Superconducting Fluctuations and the Pseudogap in the Slightly Overdoped High-T_c Superconductor $TlSr_2CaCu_2O_{6.8}$: High Magnetic Field NMR Studies

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From measurements of the ^{63}Cu Knight shift (K) and the nuclear spin-lattice relaxation rate ($1/T_1$) under magnetic fields from zero up to 28 T in the slightly overdoped high-T_c superconductor $TlSr_2CaCu_2O_{6.8}$ ($T_c = 68$ K), we find that the pseudogap behavior, i.e., the reductions of $1/T_1T$ and K above T_c from the values expected from the normal state at high T, is strongly field dependent and follows a scaling relation. We show that this scaling is consistent with the effects of the Cooper pair density fluctuations. The present finding contrasts sharply with the pseudogap property reported previously in the underdoped regime where no field effect was seen up to 23.2 T. The implications are discussed.

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The high transition temperature (T_c) superconductors have attracted enormous attention because of their high T_c and their anomalous normal-state properties. It is believed that the unusual normal-state properties are due to strong electron-electron correlation effects. On the other hand, it is also pointed out that some effects associated with the high value of T_c, such as superconducting fluctuations (SCF), may also complicate the normal-state properties [1]. Among various unsettled issues of the normal state, the so-called pseudogap (PG), which is a phenomenon of spectral weight suppression, has attracted much attention in recent years. Although the PG is observed in most underdoped materials and possibly also in the overdoped regime [2], its detailed properties remain to be characterized. Measurements under strong magnetic fields may help to discriminate between different mechanisms that are responsible for the PG [3–6].

In this Letter, we report the temperature (T) and magnetic field (H) dependence of the normal-state properties probed by the ^{63}Cu Knight shift (K) and the nuclear spin-lattice relaxation rate ($1/T_1$) measurements in the slightly overdoped superconductor $TlSr_2CaCu_2O_{6.8}$, at both zero magnetic field (nuclear quadrupole resonance) and high fields up to 28 T. It was found that the PG behavior is seen, but it depends strongly on H. We further find that the PG follows a T- and H-scaled relation, which is shown to be consistent with the Cooper pair density fluctuations. The present finding contrasts sharply with the PG property in the underdoped regime where no field effect was seen up to 23.2 T [3]. Implications of these findings are discussed.

$TlSr_2CaCu_2O_{7-\delta}$ consists of two identical CuO_2 planes in the unit cell. The doping level is controlled by changing the oxygen content by annealing [7]. The as-grown sample is nonsuperconducting with $\delta = 0.12$. Superconductivity is obtained and T_c is increased monotonically to 70 K [7,8] when δ is increased, thereby reducing the carrier concentration. The high-T electrical resistivity follows a simple power law $\rho = \rho_0 + aT^n$. The exponent n changes gradually from ~1.3 for the highest-T_c sample to 1.7 for the as-grown sample [7]. Even the sample with the highest T_c is suggested to be still in the slightly overdoped regime [8]. The sample used in this study has a zero-field critical transition temperature T_{c0} of 68 K with $\delta \sim 0.20$ and $n = 1.3$ [7]. All NMR measurements were done on the central transition (the $-1/2 \leftrightarrow 1/2$ transition) in a c-axis aligned powder sample [8]. The value $1/T_1$ was obtained from the recovery of the magnetization $[M(t)]$ following a single saturation pulse and a good fitting to $\frac{M(\infty)-M(t)}{M(\infty)} = 0.9\exp(-6t/T_1) + 0.1\exp(-t/T_1)$ [9]. The transition temperature for $H \parallel c$ axis, T_{cH}, was determined from the ac susceptibility by measuring the inductance of the NMR coil and was 59, 51, 40, and 37 K for $H = 7, 15.6, 23$, and 28 T, respectively, as indicated by the arrows in Fig. 1. Application of the Werthamer-Helfand-Hoenberg theory [10] indicates that a field of 43 T should destroy the superconductivity completely. This relatively small critical field, $H_{c2}(0)$, is another manifestation of the sample being overdoped.

