IMPURITY DISTRIBUTIONS IN CRYSTALS DUE TO GROWTH-RATE FLUCTUATIONS

S. I. Alad'ev

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Balance equations for the second moments of the velocity and temperature fluctuations are used to examine the effects of turbulent natural convection on the uniformity of impurity distributions in crystals.

A crystal grown from the melt with fluctuations in growth rate R shows a periodic impurity distribution [1, 2]. There may be various reasons for growth-rate fluctuation, one of which is turbulent natural convection. Here we consider the concentration profile for the crystal formed in that case.

The maximum deviation of the impurity concentration in the solid phase from the mean corresponding to growth at a constant rate R_0 is [2] dependent on the amplitude and frequency of the fluctuations in R, and if those fluctuations are due to turbulent convection, then the effects are related to the extent of the temperature and speed fluctuations in the melt. We restrict consideration to a crystal growing from a melt by directional crystallization in a horizontal boat. We assume that the gradients in the average temperature in the transverse directions are negligible by comparison with that in the growth direction. Then the balance equations for the second moments of the velocity and temperature fluctuations are as follows on the assumption that the density of the melt is a function of temperature only [3-5]:

$$\frac{k}{l}E^{1/2}\langle v_x' v_y'\rangle = 0,\quad \frac{k}{l}E^{1/2}\langle v_x' v_z'\rangle + \beta g\langle v_x' T'\rangle = 0, \tag{1}$$

$$\frac{k}{l}E^{1/2}\langle v_y' v_z'\rangle + \beta g\langle v_y' T'\rangle = 0,$$

$$\frac{k}{2l}E^{1/2}\left(\langle v_x'^2\rangle - \frac{2}{3}E\right) + \frac{b}{3}\frac{E^{3/2}}{l} = 0,$$

$$\frac{k}{2l}E^{1/2}\left(\langle v_y'^2\rangle - \frac{2}{3}E\right) + \frac{b}{3}\frac{E^{3/2}}{l} = 0,$$

$$\frac{k}{2l}E^{1/2}\left(\langle v_z'^2\rangle - \frac{2}{3}E\right) + \frac{b}{3}\frac{E^{3/2}}{l} + \beta g\langle v_z' T'\rangle = 0,$$

$$\langle v_x'^2\rangle\frac{\partial T}{\partial x} + \frac{k_T}{l}E^{1/2}\langle v_x' T'\rangle = 0,$$

$$\langle v_x' v_y'\rangle\frac{\partial T}{\partial x} + \frac{k_T}{l}E^{1/2}\langle v_y' T'\rangle = 0,$$

$$\langle v_x' v_z'\rangle\frac{\partial T}{\partial x} + \beta g\langle T'^2\rangle + \frac{k_T}{l}E^{1/2}\langle v_z' T'\rangle = 0,$$

$$\langle v_x' T'\rangle\frac{\partial T}{\partial x} + \frac{b_T}{l}E^{1/2}\langle T'^2\rangle = 0,$$

$$E = \frac{1}{2}\left(\langle v_x'^2\rangle + \langle v_y'^2\rangle + \langle v_z'^2\rangle\right).$$

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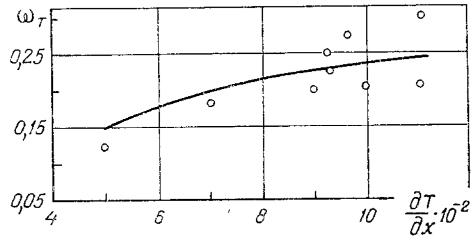

Fig. 1. Dependence of the fluctuation frequency ω_T (sec^{-1}) on the axial temperature gradient $\partial T/\partial x$ (K/m); curve from theory, points from experiment [7].

In writing (1) we have assumed that

$$\nu \langle \frac{\partial v_i'}{\partial x_k} \frac{\partial v_j'}{\partial x_k} \rangle = \frac{b}{3} \frac{E^{3/2}}{l} \delta_{ij},$$

$$a \langle \frac{\partial T'}{\partial x_k} \frac{\partial T'}{\partial x_k} \rangle = \frac{b_T}{l} E^{1/2} \langle T'^2 \rangle ,$$

$$- \langle \frac{p'}{\rho}\left(\frac{\partial v_i'}{\partial x_j} + \frac{\partial v_j'}{\partial x_i}\right) \rangle = \frac{k}{l} E^{1/2}\left(\langle v_i' v_j' \rangle - \frac{2}{3} E\delta_{ij}\right),$$

$$- \langle \frac{p'}{\rho} \frac{\partial T'}{\partial x_j} \rangle = \frac{k_T}{l} E^{1/2} \langle v_j' T' \rangle .$$

The x axis is perpendicular to the front, while the z axis is vertically upward. We solve (1) to get

$$\langle T'^2 \rangle = \frac{\alpha_1}{b_T k_T} l^2 \left(\frac{\partial T}{\partial x}\right)^2, \quad \alpha_1 = \frac{2}{3}(1 - b/k),$$
$$E = \alpha_E l^2 \beta g \frac{\partial T}{\partial x}, \quad \alpha_E = \frac{1}{k_T} \sqrt{\frac{\alpha_1}{b_T b}(1 + b_T/k)} . \tag{2}$$

