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Transition of Liquid Metals into Vapor in the Process of Pulse Heating by Current[1]

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The peculiarities of a liquid metal's nonequilibrium phase transition into vapor in the process of heating metallic wire samples by current impulse at the rate of 10^8 to 10^{10} $K \cdot s^{-1}$ are examined. A review of works by the article's author about a liquid metal's superheating up to the vicinity of the spinodal and explosive boiling under these conditions is given. In addition, the possibility of getting beyond the spinodal, realization of the unstable phase and its spinodal decay under the rate of heating of more than 10^{11} $K \cdot s^{-1}$ is analyzed.

KEY WORDS: explosive boiling; exploding wires; phase transition; pulse heating; spinodal; spinodal decay; superheated liquid.

1. INTRODUCTION

In the process of a metallic sample's heating by a microsecond current impulse with a density more than 10^6 $A \cdot cm^{-2}$ and energy adequate for the metal's evaporation, the phenomenon known as exploding wires is observed [1, 2]. Lebedev and Khaikin [3,4] examined this process for the first time by an oscillographic method. In the oscillograms of current and voltage on the wire, they registered the characteristic points defining the metal's phase boundaries in the process of its heating (Fig. 1). In particular, it was established by them that the first two points correspond to the beginning (t_1) and the end (t_2) of the wire's melting. After point t_3, a sharp rise of the liquid metal's electrical resistance begins. The exploded wire's photographs, obtained by high-speed photography using X-ray impulses, showed [5] that, after point t_3, intensive expansion of the wire begins. The

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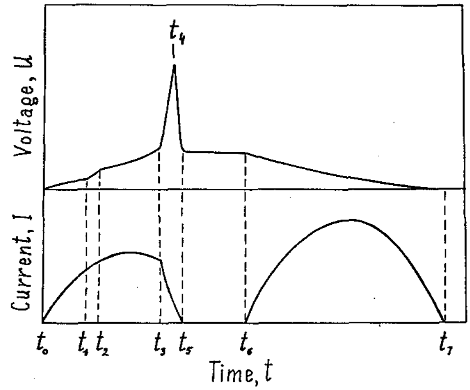

Fig. 1. Oscillogram traces of current, I, and voltage, U, for a copper sample (wire) in the process of electrical explosion in air.

initial stage of its destruction is fixed in the vicinity of the voltage peak point t_4. After point t_3, current in the conductor sharply decreases, practically to zero, as a result of which, pause of the current sets in (period t_5 to t_6). During the secondary current impulse, in the interval from t_6 to t_7, the wire material is in the plasma state. Temperature measurements by a high-speed optical pyrometry method showed [6–8] that the liquid metal's temperature in the vicinity of point t_3 is much higher than its normal temperature of boiling.

In the initial stage of the electrical explosion, transition of the liquid metal into the vapor phase plays the basic role. In the case of microsecond current impulses, the metal's rate of heating reaches values of 10^8 to $10^{10}\,\mathrm{K\cdot s^{-1}}$, and the current density reaches 10^6 to $10^7\,\mathrm{A\cdot cm^{-2}}$. So this transition has a sharply expressed nonequilibrium character. As the evaporation rate and the usual heterogeneous boiling rate are relatively small under these conditions, the liquid metal should be superheated above the phase equilibrium point until another, more intensive transition of the liquid into the vapor phase begins. Such a mechanism is explosive boiling, in which the homogeneous formation and growth of vapor bubbles, occurring in density fluctuations in the superheated liquid, take place. The concept of the liquid metal's superheating and explosive boiling (phase explosion) under the conditions of electrical explosion was expressed by Chace [9] and developed in the works of Martynyuk et al. [10–13], and Seydel et al. [6]. Further development of this concept is given in this work.

