Low-energy spin dynamics and critical hole concentrations in $La_{2-x}Sr_xCuO_4$ ($0.07 \leqslant x \leqslant 0.2$) revealed by ^{139}La and ^{63}Cu nuclear magnetic resonance

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(Received 7 July 2017; revised manuscript received 6 September 2017; published 18 September 2017)

We report a comprehensive ^{139}La and ^{63}Cu nuclear magnetic resonance study on $La_{2-x}Sr_xCuO_4$ ($0.07 \leqslant x \leqslant 0.2$) single crystals. The ^{139}La spin-lattice relaxation rate $^{139}T_1^{-1}$ is drastically influenced by Sr doping x at low temperatures. A detailed field dependence of $^{139}T_1^{-1}$ at $x = 1/8$ suggests that charge ordering induces the critical slowing down of spin fluctuations toward glassy spin order and competes with superconductivity. On the other hand, the ^{63}Cu relaxation rate $^{63}T_1^{-1}$ is well described by a Curie-Weiss law at high temperatures, yielding the Curie-Weiss temperature Θ as a function of doping. Θ changes sharply through a critical hole concentration $x_c \sim 0.09$. x_c appears to correspond to the delocalization limit of doped holes, above which the bulk nature of superconductivity is established.

DOI: 10.1103/PhysRevB.96.094519

I. INTRODUCTION

$La_{2-x}Sr_xCuO_4$ (LSCO) is a prototype of the high-T_c cuprates that exhibits very rich structural, magnetic, and electronic phases whose understanding is believed to provide a key to understanding high-T_c mechanism.

Despite intensive studies on LSCO for three decades, however, there are still nontrivial issues that are not fully understood. For example, the coexistence of magnetic and superconducting orders [1–5] and an inhomogeneous superconducting state [6] in the underdoped region have been known, but their underlying mechanism remains elusive. The strongly enhanced magnetic order [7–9] observed near a hole concentration of $x = 1/8$ is particularly interesting, as it appears to be linked to the stripe or the charge density wave (CDW) instability [10–12]. Recently, the CDW in $La_{2-x}Sr_xCuO_4$ was actually detected near $x = 1/8$ by x-ray diffraction measurements [13–15], and a high-field study of the Seebeck coefficient [16] reported that the CDW modulations cause the Fermi surface reconstruction in the limited doping range $0.085 < x < 0.15$, whose onset peaks at near $x = 1/8$. It is noteworthy that the lowest doping limit $x = 0.085$ for the CDW modulations agrees with the extrapolated value at zero temperature for the CDW onset probed by hard x-ray measurement [13].

With these latest experimental observations, several questions naturally arise: (1) how the CDW and superconductivity are related, (2) whether or how the CDW is coupled to the enhanced spin order observed near $x = 1/8$, and (3) why the CDW modulation appears at a considerably larger doping than the known critical doping $x \sim 0.05$ for superconductivity [17], above which drastic changes in the magnetic properties are observed [18–20].

In this paper, we carried out ^{139}La and ^{63}Cu spin-lattice relaxation rate (T_1^{-1}) measurements in superconducting $La_{2-x}Sr_xCuO_4$ ($0.07 \leqslant x \leqslant 0.2$), in order to investigate how the normal and superconducting properties in $La_{2-x}Sr_xCuO_4$ evolve as a function of doped holes. Based on the temperature and doping dependence of the ^{139}La relaxation rate, we propose that the unusual glassy behavior observed near $x = 1/8$ may be considered as a fingerprint of charge order. The ^{63}Cu relaxation measurements imply the presence of a critical hole concentration $x_c \sim 0.09$, above which superconductivity emerges in a full volume fraction and the Curie-Weiss temperature Θ shares a similar doping dependence with T_c.

II. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

The $La_{2-x}Sr_xCuO_4$ single crystals were grown with the traveling solvent floating zone method, as described in Ref. [21]. We confirmed the superconducting (SC) transition temperature T_c from the onset of the drop of the *in situ* ac susceptibility χ_{ac} in the nuclear magnetic resonance (NMR) tank circuit, and the resultant values are in good agreement with SQUID measurements. For $x = 0.07$, a drop of χ_{ac} was not detected down to 4.2 K, in contrast to the sharp SC transition observed for $x \geqslant 0.1$ (see the inset of Fig. 1). This indicates that superconducting volume fraction is very small at $x = 0.07$, but it becomes abruptly 100% as Sr doping exceeds a critical value x_c that is estimated to be 0.09 (see Fig. 1).

