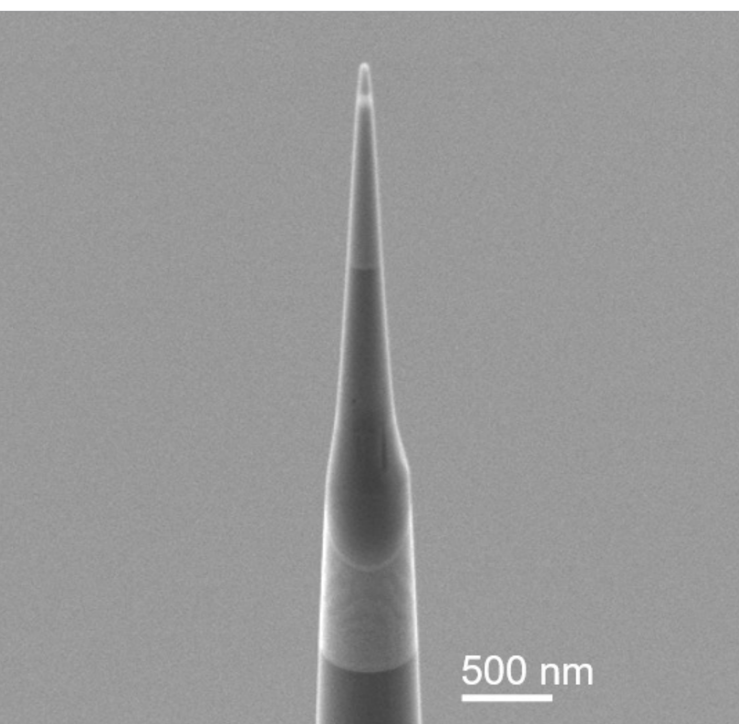
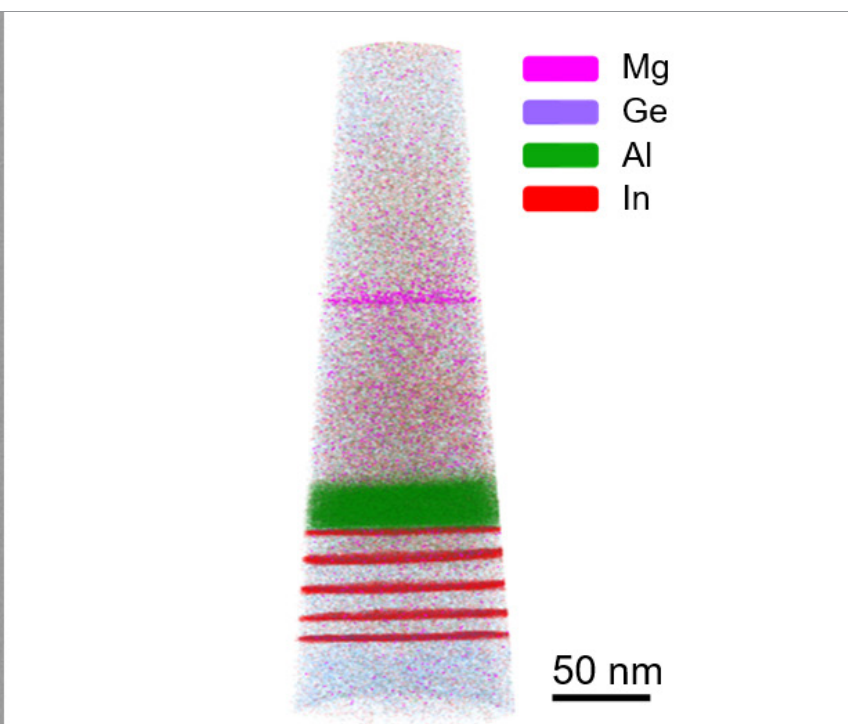
Application Note

FIB-SEM Fabrication of Atom Probe Specimens with ZEISS Crossbeam





Seeing beyond

Authors: Guillaume Audoit, Adeline Grenier
Univ. Grenoble Alpes, CEA, Leti, Grenoble, France

Fabián Pérez-Willard
Carl Zeiss Microscopy GmbH, Oberkochen, Germany

Date: March 2021

In this paper we review common protocols for the preparation of atom probe specimens using a FIB-SEM microscope. The starting point is the preparation of a 1-3 µm thick lamella. This lamella is lifted *in situ* from the bulk sample and sequentially chopped to transfer pieces of it to either free standing metallic needles or to the pre-sharpened microtips of a silicon chip, aka coupon. The choice of sample support depends on the type of instrument used for the atom probe analysis. Once attached to their support the pillars are milled into shape by FIB.

