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Heat Capacity of Thin Films at High Temperatures

E. P. Shtapenko[1] and Yu. V. Syrovatko[1,2]

¹Ukrainian State University of Science and Technologies,
2, Lazaryan Str.,
49010 Dnipro, Ukraine
²Dnipropetrovsk Branch of the State Institution
'Soil Protection Institute of Ukraine',
65A, Naukova Str.,
52071 Doslidne Settlement, Dnipro Raion, Dnipropetrovsk Oblast, Ukraine

The purpose of this paper is to develop a model, which allows determining the heat capacity of thin films at the temperatures comparable to and exceeding the Debye temperature. The model presented in the paper takes into consideration the anisotropy of vibrations of the corresponding bending waves and wave vibrations in the plane occurring with the decrease in the film thickness. Furthermore, the model is based on the quadratic dispersion law for bending wave vibrations in the normal direction of a thin film and the linear dispersion law for the wave vibrations in the film plane. In order to expand the existing model representations for the heat capacity of thin films at low temperatures, we used the Debye's method in the integral expression for the free energy. We considered this approach earlier in the model representations of the heat capacity of anisotropic quasi-crystals. Our findings show that the thin-film heat-capacity dependence on the temperature has a maximum and exceeds the heat capacity of a bulk sample. This circumstance confirms the experimental data obtained earlier by other authors. Besides, according to the experimental data collected from the literature, heat capacity of the thin films rises, compared to values of the bulk sample, when the film thickness decreases. This factor is also reflected in the model under consideration, and the calculated dependence of the increase in thin films on the number of atomic layers correlates well with the experimental data. Therefore, the proposed model allows determining the heat capacity of thin films at the temperatures exceeding the Debye temperature with sufficient accuracy without experimental investigation.

Метою даної роботи є розробка моделю, що уможливлює визначити теплоємність тонких плівок за температур, які дорівнюють і переви-

щують Дебайові температури. Представлений в роботі модель враховує анізотропію коливань відповідних хвиль вигину та коливань хвиль у площині, що виникає із зменшенням товщини плівки. Також в основу моделю покладено квадратичний дисперсійний закон для коливань хвиль вигину в нормальному напрямку тонкої плівки та лінійний дисперсійний закон для коливань хвиль у площині плівки. Для того, щоб розширити вже наявні модельні уявлення для тепломісткости тонких плівок за низьких температур, використовували метод Дебая в інтеґральному виразі вільної енергії. Цей підхід було розглянуто нами раніше у модельних уявленнях щодо тепломісткости анізотропних квазикристалів. Одержані дані показують, що залежність тепломісткости тонких плівок від температури має максимум і перевищує значення тепломісткости масивного зразка. Дана обставина підтверджується експериментальними даними, одержаними раніше іншими авторами. З використанням наведених модельних уявлень в роботі побудовано залежности тепломісткости тонких плівок алюмінію та міді від температури. Показано, що крива залежности тепломісткости має максимум і перевищує тепломісткість об'ємних зразків алюмінію та міді на 15%. Розраховано, що залежності, побудовані за допомогою наведеного моделю, справедливі для певної товщини тонких плівок, а саме, для 500 і 450 атомних шарів для алюмінію та міді відповідно. Також, згідно з літературними експериментальними даними, тепломісткість тонких плівок збільшується в порівнянні зі значеннями для масивного зразка зі зменшенням товщини плівки. Даний чинник також відображається в моделю, а розрахункова залежність збільшення тонких плівок від кількости атомарних шарів добре корелює з експериментальними даними. Таким чином, запропонований модель дає змогу визначити тепломісткість тонких плівок за температур, що перевищують Дебайову температуру, з достатньою точністю, не проводячи експериментальних досліджень.

1. INTRODUCTION

The development of microelectronic devices is currently associated with the creation of new materials of small size [1], of the order of nanometres. Microelectronic devices based on thin films are widely used in various industries [2]. Performance capabilities of microelectronic systems depend on the thermal characteristics of thin film structures [1]. Physical properties of materials of such dimensions may vary several times relative to their bulk properties [3]. Accurate measurement of the thermophysical properties of individual

thin films is important for the modelling and prediction of microsystem thermal characteristics [1]. Along with the literature data for the experimental determination of the heat capacity of thin films [4−10], there are also the theories determining the heat capacity of thin films at low temperatures [11−13].

