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Principles of the Construction and Computer Simulation of a Source of Homogeneous and Heterogeneous Cluster Ions

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Abstract—The principles of constructing a sputtering-type high-current source of homogeneous and heterogeneous cluster ions are comprehensively analyzed. The results of analysis are used to perform computer simulation of the given source. The ion-optical scheme and structural model of a new cluster ion source are developed.

Keywords: cluster ion source, ion sputtering, cascade amplification of cluster ion currents

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INTRODUCTION

Progress in ion technologies requires the advanced development of tooling backup, including cluster ion sources of solid-state elements. Their emission is most simply implemented under the condition that targets are sputtered with atomic-cesium ion beams with energies of several kiloelectronvolts under linear collision-cascade conditions [1–4]. A number of advantages of sputtering-type sources (stable currents of negative atomic and cluster ions of practically all solid-state elements, a long operating life under single-source loading with a working substance (cesium or CsCl salt), effectiveness, small overall sizes, simplicity in manufacturing and servicing, and the absence of additional means of vacuum pumping) have stimulated their application in research and analytical activities [3–6]. However, sputtering-type sources have found no application in technology due to the low intensity of cluster-ion currents. In this context, an important problem lies in creating sputtering-type high-current sources capable of generating cluster ions of solid-state elements because they are promising for obtaining new materials and coatings and modifying surfaces, including the treatment of materials and structures in the micro- and nanometer ranges.

In this work, from the analysis of known data concerning the ion sputtering of microscopic metallic targets and individual isolated nanoparticles [7], we propose methods for improving the efficiency of metal sputtering in the form of cluster ions, formulate a fundamentally new approach to designing a sputtering-type high-current source capable of producing cluster ions of solid-state elements, calculate the ion-optical system of the formation and transportation of ion beams, and also design a device model ensuring source utilization in technological problems.

PRINCIPLES OF CONSTRUCTING SPUTTERING-TYPE HIGH-CURRENT SOURCES OF CLUSTERS IONS

The essence of the proposed approach is that the following factors stimulating the enhanced emission of cluster ions are jointly employed.

Ion Sputtering of Metals in the Nonlinear Collision-Cascade Mode

Cluster-emission mechanisms are different under linear and nonlinear collision-cascade conditions. The linear mode is implemented if metals are bombarded with atomic ions with an energy of several kiloelectronvolts and characterized by the fact a low moving atom density in the collision-cascade volume [8]. Under these conditions, clusters are emitted because correlated momenta are simultaneously transferred to a group of neighboring atoms located in a surface metal layer [9, 10], and their charge is formed upon departure from a target by reason of electron exchange with the surface [11].

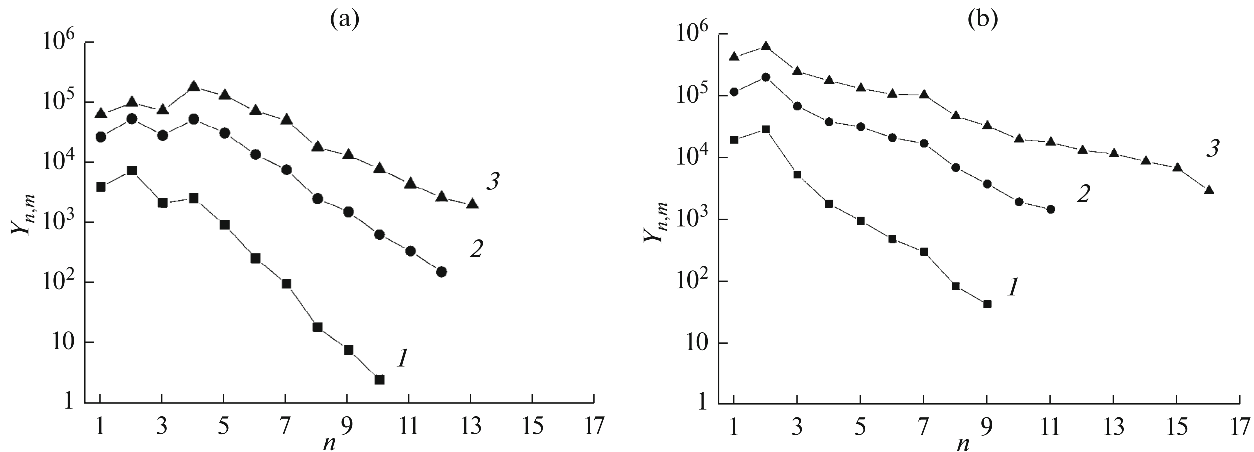

Fig. 1. Normalized mass spectra (dependences between the yield $Y_{n,\,m}$ and the number n of cluster atoms) constructed for (a) Ta_n^+ and (b) Nb_n^+ secondary ions sputtered from Ta and Nb targets with the help of Au_m^- primary ions with energies of (*1*) 6 ($m = 1$), (*2*) 12 ($m = 2$), and (*3*) 18 keV ($m = 3$).

