The Institute for Crystal Growth (IKZ) in Berlin-Adlershof
Main Working Fields and Participation in GERDA

Helge Riemann, Nikolai Abrosimov

1. About IKZ

2. Projects of the silicon- germanium group:
   - Floating Zone (FZ) Silicon
   - Monoisotopic Silicon (AVOGADRO)
   - Si_xGe_{1-x}

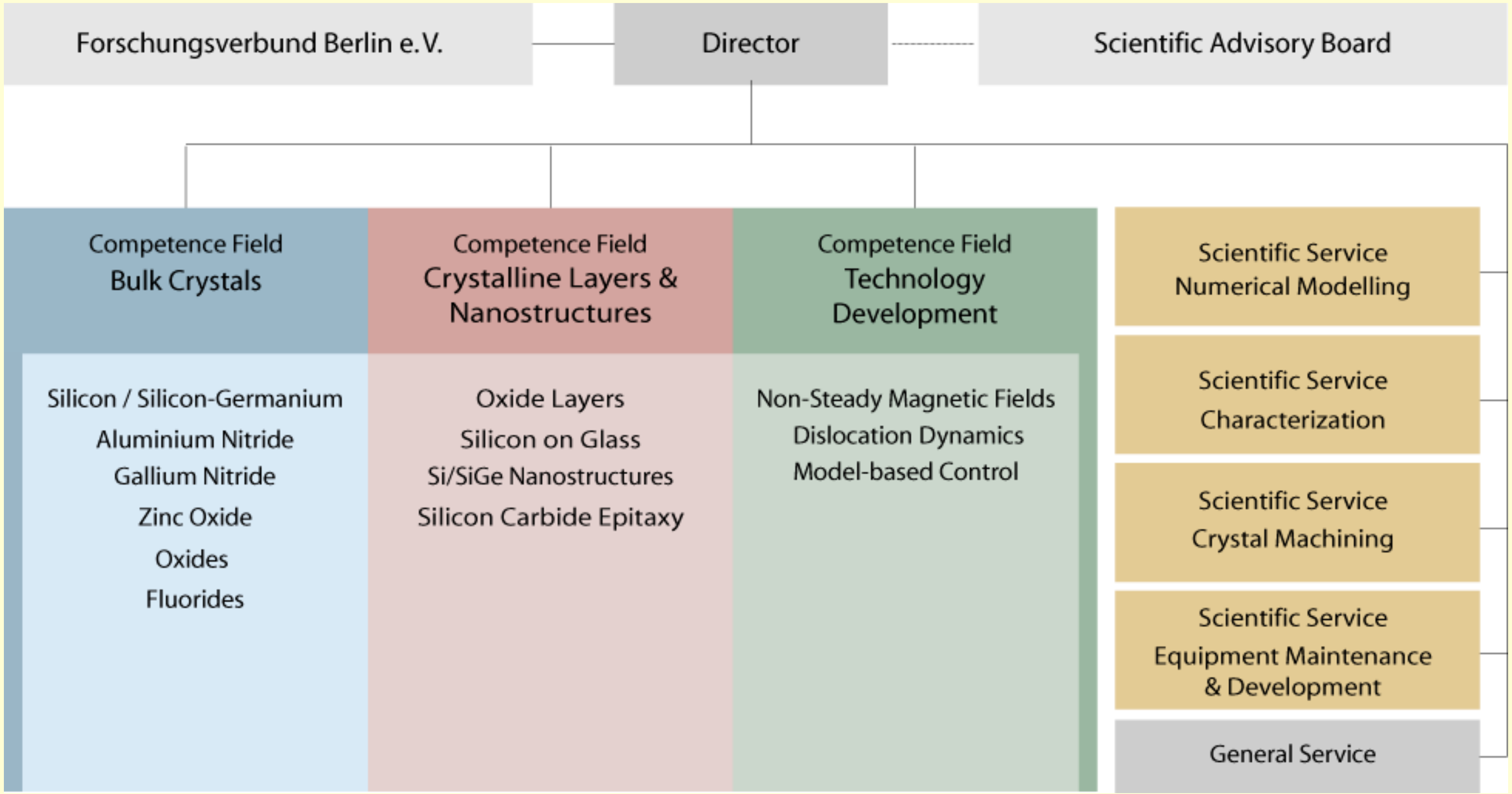
3. Germanium crystal growth, GERDA activities

The Institute for Crystal Growth in Berlin-Adlershof

- Founded in 1992, with scientific and technical staff from the Academy of Sciences of GDR and from Humboldt University
- IKZ is a member of the Leibniz association
- Research & service function
- Present staff: 95 (45 scientists, 11 PhD students, 31 technicians, 8 others)
- Budget 2007:   institutional 7.1 M Euro, external projects 2.1 M Euro

Adlershof – City of Science, Trade and Media

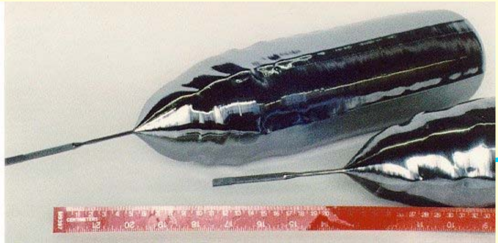
IKZ-Structure



Institute for Crystal Growth (IKZ)

materials

Three main material categories:



6“ FZ Si

Ge and GeSi

Elemental semiconductors

20 mm AlN

Compound semiconductors

6“ GaAs

Dielectric crystals

1.5“ Cr:$LiCaAlF_6$

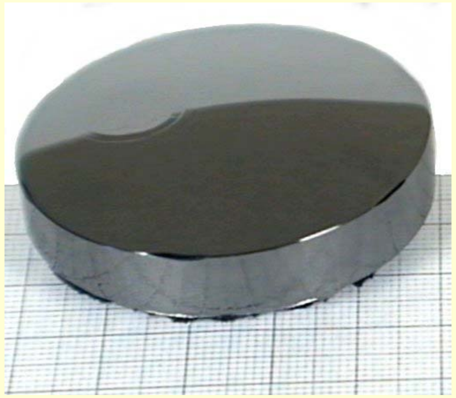
2“ $LiAlO_2$

Crystal Dimensions

Crystal dimensions

dm/cm ⟶ µm ⟶ nm

Bulk Crystals & Fibers

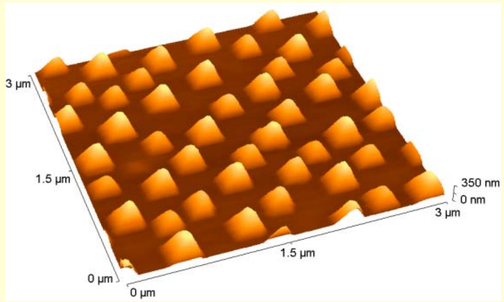


2“ SiC single crystal

Epitaxial layers

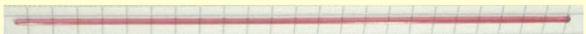
25 µm thick 4H-SiC-layer

Nano-crystals



SiGe pyramids

$Cr:Al_2O_3$ Fiber



Institute for Crystal Growth

Available growth techniques:

Melt

-**Czochralski pullers (high / low-pressure)**
-**Floating-zone furnaces (RF and laser heating)**
-**Vertical gradient freeze**
-**Micro-pulling down**

Gas phase

-**Physical vapour deposition (PVT)**
-**Chemical vapour deposition (CVD)**
-**MO Chemical vapour deposition (MO-CVD)**
-**(U)HV evaporation**

Solution

-**Liquid phase epitaxy**
-**Hydrothermal autoclaves**
-**Vapour-Liquid-Solid**
-**Top-seeded solution growth (TSSG)**

Characterisation techniques:

- Electron Paramagnetic Resonance
- **Electrical measurements (Resistivity and Hall, DLTS, Photoconductivity)**
- Optical measurements (Photolumin., Reflectivity, Absorption, Raman, **Scanning photovoltage**, elipsometry)

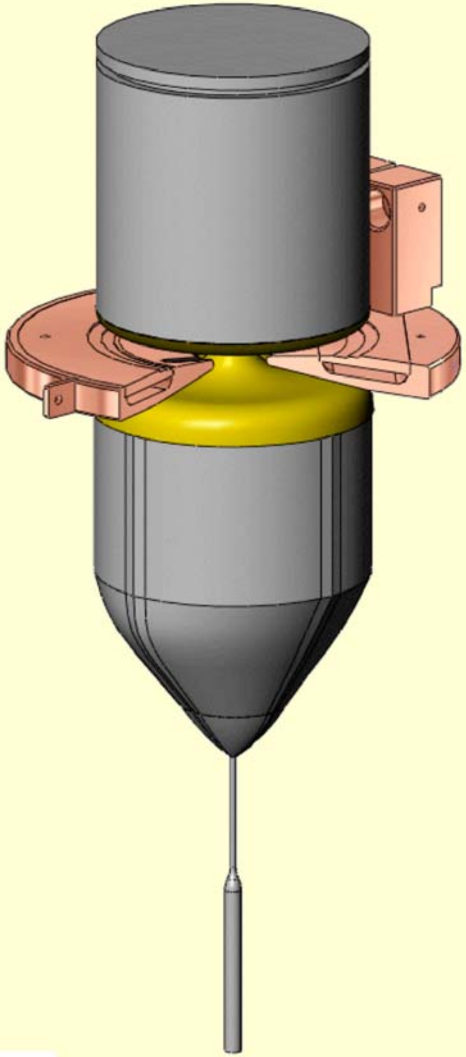
- X-Ray diffr., **X-Ray topography**, Powder diffr.
- SEM, Cathodolumin., EBIC, FIB, EDX
- AFM
- Chemical etching + optical microscopy
- **Laser scattering tomography**
- Strain determination via polaroscopy

