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Improved Focused Ion Beam Target Preparation of (S)TEM Specimen—A Method for Obtaining Ultrathin Lamellae

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Abstract: Specimen quality is vital to (scanning) transmission electron microscopy (TEM) investigations. In particular, thin specimens are required to obtain excellent high-resolution TEM images. Conventional focused ion beam (FIB) preparation methods cannot be employed to reliably create high quality specimens much thinner than 20 nm. We have developed a method for *in situ* target preparation of ultrathin TEM lamellae by FIB milling. With this method we are able to routinely obtain large area lamellae with coplanar faces, thinner than 10 nm. The resulting specimens are suitable for low kV TEM as well as scanning TEM. We have demonstrated atomic resolution by C_s-corrected high-resolution TEM at 20 kV on a FIB milled Si specimen only 4 nm thick; its amorphous layer measuring less than 1 nm in total.

Key words: FIB, target preparation, low voltage, TEM, STEM, sample preparation

Introduction

Aberration-corrected high-resolution transmission electron microscopy (AC-HRTEM) is currently undergoing revolutionary changes in its ability to image materials at the atomic level using medium energy electrons (Rose, 2008). Scientists have rediscovered the advantages of using low energies in TEM; it dramatically reduces knock-on damage for low-Z number material and enables new results in imaging and electron energy loss spectroscopy (Kaiser et al., 2011). While the instruments are constantly being improved, there has been little fundamental progress in sample fabrication techniques. This has led to the awkward situation where often sample quality is the limiting factor in TEM investigations. In particular, low voltage TEM requires extremely thin specimens virtually free of preparation artifacts. In any voltage range, thin specimens—with thicknesses smaller than half the extinction length—are required to obtain HRTEM images with weak phase contrast. In general, contrast interpretation of these images is greatly simplified for thin samples fulfilling kinematic conditions because dynamic scattering events are absent. While some nanomaterials such as graphene and nanotubes inherently fulfill these conditions, TEM investigations would be greatly advanced if high quality ultrathin specimens can be prepared from arbitrary bulk materials.

Indeed there is a large variety of classical sample preparation techniques such as fracturing, mechanical polishing, broad ion milling, etc., that can be used to prepare high quality TEM specimens (Walck, 1996; McCaffrey & Barna, 1997). However, they all share two major drawbacks: specimen faces are not coplanar and lamellae can be prepared only with limited site specificity. Problems such as failure analysis of complex samples, e.g., of a defective gate oxide in a semiconductor device, require target preparation methods to image a specific region of interest (ROI).

Focused ion beam (FIB) methods for TEM sample preparation have been developed to address these problems. They are fast replacing conventional techniques when speed and site specificity are required (Leer & Giannuzzi, 2008). Despite recent improvements, specimen thickness and quality obtainable are not satisfactory for the latest corrected microscopes, let alone low voltage TEM. Scaling down lamella thickness using conventional FIB techniques is hampered by three detrimental effects: warping, amorphization, and shrinkage. Warping is caused by intrinsic or process induced strain when a lamella is thinned below a certain threshold. Special mounting and adaptive milling techniques have been proposed to reduce this effect (Langford et al., 2002). Nevertheless mechanically stable large coplanar transparent areas have not been obtained. Amorphization is a side effect of ion milling (Mayer et al., 2007). Depending on angle of incidence and energy, the lamella is amorphized to a certain depth (e.g., ~30 nm in Si for 30 keV Ga ions at glancing milling angle). Self-evidently, amorphization depth has to be kept much smaller than half the lamella thickness. This is achieved by polishing the lamella with ions of reduced energy. In conventional processing this causes difficulties because the beam profile degrades significantly with reduced energy. This effect is usually counteracted by choosing steeper milling angles at the cost of introducing material dependent sputtering effects. Shrinking of the lamella happens during the final thinning process. With the removal of only a few tens of nanometers in thickness, the height of the lamella's transparent area can be reduced by several micrometers. This effect has to be counteracted by depositing very thick protective layers (Kang et al., 2010), a very time-consuming process at best.

