

Spin dynamics and ordering of a cuprate stripe antiferromagnet

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In $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ the ^{139}La and ^{63}Cu nuclear quadrupole resonance relaxation rates and signal wipe-out upon lowering temperature are shown to be due to purely magnetic fluctuations. They follow the same renormalized classical behavior as seen in neutron data, when the electronic spins order in stripes, with a small spread in spin stiffness (15% spread in activation energy). The La signal, which reappears at low temperatures, is magnetically broadened and experiences additional wipe out due to slowing down of the Nd fluctuations.

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Strongly correlated electron systems such as layered cuprates exhibit very unusual properties. One of the most interesting among them is the coexistence of superconductivity with local antiferromagnetism (AF)—a fingerprint of the topological effects of doping of AF insulators by holes. The charges segregate into a periodical array of stripes separating antiphase antiferromagnetic domains. Experimental evidence for stripe correlations has been provided by neutron studies in Nd-doped $\text{La}_{1.875}\text{Sr}_{0.125}\text{CuO}_4$ and in other cuprates and nickelates.^{1,2} The spatial organization of the stripe structures is a subject of much debate.^{3–8} Stripe formation is characterized by the temperatures of charge (T_{charge}) and spin T_{spin} ordering with $T_{\text{charge}} > T_{\text{spin}}$. Since these different types of order coexist on the microscopic level, local methods of analysis, like NMR or nuclear quadrupole resonance (NQR), are well suited to see their interrelation. One striking feature in the NMR data is the wipe-out effect. In Cu-NQR experiments on a number of Sr doped La_2CuO_4 samples, Hunt *et al.*⁶ showed a correlation between the amount of the intensity loss and the development of charge order of the stripe phase. Curro *et al.*⁷ found strong Cu wipe-out effect in their NMR experiments on $\text{La}_{2-y-x}\text{Eu}_y\text{Sr}_x\text{CuO}_4$ and showed that this effect could be accounted for by a wide (100%) distribution in the energy of the thermally activated correlation times that determine the relaxation processes—so-called glassy behavior.

In this communication the role of slow magnetic fluctuations is elucidated. We show that the variation of the line shape as well as wipe-out and relaxation effects probe the growing spin order in the stripe phase. Our investigation takes profit of the NQR frequency range of ^{139}La and ^{63}Cu , and especially of the low frequencies and relatively small line widths of La NQR in $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$. In this compound both Cu and La exhibit strong wipe-out effects. Because La (contrary to Cu) nuclei are relatively weakly coupled to the electronic spins in the CuO_2 planes, La NQR signals can be followed down to the spin-ordering temperature, as seen by μSR . Using the spin correlation times extracted from the activated La spin-lattice relaxation rates we are able to predict precisely these wipe-out features by introducing a spread of only 15% in the activation energy. Within experimental error this energy agrees with the value found

from neutron data, where the relation with stripe ordering was well established,² and is explained in the renormalized classical model. An additional finding is the reappearance of a magnetically broadened NQR signal at low temperatures. For the 6 MHz La transition the signal intensity is maximal around 4 K, where still about half of the La nuclei are missing.

Experimentally we measure the T dependence of the signal intensities \tilde{I} and of the relaxation rates of the three ^{139}La NQR transitions ($I=7/2$) at 6, 12, and 18 MHz for $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ and those of $^{63,65}\text{Cu}$ ($I=3/2$) around 36 MHz.⁹ The question whether spin or charge fluctuations are relevant is answered by comparison of the rates of the ^{63}Cu and ^{65}Cu and precisely monitoring the magnetization recovery curves after spin reversal for the various La transitions. All relaxation rates are purely due to spin fluctuations. Knowing that the fluctuations are magnetic, we extend the approach of the Los Alamos group^{7,8} to obtain the proper analytic description of the wipe-out effect. With a simple signal visibility criterium and the known values of the hyperfine couplings, from the wipe-out curves correlation times for the spin dynamics are calculated. At the end we show that the La linewidth increase below 20 K is due to the internal hyperfine field induced by the ordered Cu moments and that Nd fluctuations are responsible for the missing La NQR signal intensity at the lowest temperatures.

