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Observation of the adhesion of thin Ta polycrystalline films to Si wafers via *in situ* topography/radiography

By Z. B. Zhao[1], Z. U. Rek[2] and J. C. Bilello[1]

[1] *Center for Nanomaterials Science, Department of Materials Science and Engineering, The University of Michigan, Ann Arbor, MI 48109, USA*
[2] *Stanford Synchrotron Radiation Laboratory, Stanford University, CA 94309, USA*

The adhesion of thin polycrystalline metallic Ta films to Si wafer substrates was studied *in situ* under real-time conditions using white-beam synchrotron Laue transmission diffraction topography with simultaneous radiographic imaging. The observations were carried out using a newly developed experimental apparatus, which consisted of a computer-controlled mini-tensile stage, an 800 °C ancillary furnace, and a CCD X-ray imaging system. The stress and imaging data were collected simultaneously via a video recorder and also at selected intervals by frame-grabbing/storage technology on a microcomputer. This allowed direct correlation between the stress, temperature and film-failure processes. The results for sputtered Ta polycrystalline thin films deposited onto Si wafers indicated that induced stresses led to buckling delaminations along well-defined directions. These observations can be readily extended to study a variety of film-adhesion and cycle-failure problems.

Keywords: X-ray topography; radiography; synchrotron radiation; transmission, Laue; polycrystalline thin films; adhesion

1. Introduction

Pioneer work by Lang (1957, 1972) showed that defects could be directly observed on large-scale single-crystal samples using X-ray diffraction techniques. The advent of synchrotron radiation has made X-ray topographic techniques relatively simple to perform using the Laue geometry, and has the advantage of having such large photon fluxes available that data recording can be accomplished in a small fraction of the time usually required in the laboratory. In the present case, a proprietary CCD (Microphotonics™†) has been employed to collect data under real-time conditions on Beamline 2-2 at Stanford Synchrotron Radiation Laboratory. X-ray diffraction topography is an imaging technique that can be used to map variations in the gradients of the lattice distortions associated with various crystalline defects, such as dislocations, grain boundaries, stacking faults and other domains (Bilello *et al.* 1989; Tanner 1976; Bowen 1998). Numerous works have been published in recent years using a well-collimated high-intensity synchrotron radiation white beam to produce Laue topographs of single-crystal samples. Prior work has concentrated

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† Microphotonics™ CCD X-ray camera with 25 mm diameter detector with point-to-point resolution of *ca.* 25 μm.

Phil. Trans. R. Soc. Lond. A (1999) **357**, 2681–2688

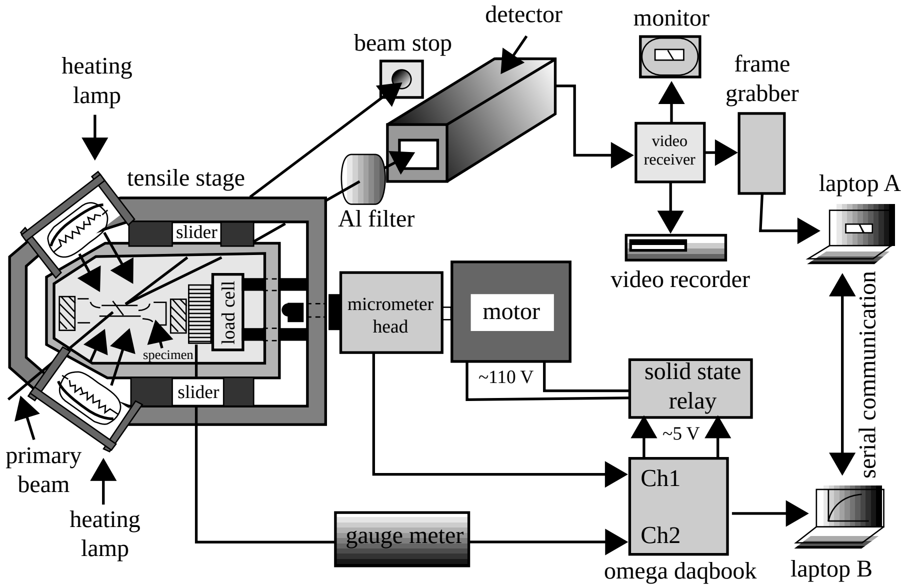

Figure 1. Schematic of *in situ* real-time X-ray imaging system coupled to a stress apparatus and the accessory high-temperature halogen quartz heaters.

on *in situ* quasi-real-time studies of many dynamic processes in materials, such as the motions of defects; crack initiation and propagation; crystal growth and phase transformations (Schmitz *et al.* 1986; Dudley 1987; Tanner 1994).