2. THERMODYNAMICS AND KINETICS OF THE LIQUID–VAPOR PHASE TRANSITION

Teaching about thermodynamic equilibrium and the stability of phases underlies the phase transition theory [14]. The phase equilibrium line (binodal) is defined by the equality of the Gibbs free energy for the liquid and vapor phases:

$$G_1(P, T) = G_2(P, T) \tag{1}$$

Thermodynamic stability of the phase is characterized by positive values of the derivatives (stability coefficients):

$$-\left(\frac{\partial P}{\partial V}\right)_T > 0, \qquad \left(\frac{\partial T}{\partial S}\right)_P = \frac{T}{c_p} > 0 \tag{2}$$

where T is the temperature, P is the pressure, V is the volume, S is the entropy, and c_p is the isobaric heat capacity. The phase stability boundary (spinodal) is defined by the vanishing of the stability coefficients:

$$\left(\frac{\partial P}{\partial V}\right)_T = 0, \qquad \text{or} \qquad \left(\frac{\partial T}{\partial S}\right)_P = \frac{T}{c_\mathrm{p}} = 0 \tag{3}$$

For the calculation of the spinodal we use the generalized van der Waals equation

$$P = \frac{RT}{V-b} - \frac{a}{V^n} \tag{4}$$

examined in Ref. 15. Here, R is the gas constant, a, b, and n are constants calculated through experimental data. From the equation of state (EOS), Eq. (4), it follows that parameter b is equal to the molar volume V_0 of the condensed phase at $T = 0$, $P = 0$:

$$b = V_0 \tag{5}$$

The sublimation heat Λ_0 of this phase at $T = 0$ is given by

$$\Lambda_0 = \int_{V_0}^{\infty} \frac{a}{V^n}\, dV = \frac{1}{n-1} \frac{a}{V_0^{n-1}} \tag{6}$$

therefore,

$$a = (n-1)\, \Lambda_0 V_0^{n-1} \tag{7}$$

At the critical point we have

$$\left(\frac{\partial P}{\partial V}\right)_T=0, \qquad \left(\frac{\partial^2 P}{\partial V^2}\right)_T=0 \tag{8}$$

Applying these conditions to the EOS, Eq. (4), produces two equations in V_c and T_c. Eliminating T_c between them gives the critical volume:

$$V_c=\frac{n+1}{n-1}V_0 \tag{9}$$

Back substitution in the two equations then gives the critical temperature:

$$T_c=\frac{4A_0}{R}\frac{n(n-1)^n}{(n+1)^{n+1}} \tag{10}$$

Substitution of V_c and T_c in the EOS gives the critical pressure:

$$P_c=\frac{A_0}{V_0}\frac{(n-1)^{n+2}}{(n+1)^{n+1}} \tag{11}$$

The critical compessibility ratio $Z_c=P_cV_c/RT_c$ is given by

$$Z_c=\frac{n^2-1}{4n} \tag{12}$$

Using the formulae for parameters a, b, T_c, P_c, and V_c, we may write the EOS, Eq. (4), in reduced variables, $\tau=T/T_c$, $\pi=P/P_c$, $\phi=V/V_c$:

$$\pi=\frac{\delta_0^2-1}{\delta_0}\frac{\tau}{\varphi-1/\delta_0}-\frac{\delta_0}{\varphi^n} \tag{13}$$

where $\delta_0=V_c/V_0=(n+1)/(n-1)$. Equations (4) and (13) described well the thermophysical properties of alkali metals (Cs, K, R*b*) with the value $n=1.46$. For the spinodal, we then want

$$\left(\frac{\partial P}{\partial V}\right)_T=0 \quad \text{or} \quad \left(\frac{\partial \pi}{\partial \varphi}\right)_\tau=0 \tag{14}$$

We thus obtain the equation of the spinodal in τ, φ coordinates

$$\tau=\left(\frac{\delta_0\varphi-1}{\delta_0-1}\right)^2\frac{1}{\varphi^{n+1}} \tag{15}$$