In consequence, conventional FIB preparation methods cannot be employed to reliably create TEM lamellae much

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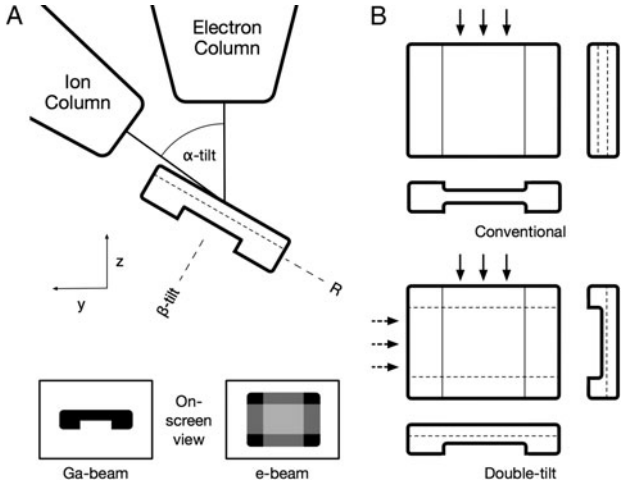

Figure 1. A: Dual beam column arrangement. **B:** Orthographic third angle projection of thinned lamellae. The direction of the Ga ions is indicated by arrows. Top: Conventional *in situ* lift-out method. Bottom: Double-tilt method. The electron transparent area is created where the two milling grooves intersect.

thinner than 20 nm. To obtain thin high quality specimens suitable for aberration-corrected TEM, a new sample preparation technique is necessary. We propose an elegant method that is a significant advancement over the conventional lift-out technique (Langford & Clinton, 2004).

Materials and Methods

Sample fabrication was performed in a Zeiss NVision 40 CrossBeam™ FIB instrument (Carl Zeiss NTS, Germany) incorporating a Ga liquid metal ion source, *in situ* scanning electron microscope (SEM) imaging using a thermal field emission source, and an Ar plasma ion source. The energy of the Ga ions can be varied in the range from 1–30 keV while the Ar source can be operated from 0.1–1 kV. Figure 1 shows a schematic drawing of the microscope.

TEM lamella preparation by the FIB lift-out method is carried out in four steps (Langford & Clinton, 2004): (1) rough milling of a lamella, (2) lift-out, (3) thinning, and (4) polishing. During rough milling, a free-standing beam containing the ROI is created by removing material around it through FIB milling. Then, for lift-out, a manipulator is attached to the beam using ion beam induced deposition (IBID). Subsequently, the beam is cut free from the bulk. Using the manipulator, the resulting lamella is transferred to a special TEM lift-out grid. Once it is firmly attached by IBID, it can be thinned further using high energy ion milling. To reduce the resulting damage layer (~30 nm for 30 keV Ga ions in Si), the Ga ion energy is reduced for one or more polishing step(s). In our FIB system a final low kV argon polishing step can be performed to remove residual Ga contamination.

Figure 1 also shows schematic drawings of the thinning and polishing process for the conventional and our new *in situ* lift-out technique. In both cases, the lamella faces are milled under a glancing angle (1°–3°, depending on milling current) to obtain coplanar surfaces. In conventional thinning, material is removed top-down from one or both sides of the lamella. Polishing is done similarly, until the desired final thickness is reached.

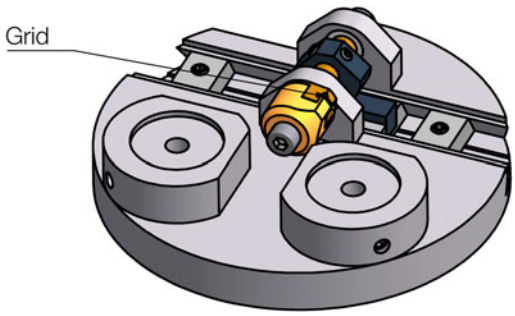

Figure 2. Schematic drawing of the rotation-tilt holder. Bulk samples are placed on the two mounting points for SEM specimen stubs. The lift-out TEM grid is mounted on a pivot pin that is attached to a cog wheel. The wheel gears into the cograil of a sliding counterweight. Movement of the counterweight drives a ±90° rotational motion of the holding fixture.

