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Temperature of thermal spikes in amorphous silicon nitride films produced by 1.11 MeV C_{60}^{3+} impacts

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Gold nanoparticles with an average diameter of 3.6 nm were deposited on amorphous silicon nitride (a-SiN) films. These samples were irradiated with 1.11 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 gold nanoparticles disappeared from a surface area with a diameter of ~20 nm around each ion track. The disappeared nanoparticles were collected by a foil placed in front of the sample. Gold particles of circular shape with a diameter of several nm were observed on the collector foil using TEM, suggesting that the gold nanoparticles were emitted as liquid droplets from the a-SiN film upon impact of the C_{60} ion. In view of the previous molecular dynamics simulations (Anders et al., 2009), this indicates that the surface temperature rises above the melting point of gold in the region with a diameter of ~20 nm around the ion impact position.

1. Introduction

When a swift heavy ion penetrates through a solid, the solid electrons are excited along the ion trajectory. The initial electronic excitation leads to the formation of a hot plasma. The electrons may be heated to 10^4–10^5 K in the vicinity of the ion trajectories in a time period of 10^{-16}–10^{-13} s. Schiwietz and his coworkers observed Auger spectra induced by bombardment with swift heavy ions [1]. They found broadening of the ion induced Auger spectra, which was attributed to the high electron temperature around the impact position. Because the typical Auger decay time is $\sim 10^{-14}$ s this allows to measure the transient electron temperature in the first stage of the thermal spike formation induced by ion impact. The electron temperature estimated from the observed broadening was about 15,000 up to 85,000 K for projectile ions with $Z > 50$ at a few MeV/u [1].

According to the inelastic thermal spike (i-TS) model [2–4], a part of the electronic energy is, then, gradually transferred to target atoms via electron–phonon coupling. The temperature of atoms increases locally and consequently ion tracks are formed if the temperature exceeds the melting temperature. Because these processes occur in a short time period ($<10^{-10}$ s) and in a localized area of nm size, it is difficult to measure the atomic temperature of the thermal spikes.

Using molecular dynamics (MD) simulations, Anders et al. demonstrated that gold nanoparticles deposited on a target surface are desorbed when they are heated to their melting point [5]. Such desorption was actually observed by Baranov et al. [6]. This may be utilized to estimate the local temperature during the track formation. Gold nanoparticles are expected to be desorbed from the vicinity of the ion impact positions if the temperature is elevated higher than the melting point of gold during the formation of the ion tracks. In the present paper, the feasibility of this method is examined in the case of 1.11 MeV C_{60}^{3+} ion impacts on amorphous silicon nitride (a-SiN) films. The formation of ion tracks in a-SiN was already observed by Canut et al. using swift heavy ions [7].

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 $Si_{0.47}N_{0.53}$ using high-resolution Rutherford backscattering spectroscopy [8], which

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is slightly Si rich compared to the stoichiometric Si_3N_4. A small amount of gold was vapor deposited on one side of the a-SiN films at room temperature. The gold-deposited a-SiN films were irradiated with 1.11 MeV C_{60}^{3+} ions to a fluence of $\sim 5 \times 10^{10}$ ions/cm^2 with the 400-kV ion implanter of JAEA/Takasaki. The irradiation was performed either on the gold-deposited surface (will be referred to as a "front surface") or on the "rear surface" at normal incidence. Note that the projected range of the monoatomic carbon ion with the same velocity (18.5 keV C ion) was calculated to be 46 nm using SRIM2011 [9], indicating that the C_{60} ions penetrate through the a-SiN films. In the irradiation on the rear surface, a self-supporting SiO_2 foil (will be referred to as a "collector foil") was placed just behind the sample (facing to the gold-deposited surface) to collect gold nanoparticles emitted from the a-SiN film during the irradiation. The samples and the collector foil were observed using a transmission electron microscope (TEM, JEOL JEM-2200FS) equipped with a field emission gun operating at 200 kV.

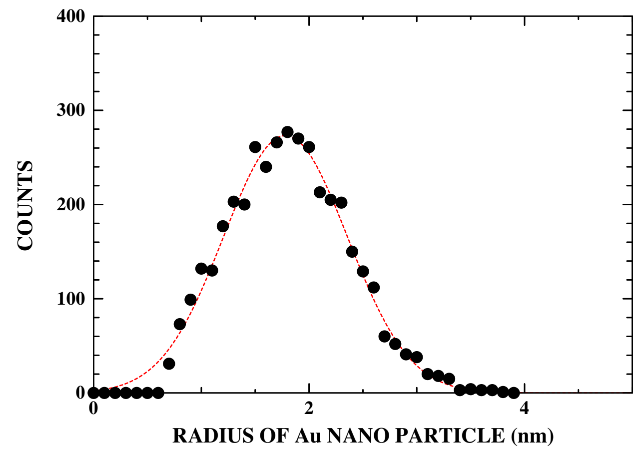

Fig. 2. Size distribution of nanoparticles deposited on a-SiN films. The curve is drawn to guide the eye.

