



Experiment title:

Inelastic X-ray scattering study of the doping and temperature dependence of electron-phonon anomalies in $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$

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The 2012 discovery with synchrotron X-rays of charge-density-wave (CDW) correlations in competition with superconductivity in hole-doped $\text{YB}_2\text{Cu}_3\text{O}_{6+\delta}$ (YBCO) has attracted much attention [1,2]. Subsequent X-ray experiments demonstrated the existence of CDW correlations in other hole-doped cuprates [3-5] and in the archetypal electron-doped cuprate $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (NCCO) [6,7]. Charge correlations have long been argued to manifest themselves as an anomalous lineshape and dispersion of the longitudinal Cu-O bond-stretching phonon mode in the hole-doped cuprates [8,9]. In pioneering inelastic X-ray scattering (IXS) work at the ESRF, M. d'Astuto *et al.* revealed similar anomalous phonon behavior in nearly-optimally-doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (NCCO) [10]. The two-dimensional wave vector $(H,K) \approx (0.2,0)$ where the anomalous phonon softening was found is consistent with the recently observed CDW wave vector at the same doping level [7]. Our experiment aimed to extend the previous work of ref. [10] to a wider doping and temperature range, and to search for a connection between CDW correlations and phonon anomalies in NCCO.

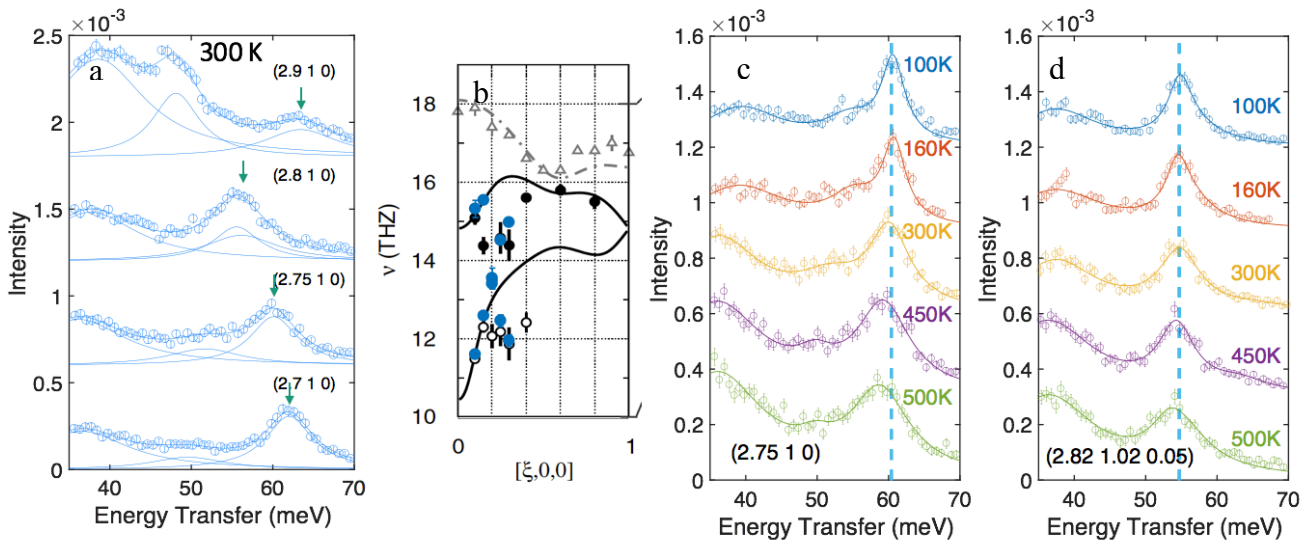


Figure1 Momentum and temperature dependence of phonons in nearly optimally doped NCCO ($x=0.145$). (a) IXS spectra of the high energy phonon modes at different momenta. Three phonon modes can be fit in the range 35 to 70 meV. The bond-stretching phonons are marked by blue arrows. (b) Dispersion of the two highest-energy modes at 300K (blue circles) are compared with the 20K prior data from ref. [10]. Temperature dependence of the phonon peaks around (c) the CDW wavevector $\mathbf{Q}_{\text{CDW}} = (2.75 \ 1 \ 0)$ and (d) phonon anomaly wavevector $\mathbf{Q}_{\text{ph}} = (2.82 \ 1.02 \ 0.05)$.

The samples investigated in the present study were pieces of the same crystals used for the recent resonant X-ray measurement of CDW correlations [7], with a-b planes parallel to a large 1 mm x 1mm surface, and polished down to 30 μm to maximize the transmission intensity. Energy transfer spectra in the range -5 to 90 meV were measured simultaneously via 8 analyzers, with the main analyzer set around \mathbf{Q}_{CDW} . In order to clearly observe the high-energy longitudinal phonon modes, we performed the measurements along $\mathbf{Q} = (3-q, 1, 0)$ with relatively long counting time.

Figure 1 shows the phonon anomaly for a nearly optimally-doped sample ($x = 0.145$). The anomalous softening at $q_{\text{ph}} \sim 0.2$ of the Cu-O bond stretching mode (Figure 1a, marked by blue arrows) reported at 20 K in ref. [10] is well reproduced at 300 K (Figure 1b). In order to search for a connection between the CDW and the phonon anomaly, we performed a comprehensive study of the temperature dependence of the phonon modes around $q_{\text{CDW}} \sim 0.25$ and $q_{\text{ph}} \sim 0.2$ (Figure 1 c and d). As for YBCO [11], if the phonon anomaly is associated with the CDW, one would expect that the soft phonon would gradually harden across T_{CDW} . However, as shown in Figure 1, the phonon energy remains unchanged between 100 K to 500 K. Given the pronounced CDW peak at the highest temperature (380 K) measured by resonant elastic X-ray scattering (REXS), it is possible that CDW correlations in NCCO are still robust even at 500 K.

