

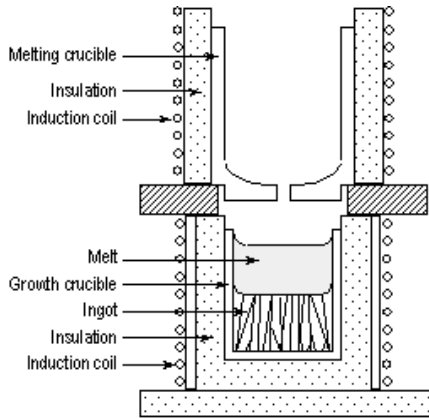
**Study of the influence of forced and natural convection on impurity segregation and coating stability in the ingot growth of multicristalline Silicon for photovoltaic applications**

**Daniel Vizman**

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**~2ST IFA-CEA SYMPOSIUM, Bucuresti 2012~**

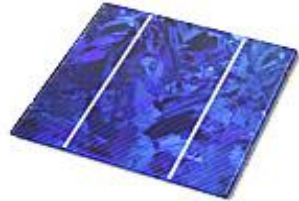
# Crystals for solar energy



Silicon ingot



Silicon wafer

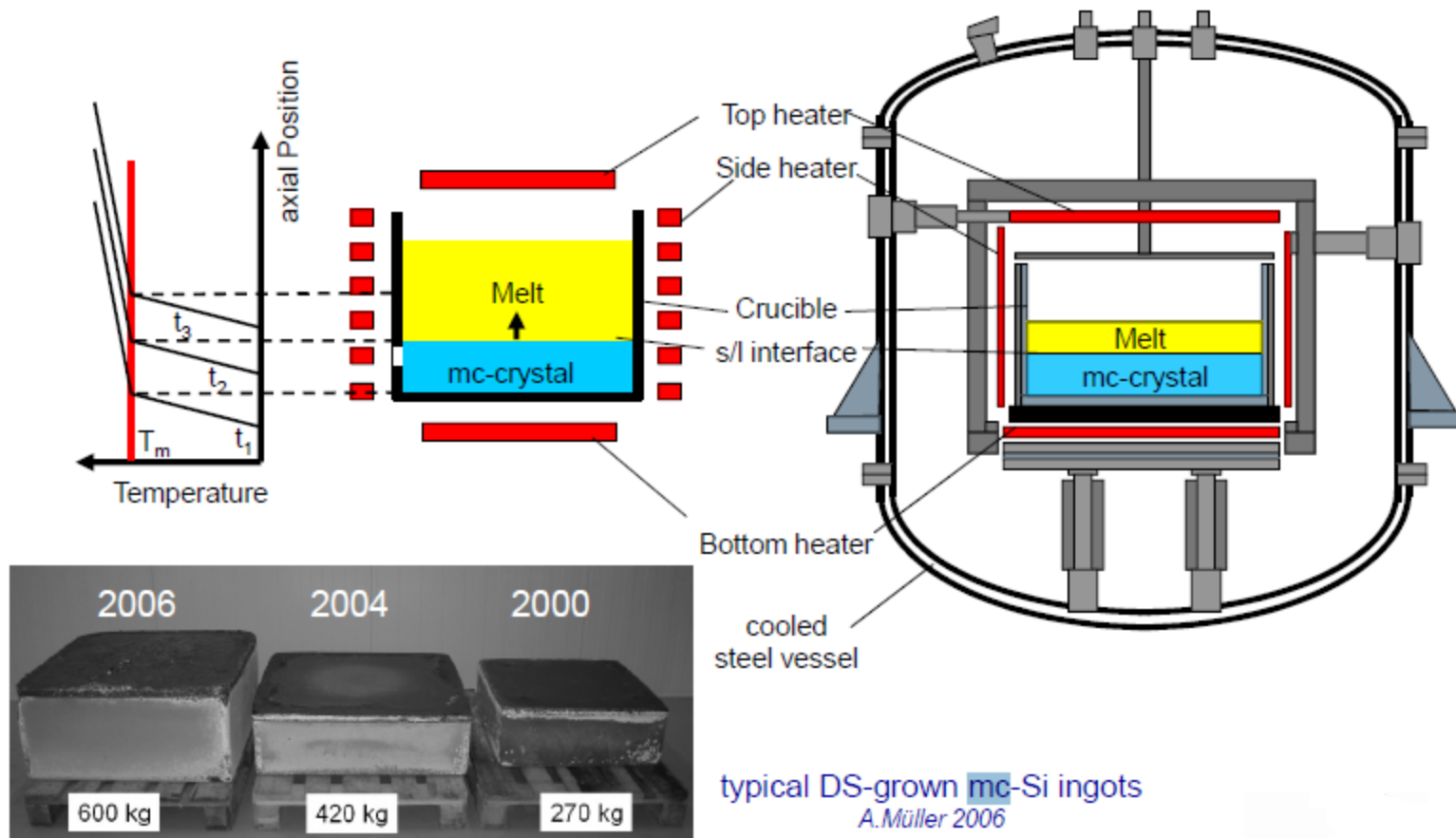


Silicon solar cell



Conversion of sunlight into electric power

# Multicrystalline Silicon ingot growth by Directional solidification/Casting



- today's research is on 800 kg ingots, but there is no reason to think that this represents an ultimate limit

