

МОДЕЛЮВАННЯ ПРОЦЕСІВ ТА ЯВИЩ В ЕЛЕКТРОНІЦІ

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MODELLING ADHESION BONDS IN COPPER CONDENSATES ON MONOCRYSTALLINE SILICON

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Using macroscopic techniques of physics of surface, theoretical research has been done regarding the changes in the energy of adhesion bonds in nanolayers at the interface between copper thin films and silicon monocrystalline substrates at different speeds of deposition. The energy of adhesion bonds and the work of adhesion turned out to be an order of magnitude larger compared with interphase energy and strain, respectively.

Based on the thermodynamic approach, the method of estimating the ratio between the electric and the mechanical components of interphase energy has been elaborated.

Keywords: copper thin films, silicon substrate, surfacial and interphase energy, work of adhesion, energy of adhesion bonds.

Introduction.

As far as the employment of copper condensates in the modern microprocessor technology is highly prospective, the issue of energy characteristics of thin films of copper on the monocrystalline silicon substrates appears to be very urgent. In particular, of great importance are the quantitative estimations of the nature of physical bonds that arise in the film-substrate structure and are counter-balanced by interaction with the substrate. This frequently causes the changes of electrical and mechanical parameters in interphase layers as well as the decrease of adhesion stability of the film.

Moreover, the problem becomes more complicated since the copper on the surface of silicon to a great extent tends to unstable behavior and is capable of plastic strains [1].

The nature of structural mechanical strains of the interphase layer at the interface of metal-semiconductor structure is insufficiently investigated [1].

There are only partial modeling approaches to the mechanisms of the occurrence of corresponding strains, such as the model of surface disordering, grain-border relaxation model, border growth model, impurity model [2-4].

It is generally accepted that internal strains are caused by peculiarities of the film formation process, by nonequilibrium process of condensation, by recrystallization as well as by interaction of condensates with residual gases that penetrate into the interphase region.

Quantitative information on mechanical strains in thin films is known to have been obtained mainly through indirect measurement techniques following the condensate formation. Therefore the results of these measurements bear solely information about the integral macrostrains that occur after the film preparation.

Moreover, there is no well-founded thermodynamic analysis of the process of forming structural stresses, energy and adhesion bonds in the film-substrate structure.

Therefore the main goal of this work was to study the interphase and adhesion bonds in copper condensates on the substrate of monocrystalline silicon at different speeds of their deposition, as well as to elaborate an appropriate thermodynamic model for analyzing the regularities of their changes.

1. Thermodynamic description of interphase bonds in the copper film-silicon substrate structure.

The studies of the characteristics of interphase interaction are based on considerations of macroscopic approach that are concluded with the model ratios of nonequilibrium thermodynamics and of the physics of solid state surface [6,7].

We will explore the change of energy and adhesion parameters of thermodynamic state of the copper film-silicon substrate structure using experimental data [1], tabulated values of physical quantities for silicon and copper [8-12] as well as a system of equations that describe mechanical and electrical processes in the surfacial layers [6,7].

1.1 The method of defining the changes in the surface energy.

Interphase energy appears to be a structurally sensitive parameter of the substrate-film structure. In order to calculate this parameter we use the procedure of transition from the surface energy to the interphase one. Let us consider here a macroscopic model of the surface layer of a solid state (fig. 1) in which $x > 0$ (V_1) region is occupied by a solid medium (metal, such a copper), $x < 0$ (V_2) is occupied by air (x, y, z stand for Decart coordinates).

