Microsc. Microanal. 13, 428–436, 2007
DOI: 10.1017/S1431927607070845

Microscopy AND Microanalysis

Review of Atom Probe FIB-Based Specimen Preparation Methods

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Abstract: Several FIB-based methods that have been developed to fabricate needle-shaped atom probe specimens from a variety of specimen geometries, and site-specific regions are reviewed. These methods have enabled electronic device structures to be characterized. The atom probe may be used to quantify the level and range of gallium implantation and has demonstrated that the use of low accelerating voltages during the final stages of milling can dramatically reduce the extent of gallium implantation.

Key words: atom probe tomography, specimen preparation, focused ion beam

Introduction

Specimen preparation with a scanning electron microscope/ focused ion beam (FIB) miller is an alternative and supplementary method to the traditional electropolishing/chemical polishing methods that have been used to fabricated the needle-shaped specimens required for atom probe analysis. These FIB-based methods enable specimens to be fabricated from site-specific regions to select microstructural features such as coarse or low volume fraction precipitates, grain boundaries, and other interphase interfaces. In addition, specimens may be fabricated from implanted regions, thin films, and coarse scale inhomogeneous materials such as dual phase materials and dendritic regions. FIB-based methods also extend the range of starting geometries to thin ribbons, sheets, fibers, powders, and films on substrates and also preparation of specimens from materials that are not easily chemically or electrochemically polished. As a result, the range of materials that can be prepared for characterization with atom probe tomography (APT) is greatly expanded through the use of FIB-based methods. A summary of these methods is presented in the current work. In addition, the deleterious effects of high energy gallium ion bombardment, including methods of ameliorating these effects, on the specimen surface are discussed.

Received December 6, 2006; Accepted June 13, 2007.
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Goal

The desired specimen shape for an atom probe specimen is a smoothly tapered needle with a circular cross section and an end radius of ~50–150 nm. The circular cross section is particularly important, as it is a major factor in proper data reconstruction. For example, the noncircular cross section in a wedge-shaped specimen results in variations in magnification along the minor and major axes (Larson et al., 1999*b*). FIB-based techniques also permit accurate selection of the specimen radius and, to a lesser extent, taper angle through the selection of appropriate inner annular diameters and focus conditions, and bitmaps or script-based slice and rotate methods, respectively. These selections may be used to tailor or to maximize the lateral extent of the volume of analysis, especially in wide field-of-view instruments. The length of the specimen should be sufficient to prevent field shielding from any sample support structure or base of the specimen. This requirement is not as critical in the case of local electrode types of atom probes (Kelly & Larson, 2000) due to the close proximity of the field-defining local electrode, and a minimum free length of ~5 μm appears to be sufficient. Bitmaps may also be used to precisely sculpt the shank of the specimen into a cusp shape to improve mechanical stability and electrical and thermal conductivity through the use of a wide-based specimen (Miller et al., 2005).

Due to the small size of an atom probe specimen, *in situ* and *ex situ* handling of the specimen are also important factors. Therefore, most methods involve the use of electro-

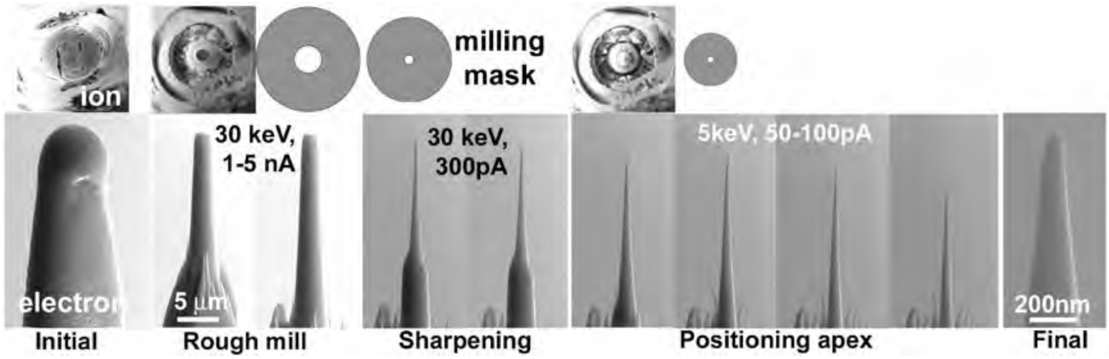

Figure 1. Example of the final annular milling stage of atom probe specimen preparation. The size of the annular mask and the ion current are decreased as shown during the annular milling procedure.

polished copper or aluminum tubes on which to mount the specimen directly or, in the case of the lift-out methods, to hold a support structure such as a needle. Common carriers for FIB and atom probe stages as well as the transmission electron microscope holders are desirable to minimize handling or remounting of the prepared delicate atom probe specimen.