- use of silicon feedstock of lesser purity than standard electronic grade material

# One major issue : Control of convection

**•Increasing  
crucible size**

**•Detrimental curvature  
of S-L interface**

**•Turbulent melt flow ??**

**•Cohesion of crucible  
coatings**

**•Feedstock of  
lesser purity**

**•Optimal segregation of  
the impurities**

**•Achievement of a total  
mixing regime**

# Research objectives

- **Understanding of melt convection particularities**
- **Study of the influence of melt convection on interface shape, impurities segregation and coating stability**
- **Investigation of different methods to tailor the melt convection (like mechanical stirring)**
- **Numerical simulation is an efficient tool to improve the understanding and the control of the heat and mass transport phenomena occurring during the crystallization process**

# CONSIL Project

## Partners:

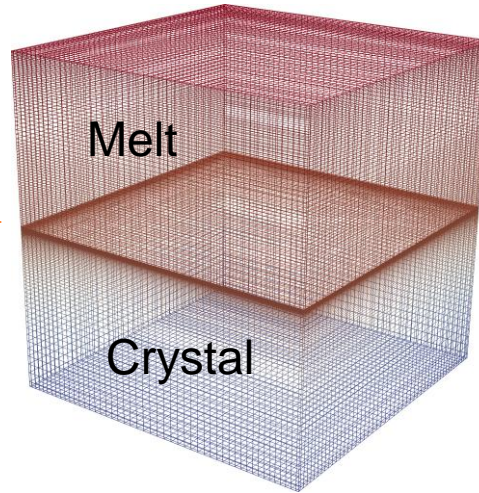
- Faculty of Physics, West University of Timisoara, Romania  
~Prof.dr. Daniel Vizman
  
- 2Laboratoire Matériaux et Procédés pour le Solaire, Institut National de l'Energie Solaire CEA/DRT/LITEN/DTS  
~Dr. Jean-Paul Garandet

# CONSIL Objectives

## Objectives:

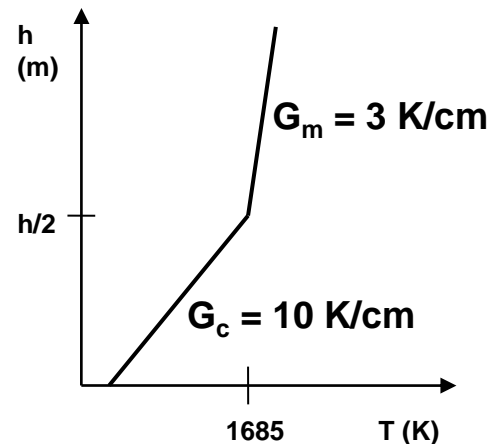
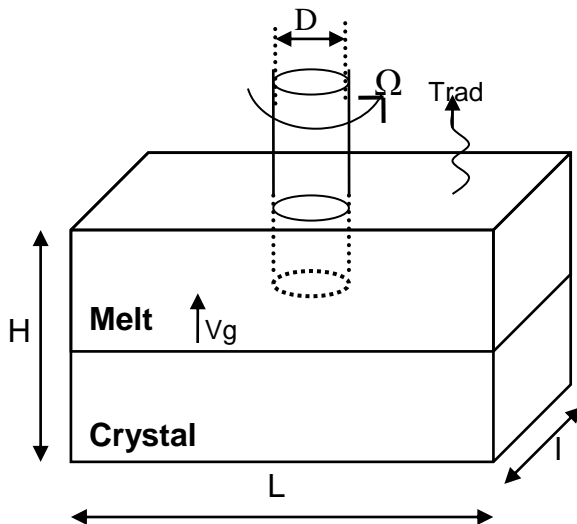
- O1** Experimental and numerical study of natural convection effect on impurity segregation and coating stability in a Bridgman method for multicrystalline Si growth (WUT, INES)
- O2** Modelling of Si crystal growth process in furnaces with stirring devices (WUT)
- O3** Growth of multicrystalline silicon in laboratory (2 kg ingots) and pilot scale (60 kg ingots) furnaces with a stirring device (INES)

# Numerical model



**Qasi steady state approximation:**

•the reaction time of the interface shape on changes in the melt flow is smaller than the total solidification time



- $L=l=38$  cm;  $h=40$  cm (G2)
- $D=8$  cm
- $T_{\text{rad}} = 1713$  K
- $v_g = 10$  mm/h
- $\Omega=0,5,10,20,30,40$  rpm

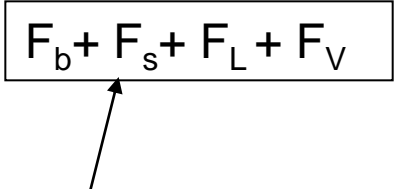


# Navier-Stokes Equations

Conservation of mass

$$\frac{\partial}{\partial y_i} (\rho u_i) = 0$$

Conservation of Momentum

$$\frac{\partial}{\partial t} (\rho u_i) + \frac{\partial}{\partial y_j} (\rho u_i u_j + \tau_{ij}) = -\frac{\partial p}{\partial y_i} + s_i^u \quad i \in \{1,2,3\}$$