Let the ratio of thermodynamic model of the metal surface layer contacting with the air be presented as follows [1]:

$$\sigma_h = \int_0^h \sigma_y dx, \quad \sigma_y = \sigma_z. \quad (1)$$

$$\sigma_y + p = 0 \quad (\text{for } x = h) \quad (p = 100 \text{ kPa Atmospheric pressure}). \quad (2)$$

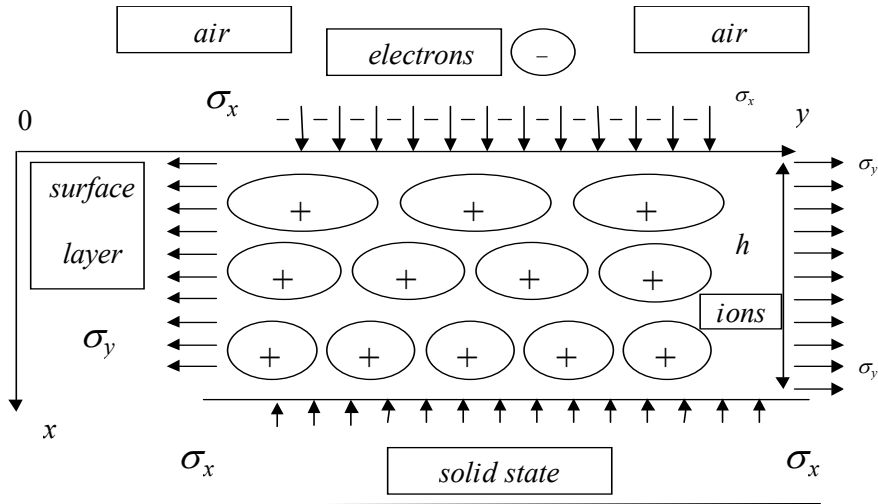
$$\gamma = \gamma_1 + \xi \gamma_2, \quad (3)$$

$$\frac{\partial \gamma}{\partial k} = \frac{\partial (\gamma_1 + \xi \gamma_2)}{\partial k} = 0, \quad (4)$$

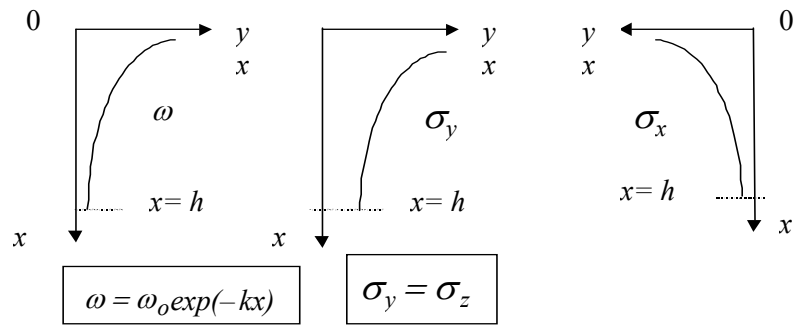
$$\sigma_{ij} = E(\nu e / (1 + \nu) - b \varphi / 3) \delta_{ij} (1 - 2\nu) + E e_{ij} / (1 + \nu), \quad (5)$$

$$\omega_v = \rho \omega = \varepsilon_0 k^2 \varphi + b E e / (3(1 + \nu)), \quad (6)$$

$$\varphi = -\Phi_0, \quad \sigma_x = -\frac{\varepsilon_0}{2} \left(\frac{\partial \Psi}{\partial x} \right)^2 \quad \text{for } x = 0. \quad (7)$$



a)



b)

c)

d)

Fig. 1. Diagram of the surface layer and the corresponding distributions of electrical charges and mechanical stresses.

Here $\gamma_1 = \int_0^h w_1 dx$; $\gamma_2 = \int_0^h w_2 dx$; $w_1 = \frac{\varepsilon_0}{2} \left(\frac{\partial \Psi}{\partial x} \right)^2$; $w_2 = \frac{\sigma_x (\sigma_x - 4\nu\sigma_y)}{2E} - \frac{(1-\nu)\sigma_y^2}{E}$; h is

effective thickness of the surface layer; σ_{ij} , e_{ij} are the components of stress tensor $\hat{\sigma}$ and strain tensor \hat{e} ($i, j = 1, 2, 3$); $\sigma_{11} = \sigma_x$; $\sigma_{22} = \sigma_y$; b, k, ξ are physical characteristics of the material; δ_{ij} are Kroneker symbol; e is the first invariant of strain tensor; ρ is material density;

ω_V , ω are spatial density and mass density of electrical charge, respectively; $\varphi = \Phi - \Phi_0$ is deviation of the modified Φ potential of electric charges from its equilibrium value Φ_0 in the volume of the body far from the surface; Ψ is scalar potential of the electric field intensity; E , ν are modulus of elasticity and Poisson coefficient.

