



# Analysis of Stress Development Mechanisms in the Coating/Substrate System

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**Abstract.** The development of nanotechnologies and the general tendency to increase the areas of use of thin coatings with the simultaneous high cost of high-tech materials makes us pay attention to the possibility of predicting the properties of the obtained coatings. This paper presents an analysis of the mechanisms of stress development of thin coatings for a deeper understanding of the possibilities of modeling the stress state of the obtained coatings with subsequent prediction of the required properties. The magnitude and sign of the internal stress in the coatings depend on many factors related to the conditions and method of coating the substrate, as well as the nature of their growth. Thermal stresses make an important contribution to the overall level of stresses in thin coatings, as well as the development of stresses in current-carrying coatings is significantly influenced by electromigration processes. No less significant contribution to the development of stresses in thin coatings can have the effects associated with the influence of the electromagnetic field: the piezoelectric effect, electro- and magnetostriction.

**Keywords:** Nanostructures · Thin coatings · Stress · Substrate · Coating properties

## 1 Introduction

The need for new functional properties in coatings and tendency to use of nanomaterials in our time leads to the emergence of a large number of new compositions of materials. As a consequence of the introduction of increasingly complex technologies for obtaining coatings requires further development of both models of their development and forecasting the properties of the obtained coatings.

The stresses that occur in the coatings can be divided into two large groups [1]. The first is the internal stresses associated with changes in the composition and structure that occur inside the coatings during their application. The second - stresses that are the result of exposure to the coating/substrate system of any external forces and occur after coating on the substrate. In addition, the relaxation processes that occur in the coatings can lead to a change in their stress state, even in the absence of any external influences. However, such a classification allows a deeper understanding of the causes and mechanisms of stress in coatings, as well as to develop methods for managing them.

## 2 Stress of Nonequilibrium in Epitaxial Coatings

The size and sign of internal stress in the coatings depend on many factors associated with the conditions and method of applying coatings on the substrate, as well as the nature of their growth. Thus, with heteroepitaxial growth, when the coating materials and substrates are different, the appearance of internal stresses is mainly caused by the nonequilibrium of their constant crystal lattices. At the initial stage of growth, a continuous heteroepitaxial coating is a biaxially stressed layer coherently linked to the substrate interface. If the parameters of the coating and substrate lattices differ slightly, the pseudomorphic growth of the coating can take a long time [2, 3]. However, since the energy of elastic deformation of the coating  $W$  is proportional to its volume  $V$ :

$$W = \frac{2 \cdot \mu \cdot (1 + \nu)}{1 - \nu} \cdot \varepsilon^2 \cdot V \quad (1)$$

where  $\mu$  – shear module,  $\nu$  – Poisson's ratio of coating,  $\varepsilon$  – flat deformation that occurs under the action stresses of nonequilibrium, the increase in the thickness of the applied layer leads to a rapid increase in  $W$ , resulting in the achievement of a certain critical thickness begins stress relaxation.

Depending on the application conditions and the characteristics of the coating and the substrate, this process may be accompanied by both plastic and elastic deformation of the coating [4].

The mechanism of surface coarsening of coatings depends on the kinetic behavior of atoms on the surface of the growing layer, which is regulated by deformation. At the boundaries of atomic clusters there is an almost complete relaxation of stresses, while in their central part the deformation energy increases significantly. Since in monocrystalline epitaxial coatings there are no internal interface, the main mechanism of mass transfer in them is diffusion on the surface. Atoms move from one place on the surface to another, leading to a change in free energy, which is determined by the gradient of chemical potential along the surface. Thus, there is a movement of growth centers along the surface in places with less free energy, ie to the tops of the protrusions, which causes the transition to the island mechanism of growth [4].

## 3 Internal Stresses in Polycrystalline Coatings

One of the main reasons for the development of internal stresses in polycrystalline thin coatings is the compaction of their structure. The nonequilibrium of polycrystalline coatings is most significant at low temperatures and/or at high rates of application, when low mobility of adatoms on the surface of the substrate leads to the formation of a large number of nuclei and does not allow accelerated growth of the crystallites with energetically favorable orientation.