A new experimental apparatus has been developed to perform real-time *in situ* observations of the deformation processes of single crystals using synchrotron white beam as a source. The apparatus is also equipped with a specimen-heating device, which enables high-temperature experiments to be performed. It is well known that the Laue topograph of a single crystal provides many individual images for each $\boldsymbol{g}$ vector satisfying the diffraction conditions. In the current experiments, a radiographic image is simultaneously recorded from the thin polycrystalline film overlay, and these data are superimposed on the image of each diffraction spot from a family of $\{hkl\}$ planes. Thus, it is possible to correlate any ruptures, cracks or delamination bands in the overlay film to possible features in the Si substrate. Since the single crystal substrate acts as an energy band-pass filter, each topography spot has radiographic information limited to the energies diffracted to that position, which, of course, includes higher-order reflections from the same zone, i.e. $\{nh, nk, nl\}$, where n is an integer. This allows unambiguous identification of features in the film overlay versus those in the substrate. The present studies will mainly focus on a magnetron sputtered Ta film deposited on Si(100) single-crystal substrates. The internal stresses in the films were varied through heating and cooling in air, and observing the concomitant cracking/delamination assessed the effect on the adhesion of the film.

2. Experiments

The experimental apparatus, which was used on Beamline 2-2 at SSRL for the *in situ* real-time X-ray topographic/radiographic experiments, is illustrated schematically in figure 1. The detailed description of the apparatus has been given elsewhere

(Zhao *et al.* 1998). The facility can be divided into two subsystems based on their respective functions—the mini-tensile device, which is used to deform the materials and to record displacements and loads and the X-ray imaging system, which is used to observe topographic images of a selected Laue reflection (*hkl*) and the radiographic picture of the polycrystalline film overlay. Load–displacement records, thermal history and X-ray topographic/radiographic images can be recorded simultaneously via coordinated computer control and data acquisition using an array of microprocessors (see schematic figure 1). Rectangular specimens *ca.* 5 mm by 20 mm with a final thicknesses of 400 μm were cut from Si(100) wafers and coated with 500 nm polycrystalline Ta. These parallelepipeds were attached to the grips of the tensile stage by mechanical interlocks.

High-temperature X-ray topographic/radiographic experiments can be performed using a specimen-heating device, which is mounted to the tensile-stage assembly. The heating device consists of two quartz lamps. Each lamp is inserted into an aluminum holder that has been shaped to direct heat onto the sample. They were designed such that both the lamp-to-specimen distance and the heat-radiating direction are adjustable. A thermocouple glued with heat sink paste onto the specimen is used for temperature measurement. For the thin-film specimens (i.e. film–substrate composites), no prior mechanical load was applied. Rather, the internal stresses in the films, including those from growth deposition and that developed during the heating–cooling cycles, were used to test the limits of film adhesion via direct observations of the associated cracking and/or delamination.

For both *in situ* topographic/radiographic experiments (deformation of crystals and film delamination), a rectangular aperture was used to illuminate only the specimen area of interest. A 26 μm thick Nb filter was put in front of the specimen to limit the spectral range of white radiation. The X-ray imaging detector was located 10 cm behind the specimen. A beam stopper and an Al filter were used to prevent the detector from saturating and to reduce fluorescence, hence optimizing the image quality. High-resolution Kodak SR-5 X-ray film was also used occasionally to provide complementary information.

3. Results and discussion

The Ta films deposited on to Si(100) substrates were heated in the air with a heating rate of 15 °C min^{-1}. The entire process of real-time variations of the topographic/radiographic images as functions of temperature were captured by the CCD detector and recorded at a rate of 30 frames per second. Here, a few representative images for a Ta/Si(100) specimen are presented in figure 2. Figure 2*a* shows the topographic/radiographic image at 375 °C from $(13\bar{3})$ reflection. From room temperature to 380 °C the image shows little observable variation as the specimen is heated. The change indicated by the topographic/radiographic image starts at 380 °C. Figure 2*b* is the image captured at 438 °C, where straight-line-like features first appear. Typically, one first notices that localized features (such as the spots labelled ‘A’ in figure 2*a*) initiate, and then they propagate along the direction shown in figure 2*b* to form linear features recorded as light contrast bands in figure 2*b*. The propagation rate of the linear features, which is not constant, is quite slow at this early stage (i.e. *ca.* 2 mm min^{-1} at 395 °C). At higher temperatures these features propagate more rapidly (i.e. *ca.* 5–10 mm min^{-1} at 420 °C). Upon further increases in temperature,