Annular Milling

The simplest forms of materials amenable to FIB-based methods are small ($\lesssim$10 μm) diameter fibers, whiskers, nanowires, or roughly electropolished needles. In these cases, the annular milling method (Larson et al., 1998, 1999*a*), illustrated in Figure 1, may be used. In this method, the specimen is mounted in a copper tube and oriented precisely end on to the ion beam. An annular mask is then used to mill away the edge of the fiber, wire, and so forth. The outer diameter of the mask is chosen to be slightly larger than the maximum diameter of the specimen to avoid the generation of multiple sharp protrusions along the shank of the specimen. This procedure is generally performed in a series of steps, with decreasing inner and outer diameters and ion currents. To maximize the milling rate, milling should be performed concentrically from the outer diameter to the inner diameter of the mask as the milling rate is greater on an exposed edge. Typically, this procedure can be completed in ~30 min. Specimens with too small an end radius can result from this procedure if too small an inner diameter is selected or if the ion beam is not properly focused. This annular milling method is particularly useful in correcting wedge-shaped electropolished specimens, improving multiple phase specimens that exhibit greatly different electropolishing rates for the different phases (Larson et al., 1998; Miller et al., 2006) or simply resharpening blunt or previously analyzed specimens. At lower accelerating energies (i.e., 2–5 keV), it may also be used to remove contamination or surface films from specimens and to prepare tips for scanning tunneling microscope probes, nanoindentors, and so forth (Vasile et al. 1991).

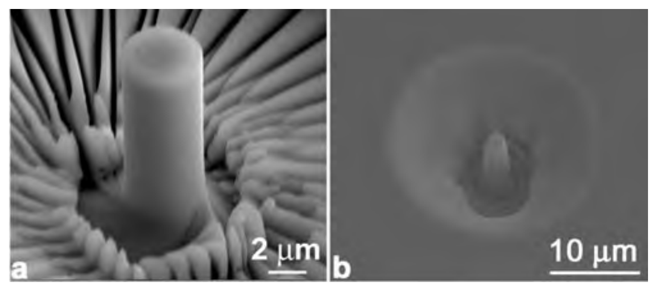

Figure 2. Examples of the script-based and circumferential milling methods to fabricate posts. **a:** Courtesy B. Schuster, U.S. Army Research Laboratory.

Annular milling may also be directly applied to multitips and posts produced by the Bosch etching process (Larson et al., 2001; Thompson et al., 2004). Prior to annular milling, these posts and multitips may be used as substrates to deposit multilayer films, and so on. Posts may also be fabricated by the slice and rotate script-based method (Uchic et al., 2004) or a circumferential milling method (Giannuzzi et al., 2006), as shown in Figure 2a,b. In these methods, milling is performed on an exposed edge to increase milling efficiency and to minimize the redeposition of material. Although moat-type posts (Miller et al., 2005) have been fabricated, the excessive milling times (>10 h) and redeposition issues generally discourage their use. In addition, these post-based methods result in embedded needles that are only suitable for local electrode configurations due to the proximity of the base of the post and the field shielding from the sides of the trench. It is considerably more efficient to extract the region of interest with a micromanipulator and mount it to a preformed needle (Thompson et al., 2007).

It is a simple extension to fabricate specimens from ribbons (Larson et al., 1998; Miller & Russell, 2007), thin

