

**Effects of charge inhomogeneities on elementary excitations in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$** S. R. Park,<sup>1</sup> A. Hamann,<sup>2</sup> L. Pintschovius,<sup>2</sup> D. Lamago,<sup>2,3</sup> G. Khaliullin,<sup>4</sup> M. Fujita,<sup>5</sup> K. Yamada,<sup>6</sup>  
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Purely local experimental probes of many copper oxide superconductors show that their electronic states are inhomogeneous in real space. For example, scanning tunneling spectroscopic imaging shows strong variations in real space, and according to nuclear quadrupole resonance (NQR) studies, the charge distribution in the bulk varies on the nanoscale. However, the analysis of the experimental results utilizing spatially averaged probes often ignores this fact. We have performed a detailed investigation of the doping dependence of the energy and linewidth of the zone-boundary Cu-O bond-stretching vibration in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  by inelastic neutron scattering. Both our results as well as previously reported angle-dependent momentum widths of the electronic spectral function detected by angle-resolved photoemission can be reproduced by including the same distribution of local environments extracted from the NQR analysis.

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**I. INTRODUCTION**

By now, it is recognized that charge inhomogeneity is an important aspect of copper oxide superconductors. Undoped copper-oxygen planes are Mott insulators due to a strong on-site Coulomb interaction. They become metallic and exhibit high-temperature superconductivity when they are doped.<sup>1</sup> Different types of charge inhomogeneities can emerge as a result of doping. Nanoscale inhomogeneity due to strong correlations between electrons, such as a stripe or checkerboard order,<sup>2-5</sup> received a lot of attention in recent years.<sup>6-8</sup> Other sources of inhomogeneity are randomly distributed heterovalent substituents or extra oxygen atoms, which introduce doped holes or electrons into the copper-oxygen planes.<sup>9,10</sup> In most cases (with some notable exceptions, such as ortho II-ordered  $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ ), the dopants act as charged impurities. They form an inhomogeneous Coulomb potential impacting the conduction electrons in the copper-oxygen planes, resulting in an inhomogeneous charge distribution. Local probes, such as scanning tunneling microscopy (STM) have provided evidence for this inhomogeneity in cuprates, such as  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  (BSCCO),<sup>11-14</sup>  $\text{Ca}_{2-x}\text{Na}_x\text{CuO}_2\text{Cl}_2$ ,<sup>15</sup> and  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO).<sup>16</sup> Bulk-sensitive nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) experiments confirmed that an inhomogeneous charge distribution is not just a property of the surface but is a feature of the bulk in LSCO.<sup>17-19</sup>

Here, we show how some doping-dependent features of the observed spectra of phonons and electronic quasiparticles in LSCO can be explained naturally in terms of an inhomogeneous charge distribution. For phonons, we base our analysis on results of neutron-scattering experiments presented here. We use previously published angle-resolved photoemission (ARPES) data for the electronic response.

It is a long-standing observation that the energy of the zone-boundary Cu-O bond-stretching (half-breathing) phonon,  $\mathbf{q} = (0.5, 0, 0)$ , depends strongly on doping. It changes from about 81 meV at  $x = 0$  to about 67 meV at  $x = 0.3$ .<sup>20-22</sup> Figure 1(b), which combines previous neutron-scattering and x-ray scattering data with our results for the frequency of the half-breathing mode, summarizes the available data. Since the phonon frequency depends strongly on the doping, especially in the underdoped region, an inhomogeneous doping should increase its observed linewidth. We have measured the linewidth of this phonon in the underdoped region to investigate this effect.

**II. EXPERIMENTAL DETAILS**

Inelastic neutron-scattering experiments were performed on large high-quality single crystals of LSCO with  $x = 0, 0.05, 0.07$ , and  $0.15$  on the 1-T triple-axis spectrometer at the ORPHEE reactor at the Laboratoire Leon Brillouin at Saclay, France. The monochromator and analyzer were the (220) reflection of copper and the (002) reflection of pyrolytic graphite, respectively. The measurements were performed at reciprocal lattice vectors  $\mathbf{Q} = (4 + h, 0, 0)$  in the tetragonal notation. In order to measure the intrinsic phonon linewidth, it is important to properly account for the experimental resolution, which depends not only on the instrument configuration, but also on the phonon dispersion in the vicinity of the measured point in  $\mathbf{Q}$ - $\omega$  space. We calculated the effective energy resolution for the bond-stretching phonon branch at each doping level based on the spectrometer resolution combined with doping-dependent dispersions calculated from the shell model, whose parameters were optimized to agree with previously published phonon-dispersion data along high-symmetry directions.<sup>23</sup> The calculated experimental resolutions at  $\mathbf{Q} = (4.5, 0, 0)$  for  $x = 0.0, x = 0.05, x = 0.07$ , and  $x = 0.15$  are 4.5, 3.6,

