Stability Limits of the Liquid Phase in the Layered Mo/Pb/Mo, Mo/Bi/Mo and Mo/In/Mo Film Systems

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The results of the study of supercooling of the fusible metals films between the continuous layers of molybdenum during the crystallization are given. Due to an original in situ technique based on the changes of resistance of samples during heating and cooling directly in the vacuum chamber the values of supercooling in Mo/Pb/Mo, Mo/Bi/Mo, Mo/In/Mo films are determined and the influence of the morphological structure of the bismuth inclusions on the temperature and nature of its supercooled melt crystallization has been found. The substrate temperature change during the condensation of samples, accompanied by a corresponding change in their microstructure, allows realizing both avalanche and diffuse crystallization and changing of supercooling value in the range of 60 to 180 K in Mo/Bi/Mo films.

Keywords: Supercooling, Multilayer films, Electrical resistance.

1. INTRODUCTION

Learning of the temperature stability limits of the liquid phase, as well as peculiar properties of its transition to the crystalline state is essential not only for the fundamental understanding of the processes that occur during crystallization of supercooled melt, but also for a variety of applications. In particular, crystallization kinetics that determines largely the structure and properties of the crystallized substance, depends substantially on the degree of supercooling [1, 2]. However, many factors have an impact on the magnitude of supercooling, the most significant of them are various kinds of impurities [3, 4], which are potential centers of crystallization. To get rid of them, there are different methods of purifying and multiple refining of the test substances, which can be combined with containerless technologies in zero gravity state or electrostatic levitation [5]. The method of micro volumes is also very effective [6], it means to divide the sample into a plurality of separate, isolated particles. As the number of external crystallization centers in the original sample is limited, there is possible presence of particles without such centers if to use sufficient granularity.

The method of micro volumes can be simply implemented due to island vacuum condensates, which can provide high purity and the required dispersibility of the test substances. Thus, the method of condensation mechanism change [4, 7] made it possible for a number of metals to obtain a record degree of supercooling, which probably corresponds to a homogeneous crystallization. In this method, the maximum temperature of supercooling during the crystallization of the liquid phase of the test substance is determined by the temperature of condensation mechanism change from vapor-crystal to vapor-liquid. To determine the temperature of condensation mechanism change it is convenient to use morphological criterion, that is the particle shape with solid and liquid phase condensation is different due to the low substances viscosity in liquid state [4, 7, 8, 9].

The easiest way to implement morphological criterion is during the study of condensed films on the substrates that are not interacting with the melt (amorphous carbon, some oxides), as in the case of condensation on the mechanism of vapor-liquid islands on the substrate become of regular spherical shape [4, 9]. In case of contact pairs with stronger interaction, i.e. a small degree of the melt contact angle of the test substance on the substrate [4], the islands are irregular in shape due to incomplete coalescence and the use of morphological criterion is difficult. Other experimental methods may also be applied for the study of supercooling, and nanocalorimetric [10] and acoustic [11] methods should be mentioned.

While studying phase transitions in the vacuum condensates, as well as the related processes (in particular, the characteristics of the formation and the interval of existence of metastable phases), an extensive empirical material on the free particles has been accumulated. A number of theoretical models has been created, which gave an opportunity to carry out computer modeling of phase transitions processes in various substances [12, 13].

The results which were obtained in [4, 7, 8, 11] as well as in similar works devoted to the study of the liquid state stability limits of metals and alloys free particles, have revealed that if the film condensation is carried out under conditions when the proportion of impurities in the sample is negligible, the limit stability temperature of the liquid phase is determined by the degree of interaction of the melt with the substrate, which has a quantitative measure of the corresponding contact angle [4, 7]. In this case the crystallization temperature for particles larger than 20 nm doesn't depend on their size [4, 14, 15].

At the same time there are less consistent and reliable data in the scientific works on the liquid state stability limits for the particles embedded in a more refractory matrix. Thus, the authors [16, 17] have studied different systems (mercury-glass, aluminum-indium) with various methods for detecting phase transitions and found no

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size dependence of supercooling. At the same time, papers [18, 19] indicate that for a wide range of contact systems the degree of supercooling during the crystallization of the embedded particles may increase for the particles of less than 20 nm size. Though papers [20] show the result which is quite different, i.e. decrease with the size of the supercooling value.

