Superconductivity and spin fluctuations in the electron-doped infinitely-layered high $T_c$ superconductor $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$ ($T_c = 42$ K)

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Abstract

We report $^{63}$Cu NMR studies on uniaxially aligned and unaligned powder samples of an electron-doped infinitely-layered high $T_c$ cuprate superconductor $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$ ($T_c = 42$ K). We measured the nuclear spin-lattice relaxation rate $1/T_1$ up to 800 K. We found from the temperature dependence of $1/T_1 T$ that the wave-vector-averaged spin susceptibility is highly enhanced by spin-fluctuations in the normal state, resulting in a Curie-Weiss behavior. Below $T_c$, we did not observe a Hebel-Slichter coherence peak of $1/T_1$, suggesting an unconventional nature of the symmetry of the superconducting order parameter. These results are quite similar to those observed for some hole-doped high $T_c$ cuprates.
I. INTRODUCTION

The mechanism of high temperature superconductivity in doped cuprate high $T_c$ superconductors is a very controversial issue. So far, no consensus has been reached. However, various experiments have established that undoped parent compounds of high $T_c$ cuprates are spin $S = \frac{1}{2}$ antiferromagnets. Microscopic experimental probes including NMR (Nuclear Magnetic Resonance), NQR (Nuclear Quadupole Resonance) [1], neutron scattering[2], and Raman scattering[3] revealed that one can describe the magnetic properties of the undoped parent compounds based on the two dimensional Heisenberg model reasonably well. The next crucial step toward the complete understanding of high temperature superconductivity is to figure out the nature of the carriers doped into the conducting CuO$_2$ planes, and the influence of mobile carriers on the electronic states of the undoped quasi 2d Heisenberg antiferromagnets. The undoped compound La$_2$CuO$_4$ is a quasi 2d Heisenberg antiferromagnet with intraplane exchange interaction $J/k_B = 1520$ K and bulk 3d Neel ordering temperature $T_N \sim 325$ K [2]. Recently, after several years of painful trials and errors, the Illinois-Kyoto collaboration reported the first successful detection of $^{63,65}$Cu NQR in the paramagnetic state of La$_2$CuO$_4$[1]. Their discovery of the NQR signal opened a way to investigate the influence of hole-doping in the entire doping range between the undoped La$_2$CuO$_4$ and the optimum hole-doped high $T_c$ superconductor La$_{1.85}$Sr$_{0.15}$CuO$_4$ ($T_c$=38 K) by directly observing the nuclear resonance in the CuO$_2$ planes. They measured the temperature dependence of the nuclear spin-lattice relaxation rate $1/T_1$ and the Gaussian component of the spin-spin relaxation rate $1/T_{2G}$. These quantities probe the imaginary and real parts of the weighted wave-vector $\mathbf{q}$-averaged dynamical electron spin susceptibility $\chi''(\mathbf{q}, \omega = \omega_n)$ and $\chi'(\mathbf{q}, \omega = 0)$, respectively, where $\omega$ is frequency and $\omega_n$ is the resonance frequency [4,5]. Theoretical calculations for the 2d-Heisenberg model based on dynamical scaling[6], high temperature expansion[7], and finite cluster calculations[8] reproduce the experimental data of La$_2$CuO$_4$ quite well without any adjustable parameters. The most surprising observation by Imai et al. is that $1/T_1$ levels off above $750$ K at approximately the same value regardless
of the doping level [1]. This finding demonstrates that at high temperatures the spin dynamics of the high $T_c$ superconductor La$_{1.85}$Sr$_{0.15}$CuO$_4$ retains essentially the same properties of the undoped antiferromagnet La$_2$CuO$_4$. In other words, in the first order approximation, copper d-spins in the superconducting phase may be viewed as localized moments at elevated temperatures [9]. More recently Sato and coworkers also reported that the Hall coefficient levels off at elevated temperatures for La$_{2-x}$Sr$_x$CuO$_4$ ($0.04 \leq x \leq 0.15$) at approximately the same value [10]. This suggests that charge dynamics in the hole-doped superconducting phase may also keep the fundamental characteristic of the insulating phase at elevated temperatures. On the other hand, King et al. reported that the dispersion relations of the bands crossing or close to the Fermi surface are very different in an electron-doped high $T_c$ superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ compared with those in hole-doped materials [11]. The temperature dependence of the penetration depth below $T_c$ in Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ reported by Wu et al. is also regarded to be an evidence for the isotropic s-wave pairing realized in the electron-doped system [12], in contrast with the result for a hole-doped system YBa$_2$Cu$_3$O$_{6.95}$ by Hardy et al. which clearly indicates an unconventional nature, most likely to be d-wave pairing[13]. In view of these results, a naive question is what happens to the NMR properties if one dopes electrons instead of holes into the CuO$_2$ planes? Unfortunately it is not easy to answer this question because of the following two reasons. Firstly, the physical properties of the well-known electron-doped high $T_c$ superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ are extremely sensitive to the oxygen content as may be seen in the results of Kambe et al.[14]. This makes preparation of high quality samples as well as reproducible studies very hard. Secondly, the presence of the large Nd moments masks the intrinsic physical properties of CuO$_2$ planes. In particular, $^{63}$Cu NMR experiments on Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ are almost completely dominated by the large Nd moments as demonstrated by Zheng et al.[15]. In this paper, we discuss our recent NMR results [16] for the electron-doped high $T_c$ superconductor Sr$_{0.9}$La$_{0.1}$CuO$_2$ ($T_c = 42$ K) [17], and compare the results with those of hole-doped materials. This material is suited for NMR experiments because unlike the case of Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ there are no unpaired spins outside the CuO$_2$ planes in Sr$_{0.9}$La$_{0.1}$CuO$_2$ [18]. This enabled
us to obtain information intrinsic to the electron-doped CuO$_2$ planes from $^{63}$Cu NMR for the first time. In addition, the infinitely-layered structure of Sr$_{0.9}$La$_{0.1}$CuO$_2$ allows us to investigate the possible influence of interlayer coupling between adjacent CuO$_2$ planes on the physical properties of high $T_c$ cuprates [19]. Our key finding is that the spin fluctuation and superconducting properties of Sr$_{0.9}$La$_{0.1}$CuO$_2$ probed by NMR exhibited very similar results to those of hole-doped compounds.