Cluster-ion emission can be enhanced if polyatomic ions are employed as bombarding particles [5, 12]. As compared to atomic ions, when metals are bombarded with polyatomic ions with the same velocity, a higher density of moving atoms is initiated in the cascade volume (in the nonlinear collision-cascade mode) and a thermal peak is formed [13, 14]. The thermal peak is understood as a local region with a high energy density, which is located near the surface and generated during the final stages of cascade evolution ($t \sim 10^{-11}$ s) if the bombarding-ion energy distributed between atoms of the given region gives rise to the excitation level (average energy per atom) corresponding to the "temperature" T_0. At $T_0 > T_{cr}$ (T_{cr} is the critical point of the target material), the material undergoes a phase transition and is converted into a dense vapor. The metal–vapor interface disappears and, consequently, the mechanism of the formation of the sputtered-particle charge is not determined by electron exchange with the surface. Under these conditions close to those existing in plasma, the atomic-particle charge is created due to the impact-ionization mechanism [15], for which the degree of ionization is specified by the Saha equation [16]. A relatively large ion concentration in the metallic vapor flux (ions play the role of condensation centers) stimulates atomic-particle condensation processes [17]. In the case of $T_0 > T_{cr}$, atomic particles will effectively condense even at high vapor temperatures when the average energy of thermal motion of atoms and ions is less than the dissociation energy of dimers and clusters. Thus, at $T_0 > T_{cr}$, thermal-peak relaxation is accompanied by the fact that a dense metallic vapor flux is ejected into free space. At the initial stage of its expansion, atoms and atomic ions are subjected to multiple collisions and a significant part of the atomic particles is condensed, ensuring an anomalously high nonadaptive increase in the cluster-ion yield intensity as compared to the linear mode [13]. For ions sputtered under linear and nonlinear collision-cascade conditions, the ion-yield intensities are compared in Fig. 1 (data were taken from [12]).

Atomic Ion Sputtering of Nanoparticles

From the viewpoint of energy, the sputtering of metals is an inefficient process under linear collision-cascade conditions because only a small part of the energy released by bombarding ions during deceleration within a target material is immediately transferred to sputtered particles (estimations indicate that the transferred energy is less than 0.1%). A greater part of the energy is spent on the creation of radiation defects, excitation of the phonon and electron subsystems, and, then, dissipates over the target volume due to thermal and electrical conduction processes. To a somewhat lesser extent, this statement refers to sputtering in the nonlinear collision-cascade mode. As was shown by computer calculations [18, 19], additional enhancement in cluster-ion emission can be implemented under the condition that isolated nanoparticles are bombarded with atomic ions. Indeed, for isolated nanoparticles, there are no channels of energy dissipation caused by thermal and electrical conduction processes. Hence, all other things being equal, the density of energy released by an atomic ion in the limited volume of a nanoparticle will exceed that liberated in the limited volume of a macroscopic target. This circumstance must increase the nanoparticle-sputtering efficiency because the sputtered-particle-yield intensity is proportional to the released energy density under linear cascade conditions. It is of interest to note that an increase in cluster-ion emission from individual nanoparticles bombarded with polyatomic ions (nanoscale effect of sputtering") was first observed experimentally in [20, 21]. It is of fundamental importance that, in the case of an individual nanoparticle sputtered with atomic ions, the released energy density

($\sim 1/d^3$) can be varied over a wide range during changes in the characteristic size d of nanoparticles. At certain d, the density of energy liberated by an atomic ion in a nanoparticle can exceed that released by a polyatomic ion in a macroscopic target. In other words, when size d decreases, not only the nanoparticle-sputtering efficiency can be enhanced but also the sputtering mechanism itself can change upon its conversion from the linear collision-cascade mode to the nonlinear one [7]. Such a conversion must be accompanied by further enhancement in cluster-ion emission. Thus, together with the bombarding-ion characteristics and the type of material of sputtered nanoparticles, the characteristic size d of nanoparticles is an additional independent parameter that determines both the efficiency and the nanoparticle-sputtering mode and, consequently, specifies the sputtered-cluster-ion yield intensity.

