Submicron X-ray diffraction

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Abstract

At the Advanced Light Source in Berkeley we have instrumented a beam line that is devoted exclusively to X-ray micro-diffraction problems. By micro-diffraction we mean those classes of problems in Physics and Materials Science that require X-ray beam sizes in the sub-micron range. The instrument is for instance, capable of probing a sub-micron size volume inside micron-sized aluminum metal grains buried under a silicon dioxide insulating layer. The resulting Laue pattern is collected on a large area CCD detector and automatically indexed to yield the grain orientation and deviatoric (distortional) strain tensor of this sub-micron volume. A four-crystal monochromator is then inserted into the beam, which allows monochromatic light to illuminate the same part of the sample. Measurement of the diffracted photon energy allows for the determination of d spacings. The combination of white and monochromatic beam measurements allow for the determination of the total strain/stress tensor (6 components) inside each sub-micron-sized illuminated volume of the sample. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

X-ray diffraction is a technique that has been used for about a century for elucidating the structure of materials on the macroscopic scale (0.1–10 mm). With the increasing need from industry to develop materials of high mechanical performance, a good understanding of their properties at the mesoscopic scale (0.1–10 μm) has become critical since many of these properties are dependant on the behavior of structural entities at this scale (grain boundaries, inclusions, intrinsic intra- and inter-granular stress distribution). There is a limited amount of experimental data at the mesoscale due to the lack of suitable techniques. This has long prevented modeling material behavior at this length scale, which in turn has prevented progress in developing a systematic link of material properties from the macroscopic to the microscopic. With the recent
availability of bright third generation synchrotron sources and progress in X-ray focusing optics, it is now practical to develop X-ray diffraction and apply it on the micron scale length to measure the mesoscopic properties of materials.

The ability to accurately characterize the orientation and strain/stress state in individual grains of a sample is a basic requirement to understand how materials behave at the mesoscopic scale. Using X-rays as a microprobe also allows the characterization of buried grains under overlying cap layers and multilayered films without the need of any sample preparation. This lack of sample preparation is especially important since the sample stress state can be greatly affected by any preparation process. This can be a major drawback of the currently used electron microprobe techniques. The accurate measurement of strain gradients at the micron and sub-micron level will be useful for solving many scientific and technological problems. These range from the strain state under nano-indenters to gradients at crack tips of turbine blades and artificial heart valves. Undoubtedly many other applications will unfold in the future as X-ray micro-diffraction is developed at other storage rings around the world.

In this work we describe a recently built beamline that can measure the grain orientation and triaxial strain in an arbitrarily orientated micro-crystal in a buried thin film. As a demonstration of this technique, we have addressed a practical and current problem in the semiconductor industry. The sub-micrometer wires which interconnect the active transistor elements in an integrated circuit can be deleteriously affected by a phenomenon called electromigration. As device structures and their interconnecting wires are made smaller and smaller, electrical current densities in the interconnects (which are increasing from $10^6$ Amp/cm$^2$ toward $10^8$ Amp/cm$^2$) actually transport the atoms of the wires themselves, inducing voids and hillocks to form and consequent failure of the device. Experiments have shown that the microstructure of these wires significantly affect this process [1,2]. In spite of much effort in this field [3–5], the relation between microstructure and electromigration-induced failure is still not understood to a great depth. Our intent is to provide new information on the electromigration phenomenon by investigating the mesoscopic physics by way of this new X-ray microdiffraction instrument.

2. Instrumental considerations

The customary way of performing X-ray diffraction is to fix the photon energy and map the Bragg reflection peaks by rotating the sample while detecting the diffracted X-rays with a detector. Such a scheme is inappropriate for micron-sized samples since current high-quality diffractometers have a sphere of confusion of $\approx 10\mu m$ and any rotation would move the sample out of the micro-beam. In our arrangement the sample remains fixed except for translation motion in the plane of the sample. The detector can be rotated into an appropriate position around the sample. The sample is illuminated with focused white radiation and the diffracted X-rays are recorded on a CCD detector as a Laue diffraction pattern. This can be indexed from a known structure to give crystal orientation. Then by scanning the photon energy and knowing the direction of the Laue beams and crystal orientation, d-spacing measurements can be made. For crystal orientation and strain measurements the X-ray optical system must fulfill the following requirements.

1. Focus both white and monochromatic light down to a sub-micron-sized focus.
2. Switch between white and monochromatic X-rays without changing the point on the sample being irradiated.
3. The ability to scan in energy whilst illuminating the same point on the sample.

