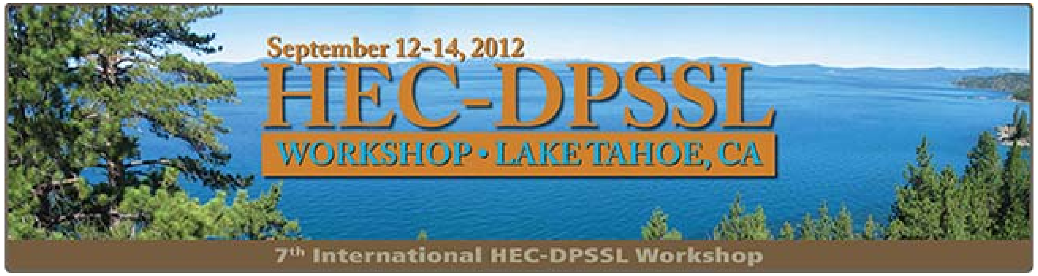

Gradually Doped and Large Diameter Yb^{3+} Doped YAG Crystals for High Power Solid State Laser Applications

M. Arzakantsyan[1,2,3], N. Ananyan[3], V. Gevorgyan[2] and J.-C. Chanteloup[1]

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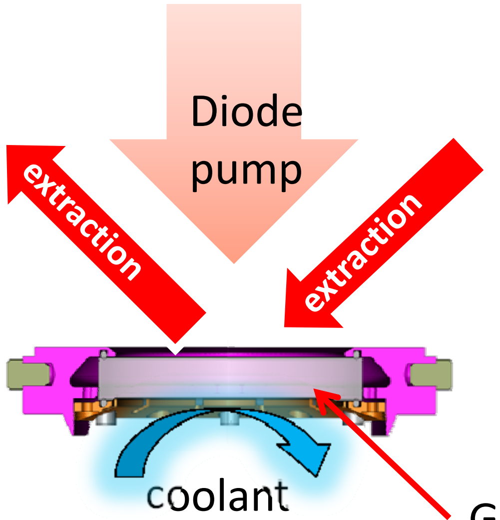
[3]Laserayin Tekhnika CSC, Yerevan, Armenia

LULI research activities in crystal growth

In 2008, LULI initiated research activities on crystal growth in collaboration with Laserayin Tekhnika, Yerevan, Armenia.
The aim is to engineer laser gain media tailored to face the requirement imposed by developpement of **HEC DPSSL** laser systems through the world:

- **Thermal** and Amplified Spontaneous Emission (**ASE**) Management
- **Larger** size gain medium

Developing YAG crystals with variable Yb ion concentration appears as a solution of choice to homogenize the stored energy distribution within the gain medium.
This is especially true in the context of Lucia amplifiers active mirror architecture



Pump and input extraction beams are reflected at the back surface of the disk

Gain medium

Outline

- Impact of gradient doped gain medium on thermal and ASE management
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- Experimental campaigns aiming at improving our model
- Large Yb:YAG crystals

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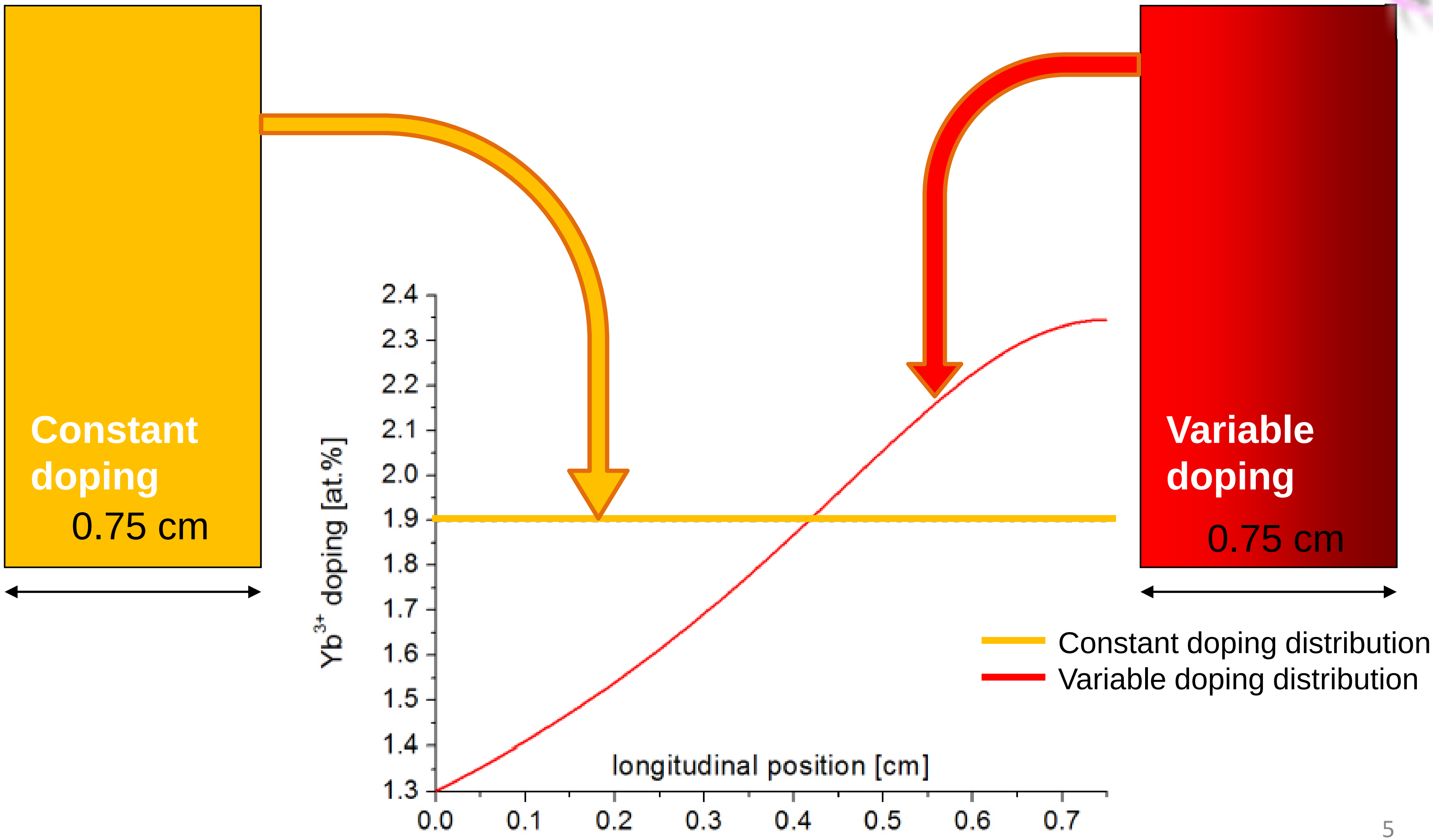
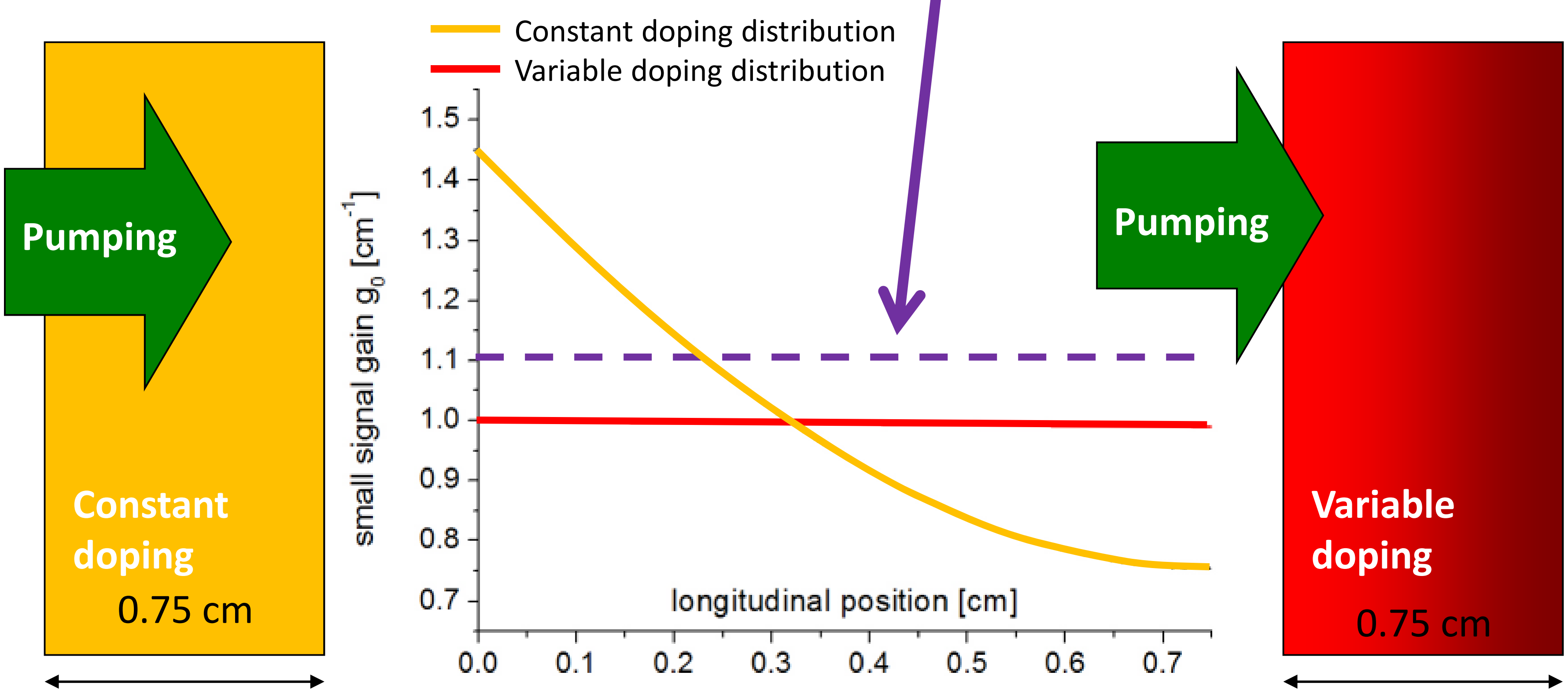
Gain medium engineering



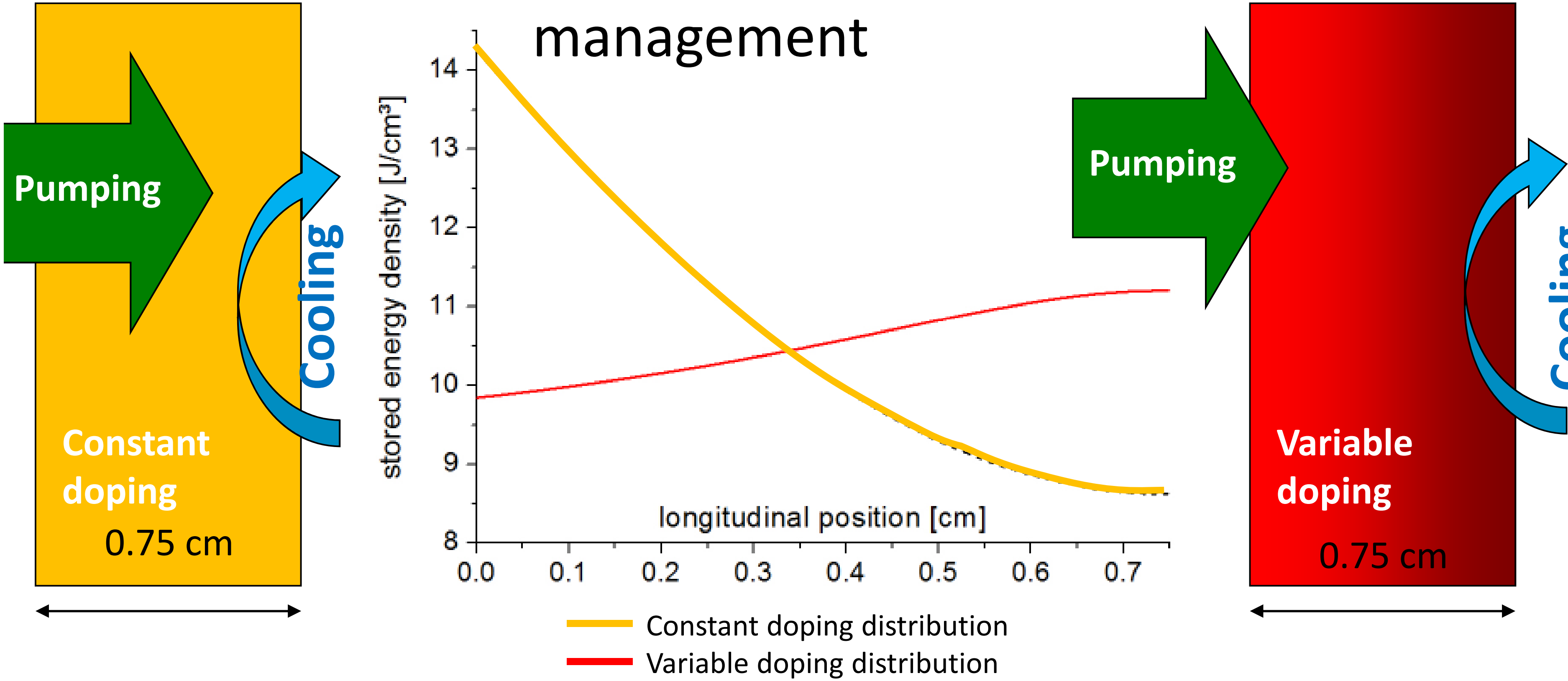
ASE & thermal managt improvt w/ gain medium engineering

Gradient doping sets gain below ASE oscillation threshold ($g_{0max}L<4$)



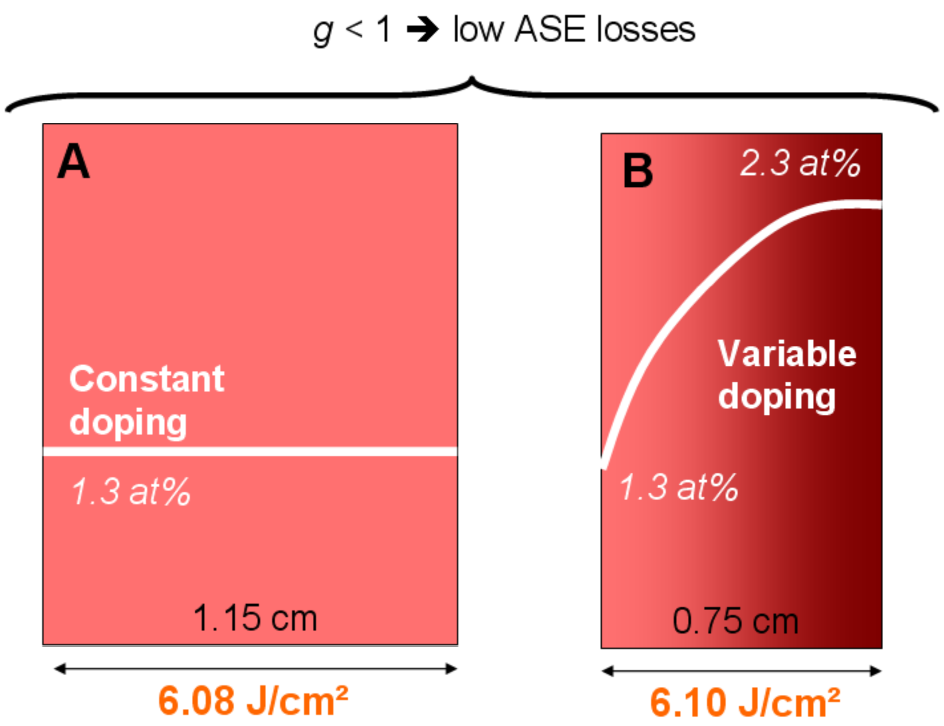
Gradient doping impact on stored energy distribution

…and therefore on thermal management



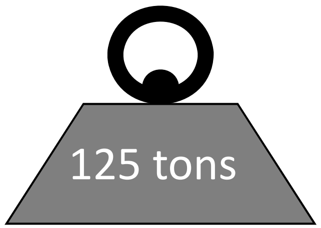
Impact on gain medium volume requirement

6 J/cm² stored energy in all cases



Such a 35% decrease on requested gain medium volume can have a dramatic impact on very large scale facility design trade….

What is the total weight of the 4320 homogeneously doped glass slabs required on LMJ facility ?...



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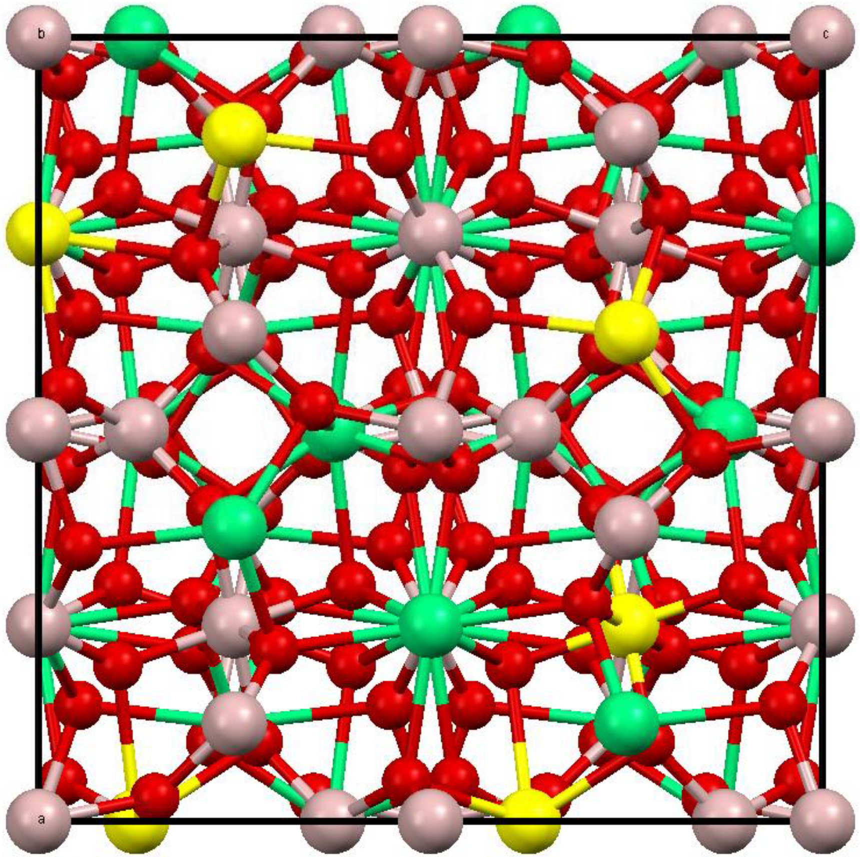
YAG structure

Yttrium Aluminum Garnet has a cubic unit cell (yellow Yb ions replace some grey Y)

Yb Y Al O



[100]
Cristallographic axis

YAG phase diagram

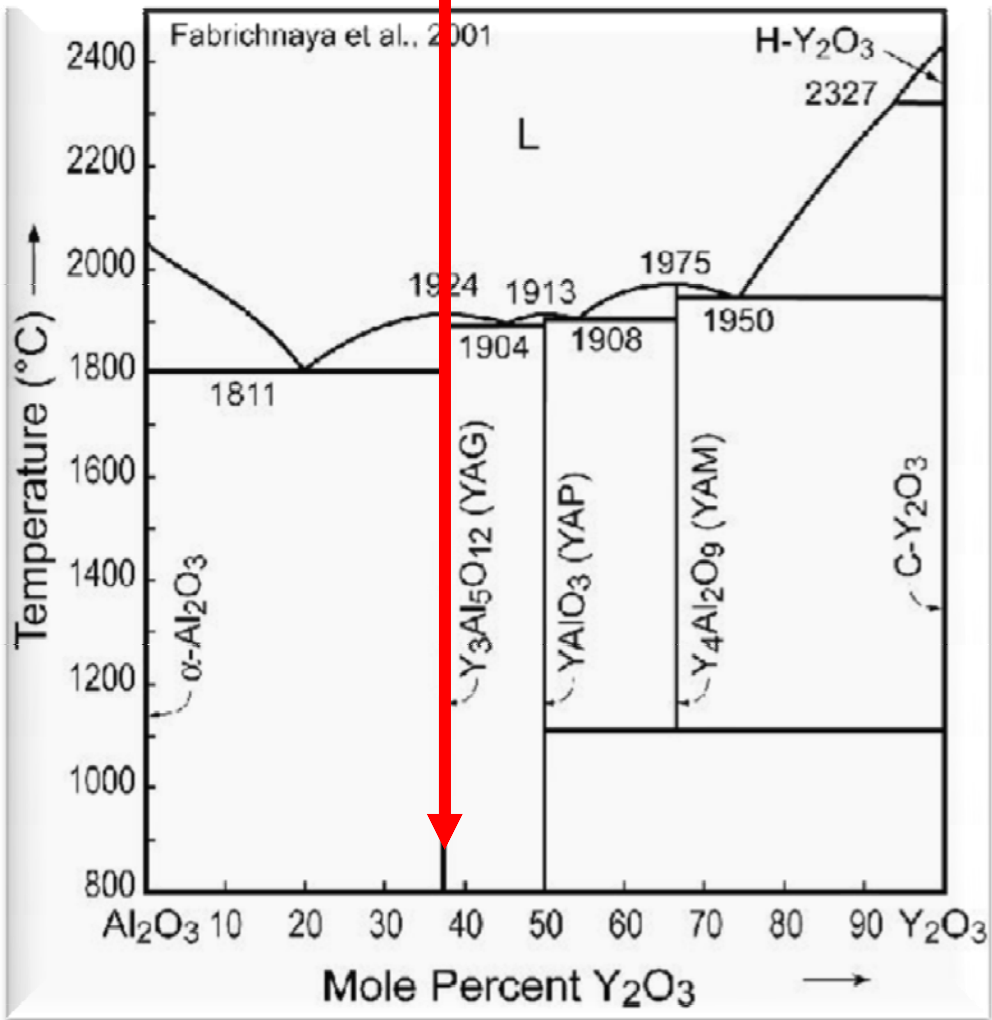
YAG is obtained by stoichiometric mixing of 3 Yttria (Y_2O_3) and 5 Alumina (Al_2O_3) : 3/(5+3) = 37.5%

5 Al_2O_3

+

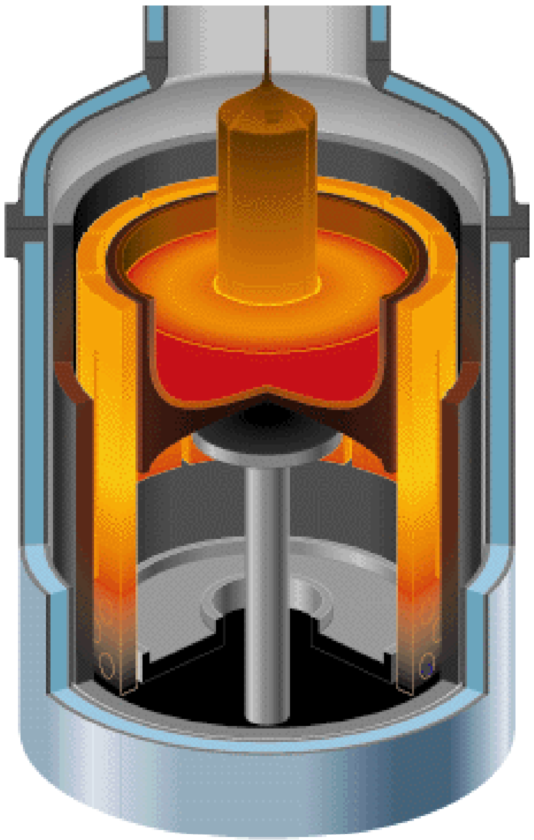
3 Y_2O_3

2 $Y_3Al_5O_{12}$



YAG growth techniques: Czochralski

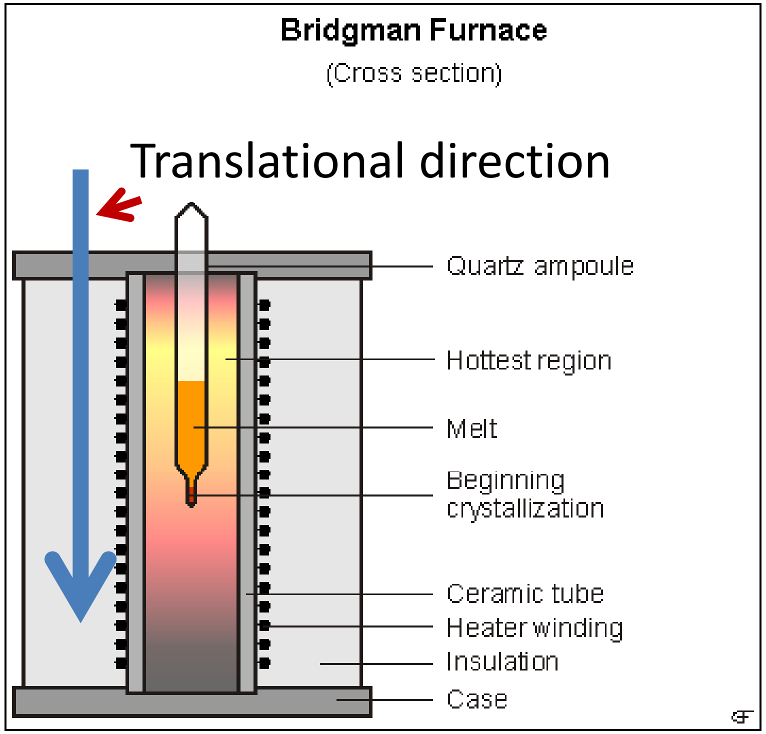
The Czochralski technique is traditionally used for YAG growth.
It is a well known technique where the grown crystal does not experience contact with the crucible. Drawbacks are :



- Cores and light scattering particles
- Large weight loss of iridium crucible
- Instability of solid-melt interface

YAG growth techniques: Bridgman

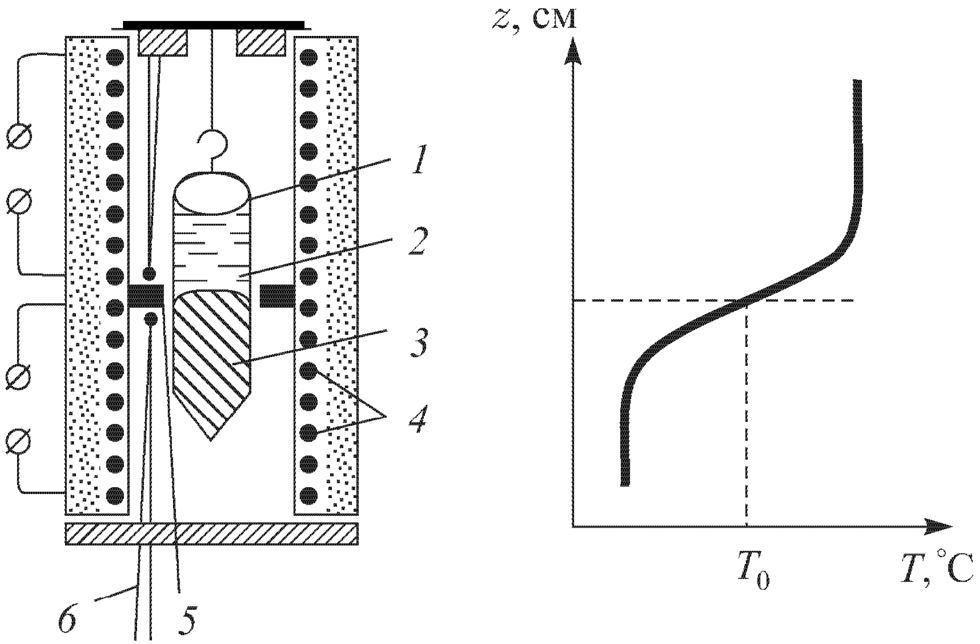
Starting material is loaded in a vertically moving crucible



