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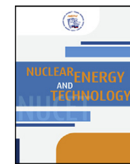

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Dynamically stable nanostructures in heavy-ion implanted silica glass

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Abstract

Previously reported experimental data have been used for kinetic analysis of the dynamically stable structure formation under implantation of 60 keV Au^- or Cu^- ions into silica glass. The kinetics has been analyzed in the frame of the one-dimensional model of evolution of the implant depth distribution, with taking into account the surface recession due to sputtering, as is also, with considering the local implant production, drift and diffusion.

The kinetic analysis has shown that the maximum number of implanted atoms retained in the irradiated substrate (maximum retained ion fluence) is independent of detailed ion-range distribution. The maximum retained ion fluence depends on ratios between the following quantities: the ion flux, the mean projectile range, the surface recession rate, the implant drift velocity and the implant diffusion coefficient. A method has been proposed for evaluation of the surface recession's role in the saturation kinetics. Estimations conducted for 60 keV Au^- ion implantation of silica glass have shown that the surface recession predominates in the saturation kinetics.

Surface recession does not dominate for 60 keV Cu^- ion implantation of silica glass. Formation of dynamically stable structures during 60 keV Cu^- ion implantation of silica glass cannot be explained if the drift of implants is excluded from considerations. Considerations have shown that the drift's contribution increases with increasing the ion flux. A mechanism of depleted region formation in the dynamically stable structures has been demonstrated. According to this mechanism, formation of a depleted region within the implanted layer is caused by expelling effect of electric field from the region where the electric field is zero. Necessary drift velocity is provided by Cu^+ solutes. A method utilizing images of the dynamically stable structures has been proposed for evaluation of the implant drift's role in the saturation kinetics.
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Keywords: Heavy-ion implantation; Radiation-induced processes; Nanostructures; Radiation resistance.

Introduction

Metal-nanoparticle composites are widely applicable to photonic devices [1–4]. To fabricate metal nanoparticles embedded into transparent insulator matrices, the high-flux heavy-ion implantation is often used [1]. The ion implantation at ion fluxes up to $10^{15}\cdot$ions/cm^{2}s is a powerful tool of overcoming the thermodynamic and kinetic limitations typical for many methods of fabrication, so that various metal nanoparticles can be quickly implanted into various matrices [5]. Recently, metal nanoparticles fabricated by the heavy-ion implantation have been used as precursors for conversion to oxide nanoparticles [6].

Optical spectroscopy measurements conducted during ion implantation have helped researchers to find the stages of nanocomposite formation, phase composition of the nanocomposites and chemical compositions of the phases [7–13]. In particular, optical spectra were measured during implantation of copper ions into silica glass. Spectra of optical absorption in the range of the surface plasmon resonance of metal nanoparticles provided information on the metal phase, and spectra of ion-induced luminescence gave information on implanted copper atoms in the solid solution. Fluence dependences of both the optical absorption and the luminescence intensity were measured at various ion fluxes, and several stages of formation of metal-nanoparticle composites were observed, namely, metallization, growth of nanoparticles and saturation. Eventually, the non-equilibrium phase diagrams,

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Peer-review under responsibility of National Research Nuclear University MEPhI (Moscow Engineering Physics Institute).

Russian text published: Izvestia Visshikh Uchebnikh Zavedeniy. Yadernaya Energetika (ISSN 0204-3327), 2016, n.1, pp. 21-29, n.2, pp. 17-25.

http://dx.doi.org/10.1016/j.nucet.2016.07.001
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with the ion flux and ion fluence taken as coordinates, were derived from these measurements [10–13].

The non-equilibrium phase diagrams consist but not entirely of various regions analogous to those known for equilibrium phase diagrams. In particular, these regions are a single phase region (the region of solid solution of implants) and a two-phase region (the region of solid solution with embedded metal nanoparticles), at low and intermediate ion fluences, respectively. In the single phase region, the implant concentration increases with increasing the ion fluence at a fixed ion flux. In the two-phase region, the portion of metal phase increases with increasing the ion fluence, but the composition of the solid solution does not vary. The non-equilibrium phase diagrams also comprise a region of saturation, which is a new region that has never been observed for equilibrium diagrams. In the state of saturation induced by irradiation at a fixed ion flux, the depth and size distributions of nanoparticles, as is also, the chemical and phase compositions of nanocomposites do not vary with increasing the ion fluence. That is, participating in the mass and energy exchange with the environment, the nanocomposites eventually reveal their dynamic stability as the saturation state. As a result, further ion implantation becomes inefficient. Very often, bimodal depth and size distributions of nanoparticles are observed for the dynamically stable nanostructures. For example, in silica glass, large nanoparticles were observed in the sub-surface layer separated by a depleted region from small nanoparticles located in the depth [14].

