IN SITU HVEM STUDY OF ION IRRADIATION-INDUCED GRAIN GROWTH
IN Au THIN FILMS*

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IN SITU HVEM STUDY OF ION IRRADIATION-INDUCED GRAIN GROWTH IN Au THIN FILMS

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ABSTRACT

In situ observations of 1.5 MeV Xe^+ ion irradiated Au films at room temperature and at 150°C reveal the evolution of grain growth: the average grain size increases by the mechanisms of grain boundary migration and grain coalescence.

INTRODUCTION

Ion irradiation induced grain growth has been observed in several materials, including Ni, Au, Si and Ge [1,2,3,4]. Similar to investigations of thermally induced grain growth, the objective is to establish a relationship between grain size and irradiation time or between grain size and ion dose when the dose rate is constant. Results from Ag^+ implanted Ni thin films [1,2] show that the average grain diameter is proportional to the ion dose. For Au, Si, and Ge, it is found, however, that the increase of grain size with the dose exhibits a power law dependence [3]. Since previous studies of ion irradiation induced grain growth are all based on observation following irradiations [1,2,3, 4], a direct observation of the ion irradiation induced grain growth is useful for understanding the mechanism, kinetics and driving force of this process. The evolution of grain growth in ion irradiated Au thin films is observed in an electron microscope, and some of the results are discussed in the present paper.

EXPERIMENTAL PROCEDURE

Au films with thickness ~55 nm were prepared by sputter deposition on NaCl substrates. The grain size distribution is uniform and the average size is about 25 nm. Moreover, grains in the as-deposited films were preferentially oriented in $\langle 111 \rangle$. Self-supporting films were then irradiated with 1.5 MeV Xe^+ ions at a dose rate of $1.7 \times 10^{11}/\text{cm}^2 \cdot \text{sec}$ to minimize Xe implantation in the films and the effect of beam heating. The experiments were carried out at the HVEM-Tandem User Facility at Argonne National Laboratory [5], where a 2 MeV Tandem accelerator is interfaced to an AEI EM-7 1.2 MV high voltage transmission electron microscope (HVEM). The ion beam is introduced into the HVEM via a 33° ion-beam access tube; i.e., the ion beam is 33° from the electron beam in the microscope. During irradiations at room temperature and 150°C, microstructural changes of the films are examined in situ with the HVEM operated at 500 kV. The evolution of grain growth is followed by recording the images continuously on video tape and sequentially on photographic film.

OBSERVATIONS AND DISCUSSION

Direct observations

Direct observation of the Au films during ion irradiation reveal a dynamic picture of the induced grain growth; i.e., a gradual increase of average grain size is associated with dynamic changes in lattice defect structures. Although individual defects were not resolved due to their high density, the change of defect structure is suggested by the continuous

change of contrast in different areas of a single grain during irradiation. An example of the evolution of a single grain is shown in the dark field images of Fig. 1. The observed defects are presumed to be dislocation loops, since it has been previously reported that high concentration of vacancies in a collision cascade can collapse to form dislocation loops under irradiation at room temperature [6,7]. Such Frank loops can expand by climb or become glissile when converted to prismatic loops by unfauling. The observed contrast change due to dislocation rearrangement during ion irradiation, therefore, can be attributed to interactions of point defects and dislocations, dislocations with one another, dislocations and grain boundaries and dislocations and the free surfaces. The excess point defect concentration accelerates atom diffusion. Dislocation movement to grain boundaries can in turn cause structure changes in boundaries and thus affect grain boundary migration and grain orientation. The actual role of dislocations in the radiation-induced grain growth process is not clear, however, and certainly warrants further study.

Growth by grain boundary migration

Dark field images of the Au films irradiated with 1.5 MeV Xe⁺ at 150°C and room temperature are shown in Fig. 2 and Fig. 3, respectively. The images cover a dose range from 5.4×10^{13} to $5.1 \times 10^{14}/\text{cm}^2$ for the 150°C irradiation, and from 1.7×10^{14} to $5.1 \times 10^{14}/\text{cm}^2$ for the room temperature irradiation. It is noticed that the majority of the grains in the films are larger than the film thickness which is about 55 nm, so that the consecutive images demonstrate the evolution of 2-dimensional growth at both temperatures. Following grain "d" in Fig. 2 very closely, we noticed that after an irradiation of $5.4 \times 10^{13} \text{Xe}^+/\text{cm}^2$ the grain had an irregular shape with diameter approximately equal to 110 nm. As the dose had increased from 5.4×10^{13} to $1.1 \times 10^{14} \text{Xe}^+/\text{cm}^2$, the size of this grain increased about 30% by consuming its surrounding smaller grains. Then, the grain shape became hexagonal by continuous consumption of neighboring grains. At the final stage, after a dose of $5.1 \times 10^{14} \text{Xe}^+/\text{cm}^2$, the grain appears to be a hexagon with sides of almost equal length and angles about 120° at the vertices. Similar development is found in Fig. 3 for grain "D" irradiated at room temperature.

Fig. 1. Dark field images of Au films irradiated by 1.5 MeV Xe⁺ at 150°C with doses of 2.6×10^{14} and $5.1 \times 10^{14}/\text{cm}^2$.