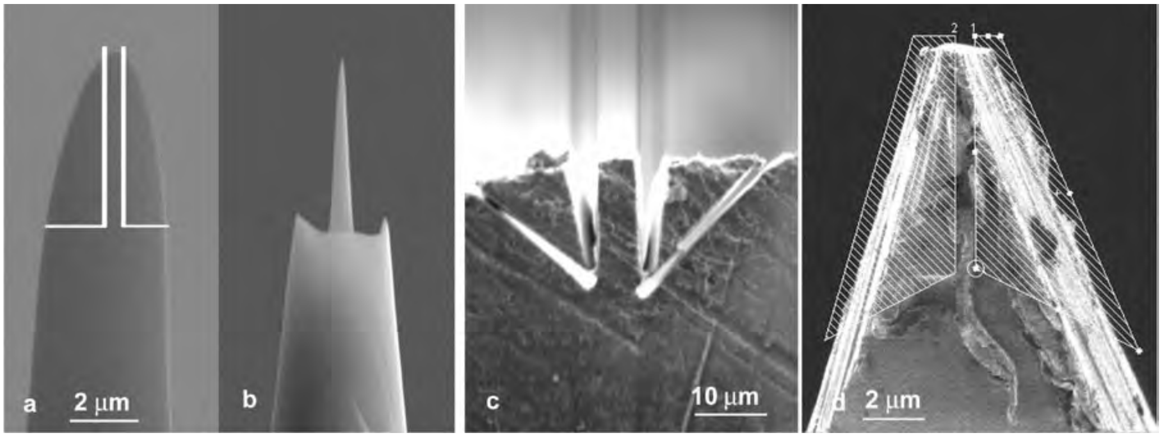

Figure 3. Examples of fabricating posts from (**a**, **b**, and **d**) ribbons and a thin sheet (**c**) with line cuts and polygon patterns.

sheets (Colijn et al., 2004; Lawrence et al., 2006), and even 3-mm-diameter transmission electron microscopy disks with a series of line cuts, as shown in Figure 3. The thin sheets with an optimal 3–5-μm end thickness are most easily produced by the tripod polishing method (Anderson and Klepeis, 1997). An *in situ* manipulator is often required to assist in the removal of the surplus material (Fig. 3c) due to redeposition or charging/electrostatic issues. It is often more efficient to use polygon patterns (Fig. 3d) to mill the entire unwanted area despite the longer milling times. Posts have also been fabricated in individual 0.5–100-μm-diameter powder particles and flakes produced by ball milling (Miller & Russell, 2006), as shown in Figure 4. In the case of powders, an individual particle is selected from particles spread out on an adhesive-coated mount and then attached *in situ* to the end of a wire needle or nanoprobe with platinum or tungsten deposit. The mounted particle is then reoriented *ex situ* and then annular milled into a post. In all these post methods, the final sharpened needle is produced by annular milling.

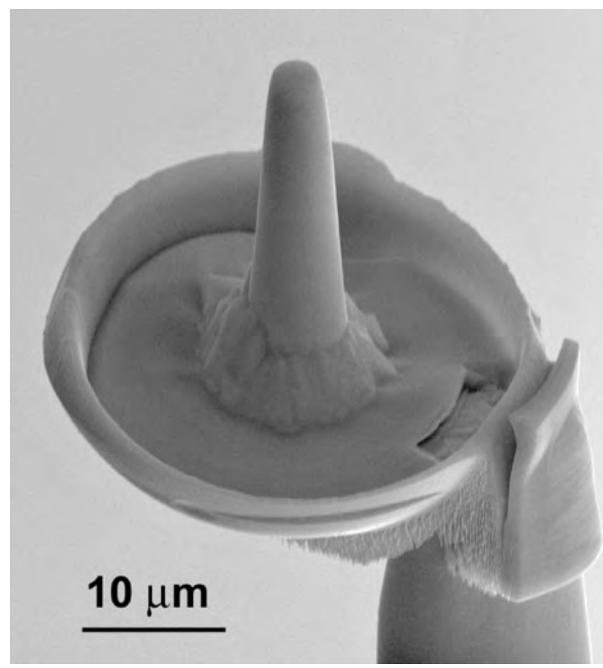

Figure 4. A post fabricated from an individual 40-μm-diameter gas atomized powder particle.

Site-Specific Methods

Although these *in situ* post methods may be performed at site-specific locations, a lift-out (LO) method is generally more applicable and efficient. Several different variants have been developed for atom probe specimens (Miller et al., 2005; Cairney et al., 2007; Miller & Russell, 2007; Saxey et al., 2007, Thompson et al., 2007). The choice of method depends on the geometry of the feature of interest as well as milling times and ease of use. These variants are generally derivatives of the methods used in the fabrication of transmission electron microscopy specimens (e.g., Giannuzzi et al., 1998; Giannuzzi & Stevie, 1999, 2004). Thin deposits of platinum or tungsten are generally used to protect the specimen during the milling. Surface damage may be minimized by initially depositing the metal layer with the electron beam followed by a thicker layer with the ion beam (Kempshall et al., 2002).

