Numerische Analyse der hydrodynamischen Stabilität in der Schmelze bei der Cz-Züchtung oxidischer Kristalle

In Zusammenarbeit mit:

H. Wilke, Mentor, Leibniz Institute of Crystal Growth
K. A. Cliffe, University of Nottingham, UK
A. Yu. Gelfgat, Tel-Aviv University, Israel

OUTLINE

- Motivation
- Fundamentals of crystal growth
- Introduction into basics of stability analysis / bifurcations
- Validating the code / software development – collaboration with TAU / UoN
- Bifurcation analysis in crystal growth (for the first time in IKZ)
- Measurement of important material properties
- Application to the Cz-oxide growth technology
- Final remarks

Motivation

Rare-earth scandate crystals ($ReScO_3$, Re=Y, La, Pr, Nd, **Sm**, Gd, Tb **Dy**, Ho, Er, Tm and Lu) showing spiral growth. These crystals are excellent candidates for substrates of ferroelectric materials (e.g. non-volatile FeRAM) or alternative gate high-K-dielectrics for MOSFETs. The Czochralski technology has been used.

Motivation

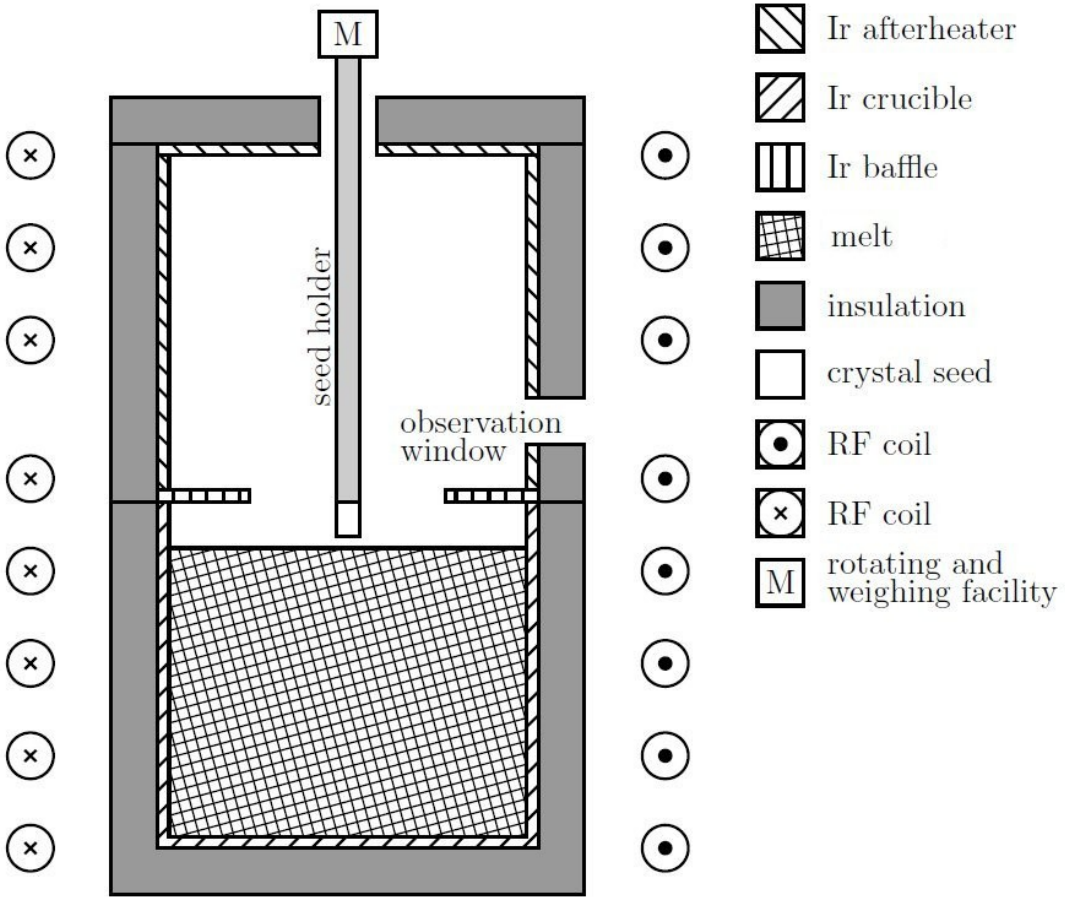
- The phenomenon of spiral crystal growth is a still unsolved problem
- There is a deep impact with commercial requirements for special oxide crystals
- The cork screw instability is a typical example of 2D symmetry breaking
- A stability analysis can be performed in terms of fluid flow interaction
- For the first time in IKZ bifurcation analysis has been used
- Experimental investigation of material properties used for numerics
- Characterizing the solution type – multiple solutions
- **Hypothesis: Heat and momentum changes initiate the spiral growth**

Fundamentals of crystal growth (1)

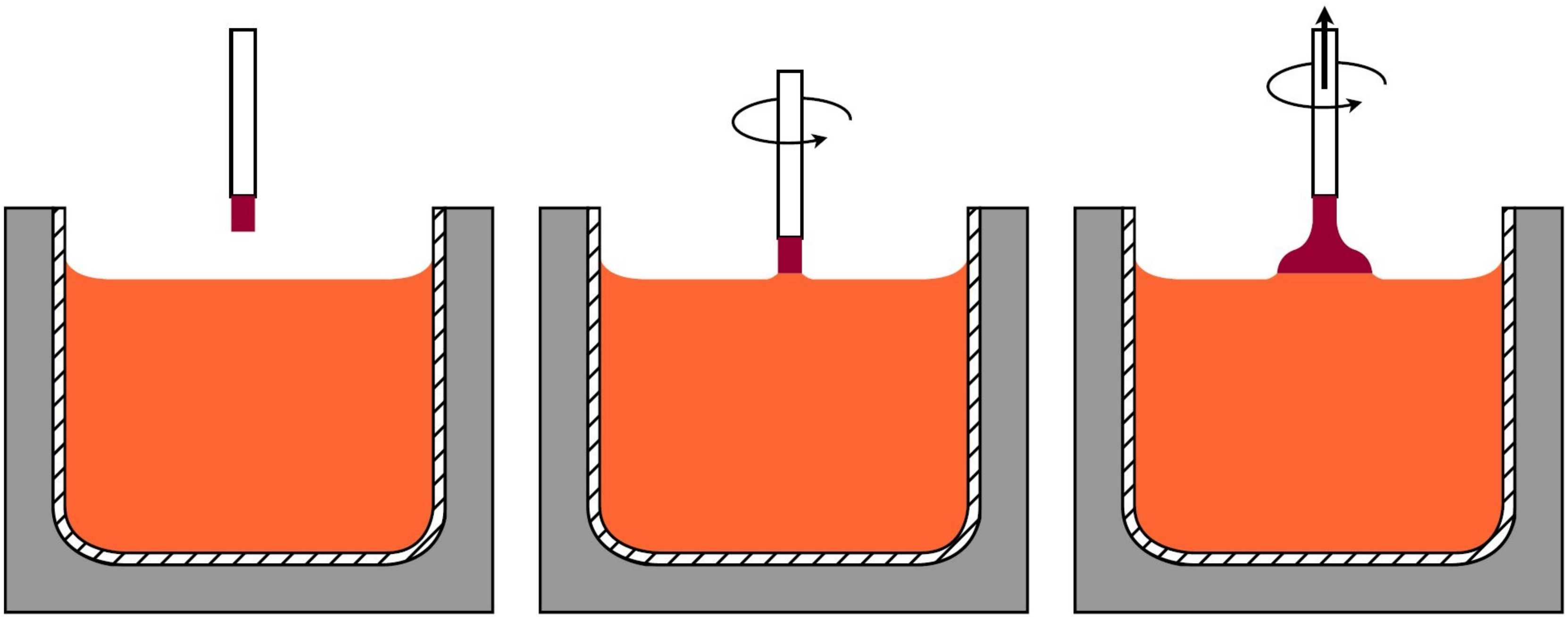
Since 1950's the growth of crystals is applied industrially using different methods

- From gas phase epitaxially (e.g. chemical vapour deposition - CVD, MOCVD)
- From chemical solution (e.g. top seeded solution growth - TSSG)
- From the melt (e.g. Bridgman, Czochralski(Cz) or floating zone(FZ) method)
- Best quality bulk crystals are achieved with Cz and FZ methods

Fundamentals of Cz crystal growth (1)



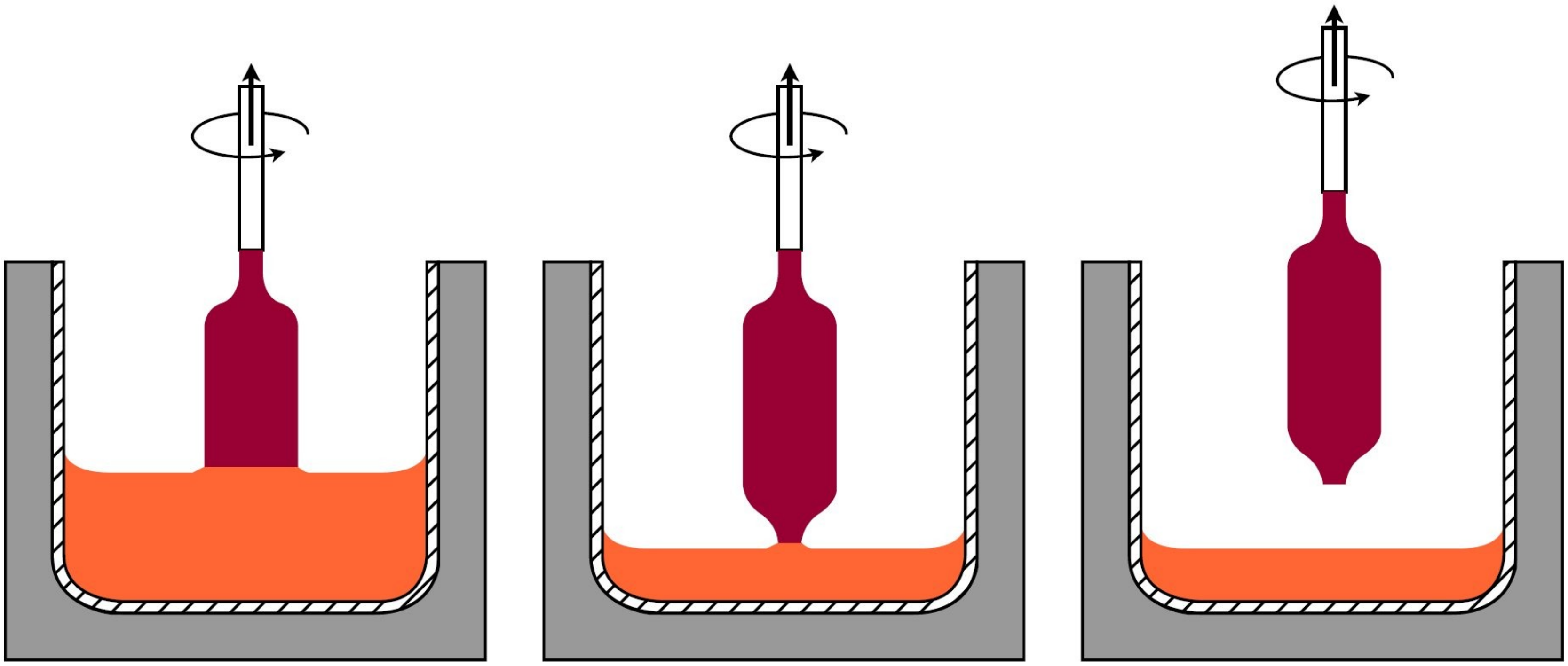
Fundamentals of Cz crystal growth (2)