The schematic layout of the beamline shown in Fig. 1. is able to perform these tasks and will now be described.

3. Beamline description and performance

As the X-ray source the beamline uses a bending magnet at the Advanced Light Source (ALS)
(1.9 GeV, 1.27 T, 400 mA). A platinum-coated silicon toroidal mirror [6] operating at a grazing angle of 5.4 mrad can collect up to $3 \times 0.2$ mrad of light and focus it in the 1:1 condition onto some $X/Y$ slits located at the front of the hutch. Beyond these slits is a 2.5 m long section within the hutch that is occupied by a time-resolved development station, followed by the micro-diffraction end station operating in air. This end station consists of a four-crystal monochromator followed by two platinum-coated orthogonal Kirkpatrick Baez (KB) micro-focusing mirrors [7], the sample and a 90 x 90 mm active area X-ray CCD. The sample and CCD are mounted on a 6-circle Huber goniometer. The schematic of the engineering model used for the end station instrument is shown in Fig. 2.

The slits at the front of the hutch act as an adjustable sized source for the KB mirrors of the micro-diffraction station. In this way spot size can be traded for flux. The 4-crystal monochromator has the property of either passing white light or monochromatic light down the same axis. This allows for easy switch over between white and monochromatic light while illuminating the same point on the sample. Some details of the individual items in the beamline will now be described in detail.

The slits consist of two orthogonal pairs of 10 mm diameter water-cooled polished tungsten rods that are separated by 2 mm. By rotating the rods on a rotary vacuum mechanism any slit size can be chosen between 0 and 2 mm with a resolution of $\pm 1$ μm. The toroidal mirror images the bending magnet source (source size = 250 x 25 μm FWHM ($H \times V$)) onto the slits with a focused spot size of 250 x 45 μm FWHM. This implies tangential figure errors of $\sim 0.5$ μrad over the used part of the mirror (15 x 600 mm). Brightness has been reduced by $\sim 2$ with the use of this toroidal mirror but this was viewed as a reasonable trade off considering the flexibility of the beamline allowing two experimental end stations.

Three and half meters downstream of the slits is the four crystal monochromator in the $++$ configuration [8]. This arrangement has the property of directing the monochromatic X-rays along the same axis as the incoming white light. The monochromator crystals to be used are two

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1SMART 6000 CCD system, Bruker AXS, Madison, WI 5373, USA
identical channel cut crystals mounted such that the rotation axis passes through the surface of diffracting surfaces 2 and 3. In this manner a Bragg angular range of 7–70° is allowed which for Ge(1 1 1) monochromator crystals corresponds to an energy range of 15574 – 2019 eV. This is a reasonable match to the cut off of the toroidal mirror (~50% reflectivity at 14 keV). The off axis mounting of the crystals allows for them to be rotated out of the way and thus allow white radiation to continue to the KB focusing mirrors. The 4-crystal monochromator is of a design similar to that of Tolentino et al. [9] consisting of two rotational stages onto which two channel cut crystals mount. For Bragg angle changes the two stages rotate in opposite directions by means of a tape drive, which is driven by a linear slide. We find the instrument is able to scan in energy and remain on the rocking curve over the available photon energy range of 6-15 keV without the additional requirement of feedback.

The KB mirror dimensions are defined by the source size, operating wavelength, spot size at the sample and working distance required around the sample. We have standardized on one design for both the horizontal and vertical focusing KB mirrors. A design spot size at the sample of 0.5 μm would be appropriate for the electromigration work. The operating wavelength should extend to ~14 keV to ensure an adequate number of Laue reflections. Taking the various factors into consideration we opted for KB mirrors of length =101.6 mm, a grazing angle =4.5 mrad and a sample to mirror end clearance distance of 60 mm. Other parameters are listed in Table 1. For the photon energies used the vertical acceptance is limited by the monochromator and is approximately the convolution of the reflectivity curves of crystal 2 and 3 in the dispersive arrangement on the Dumond diagram [10]. At 10 keV the vertical angular acceptance is ~55 μrad FWHM. In general we stop the vertical aperture down to ~65 μrad (about half the KB mirror aperture) as the monochromator prevents the full aperture of the vertical focusing KB mirror from being used. A collimating mirror before the monochromator will eliminate this vertical aperture restriction by the monochromator and is planned.