The ratios (1-4) constitute the system of four equations for defining the physical b , k , ξ and geometrical h characteristics of the surface layer. Using the equilibrium equation $\nabla \hat{\sigma} - \rho \omega \nabla \Psi = 0$ [1], the equations of state (5), (6) and the boundary conditions (7), we find the stresses in the surface layer by expanding those stresses and strains into the rows by small parameter $b_m = b \Phi_0$.

The ratios for the description of parameters of mechanical and electric fields in silicon (semiconductor) are similar (1)-(4). Since the density of free electric charges in silicon is negligible and polarization of atoms may be rather large, let us utilize the approach according to which we assume that besides mechanical surface energy there is also a component that corresponds to the bonded electric charges.

The theory of bonded electric charges for volumetric media is expounded in the monograph [13]. This theory has been applied to the formation of surface layers of dielectric in the paper [14]. Similarly to dielectrics [14], Z_c – modified chemical potential of bonded electric charges is applied to silicon. Potential Z_c in the expression of internal energy change [14] " $dU = Z_c d\omega_c + \dots$ " turns out to be a lumped parameter in relation to density ω_c of the

bonded electric charges. The same as for dielectric we can write for silicon: $w_{1c} = \frac{\varepsilon_0}{2} \left(\frac{\partial Z_c}{\partial x} \right)^2$

– specific energy of the field of bonded electric charges; $\nabla \hat{\sigma}_c - \rho_c \omega_c \nabla Z_c = 0$ – equilibrium equation; $b_{mc} = b_c Z_c$ – small parameter.

The equation of state and boundary conditions for silicon (c) can be written down as follows [14]:

$$\sigma_{ije} = E_c (\nu_c e_c / (1 + \nu_c) - b_c \varphi_c / 3) \delta_{ij} (1 - 2\nu_c) + E_c e_{ijc} / (1 + \nu_c), \quad (8)$$

$$\omega_{cV} = \rho_c \omega_c = \varepsilon_0 k_c^2 \varphi_c + b_c E_c e_c / (3(1 + \nu_c)), \quad (9)$$

$$\varphi_c = -Z_{c0}, \quad \sigma_x = -\frac{\varepsilon_0}{2} \left(\frac{\partial Z_c}{\partial x} \right)^2 \quad \text{for } x = 0. \quad (10)$$

$\varphi_c = Z_c - Z_{c0}$ – deviation of potential Z_c from its equilibrium value Z_{c0} far away from the surface in the volume of the body [4]; $k_c = \sqrt{\rho_c C_{ce} / \varepsilon_0}$, b_c – characteristics of the material; index (c) shows that this parameter or characteristic belongs to silicon; $i, j = 1, 2, 3$.

The method of estimating the surface energy and its changes at loading in the case of copper is based on the ratios of the method of atomic interactions [15] taking into account the radial-symmetric potential of central forces $u_{\alpha\beta}$ according to Born-Mayer [16]:

$$u_{\alpha\beta} = \frac{q^2}{R_{\alpha\beta}} - \frac{c_{\alpha\beta}}{R_{\alpha\beta}^6} - \frac{d_{\alpha\beta}}{R_{\alpha\beta}^8} + b_{\alpha\beta} \exp\left(-\frac{R_{\alpha\beta}}{\rho_q}\right). \quad (11)$$

Here q is the electric charge of particles; $R_{\alpha\beta}$ is the distance between the particles α and β ; $c_{\alpha\beta}, b_{\alpha\beta}, d_{\alpha\beta}$ are constants; ρ_q is the parameter of "rigidity".