Use of a Specially Prepared Target (Cluster-Ion Emitter)

During ion bombardment, the nanoparticle size d decreases (up to its complete decomposition) and the sputtered-cluster-ion yield intensity varies over time. To achieve invariable cluster emission, a model macroscopic target ("cluster-ion emitter") produced from a statistically large collection of weakly interacting nanoparticles must be sputtered instead of an isolated nanoparticle. It is assumed that, in this case, the sputtered-cluster yield intensity averaged over a large number of nanoparticles will be stable in time. The mechanism underlying the sputtering of cluster-ion emitters will not differ considerably from that responsible for isolated nanoparticle sputtering if the contact area of the nearest adjacent nanoparticles is much less than the surface area of an individual nanoparticle because, in this case, the channels of energy dissipation in a fixed nanoparticle volume are sufficiently suppressed at the expense of thermal and electrical conduction processes.

Cascade Amplification of the Cluster-Ion Current

It is important that, in the case where emitters made of nanoparticles are sputtered, the use of cluster ions, rather than atomic ones, must lead to a greater amplification of cluster emission. Such an approach is of interest because the cascade amplification of cluster emission can be performed by creating a special-purpose device ("cluster-emission multiplier") [22] involving a sequence of emitters by analogy with the technique implemented in a secondary electron multiplier. In this sequence, only the last emitter will ensure the emission of cluster ions of the given element needed to solve the assigned technological problem, while intermediate emitters only enhance the cluster-ion flux (these cluster ions can be composed of atoms of other elements) intended for sputtering of the last emitter. Hence, intermediate emitters can be constructed from nanoparticles or be specially selected

