What has NMR taught us about stripes and inhomogeneity?

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The purpose of this brief invited paper is to summarize what we have (not) learned from NMR on stripes and inhomogeneity in La$_{2-x}$Sr$_x$CuO$_4$. We explain that the reality is far more complicated than generally accepted.

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Neutron scattering experts generally declare everything static as soon as fluctuations slow down below the frequency threshold of 10$^{11}$ Hz for elastic scattering measurements. In NMR lineshape measurements, the separation time between RF excitation pulses is typically 10 $\mu$s, which sets the frequency scale of the shutter speed of the NMR picture at 10$^9$ Hz. In our quest to capture the truly static stripe phase, we developed wide-frequency zero-field NMR techniques with a top-loading He cryostat. Our measurements in La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$ and La$_{2-x}$Ba$_x$CuO$_4$ at $x \sim \frac{1}{8}$ revealed that motional narrowing effects average out the hyperfine magnetic fields from Cu spins even at 350 mK. Alas, static stripes are not really static even at 350 mK. The observed NMR intensity at 350 mK accounts for essentially 100 percent of Cu nuclear spins from the sample. This means that the NMR relaxation rates 1/$T_{1,2}$ governed by spin fluctuations at the NMR frequency ($\sim 10^{10}$ Hz) are small enough to detect the signals, which implies that the majority of the spectral weight of the spin fluctuations has slowed down to 10$^9$ Hz levels.

The dynamic nature of stripes is even stronger in La$_{2-x}$Sr$_x$CuO$_4$ ($x \sim \frac{1}{8}$), even though stripes are frequently and erroneously quoted as static below 20 K based on elastic neutron scattering data. We managed to detect a narrow, Zeeman-perturbed zero-field NMR at 350 mK in La$_{1.885}$Sr$_{0.115}$CuO$_4$ [2]. The narrow lineshape is a consequence of motional narrowing. Moreover, the integrated intensity corresponds to only a few percent of the sample. This implies that the majority of Cu nuclear spins are still under the influence of relatively fast ($>10^9$ Hz) fluctuations, and are hence undetectable.

The dynamic nature of stripes leads to an unfortunate consequence; what NMR observes in the static stripe phase is not what one would normally conceive as stripes, but slowly fluctuating magnetic entities blurred by their motion. In order to investigate the slowing of stripes through NMR techniques this forces us to rely on a somewhat indirect method which consist of measuring the Cu NQR/NMR wipeout of intensity [3]. The wipeout fraction $F(T)$ is the fraction of the Cu nuclear spins that become undetectable due to fluctuating stripes. As detailed in [1], when the fluctuation frequency falls between 10$^{11}$Hz and 10$^9$Hz, very fast NMR relaxation rates prevent us from detecting Cu NQR/NMR signals. Effectively, $F(T)$ is the volume fraction of the segments in the CuO$_2$ plane which fluctuate in the aforementioned frequency range. Quite interestingly, $F(T)$ in Nd co-doped samples closely follows the neutron/x-ray scattering intensity arising from charge order at $x = \frac{1}{8}$. This led us to equate $F(T)$ in the superconducting regime of La$_{2-x}$Sr$_x$CuO$_4$ with the volume fraction of charge-ordered segments where stripe fluctuations have slowed [3]. The onset temperature $T_{NQR}$ for $F(T)$ decreases with increasing $x$ [3] (see Fig. 1). Subsequent studies [1, 4] showed that $T_{NQR}$ precisely agrees with the onset of charge order $T_{charge}$ for $x > \frac{1}{8}$. This supports our physical picture for $x > \frac{1}{8}$. As it turned out, however, $T_{NQR}$ is always higher than $T_{charge}$ in the lower Sr doping range $x < \frac{1}{8}$ of Nd co-doped samples. Instead, it is the inflection point of the curve in the temperature dependence of $F(T)$ that agrees with $T_{charge}$ [1]. Moreover, subsequent x-ray scattering efforts resulted in no hard evidence for charge order in La$_{2-x}$Sr$_x$CuO$_4$ without Nd co-doping. We noted from the very beginning [3] that the monotonic increase of $T_{NQR}$ below $x = \frac{1}{8}$ was counterintuitive, and we were puzzled by these newer revelations for some time [1]. There must be something else involved in the mechanism of $F(T)$ below $T_{NQR}$ down to $T_{charge}$ for $x < \frac{1}{8}$. We will come back to this point below.

