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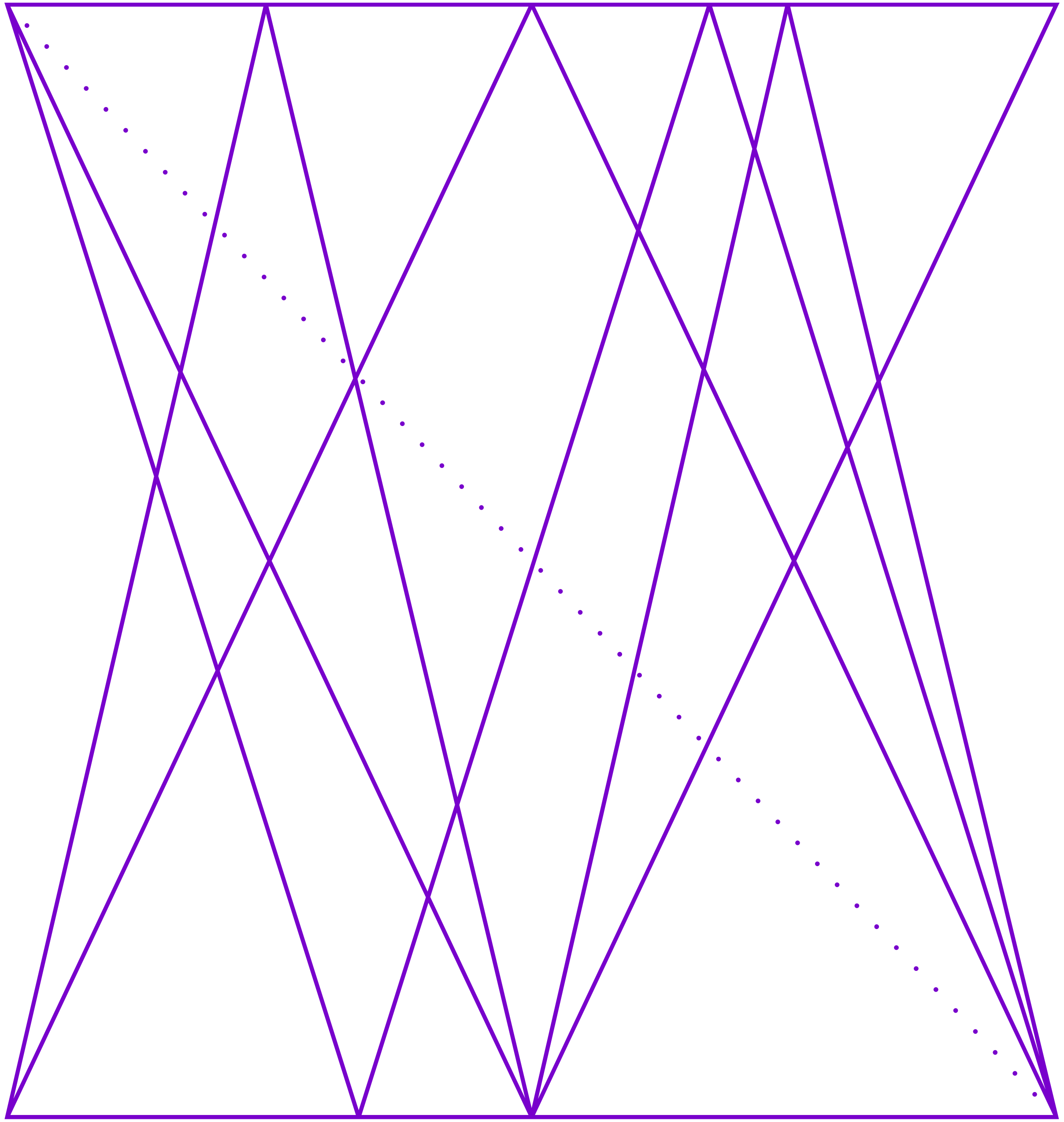
Technical Groups

Tailored Gain Materials for Visible and Infrared Lasers

Featuring Christian Kränkel, Leibniz-Institut für Kristallzüchtung
10 March 2022



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Our technical group encompasses novel laser system development for a broad range of scientific, industrial, medical, remote sensing and other directed-energy applications.

Our mission is to connect the 4400+ members of our community through technical events, webinars, networking events, and social media.

Our past activities have included:

- Webinar on Methods to Quantify Aerosol Absorption that Could Cause Laser Thermal Blooming
- Webinar on Turbulence Profile Measurement with a Dynamically Ranged Rayleigh Beacon
- Campfire Session: Breakthrough Starshot Photon Engine

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Join our online community to stay up to date on our group's activities. You also can share your ideas for technical group events or let us know if you're interested in presenting your research.

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- Email us at TGactivities@optica.org

Today’s Speaker

Dr. Christian Kränkel

Leibniz-Institut für Kristallzüchtung

Christian Kränkel studied physics and received his PhD at the Universität Hamburg, Germany with Günter Huber. After a postdoctoral stay in the group of Ursula Keller at ETH Zurich, Switzerland, he became a junior research group leader at the German excellence cluster “CUI – The Hamburg Centre for Ultrafast Imaging”. Since his habilitation in 2017 he leads the Center for Laser Materials at the Leibniz-Institut für Kristallzüchtung in Berlin, Germany and lecturer at the Humboldt Universität zu Berlin. Dr. Kränkel serves as an associate editor for Optics Express and was elected a Fellow of Optica in 2021. He is the author of more than 100 scientific papers, 300 conference contributions, and 2 book chapters. Dr. Kränkel’s research interests include the development, growth and characterization of rare-earth and transition-metal doped laser crystals for lasers with emission wavelengths from the UV to the mid-infrared spectral range in continuous wave, Q-switched and mode-locked operation.

Tailored gain materials for visible and infrared lasers

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Moritz Badtke, Elena Castellano Hernandez, Patty Eckhof, Steffen Ganschow,
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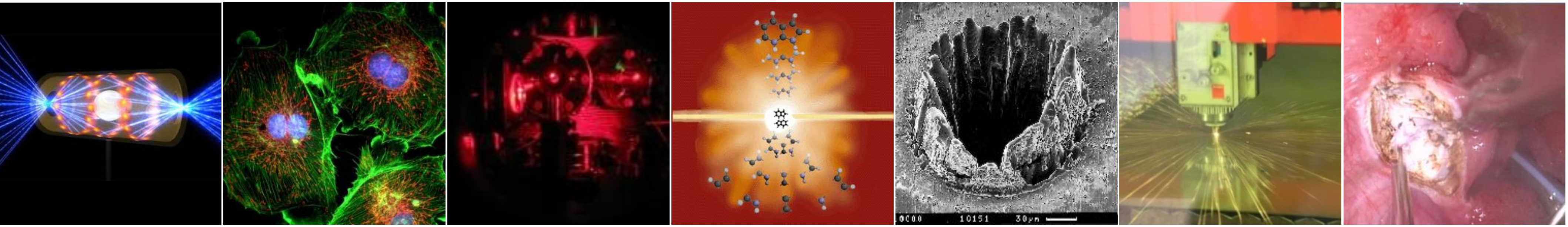
Berlin, March 10th, 2022

Outline

- **Introduction**
- Sesquioxides for high power mid-infrared lasers
- Pr^{3+} and Tb^{3+}-doped materials for visible lasers
- Conclusion

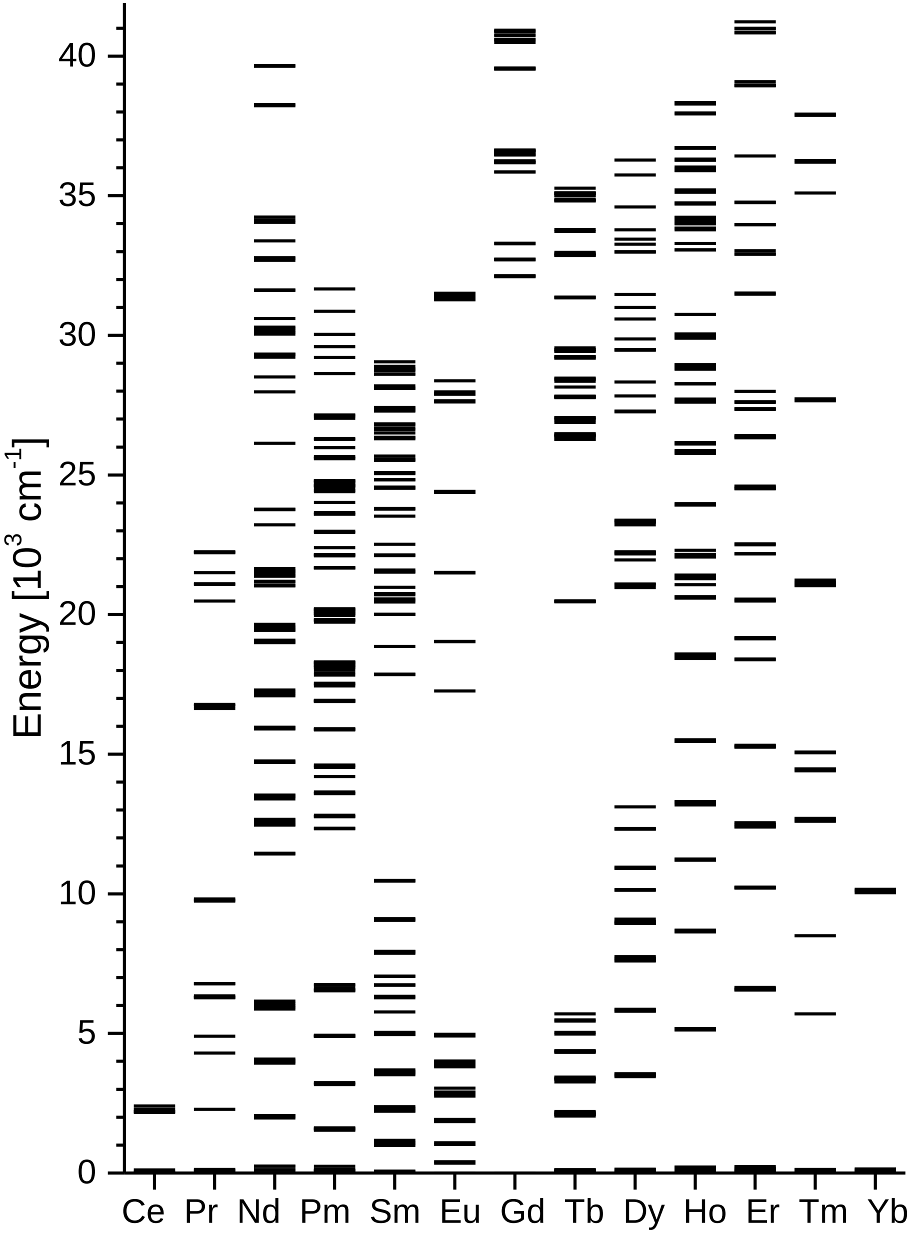
Motivation

Applications of lasers

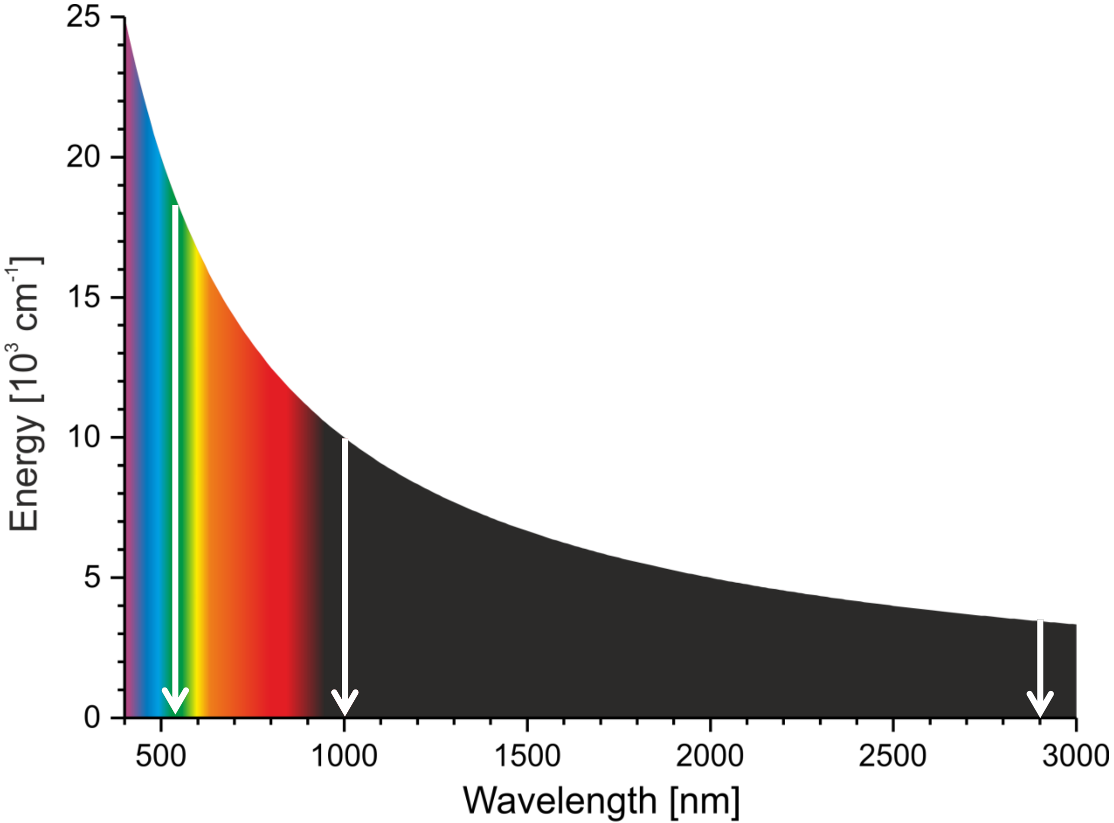


Motivation

Diecke diagram



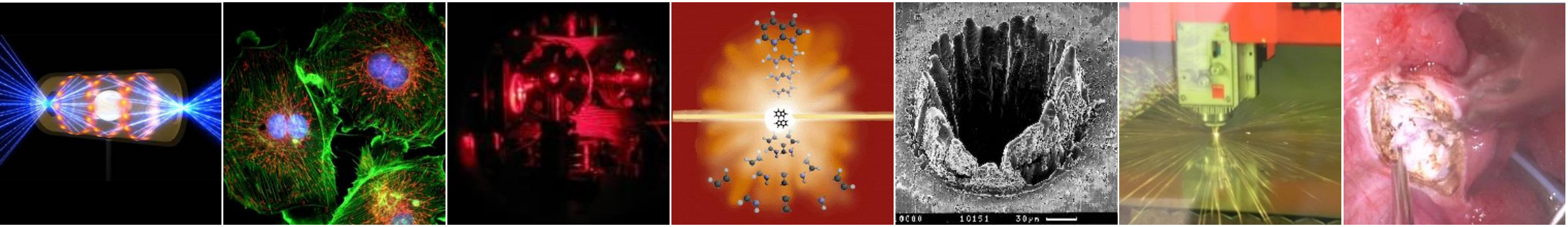
- *4f* energy levels of all trivalent rare earth ions in $LaCl_3$
- *4f* shell shielded by 5*s* and 5*d* shells: characteristic spectroscopic fingerprint for each rare earth ion

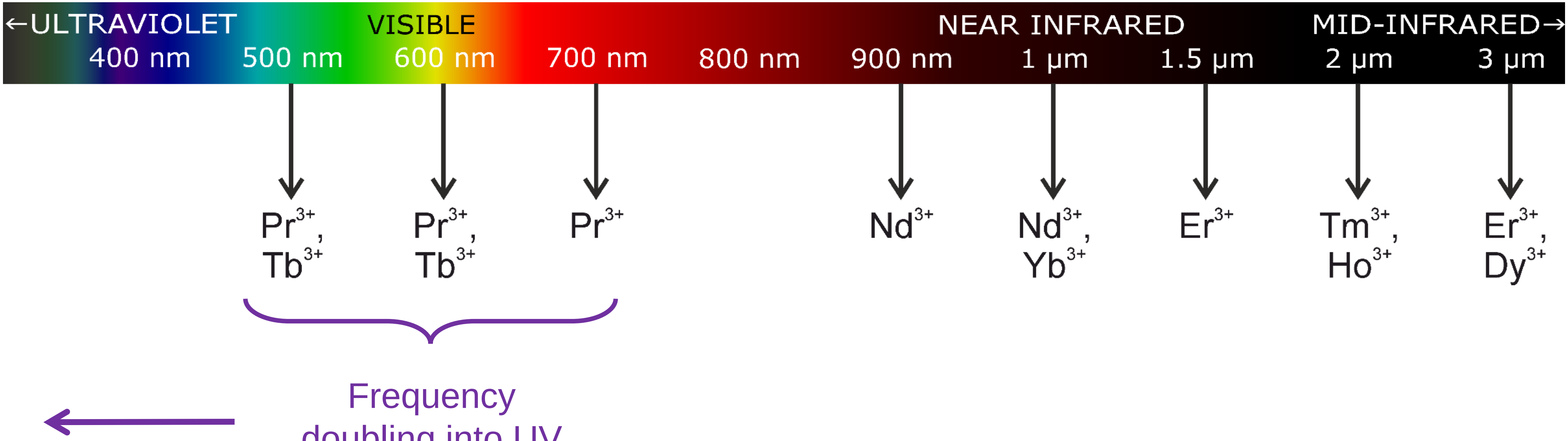


Dieke, *Spectra and Energy Levels of Rare Earth Ions in Crystals,* John Wiley & Sons (1968)

Motivation

Applications of lasers





Pr^{3+}, Tb^{3+}

Pr^{3+}, Tb^{3+}

Pr^{3+}

Nd^{3+}

Nd^{3+}, Yb^{3+}

Er^{3+}

Tm^{3+}, Ho^{3+}

Er^{3+}, Dy^{3+}

Frequency doubling into UV

Tailored host materials

Influence of the host material on the laser properties

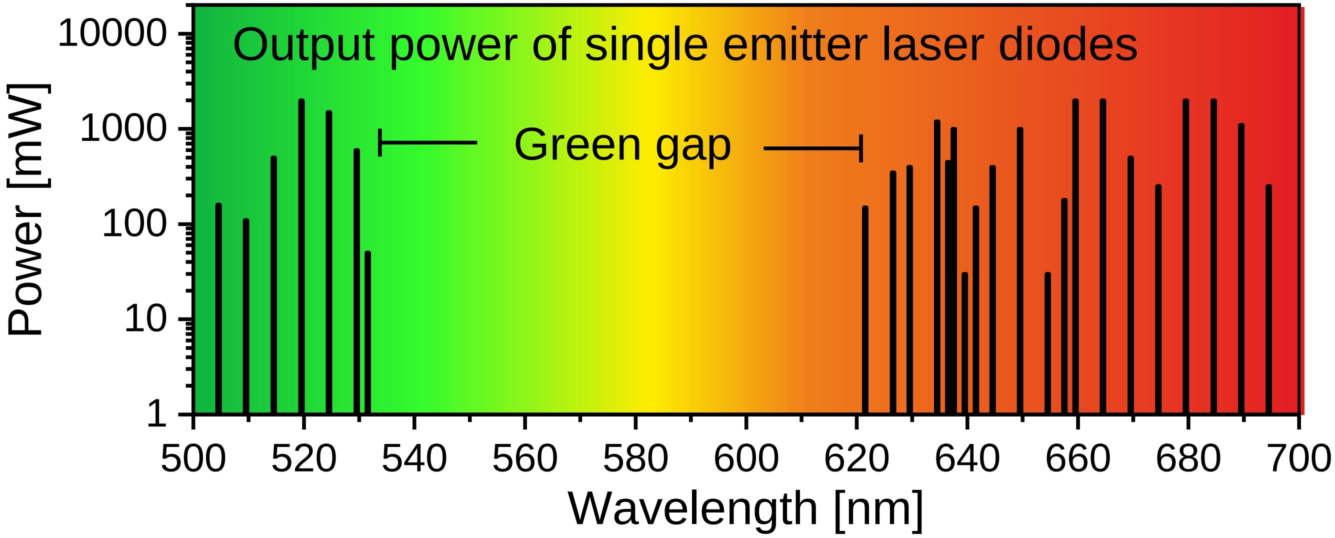
Requirement for the laser	Requirement for the host material
Parity forbidden 4*f*-4*f*-transitions	Acentric site symmetry
Non-radiative decay of mid-IR transitions	Low phonon energy host materials
ESA for high energy visible photons	Weak crystal field, high band-gap energy
Broad gain spectrum for fs-pulses	Disordered host or multiple RE^{3+}-sites
Thermal management of high power lasers	Good thermal conductivity
Need for particular laser wavelength	Crystal field shifts emission to required value

Some of these properties can be ‘tailored’
by a proper choice or composition of the host material

Outline

- Introduction
- **Pr^{3+} and Tb^{3+}-doped materials for visible lasers**
- Sesquioxides for high power mid-infrared lasers
- Conclusion

