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].