ESRF	**Experiment title:** Spin-phonon interaction in strong spin-orbit coupling Mott insulating bilayer iridate $Sr_3Ir_2O_7$	**Experiment number**: HC-2809
Beamline: ID28	**Date of experiment**: from: 28 Sep 2016 to: 05 Oct 2016	**Date of report**: 31 Aug 2017
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We investigated the lattice dynamics of the bilayer iridate $Sr_3Ir_2O_7$ with high-energy resolution inelastic x-ray scattering in our beam time on ID28. This intriguing system has a spin-orbit entangled ground state and j_{eff}=1/2 magnetic moments (pseudospins) sitting on a square lattice. Our recent Raman results have revealed pronounced Fano lineshapes of the low-energy optical phonons for $T > T_N$, reflecting strong coupling to pseudospin-fluctuations [1]. This discovery of strong coupling between lattice and magnetic degrees of freedom indicates unquenched orbital dynamics in the paramagnetic state of $Sr_3Ir_2O_7$ which, albeit apparently generic, have so far not been considered in the general description of insulating and metallic iridates and other *5d*-electron materials.

In our measurements we employed the Si(9,9,9) reflection of the high-resolution monochromator resulting in a resolution of 2.8 meV (full width at half maximum). The sample temperature was controlled by a nitrogen cryo-blower with accessible temperatures ranging from 100 K up to 500 K. The sample was a small single crystal having a thickness of about 0.1 mm (along the long, out-of-plane *c* axis of the orthorhombic unit cell). The edges in the a-b plane were about 1 mm long. The crystal was mounted in two different geometries: we measured the acoustic phonon modes with polarization patterns in the a-b plane of $Sr_3Ir_2O_7$ in transmission geometry in the Brillouin zone adjacent to the reciprocal lattice vector $\boldsymbol{\tau}$ = (4, 0, 0). The Raman-active modes observed in Ref. [1] have a significant out-of-plane component and, hence, need to be measured with a large *L* component in the scattering wavevector. Therefore, we mounted the sample in reflection geometry and measured phonons at $\boldsymbol{Q}$ = (x,0,17), where our ab-initio calculations predicted the largest structure factors for the Raman-active phonons.

Figure 1 shows raw data for the (left) transverse acoustic (TA) phonons at $\boldsymbol{Q}$ = (4, x, 0) and (right) the transverse optic (TO) modes at $\boldsymbol{Q}$ = (x, 0, 17) at temperatures above the Néel temperature T_N = 285 K. In the measurements at $\boldsymbol{Q}$ = (4, x, 0), we observe only the TA phonon mode in agreement with our structure factor calculations. In reflection, we find several peaks in the measured range of energy transfers, -5 meV $\leq E \leq$ 30 meV, which makes it more difficult to distinguish different phonon modes. All spectra were fitted by assuming a resolution limited lineshape for the elastic line. The peaks at finite energy transfers were fitted using damped harmonic oscillator (DHO) function which simultaneously approximate the peaks at the same absolute energy transfers in phonon creation (E > 0) and annihilation (E < 0) by a single function. The resulting fit functions are shown as solid (red) lines in Figure 1.

The observed dispersions of the LA (squares) and TA branches (dots/circles) are shown in Figure 2 for two temperatures, T = 100 K (open symbols) and 350 K (filled symbols), i.e., the minimum and maximum temperatures in our experiment. While the overall dispersions look rather normal, the temperature dependence reveals some peculiarities:

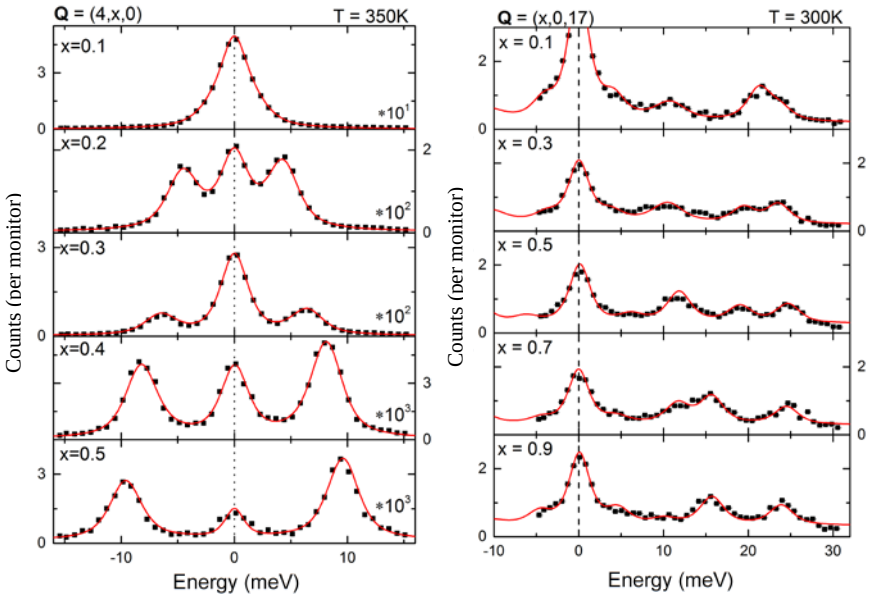

Figure 1: Raw data from inelastic x-ray scattering obtained at (left) $\boldsymbol{Q}$ = (4,x,0), $0.1 \leq x \leq 0.5$, and (right) $\boldsymbol{Q}$ = (x,0,17), $0.1 \leq x \leq 0.9$. Red lines are fits consisting of a resolution limited elastic line and damped harmonic oscillator (DHO) functions for peaks at finite energy transfers. The data taken at $\boldsymbol{Q}$ = (4, x, 0) are scaled by the factors indicated in the lower right corners of the individual panels in order to increase the visbility of the peaks at larger values of x.

(1) The longitudinal mode hardens on heating.
(2) The transverse mode softens significantly only for $x \leq 0.3$.

Regarding the LA mode, a hardening on heating is clearly anomalous. Conventionally, the thermal expansion and atomic motions at higher temperatures lead to an overall softening of phonon modes. The hardening is present over the complete wavevector range $0.05 \leq x \leq 0.5$. The detailed temperature dependence (not shown) shows that the hardening is already present at T = 200 K, increases on further heating and does not show a particular response on crossing T_N = 285 K. In contrast, the TA branch shows the generally expected softening but only for $x \leq 0.3$. Indeed, the softening increases quantitatively on reducing x, similar to the behavior of a soft phonon mode at a structural phase transition with ordering wavevector $\boldsymbol{q}$ = 0. Finally, we note that we did not observe any broadning of the acoustic phonons within the experimental error of the experiment.

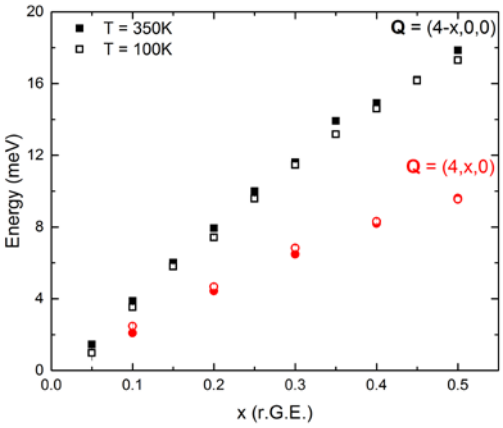

Figure 2: Dispersion of the longitudinal (squares) and transverse (circles/dots) acoustic phonon modes propagating along the [100] direction measured in the Brillouin zone adjacent to $\boldsymbol{\tau}$ = (4, 0, 0) at temperatures T = 100 K (open symbols) and 350 K (filled symbols).

We selected the wavevectors $\boldsymbol{Q}$ = (x, 0, 17) in order to be sensitive to the Raman-active mode exhibiting the anomalous broadening at $\boldsymbol{q}$ = 0 corresponding to x = 0 in our measurements. Unfortunately, the interesting peak in the range of 20 meV $\leq E \leq$ 25 meV at x = 0.1 indicates the presence of two modes and the determination of the linewidth of the inidvidual modes is not possible. At large x = 0.9, we find a single peak near 24 meV. This peak shows no broadening within the experimental error between T = 100 K and 300 K. We note, however, the reported broadening of the A_{1g} Raman-active phonon is in the order of 1 meV (FWHM) and our resolution (2.8 meV) might have been not good enough to resolve such a feature in phonon with a comparatively weak structure factor.

[1] H. Gretarsson et al., Physical Review Letters **116**, 136401 (2016).