The authors [21] have found that the melting of In-Sn nanoinclusions in the aluminum matrix consists of two stages. The first stage shows significantly lower temperature than expected following the phase diagram of the contact pair, and according to the authors, it is due to the melting temperature size effect. The second stage, when the most substance melts, occurs at a temperature determined by the phase diagram. The detected melting characteristics can be interpreted as evidence of the existence of metastable phases in the system. Similar results for the Pb-Sn particles in the copper matrix have been obtained in paper [22], which has revealed that melting in this system is also a multi-step process.

During the study of crystallization In-Sn inclusions in the aluminum matrix it has been established [21] that this process is carried out in three stages. The appearance of three discrete peaks corresponding to different temperatures on the nanocalorimetric cooling curves, was associated [21] with the grain boundary crystallization, found in paper [23], and individual crystallization of various alloy phases that are present in the sample. Similar results were obtained in Ref. [24], which found out that the crystallization of supercooled melt streches at a large temperature range. In paper [25] it is shown that the character of the crystallization (diffuse or avalanche) may be determined by the crystal structure peculiarities of the matrix which contains the fusible component.

In paper [26] it is found out that the temperature and the nature of bismuth crystallization between layers of copper is determined by low-melting inclusions' morphology. Thus, depending on the sample microstructure, bismuth crystallization may take place either avalanche at 205 °C, or at a significant temperature range, which corresponds on the average to 155 °C.

Thus, peculiarities of supercooling of the "low-melting component in the refractory matrix" systems during the crystallization in the liquid phase have been studied less than the supercooling of free particles, and the results obtained for such composite materials are much more vague. Perhaps the difficulty of studying such objects are due to the fact that many of the current methods for detecting phase transitions are limited for the study of the phenomena in the nanoparticles that are embedded in a solid matrix, as well as the technological complexity of obtaining such samples which are free of uncontrollable impurities.

For this reason it seems appropriate to determine temperature limits for the liquid phase stability of the low-melting component in multilayer films, which are a model system "fusible particle in the refractory matrix." Moreover, it is interesting to study the effect of condensation conditions that determine the microstructure and morphology of the film samples, on the supercooling value and the nature of crystallization of fusible component in such systems.

2. EXPERIMENTAL

Three-layer films were selected as the objects of the study, where the low-melting metal layer (Bi, In, Pb) 20-50 nm in thickness was between continuous layers of molybdenum. The thickness of the molybdenum layers was 20-30 nm and was determined by the necessity to achieve continuity and adequate electrical resistance values. Preparation of the samples was carried out by the vacuum condensation method from independent sources at a residual gas pressure of 10^{-6} mm Hg. The thickness control of the samples was carried out during their preparation by means of the quartz resonator. In the scientific works there are no constructed phase diagrams of contact systems data. It is noted that they exhibit a negligible solubility of the components and chemical compounds are not found.

Phase transition temperature was determined by the *in situ* method that was proposed and tested in the works [26-28]. Registration of phase transitions in this method is based on the fact that in multilayer films crystallization and melting of fusible component may be accompanied by changes in the overall electrical resistance of the samples. To implement this method, test samples were deposited on a special measuring cell with a pre-deposited electrical contact system for a four-point resistance measurement circuit and K-type thermocouple attached to the back side of the cell and providing a determination of the temperature. The measuring cell heating was carried out using a low inertia radiation heater.

Electron microscopic studies were carried out using a JEOL JSM-840 scanning electron microscope on the samples deposited on the measuring cell, and also on the specially developed samples, the deposition of which was carried out on the fresh chips of NaCl single crystals.

3. RESULTS AND DISCUSSION

While studying the dependence of resistance on temperature, it was found out that for pure Mo films, as well as for multilayered samples with the low-melting component in the first heating cycle annealing of molybdenum takes place caused by diffusion processes which are actively flowing in the thin film samples [29, 30] and it is accompanied by a sharp and irreversible decrease in the resistance. The resistance of films that consist only of molybdenum in the following heating-cooling cycles depends on the temperature almost linearly and it has no particularities at the test temperature range. Thus all the features that may be found on the temperature resistance dependence' graphs for multilayered samples are uniquely associated with the low-melting component.

