29th Annual Meeting of the American Society for Gravitational and Space Research
Orlando, Florida
November 3–8, 2013

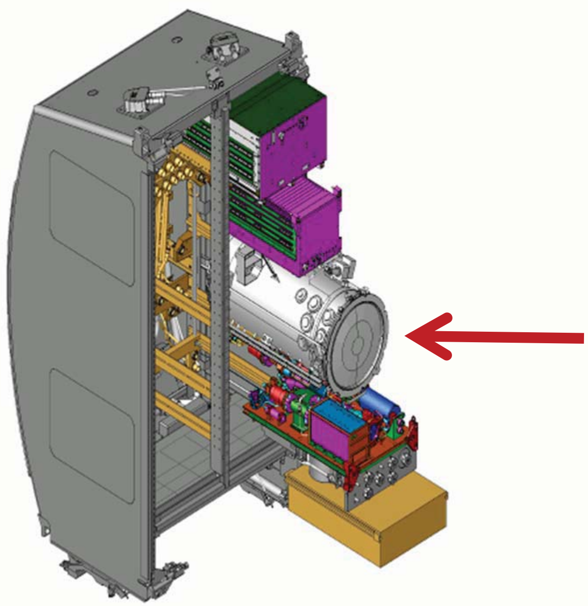
Evolution of the Shape of Detached GeSi Crystals in Microgravity

M. P. Volz[1], K. Mazuruk[2]

[1]NASA, Marshall Space Flight Center, EM31, Huntsville, Alabama, USA
[2]University of Alabama in Huntsville, Huntsville, Alabama, USA

ICESAGE Flight Investigation

- "Influence of Containment on the Growth of Silicon-Germanium" (ICESAGE) is a NASA Materials Science Flight Investigation
- 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)



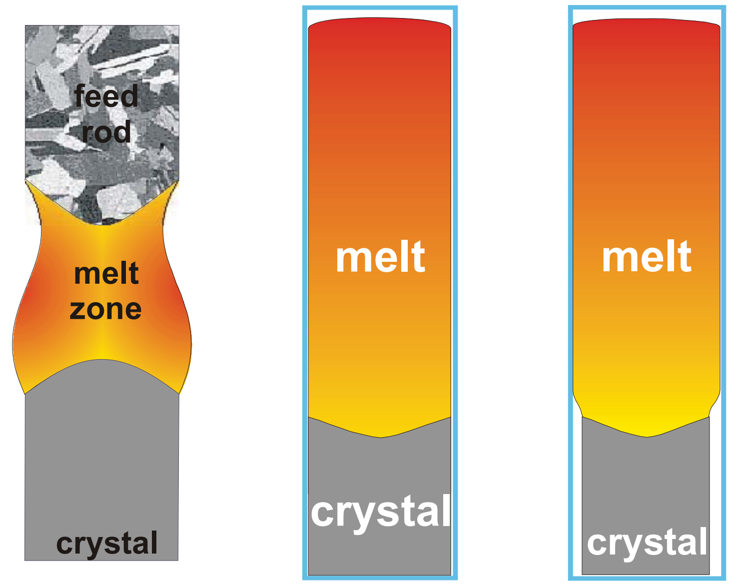
Materials Science Laboratory

MSL Low Gradient Furnace

Overview of the Investigation

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



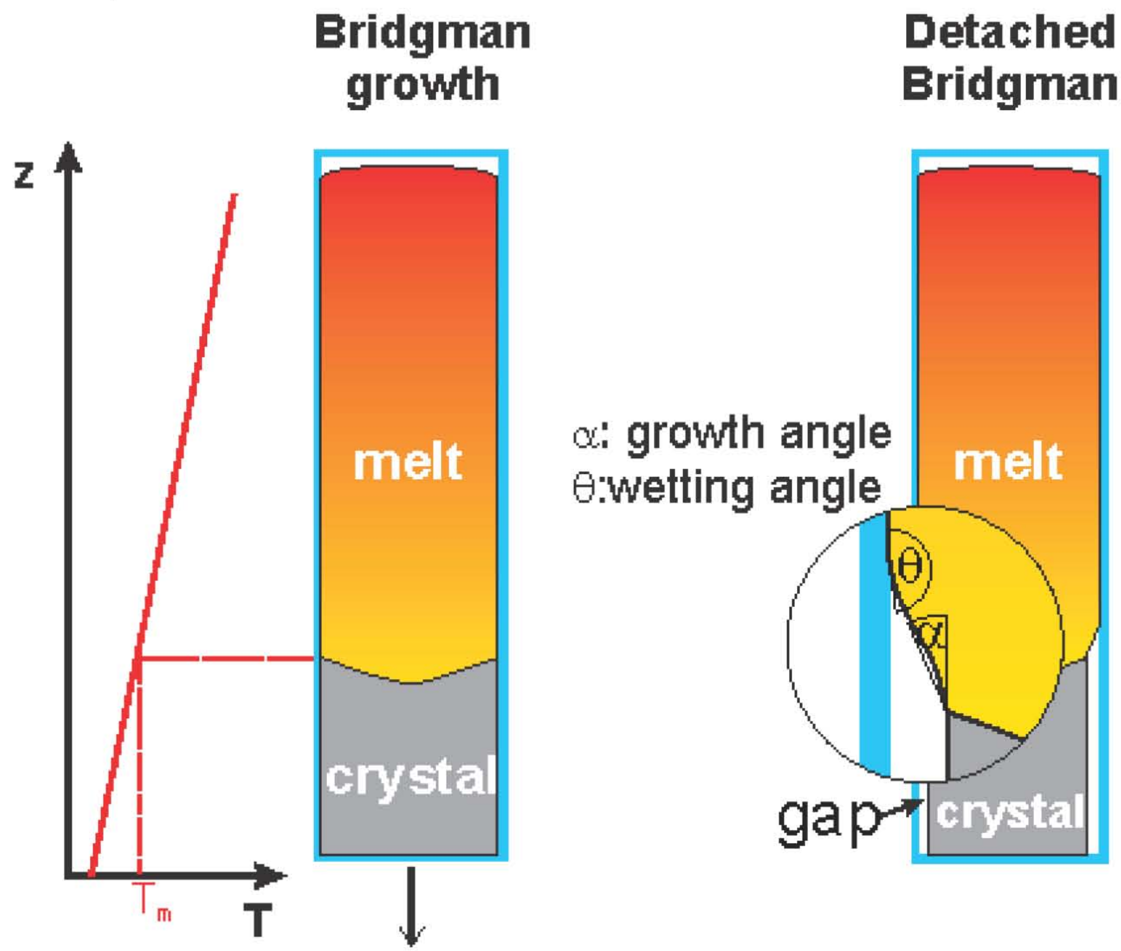
The fundamental goal of the proposed research is to determine the influence of containment on the processing-induced defects and impurity incorporation in germanium-silicon (GeSi) crystals (silicon concentration in the solid up to 5 at%) for three different growth configurations in order to quantitatively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

Why Study Germanium-Silicon Alloys?

- Technological applications
  - X-ray and neutron optics (gradient crystals)
  - High-efficiency solar cell material
  - Thermoelectric converters
  - Increased carrier mobility compared to silicon, but can still be integrated into Si technology
- Characterization methods for silicon and germanium are well-established and are applicable to the alloy crystals.
- Relatively well known material properties and material parameters
- The vapor pressure of silicon and germanium melts can be neglected; they are non-toxic materials.