(a) melting process (b) seeding process (c) crowning process

Sketch of first three principle steps in a Cz crystal growth run

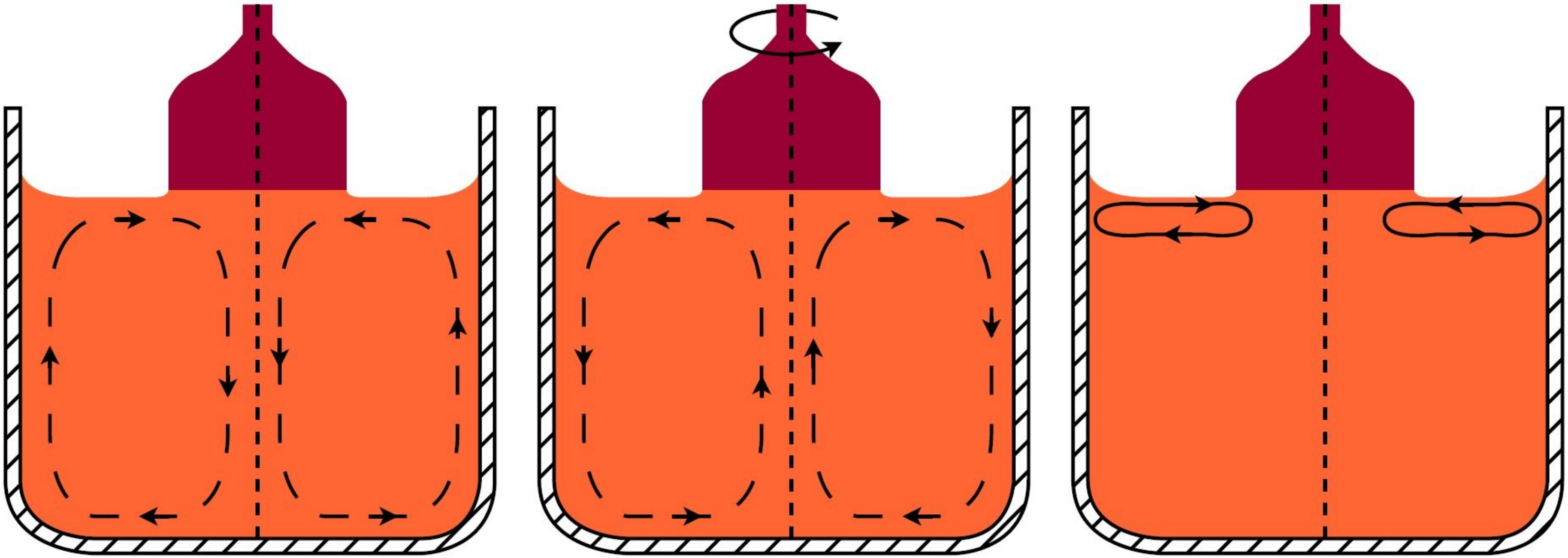
Fundamentals of Cz crystal growth (3)



(a) growing process (b) tailing process (c) removing and cooling process

Sketch of last three principle steps in a Cz crystal growth run

Fundamentals of Cz crystal growth (4)



(a) flow due to buoyancy (b) flow due to rotation (c) Marangoni flow

Cz melt flow mechanisms

Fundamentals of crystal growth – heat and mass transfer

- heat transfer

conduction $q = -\kappa \nabla T$

convection $\chi \nabla^2 T - \vec{v} \cdot \nabla T = 0$

- fluid motion

Navier-Stokes equations

$$\underbrace{\overbrace{\rho \frac{\partial \vec{v}}{\partial t}}^{\text{unsteady acceleration}} + \overbrace{\rho \vec{v} \cdot \nabla \vec{v}}^{\text{convective acceleration}}}_{\text{momentum changes per element}} = \underbrace{\rho \vec{f}}_{\text{other external forces}} - \underbrace{\overbrace{\nabla p}^{\text{pressure gradient}} + \overbrace{\nabla(\mu \nabla \vec{v})}^{\text{viscous forces}}}_{\text{stress divergence}}$$

Continuity equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0$$

$$\nabla \cdot \vec{v} = 0$$

Marangoni convection

$$\mu_1 \frac{\partial u_1}{\partial z} - \mu_2 \frac{\partial u_2}{\partial z} = \frac{\partial \gamma}{\partial x} = \frac{\partial \gamma}{\partial T} \frac{\partial T}{\partial x}$$

