On the Effect of the Crystal Structure in the Sputtering of Two-Component Single-Crystal Structures

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Abstract—The transmission sputtering of component atoms under ion bombardment of the (0001) face of a two-component VSi_2 single crystal is studied using molecular-dynamics computer simulation. The sputtering

of component atoms is calculated in the case of the sputtering of virtual VV₂ and Si'Si₂ single crystals consisting of atoms with the same masses, and the following new effect in the selective sputtering is studied: atoms from vanadium sites are preferentially transmission sputtered in the case of the same masses and binding energies of component atoms. This effect can be called the effect of nonidentity of the component sites in the complex VSi₂ lattice (of the C40 type) with respect to the momentum propagation in collision cascades, i.e., the effect of structure in the selective sputtering of two-component single crystal targets.

Keywords: sputtering, ion transmission, selective sputtering, transmission sputtering, two-component targets, VSi_2 , thin single crystal films

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INTRODUCTION

The selective sputtering of component atoms is observed in the case of the sputtering of multicomponent materials by ions in the case of small bombardment doses. The sputtering yields of component atoms turn out to be not proportional to their concentration at the surface, and atoms of one of the components are preferentially sputtered. To analyze the selective sputtering of two-component targets, the Andersen—Sigmund formula is often used [1]:

$$\frac{Y_1}{Y_2} = \frac{C_1}{C_2} \left(\frac{M_2}{M_1}\right)^{2m} \left(\frac{U_2}{U_1}\right)^{1-2m},\tag{1}$$

where Y_i is the sputtering yield of the *i*th-component atoms, U_i is the binding energy of atoms of the *i*th component, $i = 1, 2, C_i$ is the concentration of the *i*thcomponent atoms at the surface, and M_i is the mass of atoms of the *i*th component. Here *m* is the interaction potential steepness parameter, m = 0.165 corresponding to the "universal" potential, which is used successfully in many studies.

This analytical formula is used only for amorphous solids. It contains the ambiguously defined fitting parameter *m*. In addition, the dependences of the preferential sputtering of atoms of one of the components on other parameters (for example, on the energy, masses, and incidence angles of bombarding ions, and the crystal structure of the target) are revealed (for example, [2-5]).

The review of experiments gives satisfactory agreement with the Andersen—Sigmund formula with respect to the binding energy of component atoms and ambiguous results with respect to their masses [6-8].

The sputtering of targets consisting of a mixture of isotopes (the binding energies for them are assumed to be the same) were studied to separate the influence of the masses and the binding energies of component atoms. In the case of small bombardment doses, experimental studies of the selective sputtering of isotopes showed that the emission of atoms with smaller masses was preferential [9, 10]. Using the developed analytical model, the authors of [11] studied the surface mechanism of the preferential sputtering of atoms of a lighter isotope taking into account the fact that atoms of the lighter isotope deflect toward the normal to the surface more strongly when scattering at surrounding atoms of the heavier isotope in the case of emission from the surface.

However, as a rule, the isotope masses change insignificantly. A number of researchers have developed models of virtual materials and alloys (for example [2, 3] and [12, 13]). The authors of [12] studied the sputtering of a Cu single crystal (changing the atom atom interaction potential) and the sputtering of systems with different masses (they used O, Cu, Au, and their "alloys" in the upper single crystal layer) in the case of a constant binding energy. Similar computer experiments were conducted in [13]: Cu atoms and Cu atoms with double masses were taken. The binding energy ranged from 50 to 200%. The authors of both papers stressed the complicated dependence of the selective sputtering on the target-atom masses and its difference from the theoretical formula [1]: an unambiguous dependence on the target-component masses was not discovered. The preferential sputtering of atoms of light and heavy components can be observed. An approximately inversely proportional dependence on the binding energy was observed. The authors of [2, 3] simulated the sputtering of a virtual amorphous target consisting of two Mo isotopes with masses of 50 and 100 amu.

