Mechanisms of spontaneous crystallization of amorphous metallic films

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Three different mechanisms of spontaneous crystallization of pure amorphous metals (Bi, Yb, and Ga) were observed in the course of their condensation on substrates cooled with liquid helium or hydrogen. The first is the usual mechanism of homogeneous nucleation and growth of the crystal phase in an amorphous matrix. It is realized during the film condensation if the film thickness reaches the value for which the crystallization temperature becomes equal to the substrate temperature. The second and third mechanisms are connected with undamped and damped avalanche (explosive) crystallization. The avalanche crystallization sets in spontaneously when the film reaches a critical thickness in the course of condensation. The mechanism of undamped avalanche crystallization is characterized by motion of the crystallization front over the volume at a speed 10–20 m/sec. The results are discussed in the framework of the theory of thermal instability of the front of the phase transition in the decay of "frozen-in" metastable states.

1. INTRODUCTION

One of the most effective methods of obtaining amorphous metallic alloys is vapor condensation on substrates cooled by liquid helium or hydrogen. This method, first proposed by Shal'nikov,[1] is the only one that made it possible to obtain certain pure metals (Bi, Ga, Yb, V, Co, Fe, Nb, and others) in the amorphous state.[2] Amorphous metals and alloys are metastable and undergo an irreversible transition in the crystalline state (*a–k* transition) when heated above a certain temperature $T_{a\to k}$. There is also another parameter that limits the region of existence of the amorphous phase in the condensed layers. This is the critical thickness $D_{\rm cr}$ of the amorphous film, which when exceeded a jumplike crystallization of the sample occurs. The critical thickness was first found[3] and exhaustively investigated[4] in amorphous Sb films. It was later observed also in films of many amorphous metals and alloys.[2] For amorphous Sb films there was found,[4] besides the critical thickness, one other characteristic thickness (which we donote $D_{a\to k}$). Amorphous films thicker than $D_{a\to k}$ are stable at room temperature. In the interval $D_{a\to k} < D < D_{\rm cr}$ a slow crystallization of the Sb films (in contrast to the "explosive" one at $D = D_{\rm cr}$) takes place during the condensation, whereby spherulites of stable phase grow by diffusion. The crystallization rate increases with increasing film thickness. It was shown[4] that the values of $D_{a\to k}$ and $D_{\rm cr}$ depend substantially on the condensation rate and on the substrate material and temperature.

There is one other known method of crystallization of amorphous substances. Local initiation of the $a\to k$ transition in films of Ge (Ref. 5), Si (Ref. 6), (In, Ga) Sb (Ref. 7), Fe_{70} Ni_{30} (Ref. 8) and Bi and Yb (Ref. 9) can lead to avalanchelike propagation of the phase transition through the entire sample. Self-maintaining avalanche crystallization is due to intense release of the latent heat of the transition at the phase interface, which leads to a substantial self-heating of the crystallization front. The front velocity is in this case 1–20 m/sec. It was suggested that the mechanisms of spontaneous crystallization of critical-thickness films and of avalanche crystallization are identical. So far, however, this suggestion is supported by a single experimental study.[10] It has shown that spontaneous crystallization of amorphous Sb films that reached critical thickness in the course of condensation propagates from one center through the entire sample at a front velocity ≈ 32 m/sec. The dynamics of the jumplike crystallization of amorphous films of other metals and alloys that have reached critical thickness in the course of condensation[2] has not been investigated before.

The present paper is devoted to the study of the mechanisms of spontaneous crystallization of amorphous films of Yb, Bi, and Ga during their condensation at various substrate temperatures.

2. PROCEDURE

The procedure used by us to obtain amorphous films in ultrahigh vacuum ($\sim 10^{-10}$ Pa before the start of evaporation) was described in detail earlier.[2]

We investigated Yb, Bi, and Ga films condensed on polished glass surfaces cooled with liquid helium or hydrogen. During the condensation the average film temperature $T'0$ did not exceed the coolant temperature by more than 1–2 degrees.[2] The phase composition of the films was determined from the electric resistivity. The resistivities ρ of the investigated amorphous films thicker than 20 nm are (in $\mu\Omega\cdot$cm) 140 ± 20, 120 ± 20, and 26 ± 3 for Bi, Yb, and Ga, respectively. In the $a\to k$ transition, the values of ρ of Yb and Ga films decrease by a factor 5–10, and that of Bi increases by approximately a decade. Thus, crystallization of amorphous Bi, Ga, and Yb films is accompanied by a resistance jump, which can serve as an indicator of the phase transition.