Radiation $q = \sigma \epsilon (T^4 - T_a^4)$

- parameters

$$Ra = Gr\, Pr = \frac{g\beta}{\nu \chi} T_m R^3$$

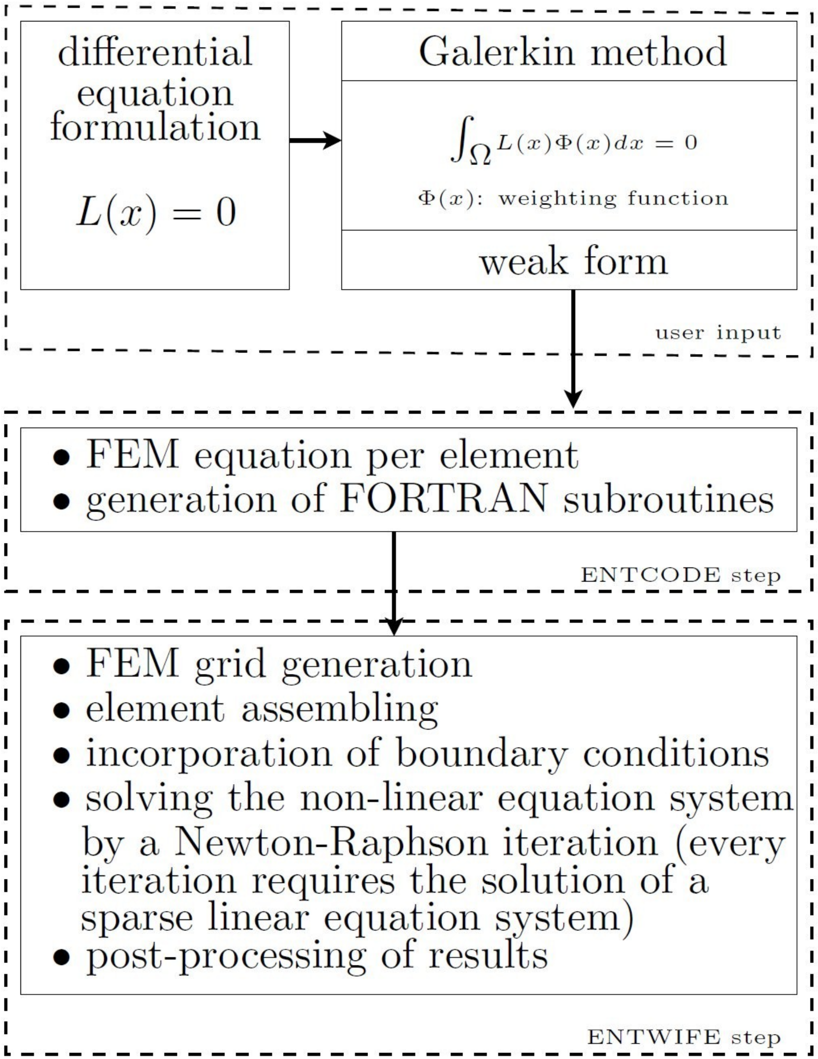
$$Gr = \frac{g \beta T_m R^3}{\nu^2}$$

$$Pr = \frac{\nu}{\chi}$$

$$Ma = \frac{\left| \frac{d\sigma}{dT} \right| T_m R}{\eta \nu}$$

$$Re = \frac{\omega R^2}{\nu}$$

FEM-Software (ENTWIFE)




```
>>ENTWIFE
    <further data>

>>MODEL DATA
    <further data>

>>SOLVER DATA
    <further data>

>>OUTPUT DATA
    <further data>

>>STOP
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further advantages:

- symbolic equation input via interface to MATHEMATICA or MAPLE
- Newton-Raphson solver convergence O(2)
- Interface to a parallel sparse direct solver (MUMPS), which allows for a simulation on a supercomputer (e.g. HLRN)
- Continuation and bifurcation algorithms (Hopf-bifurcation shows a superconvergence O(4))

What is bifurcation / path following good for?

Nonlinear dynamics: there are no fundamental solutions

- No a priori knowledge about the solution structure
- Dynamic systems often show a complex solution manifold and ambiguity
- Classical direct numerical simulation can miss important solution branches of the non linear dynamic system

The main questions are:

-What is the qualitative solution behaviour of the system?
-Which and how many different solution sets do occur?