Using the molecular-dynamics model, the author of [4] calculated the backward and transmission sputtering of component atoms under ion bombardment of the VSi₂ single crystal (0001) face. In this paper, the author calculated the sputtering of component atoms

in the case of the sputtering of virtual VV₂' and Si'Si₂ single crystals consisting of atoms of the same masses and discovered a new effect in selective sputtering: atoms from V sites were preferentially backward and transmission sputtered in the case of the same masses and binding energies of the component atoms. This effect can be called the effect of the nonidentity of component sites in the complex VSi₂ lattice (of the C40 type) with respect to momentum propagation in collision cascades, i.e., the effect of structure in the selective sputtering of two-component single-crystal targets. The discovered effect was also studied in [14].

Using the molecular-dynamics model, the authors of [5, 15] studied the role of the surface mechanism (i.e., the scattering of sputtered component atoms at surrounding surface atoms in the case of their emission from the surface) in the selective sputtering of single crystals. It was shown that the surface mechanism of the preferential sputtering of atoms of the light component is based on the stronger deflection of atoms of the light component toward the normal to the surface during their scattering at surrounding atoms of the heavy component in the case of their emission from the surface.

The authors of [16-18] studied the difference between the energy spectra of component atoms sputtered from the surfaces of two-component WSi, MoSi, and VSi crystals; it appeared as a result of the scattering of sputtered component atoms at surrounding surface atoms in the case of their emission from the surface. It was shown that the strong energy redistribution of component atoms occurs at the stage of emission; it leads to the appearance of peculiarities of the experimentally observed energy spectra of component atoms, in particular, of the maximum of the energy spectrum of sputtered atoms of the light component, which was shifted toward smaller energies as compared with the position of the energy spectrum maximum of sputtered atoms of the heavy component [19].

The large contribution of the surface mechanism, i.e., collisions between sputtered atoms and surround-

ing-surface atoms during emission, to the formation of the low-energy part of the energy spectra of sputteredcomponent atoms was revealed, and the "two-cone" structure of the emission of sputtered low-energy atoms was discovered [16-18].

Interest in effects occurring under the ion bombardment of two-component targets has significantly increased recently. In this case, the role of simulation in revealing the mechanisms determining the peculiarities observed in the experiments, including the ion bombardment of two-component targets, turned out to be very significant. We can mention recent successful studies of segregation [20–24], an effort to decrease the preferential emission of O atoms from a two-component target [25], and computer studies of disorder in multicomponent alloys under ion bombardment using the molecular-dynamics method [26, 27].

The aim of this paper is to study the dependence of the selective sputtering of a two-component singlecrystal target under ion bombardment on the targetatom mass by means of a model excluding the effect of the difference between the binding energies on the yield of component-atom sputtering and also continue to study the effect of structure [4, 14] in the selective sputtering of component atoms.

CALCULATION MODEL

The calculation model was described in [4, 5, 28]. Here we give its special features only briefly.

We calculated the characteristics of the ion transmission and transmission sputtering of a VSi₂ single crystal film and compared them with similar characteristics of ion transmission and transmission sputtering under the ion bombardment of virtual VV₂' and Si'Si₂ single-crystal films. In this case, the crystal lat-

tices for pure VV'_2 and Si'Si₂ were the same as for VSi_2 , which is similar to [4]. This gave the possibility of not only studying the dependence of sputtering on the atomic mass, but also of revealing the role of the crystal structure in selective sputtering.

The sputtering of solids under ion bombardment can be considered as a classical problem of the manybody interaction. Writing the equation of motion of Npoint particles with the masses m_i , from the second Newton equation, we obtained a system of 6N first-order differential equations with the initial conditions [29]:

$$n \mathrm{d} v_{ki} / \mathrm{d} t = F_{ki}, \ \mathrm{d} x_{ki} / \mathrm{d} t = v_{ki}, \tag{2}$$

where F_i is the resultant of all forces acting on the particle with the number *i* from all other particles (atoms and ions), i = 1, ..., N; k = 1, 2, 3. Determination of these forces is a separate complicated problem. To simplify the problem, we used approximations: the forces were pair, central. and potential. In a series of research problems in physics, the authors often used interatomic interaction potentials, the parameters of