Principles of Detached Bridgman Growth

Sufficient condition for detachment[1,2]:
$(\alpha+\theta \geq 180°)$



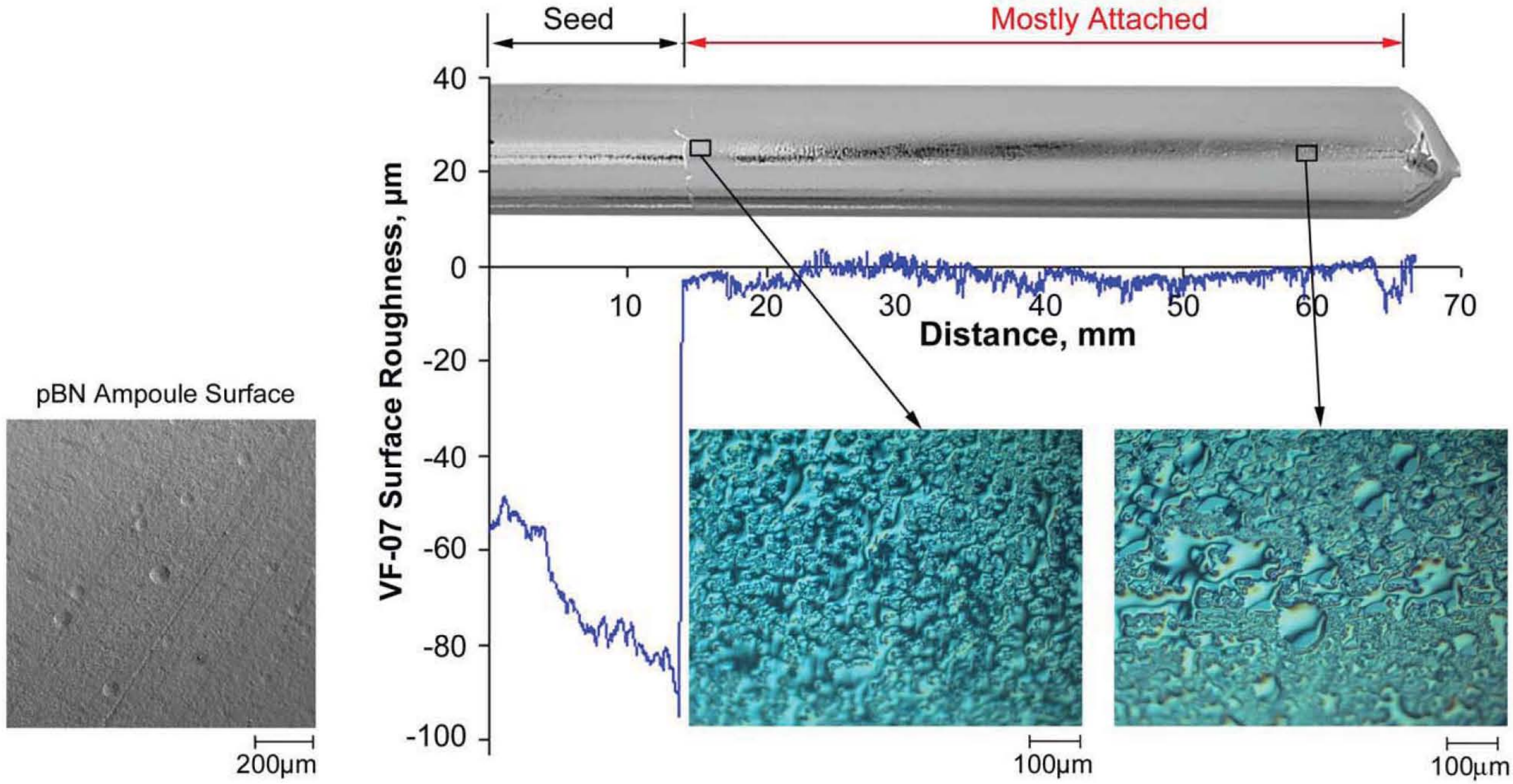
Advantages

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

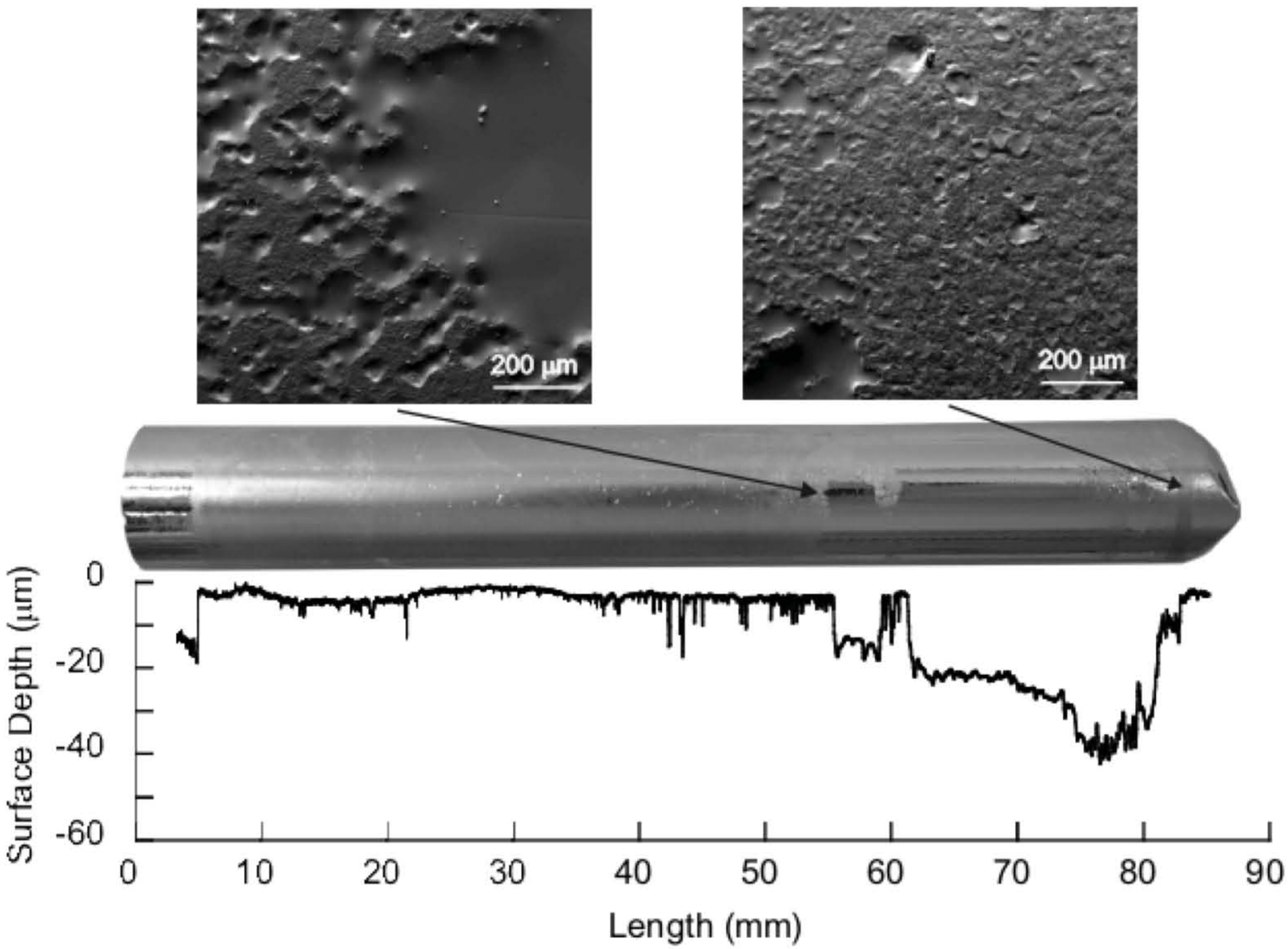
[1]V. S. Zemskov: Fiz. Khim. Obrab. Mater. 17 (1983) 56

[2]T.Duffar, I.Paret-Harter, P.Dusserre: J.Crystal Growth 100 (1990) 171.