II. EXPERIMENTAL

Polycrystalline samples of Sr$_{0.9}$La$_{0.1}$CuO$_2$ were prepared by utilizing high pressure apparatus at Texas. Details of the preparation of the samples and their characterization are discussed elsewhere [17]. The hard pellet samples were crushed and sieved to obtain fine ceramic powder samples. A part of the fine powder was uniaxially aligned in external magnetic field of 8.3 Tesla in Stycast 1266 resin at room temperature. Since Stycast reduces the high $T_c$ cuprate samples above approximately 500 K, NMR measurements above 400 K were carried out utilizing a powder sample with no epoxy sealed in a nitrogen gas atmosphere. After we completed the measurements of the NMR line shape and $1/T_1$ up to 600 K, we cooled down the sample to room temperature to check the reproducibility. We found that $1/T_1$ at room temperature was identical to that before the heat cycling. We repeated the same procedures after the measurements at 800 K, again confirming the reproducibility of $1/T_1$ at room temperature, though we found a weak $^{63}$Cu NMR signal from small amount of copper metal reduced from the sample. All the NMR measurements were carried out at Illinois using standard home made pulsed NMR spectrometers. Superconducting magnets operating either at 8.3 Tesla or at 9.4 Tesla were utilized. NMR line shape was measured by integrating the spin echo signal point by point scanning the resonance frequency. $1/T_1$ was measured by inverting the population of nuclear magnetization with a 180 degree pulse.