- No possibility of in-situ observation
- Mechanical interaction between the crucible and the crystal
- Technical issues for extracting the grown crystal

YAG growth techniques: TGT

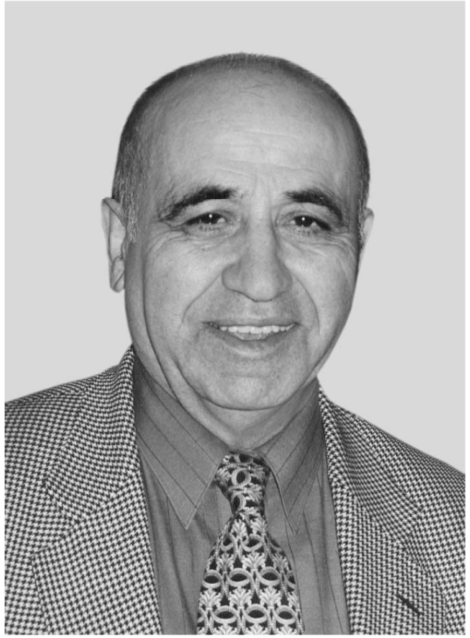
Starting material is loaded in a **static** crucible (ampoule shaped)
Heat distribution is vertically moving upwards



1 - crucible
2 - melt
3 - crystal
4 - heater spires
5 - thermal pinhole
6 - thermocouples

Drawbacks are the same as for Vertical Bridgman Technique

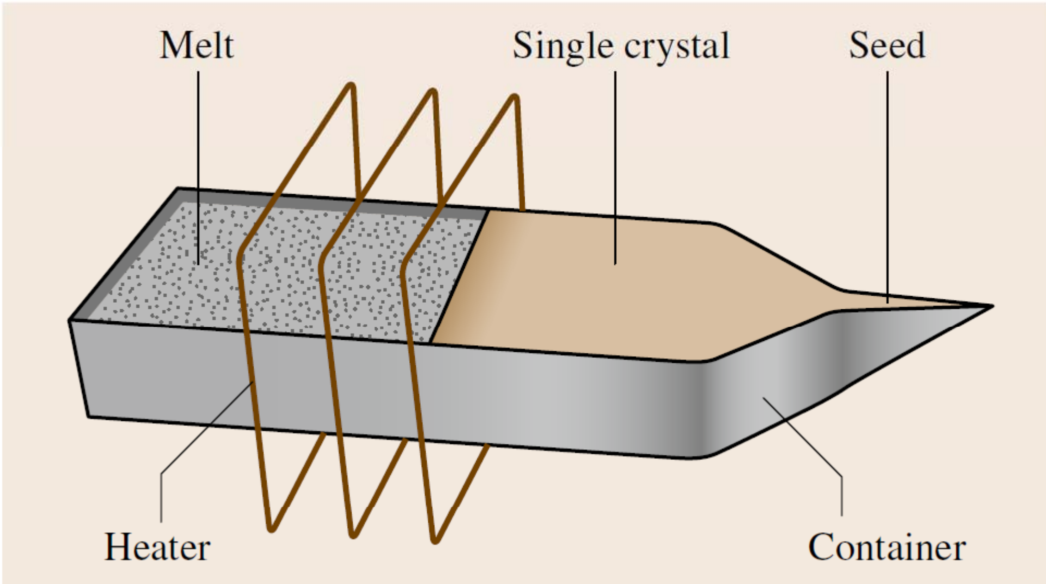
Bagdasarov technique



Kh. S. Bagdasarov

Bagdasarov technique (1964) is also known as:
Horizontal Bridgman Technique (HBT),
Horizontal Direct Crystallization (HDC),
Boat method in reference to the crucible shape,
Horizontal Direct Solidification (HDS)

The method is mainly used to produce high quality large size sapphire. It is also very effective to grow YAG crystals enriched with different dopant ions (Yb, Nd, Eu, Er, Ce…).



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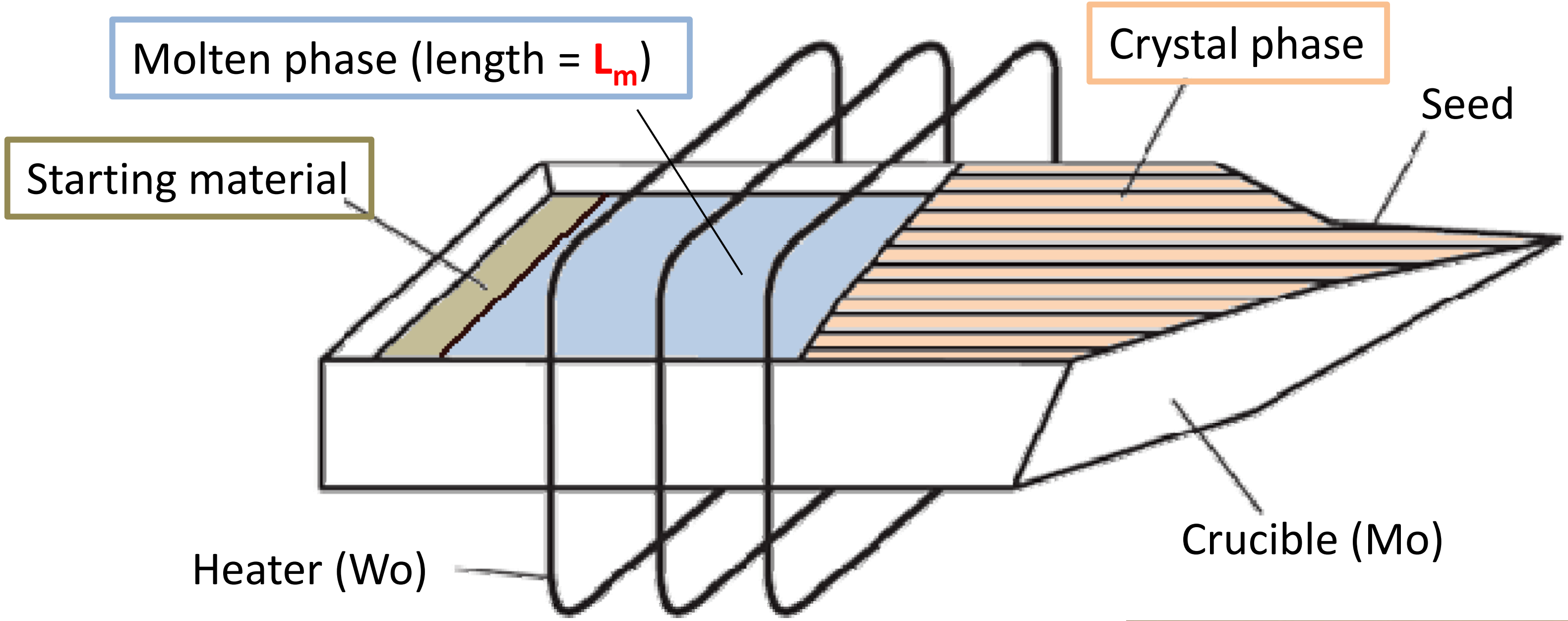
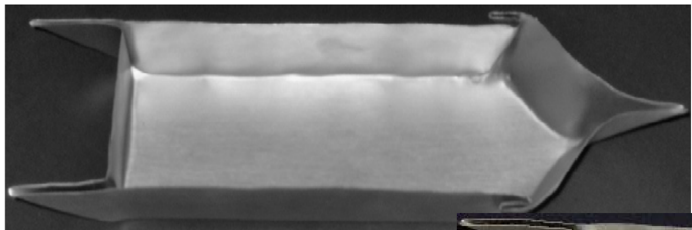
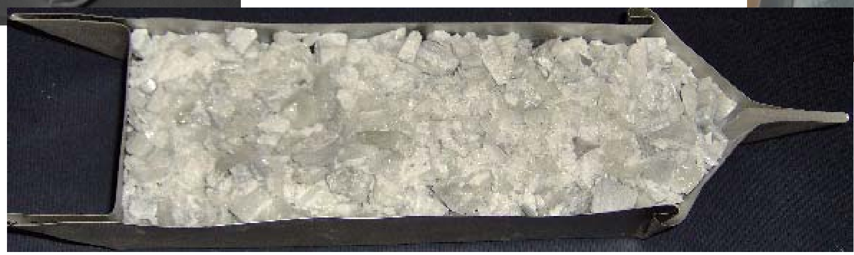
Bagdasarov technique

Starting material is loaded in a horizontally moving crucible





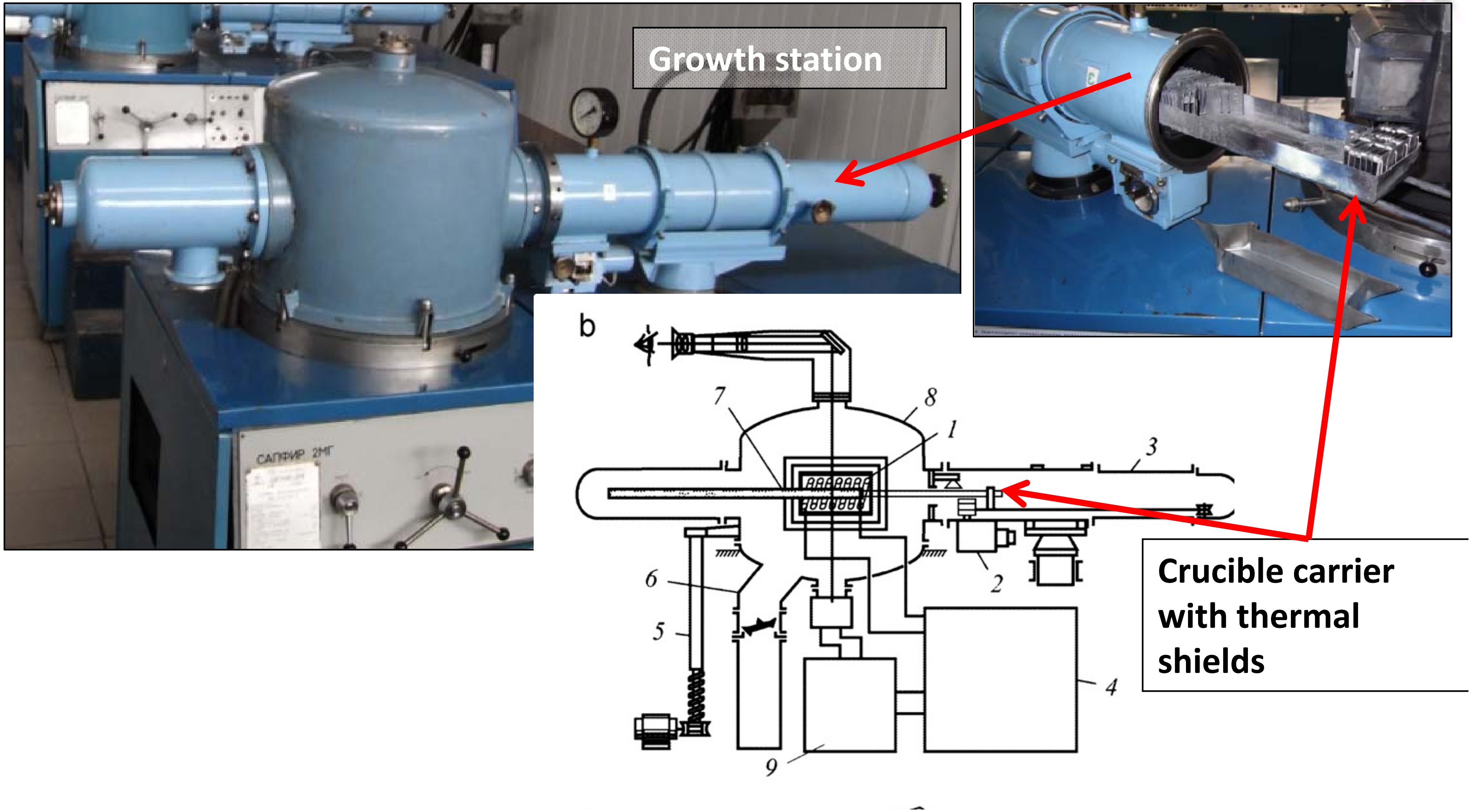
Empty crucible



Starting material loaded

Final boule

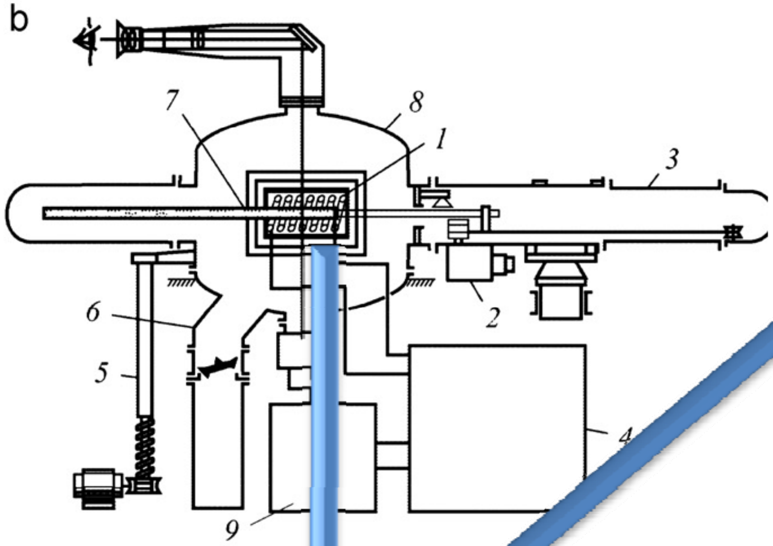
Bagdasarov furnace



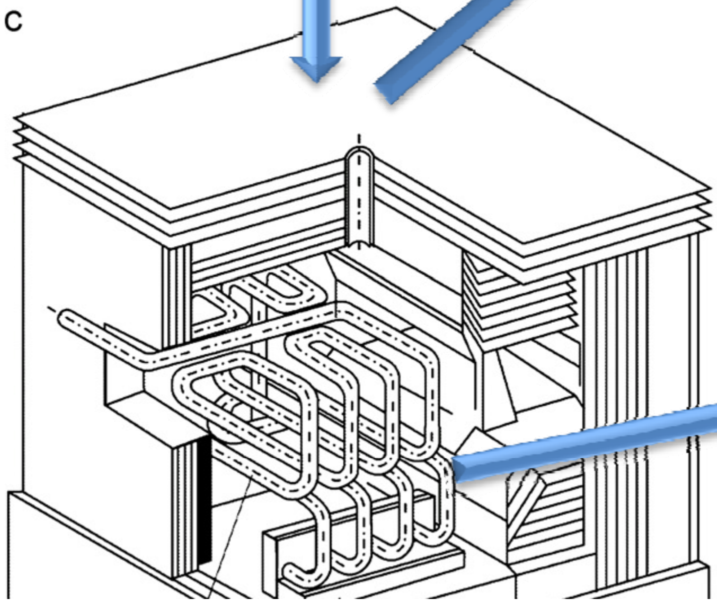
Bagdasarov furnace heater

1930°C temperature obtained with 22 Volts (1kA)

b



c



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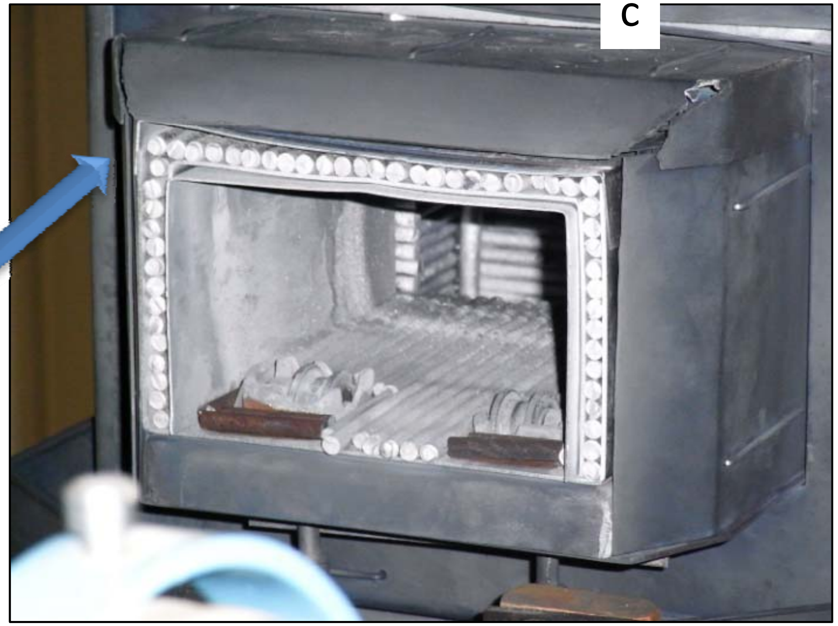

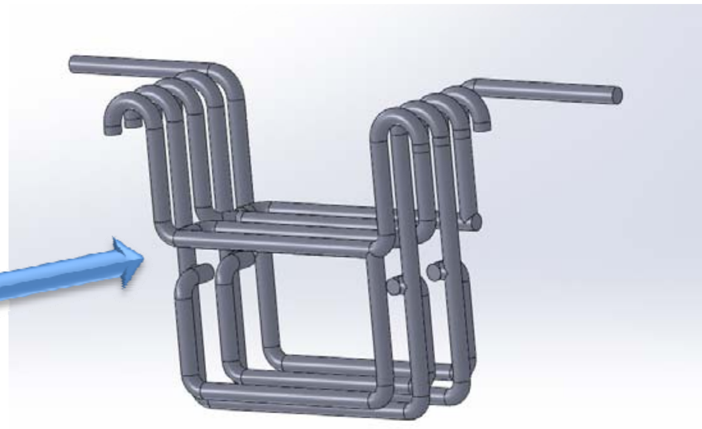

5 Tungsten spires heater

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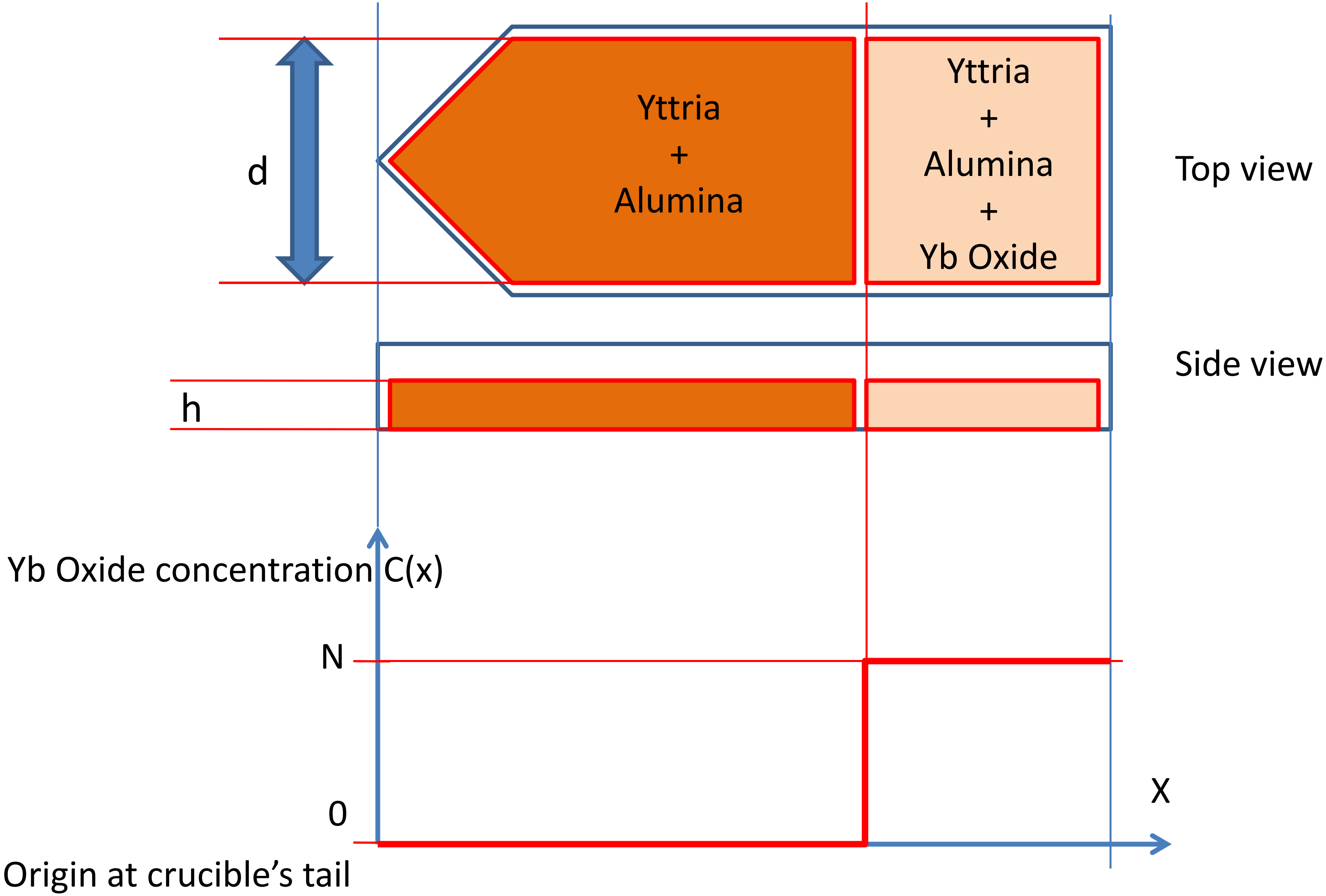
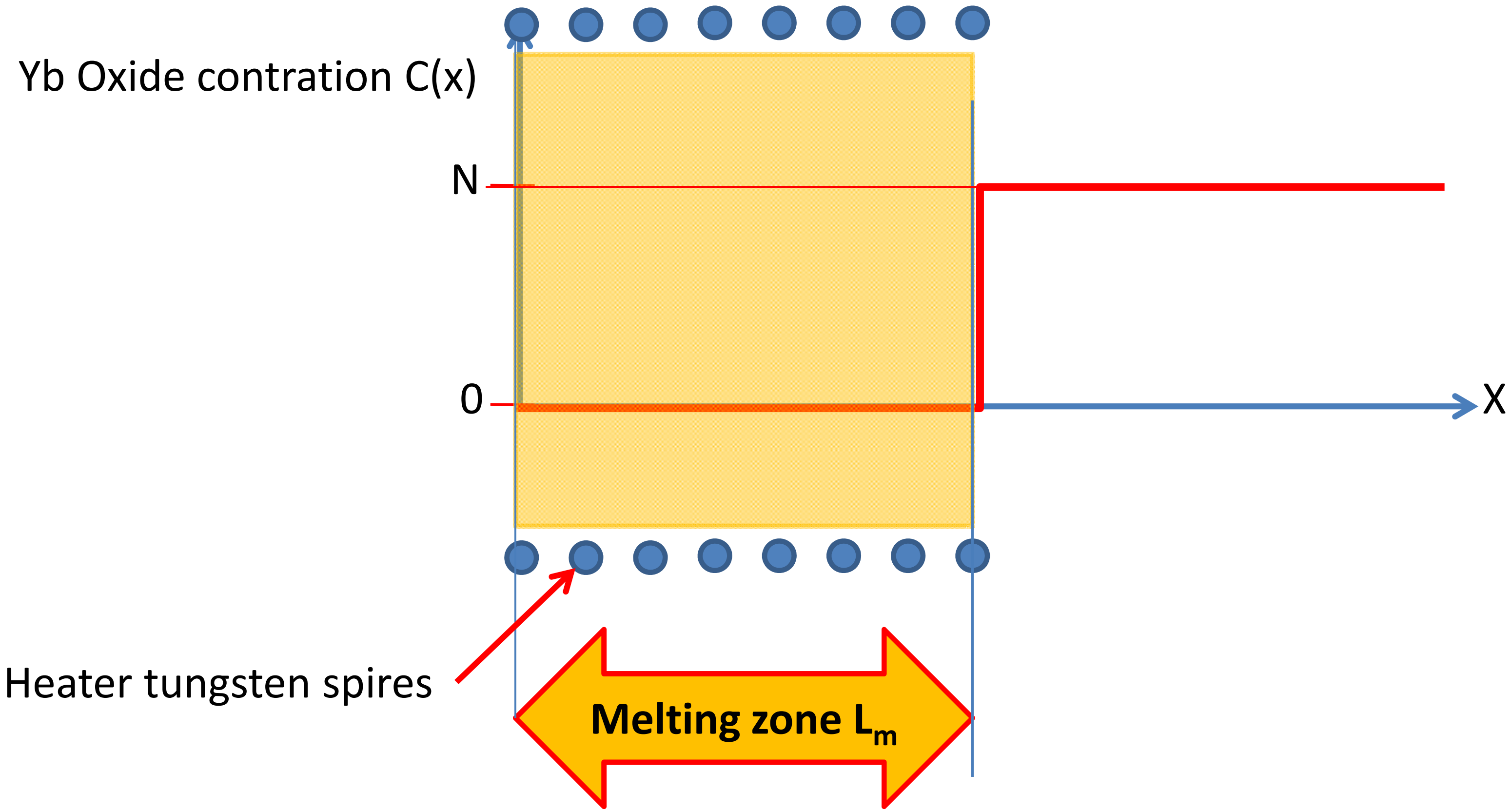
Simple model for doping ion distribution

Initial starting material distribution



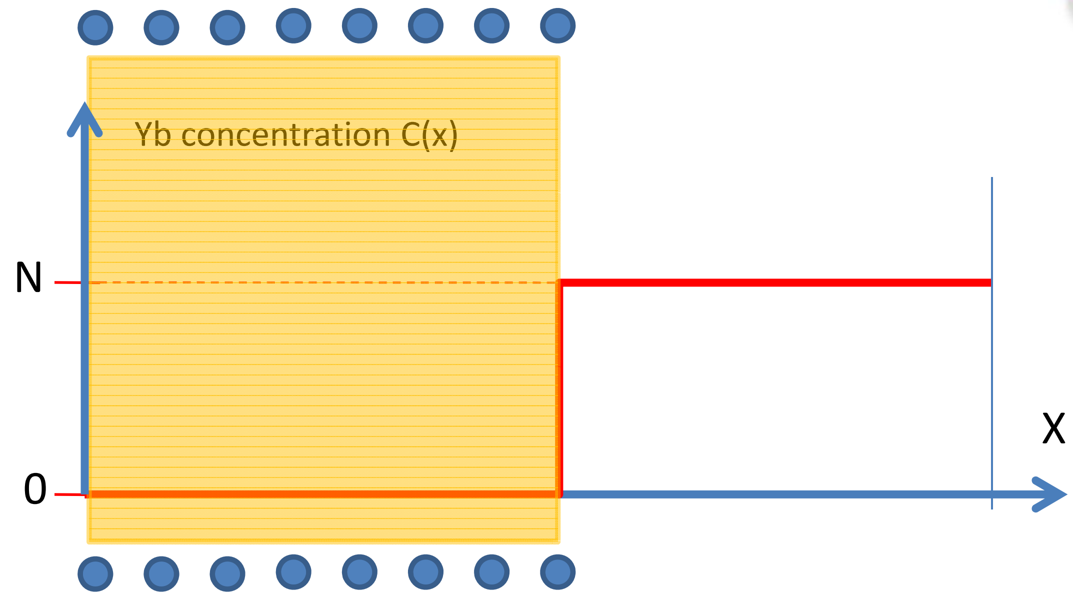
Simple model for doping ion distribution

