CALCULATION OF CONCENTRATION PROFILES OBTAINED AT THIN FILM DEPOSITION USING LOW-ENERGY ION BEAMS

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In the framework of the diffusion approximation, a mathematical model of thin film deposition which takes into account mass transport under the action of an ion beam has been developed. The bulk profiles of component concentrations at the Ni-film deposition onto a copper substrate have been calculated for ion energies of 100, 200, and 400 eV and an ion flux of 10^{15} , 10^{16} , and 10^{17} cm⁻²s⁻¹. The model parameters of the deposition process have been estimated using the SUSPRE and SRIM programs, and the corresponding profiles of component concentrations have been compared.

1. Introduction

Many technologies for fabricating the electronic devices use the treatment of materials with ion beams, in particular, film sputtering with low-energy ions. A plenty of theoretical and experimental researches dealing with this technological direction is in progress [1-3]. If an ion beam is used to deposit a film onto a substrate, there emerge the transfer processes in nearsurface regions of the substrate, in particular, at the film-substrate interface. Therefore, the development of a mathematical model for sputtering, taking the nearsurface transfer of a substance into account, is of both technical and scientific interest. The most common approach in simulating similar systems is the diffusion approximation [4–9]. In those and other works, the sputtering of films making use of low-energy (up to 500 eV ion beams at room temperature has been analyzed; as the main processes of substance transfer in near-surface layers, ionic mixing [10–15], preferential sputtering [16, 17], and deposition of the substance onto a surface [18] have been considered, the description of which in the diffusion approximation enables one to obtain realistic results. Such an approach allows one, making use of a computer program based on proposed model ideas, to construct the level-by-level concentration profiles of the deposited material, as well as their dependences on the deposition time, with regard for various parameters of the applied ion beam.

2. Description of the Model

The model proposed for simulating the process of film sputtering supposes that a substrate with a perfect crystalline lattice is subjected to the bombardment with low-energy ions. In so doing, the surface of the specimen is being sputtered as well, and some portion of ions is being simultaneously deposited on the bombarded surface. The process runs at room temperature. The concentration of lattice sites in the substrate is admitted constant, and atoms are supposed to be located at the crystal lattice sites only. The model is calculated in the case where the lattice constants of the film and substrate materials are close by value. The ratios of the film and substrate material atom concentrations to the concentration of lattice sites - hereafter, taken as dimensionless concentrations C_1 and C_2 , respectively – were normalized by the relation

$$C_1(x,t) + C_2(x,t) = 1, (1)$$

where x is the the coordinate reckoned from the instant position of the specimen surface towards the target depth, and t is the bombardment time. In what follows, the arguments of the functions will be omitted.

To describe the evolution of the concentration profiles of the deposited material and the sputtered substrate, the following system of equations was used:

$$\frac{\partial C_i}{\partial t} = D_i \frac{\partial^2 C_i}{\partial x^2} + V \frac{\partial C_i}{\partial x} + \frac{\partial}{\partial x} \left(I_c C_i \right), \tag{2}$$

where the subscript *i* distinguishes between film (i = 1)and substrate (i = 2) atoms, D_i are the coefficients of ion mixing of the film and substrate atoms, *V* is the motion speed of the surface exposed to the bombardment, and I_c is the collective flux [19, 20]. In order to take the

ISSN 0503-1265. Ukr. J. Phys. 2008. V. 53, N 2

simultaneous processes of sputtering and deposition into account, the following expression for the speed of surface motion was offered:

$$V(t) = \frac{I}{N_0} \left(-\alpha_{11}C_1 - \alpha_{12}C_2 + Y_{11}C_1 + Y_{12}C_2 \right), \qquad (3)$$

where α_{11} and α_{12} are the coefficients of film atom sticking to the substrate and to one another, respectively; Y_{11} and Y_{12} are the sputtering coefficients of film and substrate atoms, respectively; I is the ion flux density; and N_0 is the concentration of lattice sites.

The third term in Eq. (2) ensures the validity of the normalizing relation (1) throughout the whole specimen. In the framework of the given model, the collective flux allows the total concentration of the substance to be maintained at a constant level; ultimately, it is caused by the target relaxation in the cascades of atom collisions at the bombardment. For the description of the collective flux, the following relation was derived from Eqs. (1)–(3):

$$I_c = (D_1 - D_2) \frac{\partial C_1}{\partial x}.$$
(4)

The substrate surface was supposed perfectly clean at the initial moment: $C_1(x,0) = 0$ and $C_2(x,0) = 1$.