^{139}La ($I = 7/2$) and ^{63}Cu ($I = 3/2$) NMR measurements were performed on $La_{2-x}Sr_xCuO_4$ single crystals with $x =$ 0.07, 0.1, 0.125, 0.15, and 0.2 in the range of temperature 4.2–420 K and in an external field H that ranges from 6 to 16 T. The crystallographic c axis of the samples were aligned along the applied field direction using a goniometer. The spin-lattice relaxation rates T_1^{-1} were measured at the central transition ($+1/2 \leftrightarrow -1/2$) of both nuclei by monitoring the recovery of the echo signal after a saturating single $\pi/2$ pulse. Then the relaxation data were fitted to the appropriate fitting functions. For the ^{63}Cu,

$$1 - \frac{M(t)}{M(\infty)} = A(0.1e^{-(t/T_1)^\beta} + 0.9e^{-(6t/T_1)^\beta}), \quad (1)$$

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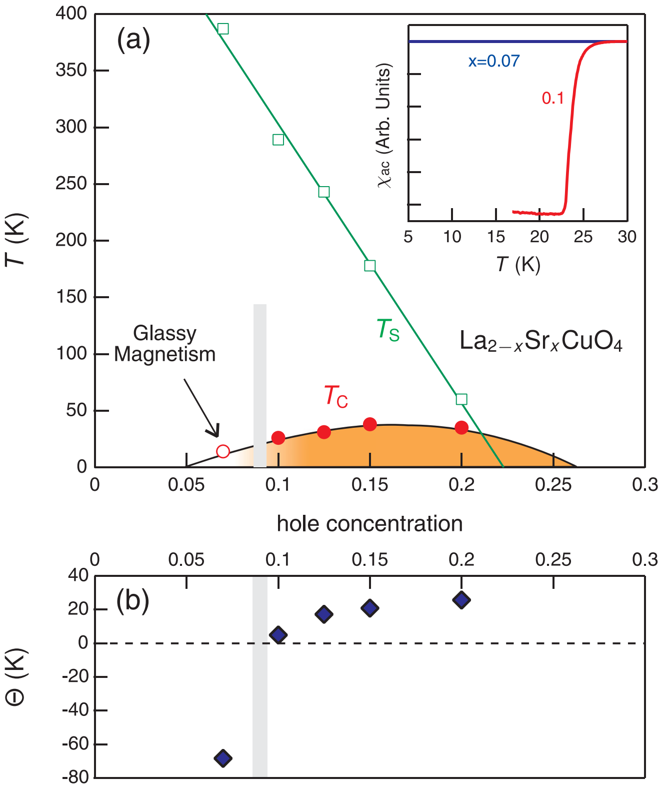

FIG. 1. (a) x-T phase diagram of $La_{2-x}Sr_xCuO_4$. T_S and T_c were all determined by our NMR measurements, except T_c for $x = 0.07$ which was determined by a weak drop in dc susceptibility measurement. As clearly shown in the inset, the SC transition for $x = 0.07$ was not observed in the *in situ* ac susceptibility χ_{ac}, in contrast to the sharp SC transition for $x = 0.1$. (b) The Curie-Weiss temperature Θ extracted from $^{63}T_1^{-1}$ measurements as a function of doping x. Above $x_c = 0.09$, Θ abruptly changes and quickly approaches a constant value.

and, for the ^{139}La,

$$1 - \frac{M(t)}{M(\infty)} = A\left(\frac{1}{84}e^{-(t/T_1)^\beta} + \frac{3}{44}e^{-(6t/T_1)^\beta} + \frac{75}{364}e^{-(15t/T_1)^\beta} + \frac{1225}{1716}e^{-(28t/T_1)^\beta}\right), \quad (2)$$

where M is the nuclear magnetization and A is a fitting parameter that is ideally one. β is the stretching exponent, which is less than unity when T_1^{-1} becomes spatially distributed due to glassy spin freezing.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. ^{139}La NMR

