

High frequency vibration and natural convection in Bridgman-scheme crystal growth

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Abstract—The problem of natural heat convection suppression in the earth gravity with the help of vibration is studied. Geometries of Bridgman vertical and Bridgman horizontal crystal growth configurations are considered. The eficiency of fluid motion damping under magnetic field and field of vibrations is compared. In the horizontal Bridgman geometry on earth the damping is more efficient with the magnetic field for semiconductor melts with relatively high dectroconductivity. For the vertical Bridgman geometry with horizontal vibration, a significant braking of fluid movement near the bottom of the crucible could be achieved (particularly in the region near the solid-liquid interface where conditions for purely diffusive mass transfer growth can be met).

1. INTRODUCTION

THE INFLUENCE of vibration on convective heat motion in a liquid was discussed earlier in the literature [l-71. In the high-frequency approach, the equations of vibro-convective motion were obtained first in ref. [1] and in the closed Form with boundary conditions by Gershuni and Zuhovitsky in ref. [2]. A few problems with simple geometries were studied including the analysis of stability of the fluid motion in the presence of vibration 12-51. In these analyses the generalized Navier-Stokes equations have been reduced because of the boundary conditions and solved separately. The simplification of the geometry gave an opportunity to apply analytical methods of analysis. The main conclusion presented in ref. [3] is that applying vibrations with specially chosen axes suppresses the convective motion in the volume of a liquid with nonuniform temperature field.

In the present work the problem of interaction between natural convection and vibration is studied for the horizontal and vertical (relatively to the gravity vector) Bridgman crystal growth geometries. The problem of convective motion suppression is of paramount importance due to the change of mass transfer mechanism from diffusive-convective to diffusive when the fluid motion is reduced, like in microgravity conditions [8, 91. Concentration of dopants incorporated in a single-crystal by a diffusion controlled mass transfer in the liquid has a smooth distribution without segregation. The vertical Bridgman crystal growth method turns out to be the best one in this sense since the small radial temperature gradients generate a low level of convective motion. Nevertheless it is very difficult to achieve the diffusive mass transfer

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in the earth gravity with the low values of crystal growth rates used (\sim 3.0 × 10⁻⁷ m s⁻¹ and diameters about a few 10^{-3} m). The well-known use of magnetic fields in crystal growth in order to suppress the convective motion [lo] is limited to melts with relatively high values of electroconductivity. Estimations presented in ref. [IO] show that diffusive mass transfer could be achieved for the materials from group A_3B_5 , with crystal diameters about 10^{-2} m with low temperature gradients and growth rates only under magnetic fields exceeding 20 T which are hardly accessible with modern industrial technology. At the same time the vibration method does not sharply depend on the particular properties of the melt and could be used cojointly with a magnetic field.

All calculations in this work were performed for 2-D geometries and therefore could be used only as zerothorder approach for the technological applications on crystal growth and estimations of vibration influence on convective motion.

2. **EQUATIONS**

As was done in refs. [2-3] we shall restrict ourselves to the case of vibrations with high frequency harmonic oscillations. In the limiting case of high frequencies the equations of the average physical values (for the period of oscillation) will take the form [3] :

$$
\frac{\partial \mathbf{V}}{\partial t} + (\mathbf{V}\nabla)\mathbf{V} = -\frac{1}{\rho}\nabla P + v\Delta \mathbf{V}
$$

+ $g\beta T\overline{\gamma} + \varepsilon(\mathbf{W}\nabla)(T\mathbf{n} - \mathbf{W})$

$$
\frac{\partial T}{\partial t} + \mathbf{V}\nabla T = \chi\Delta T
$$

div $\mathbf{V} = 0$; div $\mathbf{W} = 0$;
rot $\mathbf{W} = \nabla T \times \mathbf{n}$. (1)