Relative motion between crucible and heat zone

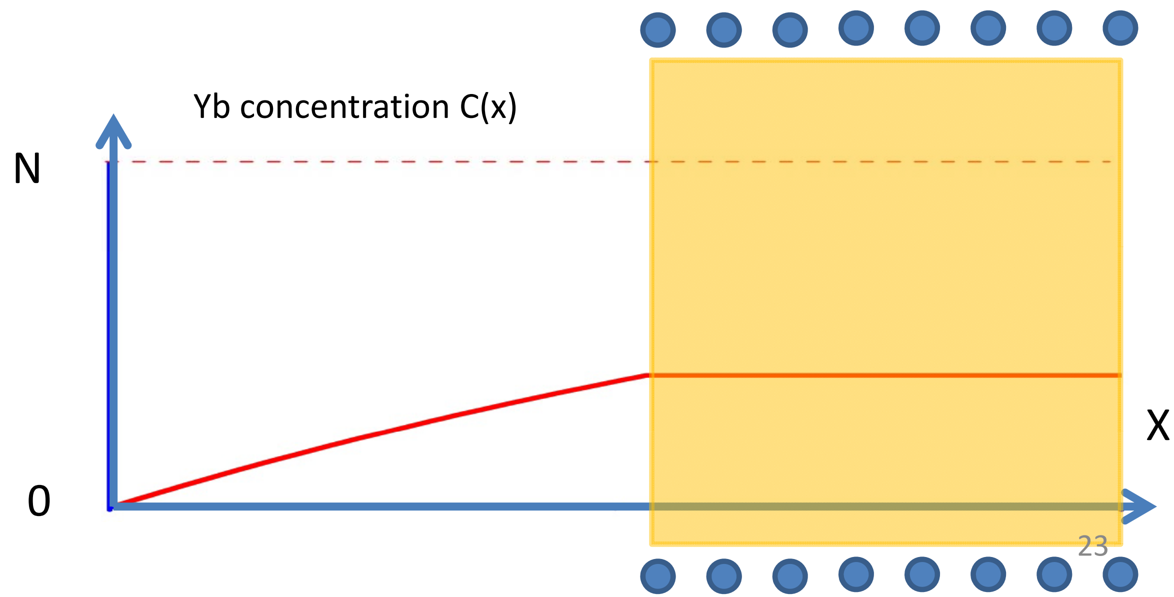


Simple model for doping ion distribution

Yb starting (t=0) distribution in the crucible referential with origin at seed



Yb final distribution



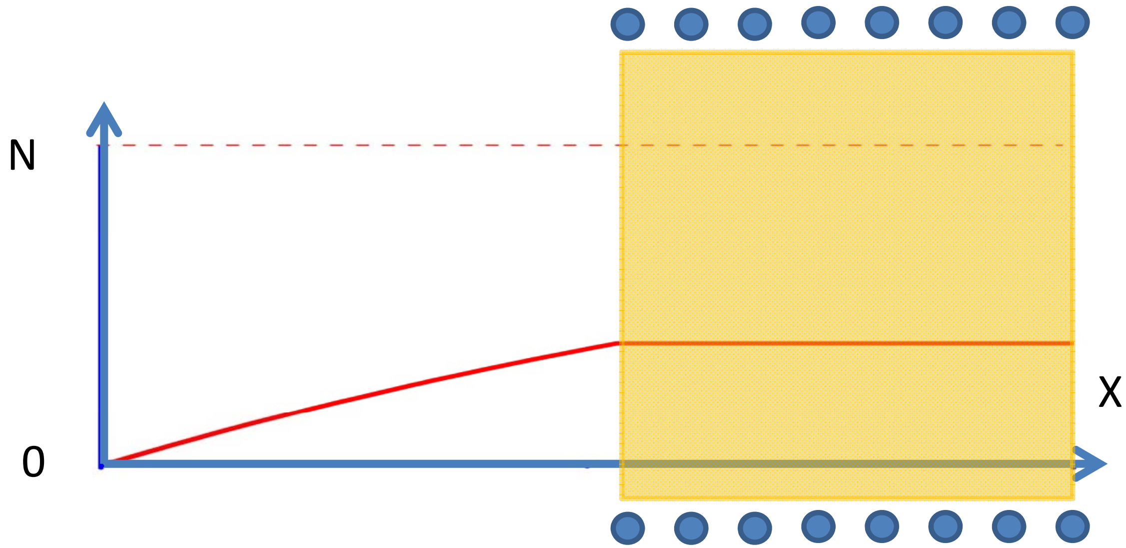
Simple model for doping ion distribution

Hypothesis

1. Segregation coefficient is considered equal to unity
2. Instantaneous Yb ions diffusion in the melt
3. Height h considered as constant over the process
4. L_m constant

Differential distribution Concentration: $$C(x+dx) = \frac{C(x)L_m - C(x)dx + Ndx}{L_m}$$

Concentration distribution: $$C(x) = N(1 - e^{-\frac{x}{L_m}})$$



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Yb concentration measurements : EPMA & ICP-MS

Electron Probe Micro Analysis (EPMA)

EPMA provides quantitative and qualitative measurements of chemical composition at the micrometer scale in solid samples without destruction

CAMECA SX 100 EPMA at the Laboratoire Magmas et Volcans

Inductively Coupled Plasma-Mass Spectrometry (ICP-MS)

1-Yb:YAG sample dissolved into liquid solution
2-solution is ionized with Inductively* Coupled Plasma (ICP)
3-Separation of the ions is performed with a Mass Spectrometer (MS)

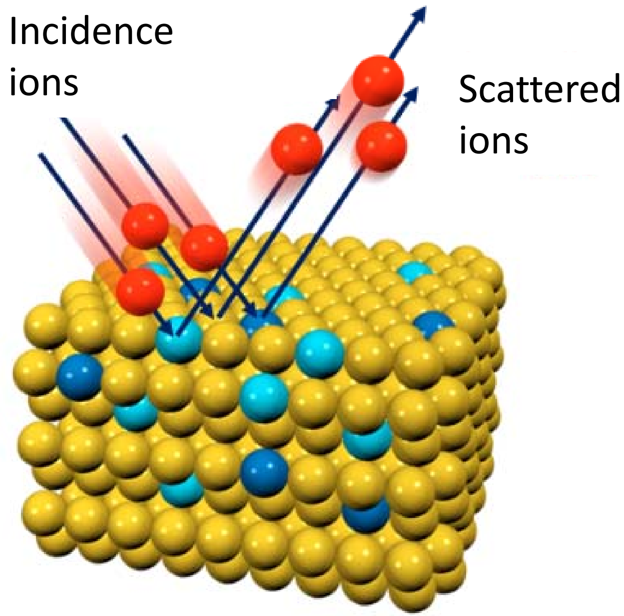
LSCE Argon ICPMS Spectrometer

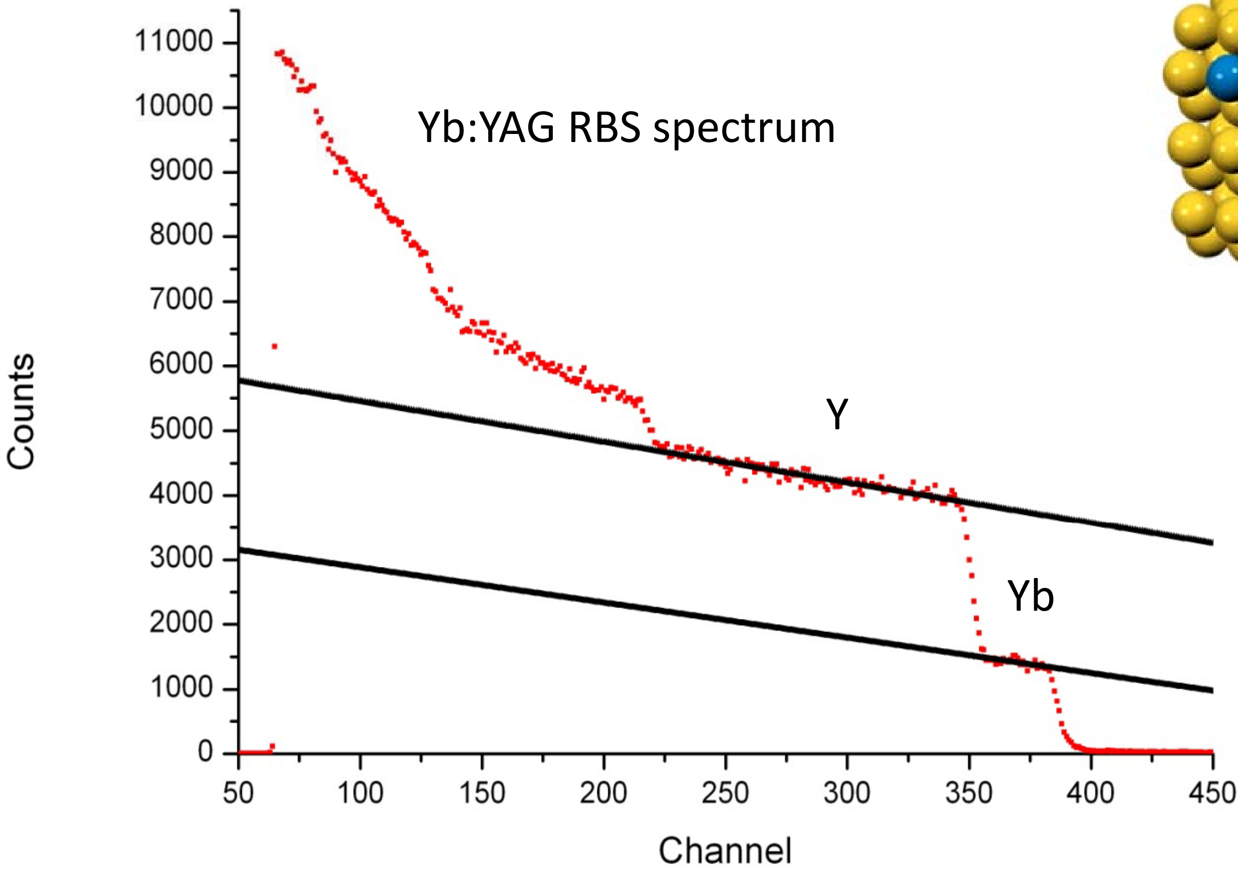
* An inductively coupled plasma (ICP) is a type of plasma source in which the energy is supplied by electric currents which are produced by electromagnetic induction, that is, by time-varying magnetic fields.

Yb concentration measurements : RBS

Rutherford Back Scatering (RBS)

RBS determines the composition of Yb:YAG samples by measuring the backscattering of a beam of high energy ions (here X^+) impinging the sample.





Van de Graaff Ion accelerator at the *Institut des Nanosciences de Paris* (INP)

EPMA, ICP-MS and RBS comparison

	Doping growth reference	EPMA	ICP-MS	RBS
Sample A [at%]	2	2.01+/-0.1	1.9 +/-0.04	2+/-0.1

Destructive and underestimating

Large facility with access time constraints

EPMA appears to be very accurate and easiest in use technique and has been choosen as a main technique

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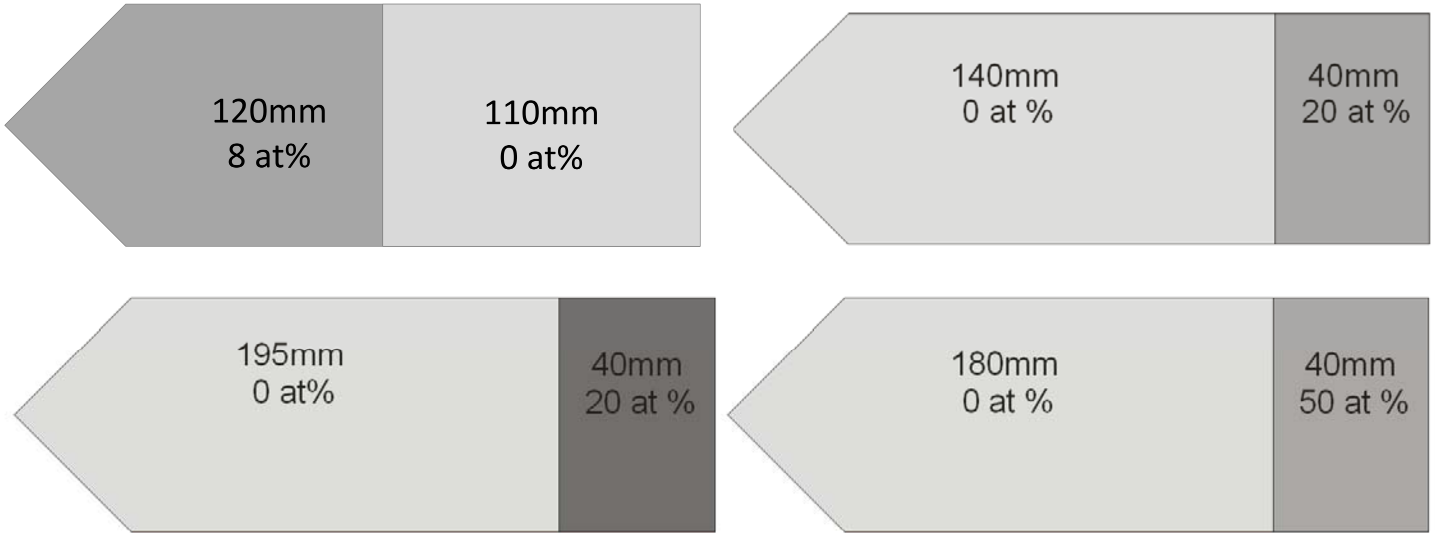
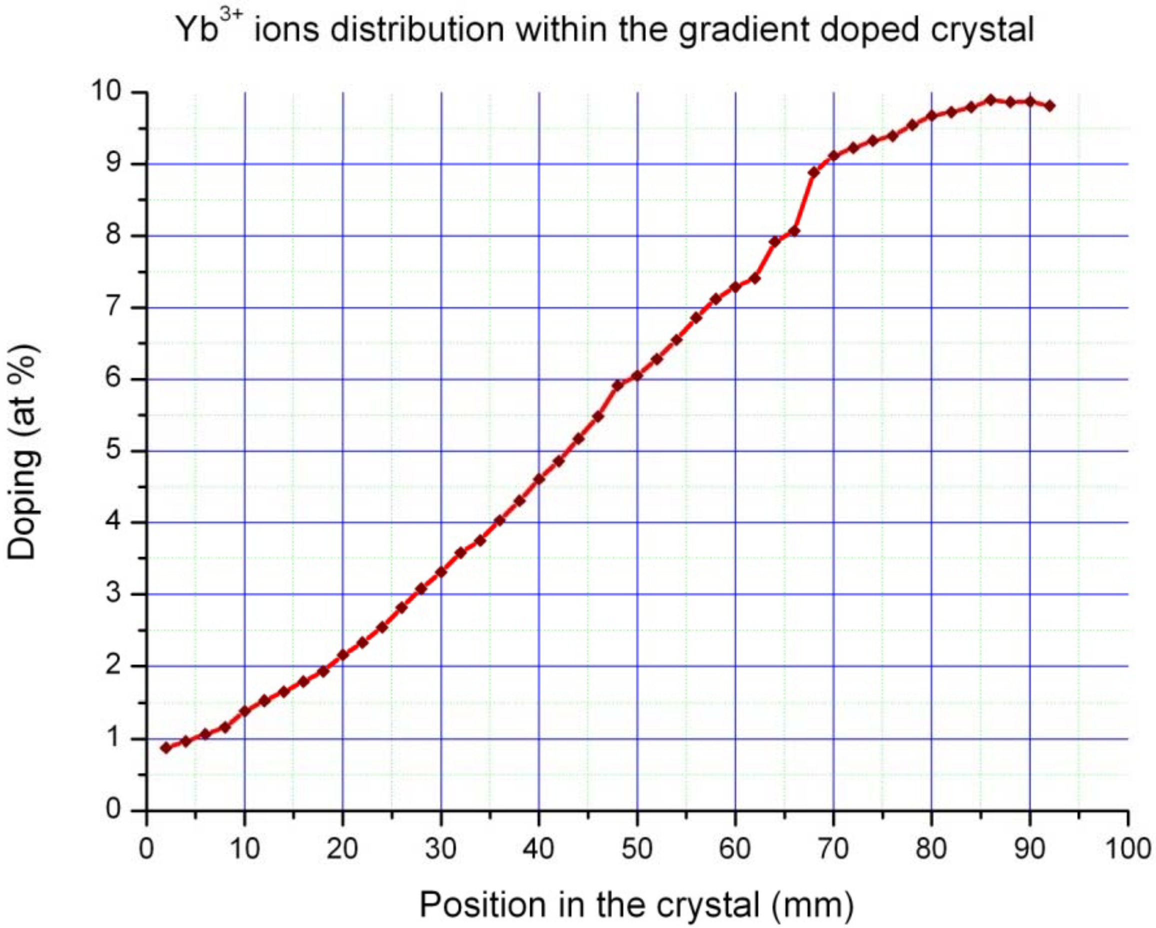
Four growth sequences performed



Different shaped crystals are extracted, polished,… for further testing

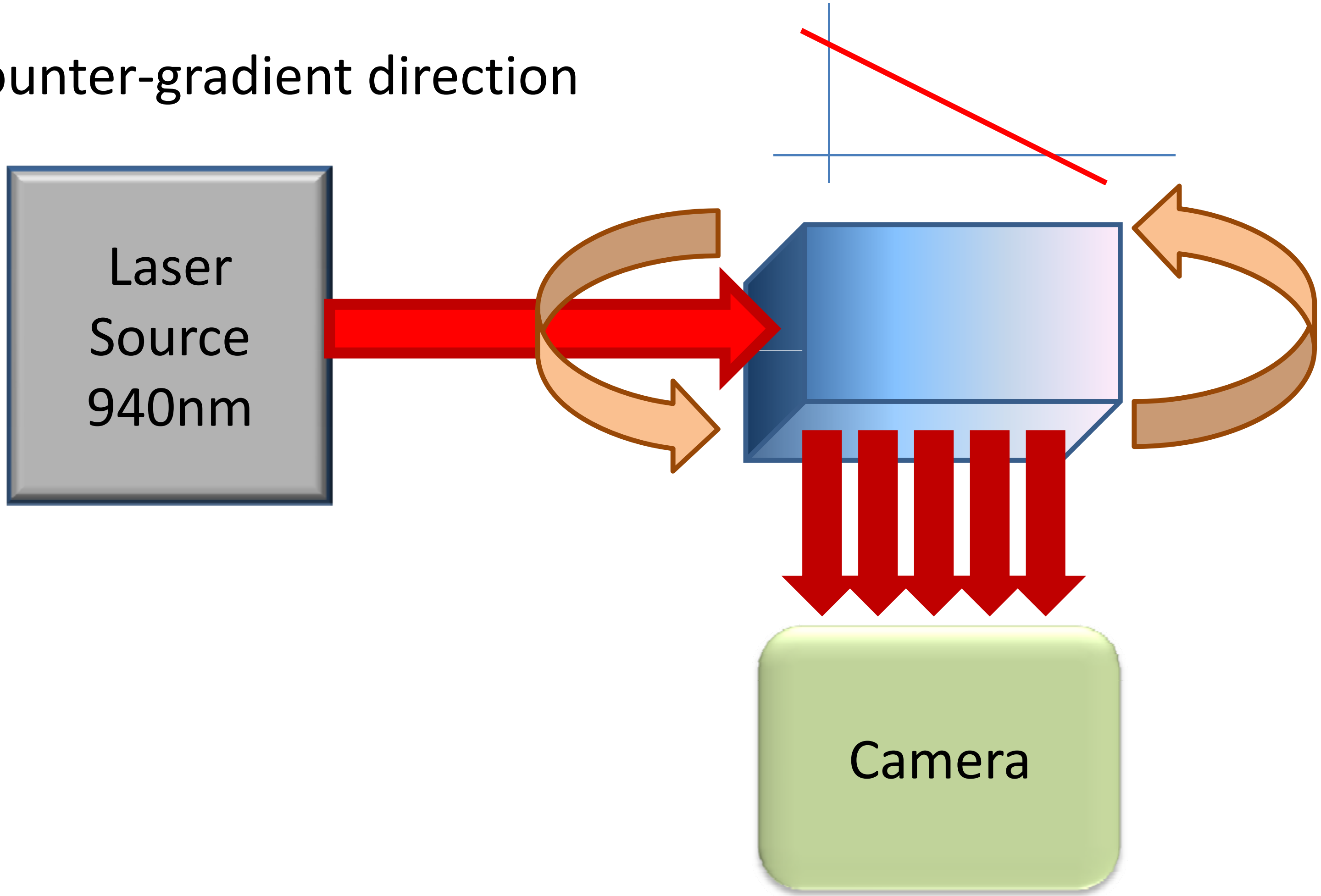
Yb experimental concentration distribution

