Systematic $^{63}$Cu NQR studies of the stripe phase in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ for $0.07 \leq x \leq 0.25$

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We demonstrate that the integrated intensity of $^{63}$Cu nuclear quadrupole resonance (NQR) in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ decreases dramatically below the charge-stripe ordering temperature $T_{\text{charge}}$. Comparison with neutron and X-ray scattering indicates that the wipeout fraction $F(T)$ (i.e. the missing fraction of the integrated intensity of the NQR signal) represents the charge-stripe order parameter. The systematic study reveals bulk charge-stripe order throughout the superconducting region $0.07 \leq x \leq 0.25$. As a function of the reduced temperature $t \equiv T/T_{\text{charge}}$, the temperature dependence of $F(t)$ is sharpest for the hole concentration $x \sim 1/8$, indicating that $x \sim 1/8$ is the optimum concentration for stripe formation.

74.25.Nf, 74.72.Dn

I. INTRODUCTION

Research into the stripe phase of the CuO$_2$ plane has continued to expand since its experimental discovery in 1995 by Tranquada et al. Static charge-stripe order was observed for La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ with $x = 0.12$ with an onset temperature $T_{\text{charge}} \sim 65$K using neutron diffraction. The same charge-order superlattice peaks were recently confirmed using hard X-ray diffraction by Zimmerman et al. and a similar $T_{\text{charge}} \sim 70$K was found. Even more recently, charge-order superlattice peaks have been observed in La$_{1.6-x}$Nd$_{0.4}$Sr$_{0.15}$CuO$_4$ by Niemöller et al. using hard X-rays, and they report a slightly lower onset temperature $T_{\text{charge}} \sim 62$K. Charge transport studies of La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ by Uchida et al. also support a static charge-stripe picture. The Hall coefficient shows a dramatic decrease starting around $T_{\text{charge}}$, reflecting the one dimensional nature of the charge transport in the striped phase. Furthermore, the decrease is sharpest around the $x = 1/8$ doping where stripe order is believed to be most robust.

The observation of spin-stripe order has been reported for an even wider range of doping. Neutron scattering by Tranquada et al. successfully detected static spin-stripe order for $x = 0.12$, 0.15 and 0.20 at $T_{\text{spin}} \sim 50$K, 45K, and 20K respectively. These findings have been confirmed and extended with more recent measurements by Ichikawa et al. The neutron scattering experiments also observe a decreasing sublattice magnetization away from 1/8 doping, and incommensurabilities that are similar to those for the La$_{2-x}$Sr$_x$CuO$_4$ series. The most interesting feature however, is that the onset temperature $T_{\text{spin}}$ for spin-stripe order in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ at $x = 0.12$ is 50K, which is lower than $T_{\text{charge}}$ of 65K, implying that charge order is a precursor to spin order. In addition, a spin ordering temperature $T_{\text{spin}} = 30$K, 25K, and $\leq 4$K for $x = 0.12$, 0.15, and 0.20 respectively has been determined by Nachumi et al. using $\mu$SR. The $\mu$SR probe has a lower inherent frequency ($\sim 10$ Hz) compared with elastic neutron scattering ($\sim 10^{11}$ Hz), so the discrepancy in the spin ordering temperature between the $\mu$SR and neutron results indicate that the spin-stripe fluctuations gradually slow down with decreasing temperature below $T_{\text{charge}}$. That is, spin-stripe ordering is a glassy transition in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$.

In this paper, we utilize Cu nuclear quadrupole resonance (NQR) to probe stripe instabilities in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$. Cu NQR gives unique information about the charge environment at the Cu nuclear site. In particular, the resonance frequency for Cu NQR, conventionally called $\nu_Q (\sim 36$ MHz), is determined by the energy splitting of the $\pm \frac{3}{2} \leftrightarrow \pm \frac{1}{2}$ levels, which in turn is directly proportional to the electric field gradient (EFG) at the Cu nuclear site. The EFG itself is very sensitive to the charge environment, and we thus expect any change in the charge distribution within the CuO$_2$ plane, such as charge density waves (CDW), to directly affect Cu NQR. Earlier work on conventional CDW systems such as NbSe$_3$ and Rb$_{0.3}$MoO$_3$ made use of the extreme sensitivity of $\nu_Q$ to the EFG. Our novel approach makes use of wipeout effects in $^{63}$Cu NQR from which we obtain unique information about charge-stripe order in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$. The fundamental difference between NQR and neutron scattering is that the former is a local probe, while the latter requires spatial coherence, thus NQR and neutron scattering provide complimentary information about stripe physics. The NQR wipeout alone cannot give details of the spatial structure of the stripes, but being a local probe, NQR is very sensitive to charge-stripe order and the charge distribution. Therefore, we use the wipeout from $^{63}$Cu NQR in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ to determine the charge-stripe order parameter, which is a complementary method to the local $\mu$SR and hard X-ray diffraction measurements.
stripes by bulk scattering has proved difficult and up to now, no direct observation of charge-stripe order has been reported other than for La$_{1.48}$Nd$_{0.4}$Sr$_{0.15}$CuO$_4$ and La$_{1.48}$Nd$_{0.4}$Sr$_{0.15}$CuO$_4$. However by confirming that the NQR wipeout fraction $F(T)$ has identical temperature dependence as the neutron and X-ray charge-stripe order parameter for $x = 0.12$ and 0.15, we claim that $F(T)$ is the charge-stripe order parameter and we extend the detection of charge-stripe order in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ to $0.07 \leq x \leq 0.25$. We also establish that the transition is sharpest at $x = 0.12$.

The rest of the paper is presented as follows: in section II we go over the experimental details and present our results. In section III we discuss our NQR data in comparison with the neutron and X-ray measurements. Section IV contains the conclusions.

II. EXPERIMENTAL PROCEDURES AND RESULTS

A. Sample preparation

Powder samples of La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ with $x = 0.07, 0.09, 0.12, 0.15, 0.20$, and 0.25 were prepared by solid-state reactions of La$_2$O$_3$ (99.999%), Nd$_2$O$_3$ (99.99%), SrCO$_3$ (99.99%) and CuO (99.995%). The materials were mixed in the desired stoichiometry and pre-reacted at 850°C. The samples were then finely ground and heated to 950°C. This cycle was repeated several times. For the final reaction, the powder was pressed (0.6 GPa) into rods of cylindrical shape and annealed in flowing oxygen. The heat cycle for this final reaction was similar to that used by Breuer et al. where a highest temperature of 1150°C was used. All the samples were confirmed by X-ray diffraction to be single phased, and the room temperature orthorhombic splitting $[b-a]$ was in good agreement with [20].

B. Characterization of NQR spectra

The $^{63,65}$Cu NQR frequency spectra, or lineshapes, were measured using a 90° − τ − 180° − τ − echo phase-cycled pulse-sequence at fixed $\tau$. The resonant circuit was heavily damped so that short $\tau$ could be used (typically $\tau = 10 \sim 12\mu s$). In addition, the damping assured that the $Q$ value of the resonance circuit changed little with temperature, therefore pulse conditions and sensitivity remained constant.

Fig. 3(a) shows the lineshapes for all the La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ materials observed at 150K. At 150K, all the materials are in the LTO (low temperature orthorhombic) phase. The two different sites, conventionally called A and B, are present for all the materials and each is further split into two lines due to the presence of $^{63}$Cu and $^{65}$Cu isotopes. Note that the intensity of the B line compared with that of the A line goes roughly as the doping $x$, in good agreement with earlier reports by Yoshimura et al. [21]. It is generally believed that the B line originates from Cu nuclei underneath a Sr atom, and the A line from the Cu nuclei away from the Sr dopants. This picture is consistent with the observed ratio of intensities. We also observed an enhanced tail at the lower frequency side of the spectrum, possibly from Cu sites underneath a Nd atom. This lower frequency tail is also observed in La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$ spectra [22], presumably from Cu sites underneath an Eu atom.

