Organic solar cell models predict how structures change properties

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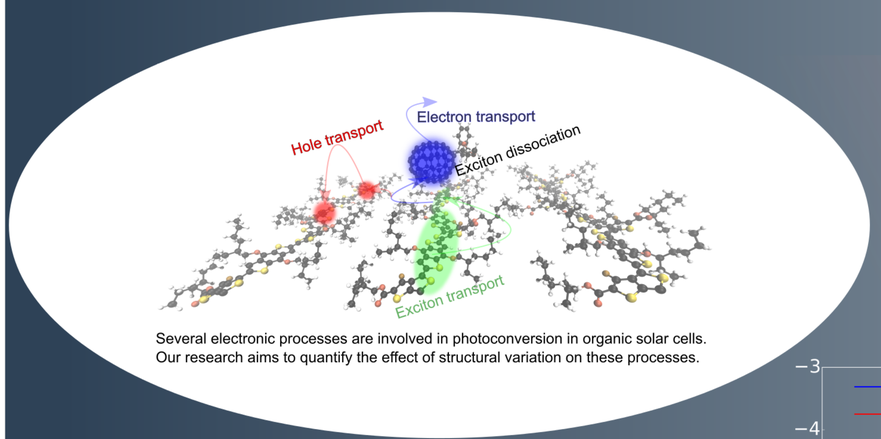
Motivation

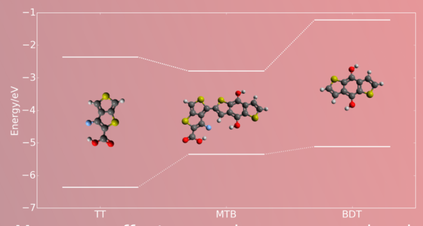
Solar cell devices composed of organic polymers are a promising new direction for solar energy conversion. These devices have chemically tunable band gaps and high absorption coefficients. Organic solar cells are thin, flexible, and comparatively inexpensive. These devices contain an active layer which is a blend of electron donor and acceptor materials, as well as electrodes for charge collection (see Fig. 1). Since charge-generation can only occur at the interface between materials, this interface is intentionally disordered, producing a bulk heterojunction.Performance depends upon electronic properties such as energy level alignment and charge carrier mobilities. Using periodic plane wave density functional theory (DFT) and many body perturbation theory (G0W0) calculations, our work studies how nanoscale structural changes affect electronic properties, such as band locations and band gaps.



Several electronic processes are involved in photoconversion in organic solar cells. Our research aims to quantify the effect of structural variation on these processes.

Monomer design

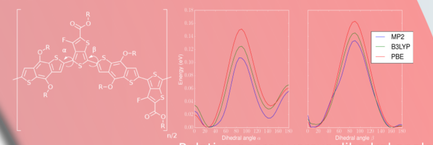
The donor polymers consist of two repeating subunits, which allow for partial localization of the highest and lowest occupied molecular orbitals (HOMO and LUMO). Using this copolymer method, the energies of these orbitals can be separately tuned, which also adjusts the fundamental and optical gap.



Monomer effects on polymer energy levels

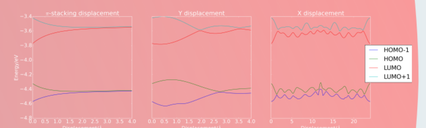
Polymer design and disorder

The connection between donor polymer subunits is flexible. Intrachain and interchain disorder changes the electronic states.



Relative energy versus dihedral angle

Different stacking motifs change how the bands delocalize, resulting in different electron and hole mobilities



Band energy versus displacement

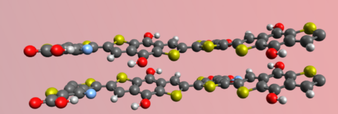
Conclusions

Our electronic structure models of organic solar cells demonstrate that structural variation changes the electronic states involved in solar energy conversion. Our lowest energy structures match experimental band gaps and locations well.

Crystalline polymorphs

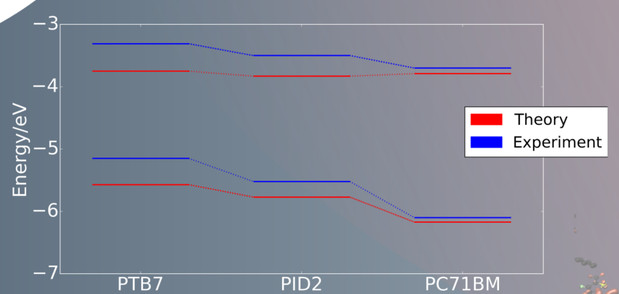
Periodic DFT models indicate significant variation of electronic states with different polymorphs.

	Relative energies (eV)	B3LYP Eg (eV)		π-stacking distance/Å
cis-trans	0.57	2.29	cis-trans	3.83
cis-cis	0.00	1.81	cis-cis	3.79
trans-cis	0.62	2.32	trans-cis	3.95
trans-trans	0.11	1.82	trans-trans	3.76



Heterojunctions

The band locations at the interface will depend on relative orientation. Our models predict the correct state ordering.



Energy of valence and conduction bands computed for a ternary solar cell composed of two donor polymers (PTB7 and PID2) and one fullerene acceptor (PC71BM). The experimental values (cyclic voltammetry) are taken from Lu, L. et al., Nature Photonics, 8(9), 716–722 (2014).

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