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NOMENCLATURE

It should be said that system (1) is valid for the con- 3. **HORIZONTAL BRIDGMAN** ditions **CONFIGURATION**

$$
L/c \ll \tau \ll \min{(L^2/v, L^2/\chi)}.
$$

For semiconductor melts like GaSb or 1nSb with $L \sim 10^{-2}$ m the last condition will take the form :

$$
2.0 \times 10^{-6} \text{ s} \ll \tau \ll \min(3.0 \times 10^{3} \text{ s}, 20 \text{ s}) = 20 \text{ s}
$$

or in terms of frequencies :

$$
5.0 \times 10^{-2} \text{ Hz} \ll \omega \ll 10^5 \text{ Hz}.
$$

The system of equations (1) could be rewritten in nondimensional form as follows :

$$
\frac{\partial \mathbf{U}}{\partial t} + (\mathbf{U}\nabla)\mathbf{U} = -\nabla P + Gr^{-1/2}\Delta \mathbf{U} + \theta
$$

$$
+ Ra_v Gr^{-1} Pr^{-1}((\theta \mathbf{n} - \nabla \phi)\nabla)\nabla \phi
$$

$$
\frac{\partial \theta}{\partial t} + \mathbf{U}\nabla \theta = Pr^{-1} Gr^{-1/2}\Delta \theta
$$

$$
\operatorname{div} \mathbf{U} = 0; \quad \Delta \phi = \operatorname{div} (\theta \mathbf{n}). \tag{2}
$$

The thermo-vibrational force in Navier-Stokes equations is not represented by the potential part ϕ of the Tn vector.

According to ref. [2], the boundary conditions for vector W have the same form as for the ideal nonviscous liquid. On the walls the condition $W_1|_G = 0$ has to be rewritten for the potential ϕ :

$$
\frac{\partial \phi}{\partial I} = TnI|_G.
$$
 (3)

U_0 character velocity, $(g\beta\Delta T h)^{-1/2}$

velocity vector

- velocity vector
- W rotational part of vector, $T_n = W + \nabla \phi$
- x nondimensional coordinate, x/h .

Greek symbols

- β volumetric coefficient of liquid expansion
 $\bar{\gamma}$ unit vector of gravity axis
- unit vector of gravity axis
- ϵ parameter of vibration, $0.5(\beta b\omega)^2$
- *0* nondimensional temperature,
- $(T-T_0)/T_0$ $\mu_{\rm c}$ system units coefficient of MHD
- interaction v kinematic viscosity
- ρ density
- σ electroconductivity of the liquid
- period of time for vibrational oscillation τ
- potential part of vector **Tn**
- $\frac{\phi}{\chi}$
- temperature conductivity in the liquid circular frequency of vibration ω
- Ω circular rotational frequency.
-

This case was considered earlier analytically [4] assuming an infinite longitudinal size of the crucible. The influence of horizontal vibration on heat convection was studied in a 2-D geometry. It was shown that horizontal vibration along the temperature gradient suppresses convective motion in the crucible. Other directions of vibrations do not affect fluid motion in the case considered.

The distribution of velocities near the vertical wall has great importance for the purposes of dopants segregation in crystal growth but could not be obtained from analytical study. Approximate solutions were derived [ll, 121. But the central question here is to know whether the convective motion can be damped with the help of vibration in typical semiconductor growth conditions. What is the efhciency of vibrational damping compared with magnetic field?

Let us consider viscous fluid motion in the crucible. Temperature boundary conditions correspond to the linear distribution of the temperature along the horizontal walls (Fig. I), and constant values of temperature on the vertical walls. Boundary conditions for velocities are non-slip on the walls. The vectorvelocity field for heat convection is shown on Fig. I in the case typical for GaSb growth in horizontal Bridgman under earth gravity. The diameter of the crucible $h = 10^{-2}$ m, the temperature axial gradient $\partial T/\partial x = 10^3$ K m⁻¹, the aspect ratio is $l/h = 4$. For the conditions with $Gr_1 \geq 10^6$ the convective motion

FIG. 1. Heat-convective motion in horizontal Bridgman crystal growth geometry. $Gr_1 = 9 \times 10^4$, $Pr = 5 \times 10^{-2}$, $U_{\text{max}} = 0.726$.

has a well-known cell structure with a few vortexes along the x axis.