Figure 2 shows the temperature and doping dependence of $^{139}(T_1T)^{-1}$ measured at 10.7 T. The sharp anomaly of $^{139}(T_1T)^{-1}$ is associated with the high-temperature tetragonal (HTT) to low-temperature orthorhombic (LTO) structural transition, giving rise to the HTT → LTO transition temperature T_S which rapidly decreases with decreasing x. The background $(T_1T)^{-1}$ is nearly independent of x [solid horizontal line in Fig. 2(a)] in a wide temperature range except an enhancement

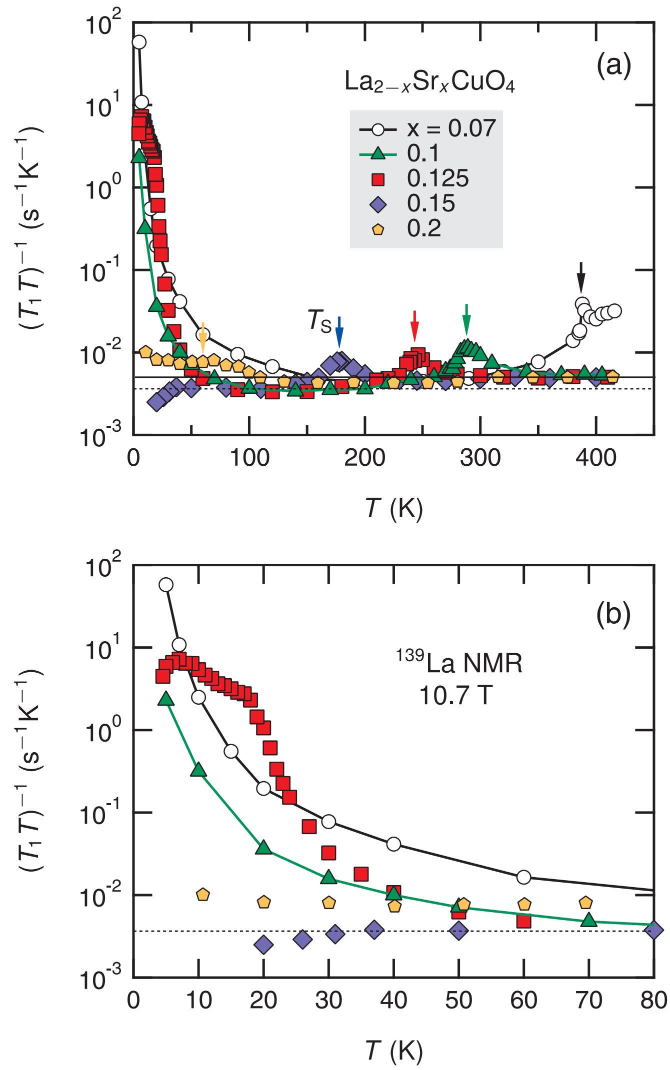

FIG. 2. (a) Temperature dependence of $^{139}(T_1T)^{-1}$ as a function of doping x measured at 10.7 T. The structural transition temperature T_S is identified from the sharp peak of $^{139}(T_1T)^{-1}$ and denoted by the down arrows. The background $^{139}(T_1T)^{-1}$ (solid horizontal line) is nearly independent of x, which is slightly reduced below T_S (dotted horizontal line). (b) At low temperatures, $^{139}(T_1T)^{-1}$ drastically changes depending on x. The prominent enhancement of $^{139}(T_1T)^{-1}$ for $x = 1/8$ is clearly shown.

observed at $T > 350$ K for $x = 0.07$ (see Ref. [21] for discussion regarding a possible origin of the enhancement). The origin of the constant $^{139}(T_1T)^{-1}$ is ascribed to the quadrupolar relaxation process due to the fluctuating electric field gradient (EFG) [22]. Interestingly, after undergoing the structural transitions at T_S, the constant value of $^{139}(T_1T)^{-1}$ drops slightly to another constant (dotted horizontal line). The small change of $^{139}(T_1T)^{-1}$ subsequent to the HTT → LTO transition is accounted for by a local tilting of the EFG with respect to the c axis [21], that is, by the misalignment between the nuclear quantization axis and the applied field direction.

