

# Doped InSb Detached Crystals by VDS Technique: Its Substrates for Infrared Devices and Physics Concept

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**Abstract**— In this paper, the vertical directional solidification (VDS) detached crystal growth process in our laboratory in which a gap exists between a growing crystal and the ampoule wall is described. However, this phenomenon is more complex due to the hydrostatic pressure, and the existence of the buoyancy convections. Important characteristics of the detached growths are the self-stabilizing gas pressure difference and self-detachment crystal growth process into VDS on earth. The hydrostatic pressure decreases during the growth, the pressure at the bottom decreases such that the liquid meniscus remains unchanged all along the growth axis. Gap formation mechanism is not totally understood yet, but experimental observations can be seen that the gas passing upwards inside ampoules for grown ingots presence of the thin oxide layer. Detachment in VDS is self controlled and the self-applied pressure difference should be of the order of the hydrostatic pressure. In our references, characterization of high quality InSb and doped InSb substrates suitable for use in the infrared devices, and VDS technologies to deliver larger substrate is explained. Here, the physics behind detached growth and technology to develop the junction devices from these substrates is highlighted.

**Index Terms**— Vertical directional solidification (VDS), InSb detached crystals, Crystal-Melt interface, Improved Crystal quality, p-n junction devices, Physics concept.

## I. INTRODUCTION

Electronic industry requires high quality semiconductor crystal with uniform dopant distribution. Non-uniformity along solid/liquid interface can lead to an interface instability which in turn can result in the formation of growth morphologies. Recently, InSb has gained interest because Intel and QinetiQ have developed an InSb-based quantum well [1]. InSb has shown as a potential device application material due to its smallest energy gap and highest mobility of the binary III-V materials for the optical spectroscopy and optoelectronic applications. Therefore it is useful in the fabrication of high-speed heterojunction transistors, low-cost solar cells with high efficiency, fabrication of infrared imaging systems, free space communications, gas phase detection systems, infrared detectors and infrared filters. InSb doping effect is producing p-n junction diodes for IR detector and Planer p-n junction photo-diodes. These materials are potential materials for room temperature infrared detectors, gas sensors and lasers operating in the region in near-infrared (0.8–1.3 $\mu\text{m}$ ), mid-infrared (2–5 $\mu\text{m}$ ) and far infrared (8–14 $\mu\text{m}$ ). Devices applications require high quality InSb and InSb:X (X = Te, Tl, In, Ga, Bi, N, Mg, Mn etc dopant) bulk

crystals [2]. However, it is very difficult to grow large single crystals of high quality, because the solute distribution and the instability in growing single crystals. There are two major problems in crystal growth. The first is the constitutional supercooling which appears in the solution ahead of the growth interface because of separation in liquids and solidus. The second is occurrence of convections in the solution result in unstable growth place due to variation of the densities. Space environment (microgravity) is ideally suited to grow detached bulk crystals with the high quality and defect free crystals as the gravity induced negative effects are absent [3].

The growth of high-quality single crystals is a challenge to the crystal growth scientists and industry; however recent crystal growths have shown that, there are two major challenges. First is the production of well-established crystalline materials with improved structural perfection and larger size at a lower cost and second is the bulk growth of new categories of materials with extreme understanding of thermodynamic process. The issue of perfect high quality crystal growth is retrograde by the presences of precipitates, inclusions, twins, grains and dislocations [4]. The high quality bulk crystal requires chemically and electrically homogeneous crystals free from extraneous secondary phases. This is one of the most difficult objectives in the growth of crystals due to thermodynamic and technological problems. For bulk crystals growth, beginning with Skylab-III and IV Mission-1974 in microgravity experiments, grown ingots diameters were less than the diameters of the crucibles by a gap. Existence of the gap provides advantages, such as no sticking of the crystal to the ampoule wall, reduced thermal, mechanical stresses, reduction in dislocations, and no heterogeneous nucleation by the ampoule [5]. Experiments performed under the microgravity / space are very useful for analyzing the growth mechanism at the solid-liquid interface, because natural convection can be suppressed and diffusion-controlled solute transport can be achieved under microgravity conditions. To understand the growth mechanisms, experiments have been performed in the International Space Station (ISS) [6-8]. It has been understood that, factors affecting detached growth are the growth angle of the crystal, the contact angle of the melt with the crucible wall and the pressure different across the meniscus [9]. The melt is supported above the lower vapor phase by a small liquid meniscus that bridges the gap between the ampoule wall and the triple-phase line at which solid, liquid, and vapor phases intersect. This upper vapor phase is not strictly necessary for detached growth to occur, but its presence facilitates manipulation of the pressure difference on the melt and to avoid a free melt surface. To achieve stable detached growth under terrestrial conditions. The detached growth retains a key advantage at appropriate thermal gradients to achieve meniscus in crystal growth [10]. Duffar