Introduction

Atom probe tomography (APT) is a quantitative analytical microscopy technique, that allows to map the spatial distribution of elements in a sample in three dimensions at the atomic scale [1,2]. It is one of the tools of choice for the analysis of current and future materials and devices in terms of morphology and composition.

APT is based on the field evaporation of a needle-shaped sample with end-radius below 50 nm. A high steady-state voltage (DC) is applied to the tip-shaped specimen which is cooled down to liquid nitrogen temperature. This induces a high electrostatic field at the tip apex that will ultimately lead to the field ionization and evaporation of surface atoms. These ions are accelerated towards a position sensitive detector (PSD) with time-of-flight (tof) detection capability. Basically, the electrostatic field at the tip apex is just below the evaporation field of chemical species. The field evaporation of surface atoms is then triggered by either high-voltage or ultra-short laser pulses applied to the sample. The field evaporated ions are projected onto the detector achieving a very large magnification (around 10^6). The chemical nature of the ions is determined by tof mass spectrometry. The tof is the time between a voltage or laser pulse and the impact of the ion on the PSD. Since the ion tof depends on its mass-to-charge (m/q ratio) ratio, its chemical nature can be determined. The position of impacts and the number of collected ions on the detector allows the analysed volume to be reconstructed in 3D at the atomic scale. The X and Y coordinates of the ion impact on the PSD are related to the initial position of the ion at the surface of the tip. The depth (Z) is determined as a function of the evaporation sequence.

The key strengths of APT are its close to atomic resolution (atomic planes can be imaged), quantitative compositional measurement and 3D mapping capability.

In the earlier days of APT, the surface atoms were evaporated using voltage pulses limiting the analysis to conductive materials. The tip-shaped specimens were mostly prepared by electrochemical polishing. While this technique remains a fast and easy method for many materials, it is difficult to apply on heterogeneous samples containing materials with different electrical conductivities. In addition, electropolishing is not site-specific. A convenient and alternative method is by ion direct machining in a FIB-SEM [3-5] addressing sample preparation of semi-conductors, insulators and heterogeneous materials and devices. Furthermore, FIB-SEM has the advantage of being site-specific to less than 10 nm. Thus, grain boundaries [6], interfaces [7,8] or single transistors [9] of the latest generation can be investigated.

Recently, with the development of cryo-transfer systems atom probe analysis has been extended to soft polymers, wet biological specimens, and even liquids [10,11]. In these cases, the sample is frozen and kept at cryogenic liquid nitrogen temperature during transfer to the FIB-SEM, FIB-SEM preparation and APT.

In this application note we review the preparation protocol for the fabrication of samples for the tomographic atom probe microscope with ZEISS Crossbeam [12].

Lamella Preparation

The first preparation steps are the same as for the preparation of a standard *in situ* lift-out TEM sample, see Figure 1 [4,5].

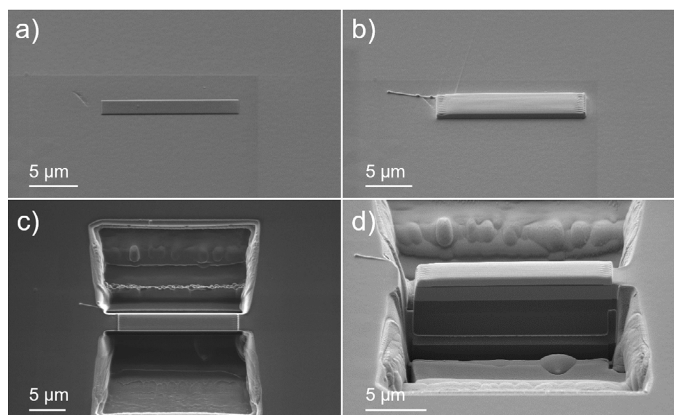


Figure 1 *a) Carbon EBID on an AlGaAs multilayer sample. b) Same sample after adding a platinum IBID on top of the carbon EBID. c) FIB image after the rough and fine milling steps. The lamella is 2.5 µm thick. d) Partially released lamella after cut-out.*

