ULTRASONIC INFLUENCE ON CONVECTION IN THE MELTS

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Summary. The influence of ultrasonic waves on convective flows in the melts on the growth of GaAs, InSb, Bi$_x$Sb$_{1-x}$, Ga$_x$In$_{1-x}$Sb single crystals by Czochralski method has been studied. Non-dimensional numbers of Rayleigh, Prandtl, Schmidt and Reynolds were calculated for the crystal growth conditions with the introduction of ultrasound at frequencies from 0.15 MHz to 5 MHz into the melts. It was found, that stationary convection is the best condition for the decrease of doping striations in pulling single crystals at the presence of ultrasound on the melts. The diffusion layer thickness for the crystal growth conditions of these single crystals was calculated. The change of the diffusion layer thickness from 0.1 mm to 0.6 mm not influenced on ultrasound effect on the striations decrease.

Key words: ultrasound, striations, non-dimensional numbers, the Czochralski method.

INTRODUCTION

It is known [1, 2], that the components inhomogeneity in semiconductors single crystals influences on the stability and reproducibility of electrophysical parameters. The formation of impurity striations is the result of a thermal convection [3, 4]. The decrease of the striations in the crystals is provided by damping of convection in the melt at the presence of the external fields: gravitation, magnetic and ultrasonic. The application of ultrasound as an external field influencing crystal growth is an alternative approach for similar fundamental investigations.

The effect of ultrasonic vibrations at a frequency 10-kHz and power up of 30 W on the growth morphology in InSb and Ga$_x$In$_{1-x}$Sb single crystals by the Czochralski method was studied in detail [5-10]. However, striations do not disappear completely in these crystals even under the action of ultrasound with a power up to 120 W.

The effect of the striations which decrease in number and disappear in the central part of InSb, GaAs, Bi-Sb Ga$_x$In$_{1-x}$Sb pulled single crystals was studied and reported in a few publications of our group [10-15]. In these experiments ultrasonic waves at frequencies of 0.15, 0.25, 0.6, 1.2, 2.5, 5 and 10 MHz were introduced into the melt through a quartz waveguide, fused to the bottom of the quartz crucible. We supposed that ultrasonic standing waves might influence the flow patterns in the melts and reduce
the striations in the growing crystals [13]. However, the interconnection of ultrasound with hydrodynamic process in the melts not studied in these experiments.

In this paper, the present work is part of a series of experimental studies of the behavior of convective flow in the fluid and the ultrasound influence in a Czochralski configuration for the crystal growth without doping striations.

**RESEARCH OBJECT**

It was found that convection in the melts decreases the positive effect of ultrasound on damping of the striations. Therefore, non-dimensional numbers of Rayleigh, Prandtl, Schmidt and Reynolds were calculated for the crystals growth conditions with the introduction of ultrasound at frequencies from 0.15 MHz to 5 MHz into the melts by Czochralski method. Using the technological parameters for crystal growth process in our experiments: the temperature gradient, melt depth, angular velocity of the crystal rotation and characteristic length of free surface - were calculated these non-dimensional numbers.

The mode of natural and forced convection can be described in the terms of non-dimensional numbers such as Rayleigh number $Ra_w$ and Reynolds number $Re_\Omega$ [15], which consider $h/d$ and $\Omega$ parameters.

The Rayleigh number for a model with an isothermal wall $Ra_w$ is defined as:

$$ Ra_w = \frac{g \cdot 2 \cdot \alpha \cdot (T_w - T_m) \cdot h^4}{d \cdot \nu \cdot a}, $$

where: $g$ is the gravity, $\alpha$ is the thermal coefficient of volume expansion, $T_w$ is the temperature of isothermal wall, $T_m$ is the temperature difference in the melt, $h$ is the height of the melt, $d$ is the diameter of the crucible, $\nu$ is the kinematic viscosity, $a$ is the thermal diffusivity.

The thermal diffusivity defined as:

$$ a = \frac{\lambda_t}{C_p \cdot \rho}, $$

where: $\lambda_t$ is the heat conduction, $C_p$ is the specific heat, $\rho$ is the density.

The Prandtl number is one of the most important factors governing the fluid flow patterns. It is dependent only on the thermal characteristics of the liquid and independent of container geometry or applied temperature gradient [15]:

$$ Pr = \frac{\nu}{a}. $$

In rotating flows, the Reynolds number is determined by the influence of the rotation of the crystal:

$$ Re_\Omega = \Omega l^2 / \nu, $$

where: $\Omega$ is the angular velocity and $l$ is the characteristic length equal of the crystal dummy radius.
The Schmidt number determines the relative contribution of linear momentum and the impurity diffusion at the solid-liquid interface:

$$Sc = \frac{\nu}{D},$$

where: $D$ is the coefficient of diffusion.