The essential innovation of the double-tilt method is that final thinning and polishing is performed with a different milling direction for each side of the lamella. Material is removed from one side until the remaining part has about half the original thickness. Then the sample is tilted around its TEM observation direction by ~90° and, subsequently, thinned from the second side. This process creates a thin window where the two milling grooves overlap. For polishing, the process is repeated in several steps, gradually reducing ion energy, until the desired thickness is reached.

Conventional FIB/SEM systems are typically equipped with a 5- or 6-axis stage. It allows the sample to be translated in the *x*, *y*, *z* directions of an orthogonal coordinate system. In addition, the sample holder can be continuously rotated in the *x*-*y* plane. For processing, the sample is brought into the coincidence point of electron and ion beam using the stage. There, the stage tilt axis (alpha-tilt) can be used to tilt the sample mount in the plane spanned by the SEM and the FIB column (optics plane). This allows the specimen surface to be oriented perpendicular to either SEM or FIB beam. To minimize apparent lateral movement of the sample during tilting, the eucentric point of the rotational axis is either placed in the coincidence point of the two beams or, in the case of a six-axis stage, is adjusted independently. Thus, the microscope stage only allows the sample to be tilted in the plane spanned by the electron and gallium beam. It does not, however, permit the orthogonal tilt (beta-tilt), which is essential for the preparation method described above. To solve this problem, a novel specimen holder was designed.

Figure 2 shows an isometric rendering of the rotation-tilt holder. The holding fixture in the center of the assembly accepts a standard lift-out TEM grid. It is mounted on a pivot pin that is attached to a semicircular pinion. The wheel gears into the cog rail of a sliding counterweight. The travel of the counterweight along the dovetail groove is limited by stops on each side. They are adjusted so that the

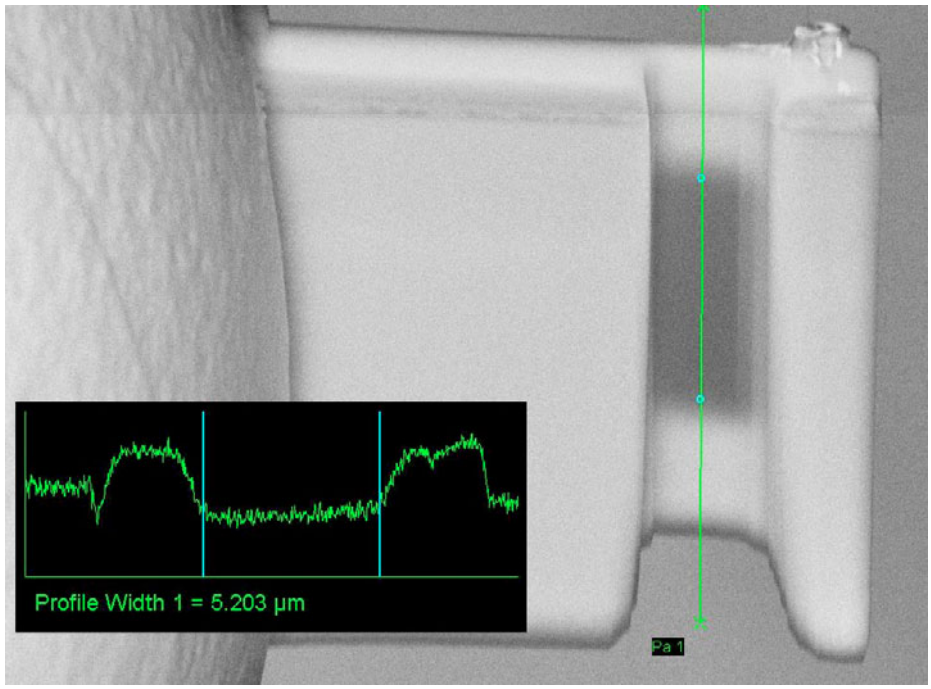

Figure 3. BSE image of the lamella during thinning recorded with the in-lens energy selective backscattered detector. The inset shows a line plot of the BSE intensity over the lamella. The electron transparent region of the lamella is encompassed by the vertical lines (circles) in the line plot (the image).

holding fixture can rotate ±45° from the normal. In addition, the holder has two mounting points for SEM specimen stubs. The entire assembly is dimensioned to fit through the instrument airlock for quick exchange and is equipped with a mating connector for the SEM/FIB stage.

