

Modelling charge transfer in molecular materials: from ferroelectrics to photovoltaics

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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 approach based on first-principles and model Hamiltonian electronic structure calculations, and classical schemes to account for electrostatic and induction 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).