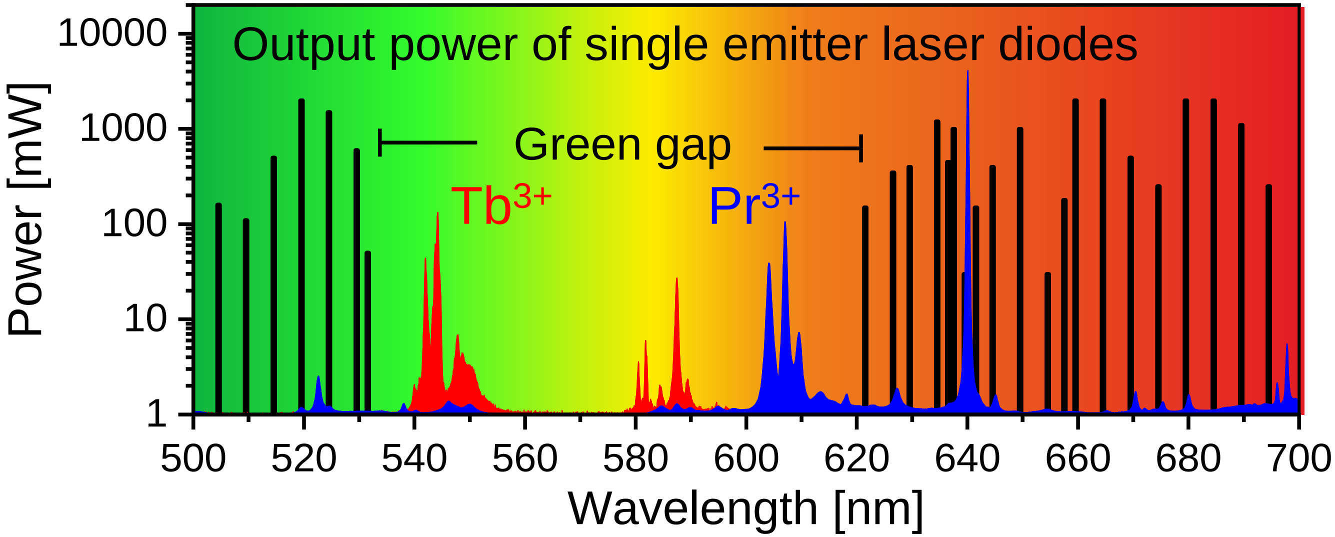
Wavelength coverage of semiconductor lasers



- (In,Ga)N-based laser diodes are commercially available up to ~535 nm
- (Al,Ga,In)P-based laser diodes are available down to ~620 nm

} **"Green gap"**

Wavelength coverage of semiconductor lasers



- (In,Ga)N-based laser diodes are commercially available up to ~535 nm
- (Al,Ga,In)P-based laser diodes are available down to ~620 nm

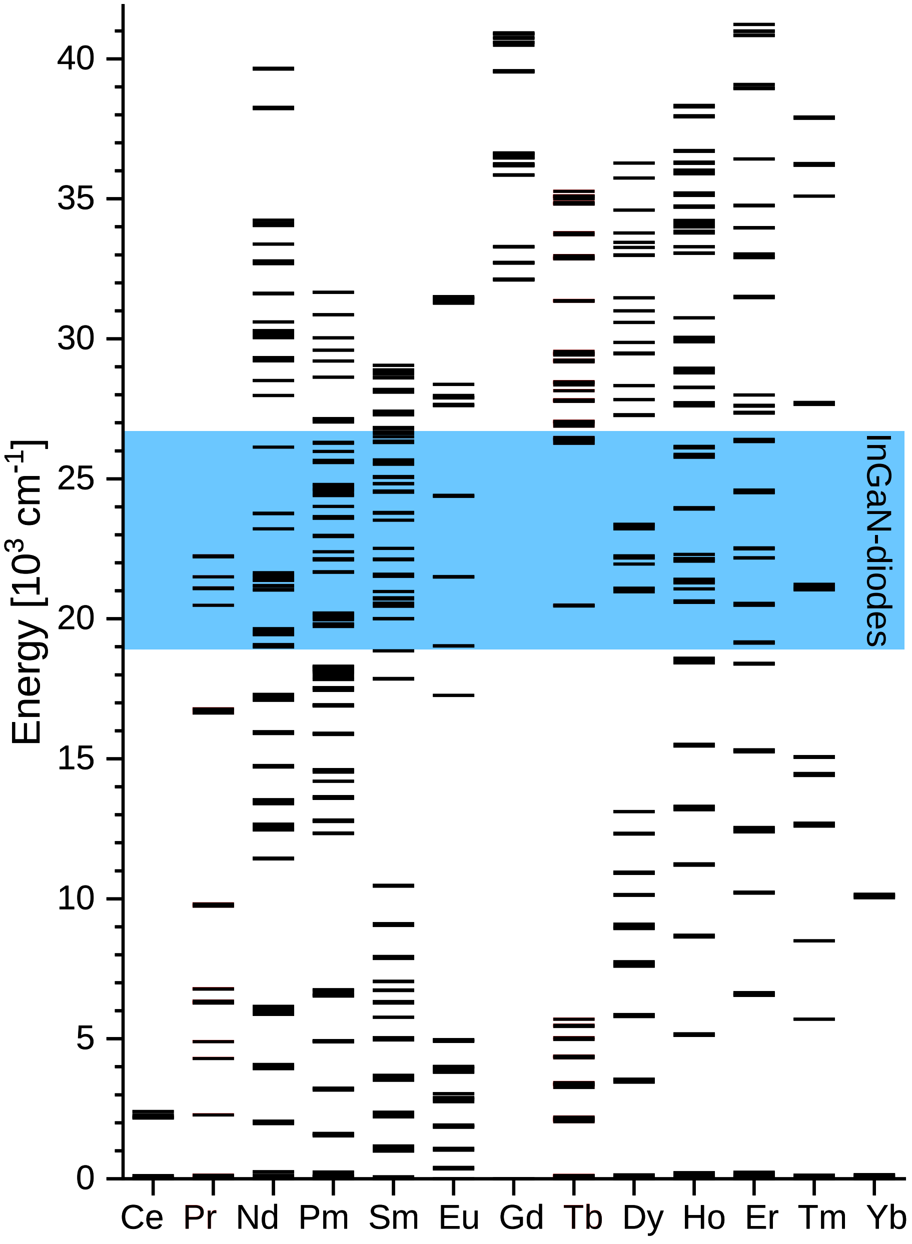
} **“Green gap”**

- Some rare earth ions provide prominent emission lines well within this gap

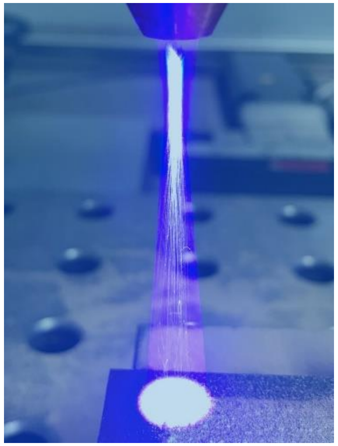
→ **(In,Ga)N-diode pumped solid-state lasers to fill the “green gap”**

Pump sources for visible lasers

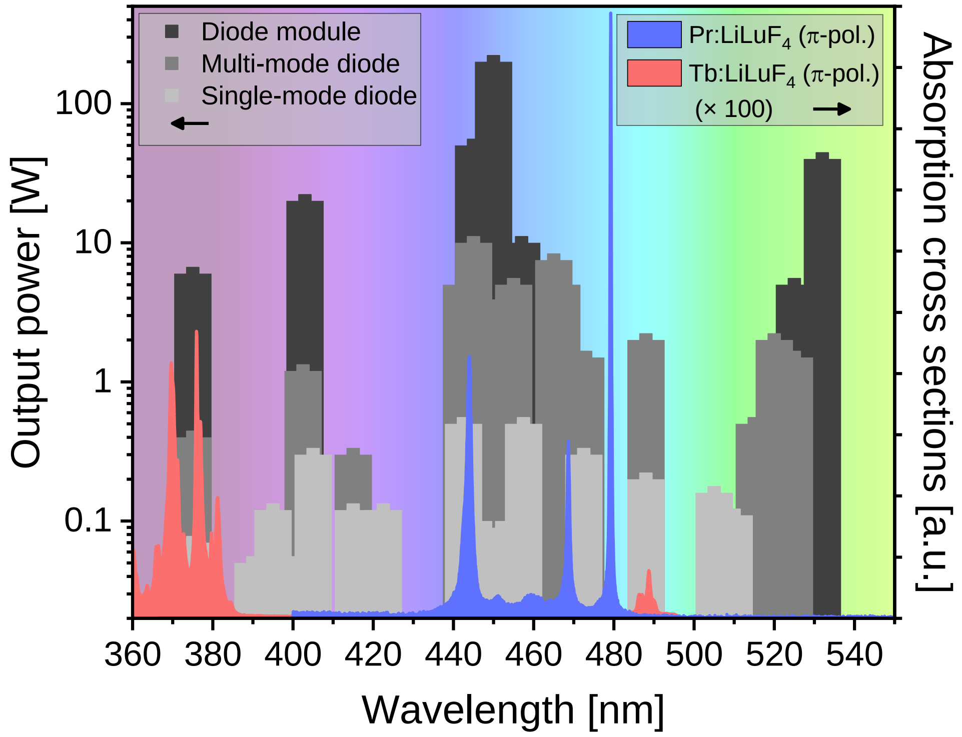
The rise of (In,Ga)N-based laser diodes



- Direct visible lasing requires short-wavelength pump sources
- Blue emitting laser diode modules are available with kW output power
- Required pump wavelengths: 444 nm or 480 nm for Pr^{3+} 485 nm or 370 nm for Tb^{3+}

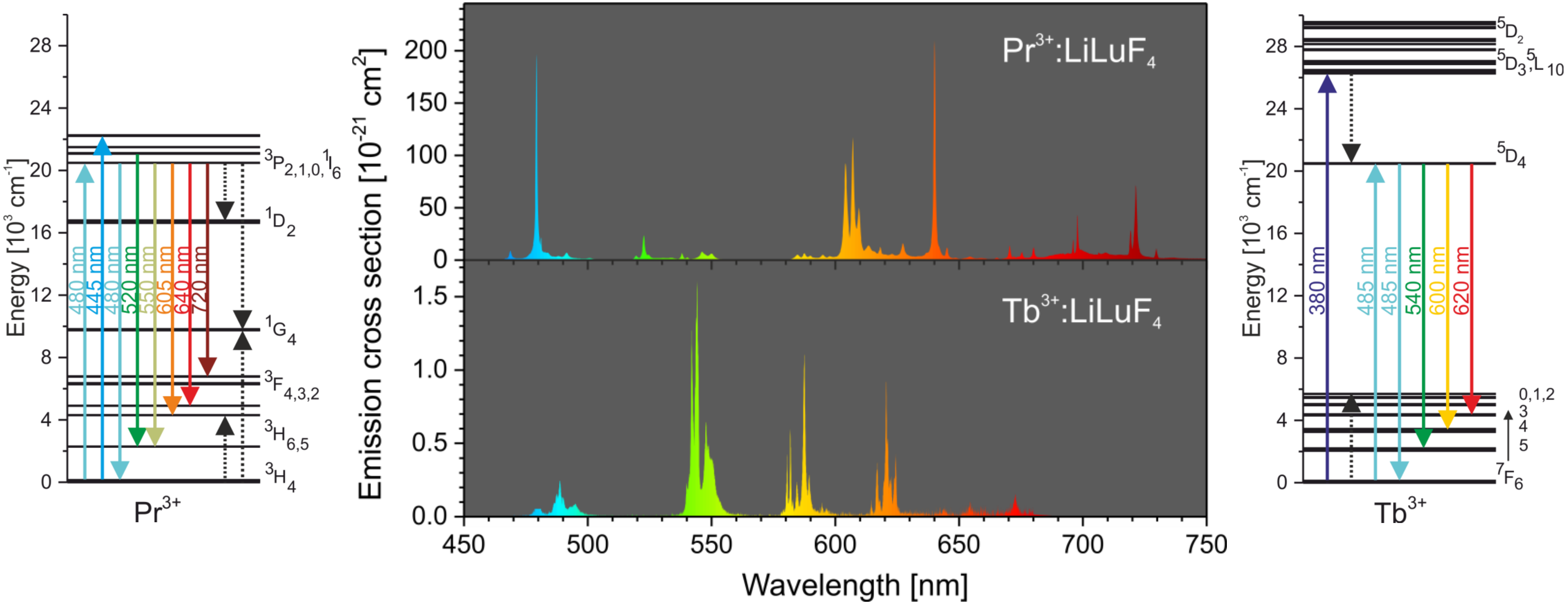


https://www.laserline.com/en-int/ldm-blue-diode-laser/

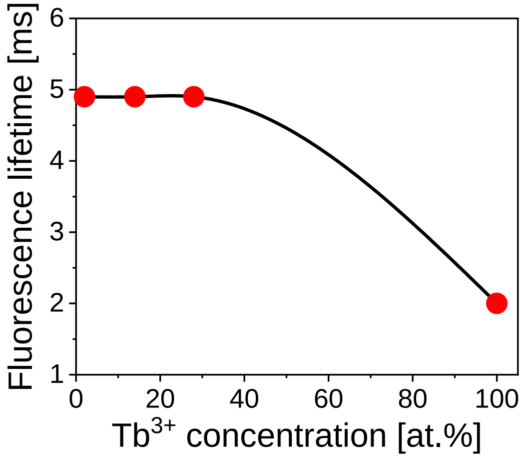


Active ions for visible lasing

Spectroscopic properties of Pr^{3+} and Tb^{3+}

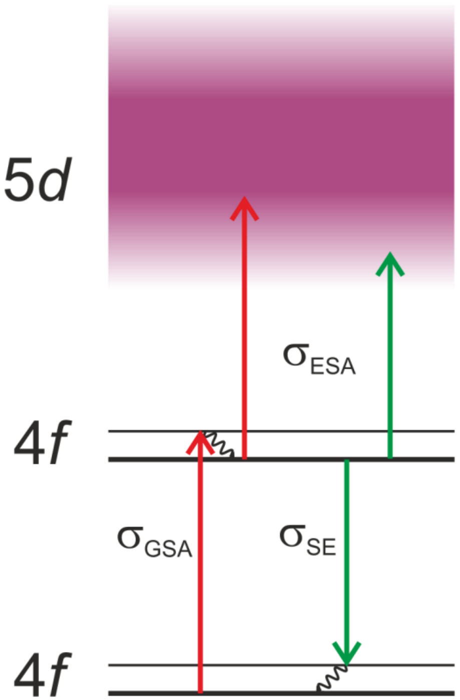


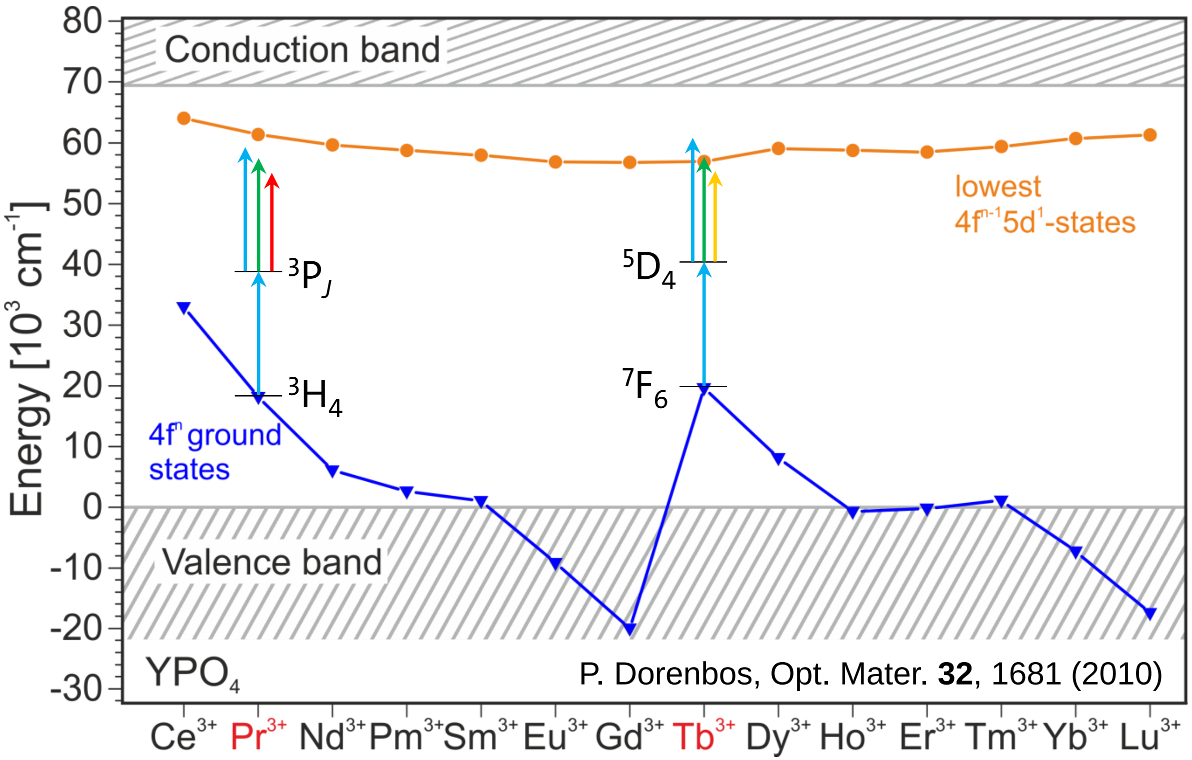
- Emission lines throughout the whole visible range for both ions
- Weak Tb^{3+}-emission compensated by high doping concentrations: No lifetime quenching up to at least 40% doping (Pr: 0.5%!)



Interconfigurational 4*f*-5*d* excited state absorption

- Visible lasers have high photon energies → Risk for parity allowed $4f^7 5d^1$-ESA
- Risk is in particular high for the most relevant ions Pr^{3+} and Tb^{3+}



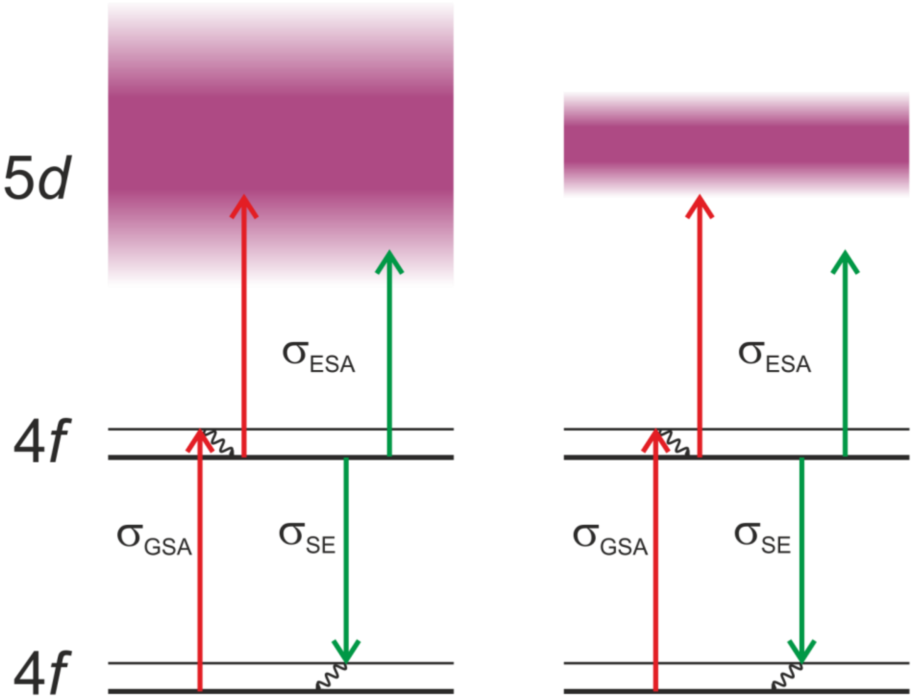


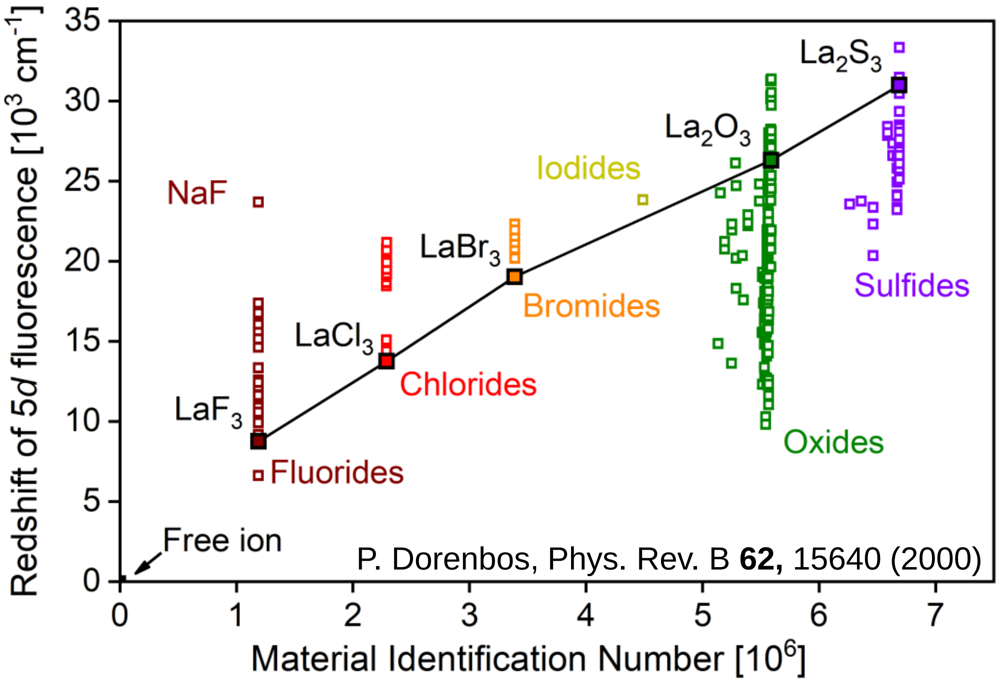
P. Dorenbos, Opt. Mater. **32**, 1681 (2010)

Host materials for visible lasers

Interconfigurational 4*f*-5*d* excited state absorption

- Visible lasers have high photon energies → Risk for parity allowed $4f^7 5d^1$-ESA
- Risk is in particular high for the most relevant ions Pr^{3+} and Tb^{3+}
- Requirement for the host material:
  - Low splitting and/or high energetic position of $4f^{n-1}5d^n$ energy bands
- **Fluorides are most suitable hosts**

