MECHANISM OF INITIATION OF ACOUSTIC EMISSION IN CRYSTALLIZATION AND MELTING OF A SUBSTANCE. II

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In [Inzh.–Fiz. Zh., **73**, No. 5, 1064–1072 (2000)], *the authors investigated one of the possible mechanisms of initiation of the acoustic effect of crystallization (melting) of substances, which was based on the assumption of a stepwise character of motion of the crystal–melt interface. In the present work, consideration is given to another mechanism of initiation of the acoustic effect of crystallization, which is based on the concept of possible enhancement of waves propagating in the liquid and in the crystal during the motion of their interface.*

In crystallization (melting) of liquids in limited volumes, systems of standing waves appear on each side of the interface, one of which is localized in the volume occupied with the liquid, while the other in the growing crystal. A distinguishing feature of these systems of waves is the fact that they are not related by the known conjugation conditions, i.e., by the requirement for the displacement and the stress at the interface to be equal. Thus, for example, in crystallization of water a thin layer of ice formed on the interface causes both phases to move apart in different directions as if the liquid and the crystal are wedged, i.e., the first condition of conjugation is violated. Whence it follows that in liquid crystallization in a tube, when the crystallization front moves along the tube, the crystal–liquid system must not be considered as a composite rod formed of heterogeneous materials.

Density disturbances emanating from the crystallization (melting) zone of the liquid propagate in different directions in both the liquid and the crystal. However, the character of propagation of these waves is observed to be significantly different.

If we restrict ourselves to a small time interval, then the thickness of the crystal H formed upon crystallization of the liquid in a tube is much smaller than the tube length $h(H \ll h)$. Therefore, for the period of time during which the disturbances propagate from the liquid-crystallization front to the rear wall of the tube and go back, a system of standing waves with frequencies multiple to the fundamental frequency $v_1 \approx a_1/2H$ succeeds in establishing itself in the crystal. By virtue of the smallness of H these waves are high-frequency ones; they exist as if independently of the waves propagating in the liquid.

Since all the time the crystal is in contact with the liquid, the oscillatory motion in it is imparted to the liquid. Thus, the forced oscillations caused by the high-frequency collisions of the crystal with the liquid surface are imposed on the forced oscillations of the liquid initiated by the slow motion of the crystal–liquid interface. Here, the oscillatory motion in the liquid and in the crystal are maintained due to the displacement of the crystallization front.

Naturally, the oscillatory motion of the liquid in turn acts as a periodic force under the action of which the crystal performs forced oscillations.

Acoustic waves initiated in the liquid and in the crystal will be considered using the liquid crystallization in a tube as an example [1].

We assume that both ends of the tube are rigidly sealed and that the internal pressure-induced change in the tube size can be neglected.
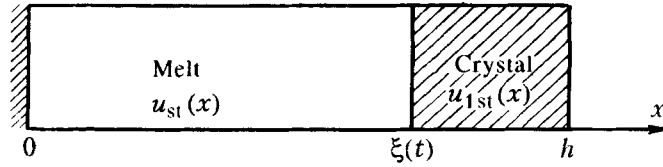
Fig. 1. Schematic of liquid crystallization in a tube.

As the crystallization front moves, the stresses in both the liquid and the crystal gradually increase – they undergo compression.

Figure 1 shows schematically the picture of water crystallization. Displacements $u_{st}(x)$ and $u_{1st}(x)$, called phase deformations, can be found by solving the equations of elasticity theory with the specifically formulated boundary conditions.

Let the crystallization front be displaced by $\delta\xi$ in the time dt. In this case, the liquid and solid phases undergo infinitesimal displacements $\delta u_{st}(x)$ and δu_{1st}:

$$\delta u_{st}(x) = \delta q x + \delta b, \quad 0 \leq x < \xi; \tag{1}$$

$$\delta u_{1st}(x) = \delta q_1 x + \delta b_1, \quad \xi \leq x < h, \tag{2}$$

where δq, δq_1, δb, and δb_1 are arbitrary constants which must be found from the boundary conditions. The first two of these conditions are of the form

$$u_{st}(0) = 0, \quad u_{1st}(h) = 0. \tag{3}$$

Now we will formulate the boundary conditions when $x = \xi(t)$. In this case, the functions $\xi(t)$ change so slowly that at each time the deformations u_{st} and u_{1st} are quasistatic.