Fig. 1. Arrangement of VSi₂ single crystal atoms (the C40 lattice) in the projection on the (0001) face surface. A crystallite consisting of 47 atoms located in three layers that are parallel to the surface is shown. The formula of alternation of layers is ABC, where A is the atomic layer at the surface, its atoms are denoted by black color; the second atomic layer B consists of atoms denoted by the grey color; and the third atomic layer C, of atoms denoted by the white color.

which are determined by fitting to some experimental data (the lattice constant, the thermal-expansion coefficient, the sublimation energy, and so on were used): Morse, Lennard–Jones, Born–Mayer potentials, and others.

Such systems of equations are solved numerically using difference schemes. The choice of the difference scheme also depends on the approximations of interaction forces. For multiparticle dynamic interaction, the second-order Runge—Kutta method is often used [30].

The calculation was carried out using the molecular-dynamics method. Kr^+ ions bombarded three ultrathin single-crystal films: VSi_2 and virtual VV'_2 and $Si'Si_2$ with the same crystal structure as that of VSi_2 . We describe the virtual crystals in more detail and clarify the notations VV'_2 and $Si'Si_2$. The virtual crystal VV'_2 consisted of V atoms located at their lattice sites and of V' atoms located at Si lattice sites. In this case, V' atoms had the same masses and binding energies at the surface as V atoms. Thus, the notation V' denoted the same V atoms, but they were located at Si lattice sites before ion bombardment. In this case, we distinguished the transmission sputtering yields of V and V' atoms in our calculations. Similarly, the virtual Si'Si₂ crystal consisted of Si atoms located at their lattice sites and of Si' atoms located at V lattice sites. In this case, Si' atoms had the same masses and binding energies at the surface as Si atoms. Thus, the notation Si' denoted the same Si atoms, but located at V lattice sites before ion bombardment. We distinguish the transmission sputtering yields of Si and Si' atoms in our calculations.

Each of the crystallites consisted of 47 atoms located in three atomic layers that were parallel to the surface (Fig. 1). The authors of [31] compared the calculated results for targets consisting of 397 and 47 atoms. Kr⁺ ions were incident perpendicularly to the (0001) face surface, their energy E_0 ranged from 50 eV to 100 keV. 1051 ion were incident on each crystallite for each energy E_0 .

The real atom-atom interaction forces (repulsion-attraction ones) were replaced with repulsion forces, and attraction to the crystal was simulated by a spherical potential barrier during atom emission from the surface. As the atom-atom and ion-atom interaction potentials, we used the Born-Mayer potential, which was smoothly conjugated to the inversely quadratic potential that was similar to the composite potential [32]. The use of the repulsion potential instead of the repulsion-attraction one gives the possibility of separating the contributions of collision cascades and the binding energy to the sputtering characteristics. The method of the "average force" (the second-order Runge-Kutta method) was used to integrate the equations of motion. Using the same potential barrier with a height of 4.64 eV (equal to the sublimation energy for Si [33]) for V and Si, we excluded the influence of the difference between the binding energies of the component atoms on selective sputtering.

We considered transmission of Kr ions and the transmission sputtering of component atoms separately from V and Si lattice sites.

RESULTS AND DISCUSSION

Ion transmission through thin single-crystal films. Figure 2 shows the dependences of the Kr⁺ ion transmission transmission coefficient on the energy E_0 . A steady increase in the ion transmission coefficient is observed for all targets as the energy E_0 increases. For a fixed energy E_0 , the coefficient of ion transmission through the Si'Si₂ crystal is larger than through the

other samples and that through the VV'_2 is smaller than through the other samples. This is explained by the difference between the Kr⁺-Si and Kr⁺-V interaction cross sections. In addition, the energy transfer from Kr⁺ to Si atoms is smaller than to V atoms because of the larger difference between the masses. Therefore, when interacting with V atoms, Kr⁺ ions lose a larger fraction of their energy than during the interaction with Si atoms. In connection with this, the larger the



Fig. 2. Dependences of the Kr^+ -ion transmission transmission coefficients on the initial energy E_0 under bombardment of the (0001) faces of the VSi₂, Si'Si₂, and VV'₂ crystals.

amount of vanadium in the target, the greater the probability for the krypton ion to lose its energy.