Experimental studies show higher values of the dependence of the thin film heat capacity compared to that of bulk samples [4, 5]. In order to determine the dependence of the heat capacity of thin films on temperatures equal to and exceeding the Debye temperature, it is expedient to use the Debye's method [14]. The integration boundaries in the integral expression of free energy were replaced by finite frequency values in the study of heat capacity of anisotropic quasicrystals in [15, 16]. Therefore, the integration took into consideration the temperatures exceeding the Debye temperatures.

Experimental data also show that, in addition to the temperature dependence, the heat capacity of thin films rises with the decrease in the film thickness [6, 7], and its values exceed the heat capacity of bulk samples.

As the thickness of thin films decreases, the key role in the formation of their heat capacity value is played by vibrations of the corresponding bending waves [11], which are described by the quadratic dispersion law. Consequently, the heat capacity of thin films will have excess values compared to the heat capacity of a bulk sample. Taking into account this circumstance and our previous studies of the heat capacity of anisotropic quasi-crystals [15, 16], this paper develops a theoretical model for the dependence of the heat capacity of thin films on the temperatures exceeding the Debye temperature.

2. MODEL REPRESENTATIONS OF THIN FILM HEAT CAPACITY

Let us consider a thin film of the crystal as a set of interconnected plates. Here, the interaction between atoms lying in the plane of the plate and atoms located in the neighbouring planes is considered. According to [11], each value of the wave vector lying in the plane of plates corresponds to three waves: two of them are polarized in the plate plane, and the third one is polarized in the direction normal to the plate plane. The linear dispersion laws are valid for the first two waves:

$$\omega_1 = c_1\chi\,,\ \ \omega_2 = c_2\chi\,,\tag{1}$$

where c_1, c_2 are group velocities in the plane; χ is wave vector $(\chi^2 = k_x^2 + k_y^2)$.

For the third branch of vibrations (so-called bending waves), the

dispersion law takes the form

$$\omega_3 = \gamma\chi^2, \tag{2}$$

where γ is group velocity in the normal direction.

Taking into consideration the contribution of sound vibrations, free energy of the body F at the temperatures $T \ll \Theta$ (Θ is Debye temperature) is determined by the formula [14]

$$F = T\sum_{\alpha=1}^{3} \int \ln\left(1 - \exp\left(-\frac{\hbar\omega_\alpha}{T}\right)\right) \frac{V dk_x dk_y dk_z}{(2\pi)^3}, \tag{3}$$

where $\hbar$ is Planck's constant; ω_α is frequency of vibrations of waves of the α-th branch; V is volume of the body; k_x, k_y, k_z are wave vectors.

According to [11], when calculating the internal energy and free energy for a film, we can limit ourselves to waves where $k_z = 0$. Vibration with the wave vector lying in the film plane will be only excited in the film. Indeed, in accordance with the Bose−Einstein distribution, we can write the expression for the free energy of an individual atomic layer of the thin film m for high temperatures in the form

$$F_m = -T \ln \sum_{n=1}^{\infty} \left(\exp\left(-\varepsilon_{nm} / T\right)\right)^n, \tag{4}$$

where n is principal quantum number of the oscillators' system in one layer. When we represent $\varepsilon_{nm} = n\hbar\omega_m$, $\exp\left(-n\hbar\omega_m/T\right) = x^n$, $\sum_{n=1}^{\infty} x^n = (1-x)^{-1}$, then, Eq. (4) can be rewritten as

$$F_m = T \ln\left(1 - \exp\left(-\frac{\hbar\omega_m}{T}\right)\right), \tag{5}$$

where $\hbar\omega_m$ is energy of transition between quantum states n. For the case $T \gg \hbar\omega_m$, after expansion of the exponent to the first term,

$$F_m = T \ln\frac{\hbar\omega_m}{T}. \tag{6}$$

For the multilayer system from (6), we can write

$$F = T\sum_{i=1}^{m} \ln\frac{\hbar\omega_i}{T} = Tm \ln\frac{\hbar\varpi}{T} = T \ln\left(\frac{\hbar\varpi}{T}\right)^m, \tag{7}$$

where the expression for the geometric mean frequency was used:

$$\ln \frac{\hbar \varpi}{T} = \left(\sum_{i=1}^{m} \ln \frac{\hbar \omega_i}{T} \right) / m .$$