Yb concentration gradients up to ~3at%/cm are obtained



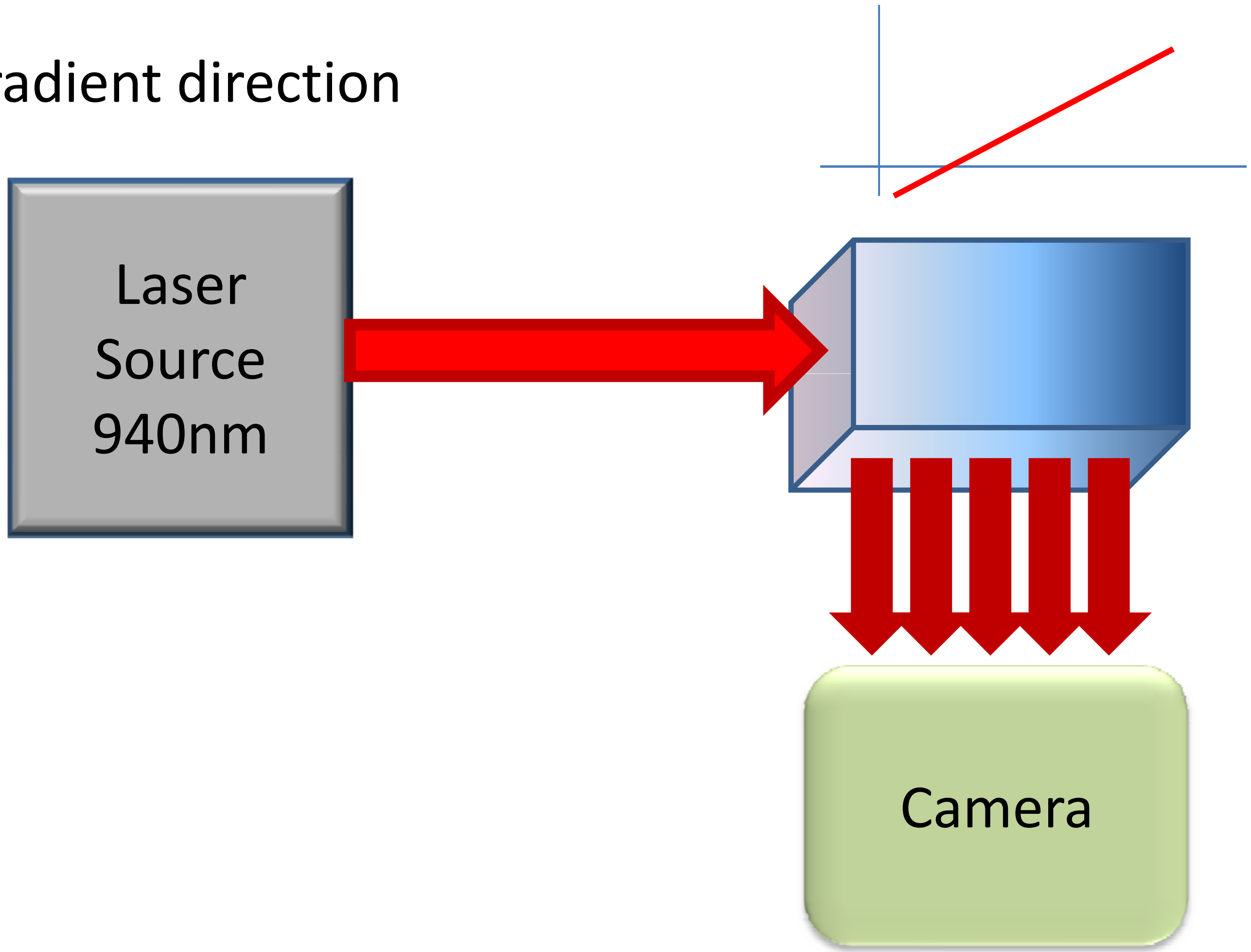
Fluorescence at 1030nm

Counter-gradient direction



Fluorescence at 1030nm

Gradient direction



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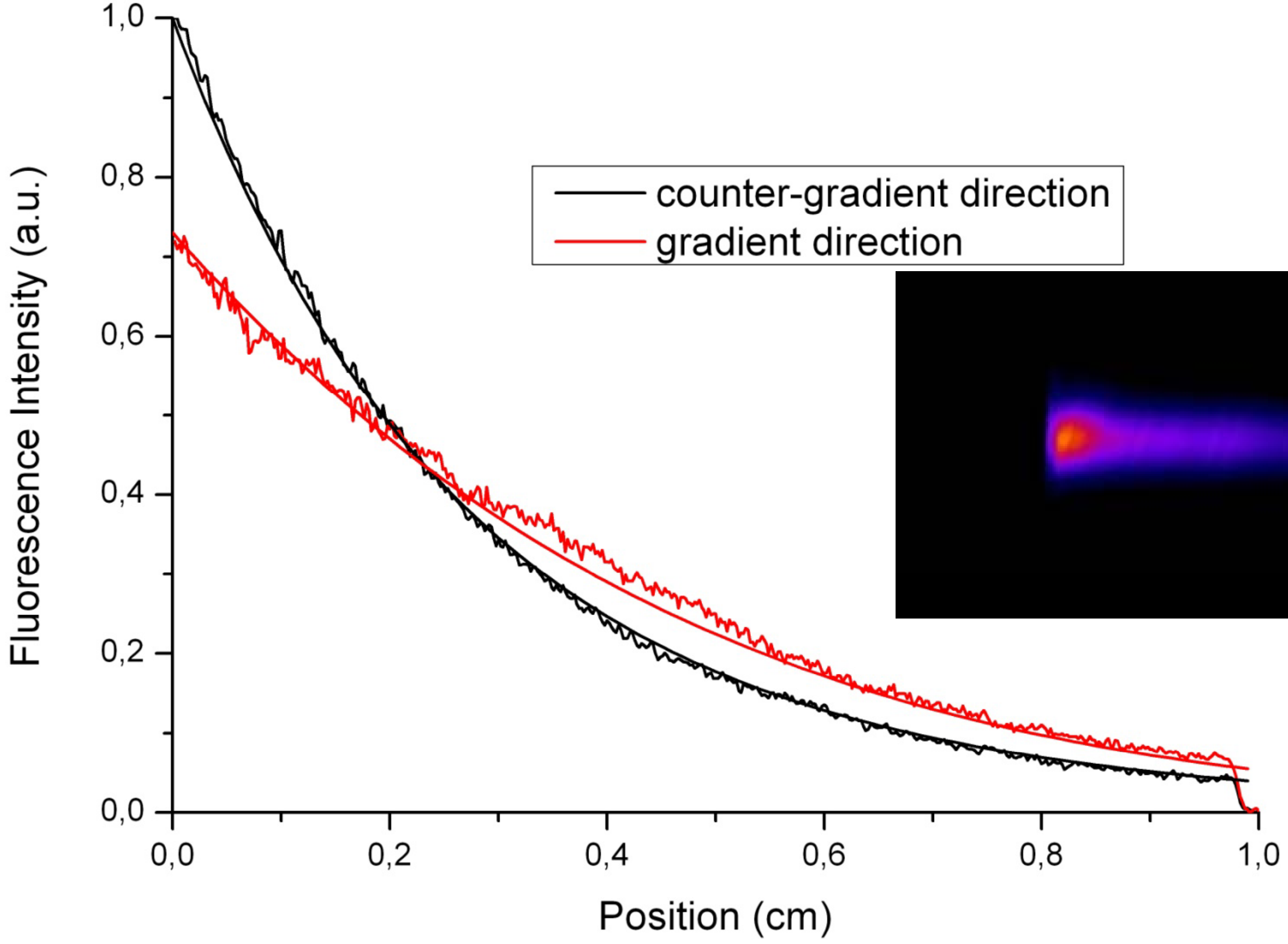
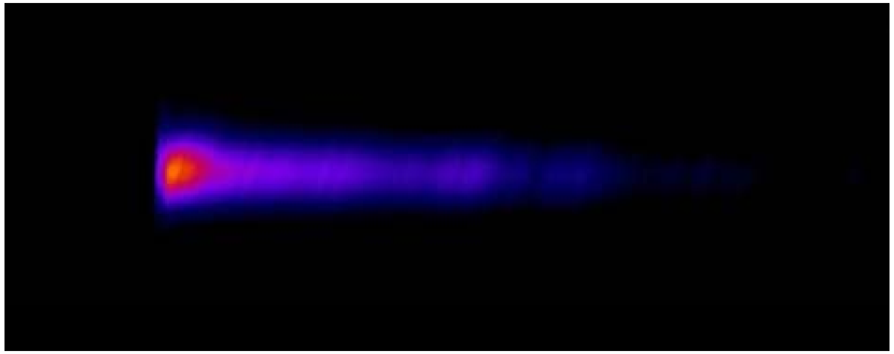
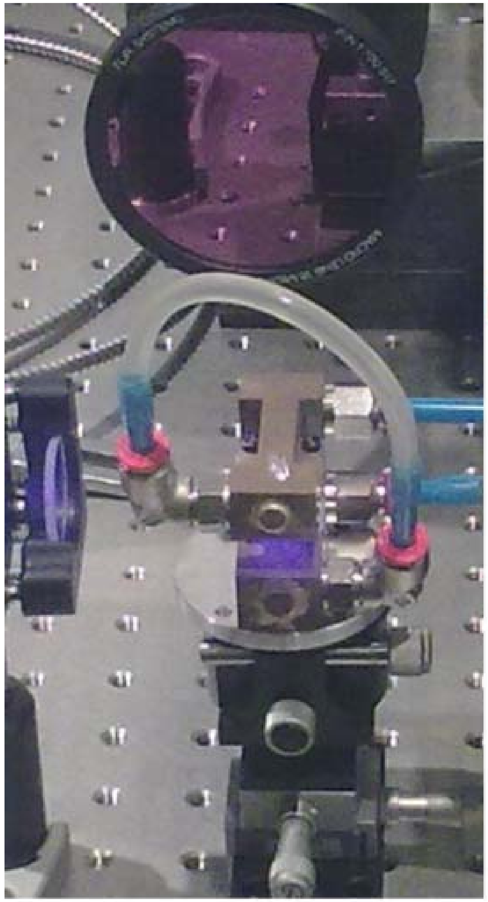
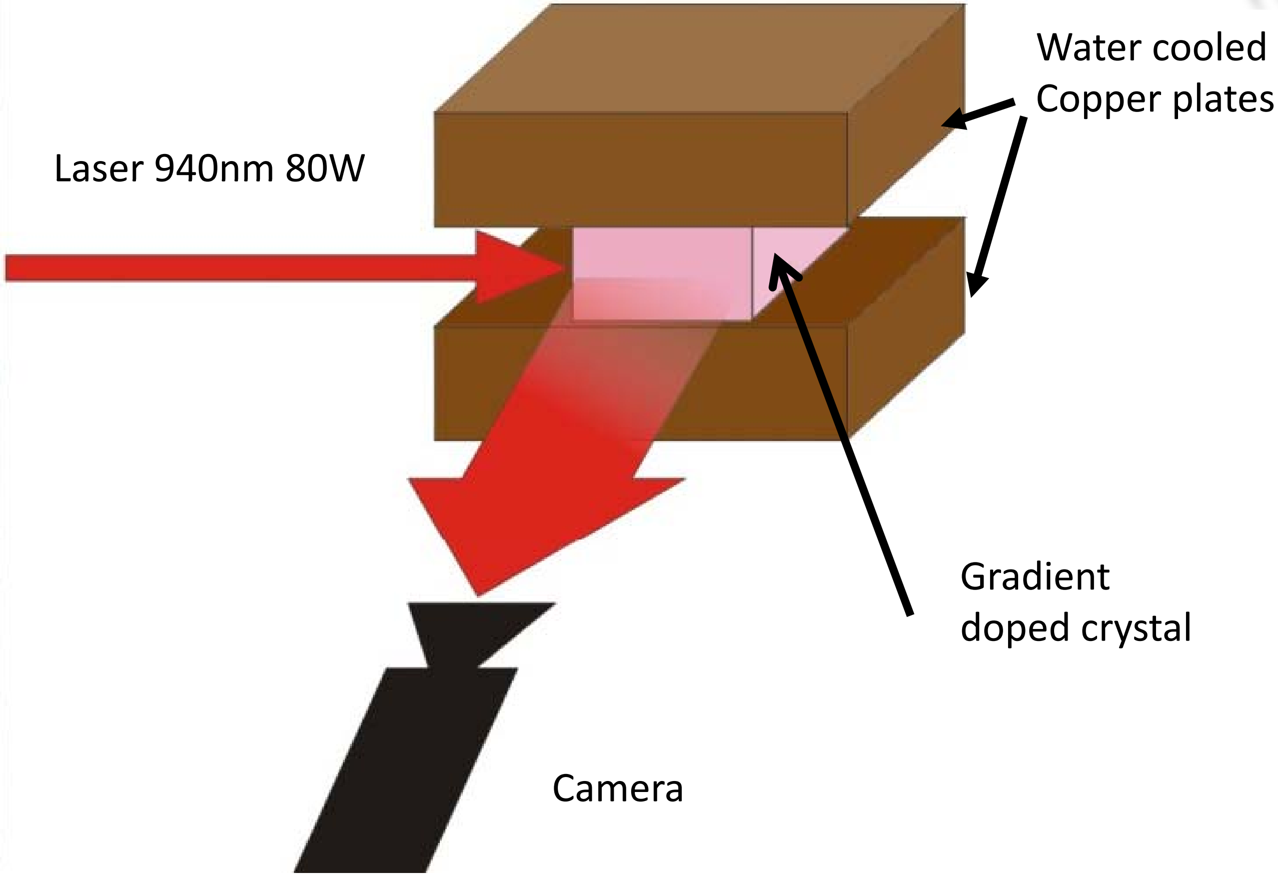




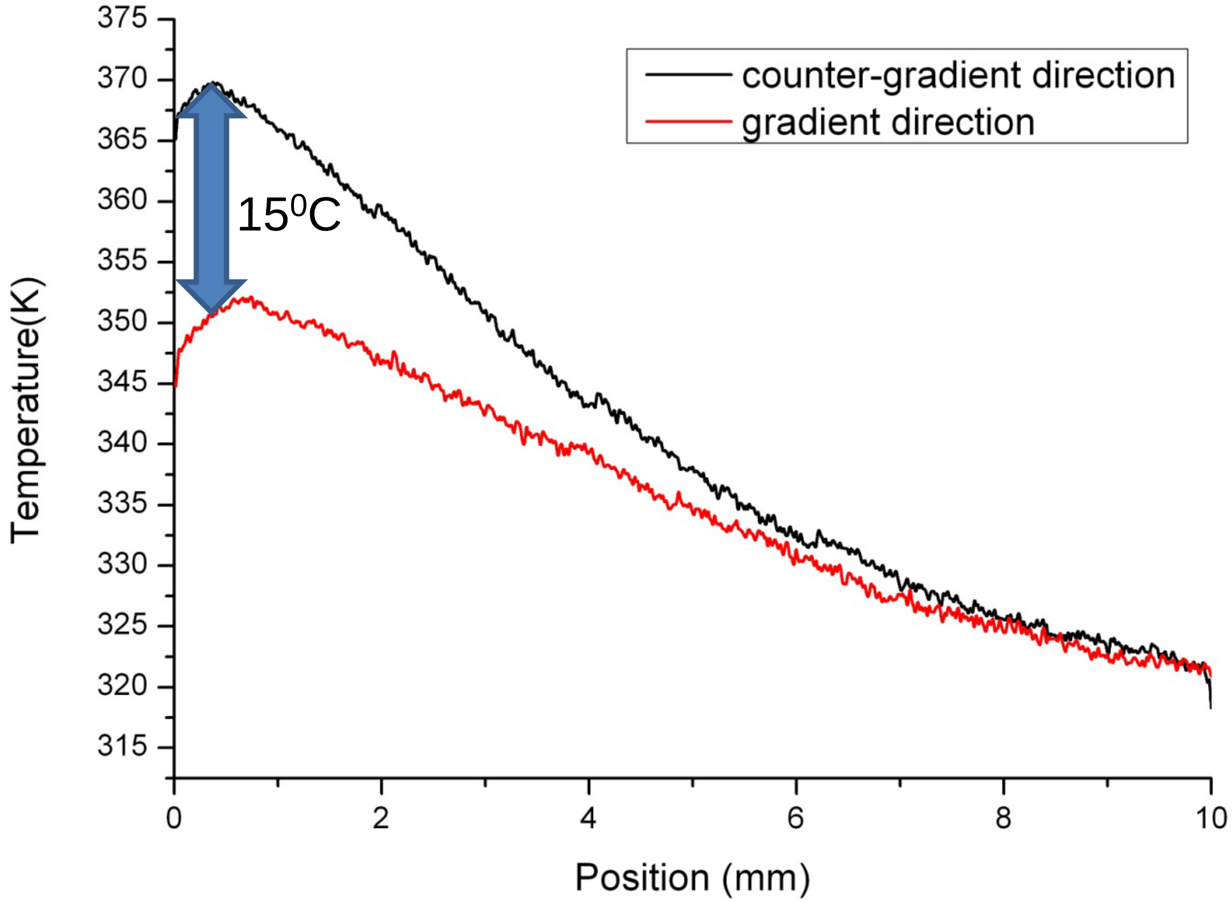
Impact of gradient doping is clearly revealed

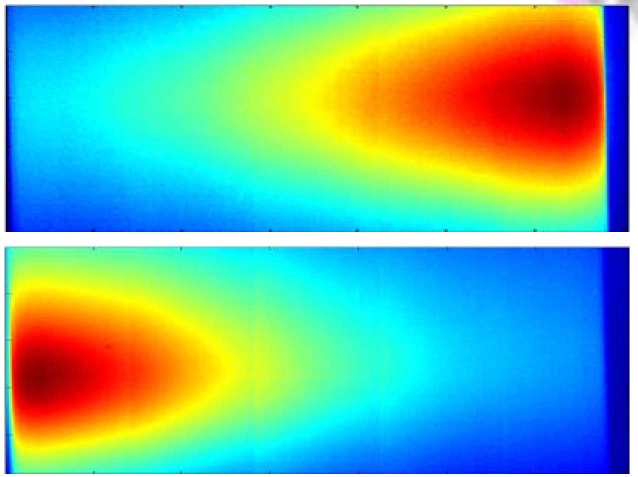
Thermal imaging: experimental setup





Thermal imaging: results



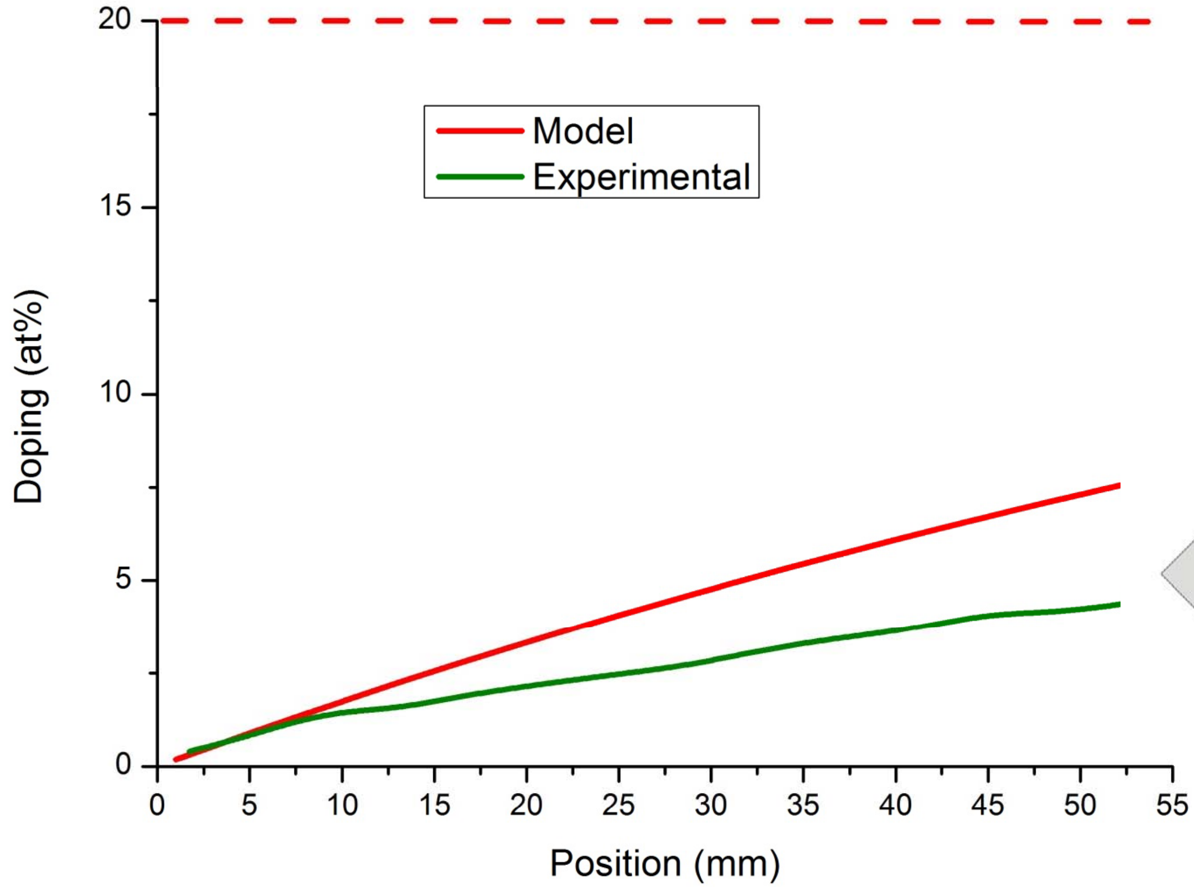


A more than 15 degrees difference is observed for opposite gradient directions

<u>Higher gradient will be required to reach an even more homogeneous T° distribution</u>

Simple model vs experimental results

Our simple model does not allow to understand the gradient value experimentally observed



Molten zone length L_m were considered as constant during the growth process

➔ Experiments were set up to evaluate Lm(x) and T(t,x)

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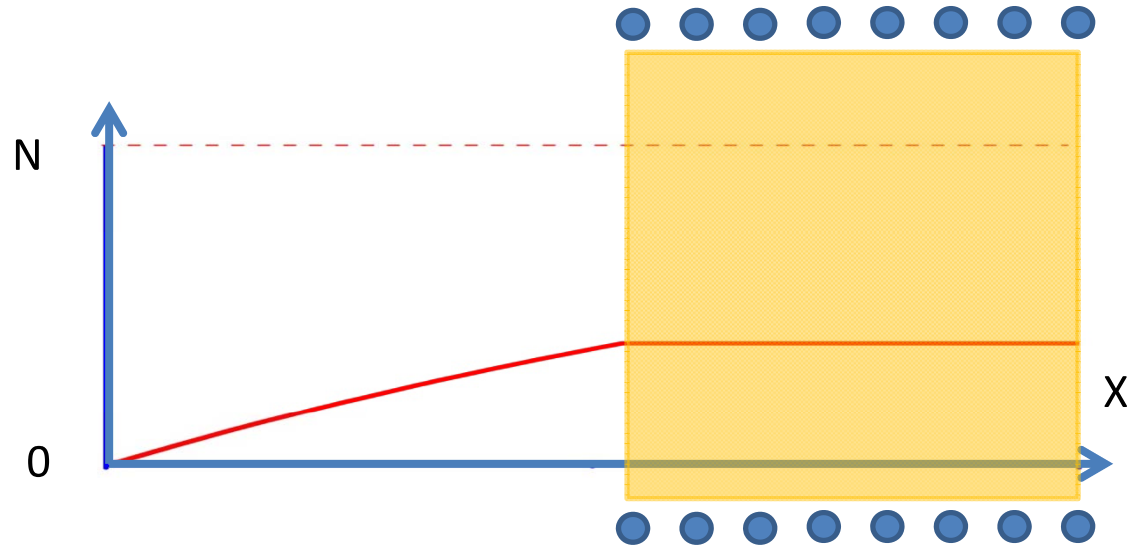
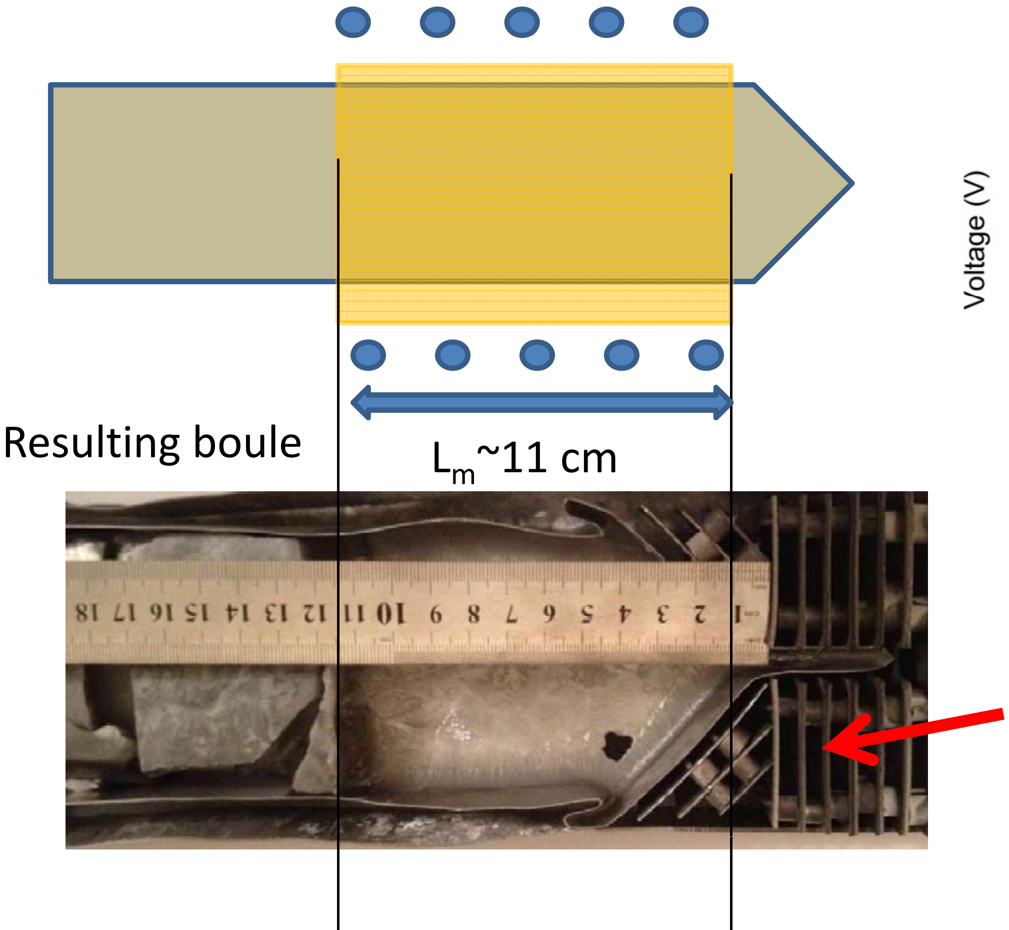
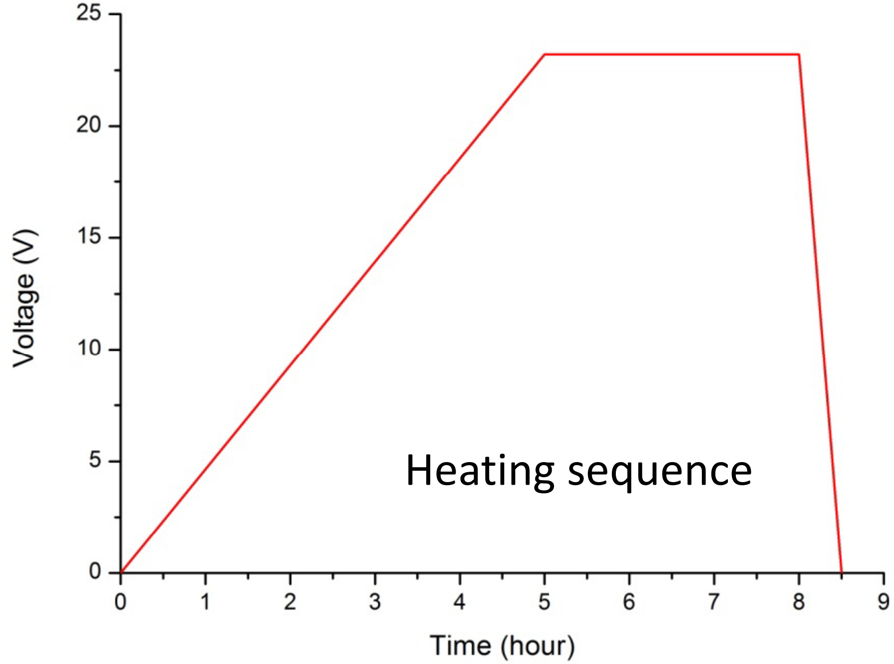


Molten zone L_m shrink experiment : 1st step

Molten zone shrinks in size within the growth process. Quantifying this shrinkage will allow avoiding constant molten zone length approximation and lead to better matching with experimentally observed gradients.

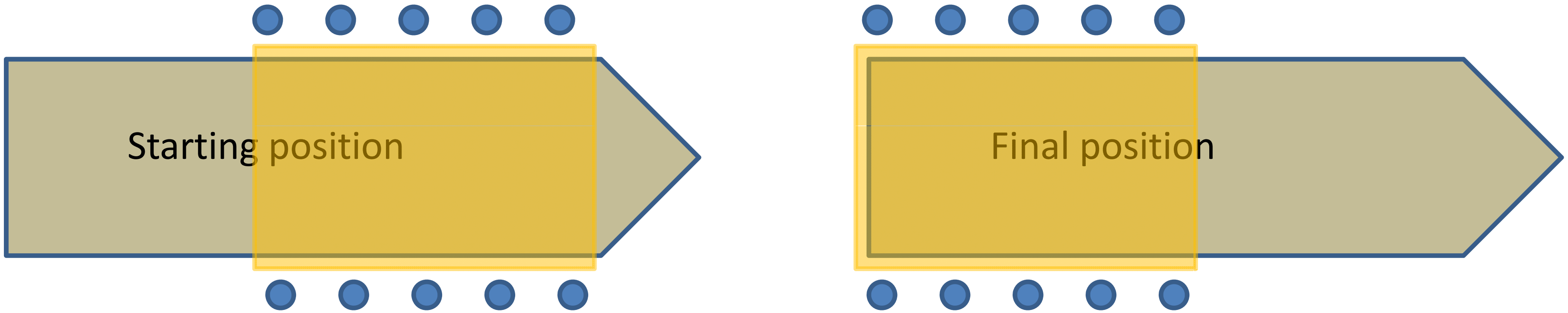
No translation of the crucible!!!



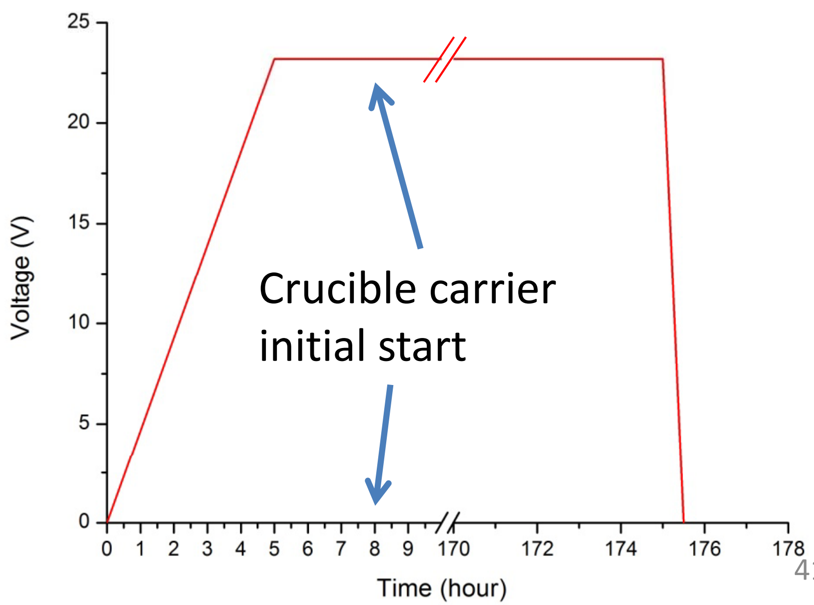


Molten zone L_m shrink experiment : 1st step

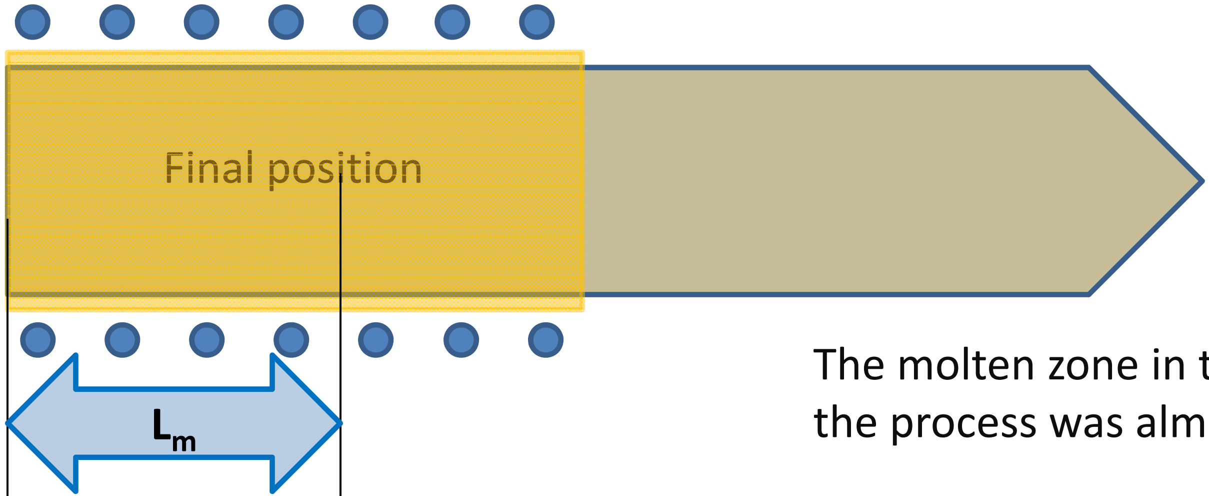
In step 2, the crucible (still loaded with resulting content of step 1) will now move slowly inside the heat zone (~1.5 mm/h)



Translation occurs with a 1.5mm/h rate like during standard growth.