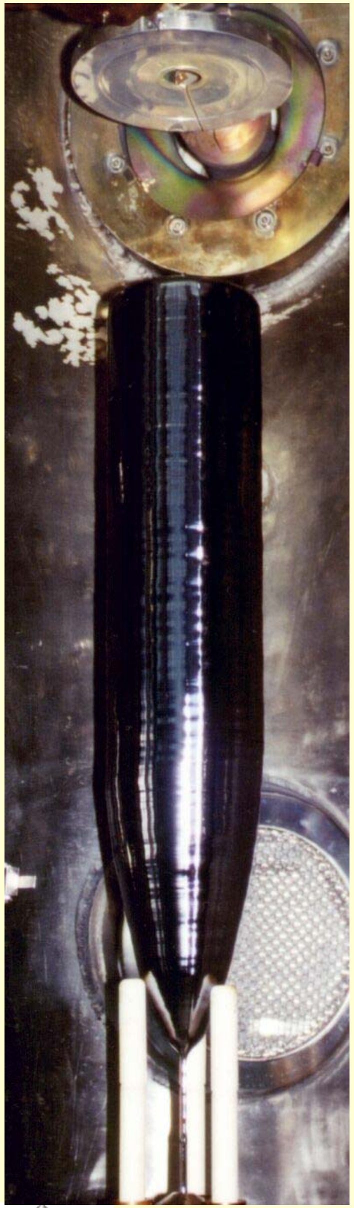
- Mass spectroscopy (ICP-OES, RF-OES), DTA, TGA

Sample Processing laboratory:

- Crystal orientation
- ID-Blade and Wire sawing
- Lapping machines
- Polishing machines
- Surface inspections

Silicon Floating Zone Crystal Growth





Floate Zone Crystal Pullers

- three FZ- machines for crystals with diameters of up to 125mm,

- one FZ- machine for 150mm (3 bar)

Dislocation-free Si FZ crystal with quadratic cross section

Quadratic FZ- silicon for high efficiency PV

Common Silicon Crystal Growth Methods

Czochralski(CZ)-Growth from crucible

floating zone(FZ) growth (crucible-free)

pedestal growth (crucible-free)

Non regarding other quality parameters, dislocation-free silicon crystals will grow if the pulling machine enables the essential growth conditions and if the polycrystalline Si feed material has a dense, pore-less structure and meets upper impurity limits markedly below solid solubility and depending on the distribution coefficients , e.g.:

C< 10^{16}cm^{-3}(k=0,07), O< 10^{17}cm^{-3}, all others < 10^{13}cm^{-3}...10^{15}cm^{-3}

Silicon-Germanium mixed crystal growth

-variable lattice constant

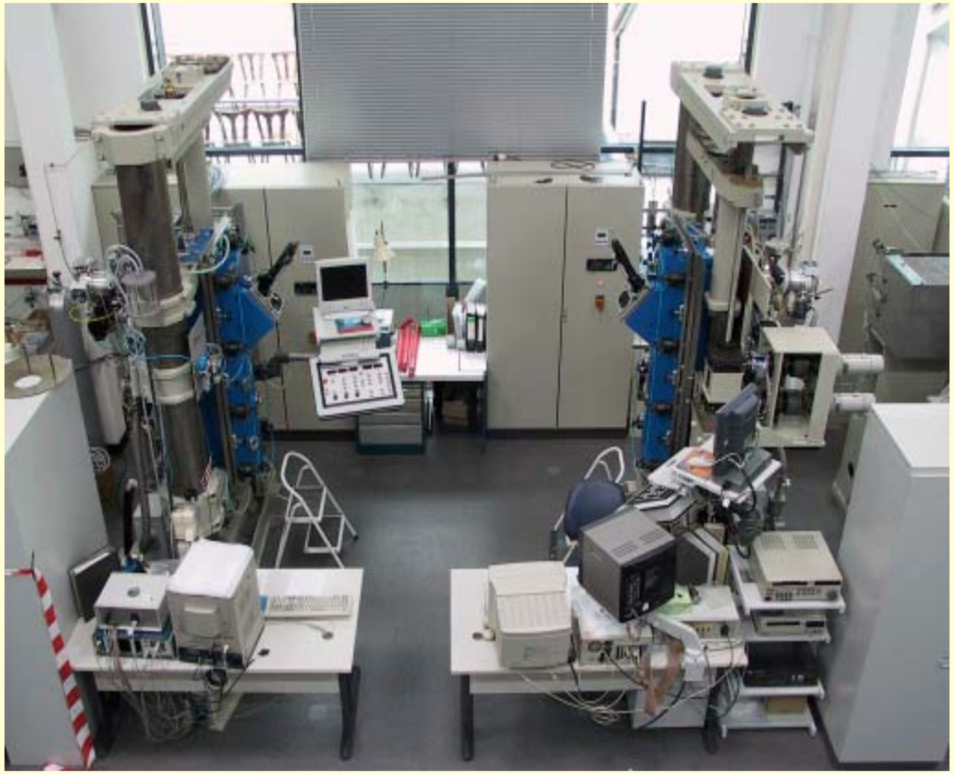
-Si- and Ge-rich crystals

-constitutional supercooling by segregation of Ge at growing interphase

-between ca.12% and 80% Si crystal growth practically impossible

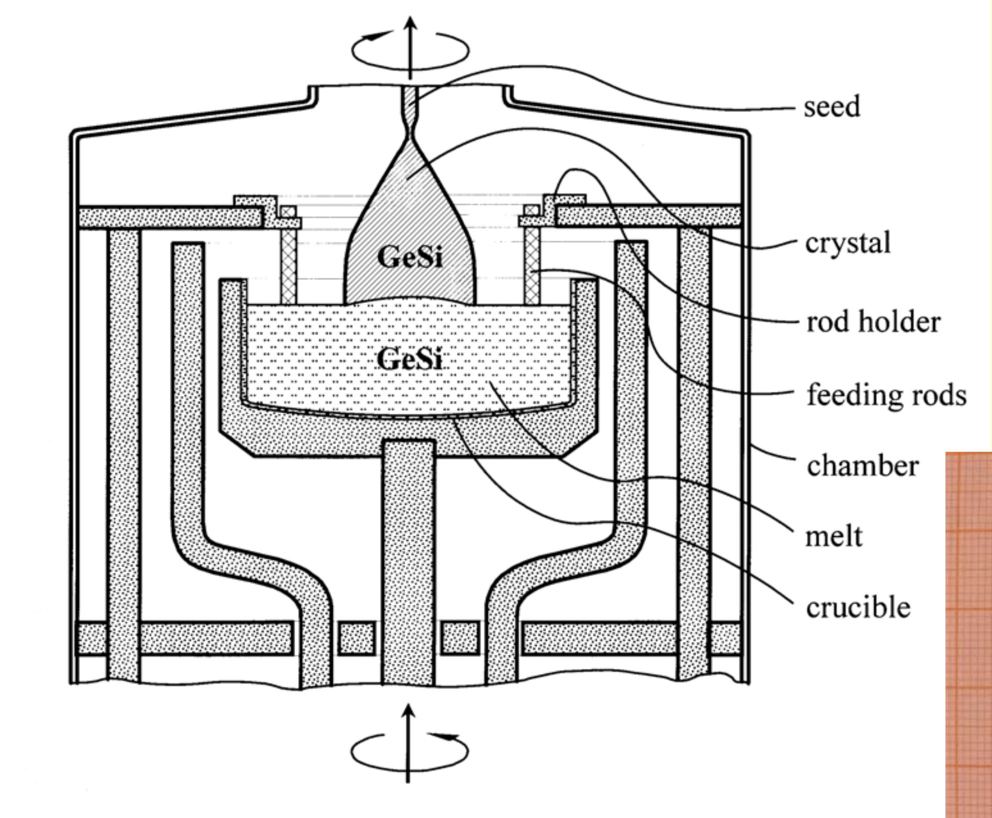
-Czochralski technique is mainly used, FZ difficult but possible