The boundary conditions on the surface for system (2) were written down as follows:

$$\begin{aligned} \frac{\partial C_1(0,t)}{\partial t} &= \left[\frac{I}{N_0} \left(\alpha_{11} C_1(0,t) + \alpha_{12} C_2(0,t) - \right. \right. \\ \left. -Y_{11} C_1(0,t) \right) + D_1 \frac{\partial C_1(0,t)}{\partial x} + \right. \\ \left. + V C_1(\Delta x,t) + C_1(0,t) I_c \right] \frac{1}{\Delta x}, \\ \frac{\partial C_2(0,t)}{\partial t} &= \left[-\frac{I}{N_0} Y_{12} C_2(0,t) + D_2 \frac{\partial C_2(0,t)}{\partial x} + \right. \\ \left. + V C_2(\Delta x,t) + C_2(0,t) I_c \right] \frac{1}{\Delta x}. \end{aligned}$$
(5)

In Eqs. (5), the terms including the coefficients of sticking and sputtring describe the fluxes of atoms which come to the surface and go away from it; the terms including the mixing coefficients and the speed of surface motion describe the diffusion (owing to the energy carried by ions) and convective fluxes of the substance, respectively. The terms of Eqs. (5) including the collective flux implicitly make allowance for the influence of substance relaxation. The quantity Δx is the minimal coordinate step used in the numerical scheme, and $C_i(\Delta x, t)$ is the concentration at the depth Δx from the surface calculated at the computation time-layer t.

For the description of the substance concentration evolution to be correct, two cases should be considered at the distant boundary of the mixing region in the specimen bulk: with either the dominant deposition or the dominant sputtering of the substance at the bombarded surface. In the former case, the system of equations for mass transfer at the distant boundary (x = L) was written down in the following form:

$$\frac{\partial C_i(L,t)}{\partial t} = \left[-D_i \frac{\partial C_i(L,t)}{\partial x} - VC_i(L - \Delta x, t) + + VC_i(L,t) + C_i(L,t)I_c \right] \frac{1}{\Delta x},$$
(6)

where $C_i(L, t)$ are the values of relative concentrations at the distant boundary of the mixing region, and $C_i(L-\Delta x, t)$ are the values of component concentrations before the distant boundary.

For the prevailing sputtering of the substance at the bombarded surface, the system of equations for mass transfer at the distant boundary was written down in the form

$$\frac{\partial C_i(L,t)}{\partial t} = \left[-D_i \frac{\partial C_i(L,t)}{\partial x} + V C_i(L + \Delta x, t) - -V C_i(L,t) + C_i(L,t) I_c \right] \frac{1}{\Delta x},$$
(7)

where $C_i(L + \Delta x, t)$ are the values of concentrations beyond the mixing region, i.e. in that part of the substrate, where there is still no ion mixing at the present time moment. The profiles of substance concentration in the region deeper than the mixing one were calculated making use of system (2), where the diffusion component and the collective flux were omitted.

3. External Parameters of the Model

The external parameters of the given model include the sticking and sputtering coefficients, the coefficients of mixing, and the depth of mixing region, as well as the density of ion flux and the energy of particles that bombard the substrate. Nickel and copper were selected as materials of the film and the substrate, respectively.

ISSN 0503-1265. Ukr. J. Phys. 2008. V. 53, N 2

Some coefficients of the model were calculated with the help of the known programs SUSPRE [21] and SRIM-2003 [22]. The former uses an analytical approach, while the latter is based on the Monte-Carlo method. The sticking coefficients were evaluated making use of the formula

$$\alpha = 1 - \frac{1}{2} \operatorname{erfc}\left(\frac{x_0}{\sqrt{2}\sigma}\right),\tag{8}$$

where the values for the parameters x_0 – the coordinate, where the concentration profile reaches its maximal value – and σ – the standard deviation of the concentration – were obtained making use of the SUSPRE program. The calculated values for the sticking coefficient at various energies of bombarding particles are listed in Table 1. One can see that the variation of the energy of particles that bombard the substrate in the investigated energy range affects the numerical values of the sticking coefficient insignificantly, and they are close to unity.

The values of the sputtering coefficient were calculated making use of both programs indicated above. For Ni atoms on the surface of a Cu substrate, the corresponding results of calculations differ from each other by approximately a factor of two, whereas the values given for the sputtering coefficient by both programs differ slightly in the case of clean copper. The results of calculations of the sputtering coefficient are presented in Table 2.

