ESRF	**Experiment title:** Inelastic x-ray scattering study of the lattice dynamics of Ca_2RuO_4	**Experiment number**:
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Shifts:	**Local contact(s)**: S.M. Souliou	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Sofia-Michaela Souliou* (ESRF)
Mathieu Le Tacon* (Max Planck Institute for Solid State Research)
GRETARSSON Hlynur* (Max Planck Institute for Solid State Research)

Report:

In the current beamtime we studied the lattice dynamics of the Mott insulator Ca_2RuO_4 by means of inelastic x-ray scattering. In addition to exploring the temperature and momentum dependence of the low energy acoustic and optical phonon modes for the first time, the purpose of the experiment was to investigate the coupling of the lattice and spin degrees of freedom across the magnetic ordering at T_N=110 K. Our Raman scattering experiments on the same system have identified pronounced Fano lineshape asymmetries of the phonon modes overlapping with magnetic amplitude mode for T < T_N [1]. Earlier inelastic neutron scattering experiments have mapped the dispersions of the phase (magnon) and amplitude (Higgs) spin excitations allowing us to monitor their crossing points with the phonon dispersions [2].

For our measurements we used high quality single crystals of Ca_2RuO_4 grown by the floating zone method and used for the earlier Raman and neutron scattering experiments. The crystals were manually thinned to needles of ~100 μm, matching the absorption length of the x-rays at the used incident energy (E_{inc}~17.7 keV with a resolution ΔE~3 meV). The needle axis was perpendicular to the crystallographic *c*-axis. The measurements were performed in the (H K 0) scattering plane. The low energy acoustic phonon dispersions were measured at the Brillouin zone attached to the strong (0 2 0) Bragg reflection. In order to optimize the structure factor for higher energy phonon modes crossing the magnon and Higgs modes dispersions we were guided by the results of DFT calculations. We collected inelastic spectra at room temperature, at 120 K (which is above the magnetic transition temperature T_N=110 K) and at base temperature 4.3 K using the closed circle helium cryostat available in ID28.

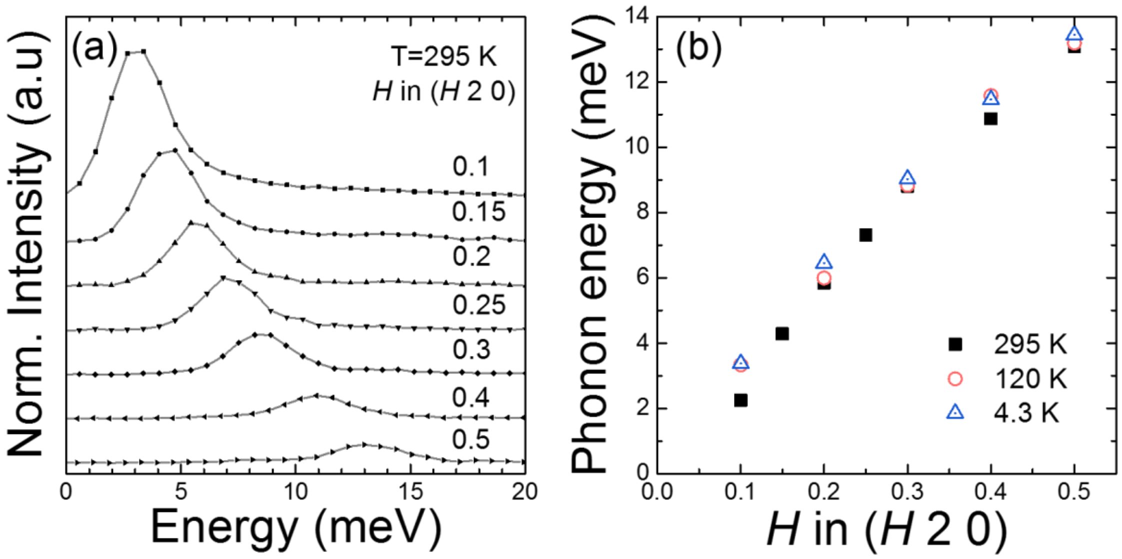

Fig.1: (a) Inelastic part of the IXS spectra recorded along the [100] direction at room temperature. (b) Temperature dependence of the transverse acoustic phonon energy.