et.al. gave explanations of dewetting of the melt from the ampoule wall by the surface roughness or poor wetting by the melt [11]. Wilcox et. al. proposed a mechanism in which gas is transported through the melt from the upper vapor space into the lower vapor space during growth. The out gassing mechanism is suitable for closed systems that have been used in space experiments [12]. The influence of gravity complicates the detached growth because the hydrostatic pressure changes with time as the height of the melt diminishes during growth and must be offset by an externally applied pressure difference to achieve detached growth in terrestrial experiments [13]. Derby et.al has proposed the theoretical studies of detached / dewetting phenomenon in terrestrial laboratory by externally applied pressure [10].

Detached growth in vertical directional solidification (VDS) technique is experimental effort for the bulk crystal growth process in terrestrial laboratory and the experimental results have been reported in earlier published paper [14] and Indian patent [15]. It showed enhancement in crystals quality: InSb [16], InSb:Te [17], InSb:Ti [18], InSb:Ga [19-20], InSb:Bi [21], InSb:N [22], GaSb [23], GaSb:Mn [24], GaSb:In [25], GaSb:Se [26] and improvement in the physical properties [27-28]. InSb is important for IR devices applications, and its constituent InSbX alloys are useful in the wavelength 2–14 $\mu$ m range. Transients in dopant distribution of the detached growth results are published in our references. Ground-based experiments have demonstrated that the level of buoyancy driven convection in the melt is significantly reduced in a detached growth, and yielded a significant improvement. Thus, ground-based VDS experiments yielded nearly-convection-free solidification, and it indicate that VDS is relatively insensitive to gravitational acceleration due presence of negative gravitational effect.

## II. EXPERIMENTAL PROCEDURE

Numerous efforts have been made to grow bulk single crystals by Czochralski (CZ), horizontal and vertical travelling heater method (THM), vertical Bridgman (VB) and vertical gradient freeze (VGF) method. *The closely spaced striations in both Czochralski and Bridgman-grown crystals have grown by buoyancy-driven convection. This leads to the thermal stress, which increases defects and dislocation density.* The difficulties in THM is that the growth rate is very low and the growth from non-stoichiometric melt produces variation in composition in the crystal due to the fluctuation in freezing rate at the solid–liquid interface produces defects. *To reduce the defect density in bulk materials, a new novel crystal growth process is necessary. VDS technique could be an innovative effort to grow bulk crystals of the high quality. It is indigenous developed method for the bulk crystals into our laboratory.*

*The source materials of high purity (5N, AlfaAeser) had been used in stoichiometric proportion for the undoped and doped InSb growth by VDS [25]. The typical seven steps furnace temperature profile has been applied and steps are -*  
i) Furnace temperature has raised in 3 hours to set temperature (150<sup>o</sup>C above m.p. of the source materials). The source materials before sealing flushed alternatively more than 10 times by vacuum 10<sup>-5</sup>torr and filled argon gas, then sealed ampoule (inside argon pressure 200-300 tor) were