In order to extract quantitative information about the lineshapes, each spectrum was fit to a convolution of Gaussians; two for the $^{63}$Cu and $^{65}$Cu A line, and two for the $^{63}$Cu and $^{65}$Cu B line (as shown for $x = 0.12$ in Fig. 3(a)). Fitting the A line with two Gaussians results in six parameters (two amplitudes, two peak positions corresponding to the NQR frequencies $^{63}\nu_Q$ and $^{65}\nu_Q$, and two widths). Using the known ratios of $^{63}\nu_Q/^{65}\nu_Q = Q^{63}/Q^{65} = 1.083$ and $^N^{63}/^N^{65} = 69/31$ for the quadrupole moments $Q$ and isotopic abundances $N$ respectively reduces the number of free parameters. The B line was fit in the same manner.

Results of A line HWHM (half width at half maximum) observed at 150K are shown in Fig. 1(b). The fitted value of the HWHM is sensitive to the lineshape data, especially at the tails, but the general trend is that the HWHM increases with $x$. The value is generally 50% higher than the HWHM observed for La$_{2.2-x}$Sr$_x$CuO$_4$ at the same doping [4]. This is possibly due to the increase in structural disorder due to Nd substitution.

Results of the $^{63}\nu_Q$ for the A and B line observed at 150K are shown in Fig. 1(c). The doping dependence of $\nu_Q$ for the A and B line in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ is consistent with previous studies on La$_{2.2-x}$Sr$_x$CuO$_4$ [21].

C. Temperature dependence of $^{63}$Cu NQR spectra

The temperature dependence of the lineshapes came in three forms:

1. The inhomogeneous linewidth (HWHM) of the $^{63}$Cu A line shown in Fig. 2(a) for $x = 0.12$ shows a smooth increase ($\sim 20\%$) from 300K to 20K. The same temperature dependence of the HWHM was found for all the samples. The HWHM data was used to estimate the integrated intensity of the NQR lineshape, so to avoid unnecessary scatter in this estimation, we typically used a linear fit to the HWHM.

2. Fig. 2(a) shows the temperature dependence of $^{63}\nu_Q$ for the $x = 0.12$ sample, and as can be seen, $^{63}\nu_Q$ shows a gradual increase of several hundred kHz from room temperature down to $T_{charge} = 70(7)K$, below which the rise in $^{63}\nu_Q$ is more dramatic. The other sam-
ples show qualitatively similar temperature dependence as discussed in section III.

(3) All of the materials showed a dramatic loss of signal intensity at temperatures below 150K. Fig. 2(b) shows the temperature dependence of the lineshapes for \( x = 0.12 \). The NQR intensity is proportional to the statistical Boltzmann factor \( e^{\Delta \nu Q/k_B T} \sim 1/T \) where \( \Delta \nu Q \ll k_B T \) in the present case. Accordingly, we multiplied each lineshape by \( T \) to take this into account. The loss of NQR intensity therefore implies that \( ^{63,65}\text{Cu} \) nuclear spins in certain segments of the sample become undetectable.

In order to quantify the loss of signal shown in Fig. 2(b), we estimated the temperature dependence of the integrated intensity for each material using a three-step process. (1) We calculated the area of the lineshape from the fitting procedure described above. (2) A Boltzmann factor was included at each temperature. (3) The lineshapes were corrected for spin echo decay as described below. We also attempted to make standard NQR frequency corrections of \( 1/f^2 \) to the lineshape intensity [14], but they did not affect our results. Fig. 3(a) shows the results of the temperature dependence of the integrated intensity for \( x = 0.12 \). When working out the lineshape area, only the integral of the A line intensity was used so that unnecessary scattering of the estimated total intensity was avoided. This was justified by previous high precision studies of the NQR intensity on a related compound \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) enriched with the \( ^{63}\text{Cu} \) isotope. As shown in Fig. 3(b), both A and B lines have identical temperature dependence of the integrated NQR intensity.

Corrections for spin echo decay, or the ‘\( T_2 \) corrections’, were made by measuring the spin-spin relaxation rate \( 1/T_2 \) at each temperature. Since \( T_2 \) exhibits slight dependence on frequency, in order to be rigorous one needs to make corrections for \( T_2 \) at every frequency rather than just at the peak of the A line. We tested whether the rigorous approach was necessary. For the \( x = 0.20 \) sample, the effect of \( T_2 \) was nearly uniform across the entire lineshape at 150K, but at 30K (shown in Fig. 4(a)), \( 1/T_2 \) increased towards the lower frequency side. Fig. 4(b) shows the integrated intensity with the ‘full \( T_2 \) correction’ at 150K and 30K compared with the intensity points deduced from correcting for \( T_2 \) just at the peak frequency. Both methods gave the same results within experimental error, so we conclude that the \( T_2 \) correction at the peak of the A line is sufficient.

As shown in Fig. 3(a) for \( x = 0.12 \), the intensity is constant within experimental error from 300K to 70(7)K, but then shows a dramatic drop to zero from 70(7)K to 10K. This is the ‘wipeout’ effect, and we will discuss the mechanism of the wipeout in section III. By taking the constant value between 300K and 70(7)K as the zero baseline and then inverting the plot, one obtains the fraction of signal lost, or the wipeout fraction \( F(T) \). As we demonstrate in section III, the onset temperature of \( F(T) \) is in good agreement with the charge-stripe ordering temperature \( T_{\text{charge}} \) determined by neutron/X-ray scattering.

Fig. 5 is a plot of the temperature dependence of \( F(T) \) for all the \( \text{La}_{1-x}\text{Nd}_x\text{Sr}_x\text{CuO}_4 \) samples. Even though \( T_{\text{charge}} \) and the transition widths are different for various \( x \), all show 100% wipeout, thus leaving no observable \( ^{63}\text{Cu} \) NQR signal by \( \sim 10K \). We emphasize that since we have taken \( T_2 \) corrections into account for the integrated intensity, the loss of observable NQR signal is not caused by a divergence of \( T_2 \) throughout the entire sample as often observed in the vicinity of magnetic long range order.

### D. \(^{63}\text{Cu} \) spin-lattice relaxation

Fig. 6 shows the spin-lattice relaxation rate \( 1/T_1 \) for \( \text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4 \), \( \text{La}_{1.45}\text{Nd}_{0.4}\text{Sr}_{0.15}\text{CuO}_4 \), and \( \text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.12}\text{CuO}_4 \) in addition to \( \text{La}_{1.88}\text{Sr}_{0.115}\text{CuO}_4 \) data from [23]. All \( ^{63}\text{Cu} \) \( T_1 \) data were taken at A line \( \nu_Q \), and all fit well to single exponential recoveries as expected for \( I = 3/2 \) nuclei in NQR. The relaxation rates for the four materials are equal within 10% from 300K to about 125K, but below about 125K, the \( ^{63}\text{Cu} \) NQR signal increases with decreasing temperature as \( 1/T_1 \). The \( ^{63}\text{Cu} \) \( T_1 \) data for \( \text{La}_{1.6-2}\text{Nd}_{0.4}\text{Sr}_{x}\text{CuO}_4 \) measured for the remaining \( ^{63}\text{Cu} \) NQR signal increases with deceasing temperature as \( \sim 1/T_1 \). For \( \text{La}_{0.68}\text{Eu}_{0.2}\text{Sr}_{0.12}\text{CuO}_4 \) [23] starts to increase at a lower temperature of 35(5)K. Wipeout effects below 30K prohibit further measurement of \( ^{63}\text{Cu} \) \( T_1 \) data for \( \text{La}_{1.6-2}\text{Nd}_{0.4}\text{Sr}_{x}\text{CuO}_4 \) and \( \text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.12}\text{CuO}_4 \). At 30K, even though \( ^{63}\text{Cu} \) \( T_1 \) for the remaining \( \sim 5\% \) of the signal is increasing with decreasing temperature, it is still measurable.