Fig. 2. Dark field images of Au films irradiated by 1.5 MeV Xe⁺ at 150°C with doses from 5.4 x 10¹³ to 5.1 x 10¹⁴/cm².

Fig. 3. Dark field images of Au films irradiated by 1.5 MeV Xe⁺ at room temperature with doses from 1.7 x 10¹⁴ to 5.1 x 10¹⁴/cm².

The evolution of grains "d" and "D" represents a process of grain enlargement during the ion irradiation. In parallel, a process of grain shrinkage is also revealed. The grain labeled "e" in Fig. 2 had a size of 55 nm after irradiation with $5.4 \times 10^{13} \text{Xe}^+/\text{cm}^2$. Afterward, its dimension did not change significantly with the ion dose until its neighboring grains became relatively larger. According to Fig. 2, the transition took place after an irradiation of approximately $1.7 \times 10^{14} \text{Xe}^+/\text{cm}^2$, when grain "e" with concave boundaries was surrounded by three larger grains. When the dose was further increased to $5.4 \times 10^{14}/\text{cm}^2$, the size of grain "e" was reduced to only 18 nm. Similarly, the shrinkage of grain "E" in Fig. 3 is also observed.

The change of morphology in this irradiated Au film resembles the two-dimensional grain growth described by Hillert [8] in his defect model, where an array of hexagons is defined as perfect structure. If a defect, e.g., a 5-side grain, is introduced into a perfect array, grain boundary migration tends to occur because of the free energy difference caused by the curved boundaries. Therefore, the in situ observations of the evolution of grains, such as the shrinkage of small grains with concave boundaries and the development of stable hexagonal structures, in the irradiated Au films indicates that the main driving force of grain growth during irradiation is reduction of grain boundary energy. Further, the grain growth in these particular areas is accomplished by the gradual migration of grain boundaries towards grains with higher free energy without any formation of nuclei.

Grain coalescence

Besides the observed grain boundary migration, it is also noticed that the contrast between some grains varies during the irradiations. Such evidence can be found in Figs. 2 and 3 of the irradiated Au films. For instance, in Fig. 2 after an irradiation of $5.4 \times 10^{13} \text{Xe}^+/\text{cm}^2$ grain "a" was distinguished from grain "b" by the contrast, and it was separated from grain "c" by a boundary which was decorated with dislocations. As the dose increased to $1.1 \times 10^{14}/\text{cm}^2$, the sharp boundary between the original grains "a" and "b" disappeared, causing a dramatic change of the geometric shape of grain "a". At this point, the boundary between grain "a" and "e" was still present, which was determined not only by the contrast but also by the existing vertices on both ends as a result of balancing the surface tension of relevant boundaries. As the irradiation continues, the boundary between grains "a" and "c" gradually faded away by a dose of $5.1 \times 10^{14}/\text{cm}^2$. The absence of that grain boundary is confirmed by the straightening of the relevant boundaries between the coalesced grain and its neighboring grains. Moreover, the contrast between grains "a" and "c" became less, indicating the two grains merged into the same orientation. It is further displayed from these micrographs that the contrast between this coalesced region relative to its surrounding grains has changed during the irradiation, which could be caused by a relative orientation change between this area and the neighboring grains or between the film and the incident electron beam. The same grain coalescence process is observed at room temperature from the merging of grains "A, B, C" in Fig. 3.

In a previous study of thermal annealing behavior of cold rolled 3% silicon-iron single crystal, Hu [9] observed in an electron microscope that recrystallized nuclei could be formed by coalescence of subgrains in a high dislocation density area. He also noticed that the orientation of the coalesced grain differed from that of the original subgrains from which it was formed. A schematic diagram of this coalescence process is given in Fig. 4 by Li [10]. At the initial stage (Fig. 4(a)), the two grains are misoriented by a small angle. During thermal annealing the low-angle boundary "CH" is eliminated by dislocation climb along this boundary and by a relative rotation of the two grains, which is achieved by short-range

Fig. 4 Schematic representation of subgrain coalescence: (a) original structure; (b) rotation of one grain; (c) coalesced subgrain and (d) rearrangement of affected boundaries. (After Li, Ref. [10])

diffusion of atoms along the boundaries from the shaded to unshaded areas (Fig. 4(b)). Then, the affected boundaries of the coalesced grain are rearranged to remove the unstable angles (Fig. 4(c) and (d)). According to his description, the coalescence of subgrains during thermal annealing is very similar to the merging of some grains in the irradiated films.

Due to the relatively low surface energy, the grains with $\langle 111 \rangle$ normal to the surface of the irradiated Au films are thermodynamically more favored than grains with other orientations. Since the grain coalescence takes place at low-angle grain boundaries, it is suggested that the grain coalescence is significant only in a process of two-dimensional grain growth where a fair number of grains are oriented in $\langle 111 \rangle$ and share low-angle boundaries. Because grain growth by coalescence is not the only mechanism of grain enlargement in the irradiated films, satisfactory kinetic data for this process were not obtained.

SUMMARY

In situ observations of 1.5 MeV Xe^+ ion irradiated Au films at room temperature and at 150°C reveal the evolution of grain growth. In the process of two-dimensional growth, the average grain size increases by the mechanisms of grain boundary migration and grain coalescence.

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