Once the region of interest has been located under SEM observation, the top surface is protected using an ion beam induced deposition (IBID). If the sample surface or its topmost layers are of interest, an electron beam induced deposition (EBID) has to be performed prior to the IBID to avoid any ion-induced damage.

For illustration, in this note we use an AlGaAs multilayer sample as an example. An area of 15 × 2 µm² of the surface was covered by a 1-minute carbon EBID at 3 kV SEM landing energy and 1 nA current (Figure 1a). This resulted in a ~100 nm-thick EBID, which was then followed by an 800 nm-thick, IBID of platinum at 30 kV ion energy, 300 pA current, and 3 minutes of duration (Figure 1b).

Trenches at both sides of the deposition were carved in two steps respectively using a 30 kV 30 nA probe for the rough milling and a 30 kV 3 nA probe for the finer milling steps (Figure 1c). For the latter a stage over-tilt of 2° was used for the polishing of the front side of the thick lamella and -2° under-tilt for the back. The additional stage tilt takes the FIB probe shape into account and ensures that the side walls are perfectly perpendicular to the sample surface. Each of the four milling steps took 90 s.

The sample was then tilted to 0° to perform the cut-out (3 nA FIB current, less than 3 minutes) step to partially release the lamella from the bulk. As can be seen in Figure 1d there is still a weak link between the chunk and bulk sample (at the right side of the lamella). At this point the lamella is ready for lift out.

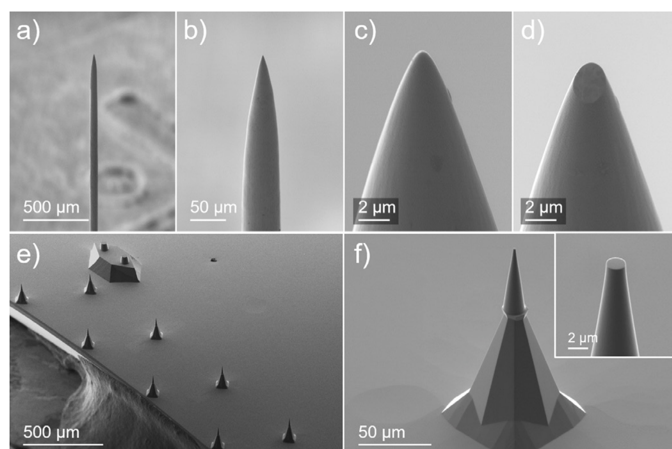


Figure 2 *a) Metallic needle imaged at low magnification. b) Same needle at slightly higher magnification. c) Apex of the needle. d) Apex after FIB milling of its top to form a plateau of 2 µm diameter. e) Silicon coupon containing several microtips and fiducials for orientation. f) Image of one of the microtips. The inset shows the apex region of the microtip, which is flat at the top.*

Sample Carrier and Lift Out

The chunk can now be lifted from the bulk using a micro-manipulator. Small square pillars will be extracted from it and transferred to suitable APT carriers sequentially, one by one.

For a standard atom probe instrument, sharp metallic needles like the one shown in Figure 2a are used as carriers. Each needle can hold one APT sample. The needles are mounted upright and ideally close to the original sample to speed up the lift outs. The top of each needle needs to be removed by FIB, see Figure 2c and d, to create plateaus of roughly 1-2 µm diameter. This is done at a stage tilt of 0 - 5°, so that the needle can be viewed and cut from the side by FIB. Thus, the resulting plateau forms an angle of 54 - 49° with the needle axis. This step is carried out either before or during the lift out.

Alternatively, when using a local electrode atom probe (LEAP) instrument, commercially available silicon chips, called coupons, will be used to hold the APT samples (see Figure 2e). Each coupon contains several pre-structured microtips (Figure 2f). The microtips are flat at the top over a circular area of ~2 µm diameter.