### E. \(^{63}\text{Cu} \) spin-echo decay

The NQR spin-echo intensity \( S(2\tau) \) depends strongly on \( 2\tau \) (where \( \tau \) is the time separation between the 90° and 180° pulses) as shown in Fig. 7 where \( S(2\tau) \) is plotted on a logarithmic scale. The data is from the \( \text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4 \) sample, and \( S(2\tau) \) is measured at \( ^{63}\nu_Q \) for the A line at each temperature. In order to correct the NQR integrated intensity for spin echo decay, one has to extrapolate the curve to the zero time \( 2\tau = 0 \). Most importantly, qualitative changes of the decay shape (such as loss of Gaussian curvature below \( T_{\text{charge}} \)) makes \( T_2 \) corrections essential in determining \( T_{\text{charge}} \) accurately. The shortest possible \( 2\tau \) is limited by the circuit dead time with a value of \( \sim 20\mu s \).

The fit used for the extrapolation has the functional form:

\[
S(2\tau) = S(0)e^{-2\tau/T_2}e^{-\left((2\tau)^2/2T_2^2\right)}
\]

where \( T_{2L} \) is the Lorentzian decay which has contributions from the Redfield \( T_1 \) process, and \( T_{2G} \) is the Gaussian decay which is dominated by the indirect spin-spin...
coupling between like spins \[16\]. In various high \(T_c\) and related copper-oxides, when the NQR linewidth is as small as \(\sim 200\) kHz, one can excite the entire \(^{63}\)Cu NQR spectrum with intense R.F. pulses. In such cases, one can give theoretical constraints on the Lorentzian contribution \(T_{2L}\) based on calculations of the spin-lattice relaxation process \[23\]. In the present case however, the full NQR linewidth (~3 MHz) is an order of magnitude larger than the strength of the R.F. pulses (~200 kHz). It is well known that this gives rise to artificial changes in the functional form of the spin-echo decay \[24\], making the Gaussian contribution more Lorentzian. Accordingly, we chose both \(T_{2L}\) and \(T_{2G}\) as free parameters. We note that use of the stretched exponential form of the fitting function:

\[
S(2\tau) = S(0)e^{-(2\tau/T_\beta)\beta}
\]

often used in the literature under similar circumstances did not affect our estimate of the temperature dependence of \(S(0)\).

At \(T_{\text{charge}}\) and below, the spin echo decay dramatically changed to a single rate Lorentzian. This can be seen by the loss of curvature in Fig. 4. As discussed in section III, this crossover provides an added signature for the onset of wipeout, however the crossover also generates more potential error in the estimate of the \(T_2\) corrected intensity \(S(0)\). Small changes of curvature in the fit creates large variations in the value of \(S(0)\) at \(T > T_{\text{charge}}\), so extra care had to be taken for data measurements and fits around \(T_{\text{charge}}\). This complexity is the reason for the larger error bars around \(T_{\text{charge}}\) in Fig. 3 (a). Consequently, \(T_{\text{charge}}\) is given a \(\pm 10\%\) error.

### III. DISCUSSION

#### A. Wipeout effects

We now compare the NQR wipeout fraction \(F(T)\) to elastic neutron diffraction studies carried out for \(\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4\) by Tranquada et al. \[1\], where charge-stripe order was discovered. Fig. 8(a) is a comparison of the temperature dependence of the square root of the transverse charge-order peak \(F\) normalized to 10K, along with our temperature dependence of \(F(T)\). We note that in general, the elastic scattering intensity represents the square of the order parameter (for example, the intensity of the magnetic Bragg scattering is the square of the sublattice magnetization \(M(T)\)) \[25\]. The identical temperature dependence of \(F(T)\) and the neutron charge-order parameter allows us to conclude that the wipeout fraction \(F(T)\) represents the charge-stripe order parameter. Also plotted is the square root of the static spin-ordered peaks \(M(T)\) for \(x = 0.12\) by neutron diffraction \[11\], and clearly indicates the wipeout fraction is triggered by charge-order and not spin-order.

Further evidence of charge-stripe order in \(\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4\) \[8\] and \(\text{La}_{1.45}\text{Nd}_{0.4}\text{Sr}_{0.15}\text{CuO}_4\) \[9\] has been reported using high energy x-ray scattering. Fig. 8(a) includes the X-ray results for \(x = 0.12\), where the square root of the longitudinal charge peaks are plotted, normalized to 10K. The X-ray data is in good agreement with both neutron and NQR data. Fig. 8(b) is a comparison of X-ray data for \(x = 0.15\) \[1\], where we have plotted the square root of the charge peaks in the transverse direction and compared it to our wipeout fraction \(F(T)\) for \(x = 0.15\). Again, the X-ray data agrees on the onset temperature \(T_{\text{charge}} \sim 60(6)\)K obtained from NQR. Both sets of X-ray data further support our identification of the wipeout fraction \(F(T)\) as the charge-stripe order parameter. Furthermore, we note that \(T_{\text{charge}}\) measured by NQR, X-ray, and neutron all agree for \(x = 0.12\) despite the different frequency scales of each probe, indicating that charge-stripes slow down to NQR time scales very quickly near \(T_{\text{charge}}\).

Given the evidence for charge-stripe ordering in \(\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4\) \[8\] and \(\text{La}_{1.45}\text{Nd}_{0.4}\text{Sr}_{0.15}\text{CuO}_4\) \[9\], we will now argue that the Cu NQR ‘wipeout’ \[19\] effect comes as a natural consequence of the ordering. Because the resonant frequency \(^{63}\nu_Q\) is directly proportional to the electric field gradient (EFG) at the Cu site, we expect NQR to be very sensitive to local charge distributions. Indeed, a spatial modulation of the hole concentration ranging from 0.5 to 0 holes per Cu atom results in as much as an \(\sim 8\) MHz \[21\] instantaneous variation of \(^{63}\nu_Q\). In the proposed stripe model \[4\] at \(x = 1/8\), rivers of hole rich CuO chains with effectively 0.5 holes per Cu are separated by bare three-leg CuO ladders with no holes. We therefore expect that below the onset temperature for charge-stripe order \(T_{\text{charge}}\), regions which contain stripe fluctuations will have instantaneously varying \(^{63}\nu_Q\) distributions, and if these fluctuations are on the NQR timescale (i.e. fluctuate at frequencies of order \(^{63}\nu_Q\)), the resonance condition in those regions will no longer be well defined. Furthermore, charge order turns on slow spin fluctuations \[26\] resulting in divergences in the spin-lattice \(^{63}\nu_{1T_1}\) \[17\] and spin-spin \(^{63}\nu_{1T_2}\) relaxation rates within the stripe ordered domains (note that the measured \(^{63}\nu_{1T_1}\) and \(^{63}\nu_{1T_2}\) in Fig. 9 and Fig. 10 do not necessarily reflect the relaxation rates in the ordered segments of the sample but they are a measure of relaxation rates of the segments that have not yet ordered). Both these effects will result in a loss of \(^{63}\)Cu NQR signal intensity and we therefore expect the fraction of the intensity loss to be a good measure of stripe order.