-Which of them are un/stable?
-What is the bahaviour of different solution sets while changing the control parameter(s) of the system?

→ **Bifurcation: appearence and disappearence of different solution sets**

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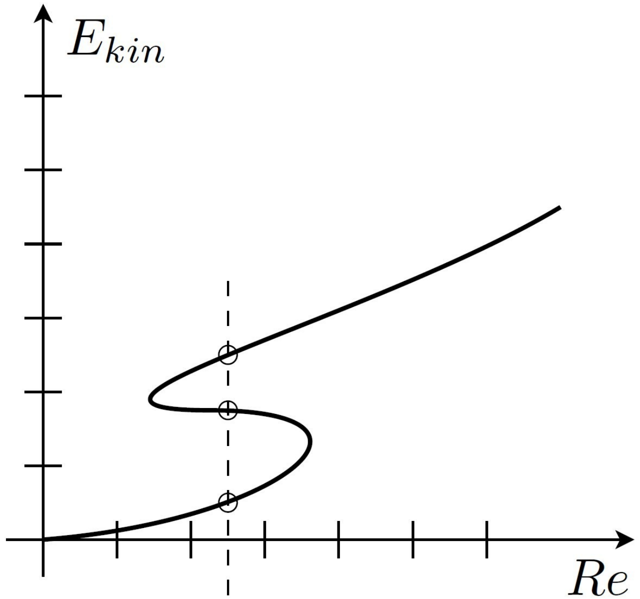
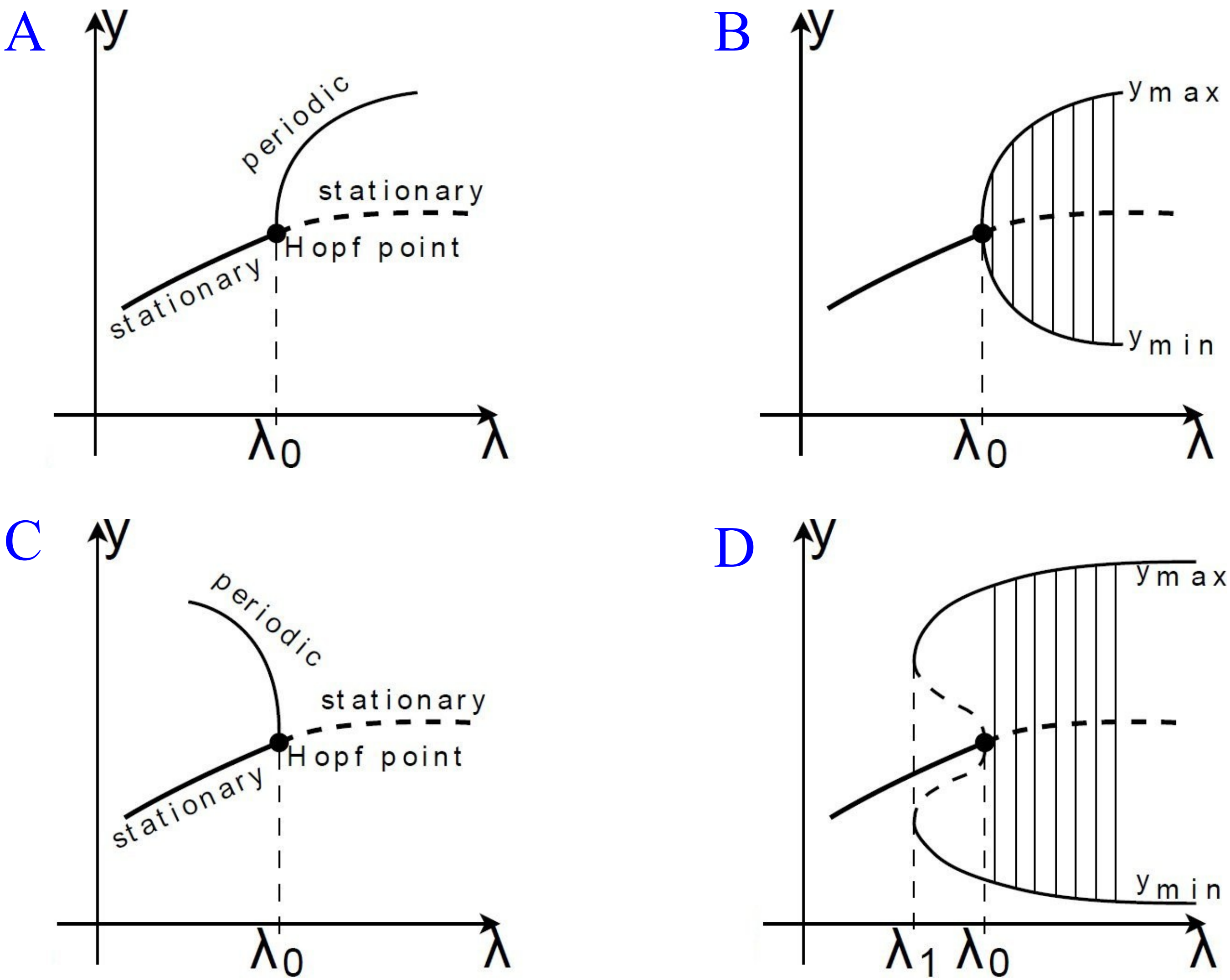
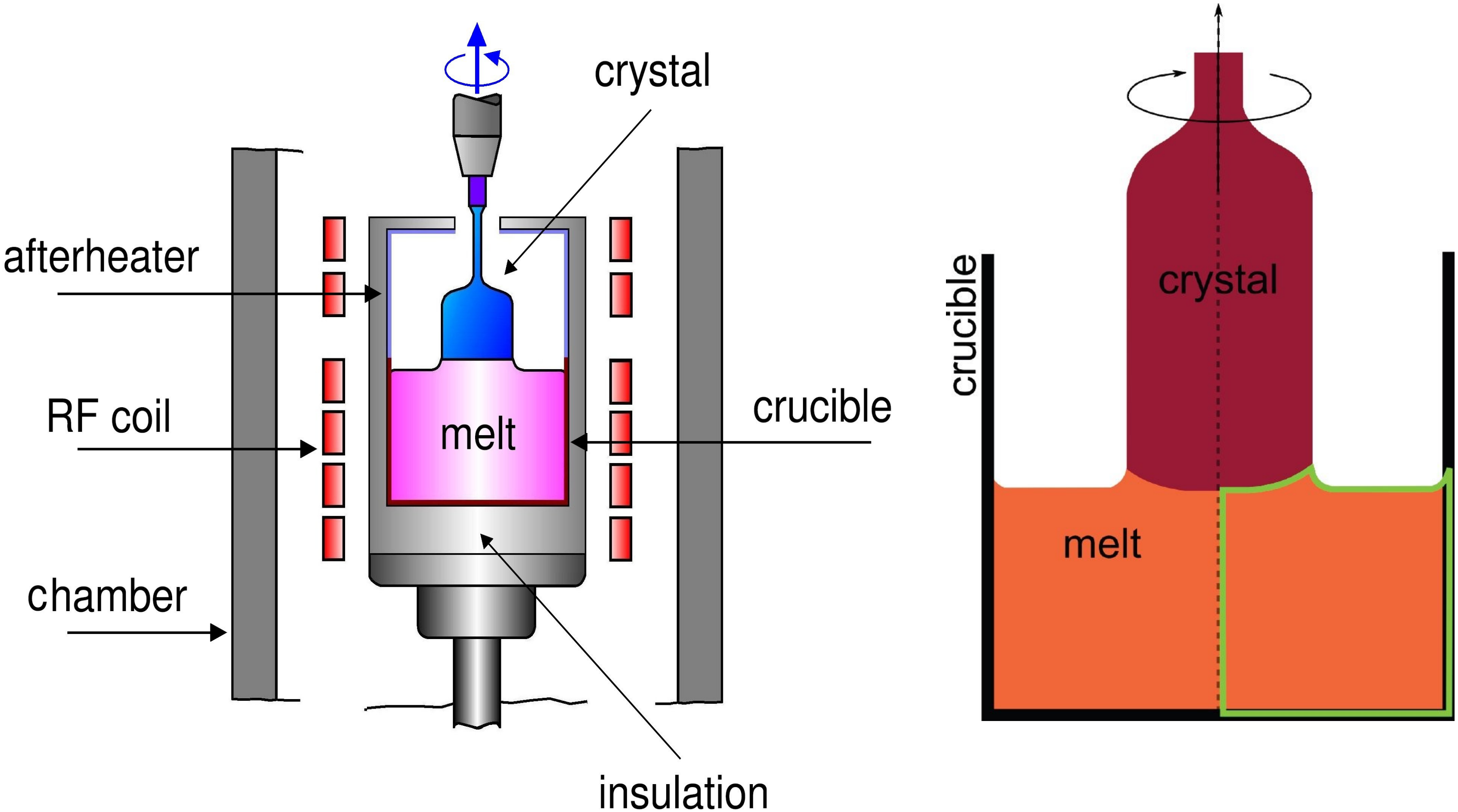


Path following: example of multiple solutions

Explanation by graphical examples (2)

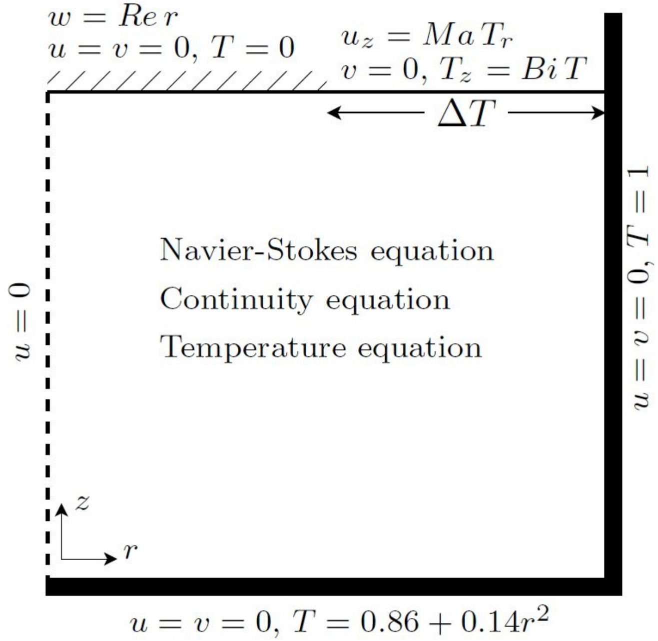


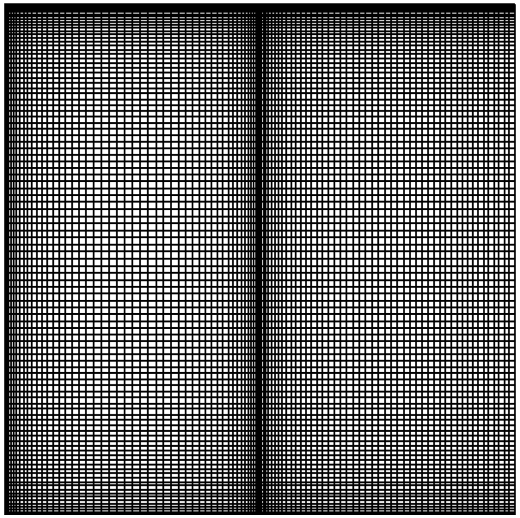
Validating numerical code – used model



Sketch of the Czochralski crystal growth technology

Non-equidistant FEM grid of 120 x 120 Q9/4 elements (ENTWIFE package)





Cylindrical coordinates scaling:

$$r := \frac{r}{R} \quad z := \frac{z}{R} \quad u := \frac{u\ R}{\nu} \quad T := \frac{T - T_m}{T_c - T_m} = \frac{T - T_m}{\Delta T}$$

Parameters: $$Gr = \frac{g\ \beta\ \Delta\ T\ R^3}{\nu^2},\quad Ma = \frac{\frac{d\sigma}{dT}\ \Delta\ T\ R}{\mu\ \nu},\quad Pr = \frac{\nu}{\kappa}$$

$$R\ e = \frac{\omega\ R^2}{\nu},\quad Bi = \frac{h\ R}{\kappa},\quad Ar = \frac{H}{R}$$

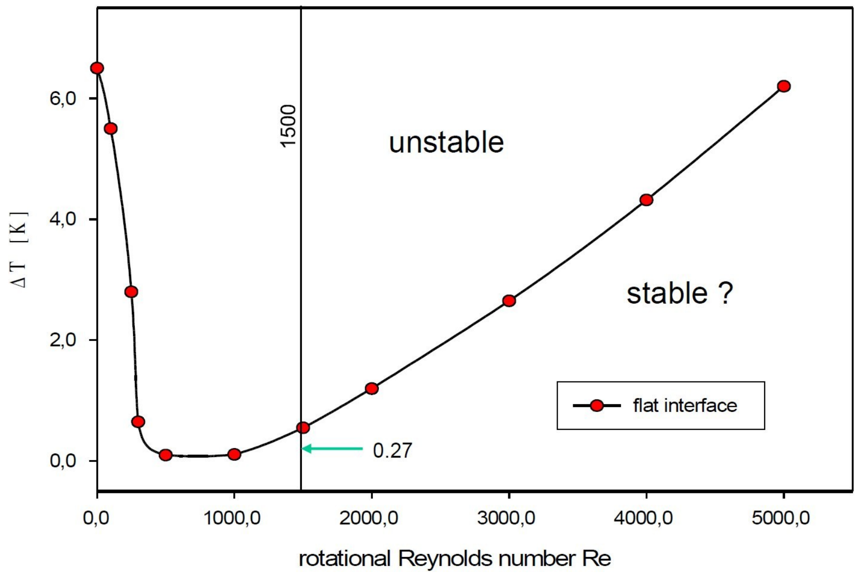
Material properties ($NaNO_3$) and geometry*:

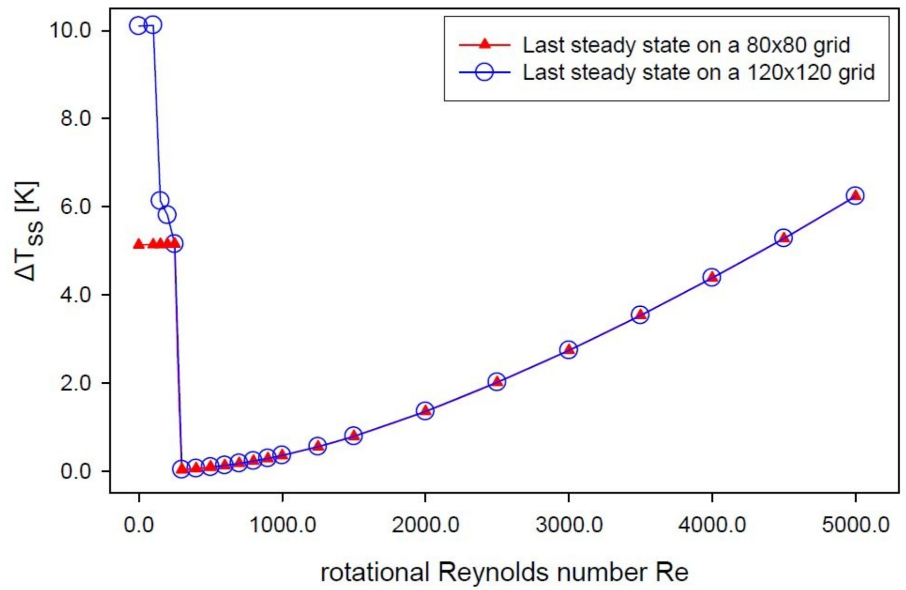
Pr=9.2
Gr=190476.0 ΔT
Ma=Mn/Pr=585.71 ΔT

Bi=0.1
H=0.92
R=1.0
$R_{crucible}$=3.8 cm

