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Comment on "Order-Disorder Structural Phase Transition in $La_{2-x}Sr_xCu_{4+\delta}$ at 150 K"

In a recent Letter, Saylor and Hohenemser [1] reported a perturbed $\gamma\gamma$ angular correlation (PAC) study of ^{111}In-^{111}Cd deposited in $La_{1-x}Sr_xCuO_{4+\delta}$. A large ($\sim 2\times$) increase of the PAC quadrupolar linewidth and a similarly large decrease of the quadrupolar asymmetry parameter were observed below ~ 150 K. These phenomena were attributed to changes in electric-field gradients at probe-nuclei sites due to a reversible order-disorder structural phase transition in the intrinsic material, *assuming that the probe is sensitive to electric-field gradients which are characteristic of the host.*

This assumption can be checked by nuclear quadrupole resonance (NQR) measurements. Both NQR and PAC spectra reflect the distributions of local electric-field gradients at nuclear sites, but NQR avoids the necessity of introducing a foreign nucleus. The present Comment describes measurements of ^{139}La NQR spectra in nearly stoichiometric single crystals of undoped $La_2CuO_{4+\delta}$ which show no 150-K anomaly in either the NQR frequency or the linewidth.

Fourier-transform spectra were obtained for temperatures between 75 and 325 K. Below the Néel temperature T_N the ^{139}La NQR line is Zeeman-split by hyperfine coupling to the antiferromagnetically ordered Cu spins [2,3] as shown in Fig. 1(a) for a sample with $T_N = 305 \pm 5$ K. Except near T_N the full width at half maximum of each line [Fig. 1(b)] is ~ 15 kHz, which is $\sim 0.1\%$ of the transition frequency. (The observation of three lines with varying linewidths near 300 K is presumably due to inhomogeneity in T_N.) It can be seen that neither frequencies nor linewidths exhibit large changes between ~ 75 K and just below T_N. The sensitivity of the present NQR linewidth measurements to such changes is $\sim 1{:}10^4$ of the transition frequency.

Thus there is no evidence in our NQR spectra for structural disorder in the end compound $La_2CuO_{4+\delta}$. It has been suggested [1] that the transition is suppressed by excess oxygen, of which the depression of T_N is a very sensitive indicator. For our most nearly stoichiometric sample $T_N \simeq 305$ K is about 23 K less than the highest observed value (328 K) [4]. This depression is about twice that of the sample used in the PAC study ($T_N = 317$ K), but there is very little excess oxygen in either sample and it is hard to see how such a small difference could induce a significant structural change.

If, as has been argued [1,5], the ^{111}In probe occupies the La site, the lack of a ^{139}La NQR anomaly is strong evidence against an order-disorder transition. The spread in asymmetry parameter η provides much of the PAC linewidth. Even though the ^{139}La η is very small, it seems unlikely that disorder could produce a 10%–20% spread in PAC frequency without also producing ^{139}La NQR frequency spreads considerably larger than our upper bound of $\sim 0.1\%$. A similar argument should hold even if the ^{111} In site were elsewhere in the unit cell. We therefore question whether the PAC behavior is intrinsic to $La_2CuO_{4+\delta}$.

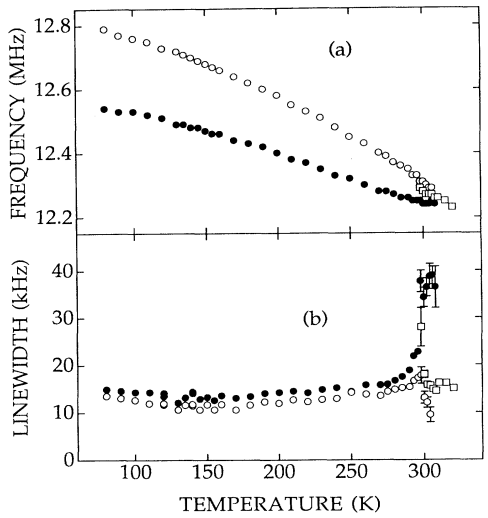

FIG. 1. Temperature dependence of ^{139}La ($\pm\frac{5}{2} \leftrightarrow \pm\frac{3}{2}$) NQR (a) frequencies and (b) linewidths in nearly stoichiometric $La_2CuO_{4+\delta}$ ($T_N \simeq 305$ K). Squares: pure NQR ($T > T_N$). Circles: Zeeman-split NQR ($T < T_N$). Effects of inhomogeneity in T_N are seen in the region 290–310 K. Neither frequencies nor linewidths exhibit an anomaly at 150 K.

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[1] J. Saylor and C. Hohenemser, Phys. Rev. Lett. **65**, 1824 (1990).
[2] Y. Kitaoka *et al.*, J. Phys. Soc. Jpn. **56**, 3024 (1987).
[3] H. Nishihara *et al.*, J. Phys. Soc. Jpn. **56**, 4559 (1987).
[4] S.-W. Cheong *et al.*, Physica (Amsterdam) **158C**, 109 (1989).
[5] J. Saylor *et al.*, Phys. Rev. B **40**, 6854 (1989).