) for the IP is smaller than that for the OP. These results in turn revealed an intimate relationship between antiferromagnetism (AFM) and superconductivity (SC) inherent to CuO$_2$ layers into which mobile hole carriers were homogeneously doped.

The recent systematic Cu-NMR studies on five-layered ($n = 5$) compounds have unraveled that AFM order, which can uniformly coexist with SC, is robust up to $N_h \approx 0.17$, a quantum critical point (QCP) where the AFM order collapses. This result significantly differs from the well-established results for mono-layered ($n = 1$) La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) and bi-layered ($n = 2$) YBa$_2$Cu$_3$O$_{6+x}$ (YBCO), in which the AFM order collapses completely by doping extremely small amount of holes of $N_h \approx 0.02$ and 0.055, respectively. These results strongly suggest that the QCP of $n \leq 4$ moves to a region of lower carrier density than that of $n = 5$. Therefore, it is likely that the interlayer magnetic coupling for an onset of AFM order is enhanced as a number of CuO$_2$ layers increases. In order to establish how the interlayer magnetic coupling affects the onset of AFM order, we deal with four-layered high-$T_c$ superconductors Ba$_2$Ca$_3$Cu$_4$O$_8$(F$_y$O$_{1-y}$)$_2$ with apical fluorine. We note that all these compounds are in an underdoped state of hole doping regime.

Ba$_2$Ca$_3$Cu$_4$O$_8$(F$_y$O$_{1-y}$)$_2$ comprises a stack of four CuO$_2$ layers as illustrated in Fig.1(a). It is known as a new family of multilayered copper oxides with apical fluorine. Substitution of oxygen for apical fluorine, i.e. a decrease in nominal fluorine content ($y$) results in doping holes into CuO$_2$ layers, increasing $T_c$ from 55 K at $y = 1.0$ to 102 K at $y = 0.6$ as indicated in Fig.1(b).

This system provides an opportunity to investigate the characteristics of CuO$_2$ layers over a wide range of carrier density, especially enabling us to focus on the interplay of SC and AFM in an underdoped region. We reported in the literature that self-doping occurred at $y = 1.0$ to realize superconductivity. For a nominal content at $y = 1.0$, if the apical site of OP were fully occupied by F$^{-1}$, the formal Cu valence would be just $+2$ and hence a Mott insulator; however, it exhibits SC. The occurrence of the SC at $y = 1.0$ was argued in terms of a self-doping model wherein charge carriers were transferred between IP and OP. These discussions, however, were based on a simple assumption that all the apical sites were occupied by F$^{-1}$. Since then, a bi-layered apical-F compound Ba$_2$CaCu$_2$O$_4$F$_2$ has also been synthesized; it exhibits SC with $T_c = 73$ K. The self-doping mechanism cannot apply to this compound because it has no IPs. We, therefore, have conducted NMR studies on the bi-
layered system to confirm that the self-doping mechanism did not occur, and that all multilayered compounds with the apical fluorine were doped by hole carriers irrespective of y. For y = 1.0, it was anticipated that a possible replacement of O−2 for F−1 and/or excess oxygen in the BaF layers resulted in doping hole carriers into CuO2 layers.26

In this paper, we report systematic 63Cu- and 19F-NMR studies on Ba2Ca3Cu4Oy(FyO1−y)2 with y = 0.6, 0.7, 0.8 and 1.0 as each nominal content. Measurements of 63Cu Knight shift (63K) have revealed that hole carrier density (Nh) increases progressively with decreasing y. The substitution of oxygen for fluorine at the apical site increases Nh and Tc from 55 K up to 102 K. The measurements of 63Cu-NMR spectra and nuclear-spin-lattice-relaxation rate of 19F-NMR (1/2T1) unravel that an AFM order, which can uniformly coexist with SC, exists up to Nh ≃ 0.15 being a QCP for the four-layered compounds. From the fact that the QCP of the four-layered compounds moves to a region of lower carrier density than that of the five-layered ones, Nh ≃ 0.17, it is ensured that the decrease in the number of CuO2 layers makes an interlayer magnetic coupling weaker.