For films comprising low-melting metal layer, it was found out that just as in the case of samples based on copper [26], carbon [27], and aluminum [28], on the graphs of electrical resistance dependence on the temperature corresponding to heating and cooling (Fig. 1), there are peculiarities that show a sudden change in temperature coefficient of resistance at limited temperature ranges. According to the works [26-2728] such jumps can be associated with the melting and crystalli-

zation of the low-melting component. Supercooling degrees that were determined in such a way during the crystallization of the liquid phase amounted to 60, 90 and 70 K for the Bi, Pb and In, respectively.

It should be noted that, as in the previously studied Cu/Bi/Cu system [26], the bismuth crystallization in the Mo/Bi/Mo films, the condensation of which was carried out on the substrate at room temperature, is almost instantaneously spread throughout the sample. This can be explained by the fact that bismuth in the Mo/Bi/Mo films forms a unique inclusions system, which due to the large size (comparable to the size of the entire sample, that is of 1 mm) crystallizes with high probability on the random impurity centers. It is obvious that the obtained supercooling degree $\Delta T = 60$ K does not correspond to the limited supercooling in bismuth crystallization in the molybdenum matrix. In order to get the true degree of the limiting supercooling in the Mo/Bi/Mo system one should split the film into individual islands of small size in accordance with the micro volumes method. This can be accomplished by changing the conditions of sample preparation, for example, by condensation of a bismuth layer to the substrate at a higher temperature.
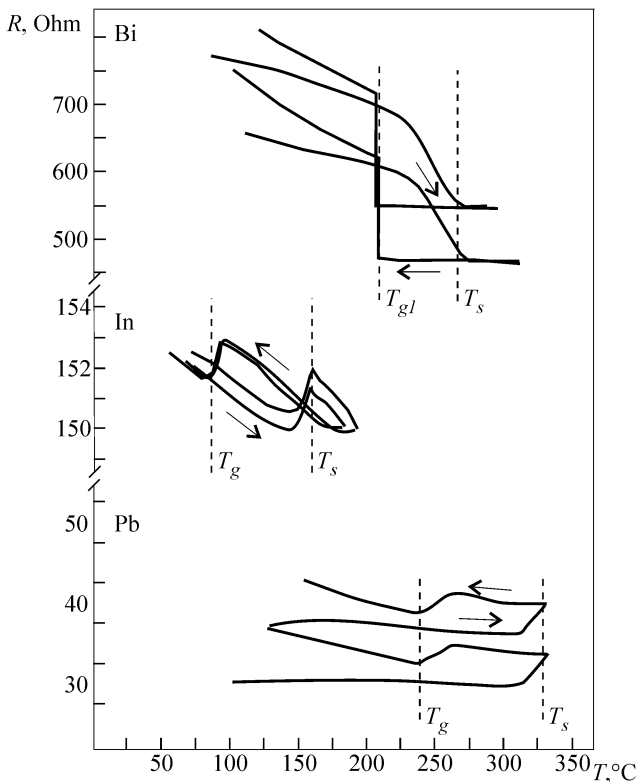

Fig. 1 – Dependence of the electrical resistance of multilayer films on temperature (low-melting component is shown on the chart)

It is known that the substrate temperature during the condensation and subsequent thermal effects have a significant impact on the microstructure and morphology of condensed films [4, 31-33]. According to [4, 7] substance condensation on substrate which temperature exceeds substance's maximum supercooling goes through a mechanism of vapor-liquid. At the same time a plurality of individual isolated particles is

formed on the substrate. On that basis the samples have been obtained where the bismuth condensation was carried out at a temperature of 230 °C, i.e. at a higher temperature than T_{g1} (Fig. 1).

The results of the study of the limiting supercooling value during the bismuth liquid phase crystallization of the samples obtained from the condensation on the mechanism of vapor-liquid, are shown in Fig. 2. As you can see, the film morphology change due to the different conditions of its condensation, significantly affects the temperature and the nature of the supercooled bismuth crystallization. Thus, the value of maximum melt supercooling increases to 180 K, and the crystallization itself is not instant, but in a certain temperature range. Temperature T_{g1} in these graphs (Fig. 2) is detected as a small leap of a few percent from the total resistance change during crystallization. It means that some fraction of bismuth inclusions still crystallizes on the external impurity centers, but now their crystallization does not cause crystallization of the entire sample.
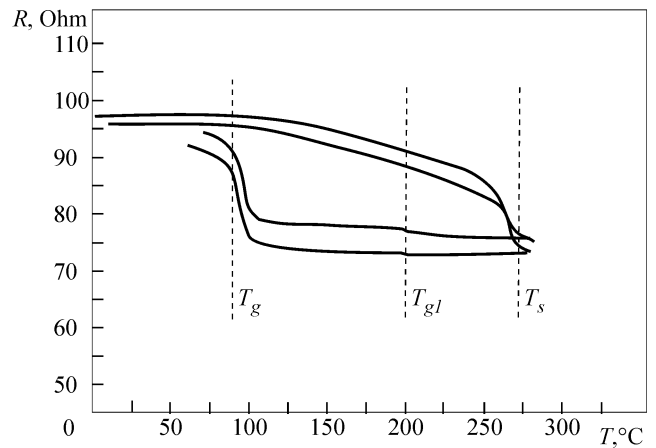