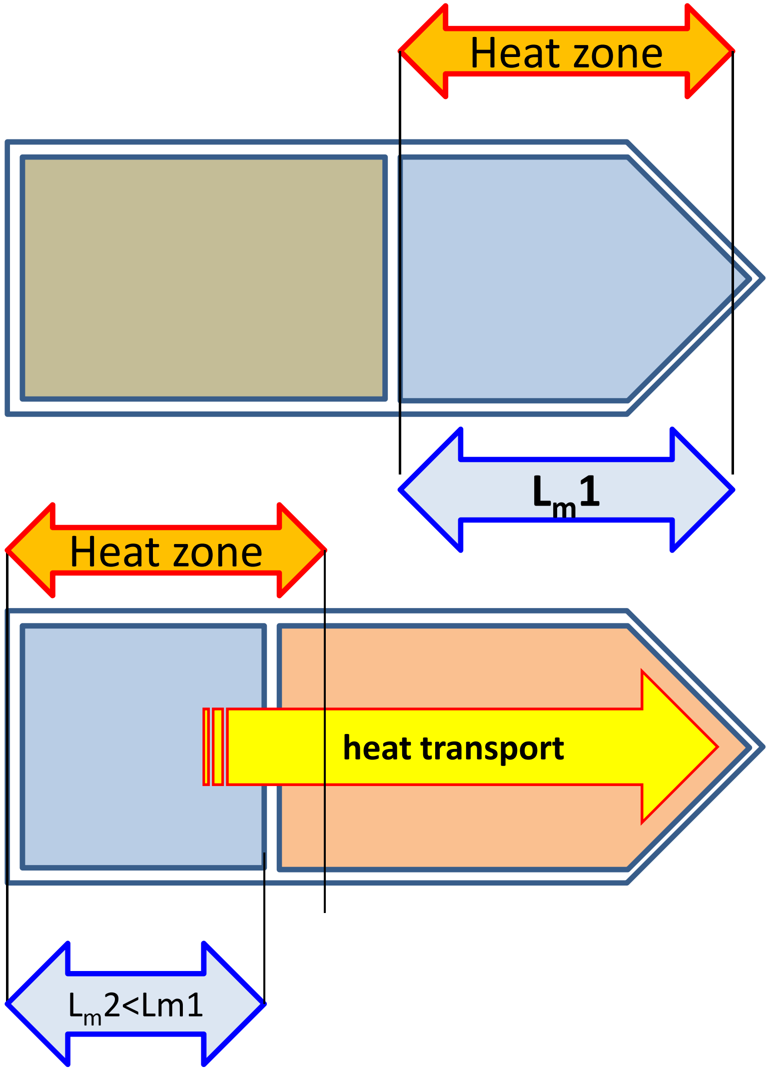
Molten zone L_m shrink experiment : 2nd step



The molten zone in the beginning of the process was almost **110 mm**.

By the end of the process, the molten zone is almost twice less in length, i.e. **~50 mm**.

Molten zone L_m shrink experiment : 2[nd] step



Explanation lies in the fact that the grown crystalline part exhibits higher thermal conductivity and therefore evacuates the heat very effectively

Molten length new expression can be linearly approximated by :

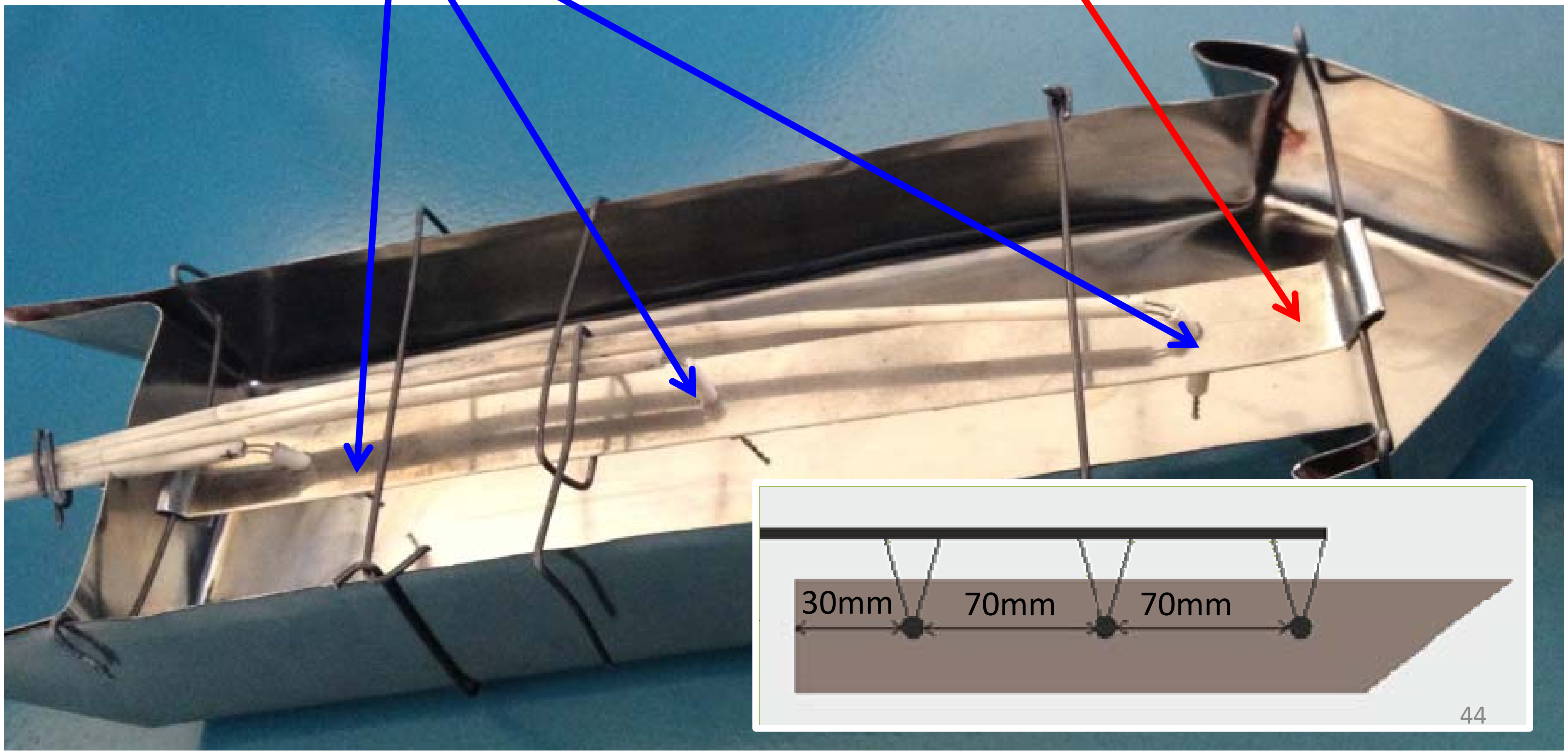
$$L(x) = L_m(0) + x\frac{L_m(final) - L_m(0)}{D}$$

Where D is a total translation

Temperature distribution evolution experiment

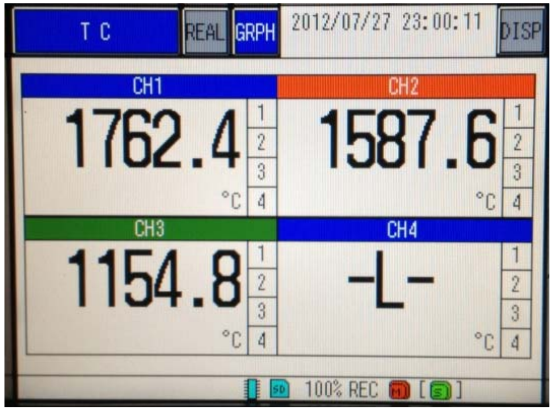
Besides the molten zone length L_m reduction during the growth process, the temperature distribution within the crucible evolves as well.

Three **C type thermocouples** are attached to the crucible via the **Mo holder** in appropriate positions and are electrically insulated from the crucible.



Temperature distribution evolution experiment

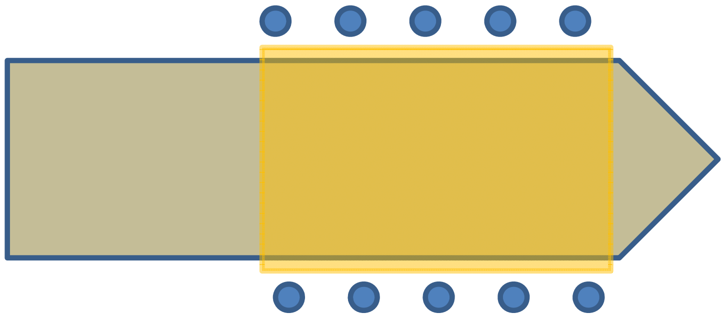
Filling the crucible with starting material



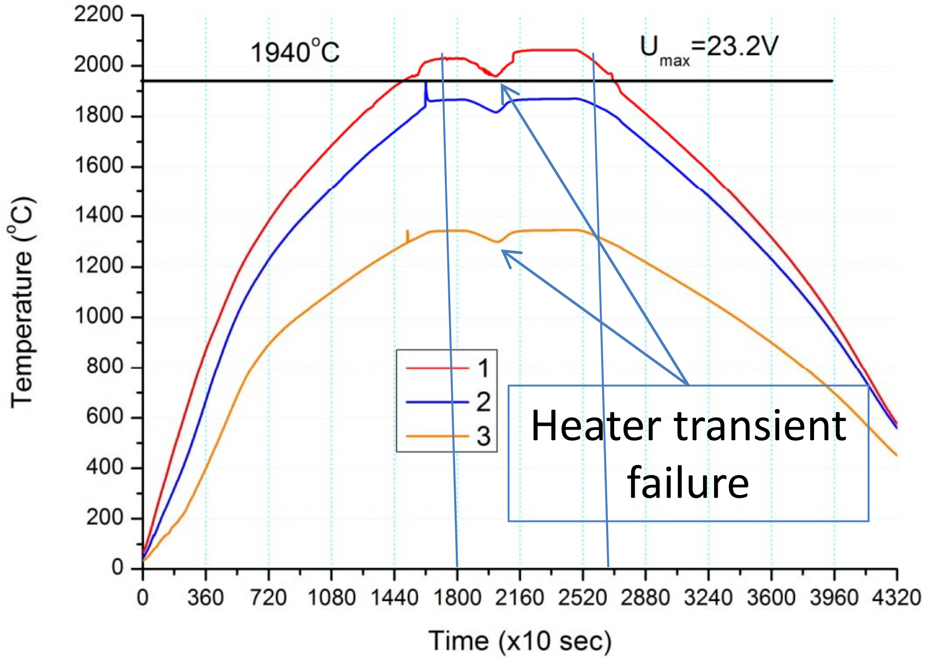
….inserting the crucible in the furnace…

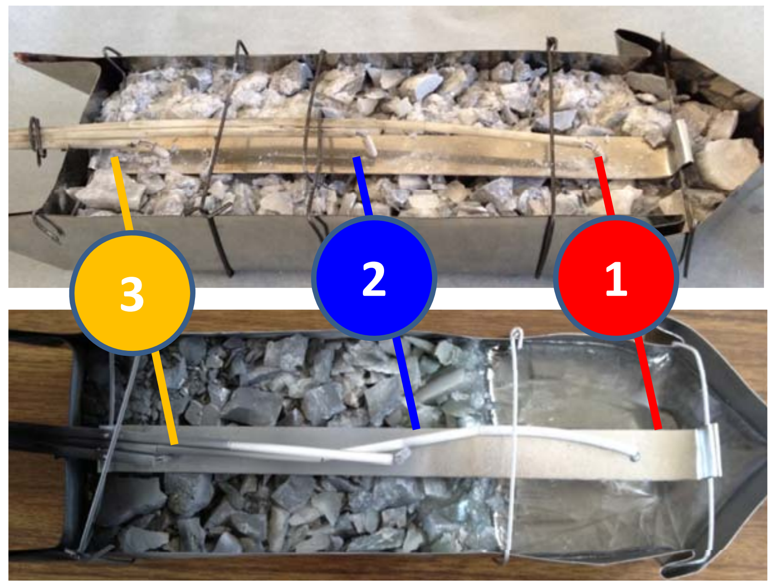
…and monitoring the temperatures with the 3 thermocouples

1^{st} experiment



Observed Temperature distribution





Input boule

Resulting boule

<u>Results :</u>

T_{1max} = 2062°C

T_{2max} = 1869°C

T_{3max} = 1344°C

T_{1max} - T_{2max} = 193°C

T_{2max} - T_{3max} = 525°C

Temperature distribution evolution experiment

The temperature in the melt, starting material and crystallized solid is measured for different positions of the crystals.
We observe different temperature distributions

These results can be used to evaluate effective thermal conductivity for the different phases.

The results are also very important in order to grow composite crystals.

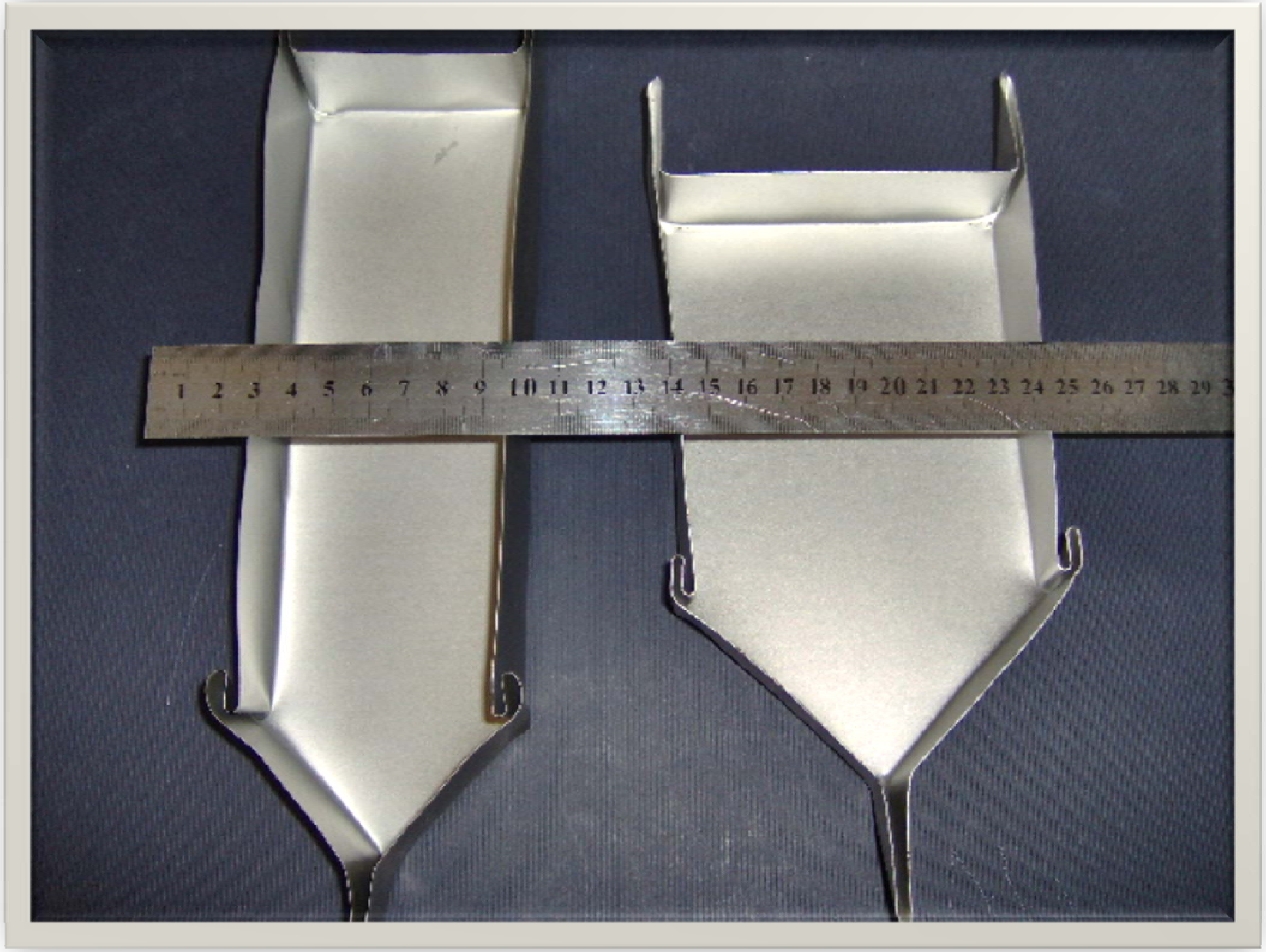
Further analysis and experiments are needed to evaluate the temperature fields in all axes to get full control over this parameter.

Outline

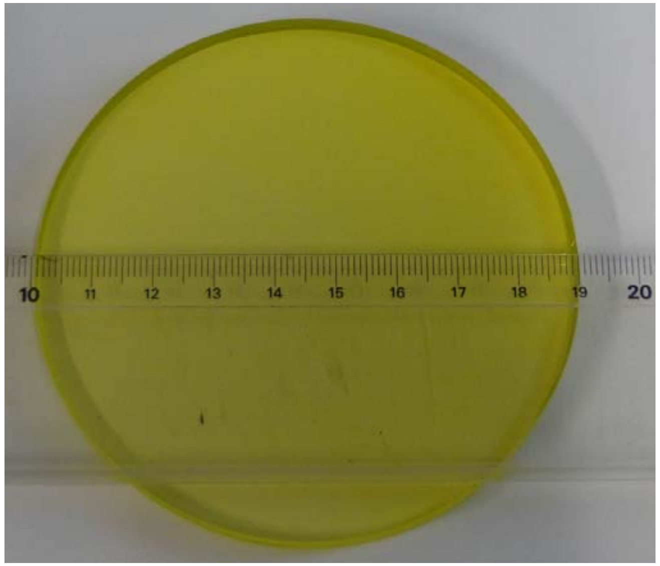
- Impact of gradient doped gain medium on thermal and ASE management
- YAG growth techniques
- Yb:YAG growth with Bagdasarov method
- Simple model for gradient doping growth
- Doping measurement techniques
- Experimental results on gradient crystal growth
- Experimental campaigns aiming at improving our model
- Large Yb:YAG crystals

The crucibles for large YAG crystals

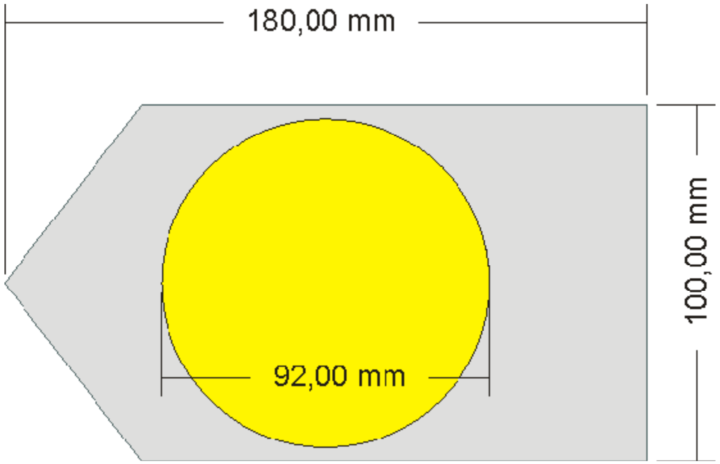
Larger crucibles were fabricated

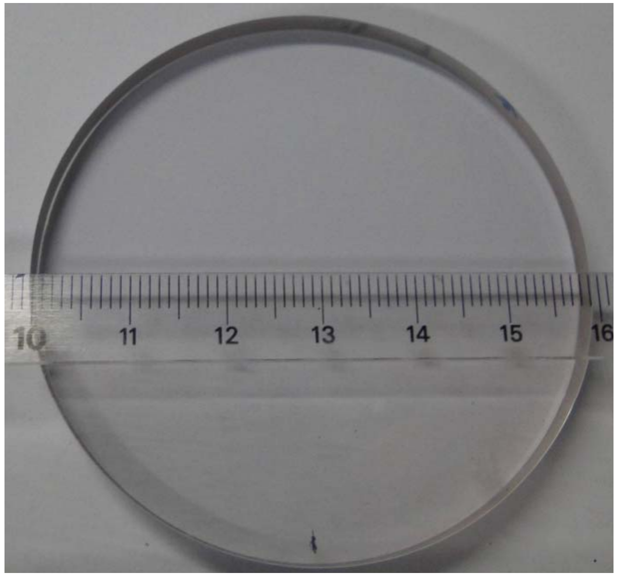


The large YAG crystals



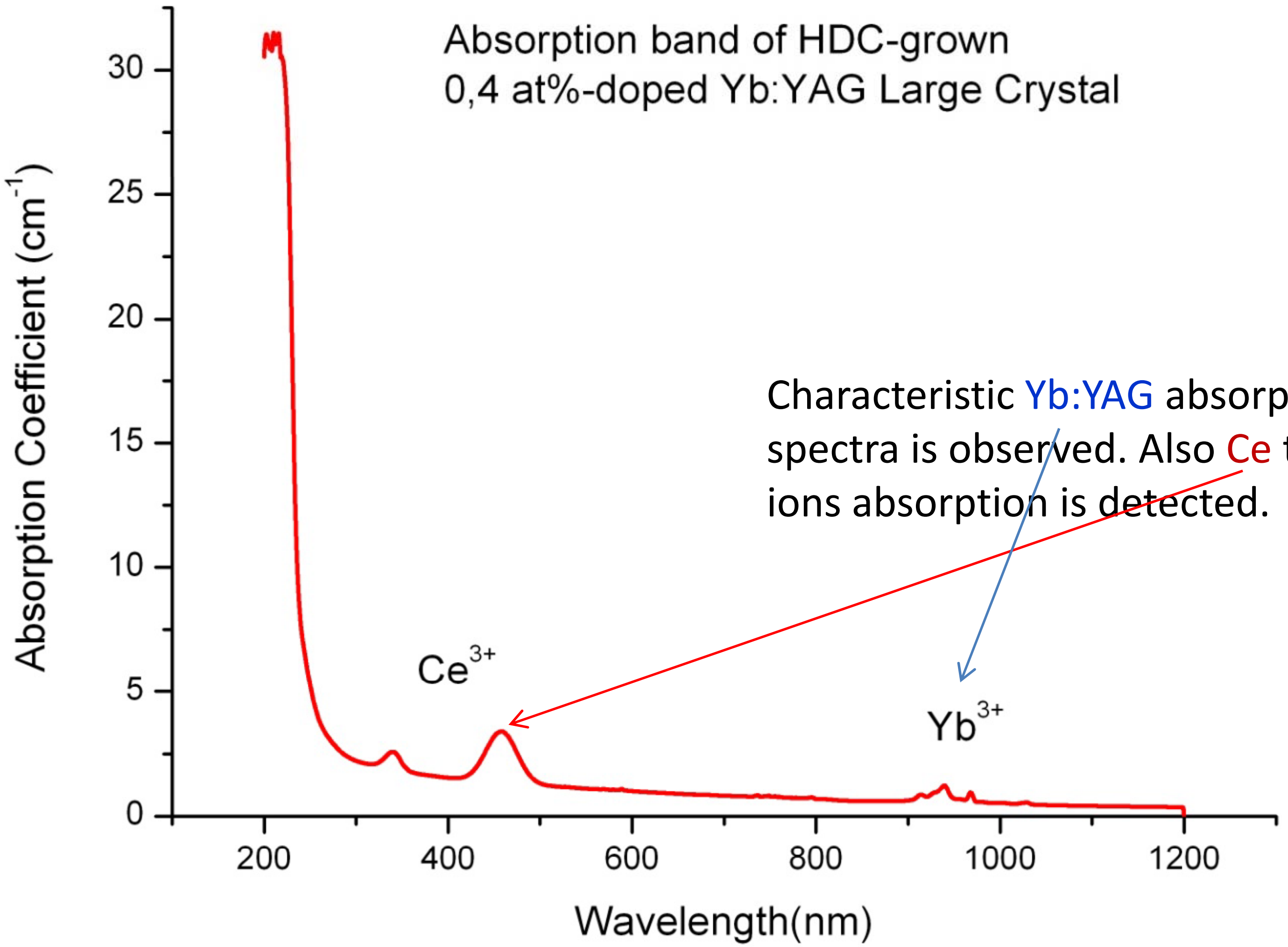
92mm Ce:Yb:YAG crystal





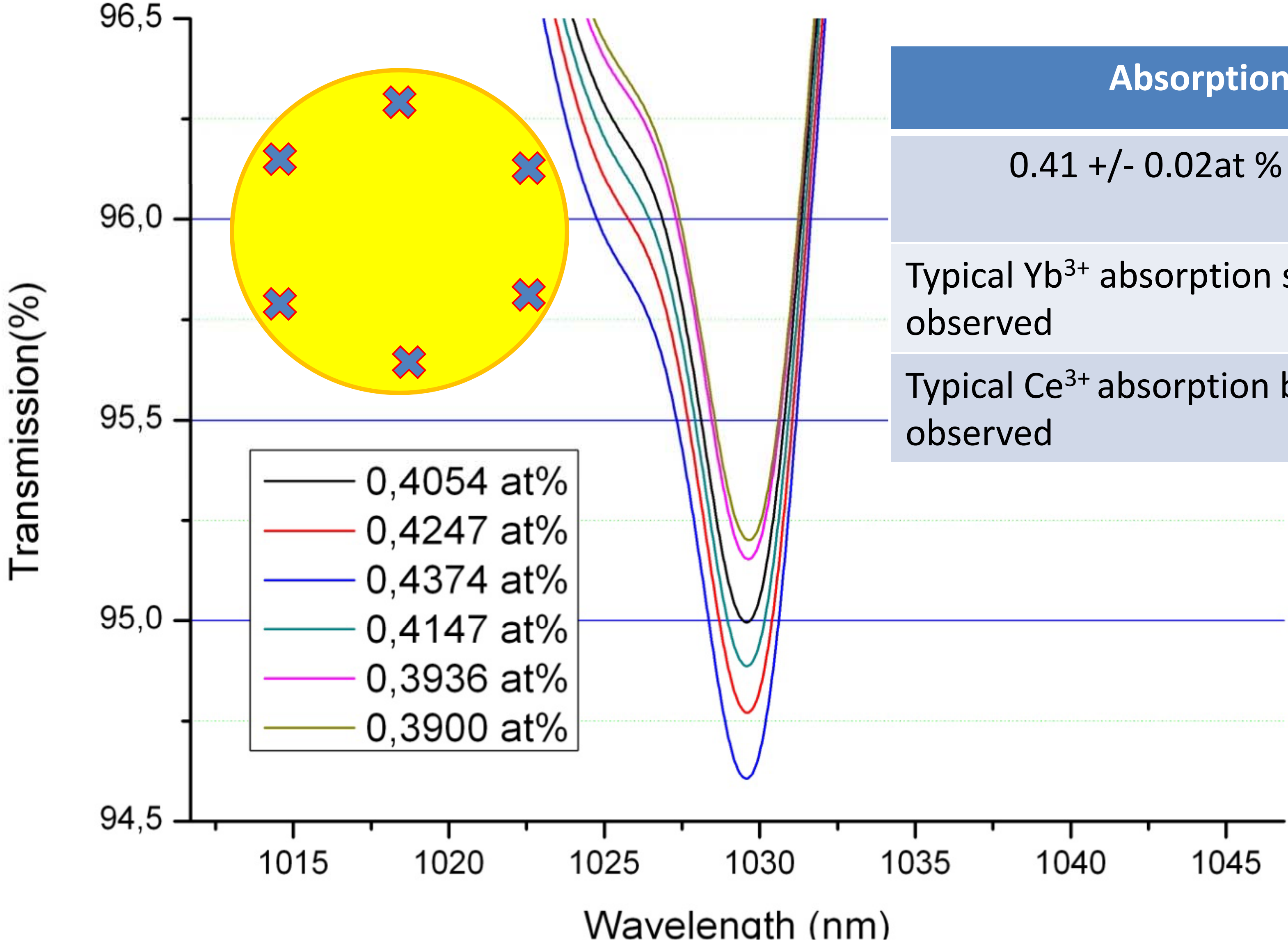
60mm Yb:YAG crystal currently used in LUCIA main amplifier

Absorption spectra



Characteristic Yb:YAG absorption lines spectra is observed. Also Ce trivalent ions absorption is detected.