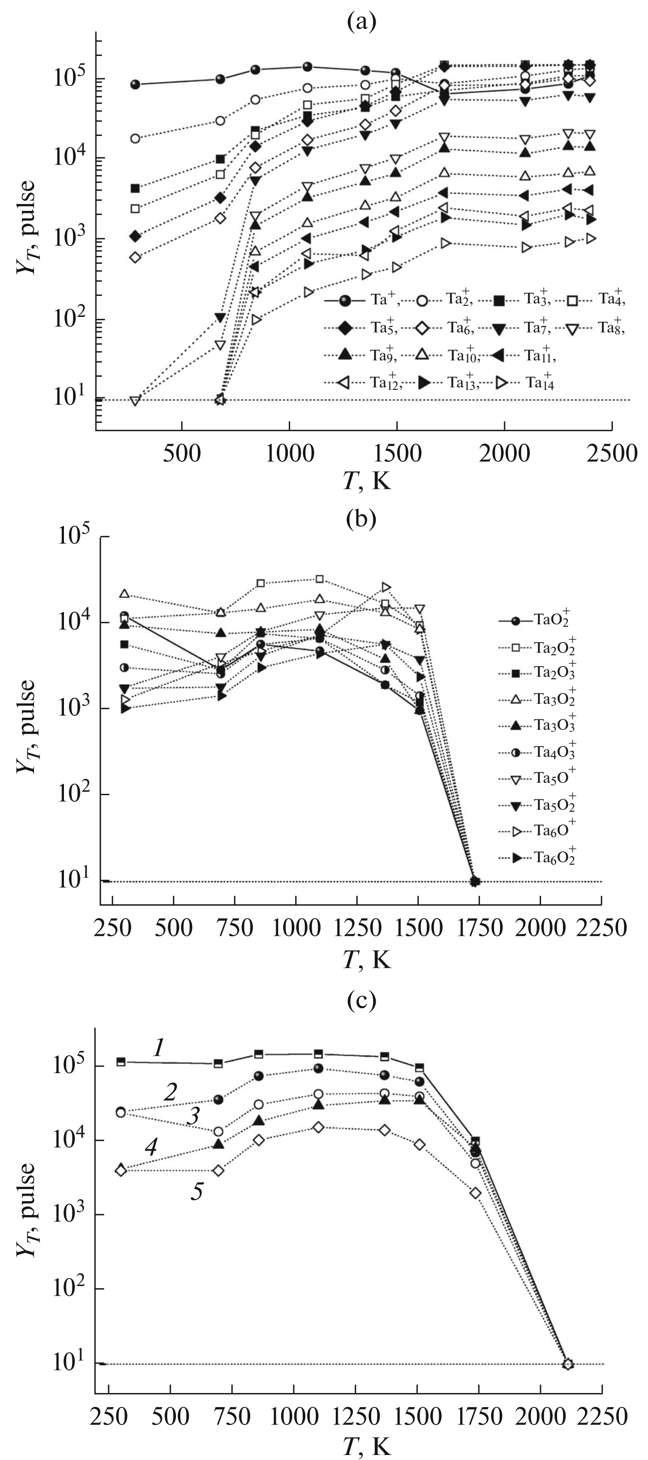

Fig. 2. Dependences between the ion yield intensity Y_T and the temperature T obtained under the bombardment of tantalum with Au_3^- ions with an energy of 18 keV: (a) Ta_n^+ ($n = 1–14$); (b) $Ta_nO_m^+$ ($n = 1–6$, $m = 1–3$); and (c) (*1*) Au^+ and TaO^+, (*2*) $TaAu^+$ and Ta_2O^+, (*3*) Ta_2Au^+ and Ta_3O^+, (*4*) Ta_3Au^+ and Ta_4O^+, and (*5*) Ta_2Au_2 and $Ta_4O_2^+$. Here, the horizontal dashed lines designate the background-signal levels.

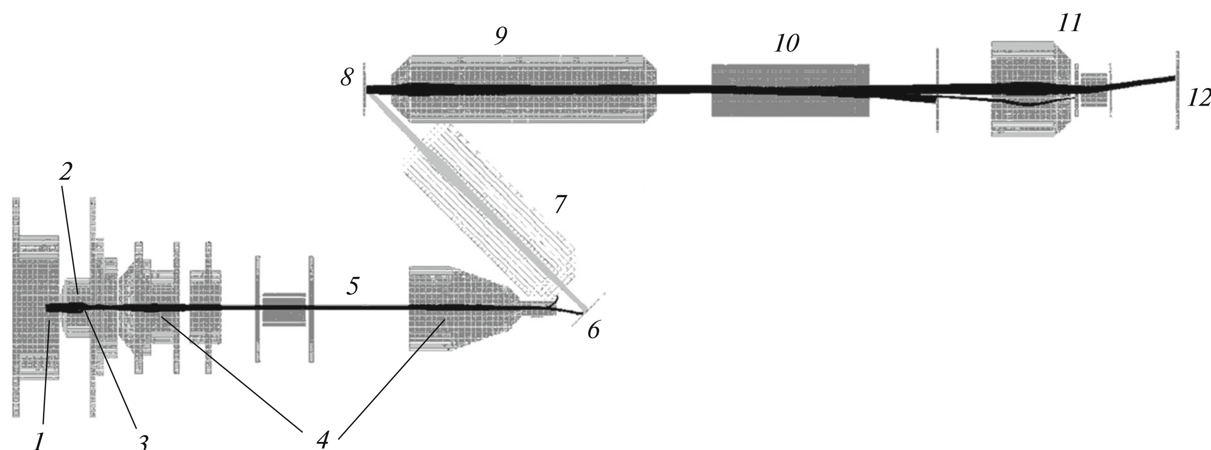

Fig. 3. Computer simulation performed with the help of SIMION 8.0 software and illustrating the formation of a cluster-ion beam upon ion passage through the three-cascade amplification system. Here, *1* is the Cs^+ ion source, *2* is the gold target no. 1, *3* is the cluster Au_n^- ion source, *4* is the optics of the formation of a Au_n^- ion beam (cascade no. 1), *5* is a Au_n^- ion beam, *6* is target no. 2, *7* is the optics of the formation of a cluster-ion beam (cascade no. 2), *8* is target no. 3, *9* is the optics of the formation of the cluster-ion beam (cascade no. 3), *10* is the Wien filter, *11* is the optics of control and focusing of the beam of cluster ions deposited onto the technological substrate of cluster ions, and *12* is the technological substrate.

composite inorganic materials whose sputtering provides the most intense generation of cluster ions.

