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Experiment Report Form

	Experiment title: Phonon dispersion and electron-phonon coupling strength at the Cu L_3 resonance in the HTS oxychloride $Ca_{2-x}Na_xCuO_2Cl_2$	**Experiment number**: HC-3884
Beamline: ID32	**Date of experiment**: from: 08/11/2018 to: 13/11/2018	**Date of report**: 20/02/2020
Shifts: 15	**Local contact(s)**: Flora Yakhou-harris	*Received at ESRF:*

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Report: During this beamtime, we performed high energy resolution resonant inelastic X-ray scattering (RIXS) measurements at the Cu L_3 edge on copper oxychloride $Ca_{2-x}Na_xCuO_2Cl_2$ (Na-CCOC). The purpose of the experiment was to map out the momentum-dependent electron-phonon coupling in Na-CCOC, and more particularly to focus on the underdoped sample (x=0.1) in where a CDW was expected.

The scattering angle of the endstation was fixed to $2\theta = 149.5°$, to maximize the momentum-transfer for the momentum-dependence measurements and different $Q_{//}$ were obtained by rotating the sample around the b-axis. We have set the energy resolution to ~ 50 meV, in order to clearly resolve the phonon excitations, with reasonable data acquisition efficiency (~ 1-2 hours per spectrum). The incident beam was polarized with linear vertical polarization (σ-polarization). We have obtained high quality data on the underdoped sample along both (0,0)-(h,0) and (0,0)-(-h,0) directions at 25 K. In addition, we also performed temperature and doping dependence measurements, by measuring at 245 K the underdoped sample and also compared our results with measurements on the undoped one.

Figure 1a shows the RIXS intensity map recorded at 25 K and on-resonance on the underdoped sample. A close examination of the RIXS intensity integrated over a small energy window around zero-energy loss is

shown in Fig. 1b. A symmetric peak is observed that can been fitted by a Lorentzian. This peak unambiguously demonstrates the existence of CDW in underdoped Na-CCOC, as previously reported by STS [1]. Upon increasing the temperature to 245 K, the CDW was no longer observed in the quasi-elastic energy region, as shown on Fig. 1 c and Fig. 1 d.

As shown on Fig. 1 e, the RIXS data at 25 K show an excitation centered at ~ 60 meV, followed by a background at higher energy originating from the paramagnon. The excitation's energy and dispersion (see Fig. 1a) agrees well with that of the Cu-O bond stretching (BS) phonon as found in the literature [2]. Importantly, the phonon dispersion gradually softens to reach a minimum near Q_{CDW}. In addition, a broadening of the peak width occurs near Q_{CDW}. These observations suggest that the CDW affects the phonon and that the electron-phonon coupling is modified in the proximity of the CDW. More surprisingly, the phonon dispersion at 245 K also softens and reaches a minimum near Q_{CDW} (Fig. 1 c), despite no clear observation of a static CDW at this temperature (Fig. 1 d). This suggest that some charge correlations persist at high temperature that still affect the lattice excitations.

Our comparison to the undoped $Ca_2CuO_2Cl_2$ sample (CCOC) at 25 K is shown on Fig. 1 f, which presents the energy-momentum intensity map of the CCOC sample with the fitted elastic peak subtracted. As expected, a good agreement is obtained by comparing the dispersion of the phonon measured by RIXS (black diamonds) and the dispersion of the Cu-O BS phonon measured by IXS on another sample having similar doping (red triangles) [3]. The Cu-O BS neither softens nor broadens in CCOC, confirming the absence of CDW modulations in the undoped compound.

In summary we have obtained interesting results in Na-CCOC consistent with the framework of the initial proposal. The direct demonstration of the existence of bulk CDW in this system along with a strong modification of the dispersion of BS phonon in the proximity of the CDW are important results, deserving to be published. Therefore, we have finished the data analysis and are working on a manuscript for publication.