The fabrication of a TEM specimen with the rotation-tilt holder is largely similar to that using a conventional holder. The notable exception being that after lift-out the prethinned lamella is typically attached to the grid at a beta-tilt angle of 45°. Next, the first side of the lamella is thinned top-down, exactly as in the conventional process. After that, the microscope stage is rotated a full 180° for causing the second side of the specimen to face the SEM beam. Since the stage is tilted toward the FIB column (alpha-tilt $> 50°$), the counterweight slides downward halfway during the rotation. This movement, in turn, rotates the pivot pin: resulting in a beta-tilt of 90°. After completing rotation and tilt movement, the lamella again ends up in the coincidence point. Then the second side is thinned orthogonally to the first milling groove but with the same alpha-tilt.

Lamella thickness is monitored by measuring changes in the backscattered electron (BSE) intensity using the in-lens energy selective backscatter detector during milling (Salzer et al., 2009). The same approach is also used to adjust the orientation of the second side, making it coplanar to the first. Figure 3 shows a BSE image of a lamella recorded during thinning. Any misalignment between the two sides can be detected by measuring the BSE intensity along two orthogonal lines across the transparent region. Horizontal deviations are adjusted by stage or FIB image rotation. Vertical misalignment is corrected using stage alpha-tilt. While the lamella is thinned, the electron beam energy is progressively reduced to provide maximum sensitivity to thickness changes. In this way, planarity of the window can be effortlessly adjusted to well below 1° deviation—enough to, in principle, obtain an atomically flat specimen over a relevant field of view. The thinning is stopped when the remaining window thickness approaches twice the thickness of the amorphization depth of the specimen material.

The desired final thickness is achieved by removing the amorphous damage layer from both sides in several polishing steps during which the Ga ion energy is gradually reduced down to 1 keV. An optional final Ar ion polishing step can be performed to remove residual Ga contamination. Switching between milling directions by rotating the sample takes place reliably, and the milled side is always facing the SEM for visual process control.

The fabricated lamellae were inspected by TEM and scanning TEM (STEM) imaging. An aberration-corrected and monochromated Libra 200 operating at 20 kV was used for HRTEM imaging of the ultrathin silicon lamella. Electron energy loss spectra (EELS) were acquired with the integrated in-column energy filter, and sample thickness was determined by Kramer-Kronig analysis (for more details, specifications, and performance of the dedicated low voltage TEM platform, see Kaiser et al., 2011). STEM images of the GaN lamella were acquired using a FEI Titan (S)TEM operating at 300 kV (FEI Company, Hillsboro, OR, USA) equipped with a Fischione high-angle annular dark-field (HAADF) detector (Fischione Instruments, Export, PA, USA). Low kV S(T)EM imaging was performed with the field-emission SEM of the NVision 40 microscope operated at voltages between 1–30 kV.

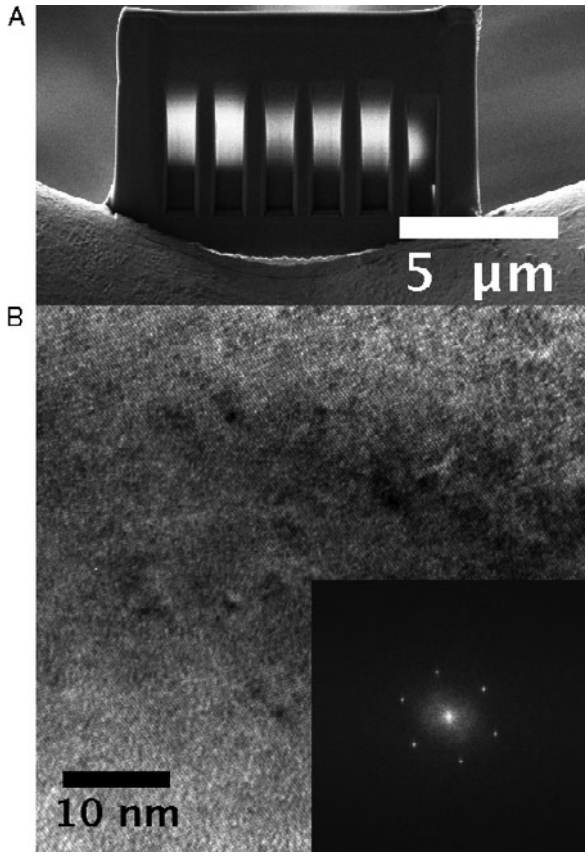

Figure 4. A: A 3 kV secondary electron SEM image of a FIB prepared windowed Si lamella. **B:** A HRTEM image of the second window (from left) demonstrating atomic resolution at 20 kV. The inset shows the corresponding power spectrum. The spotty contrast variations in the image are caused by strong dynamic diffraction contrast that cannot be avoided at 20 kV.