Besides resistivity, the superconducting properties of the Bi and Ga films can be used to determine their phase composition. The transition of these metals from the amorphous to the crystalline state is accompanied by the vanishing of the superconductivity of Bi films and by a substantial lowering of the superconducting transition temperature T_c in the case of Ga.[1)]

The connection between the electronic properties and the structure of Bi, Yb, and Ga was established by many

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workers.[11-13] Knowing this connection, it is possible to assess unambiguously the structure of the investigated films from the change of their electronic properties. This method, while not determining directly the structure states, nevertheless identifies without fail the film state (amorphous, crystalline, or mixed). To avoid erroneous assessments of the structural states of a film, attention must be paid to its shape. The usually employed film ribbons are cut using a mask placed on the substrate. Since the mask does not make perfect contact with the substrate surface, the condensed film always has wedge-shaped edges. This is immaterial for the detection of a phase transition by resistance if the sample is not superconducting during the condensation. If, however, at the temperature of the film during the condensation, the metal of the film is superconducting in the amorphous state but not in the crystalline (as is, e.g., Bi), the presence of the edges is undesirable. The thinner edge sections do not crystallize and remain superconducting while the main part of the film, having reached critical thickness, undergoes an $a \rightarrow k$ transition. This masks the change of the resistance during the transition. Therefore to investigate Bi and Ga films condensed on substrates cooled to liquid helium temperature a disk geometry of the films was used.[14,15] The film had the shape of a disk, and electrodes for the measurement of the resistance were attached to its center and edge. In our case the edge effect is minimal. In contrast to Bi, the crystallization of amorphous Ga films (even disk-shaped) cannot be detected during condensation by the vanishing of the superconductivity. The point is that amorphous Ga, whose $T_c = 8.6$ K, crystallizes with formation of metastable β-Ga, for which $T_c = 6.5$ K. Since the film temperature during condensation is lower than 6.5 K, the superconductivity is not destroyed in the $a \rightarrow k$ transition. Ga was therefore condensed in a magnetic field of intensity 16 kOe, which destroys completely the superconductivity of amorphous and β-gallium thicker than 50 nm.[16] Under this condition the start of the crystallization was identified by the resistance jump.

All the films condensed on substrates cooled with liquid hydrogen were ribbon-shaped.[2] The change of the film resistance during the time of condensation was monitored and recorded with a KSP-4 automatic recording potentiometer. The time of the jumplike crystallization of the critical-thickness films was determined with an S8-1 memory oscilloscope equipped with a special device for triggering the sweep by the investigated signal (the change of the voltage across the sample at the instant of the transition.[17] The delay in the triggering of the sweep was $\sim 3\%$ of the rise time of the investigated signal, and the amplitude loss did not exceed $\sim 1\%$.

3. EXPERIMENTAL RESULTS

3.1. Condensation on liquid-helium-cooled substrate

The behavior of the resistance R of Yb and Bi films during condensation, monitored with a KSP-4 automatic recording potentiometer, has the following features. From the instant when a Yb film becomes continuous, its resistance decreases smoothly with increasing thickness. When the film reaches critical thickness[2)] R decreases jumpwise by 75–80% (Fig. 1a). Further condensation is accompanied by a smooth decrease of the resistance on account of the increased thickness of the layer, which is now already metallic. Increasing the thickness of an amorphous Bi film from 2 to 10 nm is accompanied by an increase of its superconducting-transition temperature from 2 to 6 K.[18] Since the average film temperature T_0' during the condensation is about 5 K, the film is not superconducting until its thickness reaches ~ 6 nm, corresponding to $T_c \approx 5$ K. Up to that instant the resistance decreases smoothly with increasing layer thickness. The resistance then becomes equal to zero and remains so until the critical film thickness is reached. At $D = D_{\rm cr}$ a jumplike appearance of resistance is observed (Fig. 1b), since the $a \rightarrow k$ transition of Bi is accompanied by loss of superconductivity. Further increase of the thickness of the Bi layer (which is now crystalline) is characterized by smooth decrease of R.

