

# Glassy spin freezing and NMR wipeout effect in the high- $T_c$ superconductor $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ : Critical discussion of the role of stripes

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We report on  $^{139}\text{La}$  and  $^{63}\text{Cu}$  NMR/NQR measurements in the high- $T_c$  superconductor  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ , with  $T_c=26.5$  K. Spin fluctuations probed by  $^{139}\text{La}$  spin-lattice relaxation ( $T_1$ ) continuously slow down on cooling through  $T_c$ . We argue that spin freezing and superconductivity are bulk effects in this sample. Thus both phenomena have to coexist microscopically. The distribution of  $^{139}\text{La}$   $T_1$  values at low temperature reveals a wide spread of spin fluctuation frequencies in  $\text{CuO}_2$  planes. A simple estimate shows that  $^{63}\text{Cu}$  nuclei at sites where electronic fluctuations are the slowest are not observable (*wipeout* effect) because relaxation times are too short. This means that the  $^{63}\text{Cu}$  NQR wipeout, observed in this sample, can be explained primarily by slow magnetic, rather than charge, fluctuations. The magnetic origin of the wipeout is still compatible with a connection between wipeout and charge order [as proposed by Hunt *et al.*, Phys. Rev. Lett. **82**, 4300 (1999)], but this connection is indirect. On the other hand, since the wipeout fraction is not an intensive quantity it cannot define a proper order parameter and cannot be used by itself as a criterion for the existence of a stripe phase.

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## I. INTRODUCTION

Although  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  is probably the best studied high- $T_c$  superconductor,<sup>1,2</sup> it continues to reveal interesting phenomena that were not observed or that were overlooked in the past. This is even true for microscopic and powerful probes such as nuclear magnetic resonance (NMR), nuclear quadrupole resonance (NQR), or muon spin rotation ( $\mu\text{SR}$ ), which have already made major contributions to our understanding of these compounds (for reviews, see Refs. 3–6). Two important features were highlighted recently.

(i) The coexistence of superconductivity with a frozen magnetic state called a “cluster spin glass” at concentrations  $0.06 \leq x \leq 0.10$ , an early result which was confirmed by recent studies (see Refs. 7–9 and 10 for a concise review). In

principle, the coexistence of superconductivity with frozen spins or localized charges is rather hard to conceive. Indeed, in the cuprates, a link between superconductivity and the characteristic frequency of spin fluctuations<sup>3,5,11</sup> was inferred from NMR data at relatively high temperature,  $T \gg T_c$ . Thus it now appears equally important to characterize the evolution of magnetic fluctuations close to and below  $T_c$ , in e.g.,  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

(ii) A strong “wipeout effect” for the  $^{63}\text{Cu}$  NQR signal at concentrations  $x < 0.12$ : the number of  $^{63}\text{Cu}$  nuclei contributing to the signal decreases on cooling (even above  $T_c$ ) and the signal completely disappears at low temperature ( $T$ ).<sup>12</sup> Interestingly, a very similar phenomenology, with spin freezing and Cu NQR/NMR wipeout, is observed<sup>12–16</sup> in cuprate materials where doped holes have been shown to order in

linear single rows, known as charge *stripes*.<sup>17</sup> In fact, it was even discovered that the wipeout fraction (i.e., the fraction of unobserved Cu nuclei) has the same  $T$  dependence as the ‘‘stripe order parameter’’ (actually the intensity of superlattice peaks in neutron and x-ray scattering) in these materials.<sup>12,13</sup> As explained by Hunt *et al.*, the wipeout effect can be caused by very slow (in the MHz range) *charge and/or spin* fluctuations.<sup>12</sup> However, the presentation of the wipeout as a probe of charge dynamics<sup>12</sup> has led to the belief that the effect was predominantly caused by the charges,<sup>18</sup> and that stripe order in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  was characterized by ultraslow charge motion.<sup>19</sup> This idea was recently challenged by Curro *et al.*, who attributed the wipeout effect to slow spin fluctuations exclusively and conclude that ‘‘Cu wipeout is not a measure of the stripe order parameter.’’<sup>15</sup> Since the discovery of Hunt *et al.*<sup>12</sup> raised the hope of having a new tool to detect charge stripes (which have so far proved elusive in most materials), it is clearly important to better understand the origin of the wipeout, and to decide if this effect can be considered as a criterion for stripe order.

Here, we report on  $^{139}\text{La}$  NMR/NQR and  $^{63}\text{Cu}$  NQR measurements in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ . The main conclusions of this work are the following: (1) Bulk superconductivity coexists with frozen magnetic moments throughout the sample. (2) The slow and inhomogeneous spin dynamics characterizing the freezing process provide the most likely explanation for the Cu NQR wipeout effect.

The paper is organized as follows: first, the main magnetic properties of  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  at low  $T$  are recalled in Sec. II. Section III gives a basic NMR background, focused on spin-lattice relaxation and wipeout effects. Experimental details, including a discussion of the NMR line shape, are described in Sec. IV, with a brief account of magnetization measurements which indicate bulk superconductivity in the sample. Section V is devoted to the NMR/NQR results and to their analysis. The results are summarized in Sec. VI, together with a discussion in a more general perspective.

## II. CONTEXT OF THE WORK

We specify the context of the experiment by summarizing some magnetic properties of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $x \approx 0.1$ , focusing on relatively low temperatures,  $T \leq 100$  K. Hunt *et al.* reported a loss of the Cu NQR signal, below  $\sim 70$  K for  $x = 0.09$  and below  $\sim 50$  K for  $x = 0.115$ , in ceramic samples.<sup>12</sup> A similar wipeout effect may thus be anticipated below  $\sim 60$  K for  $x = 0.10$ .

Incommensurate elastic peaks are found in neutron scattering below  $\approx 15$  K,<sup>20</sup> i.e., in the superconducting state ( $T_c \sim 30$  K). By analogy with the results of Tranquada *et al.* in  $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ ,<sup>21</sup> this modulated antiferromagnetic order is suspected to result from the ordering of spin domains between antiphase walls formed by charge stripes. However, the corresponding charge order peaks have not been observed. On the other hand, a much lower spin-ordering temperature of 1.2 K was reported from a  $\mu\text{SR}$  study.<sup>7</sup> The difference between neutron and  $\mu\text{SR}$  results is presumably ascribed to the glassy nature of this ordering: spin fluctuations continuously slow down over a wide  $T$

range, so a dynamic measurement probes a frequency-dependent ordering temperature. Actually, the existence of frozen spins at  $x = 0.10$  was already inferred by Ohsugi *et al.* from the broadening of the NQR line at 1.4 K.<sup>22</sup> However, it was not clear if all or part of the sample was magnetic. Slowing down of spin fluctuations is also visible in electron paramagnetic resonance measurements.<sup>23,24</sup>

In conclusion,  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  lies at an interesting position in the phase diagram of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ : while being close to the  $x \sim 0.12$  composition where magnetic order is quite strong,<sup>20,25,26</sup> it shows magnetic order only at quite low  $T$  and has about two-thirds of the highest  $T_c$  achievable in this system (at ambient pressure). It should also show a wide  $T$  range of Cu NQR wipeout effect.

