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Sputtering of nano-crystalline gold by high energy heavy ions

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Abstract

In order to investigate the grain size effects and the contribution of the electronic excitation on the radiation sensitivity, we have measured the sputtering yields of nano-crystalline Au and poly-crystalline Au samples by high energy heavy ions. No meaningful difference has been observed between the sputtering yields of both samples. It also appears that the sputtering yields of both Au samples agree with those of calculation based on the linear elastic collision cascade, indicating no appreciable contribution of the electronic excitation. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Nano-crystalline materials have increased interest in applications because of their unique physical properties [1]. In applications of these materials in radiation environment, their radiation sensitivity is of importance [2]. A measure of radiation sensitivity, e.g., the sputtering yield of nano-crystalline (nano-) Au by high energy heavy ion irradiation may differ from that of poly-crystalline (poly-) Au, because of the following reasons. Firstly, the binding state near grain boundaries and inside grains of nano-Au may differ from that of poly-Au. Secondly, fission fragment tracks have been observed in Pd and Pt with the thickness smaller than 20 nm [3], and in thin (5–45 nm) Al and Au films [4]. It has been argued that the electronic energy deposition contributed to the track formation [3,4]. Thus, there is a possibility that excited electrons are confined inside a grain which might enhance the sputtering due to the electronic energy deposition. In this study, we have measured the sputtering yields of nano- and poly-Au, using a carbon-foil collector method, in order to investigate whether or not any difference of the sputtering yields exists between nano-Au and poly-Au, and also effects of the electronic energy deposition.

2. Experimental

Samples of nano-Au (~15 μm thickness) were prepared by the gas deposition method [5]. The average grain size was about 23 nm. Thickness of poly-Au films and their grain size were ~10 μm and several microns, respectively.

Both samples of nano-Au and poly-Au were irradiated with 200 MeV $^{127}I^{+12}$, 100 MeV $^{127}I^{+9}$, 70 MeV $^{58}Ni^{+6}$ and 80 MeV $^{32}S^{+7}$ ions at normal incidence through a carbon (C-) foil (~100 nm thick), which was placed in front of the samples, in vacuum of $\sim 10^{-5}$ Pa. A sketch for ion irradiation on the C-foil and sample assembly is illustrated in Fig. 1. The ion dose rate was $\sim 10^{10}$/cm^2 s and the dose was up to 5×10^{14}/cm^2.

The energy loss in the C-foil for 200 MeV I, 100 MeV I, 70 MeV Ni and 80 MeV S was calculated as 1.6, 1.3, 0.8 and 0.4 MeV, respectively [6]. The ion energy after

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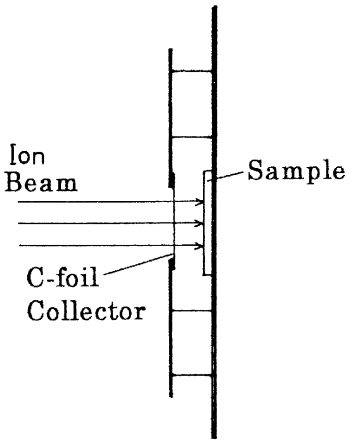

Fig. 1. Sketch of C-foil collector and sample assembly for ion irradiation. C-foil is self-supported on Al with double holes of 4 mm in diameter and samples are supported on Al.

transmission through the C-foil is given in Table 1. Bohr energy straggling in the C-foil was calculated as 0.07, 0.035 and 0.02 MeV for I, Ni and S ions, respectively. The energy loss and the energy straggling are very small, and hence do not disturb the present study. The ion charge incident on Au samples, which differs from that on the C-foils, may affect sputtering. Assuming that the ion charge reaches the equilibrium distribution after transmission of the C-foil, the ion mean charge Q after C-foil transmission was estimated from [7] and is given in Table 1. Sputtered Au atoms were collected in the C-foils and analyzed by Rutherford backscattering spectroscopy (RBS) using a Van de Graaff accelerator.