Conservation of energy or mass ( $\Phi$  – temperature or concentration)

$$\frac{\partial}{\partial t} (\rho \Phi) + \frac{\partial}{\partial y_j} \left( \rho u_j \Phi - \Gamma_\Phi \frac{\partial \Phi}{\partial y_j} \right) = s_\Phi$$

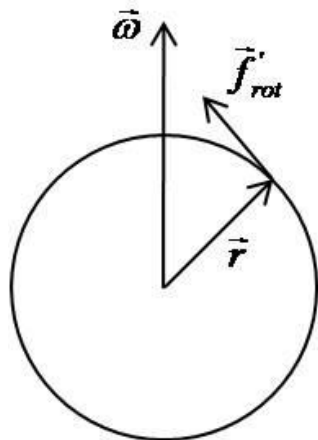
## Source terms for Navier-Stokes Equations

Bouyancy forces (gravitation):

$$s_u^i = -\rho(T_{ref})g^i\beta_t(T - T_{ref})$$

Azimuthal force:

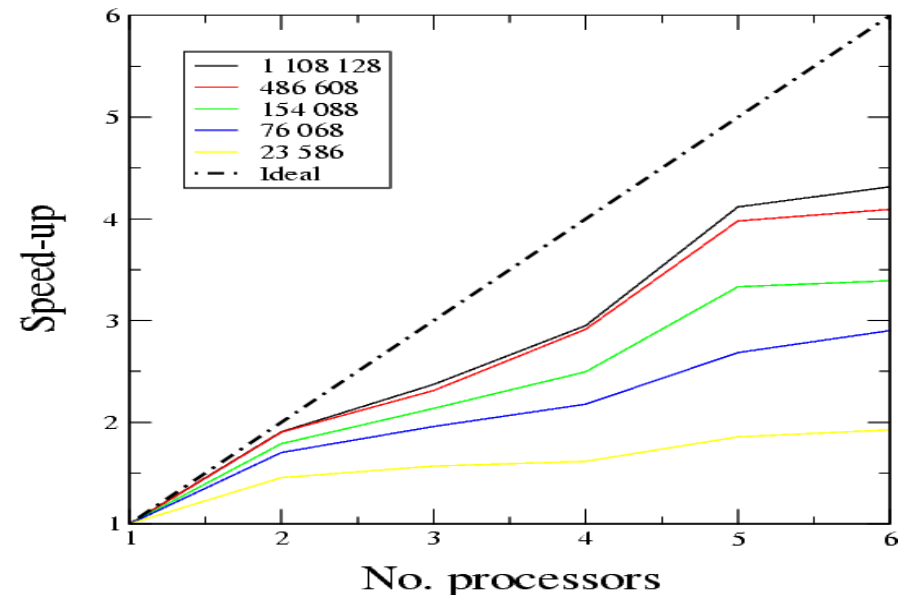
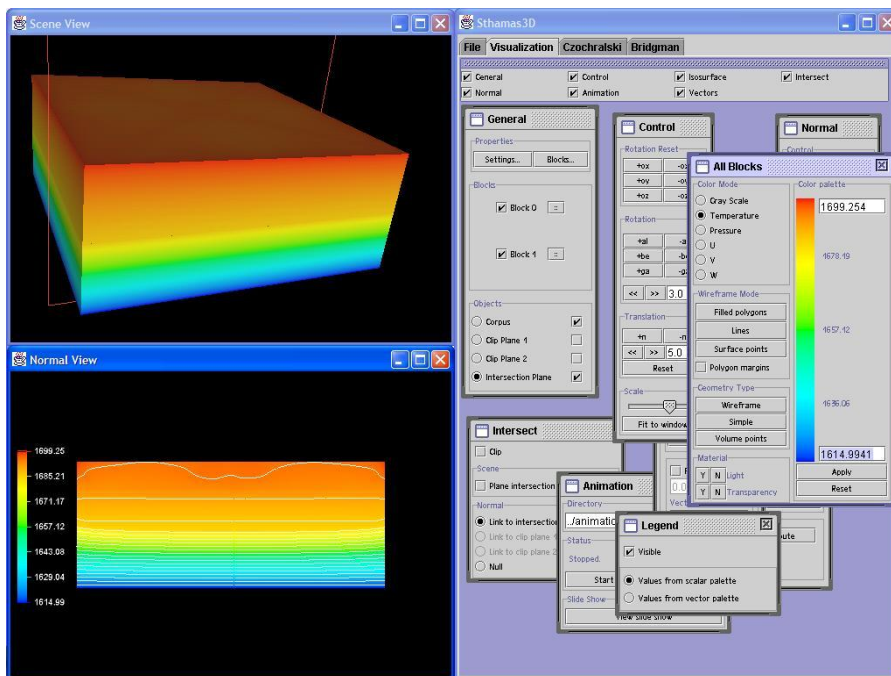
$$s_i^u = (\vec{f}_{rot})_i = ((\rho\omega \cdot \vec{\omega}) \times \vec{r})_i$$



