We use single crystal $^{63}$Cu NMR techniques to revisit the early $^{63}$Cu NQR signature of charge order observed for La$_{1.875}$Ba$_{0.125}$CuO$_4$ ($T_c = 4$ K) [A. W. Hunt et al., Phys. Rev. Lett. 82, 4300 (1999)]. We show that the growth of spin correlations is accelerated below ~ 80 K, where the inverse Laplace transform (ILT) $T_1$ analysis of the $^{139}$La NMR spin-lattice relaxation curve recently uncovered emergence of the slow components in the lattice and/or charge fluctuations [P. M. Singer et al., 101, 174508 (2020)]. From the accurate measurements of the $^{63}$Cu NMR signal intensity, spin echo decay $M(2\tau)$, spin-lattice relaxation rate $^{63}1/T_1$, and its density distribution function $P(63/1/T_1)$, we also demonstrate that charge order at $T_{\text{charge}} \approx 54$ K turns on strong enhancement of spin fluctuations within charge ordered domains, thereby making the CuO$_2$ planes extremely inhomogeneous. The charge ordered domains grow quickly below $T_{\text{charge}}$, and the volume fraction $F_{\text{CA}}$ of the canonical domains unaffected by charge order gradually diminishes by ~ 35 K. This finding agrees with our independent estimations of $F_{\text{CA}}$ based entirely on the $^{139}$La ILTT$_1$ analyses, but is in a stark contrast with much slower growth of charge ordered domains observed for La$_{1.885}$Sr$_{0.115}$CuO$_4$ from its $T_{\text{charge}} \approx 80$ K to $T_c \approx 30$ K.

I. GENERAL INTRODUCTION

A variety of phases compete or coexist in cuprate high $T_c$ superconductors, including the charge ordered phase around the magic composition at $x \sim 1/8$ (see [1] [2] for recent reviews). The charge ordered state was originally discovered a quarter century ago below $T_{\text{charge}} \sim 60$ K in the low temperature tetragonal (LTT) structure of La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ [2]. Years later, evidence for charge order based on neutron and X-ray scattering experiments also emerged in the LTT structure of La$_{1.875}$Ba$_{0.125}$CuO$_4$ ($T_{\text{charge}} \approx 54$ K) [10], followed by La$_{1.66}$Eu$_{0.2}$Sr$_{0.12}$CuO$_4$ ($T_{\text{charge}} \sim 80$ K) [2], rather than the low temperature orthorhombic (LTO) structure of the canonical superconducting phase with much higher $T_c$. Accordingly, many researchers continued to believe that the LTO to LTT structural transformation was the key to stabilizing the long range charge ordered state, which in turn suppresses superconductivity. However, recent advances in X-ray scattering techniques finally led to successful detection of charge order Bragg peaks even in the LTO structure of La$_{1.885}$Sr$_{0.115}$CuO$_4$ ($T_c \approx 30$ K) below as high as $T_{\text{charge}} \approx 80$ K [8][10].

Two decades have passed since our initial reports that all of these La$_{214}$ type cuprates undergo charge order at comparable temperatures [11][15], on the ground that they all share nearly identical NMR anomalies identified at $T_{\text{charge}}$ of La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$. During these years, NMR techniques made major advances both in the instrument technologies and data analysis methods. Owing to the reduction in the signal detection dead time of the NMR spectrometers after the application of radio frequency pulses, routine NMR measurements have become possible with the pulse separation time as short as $\tau \sim 2 \mu s$ between the 90 degree excitation and 180 degree refocusing pulses. This $\tau$ is an order of magnitude shorter than the typical value $\tau \sim 20 \mu s$ used in the 1980’s, and detection of the paramagnetic $^{63}$Cu NMR signals with extremely fast NMR relaxation rates, which arises from the charge ordered domains (represented schematically by islands with various shades in Fig. 1(b-c)), has become feasible [16][17]. Moreover, the development of the inverse Laplace transform (ILT) $T_1$ analysis technique enabled us to deduce the histogram of the distribution of the nuclear spin-lattice relaxation rate $1/T_1$ (i.e. the probability density distribution $P(1/T_1)$) [18][21], in addition to the average value of the distributed $1/T_1$ estimated from the conventional stretched exponential fit. The recent ILTT$_1$ analysis of $^{139}$La measured at the $^{139}$La sites in La$_{1.875}$Ba$_{0.125}$CuO$_4$ [19] and La$_{1.885}$Sr$_{0.115}$CuO$_4$ [20] established the continued presence even below $T_{\text{charge}}$ of the canonical domains, which exhibit canonical properties expected for superconducting CuO$_2$ planes without anomalous enhancement of Cu spin fluctuations triggered by charge order. This new finding based entirely on $^{139}$La NMR supports our original conjecture [11][13] that peculiar domain-by-domain variation emerges immediately below $T_{\text{charge}}$ due to the spatially growing charge ordered domains, as summarized in Fig. 11.

In this paper, we revisit the earlier $^{63}$Cu nuclear quadrupole resonance (NQR) report on the issue of charge order in La$_{1.875}$Ba$_{0.125}$CuO$_4$ [11][13] based on comprehensive single crystal $^{63}$Cu NMR results, and compare our findings with $^{139}$La NMR results observed for the same crystal [19]. Since La$_{1.875}$Ba$_{0.125}$CuO$_4$ has a well-defined, sharp charge order transition at $T_{\text{charge}} \approx 54$ K as determined by X-ray diffraction experiments and lacks magnetic perturbations caused by additional Nd$^{3+}$ spins, it is an ideal platform to test the NMR response that sets in precisely at $T_{\text{charge}}$. We confirmed a precur-
sor of enhanced spin correlations below ~ 80 K based on the $^{63}$Cu NMR linewidth data [22] and $1/T_1$ [23], where the ILTT$_1$ analysis of the $^{139}$La NMR data uncovered the presence of low frequency modes in the lattice and/or charge fluctuations [19]. These precursors are followed by dramatic, spatially inhomogeneous enhancement of low frequency spin fluctuations within charge ordered domains that begin to nucleate at $T_{\text{charge}} \simeq 54$ K. The volume fraction $F_{\text{CO}}$ of the charge ordered domains is not 100% immediately below $T_{\text{charge}}$, and grows only progressively below $T_{\text{charge}}$. We estimate the volume fraction $F_{\text{CA}} = (1 - F_{\text{CO}})$ of the canonical domains based on $^{63}$Cu NMR spin echo decay $M(2\tau)$ measured over a wide time range from $2\tau = 4$ to 100 $\mu$s. The temperature dependence of $F_{\text{CA}}$ (Fig.7) shows excellent agreement with the independent estimation based entirely on the ILTT$_1$ analysis of the $^{63}$Cu NMR $^{63}1/T_1$ data (Fig.5) and $^{139}$La NMR $^{139}1/T_1$ data (Fig.8) [19]. Moreover, we show that these canonical domains almost completely disappear by $\sim 35$ K, far above $T_c \simeq 4$ K of La$_{1.875}$Ba$_{0.125}$CuO$_4$. This finding is in remarkable contrast with the case of La$_{1.885}$Sr$_{0.115}$CuO$_4$, where the canonical domains still occupy nearly a half of the CuO$_2$ planes when superconductivity sets in at $T_c \simeq 30$ K [14, 19].