2. Experimental

Polycrystalline powder samples of all multilayered systems used in this study were prepared by a high-pressure synthesis technique, as described elsewhere.10-12 Powder X-ray diffraction measurements indicate that these compounds almost entirely comprise a single phase, and that the a-axis length continually changes with the nominal fluorine content y.11 The Tc was uniquely determined by a sharp onset of diamagnetism using a dc SQUID magnetometer as summarized in Table I. For NMR measurements, the powder samples were aligned along the c-axis at an external field (H) of ~16 T, and fixed using stycast 1266 epoxy. The NMR experiments were performed by the conventional spin-echo method in a temperature (T) range of 1.5 – 300 K with H perpendicular or parallel to the c-axis.

3. Results and Discussions

3.1 Knight shift and local carrier density

Figure 2 indicates typical 63Cu-NMR spectra of the central transition (1/2 ↔ −1/2) for (a) y = 0.6, (b) y = 0.7, (c) y = 0.8, and (d) y = 1.0. The field-swept NMR spectra were measured at H perpendicular to the c-axis. The NMR spectral widths for the samples at room temperature are as narrow as that of Hg-1245.5,7 Surprisingly, the NMR spectrum of OP at y = 1.0 also disappears at low temperatures due to the development of AFM correlations upon cooling as well as the case for the five-layered Hg- or Tl-based compounds.5,7 An average of the carrier density, defined as (Nh(OP) + Nh(IP))/2. Note that y is nominal fluorine contents.

Table I. Lists of Tc and Nh at the OP and IP of Ba2Ca3Cu4Oy(FyO1−y)2. Nh is estimated from the Kc(T) at room temperature (see text). Nh is an average of the carrier density, defined as (Nh(OP) + Nh(IP))/2. Note that y is nominal fluorine contents.

| y     | Nh [OP] | Nh [IP] | Nh       |
|-------|---------|---------|----------|
| 0.6   | 0.207   | 0.165   | 0.186    |
| 0.7   | 0.189   | 0.150   | 0.170    |
| 0.8   | 0.167   | 0.144   | 0.156    |
| 1.0   | 0.148   | 0.132   | 0.140    |

* Nh = 0.140 at y = 1.0 is estimated at Nh(OP) = 0.148 on a linear line in the plot of Nh(IP) vs Nh(see Fig.3). A linear extrapolation in the plot in Nh(IP) vs. Nh(IP) gives a tentative value of Nh(IP) = 0.132 at Nh = 0.140, since it cannot be estimated directly from the Kc(T) at AT = 1 (see an open circle in Fig.3 and the text).
and the averaged carrier density at room temperature and the temperature dependences of $K_s(\bar{T})$ with $y$ for (e) $y=0.6$, (f) $y=0.7$, (g) $y=0.8$, and (h) $y=1.0$, respectively. The $K_s(T)$ decreases upon cooling down to $T_c$ for all samples in association with an opening of pseudogap. 27, 28 whereas its steep decrease below $T_c$ evidences the reduction of spin susceptibility due to the formation of spin-singlet pairing. We note that the empirical relation between $K_s(T)$ at room temperature and the $N_h$ in a CuO$_2$ plane 29, 30 allows us to evaluate $N_h$s at OP and IP for the four samples, which are summarized in Table I along with the $T_c$ and the averaged carrier density $\overline{N_h}$, defined as $(N_h(OP) + N_h(IP))/2$. Figure 3 indicates the plot of $N_h$s at IP and OP against $\overline{N_h}$. The $N_h(IP)$ at $y=1.0$, however, was not directly estimated from the $K_s(T)$, because the Cu-NMR spectrum was not detected at room temperature. Instead, $\overline{N_h} = 0.140$ at $y = 1.0$ is estimated from $N_h(IP)=0.148$ on a linear line in the plot of $N_h(IP)$ versus $\overline{N_h}$ in Fig.3. Furthermore, a linear extrapolation in the plot of $N_h(IP)$ versus $\overline{N_h}$ gives a tentative value of $N_h(IP)=0.132$ at $\overline{N_h} = 0.140$. As summarized in Table I, the increase of $N_h(OP)$ and $N_h(IP)$ with increasing a nominal oxygen content at the apical site increases $T_c$ from 55 K to 102 K.

Fig. 2. (color online) $^{63}$Cu-NMR spectra of the central transition ($1/2 \leftrightarrow -1/2$) for (a) $y=0.6$, (b) $y=0.7$, (c) $y=0.8$, and (d) $y=1.0$. The temperature dependence of the Knight shift with $H$ perpendicular to the $c$-axis for (e) $y=0.6$, (f) $y=0.7$, (g) $y=0.8$, and (h) $y=1.0$.

