How does structure affect interfacial charge transfer? – isonicotinic acid on rutile(110)

Moritz Müller*†[1], Daniel Sánchez-Portal[2], He Lin[3], Gian Paolo Brivio[3], Annabella Selloni[4], and Guido Fratesi[3]

[1]CIC nanoGUNE (CIC nanoGUNE) – Tolosa Hiribidea, 76, San Sebastián, 20018, Spain
[2]Centro de Fisica de Materiales – Paseo Manuel de Lardizabal 5, 20018 Donostiá-San Sebastián, Spain
[3]Dipartimento di Scienza dei Materiali – Università degli Studi di Milano-Bicocca, Italy
[4]Chemistry Department – Princeton University, Princeton, NJ 08544, USA, United States

Abstract

Charge transfer processes at interfaces in photovoltaic systems are strongly affected by temperature. In this context we analyze the effects of heat-induced structural fluctuations from a modeling perspective. In our approach we extract lifetimes of localized states on dye molecules attached to semiconductor substrates. The atomic motions of the adsorbate and substrate are sampled by taking snapshots from *ab-initio* density-functional-theory molecular-dynamics trajectories [1]. We effectively extend the slabs utilized in our dynamical calculations to full semi-infinite substrates with the use of a Green's function approach [2,3]. At this level of theory we obtain time-averaged values of the charge injection times using several snapshots of the Car-Parinello simulations. We compare our results with core-hole-clock experiments of the same system finding a good agreement [4]. To include the perturbation inferred by the measurement technique we consider core-excited molecules in our simulations. This results in a lowered dependency on structural fluctuations of the energetic positions of the molecular level as compared to the non-excited molecule. This study enables the search for direct connections between structural parameters, resonance energies, and elastic charge transfer times scrutinizing the configurational space covered by the molecular dynamics simulations.

This project has received funding from the European Union's Seventh Framework Programme under grant agreement no. 607323 [THINFACE]

H. Lin *et al.,* J. Phys. Chem. C **7**, 3899 (2016)

D. Sánchez-Portal, Prog. Surf. Sci. **82,** 313 (2007)

G. Fratesi *et al.,* J. Phys. Chem. C **118**, 8775 (2014)

J. Schnadt *et al.,* J. Chem. Phys. **119**, 12462 (2003)

*Speaker
†Corresponding author: mllr.mrtz@gmail.com