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USE OF STANDING SURFACE WAVES AND OF A STATIONARY MAGNETIC FIELD IN GROWING CRYSTALS BY THE FLOATING-ZONE METHOD IN WEIGHTLESSNESS

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A numerical investigation of the flow and heat and mass transfer during the growth of crystals by the floating-zone method in weightlessness using standing surface waves, created by applied vibration, and a stationary magnetic field has been carried out. With crystallization of silicon serving as the illustrative example, it is shown how, by selecting optimal parameters of the standing surface waves and of the magnetic field, one can appreciably (by severalfold) reduce the micro- and macrosegregation of the phosphorus impurity and grow a crystal with a uniformity that exceeds the best specimens obtained on the ground by the Czochralski method.

Introduction. The development of microelectronics imposes higher and higher demands on the quality of single crystals: they must have a perfect crystal structure and highly homogeneous composition on micro- and macroscales. The Czochralski method, which is the main industrial method of obtaining single crystals, is not always convenient for creating promising compounds of the group A^3B^5 elements (AsGa, InP, GaP) or especially pure crystals, when fouling from a crucible is inadmissible. Among alternative methods of growing crystals the method of crucible-free zone melting (the floating zone method) is of most interest in this connection. However, the macroscopic and microscopic homogeneity of the crystals grown by this method is higher than that obtained by the Czochralski method [1]. While the latter method has been investigated and refined for a long time, the floating-zone method has been preserved virtually in its original form. A small number of experiments were carried out to study thermocapillary convection and the process of growing crystals by the floating-zone method under the conditions of microgravity [2–5], with the emphasis being placed on studying the oscillating regime of thermocapillary convection and associated microsegregation of impurities. To decrease the intensity of thermocapillary convection and temperature oscillations in experiments on rocket probes [2, 3] the surface of the specimen was covered in local zones by a film of silicon oxide, which remained solid when the specimen was melted. In the experiment aboard a space vehicle [4] a constant axial magnetic field was used for the purpose. A substantial improvement in the quality of crystals has not been attained in both cases. When a magnetic field was used, the negative effect of the increase in the radial microsegregation of impurity occurring on shift of the zone of intense thermocapillary convection to the free cylindrical surface of the liquid was not taken into account. This negative effect of a constant magnetic field was subsequently noted in the experiments in [5, 6]. The use of a rotating magnetic field led to a decrease in the microsegregation of impurity in the experiment on the ground [7]. As is shown in [8], the high speed of rotation of the liquid created by a magnetic field removes the small and spatially inhomogeneous azimuthal velocity component of the thermocapillary flow. The reason for the appearance of the oscillating regime of thermocapillary convection is thereby eliminated. It was also established in [8] that under the microgravity conditions it is possible to attain also a decrease in the macrosegregation of impurity by using a rotating magnetic field of needed intensity.

Investigations of standing surface waves (SSW) produced by vibration showed [9–11] that such waves may serve as a means of controlling the processes of heat and mass transfer. In [12], it has been demonstrated for the first time how, with the aid of SSW, one can efficiently regulate the distribution of impurities in crystals grown by the floating-zone method under microgravity conditions. In [13], the stability of thermocapillary convection under the ac-

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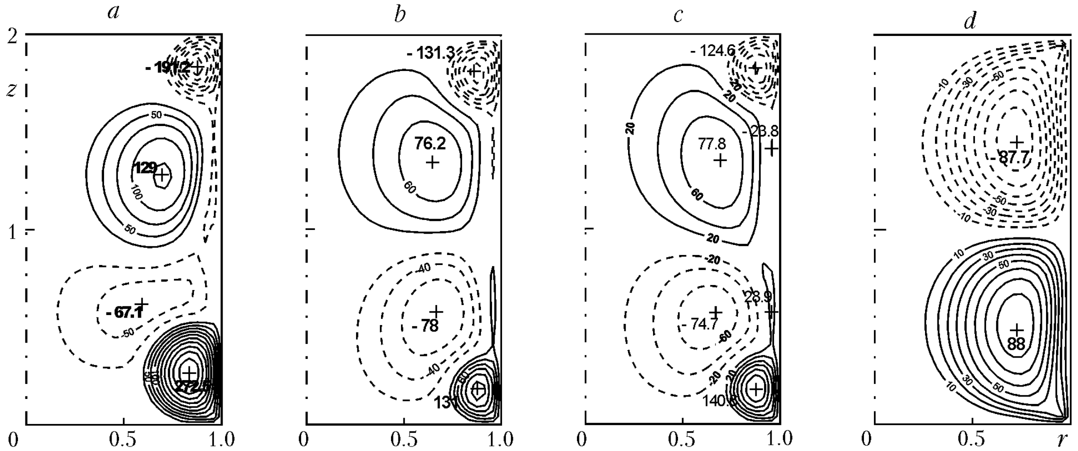