“Attached” Germanium

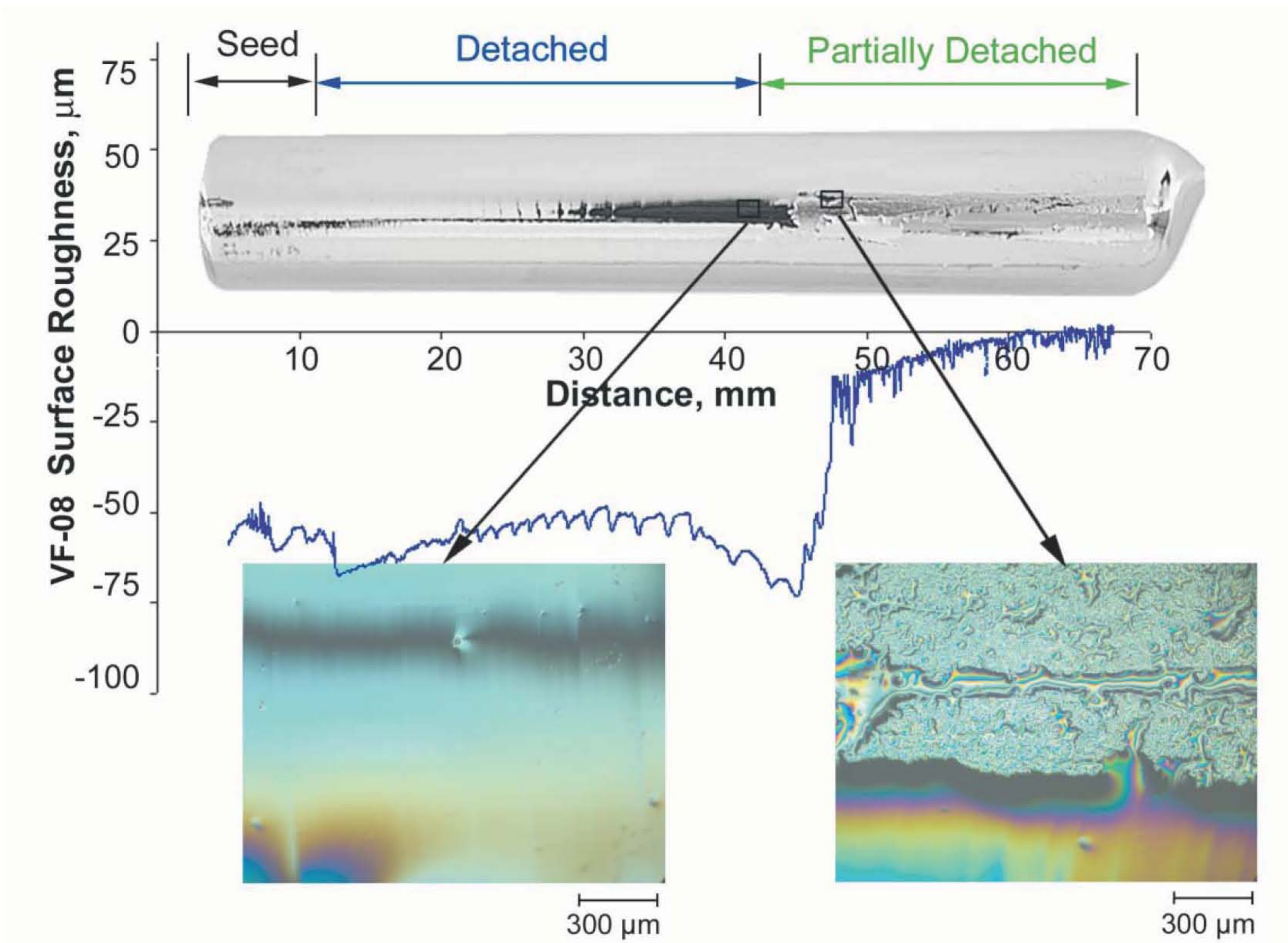


Partially Attached GeSi



M. P. Volz, M. Schweizer, N. Kaiser, S. D. Cobb, L. Vujisic, S. Motakef, F. R. Szofran, *JCG* 237-239 (2002) 1844-1848

Detached Ge in pBN Ampoule



Etch Pit Densities in Detached/Attached Crystals

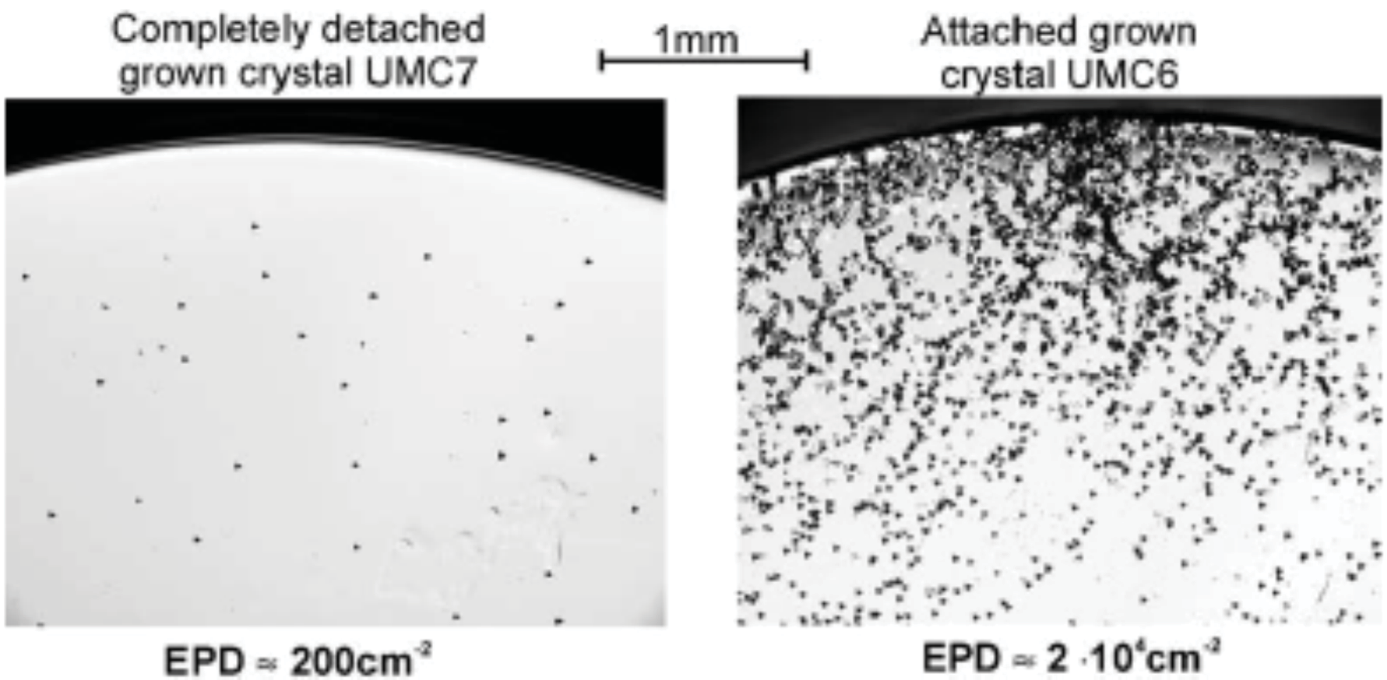


Fig. 5. Micrograph from the detached-grown sample UMC7 and from the attached-grown sample UMC6.

