The mechanism of high-$T_c$ superconductivity has been a major controversy throughout the past decade [1–3]. The complexity of the phase diagram for temperature $T$ and hole concentration $x$ makes it difficult to identify the key leading toward the superconducting mechanism. In 1995, Tranquada et al. [4–6] demonstrated that La$_{1.6-x}$Nd$_x$Sr$_x$CuO$_4$ ($x \approx 0.05$) exhibits charge-stripe order at $T_{\text{charge}} \approx 65$ K, followed by a spin-stripe order at somewhat lower temperature, $T_{\text{spin}} \approx 50$ K. The recovery of the stripe phase [7] has added a new feature to the already complex phase diagram. Initially, some researchers speculated that the stripe order was merely a by-product of the LTO-LTT (low temperature orthorhombic–low temperature tetragonal) structural phase transition and was extrinsic to the fundamental physics of high-$T_c$ superconductivity. However, more recently, spin-stripe order was observed in La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.12$) [8,9] and $x = 0.05$ [10] and La$_2$CuO$_{4+\delta}$ [11], two materials without a LTO-LTT structural phase transition. This suggests that the low energy (i.e., dynamic) incommensurate spin excitations previously observed for La$_{2-x}$Sr$_x$CuO$_4$ over the entire superconducting regime $0.05 \leq x \leq 0.25$ [12–14] may correspond to the dynamic formation of stripes. Furthermore, recent inelastic neutron scattering experiments have established similar incommensurability of spin excitations also in underdoped YBa$_2$Cu$_{3-x}$O$_6$ [15]. These developments raise new hope that understanding the mechanism of stripe instability and its dependence on $T$ and $x$ is the key to establishing a unified picture of the hole-doped CuO$_2$ planes [1–3].

Unfortunately, stripe order has proved elusive, escaping experimental detection by various techniques. For example, $^{63}$Cu nuclear spin-lattice relaxation rate $^{63}T_1$ [16,17] measured for another striped material with the LTT structure, La$_{1.88}$Ba$_{0.12}$CuO$_4$ [18], exhibits no critical divergence at the stripe transition. The origin of the elusive nature is in the glassey nature of the stripe transition [6,18,19]. In other words, the critical slowing down of spin-stripe fluctuations below $T_{\text{charge}}$ is more gradual than ordinary magnetic phase transitions involving only the spin degrees of freedom. As a consequence, the apparent critical temperature $T_{\text{spin}}$ of the spin-stripe order is lower for experimental probes with slower frequency scales, i.e., $T_{\text{spin}} \approx 50$ K for elastic neutron scattering ($\sim 10^{11}$ Hz) [4–6], $T_{\text{spin}} \approx 30$ K for muon spin rotation ($\mu$SR, $\sim 10^7$ Hz) [18], and $T_{\text{spin}} \approx 10–30$ K for NQR (nuclear quadrupole resonance, $\sim 10^6$ Hz) [17,20,21].

In this Letter, we propose a new experimental approach to investigate stripes that takes advantage of extreme sensitivity of $^{63}$Cu NQR to local charge distribution. We will demonstrate that one can accurately measure the charge-stripe order parameter in La$_{1.875}$Ba$_{0.125}$CuO$_4$ and (La,Nd,Eu)$_{1.88}$Sr$_{0.12}$CuO$_4$ based on the wipeout effects [22] of $^{63}$Cu NQR. The basic idea is quite simple. When charge-stripe order sets in at $T_{\text{charge}}$, rivers of hole-rich CuO chains separate three-leg CuO ladders without holes, resulting in spatial modulation of hole concentration between 0.5 and 0 holes per Cu atom [4]. This produces as much as $\sim 8$ MHz [23] of instantaneous spatial variation of the $^{63}$Cu NQR frequency $^{63}\nu_Q$, which is proportional to the electric field gradient [24] at $^{63}$Cu nuclei. This means that $^{63}\nu_Q$ is no longer well defined below $T_{\text{charge}}$ inside the slowly fluctuating striped domains. Moreover, charge order turns on low frequency spin fluctuations [6], and consequently the $^{63}$Cu nuclear spin-lattice and spin-spin relaxation rates $^{63}T_1$ and $^{63}T_2$, respectively, diverge in the striped domains. All of these effects should wipe out $^{63}$Cu NQR signals within stripe ordered domains even before the growth of static magnetic hyperfine fields changes the NQR frequency completely below 10 K [17,20]. Therefore, we expect that the wipeout fraction $F$ of $^{63}$Cu NQR signals (i.e., the fraction of the lost signal intensity) is a good measure of stripe order. In fact, from comparison with elastic neutron scattering data in La$_{1.88}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ we demonstrate that the wipeout fraction $F$ represents nothing but the order parameter for charge stripe. Applying the NQR approach to La$_{2-x}$Sr$_x$CuO$_4$, we demonstrate that a similar stripe instability exists over the entire underdoped superconducting regime $0.05 \leq x \leq 0.16$, but disappears right above the critical hole concentration $x_c = \frac{4}{5}$. We grew all of the ceramic samples by standard solid state reactions [17].
conducted the $^{63,65}$Cu NQR line shape measurements using the $90^\circ$–$180^\circ$ pulse sequence with a fixed separation time $t_{90-180}$, typically $t_{90-180} = \sim 10$–$12$ $\mu$sec. The $^{63}$Cu NQR intensity was estimated by integrating the Gaussian fit of the NQR line shape, with the overall magnitude corrected for the Boltzmann factor and for the spin echo decay time $T_2$ measured at the NQR peak. The latter process is essential for accurate measurements of the wipeout fraction $F$ because the spin echo decay changes dramatically at $T_{\text{charge}}$. The $^{63}$Cu NQR line shape is composed of the so-called $A$ line and $B$ line with the relative intensity of the latter proportional to $x$, in good agreement with earlier reports [23]. Since high precision measurements conducted for the $^{63}$Cu isotope enriched La$_{1.875}$Ba$_{0.125}$CuO$_4$ showed identical temperature dependence of the NQR intensity for the $A$ line and the $B$ line between 10 and 300 K, we will focus our attention on the behavior of the more intense $A$ line. The $^{63}$Cu NQR intensity decreases dramatically below $T_{\text{charge}}$ (i.e., the wipeout fraction $F$ increases from zero to a finite value), but the linewidth of the observable segments of the sample (HWHM = $\sim 0.8$–$1.8$ MHz) showed only a mild increase (up to $\sim 35\%$) with decreasing temperature with no dramatic change at $T_{\text{charge}}$.

