Towards Model-Driven Reconstruction in Atom Probe Tomography

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Reconstructions of heterogeneous material systems in Atom Probe Tomography (APT) suffer from imaging distortions due to the underlying assumptions made about specimen geometry by current reconstruction methods [1]. These distortions can seriously limit the reliability of measurements derived from reconstructions, particularly in the vicinity of microstructural interfaces [2]. While previous attempts have been made to provide a solution for correcting such distortions, these efforts have fallen short of delivering a physically motivated and general algorithm that can be deployed on experimental data.

Here we present a new model-driven reconstruction protocol capable of correcting some of these distortions. By physically modelling the underlying causes of distortions, such as sample field evaporation and ion deflection, trajectory data derived from a model is used to position ions in the reconstruction. Our continuum model, a 3D extension of that presented in [3], tracks the sample surface through a level set method and solves the electric field via a boundary element method. This modelling framework benefits from a high computational efficiency while also remaining flexible, allowing for a range of APT phenomena to be captured e.g. sample faceting and void evaporation. However, the methodology presented for calibrating our model to experiment should transfer across to other APT modelling approaches. An example reconstruction of a fin Field-Effect Transistor (finFET) dataset, originally featured in [2], using our new algorithm is given in Figure 1c. Compared to a point-projection reconstruction (Figure 1b), our reconstruction better reproduces the true microstructure (Figure 1a) while reducing density variation (Figure 1d). The algorithm can also handle more complex sample geometries through integration of Electron Tomography (ET) data. Such a reconstruction method could lead to true high resolution imagery in APT with minimal distortion.

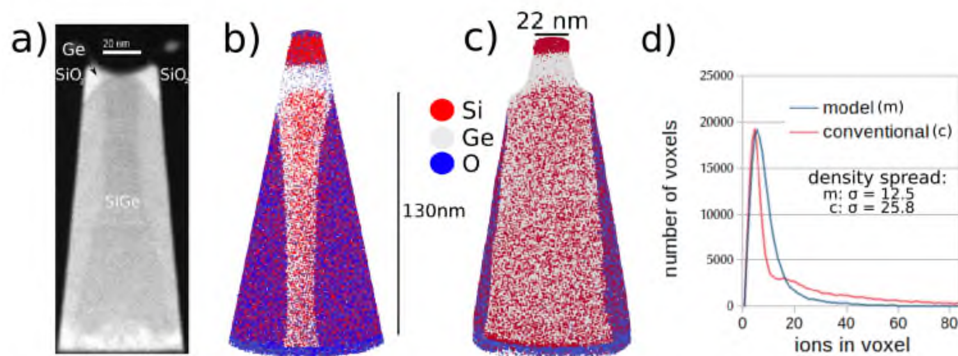

Figure 1: a) HAADF-STEM of the finFET before sample preparation (micrograph modified from [2]) b) A point-projection reconstruction of the finFET dataset. c) Our model-driven reconstruction. d) Density histogram showing an improved density profile. FinFET dataset, first seen in [2], courtesy of IMEC.

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Development of an energy-sensitive detector for the Atom Probe Tomography

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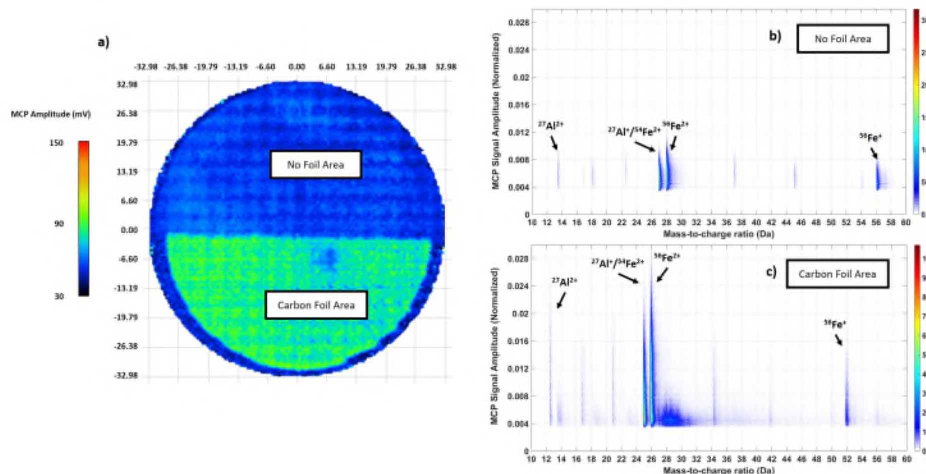
A position-energy-sensitive detector has been developed for APT instruments in order to deal with some mass peak overlap issues encountered in APT experiments. Through this new type of detector, quantitative and qualitative improvements could be considered for critical materials introducing mass peak overlaps, such as nitrogen and silicon in TiSiN systems [1], or titanium and carbon in cemented carbide materials [2].

