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Probing Spatial Correlations in the Inhomogeneous Glassy State of the Cuprates by Cu NMR

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We discuss the crossover of the form of the Cu NMR spin echo decay at the onset of Cu wipeout in lanthanum cuprates. Experimentally, the echo decay undergoes a crossover from Gaussian to exponential form below the temperature where the Cu NMR intensity drops. The wipeout and the change in behavior both arise because the nuclei experience spatially inhomogeneous spin fluctuations at low temperatures. We argue that regions where the spin fluctuations remain fast are localized on length scales of order 1-2 lattice spacings. The inhomogeneity is characterized by the local activation energy $E_a(\mathbf{r})$; we estimate the functional form of $E_a(\mathbf{r})$ for points where $E_a(\mathbf{r}) \sim 0$.

The lanthanum cuprate high temperature superconductors have received a considerable amount of attention in recent years because elastic neutron scattering experiments in rare-earth co-doped $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ and $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ have revealed ordering of doped holes into charged stripes that constitute anti-phase domain walls producing incommensurate antiferromagnetic (AF) order in the intervening undoped domains [1]. Since the initial discovery, several studies have revealed that the order is more complex than initially thought; instead of exhibiting a well-defined thermodynamic transition temperature the spin system appears to exhibit glassy behavior. [2,3] Nuclear Magnetic Resonance (NMR) and Nuclear Quadrupolar Resonance (NQR) studies have shown that this glassy behavior is accompanied by inhomogeneous slowing of the local spin fluctuations, and provide a direct measure of the distribution of activation energies giving rise to the inhomogeneous distribution. [4] This distribution can be characterized by measuring the temperature dependence of the La T_1 ; surprisingly this distribution is remarkably independent of doping (at least up to optimally doped), local structure, and doping location in the unit cell. Recent theoretical work suggests that the ubiquity of this glassy behavior may be intrinsic to doped hole systems. [5] The wipeout phenomenon of the Cu NMR spin echo intensity also arises from the development of these inhomogeneous spin fluctuations, with a broad distribution of fluctuation times. In fact, the temperature dependence of the Cu wipeout can be entirely explained by the distribution of activation energies measured by the La T_1 .

However, measurements of the La and Cu T_1 or the Cu wipeout reveal information only about the local spin fluctuations at the nucleus under observation. This allows one to determine the distribution $\mathcal{P}(E_a)$ of activation energies E_a characterizing the glassy behavior. The echo decay of the Cu, on the other hand, reveals information about not only the particular Cu nucleus under observation, but its surrounding neighbors as well. This serendipitous fact allows one to make conclusions about $E_a(\mathbf{r})$, the activation energy at position \mathbf{r} . In fact, the form of the echo decay of the Cu provides two important pieces of information: (i) the sites \mathbf{r}_i in the crystal such

$E_a(\mathbf{r}_i) \sim 0$ are separated by a distance of at least several unit cell lengths, a , and (ii) the effective radius of the minima around \mathbf{r}_i is approximately $1-2a$.

In general the decay of the spin echo intensity (the echo decay) of a nucleus is a complex, many body problem involving couplings between many neighbors. In the cuprates the echo decay of a nucleus located at the origin is determined by the Hamiltonian $\mathcal{H} = \sum_{\mathbf{r}} a_x(\mathbf{r})I_x(0)I_x(\mathbf{r}) + a_y(\mathbf{r})I_y(0)I_y(\mathbf{r}) + a_z(\mathbf{r})I_z(0)I_z(\mathbf{r})$, where the sum is over all nuclear sites. The couplings $a_\alpha(\mathbf{r})$ are given by dipolar couplings, indirect couplings involving an RKKY type mechanism, or both. [6] For arbitrary $a_\alpha(\mathbf{r})$ this problem has not been solved, although significant theoretical advances have been made recently. [7] Fortunately, it turns out that the nuclear couplings in the planes of the cuprates are dominated by the indirect channel, wherein lies the utility of Cu NMR to determine spatial properties such as the antiferromagnetic correlation length, ξ . [8] For example, studies invoking the strong O-Cu coupling in the plane have lead to important conclusions about the effects of impurities, as well as put constraints on theories of the dynamical susceptibility, $\chi'(\mathbf{q})$. [9,10] For the Cu-Cu couplings $a_z(\mathbf{r}) \gg a_x(\mathbf{r}), a_y(\mathbf{r})$. In this limiting case $I_z(0)$ and \mathcal{H} commute, allowing an exact determination of the echo decay of the nucleus at $\mathbf{r} = 0$: $M(t) = M_0 \exp[-t^2 \sum_{\mathbf{r}} a_z^2(\mathbf{r})/16]$, where the sum is over like neighbors. [8] A like nucleus is one that resonates at the same frequency as the one under observation; an unlike nucleus resonates at a different frequency. Typically, the echo decay is written in terms of a Gaussian time decay constant:

$$T_{2G}^{-2} = \frac{1}{8} \sum_{\mathbf{r}} a_z^2(\mathbf{r}). \quad (1)$$

T_{2G} gives information about the RKKY interactions giving rise to the indirect Cu-Cu coupling. [8] In fact, Pennington, and later Thelen showed that $a_z(\mathbf{r})$ can be written in terms of the real part of the dynamic susceptibility, $\chi'(\mathbf{q})$; using the MMP form for $\chi'(\mathbf{q})$ one finds $T_{2G} \sim 1/\xi$, where ξ is the antiferromagnetic correlation length. [8,11]

Studies of T_{2G} in the cuprates usually invoke the assumption that the couplings are time independent over

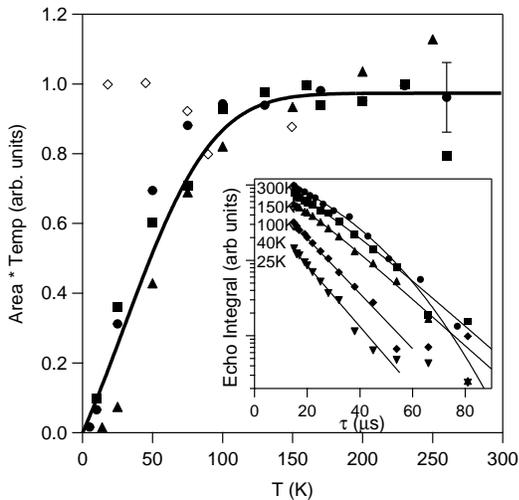


FIG. 1. NMR measurements in $\text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.15}\text{CuO}_4$ showing the wipeout of Cu signal. The solid symbols represent Cu data: (squares) 77 MHz, $H_0 \perp c$; (circles) 86 MHz, $H_0 \perp c$; (triangles) 95 MHz, $H_0 \parallel c$. The open diamonds represent La data for $H_0 \parallel c$ at 35.9 MHz. The solid line is a plot of the calculated intensity as described in the text. INSET: $H_0 \parallel c$ ^{63}Cu echo size versus the pulse spacing τ is plotted for a series of temperatures.

the time scale of the echo decay, or equivalently, $T_1 \gg T_{2G}$. In most cases this static assumption is valid. However, in cases where T_1 exhibits inhomogeneous glassy behavior (i.e., below the onset of Cu wipeout) this assumption is not necessarily valid; in fact, for wipeout to occur, $T_1 \leq T_{2G}$ locally. Therefore it is necessary to understand how the echo decay is modified when the static assumption is relaxed. This problem was studied successively by Walstedt and Cheong, [12], Recchia and Pennington, [13], and Curro and Slichter. [14] It turns out that one can write the Gaussian part of the echo decay of the nucleus at \mathbf{R} as

$$M_G(\mathbf{R}, t) = \exp \left[\frac{-t^2}{16} \sum_{\mathbf{r}} a_z^2(\mathbf{r} - \mathbf{R}) f \left(\frac{t}{T_1(\mathbf{r} - \mathbf{R})} \right) \right], \quad (2)$$

