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NMR evidence for a pseudogap in Pb-doped Bi:2201 single crystal

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A ^{63}Cu nuclear magnetic resonance (NMR) study of a Pb-doped Bi:2201 system, $Bi_{1.6}Pb_{0.4}Sr_{2.05}CuO_y$, is presented. The temperature dependencies of the NMR peak shift and the nuclear spin-lattice relaxation (SLR) rate reveal the pseudogap that opens near $T^* = 60$ K, significantly above the superconducting critical temperature $T_c \simeq 9$ K at the NMR experiment field, 7 T oriented $H \parallel c$. The noticeable disparity between T_c and T^* and the behavior of Cu SLR at $T > T^*$ imply the underdoped state of the studied system. A relatively weak effect of the magnetic field on the superconductivity evidenced from small (≈ 7 K) shift of the zero-field $T_{c0} = 16 \pm 1$ K under the applied 7 T field suggests high upper critical field, H_{c2}, unusual for compounds with as low T_{c0}.

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The "pseudogap" (PG), a phenomenon revealed in the decay of the density of states (DOS) at temperatures higher than the superconducting (SC) transition critical temperature, T_c, is a fingerprint of high-temperature superconductors (HTS). Discovered by NMR [1–3] and inelastic neutron scattering [4] as a drop of the electronic spin excitations at $T^* > T_c$, the effect was verified later by tunnel experiments [5] that tied the abnormal transport and spin properties of HTS's above T_c. Quite recently, the PG phenomenon was discovered in a XXI century HTS – a FeSe-Based Superconductor [6].

Understanding the relationship between the normal and SC properties of HTS'c, in general, and of the nature of the PG, in particular, is among key issues in unlocking the mystery of HTS'c [7]. The origin of the PG has been widely disputed over the past decades. Some attributed it to SC fluctuations [8], others employed a two-gap scenario where the PG is associated with another order of non-SC origin, that coexists and competes with the superconductivity. Scrutinized by various experimental techniques including NMR [9], tunnel spectroscopy [10–12], polarized elastic neutron diffraction [13], and angle-resolved photoemission spectroscopy (ARPES) [14], this dualism has been seemingly removed in favour of non-SC nature of the PG.

In this letter we report on ^{63}Cu NMR in a Pb-doped Bi:2201 ($Bi_2Sr_2CuO_6$), $Bi_{1.6}Pb_{0.4}Sr_{2.05}CuO_y$ ($y \approx 6$). The sample with dimensions $a \times b \times c \approx 3 \times 5 \times 1$ mm^3 was cut from a single crystal grown by the crucible-free floating zone method [15]. T_c of the sample was measured by monitoring the NMR tank circuit resonance frequency, $\nu_t \approx 80$ MHz, as shown in the inset in Fig. 1, for $H = 0$ and $\mu_0 H = 7$ T aligned with the c-axis of the crystal. At zero field ν_t diminishes with decreasing temperature below $T_{c0} = 16.5 \pm 0.5$ K, indicating the SC transition. Under the external field of 7 T, T_c shifts down to 8–9 K.

The temperature dependence of ^{63}Cu NMR line shift in the Pb-doped Bi:2201, $^{63}K = \nu/\nu_0 - 1$ is shown in the main panel of Fig. 1a. Here ν is the position of the ^{63}Cu NMR peak at $1/2 \leftrightarrow -1/2$ transition, $\nu_0 =^{63} \gamma\mu_0 H/2\pi = 79.231$ MHz, $^{63}\gamma \simeq 11.319$ MHz/T the gyromagnetic ratio. For the sake of visibility, the $\mu_0 H = 7$ T data from the inset demonstrating the SC transition in the NMR experiment field, is plotted together with ^{63}K. The decrease in $^{63}K(T)$ is observed below $T^* = 60 \pm 5$ K, appreciably higher than $T_c(7\,\mathrm{T})$.

The shift of ^{63}Cu peak is contributed from quadrupolar and magnetic shifts. For $1/2 \leftrightarrow -1/2$ transition and $H \parallel c$, the quadrupolar shift is zero (the field is along the symmetry axis of the electron gradient field). Magnetic contributions include the diamagnetic one from the core electrons, the orbital shift related to the van Vleck susceptibility, and the Knight shift resulted from contact interaction with conduction electron spins, $K_s \propto \chi_s$, where χ_s is the conduction electron static spin susceptibility. The former two contributions are usually temperature-independent while K_s follows the T-dependence of χ_s coupled to the DOS at the Fermi level. Therefore, the decrease observed in the temperature dependence of the ^{63}K shown in Fig. 1a is associated with $K_s(T)$ and denotes