The values of the mixing coefficient were calculated by the formula [23]

$$D = \frac{0.07}{N_0} I F_D \ \gamma \frac{\langle r^2 \rangle}{E_d},\tag{9}$$

where E_d is the energy needed for an atom to move from its lattice site by the distance r (about 4 Å) (as usual, it does not exceed 15 eV for the majority of materials [24]),

T a b l e 1. Sticking coefficients

enij	Energy of bombarding particle, eV					
	100	200	400			
α_{11}	0.916	0.919	0.911			
α_{12}	0.91	0.905	0.905			

T a b l e 2. Sputtering coefficients calculated with the SRIM and SUSPRE programs

Energy of bombarding	100		200		400	
particle, eV						
Program	SRIM	SUS	SRIM	SUS	SRIM	SUS
Y_{11}	0,315	0.21	0.686	0.54	1.32	102
Y_{12}	0.53	0.24	1.04	0.6	1.893	1.14
L', nm	0.4	0.414	0.6	0.556	0.8	0.761

ISSN 0503-1265. Ukr. J. Phys. 2008. V. 53, N 2

 F_D is the function that describes the distribution of energy released at elastic collisions of ions over the target depth,

$$\gamma = \frac{4m_i m_i}{\left(m_i + m_i\right)^2} \tag{10}$$

is the coefficient of energy transfer, and m_i and m_m are the masses of a bombarding ion and a matrix atom, respectively. The distribution function F_D can be approximately estimated – by neglecting inelastic losses – by the formula $F_D \approx E/L$. Here, E is the energy of bombarding particles, and L the depth of mixing region. The calculated values of mixing coefficients are given in Table 3.

The depth of the mixing region was estimated on the basis of the maximal values given by both programs for the distribution of bombarding particles over the depth L', and the values obtained turned out equal to each other to an accuracy of one Angström (Table 2). However, both computer programs were created for the case where the medium is characterized by a random distribution of atoms, and do not take into account that, if the energy is low, the dimensions of the mixing region in a crystalline medium are significantly influenced by the chains of substitutional atoms which arise in collision cascades and are directed from the central region of the cascade towards external layers [25, 26]. The chains enlarge the mixing region by approximately 10^{-7} cm. The values of L, adopted in such a way, are presented in Table 3. The energy E of ions in the sputtering beam was selected equal to 100, 200, and 400 eV. The ion flux density was taken equal to 10^{15} , 10^{16} , and 10^{17} cm⁻²s⁻¹.

4. Results and Their Discussion

On the basis of the model expounded in Sections 2 and 3, a computer program was created, which allows the

T a b l e 3. Mixing coefficients

	8						
	$L \cdot 10^{-8}$, cm						
$I,1/({ m cm}^2{\cdot}{ m s})$	14	16	18				
	E, eV						
	100	200	400				
	$D_1~{ m cm}^2/{ m s}$						
10^{17}	5.84×10^{-15}	1.02×10^{-14}	1.82×10^{-14}				
10^{16}	5.84×10^{-16}	1.02×10^{-15}	1.82×10^{-15}				
10^{15}	5.84×10^{-17}	1.02×10^{-16}	1.82×10^{-16}				
	$D_2~{ m cm}^2/{ m s}$						
10^{17}	6.30×10^{-15}	1.10×10^{-14}	1.96×10^{-14}				
10^{16}	6.30×10^{-16}	1.10×10^{-15}	1.96×10^{-15}				
10 ¹⁵	6.30×10^{-17}	1.10×10^{-16}	1.96×10^{-16}				



Fig. 1. Bulk profiles of the admixture concentration at different time moments for E = 100 eV, $I = 10^{15}$ cm⁻²s⁻¹, $\alpha_1 = 0.916$ atom/ion, $\alpha_2 = 0.91$ atom/ion, $D_1 = 5.84 \times 10^{-17}$ cm²/s, $D_2 = 6.30 \times 10^{-17}$ cm²/s, and (SUSPRE) $Y_1 = 0.21$ atom/ion and $Y_2 = 0.24$ atom/ion or (SRIM) $Y_1 = 0.315$ atom/ion and $Y_2 = 0.53$ atom/ion



concentration profiles in the deposited film to be calculated. In the presented figures, the concentration profiles are plotted for the cases of sputtering coefficients that were calculated by the SRIM (dotted curves) and SUSPRE (solid curves) programs. From Figs. 1–3, one can see that the distinctions in the concentration profiles given by those two programs are small at low intensities of the ion flux; however, they are gradually accumulated with the increase of the sputtering time.