(* Schwabe et al., JCG 265 (2004), p. 440)

Stability diagrams and their grid dependance

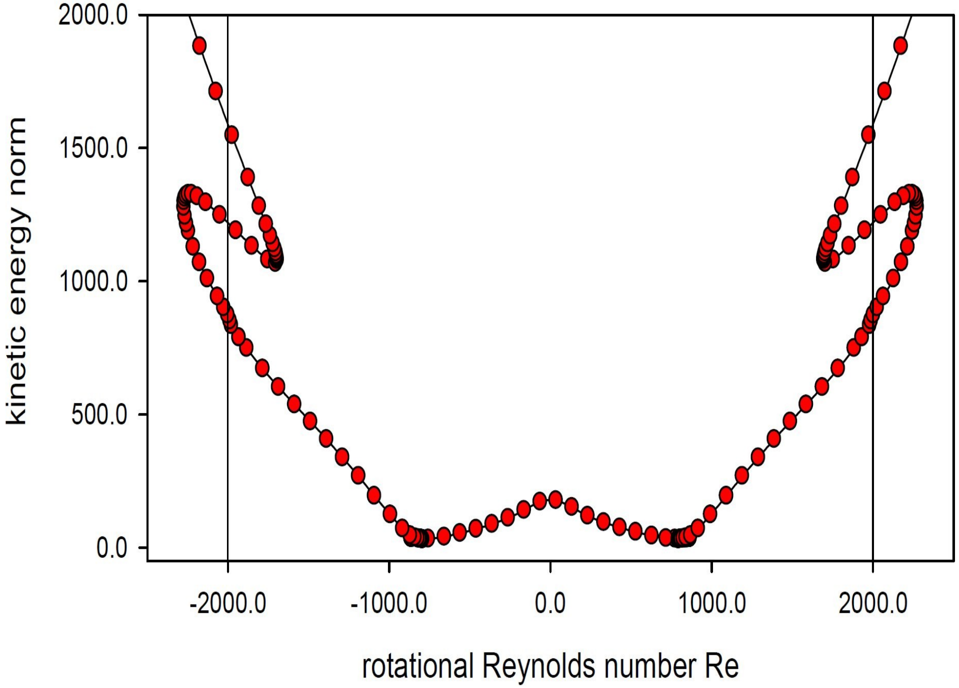




Stability diagrams of $NaNO_3$-melt flow in a Czochralski crucible (see also Gelfgat et al., J. Crystal Growth 275 (2005) e7)

Path following

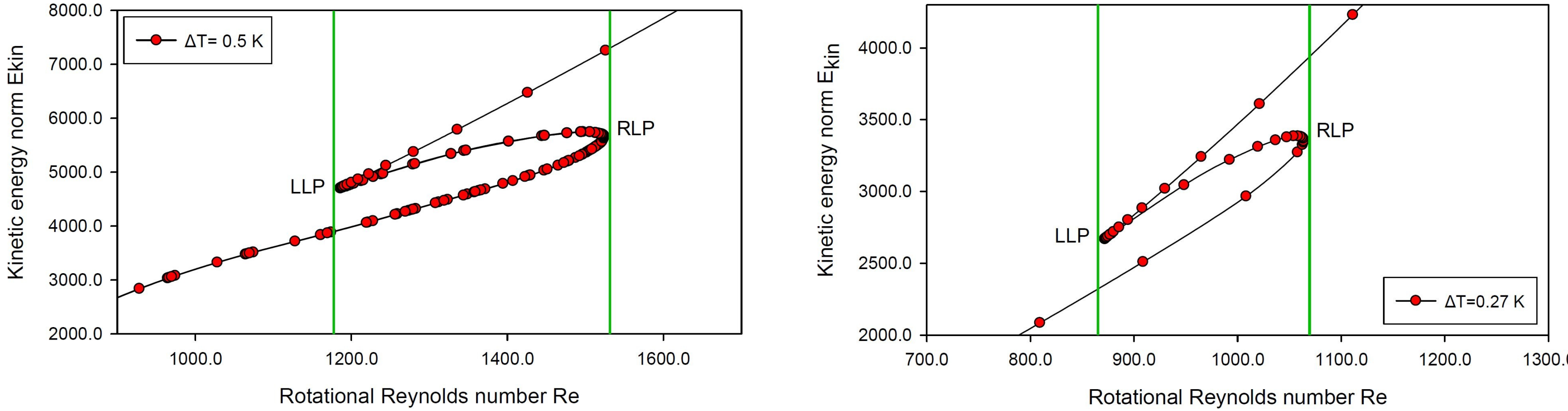
Continuation diagram for Parameter Re



Rotation direction change ($DyScO_3$)

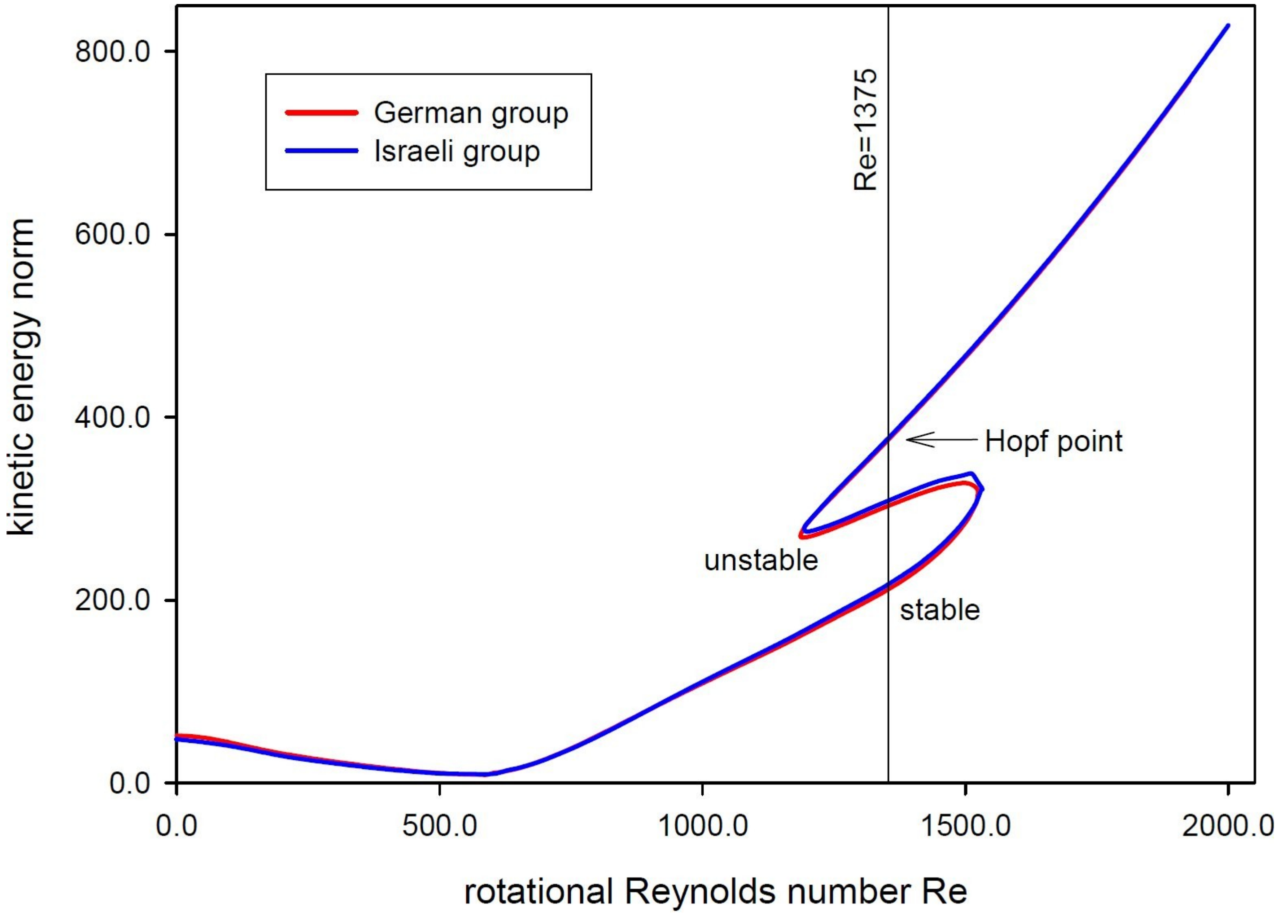
total kinetic energy norm $E_{kin} = 2\pi \int_0^H \int_0^R \left(u^2 + v^2 + w^2 \right) r dr dz$

Multiplicity of solution in 2D



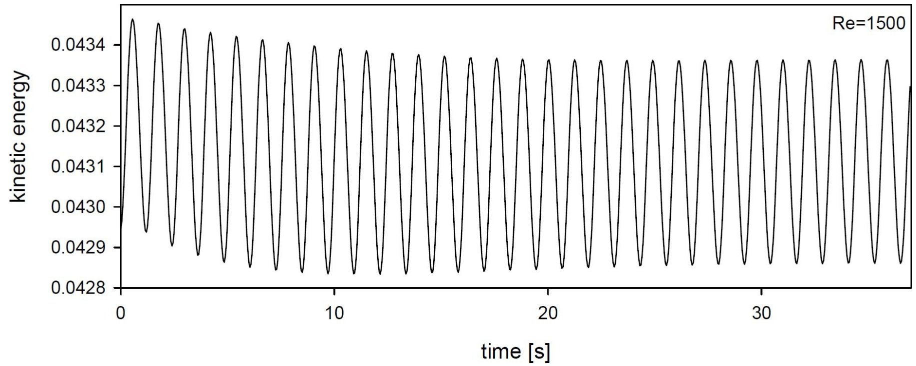
Continuation diagrams for parameter Re

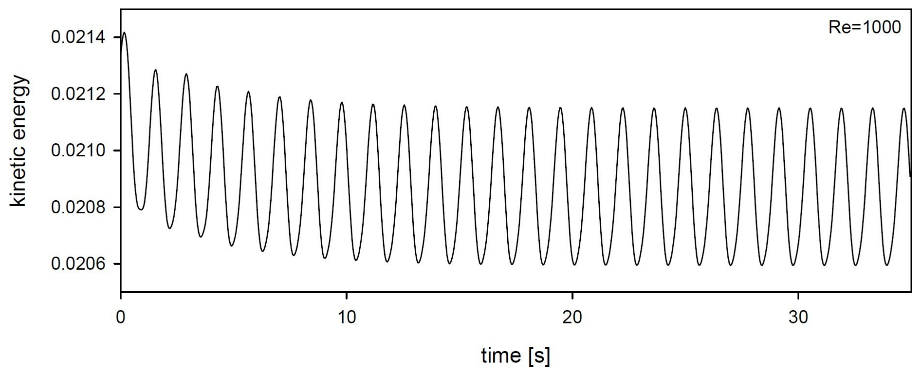
Comparison of continuation diagrams

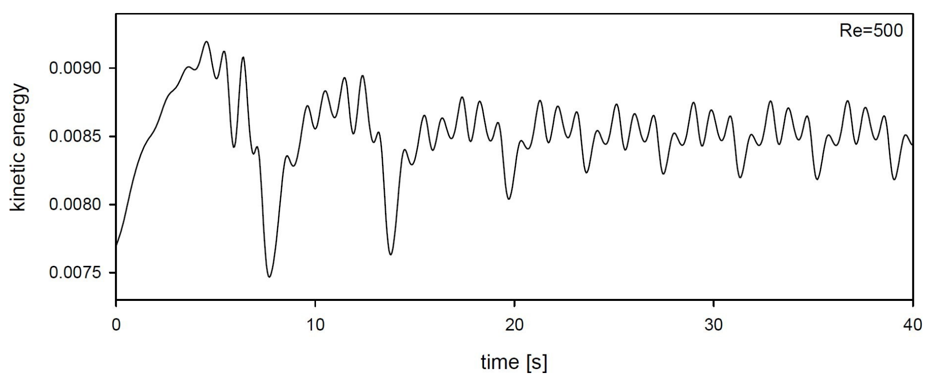


Continuation diagrams for control parameter Re with ΔT=0.27 K

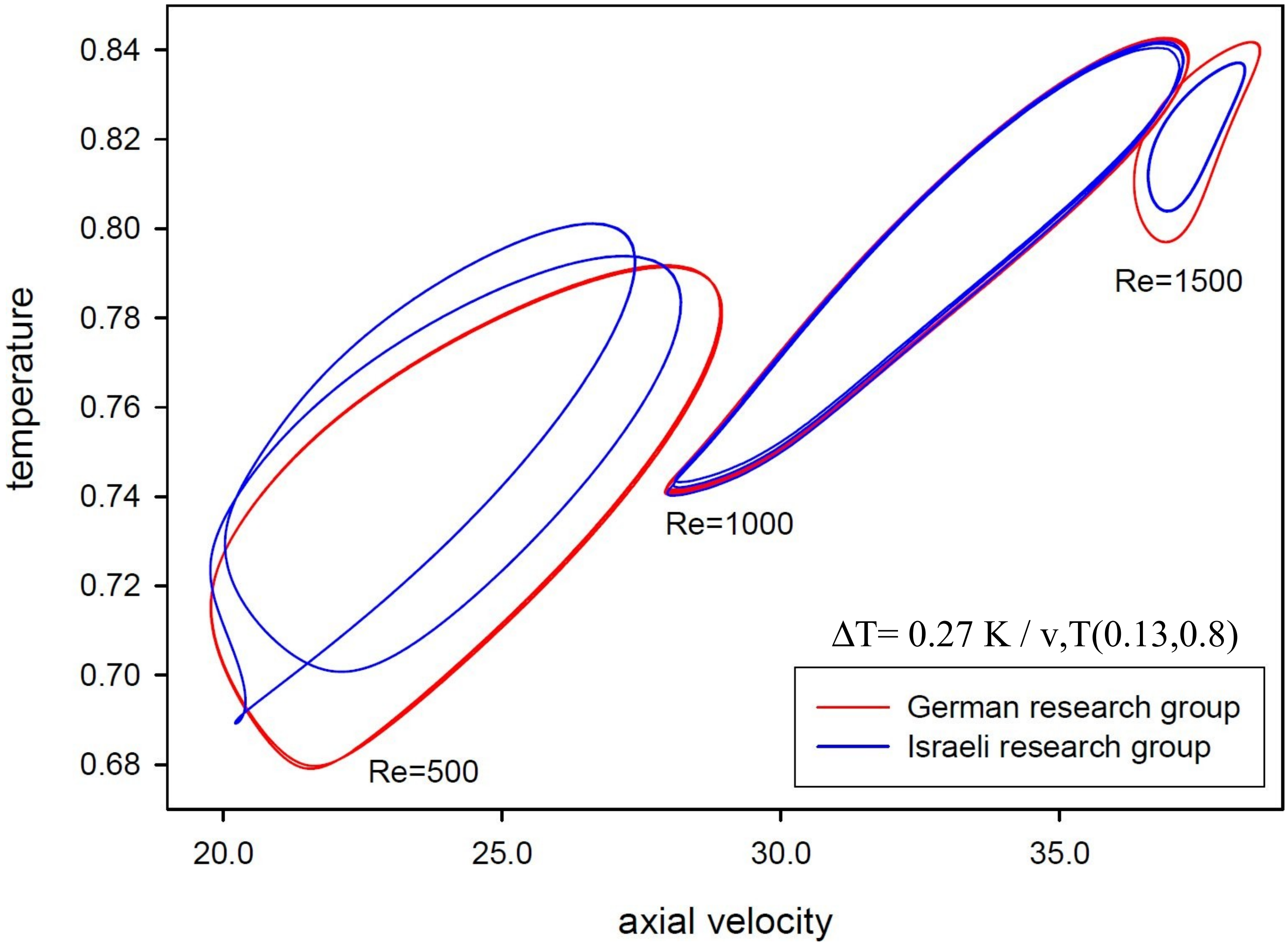
Transient simulations for different Re and constant ΔT=0.27K

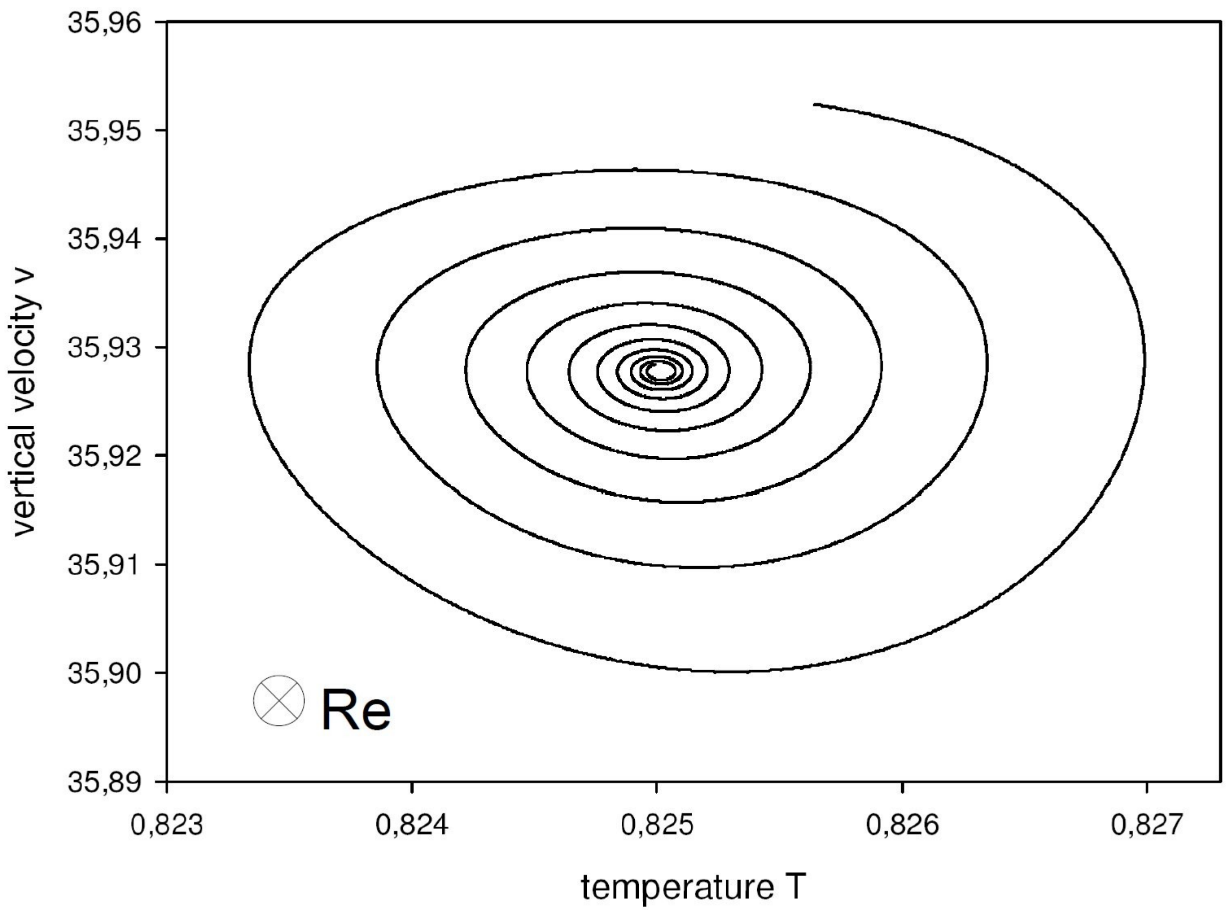






Phase plots for different Re close to the solid/liquid interface





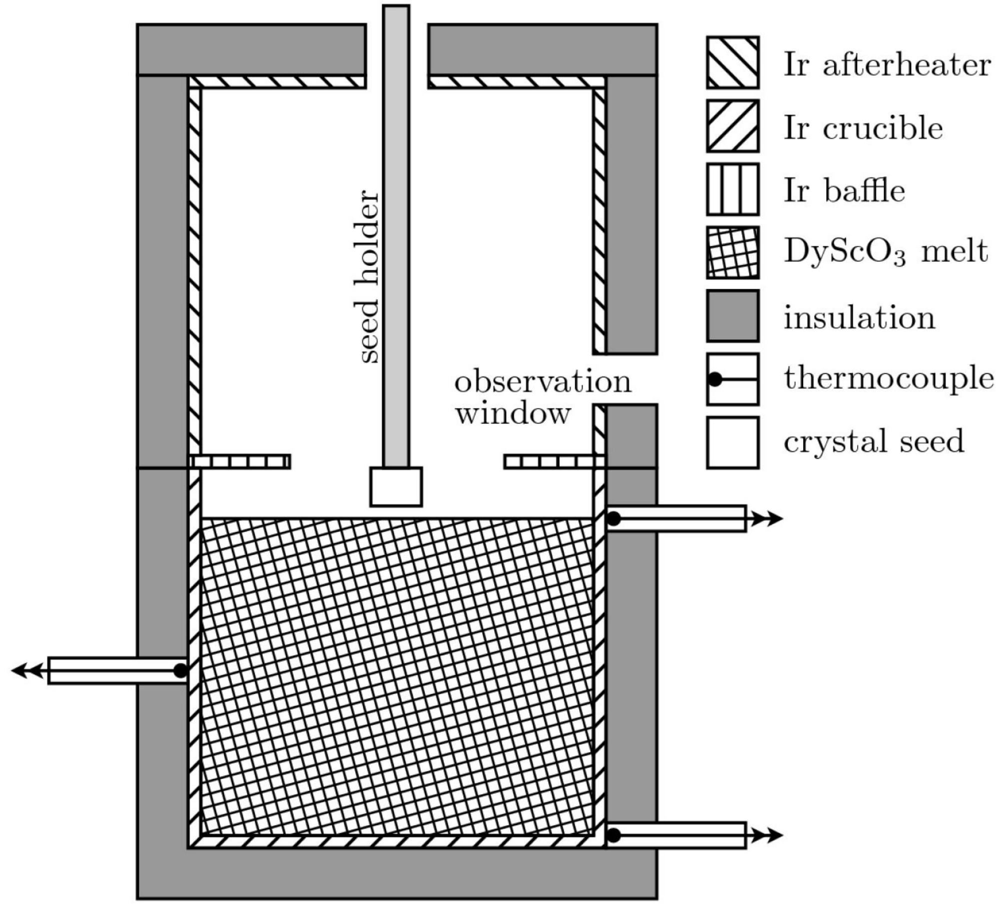
Phase portraits for $\Delta T = 0.27K$ at v, T(0.13, 0.8) while varying Re

Measurement of important material properties

Which physical quantities / material properties are interesting?

- Density
- Viscosity
- Thermal expansion coefficient
- Specific heat
- Thermal conductivity