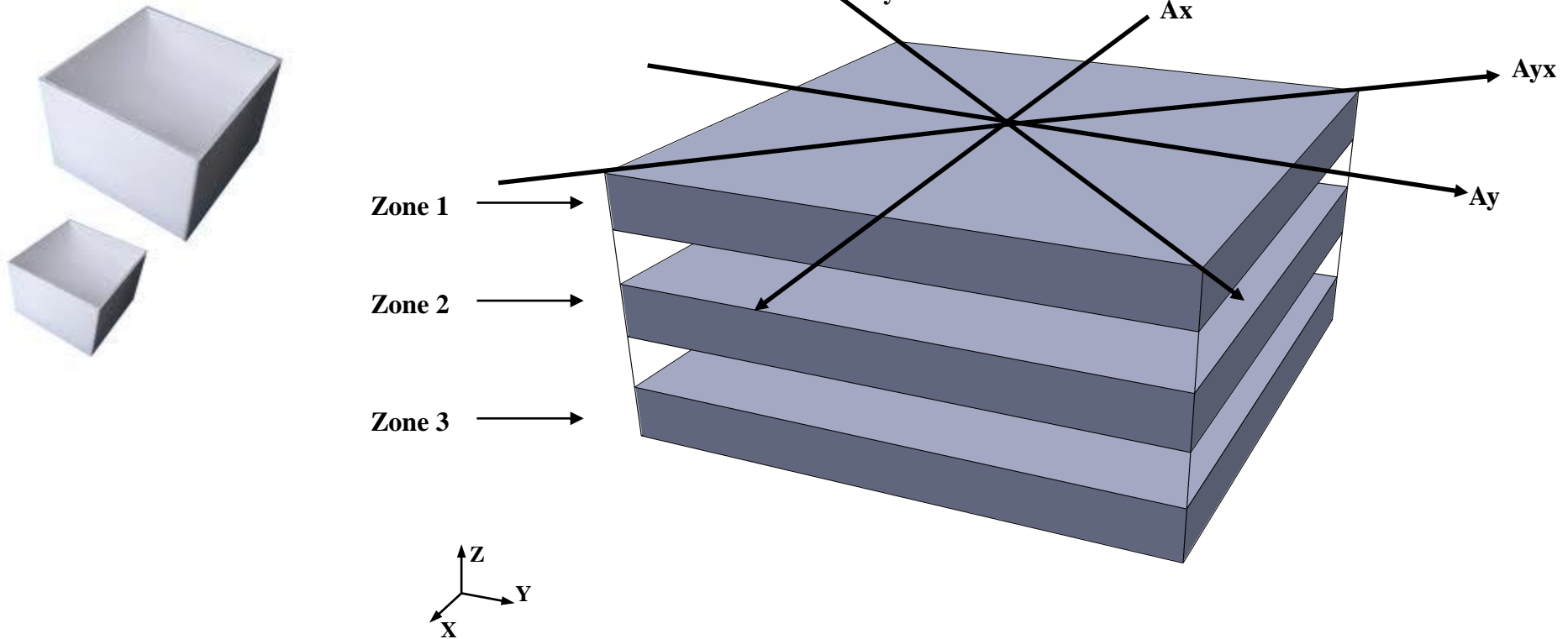
$$|\vec{f}_{rot}| = \begin{cases} \rho r \omega^2, & r \leq D/2 \\ 0, & r > D/2 \end{cases}$$

# Simulations with STHAMAS3D software

- Direct solution of Navier-Stokes equations; phase boundary tracking; Finite Volume method; parallelization with MPI
- Consideration of Lorentz force by solving additional scalar potential equation
- Higher efficiency for parallel version on a PC cluster



# Symmetries in rectangular crucibles



✓ *Regularity of a flow pattern is defined in relation with the number of symmetry planes.*

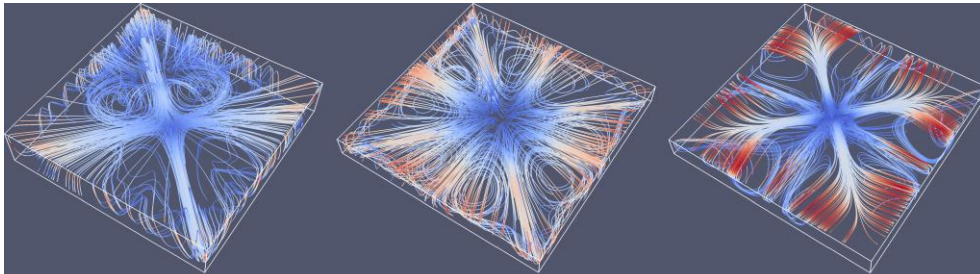
# Influence of temperature gradient in the melt on the flow pattern