III. RESULTS AND DISCUSSIONS
A. Static Properties

In Fig.1, we present the $^{63}$Cu and $^{65}$Cu NMR line shape of an unaligned powder sample of Sr$_{0.9}$La$_{0.1}$CuO$_2$ measured at 295 K. There is a small shoulder in the broad line shape at the higher frequency side of each of the $^{63}$Cu and $^{65}$Cu NMR spectra, characteristic of the NMR line shape with anisotropic Knight shift with axial symmetry. In fact we confirmed that the angular dependence of the peak position of $^{63}$Cu NMR in the aligned powder sample fits the theoretical curve expected for the case of an anisotropic Knight shift [20] quite well. In Fig. 2 (a) and (b), we present the $^{63}$Cu NMR line shapes observed for the uniaxially aligned powder sample with the magnetic field applied parallel to the crystal c-axis and the ab-plane, respectively. One notices that there is a relatively sharp transition and broad tails for both higher and lower frequency sides. We attribute the sharp peak to the transition from nuclear spin quantum number $I_z = 1/2$ to $I_z = -1/2$ (i.e. the central transition), and the broad tail to the transition from $I_z = 3/2$ to $1/2$ and from $I_z = -3/2$ to $-1/2$ (i.e. satellite transitions). We verified the assignment by comparing the nutation curves of the spin echo signal as a function of $t_w$, the width in time of the RF exciting pulse for Cu metal, the $I_z = 1/2$ to $I_z = -1/2$ transition of the planar Cu site of YBa$_2$Cu$_3$O$_7$, and the central peak of the aligned powder of Sr$_{0.9}$La$_{0.1}$CuO$_2$ under identical experimental conditions. For Cu metal, a 90 degree pulse occurs when $\gamma H_1 t_w = \pi/2$, where $\gamma$ and $H_1$ are the nuclear gyromagnetic ratio and the strength of RF magnetic field, respectively. For the $+1/2$ to $-1/2$ transition, a 90 degree pulse occurs when $2\gamma H_1 t_w = \pi/2$ due to the matrix element effects [20]. We found that the results for YBa$_2$Cu$_3$O$_7$ and Sr$_{0.9}$La$_{0.1}$CuO$_2$ were identical, as shown in the inset to Fig.1, while $t_w$ for YBa$_2$Cu$_3$O$_7$ and Sr$_{0.9}$La$_{0.1}$CuO$_2$ was 2 times shorter than that for Cu metal. From the separation of the satellite transitions, we estimate the nuclear quadrupole coupling $\nu_Q$ to be 4 MHz or below. In general the peak position of the NMR line for the $I_z = 1/2$ to $I_z = -1/2$ transition is shifted by both the NMR Knight shift, $^{63}K$, and the second or higher order effects due to the quadrupole interaction [20]. We confirmed that the relative shift of the apparent peak position is identical for both $^{63}$Cu
and $^{65}$Cu isotopes for 8.3 and 9.4 Tesla when the magnetic field is applied parallel to the aligned c-axis. This indicates that the main principle axis of the quadrupole coupling tensor is parallel to the c-axis, and the contribution from the quadrupolar effects to the NMR shift is null in the case. By taking $\nu_Q = 4$ MHz, the contribution of the quadrupole effects to the shift for the field applied parallel to the ab-plane is 0.03% at 8.3 Tesla. By taking into account these considerations, our preliminary results on the temperature dependence of Knight shifts are as follows. $^{63}K_c$ saturates at elevated temperatures and decreases below room temperature from 0.92% at 295 K to 0.86% at $T_c = 42$K. In contrast, $^{63}K_{ab} = 0.29$ % is independent of temperature. These results are in remarkable contrast with the case of underdoped region of the hole doped systems, where $^{63}K_c$ is temperature independent and $^{63}K_{ab}$ decreases with temperature [21]. The hyperfine coupling tensor in Sr$_{0.9}$La$_{0.1}$CuO$_2$ seems to be rather different from that in YBa$_2$Cu$_3$O$_x$ and La$_{2-x}$Sr$_x$CuO$_4$.