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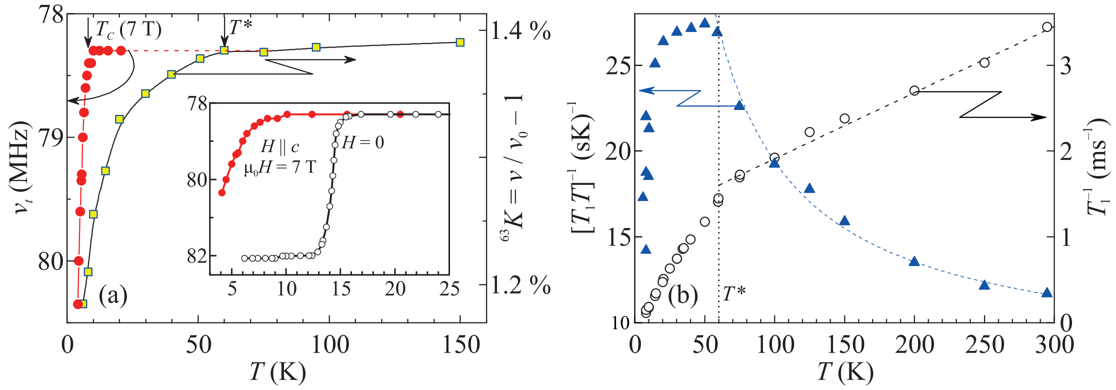

Fig. 1. (Color online) (a) – Temperature dependence of ^{63}Cu line shift (squares) and the SC transition of the sample (circles) for $\mu_0 H = 7$ T, $H \parallel c$. Inset: SC transition of the sample viewed as the shift of the NMR tank circuit resonance frequency. (b) – Temperature dependencies of the SLR rate, T_1^{-1} (circles) and the $(T_1 T)^{-1}$ product (triangles)

the decrease in DOS, hence opening of a spin gap. The temperature where it occurs, $T^* > T_c$ implying the pseudogap.

Figure 1b shows the T-dependencies of ^{63}Cu spin-lattice relaxation (SLR) rate, T_1^{-1}, and the $(T_1 T)^{-1}$ product. T_1^{-1} is linear between room temperature and 75 K, and decreases faster upon further cooling. $(T_1 T)^{-1}$ is hyperbolic down to 60 K, and decreases at $T < 50$ K, well above the $T_c \simeq 9$ K of the sample in 7 T field parallel to c-axis.

Such T-behavior of Cu SLR is typical for HTS cuprates [1, 2, 16, 17]. In nonmagnetic metals the nuclear SLR is governed by spin fluctuations of conduction electrons, generally expressed as $(T_1 T)^{-1} \propto \lim\limits_{\omega \to 0} \sum\limits_q |A(q)|^2 \chi''(q,\omega)/\omega$, where $A(q)$ is the hyperfine coupling between electronic and nuclear spins, $\chi''(q,\omega)$ the imaginary part of the wave-vector and frequency dependent dynamic electronic spin susceptibility [1]. In common metals χ'' is q-independent, so the SLR is related with the Knight shift by the known Korringa formula $(K_s^2 T_1 T)^{-1} \equiv \pi h k_B \gamma^2/\mu_B$ which assumes the T-independent $(T_1 T)^{-1}$, provided that K_s is constant. In HTS cuprates the Korringa relation is usually violated because χ'' is peaked at the antiferromagnetic (AF) wave vector $Q = (\pi, \pi)$, and $(T_1 T)^{-1}$ reproduces the $(T + \Theta)^{-1}$-like behavior of $\chi''(Q)$, caused by the development of AF correlations [16]. Below $T^* > T_c$ $(T_1 T)^{-1}$ decreases due to the pseudogap.

These features of the nuclear SLR are more pronounced in underdoped cuprates where the difference between T_c and T^* can be up to 100 K. In overdoped systems $(T_1 T)^{-1}$ is relatively flat above T^*, and the discrepancy between T^* and T_c fades away [16–18]. As for the system in study, the essential difference between T^* and T_c, viewed in both the NMR line shift and Cu SLR T-dependencies, as well as the hyperbolic T-dependence of $(T_1 T)^{-1}$ above 60 K, indicate the underdoped state of the system.

Summarizing, the Cu NMR data collected for Pb-doped Bi:2201 compound reveal the pseudogap that opens at $T^* \simeq 60$ K, way above the $T_c(7\,\mathrm{T}) \simeq 9$ K. The noticeable discrepancy between T_c and T^* and the behavior of Cu SLR at $T > T^*$ implies the underdoped state of the studied system. Unexpectedly, the 7 T magnetic field applied along the c-axis of the crystal, shifts the zero-field $T_{c0} = 16 \pm 1$ K by 7 K only. In other HTS cuprates with similar zero-field T_c the same magnetic field destroys the superconductivity either completely or shifts the transition temperature well below 4 K [19, 20].

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