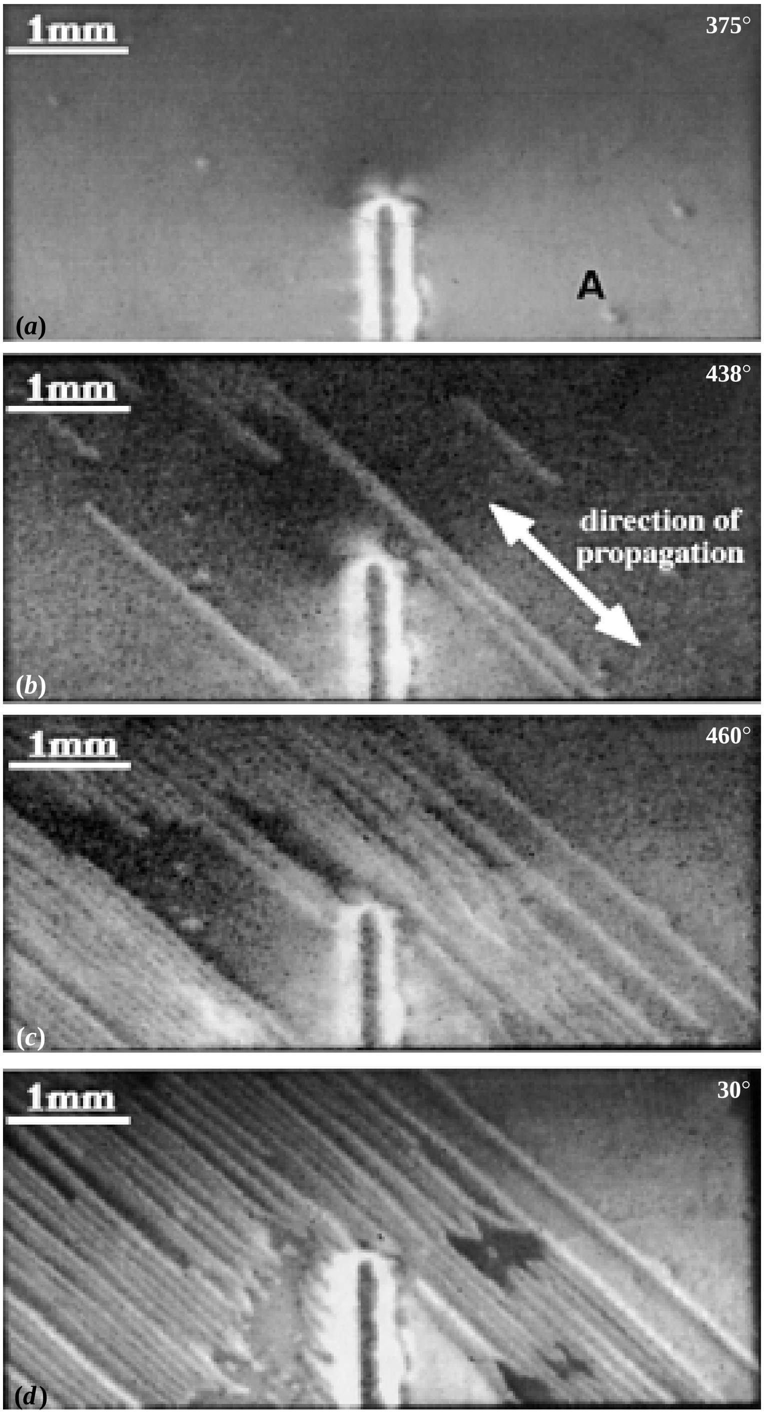

Figure 2. Selected images of a 500 nm Ta film on a Si(100) substrate at different temperatures.

more linear features appear and with increasing frequency and propagation rate. Starting from 450 °C, the formation of these linear markings becomes extensive, and almost the entire image is filled with these line-like features (figure 2*c*). The selected topographic/radiographic images shown in figure 2 can only present a rough picture of the entire failure event as captured by the real-time *in situ* observation apparatus. This is because the image selection shown in figure 2 represents only 4 out of 3600 frames recorded during the actual occurrence period in which linear contrast bands formed. To have a better picture of the entire process of image variations, the major features revealed by the recorded X-ray topographic/radiographic images can be summarized as follows.