M. Schweizer, S. D. Cobb, M. P. Volz, J. Szoke, F. R. Szofran,
JCG 235 (2002) 161-166

Etch Pit Density Variation With Attachment

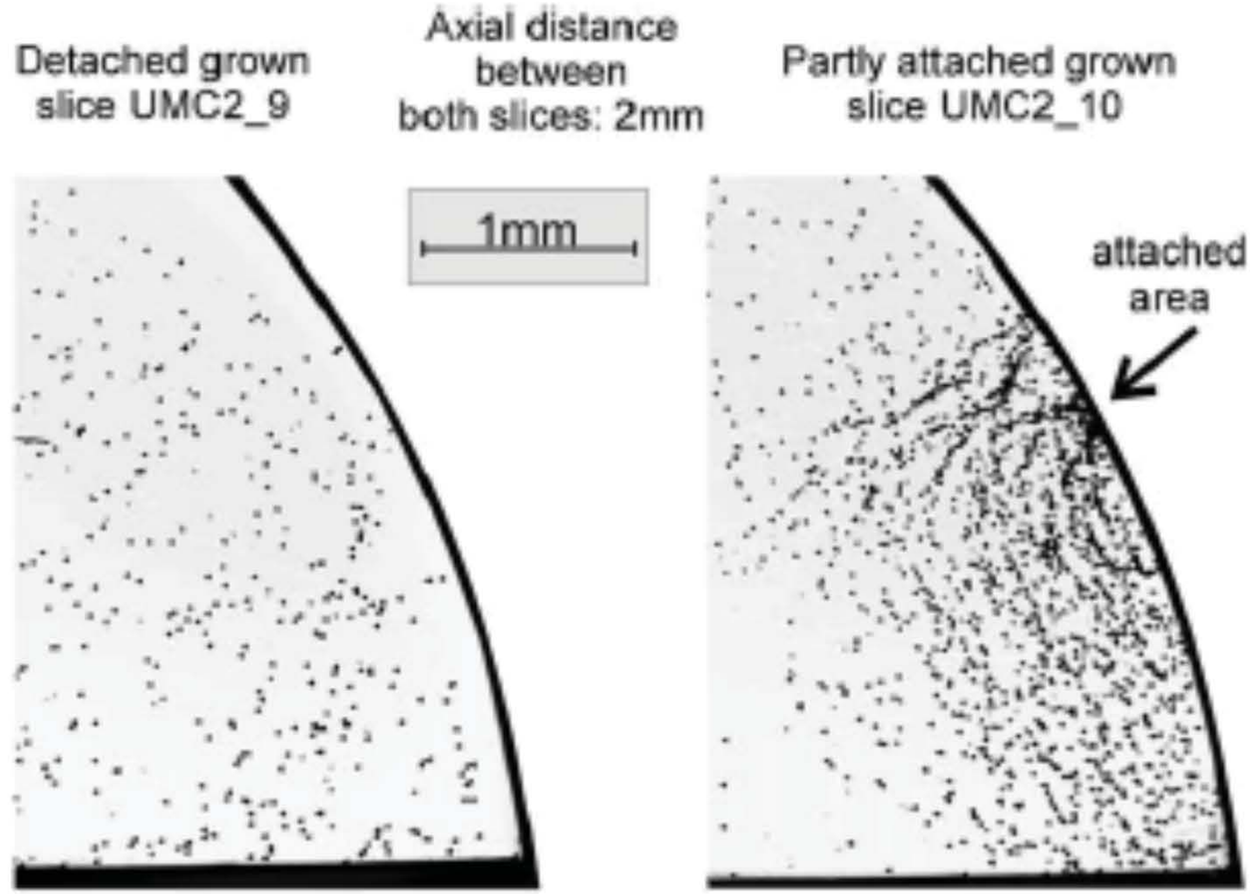
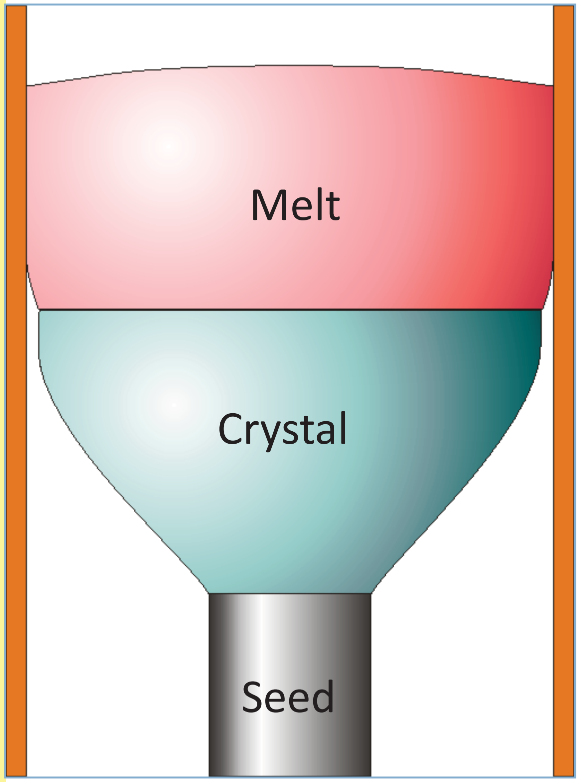


Fig. 6. Localized increased EPD after the crystal attaches partially to the wall.

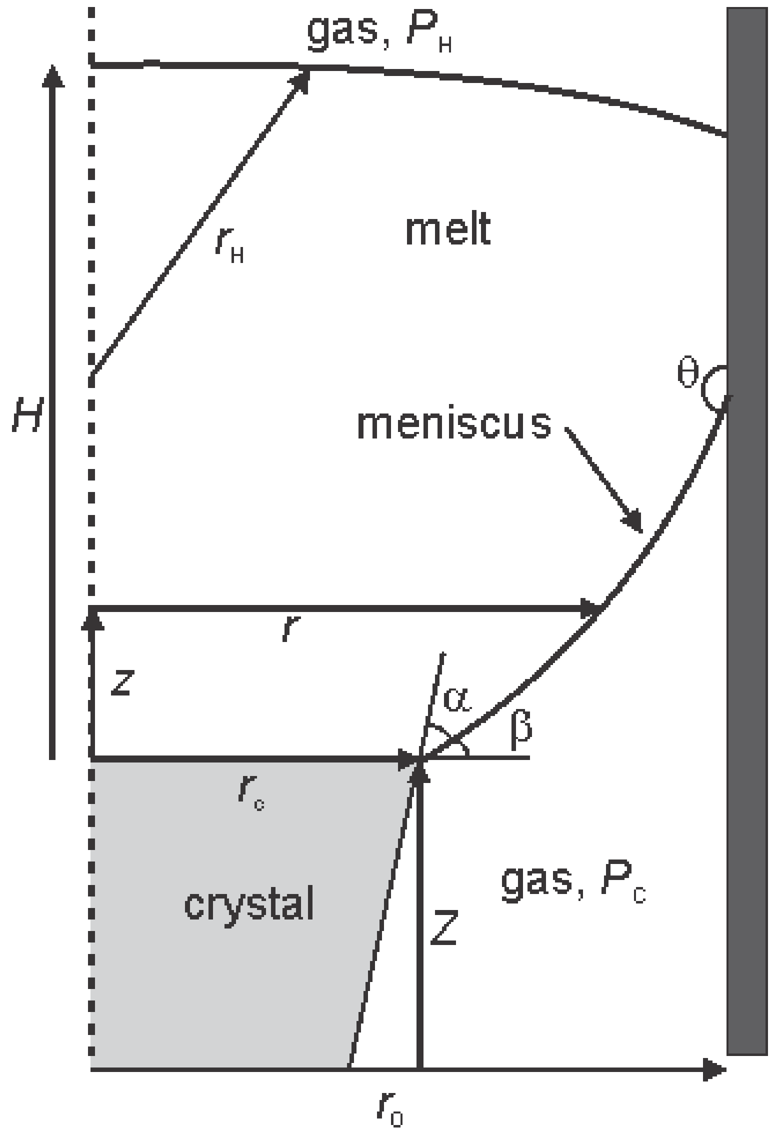
M. Schweizer, S. D. Cobb, M. P. Volz, J. Szoke, F. R. Szofran, JCG 235 (2002) 161-166