The KB mirrors are required to be plane ellipses in order to faithfully image the adjustable slit source to the sub-micron focus at the sample. In general only spheres and flats can be manufactured by the optics industry to the sub-micro-radian tolerance required. To make plane ellipses to the relevant tolerance we bend flat mirrors in a mechanical mirror bender. The schematic layout of the device used is shown in Fig. 3. It consists of a flat mirror of dimensions 101.6 × 12.7 × 5 mm thick with invar blocks attached to the mirror ends with epoxy.2 It is important to attach the blocks to the ends of the mirror rather than the mirror base as experience indicates [11] that the later sets up longitudinal strain within the mirror resulting in significant mirror surface deformation. The blocks are bolted to weak springs that are attached to slide ways that are driven by pico-motors. Moving the slide ways apart results in the leaf springs adopting a ‘S’ shape which provide the required bending moment for the mirror ends. The mirror is constrained from translating to the side by means of a thin strip of metal labeled ‘tie bar’ in Fig. 4. Asymmetric couples, when applied to the plane mirror produce a mirror shape that is a good cubic approximation to the required elliptical mirror shape. To achieve an exact fit to the required ellipse the mirror is fabricated with its width varying along its length. Details of this approach and the fundamental beam bending calculations can be found in Ref. [12]. The mirrors are assembled and then shaped using a long trace profiler [13]. Typical slope errors recorded are 0.6 μrad rms over the middle 80% of the mirror. X-ray spot sizes of 0.7 μm FWHM have been recorded by scanning the X-ray spot over a feature with a sharp edge and measuring the X-ray fluorescence. Flux levels are typically in the 107 photons/s/μm2 range with monochromatic light. The positional displacement between monochromatic and white light is <0.5 μm.

The local geometry around the sample is shown schematically in Fig. 4. The sample is inclined at

Hysol EA 9303.3NA epoxy, Dexter Aerospace Materials Div. Pittsburg, CA 94585, USA
The Laue diffraction pattern is collected by the X-ray CCD camera located above the sample. The sample can be heated up to 400°C by a heater attached to the back face of the semiconductor chip. The whole sample assembly is mounted on a piezo stage that has a range of ±50 μm in the plane of the sample. This in turn is mounted off the XYZ motorized mount of the goniometer head. This has a range of ±10 mm and allows for coarse adjustment.

The electromigration samples investigated were pure Al two level test structures 10–100 μm long, 0.7 μm wide and 0.75 μm thick. Shunt layers of Ti/TiN cover the top and bottom of the lines. The lines are buried under 0.7 μm of silicon dioxide for insulation. The test lines are connected to aluminum pads by tungsten vias of thickness 2250 Å. To determine the location of the sample we carry out an X-ray fluorescence map by rastering the sample in front of the focused white beam and detecting the Ti Kα fluorescence X-ray with a solid-state detector. Having located the sample interconnect line a Laue pattern can be recorded with a 10 s exposure with the CCD operating in 1K×1K mode. Fig. 5a shows a typical Laue pattern and is dominated by the bright spots of the silicon substrate. However as indicated by the arrows, weaker spots are also apparent in Fig. 8a. These are from the single crystal aluminum grain of the interconnect. The silicon spots can be digitally subtracted to yield the aluminum grain Laue pattern shown in Fig. 5b.

The relation of the CCD spot position to Bragg angle is calibrated by moving the CCD camera radially from the sample and recording the silicon Laue patterns at various distances from the sample. Lines drawn through the succession of the same Laue spots intersect at the sample point and this defines the sample position relative to the CCD X-ray detection surface. Using the silicon Laue pattern as a reference allows for detailed calibration of the roll, tilt and yaw of the CCD X-ray detection surface. Custom code using an algorithm similar to the one described by Chung et al. [14] is able to automatically index the Laue pattern using the CCD/sample geometry and the known crystal structure of aluminum. The code achieves this by determining the inter-ray angles for the Laue spots and comparing these to the

### Table 1: Parameters used for the KB mirrors

<table>
<thead>
<tr>
<th>Demag.</th>
<th>Source size for 0.5 μm focus</th>
<th>Mirror-sample distance (mm)</th>
<th>Maximum acceptance angle (urad)</th>
<th>Maximum convergence onto sample (mrad)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vertical focus KB</td>
<td>28.3</td>
<td>14</td>
<td>120</td>
<td>131</td>
</tr>
<tr>
<td>Horizontal focus KB</td>
<td>12.5</td>
<td>6</td>
<td>270</td>
<td>132</td>
</tr>
</tbody>
</table>

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various calculated values of indexed Laue spots until a match is found. When an indexed match is found, many other spots are naturally indexed (for aluminum about 9–15 spots). Remaining un-indexed spots belong to an adjacent grain as often several grains are illuminated which results in overlapping Laue patterns. The code is capable of dealing with this and has indexed up to 10+ overlapping Laue patterns. Automatic indexing of the Laue patterns allows the instrument to determine the orientation of the grains along the length of an interconnect line to within 0.01°. This mapping feature is important since electromigration experiments are often performed on single lines where prior knowledge of individual line structures is particularly relevant [3].

