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Evaluation of local temperature around the impact points of fast ions

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ABSTRACT

Gold and platinum nanoparticles of few-nm size were deposited on amorphous silicon nitride (a-SiN) films. These samples were irradiated with 1.1 MeV C_{60}^{3+} ions to a fluence of $\sim 5 \times 10^{10}$ ions/cm^2 and observed using transmission electron microscopy (TEM). The ion tracks were clearly seen as bright spots and the metal nanoparticles disappeared from a neighboring region (5–10 nm) around each ion track. The platinum-nanoparticle-cleared region is slightly smaller than that of gold nanoparticles. This trend can be reproduced by the u-TS calculations assuming that the nanoparticles are desorbed when the local temperature surpasses the melting point of nanoparticles as was predicted by molecular dynamics simulations (Anders et al., 2009). This indicates that the temperature distribution in a nanometer region can be evaluated by observing the desorption of nanoparticles of different metals having different melting temperatures.

1. Introduction

When swift heavy ions penetrate a solid the ions excite solid electrons. The energy of the excited electrons is gradually transferred to the lattice and eventually cylindrical damage regions, so-called ion tracks, may be created when the electronic energy loss is larger than a material dependent threshold value [1,2]. There are several models proposed for the mechanism of the track formation [2–6]. Among these models, the inelastic thermal spike (i-TS) model seems most plausible because it explains the electronic energy loss threshold for the track formation and the evolution of the track radius with the electronic energy loss [2,6]. In the thermal spike model, ion tracks are assumed to be formed if the atomic temperature surpasses a material dependent threshold energy per atom. This threshold energy per atom has been considered as the energy to reach the melting temperature plus the latent heat for solid to liquid phase transition. The heating occurs in a highly localized area of nanometer size and continues only for a short time period of $\sim 10^{-11}$ s. Such a fast and localized heating cannot be measured using conventional techniques.

Recently, we have observed desorption of gold nanoparticles from the surfaces of amorphous SiO_2 (a-SiO_2) and amorphous silicon nitride (a-SiN) upon ion impact [7,8] using transmission electron microscopy (TEM). The TEM observation showed that the gold nanoparticles were disappeared from the neighboring area around the ion impact position. Considering the result of the molecular dynamics (MD) simulations, which showed that the gold nanoparticles desorb in several ps from the surface when the nanoparticles are heated beyond the melting temperature [9], the observed result suggests that the temperature surpassed the melting point of gold in the nanoparticle-cleared region. Actually, the i-TS calculation showed that the temperature of the substrate surpasses the melting point of gold in the nanoparticle-cleared region both for a-SiO_2 and a-SiN [8]. This result suggests that detailed temperature distribution can be evaluated by observing the desorption of nanoparticles having different melting points. In the present paper, we observed desorption of gold and platinum nanoparticles from a-SiN films upon bombardment of 1.1 MeV C_{60} ions. The observed results is compared with the theoretical calculation based on the i-TS model including the effect of nuclear energy loss.

2. Experimental

Self-supporting a-SiN films (thickness 30 nm) with a nominal density of 3 g/cm^3 were purchased from Silson Ltd. The composition of the a-SiN film was determined to be $Si_{0.49\pm0.02}N_{0.51\pm0.02}$ using

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high-resolution Rutherford backscattering spectrometry [10], which is Si rich compared to the stoichiometric Si_3N_4 by several at.%. A small amount of gold or platinum was vapor deposited on the a-SiN films at room temperature. For the gold deposition, different amounts of gold were deposited to prepare different-sized nanoparticles. The prepared gold- and platinum-deposited a-SiN films were irradiated with 1.1 MeV C_{60}^{3+} ions at normal incidence to a fluence of $\sim 5 \times 10^{10}$ ions/cm^2 with the 400-kV ion implanter of Takasaki Advanced Radiation Research Institute, JAEA. The samples were observed using TEM (JEOL JEM-2200FS) equipped with a field emission gun operating at 200 kV. It is noteworthy that the nuclear stopping power for 1.1 MeV C_{60} in a-SiN is estimated to be 8.1 keV/nm using the SRIM code, which is comparable to the electronic stopping power (10.3 keV/nm).

3. Results and discussion

3.1. Desorption of nanoparticles

Fig. 1(a) shows an example of the TEM bright field images of the platinum-deposited a-SiN film observed before irradiation. There are many platinum nanoparticles formed by the vapor deposition. The areal density, N, of these nanoparticles was measured to be 8.1×10^{12} particles/cm^2. The size distribution of these nanoparticles was derived from the observed TEM images and shown by solid circles in Fig. 2. The distribution shows a Gaussian-like well-defined peak with a peak radius of 0.8 nm and a width of 0.5 nm. Similar Gaussian-like distributions were also observed for the gold nanoparticles deposited on the a-SiN films as shown in Fig. 2. The squares and triangles show the size distributions of the samples deposited with a smaller and larger amount of gold, respectively. As was expected, larger nanoparticles were formed by the deposition of a larger amount of gold. The average radiis of the gold nanoparticles are 1.8 and 2.9 nm and the areal densities of nanoparticles are 1.1 and 0.7×10^{12} particles/cm^2 for the samples deposited with a smaller and larger amount of gold, respectively.