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)?



Schematic Diagram of Detached Solidification



α: growth angle
θ: contact or wetting angle
ΔP : Dimensionless pressure differential across the meniscus
$z(r)$: meniscus shape
$Z(r)$: crystal shape

$$\Delta P = \Delta P_{external} + \rho g h + 2\frac{\gamma}{r_{tm}}$$

where

$\Delta P_{external}$: external gas pressure differential

$\rho g h$: weight of melt (pressure head)

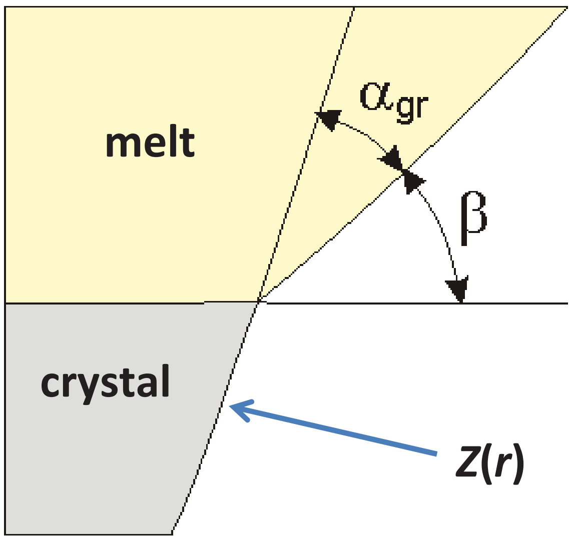
$2\frac{\gamma}{r_{tm}}$: capillary pressure from top meniscus

Equations in Zero Gravity

$$\frac{\partial z}{\partial r} = \pm \frac{\Delta P(r^2-1)-2\cos\theta}{\sqrt{4r^2-(\Delta P(r^2-1)-2\cos\theta)^2}}$$

Meniscus shape equation, *z*(*r*): 2 possible solutions for *g* = 0, *B* = 0

$$\frac{dZ}{dr} = \tan(\alpha+\beta)$$



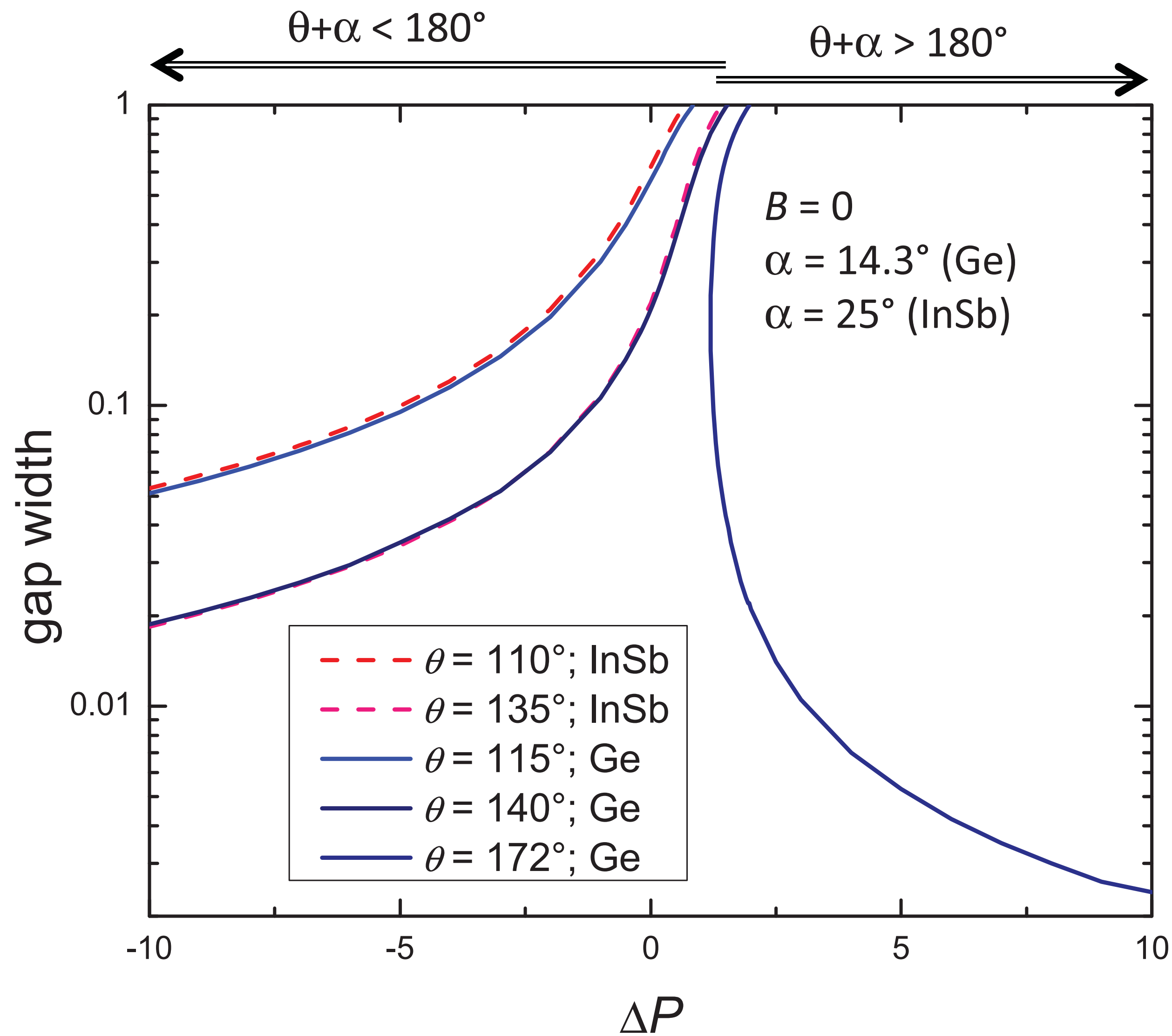
Crystal shape equation, *Z*(*r*): 2 possible solutions in zero gravity

$$\frac{dZ^{\pm}}{dr} = \frac{\sqrt{4r^2-y^2}\,\tan\alpha \pm y}{\sqrt{4r^2-y^2} \mp y\tan\alpha}, \qquad y = \Delta P(r^2-1)-2\cos\theta$$