At $x = \xi(t)$ the displacements of liquid and crystal particles occur in different directions. In our coordinate system $\delta u_{st}(\xi) < 0$, $\delta u_{1st}(\xi) > 0$, and $\delta\xi < 0$. With account for these inequalities we can write

$$\delta u_{st}(\xi) - \delta u_{1st}(\xi) = \beta\delta\xi. \tag{4}$$

Moreover, the stresses on both sides of the interface must be equal:

$$\frac{1}{K}\frac{d(\delta u_{st})}{dx}\bigg|_{x=\xi} = E_1\frac{d(\delta u_{1st})}{dx}\bigg|_{x=\xi}, \tag{5}$$

where

$$E_1 = \frac{1-\mu}{(1-2\mu)(1+\mu)}E.$$

Solutions of Eqs. (1) and (2) with boundary conditions (3)-(5) acquire the form

$$\delta u_{st}(x) = \frac{\beta K E_1 \chi}{K E_1 \xi - (h-\xi)}\delta\xi, \quad 0 \leq x < \xi; \tag{6}$$

$$\delta u_{1st}(x) = -\frac{\beta h(\chi+h)}{K E_1 \xi - (h-\xi)}\delta\xi, \quad \xi \leq x < h. \tag{7}$$

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To determine the total deformation of the system with the crystallization front displacing from $x = h$ to $x = \xi(t)$, it is necessary to integrate (6) and (7) with respect to ξ. After the corresponding simplifications we will arrive at

$$u_{st}(x) = -\beta (h - \xi) \frac{x}{h}, \quad 0 \leq x < \xi;$$ (8)

$$u_{1st}(x) = \left(\frac{\beta}{KE}\right)\left(1 - \frac{\xi}{h}\right)(x + h), \quad \xi < x \leq h.$$ (9)

Oscillatory motions of the crystal caused by the motion of the crystallization front of the liquid are described by the wave equation

$$\frac{\partial^2 u_1}{\partial t^2} = a_1^2 \frac{\partial^3 u_1}{dx^2 dt}, \quad \xi < x < h.$$ (10)

In this case, the initial and boundary conditions have the form

$$u_1(x, 0) = u_{1t}(x, 0) = 0;$$ (11)

$$u_1(\xi, t) = u_{1st}(\xi) = \frac{\beta}{KE_1 h}(h^2 - \xi^2);$$ (12)

$$u_1(h) = 0,$$ (13)

where $u_{1t} = \partial u_1/\partial t$.

A solution of problem (10)-(13) can be written as

$$u_1(x, t) = u_{1st}(x) - \frac{2\beta H}{\pi KE_1 h} \sum_{m=1}^{\infty} \frac{(-1)^m}{m} J_m \sin \frac{\pi m x}{H},$$ (14)

where

$$J_m = \int_0^t \dot{\xi}(\tau) \cos \omega_m (t - \tau) \, d\tau; \quad \omega_m = \frac{\pi m a_1}{H},$$

the variable $m = 1, 2, 3$.

Crystal oscillations described by equality (14) create a force on the surface of the liquid adjacent to it:

$$F(t) = \frac{2\beta}{Kh} \sum_{m=1}^{\infty} (-1)^m J_m \cos \frac{\pi m t}{H}.$$ (15)

In the particular case where the velocity of the crystallization front is prescribed by the formula

$$\dot{\xi}(t) = \alpha \left(\frac{\chi_1}{t}\right)^{1/2},$$

equality (15) can be written in the form

$$F(t) = \frac{\beta \alpha H}{Kh} \left(\frac{2\chi_1}{\pi a_1 H} \right)^{1/2} \sum_{m=1}^{\infty} \frac{(-1)^m}{m} (\cos \omega_m t + \sin \omega_m t) \cos \frac{\pi m \xi}{H}, \tag{16}$$

where $\alpha = \sqrt{c(T_0 - T_s)/2L}$; $T_0 - T_s$ is the temperature difference of the crystallization front and the crystal surface.

Acoustic waves initiated in the liquid consist of the sum of two waves: $u(x, t) = \vartheta(x, t) + W(x, t)$, where $\vartheta(x, t)$ describes the waves initiated by the motion of the liquid–crystal interface, while $W(x, t)$ describes the forced oscillations of the liquid caused by the force $F(T)$.