In the low temperature region, the temperature and doping dependence of $^{139}(T_1T)^{-1}$ becomes extremely strong and complicated, as shown in Fig. 2(b). For $x = 0.07$, $^{139}(T_1T)^{-1}$ is enhanced by more than three decades with decreasing temperature, representing the critical slowing down of SFs toward spin order [21]. As x is increased, this $^{139}(T_1T)^{-1}$ enhancement is greatly suppressed by more than an order of magnitude at $x = 0.1$ and disappears completely at optimal doping $x = 0.15$. Strangely, $^{139}(T_1T)^{-1}$ is enhanced below T_c for slightly overdoped $x = 0.2$, suggesting that an unusual spin dynamics arises at $x \sim 0.2$. We will return to this issue briefly in Sec. III B.

A predominant feature found in Fig. 2(b) is the strong enhancement of $(T_1T)^{-1}$ with 1/8 doping, that is distinct from the data obtained at nearby dopings. Deviating at ~75 K with respect to the temperature independent value, $(T_1T)^{-1}$ rises sharply until it bends over at ~20 K. Interestingly, it continues to increase before it drops abruptly at ~8 K. Note that the stretching exponent β in Eq. (2) starts to deviate from unity below ~75 K, as shown in Fig. 3(b). A β value less than unity, which indicates a spatial distribution of T_1^{-1}, is a key characteristic when T_1^{-1} is strongly enhanced due to glassy spin freezing as observed $x = 0.07$. Therefore, from the temperature dependence of T_1^{-1} as well as β, one can conclude that SFs are inhomogeneously slowed down below ~75 K at $x = 1/8$, being connected with the enhanced glassy spin order detected in LSCO:0.12 [4,8,9,23,24].

Such a spin-glass behavior at $x = 1/8$ is surprising, because it is caused by randomness that is usually absent in a metallic system. In lightly doped cuprates, the glassy behavior could be understood by randomly localized doped holes [25–27]. However, near $x = 1/8$, doped holes are largely delocalized and thus the origin of glassy spin order in LSCO:1/8 is hardly understood via the same mechanism as in the very underdoped regime. Furthermore, it was demonstrated that quenched disorder is not responsible for the glassy behavior in LSCO:0.12 [9], suggesting the relevance of 1/8 anomaly for the glassiness. Therefore, we conjecture that charge stripe order, although it may be still rapidly fluctuating on the NMR time scale (~μs), may generate the randomness [28,29] (e.g., localized holes) that could inhomogeneously slow down SFs.

In order to check whether the inhomogeneous slowing down of SFs and charge order are related, we performed the field dependence of $^{139}T_1^{-1}$ for LSCO:1/8, as shown in Fig. 3. Clearly, the onset temperature at which T_1^{-1} deviates from the T-linear behavior and β deviates from unity is robust against the external field strength. Taken together with a recent NMR work in LBCO:1/8 which strongly indicated that the slowing down of SFs occurs at the charge ordering temperature [30], we believe that the anomalous change of both T_1^{-1} and β at ~75 K is triggered by the charge ordering that is independent of H [31]. Remarkably, $T_{\mathrm{CO}} \sim 75$ K is in excellent agreement with the static CDW ordering temperatures, 75 K (Ref. [13]) and 85 K (Ref. [15]) for $x = 0.12$ detected by x-ray diffraction studies.

Taking it for granted that charge ordering induces the inhomogeneous slowing down of SFs for 1/8-doped La cuprates, NMR might further probe the interplay between charge order and superconductivity. For that, we examine how the T dependence of T_1^{-1} is influenced by superconductivity [see Fig. 3(a)]. At sufficiently high fields $\geqslant 13$ T, i.e., when superconductivity is nearly suppressed, it turns out that the high temperature side of the T_1^{-1} peak is independent of H.