where $f(x) = \phi_L(x) + c_1 \phi_U(c_2 x)$. Here the constant $c_1 = (\frac{65}{63} \mathcal{P}^{65} \gamma^2) / (\frac{63}{65} \mathcal{P}^{63} \gamma^2) = 0.516$, accounts for the isotopic distribution of Cu which gives rise to both like and unlike nuclear couplings, $c_2 = (\frac{65}{63} \gamma / \frac{63}{65} \gamma)^2$,

$$\phi_U(x) = x^{-2}(x + 4e^{-x/2} - e^{-x} - 3) \quad (3)$$

for unlike nuclei and

$$\phi_L(x) = 8x^{-2}(5x/2 + 9e^{-x/2} - 2e^{-x} - 7) \quad (4)$$

for like nuclei. If T_1 is spatially uniform then $f(t/T_1(\mathbf{r} - \mathbf{R}))$ can be taken out of the summation, and Eq. (2) can be written trivially as $M_G(t) = \exp[-t^2 f(t/T_1)/2T_{2G}^2]$. In homogeneous systems such as

$\text{YBa}_2\text{Cu}_4\text{O}_8$, the temperature dependence of T_1 typically leads to minor corrections to the Gaussian form of the Cu echo decay. [15]

In cuprate systems that exhibit inhomogeneous spin fluctuations the Cu echo decay can be modified significantly. Most notably, the echo decay exhibits a dramatic, sharp drop on times scales shorter than what can typically be measured by an NMR spectrometer, an effect known as wipeout, in which the number of observable nuclei effectively is reduced. The full expression for the echo decay is given by:

$$M(t) = \sum_{\mathbf{R}} M_G(\mathbf{R}, t) \exp \left(\frac{-t}{T_{2R}(\mathbf{R})} \right) \quad (5)$$

where $M_G(\mathbf{R}, t)$ is given by Eq. (1), $T_{2R}^{-1}(\mathbf{r}) = (\beta + R)T_1^{-1}(\mathbf{r})$ is the Redfield term and β and R are constants. [6] The Redfield term accounts for on-site spin lattice relaxation over the time scale of the echo decay. $T_1(\mathbf{r})$ is given by the local activation energy:

$$T_1^{-1}(\mathbf{r}) = \gamma^2 h_{\text{hf}}^2 \tau_{\infty} \exp(E_a(\mathbf{r})/k_B T), \quad (6)$$

where γ is the gyromagnetic ratio, h_{hf} is the hyperfine field at the nucleus, τ_{∞} is the spin fluctuation time at high temperatures, and T is the temperature. The spin echo signal that is left over, then, arises from Cu nuclei that remain in regions of longer T_1 , or equivalently small E_a . The activation energies E_a are distributed with distribution function $\mathcal{P}(E_a)$, as in Ref. [4]. The Redfield term leads to wipeout since at low temperatures a significant fraction of nuclei experience T_1 's that are too fast to detect with the spectrometer. One might inquire what interactions these remaining nuclei experience: are they coupled to nuclei that experience similar T_1 's, or are they coupled to nuclei with much faster T_1 's? We argue that the latter case predominates, and is the reason for the change in the character of the echo decay. Experimentally it is found that the echo decay crosses over from Gaussian to exponential character. [4,16,17] (see Fig. (1)) We have argued previously that this crossover reflects regions of small spatial extent with fast spin fluctuations; here we present a more quantitative description of the phenomenon.