1.2 Determining the interphase energy and the energy of adhesion bonds.

Interphase energy γ_m and strain σ_m are determined similarly to paper [1]:

$$\gamma_m = \gamma_4 + \xi_m \gamma_5; \quad \gamma_4 = \int_{-H}^H w_1 dx; \quad \gamma_5 = \int_{-H}^H w_2 dx; \quad \sigma_m = \int_{-H}^H \sigma_y dx. \quad (12)$$

Here ξ_m is the physical characteristic of interphase layer; $2H$ is the effective thickness of interphase layer.

Let us put the condition of interphase layer equilibrium and the approximate conditions at the interface (at $x = H$ and $x = -H$) as follow [1]:

$$\frac{\partial \gamma_m}{\partial k} = \frac{\partial(\gamma_4 + \xi_m \gamma_5)}{\partial k} = 0; \quad \sigma_{y^+} + p = 0 \quad (x = +H); \quad \sigma_{y^-} + p = 0 \quad (x = -H). \quad (13)$$

Here index (+) corresponds to the parameters of copper, and index (-) corresponds to the parameters of silicon.

Let us present the boundary conditions for the interface (metal – semiconductor) of the media according to papers [7, 14]:

$$\varphi_+ = -\Phi_0; \quad \varphi_{c-} = \varphi_c = -Z_{c0}; \quad \sigma_{x^+} = \sigma_{x-}, \quad \sigma_{y^+} = \sigma_{y-} \quad \text{for } x = 0. \quad (14)$$

Ratios (12-14) constitute a system of equations for determining physical characteristics $\xi_m, b = (b_+, b_-), k = (k_+, k_-)$ and the thickness $2H$ of the surface layer.

As is seen from the conditions at the interface (14), the problems of determining the distribution of free electric charges as well as of the bonded ones turn out to be boundary while the problem of determining the mechanical strains is the contact one. Hence, the ratios (12)-(14) appear to be the basis for the contact-interface problem. The numeric values of physical constants applied in the above problem are taken from papers [8-12, 18].

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The work of adhesion σ_{ad} of the copper-silicon structure is determined based on the known ratio [19]:

$$\sigma_{ad} = \sigma_{h^+} + \sigma_{h^-} - \sigma_m. \quad (15)$$

The energy of adhesion bonds γ_{ad} is introduced similarly to the expression (17):

$$\gamma_{ad} = \gamma_{h+} + \gamma_{h-} - \gamma_m \quad (16)$$

1.3 Numeric calculations. Discussion of results.

Numeric values of physical constants for copper (+) and silicon (-) are as follow [8-12, 18]:

$$\begin{aligned} E_+ &= 118 \text{ GPa}; \nu_+ = 0,372; \omega_+ = 8,45 \cdot 10^{28} \text{ 1/m}^3; \sigma_{h+} = 2,125 \text{ N/m}, \gamma_+ = 1,623 \text{ J/m}^2; \\ E_- &= 109 \text{ GPa}; \nu_- = 0,317; \omega_- = 5 \cdot 10^{28} \text{ 1/m}^3; \sigma_{h-} = 1,547 \text{ N/m}, \gamma_- = 1,182 \text{ J/m}^2. \end{aligned} \quad (17)$$

Here, the value of surface energy γ_- has been obtained using the method of atomic interaction [15] and the ratio [11].