(a) All the linear contrast features are nearly straight and parallel to each other.

(b) These linear features all exhibit quite similar appearances and form regular patterns on the images.

(c) The variation of topographic/radiographic images during specimen heating is not continuous. In other words, the initiation and propagation of the line features, occurring as individual events, proceeds intermittently, i.e. in stochastic bursts.

(d) The propagation rates of the linear features vary considerably depending on the particular individual line and the temperature. While there are exceptions, the general trend is that the linear features propagate more rapidly as the temperature increases.

(e) The linear features all appear to initiate from a local asperity and then increase their lengths through propagation along a unique direction. Even for those that occur abruptly, the frame-by-frame video analysis indicates that the formations of the linear features are still through propagation processes.

(f) As the Ta/Si(100) specimen is cooled from 500 °C to room temperature, the topographic/radiographic image shows little variation. Figure 2*d*, which was taken at room temperature, is nearly identical to the picture recorded at 500 °C.

These linear features, along with their appearances, suggest that the contrast of lines actually arises from the buckling of the Ta films while the substrate remains rigid. This suggestion is confirmed by viewing the topographic/radiographic images at higher magnification, which were taken with high-resolution X-ray film (see figure 3). In other words, the linear features reveal the regions where the Ta films have delaminated from the Si(100) substrate, and no attendant defect structure could be observed in the Si(100) that correlated in any way to the linear features. The ability to view both X-ray topographs of the substrate and the radiographs of the film made this correlation certain. At the temperatures used, indeed, one would not have expected defect generation in Si. Therefore, one needs to consider other possible causes for the decohesion of the film. The initiation and propagation of the linear features that were observed can be interpreted as follows: during the heating of the Ta/Si(100) specimen, the film first buckles away from the substrate in local regions where adhesion is poor, or non-existent; this process could be considered stochastic and is illustrated schematically in figure 4. Buckling then loads the

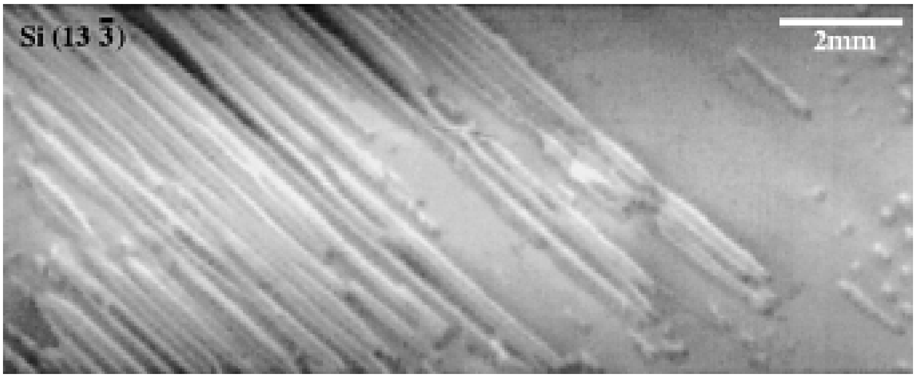

Figure 3. A topographic/radiographic image of the Ta/Si(100) specimen with higher magnification, which was taken with high-resolution X-ray film after the heating–cooling cycle was completed.