## III. NMR BACKGROUND

$^{139}\text{La}$  and  $^{63}\text{Cu}$  are complementary NMR and NQR probes.  $^{139}\text{La}$  nuclei are coupled to the magnetic moments of  $\text{Cu}^{2+}$  electrons through the hyperfine field which results from both a transferred contact interaction (via orbital overlap with the apical oxygen) and a direct dipolar interaction. The hyperfine field is estimated to be  $\approx 1 - 2$  kOe/ $\mu_B$ .<sup>27</sup> In contrast,  $^{63}\text{Cu}$  and  $^{65}\text{Cu}$  nuclei experience a much larger coupling to both the on-site  $\text{Cu}^{2+}$  spin [anisotropic hyperfine field  $A_c \approx -134$  kOe/ $\mu_B$ ,  $A_{ab} \approx 2$  kOe/ $\mu_B$  (Ref. 28)], and to the four  $\text{Cu}^{2+}$  first neighbors [isotropic transferred coupling  $B \approx 35$  kOe/ $\mu_B$  (Ref. 28)].

### A. Nuclear relaxation

Nuclear-spin-lattice relaxation occurs through temporal fluctuations of the local magnetic field (magnetic relaxation) and/or of the electric-field gradient (quadrupolar relaxation). For magnetic relaxation, the spin-lattice relaxation rate  $1/T_1$  of a given nucleus is proportional to the *square* of its gyromagnetic ratio  $\gamma_n$  and to the *square* of the transverse components ( $h_x, h_y$ ) of the local field (the  $z$  quantization axis is the direction of the external magnetic field  $H$ ),

$$\frac{1}{T_1} = \frac{\gamma_n^2}{2} \int_{-\infty}^{+\infty} \overline{\langle h_+(t)h_-(0) \rangle} \exp(i\omega_n t) dt, \quad (1)$$

where the horizontal bar denotes the ensemble average, and  $\omega_n$  is the nuclear Larmor frequency.

With  $^{63}h_{\perp}^2/^{139}h_{\perp}^2 \approx 5000$  for  $H \parallel c$  and with  $(^{63}\gamma/^{139}\gamma)^2 \approx 3.5$ , one immediately finds that fluctuations of  $\text{Cu}^{2+}$  spins will lead to  $T_1$  values for  $^{63}\text{Cu}$  which are shorter by about four orders of magnitude compared to the values for  $^{139}\text{La}$  (the exact number depends on the wave-vector dependence of spin fluctuations, which is ignored in the above estimate).

For fluctuations of the local field of the form  $\overline{\langle h_+(t)h_-(0) \rangle} = \overline{\langle h_{\perp}^2 \rangle} \exp(-2t/\tau_c)$ , where  $\tau_c$  is called the correlation time, a standard expression for  $T_1$  can be derived:<sup>29</sup>

$$\frac{1}{T_1} = \gamma_n^2 \overline{\langle h_{\perp}^2 \rangle} \frac{2\tau_c}{1 + \omega_n^2 \tau_c^2}. \quad (2)$$

Slowing down of magnetic fluctuations means that  $\tau_c$  increases on cooling ( $\tau_c^{-1} \gg \omega_n$ ), and this leads to an increase of  $T_1^{-1}$ , which eventually reaches a maximum when  $\tau_c^{-1} = \omega_n$  [Eq. (2)]. The existence of a maximum in  $T_1^{-1}$  as a function of  $\tau_c$  is a rather general feature (more general than the particular form of  $T_1$  assumed above), which also holds when the system cannot be described by a single value of  $\tau_c$ , but rather by a distribution of correlation times. The maximum of  $1/T_1$  vs  $T$  becomes broader in this case. The temperature at which  $1/T_1$  reaches a maximum defines the freezing temperature  $T_g$  at the NMR time scale. When slowing down occurs over a rather wide temperature range, the value of  $T_g$  determined by another experimental technique, with a different time scale, may differ significantly from the NMR value. With typically  $\omega_n^{-1} \sim 10^{-8}$  s,  $^{139}\text{La}$  NMR is a relatively slow probe, with a time scale comparable to  $\mu\text{SR}$ . These concepts were recently well illustrated for spin-freezing phenomena in Eu-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>13,14</sup>

The spin-spin relaxation time  $T_2$  defines the characteristic time decay of the echo height in a spin-echo sequence. Thus,  $T_2$  determines the time available for recording the NMR signal. In most solids,  $T_2$  is determined by nuclear dipole-dipole interaction. In the cuprates,  $T_2$  of  $^{63,65}\text{Cu}$  nuclei is dominated by two stronger processes:  $T_{2G}$ , which comes from indirect exchange between Cu nuclei via the non-local electronic susceptibility;<sup>30</sup> and  $T_{2R}$ , the Redfield contribution, which is a function of  $T_1$ .<sup>29,31</sup> Since both processes are proportional to squares of hyperfine fields,  $^{139}\text{La}$  nuclei have a much longer  $T_2$  ( $\sim$  ms) than  $^{63,65}\text{Cu}$  nuclei ( $\sim 1-50$   $\mu\text{s}$ ).

### B. Wipeout effects in NMR/NQR

The NMR/NQR signal is directly proportional to the population difference between adjacent nuclear levels. As such, it is proportional to  $1/T$  and to the number of nuclei in the sample. In practice, since the observation occurs at a finite time after a radio-frequency pulse, the measured signal is reduced from its maximum possible value because of the  $T_2$  process. The decrease is typically of Lorentzian or Gaussian type. So, in order to check if all nuclei are observed as a function of temperature, the signal should be renormalized by a factor  $T$  and then corrected for the  $T_2$  effect by extrapolating its magnitude at time zero.

The term *wipeout* effect was introduced in order to describe the loss of NMR signal due to non-magnetic impurity doping in metals.<sup>32</sup> The decrease of the NMR signal occurred because of the large spread of resonance frequencies out of a given spectral window. Similar loss of NMR signal has been well-documented for localized magnetic moments in metals (Ruderman-Kittel-Kasuya-Yosida oscillations).<sup>33</sup> A transition to a magnetically ordered phase may also lead to an apparent loss of signal, because the internal field causes a shift of the resonance positions, with possibly sizable broadening.

