Ionic liquid–electrode interface: from quantum capacitance to charge transfer

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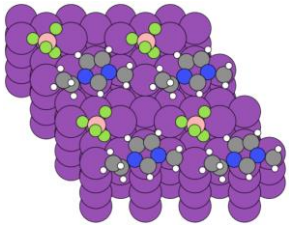
This presentation focuses on the electron density redistribution at the electrical double layer. Such redistribution – in the form of quantum capacitance and charge transfer – plays a crucial role in various applications, from ideally polarisable supercapacitors to non-polarisable catalysts for CO_2 electroreduction.

Using Density Functional Theory, we compare the behaviour of quantum capacitance and charge transfer at interfaces of metal (Au) and semi-metal (Bi and C) electrodes with imidazolium (Im^+) tetrafluoroborate (BF_4^-) and iodide (I^-) containing electrolytes. We employ electron density analysis methods, including Bader, DDEC, and Voronoi analyses, to quantify charge transfer, which ranges from modest polarisation to partial transfer with halide anions. Using Molecular Dynamics simulations, we also illustrate the contribution of quantum capacitance to the total capacitance of the electrical double layer formed by the above-listed electrodes and electrolytes. Special attention is drawn to the origin of capacitance peaks and their drifting on the potential scale upon varying electrode polarizability (in modelling).

The presentation summarises our published research[1–5] and the most recent modelling results, aiming at understanding how quantum capacitance and charge transfer can benefit the electrochemical devices for energy storage and conversion.



Scheme or Figure 1: An example Bi(111)–EMImBF4 interface model for Density Functional Calculations.

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