

## Interfacial dipole and geminate pair energetics at pentacene/C<sub>60</sub> heterojunctions

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Among the successive steps describing the light-to-electricity conversion process in organic photovoltaic cells, the splitting of the Coulomb bound electron-hole pairs at the donor/acceptor interface into free charge carriers remains an open question. The so-called electric-field assisted mechanism assumes that interfacial electric fields, arising from local interfacial dipoles, can favor the charge separation [1].

To characterize the dipole moments at pentacene/C<sub>60</sub> heterojunctions, we have coupled quantum-chemical and microelectrostatic (ME) calculations on molecular aggregates of various sizes and shapes. The results show that the interfacial dipoles mostly originate in polarization effects due to the asymmetry in the multipolar expansion of the electronic density distribution between the interacting molecules. The local dipoles are found to fluctuate in sign and magnitude over the interface and appear as sensitive probes of the relative arrangements of the pentacene and C<sub>60</sub> molecules [2].

Then, the geminate pair energetics at the pentacene/C<sub>60</sub> interface has been addressed by increasing the electron-hole separation across the interface. The results indicate that geminate pair dissociation critically depends on the orientation of the pentacene  $\pi$ -system relative to the adjacent C<sub>60</sub>. Indeed, these calculations predict that recombination of the electron-hole pairs is favored over separation at the pentacene(011)/C<sub>60</sub> interface, while the geminate pair splitting can easily occur at the pentacene(01-1)/C<sub>60</sub> interface [3].

[1] V. I. Arkhipov, P. Heremans, H. Bässler, *Appl. Phys. Lett.* **2003**, 82, 4605.

[2] M. Linares, D. Beljonne, J. Cornil, K. Lancaster, J.L. Brédas, S. Verlaak, A. Mityashin, P. Heremans, A. Fuchs, C. Lennartz, J. Idé, R. Méreau, P. Aurel, L. Ducasse, F. Castet *J. Phys. Chem. C*, **2010**, 114, 3215.

[3] S. Verlaak, D. Beljonne, D. Cheyns, C. Rolin, M. Linares, F. Castet, J. Cornil, and P. Heremans *Adv. Funct. Mat.* **2009**, 19, 3809.