3. Results and discussion

Fig. 1(a) shows an example of the TEM bright field images observed before irradiation. There are many gold nanoparticles formed by the gold vapor deposition. The areal density of these nanoparticles was measured to be 1.16×10^{12} particles/cm^2. The size distribution of these nanoparticles is shown in Fig. 2. The distribution shows a well-defined peak at a diameter of $\sim$3.6 nm. Assuming that the nanoparticle has a hemispherical shape, the average number of gold atoms in one nanoparticle was estimated to be $\sim$1000 from the measured distribution.

Fig. 1(b) and (c) show the TEM bright field images observed after irradiation with 1.11 MeV C_{60}^{3+} ions on the front and rear surfaces, respectively. In both micrographs, ion tracks are clearly seen as bright spots with a diameter of a few nm. Looking at the vicinity

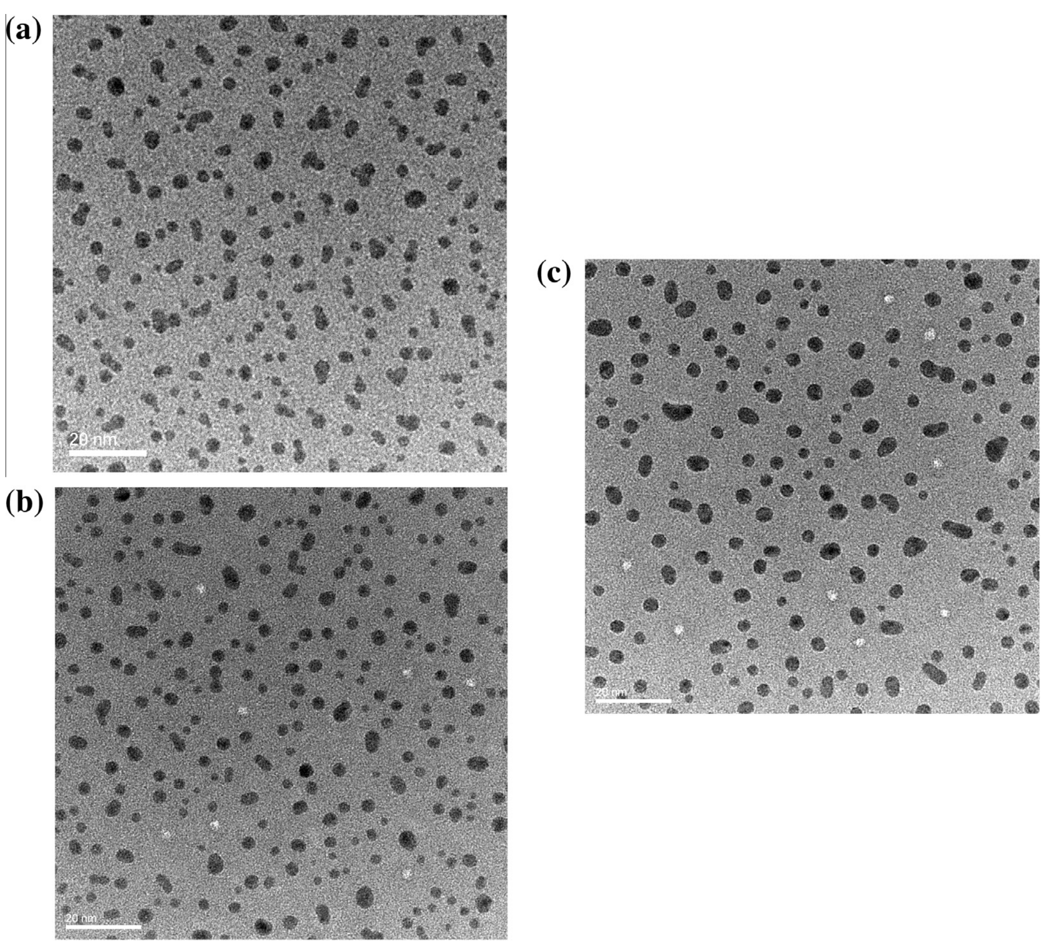

Fig. 1. TEM bright field images of gold-deposited a-SiN films. The as-grown sample (a), the samples after irradiation with 1.11 MeV C_{60}^{3+} ions on the front surface (b) and after irradiation with 1.11 MeV C_{60}^{3+} ions on the rear surface (c) are shown. The ion tracks are seen as bright spots. The gold nanoparticles disappeared from the vicinity of the ion track.