$G_m = 1 \text{ K/cm}$

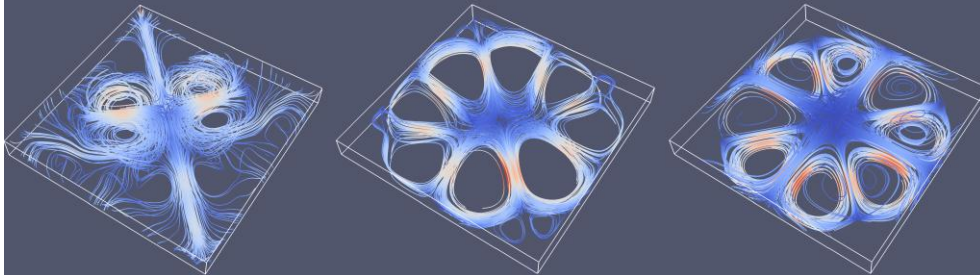
$G_m = 2 \text{ K/cm}$

$G_m = 3 \text{ K/cm}$

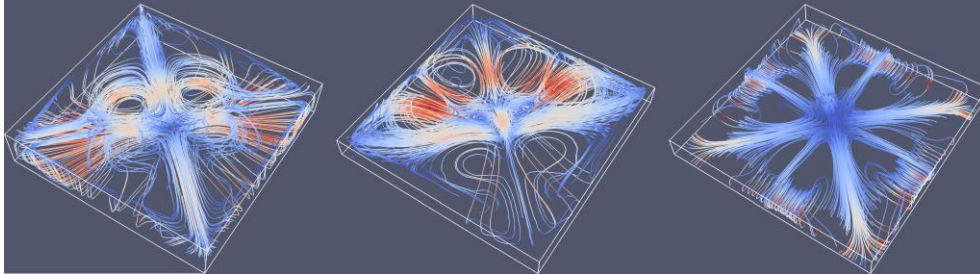
Zone 1



Zone 2



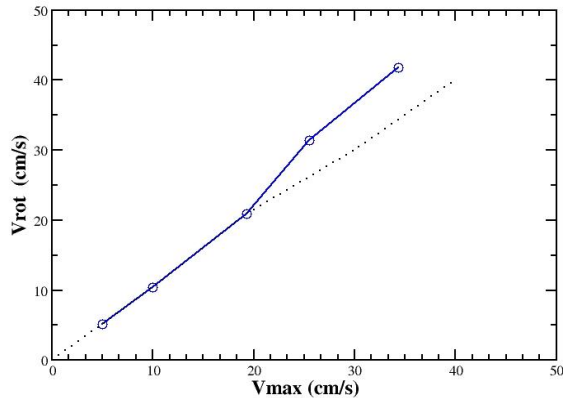
Zone 3



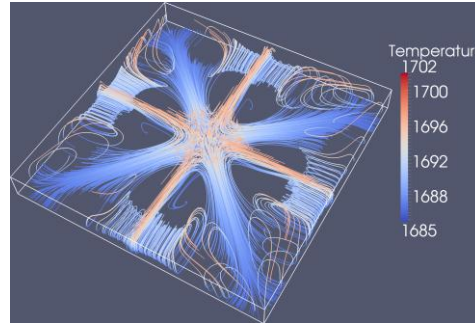
- ✓ Flow pattern regularity increase with the increase of  $G_m$
- ✓ In all zones there is a symmetry along the Axy plane
- ✓ For  $G_m = 1 \text{ K/cm}$  common flow structure appear in all 3 zones
- ✓ In zone 3 the flow structure symmetry increases along the Ax, Ay and Ayx planes
- ✓ Convection roles appear near the crucible wall for  $G_m = 3 \text{ K/cm}$
- ✓ In zone 3, regularity increases much more once with the inversion of S-L interface

$v_g = 10 \text{ mm/h}$

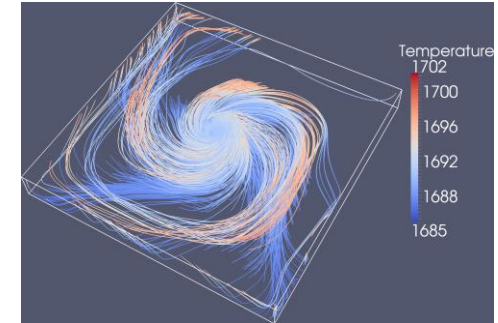
# Influence of the stirrer rotation on the melt flow



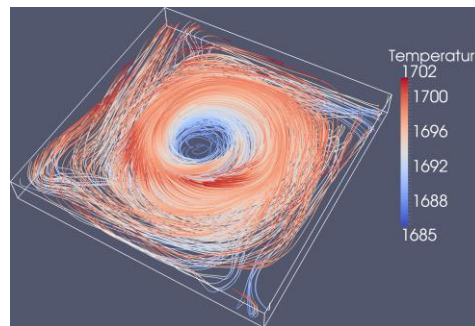
$$\omega^2 D / 2 = \rho \left| \left( \vec{V}_{\max} \cdot \nabla \right) \vec{V}_{\max} \right| \approx \rho \frac{V_{\max}^2}{D / 2}$$