Fig. 2 – Dependence of the electrical resistance on temperature in the Mo/Bi/Mo films, where condensation of fusible component was carried out on a substrate at a temperature of 230 °C

In order to find differences in the microstructure of the films that were obtained from the condensation of various mechanisms, a number of experiments was conducted where the arrangement of the substrates, evaporators and screens allows in a single vacuum cycle, i.e. under completely identical conditions, to obtain several samples in which the substances' condensation is carried out on substrates having different temperatures. Fig. 3 shows the results of electron-microscopic examination of films Mo/Bi/Mo, which fusible -melting component condensation was carried out according to the mechanisms of the vapor-liquid and vapor-crystal. After the deposition these samples were subjected to several cycles of heating-cooling, were cooled to room temperature in a vacuum chamber and were examined in a JEOL JSM-840 SEM. It is evident that their morphology is substantially different.

Thus, in the samples that were obtained by the bismuth condensation according to the vapor-liquid mechanism (Fig. 3a) it is deposited in the form of separate, isolated particles of a spherical segment shape.

The samples where the bismuth is deposited according to the vapor-crystal mechanism (Fig. 3b) show much
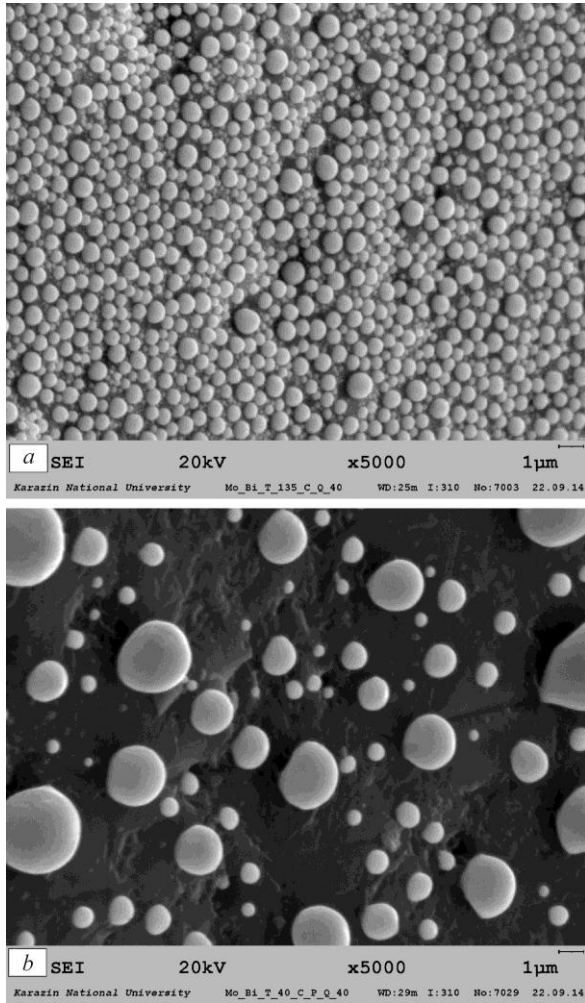
Fig. 3 – SEM images of Mo/Bi/Mo films. Samples were prepared by bismuth condensation according to the mechanism of vapor-liquid (a) and vapor-crystal (b) and subjected to several cycles of heating-cooling

much larger particles that are formed through the mechanism of liquid phase flow upon melting of continuous layer of bismuth and broke the molybdenum film. At the same time strongly deformed surface of the molybdenum film gives grounds to assume that a significant proportion of bismuth is between the layers of molybdenum and, as the nature of the crystallization shows, it forms a connected structure.