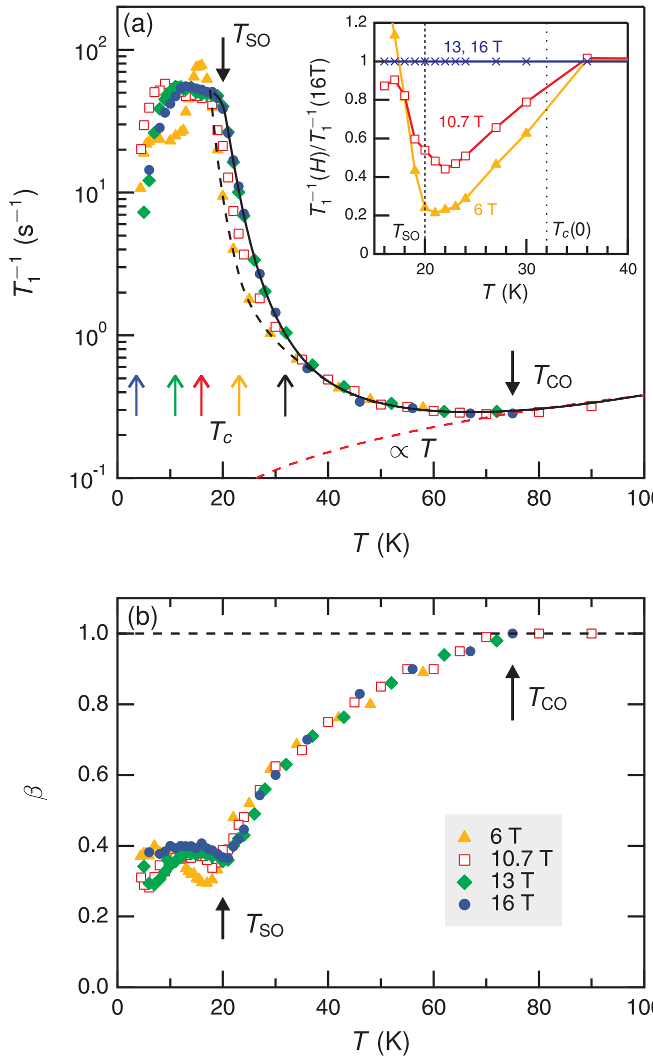

FIG. 3. (a) $^{139}T_1^{-1}$ versus T as a function of H in LSCO:1/8 shows a strong field dependence. In particular, the $^{139}T_1^{-1}$ upturn is suppressed with decreasing H, i.e., with increasing T_c which is denoted by the up arrows. Inset: $T_1^{-1}(H)$ divided by T_1^{-1} at $H = 16T$. The significant suppression of T_1^{-1} below $T_c(H = 0)$ is clearly shown. (b) Stretching exponent β versus T in LSCO:1/8. The deviation of β from one occurs near 75 K. Clearly, $\beta(T)$ is almost independent of H except for the superconducting region.

On the other hand, we find that the T_1^{-1} upturn is clearly suppressed with decreasing field, i.e., with increasing T_c. We interpret that this behavior reflects a competition between charge order and superconductivity. If so, an immediate question is then why the reduction of the T_1^{-1} upturn in Fig. 3 occurs well above the bulk $T_c(H)$. The answer may be the presence of in-plane SC correlations above $T_c(H)$ which have been shown to persist up to $T_c(H = 0)$ for moderate fields [32–35]. This idea is substantiated by the fact that T_1^{-1} is unaffected by external field above T_c at zero field (the black up arrow in Fig. 3).

In comparison, as shown in Fig. 3(b), β is nearly independent of the magnetic field above T_{SO} unlike T_1^{-1}. This indicates that although charge order itself competes with superconductivity, the spatial spin/charge inhomogeneity is robust against superconductivity. When spins order below T_{SO}, however, T_1^{-1} as well as β shows a significant magnetic field dependence. While one may suspect that it arises from the interplay between superconductivity and spin order, it is unfortunate that the complex field dependence of T_1^{-1} and β does not allow a quantitative analysis. Regardless, our data reveals that there is a peculiar interplay of superconductivity and spin order.