Thus, the free energy and, hence, the thermodynamic potential of thin films in the normal direction to the film plane have insignificant values, compared to the bulk structures, since the number of layers m in the film is several orders of magnitude less than M (the number of layers in the bulk structures), and thus,

$$\left(\frac{\hbar \varpi}{T} \right)^m \ll \left(\frac{\hbar \varpi}{T} \right)^M .$$

Consequently, wave vibrations in the orthogonal direction can be ignored for this model.

Therefore, integration in formula (3) will be carried out only by $dk_x dk_y$ in the film plane. Accordingly, (3) is rewritten as

$$F = T \sum_{\alpha=1}^{2} \int \ln \left(1 - \exp \left(- \frac{\hbar \omega_\alpha}{T} \right) \right) \frac{S dk_x dk_y}{(2\pi)^2} , \qquad (8)$$

where S is film surface area. Summation in (8) is performed in two directions, *i.e.*, vibrations in the plate plane (1) and vibrations in the direction of the normal to the plane of the bending waves' plate (2).

Integration with respect to $dk_x dk_y$ can be extended from $-\infty$ to ∞ and replaced by the integration over $2\pi\chi d\chi$ [14]. Therefore,

$$F = \frac{2TS}{2\pi} \left(\int_0^\infty \ln \left(1 - \exp \left(- \hbar c \chi / T \right) \right) \chi d\chi + \int_0^\infty \ln \left(1 - \exp \left(- \hbar \gamma \chi^2 / T \right) \right) \chi d\chi \right). \quad (9)$$

Using the Debye formalism [14], we replace the upper limit of integration ∞ by

$$y = \frac{\hbar \omega_D}{T} ,$$

where ω_D is the Debye frequency. Thus, formula (9) will be applicable for calculations at the temperatures comparable to and exceeding the Debye temperature.

Let us consider the first integral (9):

$$F_1 = 2T \frac{S}{2\pi} \left(\int_0^y \ln \left(1 - \exp \left(- \hbar c \chi / T \right) \right) \chi d\chi \right). \qquad (10)$$

Now, we make a substitution of the variable $x = \hbar c \chi / T$ and take χ out of the integral sign as a value, which varies only slightly, that is quite acceptable [17]:

$$F_1 = \frac{2T^2 S}{\hbar c 2\pi} \chi \int\limits_0^y \ln\left(1 - e^{-x}\right) dx \,. \tag{11}$$

Performing the integration by parts, we obtain two terms:

$$F_1 = \frac{2T^2 S}{\hbar c 2\pi} \chi y \ln\left(1 - e^{-y}\right) - \frac{2T^3 S}{(\hbar c)^2 2\pi} \int\limits_0^y \frac{x^2 dx}{e^x - 1} \,. \tag{12}$$

When expanding the exponent into a series in the first term in (12) to the first term and using the thermodynamic equations $S_e = -\partial F/\partial T$, $E = F + TS_e$, where S_e is entropy, E is internal energy, we obtain for the heat capacity of the first term (12):

$$C_{11} = \frac{\partial E}{\partial T} = \frac{2S}{2\pi} \chi^2 \,. \tag{13}$$

For the second term of (12), by expansion of the exponent into a series to the second term, we get

$$-\frac{2T^3 S}{(\hbar c)^2 2\pi} \int\limits_0^y \frac{x dx}{1 + x/2} \,. \tag{14}$$

After integration by parts and expansion of $\ln(1 + x/2)$ into a series to the sixth term, we obtain for (14)

$$-\frac{2T^3 S}{(\hbar c)^2 2\pi} \left(\frac{y^2}{2} - \frac{y^3}{6} + \frac{y^4}{16} - \frac{y^5}{40} + \frac{y^6}{96} - \frac{y^7}{224} \right). \tag{15}$$

