Mutual mass transfer between the target and the substrate in the process of ion sputtering. Flat and cylindrical target

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Abstract. The ion sputtering process for the deposition of coatings from alternating layers of light and heavy metals are considered, taking into account the mutual mass transfer from the target to the substrate and from the substrate to the target as the result of resputtering the coating matter by reflected fast neutral gas atoms (former primary bombarding ions, backscattered from the target towards the substrate). The resputtering effect is more important in the case of deposition of a heavy metal of Ta type on layer from a light matter of Si type since it forms the intermediate layer of mixing composition “Ta+Si” between layers of pure metals.

1. Introduction

The technology of ion sputtering of solid materials (targets) is widely used for the deposition of thin layers of different materials in manufacturing film systems for various purposes, including for optical, electronic and mechanical products [1–3]. When analysing processes in ion sputtering systems, as a rule, two sorts of atomic particles ejected by targets under ion bombardment are taken into account – sputtered target particles and fast reflected atoms (FRA) of the working gas, which are the neutralised former primary ions [4, 5]. Both kinds of the particles fall on the substrate, thereby ensuring the mass transfer of the target material with the formation of a coating and the implantation of fast reflected gas atoms into the coating. The same particles provide energy transfer to the substrate, too. Earlier, it was shown that at low gas pressures, these particles are able to resputter the coating material back towards the target and this effect has been termed as “the intrinsic resputtering” [4]. It should be distinguished from the effects of resputtering substrates in processes with ion bombardment of the substrates by positive or negative ions of the gas or target material. It is interesting to explore the intrinsic resputtering in the case of manufacturing multilayer coatings with alternating layers from light and heavy atoms in the time periods when the substrate with the external layer from light atoms is faced to the heavy target. In such periods a part of light atoms will be resputtered back onto the heavy target and then again the light atoms will be ejected towards the substrate but together with heavy target
atoms. In results, such behaviour may lead to formation of an intermediate layer from light and heavy atoms between pure layers. The self-resputtering effect of the coating by the sputtered target atoms is much weaker than the resputtering effect by FRA because of lower energy of the sputtered target atoms [4]. Therefore, we will consider the mutual mass transfer between the target and the substrate only due to the intrinsic resputtering by FRA, i.e. fast Ar⁺, in relation to the technology of deposition of coatings obtained by alternate sputtering of silicon (Si) and tantalum (Ta) or titanium (Ti) with argon Ar⁺ ions, in particular, for optical products. Such coatings often consist either of alternating layers of materials with low and high refractive index, or of a layer of variable gradient composition [2].

2. Scheme of analysed sputtering process

Figure 1 shows the schemes of particle transfer in sputtering devices with flat and cylindrical targets. Primary gas ions, accelerated in the near-cathode space charge layer of an anomalous glow discharge (for example, of magnetron type) or injected by an ion gun, bombard a target, sputtering its surface. Sputtered particles, possessing energies of the order of several units or tens of electron-volts, fly to the substrate, forming a coating. A part of the primary ions, as a result of “wandering,” in the subsurface layer of the target, comes out to the target surface and also flies (that seems as ion reflection) towards the substrate in the form of a flux of fast neutral atoms. These atoms may have energies of tens of percents of the value of the energy of primary ions [4, 5]. In certain cases, the energy of FRA is sufficient to sputter the coated surface of the substrate [4]. At low gas pressures in the target-substrate gap, which are typical for magnetron or ion beam sputtering, the probability of collisions of FRA with thermal gas molecules and the loss of FRA energy due to the collisions are not notable; therefore, in the first approximation, the energy losses of fast gas atoms can be neglected.

Figure 1. Schemes of particle transfer in sputtering devices with flat (a) and cylindrical (b) targets: T is target, Sub is coated substrate, “⁺” is for primary ions, “S” is for sputtered target particles, “0” is for fast neutral gas atoms (backscattered or reflected former primary ions), “Rs” is for resputtered coating particles.

The reflection coefficients of FRA and their energies increase with increasing the angle of incidence of ions onto the target surface [5]. From this, it follows that in the case of cylindrical target, the resputtering effect should manifest itself to a greater extent. The distribution of reflected gas atoms over the angles of departure obeys a law close to cosine one, so most of the FRA bombard the substrate along inclined paths, and this, as is known, increases the sputtering coefficient. So, we can suppose that the cylindrical geometry may promote the resputtering effect with participation of FRA.

3. Resputter simulation procedure and results

The numerical simulation of the mutual mass transfer between the target and the substrate have been performed by calculation with using the TRIM program [6] to determine the characteristics of the FRA flux by the Monte Carlo method and the table values of the ion sputtering coefficients of the matter on the substrate surface obtained with Sigmund’s theory of ion sputtering [3]. The calculations were made for the plane-parallel sputtering system with the assumption of normal trajectories of the primary ions and FRA. For the simulation, Si, Ta, and Ti have been chosen as target and coating materials. These materials are often used for multilayer film optical systems. Silicon represents the lightest matter for one sort of coating layers with low refraction index. The metals (Ta and Ti)
represent the heavy matter for the other sort of coating layers with high refraction index. Ta was chosen as a very heavy metal. Ti was chosen as a non-very heavy metal.