The function $\vartheta(x, t)$ is a solution of the wave equation

$$\frac{d^2 \vartheta}{dt^2} = a^2 \frac{d^2 \vartheta}{dx^2} + \nu \frac{d^3 \vartheta}{dx^2 dt}, \quad 0 < x < \xi,$$

and satisfies the following conditions: $\vartheta(x, 0) = \vartheta_t(x, 0) = 0$; $\vartheta(0, t) = 0$; $\vartheta(\xi, t) = u_{st}(\xi)$. This solution can be written in the form

$$\vartheta(x, t) = u_{st}(x) - \frac{\beta \alpha K E_1 \xi (2\pi \chi_1)^{1/2}}{\pi h (1 + KE)} \sum_{n=1}^{\infty} \frac{\omega_n^{*2}}{\omega_n^2 \sqrt{\omega_n^*}} \exp(-\gamma_n t) \left[\left(1 + \frac{\gamma_n^2}{\omega_n^{*2}} \right) \sin \omega_n^* t + \right.$$

$$\left. + \left(1 - \frac{2\gamma_n}{\omega_n^*} - \frac{\gamma_n^2}{\omega_n^{*2}} \right) \cos \omega_n^* t \right] \sin \frac{\pi n x}{\xi}, \quad 0 < x < \xi,$$

where (17)

$$\gamma_n = \frac{\nu \omega_n^2}{2 a^2}; \quad \omega_n^* = \sqrt{\omega_n^2 - \gamma_n^2}; \quad \omega_n = \frac{\pi n a}{\xi}, \quad n = 1, 2, 3, \ldots .$$

The function $W(x, t)$ is a solution of the inhomogeneous wave equation

$$\frac{\partial^2 W}{\partial t^2} = a^2 \frac{\partial^2 W}{\partial x^2} + \frac{F(t)}{\rho} \delta(x - \xi), \quad 0 < x < \xi, \tag{18}$$

with the homogeneous initial and boundary conditions

$$W(x, t) = W_t(x, 0) = 0, \quad W(0, t) = W(\xi, t) = 0.$$

If we neglect the influence of the liquid viscosity, then a solution of Eq. (18) can be written in the form

$$W(x, t) = \frac{2\beta \alpha H a^2}{\xi h} \left(\frac{2\chi_1}{\pi a_1 H} \right)^{1/2} \sum_{n=1}^{\infty} \frac{1}{\omega_n} \sum_{m=1}^{\infty} \frac{1}{m} \left[\frac{\omega_m' \sin \omega_n t - \omega_n \sin \omega_m' t}{\omega_m'^2 - \omega_n^2} - \right.$$

$$\left. - \frac{\omega_n}{\omega_m'^2 - \omega_n^2} (\cos \omega_m' t - \cos \omega_n t) \right] \cos \frac{\pi n \xi}{H} \sin \frac{\pi (2n + 1) x}{2\xi}. \tag{19}$$

The main contribution to $W(x, t)$ is made by the terms for which $\omega_m' \approx \omega_n$. In this case, $\omega_m' \sin \omega_n t$ in (19) can approximately be replaced by $\omega_n \sin \omega_n t$. As a result, we obtain
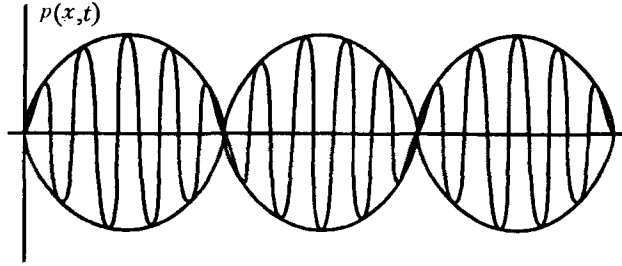
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Fig. 2. Change in the pressure amplitude near resonance.

$$W(x, t) = \frac{4\beta\alpha Ha^2}{\xi h} \left(\frac{\chi_1}{\pi a_1 H}\right)^{1/2} \sum_{n=1}^{\infty} \frac{1}{\omega_n} \sum_{m=1}^{\infty} \frac{1}{\omega'_m - \omega_n} \sin\left(\omega'_m t - \frac{\pi}{4}\right) \times$$

$$\times \sin\frac{\omega'_m - \omega_n}{2} t \cos\frac{\pi m \xi}{H} \sin\frac{\pi(2n+1)x}{2\xi}. \tag{20}$$

The corresponding pressure $p(x, t)$ is determined by the formula

$$p(x, t) = \frac{4\beta\alpha Ha}{\xi h} (\rho a^2) \left(\frac{\chi_1}{\pi a_1 H}\right)^{1/2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{1}{m(\omega'_m - \omega_n)} \times$$

$$\times \sin\left(\omega'_m t - \frac{\pi}{4}\right) \sin\frac{\omega'_m - \omega_n}{2} \cos\frac{\pi m \xi}{H} \cos\frac{\pi(2n+1)x}{2\xi}, \quad 0 \le x \le \xi. \tag{21}$$

Thus, near the resonance frequencies $\omega'_m = \omega_n$ high-frequency oscillations with amplitude modulation occur. Figure 2 shows schematically the character of change in p near resonance.