3. Results and discussion

Fig. 2 shows RBS spectra of the C-foil collector after irradiation with 198 MeV I on nano- and poly-Au samples to a dose of $5 \times 10^{14}/\text{cm}^2$. The C-foil was supported on a Be substrate for RBS analysis, which was performed using 1.8 MeV He with normal incidence and 160° scattering angle. The numbers of Au atoms in the C-foil collector show no meaningful difference between nano- and poly-Au samples. In Fig. 2(a), peaks around the channel numbers at 290, 500 and 650 are identified as O, P and Cu in the surface region of Be supports. The numbers of these atoms did not change appreciably before and after ion irradiation. That is, both RBS

Table 1
Sputtering yields Y (Au atoms per ion) of nano- and poly-Au samples by high energy heavy ion irradiation, together with the ion dose for Y evaluation

Ion	E (MeV)	Q	Y		Dose ($10^{14}/\text{cm}^2$)	S_n (keV/nm)	S_e (keV/nm)	Y_c
			Nano-Au	Poly-Au				
$^{127}I^{+12}$	198	30	2.4	2.2	0.5, 5	0.228	44	2.3
$^{127}I^{+9}$	99	25	7.3	7.1	0.4, 1.5	0.396	32	4.0
$^{58}Ni^{+6}$	69	18	1.5	1.7	1	0.082	23	0.8
$^{32}S^{+7}$	80	13	0.25	0.27	1	0.015	12	0.15

E and Q are the mean energy and mean charge after transmission through C-foil (~100 nm). S_n and S_e are the nuclear (elastic) and electronic (inelastic) energy losses, respectively. Y_c is the calculated sputtering yield due to the elastic collisions.

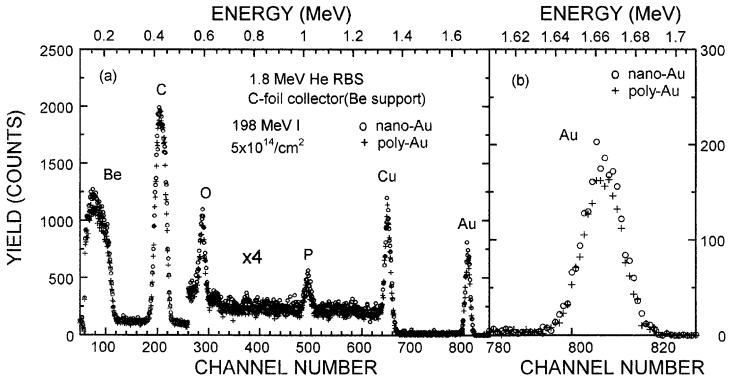

Fig. 2. (a) RBS spectra of C-foil collector after irradiation with 198 MeV I on nano-Au (○) and poly-Au (+) samples to a dose of $5 \times 10^{14}/\text{cm}^2$. (b) Expanded part for RBS spectra of Au peaks.

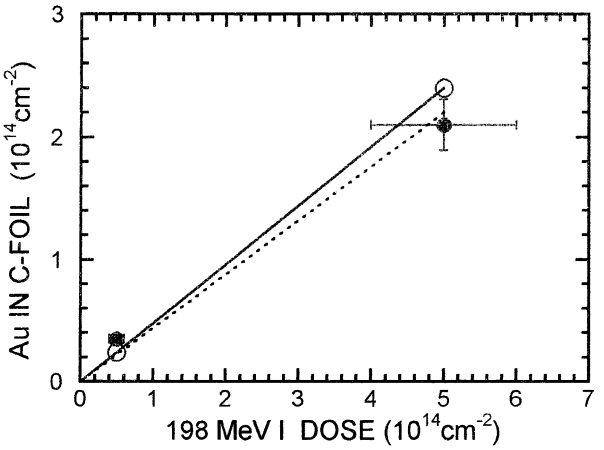

Fig. 3. Au atoms collected in C-foil vs 198 MeV I dose for nano-Au (○) and poly-Au (●) samples. Solid and dotted lines are the fits to data of nano- and poly-Au, respectively.

spectra for nano- and poly-Au are almost identical. The number of Au atoms collected in the C-foil is found to be proportional to the 198 MeV I dose as shown in Fig. 3, within an RBS analysis error of 10% and an estimated error of 20% in the ion dose. This implies insignificance of the sample contamination effects on sputtering.