On the other hand, a loss of signal may be produced by a dramatic shortening of  $T_2$  (or of  $T_1$ , through the  $T_{2R}$  term). The correct signal cannot be obtained by extrapolating the measured signal at time zero, when relaxation times for part of the nuclei become shorter than the ‘‘dead time’’ of the

spectrometer, i.e., some nuclei have relaxed so fast that the signal coming from them cannot be digitized. If this occurs for all nuclei in the sample, the signal is completely lost. As seen above, very short relaxation times occur if the spectral density of electronic fluctuations is large at the Larmor frequency.

Finally, when the Larmor frequency is directly defined by the value of the hyperfine electric or magnetic coupling (as in NQR or zero-field NMR), strong fluctuations of these couplings at the observation time scale will also cause a loss of the resonant signal. Because of the slow fluctuations involved, very short nuclear relaxation times are likely to occur in such cases. Thus the various contributions to the wipeout effect may not be distinguishable.

In summary, a loss of NMR signal can result from static effects (modification of the line shape), from dynamical effects or from both. These effects can occur homogeneously or inhomogeneously in the sample. Inhomogeneity frequently leads to a wipeout effect which is only partial, in which case careful measurements are required in order to realize that part of the signal has been lost. Not surprisingly, wipeout effects are observed in canonical spin-glasses close to the glass transition.<sup>34-36</sup>

## IV. EXPERIMENTAL DETAILS

### A. NMR measurements

Most of the experiments were performed on a crystal ( $m = 113$  mg) grown by the traveling solvent floating-zone method.<sup>37</sup> It is a piece of the large crystal used by Petit *et al.*, for neutron-scattering measurements.<sup>38</sup> In the course of the NMR experiments, it was found that the sample was not a true single crystal, as a part of it had a different orientation from the rest. While this does not affect our analysis of the NMR/NQR signal intensity (because all frequencies were integrated), it affects the line shapes. This sample was then cut into two equal pieces, one of which was confirmed to be a single crystal of very high quality from both neutron and x-ray diffraction. We then performed  $^{139}\text{La}$  NMR line-shape measurements as well as  $^{139}\text{La}$   $T_1$  and  $T_2$  measurements on a well-isolated line in this single crystal ( $\theta = -15^\circ$  in Fig. 1). The recovery laws were strictly identical to those in the original sample. Thus the distribution of  $T_1$  values, which will be discussed below, is intrinsic. Within experimental accuracy,  $T_1$  and  $T_2$  are the same on the different lines.

NMR spectra were obtained from a single Fourier transform of half of the spin echo signal when the line was sufficiently narrow. For lines broader than the frequency window of the excitation, spectra were obtained by summing Fourier transforms at equally spaced frequencies or by recording the spin-echo integral/amplitude at different frequencies.  $^{63}\text{Cu}$  and  $^{139}\text{La}$  NQR spectra were recorded with the latter method.

The  $^{139}\text{La}$  NMR spectrum with  $H \parallel c$  in the single crystal shows a single peak which splits into four peaks as the magnetic field is tilted away from the  $c$  axis (one of the lines is not well defined, but only appears as a shoulder; see Fig. 1). The angular dependence of this spectrum (Fig. 1), and the fact that the  $^{139}\text{La}$  NQR spectrum does not show well-

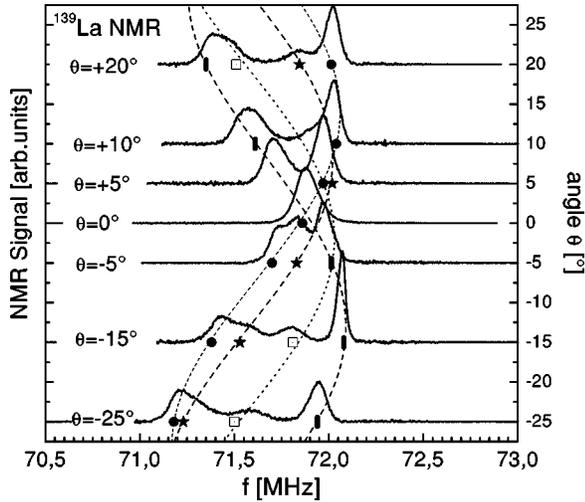


FIG. 1.  $^{139}\text{La}$  NMR spectra in a field  $H = 12$  T at  $T = 57$  K.  $\theta$  is the angle between  $H$  and the  $c$  axis. The line splitting is related to different directions of the main axis of the electric-field gradient (see text). The dashed lines represent theoretical calculations of the line positions due to second-order quadrupolar shifts. The symbols identify the positions of various lines (several symbols are omitted at small angles, for clarity).

separated lines but a single peak (Fig. 2), indicate that the splitting is due to different values of the angle between  $H$  and the direction of the principal axis of the electric-field gradient tensor  $V_{zz}$ . Since these four angles are roughly equal when  $H \parallel c$  (Fig. 1), the effect originates from different directions of  $V_{zz}$  in the crystal, with equal tilts from the  $c$  axis. Unambiguous identification of the different directions would require a full angle-dependence study within two perpendicular rotation planes. Because of sample geometry, this could not be performed here. The various directions of  $V_{zz}$  may correspond to different tilt directions of  $\text{CuO}_6$  octahedra. The  $T$  dependence of the shifts and linewidth show that the tilt angles increase smoothly on cooling from  $\approx 340$  K down to  $\approx 100$  K, with a saturation at lower  $T$ .

These results motivated us to reexamine  $^{139}\text{La}$  NMR spectra in the single crystal of  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  that we used for a previous study.<sup>8</sup> Our measurements, at higher magnetic fields and with improved experimental conditions, revealed

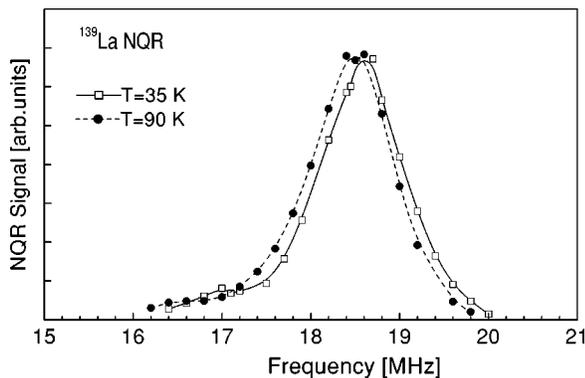


FIG. 2. High-frequency ( $3\nu_Q$ ) transition of the  $^{139}\text{La}$  NQR spectra at 90 and 35 K.

