

Phonon Excitations in $\text{Eu}_2\text{Ir}_2\text{O}_7$ Probed by Inelastic X-ray Scattering

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1. Introduction

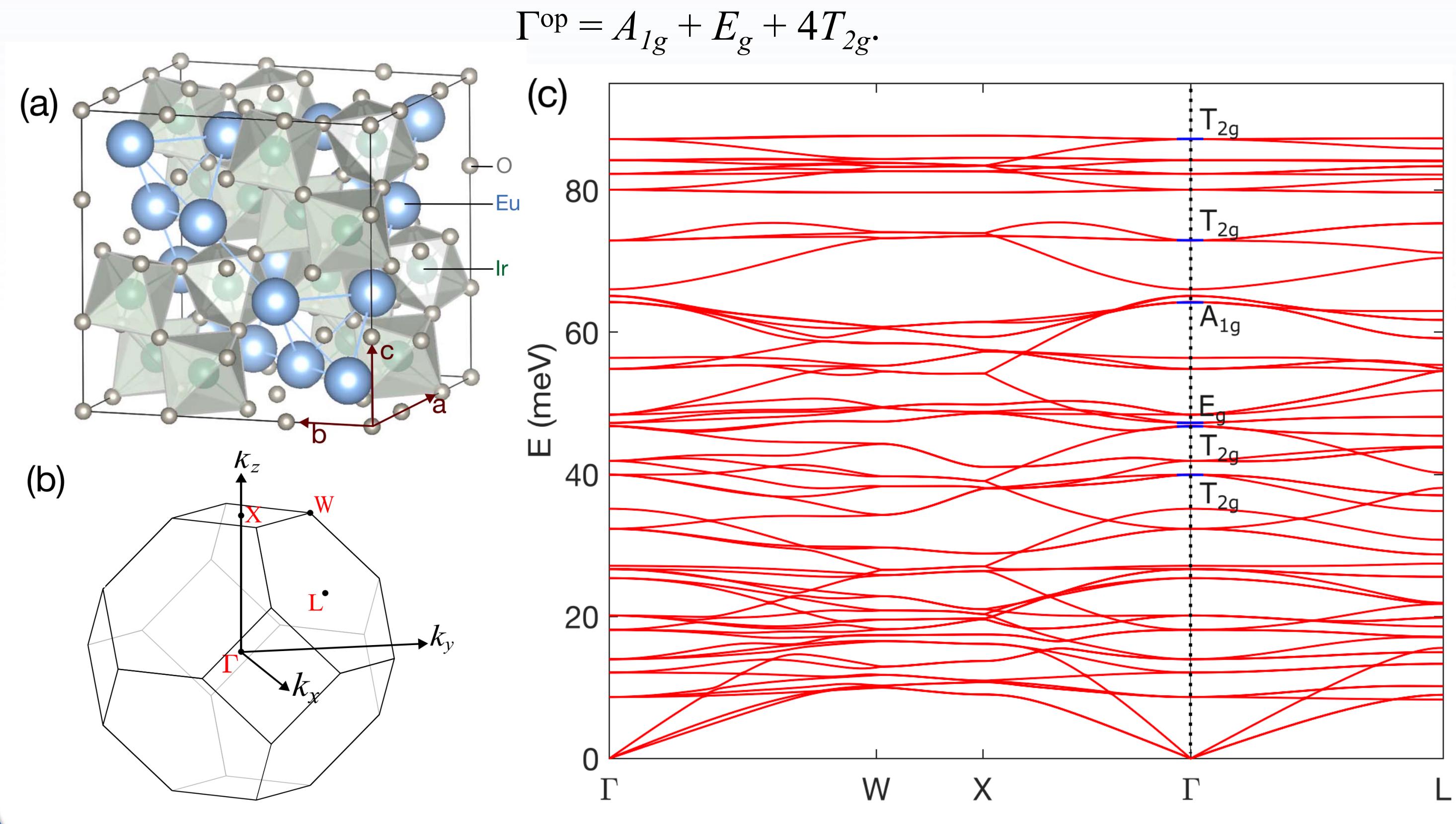
The study of phonon dynamics and its interplay with magnetic ordering is crucial for understanding the unique quantum phases in the pyrochlore iridates. Here, through inelastic X-ray scattering on a single crystal sample of the pyrochlore iridate $\text{Eu}_2\text{Ir}_2\text{O}_7$, we map out the phonon excitation spectra in $\text{Eu}_2\text{Ir}_2\text{O}_7$ and compare them with the theoretical phonon spectra calculated using the density functional theory. Possible phonon renormalization across the magnetic long-range order transition is observed in our experiments, which is consistent with the results of the previous Raman scattering experiments.