Fig. 1. Structures of flow (stream function fields: solid lines, clockwise flow; dashed lines, anticlockwise flow) with the laminar thermocapillary convection being combined with standing surface waves (a and d) at $n = 2$ and for an oscillating thermocapillary convection without an SSW (b and c); a) Ma/Pr = $6{\cdot}10^4$, $n = 2$, $\delta = 0.003$, $\Omega = 7.8{\cdot}10^4$; b and c) Ma/Pr = $1.2{\cdot}10^5$; d) Ma/Pr = $3.9{\cdot}10^4$, $n = 2$, $\delta = 0.0045$, $\Omega = 7.546{\cdot}10^4$.

tion of SSW of different configurations was investigated and effects were revealed that can be used to decrease the micro- and macrosegregation of impurities. The present work is devoted to determination of the optimal parameters of SSW and magnetic field which allow one to considerably reduce the micro- and macrosegregation of impurities in growing crystals by the floating-zone method under weightlessness conditions.

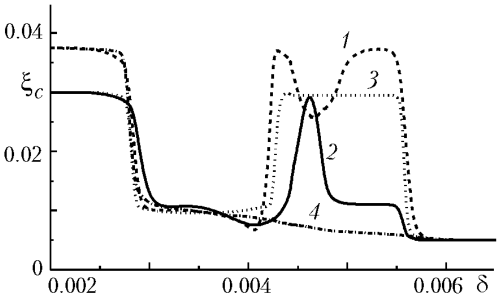
Statement of the Problem and Computational Technique. We will consider the process of liquid crystallization by the floating-zone method. Since the crystallization front moves at a rate of several orders of magnitude smaller than the characteristic velocity of the flow of liquid, mathematical simulation is carried out without displacement of the molten zone over a specimen. Exchange of impurity between the melt and the crystal is modeled by a constant value of the impurity concentration and by the mass balance at the crystallization boundary provided there is a good mixing of the liquid. The system of equations and boundary conditions, the difference approximation of differential equations, and the technique of numerical solution of the equations are described in [13]. To allow for the effect of the static axial magnetic field the Lorentz force is added into the equation of motion in the projection onto the radius of the liquid cylinder, and this equation takes the following form:

$$\frac{\partial u}{\partial \tau} + u\frac{\partial u}{\partial r} + v\frac{\partial u}{\partial z} = -\frac{\partial p}{\partial r} + \left(\frac{\partial^2 u}{\partial r^2} + \frac{1}{r}\frac{\partial u}{\partial r} - \frac{u}{r^2} + \frac{\partial^2 u}{\partial z^2}\right) - \mathrm{Ha}^2 u .$$

An expression for the Lorentz volumetric force was obtained in a noninductive approximation. The approximation applies to liquids that easily conduct an electric current and to the dimensions of a computational domain that are typical of the considered technological process, as is shown in [8]. The remaining equations and boundary conditions are the same as those in [13].

In all of the cases cited below, we consider as an example the process of crystallization of silicon with an admixture of phosphorus, Si(P). The following dimensionless parameters are used which describe the processes of heat and mass transfer in growing this crystal: Pr = 0.023, Sc = 5, k_0 = 0.35, $\mathrm{Re}_{\mathrm{cr}}$ = 0.1, and L/R = 2. The distribution of the heat flux on the cylindrical liquid surface is given, just as in [13], in the form of an exponent with two parameters which describe well the experimentally obtained distribution of temperature on the surface of the liquid zone [2, 3]. In contrast to [13], the oscillation of the standing wave was switched-on from the start of calculation. At that very moment the process of crystal growth began.