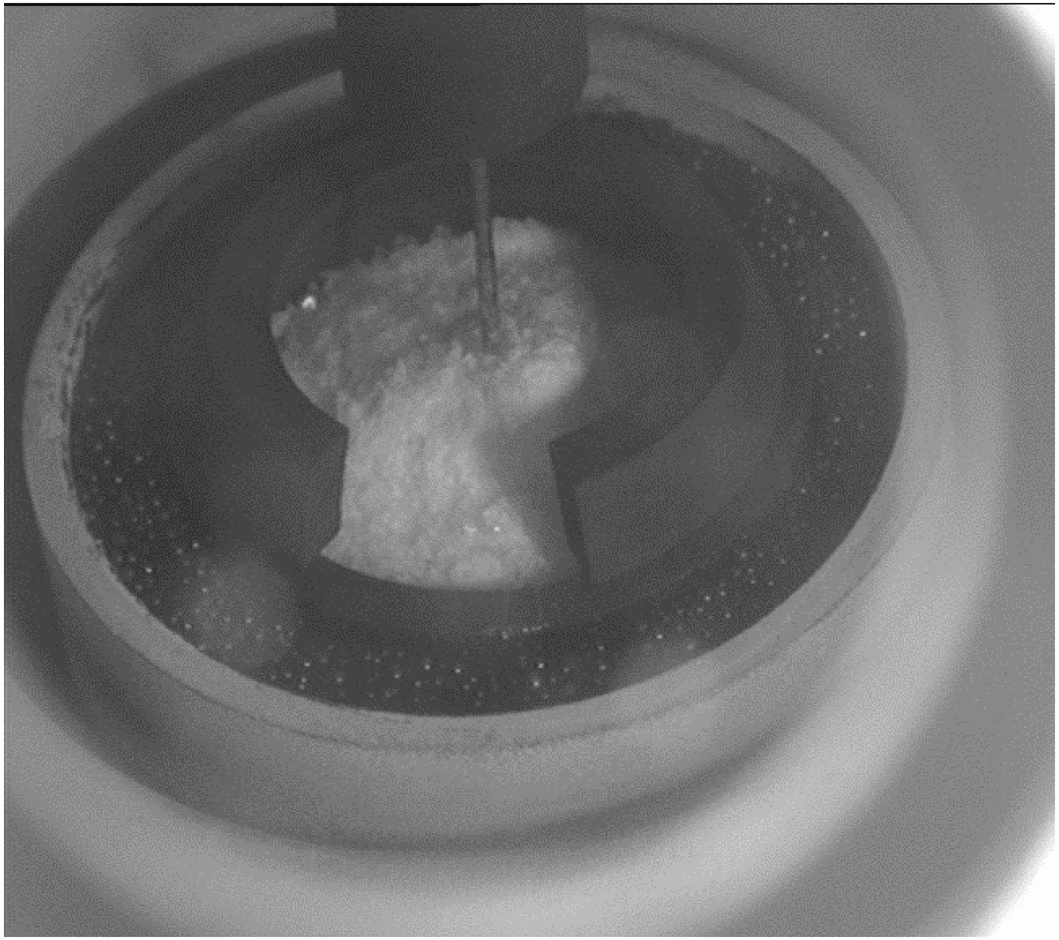




P. Dorenbos, Phys. Rev. B **62,** 15640 (2000)

Czochralski growth of laser crystals

- Most common growth method for laser crystals
- Typical growth velocities of mm/h → 10 cm long crystal needs ~4 days
- Challenge for fluoride crystals: oxide free atmosphere
- Challenge for oxide crystals: melting points up to 2200°C



Time-lapse video of fluoride growth (original duration ≈ 1 day)

Czochralski growth of laser crystals

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- Challenge for fluoride crystals: oxide free atmosphere
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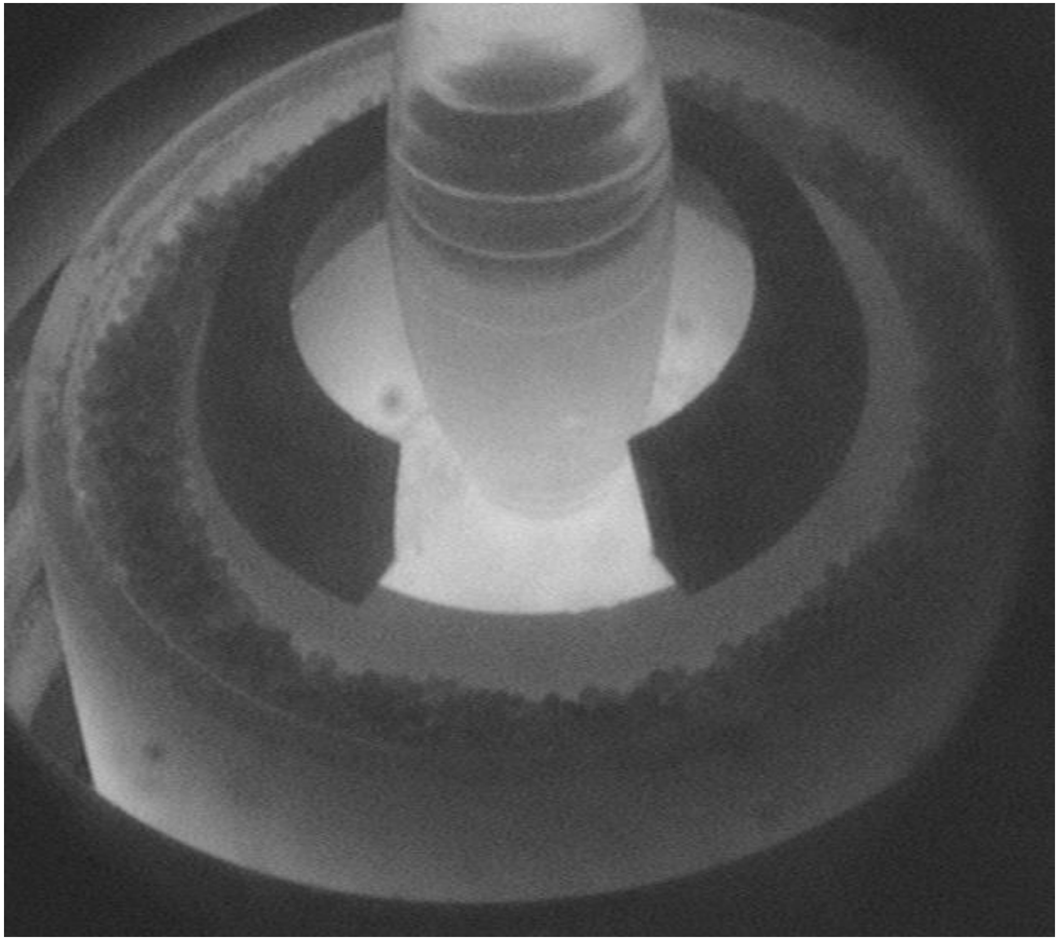
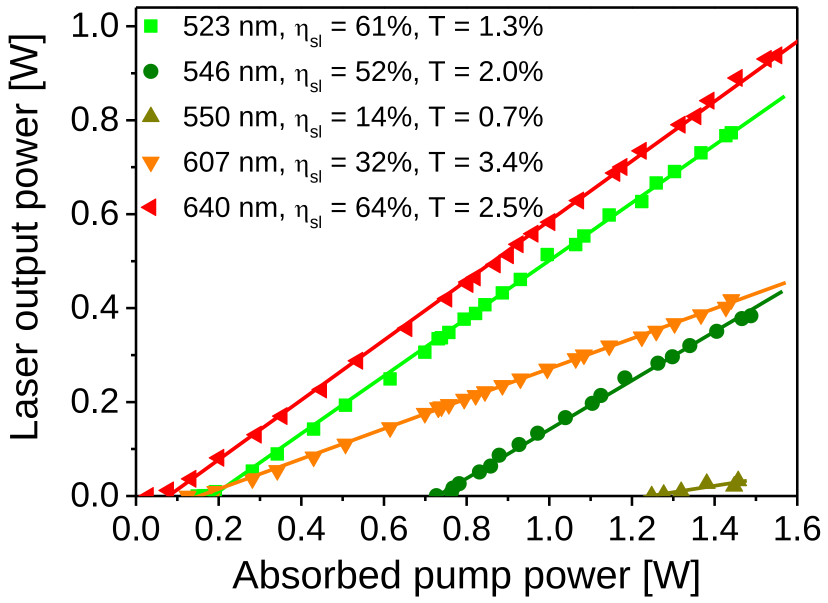
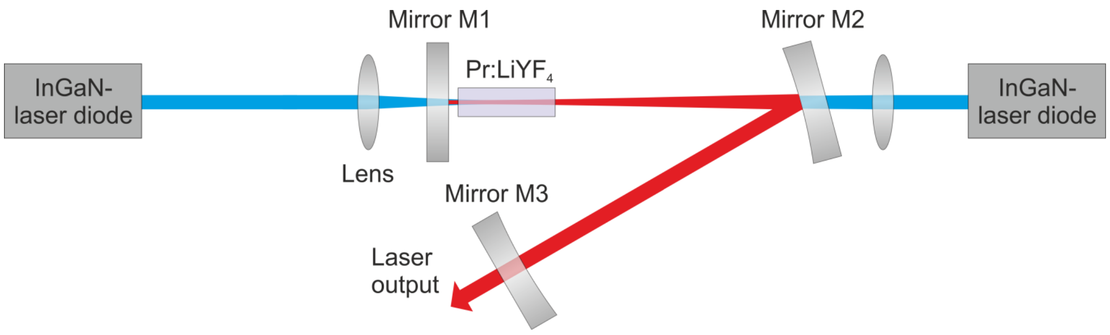


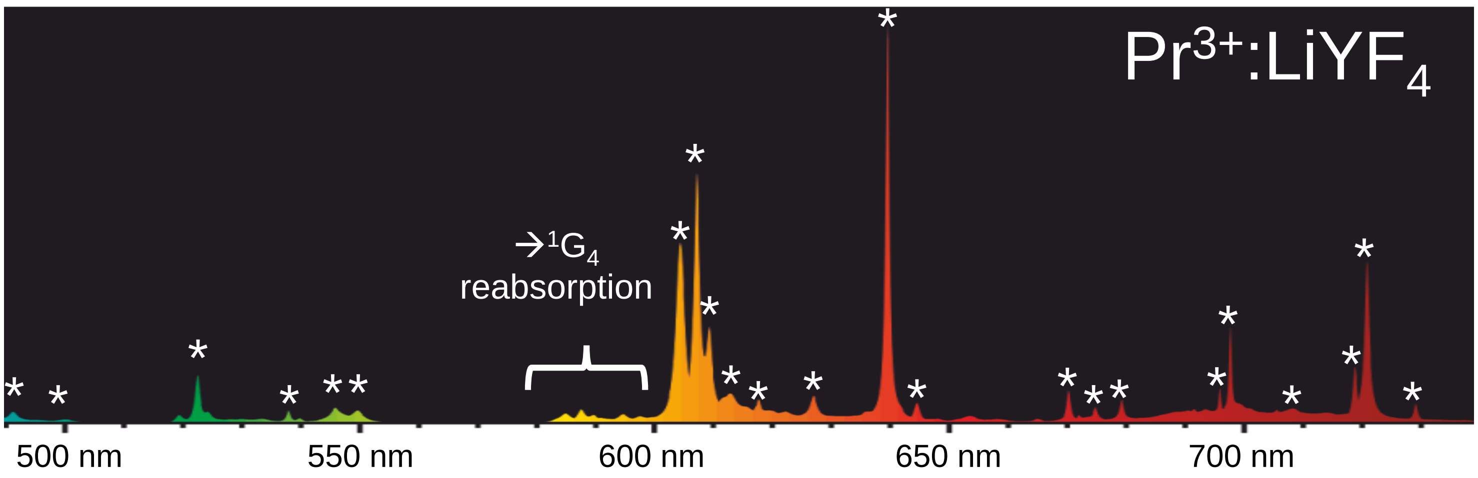
Visible Pr^{3+} lasers

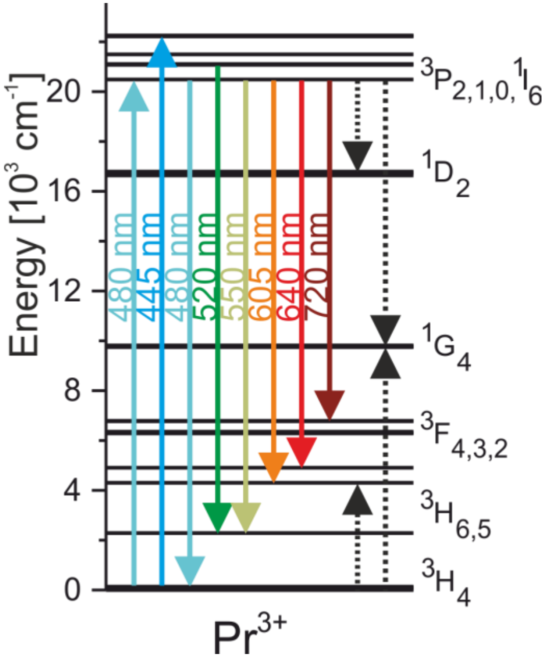
Diode pumped cw operation of $Pr^{3+}:LiYF_4$

- Pr:YLF enables efficient lasing at many visible lines
- Power scaling of fluorides is possible, but sophisticated cooling approaches are required
- **Task:** Find suitable oxide host materials with better thermal conductivity





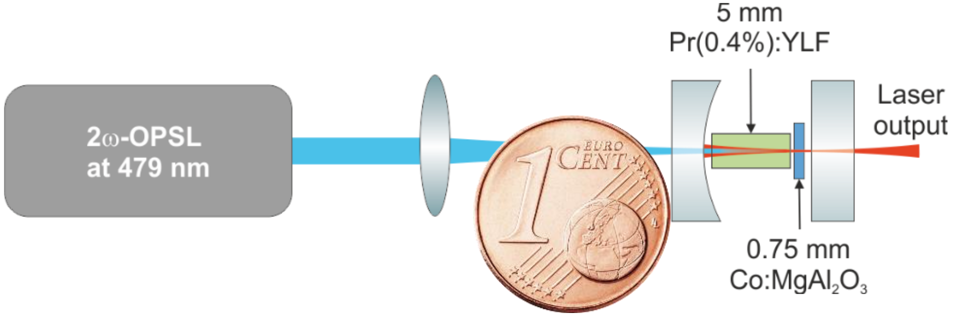


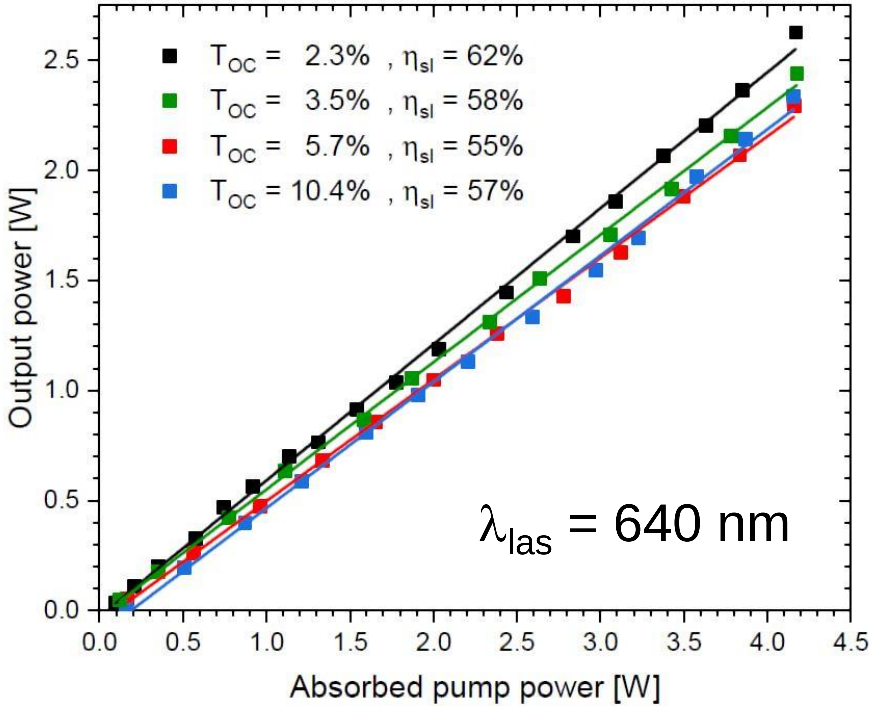


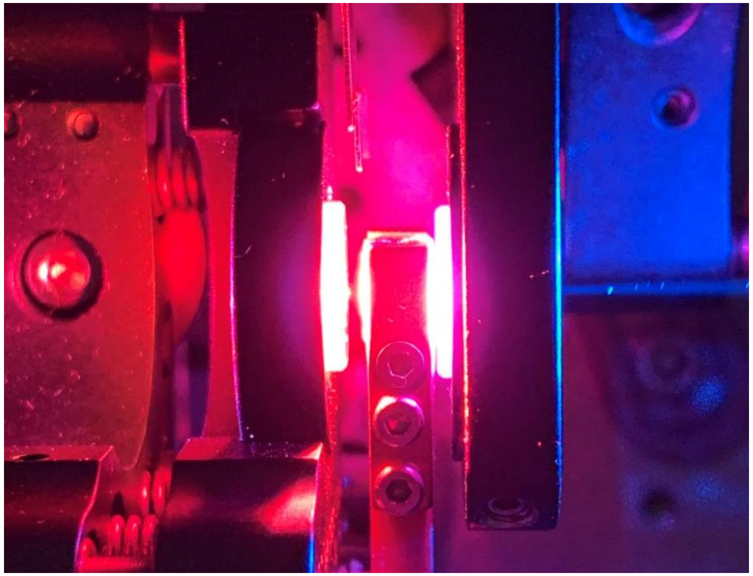
Gün *et al.*, Opt. Lett. **36** (6) 1002 (2010), Kränkel *et al.*, Las. Phot. Rev. **10** (4), 548-568 (2016)

Passively Q-switched Pr:YLF laser

- 8.5 mm short cavity
- 5 mm Pr(0.6%):YLF
- SA: 0.75 mm $Co:MgAl_2O_4$

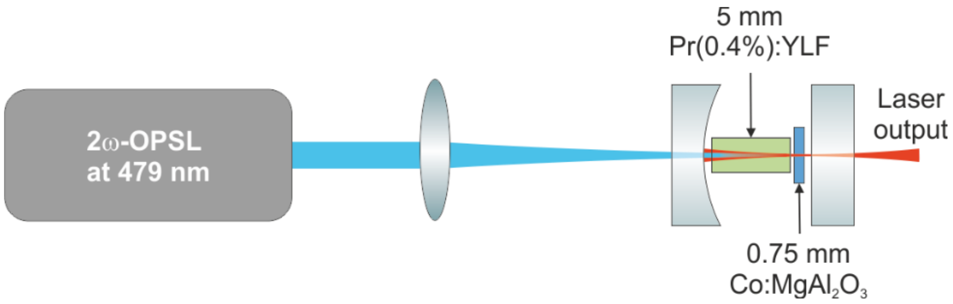






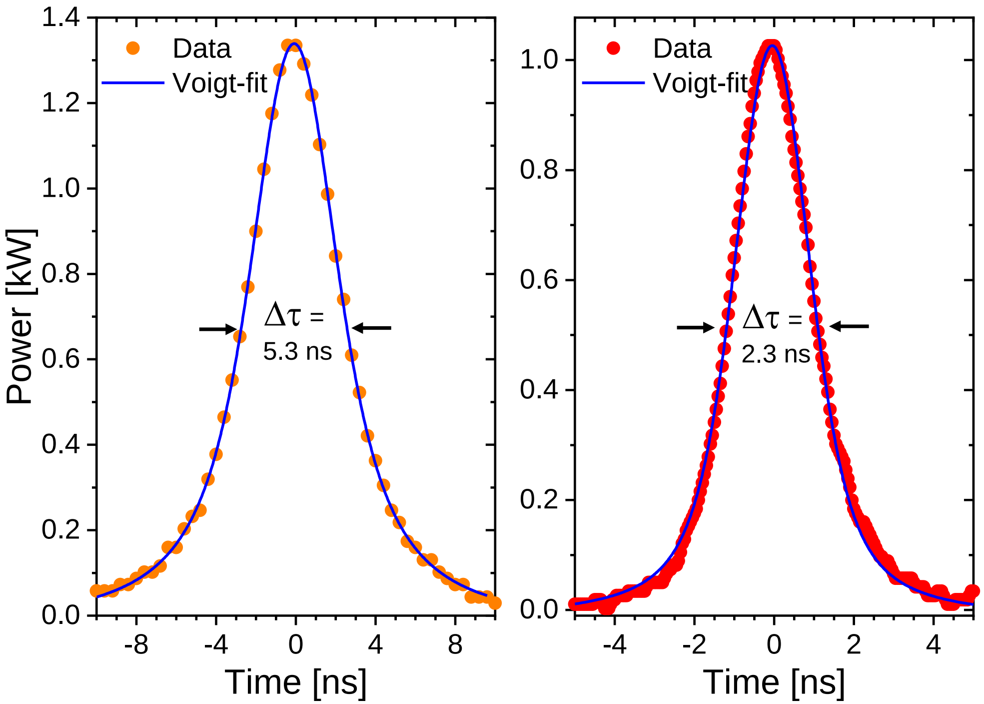
Badtke *et al.*, Appl. Phys. B **127** (6) 83 (2021)