The data obtained testify that the higher is the flux density of bombarding particles (and, accordingly, the speed of surface motion), the larger are the differences



Fig. 3. The same as in Fig. 1, but for $I=10^{17}~{\rm cm}^{-2}{\rm s}^{-1}$

between the corresponding concentration profiles. The dependences of the component concentrations on the depth for various energy values of bombarding particles are depicted in Figs. 1, 4, and 5.

One can see that the behavior of the concentration profile in the film bulk at an energy of 400 eV differs essentially from those obtained at 100 and 200 eV. This fact is associated with the circumstance that, if the ion energy equals 400 eV, the process of surface sputtering prevails over the deposition one. In this case, the film can be created on the surface only owing to ion mixing, so that the transition layer at the interface between the film and the substrate will be very narrow, because, provided that the etching of the surface dominates, the transfer of the admixture does not take place beyond the mixing region. It is worth noting that, in the framework of the given model, the transition layer does not arise in this case, because the mixing coefficients D_i are considered independent of the depth over the whole mixing region L.

The concentration profiles for the energies of bombarding particles of 100 and 200 eV (Figs. 1 and 4, respectively) are similar to each other at the initial stage of bombardment, provided that all other parameters are equal. Divergences become accumulated as the time grows, owing to a shorter time of film deposition at 100 eV. In addition, at 200 eV, the broadening of the profile is larger, and the depth of the transition layer (beyond the mixing region) is smaller. The larger broadening is associated with higher values of the mixing coefficient at 200 eV (see Table 3). The narrowing of the transition region at this energy, as well as the longer time needed for the emergency of similar profiles, is caused by



Fig. 4. The same as in Fig. 1, but for E = 200 eV, $I = 10^{15}$ cm⁻²s⁻¹, $\alpha_1 = 0.919$ atom/ion, $\alpha_2 = 0.905$ atom/ion, $D_1 = 1.02 \times 10^{-16}$ cm²/s, $D_2 = 1.10 \times 10^{-16}$ cm²/s, and (SUSPRE) $Y_1 = 0.54$ atom/ion and $Y_2 = 0.6$ atom/ion or (SRIM) $Y_1 = 0.686$ atom/ion and $Y_2 = 1.04$ atom/ion

a reduction of the rate of film deposition owing to the growth of sputtering coefficients (Table 2).

The difference between bulk concentration profiles for different intensities of the ion flux (Figs. 1-3 for the flux of 10^{15} , 10^{16} , and 10^{17} cm⁻²s⁻¹, respectively), provided that all other factors are equal, stems from a significant modification of the speed of surface motion. It was demonstrated that, if the fluxes of bombarding particles differ by an order of magnitude and the times of bombardment also differ by an order of magnitude, the resulting films are approximately identical by their depths and concentration profiles. For instance, if the time needed for sputtering a film with a required concentration is of the order of tens of seconds for the flux $I = 10^{15}$ cm⁻²s⁻¹, then, for the flux I = 10^{16} cm⁻²s⁻¹, this value will be, accordingly, less by an order of magnitude; and, for $I = 10^{17} \text{ cm}^{-2} \text{s}^{-1}$, the time of film deposition falls within the limits of a one-tenth-second interval, which enables one to speak - in the latter case - about a possibility of pulse deposition.

The variations of mixing coefficients under the action of various parameters can substantially affect the shape of concentration profiles. The calculations have been carried out, which showed that, if the mixing coefficient increases by an order of magnitude, the profiles that correspond to those demonstrated above reveal a much larger broadening, a wider transition layer, and, accordingly, a higher predicted adhesion.





Fig. 5. The same as in Fig. 1, but for E = 400 eV, $I = 10^{15}$ cm⁻²s⁻¹, $\alpha_1 = 0.911$ atom/ion, $\alpha_2 = 0.905$ atom/ion, $D_1 = 1.82 \times 10^{-16}$ cm²/s, $D_2 = 1.96 \times 10^{-16}$ cm²/s, and (SUSPRE) $Y_1 = 1.02$ atom/ion and $Y_2 = 1.14$ atom/ion or (SRIM) $Y_1 = 1.32$ atom/ion and $Y_2 = 1.893$ atom/ion

5. Conclusions

The model developed for describing the deposition of a thin film by bombarding a surface with low-energy ions allows one to evaluate the geometrical characteristics of the concentration profile of the deposited material. The concentration profiles for various values of the ion flux density have been calculated. They demonstrate that the growth of the flux of the deposited substance by an order of magnitude gives rise to a similar reduction of the characteristic time of film deposition. In the case of the prevailing deposition, the broadening of the profile for higher energies of bombarding particles was shown to increase in the mixing region and to decrease beyond it. In case of even higher energies, when the surface etching becomes prevailing, the film formation occurs owing to ion mixing, but the film thickness is practically confined at that by the depth of the mixing region. The external parameters of the model were calculated making use of the SRIM and SUSPRE programs. The results of calculations revealed substantial differences in the estimated concentration profiles of the system components, the discrepancies being more considerable for higher energies of bombarding ions.