Results

Figure 4 shows TEM and SEM images of a Si lamella with several electron transparent windows. The secondary electron SEM image shows the layout of the lamella; the transparent windows appear bright. This lamella belongs to the batch of samples first fabricated as proof-of-concept, before the development of the double-tilt holder. Thus the windows were not created by the overlap of two orthogonal milling grooves. Instead rectangular recesses were milled into the lamella at an angle of ~30°. This, however, was only possible because the material is monocrystalline.[a] Nevertheless, the acquired results are directly comparable to those of the double-tilt method. After the localized thinning, the lamella was polished using Ga ions with energies as low as 1 keV. After that, a final Ar polishing step at 0.5 keV was performed. The thickness of the resulting contamination layer—less than 1 nm in total—was determined by cross-sectional TEM measurements of a similarly prepared specimen. The HRTEM image of the lamella was obtained with a spherical aberration-corrected TEM operated at 20 kV. The associated power spectrum (inset) clearly exhibits Si 100 reflexes; diffuse intensity rings, an indicator for amorphization, are notably absent. The background contrast variation in the HRTEM image is thus primarily attributable to surface roughness creating strong Bragg contrast at 20 kV. The total window thickness at the imaging location was determined to be 4 nm by EELS measurement (for details see Kaiser et al., 2011).

Figure 5 shows SEM images of a GaAs transistor structure before and after lamella fabrication with the double-tilt method. First is an in-lens secondary electron image (V_{acc} = 1 kV) of a FIB cross section through the transistor gate. The second SEM image shows a lamella of the same transistor after lift-out, thinning, and polishing. In the center, the electron transparent region (dark) created by the overlapping milling grooves—one parallel, one perpendicular to the substrate—can be seen. The last shows the dark-field transmission scanning electron image of the gate head recorded on a Zeiss 4-quadrant solid state detector (V_{acc} = 30 kV). Compared to the SEM image, there is a striking increase in resolution as well as additional information about material composition. Note in particular that the holes in the dielectric next to the base of the gate electrode did not enlarge during lamella preparation.

Figure 6 shows a 300 kV STEM HAADF image of a lamella cut from a GaInN quantum well structure deposited on the semipolar side facets of a GaN pyramid grown by selective metal organic vapor phase epitaxy (Wunderer et al., 2009). The front side has been thinned parallel to the left pyramid surface while the back side was thinned perpendicular to it. The size of the resulting electron transparent area exceeds 12 μm^2 (1.2 $\mu m \times 10$ μm). This allowed atomic scale measurement of the quantum well dimensions by STEM (see inset) along its entire length. Unfortunately, layering of the IBID Pt layer has caused some thickness variations ("curtaining") along the lamella. In addition, some contamination is visible in the lower left corner. This was attributed to material redeposition as a result of beam profile degradation caused by the finite depth of focus. For this reason, polishing steps at energies below 5 keV were omitted.

Discussion

Mechanical stability and small thickness are irreconcilable features in a conventional lamella: the thinner the transparent area is made, the more the mechanical stability of the entire lamella is degraded. This manifests in drastic bending, especially in lamellae that are attached to the lift-out grid on one side only. However, even in double clamped specimens, bending is observed eventually. Adaptive milling techniques have been proposed that follow the contour of a bent lamella and prevent the formation of holes. This is difficult to implement and provides very little compensation for three-dimensional deformations. In contrast, the window geometry of double-tilt lamellae leaves the transpar-

[a]In samples composed of different materials or grain orientations, this process does not result in a uniform lamella due to the resulting different sputtering yields when milling at this low incidence angle.