Consider a hypothetical spatial dependence of activation energies $E_a(\mathbf{r})$ such as that shown in Fig. (2). Note that at the observable nuclei are those near the minimum in the figure. It is useful to consider how the spatial dependence of $T_1(\mathbf{r})$ affects the echo decay of the remaining observable nuclei when the system becomes inhomogeneous. To address this question we analyze Eq. (5) with a specific model for the dynamic susceptibility. In order to calculate the indirect couplings, we adopt the expression used by Haase, Morr and Slichter *et al*, for the real space χ_{MMP} : [18,19]

$$\chi'(\mathbf{r} > \mathbf{0}) = \frac{\alpha}{4\pi} \cos \left(\frac{\pi x}{a} \right) \cos \left(\frac{\pi y}{a} \right) K_0 \left(\frac{r}{\xi} \right), \quad (7)$$

$$\chi'(\mathbf{r} = \mathbf{0}) = \frac{\alpha}{4\pi} \ln(1 + 4\pi\xi^2), \quad (8)$$

where α is a temperature independent constant, ξ is the antiferromagnetic correlation length, and $K_0(x)$ is the zeroth order modified Bessel function of the second kind.

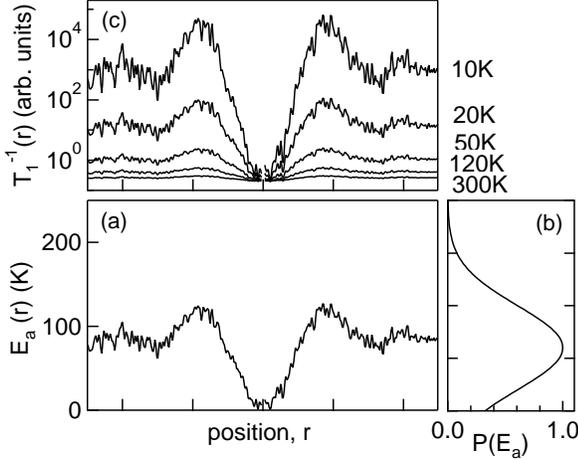


FIG. 2. (a) A hypothetical spatial distribution $E_a(\mathbf{r})$ versus r in the vicinity of a minimum in E_a . (b) The distribution $\mathcal{P}(E_a)$, with parameters typical for $\text{La}_{1.65}\text{Eu}_{0.2}\text{Sr}_{0.15}\text{CuO}_4$. (c) The resulting spatial distribution of T_1 's at a series of temperatures. Note that T_1 's get more spatially inhomogeneous as the temperature is lowered.

Note that this expression assumes translation invariance; however this assumption is not necessarily valid in the case of inhomogeneous spin fluctuations. Nevertheless this expression has been successful in describing the dynamical susceptibility in homogeneous cuprates as measured by neutron scattering and NMR. [20] Given a specific form for $\chi'(\mathbf{r})$ the indirect couplings $a_z(\mathbf{r})$ are given by: [18]

$$(g\mu_B)^2 a_z(\mathbf{r}) = A_z^2 \chi'(\mathbf{r}) + 2A_z B \sum_{m=1}^4 \chi'(\mathbf{r} - \mathbf{b}_m) + B^2 \sum_{m,n=1}^4 \chi'(\mathbf{r} - \mathbf{b}_n - \mathbf{b}_m), \quad (9)$$

where \mathbf{b}_m are the four nearest neighbor Cu sites, $A_z = 1.528\mu\text{eV}$ is the on-site hyperfine coupling and $B = -A/4$ is the transferred hyperfine coupling to the nearest neighbor Cu sites.

In order to estimate an appropriate form for the spatial dependence of T_1 in the vicinity of a minimum, we first consider the distribution function $\mathcal{P}(E_a)$ for E_a close to zero. In fact, given $E_a(\mathbf{r})$, $\mathcal{P}(E_a)$ is given by:

$$\mathcal{P}(E_a) = 1/|\nabla E_a(\mathbf{r})| = \left| r \left(\frac{dE_a}{dr} \right)^{-1} \right|, \quad (10)$$

so for $r \sim r_i$ we have $E_a(r) = (r - r_i)^2/2\mathcal{P}_0\lambda^2$, where $\mathcal{P}_0 = \mathcal{P}(E_a = 0)$, and λ is a length scale that characterizes the size of the regions in space where E_a is close to zero. If $\mathcal{P}_0 = 0$, we have $E_a(r) = \left(\frac{d\mathcal{P}_0}{dE_a} \right)^{-1/2} ((r - r_i)/\lambda)$, where $d\mathcal{P}_0/dE_a = d\mathcal{P}(E_a = 0)/dE_a$. Thus we use two test cases for the spatial dependence of T_1 , defining $W(\mathbf{r}) = 1/T_1(\mathbf{r})$:

$$W_e(\mathbf{r}) = W_0 \exp((r - r_i)/l) \quad \text{case I} \quad (11)$$

$$W_G(\mathbf{r}) = W_0 \exp((r - r_i)^2/2l^2) \quad \text{case II}, \quad (12)$$

where $l = k_B T \sqrt{d\mathcal{P}_0/dE_a} \lambda$ for case I, $l = \sqrt{\mathcal{P}_0 k_B T} \lambda$ for case II, and $W_0 = \gamma^2 h_{\text{hf}}^2 \tau_\infty$. We then calculate $M(t)$ (Eq. (5)) numerically on an $N \times N$ lattice with $N = 11$. The results are shown in Figs. (3) and (4). Note that T_{2G} is defined as in Eq. (1). Here we use $\xi = 2a$ and $\alpha = 1.0$. Although it is not possible to determine the absolute value of ξ , it is not unreasonable to expect $\xi \sim 2 - 3a$, at least in regions of fast spin fluctuations (slow T_1).

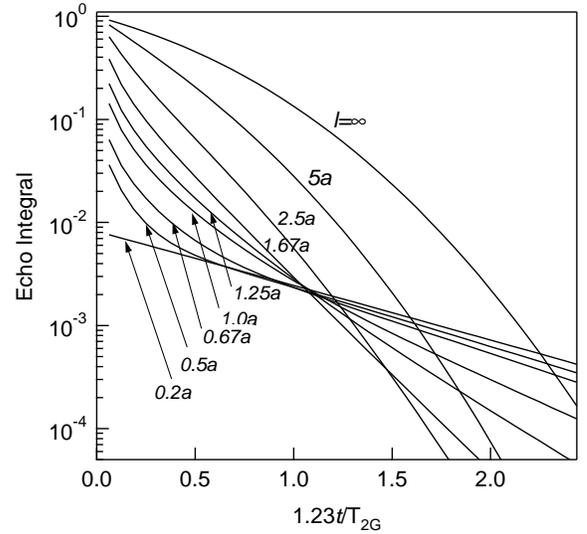


FIG. 3. Case I: The calculated $M(t)$ versus t for $\xi = 2a$, with $W(\mathbf{r}) = W_e(\mathbf{r})$ and $W_0 = 0.25/T_{2G}$.

In both cases, as the degree of inhomogeneity grows (as l shortens, or as T decreases), there are two important changes in the form of the echo decay. First, the echo decays drastically at short times due to the fast on-site relaxation from the Redfield term $\exp(-t/T_{2R}(\mathbf{R}))$. This is the Cu wipeout. Secondly, the echo decay from the remaining sites gradually becomes more exponential, as seen experimentally (see Fig. (1)). This crossover occurs when $l \leq \xi$. In other words, when the neighbors that have a significant coupling (those within a radius ξ) begin to undergo fast spin lattice fluctuations, they behave as unlike nuclei; essentially they behave as strongly coupled independent fluctuating moments. As the fluctuation rate of these neighbors gets faster, the echo decay rate becomes exponential, and eventually the fast fluctuations of the neighbors decouples from the nuclei under

observation. In the extreme limit, the nucleus at $\mathbf{r} = \mathbf{0}$ is the only one with T_1 sufficiently slow to be observable, and all of the neighbors are fluctuating so fast they do not contribute to the echo decay; thus the single nucleus at the origin decays solely due to the local Redfield term.