Substitution of Eq. (15) in the EOS, Eq. (13), then gives the equation of the spinodal in π, φ coordinates

$$\pi = \frac{n(\delta_0 \varphi - 1)}{\varphi^{n+1}} - \frac{\delta_0}{\varphi^n} \tag{16}$$

Equations (15) and (16) may be solved parametrically with ϕ ranging as a parameter between 0.593 and 1.00, with $n = 1.46$ to give the spinodal in π, τ coordinates. The phase diagram in π, τ coordinates is shown in Fig. 2 for alkali metals. In this figure, 1 is the binodal, according to experimental data for cesium [16], and 2 is the spinodal for the liquid phase, calculated according to Eqs. (15) and (16) with $n = 1.46$. The binodal 1 and the spinodal 2 merge at the critical point K. To the left of the binodal is the region of stable liquid. The region of metastable (superheated) liquid is located between the binodal and the spinodal. According to Eqs. (15) and (16), under zero pressure, the liquid phase's superheating limit (temperature at the spinodal) is equal to $\tau_{\mathrm{so}} = T_{\mathrm{so}}/T_{\mathrm{c}} = 0.902$. The volume of the liquid at this point is equal to $\varphi_{\mathrm{so}} = V_{\mathrm{so}}/V_{\mathrm{c}} = 0.593$. Under a pressure increase, the temperature and volume on the spinodal increase up to their values at the critical point. Line 3 is shown in Fig. 2. At the intersection of this line anomalous change in the stable liquid metal's properties is

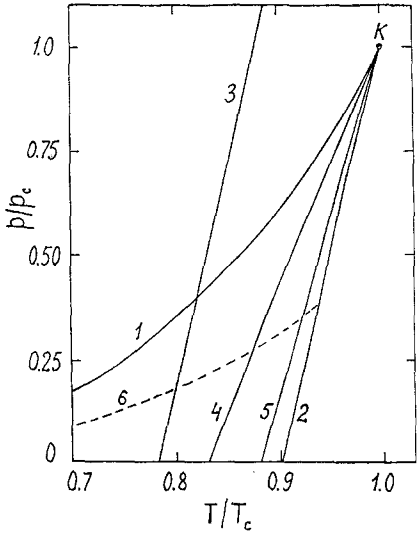

Fig. 2. Pressure–temperature phase diagram of a metal in the liquid–vapor transition region.

observed. Line 3 is the locus of points where the onset of rapid increase occurs for the specific resistance for mercury, the heat capacity of cesium, and the specific volume for cesium and rubidium [13]. It is defined by

$$\tau_3 = \frac{T_3}{T_c} = 0.095\left(\frac{P}{P_c}\right) + 0.78 \tag{17}$$

Line 3 is extrapolated to the region of metastable states. Such an extrapolation's validity is confirmed by the experimental data for properties of stable and superheated liquids [17]. Anomalous change of the metastable liquid's properties to the right of the line 3 is defined by a drastic decrease in its thermodynamic stability while approaching the spinodal 2.

Vapor bubbles can arise only in superheated liquid. Superheating is necessary for reducing the work of critical bubble formation. In the usual process of heterogeneous boiling, bubbles form at the ready-made centers (bubbles of dissolved gas in the liquid, suspended hard particles, admixture). As the work of formation of such bubbles is small, heterogeneous boiling takes place under small superheating. The concentration of ready-made centers in liquid is comparatively small, so the intensity of such boiling is small. Under a sufficiently high rate of heating, the part of the liquid evaporating through its free surface and through bubbles which formed at ready-made centers, is not significant. As a result, the great bulk of the liquid is superheated much higher than the binodal points.