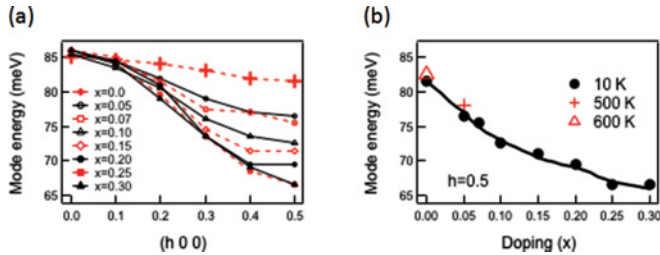


FIG. 1. (Color online) Doping dependence of the Cu-O bond-stretching mode energy. (a) Dispersion in the [100] direction at different doping levels. (b) Zone-boundary half-breathing ( $h = 0.5$ ) mode energy as a function of doping ( $x$ ).

3.2, and 3.4 meV, respectively. Electron-phonon coupling or anharmonicity should result in a Lorentzian intrinsic phonon line shape, but we found that a Gaussian gives better fits. Thus, the resolution function was convolved with a Gaussian function representing the phonon peak, with the Gaussian width (and frequency and amplitude) determined by fitting to a constant- $Q$  scan. The background was determined from the scans at nearby wave vectors as was performed in Ref. 24.

### III. RESULTS AND DISCUSSION

Figures 2(a)–2(d) show inelastic neutron-scattering spectra of the half-breathing mode of LSCO from  $x = 0$  to  $x = 0.15$ . The frequency clearly softens with increasing doping, as previously reported.<sup>20</sup> Red (solid) lines through the raw data indicate the fitted Gaussian peak, convolved with the resolution function, on top of a linear background. In the cases of  $x = 0.0$  and  $x = 0.05$ , we have included a second Gaussian to account for a small peak at 87 meV that we believe corresponds to an artifact,<sup>25</sup> based on its lack of dependence on doping, although we do not have a definitive explanation for it. We will ignore it in the following discussion of the linewidth; including it in the evaluation of the linewidth would not make a qualitative difference to the argument, as discussed below.

The fitted phonon linewidths are plotted as a function of doping in Fig. 2(f). The filled symbols show the width of the half-breathing mode at low temperatures. As one can see, the width makes a large jump from  $x = 0$  to  $x = 0.05$  and then gradually decreases with further doping. For comparison, the open diamonds show the fitted width of the zone-center mode. The absence of any significant variation with doping suggests that the results for  $h = 0.5$  are intrinsic and are not a result of a difference in sample quality. Furthermore, different samples with the same doping give identical results.

The most obvious explanation of phonon broadening at the zone boundary is electron-phonon coupling. According to the conventional theory of metals, the phonon linewidth due to electron-phonon coupling should be proportional to the density of states near  $E_F$ ,<sup>27,28</sup> which is roughly proportional to doping in LSCO.<sup>29–31</sup> The trend for the  $h = 0.5$  mode in Fig. 2(f) clearly violates a simple monotonic increase with doping. In particular, the  $x = 0.05$  sample is insulating<sup>32</sup> and should not have more conduction electrons near the Fermi surface than the  $x = 0.07$  and 0.15 samples. Thus, we can rule out electron-phonon coupling as the dominant cause of this effect.