The LO methods all entail cutting out a small volume that contains the feature of interest. To maximize throughput, the approach should also minimize the required milling time and the number of steps in the procedure. A typical block-shaped coupon may be milled with the method (Miller et al., 2005) shown in Figure 5. In this method, trenches are milled on three sides, and the specimen is then tilted to cut the underside. The partially cut coupon is then

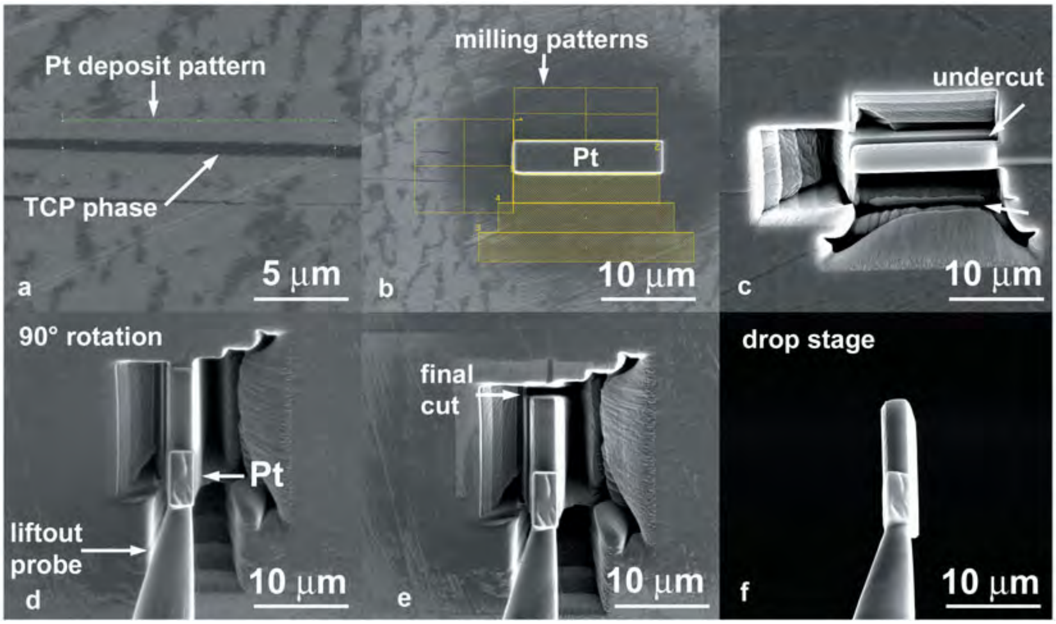

Figure 5. Lift-out method for site-specific specimen preparation. **a:** Selection of feature of interest (TCP phase). **b:** After Pt deposition over feature of interest and positioning the milling patterns for the next stage. **c:** After tilting and making the undercut. **d:** After rotating 90° and attaching the lift-out probe to the coupon with Pt deposit. **e:** After final cut. **f:** After dropping the stage to release the lift-out coupon.

attached to a support (usually a thin pointed wire) and the final side is cut free. The long axis of the coupon may be chosen to be along or perpendicular to the surface depending on the orientation of the feature of interest.

An alternative total release method, as applied to atom probe specimens (Thompson et al., 2007), is shown in Figure 6. This method entails milling two intersecting line or rectangular cuts by tilting the specimen with respect to the ion beam and rotating the specimen 180° between the cuts. The first cut is generally repeated to ensure that redeposited material from the second cut does not reattach the extracted wedge of material. One end of the wedge may be cut free. The wedge is then attached to a support and the final side is cut free. Alternatively, both ends may be cut free before attachment. This method produces a triangular shaped wedge that may be attached to a support post *in situ* (Thompson et al., 2007) or *ex situ* (Saxey et al., 2007). Multiple specimens may be cut from one long aspect ratio coupon in the case of multilayer or thin film specimens. These lift-out methods typically take ~1–2 h per coupon. As with the post methods, the final sharpened needle is produced by the annular milling procedure.

The visibility of the feature of interest in the selection of the volume and during annular ion milling is critical for these lift-out methods. The dual beam instruments have several imaging modes with both the electron and ion beams coupled with secondary electron, backscattered electron, secondary ion, and energy dispersive X-ray (EDS) detectors. All of these modes may be used to initially identify and select the feature of interest. For example, imaging with the focused ion beam can generate different contrast and highlight the grains. Secondary electron, backscattered electron, and EDS detectors may be used to distinguish and to identify phases. After the feature of interest is located on the surface, its actual three-dimensional location within the specimen must be established so that the key region of interest is located properly within the needle during the final milling process. In some cases, such as grain boundaries and plate-shaped precipitates, a trench perpendicular to the interface may be milled to establish the orientation and thereby adjust the position of the volume to be milled. In other cases, the selection is a combination of the lift-out stage and the final annular milling stage. An example of this LO procedure is shown in Figure 7. The area of interest in the device is indicated by an arrow in Figure 7a. Two orthogonal views of the sample after thinning to a size of ~1000 × 500 nm are shown in Figure 7b,c. The final selection process as produced in the annular milling stages is shown in Figure 7d–f.