Results of Calculations. As a result of the action of the thermocapillary effect and oscillations of the free surface in the form of a standing wave, from the beginning of the process the structure of flow is being formed,



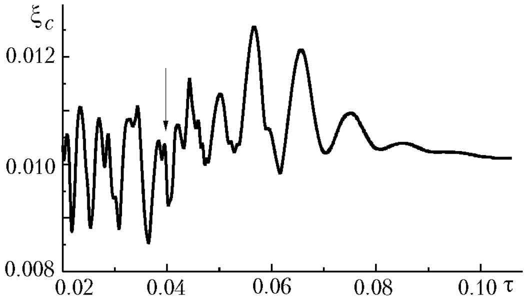
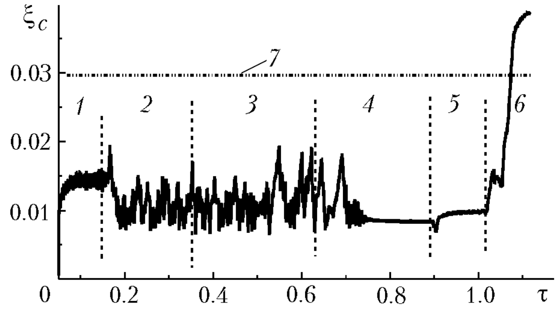


Fig. 2. Relative radial inhomogeneity of impurity vs. the SSW amplitude: 1) Ma/Pr = $3.9{\cdot}10^4$, n = 2, Ω = 75,460; 2) $6.6{\cdot}10^4$, 2, and 75,460; 3) $3.9{\cdot}10^4$, 2, and 2500; 4) $3.9{\cdot}10^4$, 4, and 2500.

Fig. 3. Relative radial inhomogeneity of impurity on exposure to a standing wave and magnetic field in the regime of initial laminar convection. Ma/Pr = $6{\cdot}10^4$; the arrow indicates the moment of the switching on of the magnetic field (Ha = 5).

which differs from that considered in [13] at the same parameters. Figure 1a demonstrates the instantaneous structure (the field of the stream function) of oscillating convection for the case where the parameter Ma/Pr corresponds to a laminar regime of convection and SSW is operative. This structure differs from the usual oscillating thermocapillary convection (without SSW) shown in Fig. 1b and c for antiphase time instants. In studying the stability of thermocapillary convection an unusual phenomenon was revealed in [13] consisting of the fact that in the case of mutual symmetry of the structure of thermocapillary convection and the configuration of an SSW, when n = 2, a zone of practically stable flow is formed in the region of the oscillating flow (a specific "resonance"). It is interesting that in the calculation procedure used in the present work this effect is also observed. Figure 1d demonstrates the structure of flow for the zone of "resonance," when the same flow as that without an SSW is formed. In the zone of "resonance," the radial macrosegregation of an impurity is the same or close to the values obtained in the absence of an SSW. This is clearly shown in Fig. 2: for δ = 0.0042–0.0057 the local maxima of the function $\xi_c(\delta)$ attain the values (curves 1, 2, 3) that were obtained when an SSW did not affect the flow (at $\delta \le 0.002$). For n = 4 (Fig. 2, curve 4) the effect of "resonance" is not observed even in a degenerate form [13], and the regime of weak oscillations smoothly goes over into a regime of a turbulent flow at $\delta \approx 0.0057$. It should also be noted that with this calculation technique the boundaries of transition from a stable flow to an oscillating one and from the latter to a turbulent one are displaced to the side of smaller values of the standing wave amplitude δ, as compared to [13]. The shape of the "resonance" region depends substantially on the frequency of oscillations of an SSW and to a lesser degree on the value of the parameter Ma/Pr that characterizes the intensity of a thermocapillary flow. For other values of the number of periods of an SSW, except for n = 1, the dependences of the parameter ξ_c on δ resemble that shown in Fig. 2 for n = 4 (curve 4). For n = 1 with increasing δ, transition from a laminar regime of flow to a turbulent one occurs bypassing the zone of weak fluctuations of parameters, as described in [13]. For $n \ge 2$ the behavior of the parameters of the liquid in the zone of weak oscillations ($\delta \approx$ 0.003–0.004) practically does not depend on Ma/Pr, Ω, and n (see Fig. 2). This remark also relates to the frequencies of fluctuations of parameters in the liquid. The sharp decrease in the value of the parameter of radial macrosegregation of an impurity ξ_c and small amplitudes of the fluctuations of parameters in the zone of weak oscillations create prerequisites for investigating the possibility of decreasing the macro- and microsegregation of impurity when an SSW is used alone or in combination with a static magnetic field. To demonstrate the way in which the optimum parameters of an SSW and of a magnetic field are selected, all the cycles of the regimes are combined in a single computational process, which makes it possible to demonstrate the dynamics of the process and duration of the transition regime.