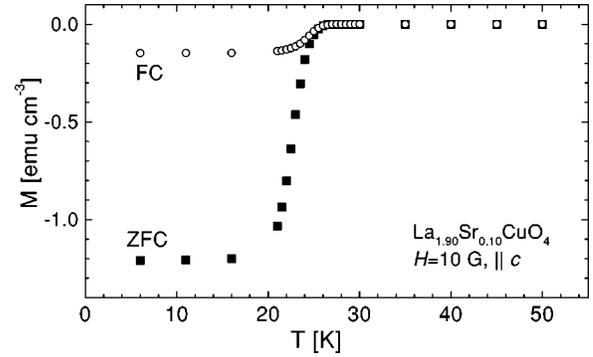


FIG. 3. Field cooled (FC) and zero-field-cooled (ZFC) magnetizations of the  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  single crystal [from Huh *et al.* (Ref. 39)].

that the spectrum is composed of at least three lines, whose separation is predominantly of quadrupolar origin, as in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ . Thus this finding invalidates the hypothesis in Ref. 8 of only two lines split by a purely magnetic effect.

Measuring the magnitude of the signal intensity requires care. In order to ensure that both the radio-frequency excitation and the detection of the signal remain identical at all temperatures, experimental conditions such as the  $50 \Omega$  matching and the  $Q$  factor of the resonant circuit should be carefully controlled. Here this was made easier by the fact that the capacitors for impedance matching and frequency tuning were outside the probe, at a constant room temperature.

Because of flux expulsion in the superconducting state, the NMR signal in a single crystal is reduced drastically below  $T_c$ . In order to study the spin dynamics down to low temperature, we have applied high magnetic fields up to  $H = 23.2$  T. Such a field is expected to reduce  $T_c$  down to a few Kelvin, although the exact  $T_c$  is not known here, and in any case hard to define given the broadening of the transition under field. The experiments at 23.2 T were carried out in a high homogeneity resistive magnet of the National High Magnetic Field Laboratory in Tallahassee, FL. Other measurements up to 15 T were carried out in superconducting coils.

## B. Superconducting properties

Magnetization measurements were performed on the polycrystalline sample and on the single crystal, with almost identical results. In Fig. 3 results for the single crystal are reported.<sup>39</sup> The sample quality can be checked from the narrow width ( $\approx 5$  K) of the superconducting transition, which has an onset at  $T_c = 26.5$  K. It is particularly difficult to establish bulk superconductivity from single-crystal measurements in fields much higher than a few G.<sup>40,41</sup> Nevertheless, a study of the high-field reversible magnetization, similar to that in Ref. 42, concludes that our sample is a bulk superconductor.<sup>39</sup>

## V. NQR AND NMR RESULTS

### A. $^{63}\text{Cu}$ NQR spectra: Wipeout effect

As a preliminary step of this study, we intended to check the presence of a wipeout effect, as previously reported by

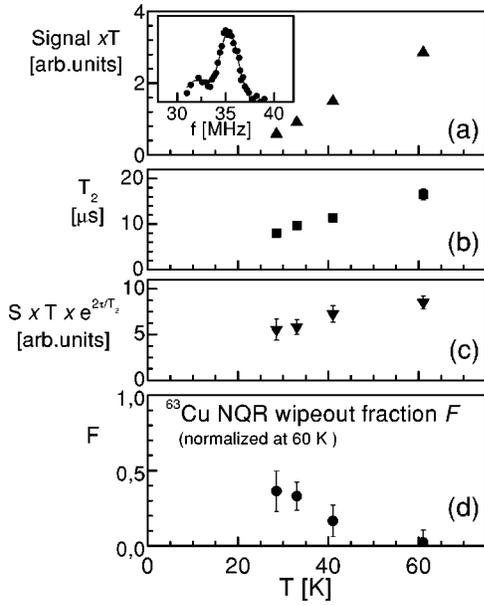


FIG. 4. (a)  $T$ -normalized signal intensity from the  $^{63}\text{Cu}$  NQR spectra, all taken in the very same experimental conditions. The inset shows a typical  $^{63,65}\text{Cu}$  NQR spectrum at  $T=34$  K. (b)  $T$  dependence of  $T_2$  for  $^{63}\text{Cu}$ . (c)  $T$ - and  $T_2$ -normalized signal intensity. (d) Wipeout fraction (see text), which is a measure of the unobserved signal.

Hunt *et al.*<sup>12</sup> The  $^{63}\text{Cu}$  signal was recorded in a NQR experiment, i.e., in zero external magnetic field. Because of the excessive loss of signal in the superconducting state, our data are limited to a narrow  $T$  range above  $T_c=26.5$  K.  $T_2$  was measured at different frequencies on the NQR spectrum, and was found to shorten on decreasing frequency, in agreement with Ref. 12. However, the frequency dependence is relatively weak, so that it is sufficient to correct the frequency-integrated signal from the value of  $T_2$  at the center of the line only.<sup>13</sup>  $T_2$  was estimated from a single exponential fit [ $s(t) \propto \exp(-2\pi t/T_2)$ ] of the echo decay (the accuracy of the data did not allow us to distinguish between Lorentzian and Gaussian forms of the echo decay).

Figure 4 shows the results for the signal intensity,  $T_2$  and the wipeout fraction  $F(T)=[s^*(60\text{ K})-s^*(T)]/s^*(60\text{ K})$  ( $s^*$  is the signal corrected for  $T_2$  and temperature, and integrated over frequencies). There is clearly a wipeout effect, which basically agrees with the results of Hunt *et al.*<sup>12</sup> (our values appear somewhat lower, presumably because of the normalization to the intensity at 60 K, our highest temperature data point; Hunt *et al.* found 20% of wipeout for  $x=0.09$ , at this temperature). We found an increase of  $300 \pm 50$  kHz of  $^{63}\nu_Q$  from 61 to 28 K, resembling the one observed by Singer *et al.*<sup>13</sup> in the stripe-ordered phase.

### B. $^{139}\text{La}$ NMR spectra: Low- $T$ magnetic broadening

As shown in Fig. 5, the  $^{139}\text{La}$  central line in a field of 23.2 T broadens on cooling below about 50 K, with a saturation of the width below  $\approx 10$  K. The same broadening is seen at 15 T (Ref. 43) (not shown), but not at 9.4 T above 15 K (Fig. 6). This strongly suggests that the broadening is of magnetic

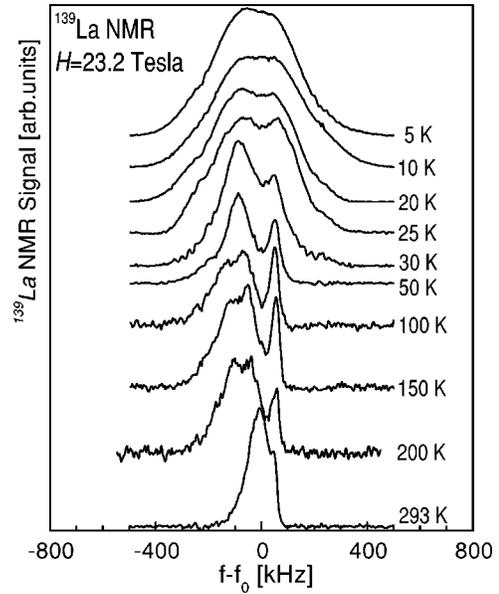


FIG. 5.  $^{139}\text{La}$  NMR spectra (central transition) in a field of  $H=23.2$  T ( $f_0=139.528$  MHz). See Sec. IV for explanation of the line splitting. Note the broadening below  $\sim 50$  K.