Fig.1(a) shows the inelastic part of the spectra up to 20 meV recorded in transverse geometry at ambient conditions. In agreement with our structure factor calculations, we observe only the transverse acoustic phonon mode. The inelastic data were fitted with damped harmonic oscillator functions, the results of which are summarized in Fig.1(b) for all the measured temperatures. Cooling results in the expecting continuous phonon hardening. The hardening is larger between room temperature and 120 K than between 120 K and 4.3 K, in agreement with the reported T dependence of the lattice parameters which change only slightly below T_N [3].

Fig.2(a) shows the spectra recorded close to the Brillouin zone attached to the (140) Bragg reflection, where in agreement with our calculations 4 phonon modes are seen. The closed symbols correspond to the data taken at base temperature and the open symbols to the ones taken at 120 K. The black (red) ticks mark the energies where the magnon (Higgs) mode was observed by neutron scattering [2]. We observe no clear change in the optical phonons dispersions at their crossing points with the spin excitations, other than the expected hardening.

The temperature dependence of the phonon spectra at momentum transfer Q=(1.2 4 0), for which the phase and amplitude magnetic excitations cross the optical phonons at ~21 and 36 meV respectively (see Fig.2(a)), are given in Fig.2(b). Interestingly, we observe some peculiarities:

(1) as the vertical lines indicate the phonon energies do not follow a monotonic change as the sample is cooled to base temperature,

(2) the phonons modes at ~21 and 36 meV show a strong intensity damping at 43 K and 35 K respectively.

Unfortunately the available beamtime was not sufficient to explore the temperature dependence in detail. Future measurements will be needed to clarify this intriguing behavior.

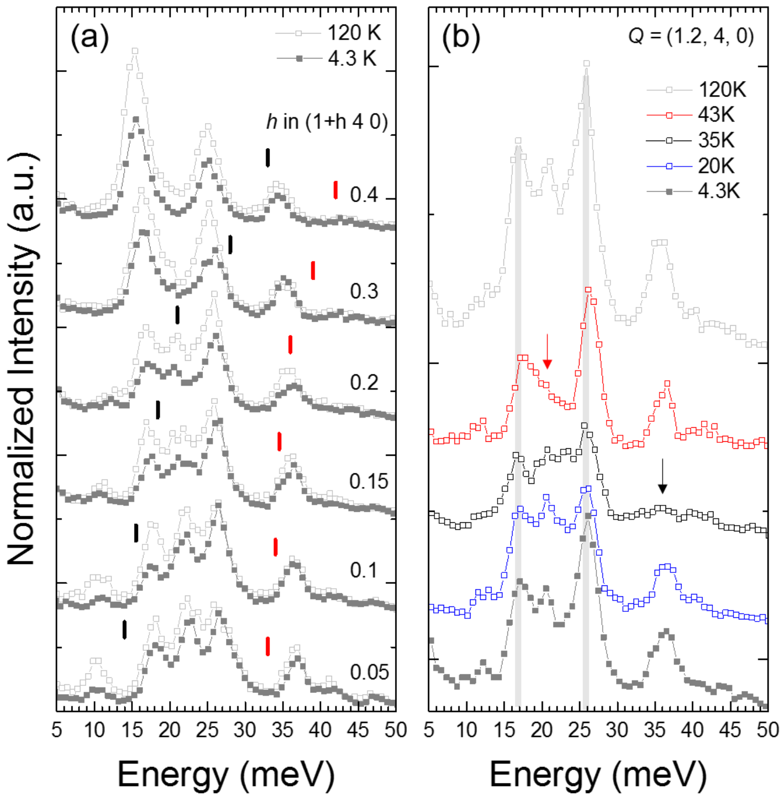

Fig.2: (a) Inelastic part of the IXS spectra recorded along the [100] direction close to the Brillouin zone attached to the (140) Bragg reflection at 120 K (open symbols) and at base temperature (closed symbols). The vertical black (red) ticks indicate the positions of the magnon (Higgs) modes [2]. (b) Temperature dependence of the IXS spectrum at Q= (1.2,4,0).

References:

[1] S.M. Souliou et al., PRL 119, 067201 (2017)
[2] A. Jain et al., Nat. Phys. 13, 633 (2017)
[3] M. Braden et al., PRB 58, 847 (1998)