Lattice dynamics of $Cs_3Cu_2I_5$ scintillator investigated by temperature-dependent inelastic neutron scattering

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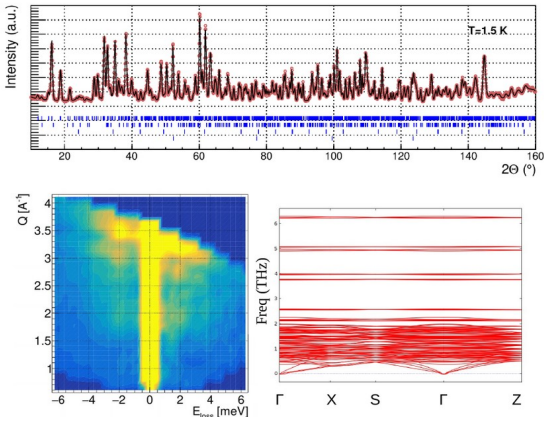
Recently, the charged-particle induced scintillation of a metal halide perovskite, $Cs_3Cu_2I_5$ (CCI325), as polycrystalline thin (~50-150 μm) layers, was investigated by irradiation with accelerated particle beams. [1]. CCI325 was found to have a high particle-induced luminescence (pRL) light yield along with quasi-linear spectroscopic response with respect to conventional activator-doped scintillators. Very recently, we measured the temperature dependence of the photoluminesence (PL) and pRL yields for CCI325 down to T=4.2 K and a striking difference between the two yields was observed. This finding could potentially be explained by the difference in ionization densities and the subsequent, phonon-assisted hot carrier diffusion and cooling. Our analysis on the temperature dependent PL yield within the Huang-Rhys theory suggested the phonon-assisted nature of self-trapped exciton (STE) dynamics. Besides, we observed an abrupt variation in the PL/RL yields of near-band and deep excitons indicating a possible structural transition at cryogenic temperatures.

The concept of STE generation in CCI325 was recently studied by Chen [2] who evaluated the crystal lattice rigidity by measuring the dynamical structure factor S(Q,E) of CCI325 via inelastic neutron scattering (INS). The phonon spectrum of CCI325 was measured up to ~30 meV, though with a moderate resolution manifesting as a broad continuum below 10 meV. Their finding is consistent with multiple self-trapped emissions due to the ultra-soft 0D structure. However, no experimental information exist on the temperature dependence, which could be the key to explain the observed temperature dependent PL and RL yields through the concept of phonon-assisted STE generation and carrier cooling and diffusion.



In the present work, we study the phonon density-of-states (PDOS) in polycrystalline CCI325 and relate it to changes in exciton generation. We investigated the structural evolution of CCI325 by temperature dependent neutron powder diffraction and the temperature dependence of the dynamics (PDOS) by INS (T=1.5K–270K) to explore the thermally activated motions and phonon modes. We also performed first principles (DFT) calculations on the phonon dispersion and PDOS to interpret the measured INS data.

1. M. Hunyadi *et al.*: "Scintillator of polycrystalline perovskites for high-sensitivity detection of charged-particle radiations", Adv. Funct. Mater., **32**, 2206645 (2022).
2. H. Chen *et al.*, "Multiple Self-Trapped Emissions in the Lead-Free Halide $Cs_3Cu_2I_5$", J. Phys. Chem. Lett. **11** (2020) 4326.