It is worth mentioning that at low enough temperatures, an NQR like signal reappears for the case of \(\text{La}_{1.88}\text{Ba}_{0.12}\text{CuO}_4\) \[28\]. Below \(\lesssim \text{10K}\), static magnetic hyperfine fields strongly perturb the Cu NQR.
spectrum causing a line broadening from low frequencies up to 80 MHz [28]. We confirmed the findings from [28] for La_{1.88}Sr_{0.12}CuO_{4}, where a broad, featureless Zeeman perturbed NQR spectrum at 1.7K was reported. We note that the low temperature spectrum for La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} is further complicated by magnetic field contributions from the Nd ions that order at \( T < 3K \).

We now discuss a possible fit of the temperature dependence of \( F(T) \). Conventional CDW ground states have many common characteristics with other broken symmetry ground states such as superconducting and spin density wave ground states. In particular, within the weak coupling limit, the thermodynamics of the phase transition and the temperature dependence of the order parameter are the same as those of a BCS superconductor [29,30]. To see if this is the case, we present Fig. 8(c) where \( F(t) \) is plotted as a function of the reduced temperature \( t = T/T_{charge} \) for all the La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} samples. The solid curve is the BCS form of the condensate density \( f_{d}(t) \) in the dynamic limit [31], and the dashed curves are a guide for the eye. The data for the \( x = 0.09, 0.15, \) and 0.20 samples look similar, and the \( x = 0.07 \) data stands out as having the broadest transition. The \( x = 0.25 \) data looks identical to the \( x = 0.20 \) data and has been omitted from this plot. The temperature dependence of \( F(t) \) for \( x = 0.12 \) appears to be the sharpest and shows the best agreement with \( f_{d}(t) \). From the wipeout mechanism we have proposed, this is expected since \( F(T) \) is related to the fraction of the CuO_{2} plane where condensate fluctuations exist. The fact that the transition is sharpest around \( x \approx 1/8 \) could indicate that the charge-stripe is most stable around \( x \approx 1/8 \).

We now present the phase diagram of La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} in Fig. 9 with \( T_{charge}, T_{spin} \) according neutron scattering [22], \( T_{spin} \) obtained by \( \mu \mathrm{SR} \) [2], the superconducting transition temperature \( T_{c} \), and the LTO to LTT (low temperature tetragonal) or LTLO (low temperature less orthorhombic) transition temperature \( T_{LTLO} \) [2]. We measured the bulk magnetic susceptibility with a SQUID magnetometer in a field of 10 Oe, and we estimated \( T_{c} \) by taking the slope at the half point in the diamagnetic response and extrapolating to zero susceptibility. The \( x = 0.12 \) and 0.07 samples show residual superconducting components, and the \( x = 0.25 \) sample shows no diamagnetic response down to 3K. The \( x = 0.09, 0.15, \) and 0.20 samples have the largest low temperature susceptibility \(-4\pi\chi\) with Meissner fractions corresponding to \( \approx 40\% \). Fig. 9 demonstrates for the first time that charge-stripe order exists throughout the entire superconducting region of La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4}. Furthermore, the 100% wipeout indicates that the charge-stripe transition involves the \textit{entire} CuO_{2} plane.

Fig. 9 clearly indicates that \( T_{charge} \) for La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} does not strictly coincide with \( T_{LTLO} \approx 60K \) except at \( x = 0.12 \), suggesting that the LTO-LTT structural transition is not the primary cause of the charge anomaly at \( T_{charge} \). Further support of this statement comes from NQR wipeout measurements we made on La_{1.8}Eu_{0.2}Sr_{x}CuO_{4} with \( x = 0.07, 0.12, 0.16, \) and 0.20 all with LTO-LTT structural transition temperature \( T_{LTLO} \approx 130K \) [53] that are higher than for La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} where \( T_{LTLO} \approx 60K \). We used exactly the same techniques to measure \( F(T) \) and \( T_{charge} \) for La_{1.8-x}Eu_{0.2}Sr_{x}CuO_{4} as described earlier for La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4}, and the results shown in Fig. 9 indicate that \( T_{charge}(x) \) for La_{1.8-x}Eu_{0.2}Sr_{x}CuO_{4} and La_{1.6-x}Nd_{0.4}Sr_{x}CuO_{4} is the same within the experimental error. This clearly shows that \( T_{charge} \) and \( T_{LTLO} \) are not strictly correlated and that the LTT structural transition is not the primary origin of the charge anomaly. We also note that spin-stripe order has been observed in La_{2-x}Sr_{x}CuO_{4} \( (x = 0.12, 0.15, \) and \( x = 0.05 \) and La_{2}CuO_{4+\delta} [31], where both materials do not even have the LTO-LTT structural phase transition.

B. \( ^{63}\text{Cu} \) spin-lattice relaxation rate \( ^{63}1/T_{1} \)

We now compare the temperature dependence of \( ^{63}1/T_{1} \) in La_{1.48}Nd_{0.4}Sr_{0.12}CuO_{4}, La_{1.45}Nd_{0.4}Sr_{0.15}CuO_{4}, La_{1.68}Eu_{0.2}Sr_{0.12}CuO_{4} and La_{1.88}Sr_{0.115}CuO_{4} [28] in light of previous \( ^{63}1/T_{1} \) measurements by Itoh \textit{et al.} [31] on a variety of the rare earth (R) doped 1-2-3 materials RBa_{2}Cu_{3}O_{7-\delta} \( [R = Y, \text{Nd, Eu}] \).

As discussed in [24], the 4f electron moments from the trivalent rare earth \( R^{3+} \) ions contribute to the \( ^{63}1/T_{1} \) primarily through a dipole interaction between the 4f moment and the Cu nuclear moment, giving the general form:

\[
^{63}1/T_{1} = (1/T_{1})_{4f} + (1/T_{1})_{Cu}
\]  

We find that the \( ^{63}1/T_{1} \) data for La_{1.88}Sr_{0.115}CuO_{4} and YBa_{2}Cu_{3}O_{7-\delta} have no \( (1/T_{1})_{4f} \) component, and \( (1/T_{1})_{Cu} \) for the observable segments of the NQR signal in La_{1.88}Sr_{0.115}CuO_{4} decreases with decreasing temperature [31].

(1) La_{1.48}Nd_{0.4}Sr_{0.12}CuO_{4}, La_{1.45}Nd_{0.4}Sr_{0.15}CuO_{4}, and NdBa_{2}Cu_{3}O_{7-\delta} all have Nd\(^{3+}\) ions which have \( J = 9/2 \) ground states, and a large \((1/T_{1})_{4f}\) contribution is apparent with decreasing temperature in both cases. The temperature dependence of \((1/T_{1})_{4f}\) depends on the details of the crystal field [31]. \((1/T_{1})_{4f}\) for NdBa_{2}Cu_{3}O_{7-\delta} is temperature independent down to at least 1K, which is the expected behaviour in the limit...
$T_N \ll T \ll \Delta_1$ where $T_N = 0.52K$ is the ordering temperature and $\Delta_1 = 140K$ is the crystal field splitting between the ground state and the first excited state. The La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_{4}$ and La$_{1.45}$Nd$_{0.4}$Sr$_{0.15}$CuO$_{4}$ data shows that $(1/T_{1}I_{f})$ increases with decreasing temperature indicating that $\Delta_1 \ll 140K$ in these materials.