3.2 Zero-field NMR evidence of AFM order

We deal with the AFM order taking place in the underdoped CuO$_2$ layers. The observation of zero-field NMR (ZF-NMR) spectra enables us to assure an onset of an AFM order, since magnetically ordered moments induce internal magnetic field $H_{\text{int}}$ at nuclear sites. Generally, the Hamiltonian for Cu nuclear spin with $I = 3/2$ is described by the Zeeman interaction due to magnetic field $H (Hz)$ and the nuclear-quadrupole interaction ($H_Q$) as follows:

$$H = H_Z + H_Q = -\gamma_N h I \cdot H + \frac{e^2 q Q}{4(2I-1)}(3I_z^2 - I(I+1)), \quad (1)$$

where $\gamma_N$ is the Cu nuclear gyromagnetic ratio, $eQ$ is the nuclear quadrupole moment, and $eQ$ is the electric field gradient (EFG) at the Cu nuclear site. Here, in the $H_Q$, an asymmetric parameter ($\eta$) is zero in the tetragonal symmetry. Note that the nuclear quadrupole resonance (NQR) frequency $\nu_Q = 3e^2 q Q/2hI(2I-1)$. The nuclear Hamiltonian given by eq.(1) is described with $H_{\text{int}}$ instead of $H$ for zero-field experiments.

Figure 4(a) indicates the Cu-NQR spectrum at $y=0.6$. Respective $^{63}\nu_Q$s are evaluated as 9.7 and 15 MHz at
The IP and OP, which are comparable to ~8–10 MHz and ~16 MHz for five-layered systems. Here, the sharp $^{63}$Cu- and $^{65}$Cu-NQR spectral widths at $^{63}\nu_Q = 22.5$ and $^{65}\nu_Q = 20.8$ MHz are as narrow as about 400 kHz. These NQR spectra qualitatively differ from those reported for various copper oxides. Integrated intensities of these NQR spectra are an order of magnitude smaller than those for the intrinsic phase, which suggests that these NQR spectra arise from some impurity phases containing copper such as starting materials prepared for the sample synthesis, and intermediate products in the high-pressure synthesis.

Figure 4(b) indicates the Cu-NQR/ZFNMR spectra at $y = 0.7$. The NQR spectrum for OP is observed with almost the same $^{63}\nu_Q$ as that of the OP at $y = 0$. Note that the respective NQR and ZFNMR spectra at IP(i) and IP(ii) arise from IP. The NQR spectrum at IP(i) is observed at 9.1 MHz that is close to $^{63}\nu_Q = 9.7$ MHz of the IP at $y = 0$, whereas the ZFNMR spectrum at IP(ii) is observed at ~18 MHz. Assuming $^{63}\nu_Q = 9.1$ MHz, $H_{int} \sim 1.5$ T is estimated for the IP(ii). The $H_{int}$ at CuO$_2$ plane is generally given by $H_{int} = |A_{hf}|M_{AFM} = |A - 4B|M_{AFM}$, where $A$ and $B$ are the on-site hyperfine field and the supertransferred hyperfine field from the four nearest neighboring Cu-AFM moments, respectively, and $M_{AFM}$ is the AFM moment. Here $A \sim 3.7$ T/µB, $B$(OP) ~ 7.4 T/µB, and $B$(IP) ~ 6.1 T/µB are assumed to be the same as those for Hg-1245. Using these values, a uniform AFM moment at the IP(ii) is estimated at $M_{AFM}$(IP) ~ 0.08 µB for an AFM phase at $y = 0.7$. The fact that IP(i) and IP(ii) originate from a paramagnetic phase and an AFM phase, respectively, suggests that the phase separation takes place because of the closeness to the QCP at which the AFM collapses. The presence of the phase separation probably imply that the AFM critical point could be close to $N_B \sim 0.15$.

Figure 4(c) indicates the Cu-NQR/ZFNMR spectra at $y = 0.8$. A spectrum observed around 14.4 MHz arises from OP since its peak frequency is almost the same frequency as $^{63}\nu_Q = 15$ MHz for the OP at $y = 0$. Accordingly, another spectrum around 28 MHz is assigned to arise from IP. Using above-mentioned parameters, $H_{int} \sim 2.4$ T and $M_{AFM}$(IP) ~ 0.12 µB are estimated for the IP at $y = 0.8$.