Under higher superheating, homogeneous formation of vapor bubbles is possible at any point of the liquid volume, induced by density fluctuations. According to the classical theory developed by Volmer, Becker, Doring, Frenkel, and Zeldovich, for a stationary process, the frequency of homogeneous formation of vapor bubbles at temperature T is equal to [18]

$$J_0 = nB\exp\left(-\frac{\Delta G_k}{kT}\right) \tag{18}$$

where n is the number of particles of liquid in unit volume; k is the Boltzmann constant; ΔG_k is the work of formation of a critical bubble, which is expressed by the approximate formula

$$\Delta G_k = \frac{16\pi\sigma^3}{3[(P_b - P)(1 - D_2/D_1)]^2} \tag{19}$$

in which σ is the surface tension of the liquid; P is the pressure in the liquid; P_b is the pressure on the binodal at temperature T; and D_1 and D_2 are the densities of the liquid and vapor on the binodal.

Value B in Eq. (18) is a function which weakly depends on temperature and pressure, compared to the exponential term. According to a theory of Kagan [19], for liquids with lower viscosities (for example, for liquid metals), this value is equal to

$$B=\beta\sqrt{\frac{2\sigma N_0}{\pi\mu}} \tag{20}$$

where β is the coefficient of evaporation, N_0 is Avogadro's number, and μ is the molar mass of the liquid.

Equations (18) to (20) express drastic dependence of J_0 on T. Results of calculations using these formulae (with $\beta=1$) for cesium are presented in Fig. 2. On line 4, the nucleation frequency $J_0=1\ \text{cm}^{-3}\cdot\text{s}^{-1}$, and on line 5, $J_0=10^{26}\ \text{cm}^{-3}\cdot\text{s}^{-1}$. The second value of the frequency corresponds to the conditions of electrical explosion, when metal is transformed into vapor within the time of 10^{-7} s by the mechanism of homogeneous nucleation of vapor bubbles. Line 5 is located nearer to the spinodal 2. The position of line 4 shows that under zero pressure, homogeneous nucleation practically starts under superheating of the liquid to the temperature 0.84 T_c; under further superheating, to a temperature of 0.88 T_c, the frequency of nucleation increases by a factor of 10^{26}.

These calculations thus show that the region of intensive homogeneous nucleation of vapor in superheated liquid metal is located in a narrow temperature interval, resulting in explosive boiling. This region is located to the right of line 3. As a result, it can be assumed that the explosively boiling liquid metal has anomalous properties: higher values of heat capacity, compressibility, and coefficient of volumetric expansion.

3. CONDITIONS FOR THE REALIZATION OF MAXIMUM SUPERHEATING AND EXPLOSIVE BOILING OF LIQUID METALS

In the process of the wire's heating by current impulse, there are a number of factors which can lead to its destruction before the liquid metal is superheated to the vicinity of the spinodal. Evaporation of the metal and development of different disturbances (instabilities) of the wire's form relate to these factors. Study of these processes gives the possibility of minimizing their impact in experiments with superheating of liquid metals.

From the theory of the kinetics of the liquid's evaporation [20], the maximum rate of evaporation with $\beta=1$ may be expressed by the formula

$$U=\frac{1}{2}a_s\exp\left(-\frac{\Lambda}{RT}\right) \tag{21}$$

where a_s is the sound velocity in the liquid, T is the temperature of its surface, Λ is the molar heat of evaporation, and R is the gas constant. Proceeding from Eq. (21), for a liquid wire rate of heating $E = dT/dt$, the condition for which evaporation from its surface will be insignificant up to temperature T_3 on line 3 (Fig. 2), is obtained in [21]

$$E \geqslant E_m = \frac{2U_3RT_3(T_3 - T_2)}{xr_0\Lambda} \tag{22}$$

where U_3 is the rate of evaporation at temperature T_3 according to (11), T_2 is the melting temperature, r_0 is the wire's radius, and x is the maximum allowed fraction of the metal evaporating in the process of heating from temperature T_2 to T_3.