Figures 2 and 3 show the TRIM histograms of the distribution of energy for FRA of Ar\(^0\) type from Ta and Ti targets, accordingly, when the targets are bombarded by Ar\(^+\) ions with energy \(E_0\) from 0.3 to 2.0 keV. The histograms show the quantity \(\Delta N_a\) of fast Ar\(^0\) atoms with energy about \(E_a\) in partial energy bands, width of which are 1/20 of \(E_0\) (the number of the partial energy bands is 20). The calculations were performed for \(N_0 = 10^4\) ions bombarding the target. These histograms, in fact, represent the entire flux of reflected Ar\(^0\) atoms to the substrate in the form of 20 partial monoenergetic fluxes of atoms. The integral reflection coefficient of Ar\(^0\) atoms is \(R = N_a/N_0\), where \(N_a\) is the entire number of reflected Ar\(^0\) atoms or the sum of \(\Delta N_a\).

**Figure 2.** Histograms of the energy \(E_a\) distribution of fast Ar\(^0\) atoms reflected from the Ta target for different energies \(E_0\) of Ar\(^+\) ions.

**Figure 3.** Histograms of the energy \(E_a\) distribution of fast Ar\(^0\) atoms reflected from the Ti target for different energies \(E_0\) of Ar\(^+\) ions.

As one can see, the form of the energy distributions weakly depends on the value of \(E_0\). The most of the atoms reflected from Ta target have energy in the range from 0.05\(E_0\) to 0.5\(E_0\). The most of the atoms reflected from Ti target have energy in the range up to 0.1\(E_0\). Besides, the entire number of FAR is much lower in the case of Ti. There are practically no fast Ar\(^0\) atoms reflected from Si target.

Using the data on the sputtering coefficients of Si, Ta, and Ti for each of the partial energy bands from [3] and the quantity \(\Delta N_a\) of reflected fast Ar\(^0\) atoms for the bands, the integral resputtering coefficients of Si, Ta, and Ti layers (coatings) upon bombardment by the entire flux of fast Ar\(^0\) atoms were calculated. The calculation results are given in table 1 and table 2, where the values of the parameters are determined by the following formulas:

\[
S = N_T/N_0, \quad S_{Sub} = N_{Sub}/N_0, \quad N_{Sub}/N_0 = N_{Sub}/N_a = S_{Sub}R,
\]

where \(S\) is the sputtering coefficient of the target matter by Ar\(^+\) ions, \(N_T\) is the number of sputtered target atoms, \(S_{Sub}\) is the integral resputtering coefficients of Si, Ta, and Ti layers by reflected Ar\(^0\) atoms, \(N_{Sub}\) is the number of atoms of Si, Ta, and Ti layers resputtered from the substrate, \(N_{Sub}/N_0\) \((S_{Sub}R)\) is the effective coefficient of resputtering of the coating matter (the ratio of the number of resputtered coating atoms to the number of Ar\(^+\) ions bombarding the target).

4. Discussion

It can be seen that the Ta target effectively reflects fast Ar\(^0\) atoms. These atoms, despite the fact that their energy is less than the energy of the primary Ar\(^+\) ions, notably resputter the Ta and Si layers on the substrate. The integral resputtering coefficients \((i.e. S_{Sub})\) of Ta and Si layers are \(\sim 30-50\%\) of the value of the ion sputtering coefficient \(S\) of Ta target in the used range of \(E_0\). Also note, the resputtering coefficients \(S_{Sub}\) for the Ta and Si layers are close each to other since their ion sputtering coefficients \(S\) are close each to other, too [3].
Table 1. Parameters of the resputtering of Ta and Si layers by fast Ar\textsuperscript{0} atoms reflected from Ta target.

| \(E_0, \text{keV} \) | \(I_{\text{Ar}/\text{Ta}} \rightarrow \text{Ar}/\text{Ta} \) | \(I_{\text{Ar}/\text{Ta}} \rightarrow \text{Ar}/\text{Si} \) |
|---|---|---|
| 0.3 | 0.5 | 1.0 | 2.0 | 0.3 | 0.5 | 1.0 | 2.0 |
| \(S \) | 0.55 | 0.80 | 1.21 | 1.73 | 0.55 | 0.80 | 1.21 | 1.73 |
| \(R \) | 0.30 | 0.28 | 0.27 | 0.25 | 0.30 | 0.28 | 0.27 | 0.25 |
| \(S_{\text{Sub}} \) | 0.15 | 0.27 | 0.53 | 0.84 | 0.1 | 0.25 | 0.53 | 0.76 |
| \(S_{\text{Sub}}R \) | 0.04 | 0.07 | 0.14 | 0.21 | 0.03 | 0.07 | 0.14 | 0.19 |

The resputtering leads to decreasing the deposition rate of Ta on the Ta layer. Accordingly, the mass transfer of resputtered Ta atoms back to Ta target leads to decreasing the resultant sputtering rate of Ta target \(S_{\text{res}} \approx S - S_{\text{Sub}}R \). So, for the Ta target and Ta coating, the value of \(S_{\text{res}} \) is less by about 10% relatively the ion sputtering coefficient \(S\).