The rest of this article is organized as follows. In section 2, we provide a brief overview of the NMR response expected in the charge ordered CuO$_2$ planes of the La214 cuprates. In section 3, we present results and discussions, followed by summary and conclusions in section 4.

II. NMR RESPONSE IN CHARGE ORDERED LA214 CUPRATES

Until fairly recently, tremendous confusions persisted since our original publications asserting the presence of charge order in La$_{1.875}$Ba$_{0.125}$CuO$_4$ and related La214 materials. This is primarily because, except for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ [3], charge order Bragg peaks were not successfully detected in many La214 cuprates for years [11, 17, 19]. This unfortunate circumstance misled a large number of researchers to argue that charge order was absent in all the cuprates but La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, and cast doubt on the link between the $^{63}$Cu NMR anomalies and charge order (see, for example [19, 27]). Moreover, many NMR experts overlooked, or failed to understand the implications of the following crucial statement in our original publication, quoted verbatim from Hunt et al. [11]:

charge order turns on low frequency spin fluctuations [28], and consequently the $^{63}$Cu nuclear spin-lattice and spin-spin relaxation rates diverge in the striped domains.

The idea outlined in this short statement about the spatial inhomogeneity of magnetic properties induced by charge order is the key to the proper understanding of the NMR data of all the La214 cuprates below $T_{\text{charge}}$. Let us elaborate with the aid of Fig.2 in which we contrast the behaviors in the charge ordered state in the left panels with those for typical antiferromagnets without any spatial inhomogeneity, such as La$_2$CuO$_4$ [29, 30], in the right panels. In Fig.2(a), we sketch the temperature dependence of the imaginary part of the dynamical local spin susceptibility $\text{Im} \chi(\omega)$ at very low energy transfer $\omega$ using solid curves. See Fig.7 in [28] for the first original data for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, and Fig.1(c) in [6] as well as Fig.8(a) in [4] for the original data for La$_{1.875}$Ba$_{0.125}$CuO$_4$. In the case of La$_{1.875}$Ba$_{0.125}$CuO$_4$, panels with typical antiferromagnets without any spatial inhomogeneity, such as La$_2$CuO$_4$ [29, 30], in the right panels. In Fig.2(a), we sketch the temperature dependence of the imaginary part of the dynamical local spin susceptibility $\text{Im} \chi(\omega)$ at very low energy transfer $\omega$ using solid curves. See Fig.7 in [28] for the first original data for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, and Fig.1(c) in [6] as well as Fig.8(a) in [4] for the original data for La$_{1.875}$Ba$_{0.125}$CuO$_4$. In the case of...
Imχ(ω) at the low energy transfer of ω = 0.5 meV begins to grow dramatically precisely at T_{charge} ≃ 54 K [3].

This inelastic glassy spin response induced by charge order below T_{charge} ≃ 54 K should not be confused with the elastic response of Bragg scattering arising from the static spin order at T_{spin} ≃ 40 K [4, 5]. Simply put, charge order creates small, finite size domains, in which Cu spins are slowly and collectively fluctuating without entering the long range spin ordered state. The long range spin order takes place when the charge correlation length grows to ∼ 20 nm at T_{spin} ≃ 40 K [6], where charge ordered domains become interconnected. This sequence of order may look similar to the nucleation of charge ordered domains become interconnected. This sequence of order may look similar to the nucleation of charge ordered domains become interconnected. This sequence of order may look similar to the nucleation of charge ordered domains become interconnected.

The long range spin order takes place when the charge correlation length grows precisely at T_{charge} due to the glassy nature of spin fluctuations that begins without any onset temperature, then the elastic response in neutron scattering sets in below T_{N} as shown in Fig.2(c). In the case of charge ordered La_{1.875}Ba_{0.125}CuO_{4}, the elastic magnetic response (Bragg peaks) sets in only below the spin ordering temperature, T_{spin} ≃ 40 K [4, 5], but strong enhancement of 1/T_{1} precedes below the clear onset temperature at T_{charge} due to the glassy nature of spin fluctuations induced by charge order.

Such an upturn of 139/1/T_{1} below T_{charge} can be easily observed at 139La sites by NQR [13], and more recent high precision NMR measurements confirmed that the onset of the drastic growth of 139/1/T_{1} is precisely at T_{charge} ≃ 54 K [14]. It is important to note, however, that these earlier 139/1/T_{1} values were estimated based on the stretched exponential fit, and probed only the average behavior of the entire sample. In fact, based on the ILTT1 analysis of the T_{1} recovery curve at 139La sites, we recently demonstrated that the fastest component of 139/1/T_{1} indeed begins to grow precisely below T_{charge} (the solid curve in Fig.2(b)), but the slower components exhibit no anomaly through T_{charge} (dashed curve in Fig.2(b)), both in La_{1.875}Ba_{0.125}CuO_{4} [19] and La_{1.85}Sr_{0.115}CuO_{4} [20].