Apart from the evaporation from the wire's surface, the evaporation inside the heterogeneous vapor bubbles should be taken into account. Accurate account of such evaporation is impossible owing to the lack of knowledge of the concentration of centers in which these bubbles originate. In any case, estimates show [6, 10, 11] that the evaporation of metal inside these bubbles is not more than the evaporation from the surface of the liquid.

Under heating of the liquid wire with radius r_0 by current I_0, the wire is compressed by magnetic pressure, whose mean value over the cross section is equal to

$$\langle P_m \rangle = \frac{\mu_0}{8\pi^2}\frac{I_0^2}{r_0^2} \tag{23}$$

where $\mu_0 = 4\pi \times 10^{-7}\ \mathrm{H \cdot m^{-1}}$.

At the points of the wire where the radius is less, the magnetic pressure is greater. This is the reason for the development of magnetohydrodynamic (MHD) instabilities of the wire's form. From the theory of this process, it follows that [22] in the case of the form's smaller changes, the time dependence of the disturbance's amplitude Δr is expressed by the formula

$$\Delta r = \Delta r_0 \exp\left(\frac{t}{\tau_m}\right) \tag{24}$$

where Δr_0 is the initial amplitude, and τ_m is the time constant of MHD instabilities' development:

$$\tau_m = \frac{2}{i}\sqrt{\frac{D}{\mu_0}} \tag{25}$$

where i is the mean current density in the wire, and D is the density of the metal. Proceeding from Eqs. (24) and (25) at $t=\tau_m$, the heating rate for which the wire's form does not change considerably while heating up to temperature T_3 was obtained:

$$E \geqslant E_m = \frac{\mu_0 c_p (T_3 - T_2)^2}{4\rho} \tag{26}$$

where c_p and ρ are the mean values of the specific heat capacity and the specific resistance of the metal in the temperature interval from T_2 to T_3.

The value of the minimum rate of heating E_m in Eqs. (22) and (26) is related to the corresponding values of the heating current's density i_m and to the time t_{23m} of the wire's heating from the melting point T_2 to temperature T_3, by the following expressions:

$$E_m c_p D t_{23m} = c_p D(T_3 - T_2) = i_m^2 \rho t_{23m} \tag{27}$$

Apart from MHD instabilities, the electrothermal instabilities becoming apparent in the development of temperature disturbances may also be the reason for the wire's destruction. For smaller periodic temperature changes along the wire's length, the amplitude of the temperature change ΔT changes with time by the law [23]

$$\Delta T = \Delta T_0 \exp\left(\frac{\alpha i^2}{c_p D} - \frac{4\pi^2 a}{\lambda^2}\right) t \tag{28}$$

where ΔT_0 is the initial amplitude, λ is the period of the disturbance, a is the thermal diffusivity of the metal, and $\alpha = d\rho/dT$ is the temperature coefficient of the metal's specific resistance. Temperature disturbances dissipate from the mechanism of heat conduction under the condition

$$\frac{\alpha i^2}{c_p D} < \frac{4\pi^2 a}{\lambda^2} \tag{29}$$

For liquid metals in the interval from T_2 to T_3, α is small. So under current densities of $i = 10^6$ to $10^7\ \mathrm{A \cdot cm^{-2}}$, $\lambda < 1$ mm, this condition is fulfilled. However, after point T_3, α drastically increases, which leads to violation of the condition given by Eq. (29) and to the destruction of the wire by electrothermal instability. Hence, temperature T_3 on line 3 (Fig. 2) is the maximum temperature up to which the wire's uniform heating by current is practically possible.

From Eqs. (22) and (26) it follows that for different metals with $x = 0.01$, $r_0 = 0.2$ mm, the minimum value of the heating rate E_m is located in the limits $E_m = (4 \text{ to } 10) \times 10^8 \text{ K} \cdot \text{s}^{-1}$. According to Eq. (17), for this value of E_m, we have $i_m = (2 \text{ to } 8) \times 10^6 \text{ A} \cdot \text{cm}^{-2}$ and $t_{23m} = (1 \text{ to } 20)\ \mu\text{s}$.