In Fig. 1(a), we present the temperature dependence of the wipeout fraction $F$ of the $A$ line in La$_{1.875}$Ba$_{0.125}$CuO$_4$, La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, and La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_4$. Within our experimental uncertainties, $F$ is zero between 300 and 65 K (i.e., we detect NQR signals from the entire sample). However, both $^{63}P_Q$ and $F$ begin to increase abruptly below $T_{\text{charge}} = 65$ K. $^{63}P_Q$ increases $\sim 1$ MHz between 65 and 10 K showing a similar temperature dependence as $F$ [17], and almost the entire NQR signal disappears by 10 K. We recall that below 10 K, the entire $^{63}$Cu resonance intensity reappears as Zeeman perturbed NQR (NQR broadened by static magnetic hyperfine field) between 20 and 80 MHz [17,20]. Coincidence of $T_{\text{charge}} = 65$ K for all three materials with different LTO-LTT transition temperatures [4,19] ($\sim 65$ K for La$_{1.875}$Ba$_{0.125}$CuO$_4$ and La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, and $\sim 130$ K for La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_4$) indicates that the LTO-LTT structural transition is not the primary origin of the observed anomaly at $T_{\text{charge}}$. In fact, La$_{2-x}$Sr$_x$CuO$_4$ exhibits qualitatively the same temperature dependence of $F$ without the LTO-LTT structural phase transition, as presented in Fig. 1(b). Accordingly, we must attribute the observed wipeout to electronic effects, as discussed above in the third paragraph. In Fig. 1(a), we compare the wipeout fraction $F$ with the charge- and spin-stripe order parameter determined by elastic neutron scattering experiments [5]. (We note that by definition the elastic neutron scattering intensity represents the square of the order parameter.) The identical temperature dependence observed for NQR and neutron data for charge ordering allows us to identify the wipeout fraction $F$ as the charge-stripe order parameter. We note that none of the samples with wipeout effects exhibit divergence of $^{139}$La nuclear spin-lattice relaxation rate $^{139}1/T_1$ at $T_{\text{charge}}$ [17,25]. Instead, $^{139}1/T_1$ exhibits gradual upturn below $T_{\text{charge}}$, showing a hump below 30 K depending on the hole concentration [17,25]. This is evidence that Cu spin fluctuations do not slow down to NQR frequencies immediately at $T_{\text{charge}}$, in agreement with $\mu$SR [18], ESR [19], and earlier NQR line shape measurements [20,21]. We also found that the temperature dependence of the charge-stripe order parameter $F$ fits reasonably well to the weak coupling BCS theory with $\Delta = 115$ K. The spin-stripe ($\circ$) and charge-stripe ($\triangle$) order parameter in La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ [3] are also plotted. (b) The wipeout fraction $F$ in La$_{2-x}$Sr$_x$CuO$_4$ for $x = 0.125$ ($\triangle$), 0.115 ($\times$), 0.09 ($\bullet$), and 0.07 ($\square$). The curves are guides to the eyes.