*kept for congruent homogeneous mixing at this temperature for 12 hours, ii) Ampoule was lowered in 3 hours to 50<sup>o</sup>C above the melting point of the InSb (525<sup>o</sup>C), iii) The temperature thermal stability was maintained for 3 hours, iv) Growth parameters: growth time  $\approx$ 20 hours (crystal diameter 10-22mm and length 65-70mm), the ampoule transition rates (3-7mm/h), and rotation speed (10-20 rpm), and it has also no downward support. In crystal growth process, ampoule filled with melt was slowly translated downward with the tapered end towards cold zone, which acts as spontaneous centre of nucleus formation. In the process, melt freezes and self-detaches from the wall of ampoule and spontaneously self-pressure difference develops into the gap due to the differential thermal dilatation. The low solidification rate assists to control the heat flow and heat transfer. v) Ampoule was lowered in 3 hours to 400<sup>o</sup>C, and vi) kept at this temperature for 3 hours for thermal stabilization, vii) Furnace set temperature was lowered to 300<sup>o</sup>C and then switched off for natural cooling. Rotation was continued during growth at the constant speed for congruent mixing of source materials. Experimental optimized growth parameters confirmed from the growth of 72 ingots (InSb, InSbX, GaSb and GasbX). It is investigated that 80% ingots slide out easily from the ampoules, 15% ingots were entrapped into conical region, and 5% ingots were attached to the wall of ampoule. Experimental observations of VDS are explained similar to that used to predict the influence of gravity on detachment.*

## III. RESULTS AND DISCUSSION

### A. Detached ingots growth

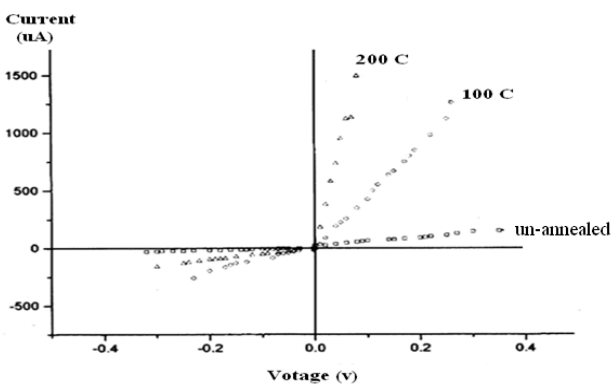
Scientists have put forth the idea of oxidation involvement in detachment; which enhances the melt pollution growing ingots by residual gases, and then an artificial increase of the wetting angle would be a possible. Therefore, semiconductor materials can present very high contact angles on ampoule due to slight amounts of pollution by residual gases. This phenomenon is proposed to explain the detachment of the crystals grown under microgravity conditions in smooth crucibles. In addition, impurities have been involved in detached solidification by increasing the melt-ampoule contact angle, which also should favor the detachment. In VDS experiments, InSb and GaSb and its doped bulk crystals were grown in a sealed quartz tube ampoules by constant backfilling pressure of an argon gas. The system contains three phases: liquid, solid and gaseous (backfilling gas). Detached phenomenon is focused on the crystal growth - *without the seed, without contact with the ampoule wall, without coating, and without applied external gas pressures.* It reveals that the process is self-detached and self-controlled pressure difference. Thus the pressure is slightly higher in the gap than the sum of the top gas pressure and hydrostatic pressures. In our case the most likely impurity that interacts strongly with molten material is believed to be oxygen. This is because numerous VDS experiments showed the thin oxide layer on the inner wall of the ampoule and at the end i.e. cap of the ingots. Another very important observation in our ingots growth is attached growth. When the ampoule is not sealed with back filled argon gas, and then ampoules were broken to take out ingots. These growths showed very poor

crystal quality with the low physical and electronic properties. The consequence of the detached phenomenon is the extraordinary improvement of crystal quality and the defect density is reduced drastically with drastic improvement in physical and electronic properties of InSb and doped InSb ingots [12-28].