Knowledge of the path of the liquid metal's heating on the diagram of states (Fig. 2) is necessary for interpreting results of the experiments. While constructing this path in the high-temperature region, apart from external pressure, it is necessary to take into account the action of the scattering metal vapor's pressure on the wire surface. According to a calculation of Nite [24], under the scattering of monatomic vapor in vacuum, this pressure is equal to $P_r = 0.55\ P_b$, where P_b is the saturated vapor pressure for the temperature of the evaporating surface. Proceeding from this, it can be assumed with pulse heating of the wire in air that a thin subsurface layer of metal is under pressure $P = 0.55\ P_b + P_{at}$, where P_{at} is the atmospheric pressure. Inside the wire, magnetic pressure is acting in addition.

Line 6 for heating under $P = 0.55\ P_b + P_{at}$ is shown in Fig. 2. In the high-temperature region, $P < P_b$, so that heating proceeds into the region of superheated liquid. Line 6 intersects the line of anomaly 3 at the point $T_3 = 0.80 T_c$, and the line of intensive explosive boiling 5 at point $T_5 = 0.92 T_c$. In the process of the wire's explosion in air, under the observance of conditions given by Eqs. (22), (26), and (29), the temperature $T_3 = 0.80 T_c$ of the liquid metal's superheating in the diagram of states (Fig. 2) corresponds to the time moment t_3 in the oscillograms (Fig. 1). After point T_3, the condition given by Eq. (29) is violated and electrothermal instability is developed in the wire. Owing to this, explosive boiling takes place not through the whole volume of the wire, but in regions of development of electrothermal instability.

The above-stated interpretation of point t_3 (Fig. 1) differs from the interpretation given in our earlier works [10, 11], where this point was identified with the spinodal point, which was not fully correct. For this reason, our first estimates of the critical temperatures of metals from the enthalpy at point t_3 [25] were understated compared to the latest more accurate estimates [21].

4. ANOMALOUS ELECTRICAL EXPLOSION AND SPINODAL DECAY

Kvartskhava and others [26] measured the dependence of the relative resistance R/R_0 of copper wire on the introduced energy W at the rate of heating of more than $10^{10} \text{ K} \cdot \text{s}^{-1}$ and t_{23} less than 100 ns. Under such conditions at higher values of energy, the dependence of the wire's resistance on the rate of heating was discovered. The nonunique dependence of R/R_0 on W under such anomalous explosion was later confirmed in the experiments of Bennett and others [27] for copper and in the experiments of

Tucker [28] for gold. In the regime of anomalous electrical explosion, considerably more energy than the heat of sublimation Λ_0 is introduced to the metal. At this time, the liquid metal's enthalpy at point T_3 constitutes only 40 to 55% of Λ_0 [10].

The concept that anomalous electrical explosion is accompanied by entering beyond the spinodal into the region of the unstable phase and its spinodal decay was expressed by us [12]. For analyzing this concept, we examine the temperature T–entropy S diagram of states for a single-component substance (Fig. 3). In this diagram K is the critical point, curve kKl is the binodal, and curve mKn is the spinodal. The critical isobar P_c passes through the critical point K. For subcritical isobars $P < P_c$, the van der Waals loop *acedb* and also the path *aeb* for transitions of liquid into vapor are shown. For this isobar T_b is the temperature on the binodal (boiling), T_s is the temperature on the spinodal, and S_1 and S_2 are the entropies of the liquid and vapor, respectively, at the boiling point T_b. The region of existence of superheated liquid is located between the branch kK of the binodal and the branch mK of the spinodal. Between branch lK and branch nK is the region of supersaturated vapor. The binodal kKl bounds the region of two-phase states. For pressures $P > P_c$, the liquid–vapor transition takes place continuously without decay of the system into two phases.