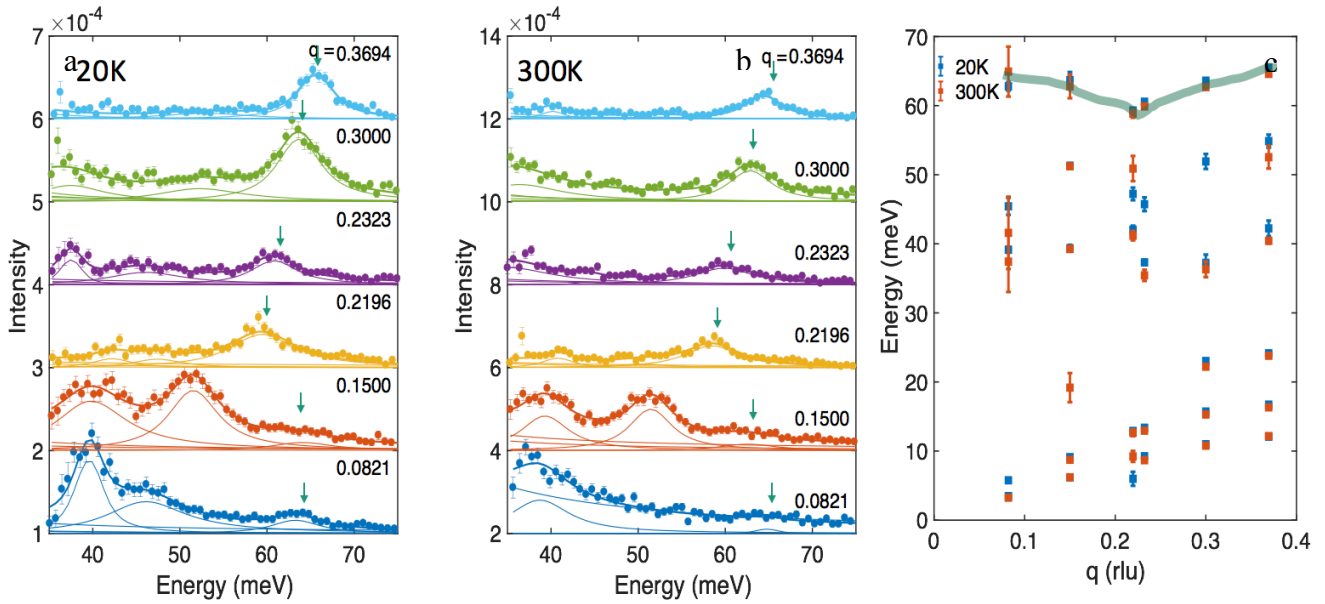


Figure 2. Momentum and temperature dependence of phonons in underdoped NCCO ($x=0.078$). IXS spectra of three high energy phonon modes at (a) 20 K and (b) 300 K. The arrows indicate the bond-stretching phonon modes. (c) Phonon dispersion at 20 K and 300 K. Solid line is guide to the eye.

We then decided to switch to a lightly-doped non-superconducting sample ($x=0.078$), for which only weak CDW peaks were detected at low temperatures, and no CDW correlations were observed at room temperature [7]. Interestingly, the phonon softening occurs in this doping as well and even persists at 300 K. Figure 2c shows that the phonon dispersions at 20 K and 300 K are nearly indistinguishable. This indicates that the phonon anomaly is not directly tied to CDW correlations in NCCO.

In summary, we performed an IXS study of bond-stretching phonons in two different samples of the archetypal electron-doped cuprate NCCO, for which distinct temperature-dependent CDW behaviors had been detected. Although both samples show phonon softening around $q_{\text{ph}} \sim 0.2$, close to the CDW wave vector $q_{\text{CDW}} \sim 0.25$, the temperature-insensitive behavior of the phonon anomaly indicate that this anomaly is not closely related to CDW correlations. The fact that $q_{\text{ph}} \approx q_{\text{CDW}}$ is, most probably, a mere coincidence. An ab-initio calculation will need to be performed in order to understand if the observed behavior is in fact anomalous, or if it is merely a regular crossing between two phonon branches.

References:

- [1] G. Ghiringhelli, *et al.*, Science **337**, 821 (2012)
- [2] J. Chang *et al.*, Nature Phys. **8**, 871 (2012)
- [3] R. Comin *et al.*, Science **343**, 390 (2014)
- [4] E. H. da Silva Neto *et al.*, Science **343**, 393 (2014)
- [5] W. Tabis *et al.*, Nat. Comm. **5**, 5875 (2014)
- [6] E. H. da Silva Neto *et al.* Science **347**, 282 (2015)
- [7] E. H. da Silva Neto *et al.*, Sci. Adv. **2**, e1600782 (2016)
- [8] D. Reznik *et al.*, Nature **440**, 1170 (2006)
- [9] D. Reznik, Physica C **481**, 75 (2012)
- [10] M. d'Astuto *et al.*, Phys. Rev. Lett. **88**, 167002 (2002)
- [11] M. Le Tacon *et al.*, Nat. Phys. **10**, 52 (2014)