B. ^{63}Cu NMR

The temperature and doping dependence of $^{63}T_1^{-1}$ measured at 8.2 T is shown in Fig. 4(a). For $x = 0.07$, the relaxation is extremely fast, and the signal intensity rapidly decreases with decreasing T due to the *wipeout effect*. As x is increased to a slightly larger doping 0.1, $^{63}T_1^{-1}$ is significantly reduced while roughly maintaining its overall temperature dependence. With further increasing doping, we find that $^{63}T_1^{-1}$ becomes nearly insensitive to x, especially at high temperatures where $^{63}T_1^{-1}$ is independent of temperature.[1]

If the spin-lattice relaxation is dominated by the staggered susceptibility $\chi(q = Q)$, where Q is the AFM wave vector, it could be described by a Curie-Weiss law, i.e., $(T_1T)^{-1} \propto \chi(Q) = C/(T + \Theta)$ [36,37], where C is the Curie constant and Θ the Curie-Weiss temperature. To confirm this behavior, we plot T_1T versus T as shown in Fig. 4(b). Indeed, at all dopings, a CW-like T linear behavior was observed at high temperatures (>250 K). The linear fits with the CW law in the high T region give rise to Θ. Here the Curie constant $C \propto J(J + 1)$ was fixed, assuming the doping independent effective spin moment J. The doping dependence of Θ is drawn in Fig. 1. It shows that Θ parallels with T_c as a function of doping in the region of $0.1 \leqslant x \leqslant 0.2$, while it changes abruptly for $x = 0.07$ where superconductivity was not detected by χ_{ac} (see the inset of Fig. 1). It should be noted that the strong wipeout effect at $x = 0.07$ is similar to that observed in the underdoped region of $YBa_2Cu_3O_y$ (YBCO) below a critical hole concentration $p_c \sim 0.1$, in which the spin-glass phase coexists with superconductivity [38]. This implies the existence of a critical hole concentration $x_c \sim 0.09$ below which glassy magnetism dominates over superconductivity, and above which AFM spin correlations increase sharply and superconductivity fully replaces the magnetic volume fraction of the sample. Interestingly, the ^{139}La relaxation rate $^{139}T_1^{-1}$ also shows an abrupt change at $T > 300$ K as x is increased through x_c—$^{139}T_1^{-1}$ is strongly enhanced for $x = 0.07$ with increasing temperature, being clearly distinguished from the data for $x \geqslant 0.1$ (see Fig. 2).

The plot of $^{63}(T_1T)^{-1}$ vs T shown in Fig. 4(c) reveals another interesting feature. $^{63}(T_1T)^{-1}$ increases with decreasing T, but drops below ~50 K (>T_c) forming a clear peak