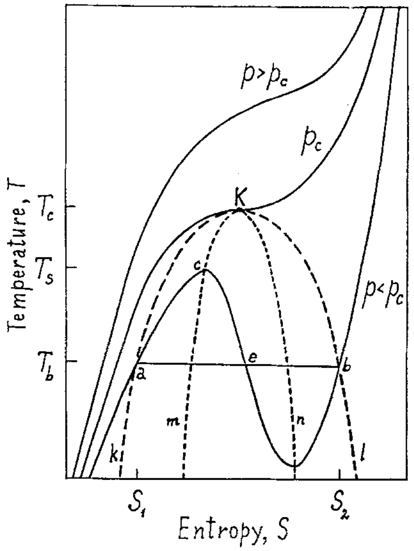

Fig. 3. Temperature–entropy phase diagram of a single component material in the liquid–vapor transition region.

The spinodal *mKn* bounds the existence region of unstable states, for which, in distinction from Eq. (2), stability coefficients are negative:

$$-\left(\frac{\partial P}{\partial V}\right)_T < 0; \qquad \left(\frac{\partial T}{\partial S}\right)_P = \frac{T}{c_p} < 0 \tag{30}$$

Zeldovich and Todes [29] for the first time indicated that under negative heat capacity c_p of the unstable phase in Eq. (30), its thermal diffusivity a also should be negative. It leads to the sign change in the heat transfer equation

$$\frac{\partial T}{\partial t} = a\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right) \tag{31}$$

Then from Eq. (31), with $a < 0$, solutions with an exponential increase in temperature difference are obtained. This means that an accidental local increase in the temperature in the unstable phase does not dissipate, but spontaneously increases. According to [29], the unstable phase decays into regions of size **I**, in which the temperature difference ΔT originates. The time of the phase's decay τ_s is equal to the time of increase Δt:

$$\tau_s = \frac{\mathbf{I}^2}{a} \tag{32}$$

The spinodal decay of the unstable phase is the nonactivation process of transition, owing to thermodynamic instability, into a continuous nonuniform system consisting of regions of dense and rarefied substance with large temperature difference. As the surface tension is zero for the unstable phase, these regions are not limited by surface partition. Under further development of spinodal decay, the system gets out of the region of unstable states (Fig. 3) and spinodal decay is replaced by the formation process of the new phase's bubbles, limited by surface partition. In this process, equalization of the temperature between the phases takes place. The process of the unstable phase's spinodal decay is experimentally observed in metallic alloys and glasses [30].

In the process of the liquid's pulse heating with $P < P_c$ (Fig. 3), its superheating up to the vicinity of the spinodal curve takes place. Here, owing to the homogeneous formation of vapor bubbles, the liquids explosive boiling takes place. This process, however, is not established at once. From theory it follows that [18] after the liquid's instant superheating, the dependence of the nucleation frequency J on time t is expressed by the formula

$$J = J_0 \exp\left(-\frac{\tau_N}{t}\right) \tag{33}$$

where J_0 is the frequency for a stationary nucleation process, expressed by Eq. (18), and τ_N is the characteristic time of establishment of the stationary process. By theoretical estimates $\tau_N = 1$ to 10 ns [18].

For getting beyond the spinodal, the realization of the unstable phase and its spinodal decay in the process of the metal's pulse heating, it is necessary for the heating time Δt_s to the vicinity of the spinodal to be less than each of the characteristic times in Eqs. (32) and (33):

$$\Delta t_s < \tau_s, \qquad \Delta t_s < \tau_N \tag{34}$$

In the experiments with anomalous electrical explosion [26–28], these conditions seem to be fulfilled as time Δt_s is around 1 ns. Proof of spinodal decay in these processes may be the registration of multicharged ions and characteristic X-ray radiation from the region of anomalous electrical explosion. Ions and X-rays can be radiated from the high-temperature regions, originating in the process of the unstable phase's spinodal decay.

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