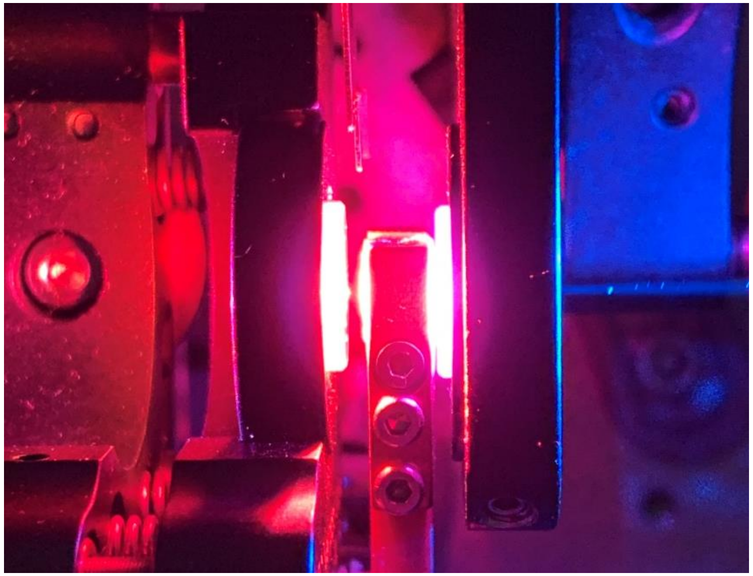
Passively Q-switched Pr:YLF laser



- 8.5 mm short cavity
- 5 mm Pr(0.6%):YLF
- SA: 0.75 mm $Co:MgAl_2O_4$

	607 nm	640 nm
τ_{pulse}	5.3 ns	2.3 ns
f_{rep}	48 kHz	98 kHz
P_{av}	0.34 W	0.29 W
P_{peak}	1.3 kW	1.0 kW

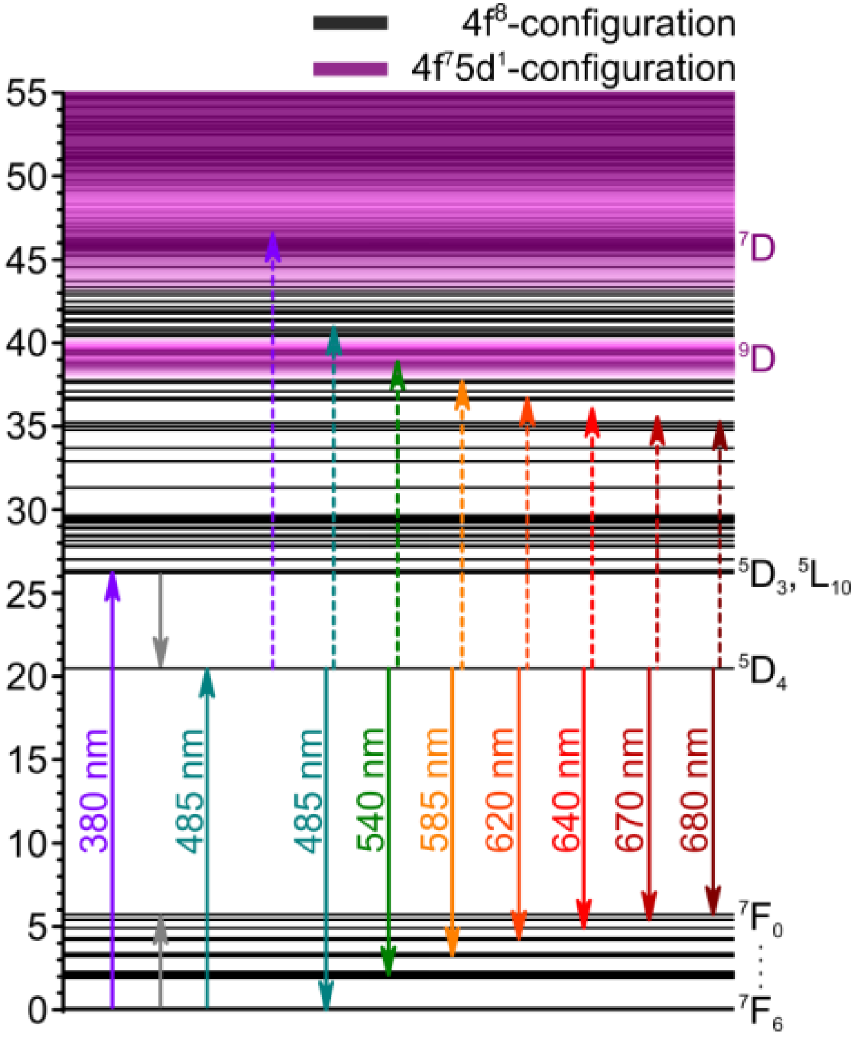


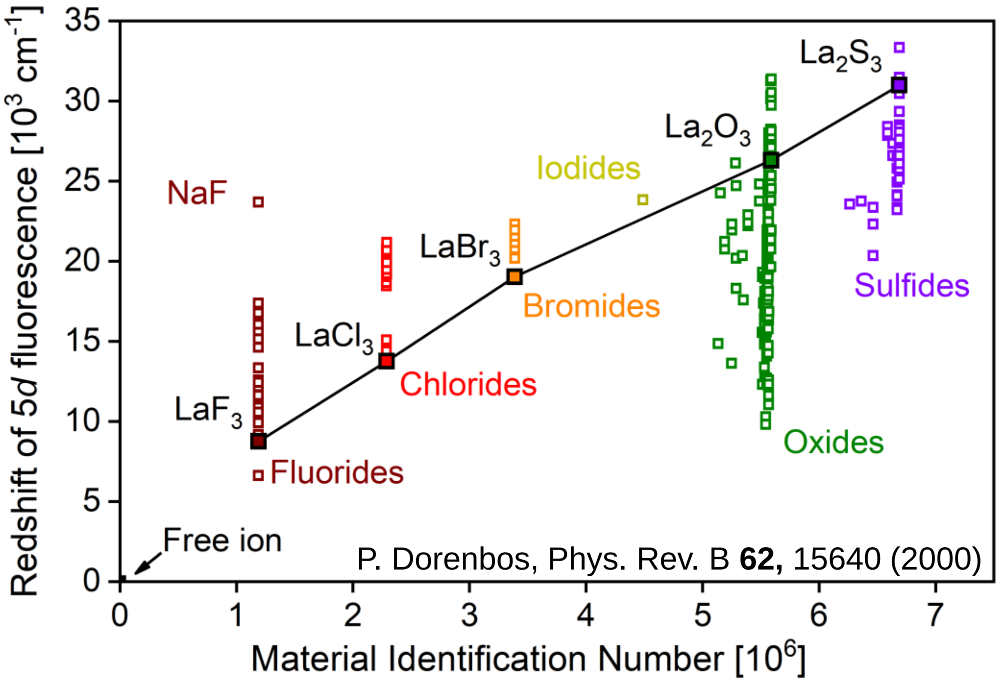


Badtke *et al.*, Appl. Phys. B **127** (6) 83 (2021)

Tb^{3+} for visible lasers

- To avoid 4f-5d ESA, Tb^{3+} requires low crystal field

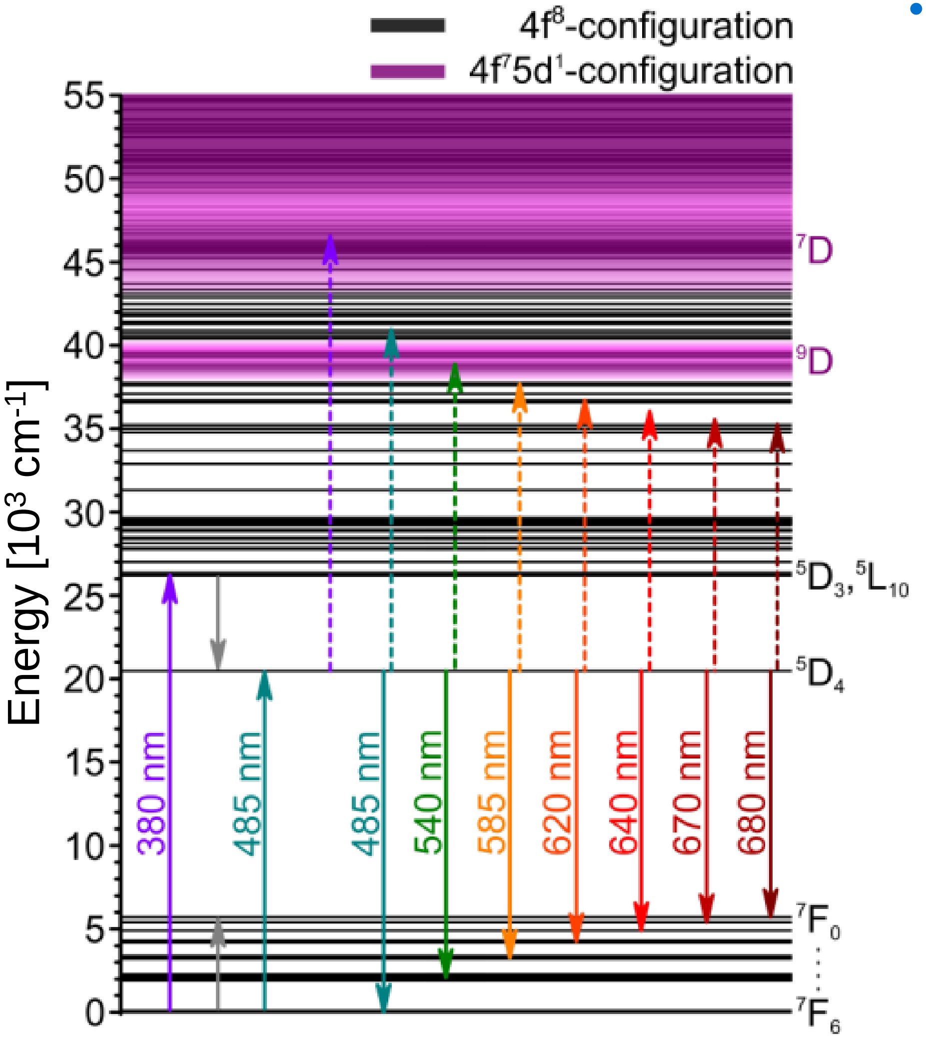


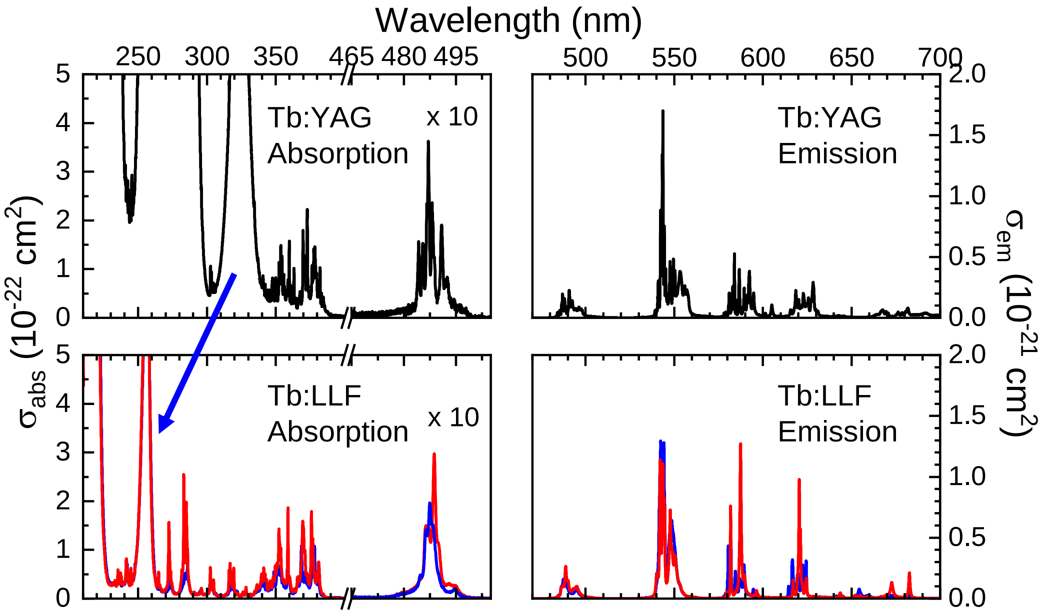


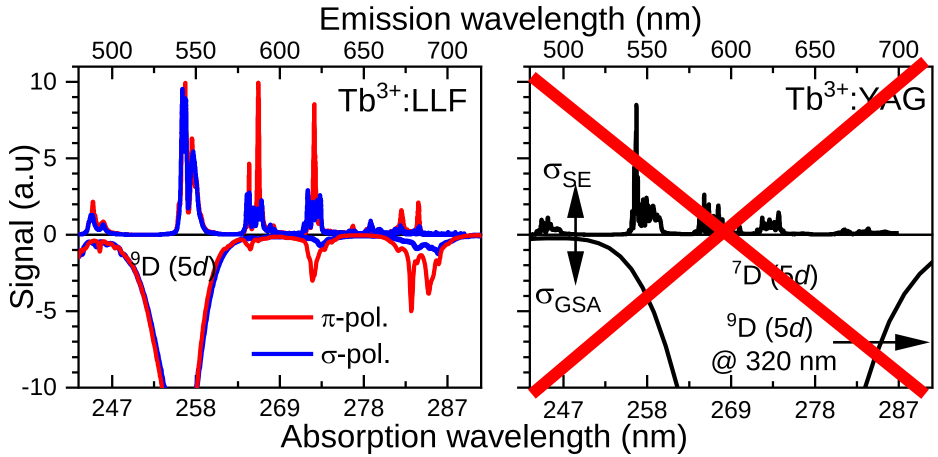
P. Dorenbos, Phys. Rev. B **62,** 15640 (2000)

Tb^{3+} for visible lasers

- To avoid 4f-5d ESA, Tb^{3+} requires low crystal field
- Compare Tb:YAG and Tb:YLF in this respect



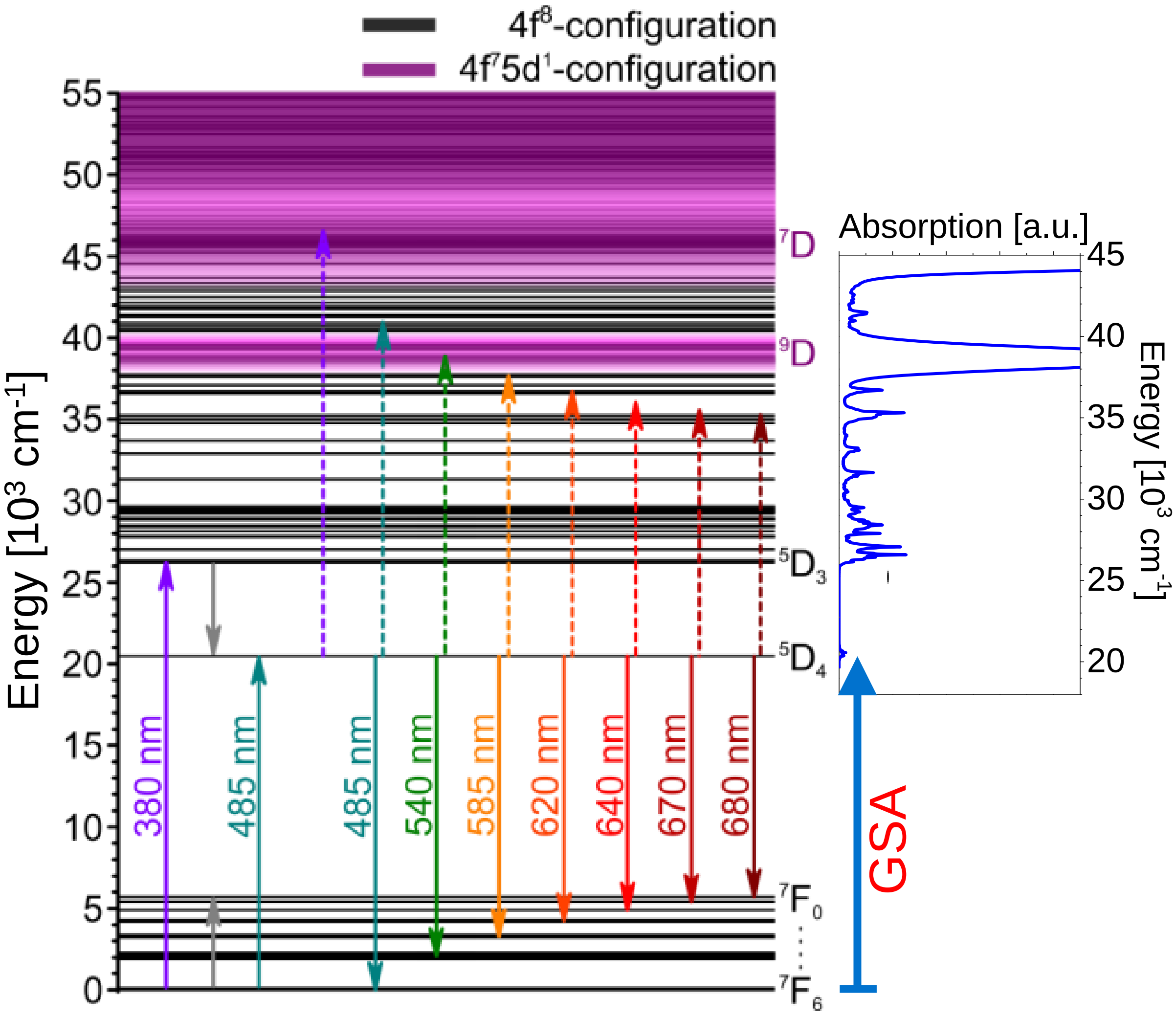




Kalusniak *et al.*, Appl. Phys. B **128** 33 (2022)

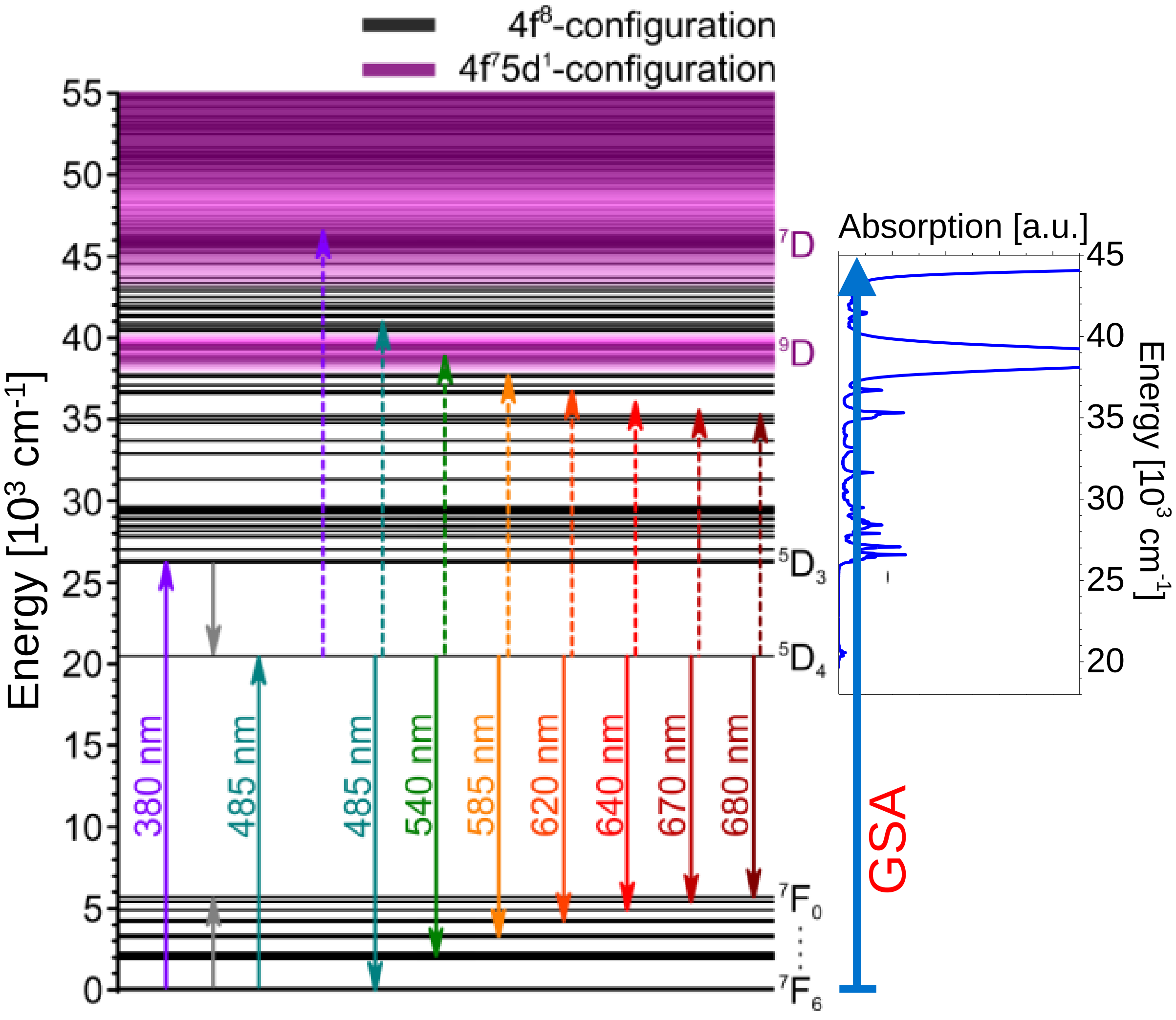
Tb^{3+} for visible lasers

Inter- and intraconfigurational GSA and ESA



Start Level	Final Level	Parity	Spin	Strength
$^7F_6$ (4*f*)	$^5D_4$ (4*f*)	↓	↓	weak