At the initial stage of formation, when the islands do not touch each other, in the coatings, compressive stresses develop, the magnitude of which can be quite high.

It is currently assumed that the main cause of compressive stresses in the island thin coatings is associated with the action of surface tension forces [7].

The increase in the size of the island during the growth of the coating should lead to a decrease in capillary pressure and, accordingly, to the gradual relaxation of the permanent lattice to the volumetric values. However, after the island reaches a certain critical size, it is rigidly fixed on the substrate, which prevents further changes within the interatomic distances in the plane parallel to the interface of the coating / substrate [8, 9]. Therefore, the long-term growth of the islands contributes to their deformation along a given plane, which according to Hooke's law is equal to

$$\varepsilon = - \frac{2 \cdot s \cdot (1 - \nu)}{E \cdot R}, \quad (2)$$

where  $s$  – is the force of surface tension,  $R$  – the radius of the island (the average curvature of its surface),  $E$  and  $\nu$  – modulus of elasticity and Poisson's ratio. Since for most low-index surfaces of metals and dielectrics  $s > 0$ , then  $\varepsilon < 0$ , that is, compressive stresses develop in the islands.

Also a significant contribution to the development of compressive stresses can be made by the interaction of adatoms with the surface of the substrate and the islands that are formed [10]. This assumption is based on the fact that the interaction of the adatom with the surface of the substrate leads to the formation in its surface layer of a field of elastic deformation, which can be considered as a "power dipole". This field causes the development of compressive stresses in the islands, the growth of which is accompanied by the continuous deposition of a significant number of new adatoms.

As the size of the islands increases, they cover most of the surface of the substrate, and neighboring islands begin to interact with each other. The result of this interaction is the fusion of the islands, which is usually accompanied by the development of tensile stresses in the coatings [11]. The maximum value of these stresses  $\sigma_{\max}$ , significantly depends on the grain size during coalescence  $d_0$  ( $\sigma_{\max} \sim 1/d_0$ ). In particular, at  $d_0 \leq 10$  nm stresses can reach several GPa [12].

The driving force of coalescence is the reduction of the total energy of the coating during the transformation of the two free surfaces of the islands into one grain boundary. The change in energy in this case can be written as

$$\Delta\gamma = 2\gamma_s - \gamma_{gb}, \quad (3)$$

where  $\gamma_s$  – surface energy,  $\gamma_{gb}$  – grain boundary energy. Because usually  $2\gamma_s > \gamma_{gb}$ , then coalescence of islands is energetically favorable.

At the initial stage of coalescence, the growth of the islands and the filling of the free space between them is carried out by diffusion of adatoms on the surface of the islands and the surface of the substrate. However, when the distance between the islands becomes less than some critical value, the diffuse mechanism of filling the cavities is difficult. It has been suggested that the final stage of coalescence (corresponding to the thickness of the applied layer 10–50 nm), during which the islands become grains of solid coating and boundaries are formed between them, is accompanied by contraction of space between the side faces of neighboring islands by their spontaneous elastic deformation [13]. This mechanism allows to explain the development in the coating of tensile stresses.

According to the model first proposed in [13], the maximum tensile stress in the coating can reach

$$\sigma_{\max} = \frac{E}{(1-\nu)} \cdot \varepsilon_{\max} = \frac{E}{(1-\nu)} \cdot \frac{\Delta_{\max}}{2a} = \left[ \frac{E}{a \cdot (1-\nu)} \cdot (2 \cdot \gamma_s - \gamma_{gb}) \right]^{\frac{1}{2}} \quad (4)$$

The above model is very simplified and contains a number of assumptions that do not correspond to reality. For example, it assumes that the walls of the islands are located vertically and can interact with each other through the gap.