The selection of the volume also defines the orientation of the final atom probe specimen. For example, it may be advantageous for the major axis of the needle to be parallel or perpendicular to surface layers, interfaces, or grain boundaries. The former orientation should yield a larger number of atoms at a particular distance from the interface to improve the counting statistics for low concentration solutes or should permit the investigation of solute fluctuations along an interface. The latter orientation should provide

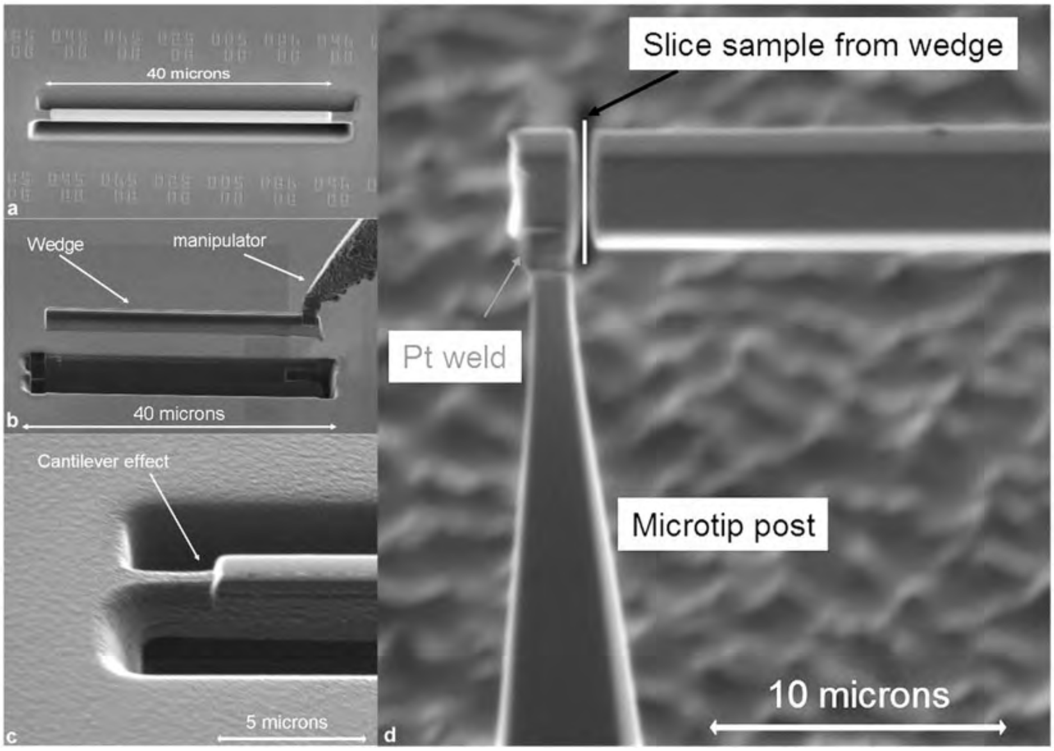

Figure 6. **a:** Initial mill cuts for wedge lift-out from a Si device wafer. **b:** Close-up image showing the wedge still attached to the Si wafer. The long wedge forms a cantilever type structure. **c:** Wedge extracted from the wafer. **d:** Sample wedge mounted to a microtip structure and sliced free from the remaining wedge (Thompson et al., 2007).