Figure 3 illustrates the lift out for our exemplary sample using a coupon. Here, the lift out was done at 5° stage tilt. The tip of the micromanipulator needle was attached to one end of the chunk by IBID (50 pA FIB current for 40 sec). The weak link holding the chunk to the bulk was then removed by a FIB cut (700 pA, 20 sec), so that the chunk could be maneuvered out of the bulk using the manipulator.

Next, the stage was moved so that a microtip of the coupon chip was situated below the lifted specimen. Using the micro-manipulator, the chunk end opposite to the micromanipulator tip was approached carefully to the microtip post and fixed to it by IBID (50 pA, 40 sec). A FIB cut across the chunk was then done to separate a square pillar of 2.5 μm × 2.5 μm cross section.

Figure 3d shows one of the pillars attached to its microtip. The previously described process can be repeated to load further microtips of the coupon. Depending on the length of the chunk, typically 5-15 APT samples can be easily extracted from it.

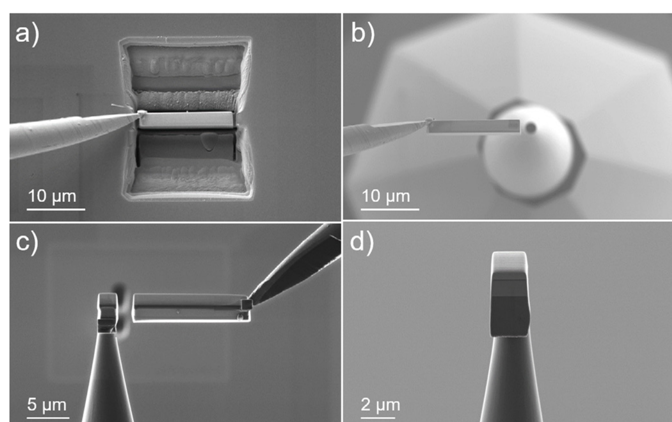


Figure 3 *Main steps of the lift out. a) The tip of the micromanipulator is welded to the chunk. The chunk is lifted, and the stage moved to b) place a microtip close to the chunk. c) One end of the chunk is welded to the tip and a piece removed. d) Image of the freestanding APT sample ready for shaping.*

Shaping the Needle

Figure 4 illustrates the APT sample shaping process. The shaping was done using successive annular FIB patterns of decreasing dimensions. Starting at 15 kV FIB accelerating voltage the beam energy was reduced stepwise to 5 keV, 2 keV and 1 keV.

Figure 4a shows a FIB top view image of the pillar with an overlay of the first annular milling object shaded in green. For the next milling steps the inner diameter of the ring pattern was reduced slightly and a strong overlap of each ring relative to its predecessor allowed. This supported the formation of a sharp tip and sculpted the needle shank.

Shaping at 15 kV FIB accelerating voltage was done using beam currents of 300, 100, and 50 pA. Figure 4b-d show the sample after execution of each of these milling steps. In Figure 4e the sample has undergone a FIB polishing step at 5kV and 20pA current. The radius of curvature at the tip apex is now 35 nm and the full cone angle 7°. The subsequent low kV polishing steps at 2 and 1 kV (20 pA and 10 pA) were kept short to maintain the overall shape of the tip whilst reducing further amorphization of the tip side walls. After the 1 kV polishing step the radius of curvature was 30 nm.