In other words, 1/T_{1} exhibits qualitatively different behaviors domain by domain. Some parts of CuO_{2} planes are not immediately affected by charge order and continue to exhibit canonical behavior expected for superconducting CuO_{2} planes even below T_{charge}, as schemat...
ically shown in Fig.1(b). These findings are also consistent with the two decade old knowledge that $1/T_1$ measured at $^{63}$Cu sites of La$_{1.875}$Ba$_{0.125}$CuO$_4$ with a relatively long $\tau = 20$ $\mu$s exhibits no anomaly at $T_{\text{charge}}$ [33], because it preferentially reflects the canonical domains with slow transverse relaxation times, whereas $1/T_1$ measured at the $^{63}$Cu sites with very short $\tau$ exhibits an upturn of $1/T_1$ below $T_{\text{charge}}$ [23].

The continued presence of $^{139}$La and $^{63}$Cu NMR signals exhibiting the canonical behavior below $T_{\text{charge}}$ implies that not all Cu electron spins are involved in the low energy upturn of $\text{Im}\chi(\omega)$ shown by the solid curve in Fig.2(a). Instead, some Cu electron spins continue the trend observed above $T_{\text{charge}}$, as shown by the dashed curve in Fig.2(a). Since inelastic neutron scattering measures only the volume integral of the spin response, one needs to rely on a local probe such as NMR to reveal the domain by domain response schematically summarized in Fig.1.

The unusual magnetic inhomogeneity induced in the charge ordered state is also reflected on the transverse $T_2$ relaxation process observed for the transverse nuclear magnetization $M(2\tau)$ at $^{63}$Cu sites, as schematically summarized in Fig.2(c) (see Fig.6 below for the actual data). Upon entering the charge ordered state, $M(2\tau)$ begins to exhibit initial fast decay in the short time regime, followed by slower decay in the long time regime. The transverse relaxation rate in the latter is comparable to that observed at $T_{\text{charge}}$ and above. The nuclear spins responsible for the fast and slow transverse relaxation in Fig.2(c) can be attributed to the $^{63}$Cu sites located in the charge ordered and canonical domains in Fig.1, respectively.

We estimate the volume fraction $F_{\text{CA}}$ of the canonical domains by extrapolating the slow decaying part of the $M(2\tau)$ curve to $2\tau = 0$, as shown by dashed lines in Fig.2(c). The intercept of the extrapolated dashed line with the vertical axis at $2\tau = 0$ yields $F_{\text{CA}}$. We emphasize that, if the charge ordered CuO$_2$ planes undergo uniform enhancement of spin correlations, then $M(2\tau)$ curve would look very different, and should be similar to the case of uniform antiferromagnets shown in Fig.2(g).

In Fig.2(d), we summarize the temperature dependence of the $^{63}$Cu NMR signal intensity at short $2\tau_1$ and long $2\tau_2$, as expected from Fig.2(c). The total intensity in the limit of $2\tau = 0$ is proportional to the number of nuclear spins that does not change with temperature, and hence always conserved, as shown by the horizontal gray line. For the finite values of $2\tau$, the intensity exhibits an anomaly at $T_{\text{charge}}$, because the signal intensity arising from the charge ordered domains is reduced by the fast transverse relaxation in the short time regime in Fig.2(c). As explained in the previous paragraph, one can estimate the volume fraction of the canonical domains $F_{\text{CA}}$ by extrapolating $M(2\tau)$ in the long time regime of Fig.2(c) to $2\tau = 0$. The end result would be the solid curve in Fig.2(d). Recalling $F_{\text{CO}} + F_{\text{CA}} = 1$, one can also estimate $F_{\text{CO}}$ as shown in Fig.1(d). This is the technique of the signal intensity wipeout effect to probe the volume fraction of the charge ordered domains, developed originally in [11-13].

### III. RESULTS AND DISCUSSIONS

#### A. NMR lineshapes

In Fig.3(a-b), we summarize the representative $^{63}$Cu NMR lineshapes for a 51 mg single crystal [31] measured with a fixed pulse separation time $\tau = 12$ $\mu$s in an external magnetic field $B_{\text{ext}} = 9$ T. The typical radio frequency pulse width was 2 $\mu$s and 4 $\mu$s for 90 and 180
degree pulses throughout this work. We confirmed both above (60 K) and below (50 K) $T_{\text{charge}} \simeq 54$ K that the lineshape hardly changes even if we use $\tau = 2 \mu s$, except that the transverse relaxation process (i.e. $T_2$) reduces the overall intensity for longer $\tau$. These lineshapes indicate that La$_{1.875}$Ba$_{0.125}$CuO$_4$ develops its charge ordered state in a fundamentally different manner from La$_{1.885}$Sr$_{0.115}$CuO$_4$. We recall that the c-axis $^{63}$Cu NMR lineshapes in the latter comprised of two distinct types of signals below its higher $T_{\text{charge}} \simeq 80$ K: (i) a narrower, canonically behaving peak with slower relaxation rates that are typical for high $T_c$ cuprates, and (ii) a much broader wing-like signal with extremely fast relaxation rates [14]. The former is gradually wiped out below $T_{\text{charge}}$, transferring the spectral weight to the latter. Accordingly, the NMR lineshapes completely change between $\tau = 2 \mu s$ and $\tau = 12 \mu s$ below $T_{\text{charge}}$, since only the canonically behaving narrower peak can be detected with $\tau = 12 \mu s$. That is not the case here for La$_{1.875}$Ba$_{0.125}$CuO$_4$, and the observed c-axis lineshapes are more uniformly broadened even for $\tau = 12 \mu s$.

In the inset of Fig.2 we also summarize the temperature dependence of the integral of these $\tau = 12 \mu s$ lineshapes. We emphasize that the intensity data are merely the integral of the lineshapes in panels (a) and (b), and have not been subjected to any data analysis. The sharp anomaly observed at $\sim 54$ K in the raw integrated intensity corresponds to that in the conceptual sketch of the dashed curve in Fig.2(d) for $2\tau_2 = 24 \mu s$, and signals a lurking phase transition in La$_{1.875}$Ba$_{0.125}$CuO$_4$.

B. Linewidth

In Fig.2(b), we summarize the temperature dependence of the half width at the half maximum (HWHM) of the c-axis lineshapes shown in Fig.2(b). For the $B_{\text{ext}} || c$-axis geometry, the second order nuclear quadrupole effect vanishes [28], and the temperature dependence of the linewidth is set almost entirely by magnetic effects [28]. On the other hand, since the lower frequency side of the broadened NMR lineshape nominally has negative frequency shifts, a large distribution of the chemical shift cannot account for the observed broadening, either. Therefore, the spin degrees of freedom must be playing the key role in the line broadening, but the exact mechanism of the broadening has long been an enigma.