From here, it follows as

$$-\frac{2TS\chi^2}{2\pi} \left(\frac{1}{2} - \frac{L}{6T} + \frac{L^2}{16T^2} - \frac{L^3}{40T^3} + \frac{L^4}{96T^4} - \frac{L^5}{224T^5} \right), \tag{16}$$

where $L = \hbar c \chi$. When entropy and energy for the heat capacity corresponding to the second term of a free energy (12) is found, we get

$$C_{12} = -\frac{S}{2\pi} \chi^2 \left(1 - \frac{3L^2}{8T^2} + \frac{2L^3}{5T^3} - \frac{5L^4}{16T^4} + \frac{6L^5}{28T^5} \right). \tag{17}$$

By summing two terms of the heat capacity (13) and (17), we obtain an expression for the found value of heat capacity corresponding to the first term of free energy of the expression (9):

$$C_1 = 3R \left(1 + \frac{3\Theta^2}{8T^2} - \frac{2\Theta^3}{5T^3} + \frac{5\Theta^4}{16T^4} - \frac{6\Theta^5}{28T^5} \right). \tag{18}$$

We took into account in the expression (18) that $\dfrac{S}{2\pi}\chi^2 = \dfrac{Sk^2 dz}{2\pi^2 dz}$.
After integration of the area over dz and k^2 over dk_z (since $k_z \sim 1/z$),
we get $\dfrac{S}{2\pi}\chi^2 = \dfrac{Vk^3}{2\pi^2 3} = 3R$, which is the Debye sphere $3R$ [14]. Coefficients of the terms of expansion into a series are similar in their physical expression to the Debye temperatures, since they are the products of the Planck's constant and complex frequencies at the boundary of the Brillouin zone, *i.e.*, Debye frequencies (determined by the limits of integration).

Now, we shall calculate the heat capacity corresponding to the second term in the expression (9) of the free energy:

$$F_2 = 2T\frac{S}{2\pi}\left(\int_0^y \ln\left(1 - \exp\left(-\hbar\gamma\chi^2 / T\right)\right)\chi d\chi \right). \tag{19}$$

After substitution of the variable $z = \hbar\gamma\chi^2/T$ and integration by parts, we get

$$F_2 = \frac{T^2 S}{\hbar\gamma 2\pi}\int_0^y \ln\left(1 - e^{-z}\right)dz = \frac{T^2 S}{\hbar\gamma 2\pi}\left(y\ln\left(1 - e^{-y}\right) - \int_0^y \frac{z\,dz}{e^z - 1} \right). \tag{20}$$

Similarly to (13), we obtain the expression for the heat capacity for the first term of (20):

$$C_{21} = \frac{\partial E}{\partial T} = \frac{S}{2\pi}\chi^2. \tag{21}$$

Then, we expand the exponent into a series in the second term of the expression (20):

$$-\frac{T^2 S}{\hbar\gamma 2\pi}\int_0^y \frac{dz}{\left(1 + z/2\right)} = -\frac{T^2 S}{\hbar\gamma 2\pi}2\ln\left(1 + z/2\right). \tag{22}$$

Further, by expansion of the logarithm function into a series to the sixth term, we obtain

$$-\frac{T^2 S}{\hbar\gamma 2\pi}\left(y - \frac{y^2}{4} + \frac{y^3}{12} - \frac{y^4}{32} + \frac{y^5}{80} - \frac{y^6}{192} \right). \tag{23}$$

From here,

$$-\frac{TS\chi^2}{2\pi}\left(1 - \frac{L}{4T} + \frac{L^2}{12T^2} - \frac{L^3}{32T^3} + \frac{L^4}{80T^4} - \frac{L^5}{192T^5} \right), \tag{24}$$

where $L = \hbar \gamma \chi^2$. After finding the entropy and energy, we obtain the expression for the heat capacity corresponding to the second term in (20):

$$C_{22} = -\frac{S\chi^2}{2\pi}\left(1 - \frac{L^2}{4T^2} + \frac{L^3}{4T^3} - \frac{3L^4}{16T^4} + \frac{L^5}{8T^5}\right). \qquad (25)$$

By summing (21) and (25), we get the expression for the heat capacity corresponding to the second integral of (9) in the free energy:

$$C_2 = 3R\left(\frac{\Theta^2}{4T^2} - \frac{\Theta^3}{4T^3} + \frac{3\Theta^4}{16T^4} - \frac{\Theta^5}{8T^5}\right). \qquad (26)$$

The heat capacity (26) is a contribution to the heat capacity from vibrations of the bending waves, which is determined by anisotropy of the dispersion laws in this model.