Let us now discuss our findings in more detail. NQR measurements were performed on a powder sample.¹⁰ The preparation is described in Ref. 11. Susceptibility χ measurements at 0.001 T show a superconducting transition temperature of 5 K. The intensity \tilde{I} multiplied by T and corrected for T_2 is shown in Fig. 1. Because the nuclear magnetization follows a Curie law, $T\tilde{I}$ is expected to be T independent. This relation is not obeyed, see Fig. 1. Instead, $\tilde{I}T$ strongly decreases with decreasing T , the so-called wipe out being different for Cu and La. In Fig. 1 arrows indicate the charge ($T_c \sim 65$ K) and spin-order ($T_s \sim 54$ K) temperatures as seen by neutrons,¹ and the magnetic transition seen by μSR ($T_m \sim 31$ K).⁵ The low-temperature orthorhombic (LTO) to low-temperature tetragonal (LTT) transition is around 68 K.

The spin-lattice (T_1^{-1}) and spin-spin (T_2^{-1}) relaxation rates for the several La-quadrupolar transitions, Fig. 2, peak

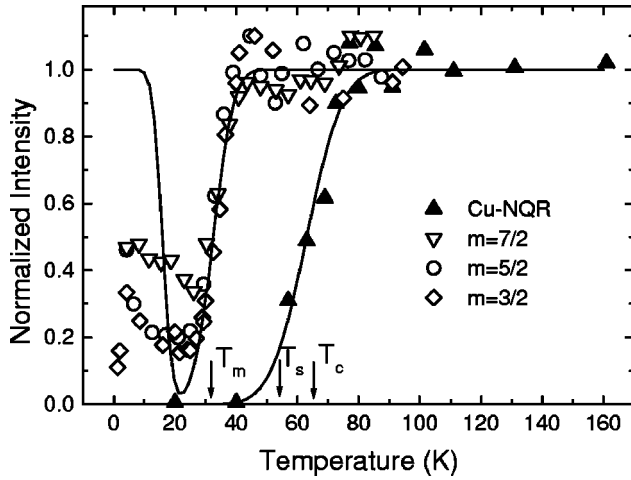


FIG. 1. Wipe out in La and Cu NQR. For both Cu isotopes wipe out starts around 70 K, while for the three satellites of ^{139}La , this temperature is around 40 K. Drawn lines are predictions from the model discussed in the text.

around 20 K, where also the wipe out has its maximum. The labels $m=7/2$, $5/2$, and $3/2$ refer, respectively, to the $(\pm 7/2, \pm 5/2)$, $(\pm 5/2, \pm 3/2)$, and $(\pm 3/2, \pm 1/2)$ transitions.¹² Fits are made with stretched exponentials ($[1-M(t)]$ -recovery is $\propto \exp[-(t/T_1)^\alpha]$, indicating the presence of a distribution in rates; the more α deviates from 1 the larger the influence of the distribution is. Here α decreases almost linearly from 1 at 300 K to 0.6 at 20 K. Down to 30 K the T dependence can be described by $T_1^{-1} = W^2 \tau / (1 + \omega^2 \tau^2)$ (characteristic for exponential time correlation between fluctuating electronic spins) with $\tau = \tau_\infty \exp(E/k_B T)$,¹³ W a matrix element and E an activation energy, see drawn line in Fig. 2. The T dependence of T_2 , see Fig. 2, is determined by the same activation law. From the fit we obtain $E = 143 \pm 5$ K. With the known hyperfine coupling,¹⁴ we estimate τ_∞ as $4 \cdot 10^{-12}$ s. The same value is found from the maximum in T_1^{-1} .

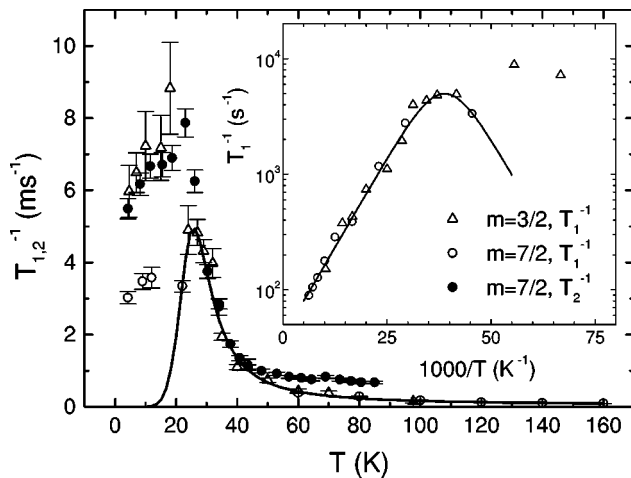


FIG. 2. ^{139}La T_1^{-1} and T_2^{-1} as function of T . The solid line is a fit based on activated behavior with $E_0 = 143$ K. The deviations below 20 K are due to the magnetic ordering.