The dashed curve overlaid on the HWHM data points above $\sim 80$ K are the best empirical Curie-Weiss fit. The HWHM begins to grow more quickly below $\lesssim 80$ K. Our single crystal result is consistent with an earlier aligned powder result [22]. This linewidth anomaly is accompanied by an analogous deviation from the Curie-Weiss growth of $1/T_1T$ at $^{63}$Cu sites [23], signaling that strong enhancement of antiferromagnetic spin correlations is playing a role in HWHM as well. Interestingly, charge order sets in for a minor volume of La$_{1.885}$Sr$_{0.115}$CuO$_4$ also at $\sim 80$ K and the aforementioned wing-like $^{63}$Cu NMR signal emerges [17], but it may be a coincidence.

We recently showed based on the ILTT$_1$ analysis of the $^{139}$La nuclear spin-lattice relaxation curve that the electric field gradient (EFG) at the $^{139}$La sites has slowly fluctuating components ($\sim$ MHz) below $\sim 80$ K [19]. The ILT cannot distinguish the origin of the slow dynamics between the lattice and/or charge degrees of freedom. Regardless of the origin, these NMR results indicate that
spin correlations begin to grow more steeply when fluctuations of the lattice and/or charge degrees of freedom slow down. It is also interesting to note that recent X-ray scattering data showed the dynamic short range charge order above $T_{\text{charge}}$ [4]. All pieces put together, the onset of charge order in La$_{1.875}$Ba$_0.125$CuO$_4$ seems to be suppressed to $\sim$ 54 K, until the LTO to LTT structural phase transition suddenly takes place.

C. $^{63}$Cu spin-lattice relaxation rate $^{63}1/T_1$

We measured $^{63}1/T_1$ at the center of the $B_{\text{ext}} \parallel c$ axis peak in Fig. 3(b) using the standard inversion recovery method by applying a 180 degree pulse prior to the spin echo sequence. The goodness of the fit of the nuclear spin recovery curve $M(t)$ with the standard formula for the central transition was similar to the case of La$_{1.85}$Sr$_{0.15}$CuO$_4$ [17], and stretching was not necessary for our purpose even below $T_{\text{charge}}$ owing to modest distributions, as shown in Appendix. This is simply because the signals arising from $^{63}$Cu sites with very fast $^{63}1/T_1$ are wiped out below $T_{\text{charge}}$ even for $\tau = 2$ µs (see Fig. 6 below), and hence hardly contribute to the $M(t)$ results. In Fig. 4(a), we summarize the temperature dependence of $^{63}1/T_1$ observed for three different values of the pulse separation time $\tau = 2$, 12, and 20 µs between the 90 and 180 degree pulses, and compare the results with $^{139}1/T_1$ observed at the $^{139}$La sites [19].

The gradual decrease of $^{63}1/T_1$ with temperature observed for $\tau = 20$ µs down to 48 K is typical for high $T_c$ cuprates [39]. The extremely broad, small NMR signal from a small single crystal made accurate measurements of $^{63}1/T_1$ difficult below 48 K. For comparison, we show the $^{63}1/T_1$ results measured with $\tau = 20$ µs for an aligned powder sample [35]. The signal intensity was large and manageable even below 48 K for the large amount ($\sim$ 300 mg) of aligned powder. The decreasing trend of $^{63}1/T_1$ continues below 48 K. These results for $\tau = 20$ µs show no anomaly through $T_{\text{charge}}$, and indicate that some parts of CuO$_2$ planes remain unaffected by charge order and the resulting enhancement of spin fluctuations even deep into the charge ordered state below $T_{\text{charge}}$. That is why we initially overlooked the lurking charge order in 1990 [35].

The $^{63}1/T_1$ results for $\tau = 2$ µs are consistently larger by $\sim$ 6% than those for $\tau = 20$ µs down to $\sim$ 80 K. This is merely because the quenched disorder caused by Ba$^{2+}$ substitution into the La$_3^{3+}$ induces a nanoscale inhomogeneity in the local hole concentration of the CuO$_2$ planes [24, 25], as represented schematically by different shades in Fig. 1(a). $^{63}1/T_1$ is generally smaller for larger values of the hole concentration $x$ [23, 24], but the $^{63}$Cu NMR peak frequency for the $B_{\text{ext}} \parallel c$ axis geometry is set entirely by the chemical shift that is independent of $x$. Moreover, the $^{63}$Cu B-sites located at the nearest neighbor of La$_3^{3+}$ sites are superposed in this field geometry [10], and their $^{63}1/T_1$ is somewhat slower than those at the main $^{63}$Cu A-sites [24]. Accordingly, a mild distribution of $^{63}1/T_1$ is always present. As shown below in Fig. 9(a), the transverse relaxation does not reduce the spin echo intensity significantly for $\tau = 2$ µs above $T_{\text{charge}}$, and hence nearly 100% of the $^{63}$Cu nuclear spins contribute to the observed value of $^{63}1/T_1$.

$^{63}1/T_1$ for $\tau = 2$ µs levels off towards $T_{\text{charge}}$, and shows qualitatively different behavior from the $\tau = 20$ µs results. This is consistent with the increased distribution of $^{139}1/T_1$ observed at $^{139}$La sites in the same temperature range [19]. $^{63}1/T_1$ measured with $\tau = 2$ and 12 µs begins to increase precisely below $T_{\text{charge}}$, in agreement with the earlier NQR report by Tou et al. [23]. Pelc et al. observed greater values of $^{63}1/T_1$ below $T_{\text{charge}}$ with NQR than Tou et al., because they used $\tau = 2$ µs and captured more nuclear spins with faster relaxation rates [16]. Our $^{63}1/T_1$ results for $\tau = 2$ µs is slower below $T_{\text{charge}}$ than Pelc et al.’s, probably because the fastest nuclear spins are pushed aside to the tail sections of the magnetically broadened NMR lineshape below $T_{\text{charge}}$ due to locally stronger spin correlations.