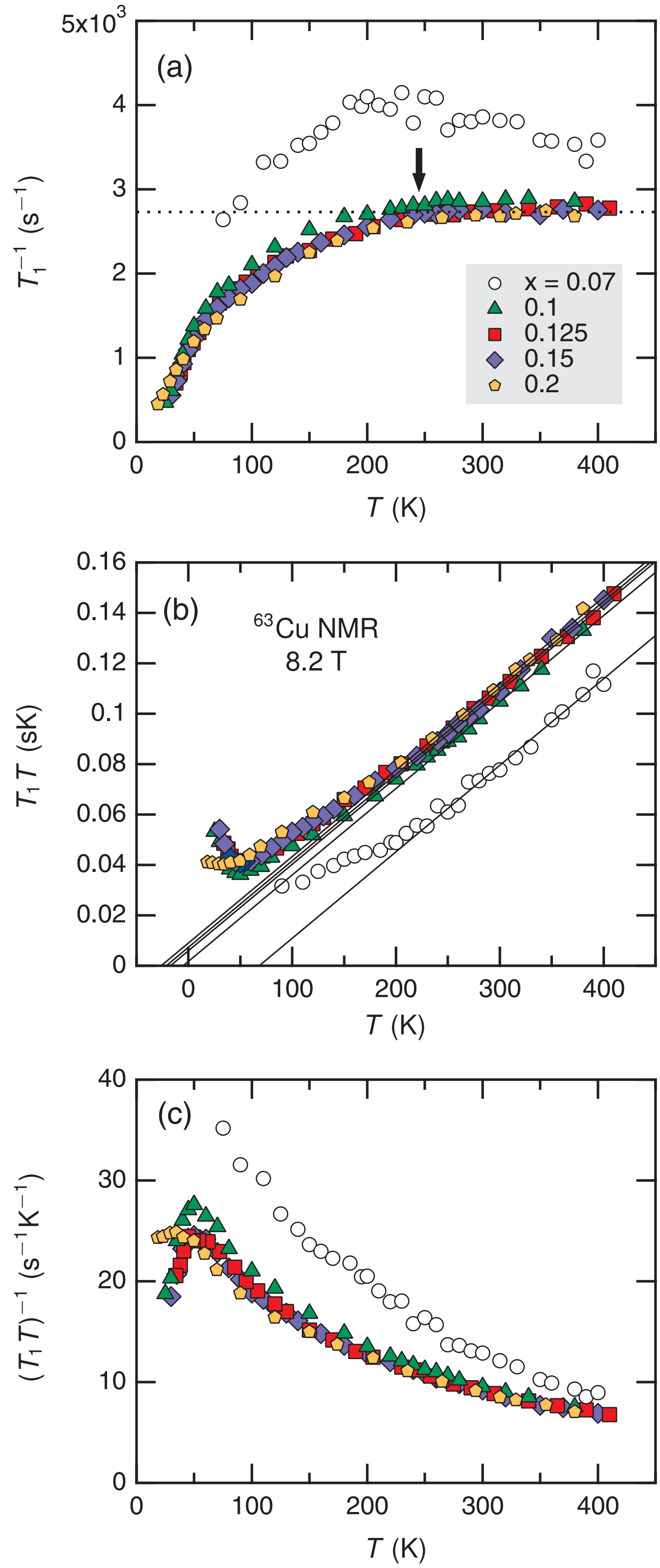

FIG. 4. (a) ^{63}Cu T_1^{-1} versus T for different x measured at 8.2 T applied along the c axis. T_1^{-1} at $x = 0.07$ is clearly distinct from other data for $x \geqslant 0.1$ which show almost x independent behavior. (b) Linear fits of T_1T at high temperatures yield the Curie-Weiss temperature Θ which abruptly changes near the critical doping $x_c \sim 0.09$. (c) $^{63}(T_1T)^{-1}$ vs T. For $0.1 \leqslant x \leqslant 0.15$, $^{63}(T_1T)^{-1}$ forms a clear peak at the same temperature ~50 K. At $x = 0.2$, the peak is notably suppressed.

[1]Our ^{63}Cu NMR results are in marked contrast to the early results obtained in powder samples two decades ago [36,37,39] in which the T dependence of $^{63}T_1^{-1}$ progressively changes with increasing x up to the overdoped region. We believe that the previous NMR studies were strongly affected by substantial disorder in the samples, such as excess oxygen which is known to affect $^{139}T_1^{-1}$ above 200 K in underdoped LSCO [48] and/or Sr impurity disorder [49].

that is insensitive to x in the range $0.1 \leqslant x \leqslant 0.15$, which is qualitatively consistent with previous results [36,37,39]. The origin of the $^{63}(T_1T)^{-1}$ peak is unclear, but it may be either the rapid reduction of the AFM correlation length or a spin gap opening at low temperatures [39].

The peak is, however, notably suppressed and moves to lower temperature at $x = 0.2$. The abrupt suppression of the $^{63}(T_1T)^{-1}$ peak at $x = 0.2$ is actually consistent with a previous ^{63}Cu NQR study in the powder sample [37]. Remarkably, at the similar doping $x \sim 0.2$, the in-plane resistivity ρ_{ab} reveals a critical behavior [40]. ρ_{ab} shows a T-linear behavior at all temperatures near $x = 0.2$ just after the pseudogap vanishes at $x = 0.19$, while it approaches purely T^2 behavior with either increasing or decreasing x [40]. Furthermore, as x exceeds roughly 0.2, the system exhibits a Curie-like paramagnetism [41–43] and a drastic increase of the residual term $\gamma(0)$ of the specific heat [44]. Therefore, we conclude that the suppression of the $^{63}(T_1T)^{-1}$ peak at $x = 0.2$ reflects the intrinsic change of the physical properties of the system. A plausible explanation could be that the quasiparticle scattering mechanism critically changes beyond $x \sim 0.2$ [40], giving rise to an unusual spin dynamics at low energies. In fact, this could naturally account for the peculiar upturn of $^{139}(T_1T)^{-1}$ in the SC state observed at $x = 0.2$ (see Fig. 2).