FIG. 1. (a) The wipeout fraction $F$ of $^{63}$Cu NQR in La$_{1.875}$Ba$_{0.125}$CuO$_4$ (A), La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ ($\bullet$), and La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_4$ ($\times$). The solid curve represents the fit to the BCS weak coupling theory with $\Delta = 115$ K. The spin-stripe ($\circ$) and charge-stripe ($\triangle$) order parameter in La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ [3] are also plotted. (b) The wipeout fraction $F$ in La$_{2-x}$Sr$_x$CuO$_4$ for $x = 0.125$ ($\triangle$), 0.115 ($\times$), 0.09 ($\bullet$), and 0.07 ($\square$). The curves are guides to the eyes.
The sample with $x = 0.125$ ($T_c = 31$ K) showed pure Lorentzian spin echo decay below $T_{\text{charge}} = 50$ K [not shown, the spin echo decay in the striped phase of La$_{2-x}$Sr$_x$CuO$_4$ is semiquantitatively the same as those shown in Fig. 2(a) [17]]. This indicates that stripe fluctuations exist in the entire sample at 50 K. Approximately 40% of the NQR signal disappears by the superconducting transition $T_c$. The sample with $x = 0.115$ ($T_c = 31$ K) also shows the onset of charge-stripe order at $T_{\text{charge}} = 50$ K, and the striped fraction $F$ exhibits a plateau near $T_c$, where elastic incommensurate magnetic scattering also sets in [9]. The behavior of samples with smaller hole concentration, $x = 0.09$ ($T_c = 29$ K, $T_{\text{charge}} = 70$ K) and 0.07 ($T_c = 19$ K, $T_{\text{charge}} = 90$ K), is qualitatively similar to the case of (La,Nd,Eu)$_{1.88}$Sr$_{0.12}$CuO$_4$ and La$_{1.875}$Ba$_{0.125}$CuO$_4$, but the stripe transition is considerably broader. The broad transition may indicate that the stripe is unstable for $x < \frac{1}{4}$. Unfortunately, we cannot determine the real space structure of the stripe [7] based on wipeout effects alone. The Meissner fraction in $x = 0.125, 0.115, 0.09, 0.07$ [17] is comparable to the observable NQR fraction, $1 - F$, at $T_c$ for each sample. Whether superconductivity coexists with stripe, or microscopic phase separation sets in at $T_c$, is a matter of debate. However, it is certainly true that a canonical superconducting transition with a large specific heat jump at $T_c$ is realized only above $x = 0.125$ [31].

In contrast to the superconducting samples, the lightly hole-doped nonsuperconducting La$_{1.96}$Sr$_{0.04}$CuO$_4$ showed a gradual decrease of the NQR intensity below 300 K. However, the spin echo decay always has a Gaussian component as shown in Fig. 2(b), and there is no evidence for stripe fluctuations. This is consistent with the neutron scattering result that spin response is commensurate below $x = 0.05$ without the signature of spin stripes [10,13,32]. Since dc resistivity for $x = 0.04$ is large and shows a gradual upturn due to weak localization below 300 K [32], we attribute the gradual loss of the NQR signal to commensurate short-range spin order in the vicinity of randomly localized holes. In this context, it is worth recalling that even conventional dilute Kondo alloys such as Cu:Fe exhibit similar gradual wipeout ($F \sim \frac{1}{x}$) above the Kondo temperature [33].

In Fig. 3 we summarize the critical temperature $T_{\text{charge}}$ and order parameter $F$ as a function of $x$. Clearly, the ground state of La$_{2-x}$Sr$_x$CuO$_4$ exhibits a sharp crossover at the critical hole concentration $x_c = \frac{1}{8}$ from the striped state to the superconducting state with low energy ($\sim$meV) stripe fluctuations [12–14]. The charge-ordering temperature $T_{\text{charge}}$ is consistently higher than the spin-ordering temperature determined by elastic neutron scattering ($T_{\text{spin}} = 30$ K for $x = 0.12$ [9], and 17 K for $x = 0.05$ [10]), $\mu$SR and $^{139}$La NQR ($T_{\text{spin}} \sim 15$ K is the largest for $x = 0.115$ [17,18,21,25]). Perhaps remnant fluctuations of charge stripes prevent spins from freezing, resulting in the glassy transition. The fact that...
T_{\text{charge}} is much higher than T_{\text{spin}} in La_{2-x}Sr_{x}CuO_{4} may indicate that the charge stripes are significantly disordered. The previously undetected phase boundary of charge stripe in Fig. 3(b) explains why insulating (i.e., underdoped) La_{2-x}Sr_{x}CuO_{4} is much higher than La_{2-x}Sr_{x}CuO_{4}. The previously undetected phase boundary of charge stripe in Fig. 3(b) explains why insulating (i.e., underdoped) La_{2-x}Sr_{x}CuO_{4} are energetically very close. These observations as well as recent neutron data[4–6,8–14] are consistent with the mechanism of high-\(T_{\text{c}}\) behavior of charge stripe in Fig. 3(b) explains why insulating (i.e., underdoped) La_{2-x}Sr_{x}CuO_{4}. Filled (open) symbols denote the phase boundary with more (less) than 50% fraction.

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