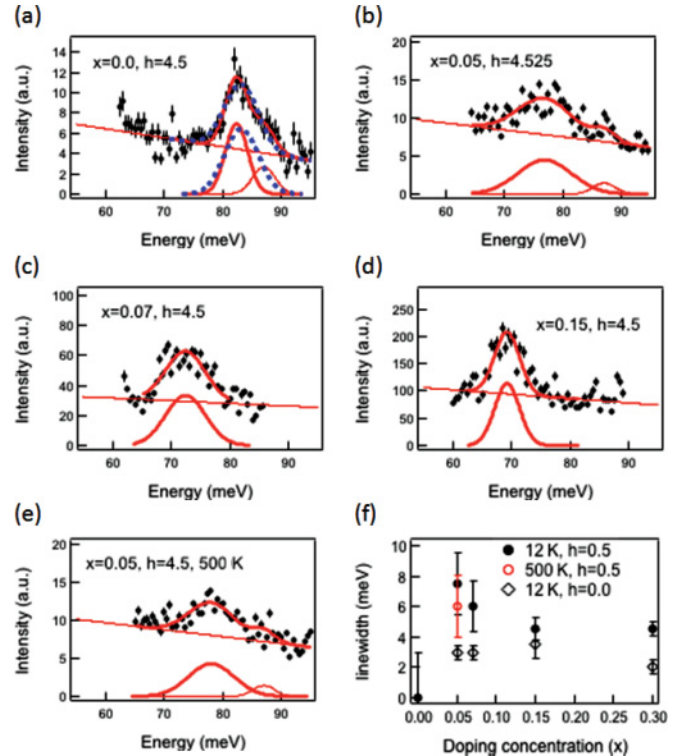


FIG. 2. (Color online) Inelastic neutron-scattering data of half-breathing zone-boundary bond-stretching modes in LSCO. (a)  $x = 0$ , (b)  $x = 0.05$ , (c)  $x = 0.07$ , (d)  $x = 0.15$  at 10 K, and (e)  $x = 0.05$  at 500 K. Red (solid) lines on raw data are composed of a linear background and two Gaussian peaks for (a) and (b) and a linear background and a single Gaussian peak for (c)–(e). Large Gaussian peaks on the bottom represent longitudinal bond-stretching modes; small Gaussian peaks at 87 meV represent the suspect artifact (see text). Full widths at half maximum (FWHM) of the main peaks of  $x = 0$ ,  $x = 0.05$ ,  $x = 0.07$ , and  $x = 0.15$  are 4.5, 10.5, 8, and 5 meV, respectively. Two Gaussian peaks of  $x = 0.0$  data and a small Gaussian peak of  $x = 0.05$  data are resolution limited (4.5 meV). Dotted blue line on  $x = 0.0$  raw data is composed of a linear background and a Gaussian peak with 6.5-meV FWHM. (f) Doping dependence of the zone boundary ( $h = 0.5$ ) and zone center ( $h = 0.0$ ) longitudinal bond-stretching mode linewidths after deconvolving the resolution function.

Another possible explanation for the broadening of the low-temperature line shape is the effect of the tilt of the  $\text{CuO}_6$  octahedra responsible for the transition from the high-temperature tetragonal to the low-temperature orthorhombic phase.<sup>33</sup> We can rule out this scenario because octahedral tilt is largest for  $x = 0$  and decreases with doping.

Here, we propose a different mechanism to explain our results. As already mentioned, NQR and NMR experiments found that the doping level in nominally homogeneous samples of the LSCO family is strongly spatially inhomogeneous.<sup>17,18</sup> Since the phonon frequency is known to depend on doping, as shown in Fig. 1, we propose that the observed broadening results from the distribution of local environments observed by NQR. To test this idea, we modeled phonon line shapes based on the distribution of dopings measured by NQR (Ref. 17) combined with the doping dependence of phonon frequencies. (As the intrinsic phonon linewidth is smaller

than the resolution, we have ignored it in the modeling.) The resulting line shapes were convolved with the resolution function. This analysis provides a very rigorous test of the above hypothesis since it contains no adjustable parameters aside from the overall peak magnitude.

One can imagine more rigorous approaches to calculate the phonon spectrum in an inhomogeneous medium, such as calculations based on the coherent potential approximation (CPA), which is usually used for calculating phonon spectra in alloys.<sup>34</sup> But it is not clear if this method would apply to phonons in solids with charge inhomogeneity as in LSCO since the CPA was originally developed for random-mass-disordered alloys.<sup>34</sup> However, we note that calculations of disorder effects in isotopically disordered Ge using CPA gave no better results (actually even slightly inferior results) than calculations using large supercells.<sup>35</sup> Somewhat surprisingly, the supercell approach reproduced the experimentally observed phonon frequencies, linewidths, and intensities very well, despite the fact that this approach was based on harmonic lattice dynamics. Therefore, we think that our ansatz for calculating linewidths in LSCO—which is equivalent to a simplified version of the supercell approach—is quite adequate.