Note that for case II the spin lattice relaxation rate changes less drastically between the origin and the nearest neighbor sites. As a result, the echo decay, which is dominated by the coupling to the nearest neighbor sites, is not as strongly affected by the onset of the inhomogeneity ($T_1(\mathbf{r})$ is locally more homogeneous). As seen in Figs. (3) and (4), $W_G(\mathbf{r})$ results in an echo decay that has a slightly more positive curvature at short times than $W_e(\mathbf{r})$. Although one cannot distinguish between the two cases experimentally, qualitatively the echo decay curves in Fig. (1) seem to match more those in case I. In other words, for $T = 25\text{K}$, the echo decay in Fig. (1) looks like the echo decay in Fig. (3) for $l \approx 1 - 2a$.

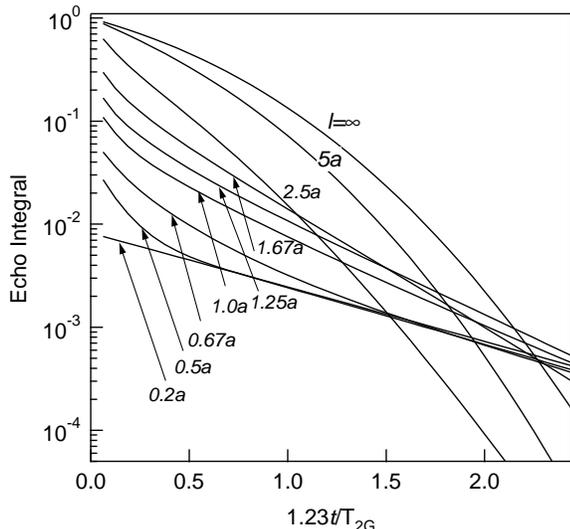


FIG. 4. Case II: The calculated $M(t)$ versus t for $\xi = 2a$, with $W(\mathbf{r}) = W_G(\mathbf{r})$ and $W_0 = 0.25/T_{2G}$.

We can now estimate λ , the effective radius of the regions in space with low activation energy. To do so, we take advantage of the fact that the wipeout fraction, $I_0(T)$ is given by:

$$I_0(T) = \int_0^{\kappa k_B T} \mathcal{P}(E_a) dE_a, \quad (13)$$

where $\kappa \approx 1.5$ is defined as in [4]. Therefore, we have:

$$\mathcal{P}_0 = \frac{1}{\kappa k_B} \left(\frac{dI_0}{dT} \right)_{T=0} \quad (14)$$

$$\frac{d\mathcal{P}_0}{dE_a} = \frac{1}{(\kappa k_B)^2} \left(\frac{d^2 I_0}{dT^2} \right)_{T=0}, \quad (15)$$

where $I_0(T)$ is measured experimentally, and is shown in Fig. (1). From the data $\mathcal{P}_0 \approx 7 \times 10^{-3} K^{-1}$, $d\mathcal{P}_0/dE_a \approx$

$4 \times 10^{-4} K^{-2}$, and $l \approx a$ at 25K. For case I, we have $\lambda \approx 2.0a$, and for case II, we have $\lambda \approx 2.3a$. Note however, that measurements of $I_0(T)$ at low temperature are difficult, and the error bars on \mathcal{P}_0 and $d\mathcal{P}_0/dE_a$ are significant. Therefore, the values given here are to be taken only as estimates of the order of magnitude.

Clearly, the echo decay is not consistent with slow spatial variations of the activation energy, but rather with disperse, localized point or line minima. A possible interpretation of the small spatial regions of small E_a is that these regions constitute domain walls between large regions of coupled spins. In the large domains, the collective fluctuation rate (τ^{-1}) would be small, and T_1 very fast. Calculations of the echo decay for a linear trough of minima in T_1 , rather than the point minima in cases I and II, are not consistent with the data. The reason is that the nuclei along the trough remain like nuclei, and the echo decay does not become exponential. Such a case is idealized; however one cannot rule out a situation in which the line minima fluctuates and neighbors along the trough decouple. Further experiments to probe $\mathcal{P}(E_a)$ and the echo decay with greater precision are likely to shed more light on the spatial dependence of the local activation energies, and their relationship to the stripe lattice. Also, a careful analysis of the temperature dependence of l may allow one to distinguish between case I and II.

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