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Inelastic Neutron Scattering on Colloidal Nanocrystal Solids: Understanding the Importance of Surfaces

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Colloidally synthesized nanocrystals (NCs) are being developed for a large number of energy-related devices, including solar cells, thermoelectrics, and batteries. To function in solid-state devices, these solution-processed NCs are assembled into 3D superlattice structures, known as NC-solids. While phonon density-of-states (g(ω)) have been measured and calculated in bulk crystalline semiconductors, phonons remain poorly understood in nanomaterials, despite the critical importance of phonons in determining how energy is gained, lost, and transported in materials.

I will present the first experimental investigation of the g(ω) of NC-solids carried out via Inelastic Neutron Scattering (INS) [1]. We determined the g(ω) of lead sulphide (PbS) NC-solids, explained the origins of their differences with the g(ω) of bulk PbS, and used our findings to gain insight into charge carrier recombination in PbS NC solar cells.

Specifically, we measured the g(ω) as a function of PbS NC size, and found low and high frequency phonon modes with large thermal displacements in nanosized PbS that are not present in bulk PbS. The experimental results were complemented with ab initio molecular dynamics (AIMD) simulations of the NCs, which showed that these modes come from the NC surface. The partial g(ω) in the core of the NCs remains as in the bulk. Using thermal admittance spectroscopy (TAS) measurements on a large number of PbS NC-based diodes, we measured the energy and rate of the electronic transitions. With the theoretical framework of Multi-Excitation Entropy, we could explain the unusually high transition rates by coupling of the quantum-confined electronic states of the NCs to the large displacement phonon-modes at the NC-surface.

To reduce the electron-phonon coupling and decrease charge recombination dynamics, we look toward the surface molecules (known as ligands) that are primarily used to tune the electric conductivity of NC-solids. We demonstrate that, contrary to previous thinking that certain ligands electronically passivate the nanocrystral and remove mid-gap electronic traps states, these best performing ligands (1) mechanically strengthen the surface of the NCs, dampening out high displacement surface modes, and (2) confine the electronic wavefunctions, decreasing their spatial overlap with the surface phonon modes.

References:

[1] D Bozyigit, N Yazdani, M Yarema, O Yarema, W M M Lin, S Volk, K Vuttivorakulchai, M Luisier, F Juranyi & V Wood, Soft surfaces of nanomaterials enable strong phonon interactions. Nature (2016) doi: 10.1038/nature16977

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