The scale l of the turbulence is $l = \frac{2}{d_0} \int_0^{d_0/2} 0.4\, t dt = 0.1\, d_0$, where $d_0 = \sqrt{y_0 z_0/\pi}$, while y_0 and z_0 are the transverse dimensions of the boat. We put $b/k = 0.125$, $b_T/k = 0.2$, $k_T/k = 0.9$, $k = 1.12$ on the basis of data on wall turbulence to get that $\alpha_1 = 0.583$ and $\alpha_E = 4.69$. Then we have the following expressions for the turbulent energy E and the amplitude of the temperature fluctuations ε_T, which is $\varepsilon_T = \sqrt{\langle T'^2 \rangle}$:

$$\varepsilon_T = 0.16\, d_0 (\partial T/\partial x), \qquad E = 0.047\, d_0^2 \beta g (\partial T/\partial x). \tag{3}$$

The frequency of the turbulent fluctuations containing the most energy is [6] given by

$$\omega_T = \sqrt{E}/d_0 = 0.22 \sqrt{\beta g (\partial T/\partial x)}. \tag{4}$$

Figure 1 compares calculations from (4) with experimental data [7] on the growth of tin crystals in horizontal boats under conditions of turbulent natural convection. The bulk expansion coefficient for tin was taken as $\beta = 0.95 \cdot 10^{-4}$ 1/K.

The relative amplitude in the growth rate $\varepsilon = (R_0 - R)/R_0$ can be found from the Stefan condition $q^* = q + r\rho R$, where q^* and q are the axial densities of the heat fluxes at the front from the solid and liquid, respectively. With $q = q_0 + \lambda \varepsilon_T / l$, q_0, $q^* = \text{const}$, we have

$$\varepsilon = \frac{\lambda}{r\rho R_0 d_0} \varepsilon_T. \tag{5}$$

In accordance with [2], the maximum deviation of the impurity concentration φ in the solid phase from the mean is determined by the reduced frequency of the growth-rate fluctuations $\omega = \omega_T D/R_0^2$; for $\omega \gg 1$, the expression for φ, becomes

$$\varphi = \frac{c_m - c_0}{c_0} = \sqrt{\frac{5}{8}}(1 - k_0)\frac{\varepsilon}{\sqrt{\omega}}, \tag{6}$$

and for $\omega \ll 1$

$$\varphi = (1 - k_0)\frac{\sqrt{\omega^4 + k_0\omega^2}}{k_0^2 + \omega^2(2k_0 + 1)}\varepsilon. \tag{7}$$

On substituting (4) and (5) into (6) and (7), we get

$$\varphi = 0.27(1 - k_0)\frac{\lambda}{r\rho\sqrt{D}}(g\beta)^{-1/4}\left(\frac{\partial T}{\partial x}\right)^{3/4},\ \omega \gg 1, \tag{6a}$$

$$\varphi = 0.035(1 - k_0)\frac{\lambda D}{r\rho R_0^3 k_0^{3/2}}\sqrt{g\beta}\left(\frac{\partial T}{\partial x}\right)^{3/2},\ \omega \ll 1,$$
$$\varphi = 0.16(1 - k_0)\frac{\lambda}{r\rho R_0}\frac{\partial T}{\partial x},\ \omega \ll 1,\ k_0 \to 0. \tag{7a}$$

These expressions show that the inhomogeneity in the impurity distribution increases in all cases with the temperature gradient, because the amplitude of the turbulent temperature fluctuations increases with $\partial T/\partial x$, and the same therefore applies to the amplitude of the growth-rate fluctuations.

The frequency dependence of φ varies with ω; for example, $\varphi \sim \omega^{-1/2}$ for $\omega \gg 1$, whereas $\varphi \propto \omega$ for $\omega \ll 1$ according to (7), while for $\omega \ll 1$ and $k_0 \to 0$, the degree of inhomogeneity ceases to be dependent on frequency, which in turn means that the inhomogeneity in the distribution decreases as the frequency of the turbulent fluctuations increases for $\omega \gg 1$, while it increases on the other hand for $\omega \ll 1$. The only exception is represented by the case $\omega \ll 1$ and $k_0 \to 0$.

In conclusion we note that one can obtain a more uniform impurity distribution over the length for $\omega \gg 1$ by conducting the process at high g, i.e., in a centrifuge. On the other hand, using a centrifuge for $\omega \ll 1$ would increase φ.

NOTATION

a, thermal diffusivity; d_0, characteristic size; c_0, c_m, mean and maximum impurity concentrations; D, diffusion coefficient; k, k_T, b, b_T, constants; k_0, impurity partition coefficient; ℓ, turbulence scale; g, gravitational acceleration; p, pressure; r, latent heat of fusion; R, R_0, instantaneous and mean growth rates; T, melt temperature; v_x, v_y, v_z, projections of the melting rate vector on the coordinate axes x, y, z; β, volume expansion coefficient; λ, ρ, ν, thermal conductivity, density, and kinematic viscosity of the melt; ε_T, ω_T, amplitude and frequency of turbulent pulsations; ε, ω, reduced amplitude and frequency of fluctuations in growth rate; φ, degree of nonuniformity; ', pulsation.

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