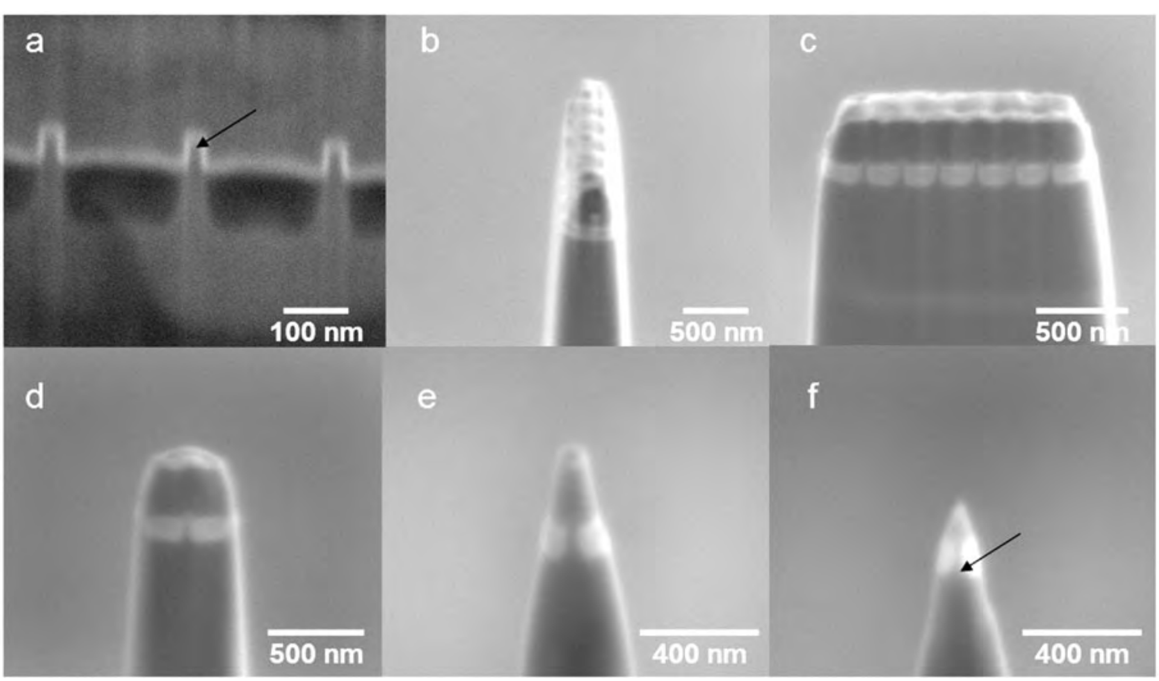

Figure 7. Sequence of SEM images showing FIB LO and sharpening from a transistor-type test device.

the highest spatial resolution with distance from the interface but with a more limited field of view. This orientation is also more susceptible to failure in brittle materials. Alternatively, the extracted material may be mounted upside down in order to reverse the order of the analysis. For example, this orientation may be beneficial if it positions a low evaporation field material before a high evaporation field material in order to minimize material loss or potential specimen fracture during the transition between the two regions of different evaporation fields. It may also enhance the analysis because one could conceivably produce nearly identical data sets analyzed in reverse (or, in fact, arbitrary) directions.

Although the blank produced by some of the LO procedures such as the total release method may be retrieved *ex situ* to the microscope, it is more efficient to use *in situ* nanomanipulators. A variety of nanomanipulators are commercially available. These nanomanipulators fall into two classes: stage mounted that can tilt with the stage and static (i.e., not tiltable) chamber mounted. Allowance for these different classes of nanomanipulators has to be taken into account in the milling and mounting/attaching procedures. Multiple nanomanipulators are also a possible, albeit expensive, option. A variety of options such as wire probes, sharpened points, and tweezers are available for attachment of the blank to the support. Some methods involve the transfer of the coupon to a support via the nanopositioning probe, whereas others actually make use of the end of the nanopositioning probe as the support (Miller & Russell, 2006). Because good electrical and thermal conductivity between the atom probe specimen and the support are essential, platinum or tungsten deposits are invariably used for all attachments.

The sample volume in the apex of the needle is generally insufficient to get a good signal-to-noise ratio image from most of the detectors during the final stage of milling. The large mass difference of the protective platinum/tungsten cap compared to most specimen materials may be used as a fiducial marker to indicate the position of multilayer films, implanted layers, and so forth. The precise position of the apex of the specimen along the shank of the needle may be adjusted with a broad beam at low ion current and accelerating voltage (Thompson et al., 2006). This operation tends to sharpen the apex of the needle. The simultaneous milling and imaging mode may be used to terminate milling at the desired location along the shank of the needle.

Gallium Implantation Issues

As with all specimen preparation techniques, some factors have to be taken into account in order to ensure reliable analysis of the microstructure and to ensure that no artifacts are introduced during the fabrication process. It is well known that FIB-based techniques implant gallium into the surface of the specimen and can produce damaged regions and amorphization of crystalline materials (Larson et al., 1998). Gallium implantation also puts atom probe specimens under additional stress due to the large size of the gallium atom compared to most elements. This additional internal stress can promote failure in brittle materials under application of the high field necessary for field evaporation. Therefore, steps are normally taken to minimize the level of gallium implanted into the specimen (Kempshall et al., 2002).