### B. Devices applications

**Ion implantation:** The detached grown VDS samples have been used for this study and samples have physical data : the size of samples  $10 \times 10 \times 0.30 \text{ mm}^3$ , complex (p-n- type) InSb, carrier concentration  $\sim 2 \times 10^{16}$  to  $1.8 \times 10^{17}$ , mobility  $\sim 6.0-1.2 \times 10^4 \text{ cm}^2/\text{V.S}$ , FWHM  $\leq 100$  arcsec, and dislocating density  $\leq 1 \times 10^3 \text{ cm}^2$ . The current-voltage (I-V) curves of two implanted doses are shown in Fig- 1, 2. *The effect of annealing on the ideality value (n) of the p-n junction formed by ion-implantation at a dose of  $2 \times 10^{13} \text{ 30 keV Te} + \text{cm}^{-2}$  and annealing temperature, R.T.  $100^\circ\text{C}$ ,  $200^\circ\text{C}$ , is shown in Fig-1* The ideality factor was  $n = 1.08$  for sample annealed at  $200^\circ\text{C}$  for 10 minutes. *The effect of annealing on the ideality value (n) of the p-n junction is shown in Fig-2 formed by ion-implantation at a dose of  $1.8 \times 10^{14} \text{ 30 keV Te} + \text{cm}^{-2}$  and annealing Temperature, R.T.  $100^\circ\text{C}$ ,  $200^\circ\text{C}$ , the ideality factor was  $n = 1.1$  for sample annealed at  $200^\circ\text{C}$  for 10 minutes.* The increase in reverse voltage drop at higher annealing temperatures in both samples may be due the reduction in generation-recombination process that indicates the detached crystal has high quality all along the growth direction, i.e. axial direction.

**Tellurium diffusion:** The p-n junction diode is fabricated by diffusion of tellurium into p-type InSb and the I-V characteristics measured of the p-n junction diodes with dot dimension  $d = 500 \mu\text{m}$  for tellurium diffusion. From reverse bias (below -200 mV), the plots show relatively flat I-V characteristics indicating a generation-recombination-limited (G-R) current transport and presence of diffusion current. The leakage current by the Shockley-Read-Hall (SRH) has been significantly reduced, which is caused by the traps in the depletion region and the surface leakage similar to the fig-1, 2. At reverse bias voltages above -200 mV, the plots exhibit some leakage current in the diodes with a large sensitive area at room temperature for the sample un- annealed.



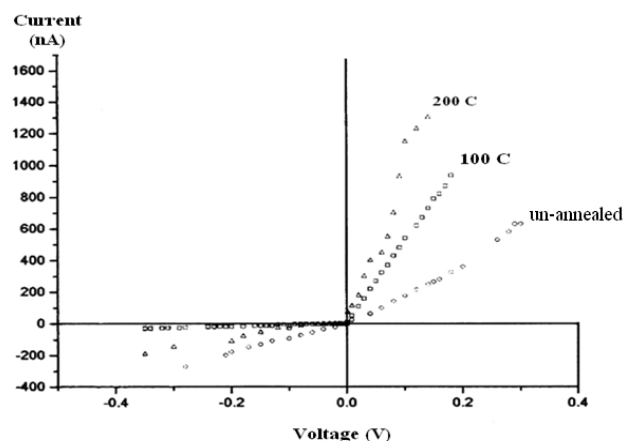
**Fig-1** The room temperature I-V characteristic of p-type InSb grown by VDS and sample was implanted  $2 \times 10^{13} \text{ 30 keV Te} + \text{cm}^{-2}$  and annealing Temperatures - un-annealed,  $100^\circ\text{C}$ ,  $200^\circ\text{C}$  for 10 minute annealing time. Ideality factor (n) improved with increase annealing temperature and the leakage current is reduced with increasing annealing time.

**MOS structure:** The Schottky diode is fabricated by anodic oxidation method. I-V characteristics of different diode fabricated from different axial positions of the ingot have been investigated. It is necessary to mention that the crystals obtained from our laboratory are p-type and tellurium doped ingot is n-type conductivity. The comparisons of I-V curves of the Schottky junction diode structure obtained from two different ingots are reported. We can observe that the I-V curves of all the detectors display Schottky contact behaviors. However, the barrier height of various diodes presents different behaviors according to the region along the ingot direction. It shows a bigger reverse saturation current than the ideal diode; however, the entire I-V curve shape is more important than one region, and the forward bias region is very similar to the ideal diode. Besides the reverse current in which growth layer process anodic oxidation method was employed, the reverse saturation current was improved.