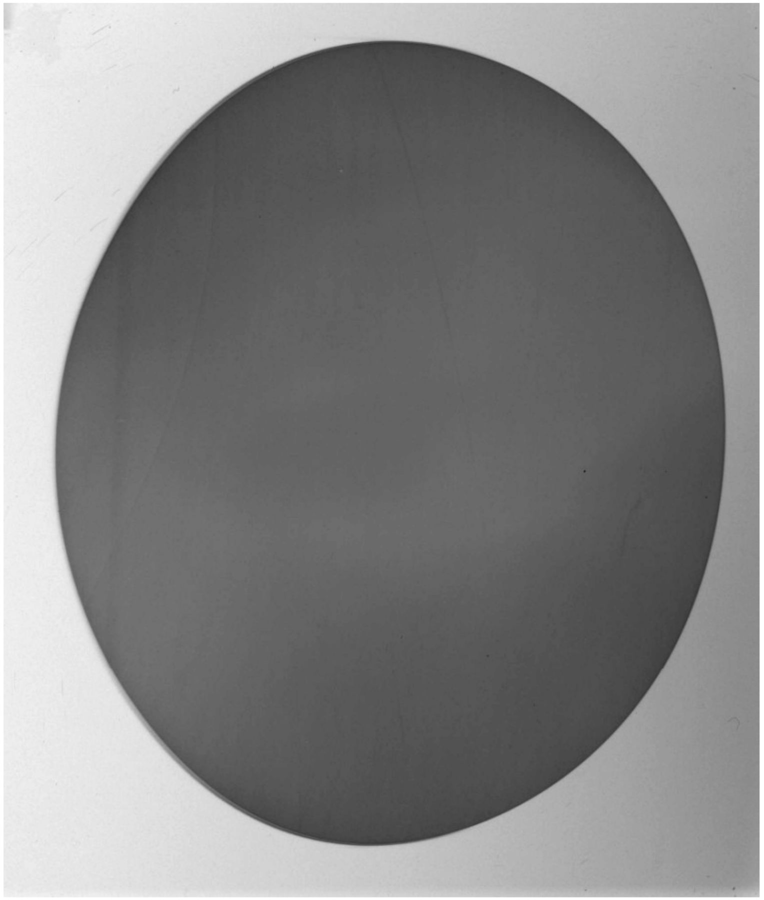
Doping homogeneity



Absorption
0.41 +/- 0.02at % of Yb^{3+}
Typical Yb^{3+} absorption spectra is observed
Typical Ce^{3+} absorption bands are observed

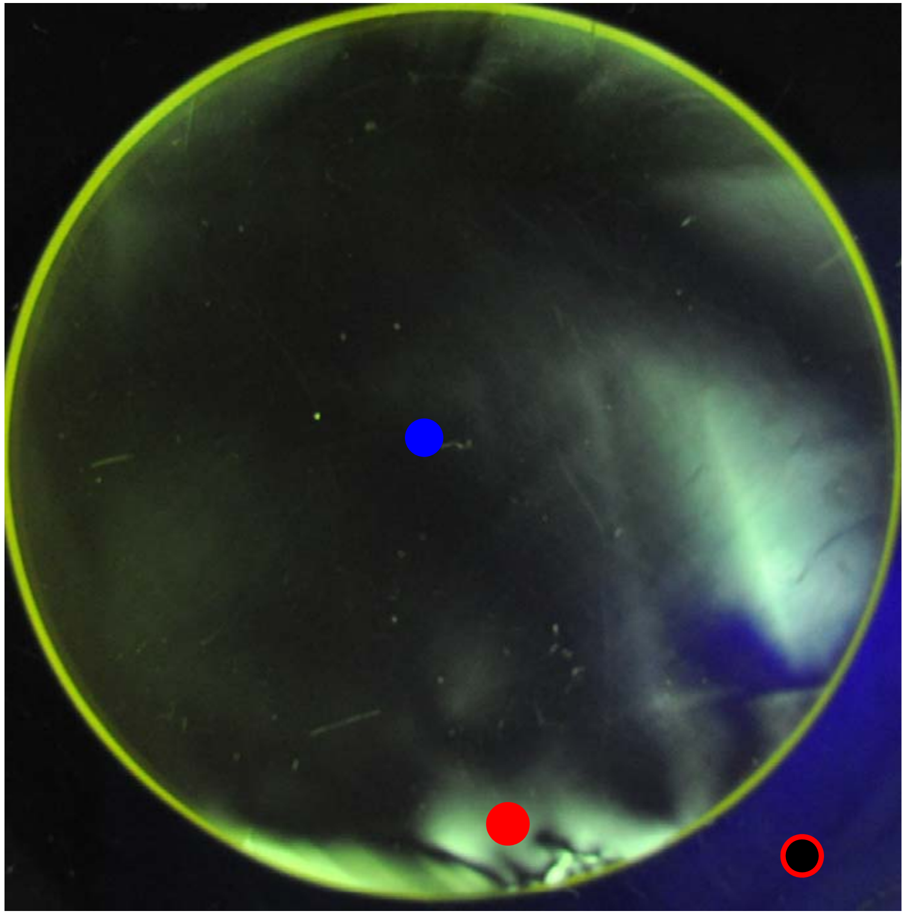
X-Ray (Cu-a) Topograms reveal no grains

Constant 2 at% doping level / 60 mm diameter crystal used for Lucia amplifier

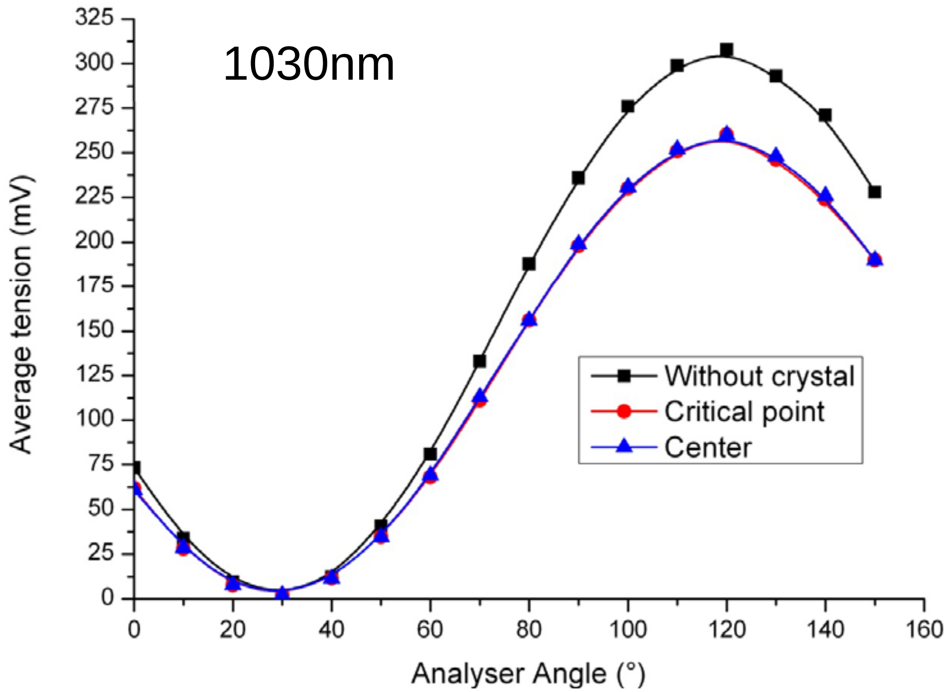


Thanks to Joachim Hein, IOQ, FSU, Jena, Germany …….

Depolarization in the crystals



White light observation



Stresses are concentrated mainly near the periphery, which is also the area of Interaction of the crystal and molybdenum crucible

Outlook

Variable doping

- Increasing the values for doping gradients through in-growth control of melt composition
- obtaining variable doping “hill” or “step” shape profiles
- growing gradient doped crystals with different dopants

Large crystals

- tailoring the furnace in order to increase the diameter of growing crystals
- growing large crystals of other garnets (for example` LuAG)

Method characterization

•Further experiments on growth characterization in order to improve the model

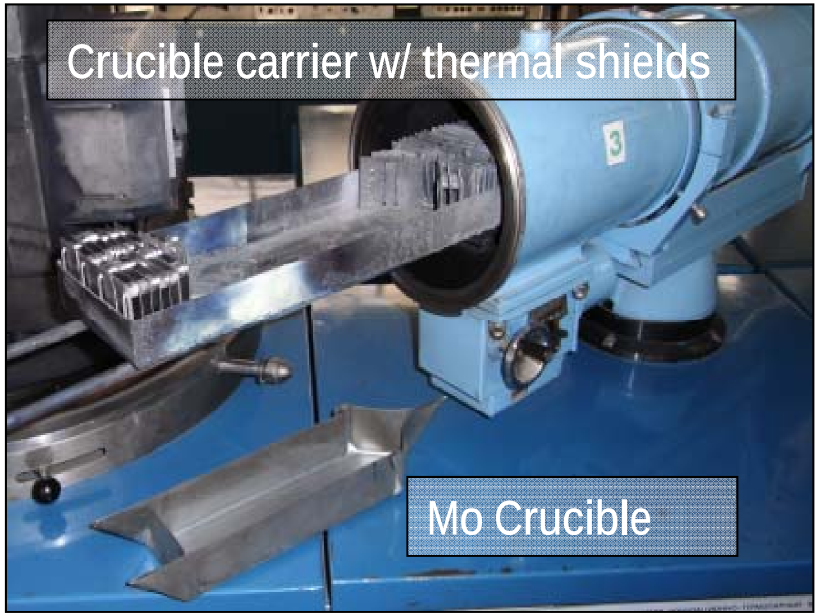
THANK YOU FOR ATTENTION

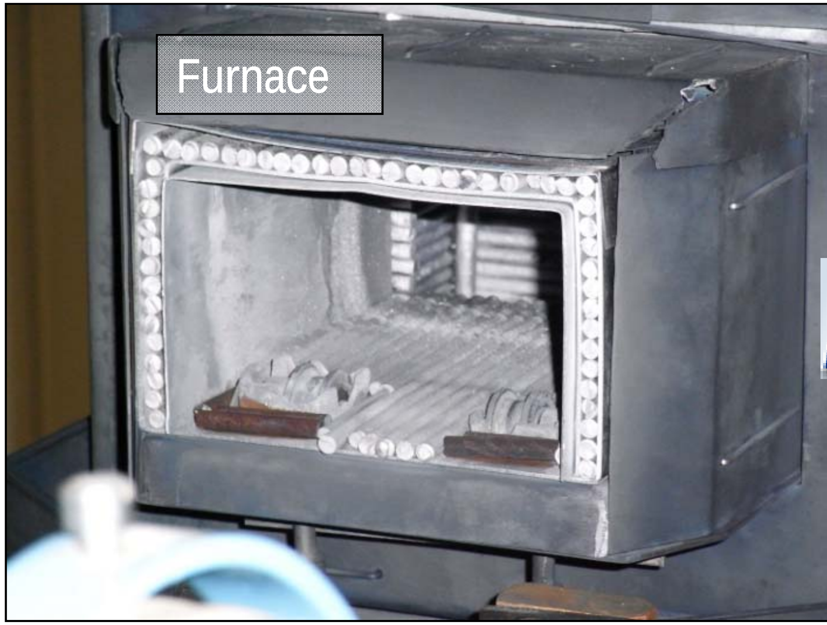
GAIN THANK YOU FOR ATTENTIO

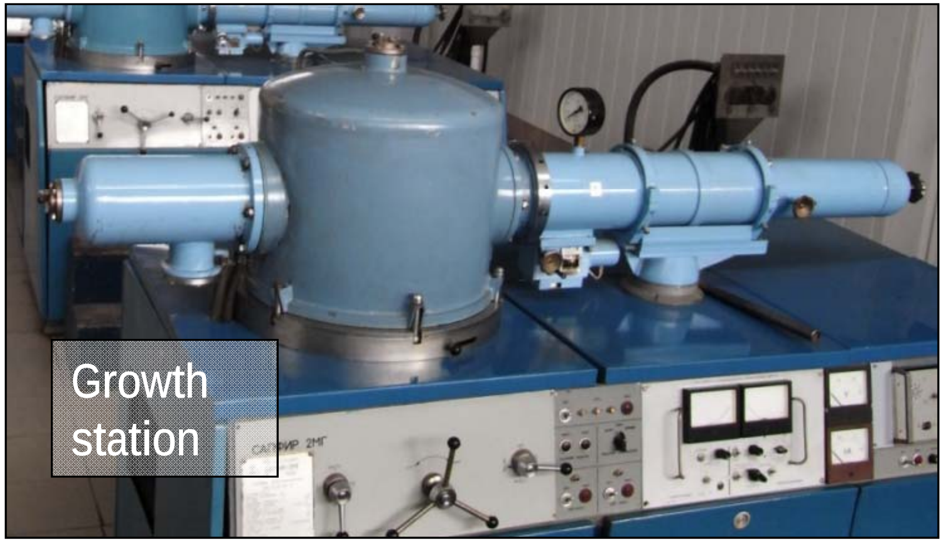
State of art

Gradient doped Yb:YAG crystals
R. Triboulet **The travelling heater method (THM) for Hg(1-x)Cd(x)TE and related materials**
Denis Freiburg and etc. **Power scaling of Diode End-Pumped Nd:YAG Lasers by Hyperbolic Dopant Concentration Profiles**
Ralf Wilhelm and etc. **Power Scaling of End-Pumped Solid-State Rod Lasers by Longitudinal Dopant Concentration Gradients**
Ralf Wilhelm and etc. **End-Pumped Nd:YAG laser with longitudinal hyperbolic dopant concentration profile**
Xiaodong Xu **Comparison of Yb:YAG crystals grown by Cz and TGT method**
Our journal of crystal growth
Our Optical materials express

Horizontal Direct Crystallization (HDC) exp^{tal} set-up

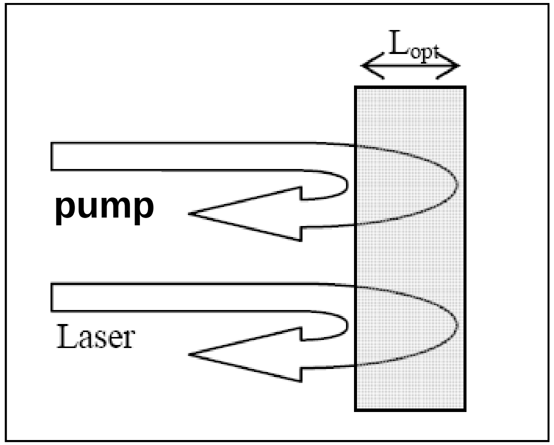






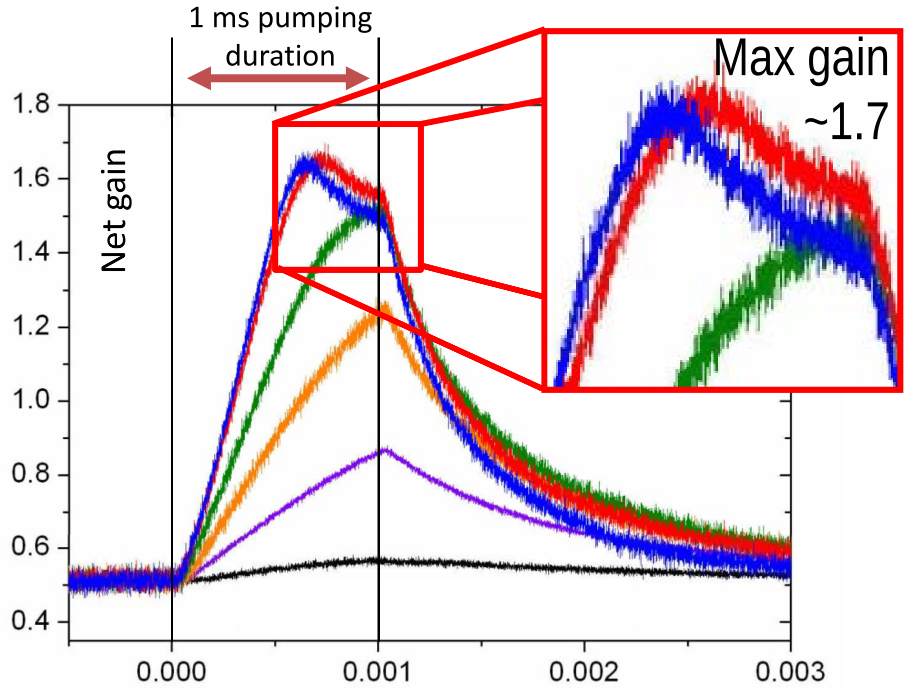
How it is calculated!

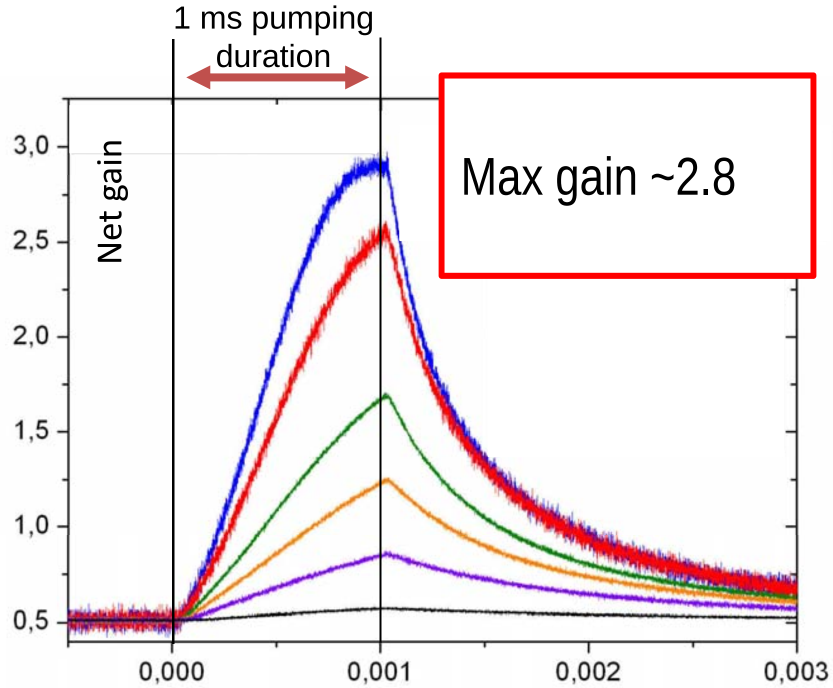
$$\begin{cases} \dfrac{d\beta}{dt} = \left\{\sigma_{abs\_P} - \left(\sigma_{em\_P} + \sigma_{abs\_P}\right)\beta\right\}\dfrac{I_P}{h\nu_P} - \dfrac{\beta}{\tau_{fluo}} \\ \dfrac{dI_P}{dz} = N_{tot}\left\{\left(\sigma_{em\_P} + \sigma_{abs\_P}\right)\beta - \sigma_{abs\_P}\right\}I_P \end{cases}$$

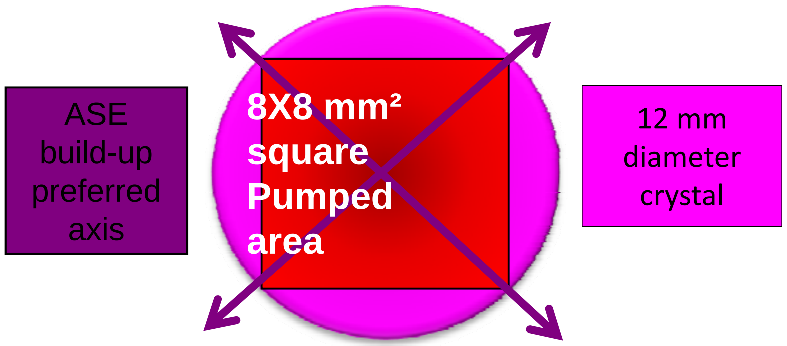


IFSA 2009 San Francisco HiPER

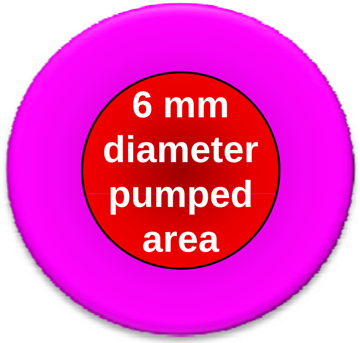
ASE losses impact on gain







1 kW/cm² 11 kW/cm²
4 kW/cm² 14 kW/cm²
8 kW/cm² 18 kW/cm²

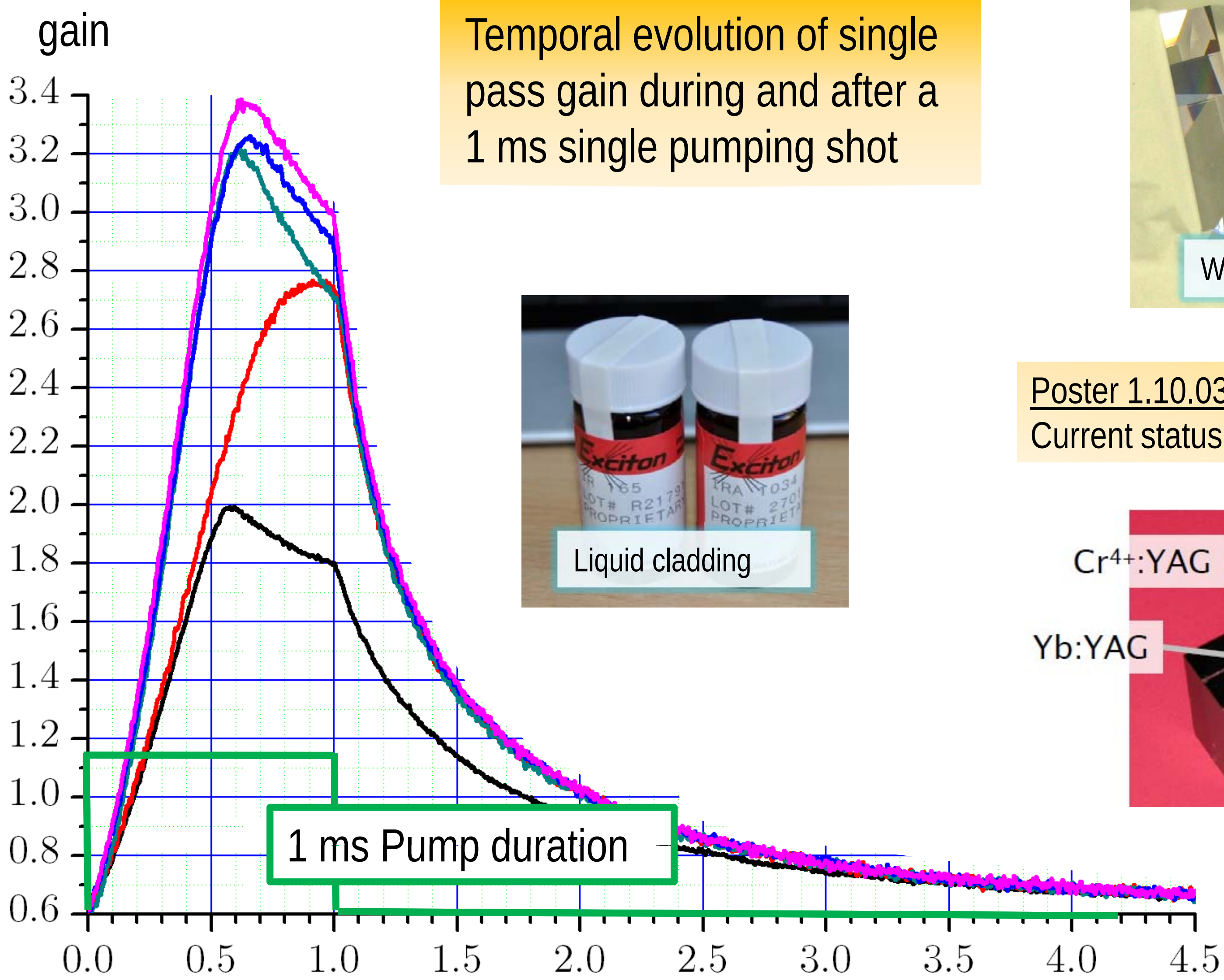


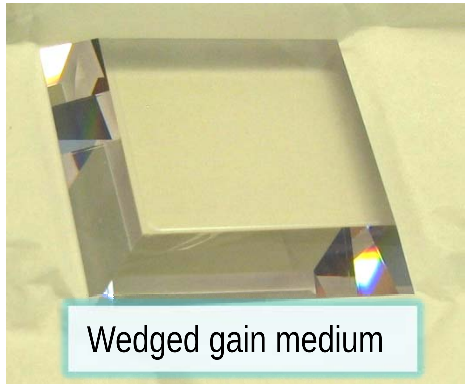
When the pump intensity exceeds 11 kW/cm², lateral losses cannot compensate for single path ASE ➔ oscillation regime depleting the gain

Taking advantage of full pumping brightness now possible with adequate cladding

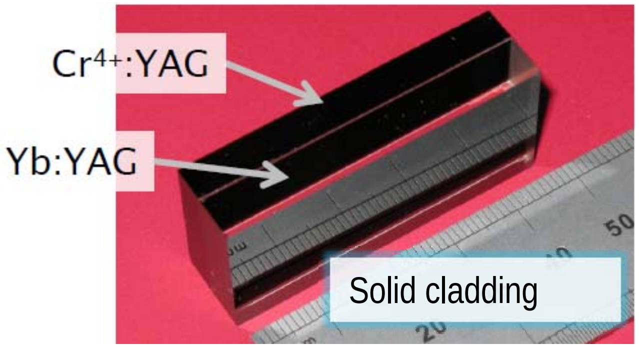
Amplified Spontaneous Emission managt

Temporal evolution of single pass gain during and after a 1 ms single pumping shot