We present $^{63}1/T_1T$ in Fig. 3(b) in comparison to the WHHM. $^{63}1/T_1T$ probes the wave vector $q$ integral of Im$\chi(q, \omega_n)$. $^{63}1/T_1T$ obeys the Curie-Weiss behavior analogous to that observed for the WHHM. The $^{63}1/T_1T$ results for $\tau = 2$ µs as well as the WHHM begin to deviate from the Curie-Weiss behavior somewhat above $T_{\text{charge}}$, in agreement with the earlier report by Tou et al. as noted above [23].

The enhancement of $^{63}1/T_1$ observed at $^{63}$Cu sites below $T_{\text{charge}}$ is only modest, compared with the steep divergent behavior observed at $^{139}$La sites for the same crystal (grey bullets) [11]. This apparent discrepancy arises from the fact that $^{139}1/T_1$ plotted for the $^{139}$La sites represents the spatially averaged (center of gravity) value of the widely distributed $^{139}1/T_1$ below $T_{\text{charge}}$. In contrast, $^{63}1/T_1$ at $^{63}$Cu sites reflects only the nuclear spins that are still observable below $T_{\text{charge}}$ owing to their slower NMR relaxation rates. For example, the observable $^{63}$Cu NMR signal intensity at 48 K is only about a half of the total intensity even for $\tau = 2$ µs, because a majority of $^{63}$Cu NMR signals is already suppressed by their extremely fast transverse relaxation rates (see the data points at $2\tau = 4$ µs in Fig. 3(b) below).

To underscore this point, we used dashed lines in Fig. 9(a) to mark the top and bottom 10% values of the distributed $^{139}1/T_1$ at $^{139}$La sites estimated from the ILTT$T_1$ analysis [19]. The comparison indicates that $1/T_1$ measured at the observable $^{63}$Cu sites below $T_{\text{charge}}$, especially for the longer values of $\tau$, reflects only the bottom end of the spatial distribution in spin fluctuations.

These findings can be corroborated by the density distribution function $P(^{63}1/T_1)$ at $^{63}$Cu sites deduced by ILT. In Fig. 5 we summarize $P(^{63}1/T_1)$ obtained from the $M(t)$ curves measured with $\tau = 2$ µs in Fig. 9(a). The integrated area underneath the $P(^{63}1/T_1)$ curve for 50 K is set to 0.63 to reflect the suppressed signal intensity observed with $\tau = 2$ µs at 50 K, as shown in Fig. 6(a) in the
FIG. 5. The density distribution function \( P(63^{1}/T_1) \) of the distributed values of \( 63^{1}/T_1 \) at \( 63^{1}\mathrm{Cu} \) sites, deduced by ILT from \( M(t) \) measured with \( \tau = 2 \mu s \) shown in Appendix. The integrated area (in a log scale) is normalized to 1 at 295 K and 100 K (so that the total probability is 1), whereas the area underneath the 50 K result is set to 0.68 in proportion to the 100 K (so that the total probability is 1), whereas the area integrated area (in a log scale) is normalized to 1 at 295 K and 50 K*0.68 CA 50K*0.68 CO 50K fit combo 295 K cg 100K cg 50K cg

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the other hand, a recent work by Pelc et al. presented \( M(2\tau) \) data only in the short time regime, and did not demonstrate the crossover to the long time regime, either \cite{16} (see their Fig. 3(a)). Pelc et al.’s limited data set might inadvertently leave unsuspecting readers with a false impression that CuO\(_2\) planes in charge ordered La\(_{1.875}\)Ba\(_{0.125}\)CuO\(_4\) is spatially homogeneous, and exhibit uniformly fast transverse relaxation, similar to the case of homogeneous antiferromagnet shown in Fig.2(g). But our new data presented in Fig.6 firmly establish that is not the case.

We also emphasize that what matters in understanding the inhomogeneous glassy state induced by charge order based on \(^{63}\)Cu NMR intensity is the extrapolation of the \textit{slowly decaying part} of \( M(2\tau) \) observed in the long time regime above \( 2\tau = 20 \mu s \) to \( 2\tau = 0 \mu s \), as explained in section 2 and shown with the dashed curves in both Fig.2(c) and Fig.6. Instead, Pelc et al. examined the extrapolation of the \textit{fast} decaying part of their \( M(2\tau) \) data in the short time regime to \( 2\tau = 0 \) (solid curves extrapolated to \( 2\tau = 0 \) in their Fig. 3(a)), only to confirm that the \textit{total intensity} arising from both the charge ordered and canonical domains is conserved. Their finding below \( T_{\text{charge}} \) summarized in the inset of their Fig. 2 corresponds to the trivial conservation law of the \textit{total intensity} \( F_{\text{CO}} + F_{\text{CA}} = 1 \), represented schematically by the gray horizontal line in Fig.2(d). It does not provide any useful insight into the nature of the glassy, charge ordered state.

E. \(^{63}\)Cu NMR signal intensity wipeout and estimation of \( F_{\text{CA}} \) based on ILT

Finally but not the least, let us return to the issue of the integrated intensity of the \(^{63}\)Cu NMR lineshapes in the inset of Fig.3. To eliminate the minor effects of the transverse relaxation on the bare integrated intensity measured at a fixed \( \tau = 12 \mu s \), we extrapolated the \( M(2\tau) \) curves to \( 2\tau = 0 \) as shown by the dashed lines in Fig.6(b), and estimated the volume fraction \( F_{\text{CA}} \) of the canonical domains. We summarize the \( B \parallel ab \)-axis results in Fig.7 using bullets \cite{39}. For comparison, we also plot our original signal intensity wipeout data measured with NQR for a \(^{63}\)Cu isotope enriched powder sample (open triangles) \cite{13}. The agreement between the new NMR and older NQR results is satisfactory, in view of the greater uncertainties in the latter arising from the extra Gaussian \( T_{2G} \) term in the spin echo decay.