origin, as previously observed in  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$ .<sup>8</sup> The broadening is not seen at 9.4 T, because the linewidth is dominated by the large quadrupolar broadening (which varies as  $1/H_0$ ). The  $^{139}\text{La}$  NMR broadening is not an artifact related to signal loss at the center of the spectrum (see next subsection), and is not of dynamic origin ( $^{139}T_2$  is much longer than the inverse linewidth in the single crystal), at least above  $\sim 10$  K. Thus, the broadening indicates a spread of local fields along the  $z$  direction. This is a purely paramagnetic effect since  $T_1$  data show no sign for frozen moments in the range 10–60 K, but a smooth evolution toward freezing at lower  $T$  (see Fig. 9 and the discussion below). Clearly, the broadening cannot be caused by frozen magnetic regions at so high temperatures. This effect is somehow similar to the one observed in Zn-doped materials<sup>44</sup> and is not necessarily seen in bulk magnetization measurements, which sums the contributions from all staggered moments.

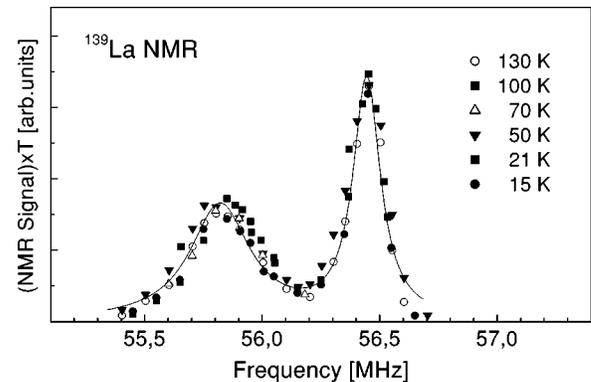


FIG. 6.  $T$ -normalized NMR signal for the  $^{139}\text{La}$  NMR central line. The solid line is a guide to the eye. Except for minor changes, the data collapse on a single curve. There is no wipeout effect in this temperature range.

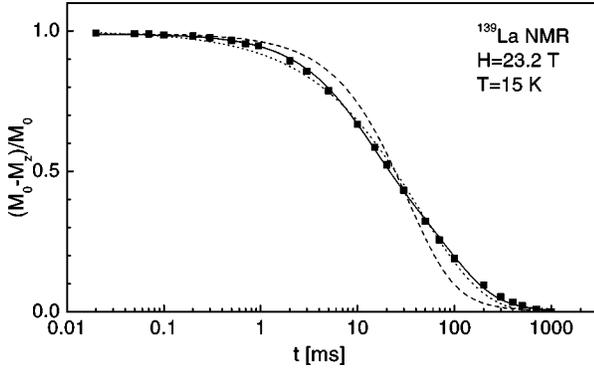


FIG. 7. Time dependence of the  $^{139}\text{La}$  NMR signal  $M_z$  after saturation of the central line at  $T=19$  K, in a field of 23.2 T [ $M_0 = M_z(t=\infty)$ ]. The dashed and solid lines are fits with one and two components, respectively, with each component following the theoretical recovery law for magnetic relaxation (see the text). The dotted line is a fit according to the stretched form (see Ref. 51).

Note that the above discussion makes clear that *the broadening of the NMR line cannot be taken as evidence for magnetic order*. This erroneous criterion has been sometimes used to define a Néel temperature in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $x \approx 0.12$ .<sup>45–47</sup>

### C. $^{139}\text{La}$ NMR spectra: Absence of wipeout effect

The  $T$  dependence of the  $^{139}\text{La}$  intensity was measured for the NMR central line ( $m_I = +1/2 \leftrightarrow -1/2$  transition) in a field of 9.4 T.  $T_2$  was observed to shorten on cooling. However, *in the range 100–15 K*,  $^{139}T_2$  of the order of ms remains much longer than the delay between  $\pi/2$  and  $\pi$  NMR pulses ( $\tau \approx 30 \mu\text{s}$ ), so the  $T_2$  correction is essentially negligible for  $^{139}\text{La}$ .

As shown in Fig. 6, the NMR signal (multiplied by a factor  $T$ ) is independent of  $T$  from 130 K down to 15 K. There is no loss of  $^{139}\text{La}$  NMR signal on cooling in the range 70–15 K, where the Cu NQR spectrum is wiped out. This contrasting behavior is not *a priori* unexpected, since  $^{139}\text{La}$  lies out of the  $\text{CuO}_2$  planes. Hence spin and charge fluctuations in these planes produce hyperfine-field and electric-field gradient fluctuations which are considerably reduced at the La site. Still, the absence of  $^{139}\text{La}$  NMR wipeout down to 15 K makes  $^{139}T_1$  measurements particularly interesting, since the whole sample is probed, including those parts where the Cu NQR signal has disappeared.

### D. $^{139}\text{La}$ NMR relaxation evidence for a distribution of spin fluctuation frequencies

A typical plot of the time dependence of the  $^{139}\text{La}$  longitudinal magnetization after a comb of saturation pulses at  $T = 15$  K is shown in Fig. 7. For a purely magnetic relaxation mechanism, the theoretical expression for the recovery of the magnetization, after fast irradiation of the central NMR transition of nuclear spins  $I = \frac{7}{2}$ , is<sup>48</sup>

$$\begin{aligned} [M_0 - M_z]/M_0 = & [8575 \exp(-28t/T_1) + 2475 \\ & \times \exp(-15t/T_1) + 819 \exp(-6t/T_1) \\ & + 143 \exp(-t/T_1)]/12012. \end{aligned}$$

However, the data points cannot be fitted to this expression (Fig. 7). This means that the recovery is modified by a distribution of  $T_1$  values or by nuclear transitions produced by electric-field gradient fluctuations. At low temperatures,  $T_1$  is entirely magnetic (see the large enhancement of  $T_1^{-1}$  due to the spin freezing below). So the deviation of the recovery from the ideal behavior is due to a distribution of  $T_1$  values in that case. Such a distribution was previously found to characterize the magnetic freezing in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $x > 0.02$ ) (Refs. 8 and 49) and in  $\text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.15}\text{CuO}_4$ .<sup>15</sup> Since the shape of the  $^{139}\text{La}$  recovery smoothly depends on  $T$  in the range 5–40 K, it is very reasonable to assume that there is a distribution of  $T_1$  values in all this  $T$  range.<sup>50</sup>