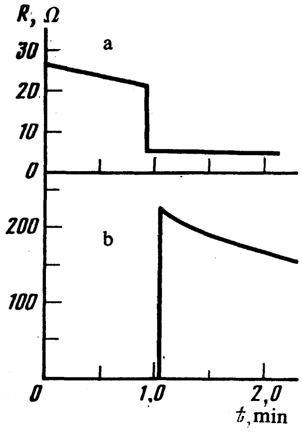

FIG. 1. Change of the resistance in the course of condensation of Yb and Bi films on substrates cooled with liquid helium: a—Yb film, $D_{\rm cr} = 110$ nm, condensation rate $v_{\rm cond} \approx 60$ nm/min; b—Bi film, $D_{\rm cr} \approx 36$ nm, $v_{\rm cond} \approx 10$ nm/min.

The phase-transition times, to which correspond the vertical sections of the curves of Fig. 1, were determined from oscillograms, typical examples of which are shown in Fig. 2. The time $t_{a \rightarrow k}$ of the complete $a \rightarrow k$ transition of amorphous Yb films (~ 10 mm long) is 500 to 1000 μsec. The crystallization time of amorphous Bi films (disk of radius ~ 3.5 mm) is 120–180 μsec.

Certain experiments, in which the resistances of separate film sections insulated beforehand from one another were monitored in the course of the condensation, have shown that the different sections do not crystallize simultaneously. The time intervals Δt between the transitions of different sections of one and the same film depend on the condensation rate, but at any rate they exceed by three to five orders the times $t_{a \rightarrow k}$. This is evidence that, under ordinary experimental conditions, a spontaneous $a \rightarrow k$ transition of some local section of the film takes place when the critical thickness is reached during the condensation of an amorphous Yb or Bi film. The crystallization process propagates next avalanchelike through the entire sample. Indeed, the values of $t_{a \rightarrow k}$ obtained in the present study are approximately equal[3)](within a factor of two) to the transition times

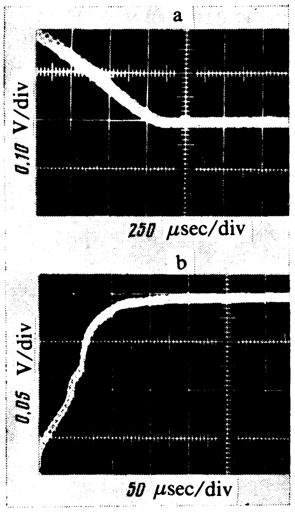

FIG. 2. Oscillograms of spontaneous crystallization of amorphous Yb (a) and Bi (b) films when they reach critical thickness in the course of condensation. The inclined sections of the oscillograms correspond to the vertical sections of the corresponding curves in Fig. 1.

in the avalanche crystallization of amorphous Yb and Bi films.[9] Thus, in both cases the transition proceeds via linear motion of the crystallization front with equal velocity ($\sim$ 10–20 m/sec), i.e., the transition mechanism is one and the same.

We note that in contrast to Sb (Ref. 4) the critical thickness of Yb or Bi films condensed at a substrate temperature $T_0 = 4.2$ K is practically independent of the condensation rate in the interval 0.35–60 nm/min. It is possible that this dependence is masked by the influence of stronger factors, for example impurities.