It is known, that near at the solid-liquid interface is the diffusion layer which can influence on the component distribution in semiconductor single crystals. The diffusion layer thickness $\delta$ is defined as [16]:

$$\delta = 1.6 \cdot D^{1/3} \cdot \nu^{1/6} \cdot \Omega^{-1/2}.$$  \hspace{1cm} (6)

Thickness of diffusive layer was also calculated, because it can affect on the distribution of components at pulling single crystals.

**RESULTS EXPERIMENTAL RESEARCH**

Based on the data of the Table 1 and Figs. 1, 2 we should note that the increase of the Schmidt number and decrease Reynolds number by 2-3 orders of magnitude, respectively, for the growth conditions of these melts. We did not observe the influence of the diffusive layer thickness on the striations. The smallest Rayleigh number $Ra \approx 15.8$ correspond conditions for growing of Bi-Sb single crystals and greatest $Ra \approx 1 \cdot 10^6$ for growing of GaAs single crystals, respectively. In Bi-Sb single crystals the initial parts of which were pulled with ultrasound we observed an aftereffect phenomenon: even after switching the ultrasound off, single crystals with constant diameters containing no layers grew over 2-2.5 h. We studied the distribution of As in the GaAs single crystals pulled with 0.15 MHz ultrasound. In the ultrasound field, the distribution of As became more uniform only in the growth zone with a constant crystals diameter. Periodic switch-offs of the ultrasonic transducer facilitated the formation of inhomogeneity in the As distribution. The positive effect of ultrasound was observed only for GaAs single crystals grown from 30 mm melt depth. When the melt depth was doubled, it was impossible to reduce the As inhomogeneity under the influence of ultrasound, which may be caused by the increase of convective mixing in the melt.

Therefore, we should note, that the Rayleigh number is a main criterion for detection of the ultrasound effect on the striation decrease in the single crystals growing by the Czochralski method. The Rayleigh number makes it possible to analyse of stationary and time-dependent convection. The ultrasonic waves introducing in the melt can decrease the striations in pulled single crystal for the Rayleigh number $Ra < 10^5$.

The comparative data on the non-dimensional numbers of Rayleigh, Prandtl, Schmidt and Reynolds, the diffusive layer thickness and hydrodynamic parameters of the melts for the growth of GaAs, InSb, Bi-Sb and Ga$_x$In$_{1-x}$Sb single crystals are listed in Table 1.

Results of calculation of the Rayleigh number for a model with an isothermal wall are presented in Fig.1 and Fig.2.
Table 1. Hydrodynamic parameters and non-dimensional numbers of melts