**B. Spin Dynamics**

In Fig. 3 we present an example of the dependence of the echo integral $M(t)$ on the delay time $t$ of the echo sequence after a 180 degree inversion pulse. The result fits very well the solution of the rate equation for the $I_z = 1/2$ to $I_z = -1/2$ transition of nuclear spin $I = 3/2$, $M(t) = A - B[0.9 exp(-6t/T_1) + 0.1 exp(-t/T_1)]$, where A and B are constants, and $T_1$ is the spin-lattice relaxation time. The temperature dependence of the spin-lattice relaxation rate $1/T_1$ measured for an aligned powder sample under the external magnetic field 8.3 Tesla applied parallel to the crystal c-axis or ab-plane is presented in Fig.4. The anisotropy of $1/T_1$, $^{63}R$, is approximately $^{63}R \sim 2.6$. This is comparable to but somewhat smaller than the value observed for undoped La$_2$CuO$_4$ [1] and hole-doped YBa$_2$Cu$_3$O$_7$ [22], $^{63}R \sim 3.7$, and much smaller than that for an antiferromagnet CuO, $^{63}R \sim 8$ [23]. The large value of $^{63}R$ in CuO is due to the absence of the transferred hyperfine interaction. Our observation suggests that the mechanism of transferred hyperfine interaction between nearest neighbor Cu sites [24] are present in electron-doped systems, too. Mild scattering of the data for $(1/T_1)_c$ caused
by the broad line width and the resulting contamination by the satellite transition make it difficult to obtain the precise value and temperature dependence of $^{63}R$. It appears that $^{63}R$ does not show any strong temperature dependence above $T_c$. Above 450 K, we found that the ceramic powder sample was reduced and destroyed in stycast 1266. To avoid this, we carried out measurements of $1/T_1$ for an unaligned powder sample at the peak position of the powder spectrum at higher temperatures. We normalized the data for powder, $(1/T_1)_{\text{powder}}$, to that measured for aligned powder, $(1/T_1)_{ab}$, at 295 K by multiplying with a factor 1.4. We plotted the data of $(1/T_1)_{ab}$ and $1.4(1/T_1)_{\text{powder}}$ together in Fig. 5. Since the normalized results showed good agreement also at 77 K, the normalization procedure should not alter any essential aspect of the data. It is clear from Fig. 5 that the results for the electron-doped compound is semi-quantitatively similar to that of various hole-doped compounds. A high relaxation rate $1/T_1$ with negative curvature in its temperature dependence indicates that the electronic states can not be described by the canonical Fermi liquid state, and strong correlation effects have to be taken into account. As shown in Fig. 5(c), the temperature dependence of $T_1 T$ follows a Curie-Weiss law, $T_1 T = c(T - T^*)$, where $c = 3 \times 10^{-4}$ sec and $T^* = -174$ K are constants, in the entire temperature range between $T_c = 42$ K and 800 K. As demonstrated by Moriya [4], $1/T_1$ probes the weighted $q$-average of the imaginary part of the electron spin susceptibility. Therefore our finding indicates that the wave vector $q$ averaged spin susceptibility in the electron doped superconductor Sr$_{0.9}$La$_{0.1}$CuO$_2$ satisfies a Curie-Weiss law. The negative value of $T^*$ means that the electron spin system in the electron-doped CuO$_2$ planes are not approaching the Neel ordered ground state. These findings are essentially the same as those previously reported for YBa$_2$Cu$_3$O$_7$ by Barrett et al. [25] We also attempted to fit $1/T_1$ to a power law, $1/T_1 \sim T^n$ ($n \sim 1/2$), as suggested by Ren and Anderson for a Luttinger liquid system [26]. Though our data is in general accord with a power-law behavior with $n < 1$, we found that the fit is not good. One unique feature of Sr$_{0.9}$La$_{0.1}$CuO$_2$ is in the strong interlayer coupling. In fact the undoped parent compound (Ca$_{0.85}$Sr$_{0.15}$)CuO$_2$ has the highest Neel ordering temperature, 540K, among various cuprates [27]. Moreover Cobb and Markert found that the superconducting anisotropy is relatively
Their observation is an evidence for strong interlayer coupling in the electron doped compound [28]. The interlayer coupling of CuO$_2$ bi-layers has been proposed to be responsible for the spin gap behavior in underdoped YBa$_2$Cu$_3$O$_{x}$ and YBa$_2$Cu$_4$O$_8$, where $1/T_1 T$ shows a significant reduction above $T_c$ [21]. Though one may anticipate that the spin gap behavior is even enhanced in the infinitely-layered compound compared with the bi-layer systems, we did not observe spin gap behavior in Sr$_{0.9}$La$_{0.1}$CuO$_2$. As presented in Fig. 5(b), $1/T_1 T$ increases monotonically down to $T_c$. The interplay between the doping level, the nature of doping (hole or electron), interlayer coupling, and in-plane anisotropy [29] needs further investigation to clarify the mechanism of spin gap behavior observed in some hole-doped systems. Another important finding in our data is that $1/T_1$ exhibits monotonic and sudden decrease just below $T_c$. Various model calculations showed that isotropic s-wave superconducting pairing results in a Hebel-Slichter coherence peak [30] just below $T_c$ even for the systems with strong antiferromagnetic spin fluctuations. The absence of the coherence peak in the magnetic field of 8.3 Tesla makes the s-wave scenario unlikely in the electron-doped superconductor Sr$_{0.9}$La$_{0.1}$CuO$_2$.