The fact that some Cu nuclear spins are observable while others are wiped-out implies a highly inhomogeneous nature of the CuO$_2$ planes, i.e. that the fluctuation frequency is different position by position. This glassy nature of the stripes led to significant confusion in the NMR community. A major source of confusion is that from old days everybody in the NMR community knew that NMR data in La$_{2-x}$Sr$_x$CuO$_4$ showed a variety of signatures for an electronic inhomogeneity, and distinguishing this intrinsic electronic inhomogeneity from the inhomogeneous magnetism arising from the glassy slow-
ing of stripes is not a straightforward task. Some authors even claim that all NMR anomalies including wipeout effects may be understood based on an analogy with conventional spin glass without invoking any spatially coherent nature for the stripes. Prior to, we had already pointed out in [3] the importance of the similarity with NMR wipeout effects in simple Cu metal with dilute magnetic Fe impurity spins. The whole point of the Los Alamos paper [6] missed this point.

In any case, these confusions concerning the inhomogeneity led us to critically reexamine the issue of electronic inhomogeneity, more specifically, the validity of the assumption that CuO$_2$ planes are electronically homogeneous even in alloyed high $T_c$ cuprates. To make a long story short, our measurement of $1/T_1$ as a function of temperature and frequency within each Cu NQR lineshape indicates that the local hole concentration in La$_{2-x}$Sr$_x$CuO$_4$ deviates significantly from nominal $x$, whether the sample is a poly-crystalline or high-quality single crystal. This finding clearly raises questions regarding theoretical debates of a “universal electronic phase diagram”, including La$_{2-x}$Sr$_x$CuO$_4$, which are based on the assumption that hole doping is homogeneous. Furthermore, our new result has several implications in our understanding of stripes and the NQR wipeout effects. First, the intrinsic electronic inhomogeneity may be partially responsible for the glassy, inhomogeneous nature of the slowing of stripes observed even at $x = \frac{1}{8}$. Second, it naturally explains why the onset temperature for wipeout jumps up to $T_{NQR} \sim 300$ K at $x \sim 0.05$. Some segments of CuO$_2$ planes become nearly undoped below 300 K. For these undoped patches, the strong short-range Néel-order results in enhanced NMR relaxation rates causing the Cu NQR signal to become undetectable. Whether nucleation of these nearly undoped patches is associated with the rotation of the stripes from the diagonal to vertical direction remains to be seen. Third, similar effects would provide a natural account for why wipeout sets in at $T_{NQR}$ above $T_{charge}$ below $x = \frac{1}{8}$. The inflection point in the $T$ dependence of $F(T)$ corresponds to the temperature where spatially coherent fluctuations of glassy stripes finally kick in.

To summarize, one needs to take into account both the intrinsic electronic inhomogeneity and the glassy, inhomogeneous slowing of stripes. Much of the confusion over La$_{2-x}$Sr$_x$CuO$_4$ stems from the failure by many authors to recognize the importance of both.

FIG. 1: The onset temperature $T_{NQR}$ of Cu NQR wipeout effects in La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ for $y = 0$ (○), and $y = 0.04$ (●). 4% Zn doping suppresses superconductivity completely.

[1] A.W. Hunt et al., Phys. Rev. B 64, 134525 (2001).
[2] A.W. Hunt et al., unpublished thesis work at M.I.T (2001).
[3] A.W. Hunt et al., Phys. Rev. Lett. 82, 4300 (1999).
[4] P.M. Singer et al., Phys. Rev. B 60, 15345 (1999).
[5] P.M. Singer et al., Phys. Rev. Lett. 88, 47602 (2002).
[6] N. Curro et al., Phys. Rev. Lett. 85, 642 (2000).
[7] T. Imai et al., J. Phys. Soc. Jpn. 59, 3846 (1990).