Inter- and intraconfigurational GSA and ESA



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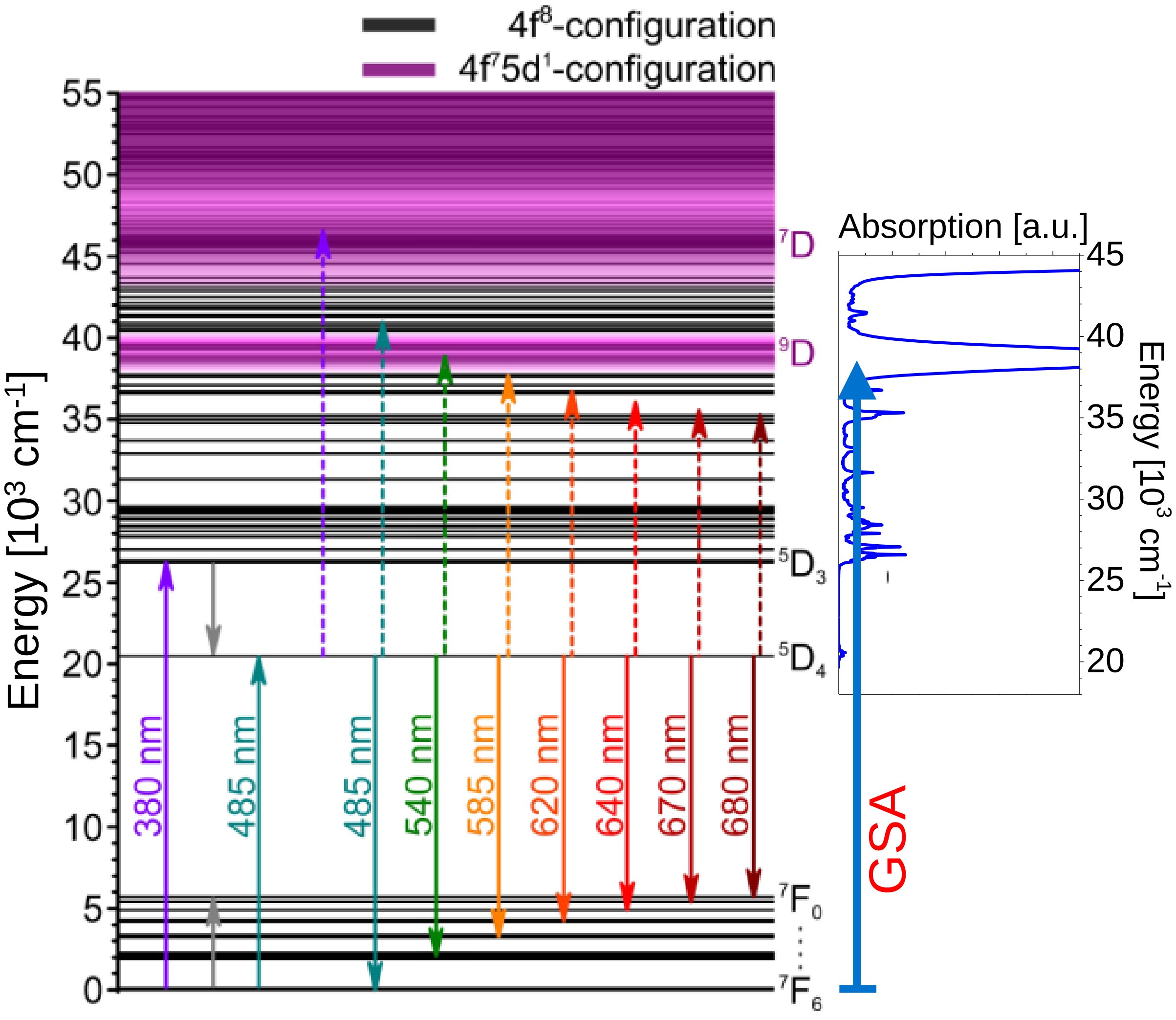
$^7F_6$ (4*f*)	^7D (5*d*)	↑	↑	strong

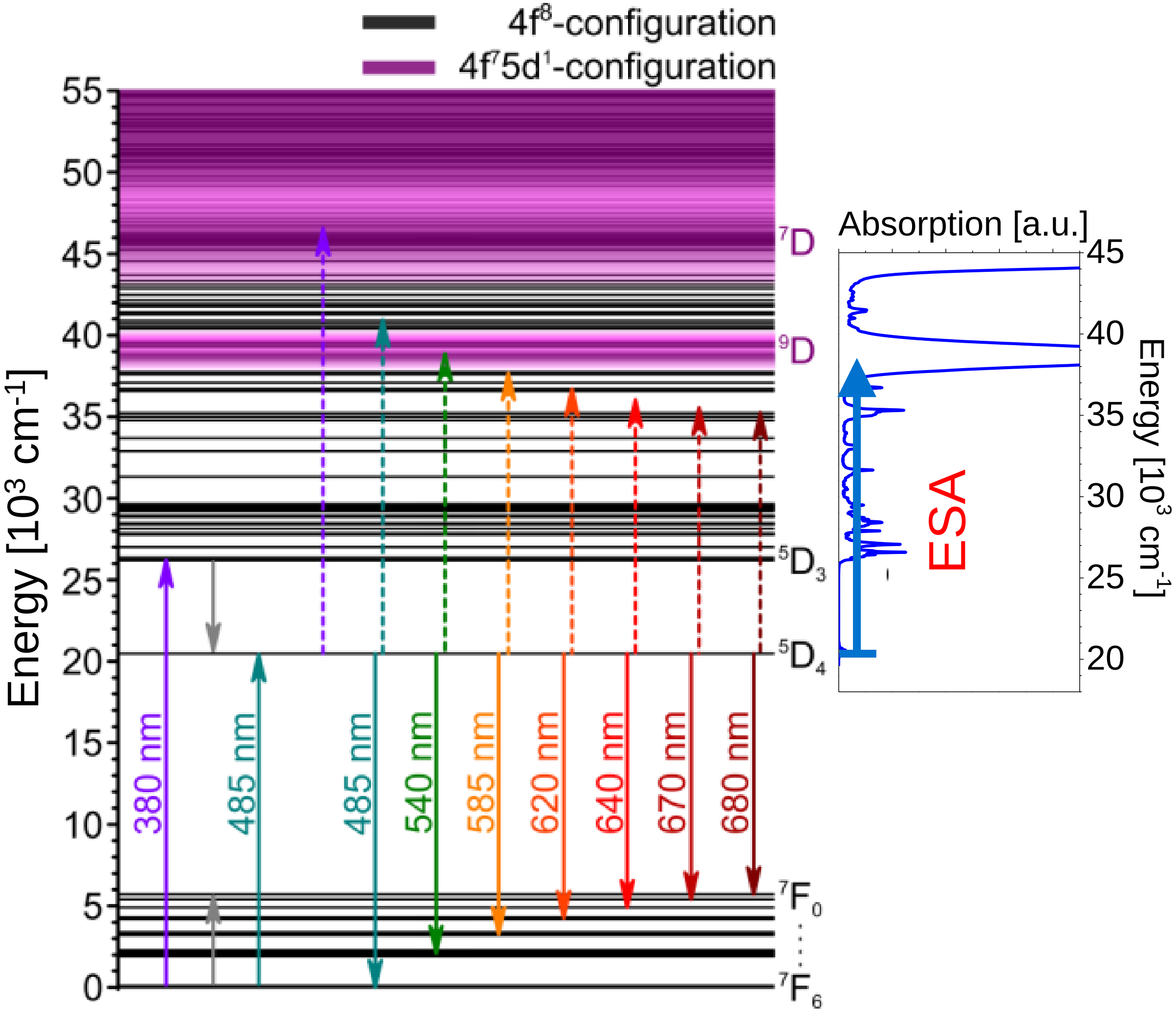
Tb^{3+} for visible lasers

Inter- and intraconfigurational GSA and ESA

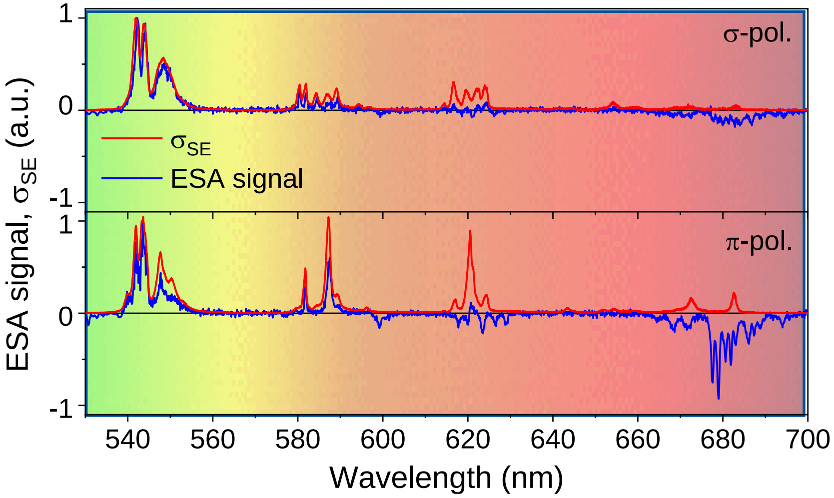


Start Level	Final Level	Parity	Spin	Strength
$^7F_6$ (4*f*)	$^5D_4$ (4*f*)	↓	↓	weak
$^7F_6$ (4*f*)	^7D (5*d*)	↑	↑	strong
$^7F_6$ (4*f*)	^9D (5*d*)	↑	↓	medium

Inter- and intraconfigurational GSA and ESA



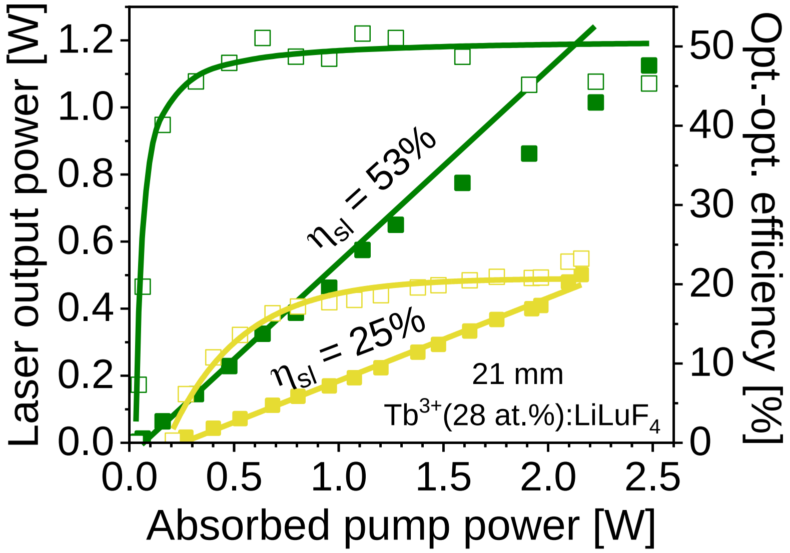
Start Level	Final Level	Parity	Spin	Strength
^{7}F$_6$ (4*f*)	^{5}D$_4$ (4*f*)	↓	↓	weak
^{7}F$_6$ (4*f*)	^{7}D (5*d*)	↑	↑	strong
^{7}F$_6$ (4*f*)	^{9}D (5*d*)	↑	↓	medium
^{5}D$_3$ (4*f*)	^{9}D (5*d*)	↑	↓↓	very weak

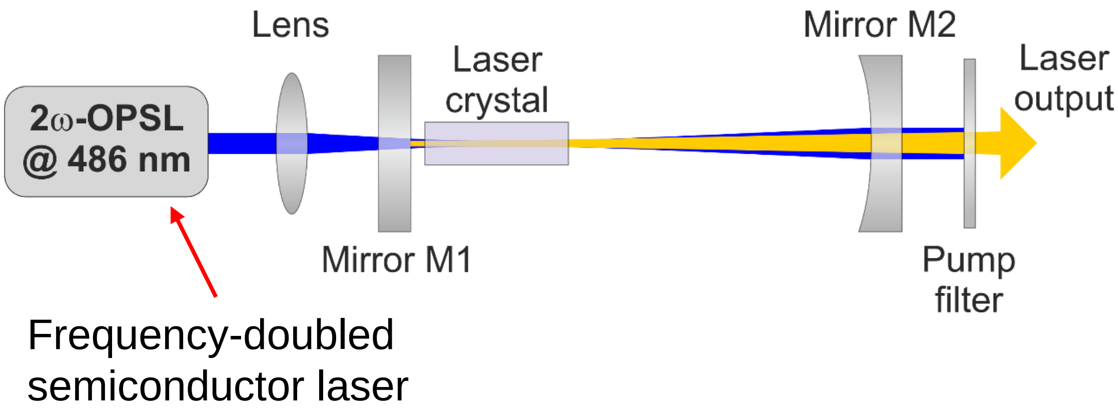


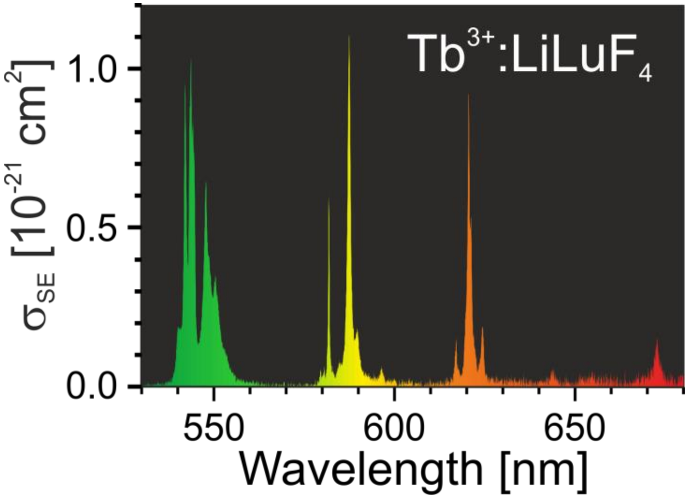
Tb^{3+} for visible lasers

Green and yellow Tb^{3+}-lasers

- Simple hemispheric linear laser setup:
  - 2ω-OPSL at 486 nm
  - Mirrors for yellow or green laser operation
- Spin forbidden absorption:
  η_{abs} only ~50% in 11 mm Tb(28%):$LiLuF_4$
- Efficient lasing at **545 nm** and at **587 nm**
  in various fluoride host materials
- Lasing in the red and orange suppressed by ESA
- Lasing even in TbF_3 → higher doping possible

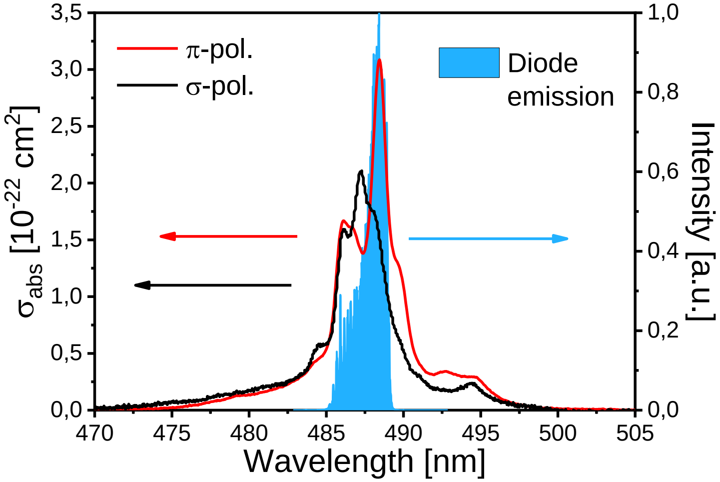


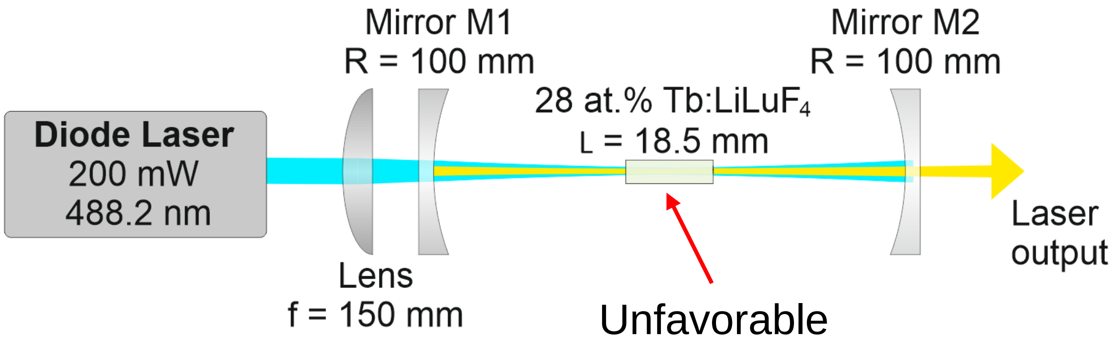




Kränkel *et al.*, Las. Phot. Rev. **10** (4), 548-568 (2016), Castellano-Hernández *et al.*, Opt. Lett. **43** (19), 4791-4794 (2018)

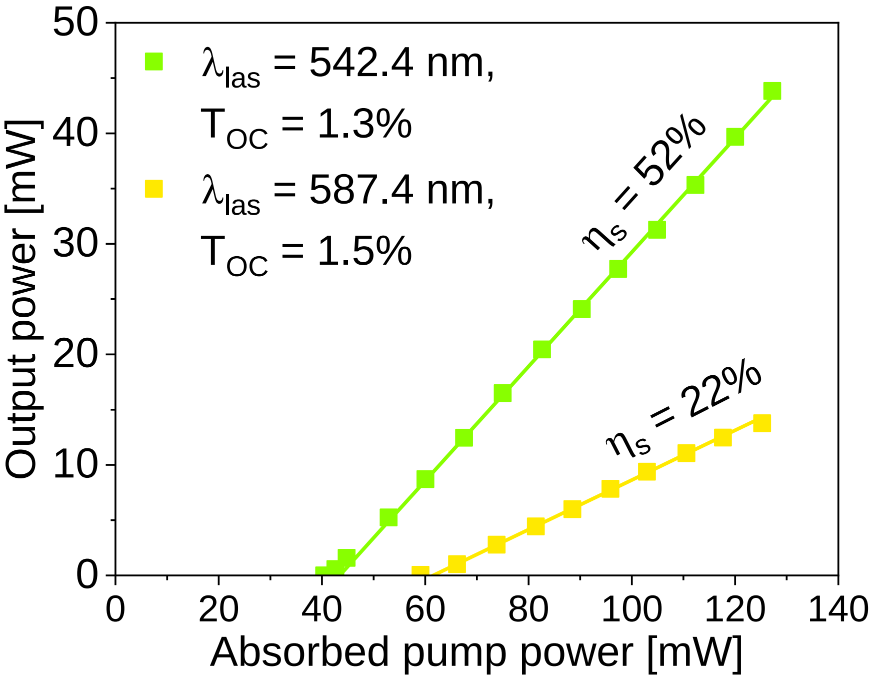
488 nm diode pumping of green and yellow Tb^{3+}:LLF lasers





First diode pumped Tb^{3+}-laser

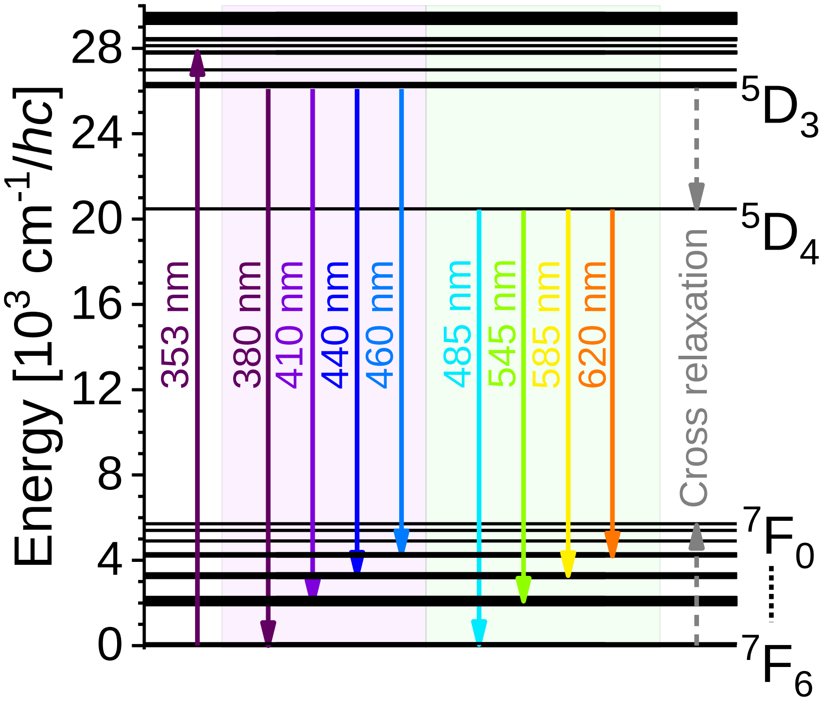
- 18.5 mm long Tb^{3+}(28 at.%):$LiLuF_4$
- 77% absorption efficiency
- Efficiency similar to 2ω-OPSL pumping
  - 52% slope at 542 nm (green)
  - 22% slope at 587 nm (yellow)
- Output power limited by available single emitter pump power of 200 mW

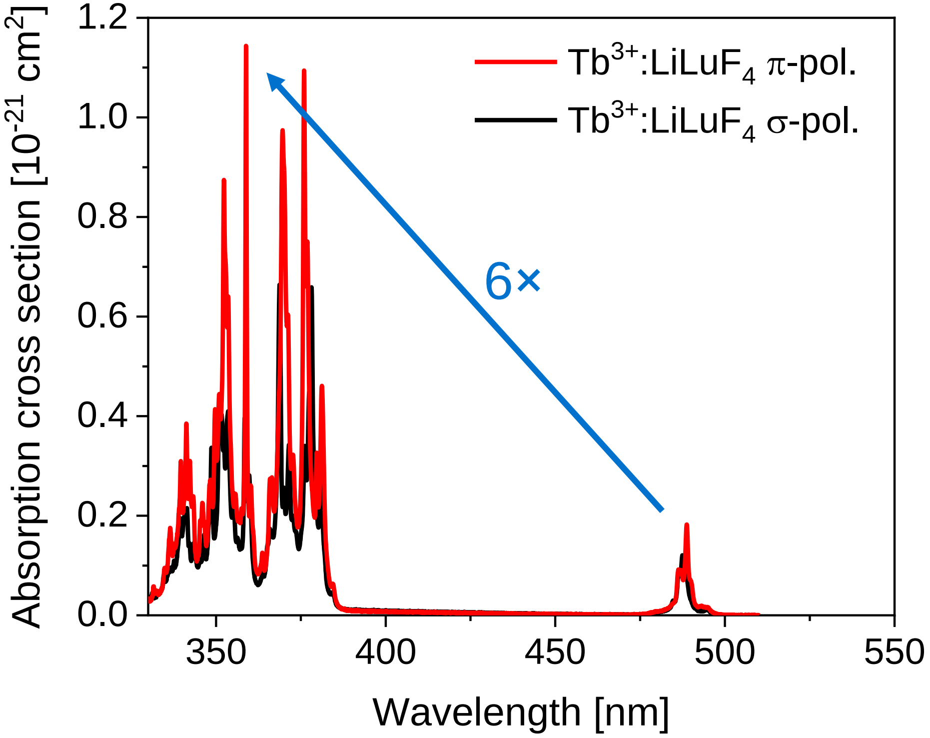


Castellano-Hernandez *et al.*, Las. Photon. Rev. **14** (2), 1900229 (2020)

UV pumping of Tb^{3+}-lasers

Pumping into the $^5D_3$ level of Tb^{3+}:$LiLuF_4$





Can the strong UV absorption be utilized for laser pumping?