The case with the presence of horizontal vibrations is represented on Fig. 2. Rayleigh vibrational number was chosen equal to 4.5×10^4 , those conditions seem to be the limiting maximal values which could be achieved in practice. In the above mentioned crystal growth conditions this corresponds to a vibrational parameter $\omega b = 10$ m s⁻¹ (for instance amplitude b) about 10^{-3} m and frequency $\omega \sim 10^4$ Hz). Intensity of the fluid motion is decreased by a factor 3 as shown on Fig. 2. For an aspect ratio larger than 10 the velocity profiles in the center of the crucible differ only slightly from the ones given analytically in ref. [4]. And near the cold vertical wall the damping is about 2.2 times for the chosen parameters.

It is interesting to point out the analogy between the influences of vibration and applied vertical magnetic field on convective motion in a horizontal crucible with electroconducting walls. If we neglect the convective fluid motion which changes the linear onedimensional temperature profile in the volume there is no influence of horizontal vibration on the movement. This state is considered as a neutral one. Therefore the curl part of *Tn* vector is generated only by the convective movement. So in the linear approach, the heat transfer equation system (1) could be written in

the form :

$$
\mathbf{W} = \mathbf{V} \frac{\partial T}{\partial x} \frac{h^2}{\chi}
$$

and the vibrational force term in (1) in the linear approach is reduced as follows :

$$
\varepsilon(\mathbf{W}\nabla)(T\mathbf{n}-\mathbf{W})=\varepsilon\mathbf{W}\frac{\partial T}{\partial x}=\mathbf{V}\left(\frac{\partial T}{\partial x}\right)^2\frac{h^2}{\chi}\frac{1}{2}\beta^2(b\omega)^2.
$$

In nondimensional form, the thermo-vibrational term has the form Ra_{v} $Gr^{-1/2}$ U. Now we can compare this term with the Lorentz force $Ha^2 Gr^{-1/2}(\mathbf{U} \times \mathbf{e}_B) \times \mathbf{e}_B$. For the vertical orientation of magnetic field and horizontal component of U, the Lorentz term will take the form Ha^2 Gr^{-1/2}U. The term

$$
\left(\frac{\partial T}{\partial x}\right)^2 \frac{h^2}{\chi} \frac{1}{2} \beta^2
$$

plays the role of electroconductivity σ and vibrational parameter (ωb) the role of external magnetic field.

For GaSb material grown in a 10^{-2} m diameter crucible with an axial temperature gradient about $10³$ K m⁻¹ and a vibrational parameter $b\omega \sim 1$ m s⁻¹, the value of $Ra_v^{1/2}$ is about 10. For the value of vertical

FIG. 2. Heat-convection motion in horizontal Bridgman crystal growth geometry in vibrational field with horizontal axis. $Gr_1 = 9 \times 10^4$, $Pr = 5 \times 10^{-2}$, $Ra_v = 4.5 \times 10^4$, $U_{max} = 0.245$.

magnetic field of 1 T the value of Hartman number (Ha) is about 140. The maximal value of velocity calculated in the presence of the magnetic field is decreased by a factor 7 (the corresponding velocity decreases given on Fig. 2 arc about 3 times for the value of the parameter $b\omega = 10$ m s⁻¹). Using magnetic fields to suppress the convective motion seems to be easier in practice for the considered case of GaSb growth. The use of vibration in horizontal Bridgman geometry could be successful for melts with low electroconductivity like materials from group A_2B_6 (CdTe) where the value of electroconductivity is $10²$ times lower than for GaSb.

4. **VERTICAL BRIDGMAN CONFIGURATION**

At first we propose an analytical study of vibration influence on convective-heat flow. Let us consider a vertical 2-D rectangular crucible in a vibrational field with an axis parallel to x (Fig. 3). Boundary conditions in the crucible correspond to a constant temperature gradient along the vertical walls. We can admit that there exists a distortion of the flat isotherms in the volume of the liquid which cause in turn convective motion in the crucible. The temperature field is described with a simple dependence :

FIG. **3.** Temperature boundary conditions and computed vector field $\mathbf{W} = T\mathbf{x} - \nabla \phi$.

where D is the vertical temperature gradient and C the local bending of the isotherms, supposed to be constant in an analytical study. Two force terms appear in the Navier-Stokes equations (2) with the presence of vibration: buoyancy and thermovibration. The absence of fluid motion in the volume could be achieved only if the applied volumetric forces are potential. The forces are balanced in this case by the pressure field.