- Melting point
- Vertical temperature gradient
- Surface tension
- Temperature dependence of material properties

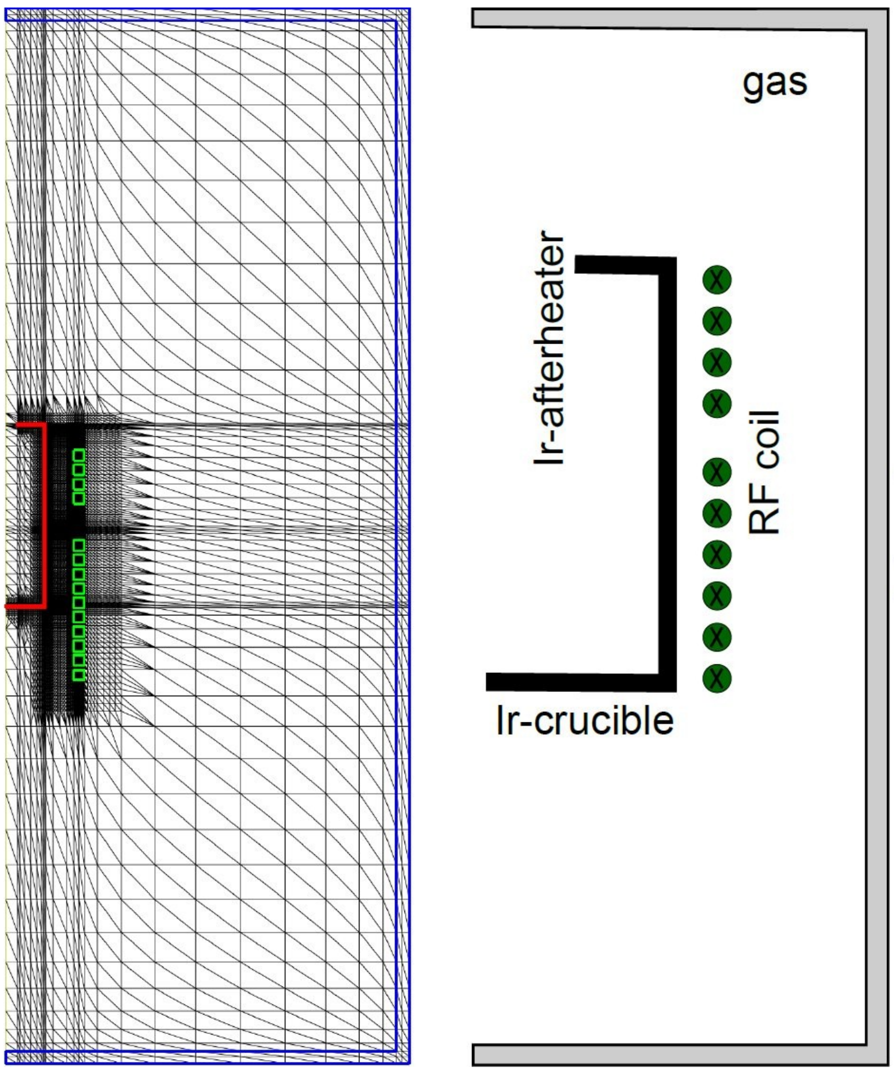
1900 °C - 2100 °C

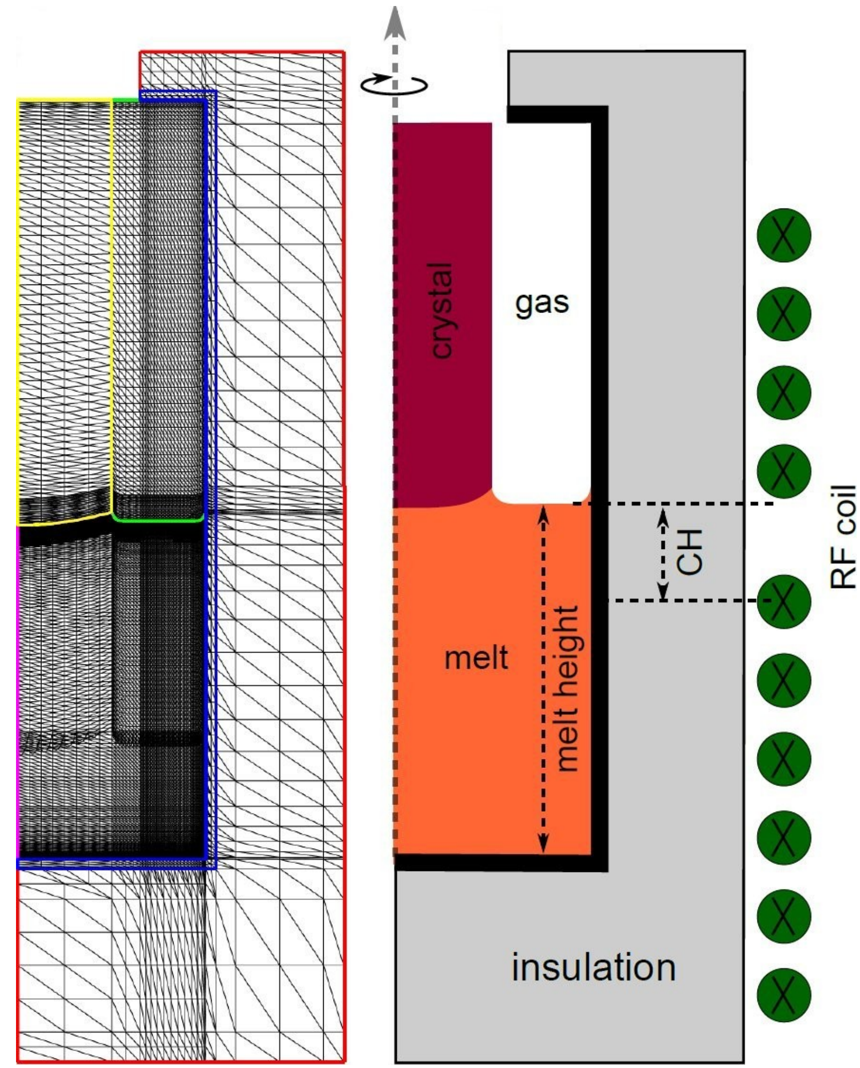


sketch of the measurement setup

Application to the Cz-oxide growth technology

Influence of the RF-heating configuration for different melt heights





Model problem and numerical method

magnetic stream function

$$\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \psi_B}{\partial r}\right) + \frac{\partial}{\partial z}\left(\frac{1}{r}\frac{\partial \psi_B}{\partial z}\right) = \mu J$$

$$\text{where} \quad J = \begin{cases} J_0 \cos \omega t & \text{in the coil} \\ -\frac{\sigma_c}{r}\frac{\partial \psi_B}{\partial t} & \text{in the conductors} \end{cases}$$

$$\text{with} \quad \psi_B = C(r,z)\cos(\omega t) + S(r,z)\sin(\omega t)$$

induced heat in metallic parts

$$Q = \frac{\sigma_c \omega^2}{2r^2}(C^2 + S^2)$$

heat and mass transfer

$$\rho \vec{V} \cdot \nabla \vec{v} = -\nabla p + \mu \nabla^2 \vec{v} + \rho \vec{g}(T - T_0)\beta$$

$$k\nabla^2 T_s + Q_s = 0 \qquad s = \text{solid regions}$$

$$\frac{k}{\rho c_p}\nabla^2 T - \vec{v} \cdot \nabla T = 0 \qquad \text{in melt and gas}$$

$$\nabla \cdot \vec{v} = 0$$

thermocapillary convection

