Influence of Containment on the Growth of Germanium-Silicon in Microgravity

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This investigation involves the comparison of results achieved from three types of crystal growth of germanium and germanium-silicon alloys:

- Float zone growth
- Bridgman growth
- Detached Bridgman growth

The fundamental goal of the proposed research is to determine the influence of containment on the processing-induced defects and impurity incorporation in germanium-silicon (GeSi) crystals (silicon concentration in the solid up to 5 at%) for three different growth configurations in order to quantitatively assess the improvements of crystal quality possible by detached growth.
What is Detached Bridgman Growth?

**Sufficient condition for detachment**: $(\alpha + \theta \geq 180^\circ)$

**Advantages**
- No sticking of the crystal to the ampoule wall
- Reduced stress
- Reduced dislocations
- No heterogeneous nucleation by the ampoule
- Reduced contamination

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Research Motivation

- What are the conditions for detachment in microgravity and how do they depend on the governing parameters?
  - Growth angle
  - Contact angle
  - Pressure differential
  - Bond number (ratio of gravity to capillarity)
- Which detached growth solutions are dynamically stable?
- How does an initial crystal radius evolve to one of the following states?
  - Stable detached gap
  - Attachment to the crucible wall
  - Meniscus collapse
- What are the effects of angular dependence on the crystal shape (faceting effects)?
Detached Crystal Growth
Etch Pit Densities in Detached/Attached Crystals

Completely detached grown crystal UMC7

 Attached grown crystal UMC6

EPD $\approx 200 \text{cm}^{-2}$

EPD $\approx 2 \cdot 10^4 \text{cm}^{-2}$

Schematic Diagram of Detached Solidification

\( \alpha \): growth angle
\( \theta \): contact or wetting angle

Calculation of Meniscus Shapes

Young-Laplace Equation

\[
\frac{d^2 z}{dr^2} + \frac{dz}{dr} \left(1 + \left(\frac{dz}{dr}\right)^2\right)^{3/2} = \Delta P - Bz(r)
\]

\[
\Delta P = \frac{\Delta P_m r_0}{\sigma}, \quad \Delta P_m = P_H - P_C + \rho g h + 2 \frac{\sigma}{r_H}
\]

\[
B = \frac{\rho g_0 r_0^2}{\sigma}
\]

\[
B = 3.248; \text{ Ge, } r_0 = 6 \text{ mm}
\]

\[
B = 4.651; \text{ InSb, } r_0 = 5.5 \text{ mm}
\]

\[
\frac{\partial r}{\partial s} = \cos \beta, \quad \frac{\partial z}{\partial s} = \sin \beta, \quad \frac{\partial \beta}{\partial s} = -\frac{\sin \beta}{r} + \Delta P - Bz
\]

Boundary Conditions

\[
z(0) = 0; \quad \beta(0) = 90^\circ - \alpha;
\]

\[
\beta(1) = \theta - 90^\circ; \quad r(1) = 1
\]

\(\Delta P\): Dimensionless pressure differential across the meniscus

\(B\): Bond number; ratio of gravity force to surface tension force

\(\alpha\): growth angle

\(\theta\): contact or wetting angle
Gap Width vs. Pressure Differential (Ge at 1g)

\[ \theta + \alpha < 180^\circ \]

\[ \theta + \alpha > 180^\circ \]

\[ \alpha = 14.3^\circ \]

\[ B = 3.248 \]
Gap Width vs. Pressure Differential (Ge at $10^{-6} \times g_0$)

$\theta + \alpha < 180^\circ$

$\theta + \alpha > 180^\circ$

$\alpha = 14.3^\circ$

$B = 3.248 \times 10^{-6}$
• “Influence of Containment on the Growth of Silicon-Germanium” (ICESAGE) is a collaborative investigation between NASA and the European Space Agency (ESA)

• The ICESAGE experiments will be conducted in the Low Gradient Furnace (LGF) in the Materials Science Laboratory on the International Space Station (ISS)

• Processing parameters will be varied to assess their affect on detachment
  • Sample Material (GeSi, Ge:Ga)
    ➢ Affects the growth angle
    ➢ Comparison of semiconductor alloy and doped element
  • Inner Ampoule surface material (SiO₂, boron nitride)
    ➢ Affects the contact angle
  • Pressure: positive, negative, or zero (vacuum) gas pressure below the meniscus
Microgravity Effects

• Microgravity reduces the pressure head ($\rho gh$) resulting from the weight of the melt.
  - Detached growth requires that capillary forces dominate over gravitational forces.
  - On Earth, gravity complicates a comparison of detached growth theory and experiment: the pressure head continuously decreases as the melt solidifies and the pressure varies along the height of the meniscus.

• Microgravity allows a larger value of the gap width.
  - On Earth, when the gap width becomes too large, gravity overcomes surface tension, a stable meniscus cannot be maintained, and the melt will flow down between the crystal and ampoule wall.
  - A large initial gap width will allow measurement of anisotropy in the growth angle.

• Microgravity enables a study of the dynamic stability of crystallization independent of thermal effects.
• A Ge$_{1-x}$Si$_x$ ingot is placed inside a pyrolitic boron nitride (pBN) tube and sealed in a SiO$_2$ ampoule.
• The ampoule is placed inside a cartridge which is inserted into the furnace.
• Thermocouples in the cartridge provide for real-time monitoring of the thermal profile.
Positive Pressure Configuration

- When the top of the ingot melts, different pressure volumes are created.
- As the ingot is inserted into the furnace, the pressure above the meniscus increases faster than below the meniscus.
• When the top of the ingot melts, different pressure volumes are created
• As the ingot is inserted into the furnace, the pressure below the meniscus increases faster than above the meniscus
Summary

- Crystals grown by the detached Bridgman method have greatly increased crystalline perfection, motivating a systematic study of the phenomenon
- A theory describing the conditions for detachment has been developed
- Only crystals where $\alpha + \theta > 180^\circ$ are expected to achieve stable detached growth in microgravity
- Reproducible detached growth has been achieved in the laboratory under limited conditions
- Microgravity will allow the study of detachment over a range of parameters not possible to achieve on Earth
- A series of Ge and GeSi crystal growth experiments are being developed for processing on the ISS