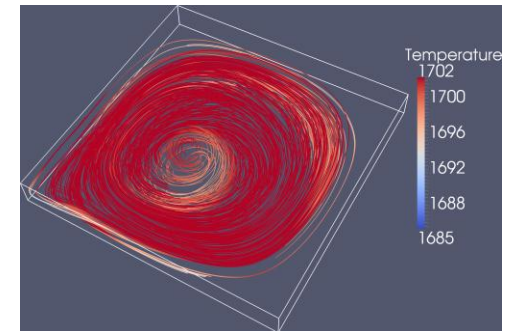
$\Omega=0\text{rpm}$



$\Omega=5\text{rpm}$



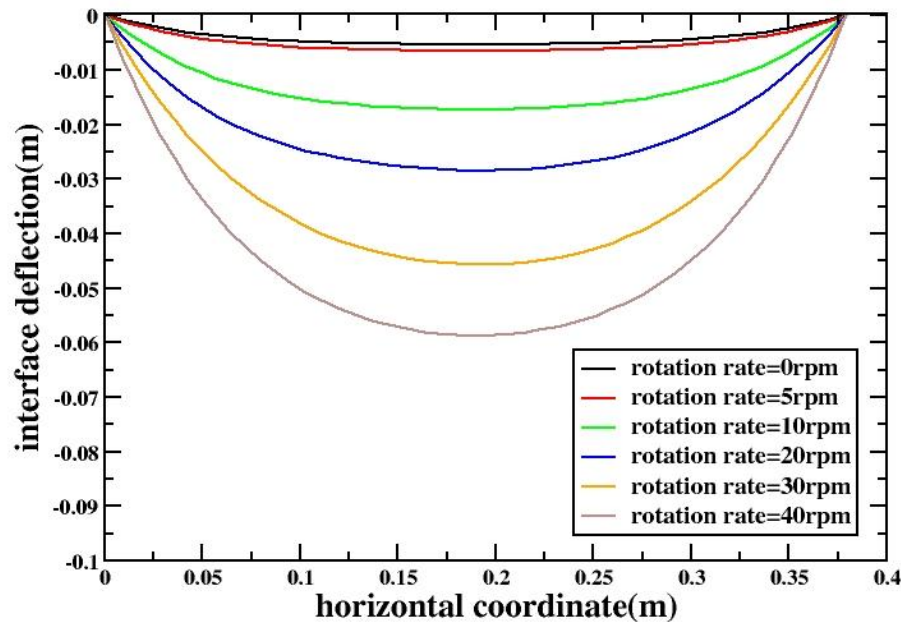
$\Omega=10\text{rpm}$



$\Omega=30\text{rpm}$

- Stirrer rotation enhance the melt convection
- For small rotation rates (less than 20rpm) a uniform azimuthal rotation is produced and the rotational driving force is balanced by inertia
- Stirrer rotation facilitate the transport of hot melt from upper parts to the S-L interface

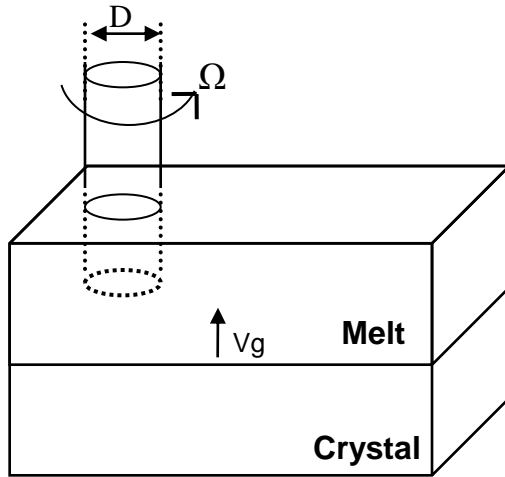
# Influence of the stirrer rotation on the interface shape



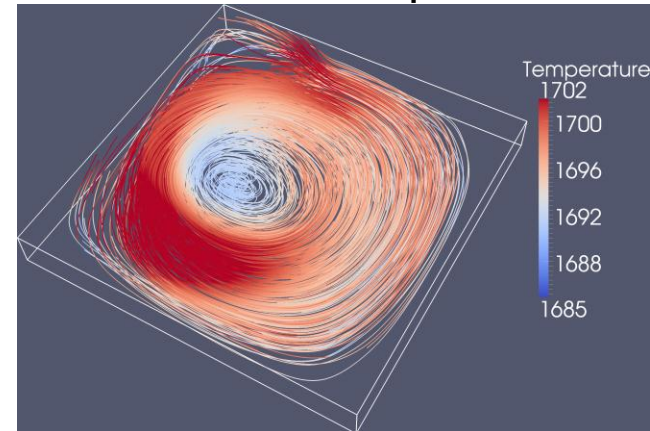
✓ With the increase of stirrer rotation the interface deflection increases