As explained in detail in section 2 using Fig.2(d), the temperature dependence of the \(^{63}\)Cu NMR signal intensity in Fig.7 indicates that charge order does not set in homogeneously in the CuO\(_2\) planes. The finite value of \( F_{\text{CA}} \) below \( T_{\text{charge}} \) implies that a significant fraction of the volume is hardly affected by charge order even below \( T_{\text{charge}} \), and exhibits the canonical behavior expected for CuO\(_2\) planes that seem destined to undergo superconducting transition at \( T_c \simeq 30 \) K. But the residual volume fraction of such canonical behaving CuO\(_2\) planes almost vanishes by \( \sim 35 \) K, where the charge correlation length saturates at \( \sim 20 \) nm \cite{5}. In addition, spin stripe order sets in at \( \sim 35 \) K at the time scale of \( \mu \)SR experiments \cite{17, 48} and the volume-averaged value of the distributed \( ^{139}\text{La}/T_1 \) is peaked at \( ^{139}\text{La} \) sites \cite{19, 34}. In contrast, the volume fraction of the canonical domains exceeds 40% at its \( T_c = 30 \) K in La\(_{1.885}\)Sr\(_{0.115}\)CuO\(_4\) \cite{17, 19, 20}.

We can achieve more quantitative understanding of the \(^{63}\)Cu NMR intensity anomaly and its relation with the unconventional nature of charge order with the aid of the ILT\(_{T_1}\) analysis of the \(^{139}\)La nuclear spin recovery curve \cite{19}. For convenience, we reproduce the key results of the probability density distribution function, \( P(\frac{^{139}\text{La}}{T_1}) \) of \( ^{139}\text{La}/T_1 \) in Fig.8. The main peak of \( P(\frac{^{139}\text{La}}{T_1}) \) has finite...
values only below $^{139}T_1/T_1 \sim 1 \text{s}^{-1}$ from 100 K down to 60 K. In other words, the upper bound of the distributed values of $^{139}T_1/T_1$ is 1 $\text{s}^{-1}$. Notice, however, that the main peak gradually broadens below 77 K, accompanied by a small split-off peak centered around $^{139}T_1/T_1 \sim 5 \text{s}^{-1}$. Analogous anomalies of $P(^{139}T_1/T_1)$ are observed also around 240 K near the high temperature tetragonal to low temperature orthorhombic structural phase transition [19]. Since the charge order transition is accompanied by a first order structural transition from low temperature orthorhombic to low temperature tetragonal phase [19], we can attribute these anomalies slightly above $T_{\text{charge}}$ to the contributions of fluctuating electric field gradient precursor to the structural phase transition and/or fluctuating charges [19].

As temperature is lowered through $T_{\text{charge}}$, $P(^{139}T_1/T_1)$ gradually transfers spectral weight to larger values of $^{139}T_1/T_1$ while broadening asymmetrically. This corresponds to the fact that a sharp divergent behavior sets in precisely at $T_{\text{charge}}$ for $^{139}T_1/T_1$ estimated from the stretched fit, which tends to be close to the center of gravity of the distributed $^{139}T_1/T_1$ [19]. We emphasize that a half of the spectral weight of $P(^{139}T_1/T_1)$ still remains below 1 $\text{s}^{-1}$ even at 50 K. This implies that the corresponding sample volume is still unaffected by charge order, and $^{139}T_1/T_1$ is as slow as at 77 K. This is consistent with our findings for $^{63}T_1/T_1$ at $^{63}$Cu sites in Fig.4 and 5. $^{63}$Cu nuclear spins that are still easily observable below $T_{\text{charge}}$ owing to slow NMR relaxation rates are located in the same domains as these $^{139}$La sites with slower relaxation rates.

In view of the fact that the ILT curve $P(^{63}T_1/T_1)$ observed at 50 K in Fig.5 has a well-defined peak associated with the canonical domains with slower $^{63}T_1/T_1$, perhaps it may be somewhat surprising to find that $P(^{139}T_1/T_1)$, which should also encompass the canonical component centered around $^{139}T_1/T_1 \sim 0.5 \text{s}^{-1}$, is broader and increasingly featureless below $T_{\text{charge}}$. But this is simply because the transverse relaxation does not suppress the faster components of $P(^{139}T_1/T_1)$ arising from charge ordered domains. In this context, we recall that the charge correlation length in the charge ordered state is known to be as short as several nm immediately below $T_{\text{charge}}$, and the spin correlation length cannot exceed this. This means that the extent of enhancement of $1/T_1$ in each charge ordered domain is set by the domain size. The highly disordered nature of the charge ordered state with varying domain sizes naturally explains the very broad distribution of $^{139}T_1/T_1$ below $T_{\text{charge}}$, ranging from the small canonical value to the upper bound set by the largest charge ordered domains. It is also worth noting that $P(^{139}T_1/T_1)$ curve exhibits somewhat more distinctive features in La$_{1.885}$Sr$_{0.115}$CuO$_4$ for the canonical and charge ordered domains[20]. That is probably because the canonical domains are more robust below $T_{\text{charge}}$ in a wider temperature range in La$_{1.885}$Sr$_{0.115}$CuO$_4$, and in agreement with the fact that $T_c$ is as high as $\sim 30$ K.

The featureless, continuous distribution of $P(^{139}T_1/T_1)$ makes it difficult to de-convolute $P(^{139}T_1/T_1)$ and estimate $F_{\text{CA}}$ from $P(^{139}T_1/T_1)$. We therefore introduce a cut off in Fig.8 at 2 $\text{s}^{-1}$, at the upper end of the distributed values of $^{139}T_1/T_1$ observed at 56 K, as represented by the upward vertical arrow in Fig.8. Then we can estimate the $F_{\text{CA}}$ of the canonical $^{63}$Cu nuclear spins as the integrated area of $P(^{139}T_1/T_1)$ below the cut-off. We summarize the temperature dependence of thus estimated $F_{\text{CA}}$ in Fig.7 using x symbols, in comparison to $F_{\text{CA}}$ estimated from the $^{63}$Cu NMR intensity. Despite the simplicity of this analysis, the estimation based entirely on the $^{139}$La NMR results reproduces the $^{63}$Cu NMR signal intensity anomaly very well.

We can also test the consistency of $F_{\text{CA}}$ with the $^{63}$Cu ILT result of $P(^{63}T_1/T_1)$ at 50 K. From the integral of the light dashed curve in Fig.8 arising from the canonical contribution with slower relaxation rate, we estimate $F_{\text{CA}} \sim 0.63$ at 50 K. We plot the result in Fig.8 with a diamond, in comparison to $F_{\text{CA}}$ estimated from two other methods, the extrapolation of $M(2\tau)$ (bullets) and cut-offs introduced for $P(^{139}T_1/T_1)$ (x). Despite the completely different methodologies between the three approaches, agreement is good.