With allowance for viscosity, the solution of Eq. (18) acquires the form

$$W(x, t) = \frac{2\beta\alpha Ha^2}{h\xi} \left(\frac{2\chi_1}{\pi a_1 H}\right)^{1/2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{1}{m\omega_n} J_{mn}(t)(\cos\omega'_m\tau + \sin\omega'_m\tau) \times$$

$$\times \sin\left(\omega'_m t - \frac{\pi}{4}\right) \sin\left(\frac{\omega'_m - \omega_n}{2}\right) t \cos\frac{\pi m \xi}{H} \cos\frac{\pi(2n+1)x}{2\xi}, \quad 0 \le x \le \xi,$$

where

$$J_{mn} = \int_0^t \exp[-\gamma_n(t-\tau)] \sin\omega_n(t-\tau)\, d\tau. \tag{22}$$

After opening the integral (22), expression (21) acquires the form

$$W(x, t) = A \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{d_{mn}}{m\omega_n} [\cos(\omega'_m t +$$

$$+\delta_{mn}) + \exp(-\gamma_n t)(A_{mn}\cos\omega_n t + B_{mn}\sin\omega_n t)] \times$$

$$\times \cos \frac{\pi m \xi}{H} \sin \frac{\pi (2n + 1) x}{2h}, \quad 0 \leq x \leq \xi, \tag{23}$$

where

$$d_{mn} = [(1 - \lambda_{mn}^2)^2 + 4\lambda_{mn} h_n^2]^{-1/2}; \quad \lambda_{mn} = \frac{\omega_m'}{\omega_n}; \quad h_n = \frac{\gamma_n}{\omega_n};$$

$$A_{mn} = (1 - \lambda_{mn})^2 + h_n^2; \quad B_{mn} = \frac{\sqrt{2}}{4} (1 - \lambda_{mn}^2)^2 (h_n + \lambda_{mn}) \frac{\omega_n'}{\omega_n};$$

$$\delta_{mn} = \arctan \frac{1 - \lambda_{mn}^2 + 2h_n \lambda_{mn}}{1 - \lambda_{mn}^2 - 2h_n \lambda_{mn}}; \quad A = \frac{8\beta\alpha H}{K\rho\xi h} \left(\frac{\gamma_1}{\pi a H} \right)^{1/2}.$$

The second term in the square brackets of the right-hand side of equality (23) is negligibly small as compared to the first term describing the forced oscillations. The pressure that corresponds to the latter is

$$p(x, t) = -\frac{A}{Ka} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{d_{mn}}{m\omega_n} \cos \frac{\pi m \xi}{H} \cos (\omega_m' t + \delta_{mn}) \cos \frac{\pi (2n + 1) x}{2\xi},$$

$$0 < x < \xi. \tag{24}$$

The coefficients d_{mn} in the oscillation theory are termed the dynamics coefficients. They show how many times the amplitude of forced oscillations of the corresponding modes under resonance conditions exceeds the static displacements of particles under the action of a constant force equal in magnitude to the amplitude of the corresponding acting force.

A plot of d_{mn} against λ_{mn} at resonance values of h_n is given, for example, in [2].

Under resonance conditions,

$$d_{mn} = \frac{a^2}{v\omega_n' \sqrt{1 - h_n^2}} \approx \frac{a^2}{v\omega_m'},$$

while the pressure amplitude is

$$p_{\max} = \frac{8\beta\alpha a H^{5/2}}{\pi^2 \xi v \sqrt{h m^2}} \left(\frac{2\chi_1}{\pi a_1 h} \right)^{1/2} (\rho a^2) \left(\frac{a}{a_1} \right) \cos \frac{\pi m \xi}{H}, \quad m = 1, 2, \ldots . \tag{25}$$

In the case of water crystallization, $\beta = 0.1$, $a = 1.5 \cdot 10^3$ m/sec, $a_1 = 4 \cdot 10^3$ m/sec, and $\chi_1 = 1.15 \cdot 10^{-4}$ m^2/sec. At $\xi = h = 0.1$ m, $H = 2 \cdot 10^{-3}$ m, and $m = 1$, the quantity $p_{\max}$ is $(10^3 - 10^4)$ Pa at a velocity of the crystallization front of about 1 mm/min. The oscillation frequency of the fundamental mode in this case is 10^6 Hz.