A more complex and physically realistic model was proposed in [4]. In it, the filling of the gap between adjacent islands and the formation of the grain boundary were considered as a process of closing the crack from the standpoint of the Griffiths destruction criterion. This model assumes that the islands have a rounded shape and their coalescence begins with the occurrence of contact at one point. Next, the area between adjacent islands is considered as a crack, the closure of which is energy efficient, because it reduces the total free energy of the system. In contrast to the above model [13], in [4] it is assumed that the process of crack closure occurs gradually, accompanied by elementary acts of elastic deformation of the islands, as long as the decrease in surface energy due to grain boundary exceeds the corresponding increase in elastic deformation. Despite the significant differences of this mechanism of coalescence of the islands from the model proposed in [13], obtained in [4] the expression for the average tensile stresses developing in the coating

$$\sigma_{aver} = \left[ \frac{(1+\nu)}{a} \cdot \frac{E}{(1-\nu)} \cdot (2 \cdot \gamma_s - \gamma_{gb}) \right]^{\frac{1}{2}}, \quad (5)$$

differs from the ratio (4) only by the factor  $1 + \nu$ . Thus, the average stresses, which are determined by relation (5), exceed the maximum stresses in (4), ie are even more inflated. Among the possible reasons leading to such overestimation, the authors [13] note the lack of consideration in the model of shear stresses occurring at the interface/substrate during the deformation of the islands, as well as the assumption that the coalescence of all islands begins simultaneously.

Another approach to the description of the mechanism of development of tensile stresses in thin coatings at the stage of coalescence of islands was proposed in [14]. The authors of this work considered the coalescence of the islands as the contact of elastic bodies between which the force of adhesion acts. This model is based on the classical Hertz theory of elastic contact of solids, taking into account the cohesive interaction between the contact surfaces [15]. For three-dimensional islands of hemispherical shape in [14] the following expression is obtained for tensile stresses averaged over the volume of the island

$$\sigma_{aver} = \frac{2 \cdot (2 \cdot \gamma_s - \gamma_{gb})}{a} \quad (6)$$

The results of this model are in better agreement with the experimentally obtained stress values. However, this model does not allow to explain the large scatter of experimental data for different coatings, as the value determined by relation (6), in fact, can change only by varying the size of the islands.

After the coalescence of the islands and the formation of a continuous thin coating, one of the main factors determining the magnitude of internal stresses in it is the diffusion mobility of adatoms, which depends on the deposition rate and homologous temperature (the ratio of the deposition temperature of the coating to its melting point  $Td/Tm$ ). In the general case, thin-film materials can be divided into two types [11, 16]. Coatings of the first type, applied at low homologous temperature ( $Td/Tm < 0,2$ ) and/or high speed. In these coatings, the tensile stresses that occur during the coalescence stage of the islands are maintained after its completion and increase with increasing thickness of the applied layer, eventually reaching saturation. Coatings of the second type are formed at higher homologous temperatures and/or low deposition rates. In these coatings with increasing thickness there is a transition of tensile internal stresses in compressive.

Another mechanism for the development of tensile stresses is associated with recrystallization and growth of grains flowing in a continuous coating, both during its deposition and after its completion [11, 16, 17]. This mechanism has a low effect on coatings of the first type, because in this case the low diffusion mobility of atoms does not contribute to significant grain growth. However, it contributes to the relaxation of compressive stresses and the re-emergence of tensile stresses in coatings of the second type, the mobility of atoms in which is high.

Since the substrate does not have a significant orienting effect on the growth of polycrystalline coatings, the location and crystallographic orientation of neighboring islands are random, which does not contribute to the formation of equilibrium boundaries when combined. In the process of grain growth there is a decrease in the number of grain boundaries and, consequently, the redistribution of free volume. As a result, the coating is compacted, which in the free state would reduce its size. The transverse dimensions of the coating remain unchanged, and it undergoes biaxial deformation, which with increasing average grain size from  $d_0$  to  $d$  is [12]:

$$\varepsilon_d = \Delta V \cdot \left( \frac{1}{d} - \frac{1}{d_0} \right) \quad (7)$$

If this deformation accommodates elastically, then the compaction of the coating during grain growth causes the development of biaxial tensile stresses in it.