$$\mu_l \frac{\partial u_l}{\partial \hat{n}} - \mu_g \frac{\partial u_g}{\partial \hat{n}} = \frac{\partial \gamma}{\partial \hat{\tau}} = \frac{\partial \gamma}{\partial T}\frac{\partial T}{\partial \hat{\tau}}$$

parameters

$$Gr = \frac{g\beta T_m R^3}{\nu^2}$$

$$Pr = \frac{\nu}{\chi}, \quad \chi = \frac{k}{\rho c_p}$$

$$Ma = \frac{\left|\frac{d\sigma}{dT}\right| T_m R}{\eta \nu}$$

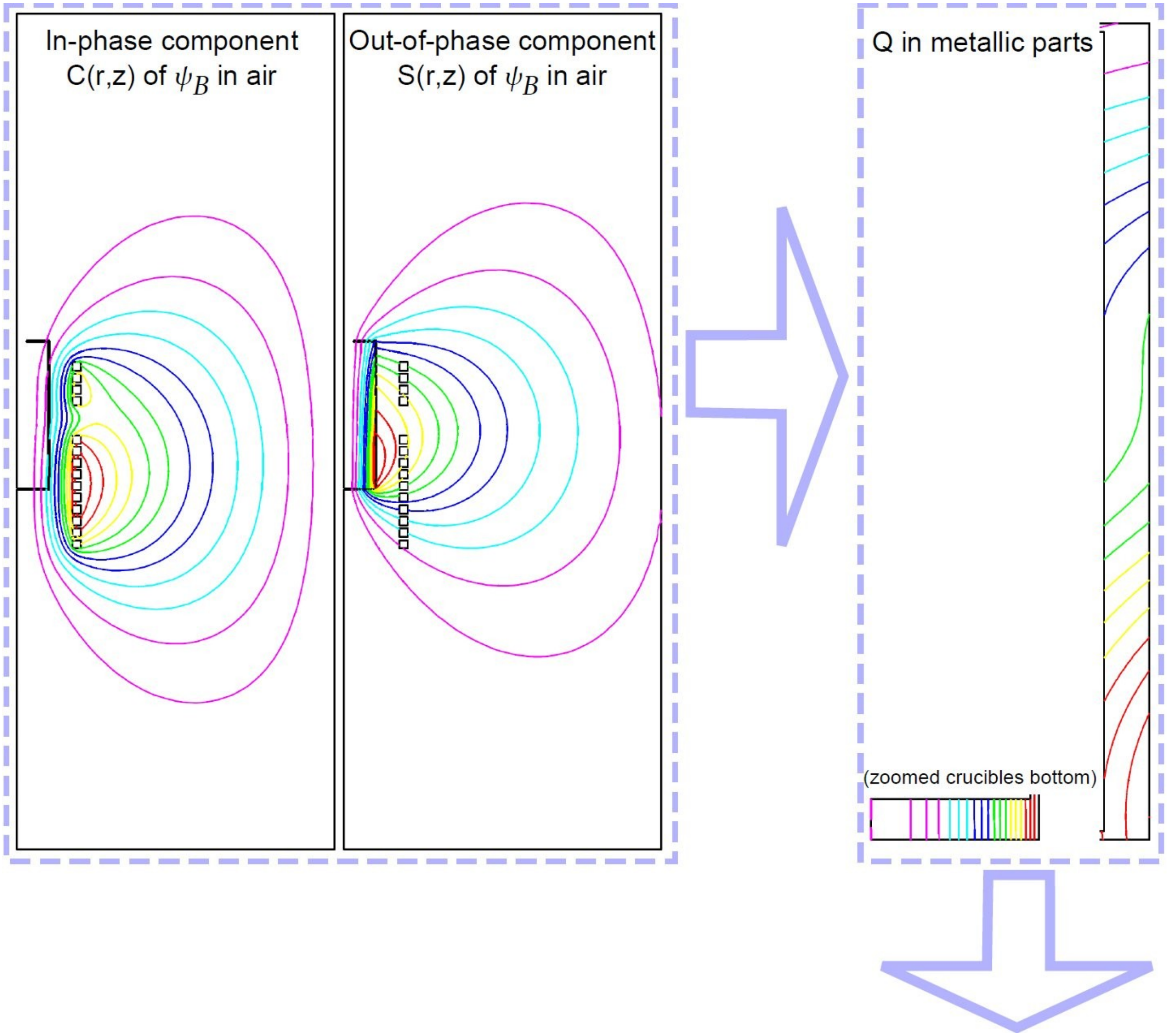
$$Re = \frac{\omega R^2}{\nu}$$

$$Bi = \frac{h_i R}{\lambda_i}$$

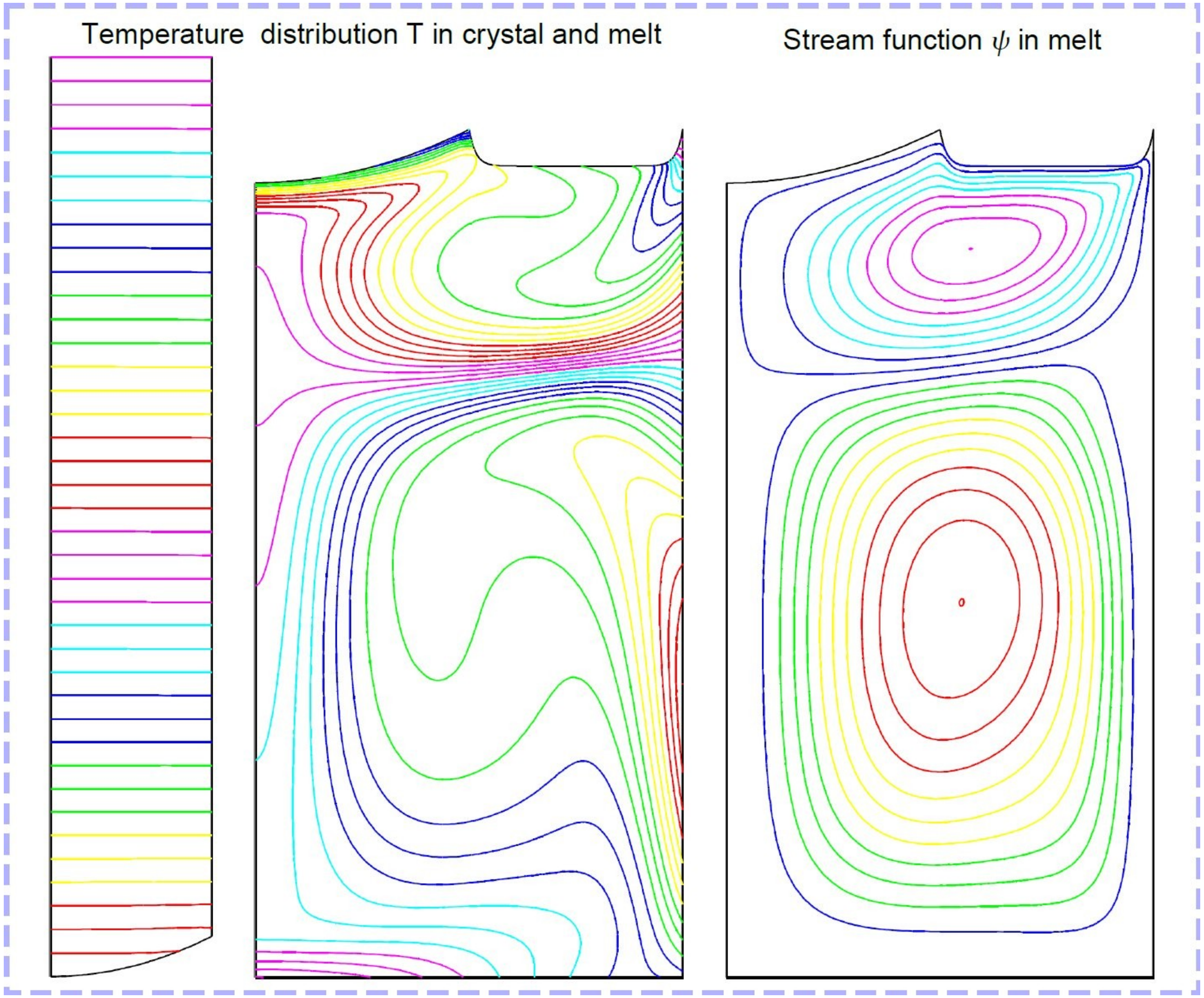
$$Rad_i = \epsilon_i T^3 R/\lambda_i$$

$$Rad_l = \epsilon_l T^3 R/\lambda_l$$

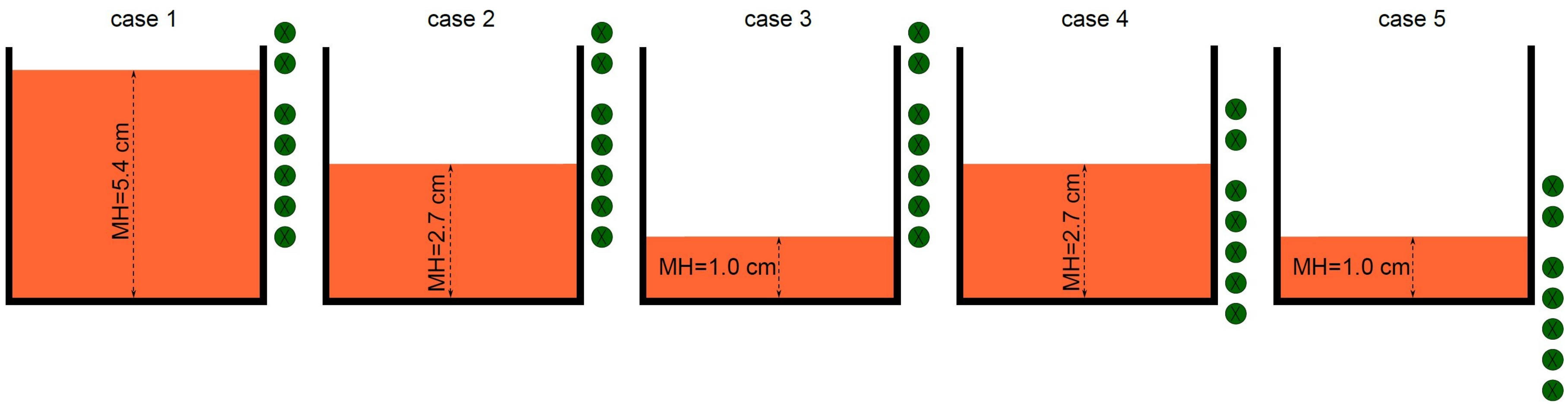
Influence of the RF-heating configuration for different melt heights (1)



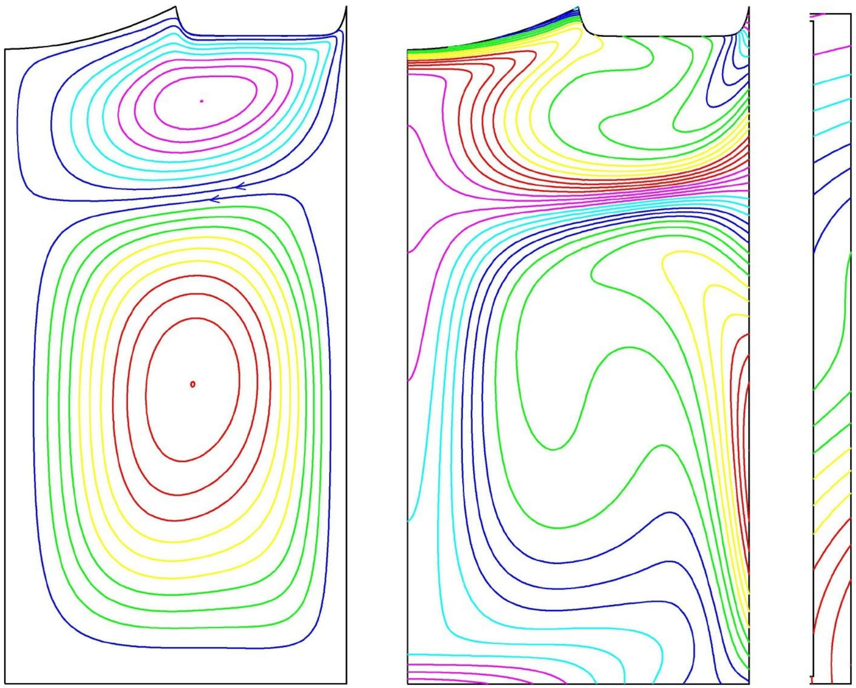
Influence of the RF-heating configuration for different melt heights (2)



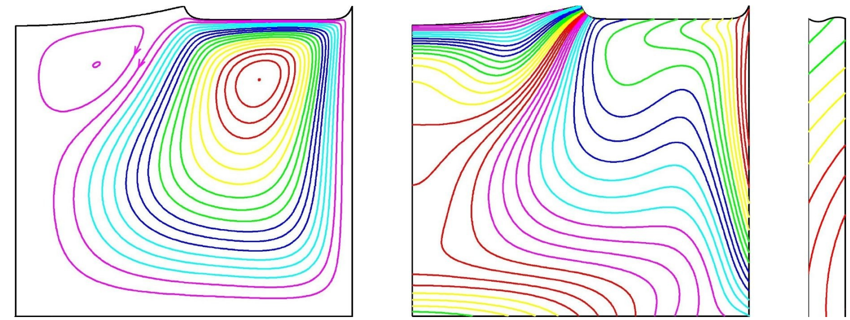
Different configurations of melt height and RF-coils



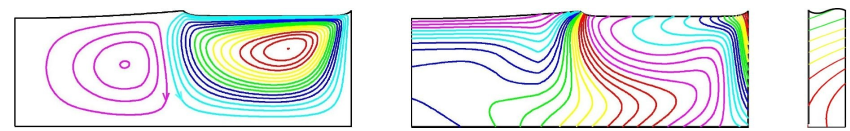
Melt isotherms and streamlines for different cases



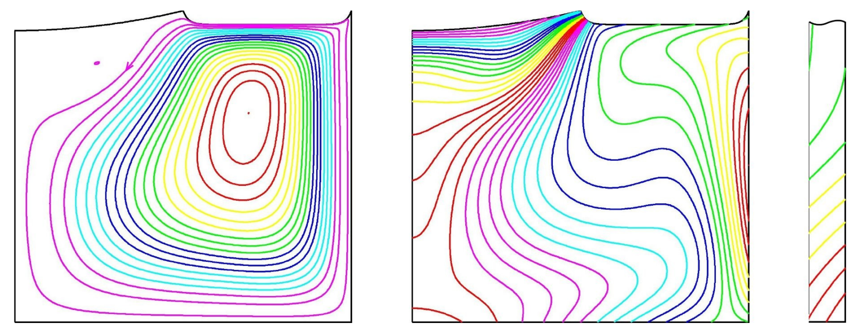
Case 1



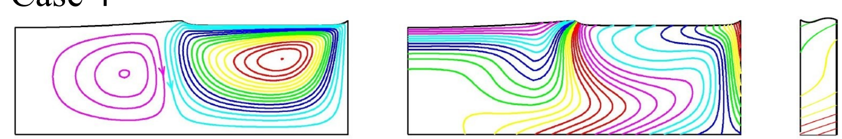
Case 2



Case 3



Case 4



Case 5

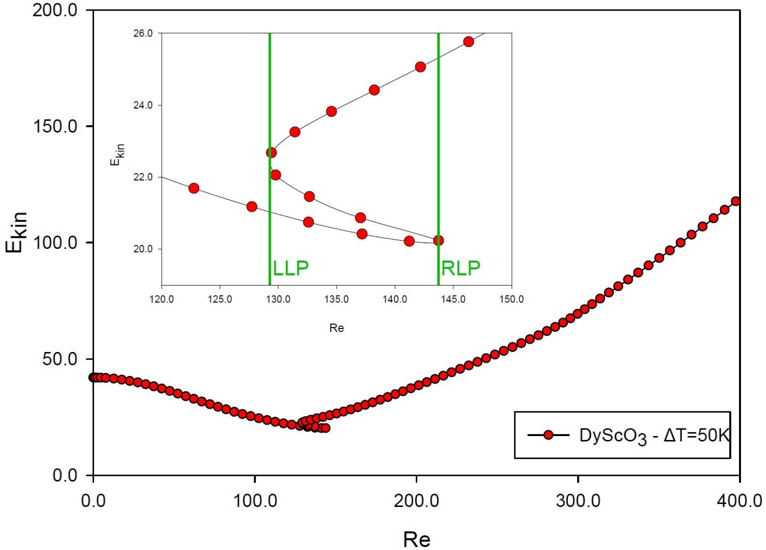
Bifurcation analysis and continuation using $DyScO_3$ melt properties

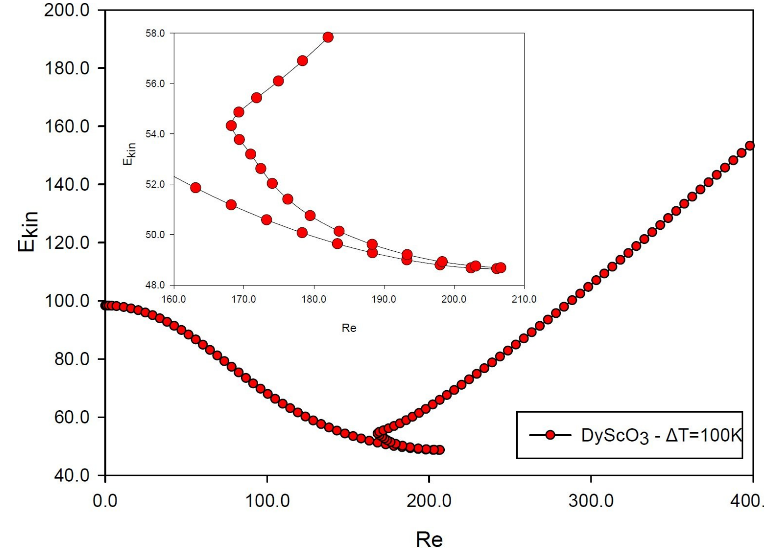
Bifurcation analysis and continuation using $DyScO_3$ melt properties

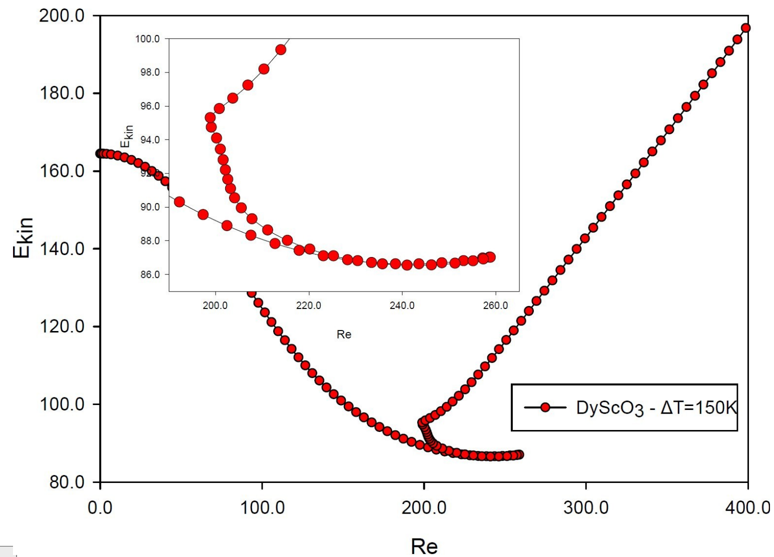