This new detector is based on a thin carbon foil positioned on the front panel of a conventional MCP-DLD detector. According to several studies, it has been demonstrated that the impact of ions on thin carbon foils has the effect of generating a number of transmitted and reflected secondary electrons that mainly depends on both the kinetic energy and the mass of incident particles. Despite the fact that this phenomenon is well known and has been widely discussed for decades [3], no studies have been performed to date for using it as a mean to discriminate particles energy. Therefore, this study introduces the first experiments on a potential new generation of APT detectors that would be able to resolve mass peak overlaps through the energy-sensitivity of thin carbon foils.



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Atom probe observation of hydrogen in steel microstructures

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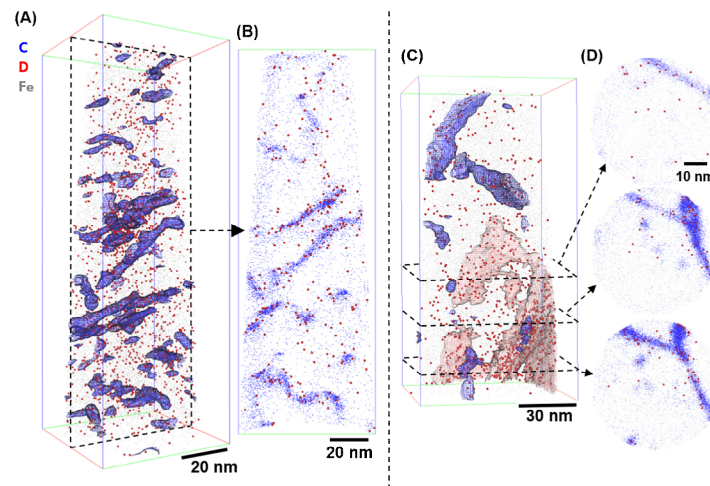
The presence of hydrogen in steels can lead to catastrophic embrittlement/early-fracture. This is a serious issue for hydrogen transportation and storage. However, consensus has not been reached on the exact mechanism of hydrogen embrittlement, mainly due to the difficulty to provide direct evidence of the hydrogen-materials interactions that underpins the hypotheses [1]. In addition, a proposed solution to hydrogen embrittlement by using steels that contain hydrogen traps such as carbide precipitates [2], is limited in its effectiveness, due to the inability to directly observe the proposed hydrogen trapping at microstructural features.

As such, we used atom probe to study the hydrogen distribution at key features, including dislocations [4], grain boundaries [4], and both incoherent [4] and coherent [3] carbide precipitates in BCC/BCT iron matrix. To enable these studies, we charged the sample with deuterium (a hydrogen isotope) to avoid ambiguity from background hydrogen, and utilised a custom cryogenic sample transfer protocol to allow sufficient signal to be retained for observations. These efforts lead to the confirmations of: i) hydrogen enrichment at dislocations (Figure A and B), providing a concrete validation of the hydrogen-enhanced dislocation mobility theory of embrittlement; ii) hydrogen enrichment at grain boundaries (Figure C and D), underpinning the hydrogen-enhanced grain boundary decohesion theory; iii) the hydrogen at the interface between large, incoherent precipitates and the surrounding steel matrix, settling a long-standing debate around whether hydrogen trapping is an interfacial effect; and iv) the hydrogen at the interior of small, coherent carbides, suggesting hydrogen can internalise into carbides under certain conditions.



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Imaging individual solute atoms at crystalline imperfections in metals

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Directly imaging all atoms constituting a material and, maybe more importantly, crystalline defects that dictate materials' properties, remains a formidable challenge. Here, we propose a new approach to chemistry-sensitive field-ion microscopy (FIM) combining FIM with time-of-flight mass-spectrometry (tof-ms). Elemental identification and correlation to FIM images enabled by data mining of combined tof-ms delivers a truly analytical-FIM (A-FIM). Contrast variations due to different chemistries is also interpreted from density-functional theory (DFT). A-FIM has true atomic resolution and we demonstrate how the technique can reveal the presence of individual solute atoms at specific positions in the microstructure. The performance of this new technique is showcased in revealing individual Re atoms at crystalline defects formed in Ni–Re binary alloy during creep deformation. The atomistic details offered by A-FIM allowed us to directly compare our results with simulations, and to tackle a long-standing question of how Re extends lifetime of Ni-based superalloys in service at high-temperature.