A behavior different from that described above for Yb and Bi films is observed when an amorphous gallium film is condensed on a liquid-helium-cooled substrate. Figure 3 shows the variation of the resistance for Ga film condensation in the critical-thickness region. Up to the curve section marked by the arrow (where $D = D_{\mathrm{cr}}$), the resistance decreases smoothly with increasing thickness of the amorphous layer. At the instant when the film reaches the value $D = D_{\mathrm{cr}}$, a small jump of the resistance is observed. Next, jumps of this kind, differing in size, repeat at unequal time intervals up to values $D \gg D_{\mathrm{cr}}$. These jumps correspond to $a \rightarrow k$ transitions of small volumes of the amorphous Ga phase into the metastable β-Ga phase. Evidence of this is, in particular, the shape of the superconducting-transition curve of layers with thickness larger than critical.[19] The steplike character of this curve indicates that both low-temperature modifications of Ga (amorphous and β-gallium) exist simultaneously in the film and their T_c values are 8.6 and 6.5 K, respectively. If the Ga condensation is stopped at the instant of the first jump on the $R(t)$ curve, the film resistance at $T_0 = 4.2$ K will practically remain unchanged with time. A noticeable change of R, corresponding to further crystallization of the sample, begins only after it is heated to $T_0 = T_{a\rightarrow k} \approx 15$ K. This result indicates that in the intervals between the resistance jumps (see Fig. 3) there is no crystallization, and the observed change of R is due only to the increase of the layer thickness.

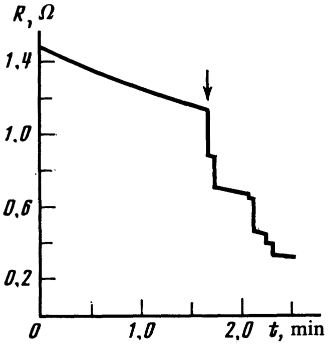

FIG. 3. Variation of resistance during the time of condensation of a Ga film on a substrate cooled with liquid helium. The arrow shows the instant when the film reaches critical thickness. $D_{\mathrm{cr}} \approx 210$ nm; $v_{\mathrm{cond}} \approx 33$ nm/min.

3.2. Condensation on liquid-hydrogen-cooled substrate

When Bi, Ga, or Yb vapor is condensed on a substrate cooled with liquid hydrogen, amorphous modifications of these metals are first produced. This is demonstrated by their resistivities, and in the case of Bi or Ga also by the values of T_{cr}, which are comparable with the values obtained for films of the same thickness obtained at $T_0 = 4.2$ K. The crystallization activation energies determined for several amorphous Bi films are likewise the same at $T_0 = 4.2$ K as at $T_0 = 20.4$ K. It can be assumed that the structural state of the films is the same in both cases.

When a certain thickness $D_{a\rightarrow k}$ is reached in the course of the condensation, the films of the investigated metals begin to crystallize slowly. By way of example, Fig. 4 shows the change of the resistance of Bi films in a region of thickness $D_{a\rightarrow k}$. The behavior of the Bi resistance is clearer, since it increases upon crystallization, in contrast to Yb and Ga films. Figure 4a describes the behavior of the resistance of a

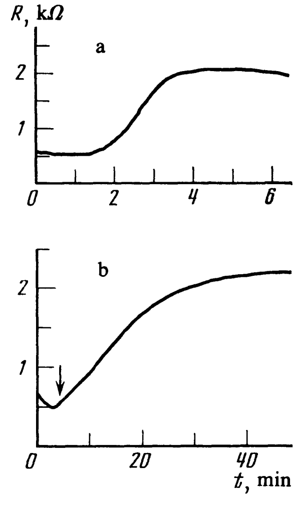

FIG. 4. Time dependence of resistance of Bi film condensed at $T_0 = 20.4$ K: a—in the course of continuous condensation; final film thickness $D \approx 21$ nm, $v_{\mathrm{cond}} \approx 1.5$ nm/min; b—with the film maintained at $T_0 = 20.4$ K. The arrow shows the instant when condensation stops at $D \approx D_{a\rightarrow k} \approx 10$ nm.