In the paper presented in [10] authors discussed roles of radiation-induced processes (surface recession, radiation-stimulated diffusion, atomic collisions, electronic excitation, etc.) in the saturation, and lacking understanding of the saturation kinetics was pointed out. In particular, the mechanism of formation of depleted regions in the ion-implanted nanocomposites has not been understood. In the present study, kinetics of the saturation is analyzed more thoroughly, aimed at finding approaches to evaluate contributions of radiation-induced processes.

Surface recession

Heavy-ion implantation results in surface recession due to sputtering. Data on surface recession for silica glass implanted by 60 keV Cu^- и Au^- ions at various ion fluxes to a fixed ion fluence of $2\cdot10^{17}\cdot$ions/cm^2 are shown in Fig. 1. Thicknesses of sputtered layers are of the order of the mean projectile ranges. These results suggest that the saturation is caused by the ion-induced surface recession competing with accumulation of implants [10]. Provided that only these two contributions are taken into account, the time variation of the implant concentration profile can be expressed by the following equation

$$\frac{\partial N(x,t)}{\partial t} = G(x) + SF\frac{\partial N(x,t)}{\partial x}, \tag{1}$$

where $N(x,t)$ is a time-dependent local implant number density, $G(x)$ is a local implantation rate (a product of the ion

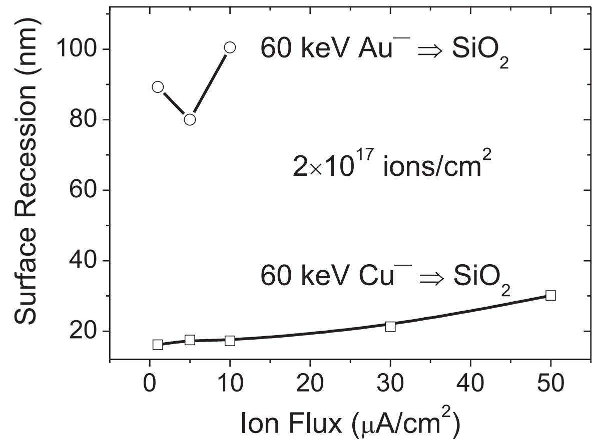

Fig. 1. Surface recession for silica glass implanted by 60 keV Au^- и Cu^- ions at various ion fluxes to a fixed ion fluence of $2\cdot10^{17}\cdot$ions/cm^2.

flux and the probability distribution of ion ranges), S is a sputtering efficiency (sputtered thickness per unit ion fluence), F is an ion flux. In this paper we assume that the ion flux is directed from the left to the right (along x axis), and the implanted surface is located at $x=0$. Accordingly, the implantation rate integrated over depth is equal to the ion flux

$$F = \int_0^\infty G(x)dx. \tag{2}$$

Measurements of depth dependences of the local implantation rate $G(x)$ is a severe experimental problem. The distribution of ion ranges and, accordingly, the local implantation rate can be calculated with the help of TRIM [15] or TRIDYN [16] codes. Let us show, however, that neither measurements nor calculations of $G(x)$ are necessary to evaluate the contribution from the sputtering.

As the first approximation, it is assumed that both the local implantation rate $G(x)$ and the sputtering efficiency S do not vary with time. Given the initial conditions of $N(x,0)=0$, solutions of Eq. (1) can be represented by the following formula

$$N(x,t) = \int_0^t G(x+SF\tau)d\tau. \tag{3}$$

The time dependence of retained ion fluence $\Phi(t)$ can be derived from Eq. (3)

$$\Phi(t) = \int_0^\infty N(x,t)dx = \int_0^t d\tau \int_0^\infty G(x+SF\tau)dx. \tag{4}$$

The retained ion fluence is a number of implanted atoms (implants per unit surface) retained in the irradiated substrate, that is $\Phi(t) \le Ft$.

The following steady-state concentration profile corresponds to the saturation

$$N(x,\infty) = \frac{1}{SF}\int_x^\infty G(x)dx. \tag{5}$$

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