Development of a wide field of view three dimensional field ion microscope and high fidelityreconstruction algorithm to investigate small defect in materials

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Field Ion Microscopy (FIM), invented in 1951 by Erwin Muller, is the predecessor of the Atom Probe Tomography (APT). It is the first microscopy technique allowing a direct imaging of individual atoms at the surface of a material [1,2]. From several decades FIM remained a 2D surface microscope principally used to select analysis regions for Atom Probe Tomography. Nevertheless, field evaporation allows to records images with a sub-angstrom precision at different depths in the material. The ability to localized the 3D positions of individual atoms has been established by Seidman et al. [3] in 1981. This pioneer work and others most recent [4-6] succeeded to show the potential of FIM to reconstruct in 3D at the atomic scale the lattice of a pure material and some defects.However, until today, there is no automatic and repeatable technique capable of mapping in 3D with an atomic resolution and wide angle the finest features in the crystallographic lattice of materials. In the present work, new experimental method and digitals algorithms, based partially on Dagan et al.[5] and Vurpillot et al. [4] studies, are proposed. This new approach adapted to large volume of data allows to localize individual atoms on FIM images. Using this developed method, 3D FIM proves to be a powerful tool and the most accurate technique to reconstruct in real space a material at the atomic scale in volumes larger than 100x100x100 nm3 (Fig.(1a)). Spatial resolution on tungsten sample is almost ten times better than that of atom probe. Detection efficiency are also improved compared to atom probe and it can reach a global efficiency of 85% and a local efficiency of almost 100%. Performances were validated through a new model of FIM images (Fig.(1b)). It is also shown that the intensity of each spot just before evaporation on the FIM images which depends of the chemical species and can be used to differentiate chemical elements and to study their distribution in the sample (Fig.(1c)).

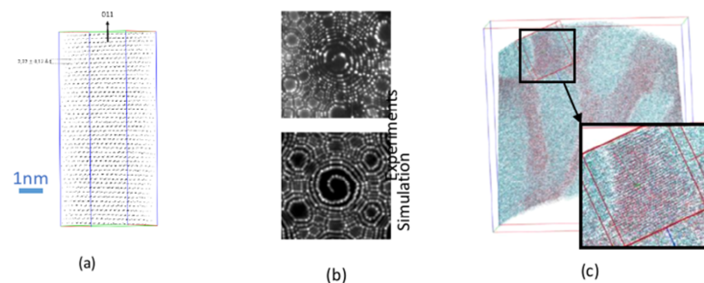

Figure 1 : Sub-volume of a tungsten analysis in 3d FIM(a). Note that no filtering or image improvement method were necessary to reconstruct data. Comparison between FIM image of a screw dislocations and with the model simulation (b). 3D FIM reconstruction of annealing FeSiB at 500°C during 1h (c), spots intensity revels the chemical nature (red FeSi, blue SiB).

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Scanning probe microscopy for atom probe tip shape monitoring

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The determination of the atom probe tip shape has been identified as key to understand, predict and possibly account for artefacts in atom probe tomography (APT) data induced by variations of the local tip surface curvature[1]. This work elaborates on the application potential of scanning probe microscopy (SPM) as a tool to monitor the tip shape of the atom probe specimen. The intrinsic challenge of this technology relates to the alignment of the apex of the SPM probe (< 25 nm) to the apex of the atom probe tip. The latter enables a measurement of the tip surface with nanometre precision by scanning across the APT tip surface while maintaining a constant interaction force between both tips. We demonstrate how the enhanced electric field around the apex of the APT tip can guide the tip-on-tip alignment. As the SPM probe is sensitive to electrostatic forces, which enables to capture the spatially rapidly decaying field strength around the tip, it is a potential candidate to experimentally map out the three-dimensional electric field distribution around the APT tip. This will be shown on dedicated examples in which the full three-dimensional electric field distribution is determined. The alignment procedure is applied to study the curvature variations in homogeneous and heterogeneous APT tips which can be traced back to local material properties. Finally, simulations based on the experimentally observed tip shapes are compared to experimental data. The strong agreement between the latter two indicates the strong potential of SPM to become a cornerstone for future reconstruction protocols.

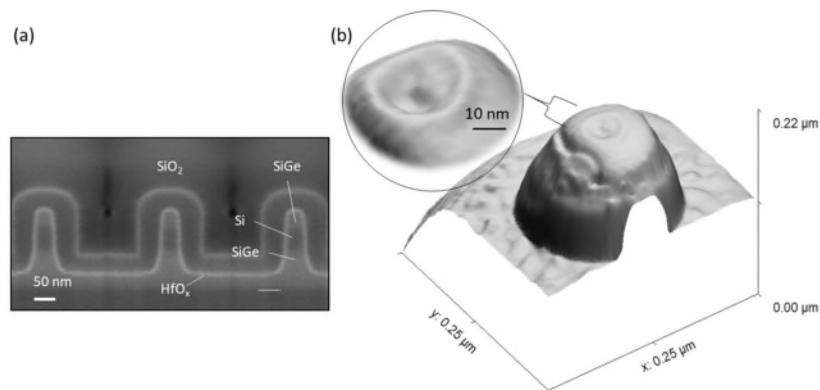

Figure 1: SPM analysis of a nanowire post APT analysis. (a) Scanning electron microscope image of the nanowire embedded in Si. (b) SPM topography image of the end form of the APT tip in which the circular region in the middle represents the protruding HfO_x film.

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