Obviously, the probability of ion transmission must depend on the concentration of components in the target, which we observe: the number of transmitted ions normalized to that of bombarding ions, i.e., the ion transmission transmission coefficient R_{\downarrow} , in the energy range of E_0 100–300 eV, depends on the component concentration approximately as follows

$$3R_{\downarrow VSi_2} = 2R_{\downarrow Si'Si_2} + R_{\downarrow VV'_2} \tag{3}$$

(Table 1). As the initial ion energy increases, a deviation of the ion transmission coefficient from linearity is observed. At high energies, the interaction cross section tends to zero, and, accordingly, the ion transmission coefficient tends to 1.0. In the range of small energies E_0 , the following peculiarity is observed: the coefficient of ion transmission through the VSi₂ crystal turns out to be larger than through the Si'Si₂ crystal. **Transmission sputtering of component atoms.** Figure 3 shows the coefficients of the transmission sputtering of atoms from different sites normalized to the stoichiometric ratio (the ratio of the component concentrations) for the VSi₂ sample that did not undergo ion bombardment (1 : 2). For all energies of Kr⁺ ions, the number of atoms transmission sputtered from the virtual Si'Si₂ sample is smaller than that from the other samples. This can be explained by the difference between the interaction cross sections and the energy transfer coefficients of Kr⁺–Si and Kr⁺–V. For energies of bombarding ions of $E_0 > 100$ eV, the maximum number of atoms is transmission sputtered from the virtual VV₂ sample, as was expected. For small energies of bombarding ions, the number of atoms trans-

mission sputtered from VV'_2 is smaller than that from VSi_2 . However, a larger number of atoms is sputtered backward, and, on the whole, the number of atoms

emitted from the virtual VV₂' is larger than that emitted from other samples for all energies E_0 . The fact that the

number of atoms transmission sputtered from VV_2' is smaller for low energies of bombarding ions is evidence of the larger "density" of the sample: the particles collide more frequently and have time to change their directions and eject backward. For this reason, the maximum of the energy spectrum of atoms trans-

mission sputtered from the VV_2 sample is shifted toward higher energies.

The crystallite transparency (at high energies E_0)

increases when passing from VV'_2 to VSi_2 and $Si'Si_2$. When passing from $Si'Si_2$ to VSi_2 , the average cross section for the interaction between a Kr^+ ion and the target atom increases, a significant number of ions gives almost all energy to the crystal atoms. Naturally, this leads to an increase in the total transmission sputtering coefficient. Although the number of true Si atoms (located in their lattice sites) does not change in the crystal, the transmission sputtering of such Si atoms increased. Obviously, this is related to the larger Kr^+-V interaction cross section (compared with the Kr^+-Si' one), to the appearance of a larger number of displaced V atoms (compared with the number of dis-

Energy of bombarding ions E_0 , eV	Ion transmission transmission coefficient R_{\downarrow}		
	VV ₂	Si'Si ₂	VSi ₂
100	0.078	0.555	0.406
150	0.462	0.906	0.749
200	0.625	0.975	0.858
300	0.776	0.999	0.952
500	0.887	1.000	0.998

Table 1. Coefficient of Kr^+ -ion transmission through ultrathin VV'_2 , Si'Si₂, and VSi₂ films as a function of the energy E_0



Fig. 3. Dependences of the transmission sputtering yields of atoms from vanadium (solid lines) and silicon (dashed lines) sites on the initial ion energy E_0 under bombardment of the (0001) faces of the VSi₂, VV₂, and Si'Si₂ crystals by Kr⁺ ions. The sputtering yields are normalized to the stoichiometric ratio for the sample not subjected to ion bombardment (1 : 2).

placed Si' atoms), and to subsequent energy redistribution in the collision cascade.