We will consider variants in which the parameter Ma/Pr corresponds to a laminar thermocapillary convection. In Fig. 3 the SSW begins to operate from the beginning of calculation, and by the time instant τ = 0.04 an oscillating regime of flow and impurity distribution sets in. By this time the average value of the parameter of radial inhomogeneity of the impurity is ξ_c = 0.00984, whereas the amplitudes and frequencies of the fluctuations of the liquid pa-



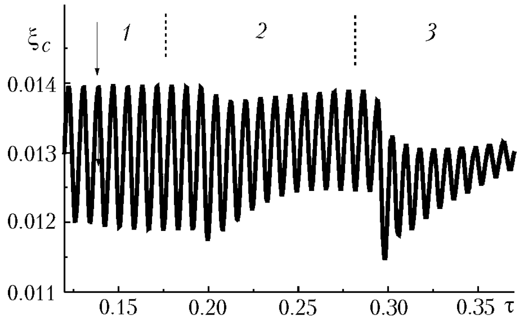


Fig. 4. Relative radial inhomogeneity of impurity on exposure to a standing wave and magnetic field in the regime of initial laminar convection: 1) Ha = 0; 2) 5; 3) 10; 4) 15; 5) 20; 6) 30 7) without a standing wave and magnetic field.

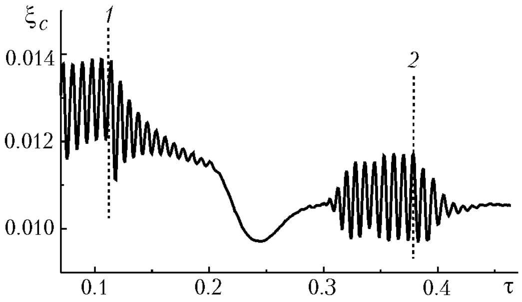
Fig. 5. Relative radial inhomogeneity of impurity on exposure to a standing wave and magnetic field in the regime of initial oscillating convection (Ma/Pr = $9.5 \cdot 10^4$); after the arrow) a standing wave (n = 2, Ω = 2500) with a different amplitude: 1) δ = 0.0026; 2) 0.004; 3) 0.006.

rameters do not change; thus, the amplitude of the fluctuation of ξ_c (microsegregation of the impurity) is equal to 12.8%. After this the magnetic field is switched on, and the calculation is continued until a new steady state ξ_c sets in. At the end of the process considered ($\tau \approx 0.11$) the average value of ξ_c is equal to 0.0105, which is somewhat higher than at the time τ = 0.04, but the fluctuations virtually disappeared (the amplitude of the fluctuations is ~0.1%). The value of the macrosegregation of the impurity obtained at the end of the process considered corresponds to a developed oscillation regime of thermocapillary convection (at Ma/Pr $\approx 1.2 \cdot 10^5$). We note that in a laminar regime of flow at Ma/Pr = $6 \cdot 10^4$, without an SSW and a magnetic field, the parameter ξ_c is constant and equal to 0.032. Thus, in this case a more than threefold decrease in the radial macrosegregation of the impurity at very small microsegregation was obtained.

It has been shown above that the liquid parameters in the regime of weak oscillations under the action of controlling SSWs depend little on the frequency of vibrations of a standing wave (Fig. 2). This allows one to carry out calculations at lower values of Ω than is required by the considered parameters of SSW, which will considerably accelerate the process of computations due to a greater step in time $\Delta\tau$. The process of determining the optimal intensity of a magnetic field (the Hartmann number) is shown in Fig. 4. The calculation was performed at Ma/Pr = $7.2 \cdot 10^4$ and the following parameters of an SSW: n = 4, δ = 0.0032, and $\Omega = 2.5 \cdot 10^3$. Without an SSW and a magnetic field ξ_c = 0.0295 (straight line 7); in the presence of an SSW without a magnetic field the average value of ξ_c is equal to 0.0144 and the amplitude of fluctuations is about 10%. At Ha = 15 the average value of ξ_c is 0.00835 and the amplitude of fluctuations is ~1%, whereas at Ha = 20 the average value of ξ_c is 0.00976 and the amplitude is ~1.6%. Finally, at Ha = 30 the fluctuations disappear but ξ_c increases up to 0.04. Thus, an optimal regime in this case is that with Ha = 15.