Kalusniak *et al.*, Opt. Lett., **45** (20), 6170 (2020)

UV pumping of Tb^{3+}-lasers

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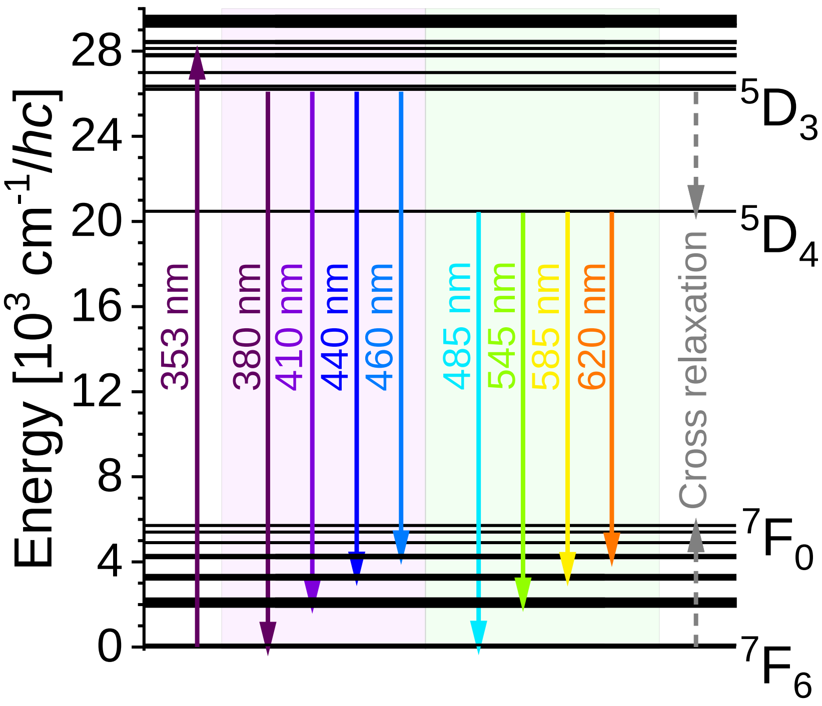
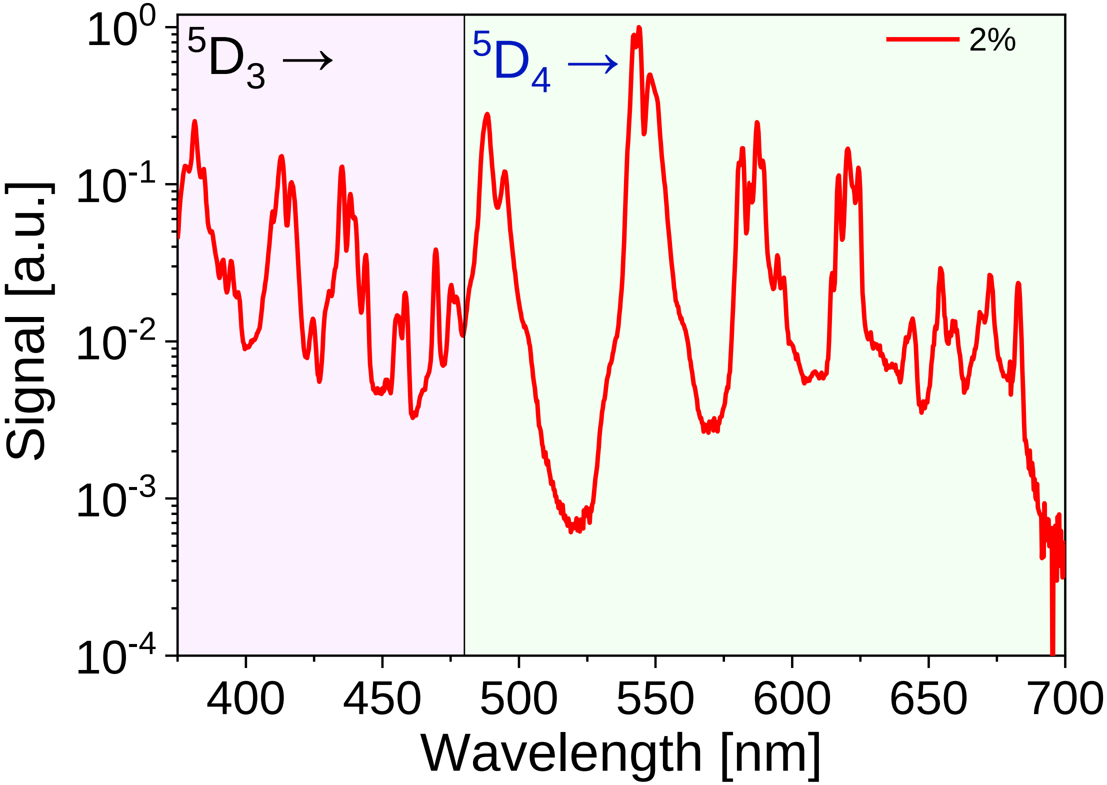


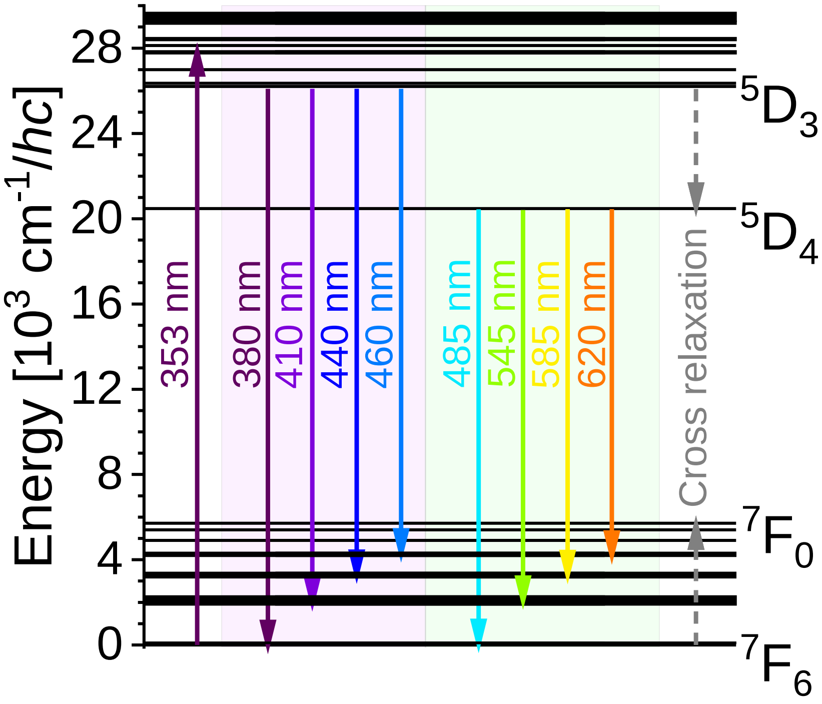


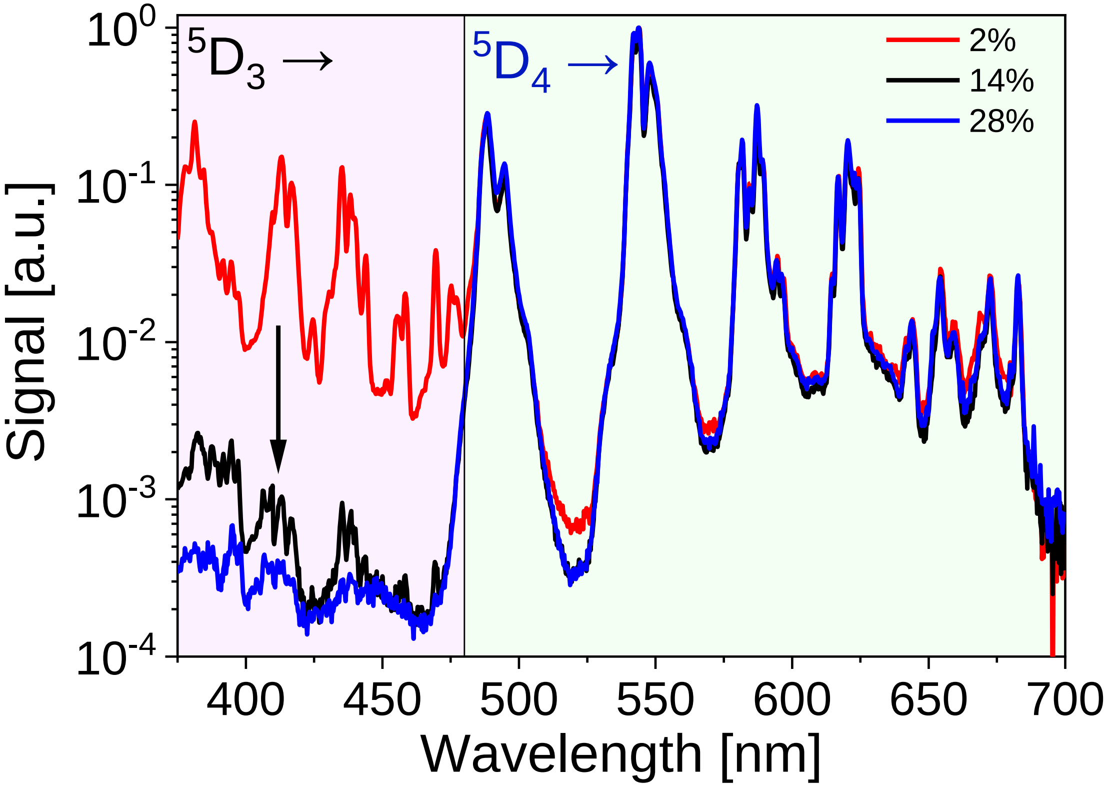
- Strong $^5D_3$ fluorescence in Tb^{3+} under UV excitation in low doped Tb^{3+}(2 at.%):YLF

Kalusniak *et al.*, Opt. Lett., **45** (20), 6170 (2020)

UV pumping of Tb^{3+}-lasers

Pumping into the $^5D_3$ level of Tb^{3+}:$LiLuF_4$



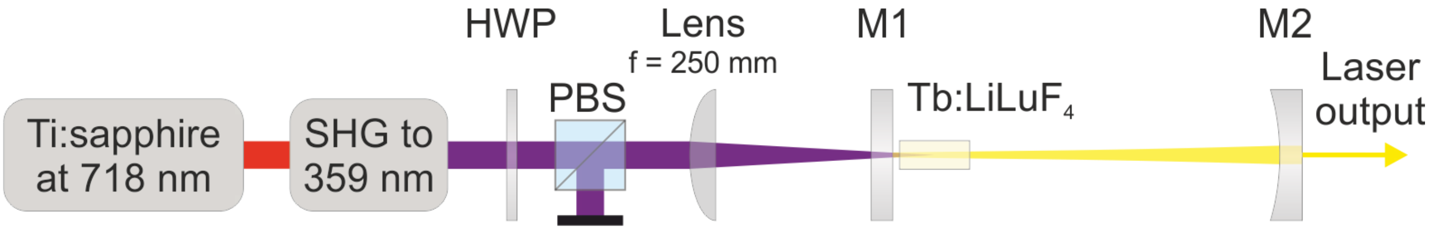


- Strong $^5D_3$ fluorescence in Tb^{3+} under UV excitation in low doped Tb^{3+}(2 at.%):YLF
- Quenching of $^5D_3$ fluorescence by 2 orders of magnitude in 14 at.% and 28 at.% Tb^{3+}:YLF

Efficient cross relaxation populates upper laser level $^5D_4$ under UV excitation

Kalusniak *et al.*, Opt. Lett., **45** (20), 6170 (2020)

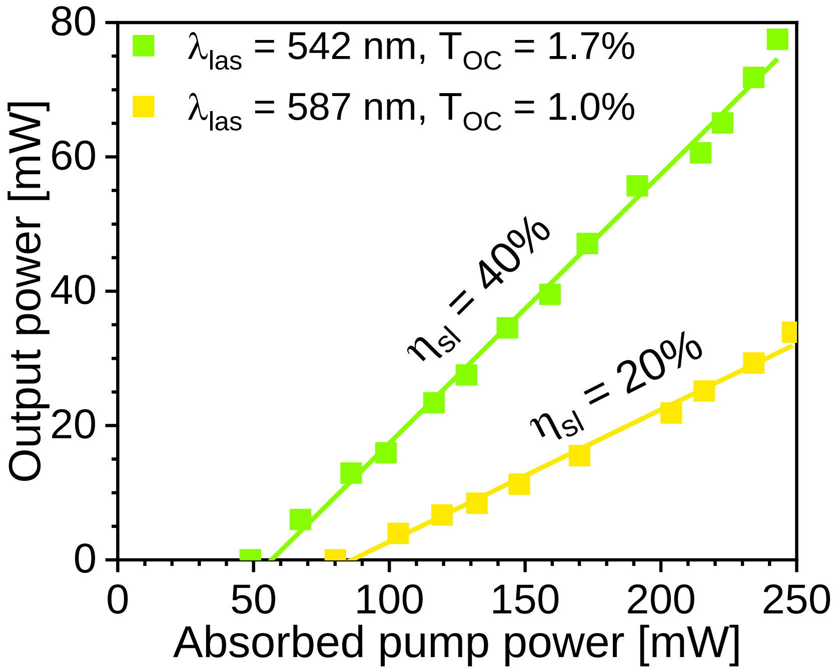
359 nm pumped green and yellow 11 mm Tb(14 at.%):$LiLuF_4$ laser



- UV-pumping: Lower η_{Stokes} and thus η_{slope}
- 3 × stronger absorption than P_{pump} = 486 nm

- **2 × higher $\eta_{opt,inc}$**

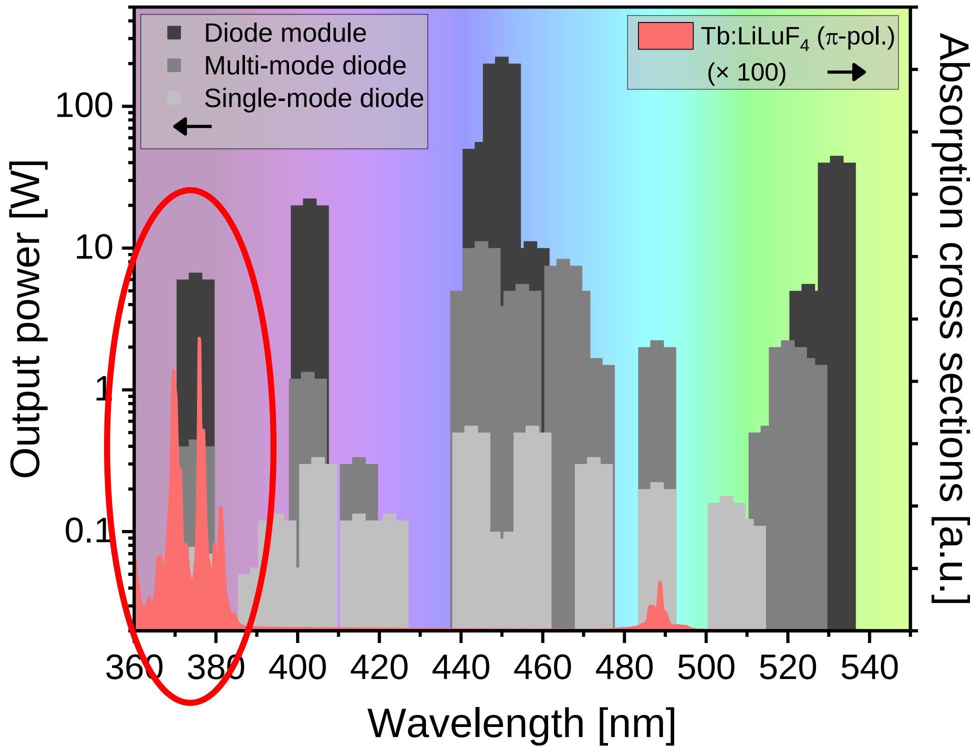
λ_{las}	542 nm		587 nm	
λ_{pump}	486 nm	359 nm	486 nm	359 nm
$P_{thr,HR}$	11 mW	15 mW	18 mW	24 mW
η_{St}	90%	66%	83%	61%
$\eta_{sl, abs}$	59%	40%	26%	20%
$\eta_{sl, abs}/\eta_{St}$	66%	61%	31%	33%
η_{abs}	32%	97%	32%	97%
$\eta_{opt, inc}$	**19%**	**39%**	**8%**	**19%**

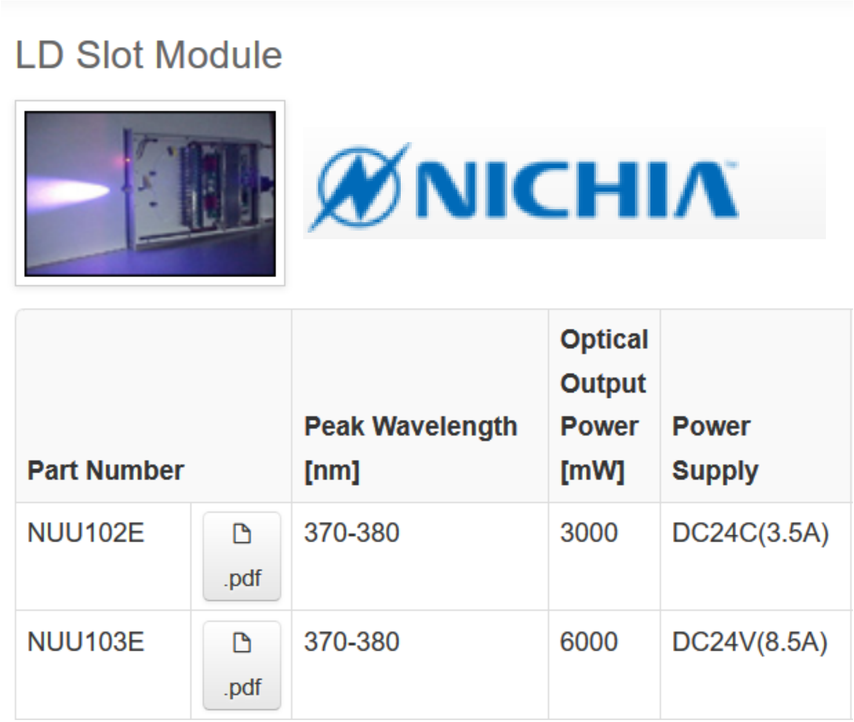


Kalusniak *et al.*, Opt. Lett., **45** (20), 6170 (2020)

UV pumping of Tb^{3+}-lasers

Prospects for high power UV diode pumping of Tb^{3+}-lasers





LD Slot Module

Part Number		Peak Wavelength [nm]	Optical Output Power [mW]	Power Supply
NUU102E	.pdf	370-380	3000	DC24C(3.5A)
NUU103E	.pdf	370-380	6000	DC24V(8.5A)

https://www.nichia.co.jp/en/product/laser.html

- Absorption at ~375 nm addressed by commercial multi-watt diode modules
- 3 mm long Tb(28%):YLF sample would absorb more than 80% of pump power
  - → Good mode matching despite low beam quality of diode modules
  - → 970 nm peak with 3 nm FWHM acceptance bandwidth

Watt-class yellow and green Tb^{3+}-based DPSSLs within reach

Kalusniak *et al.*, Opt. Lett., **45** (20), 6170 (2020)

Outline

- Introduction
- Pr^{3+} and Tb^{3+}-doped materials for visible lasers
- **Sesquioxides for high power infrared lasers**
- Conclusion