### E. Implications of $^{139}\text{La}$ $T_1$ results for the $^{63}\text{Cu}$ NQR/NMR wipeout effect

Given the distribution of  $T_1$  values, the data should be characterized by the width and the central value of this distribution. However, as is usually observed in such cases, the data may be reasonably well fitted with only two contributions, each of them following the theoretical expression for  $I = \frac{7}{2}$  (given above). Here we adopt this procedure for simplicity. From the data at 15 K, we extract two characteristic relaxation times for  $^{139}\text{La}$  nuclei, which have quite different values:  $T_1^A = 2200$  ms and  $T_1^B = 265$  ms. Since the best fit of the recovery plot according to the above equation yields two contributions with almost equal weights, the difference in  $T_1$  is a crude estimate of the width of the  $T_1$  distribution. Here, we see that the distribution is indeed sizable. With the help of Eq. (2), one readily finds that magnetic fluctuations responsible for a  $T_1$  of 265 ms at La sites would produce a  $T_1 \leq 15 \mu\text{s}$  at  $^{63}\text{Cu}$  sites and thus a  $T_2 \leq 2 \mu\text{s}$ .<sup>31</sup> The spin-echo signal from nuclei with such short relaxation times cannot be observed, while nuclei with relaxation times greater by an order of magnitude should be observable. This implies that the  $^{63}\text{Cu}$  NQR spectrum will be partially wiped out because some nuclei have relaxation times too fast to be observed, while the remaining nuclei are still detected. It could be remarked that the Cu NQR wipeout starts at least below 60 K, while  $^{139}T_1^{-1}$  is weakly  $T$  dependent down to 30–40 K (with no significant jump at the wipeout onset). However, there is no contradiction between these two observations. Indeed,  $T_1$  of  $^{139}\text{La}$  contains a background of quadrupolar relaxation which likely masks the onset for the enhancement of magnetic relaxation. Moreover, the quadrupolar relaxation channel which is active for  $^{139}\text{La}$  nuclei is not necessarily present at Cu sites.  $^{139}\text{La}$  and  $^{63}\text{Cu}$  nuclei certainly have a different ratio of magnetic to quadrupolar relaxation, and this ratio is extremely difficult to determine experimentally.<sup>51</sup> Actually,  $^{139}T_2$  shortens by a factor of 1.3 between 60 and 30 K.

Thus we conclude that *the slow and spatially distributed magnetic fluctuations are sufficient to explain the strong Cu*

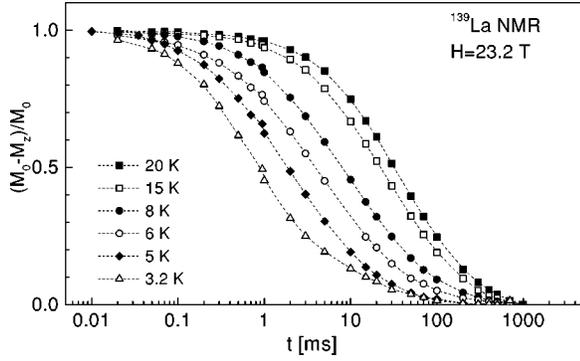


FIG. 8. Time dependence of the  $^{139}\text{La}$  NMR signal  $M_z$  after saturation of the central line in a field of 23.2 T, at different temperatures. The faster recovery of  $M_z$  at low  $T$  is due to the slowing down of magnetic fluctuations.

*wipeout effect at low temperatures in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ .* One cannot exclude that slow charge fluctuations are present, especially around the wipeout onset, but there is no evidence for this.

#### F. $^{139}\text{La}$ $T_1$ evidence for spin freezing

Figure 8 shows the recovery plots of the nuclear magnetization after a sequence of saturation pulses, at selected temperatures, in a field of 23.2 T. The overall trend clearly shows that the recovery becomes faster on cooling, i.e.,  $T_1$  shortens. This behavior holds down to about 4 K. Below 4 K, a tail appears at long times, becoming as important as the fast component at  $T=1.65$  K, our lowest  $T$  data point (not shown). This long component in the relaxation does not seem to be linked to superconductivity since it is also observed at 15 T in the same  $T$  range, and  $T_c$  is higher at this field (at 15 T, a clear change in the frequency tuning of the NMR probe at 6.5 K signals the irreversibility line). More

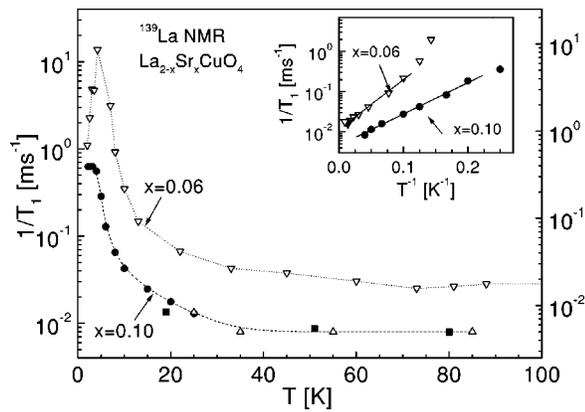


FIG. 9.  $T$  dependence of the  $^{139}\text{La}$  nuclear-spin-lattice relaxation rate  $1/T_1$ <sup>51</sup> for  $x=0.10$  (this study) and  $x=0.06$  (Ref. 8). The left scale is associated with the NMR data for  $x=0.10$ : the filled circles are data at 23.2 T, and the filled squares are data at 9.4 T. The right vertical scale is associated with NQR data (up triangles for  $x=0.10$  and down triangles for  $x=0.06$ ). Dotted and dashed lines are guides to the eye. Inset: same data vs  $1/T$ ; continuous lines are fits explained in the text.

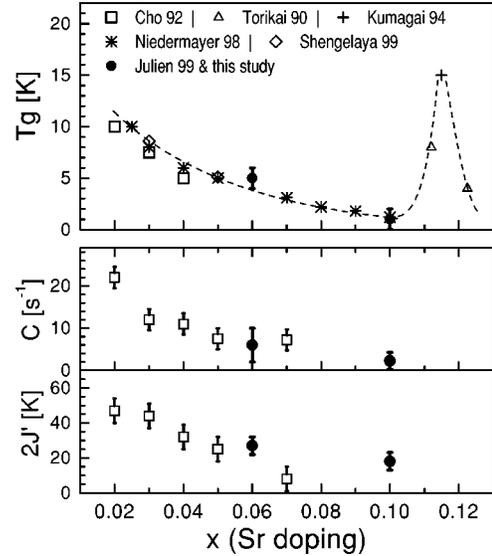


FIG. 10. Top panel: Magnetic transition temperature  $T_g$  in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $0.02 < x < 0.125$ ) from  $\mu\text{SR}$  (Refs. 7, 25, 26, and 55) and NMR/NQR (Refs. 8 and 49).  $T_g$  data from magnetization measurements can be found in Refs. 56–58. Middle and bottom panels: parameters  $C$  and  $J'$ , obtained from a fit explained in the text, and compared to the data of Cho *et al.* (squares) (Ref. 49).