The change in the amplitude of fluctuations of the parameter ξ_c in an oscillating regime of thermocapillary convection in the case of a stepwise increase in the amplitude of an applied SSW δ from 0.0026 to 0.006 is shown in Fig. 5. Without an SSW the average value of ξ_c is 0.013 and the amplitude of fluctuations is 7.27%. At a standing wave with δ = 0.006 the average value of ξ_c decreases slightly, to 0.01295, but the amplitude of fluctuations decreases to 1.47%, i.e., almost fivefold. The dimensionless frequency of the initial fluctuations F does not change here and is equal to 121.6. Note that at a very small amplitude of an SSW (δ = 0.0026), the amplitude of fluctuations ξ_c is even higher (8%) than without an SSW.

Figure 6 shows how the parameter ξ_c changes after successive switching on of the SSW and of the magnetic field. Before the switching on of the SSW the flow corresponds to the beginning of the oscillating convection (Ma/Pr = $9.6 \cdot 10^4$). In this regime of flow the average value of ξ_c is 0.0126 at a fluctuation amplitude of about 7.6%. After the start of the action of the SSW with the parameters n = 2, δ = 0.006, and $\Omega = 7.8 \cdot 10^4$ a transition regime



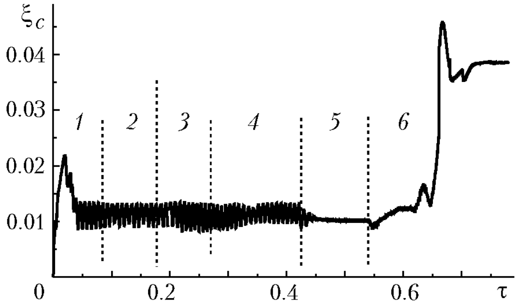


Fig. 6. Relative radial inhomogeneity of impurity on exposure to a standing wave and magnetic field in the regime of initial oscillating convection (an SSW is acting after 1, after 2 — a magnetic field). $Ma/Pr = 9.52 \cdot 10^4$.

Fig. 7. Relative radial inhomogeneity of impurity on exposure to a standing wave ($n = 2$, $\Omega = 2500$) and magnetic field in the regime of initial oscillating convection ($Ma/Pr = 1.2 \cdot 10^5$); 1) without a standing wave and magnetic field; 2) $\delta = 0.0032$, $Ha = 0$; 3) 0.005 and 0; 4) 0.005 and 5; 5) 0.005 and 10; 6) 0.005 and 15.

begins, at the end of which the average value of ξ_c decreases to 0.01074 and the amplitude of fluctuations increases up to about 9%. Thereafter the magnetic field ($Ha = 5$) is switched on, as a result of the action of which the fluctuations damp out rapidly (the residual microsegregation of the impurity is ~0.1%) and the average value of ξ_c is established at a level of about 0.01055. In this example, the decrease in the macrosegregation is slight, about 16% of the initial level, but the principal effect is the practical liquidation of the microsegregation of the impurity.

Figure 7 demonstrates the process of determination of the optimal parameters of an SSW and of a magnetic field. The initial regime of liquid flow corresponds to a developed oscillating thermocapillary convection close to a turbulent regime ($Ma/Pr = 1.2 \cdot 10^5$). In this regime of flow the average value of ξ_c is 0.0107 and the amplitude of fluctuations is about 21.5%. The amplitude of an SSW with $n = 2$ and $\Omega = 2.5 \cdot 10^3$ first takes the value 0.0032 and then 0.005. In the latter case the average value of ξ_c is set at a level of 0.0125 at an amplitude of fluctuations equal to about 19.4%. After this a magnetic field with a stepwise increase in the Hartmann number (5, 10, and 15) is switched on. In the optimal regime ($\delta = 0.005$ and $Ha = 10$) the average value and the amplitude of fluctuations of ξ_c are equal to 0.01013 and ~1%, respectively. At $Ha = 15$ the fluctuations of the parameter ξ_c decrease to 0.4%, but its average value increases sharply up to about 0.0385.