Figure 4(d) indicates the Cu-ZFNMR spectra observed around 30 and 45 MHz at $H = 0$ for $y = 1.0$. When noting that ZFNMR spectra are absent around $^{63}\nu_Q$(IP) = 8 ~ 10 MHz and $^{63}\nu_Q$(OP) = 14 ~ 16 MHz, the observation of the NMR spectra around 30 MHz and 45 MHz demonstrates that $H_{int}$s are present at the respective IP and OP with $H_{int} \sim 3.8$ T and 2.7 T. Since $N_B$(OP) > $N_B$(IP) due to the charge imbalance between OP and IP and hence $M_{AFM}$(OP) < $M_{AFM}$(IP), $M_{AFM}$(OP) ~ 0.11 and $M_{AFM}$(IP)~0.18 µB are evaluated at the OP and IP, respectively, using the relation of $H_{int} = |A_{hf}|M_{AFM} = |A - 4B|M_{AFM}$. Notably, the OP, which is mainly responsible for the SC with $T_C = 55$ K, manifests the AFM order, leading us to a conclusion that the uniform mixing of AFM with $M_{AFM} = 0.11$ µB and SC at $T_C = 55$ K occurs in the OP as well as in the three IPs of the five-layered systems.\(^7\)
3.3 $^{19}$F-NMR probe of Néel temperature

Néel temperatures ($T_{NS}$) at $y = 0.7$, 0.8 and 1.0 are determined by the $^{19}$F-$T_1$ measurement, which sensitively probes critical magnetic fluctuations developing at OP and IP as the system approaches an AFM order. Generally, $1/T_1$ is described as follows:

$$\frac{1}{T_1} = \frac{2\gamma^2 \hbar}{(\gamma_c \hbar)^2} \sum_q |A_q|^2 \frac{\text{Im}[\chi(q,\omega_0)]}{\omega_0},$$  \hspace{1cm} (2)

where $A_q$ is the wave-vector ($q$)-dependent hyperfine-coupling constant, $\chi(q,\omega)$ is the dynamical spin susceptibility, and $\omega_0$ is the NMR frequency. The $T$ dependence of $1/T_1$ shows a peak at $T_N$ because the low-energy spectral weight in $\chi(q = Q,\omega)$ is strongly enhanced around $\omega_0 \sim 0$ in association with a divergence of magnetic correlation length at $T \sim T_N$. Here $Q$ is the AFM wave vector ($\pi/a$, $\pi/a$). $^{19}(1/T_1)$ for all samples are presented in Fig.5. In the present case, the relaxation processes in $1/T_1$ compose of quasiparticle contributions probing the onset of SC and the magnetic one probing magnetic fluctuations. However, we consider that the former is negligible in the case of $1/T_1$ at the apical oxygen by $^{17}$O-NMR did not change drastically at $T_c$ because of the very small hyperfine-coupling constant with the quasiparticles in CuO$_2$ layers. In this context, $^{19}(1/T_1)$ is expected to be dominated by magnetic fluctuations.

Respective figures 5(b) and (c) show the $T$ dependences of $^{19}(1/T_1)$s at $y = 0.7$ and 0.8, exhibiting the peaks at $\sim 30$ K and 50 K. This result ensures the AFM order at $T_N = 30$ and 50 K for the IP(ii) at $y = 0.7$ and the OP at $y = 0.8$ with the spontaneous AFM moment of $M_{AFM}(\text{IP(ii)}) \sim 0.08 \mu_B$ and $M_{AFM}(\text{OP}) \sim 0.12 \mu_B$, respectively. Note that the absence of a peak in $1/T_1$ at $y = 0.6$ evidences that this compound is in a paramagnetic state down to 4.2 K. As shown in Fig.5(d) for $y = 1.0$, there are a distinct peak in $1/T_1$ at $T_N \sim 80$ K and a significant one at $T_N^* \sim 30$ K. The AFM order inherent to the OP responsible for SC is presumably developed below $T_N^* \sim 30$ K, exhibiting the spontaneous AFM moment of $M_{AFM}(\text{OP}) \sim 0.11 \mu_B$ at low temperatures. This suggests that the SC uniformly coexists with the AFM order in a single CuO$_2$ plane with $N_h \sim 0.148$. Since $N_h(\text{OP}) < N_h(\text{OP})$ and $M_{AFM}(\text{IP}) > M_{AFM}(\text{OP})$, the $T_N$ at the IP becomes larger than at the OP. It is noteworthy that $T_N^* \sim 30$ K for the OP at $y=1.0$ is comparable to the $T_N \sim 30$ K for the OP at $y=0.7$ because both layers possess almost the same $N_h$.

