Change in ion sputtering coefficients of targets due to cross-dusting during simultaneous operation of two sputters

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Abstract. The influence of cross-dusting of silicon and metal targets simultaneously sputtered by argon ions on partial sputtering coefficients of targets materials components is studied to take this effect into account in the technology of gradient optical coating deposition. The effect has been numerically simulated as series of cascades of binary collisions of bombarding ions and recoil atoms with target atoms. Addition of metal atoms (Ti, V, Zr, Hf, Nb and Ta) to the Si target enhances the partial sputtering of Si atoms but addition of Si atoms to the metal targets decreases the partial sputtering of metals atoms. Also the coefficients of reflection (back scattering) of the bombarding argon ions from the targets with different additives have been calculated. It is possible that enhanced sputtering of Si atoms from the coatings with a “heavy” component by reflected neutralized ions would lead to dusting the “heavy” target by silicon.

1. Introduction

Ion sputtering technology for applying thin layers is used for manufacturing film systems for various purposes, including optical products and metamaterials. At the same time, gradient optical film systems with a smooth change of the refractive index n in thickness and diameter [1, 2] are gaining importance. Such systems make it possible to create optical devices with improved spectral characteristics and resistance to laser, climatic and mechanical factors. To obtain intermediate values of n, the layers are applied from mixtures of at least two initial substances, one of which has a small value of n (this is most often SiO₂), and the second one has a large value (oxides of d-elements of IV and V groups of the periodic table, i.e. metals Ti, Zr, Hf, Nb and Ta). Recently, for the deposition of gradient coatings, ion sputtering of pure metals has been used, and the oxidation of condensate on the substrate is carried out in a separate part of the installation. To implement this technology, the simultaneous sputtering of two adjacent metal targets is often used [1, 2] that simplifies the design of the installations, but the sputtered metal of one target hits the surface of the neighboring target (see figure 1, particle fluxes I). The latter occurs due to the scattering of sputtered particles in the gas or when the target surfaces are inclined towards each other. The consequence of this is a change in the emission characteristics of targets, which must be taken into account when developing practical technology.

At one time [3] it was found that the presence of an impurity of a heavy metal (Pt, W, or Nb) in a target made of a lighter material (in particular, from C or Al) leads to a noticeable increase in the
sputtering coefficient of light atoms. This is a consequence of the fact that heavy atoms play the role of back-scattering centers for bombarding ions with the subsequent transfer of moments towards the surface to light atoms of the target. This also applies to recoil atoms. Similar effects take place on a heavy metal substrate during ion-assisted deposition of coatings [3]. It can also be assumed that this is the case in the above-described technology for deposition of gradient coatings.

Figure 1. Scheme of particle transfer in a sputtering device with two targets T1 and T2, S is a substrate with a deposited coating of sputtered materials, I, II are streams of particles sputtered and reflected from the targets, II is also possible streams of particles, which sputtered from the substrate by the high-energy atoms (the former ions) reflected from the targets.

Taking these circumstances into account, this work analyzes the change in the ion sputtering coefficients of two targets, from silicon and heavy metals, which are simultaneously used to obtain gradient optical coatings. It is also advisable to evaluate the possible role of high-energy atoms of the working gas, reflected (backscattered) from the surface of the heavy target and bombarding the surface of the substrate with an energy sufficient to sputter the low-refractive (“light”) component of the coating. The latter could lead to contamination of the “heavy” target with silicon and a change of sputtering coefficient of heavy metal (see figure 1, streams II).

2. Simulation procedure
The work was carried out by calculation using the widely tested TRIM (TRansport of Ions in Matter) program [4]. The calculations are based on the Monte Carlo method, a method for analyzing random walks of high-energy particles (bombarding argon ions and recoil target atoms) in the target body in the approximation of binary collisions of particles with target atoms. The trajectory of each high-energy particle is traced until it loses its energy. It is assumed that the targets have an amorphous microstructure with a random arrangement of atoms.

For calculations of sputtering, the sublimation energy is used as a parameter, which determines the possibility of emission of surface atoms from the target. For mono-component targets from one chemical element, the sublimation energies are standard reference data. In the case of two-components targets, TRIM usually uses the half-sum of the sublimation energies for the individual components. In our work, we also used the weighted average values of sublimation energy determined by the formula \( E_1C_1 + E_2C_2 \), where \( E_1 \) and \( E_2 \) are the sublimation energies of pure components (chemical elements from single-element targets), \( C_1 \) and \( C_2 \) are the relative concentrations of components in two-components targets (\( C_1 + C_2 = 1 \)). Comparison of the results of calculations with different approximations for the sublimation energy values for two-components targets showed their discrepancy is of the order of 10\%, and the use of half-sums for the purposes of this work can be considered as quite acceptable.