Evaluation of the sputtering yields requires the C-foil collection efficiency f_C, defined as the number of Au atoms collected in the C-foil divided by the number of Au atoms sputtered from Au sample. It was calibrated using 120 keV Ne^+ ions obtained from a 200 kV Cockcroft–Walton type accelerator and the assembly shown in Fig. 1. The energy loss and straggling of 120 keV Ne in the C-foils were estimated to be 60 and 13 keV. Thus, the number of sputtered Au atoms is given by the sputtering yield of Au by 60 keV Ne times the Ne dose. The Au sputtering yield by 60 keV Ne was experimentally determined as 3.6, by measuring thickness decrease of Au films deposited on Al substrate. This value agrees with the experimental value of 3.6 by 50 keV Ne [8], and the simulation values of 3.9 (TRIM 1992 version) and 3.1 (TRIM 1997 version) with the surface binding energy of 3.81 eV. The value of f_C was obtained as 0.2, independent of the Ne dose in the range of $0.5–7 \times 10^{15}/cm^2$ within an estimated error of 20%.

Using the f_C value, the sputtering yields from nano- and poly-Au irradiated with high energy heavy ions were evaluated and the results are summarized in Table 1. No meaningful difference of the sputtering yields was seen between nano- and poly-Au samples for all ions investigated.

Fig. 4(a) shows the experimental sputtering yields Y of nano- and poly-Au samples vs the nuclear (elastic) energy loss S_n. It appears that Y increases nearly proportional to S_n. An estimated error of Y is ~30%, considering errors of 20%, 10% and 20% in f_C, RBS analysis and ion dose, respectively. Next, the experimental sputtering yields are compared with the calculated sputtering yield Y_c, which is obtained on the basis of the linear collision cascade scheme that Y_c is proportional to S_n. The proportional constant was evaluated as 10 nm/keV using the experimental sputtering yields of Au by Ne, Ar, Kr, Xe at 50 keV [8], which is in reasonable agreement with the value of 6.7 nm/keV derived using Au sputtering yields by 33 keV to 2.9 MeV Au ions [9]. Y_c values by high energy heavy ions are given in Table 1. Although Y is found to be somewhat larger than Y_c, agreement of Y with Y_c is reasonable because of the following reasons. The linear collision cascade scheme might not be accurate enough for high energy regions and the proportional constant, which was determined using the data of low energy ions because the sputtering yield data are scarce for high energy ions, may not be applicable to high energy ions. Neither is it completely ruled out that the experimental error is larger than the estimation. In conclusion, the linear elastic collision cascades are dominant in the sputtering of both nano- and poly-Au by high energy heavy ions.

Furthermore, if there is any effect of the electronic energy deposition or multiple charges of ions, the sputtering yields increase with the electronic energy loss S_e or the ion mean charge Q. Such a relationship was not found between Y and S_e, neither between Y and Q as shown in Fig. 4(b) and (c). These indicate no appreciable effects of the electronic energy deposition and of multiple charges carried by ions to the sputtering. No appreciable effect by the ion charge is supported by the direct observation that the Au sputtering yield by highly charged Ar and Xe ions are independent of their charge [10].