Figure 3 shows that the calculated line shapes reproduce the experimental low-temperature phonon line shapes at three doping levels reasonably well. We also measured this phonon in the  $x = 0.05$  sample at 500 K. Heating to such temperatures increases anharmonicity and, thus, broadens the phonon. For example, this broadening already has been observed at 330 K near the zone center in similar samples.<sup>36</sup> However, the 500-K data appear to be slightly narrower than at 10 K, which is consistent with decreasing charge inhomogeneity with increasing temperature as reported by NQR.<sup>17</sup> An anharmonic contribution must be present, but it appears to be comparable to the contribution from charge inhomogeneity.

It is not completely clear why charge inhomogeneity decreases upon heating the sample. One possibility is that some electrons are trapped by the disordered chemical potential due to randomly distributed Sr cations.<sup>17</sup> Then, the real-space distribution of the trapped electrons should broaden with increasing temperature, resulting in a smoother disorder potential.<sup>17</sup> Charge inhomogeneity, resulting from electronic correlations, should also decrease at elevated temperatures due to the breakup of electronic self-organization by thermal fluctuations.

Based on the above evidence, we conclude that the phonon linewidth can be explained well by charge inhomogeneities measured by NQR. Intrinsic phonon lifetime, which is governed by anharmonicity and electron-phonon coupling, is too long to measurably contribute to the linewidth in addition to the effect of inhomogeneous doping.

If this explanation is correct, charge inhomogeneity should show up in other experiments involving spatially averaged probes. In particular, electronic quasiparticles measured by ARPES also should be affected by inhomogeneous doping since the Fermi surface in LSCO has a strong and nontrivial doping dependence: The Fermi wave vector  $\mathbf{k}_f$  is nearly doping independent at the nodes but rapidly decreases with increased doping near the antinodes due to the proximity to a Van Hove singularity [Fig. 4(c)].<sup>31,38</sup> Similarly, measurements of the momentum width of the spectral function obtained from

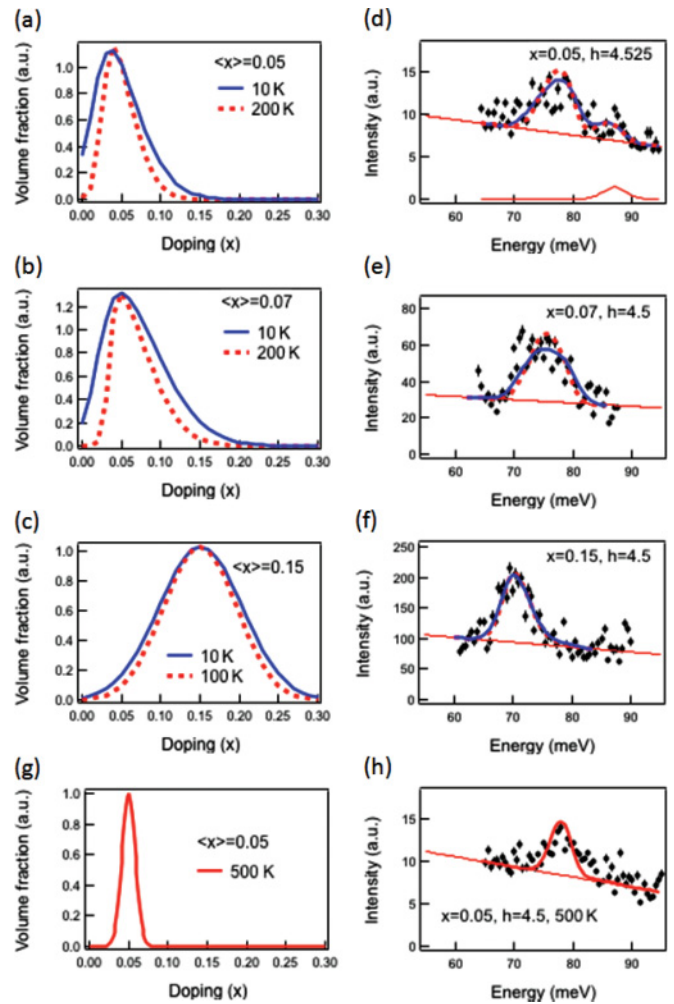


FIG. 3. (Color online) Doping distributions for (a) and (g)  $x = 0.05$ , (b)  $x = 0.074$ , and (c)  $x = 0.15$  based on the NQR data.<sup>17</sup> Inelastic neutron-scattering data from the half-breathing mode for (d)  $x = 0.05$ , (e)  $x = 0.07$ , (f)  $x = 0.15$  at 10 K, and (h)  $x = 0.05$  at 500 K. Dotted red curves and solid blue curves over data points show results calculated from the inhomogeneous doping effect at high temperatures and at 10 K, respectively. Red curve in (h) shows the simulated results at 500 K.