3. Results and discussion
Figure 2 shows the characteristic trajectories of the bombarding particles and recoil atoms within the target bodies in vicinity of the bombarded surface and the average penetration depth of Ar ions into the targets. Table 1 shows the calculated normalized partial sputtering coefficients \( S \) for two-components targets. Here \( C_1 \) refers to the concentration of Si. The normalization of the \( S \) coefficients was carried out by dividing the partial sputtering coefficients of individual elements by the sputtering coefficient of pure silicon at its concentration \( C_1 \geq 0.9 \) or by the sputtering coefficient of the corresponding pure metal at \( C_1 \leq 0.1 \). Table 1 also shows (after the slash) the calculated values of the reflection coefficients \( R \) from the target of ions (as neutralized Ar atoms) that bombard the coating on the substrate.
Average penetration depth of Ar ions into the targets:
Si target – 27 Å,
Ti target – 23 Å,
Hf target – 22 Å,
Si (90%)+Hf (10%) – 25 Å,
Si (10%)+Hf (90%) – 22 Å,
Hf (100%) – 22 Å.

**Figure 2.** Projections of the trajectories of recoil atoms (top) and Ar ions (bottom) during ion bombardment of Si target with Ti and Hf additives and metal targets with Si additive. The ion energy is 1 keV, the point of incidence of ions onto the targets is at the left and the middle of the height of the figures.

Figure 2 shows that Ar ions penetrate deeply into the Si target. Moreover, in pure Si, the role of collisions cascades with the participation of recoil atoms is very important, and ions play only the role of initiators of the cascades. But even a small additions of metals (especially heavy ones) scatter ions back to the surface, which directs the cascades also towards the surface, and significantly increases the Si sputtering coefficient (see table 1). In turn, the addition of Si to metals targets decrease the metal sputtering coefficient. It can be said that here the Si atoms are, as it were, obstacle in the path of both ions and recoil atoms.

From table 1 one sees that, when a “heavy” target is sputtered, a significant amount (> 20 %) of high-energy Ar atoms (they are the neutralized former ions) are reflected from the target towards the substrate. They are capable to sputter silicon from the coating and thus to contaminate the target surface. This process can occur even in the absence of atom transfer between the targets.

4. Conclusions
From the data obtained, one may conclude that the process of deposition of gradient coatings from low-refractive and high-refractive materials during simultaneous ion sputtering of targets from silicon and heavy metals should be considered within the framework of cross-transfer of silicon and metal atoms between the targets. Even small additions of heavy metals to the Si target significantly enhances the Si sputtering coefficient due to backscattering ions to the bombarded surface that directs the cascades collisions also towards the surface. The addition of silicon to metals targets decrease the
metal sputtering coefficient. Also, it should be taken into account the possible transfer of Si atoms sputtered by reflected Ar ions from the substrate to the “heavy” target and additional contamination of the target surface.

Table 1. Normalized partial sputtering coefficients of target components $S$ and reflection coefficients of argon ions $R$ for two-components targets containing silicon and metals.

| Si concentration ($C_1$) | Target metal component | Partial sputtering coefficients ($S$) of silicon/reflection coefficients ($R$) of argon ions | Partial sputtering coefficients ($S$) of metals/reflection coefficients ($R$) of argon ions |
|--------------------------|------------------------|---------------------------------------------------------------------------------|---------------------------------------------------------------------------------|
| 0.97                     | Ti                     | 1.06/0                       | 1.07/0                       |
|                          | V                      | 1.11/0                       | 1.12/0                       |
|                          | Zr                     | 1.175/0                      | 1.123/0                      |
|                          | Nb                     | 1.437/0                      | 1.39/0                       |
|                          | Hf                     | 1.616/0.01                  | 1.39/0                       |
|                          | Ta                     | 1.39/0                       | 1.39/0                       |
| 0.95                     | Ti                     | 1.09/0                       | 1.09/0                       |
|                          | V                      | 1.13/0                       | 1.13/0                       |
|                          | Zr                     | 1.381/0                      | 1.332/0                      |
|                          | Nb                     | 1.795/0.01                  | 1.616/0.01                  |
|                          | Hf                     | 1.39/0                       | 1.39/0                       |
|                          | Ta                     | 1.39/0                       | 1.39/0                       |
| 0.93                     | Ti                     | 1.12/0                       | 1.12/0                       |
|                          | V                      | 1.152/0                      | 1.152/0                      |
|                          | Zr                     | 1.534/0                      | 1.534/0                      |
|                          | Nb                     | 2.019/0.01                  | 1.937/0.01                  |
|                          | Hf                     | 1.39/0                       | 1.39/0                       |
|                          | Ta                     | 1.39/0                       | 1.39/0                       |
| 0.90                     | Ti                     | 1.39/0                       | 1.39/0                       |
|                          | V                      | 1.351/0                      | 1.351/0                      |
|                          | Zr                     | 1.784/0                      | 1.735/0                      |
|                          | Nb                     | 2.907/0.01                  | 2.627/0.02                  |
|                          | Hf                     | 1.39/0                       | 1.39/0                       |
|                          | Ta                     | 1.39/0                       | 1.39/0                       |

Obviously, the analogous effects might occur in sputtering systems with combined targets consisting of separated parts, which made from heavy and light metals.

The data obtained also indicate the urgent need for the introduction of continuous optical monitoring of the deposited condensate on the substrate and adaptive control of the sputtering process to obtain the specified parameters of the optical gradient coating.

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