(3) La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ and EuBa$_2$Cu$_3$O$_{7-\gamma}$ both have Eu$^{3+}$ ions which have $J = 0$ ground states and $J = 1$ first excited states split by the spin-orbit constant $\lambda$. The EuBa$_2$Cu$_3$O$_{7-\gamma}$ data shows negligible $(1/T_{1}I_{f})$ contribution at and below 50K, consistent with the $\lambda \approx 450K \gg T$. Since the first excited state is predominantly split by spin-orbit effects, one also expects negligible $(1/T_{1}I_{f})$ contributions in La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ below 50K. However, as the La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ data indicates, there is an increase in $63^1T_{1}$ below 35(5)K, suggesting that the observable $^{63}$Cu NQR signal senses diverging low frequency spin fluctuations due to critical slowing down of Cu moments toward $T_{spin} = 25 \sim 27K$ as observed by $\mu$SR [2] for La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$. Both $\mu$SR and NQR have similar inherent probing frequencies of $\sim 10^5$ Hz, thus one expects $T_{spin}$ even for a glassy transition to be comparable for both measurements, as is the case for La$_{2-x}$Sr$_x$CuO$_{4}$ according to $\mu$SR [3] and $^{139}$La NQR [4]. We also find that $63^1/T_{1}T$ in La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ decreases slightly below $T_{charge}$ prior to the onset of critical divergence of $63^1T_{1}/T$ toward $T_{spin}$. Even though reduction of $63^1T_{1}T$ is usually attributed to pseudogap in the spin excitation spectrum, it is not clear whether that is the case here. We emphasize that the reduction of $63^1T_{1}T$ is for the observable fraction of the CuO$_2$ planes, and that the unobservable fraction may have divergent $63^1T_{1}$ at $T \ll T_{charge}$.

We would like to add that although in the case of La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ below $T = 125K$, $(1/T_{1}I_{f})$ dominates over $(1/T_{1}I_{f}C_{Cu})$ and no spin-ordering can be inferred, $\mu$SR measurements report that there is spin-ordering at temperatures $T_{spin}$ similar to La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_{4}$. $\mu$SR measurements were obtained for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_{4}$ [4] where $T_{spin} \sim 30K$, and a reduced value for La$_{1.45}$Nd$_{0.4}$Sr$_{0.15}$CuO$_{4}$ of $T_{spin} \sim 25K$ has also been reported [4]. It was also shown by $\mu$SR [1] that $T_{spin} \sim 25K$ for La$_{1.85-y}$Nd$_y$Sr$_{0.15}$CuO$_{4}$ is independent of the Nd concentration $y$ for $0.3 \leq y \leq 0.6$.

Fig. 1 includes the values of $T_{spin}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_{4}$ according to neutron scattering [2] and $\mu$SR [4]. No spontaneous magnetization is detected for $x = 0.20$ by $\mu$SR down to 4K in contrast to the static stripe-observations by neutron scattering [2], where $T_{spin} \sim 20K$ for $x = 0.20$. Indeed, for $x = 0.10, 0.12, 0.15$, and 0.20, $T_{spin}$ is consistently $\sim 20K$ higher for neutron scattering [2] than for $\mu$SR measurements [4]. Further evidence of the glassy nature of the spin-ordering comes from ESR (electron spin resonance) studies by Kataev et al. [3] for La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_{4}$ at $x \sim 1/8$, where they clearly observe that the Cu spin fluctuation frequency $\omega_{sf}$ shows a strong temperature dependence below $T_{charge}$.

C. $^{63}$Cu spin-echo decay $^{63}1/T_{2}$

We now discuss the temperature dependence of the spin echo decay both above and below $T_{charge}$. Above $T_{charge}$, the spin echo decay from all the samples were fit using the two free parameters $T_{2G}$ and $T_{2L}$ in Eq. 1. We found that greatly reducing the excitation range from $\sim 200$ kHz to $\sim 50$ kHz, caused a significant reduction of the Gaussian component. This can be understood [2] in the context of inhomogeneous broadening, where reducing the frequency range has the effect of reducing the number of like spins responsible for the indirect spin-spin (Gaussian) decay. The reverse situation where the frequency range is kept fixed but the inhomogeneous line is broadened will also reduce the number of like spins. Either way, detuning the spin-spin mechanism [2] results in an apparent reduction of the Gaussian curvature in the spin echo envelope and results in the Gaussian contribution to the second moment appear more Lorentzian in character. However, even if we use weak R.F. pulses and thereby eliminate the Gaussian curvature in the spin echo decay, we found that the extrapolated value S(0) does not change.

Even though the NQR HWHM does not increase significantly around $T_{charge}$ (Fig. 2(a)) and we maintain the same strength of R.F. pulses, we still observed a dramatic reduction of the Gaussian component at and below $T_{charge}$, indicating that there is a mechanism below $T_{charge}$ that inherently eliminates the Gaussian process. The same mechanism that causes the wipeout can also be used to explain the crossover, namely that the spatial charge modulations below $T_{charge}$ deter the indirect interaction by creating site to site $^{63}$NQ variations, thereby reducing the number of like spins and thus eliminating the Gaussian decay. Below $T_{charge}$, the spin-echo decay was always Lorentzian with $T_{2L} \ll T_{1}$. We observed that reducing the spectral excitation range in this case did not change $T_{2L}$. Fig. 1 shows the temperature dependence of $63^1T_{2L}$ for a selection of the La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_{4}$ samples and La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$. The data at each doping starts at $T_{charge}$ and finishes when the wipeout is nearly complete. The doping dependence of $63^1T_{2L}$ shows a systematic decease with increasing hole doping x. If $63^1T_{2L}$ was entirely dominated by 4f moment fluctuations from the Nd$^{3+}$ ion, $63^1T_{2L}$ would not show this doping dependence. Moreover, as argued for the $63^1T_{1}$ data, La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ has a negligible 4f moment at 50K, yet within scattering, $63^1T_{2L}$ at 50K is the same as for La$_{1.48}$Nd$_{0.4}$Sr$_{0.15}$CuO$_{4}$.

The fact that $T_{2L} \ll T_{1}$, coupled with the fact that the Nd$^{3+}$ ion is not the primary source of relaxation, leads us
to believe that within the observable domains there exist certain magnetic hyperfine fields \( H_{hf} \) at the Cu site that fluctuate with a correlation time \( \tau_c \) satisfies the motional narrowing limit \( \gamma_n H_{hf} \tau_c \ll 1 \) [10] (where \( \gamma_n \) is the Cu gyromagnetic ratio). We infer that these fluctuations are primarily magnetic rather than quadrupolar by verifying that:

\[
\frac{65}{63} \frac{1}{T_{2L}} \sim \frac{65}{63} \frac{\gamma_n^2}{\gamma_n^2} = 1.15 \quad (4)
\]

The frequency dependence of \( 1/T_{2L} \) for the \( x = 0.20 \) sample was shown in Fig. 11(a) at \( T = 30K \leq T_{\text{charge}} \), and indeed, the 15% rise at lower frequency is consistent with primarily magnetic relaxation. It is also known that in the motional narrowing limit [10],

\[
1/T_{2L} \sim \frac{\gamma_n^2 H_{hf}^2}{\tau_c} \quad (5)
\]

but since \( \gamma_n H_{hf} \) is not known, one cannot estimate \( \tau_c \). We note however that a sliding motion of CDW’s in conventional CDW conduces motions of narrowing [17]. Our present case is unconventional in the sense that we are observing the fraction of the material that has not yet ordered, and that charge order turns on slow spin dynamics [20].

D. The temperature dependence of \( ^{63}\nu_Q \)

Fig. 11 shows the temperature dependence of A line \( ^{63}\nu_Q \) for \( x = 0.07 \) and 0.12. The closed symbols indicate temperatures above \( T_{\text{charge}} \), and open symbols below \( T_{\text{charge}} \). Also shown is the vertical line marking the structural transition temperature \( T_{\text{LTT}} \) from LTO to LTT (or LTLO). We define \( T_{\nu_Q} \) as the temperature below which the rise in the A line \( ^{63}\nu_Q \) for the observable part of the signal starts to increase. \( T_{\nu_Q} = 70(7)K \) for \( x = 0.12 \), and for \( T_{\nu_Q} \approx 130(13)K \) for \( x = 0.07 \). \( T_{\text{HTT}} \) is also defined as the HTT (high temperature tetragonal) to LTO transition temperature [12].