Host materials for infrared lasers

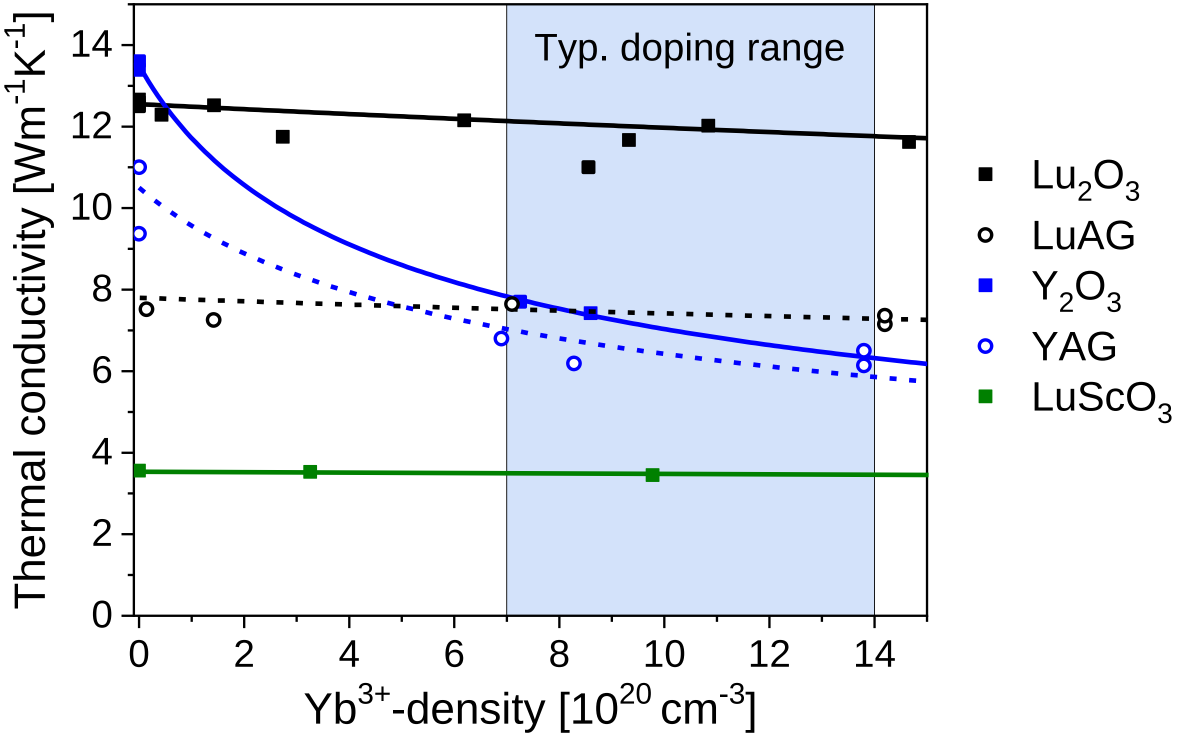
Properties of sesquioxide host materials

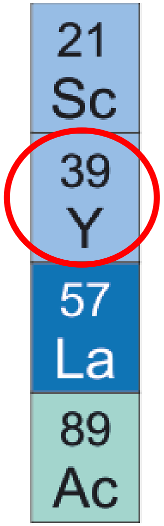
	Sc_2O_3	Y_2O_3	Lu_2O_3	YAG	YLF	
Crystal lattice	cubic	cubic	cubic	cubic	tetragonal	
Thermal conductivity [$Wm^{-1}K^{-1}$]	**16.5**	**13.6**	**12.5**	10.5	**7.2 ||a**	**5.8 ||c**
Mohs hardness	6½	6½	6½	8	4-5	
Max. phonon energy [cm^{-1}]	**672**	**597**	**618**	**857**	460	
Melting point [°C]	**~2430**	**~2430**	**~2450**	~1930	~830	
Thermal expansion [10^{-6} K^{-1}]	5.9	(8.6)	(8.5)	8	8.3 ||a	13.3 ||c
Thermo-optical coefficient [10^{-6} K^{-1}]	6.9	6.7	7.5	8	-4.3 ||a	-2.0 ||c

- Band-gap and 5*d* energy levels are not crucial for low-photon energy mid-infrared lasers
- Thermal conductivity is essential for heat management in high power lasers
- Low phonon energies avoid non-radiative multi-phonon decay in mid-infrared lasers

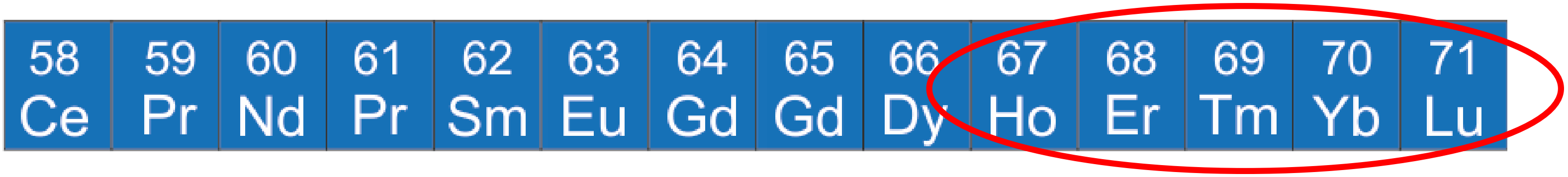
Sesquioxides combine many favorable properties

Lu_2O_3 as a high thermal conductivity host for high power lasers





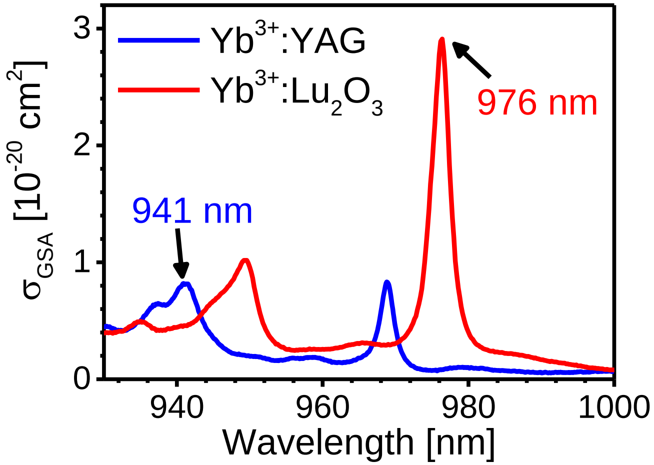
- Thermal conductivity in insulator crystals depends on phonon propagation
- Large Δm between lattice and doping scatters phonons
  → Lu^{3+} is best suited host cation for heavy rare earth ions (Ho, Er, Tm, Yb)

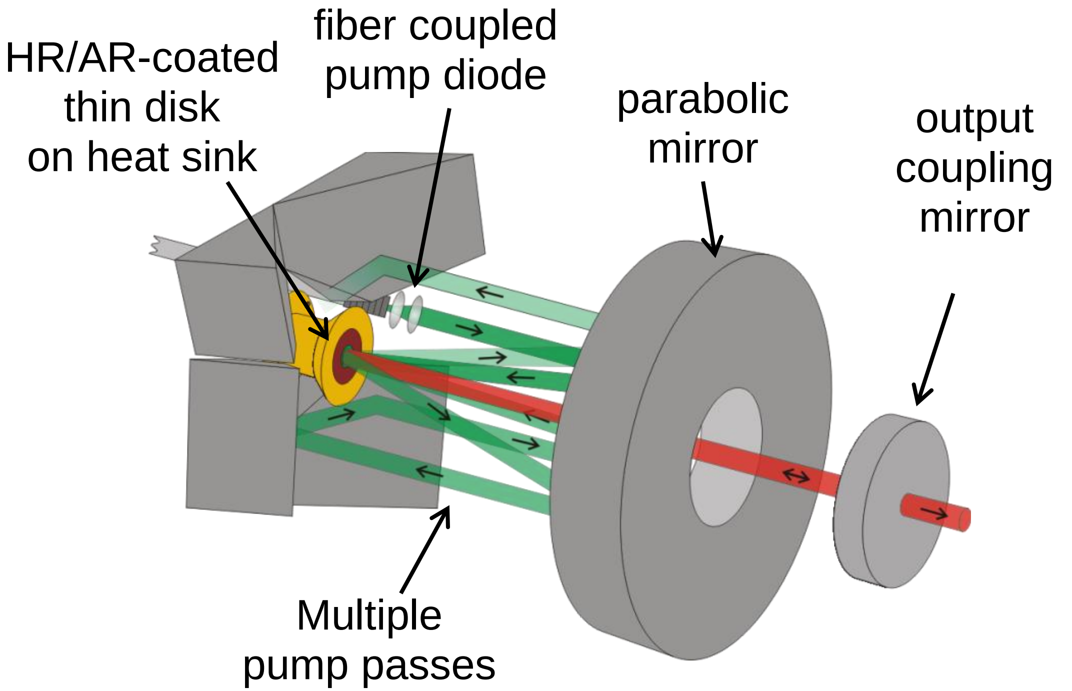


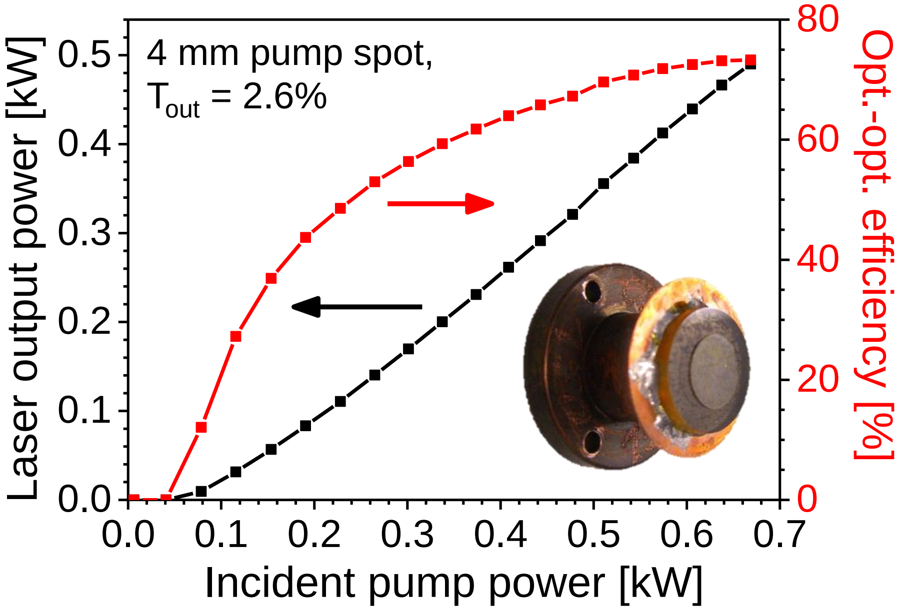
Peters *et al.*, Appl. Phys. B **102** (3), 509-514 (2011)

Continuous wave Yb:Lu_2O_3 thin disk laser

- Yb^{3+}:Lu_2O_3 provides > 3 times higher absorption cross sections compared to Yb^{3+}:YAG
- cw thin disk laser using 0.2 mm Yb(3.5%):Lu_2O_3
  - 0.5 kW output power
  - 90% slope efficiency
  - 73% opt.-opt. efficiency







Kränkel, IEEE JSTQE **21** (1), 1602013 (2015)

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- cw thin disk laser using 0.2 mm Yb(3.5%):Lu_2O_3
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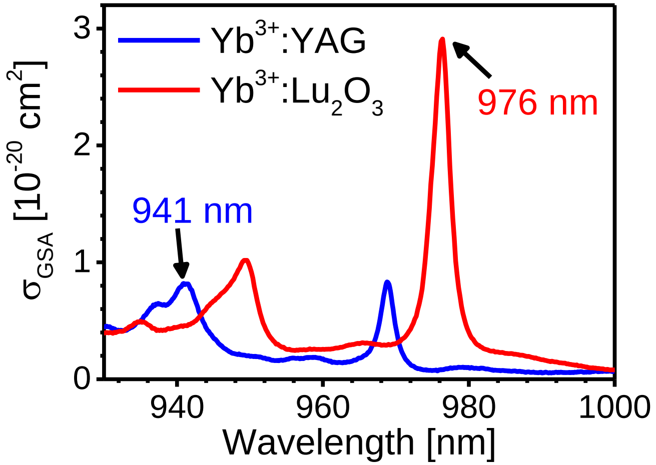
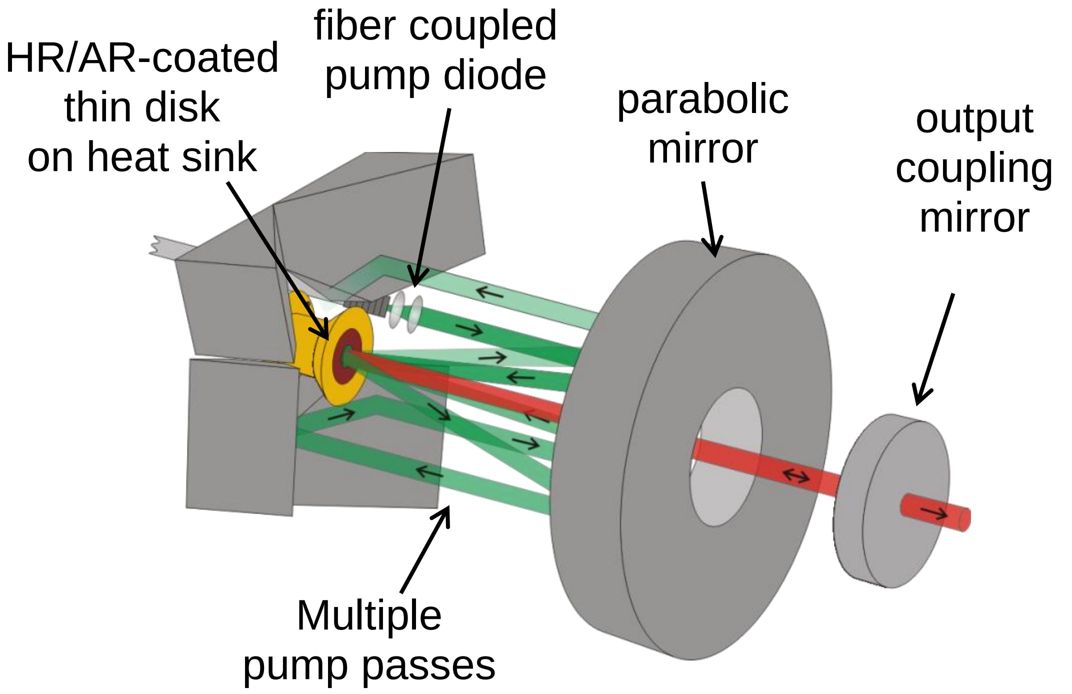
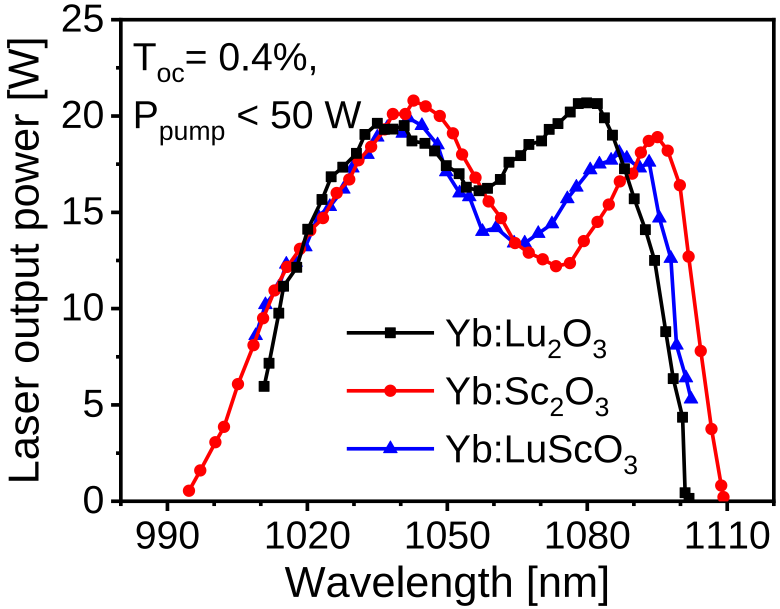
  - 90% slope efficiency
  - 73% opt.-opt. efficiency
  - wavelength tuning range of more than 100 nm



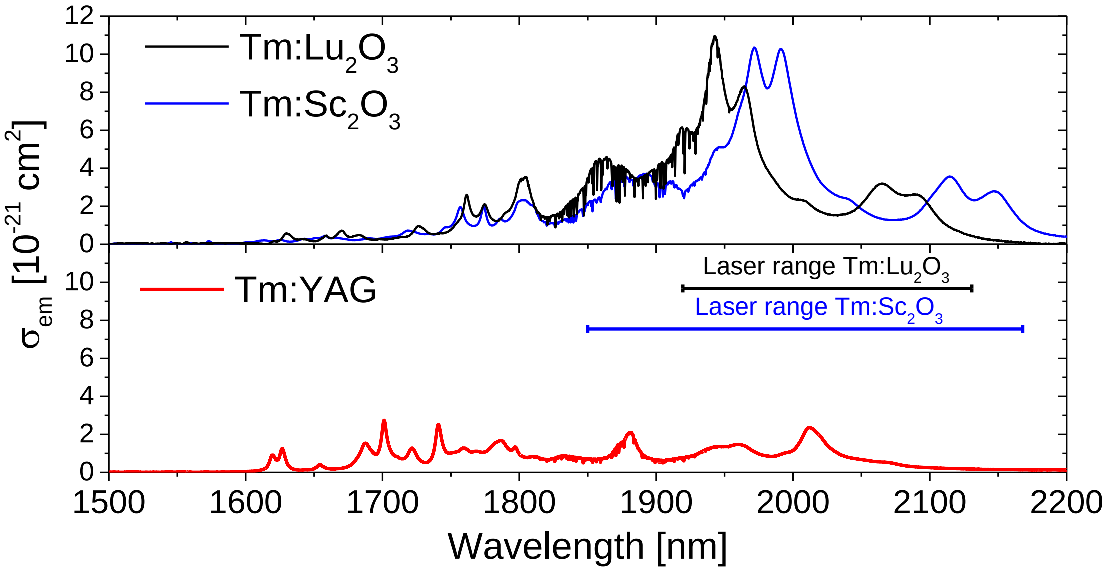


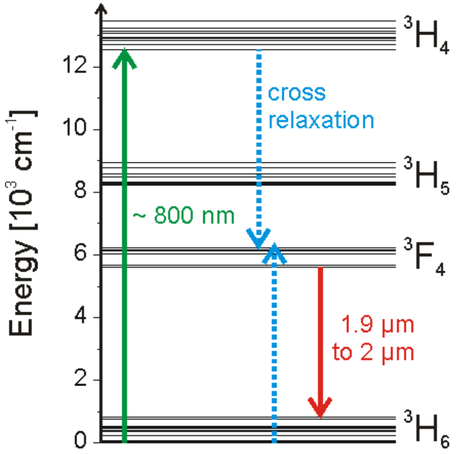


Peters *et al.*, Opt. Express **15** (11), 7075-7082 (2007)

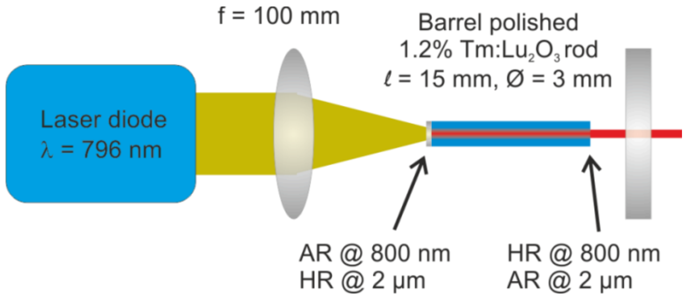
Tm^{3+}-doped sesquioxides for 2 µm lasers

High-power diode pumped $Tm:Lu_2O_3$ laser



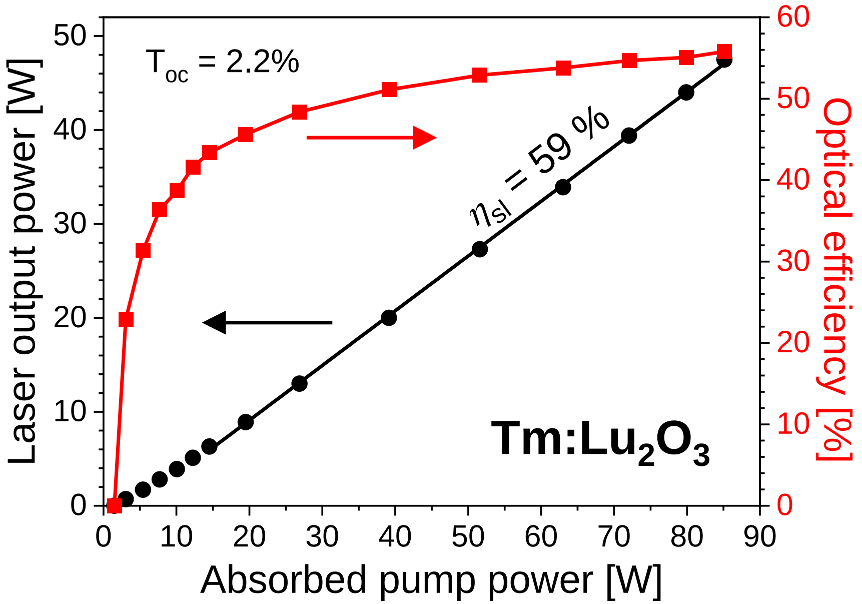


Two-for-one-pumping at 976 nm



Diode pumped cavity:

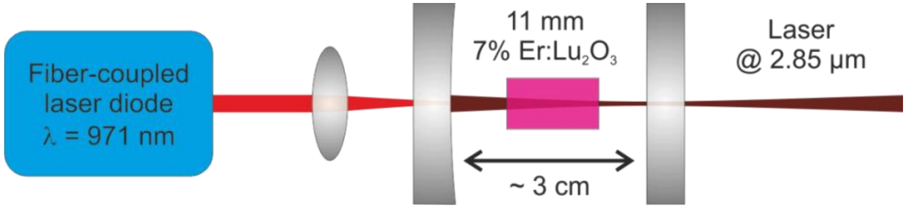
- 15 mm long Tm(1.2%):Lu_2O_3 rod
- 47 W at η_{sl}= 59 %
- λ_{las} > 2 µm

