Modelling charge transfer in molecular materials: from photovoltaics to ferroelectrics

Gabriele D'Avino

Laboratory for Chemistry of Novel Materials, University of Mons

Charge transfer (CT) interactions between electron donor (D) and acceptor (A) molecules are central to many physical phenomena in chemistry, biology and materials science. In this talk I will report about our modelling of different charge transfer systems, for which we propose a multiscale modelling approach based on first-principles and model Hamiltonian electronic structure calculations, and classical schemes to account for electrostatic and polarization interactions at the mesoscale.

I will first focus on mixed-stack CT crystals (e.g. TTF-CA, TTF-BA), a class of quasi 1D-materials presenting competing phases, strong electronic ferroelectricity and magnetoelectric phenomena. Our calculations, based on the Peierls-Hubbard model, shed light on this complex phenomenology [1] and dismantle the recent claim of room-temperature ferroelectricity in a novel series of hydrogen-bonded systems [2]. In a second instance I will consider D/A interfaces for photovoltaic application, and discuss the crucial role of microscopic electric fields, charge delocalization and excitons' hybridization, in the formation of free carriers upon light absorption [3].

[1] D'Avino, Girlando, Painelli, Lemèe-Cailleau, Soos, Phys. Rev. Lett. 99, 156407 (2007); D'Avino, Masino, Girlando, Painelli, Phys. Rev. B, 83, 161105R (2011).
[2] D'Avino and Verstraete, Physical Review Letters 113, 237602 (2014).
[3] D'Avino *et al.*, J. Phys. Chem. C, 117, 12981 (2013); Castet, D'Avino, Muccioli, Cornil, Beljonne, Physical Chemistry Chemical Physics, 16, 20279 (2014).