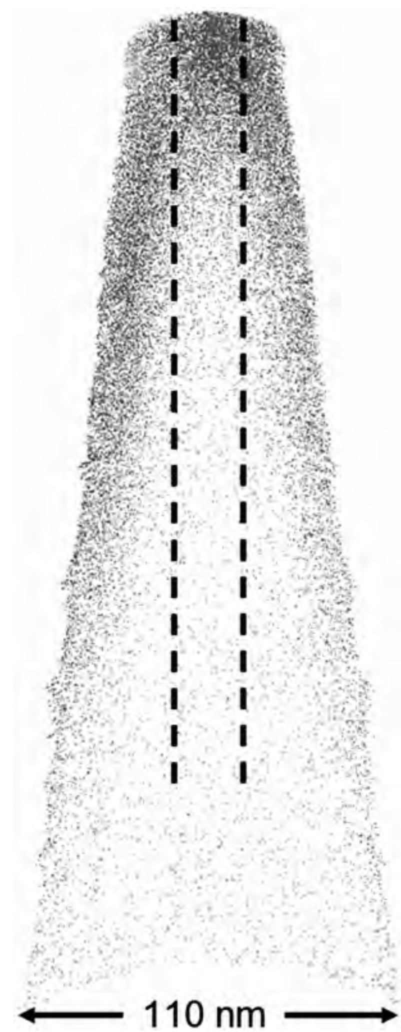

Figure 8. Atom map of a Si specimen sharpened in a FIB with 30-keV Ga ions. For clarity, only the Ga atoms are shown. This data set was obtained with an 80-mm detector. The dashed lines represent the field of view attainable with a 40-mm-diameter detector.

The extent and level of the gallium implantation may be directly measured in an atom probe analysis. An example of gallium implantation in a Si specimen obtained in a wide field-of-view atom probe is shown in Figure 8. It is evident that the gallium is located in a thin shell around the edge of