**IV. SUMMARY**

We reported our $^{63}$Cu NMR measurements for an electron-doped infinitely-layered high $T_c$ superconductor Sr$_{0.9}$La$_{0.1}$CuO$_2$. This material is unique in the sense that the doping is made by adding electrons into CuO$_2$ planes instead of holes, and the interlayer coupling might be strongest among various cuprates. Our experimental results indicate that the spin dynamics in the electron-doped Sr$_{0.9}$La$_{0.1}$CuO$_2$ show essentially the same temperature dependence as those in hole-doped compounds. The symmetry of the orbital pairing in the superconducting state is unlikely to be isotropic s-wave, again analogous to the case of hole-doped systems. Investigation of further details of some aspects of our experiments including NMR shifts, hyperfine coupling, and the comparison with the undoped insulating antiferromagnet Ca$_{0.85}$Sr$_{0.15}$CuO$_2$ are under way.
V. ACKNOWLEDGMENT

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VII. FIGURE CAPTIONS

Figure 1

$^{63}\text{Cu}$ and $^{65}\text{Cu}$ NMR spectra observed for the unaligned powder sample at 295 K in 8.3 Tesla. Inset: Echo intensity of the central peak of $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$ observed for the aligned powder sample plotted as a function of the width in time of the pulse width.

Figure 2

$^{63}\text{Cu}$ NMR spectra observed at 77 K for the uniaxially aligned powder sample of $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$. The magnetic field of 8.3 Tesla is applied parallel to (a) aligned c-axis, and (b) aligned ab-plane.

Figure 3

Plot of the normalized intensity of echo integral $M(t)$ at time $t$ after inversion of the population of spins. $M_0$ is the saturated value of the echo intensity for infinitely large $t$. Solid line is the fit to the solution of the rate equation.

Figure 4

Temperature dependencies of $^{63}(1/T_1)_{ab,c}$ measured for the uniaxially aligned powder sample.

Figure 5

(a) Temperature dependence of $^{63}(1/T_1)_{ab}$ obtained from the measurement for aligned powder sample (filled circles) and for unaligned powder sample (unfilled circles, normalized by multiplying a factor 1.43). (b) $^{63}(1/T_1T)_{ab}$. (c) $^{63}(T_1T)_{ab}$. Solid line is a fit to Curie-Weiss law, $^{63}(T_1T)_{ab} = 3 \times 10^{-4}(T + 174)$. 

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Fig. 1
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