2. Methods

- A piece of KF-flux grown $\text{Eu}_2\text{Ir}_2\text{O}_7$ single crystal, with a dimension of $0.4 \times 0.4 \times 0.1$ mm³ were used for IXS experiments on the RIKEN Quantum NanoDynamics Beamline, BL43LXU, at the SPring-8 synchrotron light source in Japan.
- The polished (111) crystal surface was aligned, using a 24-analyzer array for parallel momentum transfer measurements with a Si (11,11,11) setup and a resolution of $\gtrsim 1.3$ meV.
- Theoretical phonon spectra for $\text{Eu}_2\text{Ir}_2\text{O}_7$ were calculated using the first-principles DFT as implemented in the Vienna ab initio simulation package (VASP).
- Force constants were calculated by density functional perturbation theory as implemented in VASP. Phonon dispersion relations were obtained using the PHONONPY software in a primitive unit cell.

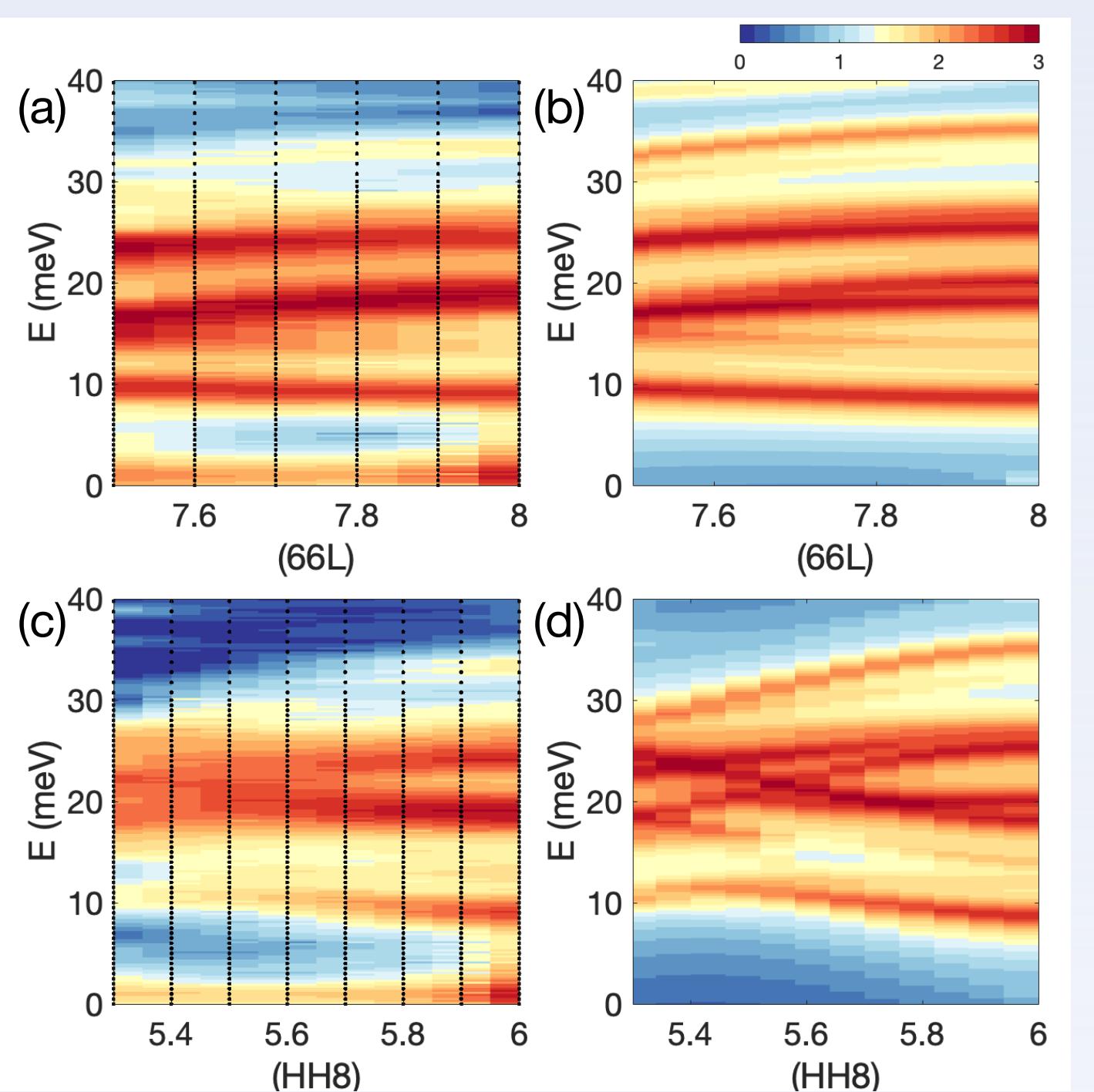
3. Crystal structure and phonon dispersion