The measurement of the temperature dependence for the A line \( ^{63}\nu_Q \) for \( x > 0.12 \) has added complexity to it. As shown in Fig. 11(a), the B line increases in amplitude with Sr doping, and unfortunately, the B line \( ^{63}\text{Cu} \) frequency coincides with the main A line \( ^{63}\text{Cu} \). This is evident in Fig. 11(a) for \( x = 0.20 \) where the shape of the main peak is largely due to the \( ^{63}\text{Cu} \) B line. Accurate determination of the A line \( ^{63}\nu_Q \) temperature dependence therefore implies taking very careful lineshapes of a decreasing signal intensity below \( T_{\text{charge}} \). We have however been able to determine that the A line \( ^{63}\nu_Q \) for all the samples increases with decreasing temperature down to \( \sim 10K \) and that the rise below \( T_{\nu_Q} = 70(7)K \) is sharp for \( x = 0.12 \, x = 0.09 \) and 0.12 have qualitatively the same temperature dependence of the A line \( ^{63}\nu_Q \).

There are at least two possible explanations for the sharp rise in \( ^{63}\nu_Q \) below \( T_{\nu_Q} \) for \( x = 0.12 \). One is based on electronic effects. If we assume that \( T_{\nu_Q} \) is in the vicinity of \( T_{\text{charge}} \) which is certainly true for \( x = 0.12 \), one can postulate that regions with lower hole concentrations wipe out at higher temperatures in such a way that \( ^{63}\nu_Q \) will appear to increase with decreasing temperature (recall that regions with higher hole concentrations have higher \( ^{63}\nu_Q \) values).

Another possible explanation for the large rise in \( ^{63}\nu_Q \) arises from structural distortion. In order to obtain a semi-quantitative idea of how structural effects change \( ^{63}\nu_Q \), we calculate the electric field gradient (EFG) at the Cu site by a point charge lattice summation method similar to that reported in [16]. The two inputs necessary for the EFG calculation are the positions of the ions \( (r_i) \) relative to the Cu nucleus and the point charge of the ions \( (e_i) \). The lattice components of the EFG are calculated from summations such as:

\[
e_{i,\text{latt}}^{\text{eq}} = \sum_i e_i \left( \frac{3z_i^2 - r_i^2}{r_i^4} \right) \quad (6)
\]

where we included all the ions within a sphere of radius 100Å from the origin. The ionic charges for the A line are assigned as follows:

\[
La(\text{Nd}) : +3 \quad O_p : -(2 - \frac{x}{2}) \quad O_a : -2 \quad Cu : +2
\]

where \( O_p \) is the planar oxygen, \( O_a \) the apical oxygen. The room temperature values of the lattice constants are taken from high resolution X-ray diffraction [14]. With decreasing temperature, the percentile change of the lattice constants are taken from [15], where the orthorhombic splitting is reported to go as:

\[
[b - a](x, T) \sim [b - a]_o(x) \times (1 - T/T_{\text{HTT}})^{2\beta} \quad (8)
\]

where \( \beta = \frac{5}{3} \). The oxygen octahedron tilting angle is taken to have the form [13]:

\[
\Phi(x, T) \sim \Phi_o(x) \times (1 - T/T_{\text{HTT}})^{2\beta} \quad (9)
\]

also with \( \beta = \frac{5}{3} \). Below \( T_{\text{LTT}} \), \( [b - a] \) is set to zero, and \( \Phi \) is kept at its maximum angle \( \Phi_o(x) \) [40] whose value is given below.

For \( ^{63}\nu_Q \) in units of MHz and \( e_{i,\text{latt}}^{\text{eq}} \) in units of \( \text{emu} \times 10^{-14} \), \( ^{63}\nu_Q \) has the general empirical form [43]:

\[
^{63}\nu_Q = A(x) - B \times e_{i,\text{latt}}^{\text{eq}}(x, T) \quad (10)
\]

where \( A(x) \approx 70 \text{ MHz} \) is the contribution from the hole in the 3d_{x^2-y^2} orbital and its value was chosen to match the room temperature value of the A line \( ^{63}\nu_Q \). B is determined by antishielding effects [15] and the value is estimated to be 14.1 MHz from an empirical fit performed
by Shimizu et al. on a large pool of data for different superconducting materials. Our calculations typically give $eq_{tilt} \approx 2.7$, consistent with lattice EFG's for high $T_c$ cuprates [13]. We note that the only parameter we have adjusted to best fit the data is the constant $A(x)$. All the other parameters have been taken from other experimental sources.

We can check that the calculation is in semi-quantitative agreement with the data by confirming the monotonic rise in $63\nu_Q$ with decreasing temperature in the LTO phase (without tilting the octahedra, the calculation predicts that $63\nu_Q$ decreases due to lattice shrinking). Indeed the calculation does show semi-quantitative agreement with the data above $T_{\nu_Q}$.

An important result from the calculation is that the change from LTO to LTT has little effect on $63\nu_Q$, i.e. $63\nu_Q$ is insensitive to azimuthal rotations of the octahedra about the c-axis. This justifies the fact that we have neglected any intermediate LTLO phases [12]. However, $63\nu_Q$ is sensitive to the angle that the octahedra are tilted from the c-axis. Thus in order to try to account for the dramatic increase of $63\nu_Q$ below $T_{\nu_Q}$ for the observable NQR signal just from structural effects, we further tilted the octahedra from the c-axis starting at $T_{\nu_Q}$ and we took the same temperature dependence of the angle in the LTO phase:

$$\Phi(x, T < T_{\nu_Q}) \sim \Phi_0(x) + \Phi_1(x) \times (1 - T/T_{\nu_Q})^b \quad (11)$$

The only additional parameters now are the maximum tilt angle of the extra tilt called $\Phi_1(x)$, and $T_{\nu_Q}$. It is clear for $x = 0.12$ that $T_{\nu_Q} \sim T_{\text{charge}} = 70(7)$K, and for $x = 0.07$ we have also used $T_{\nu_Q} \sim T_{\text{charge}} = 130(13)$K. The lower line in each plot is with $\Phi_1(x) = 0$ and the upper line is for finite $\Phi_1(x)$. The rise in $63\nu_Q$ can now be reproduced, and the angles used were as follows:

We point out that the values of $\Phi_1(x)$ that best reproduce the data should only be taken as estimates. The calculation so far described is naturally very sensitive to the exact ion positions and to the constant $B$. For instance, using a larger $B$ will make $63\nu_Q$ more sensitive to changes in $eq_{tilt}$, and one would then use lower values of $\Phi_1(x)$ to reproduce the data.

There is no conclusive evidence either way. We are reminded however that neither possibility has to involve all segments, just those that have not yet been wiped out. Even though there have been no reports from bulk scattering experiments [11,12] that there is further octahedron tilting at temperatures below $T_c$, bulk scattering experiments [14,15] measure the spatial average of the tilting angle while $63\nu_Q$ is a local probe of the observable segments.

Using the same method as described above, we also calculated the effects charge-stripe formation alone would have on the EFG for $x = 1/8$. Firstly, we confirmed that adding 0.5 holes uniformly on the planar oxygens can account for the rise in the A line $63\nu_Q$ of $\sim 8$ MHz, in agreement with experimental observations in La$_{2-x}$Sr$_x$CuO$_4$ [21]. Next, we took the conventional stripe picture where the holes lie uniformly in the river of holes across one Cu chain separated by bare three leg ladders, and we predicted an NQR line splitting into the three peaks corresponding to the three distinct Cu sites (actually each peak was further split $\sim 300$ kHz into two due to perpendicular stripes from neighbouring planes). In the calculation, we interpreted one hole on a Cu site to mean one hole distributed evenly around its four surrounding planar oxygens. The highest frequency peak corresponding to the Cu site on the river shifted $\sim 6$ MHz above the uniformly doped case, while the lowest peak corresponding to the Cu at the centre of the 3-leg ladder shifted $\sim 3$ MHz below the uniform case. We also tried various charge-stripe configurations [16], and we found that any smoothing of the charge distribution reduced the peak splitting.