It has been mentioned that grains are isolated in the ultra thin metal films [4]. A possible explanation for the

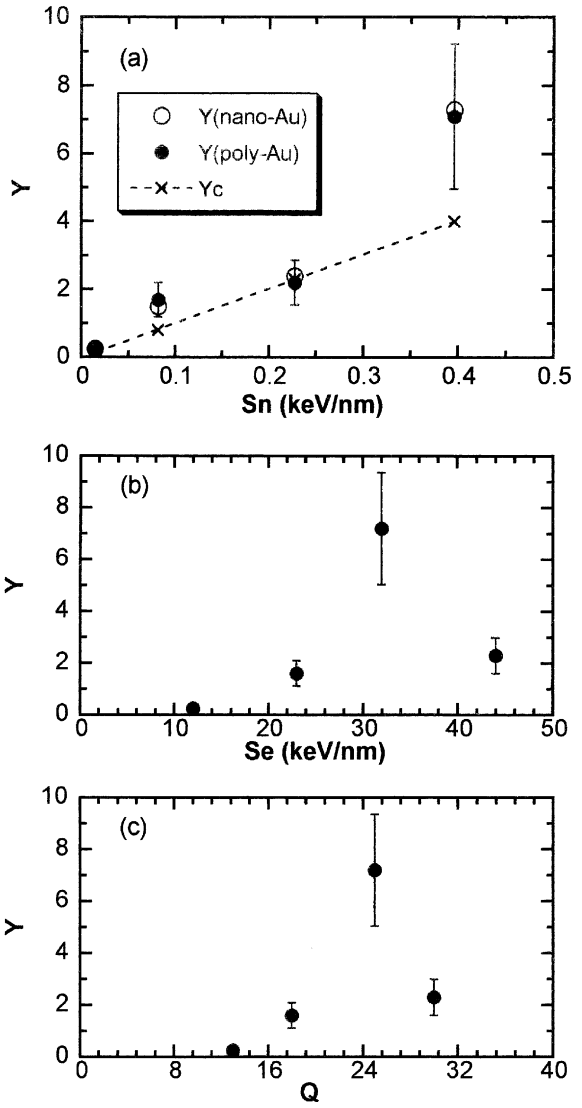

Fig. 4. (a) Sputtering yield Y of nano-Au (○) and poly-Au (●) samples vs nuclear energy loss S_n, together with the calculated sputtering yield Y_c (×). Dashed line is drawn through Y_c. (b) Y vs S_e (keV/nm) and (c) Y vs Q, where Y is the average of nano- and poly-Au sputtering yields, S_e the electronic energy loss and Q the mean ion charge.

fission track formation in ultra thin metal films due to the electronic energy deposition is that the electronic energy deposition is confined inside grains due to poor contact between grains and between grains and NaCl substrate in the ultra thin films, and hence, it is converted efficiently to the heat leading to evaporation of materials. This reveals that contact between grains may play important role in dissipation of the deposited energy.

Because TRIM simulation has been widely used, it would be interesting to see applicability of TRIM simulation to high energy heavy ions. It is found that the sputtering yields of TRIM (both 1992 and 1997 versions) simulation are smaller by a factor of ~6 than Y_c for the ions with the energy listed in Table 1. Thus, one should be careful for applying TRIM to estimate the sputtering yields by high energy heavy ions.

Finally, the ion irradiation effects on the C-foil collection efficiency was examined. The collection efficiency of the part of the C-foil supported on Al, which was not exposed to Ne ion irradiation, was obtained as a quarter to a half of $f_C = 0.2$ and this variation can be explained by the angular dependence of the sputtering yields. A similar situation was observed for high energy heavy ion irradiation. These results and the Ne dose independence of f_C described in the second paragraph of Section 3 reveal that the ion irradiation does not affect the C-foil collection efficiency of sputtered Au atoms.

4. Summary

We have measured the sputtering yields from nano- and poly-Au samples by high energy heavy ions. No meaningful difference of the sputtering yields has been observed between nano- and poly-Au samples. It also appears that the sputtering yields of both Au samples agree with those of calculation based on the linear elastic collision cascade, indicating no appreciable contribution of the electronic excitation, in contrast with the observation of the fission fragment tracks in ultra thin metallic films.

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