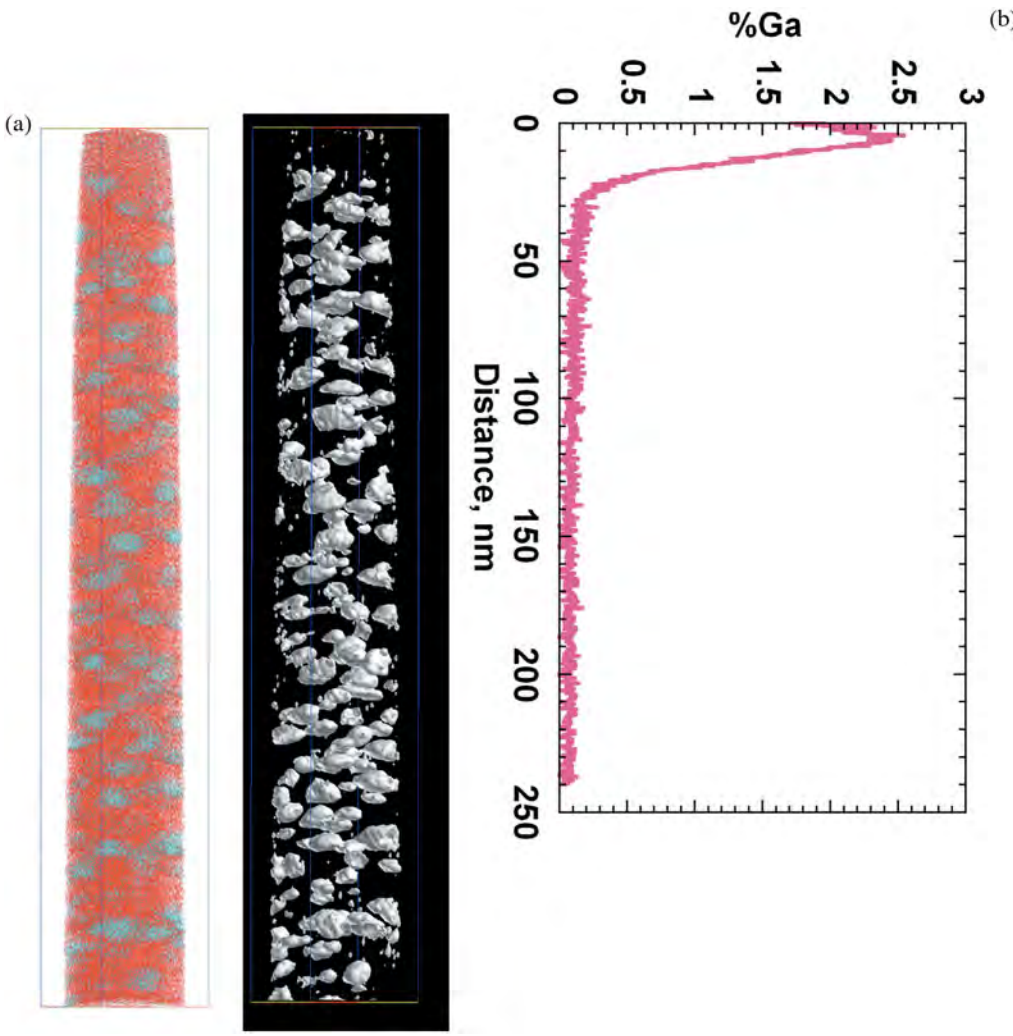

Figure 9. Example of the level of implanted gallium and the dissolution of TiC particles in the surface layer of an $Fe_{14}Nd_2B$ magnet. Courtesy Y.Q. Wu, Ames Laboratory.

the needle. The range of the gallium ions for different accelerating voltages and incident angles may be predicted for different materials with the stopping and range of ions in matter (SRIM) simulation (Ziegler et al., 1977–1985). For example, the damage that gallium may produce is shown by the dissolution of the TiC precipitates in the top 10 nm of a $Fe_{14}Nd_2B$ magnet material (Ziegler et al., 1977–1985) in Figure 9a. The high level of gallium in the damaged region is clearly shown in the concentration profile in Figure 9b. In transmission electron microscopy, the electron beam has to penetrate the full thickness of a specimen, and, therefore, the gallium implanted regions are an inherent part of the specimen from which data are collected. In atom probe tomography, this region of the specimen may simply be discarded from further analysis without affecting the remainder of the data.

Dual-beam instruments enable the level of gallium implantation to be minimized as the majority of the imaging may be performed with the electron beam rather than the gallium ion beam. However, some imaging with the gallium beam, such as focusing and imaging to select the milling location, is unavoidable. The use of low accelerating voltages (i.e., 2–5 keV) during the final stages of milling can dramatically reduce the level of gallium implantation (Fig. 10; Thompson et al., 2006, 2007). A protective platinum or tungsten cap can also be deposited on the surface of the specimen prior to milling so as to remove the gallium implanted region in the centerline of the specimen to a benign region in the cap. The surface layer of the specimen may also be removed by milling with a low energy argon ion beam (Larson et al., 2000). This final ion milling step usually has to be performed in another system.

Several challenges still remain with FIB-based atom probe specimen preparation. Of particular concern is the relatively low success rate with the lift-out procedures. Routine methods are required to ensure that the region of

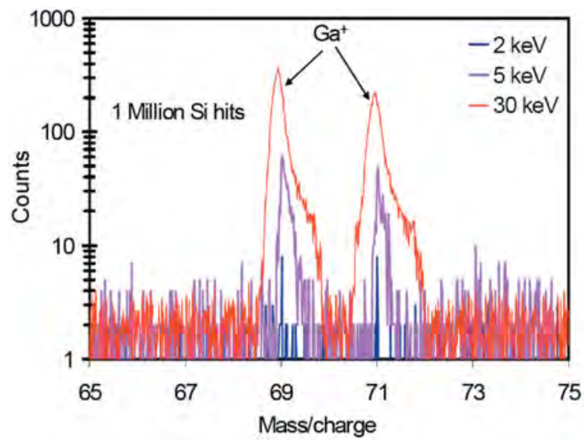

Figure 10. Mass spectrum in the Ga region for 2, 5, and 30 keV accelerating energies. One million Si atoms were sampled for each spectrum once the tip was properly aligned to the local electrode.

interest is positioned precisely in the apex region of the tip and to prevent preferential channeling of gallium (and the subsequent embrittlement) along grain boundaries or interfaces. A better understanding is required of the mechanical and electrical conductivity issues associated with the attachment of the lift-out coupon to the support.

Summary

A variety of FIB-based methods have been developed to produce the needle-shaped atom probe specimens from a variety of forms of materials and from site-specific locations. Gallium implantation is minimized in these methods through the use of platinum or tungsten FIB-deposited capping layers and by the use of lower ion energies during the final milling stages. Lift-out methods enable the preparation of atom probe specimens from very specific regions. These methods have been applied to electronic device structures and are thus enabling the successful atom probe analysis of these structures for the first time.

Acknowledgments

The authors thank L. A. Giannuzzi and R. Young of FEI, Y. Q. Wu, Ames Laboratory, B. Schuster of the U.S. Army Research Laboratory, and D. Lawrence, R. Ulfig, and S. Gerstl of Imago Scientific Instruments for their assistance and many helpful discussions. Research at the Oak Ridge National Laboratory SHaRE User Facility was sponsored by Basic Energy Sciences, U.S. Department of Energy.

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