Contrary to these calculated results of $63\nu_Q$ in stripes, no NQR line splitting for the observable part of the signal was observed experimentally at or below $T_{\text{charge}}$. This implies that there is no static stripe order in the observable part of the CuO$_2$ plane. The signal we can observe below $T_{\text{charge}}$ is either from islands that have not yet ordered, or from islands with quasi-static order but with NQR lines that are motionally narrowed.

**IV. CONCLUSIONS**

In this paper, we have reported a systematic study of the temperature and doping dependence of stripe instabilities in La$_{1.6-x}$Nd$_{0.4}Sr_x$CuO$_4$ throughout the superconducting regime based on $63\nu$ NQR. Our novel approach takes advantage of the extreme sensitivity of $63\nu$ Cu NQR to charge-stripes. We have confirmed that the NQR wipeout fraction $F(T)$ is a good measure of the charge-stripe order parameter [14,15], and we have extended earlier measurements of the charge-stripe order parameter based on diffraction techniques beyond $x = 0.12, 0.15$ to $0.07 \leq x \leq 0.25$ and in doing so, obtain an extended phase diagram of the La$_{1.6-x}$Nd$_{0.4}Sr_x$CuO$_4$ system. We have also presented and discussed the temperature and doping dependence of the NQR parameters $63\nu_Q$, $631/T_1$, and $631/T_2$.

We have shown that robust charge-stripe order continues to hold up to $x = 0.25$, where no static hyperfine fields have been reported by $\mu$SR. This implies that completely static spin-ordering is not a necessity for charge-ordering. On the other hand, the Lorentzian spin-spin relaxation rate $631/T_2$, observed below $T_{\text{charge}}$ suggests that charge-stripes continue to fluctuate slowly even below $T_{\text{charge}}$.

Our observation that $T_{\text{charge}}$ is higher for lower doping $x$ is counterintuitive, given that the charge-stripe transi-
tion is sharpest for $x = \frac{1}{3}$. On the other hand, the lower hole concentration $x$, the stronger the tendency towards charge localization. This might explain why the onset of charge order is as much as a factor of two higher in temperature for $x=0.07$ than for $x=0.12$. We should also recall that NQR is a local probe sensitive to stripes no matter how they are disordered.

Comparison of the stripe-superconductivity phase diagram of La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ with our results obtained for La$_{2-x}$Sr$_x$CuO$_4$ reveals a possibly striking feature: it would appear that charge-stripe order stops when $T_{\text{charge}}$ becomes lower than $T_c$, which for La$_{2-x}$Sr$_x$CuO$_4$ happens to occur at $x \sim 1/8$, but for La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ does not happen due to the highly suppressed $T_c$ and the larger $T_{\text{charge}}$. A natural question to ask is whether the stripe instabilities and superconductivity compete. If one looks at the La$_{2-x}$Sr$_x$CuO$_4$ with $x > 1/8$, where static-stripe ordering is suppressed and superconductivity is robust, one may conclude they compete with each other. However, inelastic neutron scattering measurements indicate that low energy ($\gtrsim 2\text{meV}$) dynamic stripe fluctuations extend beyond $x = 1/8$, and perhaps they coexist even in YBa$_2$Cu$_3$O$_{6.6}$ [5]. Furthermore, neutron scattering on La$_{2/3}$CuO$_{4+\delta}$ by Y.S. Lee et al. report that the elastic spin-order intensity appears at the same temperature as the superconductivity, suggesting that the two phenomena are strongly correlated. Our new stripe-superconductivity phase diagram for La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ in Fig. 4 also clearly indicates that the superconductivity phase boundary exists within the charge stripe phase boundary.

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[1] A.W. Hunt, P.M. Singer, K.R. Thurber, and T. Imai, Phys. Rev. Lett. 82, 4300 (1999).
[2] T. Suzuki, T. Goto, K. Chiba, T. Shinoda, T. Fukase, H. Kimura, K. Yamada, M. Ohashi, and Y. Yamaguchi, Phys. Rev. B 57, R5229 (1998).
[3] H. Kimura et al., to be published.
[4] S. Wakimoto, G. Shirane, Y. Endoh, K. Hirota, S. Ueke, K. Yamada, R.J. Birgeneau, M.A. Kastner, Y.S. Lee, P.M. Gehring, and S.H. Lee, cond-mat/9902201.
[5] Y.S. Lee, R.J. Birgeneau, M.A. Kastner, Y. Endoh, S. Wakimoto, K. Yamada, R.W. Erwin, S.-H. Lee, G. Shirane, cond-mat/9902157.
[6] J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura, and S. Uchida, Nature 375, 561 (1995).
[7] J.M. Tranquada, J. D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B 54, 7489 (1996).
[8] M.v. Zimmermann, A. Vigliante, T. Niemöller, N. Ichikawa, T. Frello, J. Madsen, P. Wochner, S. Uchida, N.H. Andersen, J.M. Tranquada, D. Gibbs, and J.R. Schneider, Europhys. Lett. 41, 629 (1998).
[9] T. Niemöller, H. Hünnefeld, J.R. Schneider, N. Ichikawa, S. Uchida, T. Frello, N.H. Andersen, and J.M. Tranquada, cond-mat/9904385.
[10] S. Uchida, N. Ichikawa, T. Noda, and H. Eisaki, to be published in vol. 125 Springer Series in Solid State Sciences.
[11] J.M. Tranquada, J.D. Axe, N. Ichikawa, A.R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. 78, 338 (1997).
[12] N. Ichikawa, S. Uchida, J.M. Tranquada, T. Niemöller, P.M. Gehring, S.-H. Lee, and J.R. Schneider in preparation; N. Ichikawa, thesis, University of Tokyo (1999).
[13] K. Yamada, C.H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R.J. Birgeneau, M. Greven, M.A. Kastner, and Y.J. Kim, Phys. Rev. B 57, 6105 (1998).
[14] B. Nachumi, Y. Fudamoto, A. Keren, K.M. Kojima, M. Larkin, G.M. Luke, J. Merrin, O. Tchernyshyov, Y.J. Uemura, N. Ichikawa, M. Goto, H. Takagi, S. Uchida, M.K. Crawford, E.M. McCarron, D.E. MacLaughlin, and R.H. Heffner, Phys. Rev. B 58, 8760 (1998).
[15] A. Abragam, Principles of Nuclear Magnetism (Oxford University Press, 1978).
[16] C.P. Slichter, Principles of Magnetic Resonance (Springer-Verlag, New York, 1989), 3rd ed.
[17] J.H. Ross, Jr., Z. Wang, and C.P. Slichter, Phys. Rev. Lett. 56, 663 (1986).
[18] K. Nomura, T. Sambongi, K. Kume, and M. Sato, Physica B 143, 117 (1986).
[19] J. Wada, Magnetic Resonance in Metals (Oxford University Press, Oxford, 1971).
[20] M. Breuer, B. Bünchner, R. Müller, M. Cramm, O. Heymer, and D. Wohlleben, Physica C 208, 217 (1993).
[21] K. Yoshimura, T. Uemura, M. Kato, K. Kosuge, T. Imai, and H. Yasuoka, Hyperfine Int. 79, 876 (1993).
[22] A.W. Hunt et al., in preparation.
[23] C.H. Pennington, D.J. Durand, C.P. Slichter, J.P. Rice, E.D. Bukowski, and D.M. Ginsberg, Phys. Rev. B 39, 274 (1989).
[24] D. Hone, V. Jaccarino, T. Ngwe, and P. Pincus, Physical Review 186, 291 (1969).
[25] S.W. Lovesey, Theory of Neutron Scattering from Condensed Matter. (Oxford Univ. Press, 1984).
[26] J.M. Tranquada, N. Ichikawa, and S. Uchida, cond-mat/9906141.
[27] K.R. Thurber, T. Imai, T. Saitoh, M. Azuma, M. Takano, and F.C. Chou, cond-mat/9906141.
[28] H. Tou, M. Matsumura, and H. Yamagata, J. Phys. Soc. Jap. 61, 1477 (1992).
FIG. 1. (a) Lineshapes for La$_{1.6-x}$Nd$_x$Sr$_2$CuO$_4$ where the value $0.07 \leq x \leq 0.25$ is shown for each line. All lineshapes were taken at 150K and all are normalised to equal heights for purposes of comparison. The raw data points are shown along with their fits (see text) and the decomposition of the fit for $x = 0.12$ is shown. (b) $^{63}$Cu A line HWHM (▲) obtained by fitting the 150K lineshapes. (c) A line $^{63}$Hy (■), B line $^{63}$Hv (○) both at 150K.