Because the bombarded sample had finite dimensions, the V and Si concentrations in the atomic layers were different. In the upper layer, the number of V atoms was slightly larger than in the ratio of 1:2 (0.58 instead of 0.5). The second and third layers were depleted with vanadium (each had 0.4 instead of 0.5). On the whole, the VSi₂ sample contained a number of V atoms that was slightly smaller than the norm (0.47 instead of 0.5). Nevertheless, for all three samples, we observed the preferential transmission sputtering of atoms from V sites even in the case where atoms with the same masses and binding energies were located at all sites.

Figure 4 shows the ratios of the coefficients of the transmission sputtering of atoms from V and Si sites as functions of the initial energy E_0 in the case of the bombardment of VSi₂, Si'Si₂, and VV₂ crystals by Kr⁺ ions. At low energies, the difference between the atom interaction cross sections plays a large role. If a sample of virtual vanadium is considered, then the V' atom cannot fit into the Si site. However, during the period of collision cascade propagation and sputtering, it has no time to depart (without additional acceleration) from the block of atoms (the interaction region). It is natural to expect that vanadium must be sputtered transmission from Si sites preferentially. This was observed at low energies E_0 of bombarding ions. However, at energies of $E_0 \ge 200$ eV, the preferential transmission sputtering of V atoms from their own lattice sites was observed. This result agrees with the conclu-



Fig. 4. Dependences of the ratio of the transmission sputtering yields of atoms from vanadium and silicon sites on the initial ion energy E_0 under bombardment of the (0001)

faces of the VSi₂, VV₂, and Si'Si₂ crystals by Kr^+ ions. The ratios are normalized to the stoichiometric ratio for the sample not subjected to ion bombardment (1 : 2).

sions in [4]. In this case, it is necessary to note that we had qualitative and quantitative (to a significant extent) correspondence between the ratios of atoms

sputtered from V and Si sites for all three VV₂', VSi₂, and Si'Si₂ samples; it is especially pronounced at energies E_0 in the range from 1 to 10 keV.

It is likely that this is related to the peculiarities of collision-cascade propagation in single crystals with complex structures (to the C40-type of the lattice in our case) and to the difference between the momentum-transfer processes in the V and Si single-crystal sublattices. We can conclude from this that the spatial structure of the target plays the determinative role in the selective sputtering of single crystals with the C40 lattice at energies of $E_0 = 1-10$ keV at least as compared with the role of the target-atom masses. The type of the crystal lattice depends on the interaction of atoms in the crystal. Therefore, it is reasonable to seek the dependence of the selective sputtering on the potential of interaction of atoms between one another (the binding energy of atoms, which is contained in theoretical formulas for estimating selective sputtering, also depends on the atom interaction potential).

CONCLUSIONS

In this paper, we have studied the ion transmission coefficients and the transmission sputtering yields of component atoms under the Kr⁺-ion bombardment of VSi₂, VV₂, and Si'Si₂ crystallites.

We have discovered and studied the peculiarities of the transmission transmission coefficient of Kr⁺ ions as functions of the initial ion energy and the target "density". As a rule, when passing to a "denser" VV_2 target, a decrease in the ion transmission coefficient was observed.

Our calculations showed that, for not very small energies of bombarding ions, the preferential transmission sputtering of atoms from vanadium sites was observed. This can be related to the nonidentity of vanadium sites with respect to (transmission) sputtering. The mechanism for the sputtering of atoms from vanadium sites can differ from that from silicon sites because of the complex structure of the VSi₂ single crystal (of the C40 type) and different mechanisms for momentum propagation through the V and Si sublattices in VSi₂.

It is obvious that this effect of VSi₂ site nonidentity with respect to sputtering must be taken into account when analyzing the elemental composition of multicomponent single crystals by means of secondary-ion mass spectrometry (SIMS) and secondary neutral mass spectrometry (SNMS).

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