equipment

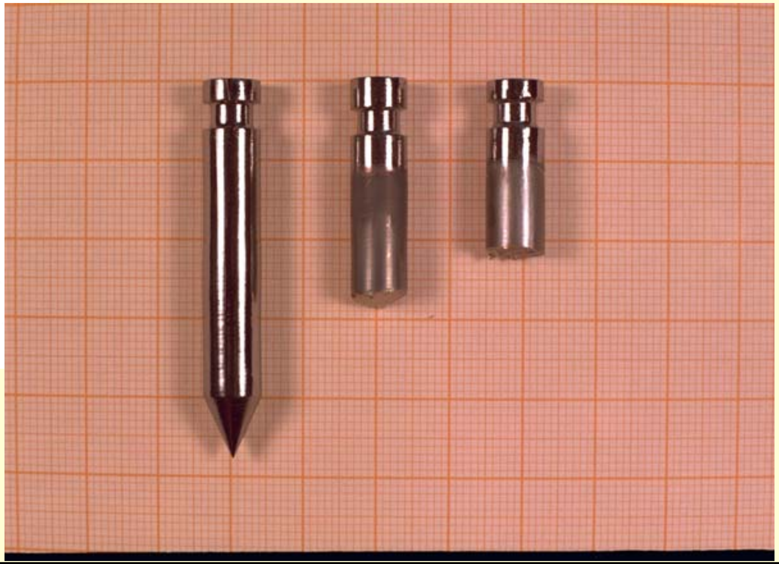


2 Czochralski furnaces
for SiGe and GeSi
crucible charge 1,5kg

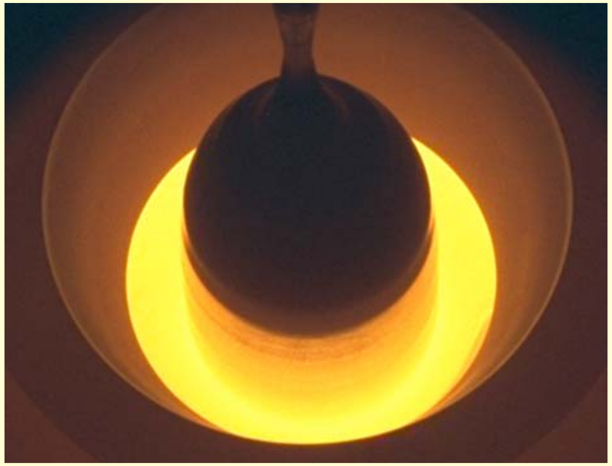
Czochralski growth of GeSi single crystals



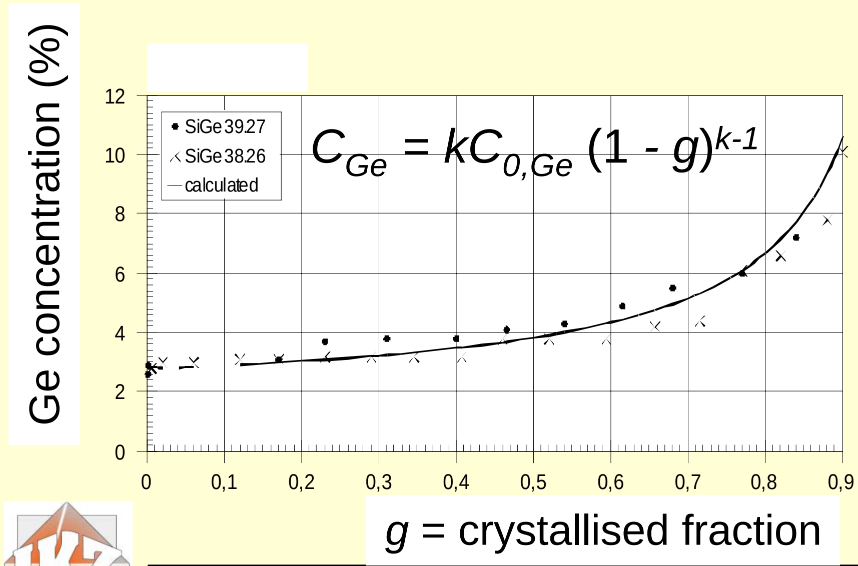
feeding rods before and after growth process

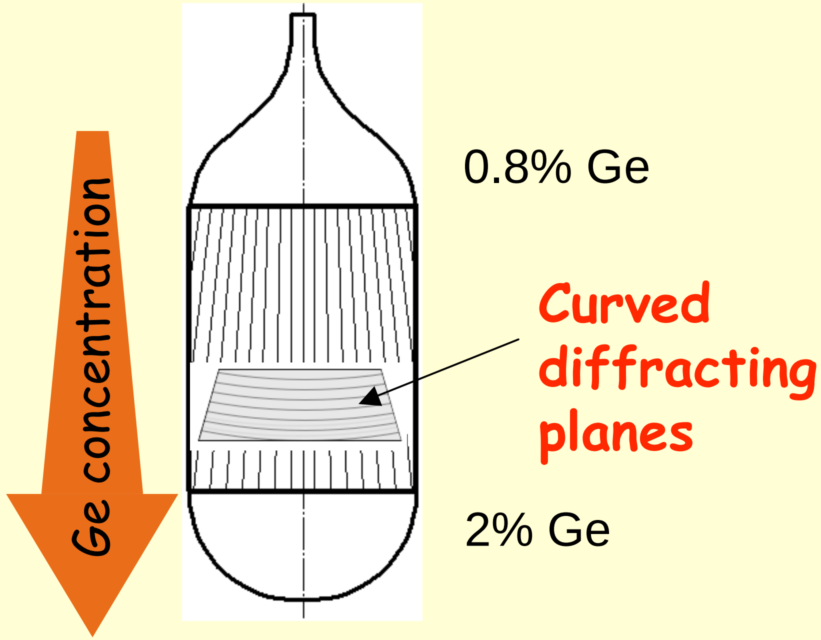


$Si_{1-x}Ge_x$ gradient crystals

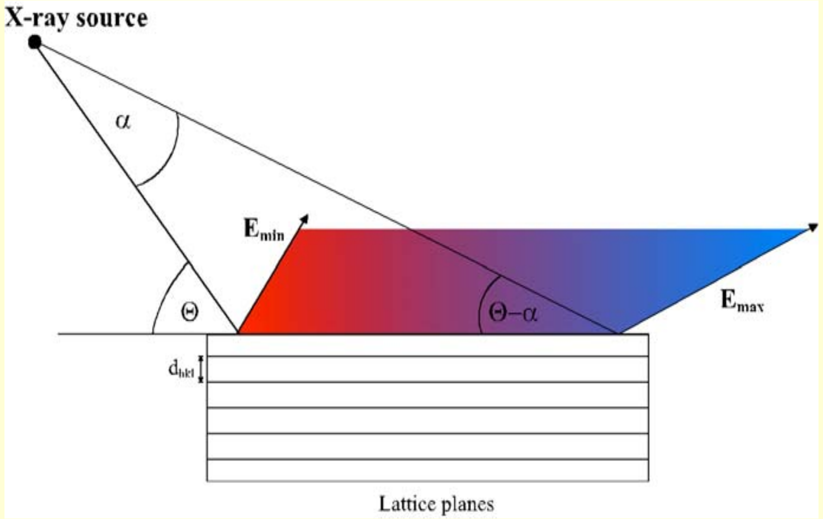


Growth of a Si_{1-x} Ge_x ingot



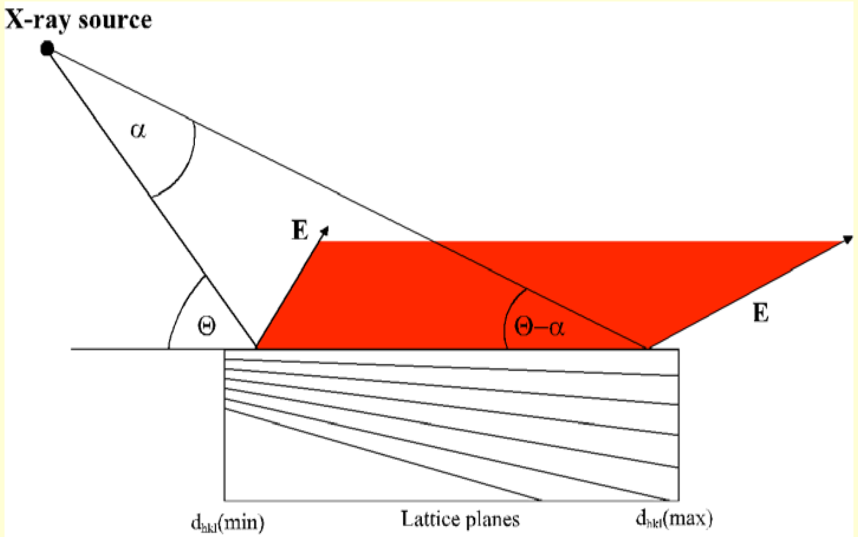


SiGe gradient crystal as monochromator for X-rays



Bragg‘s law :