- 1. Daeil Kim, Appl. Surf. Sci. 253, 2127 (2006).
- Maozhi Li, M.C. Bartelt, and J.W. Evans, Phys. Rev. B 68, 121401 (2003).

- C.-H. Ma, J.-H. Huang, and Haydn Chen, Thin Solid Films 446, 184 (2004).
- P. Varga and E. Taglauer, J. Nucl. Mater. 111/112, 726 (1981).
- 5. P.S. Ho, Surf. Sci. 72, 253 (1978).
- B.V. King and I.S.T. Tsong, Nucl. Instrum. Methods B 7/8, 793 (1985).
- B.V. King, S.G. Puranik, M.A. Sobhan, and R.J. MacDonald, Nucl. Instrum. Methods B 39, 153 (1989).
- A.A. Volokonskaya and S.V. Teplov, Poverkhnost N 12, 31 (1994).
- B.V. King and I.S.T. Tsong, J. Vac. Sci. Technol. A 2, 1443 (1984).
- 10. R.S. Averback, Nucl. Instrum. Methods B 15, 675 (1986).
- G.V. Kornich, L.O. Kornilova, V.Yu. Zhad'ko, and S.V. Teplov, Poverkhnost N 6, 15 (1991).
- 12. W. Xia et al., Nucl. Instrum. Methods B 37/38, 408 (1989).
- B.M. Pain and R.S. Averback, Nucl. Instrum. Methods B 7/8, 666 (1985).
- S. Abedrabbo, D.E. Arafah, O. Gokce, L.S. Wielunski, M. Gharaibeh, O. Celik, and N.M. Ravindra, J. Electron. Mater. 35, 834 (2006).
- A.K. Srivastava, S. Amirthapandian, B.K. Panigrahi, A. Gupta, and R.V. Nandedkar, Nucl. Instrum. Methods B 244, 359 (2006).
- 16. G. Betz, Surf. Sci. 92, 283 (1980).
- 17. S.P. Linnik and V.E. Yurasova, Poverkhnost N 3, 25 (1982).
- 18. N.N. Kudryavtsev, Usp. Fiz. Nauk 163, 75 (1993).
- 19. R. Collins, Radiat. Eff. 98, 1 (1986).

- 20. R. Collins, Nucl. Instrum. Methods B 2, 809 (1984).
- 21. http://www.ee.surrey.ac.uk/ibc/index.php?target=6:35
- 22. http://www.srim.org/
- 23. B.V. King and I.S.T. Tsong, Ultramicroscopy 14, 75 (1984).
- R.P. Webb and I.H. Wilson, in Proceedings of the 2-nd International Conference on Simulation of Semiconductor Devices and Processes (Pineridge Press, Swansea, UK, 1986), p. 249.
- 25. G.V. Kornich, G. Betz, and A.I. Bazhin, Nucl. Instrum. Methods B **152**, 437 (1999).
- A.J.E. Foreman, W.J. Phythian, and C.A. English, Philos. Mag. B 180, 211 (1981).

Received 25.07.07. Translated from Ukrainian by O.I. Voitenko

ОБЧИСЛЕННЯ КОНЦЕНТРАЦІЙНИХ ПРОФІЛІВ ПРИ ОСАДЖЕННІ ПЛІВОК З НИЗЬКОЕНЕРГЕТИЧНОГО ІОННОГО ПУЧКА

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Резюме

На основі дифузійного наближення опису перенесення речовини під дією іонного пучка розроблено математичну модель процесу напилення тонких плівок. Розраховано об'ємні профілі концентрацій компонентів, що формуються при напилюванні плівки нікелю на мідну підкладку для енергій іонів 100, 200 і 400 еВ і щільності іонного потоку $I = 10^{15}$, 10^{16} і 10^{17} 1/(cm² · c). Оцінку модельних параметрів напилення отримано за допомогою програм SUSPRE і SRIM. Проведено порівняння одержаних профілів концентрацій компонентів.