Cu-NMR Study of Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$
Superconductor in Very High Magnetic Fields

S. Kawasaki$^1$, C. T. Lin$^2$, P. L. Kuhns$^3$, A. P. Reyes$^3$ and Guo-qing Zheng$^1$

$^1$ Department of Physics, Okayama University, Okayama 700-8530, Japan
$^2$ Max-Planck-Institut fur Festkorperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany
$^3$ National High Magnetic Field Laboratory, Tallahassee, Florida 32310, USA
E-mail: kawasaki@science.okayama-u.ac.jp

Abstract. We report the results of $^{63,65}$Cu-NMR measurements on single-layered copper-oxide Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ ($T_c = 32$ K) conducted under very high magnetic fields up to 45 T. The high magnetic field suppresses superconductivity completely, and the pseudogap ground state is revealed. The $^{63}$Cu-NMR Knight shift shows that there remains a finite density of states at the Fermi level in the zero-temperature limit, which indicates that the pseudogap ground state is a metallic state with a finite volume of Fermi surface.

1. Introduction
The mechanism of the high transition temperature ($T_c$) superconductivity in copper oxides (cuprates) [1] still remains unclear, largely because the relationship between the normal-state properties and superconductivity is still unclear. In conventional metals, superconductivity develops out of a Fermi liquid state. This is also true in the electron-doped ($n$-type) cuprates[2]. However, in the normal state of hole-doped ($p$-type) cuprates, there is an intriguing phenomenon called a pseudogap state, in which the density of states is depleted upon decreasing temperature ($T$) below a characteristic temperature $T^*$[3]. Several measurements have suggested that the pseudogap and superconductivity are coexisting states of matter[4, 5, 6], but the detailed properties and the origin of the pseudogap are still under debate[7, 8, 9, 10, 11]. Previous measurements have suggested that the pseudogap is either associated with disconnected Fermi-arcs[5, 6], or small Fermi pockets[12, 13], or associated with coexisting Fermi arc and small Fermi pockets[14]. In contrast, there was also a proposal that the Fermi surface shrinks to a nodal point when cooled to $T = 0$[15].

Experimentally, this is difficult to observe the pseudogap ground state without superconductivity due to the high upper critical field ($H_{c2} \sim 100$ T). Bi$_2$Sr$_{2-x}$La$_x$CuO$_{6+\delta}$ is one of the ideal systems to study the subject. It has a single CuO$_2$ layer, highly two- dimensional structure[16], and much lower $T_{c\text{max}} \sim 32$ K for optimally doped compound Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ compared to other cuprates.

In this paper, we report the results of the spin susceptibility via the $^{63}$Cu-NMR Knight shift measurements on optimally doped Bi$_2$Sr$_{2-x}$La$_x$CuO$_{6+\delta}$ ($x = 0.4$) carried out under very high magnetic fields up to 45 T. When the quantity $1/T_1T$ has a strong $T$-dependence[4], where $T_1$ is
the spin-lattice relaxation time, it is difficult to extract the value of density of states from this quantity. In contrast, the Knight shift is directly proportional to the density of states, which allows us to evaluate the residual density of states in the pseudogap ground state. We find that the pseudogap ground state is a metallic state with a finite volume of Fermi surface.

2. Experimental Procedure

Single crystalline Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ samples are grown by the traveling solvent floating zone method as reported elsewhere\cite{17, 18}. Black and shiny single crystal platelets sized up to $\sim 10 \times 3 \times 0.4$ mm$^3$ cleaved from the as-grown ingot were used. For all measurements, the magnetic field is applied along the $c$-axis. High magnetic fields are generated by the Bitter magnet (21.7 - 30 T) and the Hybrid magnet (44 and 45 T), respectively, in the National High Magnetic Field Laboratory, Tallahassee, Florida. A standard phase-coherent pulsed NMR spectrometer was used to collect data. The NMR spectra were obtained by sweeping the magnetic field at a fixed rf frequency and recording the size of the spin echo area.

3. Experimental Results and Discussion

Figure 1 shows a typical example of the Cu-NMR spectra for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ with the magnetic field applied along the $c$-axis ($H \parallel c$). A sharp central transition line accompanied by two satellites due to the nuclear quadrupole interaction for both $^{65}$Cu and $^{63}$Cu-nuclei are observed, respectively. This sharp central line allows us to determine precisely the temperature dependence of $^{63}$Cu Knight shift.

Figure 2 shows the temperature dependence of the Knight shift ($K_c(T)$) for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ measured at $H = 7$, 30, 44, and 45 T, respectively. Here, the obtained $K_c(T)$ is written as $K_c(T) = K_s(T) + K_{\text{orb}},$ where $K_s$ and $K_{\text{orb}}$ are the shifts due to the spin and the orbital susceptibility, respectively. Generally, $K_{\text{orb}}$ does not depend on temperature and applied magnetic fields. Most importantly, $K_s$ and $K_{\text{orb}}$ are expressed as $K_s(T) = A_{hf}^s \chi_s(T)$ and $K_{\text{orb}} = A_{hf}^{\text{orb}} \chi_{\text{orb}},$ where $A_{hf}$ is the hyperfine coupling constant.