$$2\, d \sin \Theta = \lambda$$



for gradient crystals

$$2\, d\, (1 + \Delta d/d) \sin (\Theta\text{-}\alpha) = \lambda$$

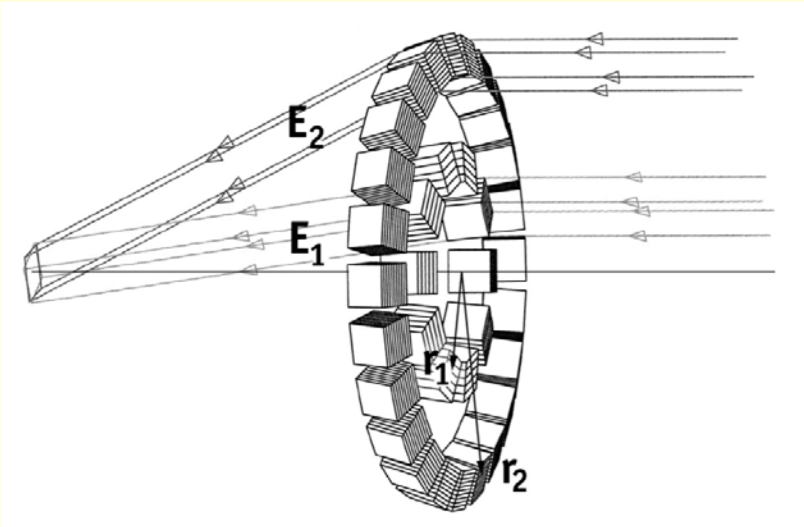
optimized Gradient

$$a=(\Delta d/d)/L=\cos\Theta_0/R$$

Principle of a Laue lens

Bragg's law: $2\ d_{hkl}\ \sin\theta_B = n\ \lambda$

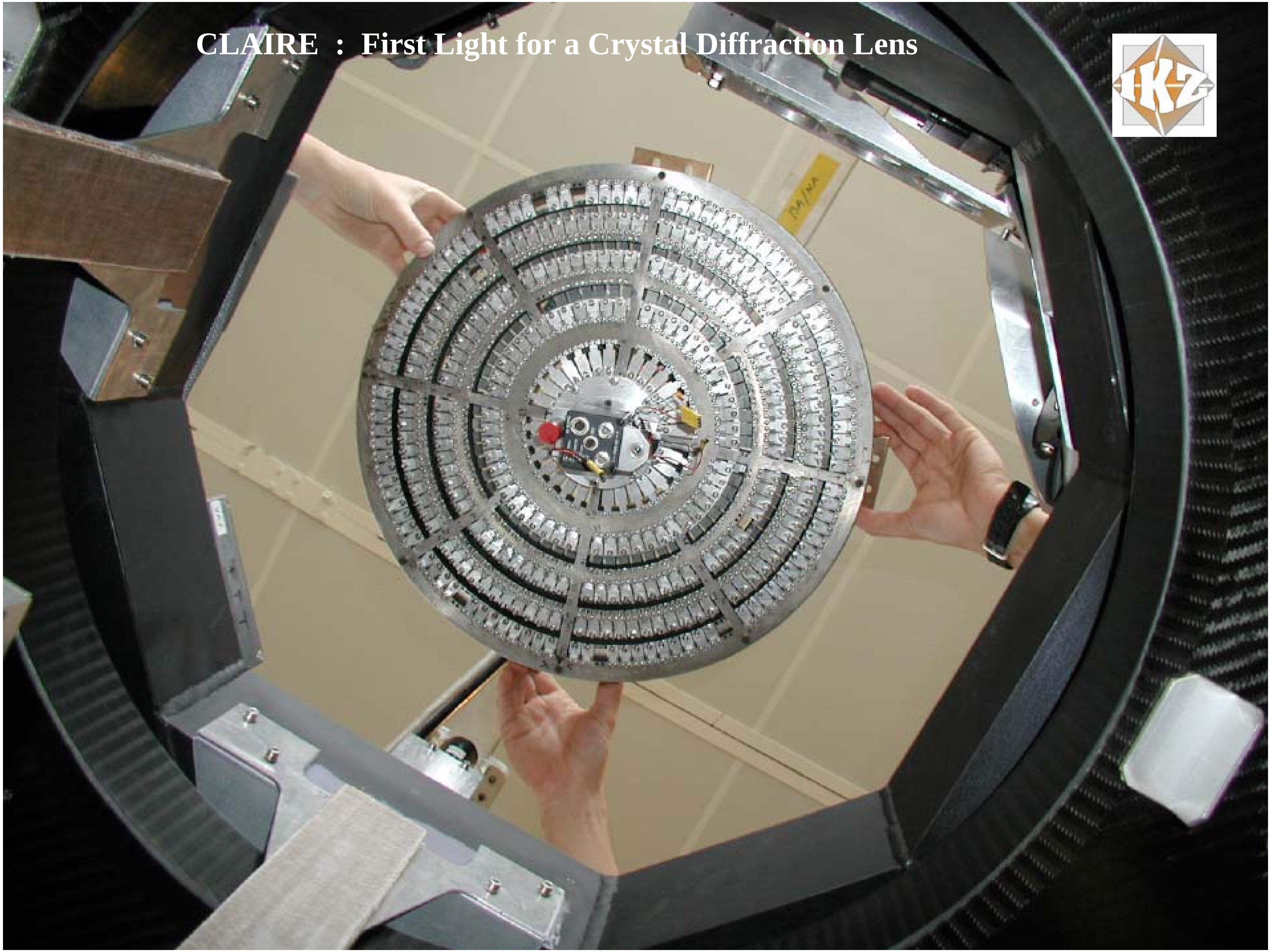
Focus: detector



γ rays from sources at infinity

Diffracting elements: crystals

CLAIRE : First Light for a Crystal Diffraction Lens



CLAIRE 2001

Launch : 14 june 2001, 8h15 UT, CNES balloon base, Gap-Tallard
Balloon : Zodiac Z600 (600.000 m^3)
floating altitude : > 41 km (3.8 g/cm2 residual atmosphère), during 5h 30’
Landing : 14 june 2001, 17 h UT, Bergerac, Acquitane (~Bordeaux region)

Monoisotopic Silicon Crystal Growth

-isotope enrichment as SiF_4 gas

-Isotpes 28Si, 29Si, 30Si

Special Demands for Mono Isotopic Si-Crystals

Basically, mono isotopic silicon crystallizes in the same way as natural silicon. Today, highly pure, perfect(dislocation-free) silicon crystals of 5-500kg weight are grown.

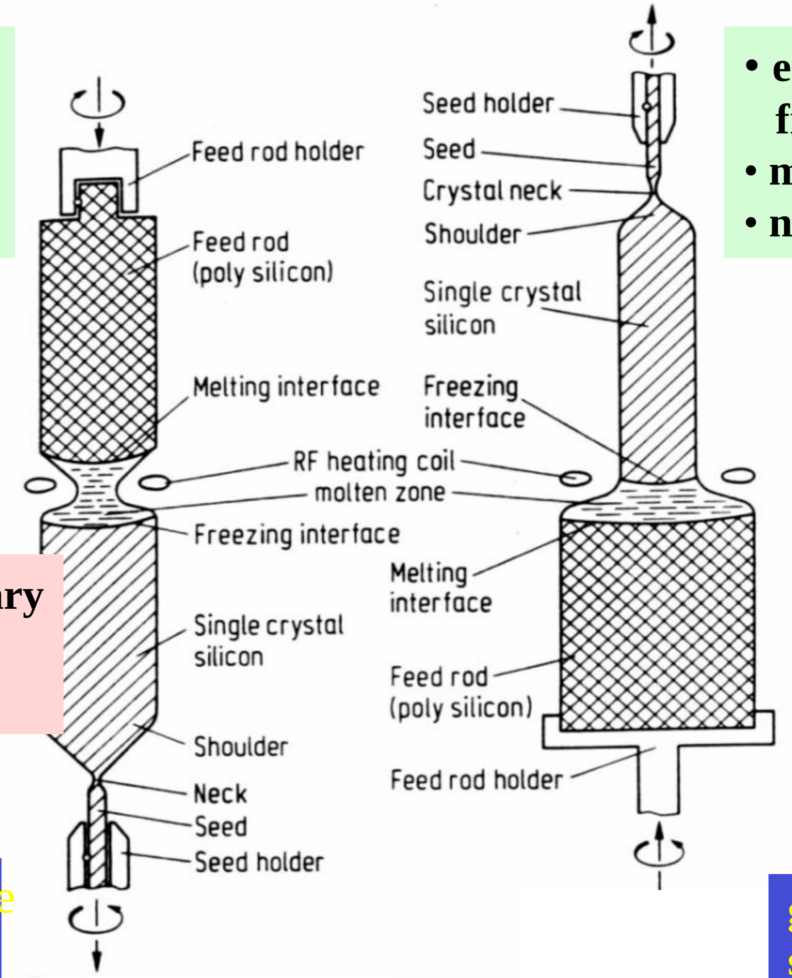
In contrast, only very small amounts of mono isotopic silicon are available. Down scaling of the silane purification, of the chemical vapor deposition(CVD) and last but not least of crystal growth methods causes new problems.

Therefor, qualified crystal growth techniques are needed in order to minimize material lost and to exploit effectively impurity segregation for the final goal of growing pure, perfect, mono isotopic Si crystals.