Obtainment of Homogeneous and Heterogeneous Ion Beams

For ion technologies, it is interesting to obtain both homogeneous M_n^+ and heterogeneous $M_nX_m^+$ cluster ions, where M is the metal atom and X is either an impurity metal (e.g., niobium or gold [23]) atom or gas (e.g., oxygen and nitrogen) atoms. It was demonstrated [23] that, in the case of homogeneous and heterogeneous cluster ions, their generation depends strongly on the conditions implemented on the sputtered target surface. When a target heated to high temperatures is bombarded, the degree to which its surface is covered with the adsorbed molecules of residual gas decreases appreciably and homogeneous ions contribute predominantly to cluster-ion emission. During bombardment of a target subjected to room or lower temperatures, heterogeneous cluster ions contribute mainly to cluster emission. The dependences between the yield intensity of homogeneous and heterogeneous cluster ions sputtered from a tantalum target by means of Au_3^- ions with an energy of 18 keV and the target temperature are presented in Fig. 2 (data were taken from [23]). This circumstance enables us to control cluster-ion emission and, if necessary, initiate the emission of homogeneous M_n^+ or heterogeneous $M_nX_m^+$ ions of the given type, which is attained by heating the last emitter of the cluster-ion multiplier or supplying gas (e.g., oxygen or nitrogen) fluxes to its surface.

CALCULATION AND DESIGN OF THE CLUSTER-ION SOURCE

Computer Simulation of the Cluster-Ion Source

The proposed cluster-ion-based methods for increasing the metal sputtering efficiency was employed to structurally simulate a sputtering-type high-current source of cluster ions of solid-state elements. The basic components of the given source are a high-current source of bombarding atomic-cesium ions [1, 3, 24]; a cascade amplification system incorporating a sequence of three model macroscopic targets (cluster-ion emitters), each constructed from a statistically large set of weakly interacting nanoparticles, which is used to enhance cluster ion fluxes; the ion-optical system whereby cluster-ion beams are accelerated, formed, and focused in each amplification cascade; and a system for the mass separation of cluster ions sputtered from the last emitter (cluster-ion multiplier), i.e., a Wien filter. During simulation, the electrical potential distribution over electrodes of the ion-optical system was selected so that the energy of cluster ions sputtered from the last emitter (cluster ion multiplier) was in the range of 20–500 eV. Such an energy level ensures both the best mass resolution when cluster ions pass through the Wien filter and the nondestructive deposition of clusters on the sample surface.

Figure 3 presents the results of computer simulation of the formation of a cluster-ion beam penetrating through the three-cascade amplification system. Calculations were carried out using SIMION 8.0 software [25].

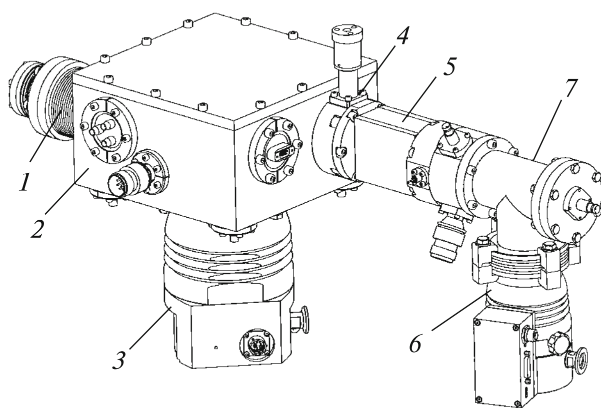

Fig. 4. Overall view of the device intended for cluster-ion source utilization in applied problems: *1* is the Cs^+ ion source, *2* is the vacuum chamber, *3* is the turbopump, *4* is the vacuum lock, *5* is the Wien filter, *6* is the turbopump, and *7* is the chamber where the technological substrate is located.

Model of the Device with the Help of which Sputtering-Type Sources of Cluster Ions of Solid-State Elements Can Be Employed in Applied Problems

The computer simulation results were used to develop a device by which the ion source can be employed in applied problems concerning cluster-ion deposition onto a sample surface (Fig. 4). The device includes a vacuum chamber in which the source and the three-cascade system of amplification of cluster-ion currents are located, a chamber intended for sample arrangement, a vacuum lock enabling rapid sample replacement without depressurization of the chamber with the source, a cluster-ion-beam mass separation system (Wien filter), and a vacuum-pumping system.

CONCLUSIONS

Due to the analysis of methods whereby the sputtering efficiency of solid-element targets made of cluster ions is improved using polyatomic ions as bombarding particles, creation of conditions under which the nanosize effect is implemented during ion sputtering, application of model targets constructed from a statistically large collection of weakly interacting nanoparticles, and employment of the principle of the cascade amplification of cluster-ion currents, the structure of a high-current source based on the sputtering of homogeneous M_n^+ and heterogeneous $M_nX_m^+$ cluster ions of solid-state elements has been developed and a model of the device by which this source can be employed in applied problems has been created.

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