FIG. 2. (a) Temperature dependence of the $^{63}$Cu A line HWHM for $x = 0.12$ (○) along with a linear extrapolation, and the temperature dependence of A line $^{63}$Hy for $x = 0.12$ (▲). (b) $x = 0.12$ lineshapes at 150K (○), 77K (■), 60K (△), 40K (▲), all corrected for Boltzmann factor (see text).

FIG. 3. (a) Temperature dependence of the integrated $^{63}$Cu NQR intensity for La$_{1.4}$Nd$_x$Sr$_{0.12}$CuO$_4$ in arbitrary units. Also shown, the onset temperature for wipeout $T_{\text{charge}}$, and the (○) show Gaussian curvature in the spin echo decay, (●) do not (see text). (b) Temperature dependence of the integrated intensity for La$_{1.875}$Ba$_{0.125}$CuO$_4$ for the A line (△) and B line (▲).

FIG. 4. (a) $^{63}$I/T$_1$ in ms$^{-1}$ (●) as a function of frequency at 30K for the $x = 0.20$ sample. Also shown, the 30K lineshape in arbitrary units measured at fixed $\tau = 0.12\mu$s (○) along with its fit and the decomposition of the fit. (b) Temperature dependence of the integrated intensity for $x = 0.20$ measured by the full $T_2$ correction (●), or by correcting for $T_2$ just at the peak of the A line (x).

FIG. 5. Wipeout fraction $F(T)$ in La$_{1.6-x}$Nd$_x$Sr$_2$CuO$_4$ for $x = 0.07$ (▲), $x = 0.09$ (△), $x = 0.12$ (●), $x = 0.15$ (○), $x = 0.20$ (▼), and $x = 0.25$ (◦). The solid lines through the points are a guide for the eye.

FIG. 6. $^{63}$I/T$_1$ temperature dependence for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ (●), La$_{1.45}$Nd$_{0.4}$Sr$_{0.15}$CuO$_4$ (△), La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_4$ (○) and the solid line La$_{1.88}$Sr$_{0.15}$CuO$_4$ (22), all measured in ms$^{-1}$.

FIG. 7. Spin echo decay for La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ at the various temperatures normalized by the Boltzmann factor. Also shown, are spin echo envelopes fit with Gaussian curvature (○), and without (●).

FIG. 8. (a) Plots of the wipeout fraction $F(T)$ (○), the neutron charge order parameter $^7$ (●), the X-ray order parameter $^8$ (▲), and the neutron spin order parameter $^7$ (x) for $x = 0.12$. Each data set is normalized to its 10K value and the dotted lines are guides for the eye. (b) Wipeout fraction $F(T)$ (○) along with the X-ray order parameter $^9$ (●) for $x = 0.15$. Each data set is normalized to its 10K value and the dotted line is a guide for the eye. (c) Wipeout fraction $F(t)$ as a function of the reduced temperature $t = T/T_{\text{charge}}$ for $x = 0.07$ (▼), $x = 0.09$ (○), $x = 0.12$ (●), $x = 0.15$ (△), and $x = 0.20$ (▲). The solid line is $F(t)$, and the dashed lines are guides for the eye.
FIG. 9. Phase diagram with $T_{\text{charge}}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ (●), $T_{\text{charge}}$ for La$_{1.8-x}$Eu$_{0.2}$Sr$_{x}$CuO$_{4}$ (○), $T_{\text{spin}}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ according to neutron scattering (×) $^{12}$, $T_{\text{spin}}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ according to μSR (+) $^{14}$, and $T_{c}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ (▲) along with a shaded region to highlight the superconducting phase. Also shown is the LTO to LTT (or LTLO) boundary (broken line) for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ $^{32}$, and two dashed lines joining the (×) and (+) points as a guide for the eye. Near the solubility limit, we find $T_{\text{charge}} = 40(10)$K for La$_{1.35}$Nd$_{0.4}$Sr$_{0.25}$CuO$_{4}$ (not shown) to be about the same as $T_{\text{charge}} = 40(6)$K for La$_{1.40}$Nd$_{0.4}$Sr$_{0.20}$CuO$_{4}$.

FIG. 10. Temperature dependence of $^{63}1/T_{2}$ in $ms^{-1}$ for La$_{1.6-x}$Nd$_{0.4}$Sr$_{x}$CuO$_{4}$ with $x = 0.07$ (●), $x = 0.12$ (○), $x = 0.25$ (▲), and also for La$_{1.68}$Eu$_{0.2}$Sr$_{0.12}$CuO$_{4}$ (■).

FIG. 11. Temperature dependence of $^{63}\nu_{Q}$ A line for (a) $x = 0.07$ and (b) $x = 0.12$. (●) corresponds to data for $T > T_{\text{charge}}$ and (○) to $T < T_{\text{charge}}$. The vertical line indicates the LTO-LTT transition temperature. The upper curve of the fit corresponds to finite $\Phi_{t}(x)$, and the lower curve to $\Phi_{t}(x) = 0$.

TABLE I.

| $x$   | $\Phi_{o}(x)$ | $\Phi_{t}(x)$ |
|-------|---------------|---------------|
| 0.07  | 5.5°          | 4.5°          |
| 0.12  | 4.8°          | 4.5°          |
Spin Echo Intensity (arb. units)

- A line
- B line

(a) 0.07 0.09 0.12 0.15 0.2 0.25 f (MHz)

(b) HWHM (MHz)

(c) Sr (x)

63 Q

36 38.4 (MHz)

35.2 37.6 40

0.05 0.1 0.15 0.2 0.25
Figure (a) shows the variation of $\delta_{\omega}^A$ (MHz) with temperature $T$ (K). The data points are accompanied by error bars. Figure (b) displays the spin echo intensity (arb. units) as a function of frequency $f$ (MHz) at different temperatures: 40K, 60K, 77K, and 150K.
Figure (a) shows the spin echo intensity as a function of frequency ($f$) in MHz, with the data points connected by a line. The y-axis represents $\frac{^{63}I}{T_{2L}}$ (ms$^{-1}$). The data points are accompanied by error bars indicating uncertainty.

Figure (b) illustrates the integrated intensity as a function of temperature ($T$) in K. The data points are shown with error bars, and the x-axis represents temperature. The graph includes a fitted line to the data points.
\[
\frac{63}{T_1} \text{ (ms}^{-1})
\]

\[
T \text{ (K)}
\]