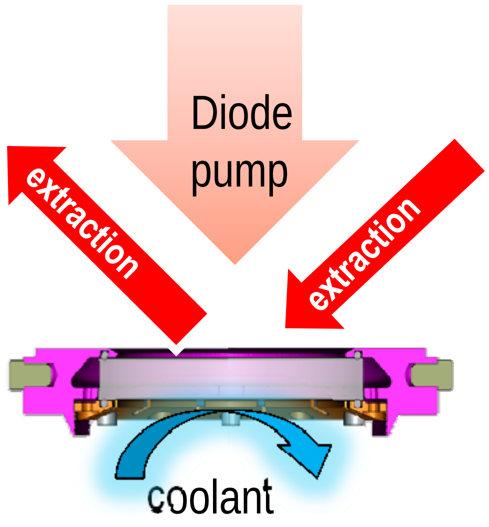


Poster 1.10.031: D.Albach
Current status of the Lucia laser system

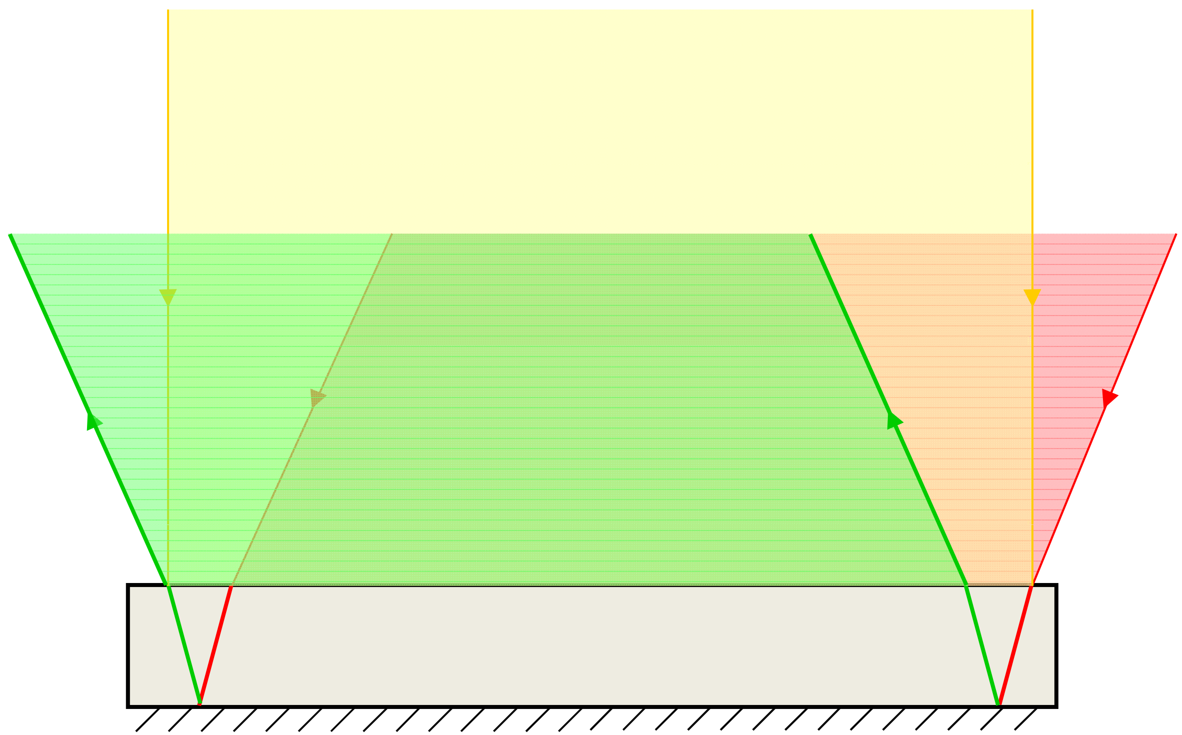


LUCIA System: Active mirror concept

LUCIA amplifying stages rely on active mirror architecture



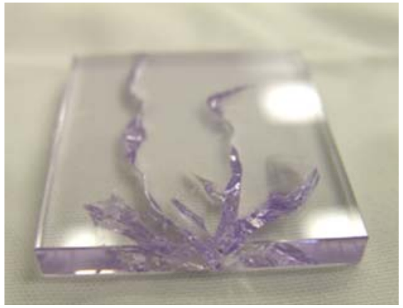
Pump and input extraction beams are reflected at the back surface of the disk



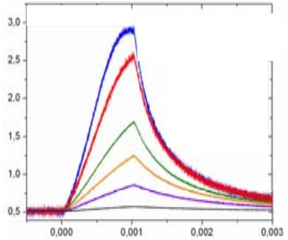
Crystal or ceramic	Incident beam } at 1030 nm
Pump beam at 941 nm	Reflected beam } at 1030 nm

Gradient doped crystals: Numerous positive impacts

Thermal
Management



Amplified Spontaneous Emission
Management



Gain
distribution

Positive
Impacts

Stored Energy Distribution

125 tons

Gain medium
volume requirement

Slide title

Outline

- Laser materials for LUCIA system: Yb:YAG
- Gradient doped materials and large crystals
- Yb:YAG growth with Bagdasarov method
- Doping verification different methods
- Gradient crystal growth
- Results on gradient crystals
- Large Yb:YAG crystals
- Other YAG crystals grown by Bagdasarov method

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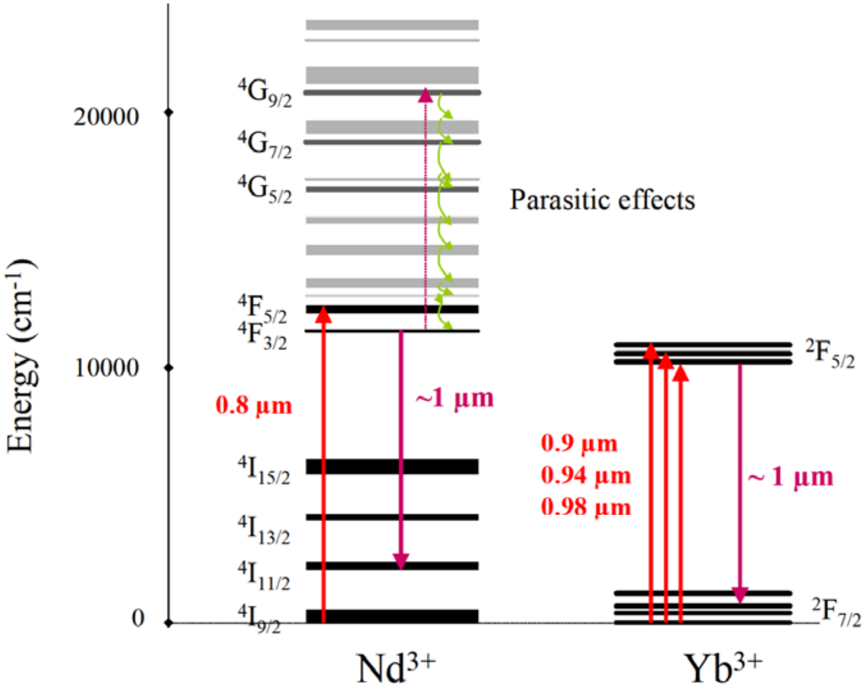
Yb:YAG as a gain medium for DPSSL

- Simple energy level system
- Long fluorescence lifetime (about 1ms)
- Suitability to high performance InGaAs/GaAs diode pumps
- No upconversion, cross relaxation and excited-state absorption



Energy levels of Yb and Nd ions. Typical laser transition lines are represented for both pump absorption and laser emission.

** Sébastien Chénais et al., Progress in Quantum Electronics 30 (2006) 89*

Outline

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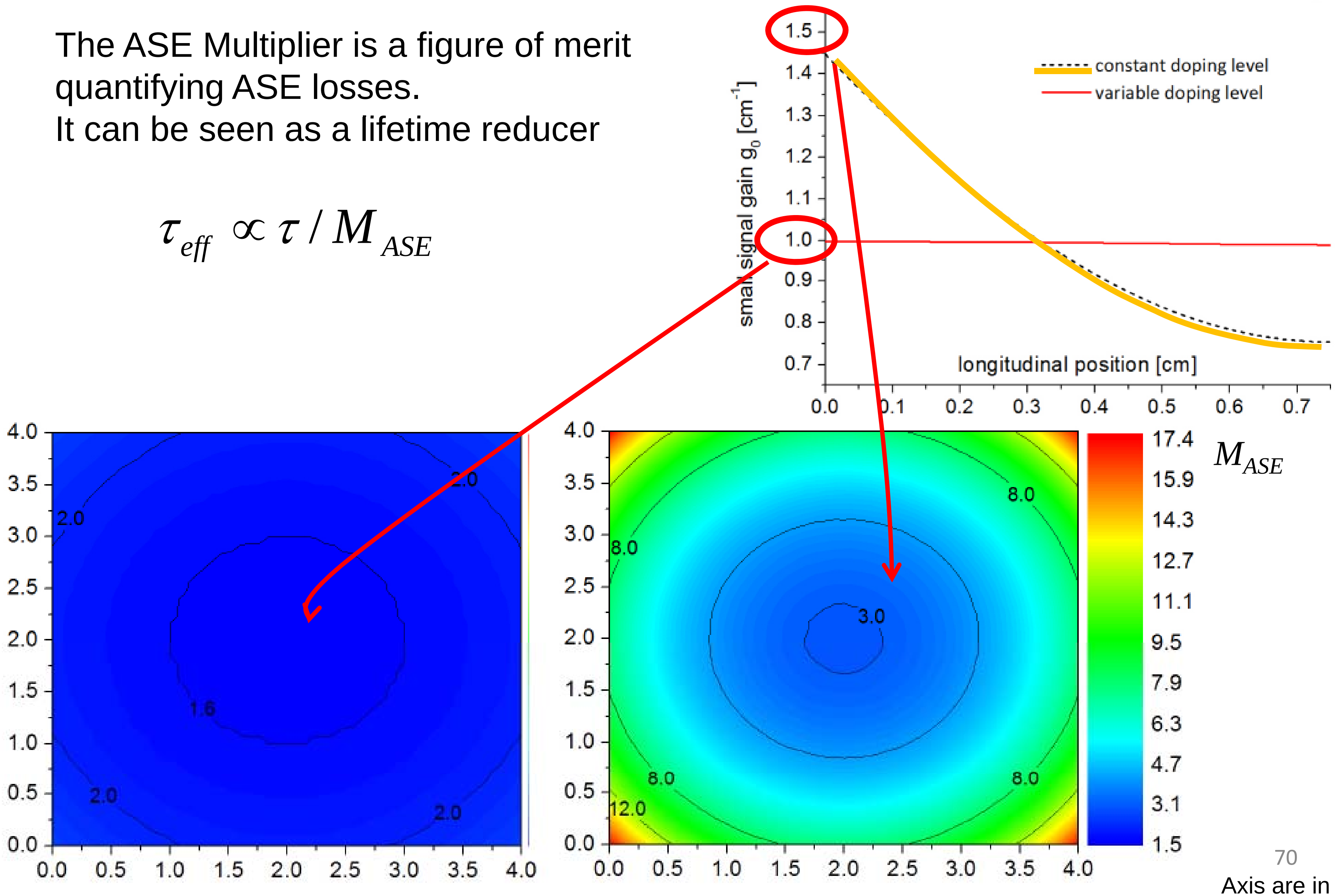
Impact on ASE

The ASE Multiplier is a figure of merit quantifying ASE losses.
It can be seen as a lifetime reducer

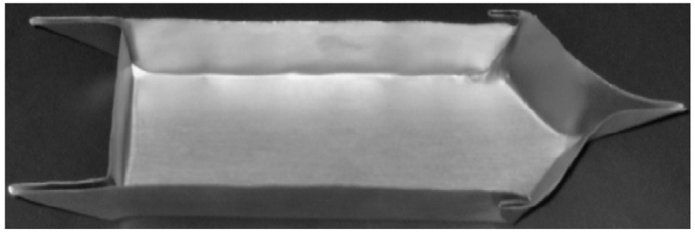
$$\tau_{eff} \propto \tau / M_{ASE}$$



Axis are in cm

Bagdasarov technique: growth conditions

- Crystallization atmosphere: *vacuum*, recovering, neutral
- Crucible material: *molybdenum* etc…



- Thermal shields: *tungsten and molybdenium,* graphite etc…

- Heater: tungsten

- Growth rates: 1-10mm/hour

Bagdasarov technique: growth steps

1-Powder weighting with appropriate mass ratio of Yb_2O_3 (or other dopant), Y_2O_3 and Al_2O_3 and placing it in special crucible

2-Melting the content by translating powder loaded crucible through heat zone under vacuum

3-Extracting the solid solution, preparing crackle

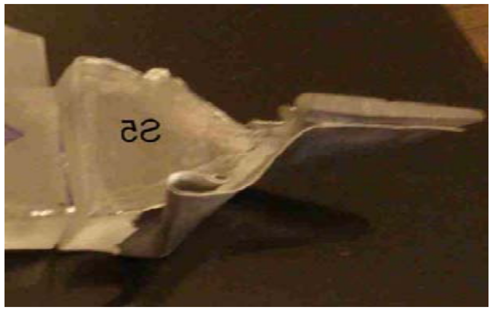
4-loading it into seeded boat-shaped crucible

Bagdasarov technique: growth steps

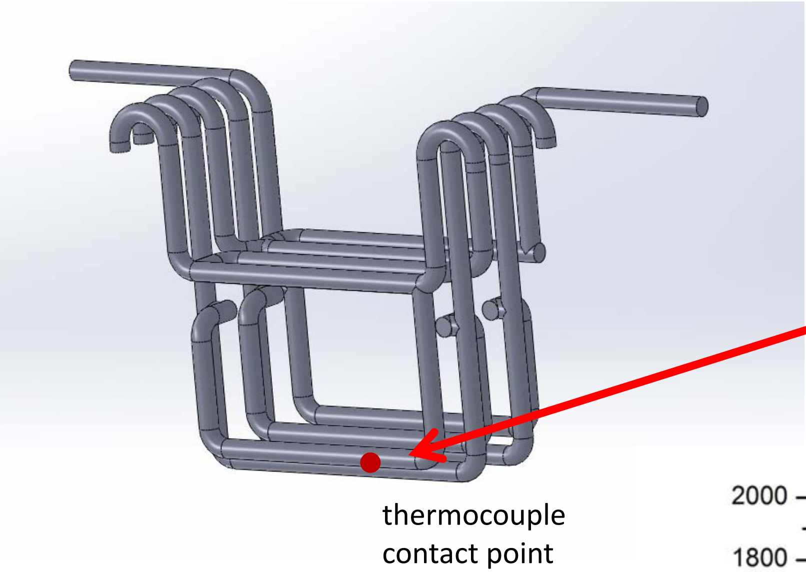
5-Placing the crucible in the furnace (with seed partially out of heat zone), pumping the vacuum, then rising the voltage on the heater. As soon as the zone under heater is molten translation starts.

Non-stoichiometric tail.

5-Mechanically extracting the boule by destroying the crucible

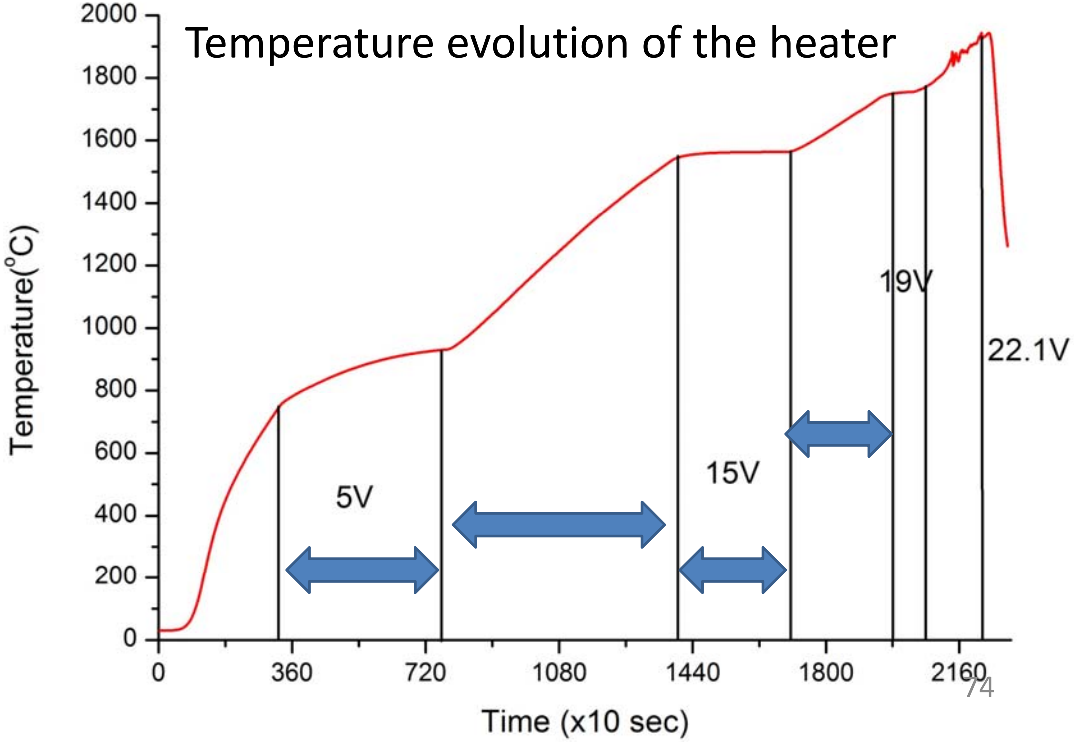


Characterization of the heater temperature

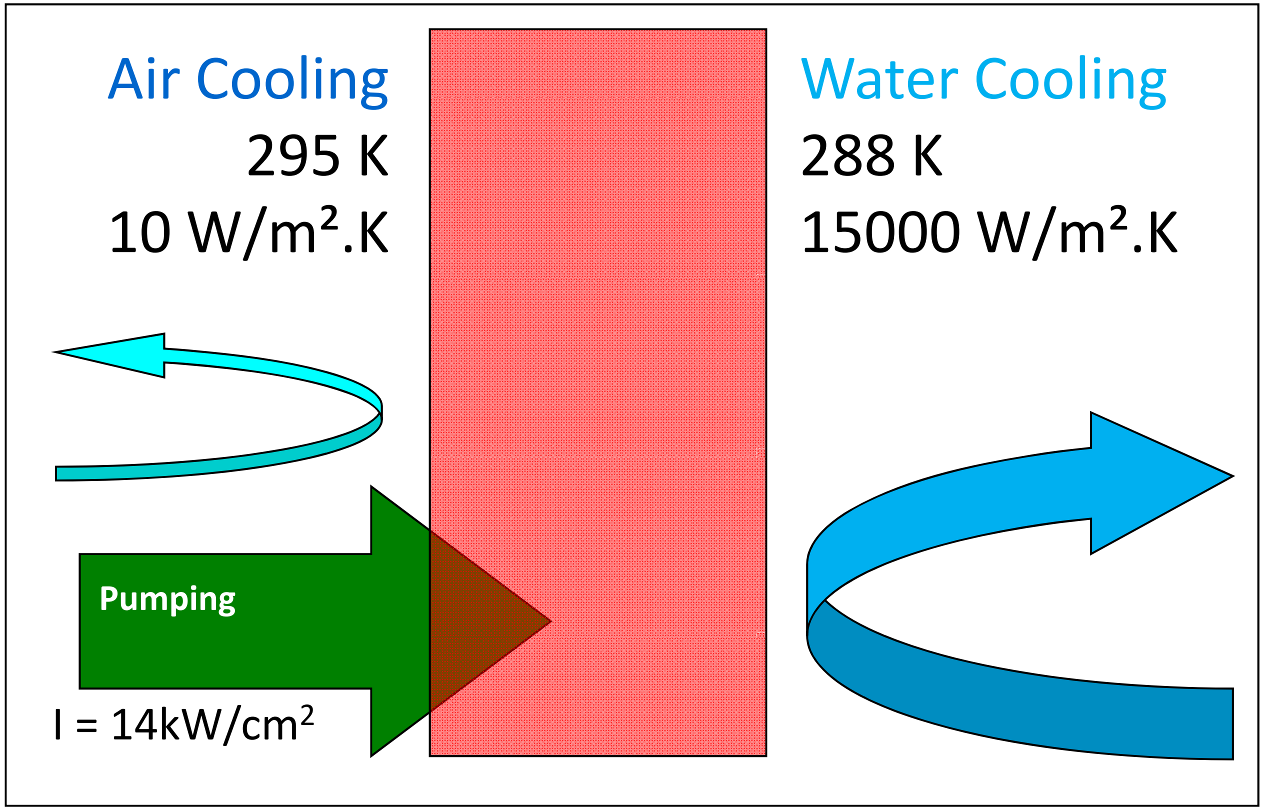


Tungsten-Rhenium thermocouple with open head and protected by ceramic substrates in order to attach to the heater

Straight vertical lines defines the limits of the areas with constant voltage.



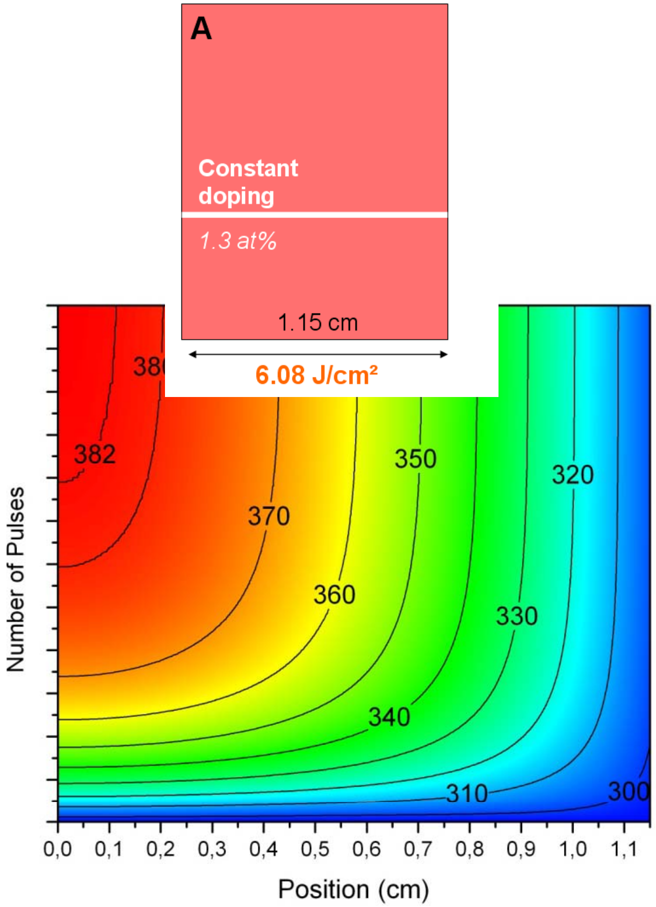
Impact on thermal management: Cooling scheme

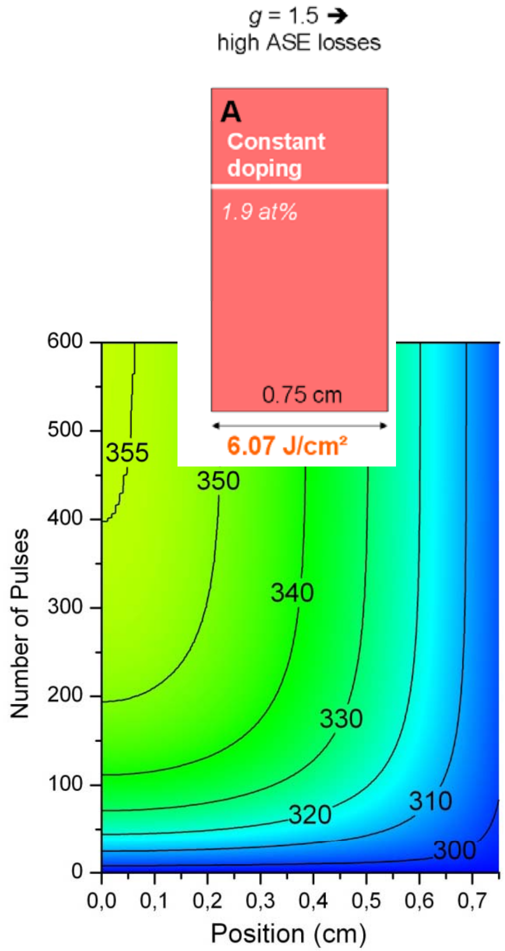


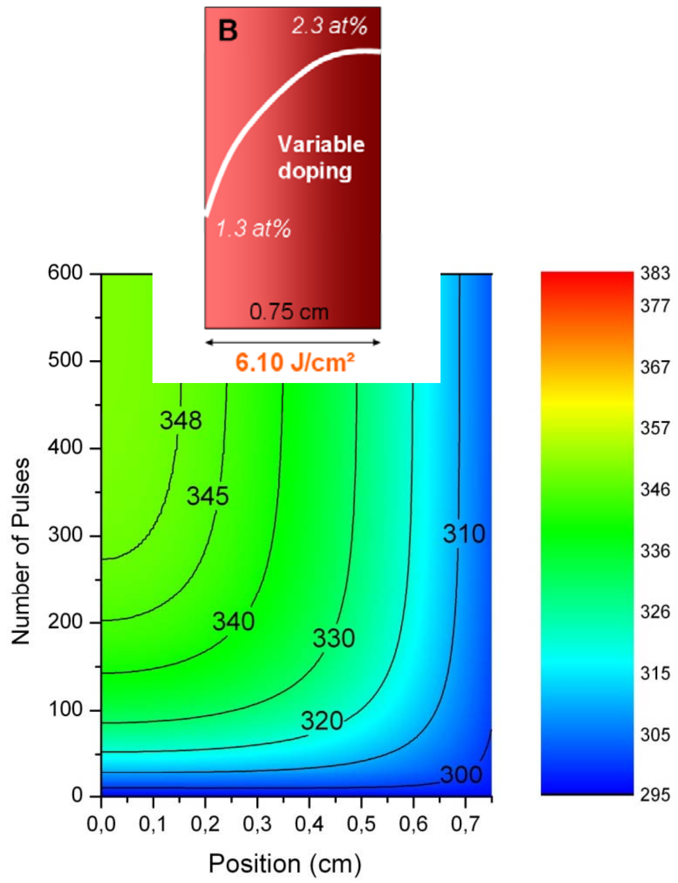
Pumping-cooling sketch used in our model
and
calculation parameters

Impact on thermal management

Axial temperature distribution evolution over 1min at 10hz





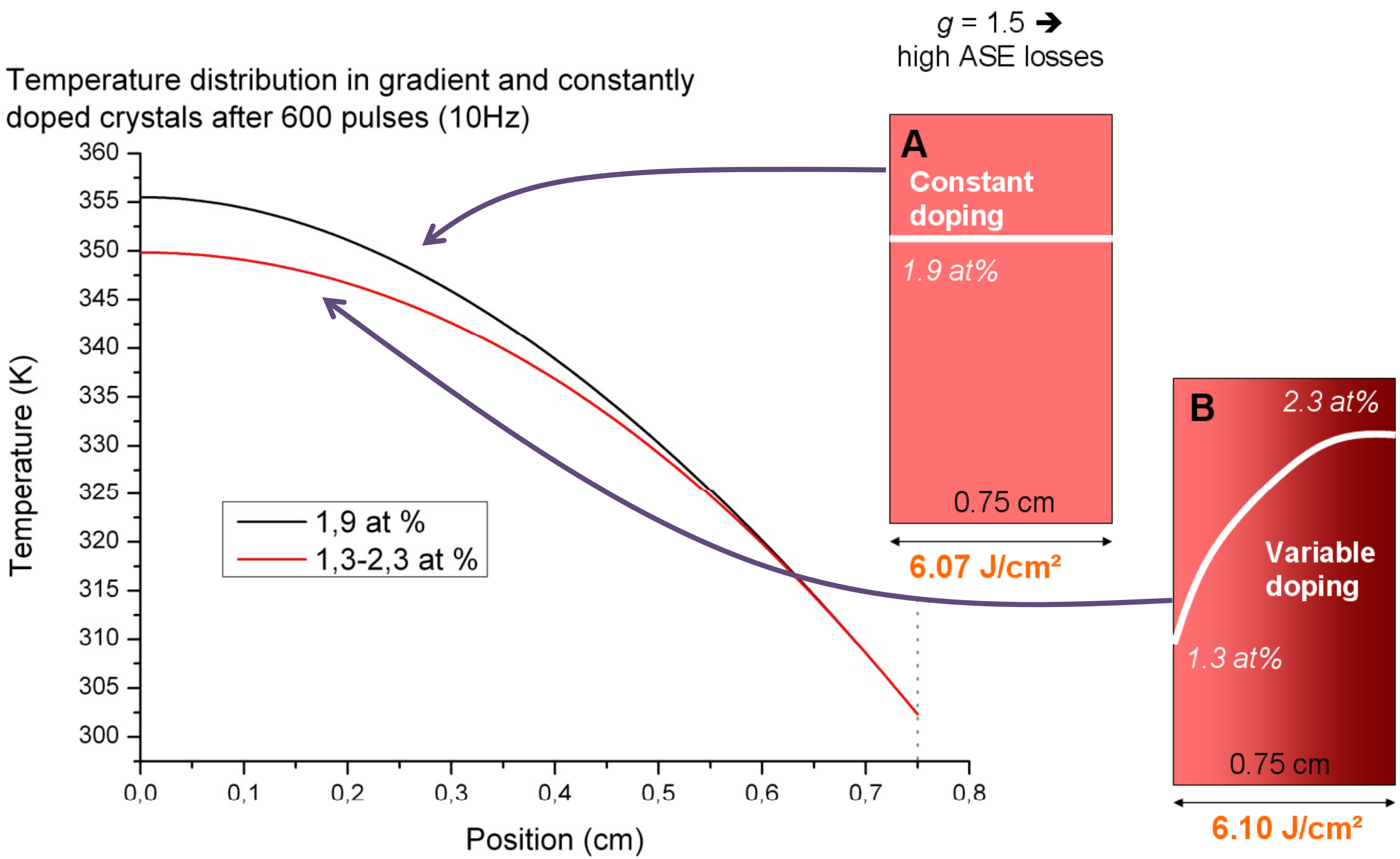


<u>1.3 at % Constant doping level</u>
<u>1.15cm crystal:</u> Max temp. is **383** K and continues rising after 1min at 10 Hz operation