Crucible-less Silicon Growth Techniques

- natural silicon seed possible
- evaporation of impurities
- crystal diameter free
- no crucible contamination

- easy growth of thin filaments from thick feed rod
- mechanically stable
- no crucible contamination



Floating Zone(FZ) Pedestal

- cylindrical feed rod necessary
- weak segregation effect
- mechanically instable

- crystal thinner than feed
- cryst. diameter < 40mm
- weak segregation effect

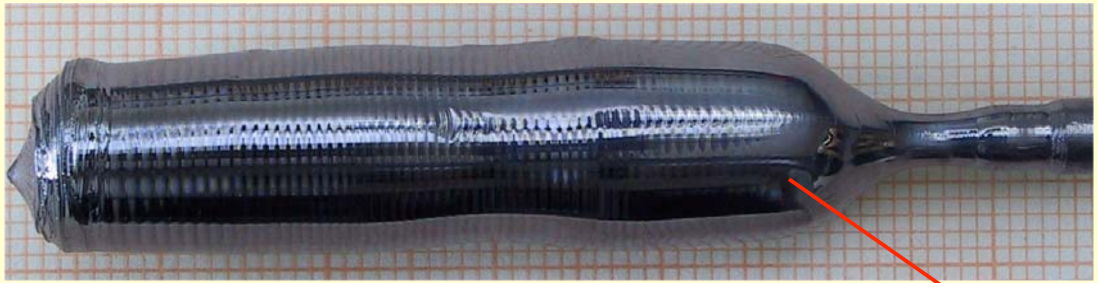
Growth of high purity, large dislocation-free crystals

growth of mono isotopic seeds and filaments for CVD

^{29}Si

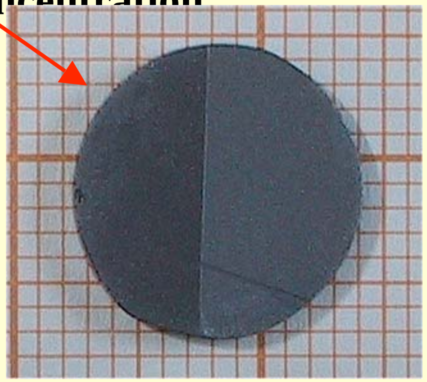
- CZ-grown crystal
- isotopic enrichment: 96,8 at% ^{29}Si
- 0.4 at% concentration loss of ^{29}Si due to quartz crucible erosion
- weight of crucible charge 4.0 g granulate, crystal weight: 3,8 g
- total impurities < 7 ppm (preferably carbon and oxygen)
- electrical resistivity: 3-5 Ωcm

^{30}Si

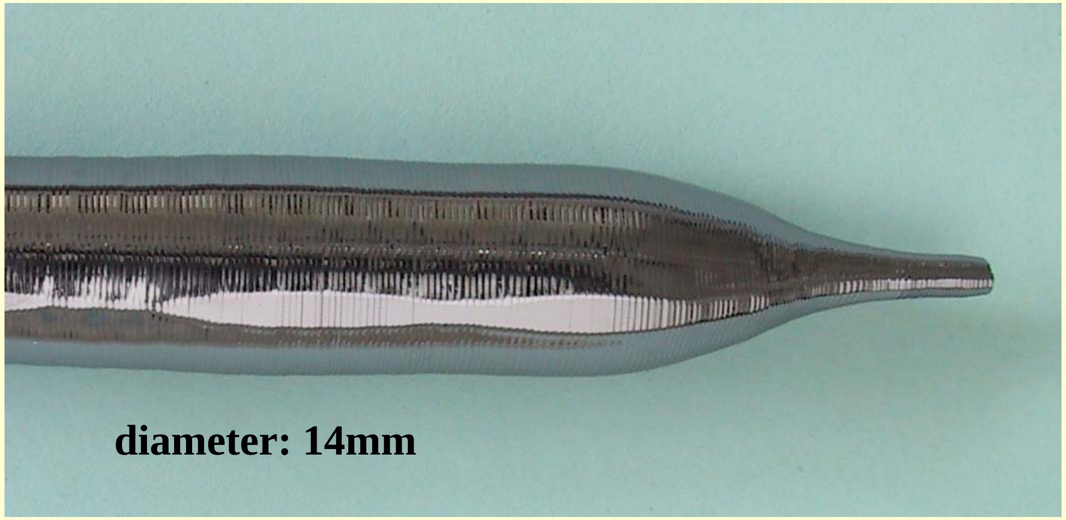


- mini CZ grown crystal
- isotopic enrichment: 99,5 at% ^{30}Si
- crucible charge: **13.3 g**
- **crystal weight 12,0g**
- **carbon content ≥ $3*10^{17}$ cm^{-3}**
- **resistivity: 1.5-2 Ω cm, p-type**

Twin boundaries due to high carbon concentration



^{28}Si



- 99,93 at% ^{28}Si dislocation- free single cystal, <211>- orientation,

- mass of disl.-free part: 18g, (mass of CVD-poly Si rod:32.0 g)

- impurities :B - <3×10^{13} cm^{-3}, C - < 3.0×10^{16} cm^{-3}, O - 3.2×10^{16} cm^{-3}

- electrical resistivity: 460 Ωcm

AVOGADRO– the new natural kg-standard ?

23.05.2007:

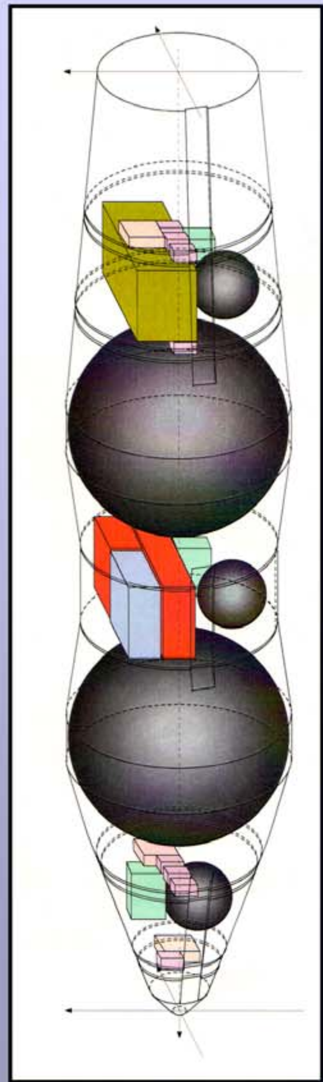
Growth of the final ^{28}Si isotope dislocation-free FZ-Si crystal

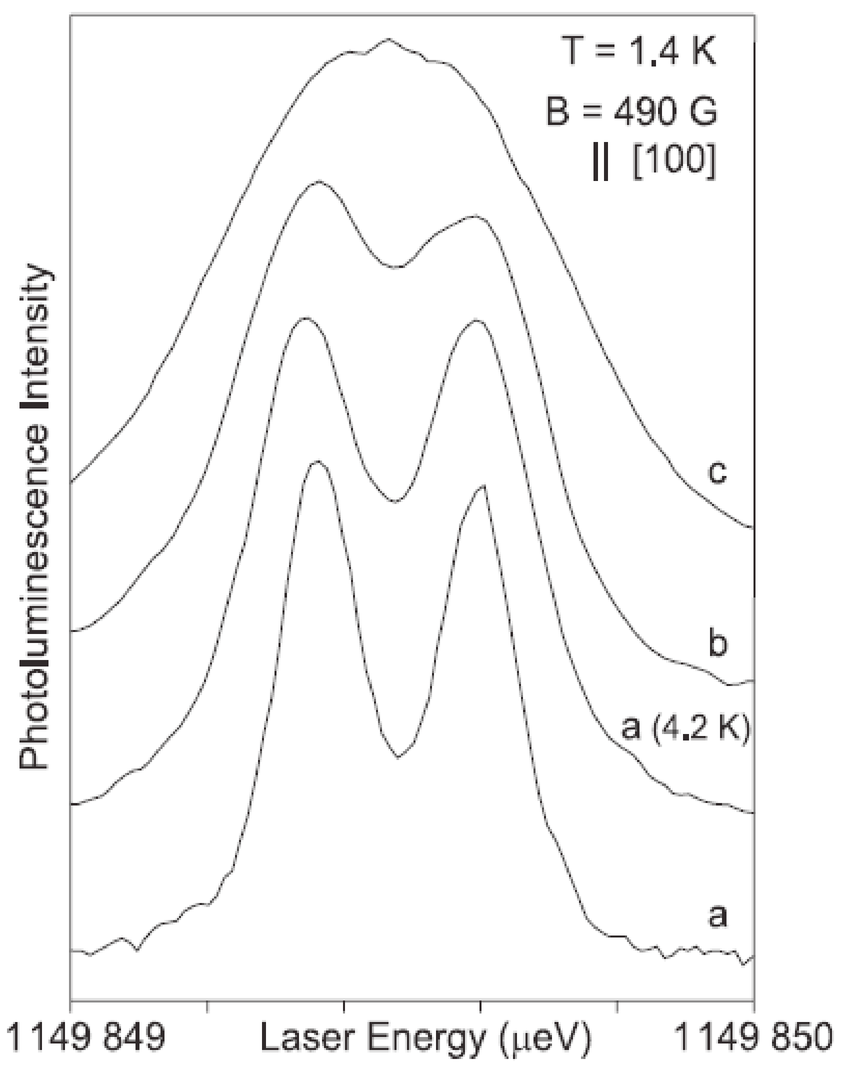
Feed rod: 6 kg polycrystalline ^{28}Si (99,994%), 65mmØ, 850mm length, made in the Institute for High Purity Substances (Nizhni Nowgorod, Ru) by CVD of Silan ($^{28}SiH_4$) synthesised from $^{28}SiF_4$ enriched by Centrotech St. Petersburg

Physical purification at IKZ:
- 11 Floating-Zone-runs (4 runs in vacuum)
- carbon reduction $<3 \cdot 10^{14}$ cm^{-3}

Final crystal(4,6kg) grown with diameter variation to enable two instead of only one 1kg-spheres (fig. right)