- In $\text{Eu}_2\text{Ir}_2\text{O}_7$ (space group $Fd\bar{3}m$), the Ir⁴⁺ ions form a pyrochlore lattice with corner-sharing tetrahedra.
- Altogether 66 phonon bands are observed in our DFT calculations, which is consistent with the number of phonon modes for 22 atoms per primary unit cell.
- At the Γ point, there are 6 Raman-active phonon bands according to group-theoretical analysis,

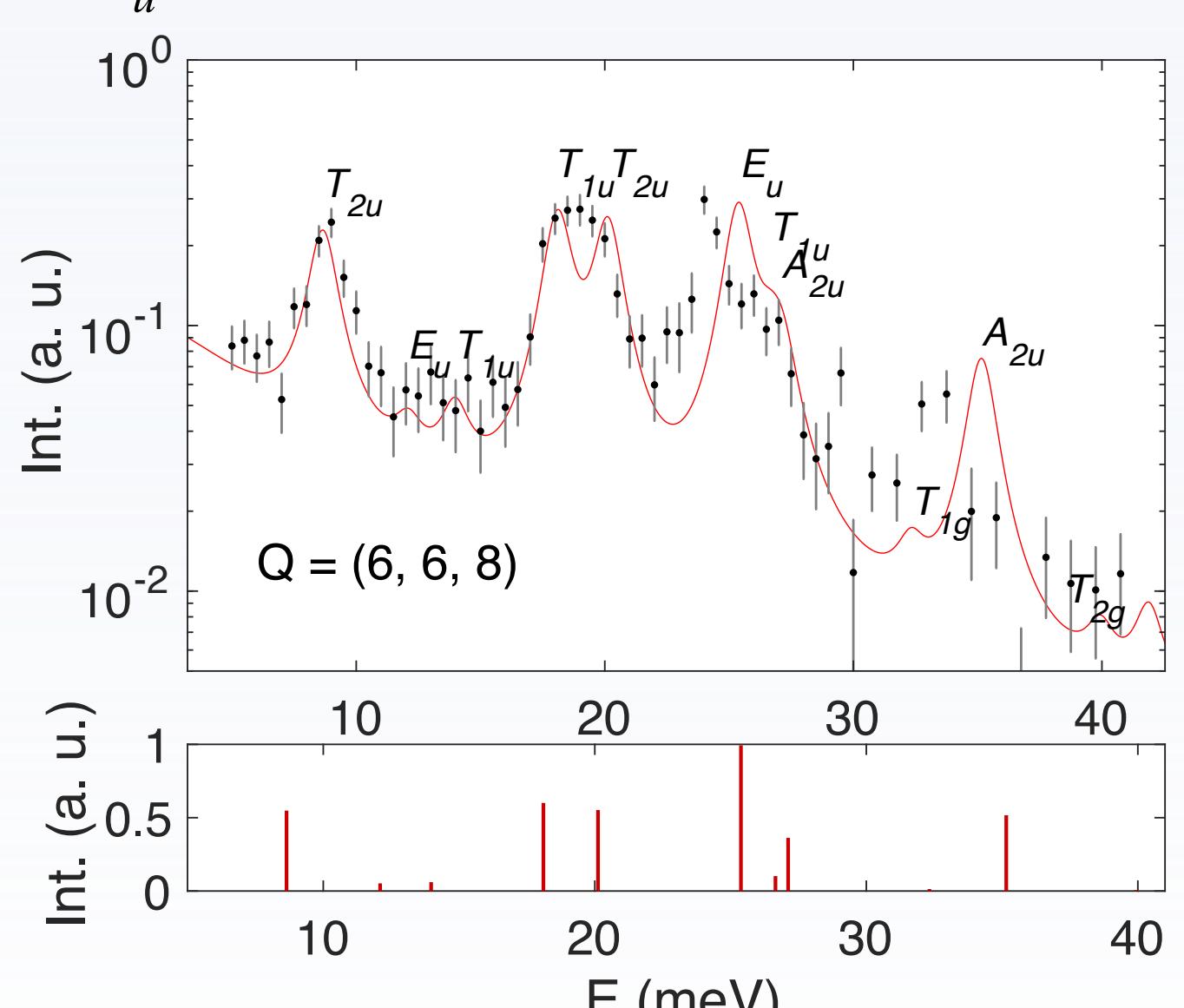


4. Experimental and calculated spectra

- The comparison of the experimental and calculated IXS cross sections along the (66L) and (HH8) directions around (668) r.l.u. in reciprocal space.