<u>1.9 at % Constant doping level</u>
<u>0.75cm crystal:</u> Max temp. is **357** K not fully stabilized after 1min at 10 Hz operation

<u>Variable 1.3-2.3 at % doping level</u>
<u>0.75cm crystal:</u> Max temp. is **349** K with thermal equilibrium after 1 min at 10 Hz

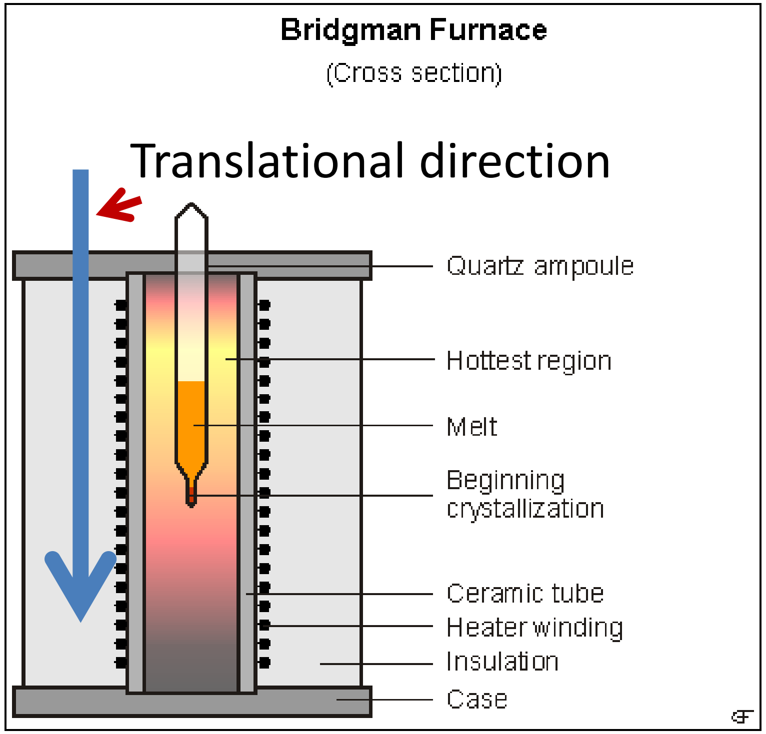
Impact on thermal management



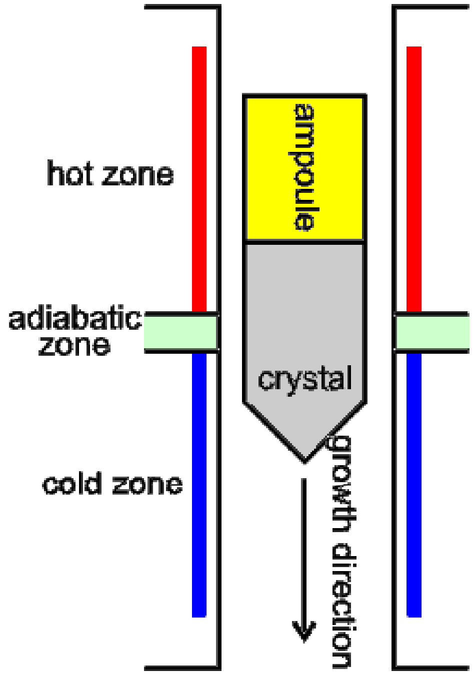
Besides experiencing much less ASE losses the gradient doped crystal also exhibit a slightly better thermal distribution

YAG growth techniques: Bridgman and Stockbarger

Starting material is loaded in a vertically moving crucible



Bridgman-Stockbarger furnace

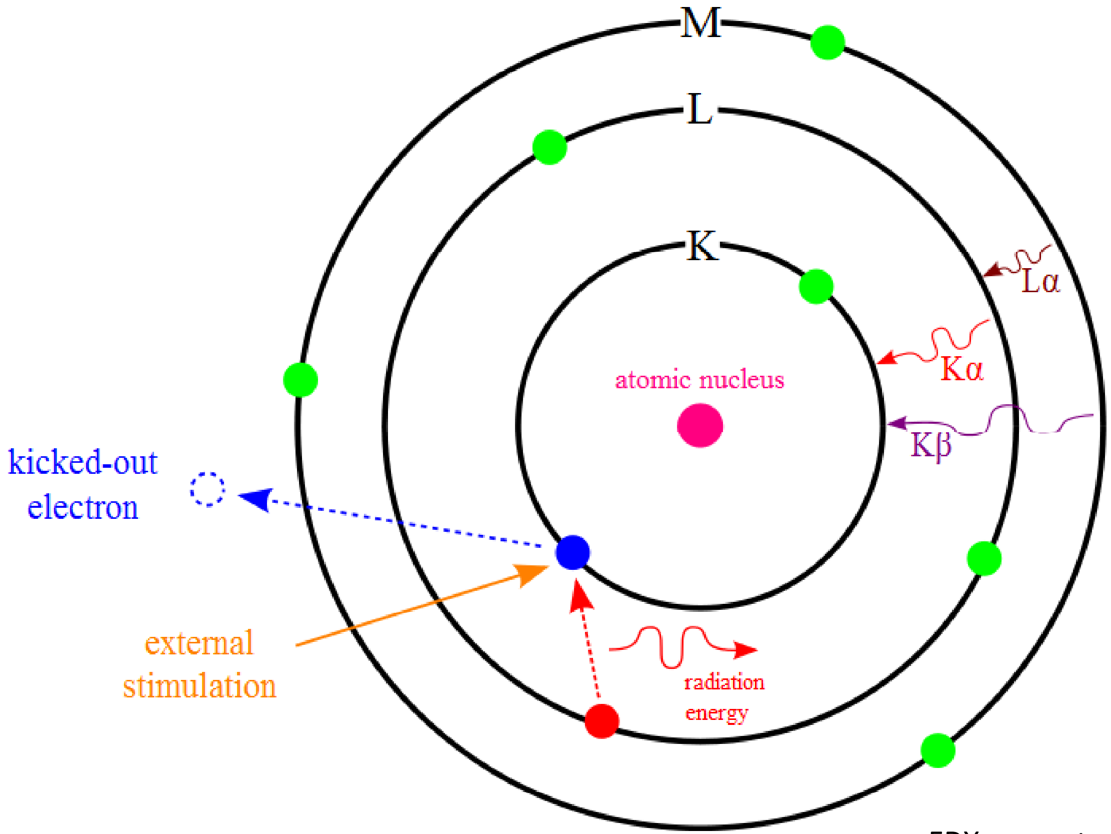


- No possibility of in-situ observation
- Mechanical interaction between the crucible and the crystal
- Technical issues for extracting the grown crystal

Yb concentration measurements : EDX

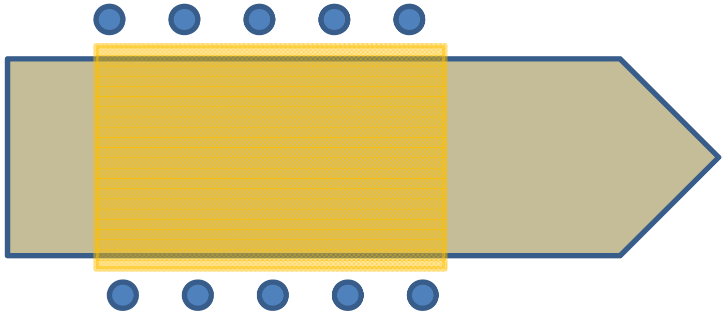
Energy Dispersive X-ray spectroscopy (EDX)

1-A high-energy electron beam is focused into the sample to stimulate the emission of characteristic X-rays from YAG
2- The composition of the specimen can be retrieved



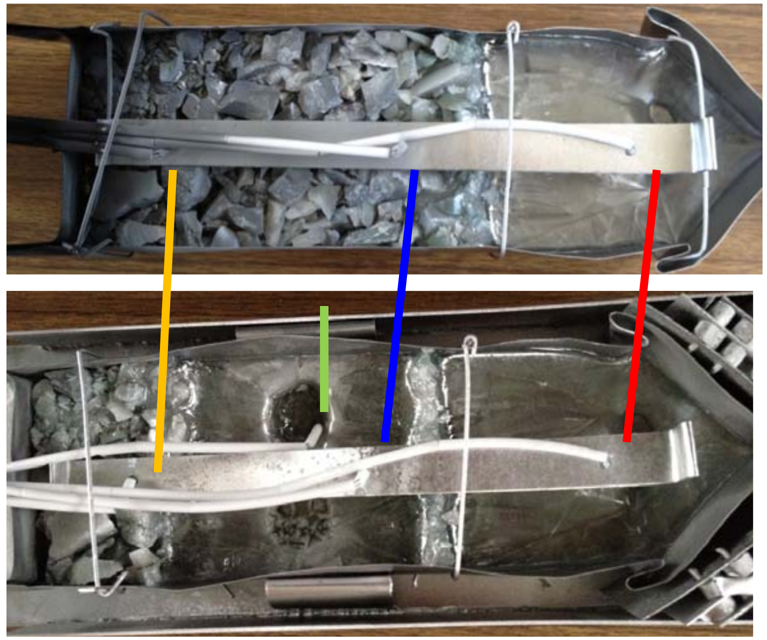
EDX apparatus used for our sample at HTSC, Armenia

2^{nd} experiment

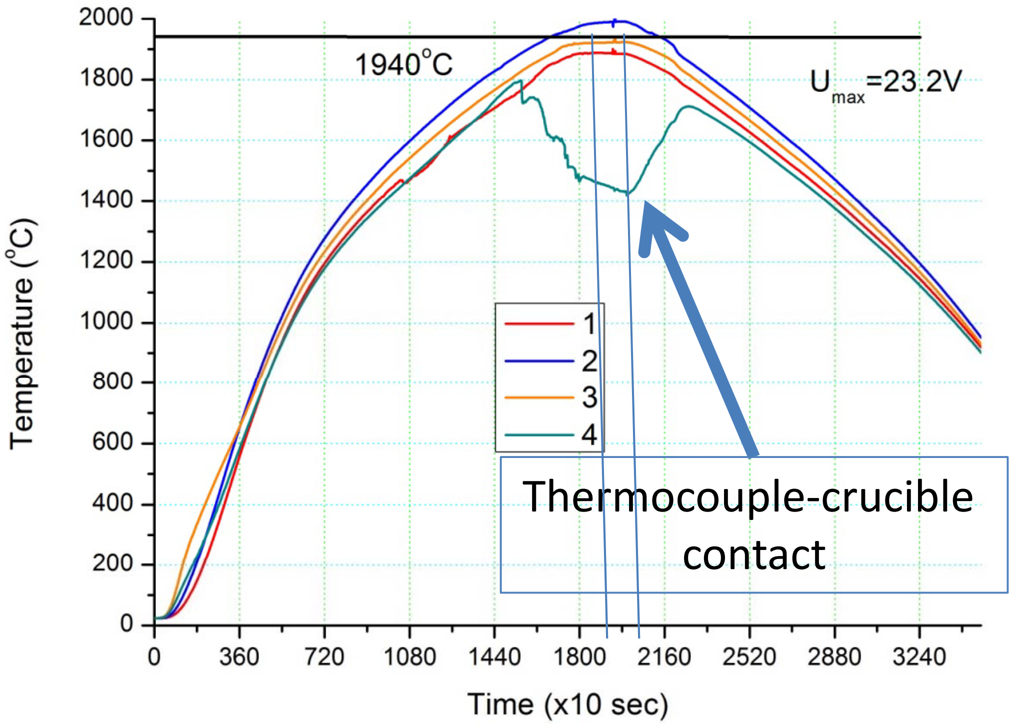


Input boule = 1^{st} experiment

Resulting boule



Observed temperature distribution



<u>Results :</u>

$T_{1max} = 1887^0C$

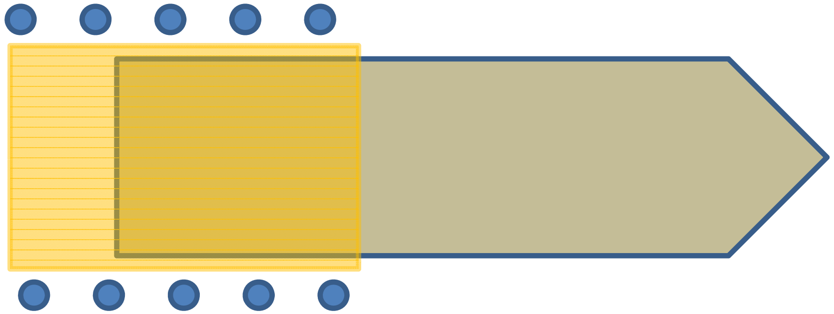
$T_{2max} = 1989^0C$

$T_{3max} = 1921^0C$

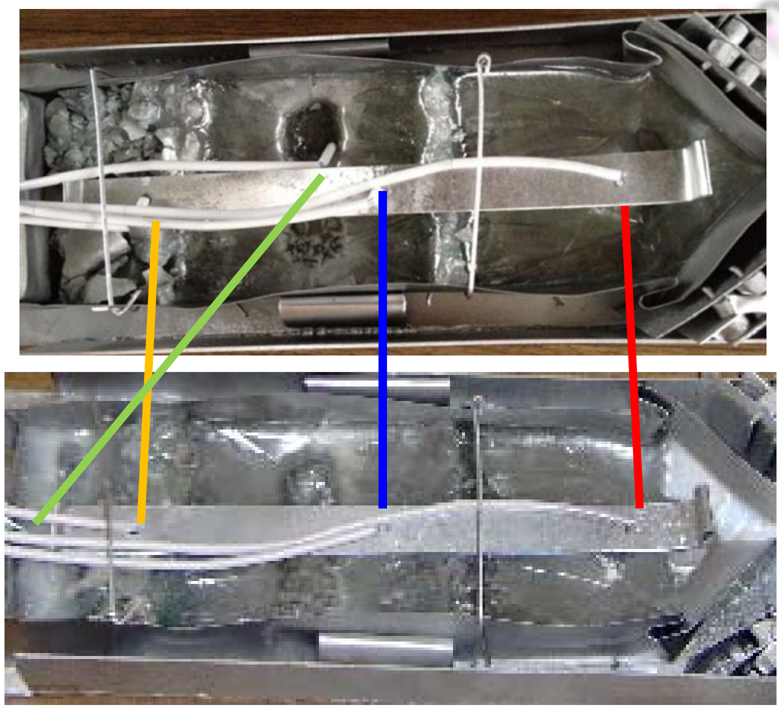
$T_{1max} - T_{2max} = -102^0C$

$T_{2max} - T_{3max} = 78^0C$

3^{rd} experiment

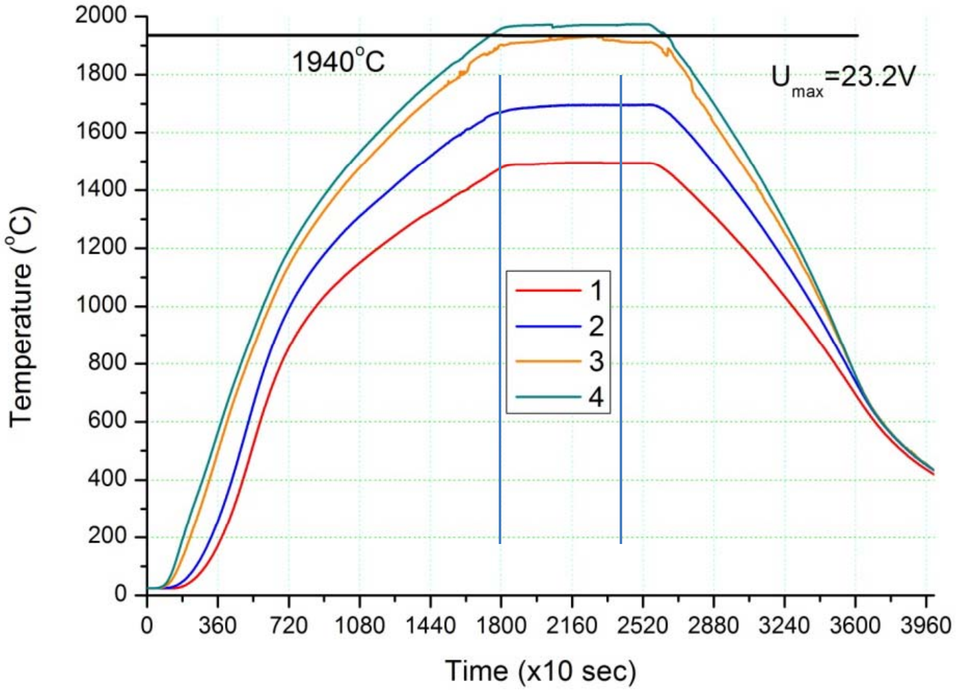


Input boule



Resulting boule

Observed temperature distribution



Conclusion :

T_{1max} = 1495^0C

T_{2max} = 1696^0C

T_{3max} = 1930^0C

T_{4max} = 1980^0C

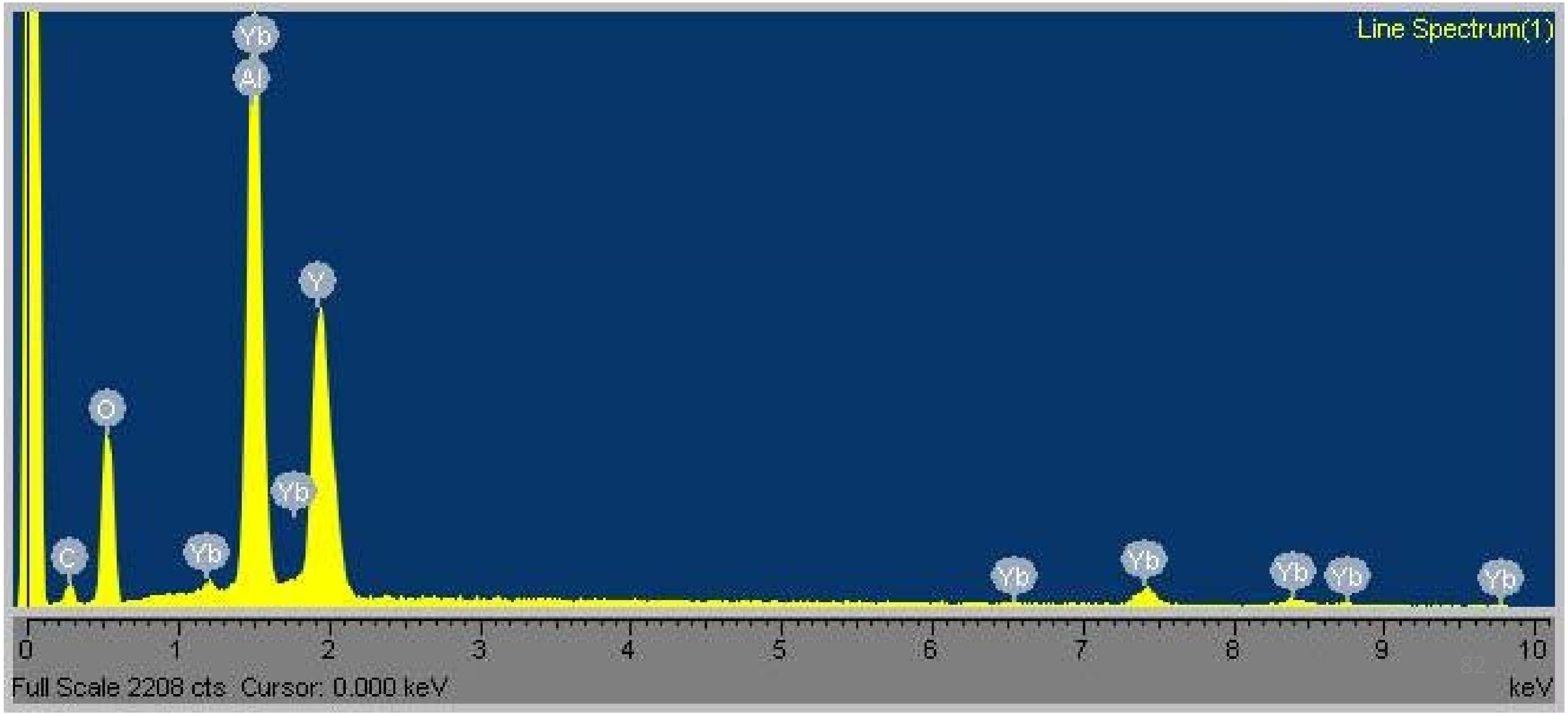
T_{1max} - T_{2max} =-201^0C

T_{2max} - T_{3max} =-234^0C

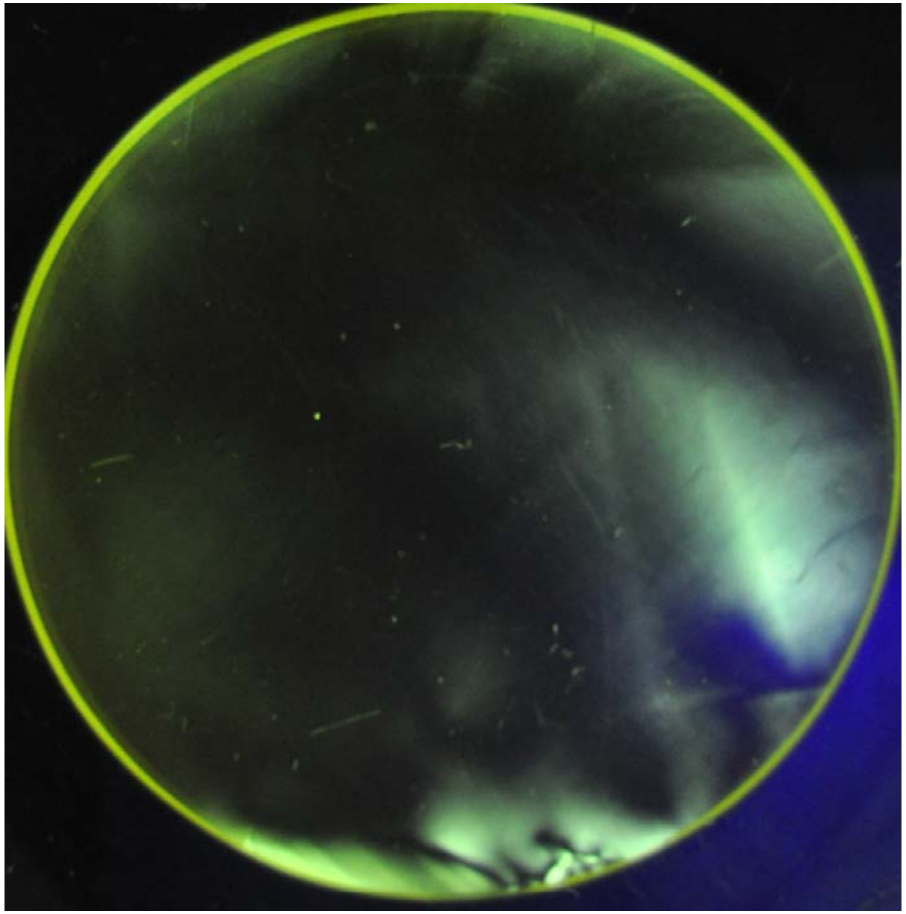
T_{4max} - T_{3max} =60^0C

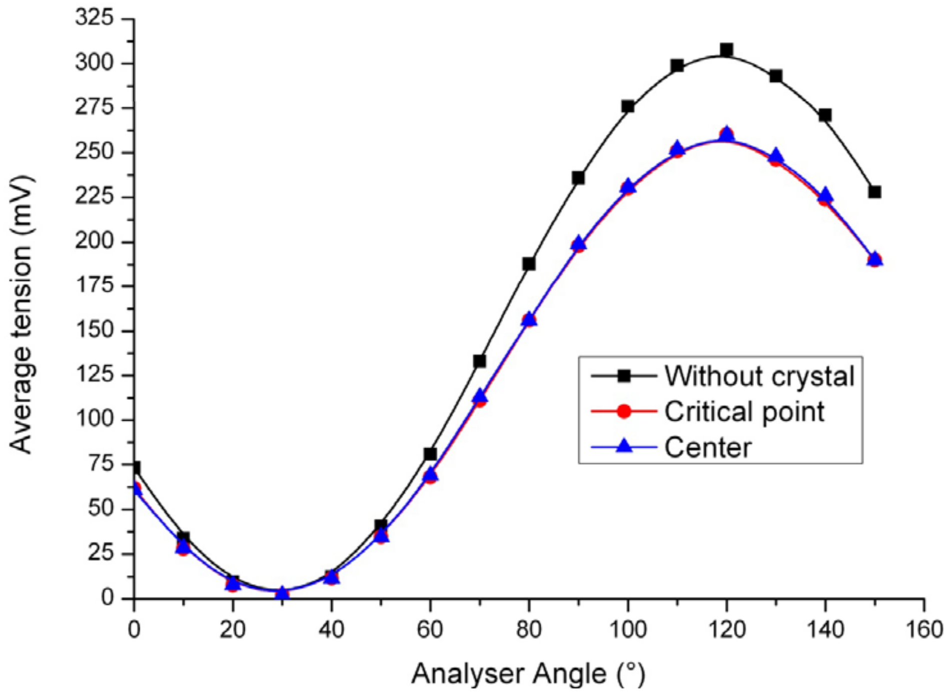
Doping level measurement for the 90mm crystal

Energy-Dispersive X-Ray Analysis
0.39+/-0.17 at% of Yb
No Ce^{3+} detected



Depolarization in the crystals





Stresses are concentrated mainly near the periphery, which is also the area of Interaction of the crystal and molybdenum crucible

Outline

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Defects in the crystals

- A few inclusions are detected near the interface of boule and container with microscope analysis

- no bubbles were detected

✓ The final product crystal was cut in the part not influenced by this defects