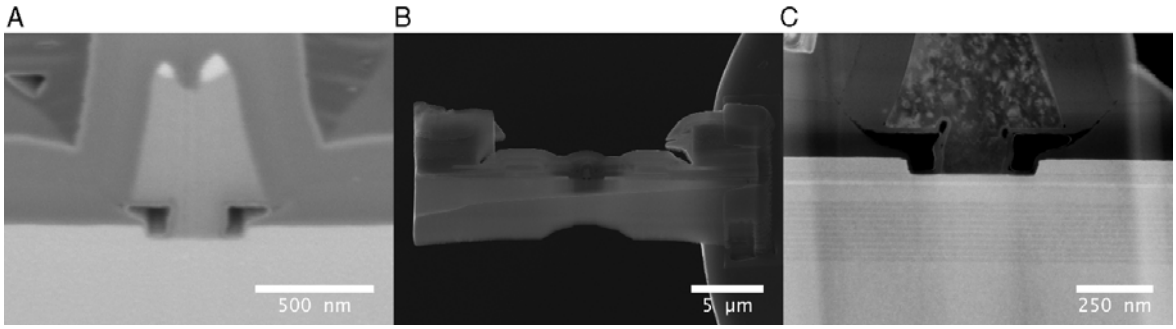

Figure 5. SEM images of a transistor gate structure before and after lamella fabrication with the double-tilt method. **A:** In-lens secondary electron image of a FIB cross section through the transistor ($V_{acc} = 1$ kV). **B:** Secondary electron image of the polished TEM lamella. In image **B**, the electron transparent region created by the overlapping milling grooves appears dark. **C:** Dark-field transmission scanning electron image of the gate head ($V_{acc} = 30$ kV).

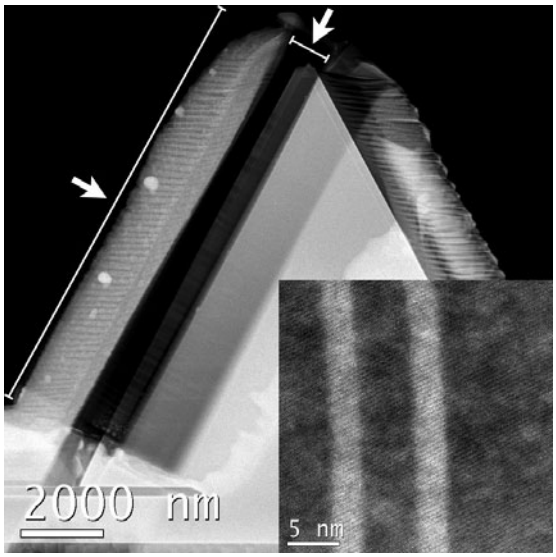

Figure 6. A 300 kV STEM HAADF image of a lamella cut from a GaInN quantum well structure deposited on the side facets of a GaN pyramid. The thinning and polishing directions of front and backside are parallel/perpendicular to the surface as indicated by the arrows. The width of the respective cuts is denoted by white indicators; the size of the resulting electron transparent area exceeds 12 μm^2. Inset: Atomic resolution STEM image of the quantum well structure.

ent area supported on all four sides by a sturdy frame. In result, we observe that even a very large ROI can be milled extremely thin without noticeable bending.

In conventional thinning and polishing, severe shrinkage of the lamella occurs. The effect is rooted in the finite sharpness of the beam rounding the upper edges of the lamella. When the lamella becomes progressively thinner, the ("missing") rounded regions start to overlap and the lamella shrinks in z direction. Geometrical sputtering effects amplify this shrinkage. First of all, sputtering yield depends very strongly on the ion incidence angle (Giannuzzi et al., 2005). Starting from normal ion incidence (90°), it increases to a maximum of around 5–15° and then decreases to almost zero for glancing angles. As a result sputtering off the top of the lamella is greatly increased: exacerbating the rounding/shrinking effect. Second, forward sputtering yield increases significantly when feature size becomes comparable to the ion penetration depth. This is important because even when the thinned part is still much thicker than the penetration depth—as it always should be to prevent total amorphization—parts of the rounded top of the lamella can satisfy the condition.[b]

These three effects combined cause strong shrinkage along the milling direction of a thin lamella, once its top becomes narrow compared to the penetration depth or beam diameter. For double-tilt lamellae the frame of the transparent window is always thick enough to prevent these effects from manifesting, and shrinkage is not observed.