**Detached growth process in VDS:** Argon filled sealed ampoule is placed in hot zone of furnace vertically. When ampoule is lowered from hot zone and reaches the melting point of materials, tiny melt freeze and form solid; due to capillary effect, gets self-detachment from the wall of the ampoule, and it acts as a seed. In this position, the trapped argon gas fills this space and creates self-pressure difference. However this process continues till all melt solidify.

### C. Physics behind the detached growth

Detached growth was first investigated by NASA Skylab Mission III and IV (1974) in microgravity, but yet its mechanism is poorly understood and still it is assumed that it happen spontaneously. The results of experimental microgravity are used to obtain the detached growth in terrestrial laboratory since last decade. In the process of crystal growth by VDS, the detached growth has been observed since 1994. The physics behind VDS grown detached crystal is discussed in this paper.



**Fig-2** The room temperature I-V characteristic of p-type InSb grown by VDS and sample was implanted  $1.8 \times 10^{14} \text{ 30 keV Te} + \text{cm}^{-2}$  and annealing Temperatures - un-annealed,  $100^\circ\text{C}$ ,  $200^\circ\text{C}$  for 10 minute annealing time. Ideality factor (n) improved with increase annealing temperature and the leakage current is reduced with increasing annealing time.

In addition, when tiny melt solidify, then its volume increase as density of the melt is larger than solid, then, increased volume try to go away from interface into melt. Therefore, this process of solidification is against the gravity and



molecules are under negative gravitational effect. When the solid is away from the wall, the gap is filled with trapped gas then meniscus forms between the wall of the ampoule and solid material due to capillary effect. In the beginning of growth, shape of meniscus is concave (seen from melt top). The meniscus concavity decreases as the melt height decrease and gas pressure into gap increases, then meniscus shape become straight.

In this position, further decreases of melt column, meniscus convert straight then into convex shape. Thus the meniscus conversion concave to straight to convex shape is a self conversion process for the detached crystals under terrestrial conditions. The meniscus shape is predicted on experimental evidences because it cannot be seen apparently. All ingots showed the convex cap with thin blackish layer at the end growth and also inside on the inner wall of ampoules as described in our references. *In the presence of gap, thermal gravitational convection (Rayleigh) during ingots growth in terrestrial laboratory decreases, which is key factor during ingots growth, thus impurity in-homogeneities distribution at the micro and macro level growth is reduces in crystal grown under these conditions. In view of the gap, crystals physico-chemical properties in the absence of contact between the ampoule wall and ingots reduce the negative effect of the ampoule wall on the growth, crystal purity and structure of the grown crystals. However, defects and dislocation density decreases significantly in detached grown ingots.* Therefore, detached crystals grown by VDS showed that the high quality substrates suitable for the device construction at the ambient conditions. The three types of gap variations are described in subsequent paragraphs.

**Gap width variation:** The three gap width variations along the growth direction have been investigated, i) constant gap, ii) increases gap and iii) decreases gap. Physics behind these variations is reported here. *The laws of physics are similar within same inertial frame.* Therefore, our experimental data and growth results are correlated with models proposed by William Wilcox, Thierry Duffar, Jeffrey Derby and their co-workers in [7]-[11]. However, the physics behind this concept is discussed on the basis of physical qualitative model -*The steady and stable meniscus and interface model: A model derived from experimental results of the VDS experiments.* The experimental results of crystals grown by VDS are compared with theoretical models based on the principals such as gas enters into gap, oxide thin layer, thermal effect, interface shape, and thermocapillary effect. These basic principles of the concepts are in good agreement with the VDS experimental results of the detached growth. VDS model is entirely based on growth results obtained from the 72 ingot grown since 1994. The gaps thicknesses (e) and convexity height i.e. cap of the ingot were measured physically. The gaps thickness (e) is used to determine the contact angle ( $\Theta_c$ ), pressure difference ( $\Delta p$ ) and meniscus height (h) by following equations cited in references.