At high temperature region, $K_c$ does not depend on temperature, which is consistent with the observation of a full Fermi-surface above $T^*$\cite{19}. As indicated in the figure, $K_c$ starts to decrease below $T^* \sim 160$ K. Notably, $1/T_1T$ for $x = 0.40$ also starts to decrease below $T^*$\cite{4}. These results indicate a certain loss of the density of states at the Fermi surface taking place below $T^*$, i.e. the opening of a pseudogap. This temperature dependence of $K_c$ is consistent with other high-$T_c$ cuprates\cite{20}. As seen in the figure, $T^*$ is almost field independent. At $H = 7$ T, $K_c$ decreases abruptly below $T_c(H) \sim 30$ K due to the reduction of the spin susceptibility as a result of a spin-singlet Cooper pair formation. However, at $H = 30$ and 44 T, no signature of superconducting transition is observed down to $T = 1.7$ K, as in previous $T_1$ measurement\cite{4}. Also, the value of $K_c$ at the lowest temperature is $H$-independent between $H = 30$ and 44 T. This indicates that the superconductivity is suppressed completely by fields $H \geq 30$ T and that the temperature dependence of $K_c (H \geq 30$ T) represents the property of the pseudogap ground states. Importantly, $K_c$ is quite large in the $T = 0$ limit. Namely, the pseudogap ground state has a finite residual density of states. Furthermore, no internal magnetic field is found when superconductivity is destroyed.

In order to quantitatively evaluate the residual density of states in the pseudogap ground state, one needs to estimate the value of $K_{\text{orb}},$ which we find to be 1.21\% as elaborated below. In the superconducting state, $K_c = K_s + K_{\text{orb}} + K_{\text{dia}}.$ $K_{\text{dia}} = -H_{\text{dia}}/H$ is the contribution due to the diamagnetism in the vortex state, which is estimated by using the relation $H_{\text{dia}} = (\phi_0/4\pi \lambda_c \lambda_v) \ln(\beta \sqrt{\xi_{ab} \xi_c} d),$ where $\phi_0$ is the flux quantum, $\lambda$ is the penetration depth, $d$ is the vortex distance, and $\beta = 0.381.$ We have used $\lambda_{ab} = 400$ A, $\lambda_c = 10 \xi_{ab}$ \cite{22}, $\xi_{ab} = 35$ A, and the relation $\lambda_{ab}/\lambda_c = \xi_{ab}/\xi_c.$ The value of $\xi_{ab}$ is extracted from $H_{c2}(H \parallel c) \sim 26$ T obtained in the present work.
The temperature dependence of the superconducting transition temperature $T_c$ for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ is shown in Fig. 2. The dotted and solid curves are fittings to the experimental data. The solid line is an eye guide.

Figure 3 shows the field dependence of $K_c$ and $K_{dia}$ for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$. The dotted and solid curves are fittings assuming $K_c(H) - K_{dia} \propto \sqrt{H}$ in the absence of impurity scattering, and $K_c(H) - K_{dia} \propto \frac{H}{T_c} \ln\left(\frac{H}{T_c}\right)$ in the presence of impurity scattering, respectively.

Figure 1. NMR spectra for the single crystalline Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ ($H \parallel c$).
obtained the value of $K_{orb} = 1.21 \%$. Here, $\delta n_{imp}/n_0 = 0.468$ is the value extracted from $T$ dependence of $1/T_1T$ at $H = 0[4]$ using theoretical calculation[26, 27], and 1.38 % is the value of $K_c$ at $T_c = 32$ K. The obtained value of $K_{orb} = 1.21 \%$ is comparable to other high $T_c$ cuprates[20]. Thus, we can extract the residual density of states, $N_{res}(E_F) \propto K_s(T = 0) = K_c(T = 0) - K_{orb}$ from the results in Fig. 2.

The relative density of states, $N_{res}/N_0$, at $T = 0$ as defined by $N_{res}/N_0 = K_s(T = 0)/K_s(T = T^*)$ for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ is about 0.49. This quantity contains the contribution released by suppressing superconductivity, and the contribution by impurity scattering[23]. The latter contribution is 0.08 as estimated from the results in Fig. 2. The relative density of states for the pseudogap ground state we found in this experiment is much larger than that inferred from the quantum oscillation ($\sim 0.03$) for the pseudogap ground state[12].

In conclusion, NMR measurements under very high magnetic fields up to 45 T reveal the pseudogap ground state for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$. Our result indicates that the pseudogap ground state is a metallic state with a finite density of state. We believe present results open new insight into the physics in high-$T_c$ cuprates.

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