Department of Chemistry and Biochemistry
Physical Chemistry Seminar Series

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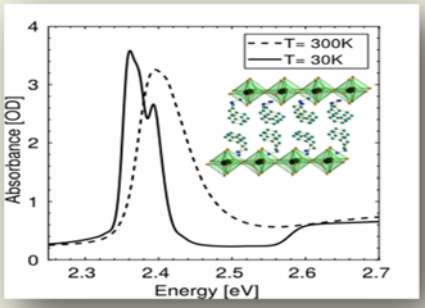
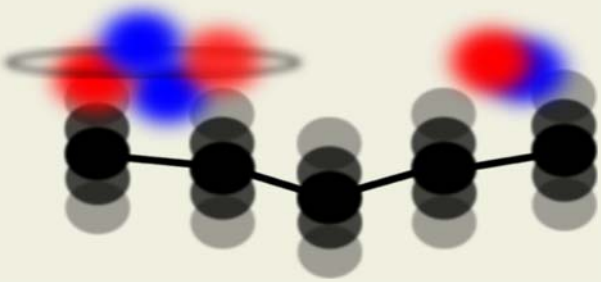
Monday, February 26 ~ 2:00 pm 331 Klamath Hall

Excitons, biexcitons, and the role of dynamic disorder in two - dimensional lead-halide perovskitoids

Owing to both electronic and dielectric confinement effects, two-dimensional organic-inorganic hybrid perovskites sustain strongly bound excitons at room temperature. In this presentation, we demonstrate that there are non-negligible contributions to the excitonic correlations from the peculiar lattice structure and its polar fluctuations, both of which are controlled via the chemical nature of the organic counter-cation. We present a phenomenological, yet quantitative framework to simulate excitonic absorption lineshapes in single-layer organic-inorganic hybrid perovskites, based on the two-dimensional Wannier formalism. We include four distinct excitonic states separated by 35 ± 5 meV, and additional vibronic progressions. Intriguingly, the associated Huang-Rhys factors and the relevant phonon energies show substantial variance with temperature and the choice of the organic cation. This points to the hybrid nature of the lineshape, with a form well described by a Wannier formalism, but with signatures of strong coupling to localized vibrations, and possible polaronic effects. This complex spectral structure depends strongly on crystalline distortion induced by the interlayer organic cation. By means of two-dimensional coherent spectroscopy, we examine excitonic many-body effects in these materials. We determine the binding energy of biexcitons — correlated two-electron, two-hole quasiparticles — to be 44 ± 5 meV at room temperature. The extraordinarily high values are similar to those reported in other strongly excitonic two-dimensional materials such as transition-metal dichalchogenides. Importantly, we show that this binding energy increases by ~ 25% upon cooling to 5 K. Our work highlights the importance of multi-exciton correlations in this class of technologically promising, solution-processable materials, in spite of the strong effects of lattice fluctuations and dynamic disorder.





Refreshments served at 1:45 pm 331 Klamath Hall

Hosted by Andy Marcus