

Atomistic-Simulation Based Modeling of Atom Probe Tomography

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Atom probe tomography (APT) promises atom-by-atom material analysis with spatial resolution and chemical sensitivity that no other technique can achieve [1,2]. It is increasingly used in research as well as in industry to explore 3D chemistry of engineering materials, semiconductors, and even organic samples at the nanoscale [3]. Technological advances in ion imaging, particle detectors, and fast laser pulsing over the past decade have positioned APT on par with other high-resolution imaging techniques in terms of ease of specimen preparation and size of the volume analyzed.

While the promise of full chemical information and outstanding spatial resolution have given APT the role of a transformative characterization tool, only moderate advances have been achieved in the processing of APT measurement data. The simplistic algorithm to generate reconstructions of a 3D volume from an analyzed specimen has not changed much since originally proposed, although it is known to produce a long list of known artifacts in the reconstructed model [4]. Even by virtue of other complementary techniques, e.g. correlative TEM microscopy, the continuous observation of the evolving sample shape under field evaporation is only possible to a limited extent, especially at close to atomic resolution, which is probably necessary to fill in the so far missing information needed for accurate subsequent reconstruction.

In order to fill this void left by experimental limitations, complementary modeling techniques of field evaporation have been developed by us and others to address fundamental questions. The most impactful methods here are based on forward simulation, where atoms are computationally evaporated from a virtual sample in a simulated field and propagated to a virtual screen. If all the evaporation and ion-flight physics are treated sufficiently well, such simulations provide realistic detector signals as well as realistic tip-shape evolution without simplifying assumptions. Foundation for that is the numeric solution for the field enabled by finite elements (FEM). The first highly simplified APT simulation approach along these lines was introduced about 20 years ago [5]. Despite its simplicity, the emitter structure was modeled on a regular mesh of fairly limited size and with “cubic” atoms, it showed to be surprisingly successful. For instance, artifacts due to local magnification effects in the APT analysis of heterogeneous emitter structures (e.g. interfaces or precipitates) could be well explained. Motivated by this success, several other approaches have been developed subsequently that removed existing restrictions of earlier predecessors. Current state-of-the-art simulations [4] are (virtually) meshless and can handle significantly larger emitter samples of arbitrary lattice structure, e.g. of 20 nm in diameter, 100 nm in length with overall one million atoms, close to the dimensions of small experimental samples.

In addition to the increased flexibility, a number of improvements were made with respect to the physics involved and thus towards more realistic simulations, e.g. by including the statistical effects from finite temperature [6] or site-specific evaporation fields [7]. Our recent work has targeted to remove the last major and rather significant restriction of atoms being fixed to their position and allows now the atoms

in the sample to relax when atoms are evaporated. To this end, the regular electrostatic solver is combined with molecular dynamics (MD) [8] [9] and the mechanical response of the emitter structure due to field induced forces is calculated (**Fig. 1**). Results from this approach include new insights into the origin of unexpected solute clustering around certain poles in experiments (e.g. for Cu or Si in Al), which we can show to be due to significant solute migration from athermal relaxation. Also, our results show that the applied load onto the surface atoms by the so-called Maxwell stresses is on the order of several 100 MPa and thus cannot be neglected as it is responsible for notable strain at the surface and within the emitter already before the onset of field evaporation. While such a sophisticated combined MD-FEM approach is computationally demanding, our results suggest it is an important key in the APT simulation toolbox that will allow for a significantly better understanding of the fundamentals of field evaporation on the atomistic level and paves the way to include field dependent parameterizations based on functional theory (DFT) in the future.

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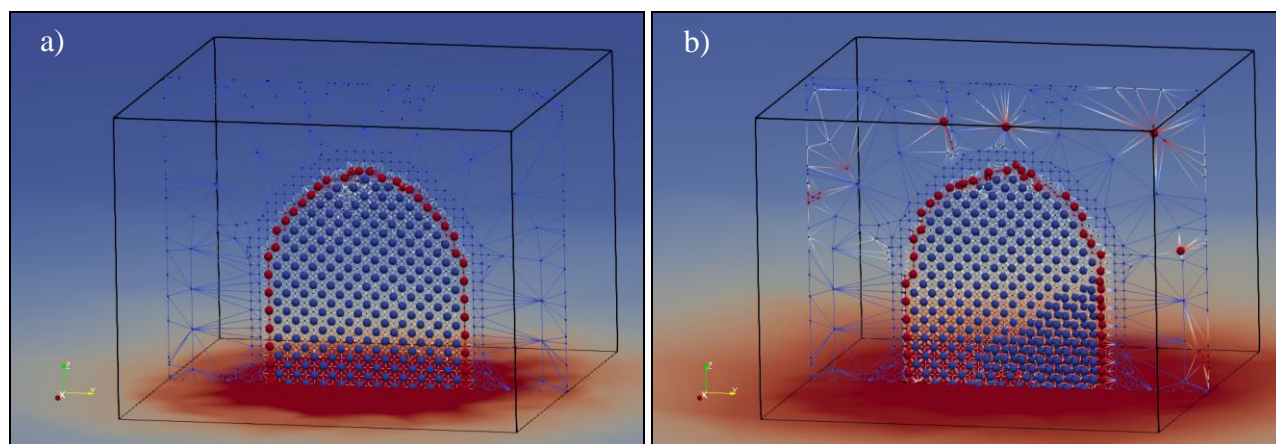


Figure 1. Slice through an APT tip from LAMMPS [10] MD simulations with added field-induced forces: The overlay in the background shows the field solution obtained from the FEM mesh in the foreground. Spheres depict atoms of the emitter tip (blue bulk, red surface). (a) At very low field the resulting atomic positions are still coincident with the surrounding mesh. (b) At elevated field the emitter lattice is severely strained and atoms desorb from the surface.