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Letter to the Editors

Accumulation and recovery of defects in ion-irradiated nanocrystalline gold

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Abstract

Effects of 60 MeV ^{12}C ion irradiation on nanocrystalline gold (nano-Au) are studied. The experimental results show that the irradiation-produced defects in nano-Au are thermally unstable because of the existence of a large volume fraction of grain boundaries. This suggests a possibility of the use of nanocrystalline materials as irradiation-resistant materials. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Nanocrystalline materials, which consist of nanometer-sized crystal grains, are of great interest in practical use owing to their novel properties, e.g., increased strength/hardness, improved ductility/toughness, reduced elastic modulus, enhanced diffusivity, higher heat capacity, enhanced thermal expansion coefficient and superior soft magnetic properties as compared with conventional polycrystalline materials [1]. Moreover, since nanocrystalline materials have a large volume fraction of grain boundaries which might work as effective sinks for irradiation-produced defects, they are expected to be irradiation-resistant. In our knowledge, however, few irradiation experiments have so far been conducted for nanocrystalline materials. In this work, we have performed energetic ion irradiation to nanocrystalline gold (nano-Au), and have studied irradiation behaviors, i.e., accumulation and recovery of irradiation-produced defects. The reason why we have chosen Au as a specimen is to avoid a possible complexity caused by surface or interface oxidation.

2. Experimental procedure

A foil of nano-Au prepared by the gas deposition method [2] was used as a specimen. Previous studies [3] show that this kind of specimen has high mechanical strength and high thermal stability. Width and length of the specimen were 0.50 and 10 mm, respectively, and the effective thickness was 5.3 μm, which was estimated by gravimetry assuming that nano-Au had the same specific gravity as the ordinary polycrystalline gold (poly-Au). The average grain size of the specimen, which was determined by X-ray diffraction method [4], was 23 nm. Before irradiation, in order to characterize the specimen, the electrical resistivity of the specimen was measured as a function of temperature by a conventional four-probe technique. Then, the specimen was irradiated at ~15 K with 60 MeV ^{12}C ions. The ions passed through the specimen without remaining as impurities. At appropriate fluence intervals, the electrical resistivity of the specimen was measured in situ at 12 K. It is well known that the resistivity measurement has a high sensitivity for defect accumulation during irradiation [5]. After the irradiation at ~15 K, defect recovery was measured up to room temperature. After that, the same specimen was irradiated then at 300 K and the change in resistivity was measured at 300 K as a function of ion fluence in the similar way as the irradiation at ~15 K. For comparison, a poly-Au foil of 10 μm thick, which had been

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annealed at 873 K for ~1 h in vacuum below 2.7×10^{-5} Pa before irradiation, was placed adjacent to the nano-Au foil, and the same irradiation and resistivity measurement as mentioned above were performed simultaneously. Since there was a strong preferred crystal orientation (⟨111⟩ for the nano-Au and ⟨100⟩ for the annealed poly-Au normal to the specimen surface), the normal direction of the specimen was rotated by ~5° relative to the ion-beam axis in order to eliminate the possibility of ion channeling.

3. Results and discussion

Fig. 1 shows the electrical resistivity, ρ, of the nano- and poly-Au specimens as a function of temperature. The resistivity for nano-Au at 300 K is 3.0 μΩ cm. This value is larger than that for poly-Au of 2.3 μΩ cm. The difference in resistivity between nano- and poly-Au is mainly attributed to the increase in scattering of conduction electrons by the grain boundaries in nano-Au. The residual resistivity ratio (RRR), i.e., the ratio of resistivity at 300 K to that at 12 K, is 5.6 for nano-Au and 264 for poly-Au.

Fig. 2 shows the increase in electrical resistivity, $\Delta\rho$, of the nano- and poly-Au specimens as a function of ion fluence, Φ, during irradiation at ~15 K. The dependence of $\Delta\rho$ on Φ indicates the defect accumulation behavior. For nano-Au, the resistivity change rate, $d(\Delta\rho)/d\Phi$, i.e., defect accumulation rate, is larger than that for poly-Au. We will discuss this interesting result later.

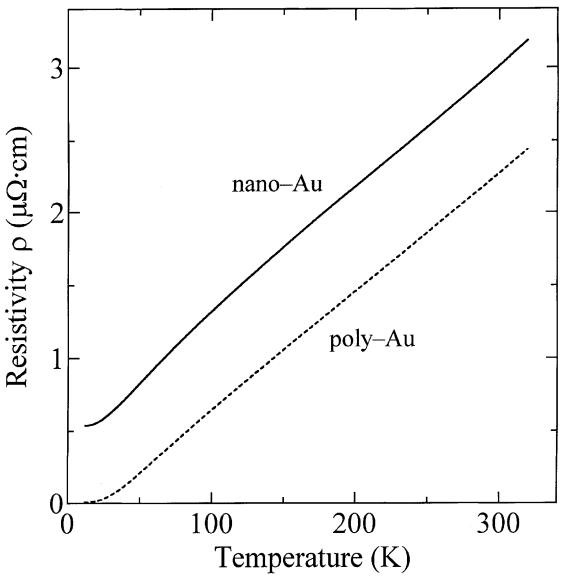

Fig. 1. Electrical resistivity, ρ, of the nano-Au (solid curve) and poly-Au (dotted curve) specimens as a function of temperature.