To see whether the relaxation processes are determined by magnetic or electric fluctuations, we compared the rates for ^{63}Cu and ^{65}Cu . The Cu rates at 71 K were 8.1 (^{63}Cu) and 9.8 ms^{-1} (^{65}Cu) and at 130 K, respectively, 7.3 and 10.1 ms^{-1} . If $\omega \tau \ll 1$, T_1^{-1} is proportional to $W^2 \tau$. For the magnetic case the ratio of the ^{63}Cu and ^{65}Cu transition rates is proportional to $(\gamma_{63}/\gamma_{65})^2 = 0.87$, while in case of electric transitions it is the ratio between the quadrupolar moments squared, which equals 1.14. The found ratio's show Cu relaxation to be magnetic. For La only the rates for the various quadrupolar transitions are available. Here we make use of the fact that the fundamental transition probability, that appears in the exponents of the relaxation expression is weighted by well defined factors,¹⁵ that are different for magnetic or electric processes. At 130, 60, 33, 28, and 4.2 K, the magnetization recovery curves after application of a π pulse follow stretched exponentials with rates that were a factor 1.8 ± 0.15 faster for $m=5/2$ than for $m=7/2$. This value agrees with the magnetic ratio of 1.9.¹⁶

How to explain the pronounced wipe-out features? Hunt *et al.* suggested that the intensity loss might be directly or indirectly related to the growth of the stripe order parameter with decreasing temperature.¹⁷ Let us restrict ourselves to the case of direct relaxation and to simplify the argument, suppose that the fluctuating stripe order leads to random jumps between the two NQR frequencies which correspond to the extremal values of charge distribution and differ by a value $\delta\omega$. The signal decay for $\delta\omega \tau_{\text{ch}} \ll 1$ can be obtained in a motional narrowing approach¹³ and is given by the $\exp\{-t[1/T_2 + (\delta\omega \tau_{\text{ch}})^2 / (8\tau_{\text{ch}})]\}$. The resulting decay is not only determined by the dephasing due to magnetic ($1/T_2$), but also due to electric fluctuations (lifetime τ_{ch}). Since the experimental relaxation rates turn out to be governed by magnetic fluctuations, charge fluctuations can at most weakly contribute to the wipe-out phenomena.¹⁸

More generally, wipe-out effects have been shown to be linked to charge/spin fluctuations having a distribution $P(E)$ in activation energies E and hence in correlation times.^{19,7} In case of a Gaussian distribution of E , the normalized intensity $\tilde{I}(t)$ is given by

$$\tilde{I}(t) = (1/\sqrt{2\pi}\Delta) \int_0^\infty \exp[-(E-E_0)^2/2\Delta^2] \times \exp[-t/T_2(E)] dE,$$

with E_0 the mean activation energy, and Δ the width of the distribution and $\tilde{I}(0) = 1$. In the echo pulse sequences $\pi/2-t_r-\pi-t_r$, the delay time $2t_r$ allows a registration in the echo of only those nuclei, that do not relax too fast. Let us assume that we are only seeing those nuclei of which the signal has decayed by a factor of f or less at time $2t_r$, i.e., for which $1/T_{2R} = \beta/T_1 = (\Omega^2 \tau) / (1 + \omega^2 \tau^2) \leq A$.²⁰ For magnetic fluctuating fields $\Omega^2 = \beta \gamma^2 h_0^2$, $A = (\ln f) / 2t_r$, h_0 denotes the hyperfine field probed by the nuclei and $\beta = (2+r)/3$,²¹ with the anisotropy factor $r = 3.6$ for Cu and $\beta = 6$ for La (as deduced from our own relaxation data). The above mentioned inequality determined the boundary values of τ , which follow from $A \omega^2 \tau^2 - \Omega^2 \tau + A = 0$ and are given by

$\tau_{1,2} = (\Omega^2 \pm \sqrt{\Omega^4 - 4A^2\omega^2})/2A\omega^2$. This introduces cut-off's $E_i = k_B T \ln(\tau_i/\tau_\infty)$ in the expression for $\tilde{I}(t)$, reflecting that part of the nuclei do not contribute to the signal. As a result the extrapolation of $\tilde{I}(2t_r)$ to $t=0$ gives

$$\tilde{I}_{2t_r}(0) \propto \int_0^{E_2} e^{-(E-E_0)^2/2\Delta^2} dE + \int_{E_1}^{\infty} e^{-(E-E_0)^2/2\Delta^2} dE. \quad (1)$$

$\tilde{I}_{2t_r}(0)$ is proportional to the number of nuclei influenced by the magnetic fluctuations with lifetimes outside the interval between τ_1 and τ_2 . There appear two bands in the solution, which contribute to the signal: a band of high-frequency fluctuations (smaller activation energies $E < E_2$) and a band of low-frequency fluctuations (larger activation energies $E > E_1$). The values of E_1 and E_2 are linear functions of T and the gap $E_1 - E_2 = k_B T \ln(\tau_1/\tau_2)$ between them is the NMR wipe-out gap. The condition for the gap to exist is very simple $\Omega^2 > 2A\omega$. The presence of two bands, see Eq. (1), gives rise to the reentrant behavior of the echo-amplitude with lowering T .