<table>
<thead>
<tr>
<th>№</th>
<th>Parameters</th>
<th>Melt Ga-As</th>
<th>Melt In-Sb</th>
<th>Melt Ga-In-Sb (Ga$<em>{0.03}$In$</em>{0.97}$Sb crystal)</th>
<th>Melt Bi-Sb (Bi$<em>{0.98}$Sb$</em>{0.02}$ crystal)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Melt depth, $h$, m</td>
<td>$3\cdot10^{-2}$</td>
<td>$2\cdot10^{-2}$</td>
<td>$2\cdot10^{-2}$</td>
<td>$2\cdot10^{-2}$</td>
</tr>
<tr>
<td>2</td>
<td>Difference of temperatures in melts along growth axis, $\Delta T$, К</td>
<td>12</td>
<td>4,6</td>
<td>4,6</td>
<td>4,6</td>
</tr>
<tr>
<td>3</td>
<td>Kinematic viscosity, $V$, m/s</td>
<td>$4,89\cdot10^{-7}$</td>
<td>$3,4\cdot10^{-7}$</td>
<td>$3,6\cdot10^{-7}$</td>
<td>$1,31\cdot10^{-7}$</td>
</tr>
<tr>
<td>4</td>
<td>Thermal diffusivity, $a$, m$^2$/s</td>
<td>$0,13\cdot10^{-4}$</td>
<td>$8,4\cdot10^{-8}$</td>
<td>$0,97\cdot10^{-3}$</td>
<td>$7,4\cdot10^{-4}$</td>
</tr>
<tr>
<td>5</td>
<td>Coefficient of thermal expansion, $\alpha$, 1/К</td>
<td>$1,87\cdot10^{-4}$</td>
<td>$5,2\cdot10^{-5}$</td>
<td>$9,8\cdot10^{-7}$</td>
<td>$1,22\cdot10^{-4}$</td>
</tr>
<tr>
<td>6</td>
<td>Thermal conductivity, $\lambda$, Vt/(m·K)</td>
<td>17,8</td>
<td>9,23</td>
<td>9,27</td>
<td>$2,6\cdot10^{-3}$</td>
</tr>
<tr>
<td>7</td>
<td>Heat capacity $C_p$, J/(kg·K)</td>
<td>434</td>
<td>263</td>
<td>263,325</td>
<td>178</td>
</tr>
<tr>
<td>8</td>
<td>Melt density, $\rho$, kg/m$^3$</td>
<td>5720</td>
<td>6430</td>
<td>6,48·10$^3$</td>
<td>9950</td>
</tr>
<tr>
<td>9</td>
<td>Coefficient of diffusion, D, m$^2$/s</td>
<td>$2,6\cdot10^{-8}$</td>
<td>$4,4\cdot10^{-9}$</td>
<td>$4,52\cdot10^{-10}$</td>
<td>$2,8\cdot10^{-9}$</td>
</tr>
<tr>
<td>10</td>
<td>Length of free surface, $l_c$, m</td>
<td>$4\cdot10^{-2}$</td>
<td>$2,3\cdot10^{-2}$</td>
<td>$2,3\cdot10^{-2}$</td>
<td>$2,3\cdot10^{-2}$</td>
</tr>
<tr>
<td>11</td>
<td>Angular velocity of rotation of the crystal, $\Omega$, s$^{-1}$</td>
<td>0,5</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>12</td>
<td>Rayleigh Number, $Ra$</td>
<td>$6\cdot10^7$</td>
<td>$2\cdot10^6$</td>
<td>$1,53\cdot10^4$</td>
<td>$1,55\cdot10^4$</td>
</tr>
<tr>
<td>13</td>
<td>Prandtl Number, $Pr$</td>
<td>0,038</td>
<td>0,04</td>
<td>0,037</td>
<td>0,017</td>
</tr>
<tr>
<td>14</td>
<td>Schmidt Number, $Sc$</td>
<td>18,8</td>
<td>77,3</td>
<td>796,5</td>
<td>4670</td>
</tr>
<tr>
<td>15</td>
<td>Reynolds Number, $Re$</td>
<td>1635,9</td>
<td>1542,4</td>
<td>1456,7</td>
<td>40,03</td>
</tr>
<tr>
<td>16</td>
<td>Diffusive layer thickness of, $\delta$</td>
<td>0,00059</td>
<td>0,00022</td>
<td>0,00104</td>
<td>0,00035</td>
</tr>
</tbody>
</table>

Fig. 1. Dependence of non-dimensional Rayleigh number from h/d for Ga-As and In-Sb melts
CONCLUSION

In order to determine special growth conditions in ultrasonic field for striation-free GaAs, InSb, Bi,Sb, and GaInSb single crystals the experimental results were studied and analyzed using non-dimensional numbers of Rayleigh, Prandtl, Schmidt and Reynolds. It has been shown, that Rayleigh number is a main factor for determining effect of ultrasound in the growth of semiconductor single crystals by the Czochralski method. It was found, that stationary convection for the Rayleigh number less than $10^5$ are the best conditions for the decrease of doping striations in pulling single crystals at the presence of ultrasound on the melts. It was found, that behavior of the effect of ultrasonic vibrations in the melts is increase, when the increase of the Schmidt number and decrease Reynolds number by 2-3 orders of magnitude, respectively, for the growth conditions of these melts. The diffusion layer thickness for the crystal growth conditions of GaAs, InSb, Bi,Sb, and GaInSb single crystals was calculated. The value of the diffusion layer thickness is varied from 0.1 mm to 0.6 mm not influenced on ultrasound effect on the striation decrease.
REFERENCES


УЛЬТРАЗВУКОВОЕ ВОЗДЕЙСТВИЕ НА КОНВЕКЦИЮ В РАСПЛАВАХ

Кожемякин Г.Н., Дегтярева А.А.

Аннотация. Изучено влияние ультразвука на конвективные течения в расплавах при вытягивании монокристаллов GaAs, InSb, Bi-Sb, GaIn_xSb методом Чохральского. Рассчитаны безразмерные числа Рэлея, Прандтля, Шмидта и Рейнольдса для условий выращивания данных монокристаллов при воздействии на расплав ультразвука с частотой от 0,15 до 5 МГц. Установлено, что лучшим условием для снижения слоистости в монокристаллах при воздействии ультразвука на расплав является наличие стационарной конвекции. Выполнен расчет толщины диффузионного слоя для условий вытягивания этих монокристаллов. При изменении толщины диффузионного слоя от 0,1 до 0,6 мм не установлено влияние его величины на эффективность ультразвукового воздействия с целью снижения слоистости.

Ключевые слова: ультразвук, слоистость, безразмерные числа, метод Чохральского.