To calculate the thermo-vibrational force according to (1) we have to subdivide vector Tx into potential and rotational parts

$$
T\mathbf{x} = \mathbf{W} + \nabla\phi
$$

taking into account boundary conditions (3) for vector $\mathbf{W}: \mathbf{W}_x|_G = 0.$

Computed vector W for constant temperature values on the horizontal walls is presented on Fig. 3, and could serve as an illustration to the further study.

Near the vertical walls, the vector Tx could be subdivided as follows :

$$
T\mathbf{x} \equiv \begin{pmatrix} Dy + Cx \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ -xD \end{pmatrix} + \begin{pmatrix} Dy + Cx \\ xD \end{pmatrix}.
$$

The boundary conditions on vertical walls are valid for vector W. The thermo-vibrational force term $\varepsilon(W\nabla)\nabla\phi$ in this case is the potential one :

$$
\varepsilon(\mathbf{W}\nabla)\nabla\phi=\varepsilon\bigg(\frac{-xD^2}{0}\bigg).
$$

There is no other term to suppress the rotational part of the buoyancy force. So there is no direct influence of horizontal vibration on the fluid near the vertical walls. This result is also confirmed by the conclusions of ref. [3], that horizontal vibration does not suppress fluid motion in an infinite crucible.

The vector *TX* has to be subdivided into potential and rotational parts in the vicinity of horizontal walls in another way :

$$
Tx \equiv \begin{pmatrix} Dy + Cx \\ 0 \end{pmatrix} = \begin{pmatrix} yD \\ 0 \end{pmatrix} + \begin{pmatrix} Cx \\ 0 \end{pmatrix}.
$$

The thermo-vibration force term takes the form :

$$
\varepsilon(\mathbf{W}\nabla)\nabla\phi=\varepsilon\begin{pmatrix}yCD\\0\end{pmatrix}.
$$

With the buoyancy force, it forms the resulting volumetric force :

$$
\mathbf{g}\beta T\mathbf{y} + \varepsilon(\mathbf{W}\nabla)\nabla\phi = \begin{pmatrix} 0 \\ \mathbf{g}\beta Dy \end{pmatrix} + \begin{pmatrix} \varepsilon yCD \\ \mathbf{g}\beta Cx \end{pmatrix}.
$$
 (4)

The first term in the right-hand part of (4) is curl free. The second one can be converted in potential form only if $\epsilon D = g\beta$, or in nondimensional form:

$$
\frac{Ra_{v}}{Gr\,Pr}\left(\frac{h}{l}\right) = 1.\tag{5}
$$

The last equality is a condition of natural convection damping by horizontal vibration in a finite crucible. The exact value of isotherms bending C is not important in the analytical study. In Fig. 4 the field of thermo-vibration forces for the full computer calculated problem is represented. It can also be seen that the influence of horizontal vibration on convective fluid motion takes place only due to the forces generated near the horizontal bottom and top walls.

The same analysis can be carried out for vibration along the vertical axis. In this case, the nonpotential part of the buoyancy force only increases with the thermo-vibrational force.

In the analytical study, we assumed a constant bending of isotherms C and we also neglected the influence of remote parts of the volume by means of pressure and fluid flow. So the condition (5) for convective fluid motion damping could be considered only like a 'zero-order' approach to the problem. Let us study the full 2-D problem. Temperature boundary conditions are :

$$
x = \pm 0.5 \quad T = y/l
$$

\n
$$
y = l \qquad T = 1
$$

\n
$$
y = 1 \qquad \frac{\partial T}{\partial y} = Q + f(x)
$$

where O describes the latent heat source on the surface during the crystal growth process and $f(x)$ the crystal cooling (symmetrical function, increasing with $|x|$).