In case of La NQR, Ω_{La} is rather small, and the condition for the wipe-out gap is realized for τ lying in the narrow interval around $\tau = \Omega^2/2A\omega^2$. Using $A_{\text{La}} \sim 10^5 \text{ s}^{-1}$ ($f \sim e^5$ and $t_r = 30 \mu\text{s}$), the ^{139}La hyperfine coupling constant ($1.7 \text{ kOe}/\mu_B$)¹⁴ and $\Omega_{\text{La}} \sim 6 \cdot 10^6 \text{ s}^{-1}$ we obtain that for this interval the typical fluctuation times are $\tau \sim 10^{-8} \text{ s}$. For the Cu nuclei $\Omega^2/A\omega^2 \leq \tau \leq A/\Omega^2$. With $A_{\text{Cu}} \sim A_{\text{La}} \sim 10^5 \text{ s}^{-1}$, the ^{63}Cu hyperfine coupling constant of $139 \text{ kOe}/\mu_B$, and $\Omega_{\text{Cu}} \sim 6 \cdot 10^8 \text{ s}^{-1}$, it follows that the wipe-out at 75 K is due to the fluctuations with $\tau \sim 10^{-11} - 10^{-12} \text{ s}$. Neglecting effects of the magnetic ordering of Cu (and the Nd) moments, the reappearance of the Cu NQR signal will take place for extremely slow fluctuations with $\tau \sim 10^{-6} \text{ s}$, realized only at very low temperatures.

The drawn lines in Fig. 1 are fits to the wipe-out behavior with the numerical constants calculated above. The free parameters are in principle E_0 , Δ , and $\ln(\tau_i/\tau_\infty)$, with $\tau_{1,2}$ being fixed by A and Ω . If for E_0 the same value is used as for the relaxation data, i.e., $E_0 = 143 \pm 5 \text{ K}$, the fit to the Cu and La data gives mutually consistent values for the other free parameters: $\Delta = 21 \pm 3 \text{ K}$ and τ_∞ equals the value found from the relaxation data. Note that for the low frequency La transitions the wipe-out is more pronounced, since the wipe-out gap is $\propto 1/\omega^2$.

An activated T dependence of τ can have many causes. However, a most natural interpretation is in terms of the behavior of the relaxation time of a classical quasi-two-dimensional (2D) Heisenberg antiferromagnet which is on its way to its 3D phase transition. The relaxation time is set by the magnetic correlation length ξ ,²² and the latter behaves like $\xi(T) = e^{T^*/T}/(2T^* + T)$ where $T^* = 2\pi\rho_s$ in terms of the spin stiffness ρ_s . According to our relaxation and wipe-out data $T^* = 143 \pm 5 \text{ K}$ which is consistent with the $T^* = 200 \pm 50 \text{ K}$ as deduced by Tranquada *et al.*² from the T dependence of ξ as measured by neutron scattering. This spin stiffness associated with the stripe antiferromagnet is an order of magnitude smaller than the one of the pure antifer-

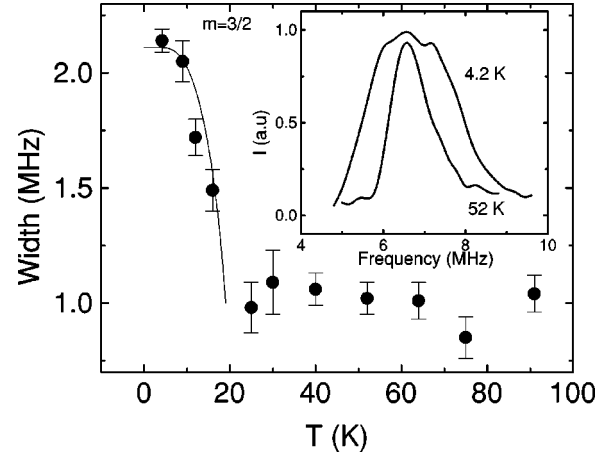


FIG. 3. T dependence of the ^{139}La linewidth for $m=3/2$. Magnetic ordering sets in below 20 K (drawn line: mean-field fit). The inset shows the changes in the line profile.