$$\sigma = \frac{E}{(1 - \nu)} \cdot \varepsilon_d, \quad (8)$$

and leads to an increase in its average energy density (per unit volume of coating) by

$$\Delta F_d = \frac{E}{(1 - \nu)} \cdot \varepsilon_d^2 \quad (9)$$

From the above, it follows that the tensile stresses in the coatings can inhibit the growth of grains in them, because it is energetically unfavorable due to the additional increase in stresses and the growth of elastic energy. At rather small  $d_0$  ( $d_0 \leq 4$  HM) nm) decrease in energy density associated with a decrease in the number of grain boundaries per unit area of coating [12]:

$$\Delta F_{gb} = \gamma_{gb} \cdot \left( \frac{2}{d_0} - \frac{2}{d} \right), \quad (10)$$

may be less than the value  $\Delta F_d$ . In this case, each stress value corresponds to a certain grain size limit, at which time the growth of grains in the coating stops. If  $\Delta F_{gb} \geq \Delta F_d$  then the growth of grains will continue, leading to the development of strong deformations. In the general case, the presence of tensile stresses promotes the formation of fine-grained coatings, and compressive stresses, on the contrary, promote the growth of large grains.

In contrast to tensile stresses, the main mechanism of which in polycrystalline coatings is currently considered to be the above-described process of grain boundary formation, today there is no complete understanding of the mechanisms of development of compressive stresses. Many studies suggest that compressive stresses in solid coatings are inherited from island coatings, in which they are the result of capillary forces [7, 16, 18]. In other work [10], it is believed that, as in island coatings, in the process of growth of solid coatings there is a mechanism of “power dipole”, which leads to compression of the coating due to the interaction of adatoms with its surface. It is also assumed that the compressive stresses in thin coatings may be the result of the introduction of excess atoms between adjacent layers on the growing surface [19]. The recently proposed model assumes that stresses in solid polycrystalline coatings arise as a result of the interaction of adjacent grains, which causes their deformation, including grain boundary slippage [20].

It should be noted that the model that linked the mechanism of development of compressive stresses with the diffusion flux of atoms from the free surface of the growing coating at the grain boundary [7, 21–23].

The advantage of this model is the fact that it directly links the development of compressive stresses in polycrystalline coatings with the process of their growth, as it considers as the main reason for this the nonequilibrium nature of the surface of the growing coating.

Another advantage of this approach is that it also explains the relaxation of compressive stresses in the coatings after the termination of the application process. As soon as the flux of atoms deposited on the free surface of the coating becomes zero, the chemical potential there decreases to equilibrium. At the same time, the chemical potential at the grain boundaries remains high due to the accumulation of excess atoms. As a result, there is an oppositely directed gradient of chemical potential, which causes the reverse flow of atoms from the grain boundaries to the free surface, which leads to the relaxation of compressive stresses. Restoration of coating deposition again causes an increase in the chemical potential on the free surface of the coating and the flow of atoms at the grain boundary.

## 4 Factors Influencing the Occurrence of Internal Stresses in Thin Coatings

Along with the above processes underlying the formation of thin coatings, the development of internal stresses in them can also be caused by the formation and annihilation of point defects, the introduction of impurities, phase transformations, the formation of precipitates, absorption and desorption of moisture and volatile substances, and structural damage due to the effects of high-energy processes (for example, ion bombardment).

Point defects, which are formed in the process of applying thin coatings, contribute to the development of internal stresses. One of the important mechanisms for the development of stresses in coatings obtained by spraying is the so-called “atomic peening” [24]. The introduction of atoms sprayed into the internodes of the crystal lattice leads to distortion of the crystal lattice and excessive compaction of the coating. As a result, it has compressive stresses, the magnitude of which is directly proportional to the molar volume and biaxial modulus of elasticity of the coating [25]. In addition to the atoms of the atomized material in the interstitial of the crystal lattice can be introduced and impurity atoms and ions, such as inert gas ions used to bomb the target. Another important parameter that affects the concentration of internodal atoms is the pressure in the working chamber [26]. Under conditions of low pressure, most of the deposited atoms do not have collisions with each other and when in contact with the surface of the coating have a high energy, which facilitates their introduction into the internodes. Also, a significant influence on the development of compressive stresses in the coatings due to “atomic peening” has a shear stress applied to the substrate [27]. At high shear stresses, a strong electromagnetic field is created near the surface of the substrate, which accelerates the ions bombarding it. This leads to an increase in their kinetic energy and, as a consequence, increases the concentration of point defects in the coating.