Final polishing of the lamella is the most critical step in the fabrication process, mainly because at low ion energies beam profiles degrade significantly. While in modern FIB systems, sub-10 nm beam diameters are readily obtainable at 30 kV; the full-width at half-maximum of a 1 keV Ga beam can easily exceed several micrometers.

As a consequence, steeper polishing angles (>3°) must be used in conventional lamella fabrication methods to avoid excessive removal of material from the top of the lamella. Additionally, very thick sacrificial layers have to be used to achieve thin lamellae (Kang et al., 2010). Worse still, for larger milling angles the sputtering yield becomes material dependent (Giannuzzi et al., 2005). Therefore, creating very thin lamellae of homogeneous thickness from a composite material becomes impossible.

The suppression of geometric sputtering effects in the double-tilt method means that even thin protective IBID layers, typically ~1 μm thick, are sufficient for the thinnest of lamellae. More important, the double-tilt geometry lifts milling angle restriction for low kV polishing with broadened beam diameter. Being able to polish at 1°–3° incident angle reduces material selective sputtering effects as well as amorphization depth. Very thin, uniformly thick lamellae

[b]The effect is more commonly observed if a hole is present in a lamella. During thinning the hole grows much faster in diameter than the lamella shrinks in thickness. In analogy, the entire lamella can then be thought of as an inverse hole.

can be created even for material combinations with strongly differing sputtering yield, e.g., integrated circuits components.

The double-tilt method is able to both prepare high quality specimens from difficult sample geometries and improve specimen quality. Nonplanar, high aspect ratio specimens are notoriously difficult to prepare by conventional FIB techniques. The glancing angle milling requires a flat lamella top to obtain coplanar sidewalls. Thus corrugated samples have to first be flattened using fillers or IBID. In contrast, thinning with the double-tilt method is straightforward if milling angles can be adjusted to run parallel to the surface geometry. Samples fabricated with the double-tilt method also show improved thermal and electrical behavior in addition to their superior mechanical stability. The window-frame geometry suppresses heating/charging effects more efficiently due to the improved thermal/electrical contact with the thicker parts of the sample; though typically not a major concern, it can be a relevant factor in low kV TEM.

Thinning (and polishing) of one lamella side can be performed before lift-out and thus be automated using conventional TEM preparation packages. The same procedures, without much difficulty, could be adapted to automate the post lift-out processing of the other side. In principle, the double-tilt method can also be used to improve existing conventional lamellae, if they are still thicker than the high-kV amorphization thickness.

A possible advantage of conventional TEM sample preparation is that only a single protective IBID layer is used because both sides of the lamella are thinned from the top. Thinning by the double-tilt method is performed at perpendicular angles for front and back side. Consequentially, a second IBID layer on the lamella sidewall might be necessary, slightly increasing the processing time. This is, however, mainly of concern for specimens with structures layered parallel to the surface. Fortunately, by choosing the beta-tilt angle a bit smaller or larger than 90°—thus not milling exactly parallel to the surface—curtaining of the ROI can be largely suppressed.

Conclusions

FIB is currently the only method for efficiently preparing a TEM specimen from an arbitrary location in bulk materials. However, conventional FIB preparation methods are not suitable for producing high quality specimens necessary to achieve ultimate resolution in a modern aberration-corrected TEM, much less for low kV (S)TEM. We have demonstrated that introducing a second tilt axis results in a significant improvement over conventional FIB sample preparation techniques. The new method compares in speed and universality but excels in terms of yield. It allows us to routinely obtain large area coplanar lamellae thinner than 10 nm. For medium voltages of 200 to 300 kV where sub-100 nm thicknesses are sufficient for HRTEM imaging or atomic resolved STEM imaging, coplanar lamellae with areas of some tens of μm^2 can be produced. The resulting specimens are superior for high-resolution (S)TEM, even at very low voltages. We believe that this technique will enable new developments in TEM and reinvigorate STEM techniques inside the SEM. With ever decreasing component size in integrated circuits, the proposed method can help to meet increased demand for high-resolution TEM inspections and enable low kV STEM to bridge the gap between SEM and STEM.

Acknowledgments

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