Based on the calculations within the equation system (1)–(14) for copper (+) and for silicon (-), the values of physical characteristics of the interphase layer material have been determined in the equations of state:

$$\begin{aligned} k_+ &= 2,431 \cdot 10^{10} \text{ 1/m}, \quad \xi_+ = 3,411, \quad b_+ = -0,487 \text{ 1/V}; \Phi_0 = -1,872 \text{ V}; \\ k_- &= 1,874 \cdot 10^{10} \text{ 1/m}, \quad \xi_- = 5,596, \quad b_- = -0,367 \text{ 1/V}; Z_{co} = -1,575 \text{ V}; \\ z_+ &= \gamma_{1+} / \gamma_+ = 0,221; \quad z_- = \gamma_{1-} / \gamma_- = 0,232; \quad \sigma_m = 0,3293 \text{ N/m}, \\ \gamma_m &= 0,2396 \text{ J/m}^2; \quad z_m = \gamma_4 / \gamma_m = 0,0672; \quad z_z = (z_+ + z_-) / 2z_m = 3,37. \end{aligned} \quad (18)$$

In view of three approximations of distribution by small parameter $b_c = b_c Z_{co} = 0,579$, the estimation has been carried out of the potential of bonded electric charges for silicon Z_{co} , interphase energy γ_m , interphase strain σ_m , as well as the work of adhesion bonds σ_{ad} and the energy of adhesion bonds γ_{ad} in the Cu-Si structure:

$$\sigma_{ad} = 3,34 \text{ N/m}; z_a = \sigma_{ad} / \sigma_m = 10,15; \gamma_{ad} = 2,57 \text{ J/m}^2; z_{ad} = \gamma_{ad} / \gamma_m = 10,71. \quad (19)$$

The analysis of the obtained data makes it possible to determine certain ratios:

$$z_+/z_m = 3,30; \quad z_-/z_m = 3,46; \quad \sigma_{ad} / \gamma_{ad} = 1,303. \quad (20)$$

It has been experimentally determined that of greatest interest from the viewpoint of changes of physical and mechanical characteristics is the range of speeds of depositions [1]:

$$w_n = [w_{n1}; v_{n2}] = [0,2 \text{ nm/sec}; 0,5 \text{ nm/sec}], \quad (21)$$

The following parameters of copper film [1] correspond to the above speed range:

$$D_n = [D_{n1}; D_{n2}] = [400; 1600 \text{ nm}]; \quad h_n = [h_{n1}; h_{n2}] = [56; 110 \text{ nm}]. \quad (22)$$

Here D_n is the size of grains that correspond to the thickness of the film $[h_{n1}; h_{n2}]$. According tot the method [1] for the range (21) it has been determinated:

$$E_+ = [E_{1+}; E_{2+}] = [118 \text{ GPa}; 128 \text{ GPa}]; \quad \nu_+ = [\nu_{1+}; \nu_{2+}] = [0,372; 0,347]. \quad (23)$$

In order to determine the relative changes in physical values σ_m, γ_m as well as in σ_{ad}, γ_{ad} (corresponding symbol δ) for the range (21), we have used the numerical constants (23) and based on (12)-(14) obtained the following:

$$\begin{aligned}\sigma_m &= [0,329; 0,347 \text{ N/m}]; & \delta\sigma_m &= 2(\sigma_{m1} - \sigma_{m2}) / (\sigma_{m1} + \sigma_{m2}) = 0,0512; \\ \gamma_m &= [0,240; 0,250 \text{ J/m}^2]; & \delta\gamma_m &= 2(\gamma_{m1} - \gamma_{m2}) / (\gamma_{m1} + \gamma_{m2}) = 0,0425; \\ \delta\sigma_{ad} &= 0,0052; & \delta\gamma_{ad} &= 0,0041.\end{aligned}\quad (24)$$

Thus, the obtained numerical results (17)-(20), (24) contain information concerning the peculiarities of the surface, interphase and adhesive interactions between metal (copper condensate) and semiconductor (silicon monocrystal).

Conclusions.