Bi film during its condensation to a thickness exceeding $D_{a\to k}$. Figure 4b shows the behavior of the resistance of a film of thickness close to $D_{a\to k}$ when it is maintained at $T_0 = 20.4$ K. In both cases the resistance growth is due to the $a\to k$ transition. It can be seen from Fig. 4 that the crystallization is not jumplike but slow, over a long time interval. The same holds for the crystallization of amorphous Yb and Ga films if in the course of condensation on a liquid-hydrogen-cooled substrate they reach the thickness $D_{a\to k}$. The values of $D_{a\to k}$ are approximately 10, 12, and 8 nm for Bi, Ga, and Yb, respectively (at $T_0 = 20.4$ K). Films thicker than $D_{a\to k}$ are crystallized in the entire volume when kept at $T_0 = 20.4$. The thicker the film the shorter the time needed to complete the $a\to k$ transition. Bi, Ga, and Yb films of respective thickness 20, 25, and 15 nm are completely crystallized by the instant when their condensation is completed at $T_0 = 20.4$ K. This is attested by the absence of resistance jumps on the heating curves, as well as by the absence of superconductivity at the corresponding temperatures in the case of Bi and Ga.

The values of $T_{a\to k}$ of films of pure amorphous metals[2,18] and of at least a few metallic alloys[20,21] increase substantially with decreasing film thickness. Figure 5 shows the $T_{a\to k}(D)$ dependence for amorphous Bi films obtained at $T_0 = 4.2$, 14.3, and 20.4 K. It can be seen from Fig. 5 that independently of the condensation conditions all the values of $T_{a\to k}$ are satisfactorily described by one curve. This is obviously connected with the action of one and the same crystallization mechanism. In particular, the spontaneous transition of films condensed at $T_0 = 20.4$ (or 14.3) K is due to the fact that a thickness $D_{a\to k}$ corresponding to the value $T_{a\to k} = T_0$ is reached during the time of condensation. If the temperature rise of the film above the substrate is small ($\Delta T_0 = T_0' - T_0$ is small), the crystallization can continue also after the condensation stops.

An analysis of the behavior of the resistance of individual sections of the same films but insulated from one another in the course of an isothermal $a\to k$ transition shows that the transition begins practically simultaneously in all the sections. This indicates that the transition mechansim constitutes homogeneous formation and growth of the crystalline phase in the amorphous matrix of the sample.

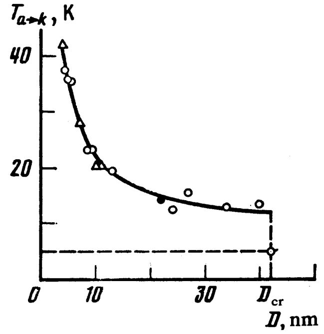

FIG. 5. Dependence of crystallization temperature of amorphous Bi films on their thickness ○—for $T_0 = 4.2$ K; △ – $T_0 = 20.4$ K; ●–$T_0 = 14.3$ K. The horizontal dashed line corresponds to the average film temperature T'_0 in the course of their condensation; the vertical dashed line corresponds to the critical thickness (both lines are for $T_0 = 4.2$ K).

One final observation: attempts to initiate avalanche crystallization of films condensed at $T_0 = 20$ or 14.3 K by using a local current pulse[9] were unsuccessful. It was found that no avalanche crystallization is realized in such films all the way to $D \approx D_{a\to k}$.

4. DISCUSSION OF RESULTS

Our results can be qualitatively explained within the framework of the theory of thermal instability of a phase-transition front upon decay of "frozen-in" metastable states.[22] The expression, known from the classical theory of crystallization of a melt, for the front velocity of a diffusionless phase-transition via the normal-growth mechanism, is considered in the theory jointly with the heat-transfer equation. A graphical solution of the system of these two equations made it possible to obtain the dependence of the crystallization-front velocity V of an amorphous metal on the substrate temperature T_0. It was shown in Ref. 22, in particular, that typical of the region of "frozen-in" (amorphous) states is the S-shaped $V(T_0)$ dependence shown schematically in Fig. 6a. As can be seen from the figure, at $T_0 < T_{\text{lim}}$ or $T_0 > T_{\text{cr}}$ this dependence is single-valued. In the case $T_{\text{lim}} < T_0 < T_{\text{cr}}$, three values of V correspond to each value of T_0, and the stable ones are V_1 and V_3. The value V_1 corresponds to a slow, almost isothermal ($T_f = T_0$) motion of the crystallization front; V_3 corresponds to a fast almost adiabatic ($T_f = T_0 + Q/C$) front motion[4)] effected by its appreciable self-heating. In the latter case, since the crystallization front is a moving thermal domain whose cooling length l_f is of the order of several microns,[22] the average temperature of a sample of length $l \gg l_f$ is close to T_0. The velocity V_1 is typical of slow heating of amorphous substances, and V_3 is realized in avalanche (explosive) crystallization. An equation satisfied by the critical substrate temperature ($T_0 = T_{\text{cr}}$) is obtained in Ref. 22:

$$\exp(-E/RT_{\text{cr}}) = (v/eV_0)(RT_{\text{cr}}^2/ET_Q). \tag{1}$$

Here E is the crystallization activation energy (J/mol);

$$T_Q = Q/C, \quad v = 2(\alpha k/DC)^{1/2}, \quad V_0 = S\exp(-\Delta H/RT_{\text{melt}}),$$

where α is the effective transparency of the film–substate interface for phonons, k is the thermal diffusivity of the film,

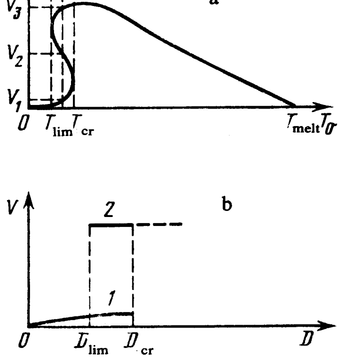

FIG. 6. Schematic plots of the crystallization front velocity vs the substrate temperature (a) and the film thickness (b).

S is a pre-exponential factor of the order of the speed of sound, ΔH is the latent heat of the phase transition at the melting temperature T_{melt}, R is the gas constant, and $e = 2.718\ldots$.

Expression (1), which is suitable for pure amorphous metals, can be regarded also as an equation for the critical thickness D_{cr} (at constant T_0), assuming that at thermal instability sets in at $D > D_{\text{cr}}$.[22] It can be seen that

$$D_{\text{cr}} = \frac{4R^2T_0^4\alpha kC\exp(2E/RT_0)}{e^2V_0^2E^2Q^2}. \quad (2)$$

We note, however, that the dependence, obtained in our experiments, of the critical thickness on the principal parameters that characterize the thermophysical properties of the film + substrate system can be described by Eq. (2) only qualitatively. The theory does not take into account certain factors that influence the formation of the condensed films and the initiation of the jumplike crystallization in them. For example, no account is taken of the heat of condensation and of the energy of the radiation from the evaporator. No allowance was made also for the influences of the stresses that can lead to severalfold change of D_{cr} while the other conditions remain unchanged.[23] Essential, however, for the understanding of the results is that the theory explains the very possibility of the appearance of a thermal instability of the crystallization front at a definite thickness of the amorphous layer.

In addition to (1) and (2), it is possible to obtain from the equations of Ref. 22 relations for the limiting temperature T_{lim} and the limiting thickness D_{lim}, parameters that restrict the possible realization of avalanche crystallization. The dependence of the crystallization front velocity on D is shown schematically in Fig. 6b. If the film thickness is less than D_{lim}, only slow motion of the crystallization-front is possible. The dependence of the crystallization-front velocity on D is shown in Fig. 6b. If the film thickness is less than D_{lim}, only slow crystallization-front motion is possible (curve 1). This was demonstrated experimentally[9] for amorphous Bi and Yb films obtained by condensation at $T_0 = 4.2$ K. The value of D_{lim} was 25–35 nm for Bi and 35–45 nm for Yb. It appears that the absence of avalanche crystallization in Yb and Bi films condensed at $T_0 = 20.4$ K and $T_0 = 14.3$ K is also connected with the fact that their values of D_{cr} are less than D_{lim} at the condensation temperature.

According to Fig. 6b, both the slow regime (curve 1) and the fast one (curve 2) are possible at $D_{\text{lim}} < D < D_{\text{cr}}$. The validity of this statement was demonstrated experimentally earlier[9] likewise for amorphous Yb and Bi films. At a thickness $D_{\text{lim}} < D < D_{\text{cr}}$ they crystallize either by slow isothermal heating (after an arbitrarily long time interval, depending on the temperature), or avalanche crystalization can be initiated in them.