Figure 2(a) shows $1/T_1T$ as a function of T for $0 \leq H \leq 28$ T parallel to the c axis. Figure 2(b) shows the T variation of the Knight shift (K_c) for various $H \parallel c$ axis. Figure 3 emphasizes the data near T_{cH}. The arrows, from right to left, indicate T_{cH} as H is increased. At $H = 0$, $1/T_1T$ increases with decreasing T down to $T^* = 85$ K. The curve in Fig. 2(a) is a fitting of the data above $T = 90$ K to the relation of $1/T_1T = \frac{C}{T+\theta}$, with $C = 4.7$ msec^{-1} and $\theta = 235$ K. This Curie-Weiss (CW) relation of $1/T_1T$ was reported in many other high-T_c cuprates such as $La_{2-x}Sr_xCuO_4$ [11] and is explained theoretically as caused by antiferromagnetic (AF) spin

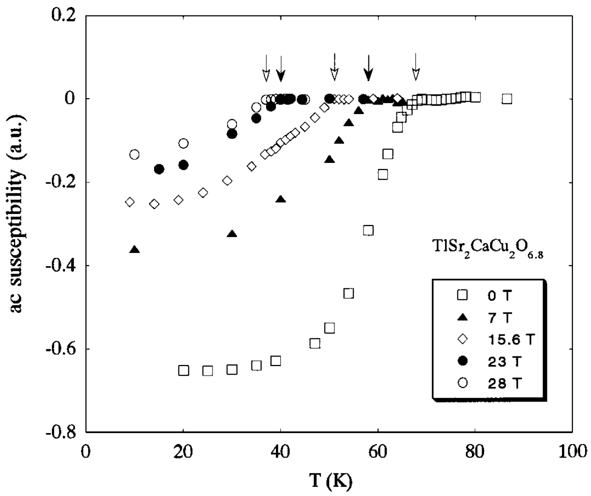

FIG. 1. ac susceptibility measured at 330 MHz under various fields. The sample was field cooled.

fluctuations [12,13]. The deviation of $1/T_1T$ from the CW relation below $\sim T^*$ has been widely attributed to the loss of low-energy spectral weight, i.e., the opening of a PG. Applying a magnetic field shifts T^* to lower T; $1/T_1T$ is strongly field dependent below $T \sim T^* = 85$ K.

K_c is the sum of an H- and T-independent orbital part K_{orb} and the spin part K_s, which is proportional to the uniform spin susceptibility χ_s. In Y- and La-based cuprates K_s of ^{63}Cu for $H \parallel c$ axis is negligibly small due to accidental cancellation of the hyperfine field, which prevents investigation of the T and H dependence of χ_s at this field alignment. K_s is finite in the present case, likely due to larger transferred-hyperfine field coming from the nearest Cu [8]. At high T, as seen in Fig. 2(b), K_c increases slightly with decreasing T, but remains constant at 1.42% for $85 \leq T \leq 150$ K. Such a T-independent K is a common feature of the optimally doped [14] or overdoped materials [15]. As seen in Fig. 3(b), K_c starts to decrease at temperatures above T_{cH}, with no apparent singularity at T_{cH}. Like $1/T_1T$, the temperature at which K_c starts to deviate from a constant value depends on H.

The key experimental results are the following: (1) The PG temperature at which K_c starts to deviate from a constant value and $1/T_1T$ deviates from that expected by the relation of $1/T_1T = \frac{C}{T+\theta}$ is lowered progressively by H. (2) The reduction of K_c and $1/T_1T$ at T_{cH} become larger as H is increased. Thus, at a first glance, it appears that the PG behavior becomes prominent at high fields, although it starts at lower T.

We find that the T and H dependence of these reductions follow a scaling relation. In Fig. 4(a), we show the normalized reduction of $1/T_1T$, $\delta(T_1T)^{-1} = \frac{(T_1T)_N^{-1} - (T_1T)_{\text{obs}}^{-1}}{(T_1T)_N^{-1}}$, divided by $\sqrt{T_{c0} - T_{cH}}$, as a function of the reduced temperature difference, $\frac{T - T_{cH}}{T_{c0} - T_{cH}}$. Here $(T_1T)_N^{-1} = \frac{4.7}{T+235}$ in msec^{-1} K^{-1}. It is seen that the data for four fields are collapsed onto a universal curve down to well below T_{cH}. In Fig. 4(b), the reduction of K, $\delta K = 1.42\% -$