Turning our attention to the low temperature side below 30 K, Zeeman perturbed NQR signal is known to

![FIG. 7. The volume fraction $F_{\text{CA}}$ of the canonical domains below $T_{\text{charge}}$ estimated by three different methods. (bullets): the estimation from $^{63}$Cu NMR signal intensity of the canonically behaving sites with slow relaxation rates, based on the extrapolation of $M(2\tau)$ curves from $2\tau = 24 \mu s$ and greater. For comparison, we also reproduce the powder NQR intensity (black triangles, adopted from [19]). The increase in the NQR intensity below $\sim 15$ K is due to the freezing of the fluctuations of the hyperfine magnetic fields from Cu electron spins; we multiplied a factor of 1.63 [19] for the Zeeman perturbed NQR results below 15 K to account for the missing contribution below 25 MHz [19]. (red diamond): $F_{\text{CA}}$ at 50 K estimated from the $^{63}$Cu ILT result in Fig.6. (Purple x): $F_{\text{CA}}$ estimated from the ILT; analysis of the $^{139}$La ILT results in Fig.6. Also shown with + symbols is $F_{\text{CA}}$ estimated from $P(^{139}T_1/T_1)$.](image-url)
reemerge when the hyperfine magnetic field from frozen Cu electron spins become static at the NMR measurement time scale below ~ 15 K [13, 49]. In general, 1/T_1 is proportional to the dynamical spin susceptibility Im(\omega) multiplied by temperature T, and hence the cut-off of 139/\T_1 = 2 s^{-1} for the same magnitude of Im(\omega) needs to be scaled down to 139/\T_1 = 0.6 s^{-1} by the ratio between 15 K and T_charge. This second cut-off is shown with a downward vertical arrow in Fig.8. We can estimate the fraction of frozen Cu electron spins as the area integral below this cut-off. The results, also shown in Fig.7 using ×, reproduce the qualitative aspects of the signal intensity recovery observed by Hunt et al. [13]. The agreement can be improved if we estimate F_CA based on P(139/\T_1) (i.e. the distribution of 139/\T_1 divided by T) by introducing a single cut-off at 139/\T_1 T = 0.036 s^{-1}K^{-1} for both above and below 30 K; this cut-off value corresponds to 139/\T_1 = 2 s^{-1} divided by T_charge. We present these estimations using + symbols also in Fig.7.

Strictly speaking, the cut-off for dividing the canonical and charge ordered domains at 2 s^{-1} for 139/\T_1 should be slightly temperature dependent, because 139/\T_1 in canonically superconducting compositions with x ~ 0.15 decreases slightly below 54 K toward T_c = 38 K [40, 50]. But the observed decrease is weak, and the varying cut-off hardly affects our estimation of F_CA. In fact, the cut-off 139/\T_1 T = 0.036 s^{-1}K^{-1} effectively incorporates such temperature dependent shift of the cut-off in 139/\T_1, but the F_CA results in Fig.7 show no significant changes.

IV. SUMMARY AND CONCLUSIONS

We reported new comprehensive single crystal 63Cu NMR results for La_{1.885}Ba_{0.125}CuO_4, and compared the results with our recent report on 139La NMR. We confirmed the precursors of enhanced growth in spin correlations below ~ 80 K based on HWHM and 1/\T_1, in agreement with earlier reports [22, 23]. This is the same temperature range, where our recent ILTT analysis of 139/\T_1 at the 139La NMR sites identified the presence of slow lattice and/or charge fluctuations [19]. We demonstrated that the apparently contradictory reports of 1/\T_1 and spin echo decay curves M(2\tau) at the 63Cu sites near and below T_charge, as well as the apparently different behavior between 63Cu and 139La sites below T_charge, are the consequence of a large spatial distribution in the enhancement of spin fluctuations.

Our findings of the sudden onset of NMR anomalies precisely at T_charge for M(2\tau), F_CA, 63/\T_1, and 139/\T_1 and its asymmetric distribution are consistent with the earlier inelastic neutron scattering experiments with very small energy transfer, i.e. charge order turns on glassy spin dynamics precisely at T_charge before the static magnetic order sets in at T_spin ~ 40 K, and hence the low frequency Cu spin fluctuations begin to undergo a dramatic enhancement at T_charge [4, 5, 28].

We revisited our earlier report of the intensity anomaly of 63Cu NMR below T_charge of La_{1.885}Ba_{0.125}CuO_4. We reproduced our original discovery of the intensity anomaly at T_charge [11] with much higher precision, by taking advantage of the convenient magnetic field geometry of B_{ext} || ab axis. We demonstrated once and for all that glassy spin dynamics induced within charge ordered domains begins to suppress the 63Cu NMR intensity exactly at T_charge, where T_charge has already been determined independently by diffraction experiments. We also explained in detail why the observable fraction F_CA of the 63Cu NMR signal intensity provides a good measure of the canonical domains that have not been affected significantly by charge order. Our finding was corroborated by a completely different approach based on the ILTT analysis of the distributed 63/\T_1 and 139/\T_1 [19]. We recall that we recently achieved the same for La_{1.885}Sr_{0.115}CuO_4 based on single crystal 63Cu NMR and the ILTT analysis of 139La NMR [17, 20].
We identified a key difference between La$_{1.875}$Ba$_{0.125}$CuO$_4$ with $T_c = 4$ K and La$_{1.885}$Sr$_{0.115}$CuO$_4$ with much higher $T_c = 31$ K; charge order enhances spin fluctuations in nearly 100% volume of the CuO$_2$ planes in the former by $\sim 35$ K, while nearly a half of the volume fraction is still hardly affected when superconductivity sets in at higher $T_c$ in the latter [17, 20]. On the other hand, in view of the fact that the aforementioned anomalous enhancement of spin correlations are commonly observed below $\sim 80$ K for both La$_{1.875}$Ba$_{0.125}$CuO$_4$ [23] and La$_{1.885}$Sr$_{0.115}$CuO$_4$ [13, 51], it is not clear why $T_{\text{charge}} \approx 54$ K is much lower in La$_{1.875}$Ba$_{0.125}$CuO$_4$ than $T_{\text{charge}} \approx 80$ K in La$_{1.885}$Sr$_{0.115}$CuO$_4$. The only signature of charge order for La$_{1.875}$Ba$_{0.125}$CuO$_4$ observed to date above $T_{\text{charge}}$ is dynamic in nature [6]. It seems as if charge order in La$_{1.875}$Ba$_{0.125}$CuO$_4$ is suppressed from 80 K to 54 K, until the first-order low temperature tetragonal structural transition sets in.

