

CHEMICAL ENGINEERING

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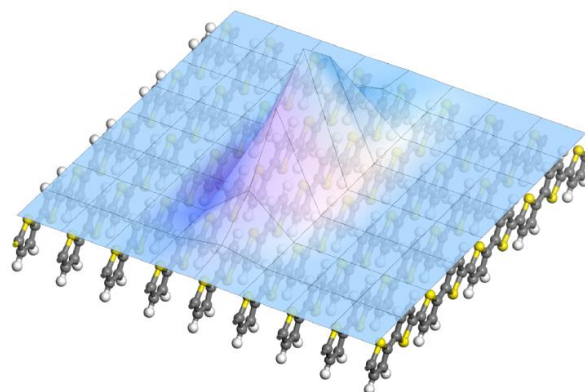
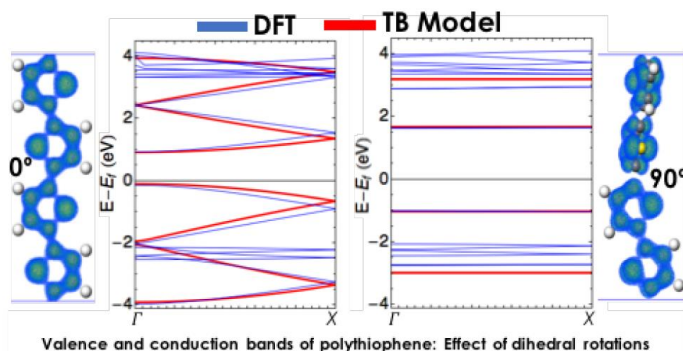


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Pennsylvania State University

Tight-Binding Model of Charge Carrier Localization and Excitons in Conjugated Polymers



ABSTRACT: In polymer semiconductors, the interaction between electronic and nuclear degrees of freedom leads to the localization of charge carriers, and strongly affects charge transport. Understanding the mechanisms that cause these localizations is necessary for establishing a clear link between the polymer structure and electronic properties. We use a coarse-grained approach based on the tight-binding approximation to model the electronic degrees of freedom of poly(3-hexylthiophene) (P3HT) as case study, with an effective account of conformational degrees of freedom. The relevant parameters of our model are determined using density functional theory (DFT) calculations. First we consider localization by disorder, particularly, dihedral disorder, which disrupts extended electronic states. Our tight binding model for charges hopping between adjacent rings, coupled to inter-ring dihedral rotations, predicts the formation of strongly localized valence and conduction states due to dihedral disorder along single polymer chains. We show that dihedral disorder alone accounts for most of the broadening of the optical absorption spectrum of P3HT chains in solution. Next, we consider localization by polaronic effects. Polarons can form when charges are able to induce deformations of the surrounding medium, including local vibrational modes or dielectric polarization. These deformations then

interact attractively with the charge, tending to localize it. Here we first look at vibrational polaron formation in P3HT, with a tight-binding model in which charges are coupled to ring distortions. On single chains, we find only broad, weakly bound polarons by this mechanism. In 2d crystalline layers of P3HT, even a rather weak transverse hopping between chains destabilizes this polaron. Then, we consider polarons stabilized by dielectric polarization, described semiclassically with a polarizable continuum interacting with the carrier wavefunction. In contrast to vibrational polarons, we find dielectrically stabilized polarons in P3HT are narrower, more strongly bound, and stable in 2d crystalline layers. Finally, we use the same tight binding methodology for single polymer chains to describe excitons, which are excited electron and hole pairs bound by a Coulomb interaction. We focus on their stability and the energetics of their dissociation and separation into individual charge carriers, as crucial to the performance of photovoltaic devices. We show that applied electric fields and donor-acceptor junctions help pull excitons apart by altering their dissociation potential, rendering them easier to unbind.

BIOGRAPHY: I am currently completing my PhD in the Department of Chemical Engineering with Scott Milner and Michael Janik as advisors at The Pennsylvania State University, which I joined since August 2013. My PhD work focused on modeling the electronic transport properties of semiconducting polymers with the aim of elucidating the structure-property relationship, as necessary to inform the design of materials with improved performance. I received my Bachelor of Science in Engineering in Chemical Engineering at the University of Cape Town in South Africa in 2012. I then worked as an assistant lecturer and research assistant in the process modeling and optimization group within the same department, before I joined Penn State. My interest in chemical engineering was formed in a mining town of the Democratic Republic of the Congo, where I grew up.

LECTURE 4:00 - 5:00 (PAA) A110
Happy Hour in Benson Hall Lobby Following



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