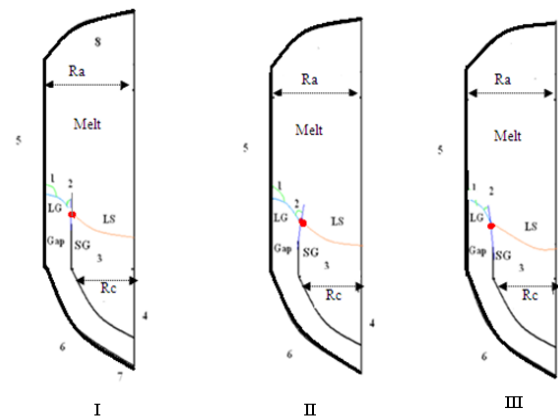
$$i) e = \frac{Ra}{\cos \theta_c} (\cos \theta_c + \cos \alpha e),$$

$$ii) e = \frac{2\gamma}{\Delta p} (\cos \theta_c + \cos \alpha e),$$

$$iii) h = \frac{2\gamma \cos \theta_c}{\rho g Ra}$$

Where  $e = R_a - R_c$ : the gap between the ampoule radius ( $R_a$ ) and the crystal radius ( $R_c$ ),  $\theta_c$ : Young' thermal contact angle on the surface of the ampoule,  $\alpha$ : the growth angle on the surface of the ingot,  $\gamma$ : the melt surface tension,  $\Delta p$ : pressure difference between top and bottom in the ampoule, h: the meniscus height,  $\rho$ : the melt density, g: the gravitational acceleration. The schematic geometric drawing model and all the parameter description is shown in Fig-3, 4. The symbols used in this model are explained here. The three a, b, c vector (dark arrow) meet at the triple point, where crystal, melt and gas phase intersect and forms the Triple-Phase-Line (TPL) i.e. tirade point, red solid dot in Fig-3 and as hollow point 'O' in Fig-4. Let the vector lengths a, b, c are interfaces free energies. The phase angles between these interfaces vectors are  $\Theta_1$ ,  $\Theta_2$  and  $\Theta_3$ . The co-existence of these tirade vectors occurs at crystal surface along the circular TPL at the point O, where the meniscus and interface meet circular shape or periphery of the growing ingot. The phase angles gives  $2\pi = \Theta_1 + \Theta_2 + \Theta_3$ . Let us call this as dynamic phase frame and the total interfacial energy of this frame is constant with respect to the movement of the point O along the growth direction (z-axis) and radius of the crystal (r-axis). However, the meniscus and interface are steady and stable at the crystal–melt interface boundary, because the set temperature maintained constant throughout experiment. Growth mechanism at micro level is not steady and stable, but produces movement effect at tirade point O. Thus the shift at the tirade point O along the TPL is predicted by the change of phase frame in three dimensions as shown in Fig-3, 4. From experimental data, three gap width variation occurrences have been investigated. Three process of the gap variation for InSb and GaSb detached ingots are i) the gap constant, ii) the gap increase and iii) the gap decrease as shown in Fig-3, 4. The established movements are i) Gas pressure in a gap, ii) Thermal capillary effect, iii) Thermal field, iv) Thermocapillary effect v) Reduction of a free surface at the melt (meniscus), which reduce the Marangoni convection, and v) Externally applied growth conditions. Thus, Young's thermal contact angles, gap width, radius of the crystal and pressure difference into gap are involved. However, physics behind gap variation is discussed.