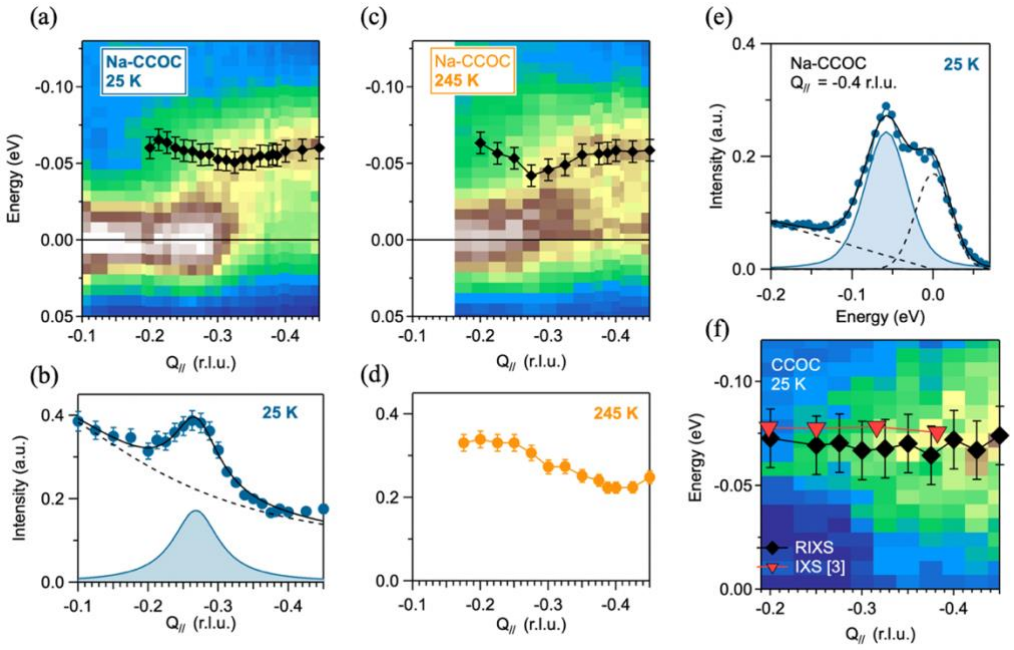

Figure1 : (**a**) RIXS intensity map recorded at 25 K as function of the energy loss and $Q_{//}$. The phonon dispersion is shown with black diamonds. (**b**) Averaged quasi-elastic RIXS intensity at 25 K. The fit of the CDW peak is highlighted is blue. The data taken at 245 K are shown in (**c**) and (**d**) with the same plotting format and notations used in panels (**a**) and (**b**). (**e**) Energy-loss spectrum at a representative momentum at 25 K (demonstrating the quality of the data), the BS-phonon is highlighted in blue. (**f**) Elastic peak-subtracted intensity map of the undoped CCOC sample at 25 K. The dispersion of the phonon extracted from the RIXS data is displayed with black diamonds. The red triangles show the dispersion of the Cu-O bond-stretching phonon from IXS measurements [3].

References:

[1] Hanaguri, T. *et al.* A 'checkerboard' electronic crystal state in lightly hole-doped $Ca_{2-x}Na_xCuO_2Cl_2$. *Nature* **430**, 1001–1005 (2004).

[2] Reznik, D. *et al.* Electron–phonon coupling reflecting dynamic charge inhomogeneity in copper oxide superconductors. *Nature* **440**, 1170–1173 (2006) ; Graf, J. *et al.* Bond Stretching Phonon Softening and Kinks in the Angle-Resolved Photoemission Spectra of Optimally Doped $Bi_2Sr_{1.6}La_{0.4}Cu_2O_{6+\delta}$ Superconductors. *Phys. Rev. Lett.* **100**, 227002 (2008).

[3] Lebert, B. W. *et al.*, Doping-induced in-plane anisotropy of bond-stretching phonon softening in oxychloride Ca2−xCuO2Cl2 compounds. *Phys. Rev. B* **101**, 020506(R) (2020).