Charge Transfer at ZnO/Organic Interfaces: A computational perspective

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Inorganic/organic semiconductor heterojunctions have opened up new opportunities for (opto-) electronic devices due to their potential for combining the favorable properties of two distinct material classes. Yet a condition for all optoelectronic applications is to employ at least one electrode that is optically transparent, i.e., a thin conductive film. The current material of choice, indium-doped tin oxide, is increasingly being replaced by the less toxic, more benign zinc oxide.

Due to its unique physics and chemistry, ZnO poses several unique challenges for experiment and theory alike. In the present contribution I will use the adsorption of the organic electron donor pyridine [1] and the electron acceptor F4TCNQ [2] on different ZnO surfaces as examples to highlight some of the challenges that standard band-structure calculations face. I will discuss how these problems can be tackled, and what the underlying assumptions and limitations imply when comparing density functional theory calculations to experimental results. In particular, the impact of the nature and concentration of doping on the amount and distribution of charge transferred across the interface will be addressed [3]. I will demonstrate that the charge distribution is very sensitive to the local chemistry. For adsorbates which show a complex bonding behavior with both covalent and ionic character, it is even possible to construct situations where, counterintuitively, most of the adsorbed electron acceptors are, in fact, positively charged.

[1]: Hofmann et al., The Journal of Chemical Physics, 139. 174701 (2013)
[2]: Xu et al., Phys. Rev. Lett., 111, 226802 (2013)
[3]: Sinai et al., Phys. Rev. B, 91, 075311 (2015)