When studying the stability limits of the liquid phase of the Mo/Pb/Mo and Mo/In/Mo films (Fig. 1), it was found out that, in contrast to films with bismuth as the fusible component, even when deposited on the substrate at room temperature, under the vapor-crystall mechanism, their crystallization streches in a certain temperature range. Accordingly, the influence of the substrate temperature during the condensation of these low-melting metals on their supercooling value were not detected. This suggests that in the case of contact pairs of data a single system of inclusions in the molybdenum matrix is not formed.

Thus, among the tested metals only bismuth during the condensation according to the mechanism of vapor-crystal forms a unique inclusions system in a metal matrix, the characteristic size of which is probably

comparable to the size of the measuring cell.

Fig. 4 can provide some idea of the effect of impurities on bismuth melt supercooling and the nature of its crystallization, which shows the graphs of dependence of electrical resistance on temperature for films of Mo/Bi/Mo that were obtained by condensation according to the mechanism of vapor-liquid and kept at a residual gas pressure of about 0.1 mm Hg. for 10 (Fig. 4a) and 15 (Fig. 4b) days respectively. The graphs show that the maximum supercooling temperature at the bismuth melt crystallization does not depend on the duration of exposure and its value generally corresponds to the value of the abovementioned samples, the study of which was carried out immediately after their preparation. However, the relative magnitude of the corresponding maximum supercooling jump decreases with the holding time. In addition, in the samples one more jump is observed ($T_{g2} \approx 130$ °C), the beginning of which corresponds approximately to the temperature of bismuth crystallization with its oxides [4]. The explanation can be as follows. In the studied films bismuth condensed according to the mechanism of vapor-liquid, which, as was noted above, leads to the formation of an array of individual, isolated particles of fusible material. Since each of these isolated particles becomes solid at its temperature, the crystallization of the entire sample is in a certain temperature range. The key factor that determines the crystallization temperature of samples subjected to prolonged exposure, are insoluble impurities (primarily oxides), causing the melt crystallization long before reaching the temperature of maximum possible supercooling. During holding time the
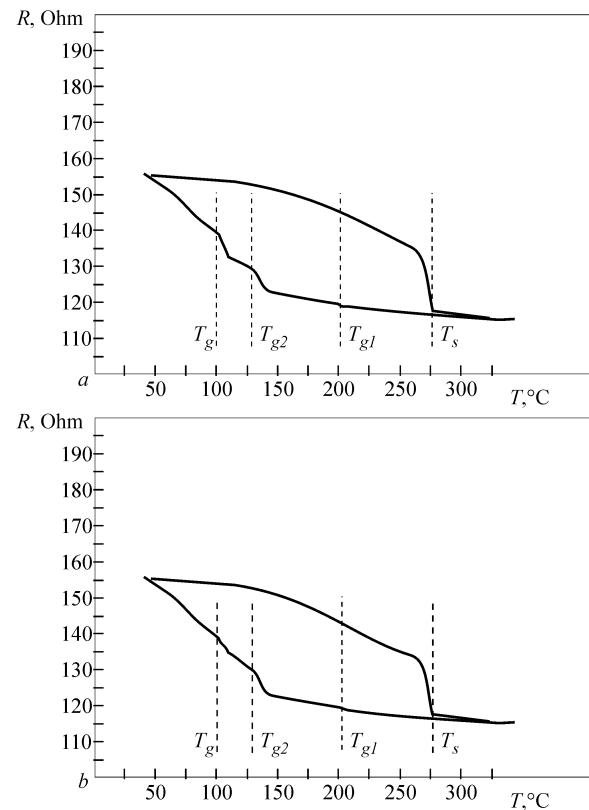

Fig. 4 – Dependence of the electrical resistance on temperature for Mo/Bi/Mo films after keeping at a pressure of 0.1 mm Hg, for 10 (a) and 15 (b) days

proportion of particles that are subjected to oxidation gradually increases, and thus the relative height of the jump increases, and it corresponds to the crystallization stimulated by oxides. The temperature of this jump is 80 K below than T_{g1} – bismuth crystallization temperature during its condensation under the mechanism of vapor-crystal (Fig. 1). This suggests that in the latter case, bismuth avalanche crystallization is not caused by oxides, but by other more active impurities, which cannot be specified yet.

The most obvious reason that can cause a change in resistance of the tested film systems, is a jump in the specific electrical resistance of low-melting component which accompanies the phase transitions. Thus, according to the table data while melting bismuth specific resistance varies 0.43, and lead and indium specific resistance doubles.