Remainig material ... sic research effords





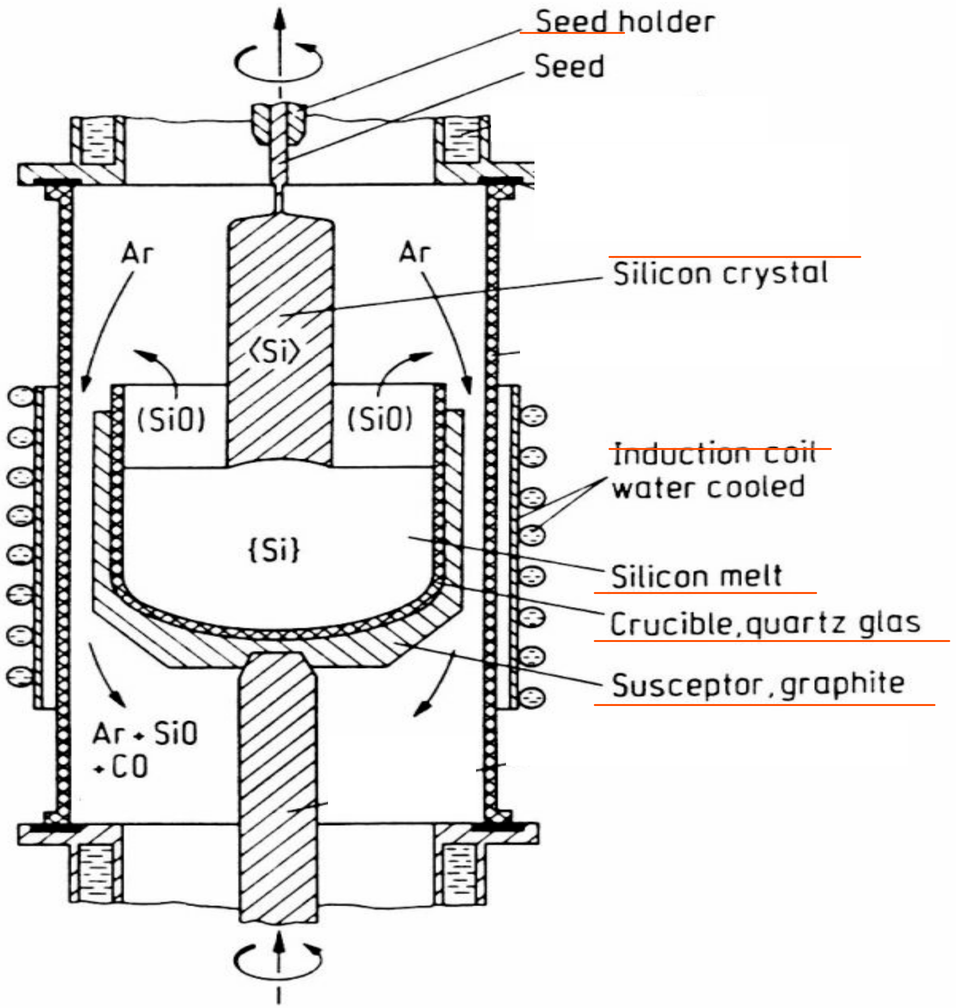
Dependence of the phosphorus bound exciton PLE line shapes on temperature and isotopic enrichment.

The spectrum labeled b is for a sample enriched to 99.983% 28Si, and the spectrum labeled c is for a sample enriched to 99.92% 28Si, both at 1.4 K.

Source: M.Thewalt

Germanium crystal growth, GERDA activities

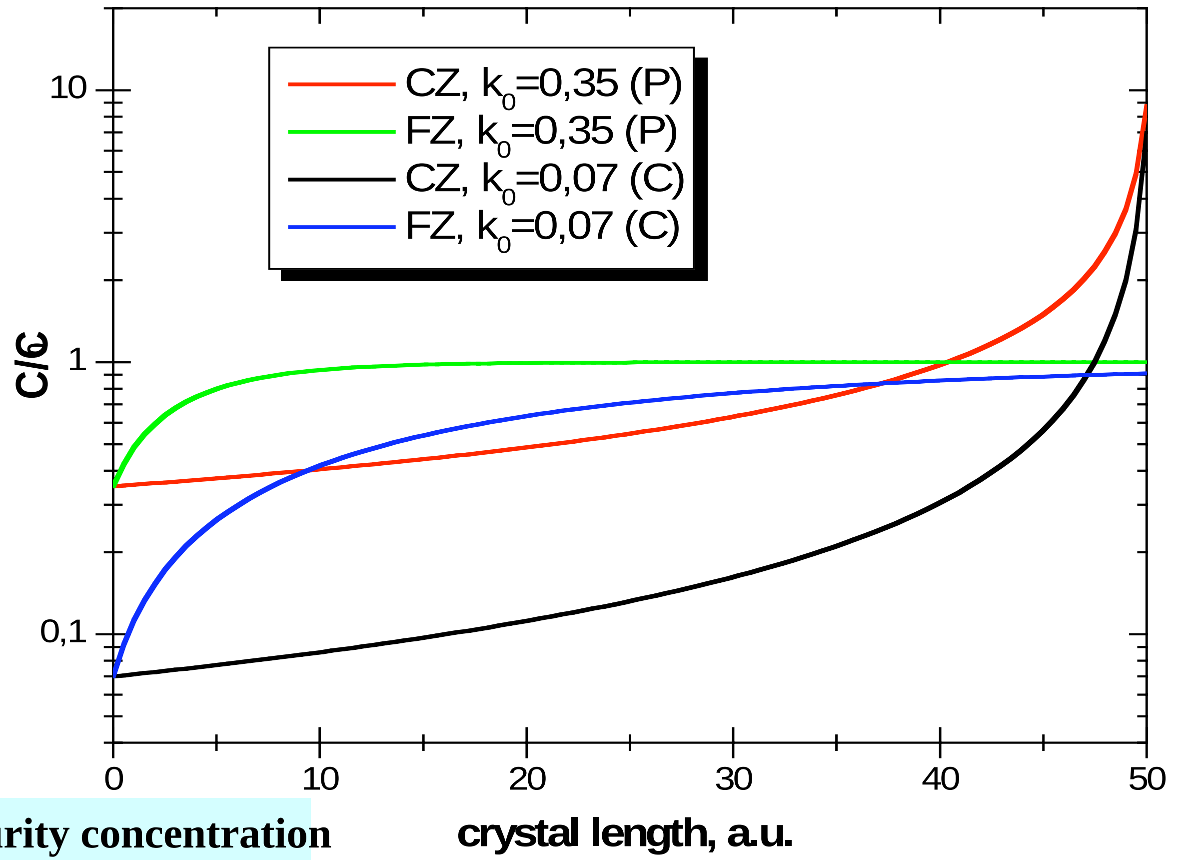
Inductively heated Czochralski (CZ) technique



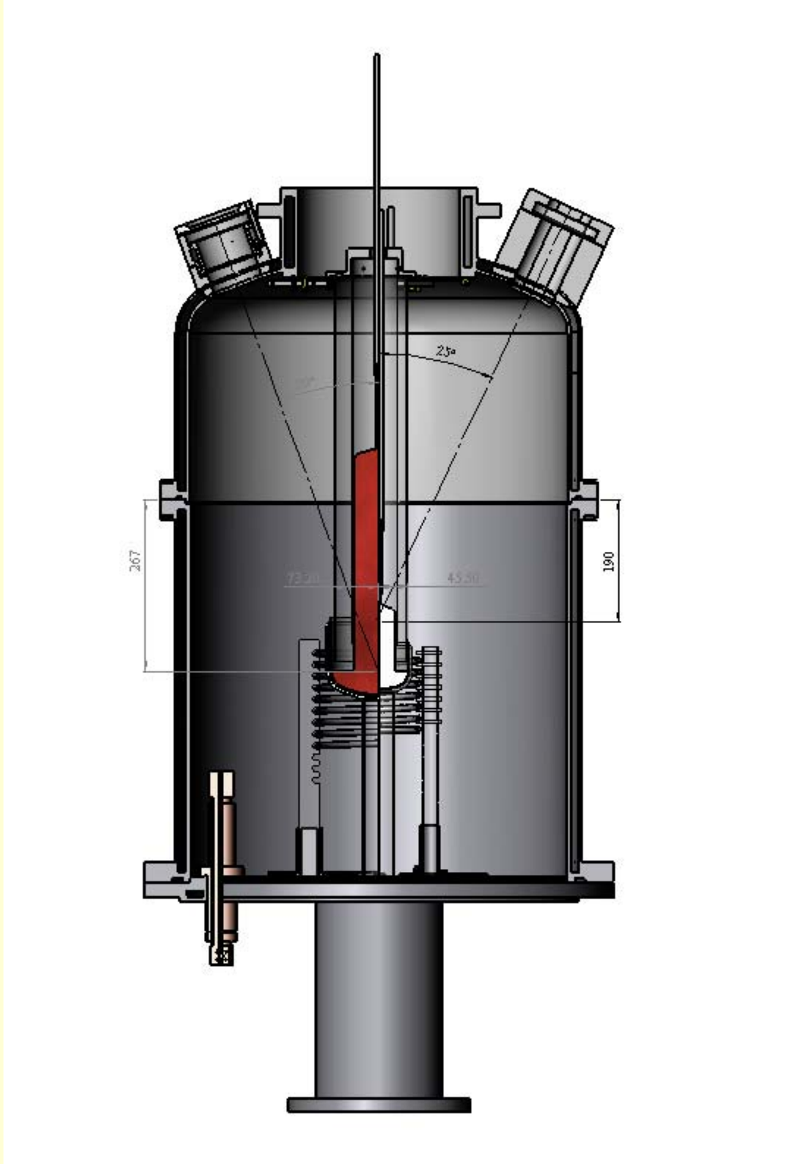
- **Starting material of arbitrary shape**
- **Effective purification by segregation**
- **Down scaling possible (mini-CZ)**