romagnet of half filling. If the spin system would be classical the implication would be that the exchange interactions mediated by the charge stripes would be smaller by two orders of magnitude as compared to the exchange interaction inside the magnetic domains. This is inconsistent with the persistence of antiphase correlations up to rather high energies as seen by inelastic neutrons scattering. Moreover, there is no doubt that the spin system is highly quantum-mechanical at short length scales and the stripe antiferromagnet should exhibit *renormalized* classical behavior.²³ This implies that the spin system should be in the proximity of a quantum-phase transition to a disordered state and it is well understood that the renormalized stiffness diminishes when this transition is approached, while the spin velocity is barely changing. Hence, the small spin-stiffness of the stripe antiferromagnet signals that this system is much closer to the quantum phase transition than the half-filled antiferromagnet, in agreement with theoretical expectations.^{24,25}

To evaluate the role of the Nd ion on the correlation times, we also determined the relaxation rates in $\text{La}_{1.71}\text{Eu}_{0.17}\text{Sr}_{0.12}\text{CuO}_4$.^{6,8,26} The ^{139}La relaxation rates were about a factor 10 lower than in the 0.4Nd compound. With the hyperfine coefficients used in 0.4Nd, the correlation times found for the fluctuating fields derived from $T_1^{-1}(^{63}\text{Cu})$ and $T_1^{-1}(^{139}\text{La})$ above 20 K in the Eu sample were the same, but compared to the Nd sample, the values of τ at comparable temperatures are different. It likely reflects the different pinning strength of stripe structure with the inhomogeneities of LTT phase induced by the Nd and Eu ion, whereas the equal hyperfine constants show that above 20 K Nd does not influence the La nuclear relaxation rates directly.

To determine whether spins or charges are responsible for the final La line shapes, we have followed the line profiles for various satellites (due to its small splitting, $m=3/2$ is the most sensitive) as function of T , see Fig. 3. Above the wipe-out regime the La line widths scale with their splitting, which show them to be electric. Below 20 K the linewidths increase due to the presence of an internal magnetic field, see Fig. 3. The drawn line represents the mean-field staggered magneti-

zation for $S=1/2$. The saturated value of the additional width (full width at half intensity), obtained by taking the square root of the difference in second moments of the broadened and unbroadened line, amounts to 2.0 MHz, close to the splitting seen in undoped La_2CuO_4 , where the $m=3/2$ splitting is 2.5 MHz.²⁷ In the undoped compound (with an ordered moment in the Neel state of $\sim 0.55\mu_B$) the splitting can be reproduced by a field of 0.11 T perpendicular to the electric field gradient (with anisotropy parameter $\eta=0.02$ and in plane field angle $\phi=0$). Here the saturated splitting seen for the $m=3/2$ line can be simulated by an external field of 0.08 T, again applied perpendicular to the electric field gradient ($\eta=0.13$ is fixed by the line positions above the magnetic ordering and $\phi=\pi/4$). As (see below) Nd moments are not yet involved, we estimate the Cu ordered moment in 0.4Nd to be $\sim 0.4\mu_B$. The missing spectral weight of about 50% at 4.2 K for $m=3/2(\text{La})$ might be explained by an internal field of the same order as the quadrupolar splitting of 6 MHz felt by the unseen La sites. Such a scenario agrees with the μSR finding⁵ that most or all μSR sites are magnetic. However, 0.1 T (2 MHz) at 4.2 K is about the maximum field at the La sites (even with $\text{Nd}^{2,4,10}$) one might expect. The T dependence of $\tilde{I}(m=3/2)$ below 4.2 K shows that we deal with additional wipe out caused by slow

Nd-spin fluctuations. This extra channel in $T_2^{-1}(\text{La})$ becomes important close to the ordering temperature of 1 K of the Nd moments¹⁰ and partially destroys the recovery of echo-signal predicted by Eq. (1).

In summary, the wipe-out and relaxation features of Cu and La in the temperature regime above the spin-ordering transition find a natural explanation in terms of the well understood fluctuations of a quantum antiferromagnet which is approaching its ordered state. For the 0.4Nd compound this “ordered” state is not straightforward as wipe out persists down to 1 K for the majority of La spins. Here La wipe out proceeds in two stages, of which the first is due to slowing down of Cu spins and the second below 4 K is dominated by fluctuations of Nd magnetic moments. From the La NQR lineshape we estimate an ordered Cu moment of $0.4\mu_B$ in the stripe phase.

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¹⁸The decay by τ_{ch} is the strongest for $\delta\omega\tau_{\text{ch}}\sim 1$, when the echo signal is already suppressed by magnetic fluctuations.

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