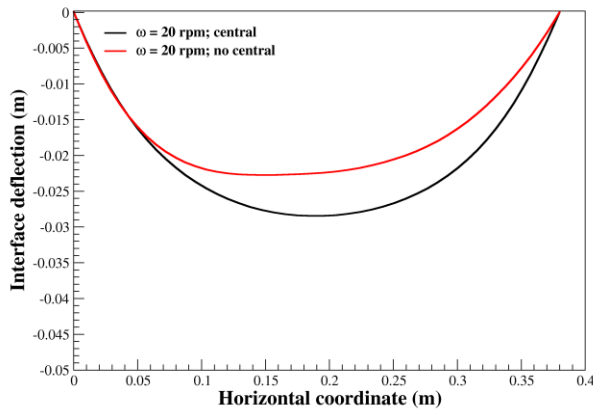
# Influence of the stirrer position



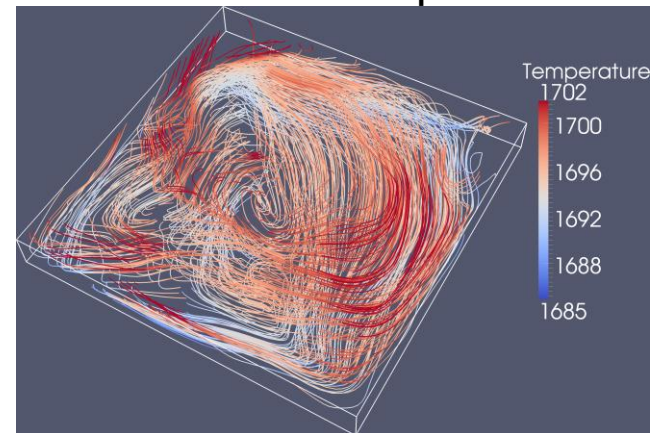
Central stirrer position



$\Omega=20\text{rpm}$



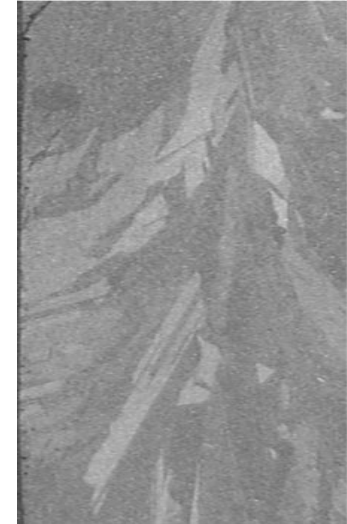
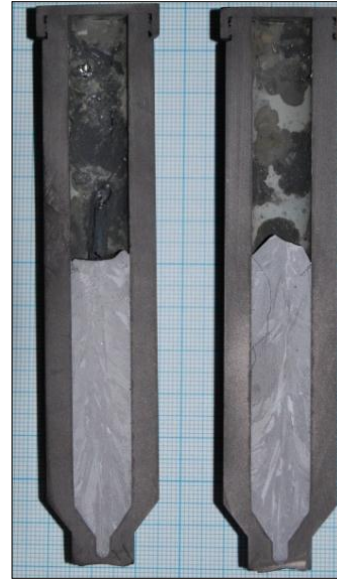
Lateral stirrer position



✓ Stirrer position can influence the symmetry of the interface shape.



# Bridgman Method for growth of mc-silicon

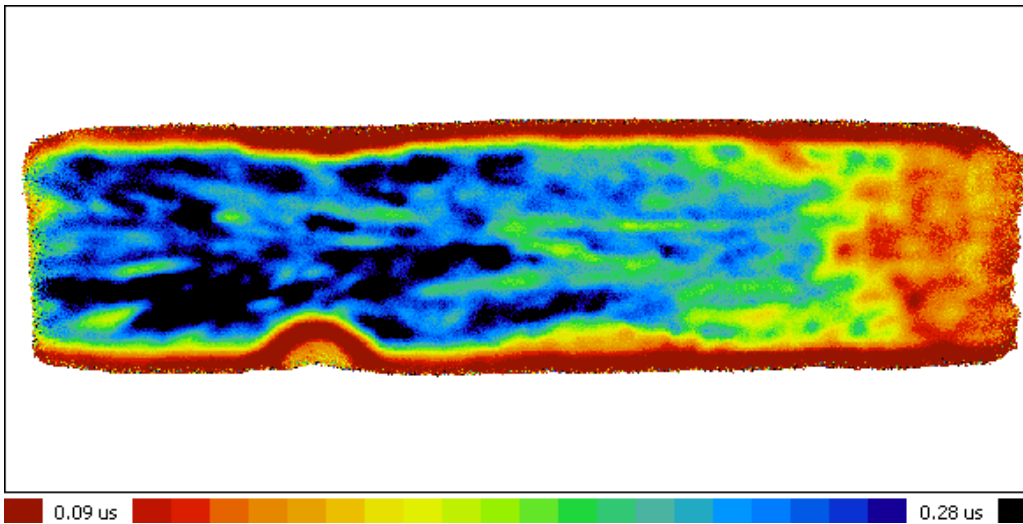


- Multicrystalline silicon have been obtained for various growth parameters (growth velocities, temperature gradients, etc)

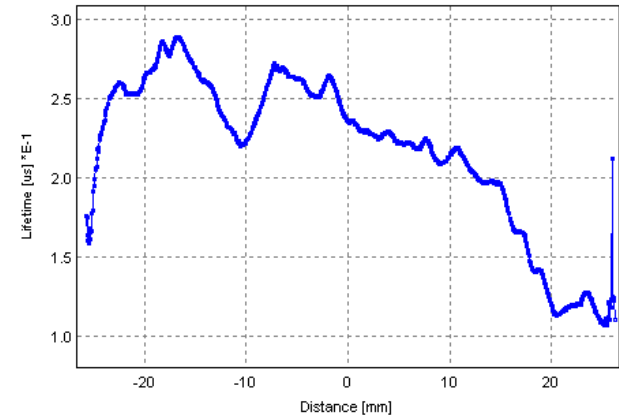
# Lifetime measurements

MW-PCD technique

(„ microwave reflectance photoconductivity decay”)