- Impurities from crucible (O,Al)
- Input of common silicon by crucible erosion (ca.50 µm/h)
- Common silicon from seed if it is not mono isotopic

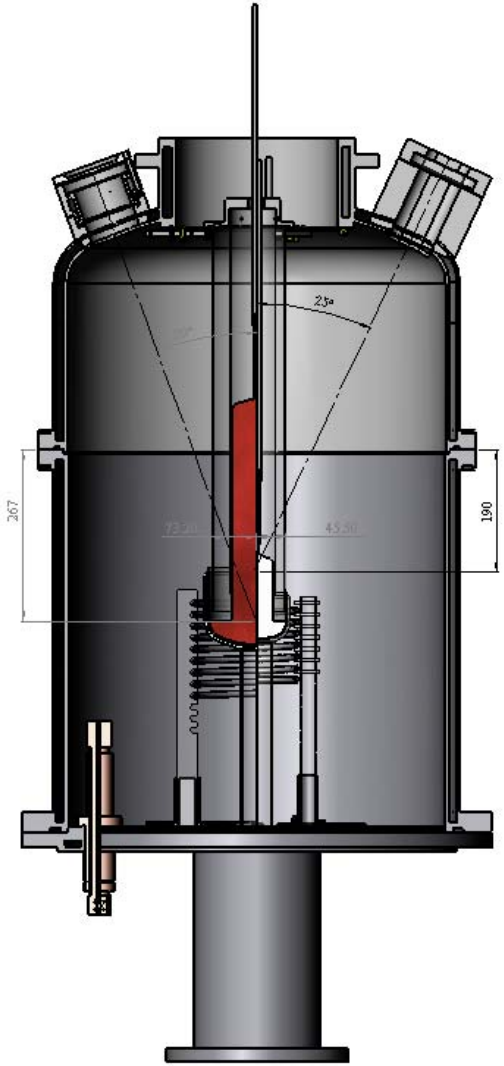
Impurity segregation for CZ and FZ growth