The Physics behind the detached growth in VDS



**Fig.3** The geometrical representation into ampoule during the growth and its schematic description of model geometry 1:  $\Theta_c$  – Young thermal contact angle, 2:  $\alpha$  – Ingot growth

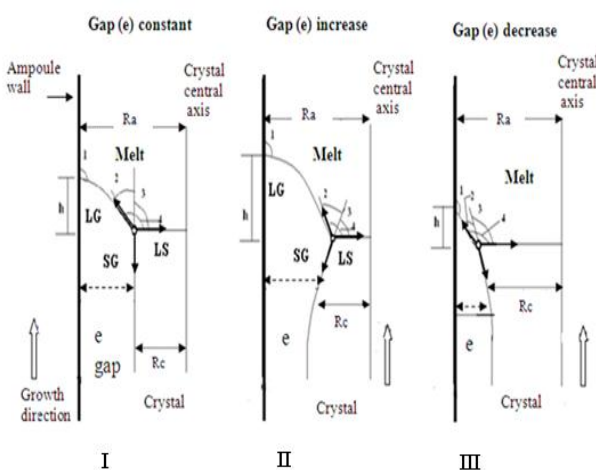
angle, 3: Rc- Ingot radius, 4: Ampoule wall, 6: Conical region, 7: Cone angle, 8: Filled argon gas, Ra: Ampoule radius, L: Liquid, G: gas, S: Solid then the tirade vectors at LG (Liquid-Gas surface), LS (Liquid-Solid surface), SG (Solid-Gas surface) at triple phase line (TPL). Gap: e, Ampoule radius (Ra), Crystal radius (Rc). Let vector along the SG – a (along crystal surface), LS – b (along interface) and LG - c (along meniscus),  $\Theta$ : Angle (in melt) between SG and LS,  $\Theta_1$ : Angle (into melt) between LG and LS,  $\Theta_2$ : Angle (into gap) between LG and SG,  $\Theta_3$ : Angle (grown crystal side) between SG and LS.

**Gap constant:** Fig.4 (I), Medium growth rate,  $\Theta_c = \Theta_d$ , steady state growth then pressure above the melt ( $P_m$ ) = pressure changes in gap ( $P_{gap}$ ) = constant; then  $\alpha = \Psi - \Theta$  if  $\Theta = 90^\circ$  then  $\Psi = 90^\circ + \alpha$  crystal grows with a constant diameter,  $h =$  constant meniscus height. Therefore TPL phase frame which is steady and no movement along the growth direction. Gap width is nearly constant of the order of 145 $\mu$ m in InSb and 160 $\mu$ m in GaSb ingots. As there is no shift of point O along z-axis or r-axis, then the surface changes related to the induced movement of the TPL has no effect on the surface variation. No any shift of the O at macro-level means extreme stability of mutual orientation of phase boundaries by vector a, b, c. This condition is equilibrium at TPL by three forces a, b, c., where the temperature gradient and surface tension gradient are also stable. At this position,  $\Theta_c + \alpha > \pi$ , this condition could be a stable gap width shown in Fig.4 (I). The crystalline quality along TPL at O gives the surface shape of the growing ingot related to the surface behavior. **Gap increase:** Fig.4 (II), Slow growth rate,  $\Theta_c < \Theta_d$ , gap exceeds then  $P_m < P_{gap}$ ,  $\alpha = \Psi - \Theta$  if  $\Theta =$  decrease  $< 90^\circ$  then  $\alpha$  varies from  $0^\circ$  to  $30^\circ$  crystal grows with increase in diameter, hence increase in meniscus height (h). Therefore TPL phase frame is dynamic and movement in the clock wise along the growth

relation depends on the movement of phase frame. The upward arrow indicates the growth direction. The bold dark line represents the ampoule outer surface.