In the case of sputtering Ta target for deposition of Ta atoms onto Si layer on the substrate, silicon, resputtered from the substrate surface, contaminates Ta target. Then both Ta and Si are sputtered simultaneously by Ar\textsuperscript{+} ions from the target until tantalum totally covers the Si layer on the substrate. Thus, the resputtering effect in relation to the silicon on the substrate does not allow forming the sharp boundary between the Si and Ta layers on the substrate. Obviously, the higher the resputtering coefficient for Si the thicker the intermediate layer of mixing composition is between Si and Ta layers.

One may suppose the following mechanism of intermediate layer formation between Si and Ta layers on the substrate. At the beginning of Ta target ion sputtering, the Si layer surface on the substrate is practically free from Ta atoms; therefore only silicon is resputtered by FRA and deposited onto Ta target. Then under ion bombardment Si atoms together with Ta target atoms return to the substrate, from which they again partially resputtered by FRA back to the target. So the mutual mass transfer between target and substrate occurs. It is known the so-called sputter yield amplification effect with the preferential sputtering of light atoms from the mix with heavy atoms [7]. According with this effect a small concentration of a heavy metal impurity promotes sputtering light atoms due to back scattering of the bombarding species by heavy atoms. Factually, such situation occurs during deposition of Ta atoms onto Si layer on the substrate. The data on calculations of the details of this effect in relation to Si sputtering may be found in [8]. Here it was also shown the presence of Si atoms on heavy metal targets, including Ta, decreases sputtering rate of the heavy metal that is applicable to our case. At last, because ion sputtering coefficient \(S\) for Ta target is bigger than \(S_{\text{Sub}}\) and \(S_{\text{Sub}}R\) for Si layer on the substrate, tantalum totally covers \((i.e. \text{buries})\) the Si layer on the substrate with formation of the intermediate layer "Ta+Si" of limited thickness.

In the case of sputtering Ti target the values of reflection coefficient \(R\) is much lower. Also, energy of fast reflected Ar\textsuperscript{0} atoms is much lower than in the case of the heavier Ta target (compare figure 2 and figure 3). This may be explained by weak ability of Ti atoms to backscatter the bombarding Ar\textsuperscript{+} ions due to the feature mechanical law at binary elastic collision between atomic particles of close
mass [9] and deeper penetration of primary ions into Ti target. These circumstances lead to practically zero or the very small resputtering effects. The like situation with practically no resputtering effect occurs when Si target is sputtered with Si deposition on Ta or Ti layer.

Since the analysis of the mutual mass transfer during ion sputtering is based on the use the sputtering coefficients of the target by gas ions and the substrate by fast gas atoms reflected from the target, the question is about choosing the correct values of the sputtering coefficients. The theoretical values of the coefficients $S$, $S_{Sub}$ and $S_{Sub}R$ for clean surfaces have been used; however, in practice, surfaces may be contaminated, primarily with oxides, which reduces the sputtering rate. A favourable factor is the permanent renewal, due to the sputtering, of the target and coating surfaces. Taking these circumstances into account, the obtained data should be considered as the upper estimate of the effect of mutual mass transfer. Using the experimental data on ion sputtering [3], the estimate of the effect of mutual mass transfer is proposed to halve for the case of possible contamination of the surfaces.

5. Conclusions

The mutual mass transfer from the target to the substrate and back to the target as the result of resputtering the substrate by the fast reflected neutral gas atoms (former primary bombarding ions, backscattered from the target) has been estimated. The resputtering is more important in the case of sputtering and deposition of heavy metal of Ta type on layer of light matter of Si type since it forms the intermediate layer of mixing composition “Ta+Si” between layers of pure metals.

Thus, the sharp boundary between the Si and Ta layers will not form on the substrate, which will undoubtedly affect the optical characteristics of the multilayer coating. In the case of a gradient coating, this effect will obviously be less critical.

Calculations for Ti and Si targets have shown that they reflect few fast Ar$^0$ atoms and, accordingly, their role in the resputtering of coatings is not so great.

The cylindrical target is likely to contribute to the resputtering effect by fast reflected gas atoms due to their oblique trajectories, but this requires additional research.

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