Density Functional Modelling of Field Evaporation

Christoph Freysoldt[1], Arpit Mishra[1], Michael Ashton[1], and Jörg Neugebauer[1]

Corresponding Author: freysoldt@mpie.de

[1.] Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Str. 1, 40237 Düsseldorf, Germany.

At the heart of atom probe tomography (APT) is the evaporation of single atoms from a very sharp tip exposed to huge electric fields (10^{11} V/m). Field evaporation is a complex process in which not only the atom must break one or more bonds to its chemical environment, but the atom is also ionized at some point in the trajectory (or even multiple times) before it follows the electric-field lines away from the surface. The electric fields, in turn, depend on the local surface morphology. Commonly used geometric reconstruction algorithms for APT fail to take into account any details of the evaporation mechanism, such as differences in evaporation between chemical species or from different sites. Indeed, the atomistic details of field evaporation remain largely obscure, both qualitatively and quantitatively. This significantly hinders the development of improved reconstruction algorithms beyond the state of the art.

Density-functional theory (DFT) may provide important insights. DFT has been shown to reproduce barriers and critical fields in reasonable agreement with experiment for adatoms on Al surfaces [1]. Unfortunately, this seminal study has not been followed by systematic investigations for other sites or other materials because common computer codes must be adapted to account for large electric fields. We have therefore implemented an efficient approach to incorporate electric fields for slab models of the surface into our DFT code SPHInX [2].

To shed light on the factors influencing field evaporation, we study desorption from various sites (ad-atom, steps, corners) on prototypical metal surfaces (Al, W). From these calculations, we obtain the field-dependent evaporation barriers, which may be used in subsequent APT simulations. We further compare the outcome of explicit-field calculations with the prediction of traditional models based on zero-field work functions and binding energies. We also find evidence for the roll-over effect at steps, that has been proposed to explain orientational bias in evaporation trajectories.

References:

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[2] S. Boeck *et al*, Comp. Phys. Commun. 182 (2011), 543-554.