direction inside melt. Therefore gap width increase, and also temperature gradient increase, surface tension force moves from higher to lower inside melt. In principal, the prediction of the detached growth, we account the  $P_{gap}$  and  $P_m$  changes with the decrease in melt weight, pressure above top ( $P_{top}$ ) filled argon gas, which is a constant, and hydrostatic pressure ( $P_{hyd}$ ) changes with the decrease in melt column height. The hydrostatic pressure  $P_{hyd} = \rho_1 g H$ , where, H: height of melt column in ampoule,  $\rho_1$ : density of the melt, g: acceleration due to gravity. Let us predict the relation of the gap width with pressure changes and its effect on the meniscus (melt-gas interface) and crystal-melt interface. As explained in earlier paragraph, there is entrapped argon gas and dissolved gases enter into gap, then pressure into gap increases. Besides this, the decrease in melt weight as well as hydrostatic pressure, thus the pressure into gap increases and also the concavity of the crystal- melt interface decreases. An increase of this pressure has influence on the tirade phase frame at TPL, which in turn induces the rotation effect on the meniscus at point O. This clock wise rotation moves towards the c-axis inside the melt, which increase the gap width as shown second diagram in Fig.4-II. There are other effects on the meniscus, first the temperature gradient increases and second the surface gradient also increases when thermo-capillary effect increases then Marangoni convection become more dominant and it would push inside the melt. It is observed that the as curved microstructures  $> 2$  mm away from the periphery of the samples. Further increases of Marangoni convection is resulted into melt flow patterns and impurity striation into centre of the some samples. For typical ingots, the increase in gap width from the first transition state (after conical shape) to final transition state (convex cap) is measured 65 to 238 $\mu$ m for InSb and 95 to 265  $\mu$ m for GaSb ingots. In detached some ingots seen with the bubbles on the surface, which reveals the high pressure into the gap. It may be the capillarity effect (reduced gravity) into gap.

Gap variation in detached ingot growth into VDS technique



**Fig.4** Enlarged sections of tirade i.e. triple point O (red solid dot in Fig-3). The three process of gap variation are predicted in VDS on the basis of experimental measured gap width. Let  $P_m$ : pressure above melt,  $P_{gap}$ : pressure in gap, 1:  $\Theta_c$ - equilibrium contact angle,  $\Theta_d$ : dynamic contact angle, 2:  $\alpha$  – Ingot growth angle,  $\Theta$ : Angle (in melt) between SG and LS, 4:  $\psi$  - Angle into melt between LG and LS.  $\Psi = \Theta + \alpha$ , this

**Gap decrease:** Fig. 4 (III), Fast growth rate,  $\Theta_c > \Theta_d$ , gap exceeds then  $P_m > P_{gap}$ ,  $\alpha = \Psi - \Theta$  if  $\Theta =$  increase  $> 90^\circ$  then  $\alpha$  varies from  $30^\circ$  to  $0^\circ$  crystal grows with a decrease in diameter,  $h =$  decrease in meniscus height. Therefore TPL phase frame is dynamic and as movement in the anti-clock wise along the growth direction inside gap. The decrease in the gap pressure along the meniscus would result into decrease in gap width, which will move closer to the ampoule wall. In this case, temperature gradient and surface tension gradient decrease fast as the growth rate is fast. For typical ingots, the decrease in gap width from the first transition state (after conical shape) to final transition state (convex cap) is measured 210 to 85 $\mu$ m for InSb and 257 to 95 $\mu$ m for GaSb ingots. Therefore gap width decrease, thermal stresses at the surface of the ingot develops the microstructures (grain, twin and dislocation), when the thermal stress induces on the periphery of the sample. Further fast decrease should result in attached growth.

#### IV. CONCLUSION

The crystal growth by VDS has been initiated in 1994 and 72 bulk detached ingots have been studied for involvement of VDS crystal growth process. Experimental results in our laboratory reveal that the quality of the crystal has been drastically enhanced in detached growth. Moreover, the VDS detached growth crystal showed similar properties of the crystal grown in space. In VDS, the triple phase line (tirade point) at the meniscus moves freely under detached condition in response to dynamic movement of this phase point at the crystal–melt interface. The measured three gaps and similar prediction based on the physics behind the gap variation more relevant and is better agreement with existed models. The grown crystals are more homogeneous, better crystal structural quality and present fewer defects / dislocations. The attempts to prepare electronic devices on substrates cut from these crystals have systematically shown highest improvement and better stability of the electronic and optical properties. The p-n junction reverse bias I-V characteristics showed significant low dark current at the ambient and forward bias showed the ideal diode nature. Ingot growths showed that it is reproducible, reliable and ensures the enhancement in crystal physical properties and useful for devices application with high yield. Encouraging experimental result conclusion foreseen is that VDS would be a truly industrial crystal-growth production system.

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