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Exciton self-trapping and Stark effect in the optical response of pentacene crystals from first principles[1] DAVID A. STRUBBE, Department of Physics, University of California, Berkeley; Materials Sciences Division, Lawrence Berkeley National Laboratory, SAHAR SHARIFZADEH, JEFFREY B. NEATON, Molecular Foundry, LBNL, STEVEN G. LOUIE, Physics Dept., UC Berkeley; MSD and MF, LBNL — Pentacene is a prototypical organic semiconductor with optoelectronic and photovoltaic applications. It is known that the lowest-energy singlet excitation has a Stokes shift between absorption and emission of about 0.14 eV, but the deformation associated with this self-trapped exciton remains unknown. We begin with a calculation of the optical properties via the first-principles GW/Bethe-Salpeter (BSE) theory [ML Tiago, JE Northrup, and SG Louie, Phys. Rev. B 67, 115212 (2003); S Sharifzadeh, A Biller, L Kronik, and JB Neaton, arXiv:1110.4928 (2011)]. We then study the self-trapping phenomenon via our reformulation of the Bethe-Salpeter excited-state forces approximation of Ismail-Beigi and Louie [Phys. Rev. Lett. 90, 076401 (2003)], which can describe the structural relaxation after optical excitation. Whether excitons in pentacene have charge-transfer character has been controversial in electro-absorption experiments. We use the same BSE analytic derivatives approach to calculate the changes in excitation energies due to an applied electric field to understand this experimental controversy.

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David A. Strubbe
Department of Physics, University of California, Berkeley;
Materials Sciences Division, Lawrence Berkeley National Laboratory

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