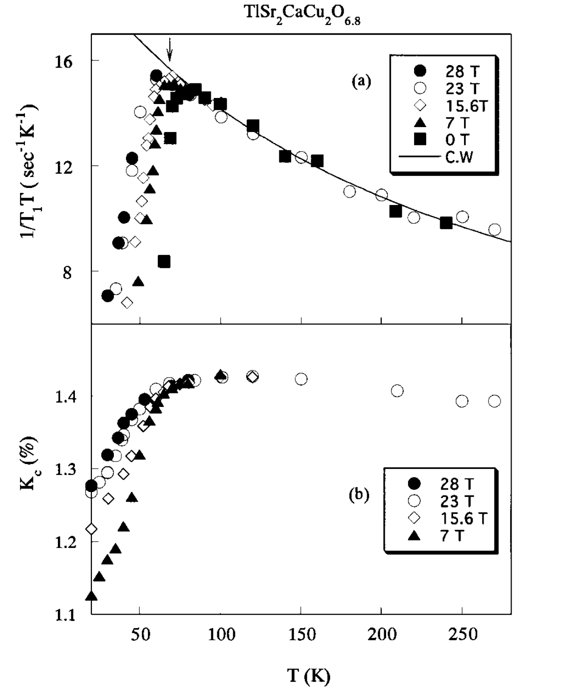

FIG. 2. (a) $1/T_1T$ of ^{63}Cu as a function of temperature T at various magnetic fields parallel to c axis. The arrow indicates T_{c0}. The curve is a fitting of the data above $T = 90$ K to a Curie-Weiss relation. (b) T variation of the Knight shift, K_c, at various fields. The typical uncertainty of the data points is about $\pm 2\%$, which is slightly larger than the size of the symbols.

K_c divided by $\sqrt{T_{c0} - T_{cH}}$ is plotted against the same normalized temperature difference. A scaling relation is also evident. In both cases, the scaling has the same dependence on $\frac{T - T_{cH}}{T_{c0} - T_{cH}}$.

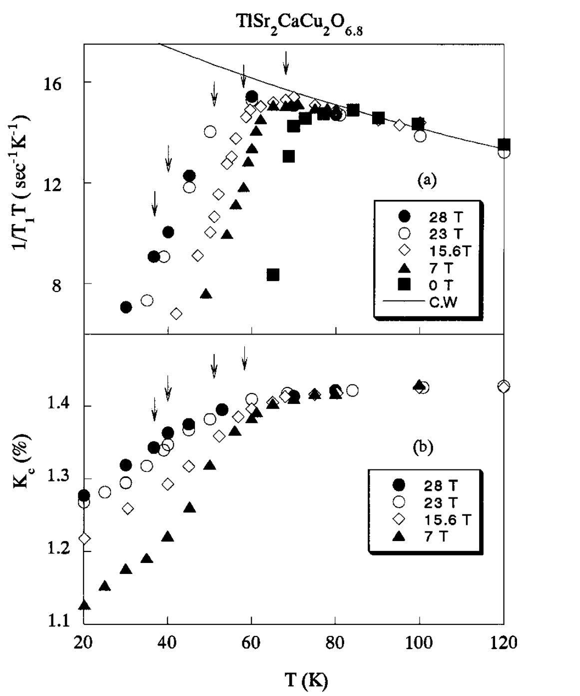

FIG. 3. Enlarged part of Fig. 2 around T_{cH}. The arrows, from right to left, indicate T_{cH} at elevated fields.

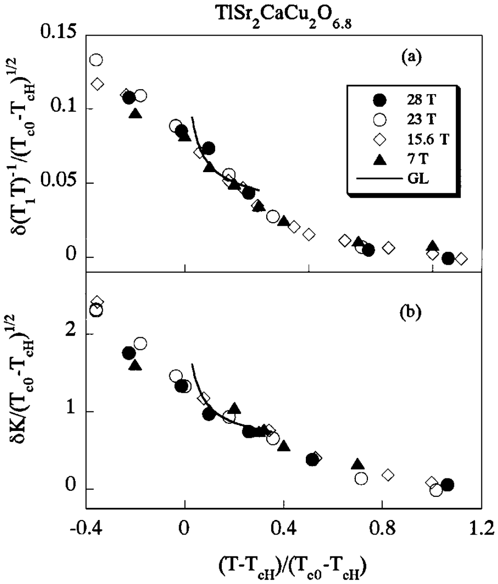

FIG. 4. (a) Normalized reduction of $1/T_1T$ from that expected from the high-T Curie-Weiss relation, divided by $\sqrt{T_{c0} - T_{cH}}$, in $K^{-1/2}$, plotted as a function of $\frac{T-T_{cH}}{T_{c0}-T_{cH}}$. (b) Reduction of the Knight shift from the constant value, divided by $\sqrt{T_{c0} - T_{cH}}$, in 10^{-4} K$^{-1/2}$, versus $\frac{T-T_{cH}}{T_{c0}-T_{cH}}$. The symbols are the same as those in (a). The curves are fits to $\frac{N_{\rm CP}}{\sqrt{T_{c0}-T_{cH}}}$ (see text).