High-energy atoms and ions can lead to the appearance in coatings not only of internodal atoms, but also of substitution atoms [27]. In this case, if the substitution occurs by an atom having a larger radius than the lattice atom, then compressive stresses develop in the coating. Otherwise, tensile stresses occur.

Another type of point defects - vacancies - can also be a source of internal stresses in thin coatings. The development of internal stresses, sign and magnitude depend on the partial molar volume of vacancies, as well as the location of the annihilation [17].

In practice, different types of point defects are present in the coatings at the same time. Therefore, their influence on the development of internal stresses is complex, as different types of defects not only cause different mechanisms of stress, but also interact with each other, which can also change the stress-strain state of the material.

Internal stresses in coatings can also occur due to phase transformations that occur inside them in the process of deposition, the formation of new phases and precipitates, as well as the introduction of impurities.

Another type of phase transformations that occur during the deposition of coatings is associated with the course of chemical reactions that lead to the formation of new phases. These can be reactions caused by mutual diffusion of the coating atoms and the substrate, or diffusion between different layers. In addition, chemical reactions of coating atoms with residual gas molecules or with embedded impurity atoms are possible.

The result of the interaction of the coating with the residual gases in the working chamber in a complex way depends on their partial pressure.

## **5 Stresses that Develop in Thin Coatings Under External Influences**

Thermal stresses make an important contribution to the overall stress level in thin coatings. Thermal stresses are external because they are caused by changes in temperature after the deposition of coatings. But they are also directly related to the deposition process of coatings, as many technologies for their application require high temperatures.

In addition, thermal stresses can develop during heat treatment of coatings after their application, as well as due to changes in temperature during operation of coatings.

The reason for the development of stresses in thin-film structures when the temperature changes is the difference between the coefficients of thermal expansion of the coating and the substrate.

If the composition coating/substrate is cooled or heated from temperature  $T_1$  to temperature  $T_2$ , the biaxial deformation of the coating that develops is determined as follows:

$$\varepsilon = -(\alpha_d - \alpha_s) \cdot (T_2 - T_1) = -\Delta\alpha \cdot \Delta T, \quad (11)$$

where  $\alpha_d$  and  $\alpha_s$  – coefficients of thermal expansion of the coating and the substrate, respectively. If the temperature changes during the deposition of the coating, then different layers of the coating will experience different deformation. In this case, its average biaxial deformation can be calculated using the ratio [17]:

$$\varepsilon \approx -\Delta\alpha \cdot \left( T_2 - \frac{1}{h_f} \cdot \int_0^h T(z) dz \right), \quad (12)$$

where  $h$  – coating thickness,  $dz$  – thickness increase. Since biaxial stresses are associated with deformation as

$$\sigma = \frac{E}{(1 - \nu)} \cdot \varepsilon, \quad (13)$$

then from (11) and (12) we obtain that the thermal stresses are directly proportional to the temperature changing:

$$\sigma = -\frac{E}{(1 - \nu)} \cdot \Delta\alpha \cdot \Delta T \quad (14)$$

Since, in thin coatings both thermal and internal stresses develop at the same time as a result they can both strengthen, and weaken each other.

A significant influence on the development of stress in current-carrying coatings, have electromigration processes.

Thus, according to [28], under the action of current, the interatomic distances in Al conductors increased near the anode and decreased near the cathode.

It should be noted that thermal stresses are always present in conductive coatings, because high-density electric current causes intense release of Joule heat, which leads to heating of the coatings. This complicates the precise determination of the magnitude of the stresses caused directly by electromigration in the coatings applied to the substrate.

Significant contribution to the development of stresses in thin coatings can have effects associated with exposure to electromagnetic fields. First of all, they include the piezoelectric effect, electro- and magnetostriction.

The piezoelectric effect under the influence of an external electric field causes the deformation of the coating crystals. And since the coatings can not be freely deformed due to the restrictions imposed by the substrate, they have mechanical stresses.