total kinetic energy norm

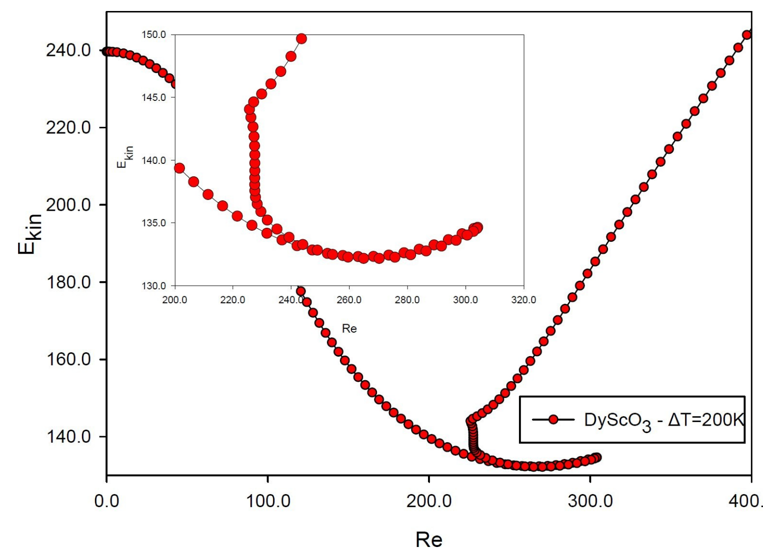
$$E_{kin} = 2\pi \int_0^H \int_0^R \left(u^2 + v^2 + w^2\right) r dr dz$$

Continuation for control parameter Re with fixed ΔT

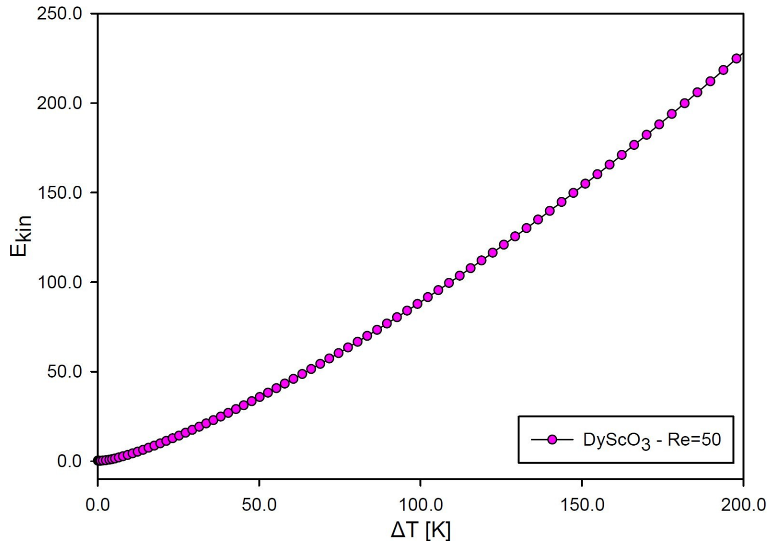


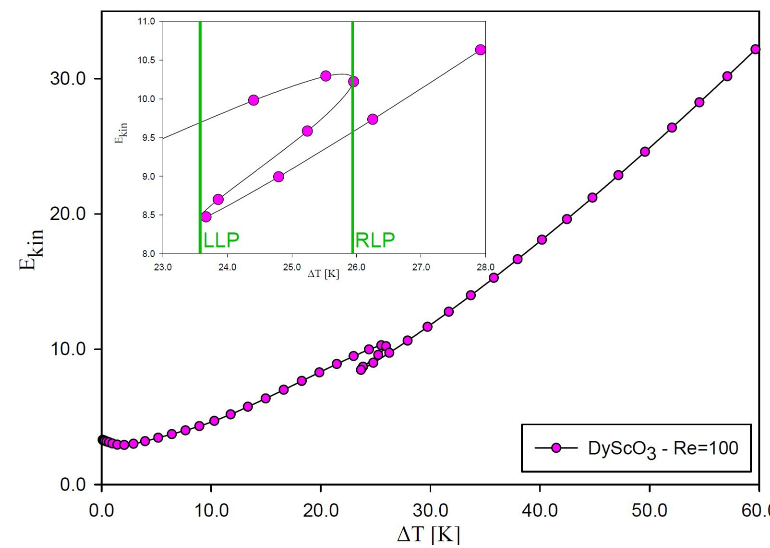


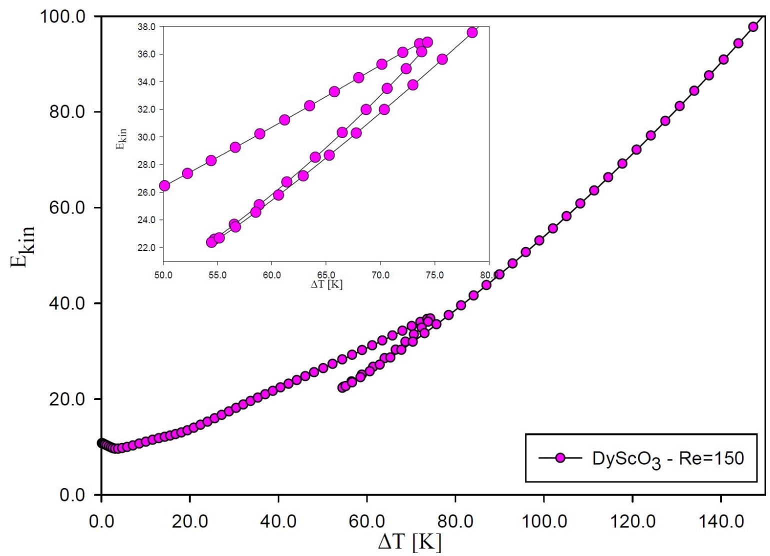


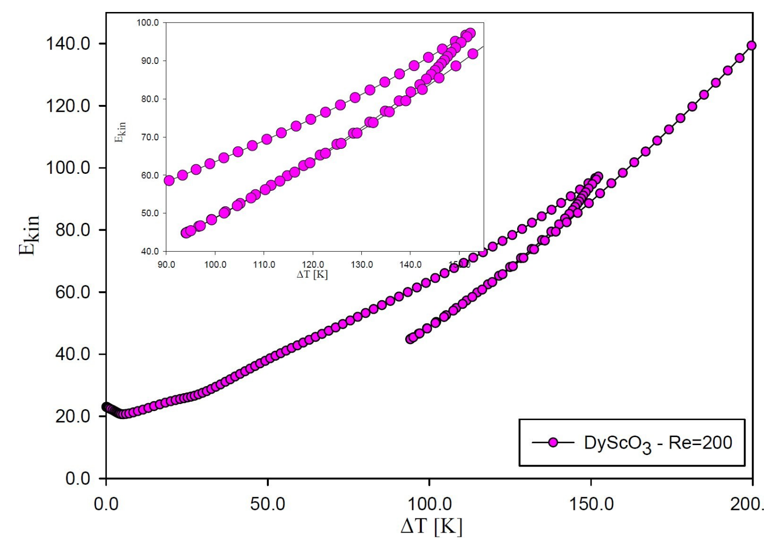


Continuation for control parameter ΔT with fixed Re









Final remarks

- Code cross validation has been done
- There is a complicated fluid flow and heat transfer interaction
- Different types of instability are already possible in 2D
- Measuring important material properties of the $DyScO_3$ melt
- We are working with a model which is close to real crystal growth conditions, but:

→ Taking into account inner and wall-to-wall radiation and latent heat
→ Transition to a full 3D approach, i.e. steady state solution and/or stability analysis

- Multi parameter bifurcation analysis / path following is possible
- Stability analysis for more complex geometry
- It is possible to get knowledge about the complexity of the system and to review
- the influence and interplay of technologicaly adjustable parameters (pull rate, rotation rate, pressure, temperature)

→ Bifurcation and continuation techniques can help to get such information

Thank you very much for your attention.