data are necessary in order to understand if this feature, which might be caused by a distribution of freezing temperatures, and is intrinsic or related to the sample purity. Because of this uncertainty and of the limited experimental  $T$  range, it is not possible to determine a precise freezing temperature  $T_g$  in this sample. Nevertheless, it is clear that a slowing down phenomenon starts below  $\sim 30$ – $40$  K, as is shown in Fig. 9 by the  $T$  dependence of  $T_1$ .<sup>52</sup> An important feature of the freezing process is that it involves the vast majority of sites, if not all, in the sample: there is no long tail in the recoveries (except the one very close to  $T_g$ ) which could be attributed to nonfreezing areas. In contrast, the recovery curves in Fig. 8 shift continuously toward short times on cooling, without a strong modification of their shape. Note that this conclusion is not affected by a possible loss of  $^{139}\text{La}$  nuclei below 15 K, since this would precisely originate from freezing zones, while nonfreezing ones have longer  $T_1$ .

#### G. Remark on high magnetic fields

It is important to realize that spin freezing is not induced by the magnetic field. In fact, (i) a similar enhancement of  $1/T_1$  was found in fields of 23.2, 15, and 9 T (not shown). (ii)  $\mu\text{SR}$  measurements (in zero field) already reported a bulk spin-freezing at  $T_g \approx 1.2$  K for  $x=0.10$  (Ref. 7) (also see (Ref. 22)). Thus magnetic fields as high as 23 T do not seem to modify spin dynamics in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  for temperatures in the range 5–40 K (an influence of the field on the freezing temperature  $T_g$  cannot be excluded).

It is interesting to consider these findings in comparison with the insulating behavior of the  $ab$ -plane resistivity under strong magnetic fields. Boeinger *et al.*<sup>53</sup> found  $\rho_{ab} \propto \log 1/T$ , which they considered as an indication of a non-

metallic ground state in zero field when superconductivity is destroyed, while Malinowski *et al.*<sup>54</sup> suggested that it is the field itself which induces localization of the charges. Since spin dynamics are likely to be affected by charge localization, our result that spin dynamics in the freezing regime do not change appreciably with the field would rather tend to support the former view.

### H. Comparison to other works

The top panel of Fig. 10 shows a selection of spin-freezing temperatures ( $T_g$ ) determined from  $\mu$ Sr and NMR/NQR experiments in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (see Refs. 7, 25, 26, and 55;  $T_g$  data from magnetization measurements can also be found in Refs. 56–58). Given the uncertainties discussed above, we plotted  $T_g = 1 \pm 1$  K for our experiment in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ , which is consistent with previous works, in particular  $T_g \approx 1.2$  K from Niedermayer *et al.*<sup>7</sup>

Cho *et al.* remarked that  $T_1$  data for  $T > 2T_g$  can be fitted to the expression<sup>49</sup>

$$\frac{1}{T_1} \propto c(x) \exp\left(\frac{2J'(x)}{T}\right), \quad (3)$$

where  $2J'$  can be considered as a coupling constant. The values of the parameters  $c(x)$  and  $J'(x)$ , obtained by fitting our data for  $x=0.10$  and  $x=0.06$  (see the inset to Fig. 9) to Eq. (3) are in good agreement with those obtained by Cho *et al.* (Fig. 10).<sup>59</sup> It is remarkable that  $J'(x)$  is reduced by less than a factor 2.5 between  $x=0.02$ , the border of the long-range antiferromagnetic (AF) phase, and  $x=0.10$ , while  $c(x)$  is reduced by about a factor of 10. Inspired by the cluster spin-glass idea, Cho *et al.* related the behavior of  $c(x)$  to the size of AF clusters that shrink with doping, and  $2J'$  to the energy barrier for the reorientation of the staggered moments.<sup>49</sup> In order to gain more physical insight into the parameters  $J'$  and  $c$ , and into the details of spin freezing, it would be interesting to have theoretical predictions from different models (various forms of cluster spin glass, stripes, impurities, etc.).

## VI. SUMMARY OF MAIN RESULTS AND DISCUSSION

### A. Coexistence of magnetic order with superconductivity

Although we could study only the freezing process and not the frozen state ( $T < T_g$ ), our  $^{139}\text{La}$   $T_1$  measurements confirm previous microscopic evidence of frozen magnetic moments below  $\sim 1$  K in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ .<sup>7</sup> In our sample, which is a bulk superconductor according to Huh *et al.*,<sup>39</sup> we observe that the magnetic freezing is visible at all  $^{139}\text{La}$  sites in the sample. This is again in agreement with Niedermayer *et al.* finding all muons probing an internal field at low  $T$ .<sup>7</sup> Thus one must conclude that superconductivity coexists with frozen magnetic moments<sup>60</sup> (which we know to be locally staggered<sup>8</sup>), and this coexistence has to occur at a microscopic scale. It is noted that the nature of the magnetic freezing found in the superconducting phase ( $x \geq 0.05$ ) appears to be of the same kind of the cluster spin-glass freezing reported earlier<sup>49</sup> in nonsuperconducting materials.

In  $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ , equal magnetic and superconducting transition temperatures were reported, based on neutron scattering and  $^{139}\text{La}$  NMR lineshape measurements.<sup>45–47</sup> However, we have shown above that the broadening of the  $^{139}\text{La}$  NMR line occurs well above the freezing temperature  $T_g$  (as determined by  $^{139}\text{La}$  NMR  $T_1$  or  $\mu$ SR). We suspect that the real  $T_g$  in  $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$  is much lower (see Refs. 25 and 26), and thus that the onset of NMR line broadening coincides with  $T_c$  only by chance.

The coexistence found here resembles previous works in, e.g., Nd-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  where bulk superconductivity is claimed by several groups (see Refs. 41, 42, 61, and 62 for an opposite point of view). Nevertheless, more experimental work remains to be performed in order to fully characterize the superconducting properties of these materials with spin-glass-like freezing. From a theoretical viewpoint, it is clear that the cluster spin-glass freezing deserves intensive consideration: the existence of superconductivity in such a context and the relation to stripe physics need being addressed more accurately (see Ref. 63 for recent views).