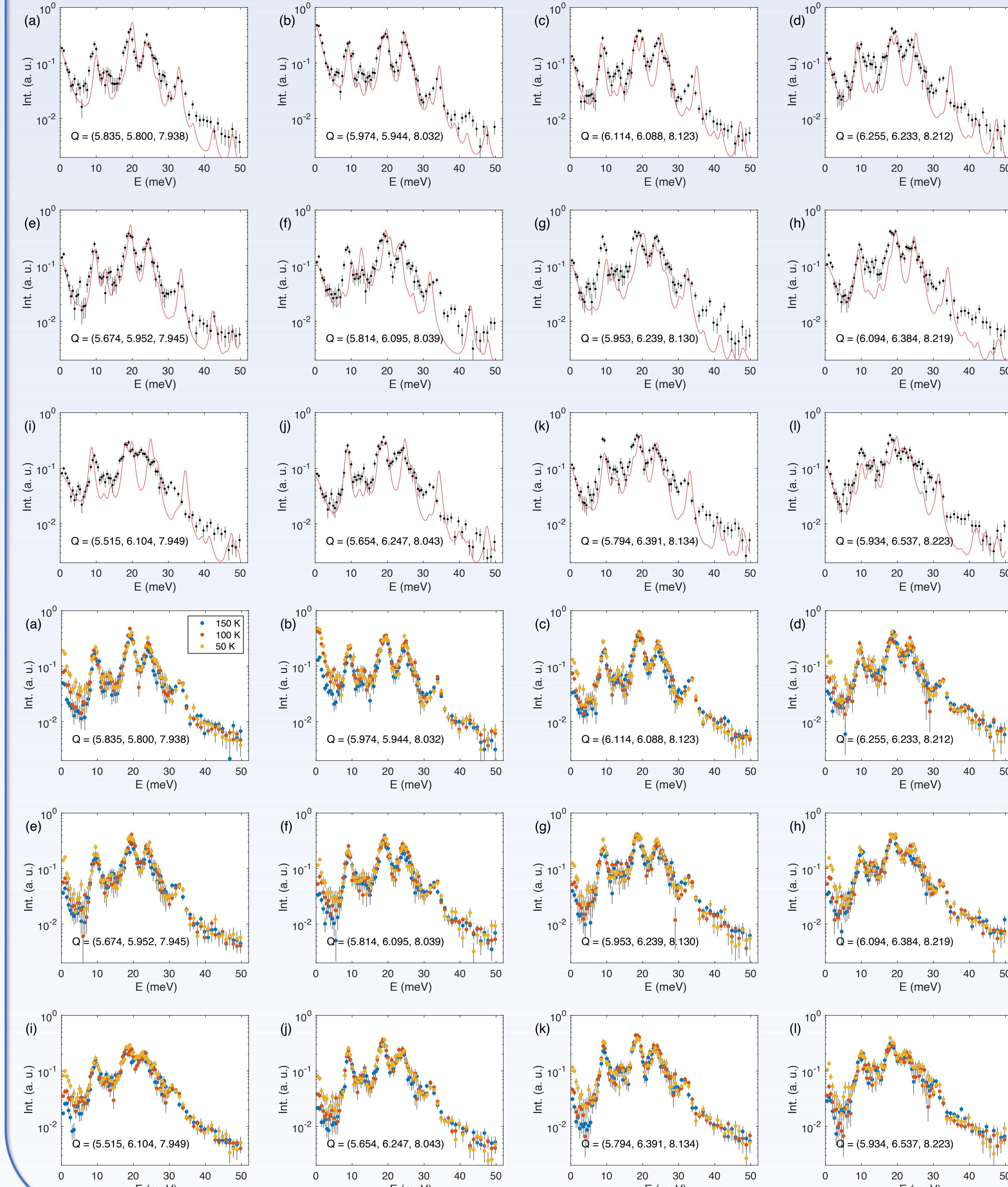
- Experimental (black dots) and calculated (red line) phonon spectra at momentum transfer $Q = (6, 6, 8)$ measured at $T = 50$ K. The calculated IXS cross section is convoluted by a pseudo-Voigt instrumental resolution function.
- The height of the vertical bars corresponding to the calculated IXS cross sections are normalized by the cross section of the E_u mode at $E = 25.4$ meV.



7. Conclusion & Acknowledgements

- In conclusion, phonon excitations of the pyrochlore iridate $\text{Eu}_2\text{Ir}_2\text{O}_7$ have been investigated using the non-resonant IXS technique.
- Comparisons to the DFT calculations reveal that the cross sections are dominated by the Raman-inactive modes in an energy transfer range of [10, 30] meV.
- Analysis of the IXS spectra reveals possible phonon renormalizations across the longrange-order transition as reported in the previous Raman scattering experiments.
- Works at the USTC was supported by the National Natural Science foundation of China (NSFC) (Grant No. 12374152).

5. The IXS spectra collected at 12 representative Q positions



- Representative IXS spectra of $\text{Eu}_2\text{Ir}_2\text{O}_7$ measured at $T = 50$ K with the target detector fixed at $Q = (6.114, 6.088, 8.123)$ r.l.u. Solid red lines represent the calculated phonon cross section co-convolved by a pseudo-Voigt instrumental resolution function.
- Temperature-dependence of the IXS spectra for $\text{Eu}_2\text{Ir}_2\text{O}_7$ measured at $T = 50$ K, 100 K, and 150 K.
- No strong variation is observed for the main phonon modes in the energy range of [10, 30] meV, which confirms the absence of structural distortion at $T_N \sim 115$ K as revealed in the neutron and x-ray diffraction experiments

6. Possible phonon renormalization

- Focusing at $Q = (6.114, 6.088, 8.123)$ r.l.u., the weak peak observed at $E = 38.8$ meV at $T = 50$ K gradually shifts to $E = 42.8$ meV at $T = 150$ K, leading to a double peak feature at $T = 100$ K, corresponding to the previous Raman scattering experiments [PhysRevB.100.115157].
- The energy shift of the weak peak can be ascribed to the renormalization of the E_g mode. It can be explained by the magnetoelastic effect.