A parallel conductors' model may be used to assess the impact of this effect on the total resistance of multilayer films in the first approximation [26]. According to [26] the estimate shows that the relative change in the resistance of the samples in the phase transition must be ± (0.01-0.02). At the same time the actual value during the crystallization is 0.3, − 0.013 and − 0.02 for bismuth, indium and lead, respectively. Note that for all tested multilayer films the jump direction during melting and crystallization coincides with a change in the specific resistance of the corresponding fusible metal. As one can see the parallel conductors' model predicts the experimental results in the case when the fusible metal is indium or lead. Thus, it is reasonable to assume that in the films of Mo/Pb/Mo and Mo/In/Mo resistance jumps during the melting and crystallization are due primarily to a change in the specific resistance of the fusible component at the phase transition.

Meanwhile, in the case of bismuth, as well as in the Cu/Bi/Cu, C/Bi/C films [26, 27] the actual resistance change significantly exceeds the estimated one. This may be due to the fact that bismuth during the condensation process through condensation-stimulated diffusion and, possibly, during the heating-cooling cycles, is concentrated within the area of grain boundaries of the molybdenum film. Since the specific volume of bismuth, unlike most metals, increases upon melting, this leads to the fact that during its crystallization in a molybdenum film, stresses arise which contribute to a sharp increase in its resistance. The defects in whole or in part are annealed during the subsequent heating cycle, which is accompanied by a decrease in resistance of the samples.

4. CONCLUSIONS

Due to the original in situ technique the supercooling values have been determined in the crystallization of the liquid phase of fusible component in the three-layer Mo/Bi/Mo, Mo/In/Mo and Mo/Pb/Mo films.

It was found out that in the case of bismuth the transition from the condensation of vapor-crystal to vapor-liquid, which is accompanied by changes in the microstructure of the films, significantly affects the degree of supercooling and nature of melt crystallization. At the same time for other studied fusible metals, the condensation conditions do not affect the temperature of supercooling of the liquid phase.

It is shown that the main reason which causes the change in electrical resistance at the phase transitions in Mo/In/Mo and Mo/Pb/Mo films, is jumping resistivity of a low-melting component in the phase transition. At the same time, the resistance change in Mo/Bi/Mo films is due primarily to mechanical stress at the interphase limits.

Границы стабильности жидкой фазы в слоистых пленочных системах Mo/Pb/Mo, Mo/Bi/Mo и Mo/In/Mo

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Представлены результаты исследования переохлаждения при кристаллизации пленок легкоплавких металлов, находящихся между сплошными слоями молибдена. С помощью оригинальной in situ методики, основанной на измерении сопротивления образцов при их нагреве и охлаждении непосредственно в вакуумной камере, определены величины переохлаждений в пленках Mo/Pb/Mo, Mo/Bi/Mo, Mo/In/Mo и обнаружено влияние морфологической структуры включений висмута на температуру и характер кристаллизации его переохлажденного расплава. Изменение температуры подложки при конденсации образцов, сопровождающееся соответствующим изменением их микроструктуры, позволяет реализовать в пленках Mo/Bi/Mo как лавинообразную так и диффузную кристаллизацию и изменять величину переохлаждения в пределах от 60 до 180 K.

Ключевые слова: Переохлаждение, Многослойные пленки, Электросопротивление.

Межі стабільності рідкої фази у шаруватих плівкових системах Mo/Pb/Mo, Mo/Bi/Mo і Mo/In/Mo

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Наведено результати дослідження переохолодження при кристалізації плівок легкоплавких металів, що знаходяться між суцільними шарами молібдену. За допомогою оригінальної in situ методики, що заснована на вимірюванні опору зразків під час їх нагрівання та охолодження безпосередньо у вакуумній камері, визначено величини переохолоджень у плівках Mo/Pb/Mo, Mo/Bi/Mo, Mo/In/Mo і виявлено вплив морфологічної структури включень вісмуту на температуру та характер кристалізації його переохолодженого розплаву. Зміна температури підкладки під час осадження плівок, що супроводжується відповідною зміною їх мікроструктури, дозволяє отримати у плівках Mo/Bi/Mo як лавиноподібну, так і дифузну кристалізацію та змінювати величину переохолодження в межах від 60 до 180 K.

Ключевые слова: Переохолодження, Багатошарові плівки, Електричний опір.

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