Gap Width vs. Pressure Differential (Microgravity)

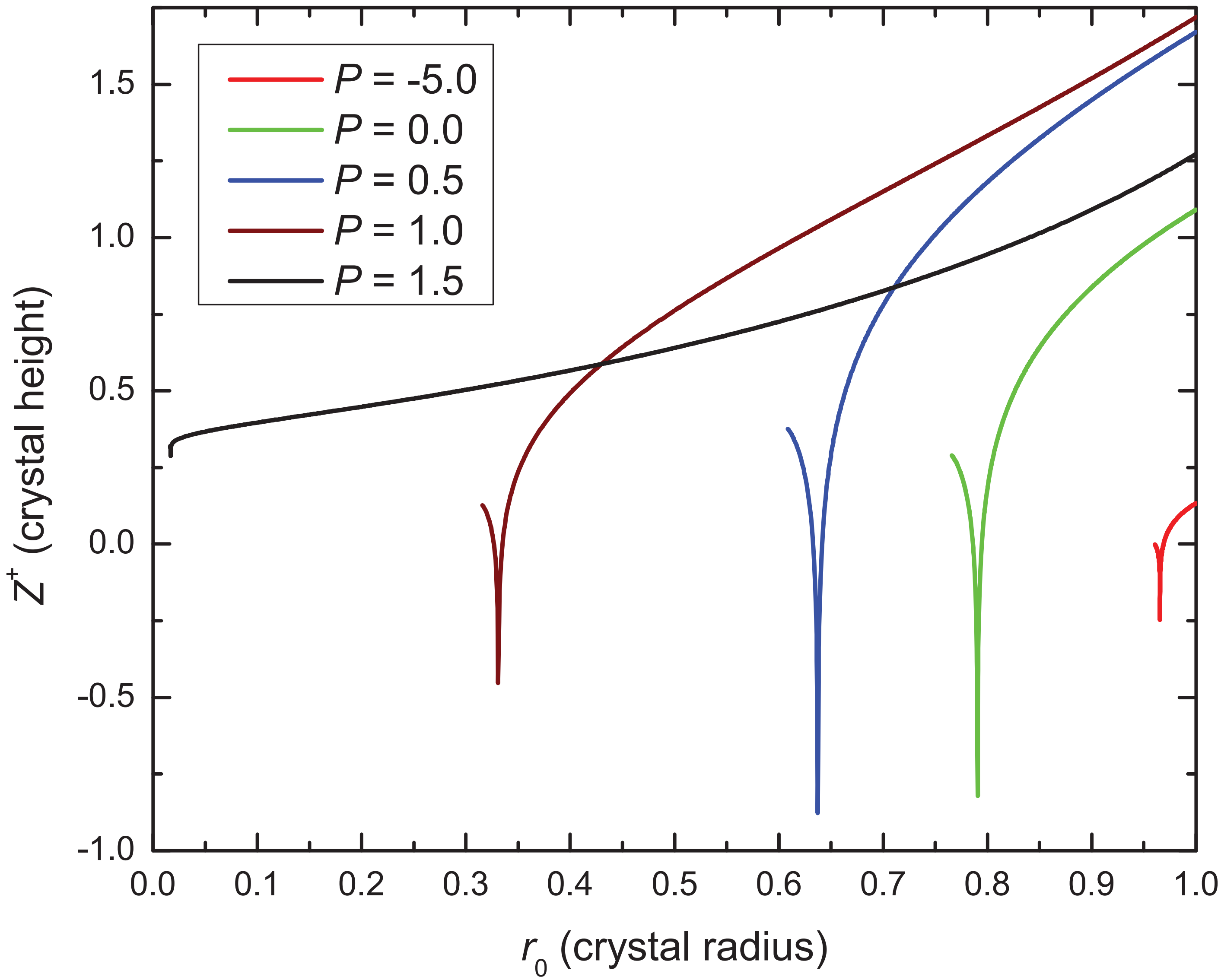
Equilibrium solutions

$$\frac{dr}{dZ} = 0$$



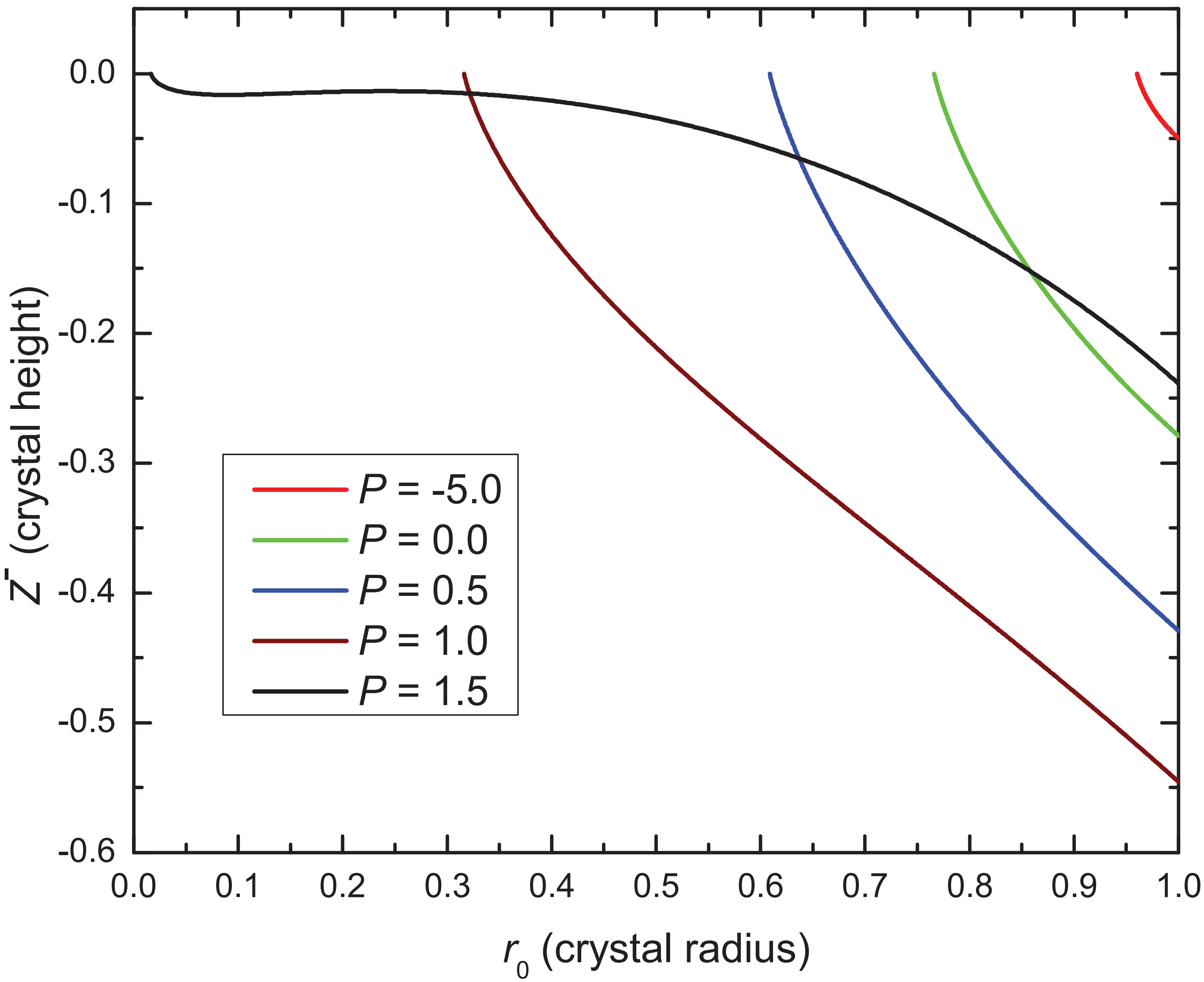
Ge Crystal Evolution for θ = 140°; Z^{+} solution