Fig. 1(b) shows an example of TEM bright field images of the platinum-deposited a-SiN film observed after irradiation with 1.1 MeV C_{60}^{3+} ions. Ion tracks are clearly seen as bright spots with a diameter of about 4 nm. It is seen that the platinum nanoparticles disappeared from the surrounding area of the ion tracks. Such a disappearance of the nanoparticles was also observed for the gold-deposited a-SiN films after irradiation of 1.1 MeV C_{60}^{3+} ions. The distance between the ion track and the closest nanoparticle

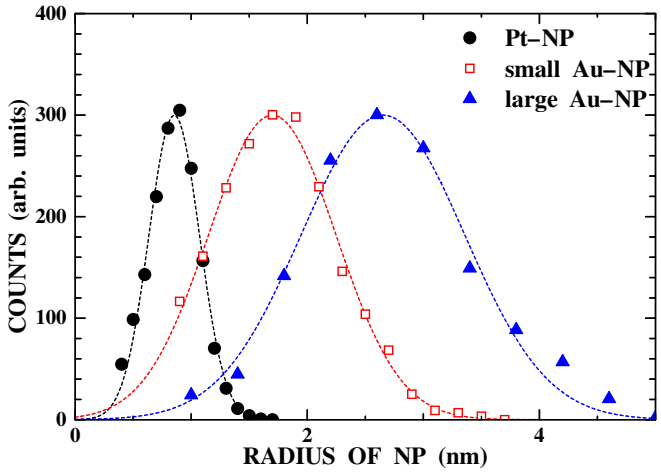

Fig. 2. Size distribution of platinum nanoparticles prepared by vacuum vapor deposition on a-SiN film (closed circles). The distributions of gold nanoparticles prepared by deposition of different amounts of gold are shown by squares and triangles. Larger nanoparticles were prepared by deposition of larger amount of gold (triangles). The lines show the results of Gaussian fitting.

was measured for each ion track. The observed closest distance represents the radius of the region where nanoparticles disappeared. The distributions of the measured closest distances for the platinum-deposited sample irradiated with 1.1 MeV C_{60}^{3+} are shown in Fig. 3 (circles). The distribution shows a Gaussian-like well-defined peak with a peak radius of 5.6 nm and a width of 1.6 nm. If the nanoparticles are desorbed from the circular region with a radius of R_c, the distribution of the closest distance R is given by [8]

$$\begin{aligned} P(R)dR &= 2\pi RN \exp\{-N\pi(R^2 - R_c^2)\}dR \quad \text{for } R > R_c \\ &= 0 \quad \text{for } R \leqslant R_c, \end{aligned} \tag{1}$$

where $P(R)dR$ denotes the probability that R is found in the interval $[R, R + dR]$. The average of the closest distance can be calculated by

$$\langle R \rangle = \int_0^\infty P(R)RdR = R_c + \frac{\exp(\pi N R_c^2)}{2\sqrt{N}} erfc(\sqrt{\pi N} R_c). \tag{2}$$

Substituting the observed results, $\langle R \rangle = 5.6 \pm 0.6$ nm and $N = 8.1 \times 10^{12}$ particles/cm^2, into Eq. (2), R_c was estimated to be 5.3 ± 0.6 nm. The distribution calculated using Eq. (1) with the determined R_c and N is shown by a dashed line in Fig. 3.

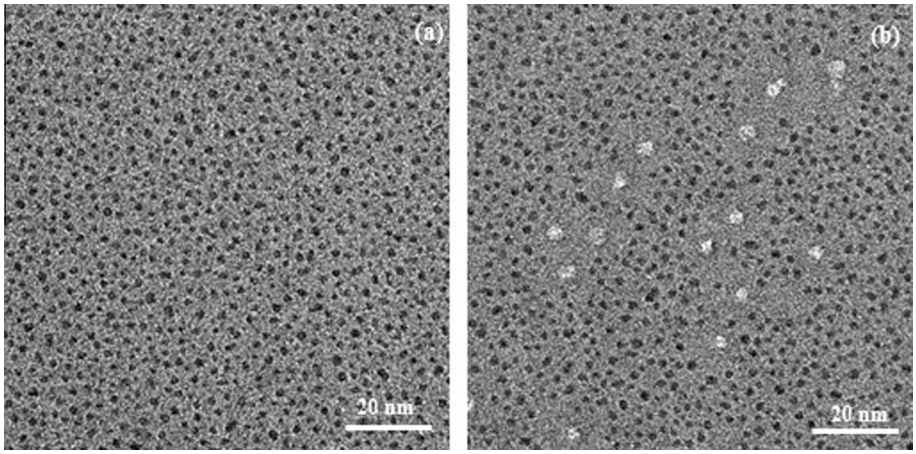

Fig. 1. TEM bright field images of platinum-deposited a-SiN films. The images observed (a) before and (b) after irradiation with 1.1 MeV C_{60} ions are shown. The ion tracks are seen as bright spots. The platinum nanoparticles disappeared from the vicinity of the ion track.