1. Based on thermodynamic approach to the study of mechanical and electrical processes in nanolayers near the metal-semiconductor interface, a ratio has been formed of a contact-interface problem for the purpose of determining the distribution of free electric charges in metal, bonded electric charges in semiconductor and mechanical strains.
2. It has been established that the components of surface energy both in metal (Cu) and in semiconductor (Si) are slightly different and make up 22-23 percent.
3. The electric component of interphase energy between metal (Cu) and semiconductor (Si) turned out to be 6,7 percent, that is 3,7 times lower than the electric component of surface energy. This testifies to a corresponding decrease of the capacity of the double electric layer in the vicinity of the media interface.
4. Specific numerical values have been obtained for Cu-Si structure concerning interphase strain, interphase energy, work of adhesion and the novel energy characteristic of interphase layer, that is the energy of adhesion bonds which is about 11 times higher comparative to the interphase energy. The work of adhesion is about the same magnitude higher than the interphase strain (for the contacting Cu-Si).
5. The range of deposition speed ($0,2 \div 0,5$ nm/sec) of copper condensates onto monocrystalline silicon substrates is characterized by abnormal changes of mechanical strains which is caused by the changes of mechanical modules and the sizes of the copper condensate grains [1]. However, these corresponding changes almost have no effect on the changes of the work of adhesion (Cu-Si) and on the energy of adhesion bonds since they do not exceed 0,5 percent. The calculations have been made concerning the possibility of somewhat higher changes in the process of deposition for interphase strain and interphase energy (of the order of 4-5 percent). However, such changes remain within the experimental error.
6. Based on computational experiment for the copper-silicon structure, the thermodynamic approach to the estimation of energy and adhesion characteristics of the surface layer turned out to be less cumbersome than the method of atomic interactions. Moreover, it permits the electric component of interphase energy to be taken into account. The proposed method makes it possible to calculate the interphase energy, interphase strain and the energy of adhesion bonds at the interface between the metal film and semiconductor substrate for other contacting systems employed in microelectronics (for example Al-Ge).

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**МОДЕЛЮВАННЯ АДГЕЗІЙНИХ ЗВ'ЯЗКІВ У КОНДЕНСАТАХ МІДІ
НА МОНОКРИСТАЛІЧНОМУ КРЕМНІЇ**В. Юзевич¹, Б. Коман²¹*Фізико-механічний інститут ім. Г. Карпенка НАН України,
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Теоретично, із застосуванням макроскопічних методів фізики поверхні, досліджено зміни енергії адгезійних зв'язків у наночастицях поблизу границі розділу між тонкими плівками міді та кремнієвими монокристалічними підкладками при різних швидкостях осадження. Встановлено, що енергія адгезійних зв'язків і робота адгезії на порядок більші порівняно з міжфазними енергією та натягом відповідно.

На основі термодинамічного підходу розроблено методику оцінки співвідношення між електричною і механічною складовими міжфазної енергії

Ключові слова: тонка плівка міді, кремнієва підкладка, поверхнева та міжфазна енергії, робота адгезії, енергія адгезійних зв'язків.

**МОДЕЛИРОВАНИЕ АДГЕЗИОННЫХ СВЯЗЕЙ В КОНДЕНСАТАХ
МЕДИ НА МОНОКРИСТАЛЛИЧЕСКОМ КРЕМНИИ**В. Юзевич¹, Б. Коман²¹*Фізико-механічний інститут ім. Г. Карпенка НАН України,
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Теоретически, с применением макроскопических методов физики поверхности, исследованы изменения энергии адгезионных связей в нанослоях вблизи границы раздела между тонкими пленками меди и кремниевыми монокристаллическими подложками при разных скоростях осадения. Установлено, что энергия адгезионных связей и работа адгезии на порядок больше сравнительно с межфазными энергией и натяжением соответственно. На основе термодинамического подхода разработана методика оценки соотношения между электрической и механической составляющими межфазной энергии.

Ключевые слова: тонкая пленка меди, кремниевая подложка, поверхностная и межфазная энергии, работа адгезии, энергия адгезионных связей.

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