Ge
α= 14.3°
θ = 140°



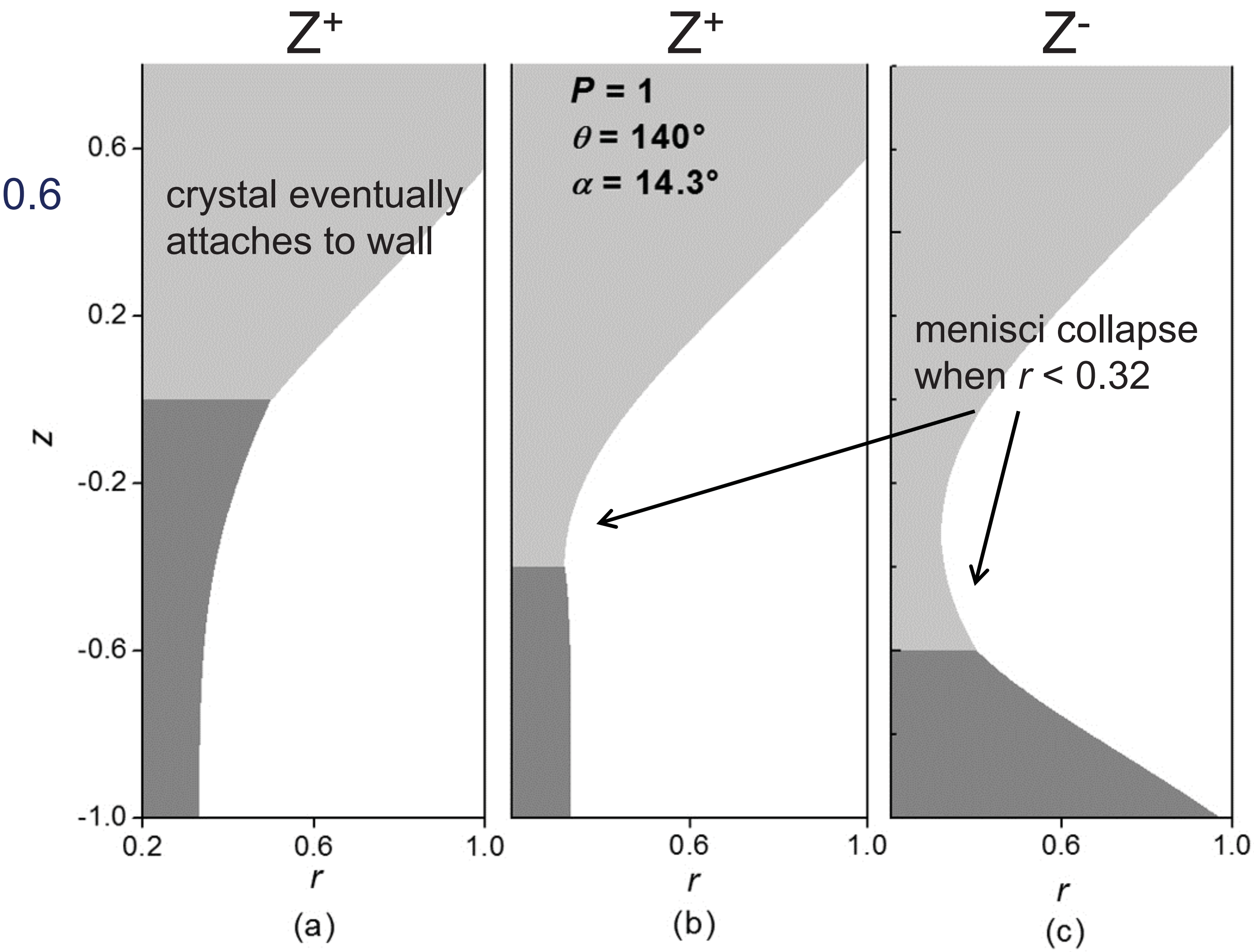
Ge Crystal Evolution for θ = 140°; Z^- solution

Ge
α= 14.3°
θ= 140°



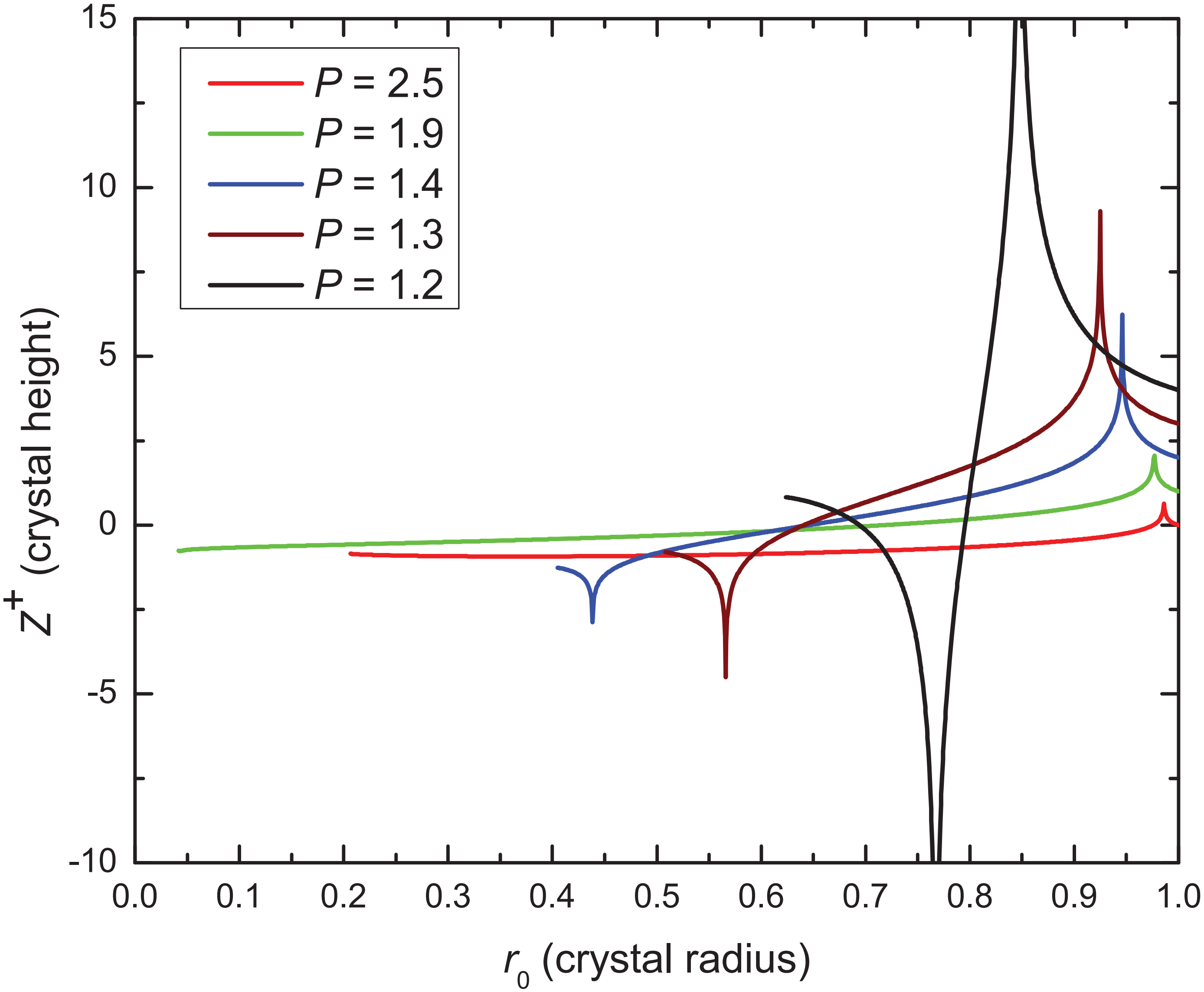
Evolution of Crystal Radii for $\theta = 140°$