The boundary conditions for the fluid velocities are non-slip conditions on the solid walls. Nondimensional numbers in the system of equations (2) were taken as follows: $Pr = 0.05$, $Gr₁ = 9 \times 10⁴$ $(Gr = 3.6 \times 10^5, l/h = 4)$ which correspond to GaSb crystal growth for the horizontal Bridgman configuration. In Fig. 5 the velocity field of natural convective motion is plotted. Two cases can be distinguished depending on the convective motion direction. For the relatively high values of Q (high rates of crystal growth) isotherms in the bottom are concave. For the low values of Q (corresponding to the low rates of crystal growth) isotherms have convex form (like in Fig. 5). At the same time, the horizontal vibration generates a fluid motion similar to the one obtained with concave isotherms in the bottom of crucible (Fig. 6, where all calculations were done without the buoyancy term in equation (2)). This points out that horizontal vibration can only damp convective motion created when the isotherms (interphase boundary) in the bottom are convex.

For practical applications, only liquid motion in the region near the bottom is important because of its influence on solute redistribution at the solid-liquid

T=l

 $T = y/1$

FIG. 4. Computed vector field of volume thermo-vibrational forces.

FIG. 5. Fluid motion with concave bottom isotherm $Gr_1 = 9 \times 10^4$, $Pr = 5 \times 10^{-2}$, $(\partial T / \partial x)|_{y=0} = 0.2 + 0.2x$ $U_{\text{max}} = 0.66 \times 10^{-2}$.

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FIG. 6. Fluid motion generated by thermo-vibrational forces without buoyancy term. $Pr = 5 \times 10^{-2}$, $(\partial T/\partial x)|_{y=0} =$ $0.2+0.2x^2$, $g\beta Ty \equiv 0$, $Ra_v = 1.2 \times 10^4$, $U_{\text{max}} = 0.99 \times 10^{-3}$.

interface. Therefore, the increase of convection in the upper part of the crucible can be neglected from a practical point of view.

The resulting flow field is shown on Fig. 7 taking into account both buoyancy and thermo-vibration force terms. Convective motion is subdivided into three regions with vortices rotating in opposite directions. Increasing or decreasing the critical *Ra,* number also changes the sizes and intensities of the vortices in the fluid. For the critical *Ra,* number (maximum of convective motion damping) the level of normal velocity component is damped by a factor 20, the transversal component by a factor 10. In dimensional values the level of y-components of velocities near the bottom changes from 2.0×10^{-5} to 10^{-6} m s⁻¹ for the chosen boundary conditions with a low rate growth (value of $Q = 0.2$ used in calculations corresponds to 3.0×10^{-7} m s⁻¹ rate of crystal growth). It is also interesting to note that the value of the critical *Ra,* obtained in the analytical study is 10 times higher than the calculated one $(Ra_v = 0.6 \times 10^4)$.

Comparison of fluid motion damping near the bottom of the crucible by means of horizontal vibration and vertical magnetic field is another interesting feature of our analysis. For the vertical magnetic field $B = 1$ T ($Ha = 140$) with crucible electroconducting

FIG. 7. Heat convection in vibrational field with horizontal axis. $Gr_1 = 9 \times 10^4$, $Pr = 5 \times 10^{-2}$, $(\partial T/\partial x)|_{y=0} =$ $0.2+0.2x^2$, $Ra_v = 0.6 \times 10^4$, $U_{\text{max}} = 0.34 \times 10^{-2}$, U_{max} (in the bottom) = 0.56×10^{-3} .

walls, the maximum value of velocities is decreased by a factor 6 (compared with 10-20 times by vibration).

5. COMPUTATION TECHNIQUE

The powerful FIDAP package (Fluid International Dynamic Application Package) was used to compute the problem. The additional equation in the generalized Navier-Stokes system was written for the potential part ϕ of vector Tn. All the source terms were incorporated by means of subroutines in the FIDAP package. The whole system was solved in a transient state. As time proceeded the solution became stationary.