MDCs show that the width at the Fermi level is substantially larger in the antinodal region compared to the nodal one.<sup>37,39</sup> We find that combining the chemical-potential-distribution model with the observed doping dependence of  $\mathbf{k}_f$  provides a reasonable explanation for the  $\mathbf{k}$  dependence of the momentum widths.

Figure 4(c) shows the doping-dependent Fermi surfaces extracted from the ARPES data.<sup>31</sup> We simulate the ARPES data by introducing a Lorentzian broadening in energy of  $\Delta E = 70$ -meV FWHM, which translates into  $\Delta E/v_F$ , broadening in  $\mathbf{k}$ , at each  $\mathbf{k}$  point and assuming a distribution of the electronic band structures weighted by the inhomogeneous doping distribution, as we had done for the zone-boundary phonon. A 70-meV FWHM is a reasonable value based on the measured quasiparticle peak width near the Fermi energy in LSCO.<sup>40</sup> We use tight-binding bands with first-, second-, and third-neighbor hopping parameters fitted to the ARPES data as the electronic

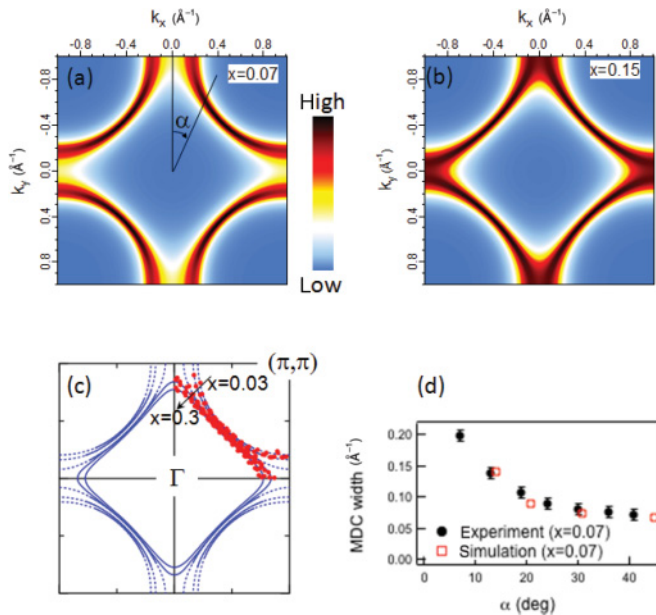


FIG. 4. (Color online) Simulated Fermi surfaces for (a)  $x = 0.07$  and (b)  $x = 0.15$ . (c) Doping-dependent Fermi surface evolution extracted from the ARPES data.<sup>31</sup> (d) Momentum distribution curve (MDC) widths at Fermi energy from the experiment<sup>37</sup> and simulation from antinodes to nodes.

band structures for each doping.<sup>37</sup> Figures 4(a) and 4(b) show the simulated results for the Fermi surfaces of  $x = 0.07$  and  $x = 0.15$ , respectively. Momentum widths of the quasiparticles near the Fermi surface broaden on going from the nodal to the antinodal directions for both dopings, as observed in the experiments.<sup>41</sup> Figure 4(d) demonstrates good quantitative agreement between the MDC widths calculated from our model and the data<sup>37</sup> for  $x = 0.07$ . So far, the linewidth of ARPES data at the Fermi energy usually has been interpreted as the self-energy effect of the quasiparticle due to impurity scattering. There is, however, no reason that the linewidth near the antinodal direction is much broader than the linewidth near the nodal direction due to impurity scattering, which is believed to be isotropic. Yet, our analysis provides a natural explanation about broader linewidth near the antinodal direction.

An alternative model is to apply a broadening of fixed width in momentum rather than energy; however, it was found that uniform momentum broadening gives a much poorer agreement with the data. The physical significance of this result is unclear.