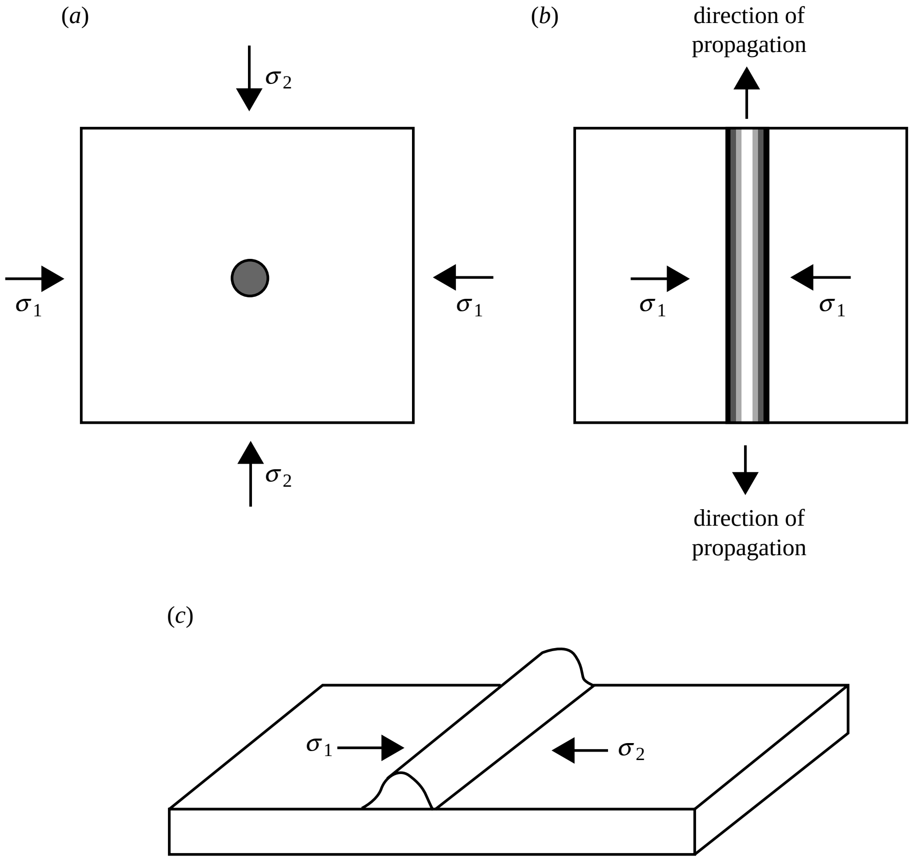

Figure 4. A schematic model showing the formation of the buckling-related delamination of a film from its substrate. (*a*) Initiation of a local blister at the regions where the adhesion is poor or non-existent. (*b*) Plane view of the delamination through buckling propagation. (*c*) A three-dimensional illustration of buckling-related delamination.

edge of interfacial cracks between the film and the substrate causing them to propagate (Hutchinson & Suo 1992). The driving force for this buckling delamination must be related to the development of internal stress in the film during heating of the Ta/Si(100) specimen. We have studied the variation of the stress in a radial direction under a heating–cooling cycle. The results indicated that this stress is compressive and increases dramatically at higher temperature (380–500 °C; see Zhao *et al*. (1999)). Other investigators have also observed such an increase in the compressive stress in Ta films upon annealing in the air (Cabral *et al*. 1994; Oda *et al*. 1997). For example, Cabral *et al*. (1994) observed that 50–200 nm thick sputtered Ta films undergo repeated compressive stress increase when thermally cycled to 400 °C. The general consensus for such a build-up in compressive stress is the affinity of Ta to oxygen atoms at annealing temperatures (Cabral *et al*. 1994; Oda *et al*. 1997). It has been suggested that oxygen atoms can readily diffuse into the grain boundaries in the polycrystalline Ta films. This results in the volume expansion of Ta films, thus leading to the increase in compressive stress. The present work does not presume to verify this impurity model; nonetheless, the buckling and delamination are closely associated with the build-up in compressive stress in the Ta films as they are heated in the air. During the heating of the specimen, the Ta film first forms blisters at the local regions with poor adhesion (see figure 4). When the compressive stress reaches certain critical levels, film delamination takes place to release the accumulated elastic energy. Associated deformation in the substrate is not required to produce film failure.

4. Summary

This work has presented the application of white-beam transmission Laue X-ray topography and concomitant radiography to the study of film/substrate decohesion. An experimental apparatus was used, which was developed for performing *in situ* real-time X-ray topographic/radiographic observations on the failure processes of a Ta polycrystalline metallic film on a Si(100) crystalline substrate. A unique advantage of the system is the simultaneous acquisition of stress data, X-ray topographs, and thin-film overlay radiographs so that their correlations may be established. Using this *in situ* real-time facility, the entire failure process of a Ta polycrystalline film on a Si(100) substrate has been observed. Accordingly, a model was created for the Ta film buckling/delamination from the interface with a Si(100) wafer, which showed that residual stresses in the metal overlay can lead to failure independent of any defect nucleation or propagation in the substrate.

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