It may be worthwhile to caution that the $^{63}$Cu NMR intensity anomaly is not always entirely related to static charge order. In the case of La$_{2-y}$Sr$_y$CuO$_4$ with [12] and without [11] Nd co-doping, we initially attributed the onset of the $^{63}$Cu NQR intensity anomaly to charge order not only for the optimal charge order composition of $x = 1/8$, but also for above and below $x = 1/8$. Subsequent X-ray diffraction experiments for La$_{1.6-x}$Na$_{0.4}$Sr$_x$CuO$_4$ [52] showed that our estimation of $T_{\text{charge}}$ was accurate for $x = 1/8$ and above, but we overestimated $T_{\text{charge}}$ for $x = 0.10$ and below. Our overestimation for $x = 0.10$ resulted from the fact that the inflection point in the temperature dependence of $F_{\text{CO}}$ at a lower temperature corresponds to $T_{\text{charge}}$ for $x < 1/8$. We refer readers to Fig. 15(a), Fig. 18 and related discussions in [13] for details.

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T. Moriya, J. Phys. Soc. Jpn. 18, 516 (1963).

V. Jaccarino, Nuclear Relaxation in Antiferromagnets, edited by G. T. Rado and H. Suhl, Vol. Magnetism IIA (Academic Press, 1965).

S.-H. Baek, Y. Utz, M. Hücke, G. D. Gu, B. Bühchner, and H.-J. Grafe, Magnetic field induced anisotropy of \(^{139}\)La spin-lattice relaxation rates in stripe ordered \(^{139}\)La\(_{1-x}\)Ba\(_{x}\)Cu\(_2\)O\(_4\), J. Phys. Soc. Jpn. 75, 1254 (1993).

C. J. Arguello, S. P. Chockalingam, E. P. Rosenthal, L. Zhao, C. Gutierrez, J. H. Kang, W. C. Chung, R. M. Fernandes, S. Jia, A. J. Millis, R. J. Cava, and A. N. Pasupathy, Visualizing the charge density wave transition in 2h-nbse\(_2\) in real space, Phys. Rev. B 89, 235115 (2014).

T. Moriya, J. Phys. Soc. Jpn. 18, 516 (1963).

V. Jaccarino, Nuclear Relaxation in Antiferromagnets, edited by G. T. Rado and H. Suhl, Vol. Magnetism IIA (Academic Press, 1965).

S.-H. Baek, Y. Utz, M. Hücke, G. D. Gu, B. Bühchner, and H.-J. Grafe, Magnetic field induced anisotropy of \(^{139}\)La spin-lattice relaxation rates in stripe ordered \(^{139}\)La\(_{1-x}\)Ba\(_{x}\)Cu\(_2\)O\(_4\), J. Phys. Soc. Jpn. 75, 1254 (1993).

C. J. Arguello, S. P. Chockalingam, E. P. Rosenthal, L. Zhao, C. Gutierrez, J. H. Kang, W. C. Chung, R. M. Fernandes, S. Jia, A. J. Millis, R. J. Cava, and A. N. Pasupathy, Visualizing the charge density wave transition in 2h-nbse\(_2\) in real space, Phys. Rev. B 89, 235115 (2014).

T. Moriya, J. Phys. Soc. Jpn. 18, 516 (1963).

V. Jaccarino, Nuclear Relaxation in Antiferromagnets, edited by G. T. Rado and H. Suhl, Vol. Magnetism IIA (Academic Press, 1965).

S.-H. Baek, Y. Utz, M. Hücke, G. D. Gu, B. Bühchner, and H.-J. Grafe, Magnetic field induced anisotropy of \(^{139}\)La spin-lattice relaxation rates in stripe ordered \(^{139}\)La\(_{1-x}\)Ba\(_{x}\)Cu\(_2\)O\(_4\), J. Phys. Soc. Jpn. 75, 1254 (1993).

C. J. Arguello, S. P. Chockalingam, E. P. Rosenthal, L. Zhao, C. Gutierrez, J. H. Kang, W. C. Chung, R. M. Fernandes, S. Jia, A. J. Millis, R. J. Cava, and A. N. Pasupathy, Visualizing the charge density wave transition in 2h-nbse\(_2\) in real space, Phys. Rev. B 89, 235115 (2014).
Phys. Rev. Lett. 85, 1738 (2000)

Appendix. The recovery curve $M(t)$

We measured $^{63}1/T_1$ at the $^{63}$Cu sites using the central transition. The standard formula for the relaxation recovery $M(t)$ calculated from the coupled rate equations is

$$M(t) = M_o - A[0.9e^{-(63T_1/3)\beta} + 0.1e^{-(t/63T_1)^\beta}], \quad (1)$$

where $M_o$, $A$, and $^{63}1/T_1$ are the free parameters, and the stretched exponent $\beta$ should be normally set to 1. We present examples of the normalized $M(t)$ curves measured with fixed $\tau = 2 \mu s$ in Fig. 9(a), together with the best fit with Eq.(1) for fixed $\beta = 1$. The standard fit with $\beta = 1$ is sufficiently good even at 50 K below $T_{\text{charge}}$ to illustrate the key aspects summarized in Fig. 4.

Also summarized in Fig. 9(b) is the $\tau$ dependence of $M(t)$ curves observed at 50 K with the phenomenological stretched fit. Lifting the constraint of $\beta = 1$ only marginally improves the fit, and the fitted value of $^{63}1/T_1$ hardly changes. The overall relaxation becomes faster for shorter $\tau$, accompanied by greater distribution, as evidenced by the smaller value of $\beta$. But the deviation of $\beta$ from the non-distributed case of 1 is not very significant. Since the signal intensity becomes very small below $T_{\text{charge}}$, especially for longer $\tau$, we fixed $\beta = 1$ for $^{63}$Cu sites to reduce the number of free fitting parameters in Eq. (1), and thereby reducing the scattering in the $^{63}1/T_1$ results.