It is interesting to note that the T dependence of $^{63}(T_1T)^{-1}$ remains nearly intact at low temperatures for $x \geqslant 0.1$, in stark contrast to the drastic change of that of $^{139}(T_1T)^{-1}$, in particular for $x = 1/8$. It may be due to the fact that the relaxation of the ^{63}Cu is three orders of magnitude faster than that of the ^{139}La at high temperatures [see Figs. 2(a) and 4(c)], and thus the ^{63}Cu relaxation is not strongly affected by the slowing down of SFs associated with glassy spin freezing. In support of this, the stretching exponent β for the ^{63}Cu in Eq. (1) remains close to unity down to low temperatures for $x \geqslant 0.1$.

C. Phase diagram of $La_{2-x}Sr_xCuO_4$

Figure 1 shows a T-x phase diagram determined from our NMR results. From the abrupt change of Θ (lower panel in Fig. 1), one could identify a critical hole concentration $x_c \sim 0.09$. Whereas the glassy magnetism dominates at $x < x_c$, causing the extremely fast upturn of $^{139}T_1^{-1}$ (Fig. 2) and the strong wipeout of the ^{63}Cu signal at low temperatures, it is significantly suppressed and superconductivity soddenly appears in a full volume fraction as x exceeds x_c. The underlying cause for the critical behavior at $x_c \sim 0.09$ may be attributed to the localization limit of doped holes. For $x < x_c$, doped holes are strongly localized and there are not enough carriers to be paired, accounting for the absence of the SC transition in χ_{ac} for $x = 0.07$. As x reaches x_c, the metal-insulator transition takes place providing free charge carriers [45].

The critical hole concentration $x_c \sim 0.09$ is very close to the value of $\sim$0.1 observed in YBCO [38], suggesting that it could be the universal limit of delocalization of doped holes in cuprates. Furthermore, the fact that x_c nearly coincides with the hole concentration $\sim$0.85 above which the Fermi surface reconstruction is induced by the CDW modulations [16] may suggest that the CDW and superconductivity are governed by the same criticality, despite the competing relationship between them inferred from the ^{139}La NMR.

The phase diagram further shows that the structural transition temperature $T_S(x)$ goes to zero at $x \sim 0.22$, which is in good agreement with previous studies [17,46]. It is worthwhile to mention that, although the structural phase boundary exists inside the SC dome, it was demonstrated that superconductivity remains intact even if the structural boundary is shifted to much higher doping level by Pr doping [47]. Nevertheless, the peculiar behavior of the spin-lattice relaxation rates observed at $x = 0.2$ suggests that the structural instability may have a large influence on the quasiparticle scattering process, resulting in an unusual low-energy spin dynamics.

IV. CONCLUSION

In summary, we presented ^{139}La and ^{63}Cu NMR studies in a series of $La_{2-x}Sr_xCuO_4$ single crystals in the range $0.07 \leqslant x \leqslant 0.2$. The unusual glassy behavior observed by the ^{139}La spin lattice relaxation $^{139}T_1^{-1}$ at $x = 1/8$ is ascribed to the randomness generated by charge order, which triggers the critical slowing down of spin fluctuations toward spin order. The field dependence of $^{139}T_1^{-1}$ further suggests that the charge order competes with superconductivity.

The ^{63}Cu spin lattice relaxation $^{63}T_1^{-1}$ at high temperatures is governed by the staggered susceptibility that obeys the Curie-Weiss law. Data show that the Curie-Weiss temperature Θ changes abruptly at the critical doping $x_c \sim 0.09$ and becomes weakly dependent on further doping in the optimally doped regime, yielding a similar doping dependence of Θ and T_c.

ACKNOWLEDGMENT

This work has been supported by the DFG Research Grant No. BA 4927/1-3.

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