Finally, it is important to note that the coexistence of frozen moments with superconductivity does not mean that both phenomena are somehow related or even cooperative. There is ample evidence that they compete (see Ref. 64 for a recent perspective and references). In fact, the internal static field, existing at  $T \ll T_g$ , is very small in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ , certainly less than 10% of the value in  $\text{La}_2\text{CuO}_4$ .<sup>7</sup>

### B. What is the evidence for stripes in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ?

Looking at the whole body of experimental data in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $0.06 \leq x \leq 0.12$  (see Refs. 7–10, 12, 23, 24, 53, and 64 and references therein), it is now quite clear that this part of the phase diagram shows (besides superconductivity) three mutually related phenomena at low temperature ( $T \approx 40$ – $100$  K, depending on doping): charge localization tendencies, glassy spin freezing and Cu NMR/NQR wipeout. It is very tempting to attribute these three features to the presence of charge stripes. There are two principal arguments in support to this view.

(1) Magnetic order issued from glassy spin freezing and Cu NQR wipeout are observed in a very similar way in rare earth-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , where they are understood as a consequence of stripe order.<sup>12–16,65,61,62</sup> Transport properties of these materials also bear strong similarities to those of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>66,67,64</sup> Of course, another important piece of the argument is the identical wave vector for magnetic scattering in Nd-free and Nd-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>20,68</sup>

(2) It is difficult to imagine how a magnetic state with local AF order could exist without charge segregation at high hole-doping level. Charge stripes represent an ideal form of such segregation. If doped holes were randomly distributed the mean distance between them would be about three lattice spacings in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ . On the other hand, the mean distance between charge stripes is of five lattice spacings at  $x=0.10$  (assuming one charge per two sites along the stripe). The charge stripe picture clearly generates much larger hole-poor regions (magnetic domains), and is naturally much more favorable to spin order: charge order eliminates spin

frustration, which is otherwise large if holes are uniformly distributed. This interpretation of the data in the “cluster spin-glass” phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  was repeatedly put forward by Emery and Kivelson.<sup>69</sup>

### C. Is the wipeout fraction a measure of stripe order?

We have found that the Cu NQR wipeout effect can be explained by the glassy nature of the magnetic freezing in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$ : slow spin dynamics shorten the nuclear relaxation times  $T_1$  and  $T_2$  of  $^{63,65}\text{Cu}$ , and these nuclei become unobservable below a threshold value of  $T_2$ . Because the dynamics is spatially inhomogeneous and the freezing occurs on a wide  $T$  interval, the NMR/NQR signal disappears only gradually on cooling. This interpretation of the wipeout effect was also proposed by Curro *et al.*,<sup>15</sup> who deduced the distribution of  $T_1$  from its  $T$  dependence, while we infer it from the recovery plots. These authors also argued that the crossover from Gaussian to Lorentzian  $T_2$  is explained by the same arguments. The magnetic origin of the wipeout was taken by Curro *et al.* as a strong argument against the wipeout fraction being a measure of the “stripe order parameter.” Our point of view is somewhat less radical, although we come to the same conclusion, as explained below.

The fact that the wipeout can be explained by glassy spin freezing only indicates that the relationship with stripe order, if any, is not straightforward. Actually, this was not excluded by the authors of Refs. 12 and 13. Their data in  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_{0.09\leq x\leq 0.15}\text{CuO}_4$  suggest a sharp wipeout onset, coinciding with the charge order temperature detected by neutron scattering.<sup>13</sup> In this sense, the wipeout effect must be somehow related to stripe order. A reasonable scenario is that charge motion generates spin flips, so it prevents magnetic ordering. Magnetic moments start to freeze only when charge-stripe order, i.e., when magnetic stripes become well defined. As a matter of fact, charge order is always followed by magnetic order. The slowing down of spin fluctuations on approaching the magnetic transition is in turn responsible for the wipeout effect. However, the argument holds only because we know that there are stripes in these materials. One should keep in mind that any other situation with slow and inhomogeneous spin dynamics (such as impurity doping) could produce a similar wipeout effect.

On the other hand, it is the very nature of the wipeout effect that makes the identification with a stripe *order parameter* questionable. The wipeout fraction measures a kind of volume fraction in which the spin dynamics is slowed down below some threshold value that clearly depends on experimental conditions. This is typically an extensive quantity, while an order parameter is usually associated with an intensive quantity like the amplitude of a field or a distortion. Moreover, it is even not *a priori* obvious to consider the amplitude of charge order peaks in neutron or x-ray scattering as an order parameter. If the order is not homogeneous, the peak intensity may also reflect the volume fraction of well-ordered stripe segments in the crystal. Finally, we note two contradictions which need to be clarified. First, the wi-

peout onset in  $\text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.15}\text{CuO}_4$  occurred at a much higher temperature in the experiments of Curro *et al.* [ $\approx 100$ – $150$  K (Ref. 15)] than that of Singer *et al.* [ $\approx 50$ – $70$  K (Ref. 13)]. Second, the onset of Cu NQR wipeout seems to occur prior to the neutron charge ordering temperature in  $\text{La}_{1.50}\text{Nd}_{0.4}\text{Sr}_{0.10}\text{CuO}_4$ .<sup>13,64</sup>

## VII. CONCLUSION

While the Cu NQR wipeout effect is certainly related to stripe order in  $\text{La}_{2-x-y}(\text{Nd},\text{Eu})_y\text{Sr}_x\text{CuO}_4$ ,  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  and  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , it would not be reasonable to use it as a criterion for the presence of charge stripes in other materials and it is *not* possible to identify the wipeout fraction as a stripe *order parameter*. In superconducting  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , direct evidence for charge stripes is still lacking but the accumulation of indirect hints is rather overwhelming. Since it is extremely difficult to probe the charges experimentally (fluctuations and disorder can readily render diffraction methods inoperative, and local probe such as Cu or La NQR do not show any spectacular anomaly at the wipeout onset), it will be important as a next step to perform detailed measurements of spin dynamics and of the wipeout fraction in the vicinity of the stripe ordering temperature, in samples where stripe order is established by diffraction techniques. A better understanding of the impact of stripe order on spin dynamics will be particularly helpful in materials with no direct signs of stripes.

*Note added.* After completion of this manuscript we became aware of a NQR work in  $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ .<sup>70</sup> A  $^{139}\text{La}$  wipeout effect is found, but starting at a much lower temperature than the Cu wipeout. The authors argued that  $^{139}\text{La}$   $T_1$  is entirely due to magnetic fluctuations, and they found in agreement with Ref. 15 and our work, that slow and distributed spin fluctuations explain the Cu NQR wipeout effect.

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- <sup>60</sup>Each  $^{139}\text{La}$  nucleus is coupled to several Cu sites. If these Cu sites have different fluctuating frequencies,  $T_1$  is determined by the fastest relaxation channel. This means that  $T_1$  may only probe the slowest magnetic moment to which it is coupled. Thus we cannot exclude, in principle, that some individual  $\text{Cu}^{2+}$  moments do not freeze. The data show, however, that “nonfreezing regions” of typical sizes larger than a few lattice spacings are not present in the sample. Furthermore, a situation with very different fluctuating frequencies on neighboring sites is quite unlikely.
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