Therefore, full expression for the heat capacity of a thin film at high temperatures will be as follows:

$$C = C_1 + C_2 = 3R\left(1 + \frac{5\Theta^5}{8T^2} - \frac{13\Theta^3}{20T^3} + \frac{\Theta^4}{2T^4} - \frac{19\Theta^5}{56T^5}\right). \qquad (27)$$

Figures 1, 2 shows the constructed temperature dependences of the heat capacity for thin copper and aluminium films.

3. RESULTS AND DISCUSSION

As shown in Figs. 1, 2, temperature dependence of the heat capacity has a maximum of about 29 J/(mol·K). The heat capacity increase above the value of $3R$ can be explained by the contribution of vibrations of bending waves, obeying the quadratic dispersion law, to the heat capacity. Consequently, the heat capacity of thin crystal films will have increased values at high temperatures and differ from the heat capacity of crystalline bodies, which obeys the Dulong−Petit law. The papers [4, 6] deal with the experimental study of the heat capacity of thin aluminium and copper films in the temperature interval of 300−420 K. It is shown that, in this temperature range, the heat capacity of thin aluminium films of 430 nm thick [6] is higher than the heat capacity of bulk samples by $\cong 20\%$. Thin copper films' heat capacity [4] with the decrease in the film thickness below 340 nm will also rise compared to the heat capacity of bulk samples in this temperature range. The results of dependences of the heat capacity of aluminium and copper on the temperature constructed with the use of the model proposed in this paper are given in Figs. 1, 2. The figures show that, with the increase in
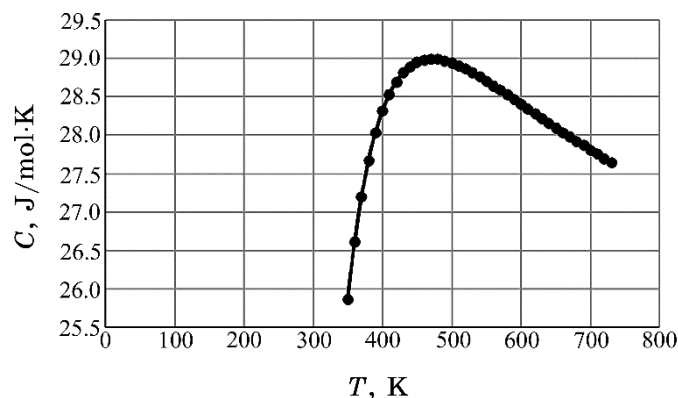
Fig. 1. Temperature dependence of the heat capacity of thin aluminium film of 500 atomic layers thick.
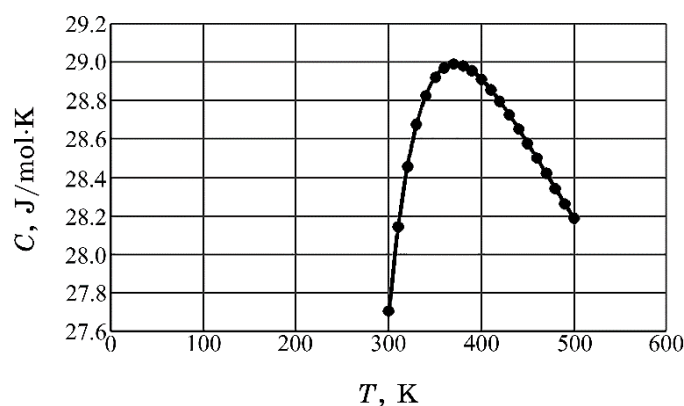

Fig. 2. Temperature dependence of the heat capacity of thin copper film of 450 atomic layers thick.

the temperature, the heat capacity of thin films rises by 15%, on average, compared to the heat capacity of bulk samples.