We now argue that these scaling relations are consistent with the effects of the fluctuating Cooper pair density. The spin Knight shift is written as $K_s \propto \chi_s$; $1/T_1T$ can be written as $1/T_1T \propto \sum_q \frac{\mathrm{Im}\chi(q,\omega)}{\omega}|_{\omega\to 0} \simeq \chi_s \sum_{q\sim Q} \frac{\xi_M^4}{(1+q^2\xi_M^2)^2}$, where Im means imaginary part, ξ_M is the magnetic correlation length, and Q is the AF wave vector [12,13]. χ_s is proportional to the density of states (DOS) or the number of the normal-state electrons. SCF modify χ_s, and also ξ_M in general. The effects of SCF on various physical quantities have been extensively studied in the past in conventional superconductors [16] and recently also in high-T_c superconductors [17]. Three processes are known: (1) the Aslamazov-Larkin (AL) term, which is a direct effect of the Cooper pairs formed above mean-field T_c [18], (2) the Maki-Thompson (MT) term, which is due to the coherent scattering of two counterparts of a Cooper pair on the same elastic impurities [19], and (3) the DOS term due to the reduction of one-electron DOS because a part of electrons form Cooper pairs [20,21]. The AL contribution to the Knight shift and the relaxation is negligible because of singlet electron pairing. The MT term is sensitive to the pairing symmetry, but the DOS term is not. When the Cooper pair is of d-wave symmetry, which is believed to be realized in the high-T_c superconductors, the MT term is much smaller than the DOS term [22,23]. Under these circumstances, the predominant effect of the SCF on K and $1/T_1$ is to reduce the contributions due to the DOS.

This reduction can be modeled by calculating the corresponding number of the fluctuating Cooper pairs, $N_{\rm CP}$, which gives rise to the reduction of the DOS. The static fluctuation of the Cooper pair density above T_{cH} can be estimated from the Ginsburg-Landau (GL) theory. The GL free energy density relative to the normal state is $f = \alpha|\psi|^2 + \frac{1}{2m^*}|(\frac{\hbar}{i}\nabla - \frac{e^*}{c}\vec{A})\psi|^2 + \frac{\beta}{2}|\psi|^4$, where ψ is the order parameter, α and β are constants, and m^* and e^* are the mass and charge of the Cooper pair, respectively. The probability for each $\psi(r)$ is proportional to $\exp(-f/k_BT)$. Consider T far enough above T_c that the $|\psi|^4$ term can be neglected. Suppose $H \parallel z$ and expand ψ in terms of the wave function of the Landau orbit φ_{n,k_z}, $\psi(r) = \sum C_{n,k_z}\varphi_{n,k_z}$. It can be shown [16] that $f = \sum \frac{\hbar^2}{2m^*}[k_z^2 + \frac{1}{\xi^2} + (n + \frac{1}{2})\frac{4\pi H}{\phi_0}]|C_{n,k_z}|^2$, where ϕ_0 is the flux quantum and $\xi \equiv \sqrt{\frac{\hbar^2}{2m^*\alpha}} \equiv \xi_0/\sqrt{\varepsilon}$ is the GL coherence length, with $\varepsilon = \log(T/T_{c0}) \approx \frac{T-T_{c0}}{T_{c0}}$. Therefore, the averaged fluctuation is $\langle|\psi_{k_z,n}|^2\rangle = \frac{k_BT}{\frac{\hbar^2}{2m^*}[k_z^2+\frac{1}{\xi^2}+(n+\frac{1}{2})\frac{4\pi H}{\phi_0}]}$, where n labels the Landau level. By introducing $\varepsilon_H = \frac{T-T_{cH}}{T_{c0}}$, where T_{cH} is the mean-field transition temperature at field H, and $\tilde{H} = H/H_{c2}(0)$, the averaged fluctuation becomes

$$\langle|\psi_{k_z,n}|^2\rangle = \frac{k_BT}{\frac{\hbar^2}{2m^*\xi_0^2}[\varepsilon_H + \xi_0^2k_z^2 + 2n\tilde{H}]}. \qquad (1)$$

