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A simple approach to atom probe sample preparation by using shadow masks

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ABSTRACT

Here, we present a new method that utilises shadow masks in a broad ion beam system to prepare atom probe samples. It is particularly suited to non-conductors and materials with surface layers such as surface oxides, implanted layers or thin films. This new approach bypasses the focused ion beam (FIB) lift-out step, increasing the sample throughput, dramatically reducing the required FIB beam time and decreasing the complexity of sample preparation.

1. Introduction

Atom probe tomography is a high resolution 3D mass spectrometry/microscopy method, where single ion or small ionic complexes are removed from solid sample surfaces by a high field, sometimes combined with a laser pulse and are detected on a 2D detector. These 2D coordinates are then reconstructed into 3D [1]. In order to produce the high fields that are required, the samples need to be in the form of sharp needles (radius < 100 nm). This was originally achieved by electropolishing rods cut from metallic samples, allowing high-throughput sample preparation with inexpensive equipment [2]. In recent years, the capabilities of atom probe have been extended by the utilisation of laser pulsing [3] and focused ion beam (FIB) based sample preparation [4,5] to allow new experiments on thin films, non-conductive samples and site-specific features that need to be prepared with high precision.

However, the FIB based methods rely on expensive machinery and expert staff, and in many laboratories, access to instrument time and training is a major bottleneck for the throughput of atom probe experiments. This is mainly because the majority of FIB based projects used in-situ lift-out processes to fabricate the samples. While efforts have been made to reduce the complexity of the process by fabricating samples by cutting sections into thin wedges of material [6,7], or by cutting trenches, these methods have not gained significant popularity because they often involve even longer FIB processing times than lift-out methods.

In an early attempt to prepare thin film samples for atom probe analysis by using a broad ion beam (BIB), Liddle et al. [8] distributed small, micron-sized spheres of diamond on a film substrate to be used as a shadow mask for ion milling. Larson et al. [9,10] and later O'Neil et al. [11] developed this technique further, producing field emitters raised several tens of μm above the bulk material. While the earlier attempts by Liddle et al. and Larson et al. yielded no or only small amounts of data since the tips were not protruding far enough above the surface to be analysed by the atom probes used (no micro-electrode setup), O'Neil successfully acquired data from Aluminium specimens using a micro-electrode setup. This highlights how broad ion beams can be used to micromachine multiple atom probe tips in parallel, in order to achieve high sample preparation throughput.

In order for a sufficient electric field to be produced to trigger field evaporation at technically feasible voltages (~1–10 kV), the sample needs to protrude from its support by much larger distances. A schematic of a typical configuration in a micro-electrode setup [12,13] is shown in Fig. 1. With counter electrode aperture diameters commonly around 30–100 μm (the commercial LEAP system uses a~45 μm aperture), the required clearance of the sample above a support structure is typically > 50 μm. This also allows for the laser beam to access the sample tip (where laser assisted field evaporation is used). When using FIB-based preparation, such a shape is usually achieved by separately preparing a support structure [5,14], to which a small piece of material, a few microns in size, is attached.

Here, we present a method where we use a BIB system to

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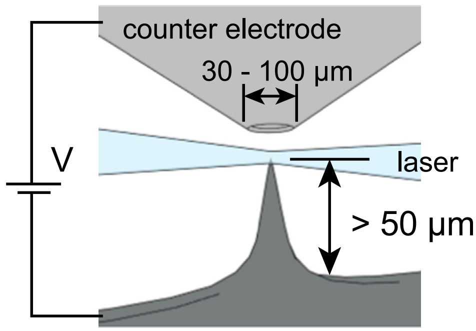

Fig. 1. Schematic of a micro-electrode atom probe and the geometrical constraints it poses on the sample.

produce similar structures from thin sections of material. By combining BIB together with a shadow mask made from low sputter yield material such as W or Mo, we are able to produce the rough shape of an atom probe sample that can then be further processed by established FIB based approaches [1,4] to form a number of sharp tips. This has the advantage that samples with surface films, or any materials that cannot be electropolished, can be processed into a shape only requiring a minor amount (typically less than 30 min per tip on the oxide samples in this paper) of FIB processing time, and much less operator training.