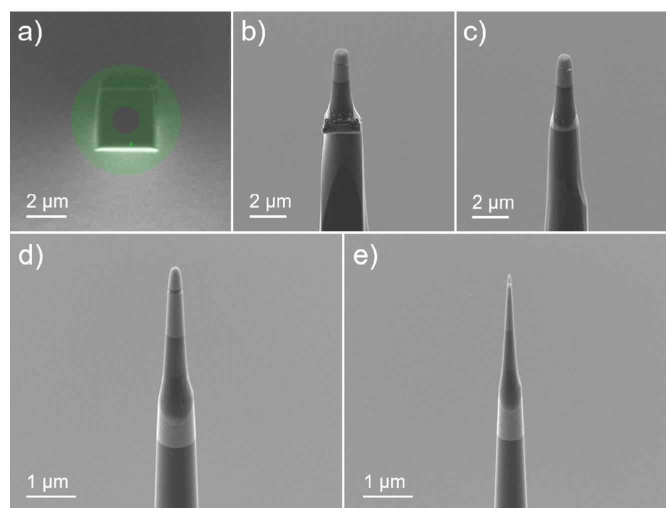


Figure 4 *Shaping of the APT specimen by FIB milling. a) FIB image with an overlay of the first annular milling object. b) to d) SEM images after shaping at 15 kV accelerating voltage and 300 pA, 100 pA, and 50 pA current. e) Tip after the 5 keV polish. At this stage the curvature radius at the apex is 35 nm and the full cone angle 7°.*

FIB performance for voltages below the standard operating voltage of 30 kV is key for the fabrication of robust and damage-free APT samples [8]. In this example, ion energies of 15 keV down to 1 keV were used, to minimize Ga implantation and amorphization. Further, as described above the corres-ponding low-kV milling steps define the final shape of the needle. Radius of curvature and shank angle are important geometrical factor that influence the intensity of the voltage or laser pulses needed to extract ions from the sample during the APT experiment. Figure 5 shows FIB images acquired with ZEISS Ion-sculptor column at 15, 5, 2, and 1kV during the preparation of the APT sample. Even at very low kV, the tip position could be easily resolved. Thus, the milling shapes could be precisely centered around the tip. Together with a well-focused beam this avoided material redeposition from the neighborhood onto the tip.

Another important aspect is the capability of imaging the sample with the SEM live while shaping it. Thanks to the non-immersion SEM objective lens design on ZEISS Crossbeams this can be done with the instrument's best resolution and with any of the in-column and chamber SE detectors. This allows to fully control the shaping process and is particularly important when targeting very small features, as these need to be contained very near the tip apex due to the limited field of view of the AP.

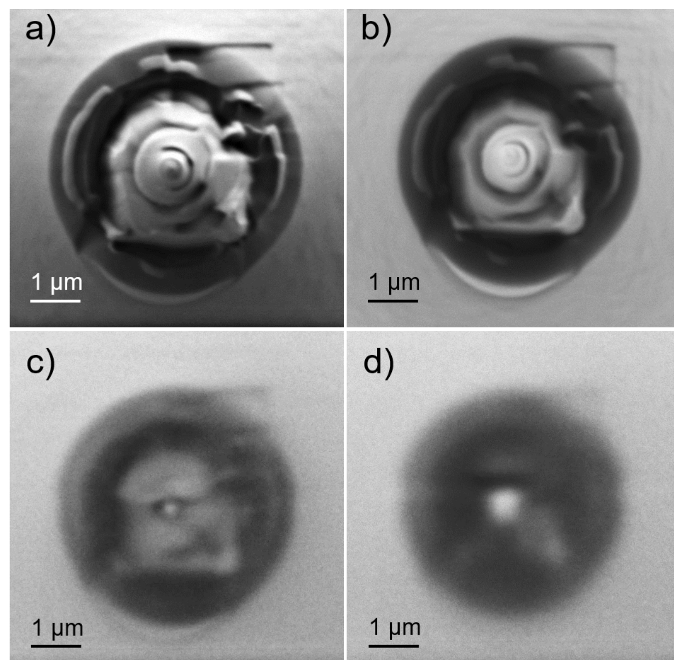


Figure 5 *FIB top view images of the specimen at different landing energies. a) 15 kV (50 pA), b) 5 kV (20 pA), c) 2 kV (20 pA), and d) 1 kV (10 pA). The FOV is 6.7 µm for all images.*

As an example, a 3D APT reconstruction of a hybrid tunnel junction InGaN light-emitting diode (LED) structure is shown in Figure 6. Doping and III-site elemental fraction can be investigated allowing to understand better the physical properties and performance of the device.