As an example of the detailed mapping capability we show in Fig. 6 the mapping of a 4 μm long aluminum grain. The (1 1 1) reflection spot is shown in detail in the sequence in the top part of the figure as the micro-beam is scanned down one side of the wire in 0.5 μm steps. The diffraction spot clearly moves in position and can be interpreted as a sequence of 3 sub-grains with slight misorientation. The angular misorientation between them is less than 0.5°. Not only is the instrument able to provide the overall grain orientation, it is able to look in detail within grains to show grain sub-structure.

The indexing code is also capable to accurately determine the centroid position of the Laue spot to 1/10th of a pixel (pixel size = 87.9 μm). The slight displacement of the spot from the calculated position is able to yield the deviatoric (distortional) part of the strain tensor [14–16]. This represents the distortion of the basic shape of the unit cell. Strain sensitivities of 2 × 10⁻⁴ are typical. In order to get the total strain tensor, the determination of the energy of one Laue spot is necessary in order to get the dilatational component of the tensor (for an isotropic compound, this component is directly related to the hydrostatic stress—the volume change of the unit cell). This is done in the following way.

From previous white beam measurements we have determined the Bragg angle of the Laue reflections from their pixel coordinates on the CCD. From the indexed Laue pattern, the
approximate energies of the reflections are calculated. To measure the d spacing we concentrate our attention on a single Laue spot say (242) and its relevant pixel region on the detector. We now insert the 4-crystal monochromator into the path of the white beam and scan energy over the appropriate energy range. In the monochromatic beam experiment the incident spot size is enlarged to about $\sim 10 \times 5 \, \mu\text{m}$, a procedure that we have found advantageous since it increases the total flux on the sample and allows for parallel recording of the diffraction spots of the sub-grains. Exposure times under these conditions amount to a few seconds. From the white beam Laue pattern we know the exact CCD pixel position where the Laue reflection for a particular sub-grain is located. An example of an energy scan for the (242) reflection of a 4 $\mu$m long aluminum grain at 225 $^\circ$C is shown in Fig. 7. The peaks correspond to different regions (sub-grains) within the long grain. From the variation in the energy and pixel position we can obtain the d spacing variation of the (242) planes along the grain. Furthermore from its variation with location in the grain we can obtain the change in strain or the local strain gradients within a grain. The relative change in strain along the grain length is shown in Fig. 7(b). Gradients of strain as large as $3.5 \times 10^{-4}/\mu\text{m}$ in single grains are evident from Fig. 7(b).

Our initial in-situ electromigration experiments were carried out on a passivated aluminum line 100 $\mu$m in length. The applied current density was $7.6 \times 10^5 \, \text{A/cm}^2$ at a temperature of 200 $^\circ$C in order to speed up the phenomenon of electromigration so that experimental observation times are reasonable. Measurements were made on each end of the wire for a distance of $\sim 6 \, \mu\text{m}$. Two grains were
monitored at the anode end (labeled A and B in Fig. 8). At the cathode end one long grain (labeled C in Fig. 8) was monitored. After 10h the total hydrostatic stress at the wire ends is shown in Fig. 8 for both ends of the wire. We observe a slight hydrostatic stress gradient along the wire length of 0.8MPa/m consistent with material removal from the cathode end and deposited at the anode end. We also observe large local stress gradients within the grains and across the grain boundaries of up to 80MPa/m. These local stress gradients are likely to play an important role in the electromigration phenomena, but to date have been ignored because of the lack of experimental strain data at the micron level.

4. Conclusion

In summary, we have developed a new X-ray-micro-diffraction technique capable of resolving sub-micron variations of texture and strain with an accuracy of about 0.01° in orientation and about 2 x 10^-4 in strain, respectively. For aluminum this corresponds to a stress sensitivity of 20 MPa. We have successfully applied the technique to in situ electromigration experiments in sub-micron-sized aluminum interconnect test structures. Our initial results show large local stress gradients within grains. We believe that detail on this scale in buried structures has not been reported before.

The X-ray micro-diffraction technique will provide key information on the behavior of materials at the micron and sub-micron level. This will provide us with a new and unique approach, which will broadly enhance our understanding of the complex and intricate links between the macroscopic and the microscopic properties of thin and bulk polycrystalline materials.

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