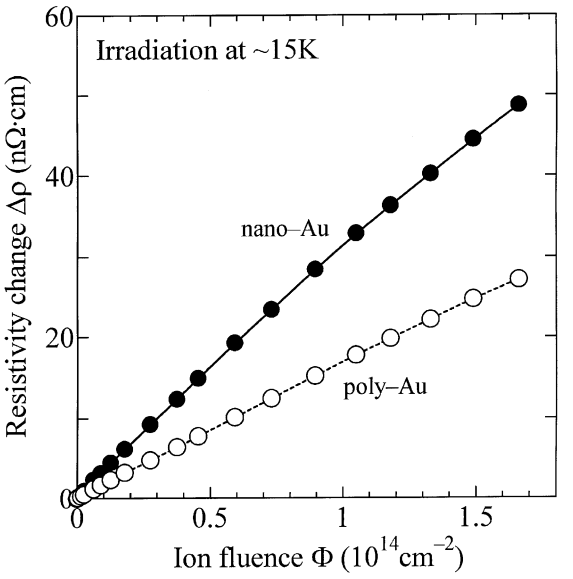

Fig. 2. Change in electrical resistivity, $\Delta\rho$, of the nano-Au (solid circles) and poly-Au (open circles) specimens as a function of ion fluence, Φ. Irradiation temperature is ~15 K and measuring temperature is 12 K.

For the subsequent irradiation at 300 K, $\Delta\rho$s of nano- and poly-Au specimens are plotted against Φ in Fig. 3. In contrast to irradiation at ~15 K, the defect accumulation rate for nano-Au is shown to be much smaller than that for poly-Au. This result is just what we expected; the irradiation-produced defects in nano-Au are thermally more unstable than those in poly-Au due to the existence of grain boundaries in high density that act as effective sinks for the defects. The result in Fig. 3 would show a promising irradiation-resistant property of nanocrystalline materials.

Fig. 4 shows the resistivity recovery curves for nano- and poly-Au following 60 MeV ^{12}C ion irradiation at ~15 K. The amount of recovery for nano-Au is rather larger than that for poly-Au. This tendency would be preserved up to room temperature, though the resistivity recovery for poly-Au could not be obtained above 150 K accidentally. This result is consistent with the irradiation behavior at 300 K shown in Fig. 3, i.e., with increasing temperature, the irradiation-produced defects in nano-Au become thermally more unstable than those in poly-Au.

Finally, we discuss the result obtained at ~15 K. The following three factors can be considered as candidates for the cause of the larger defect accumulation rate in nano-Au during irradiation at ~15 K: (a) low threshold energy for defect production near grain boundaries, (b) short focusing collision sequence due to small size of the grains, and (c) trapping of the

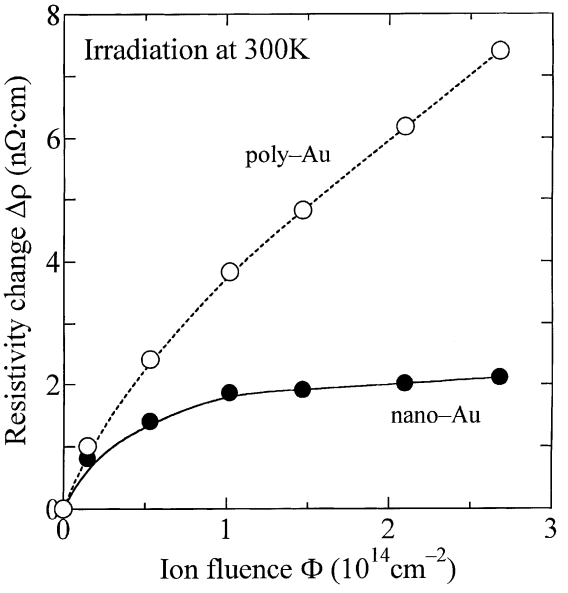

Fig. 3. Change in electrical resistivity, $\Delta\rho$, of the nano-Au (solid circles) and poly-Au (open circles) specimens as a function of ion fluence, Φ. Irradiation and measuring temperature is 300 K.

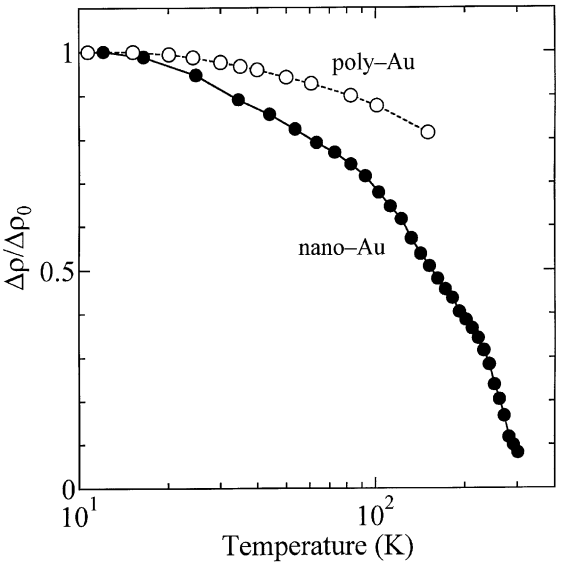

Fig. 4. Resistivity recovery curves for nano-Au (solid circles) and poly-Au (open circles) following 60 MeV ^{12}C ion irradiation at ~15 K.

stage-I defects, which cannot be seen in poly-Au even below 4.2 K [6], by grain boundaries. The true mechanism of the present interesting phenomenon, however, remains uncertain at present. Further study on grain-size dependence of irradiation effects is now in progress.

4. Summary

We have studied the defect accumulation and recovery in ion-irradiated nano- and poly-Au specimens. The defect accumulation rate in nano-Au is larger than that in poly-Au for irradiation at ~15 K (I), but for irradiation at 300 K, it becomes much smaller than that in poly-Au (II). Item (II) is expected qualitatively, and proves the promising property of nano-Au as irradiation-resistant materials. On the other hand, item (I) is not expected and possible mechanisms are proposed.

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