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On the peculiar deformation mechanism of ion-induced texture rotation in thin films

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Abstract

Ion-beam irradiation is conventionally used to tune the electronic properties of semiconductors or as a “surrogate” for the study of radiation damage. Recently, it has also been employed for microstructure engineering, namely non-selective and selective grain growth. An even more interesting phenomenon is ion-induced crystal or texture rotation in thin metallic films. This couples an extensive crystal rotation with no significant modification of the grain/film morphology. The present work concentrates on the study of microstructural mechanisms of the phenomenon. A combination of in situ transmission electron microscopy irradiation experiments and finite element simulations reveals that the effect stems from a directional motion of gliding dislocations that is driven by the anisotropic stress field induced in the material surrounding the ion track. The hindrance of dislocation glide at the grain boundaries forces the crystal to relax through a crystal rotation.

Keywords: Ion irradiation; Crystal rotation; Thin films; In situ TEM

1. Introduction

The functionality of modern devices relies heavily on the synergistic interaction of their individual components, whose performance and reliability are intrinsically controlled by the microstructure of the constitutive materials. Metals and alloys, in the form of either thin films or one-dimensional nanostructures, represent one of the major research fields in materials science, where higher performance and improved reliability can be achieved by gaining control over the microstructure. For this purpose, post-production heat treatments and/or particle radiation treatments have been employed to tune microstructure qualities such as the crystallite morphology, size, orientation and defect content, which in turn affect the material’s physical properties [1].

Importantly, while heat treatments of thin films result mainly in isotropic grain growth and in a decrease of stored energy [1], ion-beam radiation offers more means to microstructure engineering by enabling athermal, isotropic grain growth [2,3] and grain boundary migration [4] as well as selective grain growth and texturing [5–7], to such an extent that even a single-crystal-like microstructure configuration can be achieved [8–10]. A fundamental understanding of how radiation-induced damage affects the properties of metal films is of vital importance to prevent degradation of the material characteristics or unwanted side-effects, such as creep, plastic deformation and, in the case of metal interconnects, premature electromigration.

Owing to their high atomic mobility and long-range order, metal films are rather tolerant to ion-induced damage, since during the thermalization phase of the collision

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cascade (i.e. the thermal spike) efficient annealing of the ion-induced defects occurs, promoting an almost complete recovery of the original atomic arrangement [11]. Nevertheless, phenomena of ion-induced plasticity in metals such as radiation-induced creep [12–14] and grain rotation [15–17] have been reported in the literature. While the first is the result of ion-induced point defects absorption through dislocation climb under the effect of an external stress field, the second raises more stimulating questions on the process dynamics since it involves crystal rotation without significant modification of the grain/film morphology. The controlled rotation of the grains in one direction is exclusively due to the ion-irradiation process and can therefore result in a complete change of the film crystallographic texture [15]. The spatial localization of this phenomenon by lithography may allow for a novel design route of nanostructured materials, in which the mechanical properties such as stiffness and strength as well as the electronic properties such as resistivity and reflectivity [18] can be spatially tuned by changing the film texture [9].

The current understanding of ion-induced crystal rotation in metals relies on the formation of shear stresses during the thermal spike phase, which cause dislocations to pile up and climb at grain boundary triple junctions, resulting in grain rotation and shear [19,20]. However, this theory is only applicable to the specific case of nanocrystalline metals with a large volume fraction of grain boundaries and the absence of an interface with the substrate.

The aim of the present paper is to analyze and interpret ion-induced crystal rotation at the micro- and nanoscale in a thin metallic film by in situ transmission electron microscopy (TEM) with a direct crystallographic correlation of the evolution of the film microstructure with respect to the irradiation geometry. As a result, we present a model which describes ion-induced crystal rotation in self-ion-bombarded, textured gold films deposited on a rigid substrate. The experimental evidence is supported by finite element modeling (FEM) of the ion-induced thermal stress evolution in the material surrounding the ion track. The proposed model can be readily applied to the reported studies describing ion-induced texture rotation [10,15–17]. We show that crystal rotation is the result of both the anisotropic stress field stemming from the ion–matter interaction and the film crystallography itself. Thanks to the textured nature of the gold films under analysis, the phenomenon of ion-induced crystal rotation will be described by its macroscopic manifestation and hence referred to as ion-induced texture rotation (ITR).

2. Experimental

Polycrystalline gold thin films were deposited by direct current (DC) magnetron sputtering (PVD Products, Inc.) at nominal room temperature or at 400 °C on 3 in. (1 0 0) silicon wafers coated with 50 nm of SiO_2 and 50 nm of Si_3N_4, used for ex situ irradiation experiments; and on 50 nm thick Si_3N_4 membranes, used for in situ irradiation experiments. A 10 nm thick titanium layer was also DC sputter-deposited onto the ex situ samples in order to improve the adhesion of the gold film. The chamber base pressure was lower than 10^{-6} mbar. The as-deposited films presented a strong (1 1 1) fiber texture normal to the sample surface and columnar grain morphology. The gold film thickness was either 500 nm or in a range between 50 and 100 nm for ex and in situ experiments, respectively.

The ex situ irradiation was performed using a Van de Graaf EN tandem accelerator at ETH Zurich. The films were irradiated with a 7 MeV gold ion beam (self-ion bombardment) set at an angle of 35° with respect to the surface normal. The ion fluence was in the range of 10^{15} to 10^{16} ions cm^{-2} with a target current on the order of 100 nA. The irradiation temperature was measured by means of a thermocouple attached to the back side of the copper sample holder and was kept below −130 °C by flowing liquid nitrogen through a custom-made cooler. The microstructure of the films along the film cross-section was analyzed by cutting lamellae of the irradiated samples by a dual beam station (Zeiss NVision 40) operated at 30 kV of the Ga ion beam and 5 kV of the electron beam.

The in situ irradiation was carried out in a 200 kV transmission electron microscope (TEM) (Tecnai G_2 20) at the JANNuS-Orsay facility [21], which allowed us to directly monitor the evolution of the film microstructure by TEM upon irradiation. The gold ion beam was produced by a 2 MV Tandem accelerator coupled to the TEM. Gold ion energy ranged between 1.5 and 3 MeV and the impinging direction was set at an angle of 67° from the sample normal. The ion fluence was in the range of 10^{13} to 10^{15} ions cm^{-2} with a target current on the order of 50 nA. Similar to the ex situ experiments, the low irradiation temperature was ensured by a liquid-nitrogen-cooled TEM sample holder. The ex situ TEM studies of as-deposited and irradiated thin films on Si substrates were performed on FEI Tecnai F30 (operated at 300 kV) at EMEZ ETH Zürich. The evaluation of the nanodiffraction patterns was performed using WebEMAPS (University of Illinois) [22] and the ASTAR® software.

3. Results

(1 1 1) out-of-plane oriented gold films with a thickness of 50 or 100 nm were sputter-deposited on electron-transparent, 50 nm thick silicon nitride membranes and irradiated in a TEM by a 1.5 or 3 MeV Au^+ beam, up to a fluence of $\sim 9 \times 10^{14}$ ions cm^{-2}. The in situ TEM irradiation was carried out at a temperature T_{irr} of 77 K and the ion beam was set at an angle Ω of ∼67° with respect to the normal to the sample surface. As a result, the normal to the film surface was always parallel to the electron beam, making possible the direct interpretation of the microstructure evolution upon irradiation. Videos were acquired either in diffraction or in bright-field imaging modes.

A sequence of snapshots extracted from the diffraction movies shows the evolution of the diffraction rings indicative