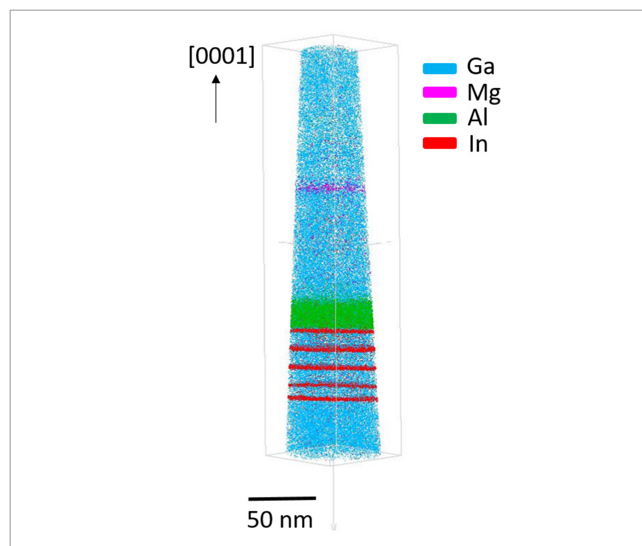


Figure 6 *3D APT reconstruction of a hybrid tunnel junction LED structure [13].*

Summary

In this application note the preparation of atom probe specimens using ZEISS Crossbeam was described. As an example, the preparation on an AlGaAs multilayer sample was shown.

Many variations of the presented workflow are possible. For instance, depending on the application, instrument configuration or even individual preferences, users might want to use a different GIS precursor to either protect the region of interest or to perform the welding to the support post. Also, instead of annular milling, other shaping strategies [5] or combinations might be preferred.

In any case, superb low-kV FIB performance and the ability to control the APT sample shaping process live by high-resolution SEM imaging facilitate the site-specific preparation of the highest-quality APT samples.

References

[1] D. Blavette et al., An atom probe for three-dimensional tomography, Nature 363 (1993), pp. 432-435.

[2] T. F. Kelly et al., Invited review article: Atom probe tomography, Rev. Sci. Instrum.78(3), (2007), pp. 031101–031120.

[3] D.J. Larson et al., Field-ion specimen preparation using focused ion-beam milling, Ultramicroscopy 79 (1999), pp. 287-293.

[4] M.K. Miller et al., Strategies for fabricating atom probe specimens with a dual beam FIB, Ultramicroscopy 102 (2005), pp. 287-298.

[5] F. Pérez-Willard et al., Focused ion beam preparation of atom probe specimens containing a single crystallographically well-defined grain boundary, Micron 39 (2008), pp. 45-52.

[6] Zhao et al., Segregation assisted grain boundary precipitation in a model Al-Zn- Mg-Cu alloy, Acta Mater. 156 (2018) 318-329.

[7] B. Bonef et al., Composition Analysis of III-Nitrides at the Nanometer Scale: Comparison of Energy Dispersive X-ray Spectroscopy and Atom Probe Tomography, Nanoscale Research Letters 11 (2016), pp. 461-466.

[8] P.J. Felfer et al., A Reproducible Method for Damage-Free Site-Specific Preparation of Atom Probe Tips from Interfaces, Microsc. Res. Tech. (2011).

[9] A. Grenier et al., Three dimensional imaging and analysis of a single nano-device at the ultimate scale using correlative microscopy techniques, Appl. Phys. Lett. 106 (2015), 213102.

[10] I.E. Carroll et al., New frontiers in atom probe tomography: a review of research enabled by cryo and/or vacuum transfer systems, Materials Today Advances 7 (2020), 100090.

[11] A.A. El-Zoka et al., Enabling near-atomic-scale analysis of frozen water, Sci Adv. 6 (2020), pp. 1-11.

[12] This work was performed on the NanoCharacterisation PlatForm (PFNC) of CEA-Grenoble, and was supported by the "Recherches Technologiques de Base" Program of the French Ministry of Research.

[13] Samples were fabricated by CNRS-CRHEA. This work was funded by DUVET ANR-17-CE08-0024-02.

Not for therapeutic use, treatment or medical diagnostic evidence. Not all products are available in every country. Contact your local ZEISS representative for more information.
EN_42_013_330 | CZ 03-2021 | Design, scope of delivery and technical progress subject to change without notice. | © Carl Zeiss Microscopy GmbH

Carl Zeiss Microscopy GmbH
07745 Jena, Germany
microscopy@zeiss.com
www.zeiss.com/microscopy