Material: Ge
Starting position, r_0: 0.6
Growth Angle: 14.3°
Contact Angle: 140°
$\Delta P = 1$



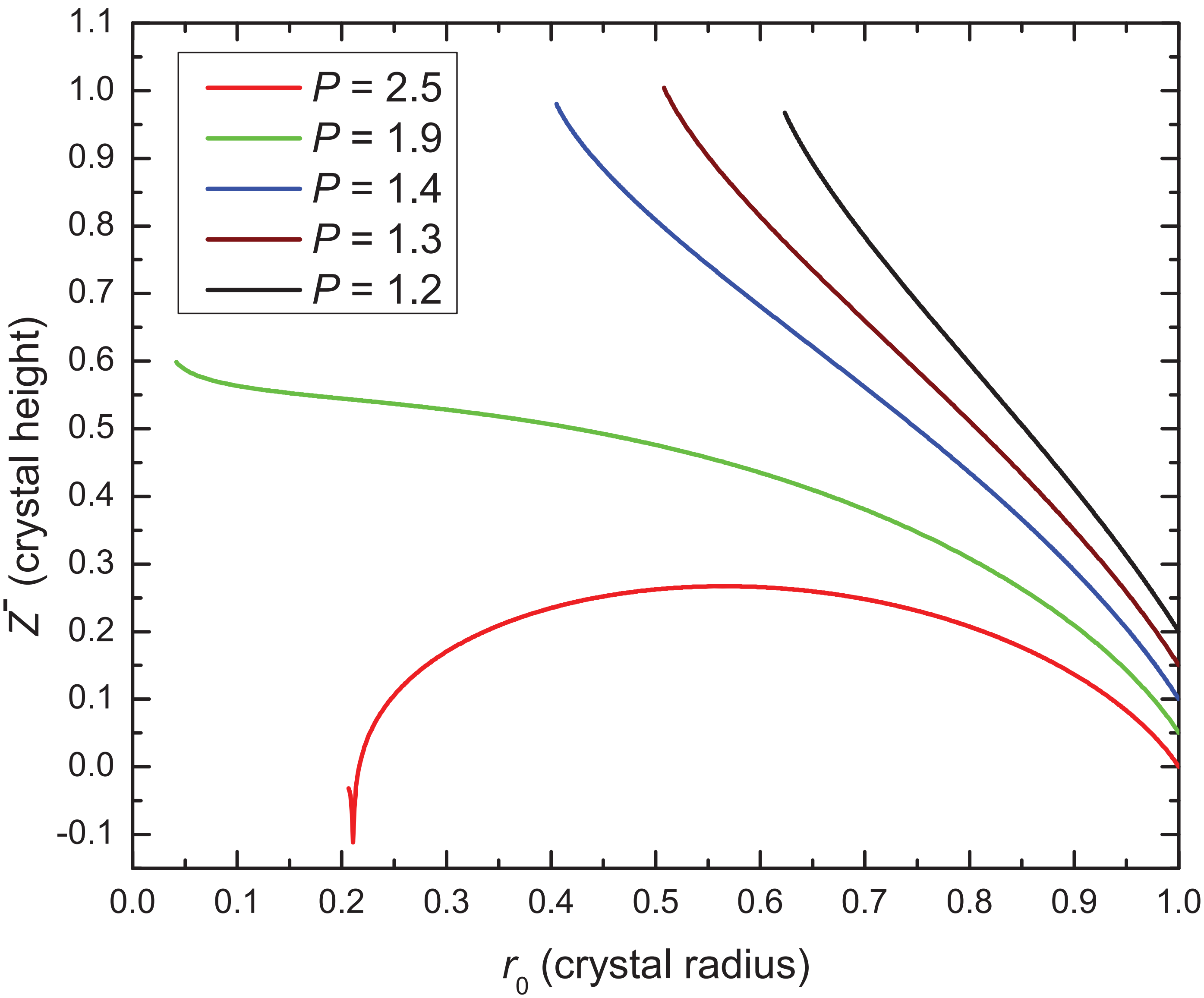
Ge Crystal Evolution for $\theta = 172°$; Z^+ solution

Ge
$\alpha = 14.3°$
$\theta = 172°$



Ge Crystal Evolution for $\theta = 172°$; Z^- solution

Ge
$\alpha = 14.3°$
$\theta = 172°$



Summary

- A theory describing the shape evolution of detached Bridgman crystals in microgravity has been developed
- A starting crystal of initial radius r_0 will evolve to one of the following states:
  - Stable detached gap
  - Attachment to the crucible wall
  - Meniscus collapse
- Only crystals where $\alpha + \theta > 180°$ will achieve stable detached growth in microgravity
- Results of the crystal shape evolution theory are consistent with predictions of the dynamic stability of crystallization (Tatarchenko, *Shaped Crystal Growth*, Kluwer, 1993)
- Tests of transient crystal evolution are planned for ICESAGE, a series of Ge and GeSi crystal growth experiments planned to be conducted on the ISS

Contributing Personnel

Prof. Arne Cröll (ESA Team Lead), Kristallographisches Institut, Freiburg, Germany
Dr. Frank R. Szofran (original U.S. PI), NASA MSFC, Huntsville, AL USA
Dr. Peter Dold (original German PI), Kristallographisches Institut, Freiburg, Germany
Prof. Dr. Klaus Werner Benz, Kristallographisches Institut, Freiburg, Germany
Dr. Sharon D. Cobb, MSFC, Huntsville, AL USA
Dr. Natalie Kaiser, Kristallographisches Institut, Freiburg, Germany
Mr. Uwe Kerat, Kristallographisches Institut, Freiburg, Germany
Ms. Alina Mitric, Kristallographisches Institut, Freiburg, Germany
Dr. Shariar Motakef, CAPE Simulations, Inc., Newton, MA USA
Dr. Witold Palosz, USRA/MSFC, Huntsville, AL USA
Ms. Penny Pettigrew, Morgan Research/MSFC
Dr. Markus Schweizer, USRA/MSFC, Huntsville, AL USA
Dr. Ljubomir Vujisic, Cape Simulations, Inc., Newton, MA USA
Prof. John S. Walker, University of Illinois at Urbana-Champaign, Urbana, IL USA

Technical Support
C. Bahr/D. Lovell/J. Quick, Morgan Research/MSFC
P. Carpenter/C. Cochrane, USRA/MSFC
S. Gallop, Tec-Masters/MSFC