6. DISCUSSION

The important question is to know if there exists a possibility to achieve the diffusive mass-transfer conditions in the vicinity of the interface under earth gravity. For this purpose the convective motion has to be damped significantly. In ref. [IO] it was shown that for semiconductor melts with relatively high electroconductivities and diameters of grown crystals about 10^{-2} m these conditions could be achieved with magnetic fields stronger than 20 T. Until now, this fact was not confirmed experimentally. The suppression of natural convection in vertical Bridgman crystal growth with the help of vibration is a new possibility. It is shown that horizontal vibration in a temperature field with a vertical gradient generates a field of forces. For the conditions which correspond to a concave interphase boundary the temperature and thermovibrational convections have opposite directions ; their interaction leads to a significant decrease of the velocities. For the growth rates used $\sim 10^{-6}$ m s⁻¹ in the Bridgman technique, the velocities of convection have to be lower in order to achieve a mainly diffusive mass transfer regime to the growing surface. The lowest values of the temperature gradients used in this technique are about 7 K cm^{-1} to avoid solidification processes in volume. The maximal level of velocity near the front in this case in dimensional values is about 2.0×10^{-5} m s⁻¹. With the horizontal vibration this value could be decreased 20 times in normal direction to the front and in 10 times in transversal direction ; the diffusive mass transfer conditions can then be attained. The resonance value of the vibrational parameter obtained is $b\omega \sim 3$ m s⁻¹. With the increasing of this critical value, the vibro-convection appears to be stronger than the natural one, and with the decreasing there is an opposite situation. The value of this parameter is high but could probably be attained in practice with the amplitude of harmonic oscillations of about 3.0×10^{-3} m and frequencies of about 10^3 Hz. In any case the application of vibration to suppress the heat convection could be valuable for the low gravity conditions where the levels of velocities in convective motion are significantly lower than in the earth gravity.

Another question : how could these results be transferred to axisymmetric or 3-D cases? It could be proposed to rotate an axis of vibration in a horizontal plane with the frequency $\Omega : \tau^{-1} \ll \Omega \ll \omega$ (where τ is the characteristic time of vibro-heat convective motion). In this case, the problem could be considered axi-symmetrical.

7. **CONCLUSION**

The usage of vibration to damp the heat convection in the horizontal Bridgman configuration could be successful for relatively high *Ra,* (Rayleigh vibrational number), and low *Gr* (Grashof number). For the real conditions on earth for semiconductor materials grown in horizontal Bridgman such as Si, GaSb, GaAs, etc. with relatively high values of electroconductivity, application of magnetic fields appear to be more realistic and simpler to achieve the same effect of movement braking in the melt compared with the application of vibration.

For the vertical Bridgman crystal growth geometry, suppression of convection with the horizontal vibration could be achieved when the buoyancy and vibro-convective forces have opposite directions. The effect of fluid motion suppression has a resonant character and takes place in the bottom zone of the

crucible near the growing surface. Calculations show that the vertical velocity near the front could be decreased about 20 times and the horizontal one about 10 times. In these conditions it is possible theoretically to achieve diffusive mass transfer of solutes to the growing surface, which was only possible till now in microgravity conditions. The rate of crystal growth in the last case can be as low as $(0.7-1.0) \times 10^{-6}$ m s⁻¹. For the low-convective motion of semiconductor melts, the usage of vibration seems to be effective mean in comparison with medium-range magnetic field $(1-5T)$.

8. **FURTHER PROBLEMS**

The application of horizontal vibration seems to be fruitful in the case of vertical Bridgman crystal growth and gives an opportunity to suppress convective motion significantly. Another problem then arises : how to choose the temperature distribution along the vertical walls to attain the maximal damping of the fluid near the front of crystal growth front. Another question is: can we suppress the convective motion further if both magnetic field and vibration are applied simultaneously?

In the previous studies, the transfer coefficients (diffusive .coefficient and temperature conductivity) were considered nondependent of the frequency of vibration, but more work would be necessary on this point.

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