In contrast to the piezoelectric effect, electrostriction is observed in absolutely all dielectrics and is also due to their polarization in an external electric field. At the same field strength, the deformation of bulk materials caused by electrostriction is several orders of magnitude smaller than in the piezoelectric effect. But in thin-film materials, electrostriction can be an important source of stress. In particular, strong stresses caused by electrostriction develop in coatings of anode oxides of metals, the thickness of which can be several nanometers [29, 30].

It should be noted that the development of stresses in dielectric coatings under the action of an external electric field is due to two different factors. First, as a result of polarization on the surface of the coating and at its interface with the substrate, uncompensated charges appear. Which leads to the exploration of the so-called Maxwell stresses [29]:

$$\sigma_M = -\frac{\nu}{1-\nu} \cdot \frac{\varepsilon_0 \cdot \varepsilon_{di}}{2} \cdot E_e^3, \quad (15)$$

where  $\varepsilon_{di}$  – dielectric permittivity,  $\varepsilon_0$  – dielectric constant,  $E_e$  – electric field strength. As can be seen from expression (15), Maxwell's stresses have a minus sign, ie they are always compressive, which follows from the geometry of the Coulomb interaction.

Secondly, under the action of an external electric field, molecular dipoles tend to line up along its lines of force, which also causes the deformation of the coating and the occurrence of stresses in it. This effect is sometimes called dielectrostriction. According to [29, 31], the magnitude of these stresses can be recorded

$$\sigma_{ES} = -\frac{\nu}{1-\nu} \cdot \frac{\varepsilon_0}{2} \cdot [\varepsilon_{di} - (\beta_1 + \beta_2)] \cdot E_e^3, \quad (16)$$

where  $\beta_1$  and  $\beta_2$  – electrostriction parameters that depend on the dielectric permittivity of the coating. As can be seen from expression (16), the sign of the stresses caused by electrostriction is determined by the ratio  $\varepsilon$  and amounts  $\beta_1 + \beta_2$ , that is, depending on the dielectric properties of the coating, these stresses can be both compressive and tensile. The final value of the stresses caused by electrostriction depends on the ratio  $\sigma_M$  and  $\sigma_{ES}$ .

This effect of deformation of the material when changing the state of its magnetization under the action of an external magnetic field is called magnetostriction. The mechanism of stress development in thin-film structures due to magnetostriction is similar to the cases of piezoelectric effect and electrostriction described above. [32, 33].

Finally, thin coatings in the process of their operation can be directly exposed to various mechanical influences, which lead to the development of stresses in them. Thus, in recent years, researchers are of great interest to thin-film structures on flexible substrates, which have broad prospects for use in the production of flexible LED screens, solar panels, various microelectromechanical systems, etc. [34, 35]. During operation, such flexible thin-film structures experience multiple bending deformation, which leads to the development of inhomogeneously distributed mechanical stresses, which causes their high gradients.

## 6 Conclusions

The analysis showed that the study of the mechanism of stress development in the coating/substrate system and their subsequent relaxation is of great interest. The high level of mechanical stresses in thin coatings is primarily due to the rigid connection with the substrate, which prevents changes in the size of the coating in the plane parallel to the interface between them and thus prevents elastic stress relaxation. In turn, the small thickness of the coatings and the presence of the interface with the substrate impose restrictions on the sliding of dislocations, complicating the development of plastic deformation. Therefore, either there is a coherent elastic deformation of the coating/substrate system due to its bending, which provides partial relaxation of stresses, or there is a destruction of the system.

To date, there is no consensus on the adequacy of the proposed models and their agreement with the available experimental results.

Obviously, to understand and take into account all the mechanisms of occurrence and evolution of stresses in thin coatings requires further experimental and theoretical studies.

Further research is needed to fully understand the wide range of factors that control the evolution of internal and external stresses in thin coatings. The contribution of each of the mechanisms to the total amount of stresses significantly depends on the growth conditions of the coatings. Therefore, to obtain coverage with the required level of stress, it is necessary to conduct research in a wide range of parameters of their deposition.

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