Abstract Submitted
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**Exploring the nature of low-lying excited-states in molecular crystals from many-body perturbation theory beyond the Tamm-Dancoff Approximation**[1] TONATIUH RANGEL, Lawrence Berkeley Natl Lab; UC Berkeley, SAHAR SHARIFZADEH, Boston University, ANDRE RINN, Philipps-Universitt Marburg, FELIPE H. DA JORNADA, UC Berkeley; Lawrence Berkeley Natl Lab, MEIYUE SHAO, Lawrence Berkeley Natl Lab, GREGOR WITTE, Philipps-Universitt Marburg, CHAO YANG, Lawrence Berkeley Natl Lab, STEVEN G. LOUIE, UC Berkeley; Lawrence Berkeley Natl Lab, SANGAAM CHATTERJEE, Philipps-Universitt Marburg, LEEOR KRONIK, Weizmann Institute of Science, JEFFREY B. NEATON, Lawrence Berkeley Natl Lab; UC Berkeley — Organic semiconductors have attracted attention due to their potential for optoelectronics and novel phenomena, such as singlet fission. Here, we use many-body perturbation theory to simulate neutral excitations in acene and perylene crystals. By diagonalizing the full Bethe-Salpether (BSE) Hamiltonian beyond the Tamm Dancoff approximation (TDA) [1], we find that both low-lying excitation energies and oscillator strengths are in improved agreement with experiments relative to the TDA. We characterize the low-lying excitons, focusing in the degree of charge-transfer and spatial delocalization, connecting their relevance to singlet fission. [2] For perylene, we find overall good agreement with absorption measurements, and we see evidence for the formation of an exciton-polariton band in $\beta$-perylene. 1. da Jornada, F. H. et al., to be submitted. 2. Sharifzadeh. S. et al., J. Phys. Chem. Lett. 4, 2197 (2013).

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