Further, it is expedient to form the estimated characteristics of the film thickness, for which this model is valid, and to evaluate the nature of changes in the heat capacity of thin films when the film thickness is decreased. We neglect vibrations in the direction normal to the film plane for this model. In accordance with (5), the exponential term in the expression for the free energy of one layer can be represented as

$$\exp\left(-\frac{\hbar\omega}{T}\right) \simeq \exp\left(-\frac{\Theta}{mT}\right), \tag{28}$$

where Θ is the Debye temperature of the multilayer system, m is number of atomic layers. In this case, the expression under the logarithm sign, which determines the free energy value in accordance with thermodynamic concepts, $-NT + T\ln\left(1 - \exp\left(-\dfrac{\Theta}{mT}\right)\right)$, is to be positive, *i.e.*, $\simeq \exp\left(-\dfrac{\Theta}{m_k T}\right) \approx 0.998$ (less than unity),

$$N = \frac{1}{\exp\left(\dfrac{\Theta}{T}\right) - 1}. \qquad (29)$$

Consequently, the number of atomic layers m_k for aluminium films at the considered temperatures (400 K) will be of about 500 layers. In this case, as shown above, the excess heat capacity of aluminium films will be 15% of the heat capacity of a bulk sample. The excess heat capacity of a thin film $\Delta C = C - 3R$, according to thermodynamic concepts [14], will depend on the number of atomic layers at a certain temperature as follows:

$$\Delta C_m = 3R\left(\exp\left(-\frac{\Theta}{mT}\right)\left(1 + \frac{\Theta}{mT}\right) + \frac{\exp\left(\dfrac{\Theta}{T}\right)\left(\dfrac{2T}{\Theta} - 1\right)}{m} - 1\right). \quad (30)$$

The excess heat capacity of thin film will rise with regard to the excess heat capacity of a film with the number of atomic layers m_k, respectively, by $\Delta C_m / \Delta C_{m_k}$ times. When calculating, we expand the exponential function (30) in a series up to the fourth term.

Thus, thin aluminium film of 20 nm thick will have 30 atomic layers, and its excess heat capacity will be higher than the excess heat capacity of aluminium film with 500 layers and by 2.4 times $(0.15\,\Delta C_m / \Delta C_{m_k})$ higher compared to the heat capacity of a bulk sample.

The number of atomic layers m_k for the copper film at the temperatures of $\cong 350$ K, based on (29), will be about 450. The heat capacity in this case is 1.15 times higher than the heat capacity of a bulk copper sample. Thin copper film of 20 nm thick will have 33 atomic layers and it heat capacity exceed the heat capacity of a bulk sample by a factor of 3.2.

The paper [7] considers the temperature dependence of thin platinum films of 40 nm thick. It is shown that, at the temperature of 300 K, the heat capacity has maximum values and exceeds the val-

ues of a bulk sample by 1.8–2.3 times. Further, as the temperature rises, the heat capacity decreases. According to our calculations based on (29), $m_k = 380$ layers. At the thickness of 40 nm, thin platinum film will have about 60 layers. Accordingly, at the temperature of 300 K, the heat capacity of thin platinum film of 40 nm thick will exceed the heat capacity of a bulk sample by 2.0, which is comparable with [7].

Therefore, the excess heat capacity of thin films described by this model, compared to the heat capacity of a bulk sample, agrees with the results of experimental studies on the heat capacity of thin films [4–7]. It should be noted that the proposed model could be used to calculate the heat capacity of thin films based on the input parameters: the Debye temperature, film thickness, and temperature. We should also indicate that the model considers the heat capacity for thin films of pure metals only. The development of a model to calculate the heat capacity for the films comprising several layers of different materials will be the subject of our further research.

4. CONCLUSIONS

The paper presents the model of calculation of the heat capacity for temperatures comparable to and exceeding the Debye temperature. The model describes thin films of the maximum thickness, at which the contribution of wave vibrations in the normal direction to the film plane to the integral expression of the free energy can be neglected. It is found that the temperature dependence of the heat capacity of thin films will have a maximum and increased values compared to bulk samples. It is shown that, at film thickness of 500 and 450 atomic layers, respectively, for aluminium and copper at the temperatures of about 400 K and 350 K, the heat capacity of thin films exceeds the heat capacity of bulk samples by 1.15 times that is confirmed by experimental studies of the other authors.

The proposed model takes into consideration the experimental data regarding the dependence of the heat capacity of thin films on the film thickness collected from the literature. It is found that, with the decrease in the film thickness, the heat capacity will rise and, at 40 nm for platinum, it will exceed the heat capacity of bulk samples by $\cong 2.0$ times that agrees with the experimental data.

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