The factor of T shown in Eq. (1) is canceled out when one includes the effect of dynamic fluctuations (nonzero frequencies). Dynamical fluctuations suppress the order parameter modulus. This suppression is larger for higher T [16,20,23]. Calculating these fluctuations is rather elaborate and has not been derived analytically. However, by using the Matsubara Green's function and numerical calculation, Heym [20] found that the inclusion of dynamical fluctuations is equivalent to multiplying the static fluctuation term by the factor $1/T$ in the temperature range of $1.05 \le T/T_c \le 1.6$, which corresponds to $0.1 \le \frac{T-T_{cH}}{T_{c0}-T_{cH}} \le 1$ in the present case. On the basis of this argument, we drop the factor T in Eq. (1) from further consideration. Then, $N_{\rm CP}$ is

$$N_{\rm CP} = \sum_k\langle|\psi_k|^2\rangle = \int \frac{dk_z}{2\pi}\frac{H}{\phi_0}\sum_n\langle|\psi_{k_z,n}|^2\rangle$$
$$\propto \sum_n \frac{\tilde{H}}{\sqrt{\varepsilon_H + 2n\tilde{H}}}. \qquad (2)$$

By dividing the two sides of Eq. (2) by $\sqrt{\tilde{H}}$, one obtains a scaling relation between $N_{\rm CP}/\sqrt{\tilde{H}}$ and $\varepsilon_H/\tilde{H}$. By noting that $H_{c2}(T)$ is linear near T_c so that $\tilde{H}$ can be written as $\tilde{H} = \frac{1}{0.69}\frac{T_{c0}-T_{cH}}{T_{c0}}$ [10], we obtain

$$\frac{N_{\rm CP}}{\sqrt{T_{c0} - T_{cH}}} \propto \sum_n \frac{1}{\sqrt{\frac{T-T_{cH}}{T_{c0}-T_{cH}} + \frac{2n}{0.69}}}. \qquad (3)$$

Equation (3) reproduces the scaling found experimentally. The solid curves in Fig. 4 are fits to the right-hand side of Eq. (3) by taking $n_{\max} = 1/\tilde{H} = 5$ following Ref. [24]. Taking larger $n_{\max}$ is found to result only in a shift of the fitted region to lower temperature, as found by Eschrig *et al.* [23]. We have limited the fitting to the vicinity of T_{cH} where the GL theory is valid. In the range of

$0.1 \leq \frac{T-T_{cH}}{T_{c0}-T_{cH}} \leq 0.35$, the fitting is reasonably good. The deviation from the GL theory near (and below) T_{cH} is not surprising since the fluctuations there (i.e., in the critical region) are strong so that the $|\psi|^4$ term cannot be neglected.

Thus, the H and T dependences of $1/T_1T$ and the Knight shift can be understood as due to Cooper pair density fluctuations of both static and dynamic origins. The reduction of both K and $1/T_1T$ are proportional to N_{CP}, as expected by the theory to the first order of approximation (i.e., neglecting the change in ξ_M). The H-enhanced fluctuation near T_{cH} is attributed to the increase of the density of the fluctuating pairs due to the degenerated Landau level.

To our knowledge, this is the first report of the observed scaling relation for the NMR quantities. It is observed over a wide T range that extends well below T_{cH} and up to as high as $2T_{cH}$. Although the above simplified argument gives an intuitive account for the scaling above (and near) T_{cH}, theories which can describe the whole T range including the critical and the high-T regions are needed.

Finally we discuss the implications of the present finding. First, the present experiment warns that care should be taken when making quantitative arguments about the PG phenomenon under magnetic field. As seen in our experiment, applying a magnetic field gives a result that is consistent with the quantization of the orbital motion of the electrons which in turn lowers T^* and makes the PG appear more pronounced than at zero field. Second, the PG behavior in the overdoped regime contrasts sharply with what is seen in the underdoped material $YBa_2Cu_4O_8$ reported previously, where no field dependence was found up to 23.2 T, even though the field reduces T_c by 20 K (26% of T_{c0}) [3]. There are a wide variety of interpretations for the PG [25] and its H dependence [26]. The present findings imply that any electron correlation-driven pseudogap, if realized in the underdoped regime, would terminate at some doping level [27], before entering the overdoped regime.

In summary, we have found in the slightly overdoped superconductor $TlSr_2CaCu_2O_{6.8}$ that the reduction of the Knight shift and $1/T_1T$ above T_c, i.e., the pseudogap behavior, is strongly field dependent and follows a simple scaling relation. Based on the GL theory we argued that this scaling is consistent with the effects of the Cooper pair fluctuations above mean-field T_{cH}. The present results contrast sharply with the pseudogap property in the underdoped regime, which is not affected by a field up to 23.2 T [3]. These results imply that an electron correlation-driven pseudogap would terminate at some doping level before entering the overdoped regime.

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