Comment on order-disorder structural phase transition in La$_{2-x}$Sr$_x$Cu$_{4+}$ at 150 K

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Comment on "Order-Disorder Structural Phase Transition in La$_2$-$\delta$Sr$_x$Cu$_{4+\delta}$ 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$_x$CuO$_{4+\delta}$. A large (~2×) 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$_2$CuO$_{4+\delta}$ 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 ± 5 K. Except near $T_N$ the full width at half maximum of each line [Fig. 1(b)] is ~15 kHz, which is ~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 ~1:10$^4$ of the transition frequency.

Thus there is no evidence in our NQR spectra for structural disorder in the end compound La$_2$CuO$_{4+\delta}$. 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 $T_N$ =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 $\eta$ provides much of the PAC linewidth. Even though the $^{139}$La $\eta$ 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 ~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$_2$CuO$_{4+\delta}$.

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