As is seen from formula (25), the dependence of $p_{\max}$ on the crystal thickness is determined by the product $H^{5/2} \cos (\pi m \xi / H)$, which fluctuates with change in H.

We consider in more detail the behavior of the function $p(x, t)$. At the initial instant of formation of the solid phase the frequencies v_m' are great; however the corresponding modes of oscillations are virtually momentarily damped due to the liquid viscosity. After the cessation of the initial period of formation of a stable layer of the crystal the process of its growth stabilizes. The oscillation frequencies ω_m' gradually decrease. The main contribution to $p_{\max}$ is made by the mode with the frequency $\omega_1 (m = 1)$.

As the solid phase grows, the frequencies ω_n and ω'_m are displaced to the left. Here, the velocity of displacement of the point ω'_m is substantially larger than that of displacement of the points ω_n and ω_{n-1} and so on and the point ω'_m will obligatorily be brought into coincidence with the nearest point ω_n. Whence it follows that resonance inevitably occurs.The dependence of p and t is the same as the dependence of the dynamics coefficient a_{ln} on h_n. Here, amplitude modulation takes place. The next resonance event occurs already at coincidence of the points ω'_1 and ω_{n-1} and so on.

Now we will find the time interval between two successive resonance events at the fundamental frequency ω'_1.

At the instant t, $\omega'_1 = \pi a/H(t)$, while after Δt we already have $\omega'_1(t + \Delta t) = \pi a_1/H(t + \Delta t)$. The difference of these two frequencies must be equal to the frequency $\omega_1 = \pi a/\xi$, i.e.,

$$\frac{1}{H(t)} - \frac{1}{H(t + \Delta t)} = \frac{a}{a_1\xi} \approx \frac{a}{a_1 h}.$$

Taking into account that $H(t) = 2\alpha\sqrt{\chi t}$, we obtain

$$\frac{\Delta t}{t} \approx \frac{2aH(t)}{a_1 h}, \quad \Delta t \sim t^{3/2}.$$

Since the ratio $aH/a_1 h \ll 1$, in the initial period of liquid crystallization the emissions will often be repeated.

Thus, due to the influence of high-frequency oscillations of the crystal that is directly adjacent to the crystallizing liquid, in the latter, high-frequency oscillations with amplitude modulation occur; the fundamental frequencies of these oscillations and the amplitude values of the pressure are, approximately, the same as those measured in [3]. Therefore, there are grounds to suggest that the above theory sufficiently well reflects the basic features of the process of initiation of the acoustic effect of crystallization of substances.

However, there are two aspects in the model described above which, physically, are contradictory:

1) the velocity of the crystallization front at $t = 0$ turns into infinity, which leads to artificial overestimation of the peak pressure amplitudes to $(10^4 - 10^3)$ Pa;

2) the natural frequencies of crystal oscillations $\omega'_m = \pi a m/H$ can take as large a value as desired at small H.

Both these circumstances are a consequence of the idealization of a mathematical formulation of the problem. With more rigorous account for the crystallization conditions of the liquid and the influence of the gate at the right-hand end of the tube on the liquid-crystallization process, both aspects, as shown by calculations, are easily eliminated: the peak values of p obtained by formula (25) decrease, approximately, by an order of magnitude, and instead of H a quantity that is approximately equal to the sum $H + H_1$ appears.

NOTATION

ν, kinematic viscosity of the liquid; $u_{st}(x)$ and $u_{1st}(x)$, statistical deformations of the liquid and the crystal, respectively; $x = \xi(t)$, crystal–melt interface; H, crystal thickness; h, length of the crystallization cell; a_1, velocity of sound in the crystal; a, velocity of sound in the liquid; β, volume expansion coefficient of the liquid under crystallization; K, compressibility coefficient of the liquid; E_1, elasticity modulus of the plane wave; E, Young's modulus; μ, Poisson coefficient; $\xi(t)$, velocity of the crystallization front; T_0, temperature of stable equilibrium of the liquid–crystal phases; T_s, surface temperature of the growing crystal; L, specific heat of liquid crystallization; χ_1, thermal diffusivity of ice; ω'_m, circular frequency of oscillations of the n-th mode of the solid phase; ω_n, circular frequency of oscillations of the n-th mode of the liquid phase; γ_n, absorption coefficient of waves of the frequency ν_n; p, pressure in the acoustic wave; t, time; τ, delay time; H_1, gate thickness; c, heat capacity of the crystal; δ, infinitesimal increment; ν_n, oscillation frequency of the n-th mode.

Subscripts: n and m, numbering of the quantities; s, surface; points above a symbol, differentiation with respect to time.

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