Conclusions. The investigation carried out shows that the use of an SSW as well as of an SSW in combination with a static magnetic field allows one to obtain a considerable decrease in the micro- and macrosegregation of impurity in crystals grown by the floating-zone method under microgravity conditions. An especially great decrease in the macrosegregation of an impurity is obtained when the initial condition is characterized by the small intensity of thermocapillary convection in a laminar regime of flow. A positive result can be obtained when the optimal parameters of the SSW (the number of the periods of a standing wave over the liquid zone length and the amplitude) and of the magnetic field are selected exactly, since otherwise there is a danger of occurrence in the zone of a practically stable flow, the "resonance" zone for $n = 2$, or of slip into the zone of a developed turbulent regime. The overshooting of the optimal intensity of a magnetic field may lead to a catastrophic increase in the macrosegregation of the impurity. An optimal regime can be chosen for a specific material and conditions of growing a crystal by the method of mathematical simulation. Thus, the program of calculation of flow and heat and mass transfer must be an integral part of a technological setup. In the figures presented the attainment of an optimal segregation of impurity is shown together with a transition process of flow development. In the actual process of growing a crystal this transient period should be held prior to growing a crystal. In this case, the period of the development of the impurity concentration profile takes a time not longer than $\tau = 0.1$. The length of the crystal in the regime of establishment of the concentration field determined from $Re_{cr}\tau$, i.e; $Re_{cr} = 0.1$ is not longer than 0.01 of the crystal radius. For practical application of the results obtained it is important to determine exactly the relationship between the vibration parameters and the standing surface waves generated by it. This kind of relationship was determined computationally in [11, 12]. Experimental verification of the relations obtained is needed. The experiment "Ivolga" included in 1998 into the program of scientific experiment on the Russian segment of the International Space Station was devoted to that problem.

Under the joint effect of thermocapillary and thermal gravitational convection in on-the-ground conditions the structure of liquid flow is nonsymmetrical relative to the plane passing through the middle of the liquid column ($z = L/2$). Such a structure of flow arises when an SSW acts on thermocapillary convection in weightlessness (see Fig. 1a). This permits us to hope that the positive effect of using an SSW separately or in combination with a static magnetic field can be obtained also in growing crystals by the floating-zone method on the ground.

NOTATION

B, magnetic induction, V/m; b, standing wave amplitude, m; c, impurity concentration in a liquid, kg/m^3; $C = c/c_0$, dimensionless concentration of impurity; D, diffusion coefficient of impurity in a liquid, m^2/sec; f, frequency, Hz; F, dimensionless frequency; $\mathrm{Ha} = BR(\lambda/\rho\nu)^{1/2}$, Hartmann number; L, length of a liquid column, m; k_0, equilibrium coefficient of impurity distribution; $\mathrm{Ma} = -(\partial\sigma/\partial T)R\Delta T/(\rho\nu\chi)$, Marangoni number; n, number of periods of a standing wave over length L; $\mathrm{Pr} = \nu/\chi$, Prandtl number; p, dimensionless pressure; R, radius of a nonperturbed liquid column, m; $\mathrm{Re}_{\mathrm{cr}} = v_{\mathrm{cr}}R/\nu$, dimensionless rate of growth of a crystal; $\mathrm{Sc} = \nu/D$, Schmidt number; T, temperature, K; t, time, sec; u and v, dimensionless velocities along the axes r and z; χ, thermal diffusivity m^2/sec; $\delta = b/R$, dimensionless amplitude of a standing wave; $\Delta T = T_{\max} - T_0$, characteristic temperature difference in the system, K; $\Delta C_{\mathrm{s}} = C_{\mathrm{s,max}} - C_{\mathrm{s,min}}$, radial difference of concentration in the liquid; λ, specific electrical conductivity, $1/(\Omega\cdot\mathrm{m})$; ν, kinematic viscosity, m^2/sec; ρ, density, kg/m^3; σ, surface tension, N/m; ω, circular frequency in a standing wave, sec^{-1}; $\Omega = \omega R^2\nu^{-1}$, dimensionless circular frequency; $\xi_c = \Delta C_{\mathrm{s}}/\bar{C}_{\mathrm{s}}$, dimensionless relative radial difference of the impurity concentration; $\tau = t\nu/R^2$, dimensionless time. Subscripts: 0, initial value; cr, crystallization; max and min, maximum and minimum values of the parameters; s, crystallization boundary. Superscript: overbar, mean value at the crystallization boundary.

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