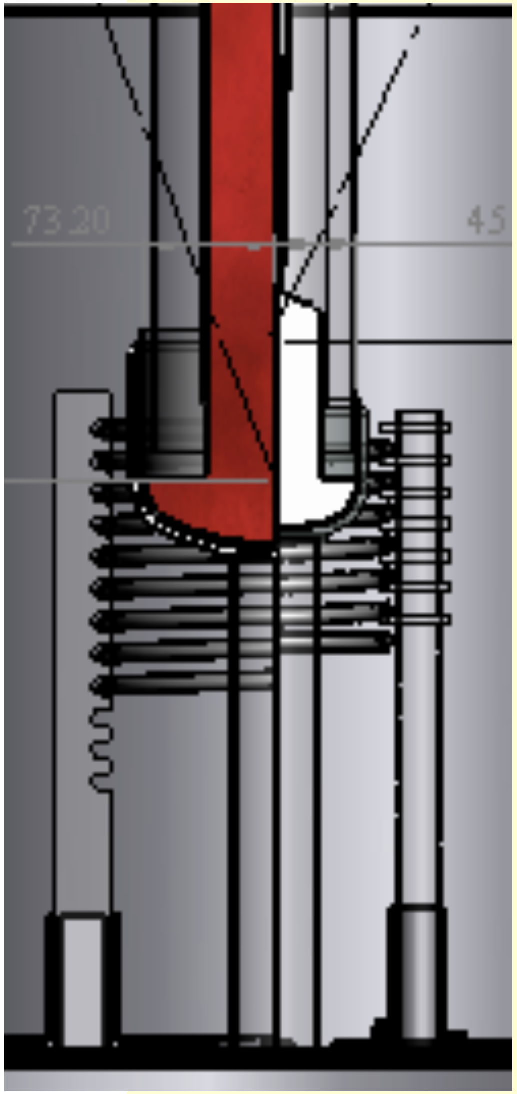


C - impurity concentration
C_o- concentration in feed

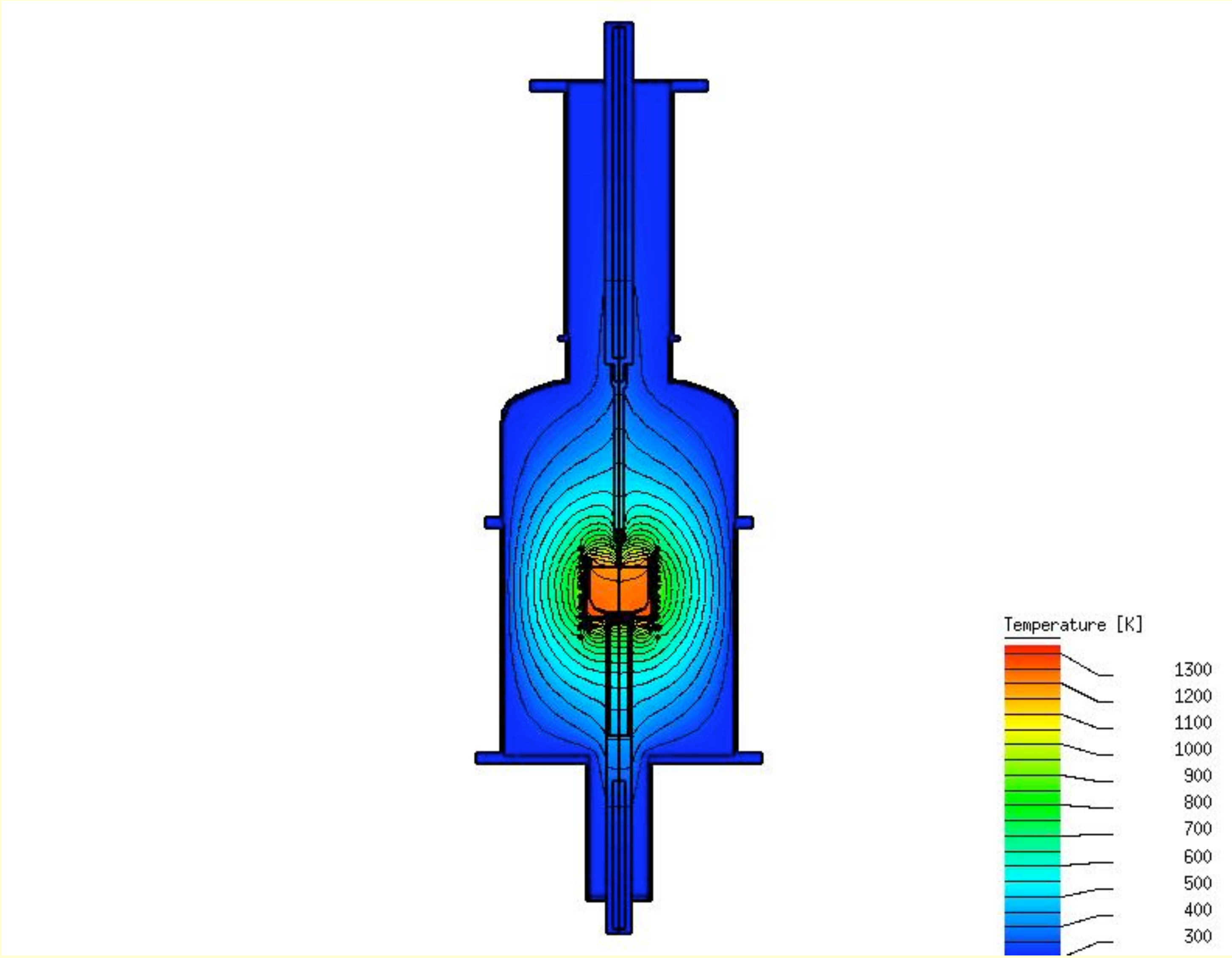


267
190

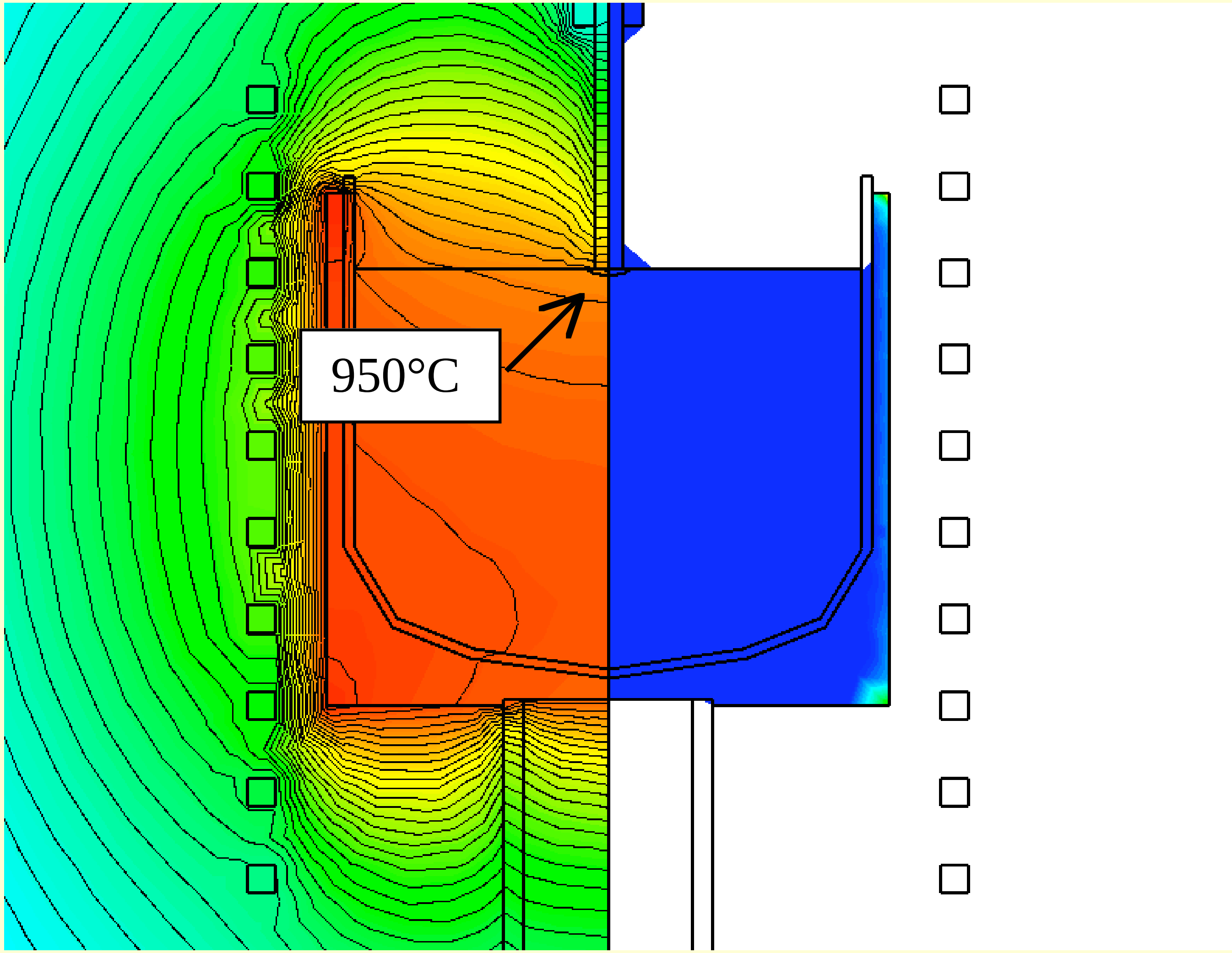


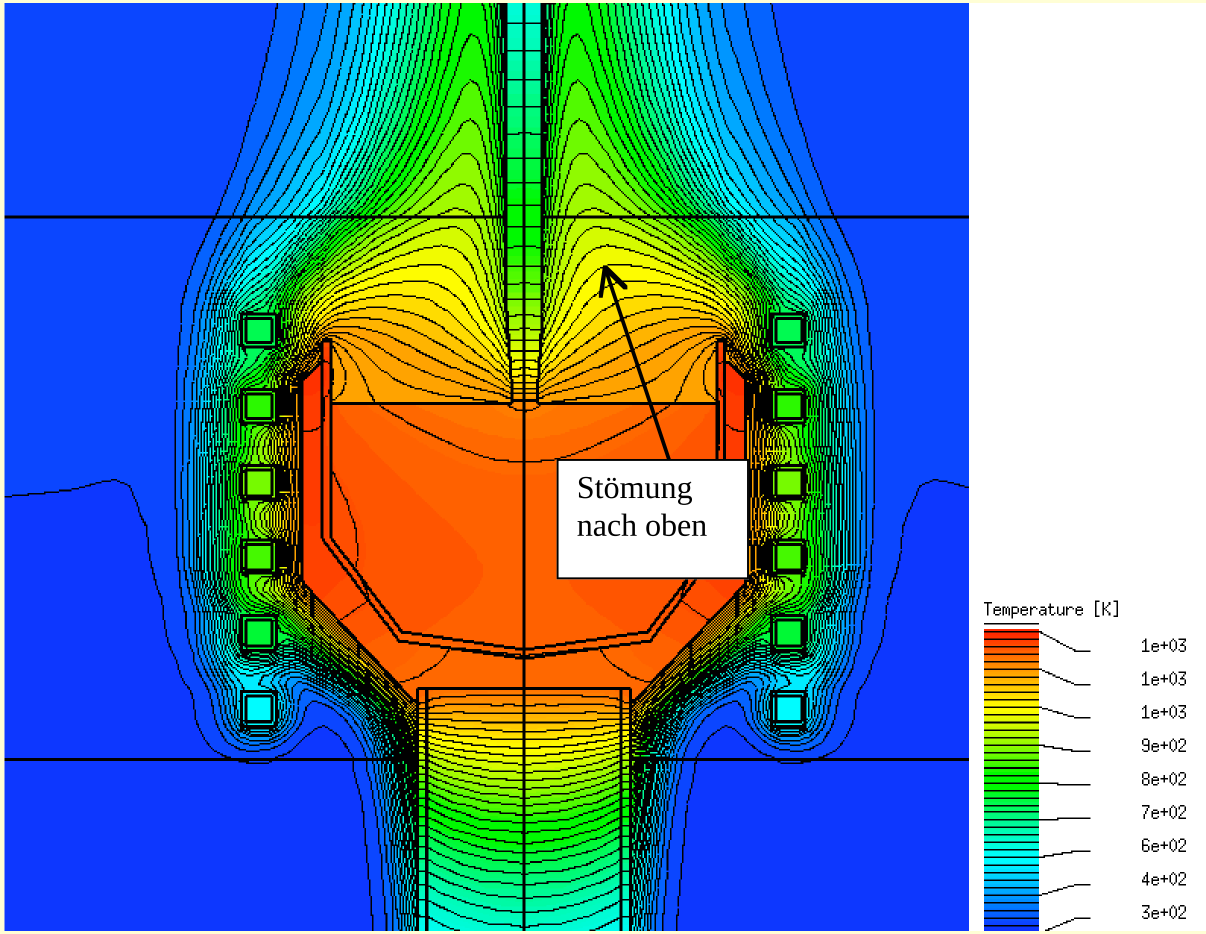


Spule	→	Silberrohr
Stütztiegel	→	Mo, TZM
Spulenhalter	→	Quarz
Tiegel	→	Quarz
Gasführung	→	Quarz
Keimstange	→	TZM



simulated global temperature field, Mo-susceptor (150 mm), ΔT=50 K, without convection – P ≈ 7,1 kW

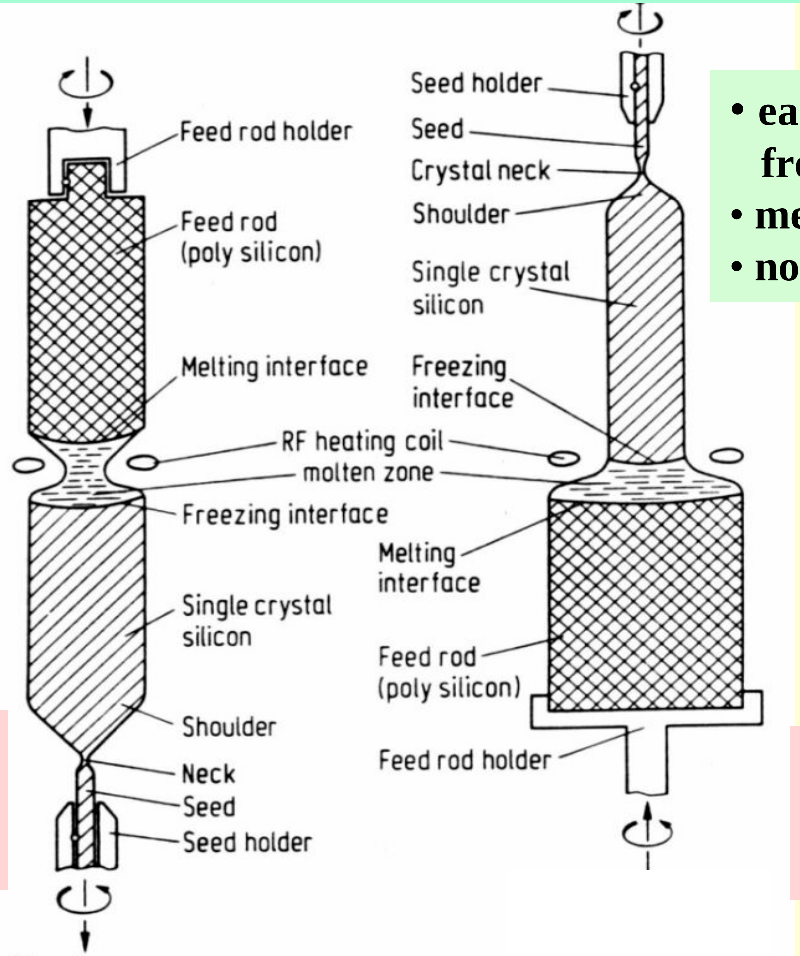

950°C



Calculated with gas convection

Crucible-less Growth Techniques also for Germanium??

- natural silicon seed possible
- evaporation of impurities
- crystal diameter free
- no crucible contamination



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Thank You for Your Attention!