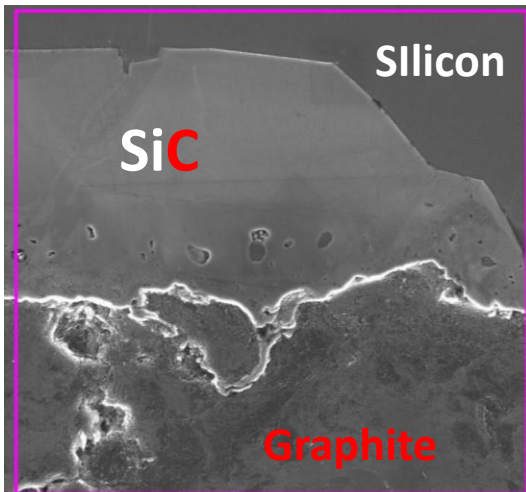


### Axial distribution

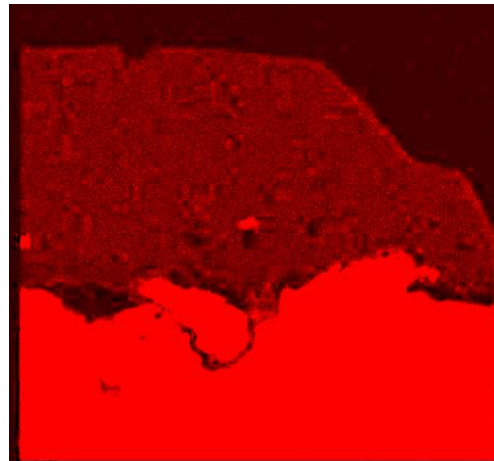


•Lifetime decreases in the upper part of the crystal because of the impurities agglomeration due to segregation

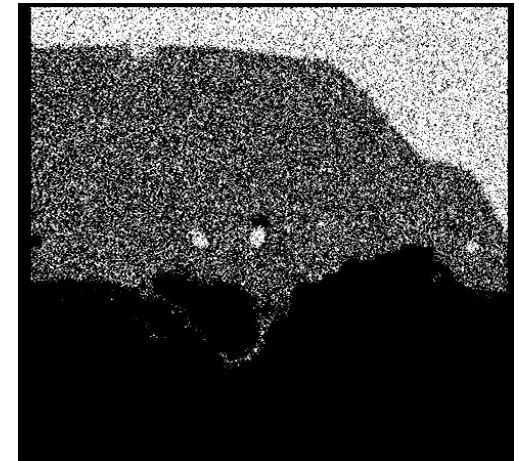
## EDX element charts



Electron scanning image



Carbon concentration  
(red dots)



Silicon concentration  
(white dots)

- SiC precipitate formed at the crystal crucible edge

## Other results

- **Master thesis (2011):**

**Characterization of photovoltaic silicon ingots by measurements of lifetime of minority carriers.**

**Author: Radu Negrila (6 months at INES)**

- **PHD – thesis (2012):**

**Study of the directional solidification process by means of numerical methods**

**Author: Popescu Alexandra**

- **12 participations at international conferences**

- **4 participations at national conferences**

## Other results

### ISI-papers

**A. Popescu, D. Vizman** - Numerical study of the influence of melt convection on the crucible dissolution rate in a silicon directional solidification process, *International Journal of Heat and Mass Transfer* 54 (2011), 5540-5544; **AIIS:0.823, IF:2.407**

**A. Popescu, D. Vizman** - Numerical Study of Melt Convection and Interface Shape in a Pilot Furnace for Unidirectional Solidification of Multicrystalline Silicon, *Crystal Growth & Design* 12 (2012), 320-325; **AIIS:0.912, IF:4.720**

**S. Dumitrica, D. Vizman, J.-P. Garandet, A. Popescu** - Numerical studies on a type of mechanical stirring in directional solidification method of multicrystalline silicon for photovoltaic applications, *Journal of Crystal Growth* (2012), *doi:10.1016/j.jcrysgr.2012.01.011*; **AIIS: 0.490, IF: 1.710**

### AIP- conference proceedings

**O. Bunoiu, M. Stef, A. Popescu** - Interface shape studies in bridgman growth of multicrystalline silicon, *AIP Conference Proceedings* 1387 (2011), 226-231

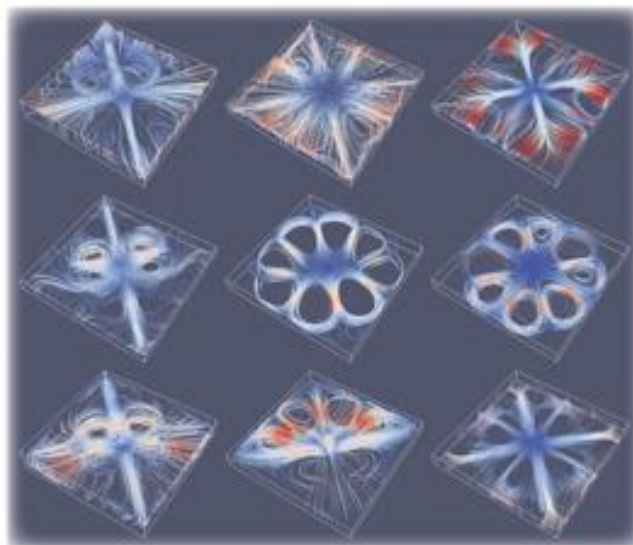
**V. Pupazan, A. Popescu, O. M. Bunoiu, D. Vizman** - Influence of growth rate on interface shape and grains structure in multicrystalline silicon growth by bridgman method, *AIP Conference Proceedings* 1472 (2012), 210-214

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