Broader ARPES linewidths near the antinodal directions were interpreted in terms of stronger scattering,<sup>41</sup> but we quantitatively reproduced the experimental data with a large enhancement of the MDC widths toward the antinodal direction by the inhomogeneous doping effect. Our simulation, which only includes doping inhomogeneity, cannot reproduce the entire ARPES spectrum, especially in the antinodal region, because of the pseudogap and the incoherent spectrum at higher binding energies.<sup>42</sup> However, we can conclude that the long-standing observation of broader peaks near antinodes in LSCO is due, in a large part, to inhomogeneous doping. We find further evidence for the mechanism proposed here from another cuprate superconductor. Quantum oscillation

measurements showed that an overdoped  $\text{Ti}_2\text{Ba}_2\text{CuO}_{6+d}$  (Ti2201) had a much more homogenous chemical potential<sup>43</sup> because the dopants in Ti2201 were farther from the copper oxide plane than the dopants in LSCO and BSCCO.<sup>9</sup> Recent ARPES data on this material show that the spectral functions near the antinodes are even sharper than those at the nodes.<sup>44</sup> These results are consistent with the conclusion that the broader line shape near the antinode than near the node is not universal in the cuprates but is a material-dependent effect resulting from doping inhomogeneities.

Crystal momentum is generally a good quantum number for phonons and the electronic quasiparticles, whereas, the length scale of charge inhomogeneities in real space is only a few nanometers.<sup>11,14-17</sup> Thus, it may be surprising that the spectra of phonons and of the electronic quasiparticles are reproduced well by a linear sum of spectral peaks for each doping weighted by the inhomogeneous doping distribution function. For the electronic quasiparticles, a 70-meV FWHM intrinsic linewidth, combined with the measured Fermi velocity, translates into about a 3-nm mean-free path for the nodal direction, which is almost the same as the length scale of the doping variation from NQR (Ref. 17) and STM.<sup>16</sup> It is even shorter for the antinodal quasiparticles because they have a smaller Fermi velocity. Therefore, we can think that electronic quasiparticles reside in each patch and simply add all spectral peaks from corresponding patches with different dopings.

For the phonon, we do not need additional linewidth broadening to fit the experimental data as shown in Fig. 3. This, however, does not mean that the phonon has a much longer correlation length than the electronic quasiparticles. If the wavelength of any waves in an inhomogeneous medium is smaller than the length scale of the inhomogeneity of the medium, the waves tend to be localized inside a domain of the inhomogeneous medium.<sup>45-48</sup> The wavelength of zone-boundary phonons is two unit cells, which is about four times shorter than the length scale of the inhomogeneity measured by STM and NQR.<sup>16,17</sup> Thus, the distribution of the zone-boundary phonon frequencies in a medium with inhomogeneous force constants directly reflects the distribution of the force constants.

To summarize, we have found evidence that elementary excitations in LSCO, such as phonons and electronic quasiparticles, are strongly influenced by inhomogeneous doping previously reported by purely local probes, NQR, NMR, and STM. Phonon and electronic spectra are reproduced well by just averaging over spectra from patches having various doping concentrations, even though the typical length scale of a patch is a few nanometers. Good agreement between the model based on inhomogeneous doping with both ARPES and neutron results, despite almost no adjustable parameters, provides compelling evidence that inhomogeneous doping must be included into any analysis of experimental results in the cuprates. Recently, iron pnictide superconductors also have turned out to have chemical potential inhomogeneities due to a random dopant distribution.<sup>49</sup> Therefore, it is necessary to take account of these charge inhomogeneities when interpreting measurements on iron pnictide superconductors as well.

Inhomogeneity most likely originates from a random potential created by introduced dopants (in LSCO, it would be Sr), rather than from the self-organization of conduction electrons,

for example, into stripes. Yet, it is essential to gain further insight into interactions of dynamic stripes with phonons, which is at the focus of a different project. If stripes interact with phonons via a mechanism discussed in Refs. 36 and 50, then the effect of dynamic stripes should occur at a different wave vector,  $\mathbf{q} = (0.25, 0, 0)$ , not at the zone boundary. In fact, we find that the huge phonon linewidths reported near this wave vector in Ref. 36 cannot be reproduced by inhomogeneous doping alone for  $x \geq 0.07$ , and large additional broadening is required near optimal doping. These results will be presented in a subsequent publication.

## ACKNOWLEDGMENTS

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