3.4 Phase diagram of AFM and SC

Figure 6 reveals a phase diagram of AFM and SC as a function of $N_h$ where $T_c$ and $T_N$ ($T_N^*$) are plotted against $N_h$ for the OPs and IPs of the four-layered superconductors Ba$_2$Ca$_3$Cu$_4$O$_8$ (FCO$_{1-y}$) at $y = 0.6, 0.7, 0.8$, and 1.0. We remark that the uniform mixing of AFM ($T_N = 30$ K) and SC ($T_c = 55$ K) was observed for the OP at $y = 1.0$, which strongly suggests that it is a general property inherent to a single CuO$_2$ plane in the underdoped regime for hole-doping. It has been reported in the literatures\textsuperscript{4-7} that a bulk $T_c$ in multilayered compounds was determined by the $T_c$ of OP and that the $T_c$ of IP was significantly lower than a bulk $T_c$ due to the lower $N_h$ at IP. In the phase diagram in Fig.6, the QCP in the four-layered system is obtained at $N_h \approx 0.15$ smaller than $N_h \approx 0.17$ for the five-layered system\textsuperscript{7} suggesting that the interlayer magnetic coupling of the four-layered compound is smaller than that of the five-layered compound. The phase diagrams of AFM and SC in multilayered systems are remarkably different from the well-established ones for LSCO ($n = 1$)$^8$ and YBCO ($n = 2$)$^9$ where the AFM order totally collapses by doping very small amount of holes with $N_h \sim 0.02$ and $N_h \sim 0.055$, respectively. The reason that the AFM phase exists up to $N_h$
\[ \simeq 0.15 \text{ and } 0.17 \] in the four- and five-layered compounds, respectively, is because the interlayer magnetic couplings are stronger than in LSCO or YBCO due to the existence of the homogeneously underdoped IPs.

**Fig. 6.** (color online) A phase diagram of AFM and SC as a function of hole carrier density \( N_h \), \( T_c \) (indicated by open circle) and \( T_K \) (closed circle) are plotted against \( N_h \) for the OPs and IPs of Ba\(_2\)Ca\(_3\)Cu\(_4\)O\(_{8+y}\) at \( y = 0.6, 0.7, 0.8 \) and 1.0. \( N_h \) was determined from the Knight shift measurement (see the text). The \( T_K \) \( \sim 70 \text{ K} \) for the IP at \( y = 0.6 \), that is shown by black cross (+) was determined from a small peak at \( T \sim 70 \text{ K} \) in the \( T \) derivative of the Knight shift (\( dK/dT \)) as well as in the literatures.\(^{3,7} \) Note that the uniform mixing of AFM (\( T_N = 30 \text{ K} \)) and SC (\( T_c = 55 \text{ K} \)) takes place at the OP at \( y = 1.0 \). This result strongly suggests that it is a general property inherent to a single CuO\(_2\) plane in the underdoped regime for hole-doping.\(^{6,7} \)

**4. Summary**

The extensive Cu-NMR/NQR and F-NMR measurements on the four-layered high-\( T_c \) superconductors Ba\(_2\)Ca\(_3\)Cu\(_4\)O\(_{8+y}\) have unraveled the systematic evolution of AFM and SC as the function of hole carrier density \( N_h \); \( T_c \) and \( T_K \) are controlled by the substitution of oxygen for fluorine at the apical site. It is demonstrated that the AFM order, which can uniformly coexist with SC, exists up to \( N_h \simeq 0.15 \), reinforcing that the uniform mixing of AFM and SC is a general property inherent to a single CuO\(_2\) plane in the underdoped regime for hole-doping. \( N_h \simeq 0.15 \) at QCP for the four-layered compounds is somewhat smaller than \( N_h \simeq 0.17 \) for the five-layered compounds. The fact that the QCP for the four-layered compounds moves to a region of lower carrier density than for the five-layered compounds ensures that the decrease in the number of CuO\(_2\) layers makes an interlayer magnetic coupling weaker. The present studies have highlighted the intimate evolution of AFM and SC in the phase diagram inherent to the homogeneously doped CuO\(_2\) plane, which depends on the interlayer magnetic coupling significantly.

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