2. Materials and methods

A schematic of the sample preparation process is shown in Fig. 2. The first step is to produce a sliver of material that is thin enough for a broad ion beam to mill through within a reasonable amount of time. In the system used in this work, typical removal rates are 8 μm/h for Si and 1 μm/h for W, meaning that the starting samples are required to be less than ~50 μm thick. For bulk materials or thin films on a substrate, this can be achieved by tripod polishing, a commonly used technique in the preparation of lamellae for transmission electron microscopy [15]. This results in a thin wedge of the material. In the case of brittle materials such as

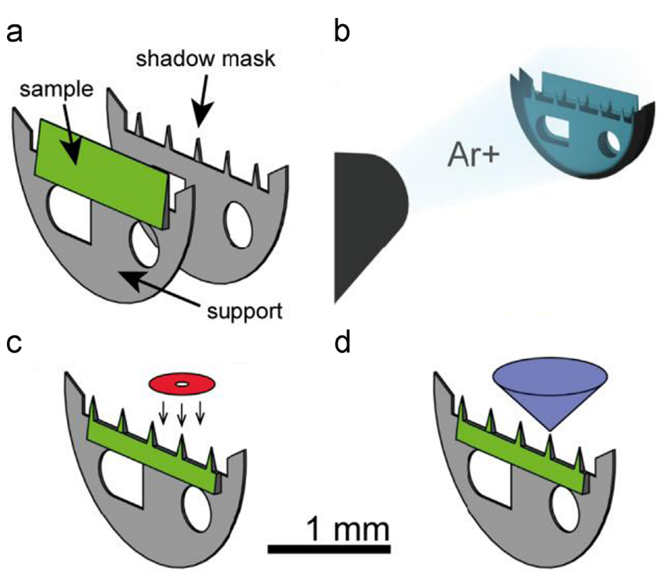

Fig. 2. Schematic of the process of fabricating an atom probe sample through broad ion beam shadow masking. (a) A sliver of material is attached to a carrier grid with no tips and a shadow mask is applied. (b) The sample is milled in a broad ion beam. (c) The tips formed after BIB milling are sharpened with a focused ion beam. (d) The atom probe experiment is carried out. The entire process is shown in the supplementary movie.

oxides or Si, or samples in which deformation in the area of interest is likely to pose an issue, the tripod-polished wedge can be further be thinned by ion polishing. For thin films, depositing directly into ultra-thin wafers (commercially available with < 10 μm thickness) would allow for this technique to be used without further thinning before the shadow mask milling. In the case of metallic samples, we have also produced thin wedges directly from 3 mm discs.

The thin sliver of material is attached to a custom-designed support grid with no tips. In this step, the glue is applied to the support grid only, in order to avoid having to remove glue residues from the tip areas later. The glue needs to be vacuum compatible and free of air bubbles. We used both cyanoacrylate (superglue) and silver filled epoxy. The sample on the support grid is then combined with a shadow mask of similar shape, made from a slow-milling material such as W or Mo (Fig. 2a). This mask is produced by electropolishing a custom grid to create the sharp tips, published in [14]. In the case of thin films, the film (green in Fig. 2 and the Supplementary movie) is positioned so that it is facing away from the ion beam's incident direction, so it will be the last part of the sample to be milled away. A shadow mask is clamped to the sample. We used a specimen holder that allows easy transfer between BIB, FIB, TEM and atom probe [14]. The whole assembly is then inserted into the ion beam processing chamber (Fig. 2b). The entire process is shown in detail in the Supplementary movie. The shadow mask can also be glued into place but this can result in glue residues that need to be removed by the ion beam. Gluing also fixes the shadow mask to the sample, preventing further adjustments of the position. The grid we have used here has been designed with tweezer holes that make handling easier and which can also be used to apply a second shadow mask after the first one is consumed. This is especially helpful for the preparation of low sputter-yield materials such as oxides.

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For the specimens used as examples in this paper, BIB milling was carried out on a South Bay IBSe ion coating/milling unit, using an Ar ion beam at 5 kV, with a beam current of 3 mA in a spot of several cm^2, with the high intensity part being around ~1 cm^2. The thinning of the oxide was carried out in a Gatan precision ion polishing system (PIPS®), using a 5 kV Ar ion beam of 10 mA. The final FIB milling was carried out on an FEI Quanta 200 3D FIB-SEM system (oxide sample) and on a Zeiss Auriga® FIB-SEM (oxide layer).

3. Results

3.1. BIB preparation of a bulk oxide

In recent years, the analysis of oxides and other ceramic materials [16–21], including geological materials [22] has gained significant popularity. In the following section, we demonstrate the application of this new method to prepare samples from these brittle, low-sputter yield materials.

Fig. 3 shows the whole preparation procedure for a polycrystalline sample of Zirconium-, Yttrium-, Tantalum oxides, as an example of a material that would normally be prepared by FIB liftout. The sample has a nominal composition of 28 mol% Yttria ($YO_{1.5}$), 28 mol% Tantala ($TaO_{2.5}$), balance Zirconia (ZrO_2). It contains two phases; a tetragonal ZrO_2-rich phase and a monoclinic $YTaO_4$ phase in which 21 mol% ZrO_2 is soluble. The process starts with the production of a thin wedge by tripod polishing, which is then attached to a support grid. Due to the brittle nature of the material, the remaining thickness of the material before it starts