Finally, a possibility exists of only a fast avalanchelike transition regime, which occurs spontaneously when the film reaches the critical thickness during its condensation. Such a behavior is typical, for example, of Yb and Bi layers condensed at $T_0 = 4.2$ K (see Figs. 1 and 2). The velocity of the crystallization front will be equal in this case to a certain value V_3 corresponding to the upper part of the S-shaped curve (Fig. 6a). The maximum values of V_3 observable for Yb and Bi films are ~10 and ~20 m/sec, respectively.

As shown in the present paper, when the critical thickness is reached in the course of condensation the $a \rightarrow k$ transition always begins locally, obviously with an onset of a crystalline domain in the amorphous matrix of the film. The mechanism of this onset is not clear, and has possibly a fluctuating character. It is possible that density fluctuations, which always occur in an amorphous layer, are significant here. Since the heat transfer from the film decreases with increasing thickness,[22] the released heat of condensation may be sufficient for crystallization of the least stable section of the film. The transition latent heat Q released thereby initiates then avalanche crystallization of the entire volume of the amorphous layer. It can thus be stated that the conditions for the existence of a critical thickness of condensed amorphous films are determined principally by the conditions under which avalanche crystallizations are realized in them.

Besides cases of undamped avalanche crystallization, we have demonstrated in the present paper the feasibility of rapid damping of this process. Thus, an avalanche crystallization process obviously occurs when a condensed Ga film reaches critical thickness, as attested by resistance jump shown by the arrow in Fig. 4. The transition, however, is damped rapidly. With further condensation, an avalanche transition sets in at another point of the sample, is again damped, and so forth. It was shown earlier[19] that residues of the amorphous phase are preserved in Ga films of thickness three times critical. The possible cause of the stabilization of the amorphous state may be the stresses that occur, for example because of the difference between the specific volumes of the amorphous and crystalline phases.[4] In addition, a kinetic explanation of the damping of the avalanche transition is possible in the case of nonstationary motion of the crystallization front.[24]

5. CONCLUSION

Summarizing the foregoing, we note that there exist at least three mechanisms of spontaneous crystallization of amorphous metallic films in the course of their condensation.

The first mechansim is the usual homogeneous formation and growth of a crystalline phase in the amorphous matrix of the sample. This mechanism is realized for all amorphous substances, without exception, when they are heated. Spontaneously, however, it arises only when the thickness of the amorphous region reaches during the condensation time a value corresponding to a crystallization temperature equal to the temperature of the condensed layer. Such a spontaneous crystallization can be observed if $T_{a \rightarrow k}$ of the given amorphous substance has a D dependence similar to that shown in Fig. 5 for Bi. The $a \rightarrow k$ transition can then proceed both with and without diffusion.

The second mechansim is undamped avalanche crystallization, which sets in spontaneously at the instant when the condensed film reaches the critical thickness. The realization of this mechanism is determined by the theoretically considered[22] conditions for the onset of thermal instability of the crystallization front. One of the most important conditions is apparently the feasibility of a diffusionless transition.

Besides the first two mechanisms, which are characterized by a stationary or quasistationary state of motion of the crystallization front, there exists also a third, connected with a nonstationary (damped) regime of avalanche crystallization.

The authors are grateful to V. A. Shklovskiĭ for helpful discussions and for an evaluation of the results.

1) The values of T_c for amorphous Bi and Ga films are 6 and 8.6 K, respectively.

2) The values of D_{cr} are 30–60 nm for Bi, 60–120 nm for Yb, and 180–230 nm for Ga.

3) The transition can begin at the center of the film and propagate toward its ends. In this the transition time is shorter by one-half than when the process propagates from one end of the film to the other (at equal velocity of the crystallization front).

4) In the equations cited, A is the heat of the $a \rightarrow k$ transition (J/m^3), C is the heat capacity per unit volume of the film, and T_f is the temperature of the crystallization front.

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