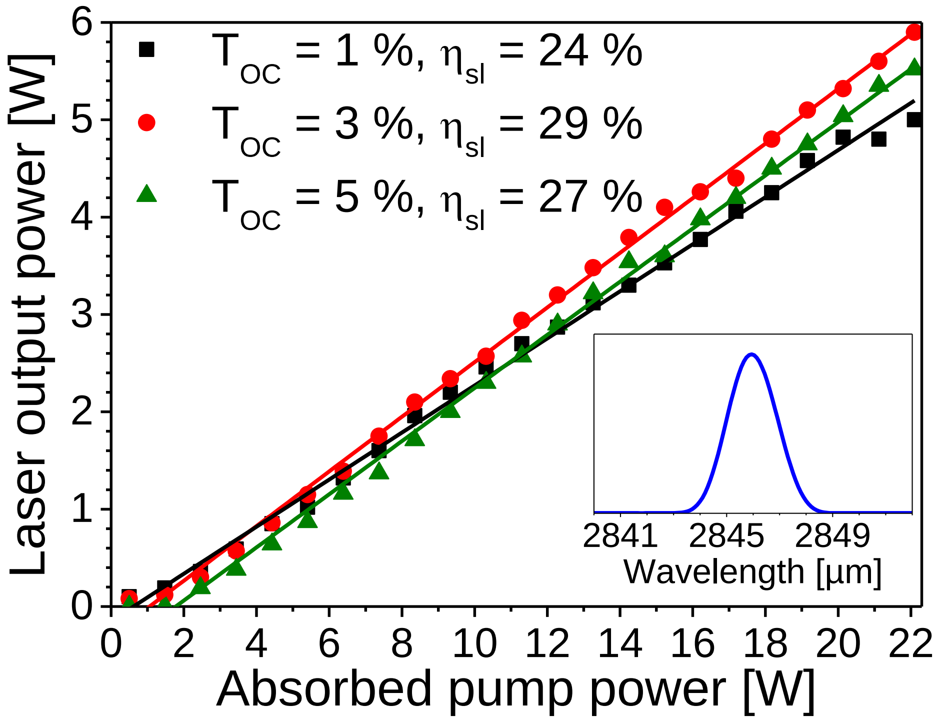


P. Loiko *et al.*, IEEE JSTQE **24** (5), 1600713 (13pp) (2018)

High power 3 µm $Er^{3+}:Lu_2O_3$-laser

- Highest room temperature output power: 5.9 W
- Short resonator prevents water absorption
- Center emission wavelength 2845 nm
- Recent result: > 10 W two-side diode pumped



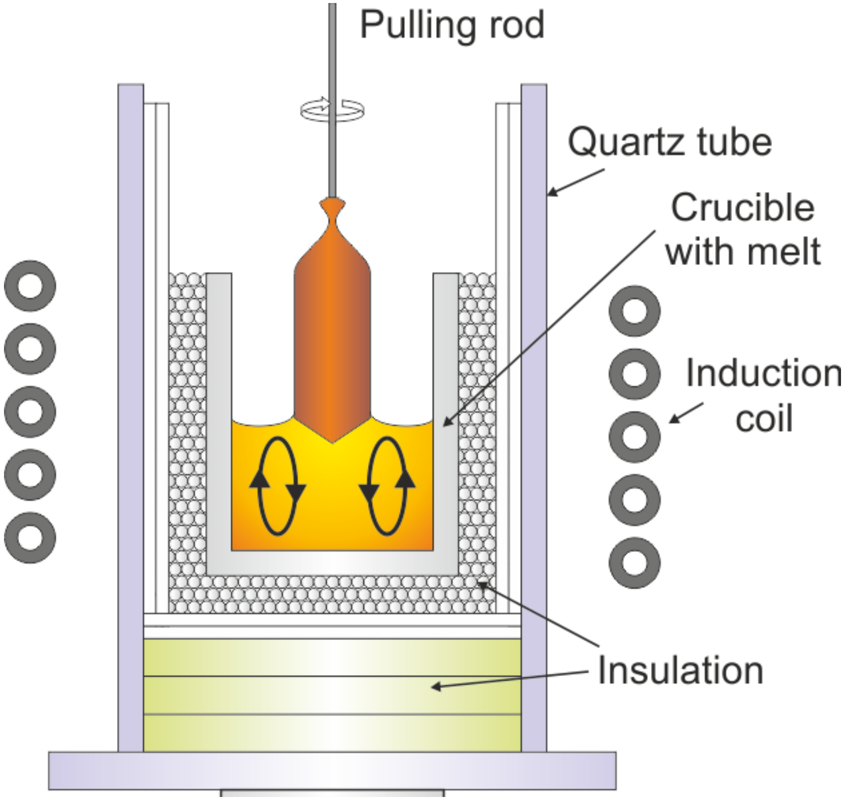


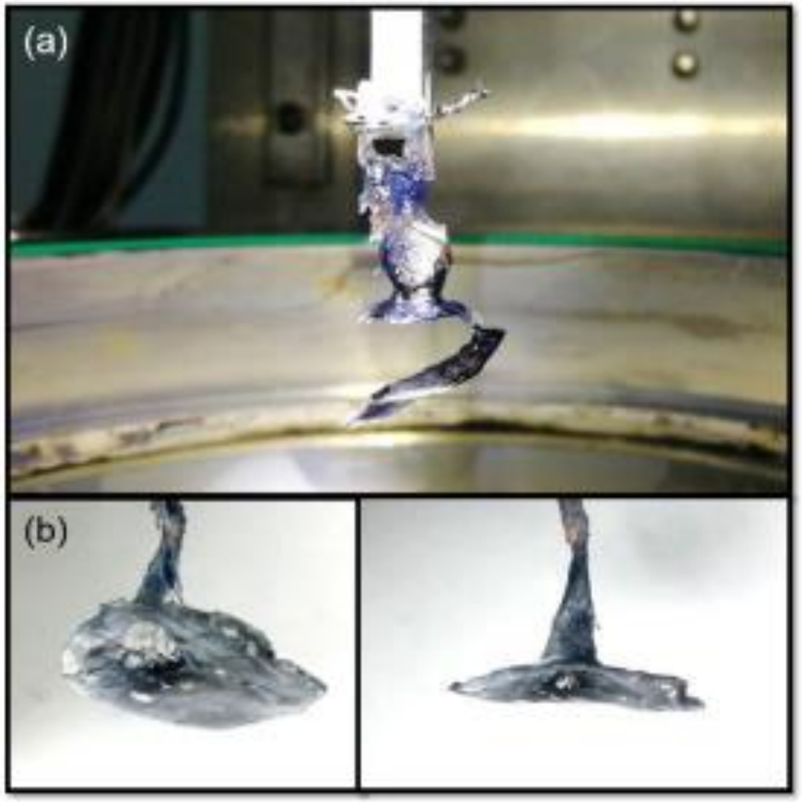
Li *et al.*, Opt. Lett. **37** (13), 2568-2570 (2012)

Czochralski growth of sesquioxides

Sesquioxide melting points of >2400 °C too high for Czochralski growth:

- Expensive rhenium crucibles required
- Growing crystal absorbs heat radiation and acts as a heat shield
  → no report on the laser quality growth of sesquioxide crystals by Czochralski

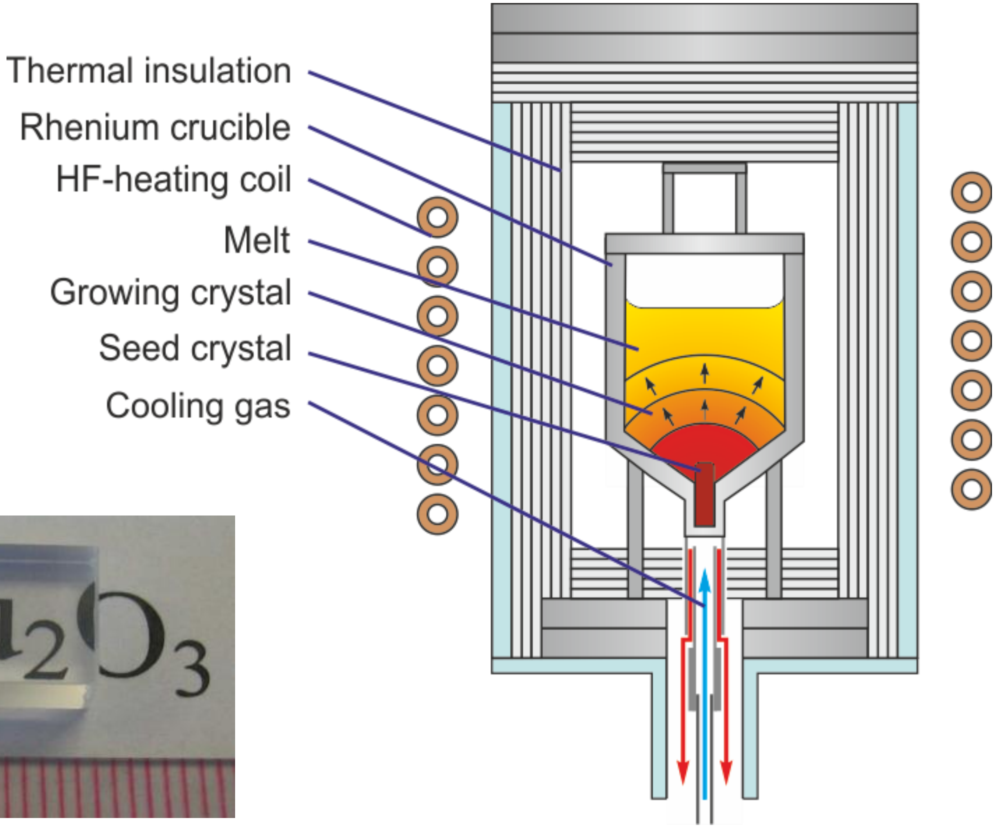




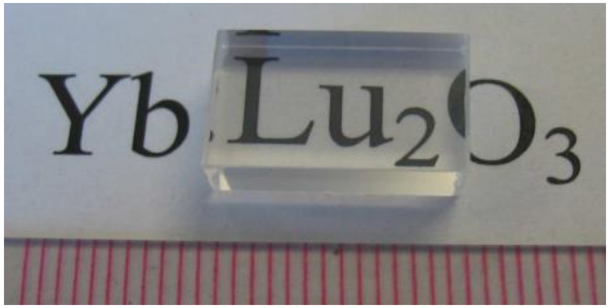
Yin *et al.*, CrystEngComm **22** (39), 6569-6573 (2020)

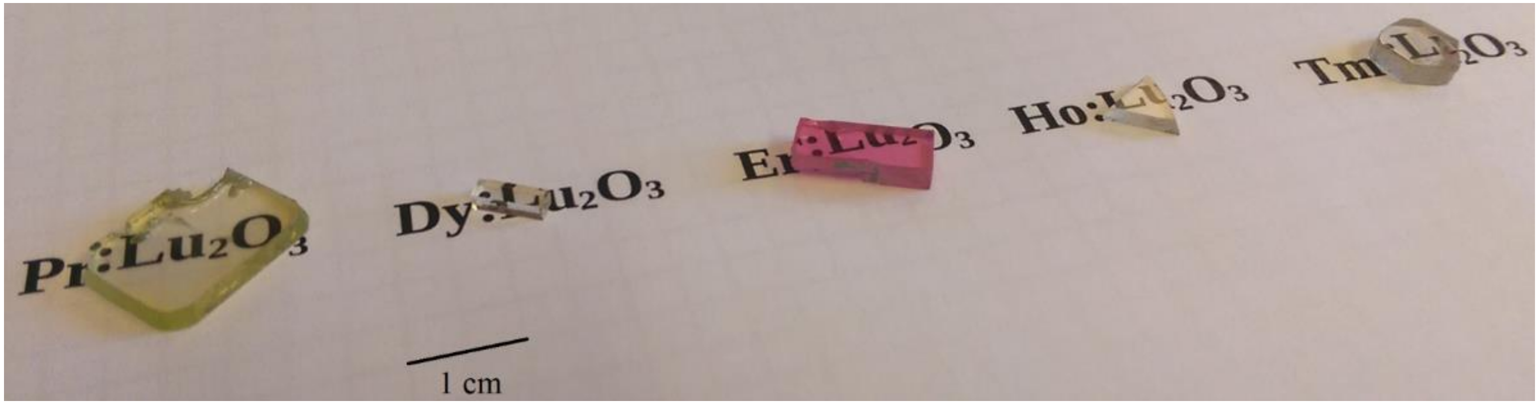
Growth of sesquioxides by the heat exchanger method

- Melting points > 2430 °C: very demanding growth
- Rhenium crucibles require precise growth atmosphere (low oxygen)
- **Good results, but high crucible cost & high damage risk**



Thermal insulation
Rhenium crucible
HF-heating coil
Melt
Growing crystal
Seed crystal
Cooling gas

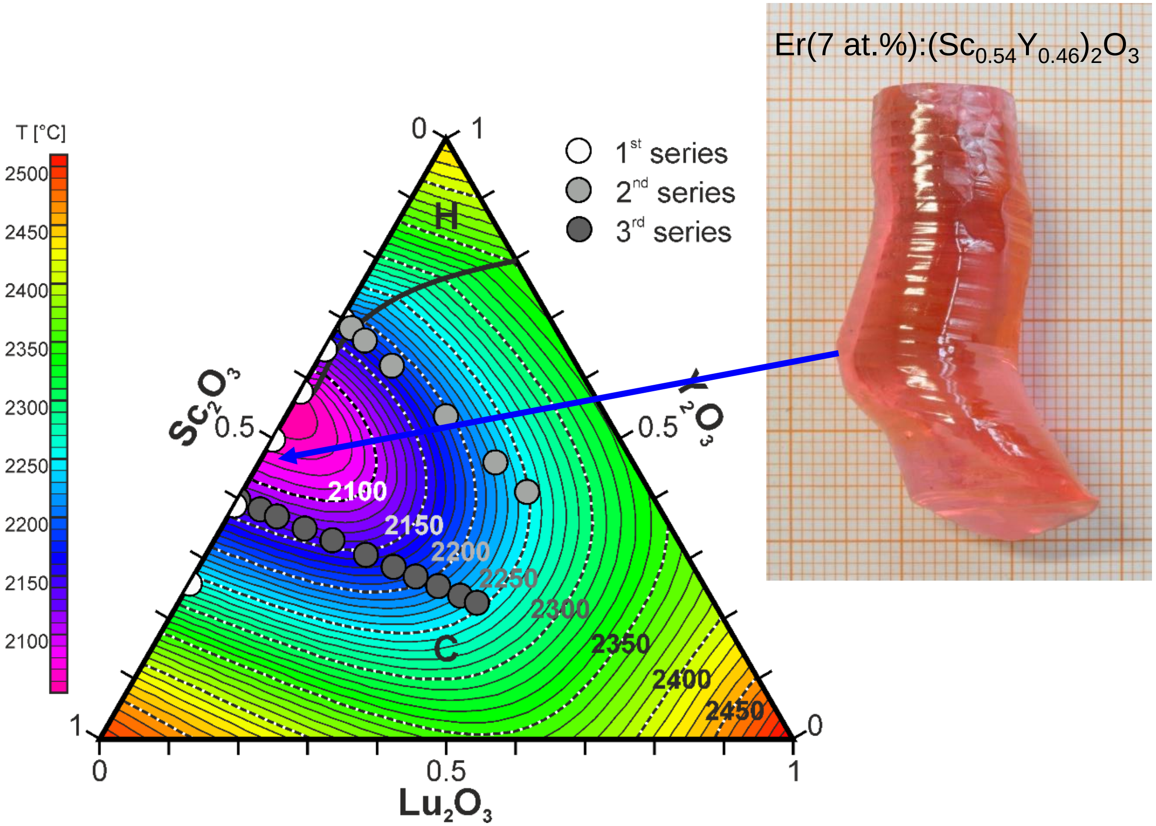


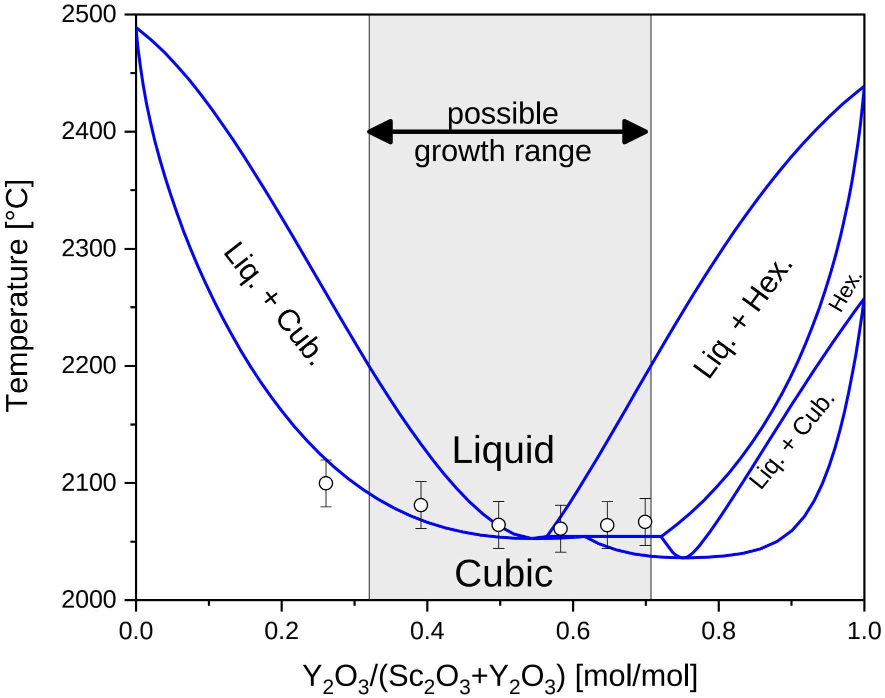


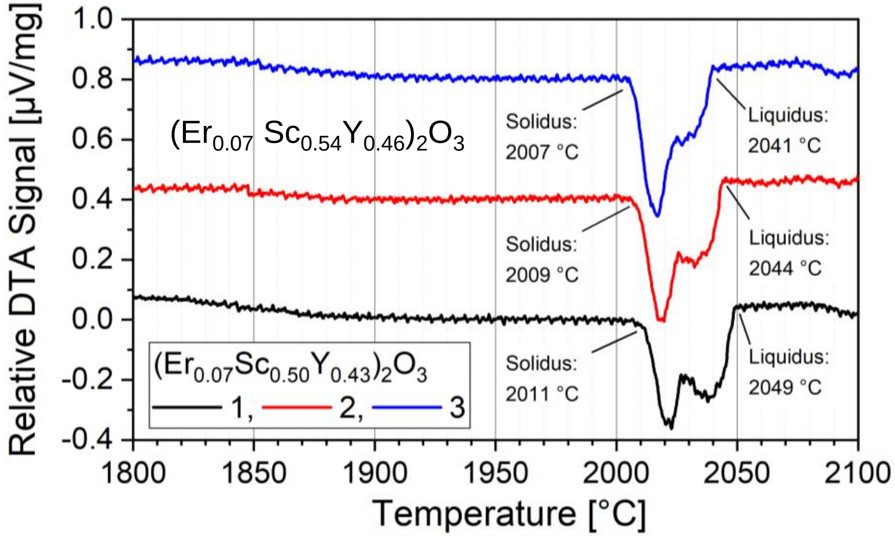
Peters *et al.*, J. Cryst. Growth **310** (7-9), 1934-1938 (2008)

Reduced melting point and Czochralski growth of mixed sesquioxides

- Liquidus of 2053°C for $(Sc_{0.45}Y_{0.55})_2O_3$
- Hexagonal phase for > 55% Y_2O_3
- Further reduced melting point and congruent melting for $(Er_{0.07}Sc_{0.50}Y_{0.43})_2O_3$
- Czochralski growth from Ir-crucible possible



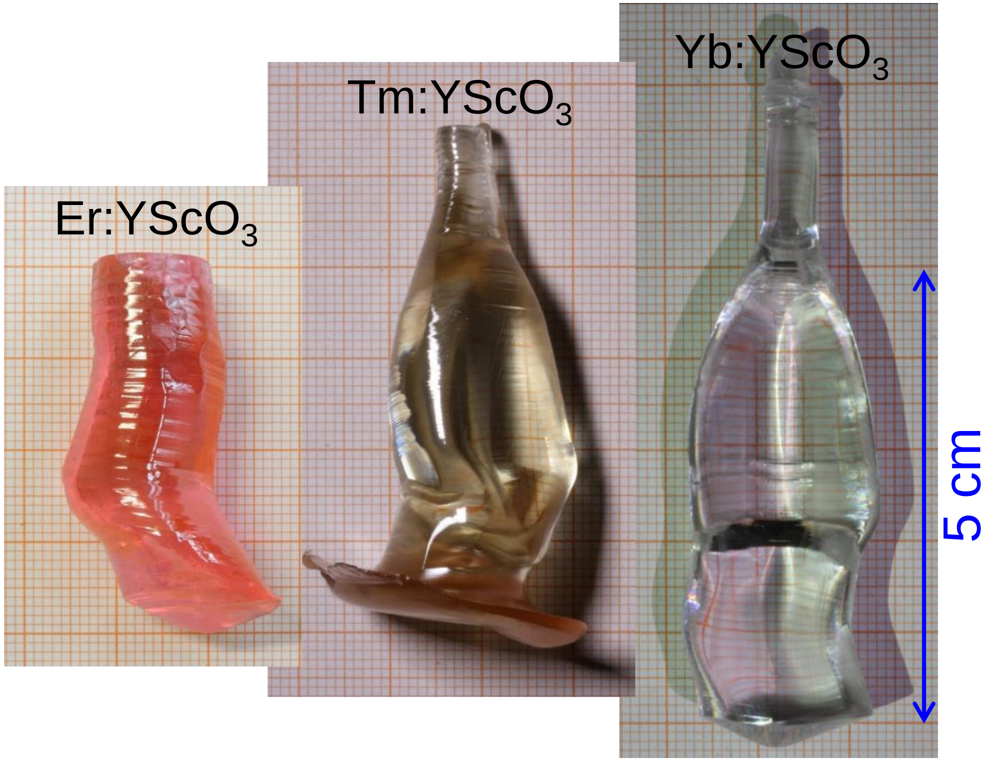


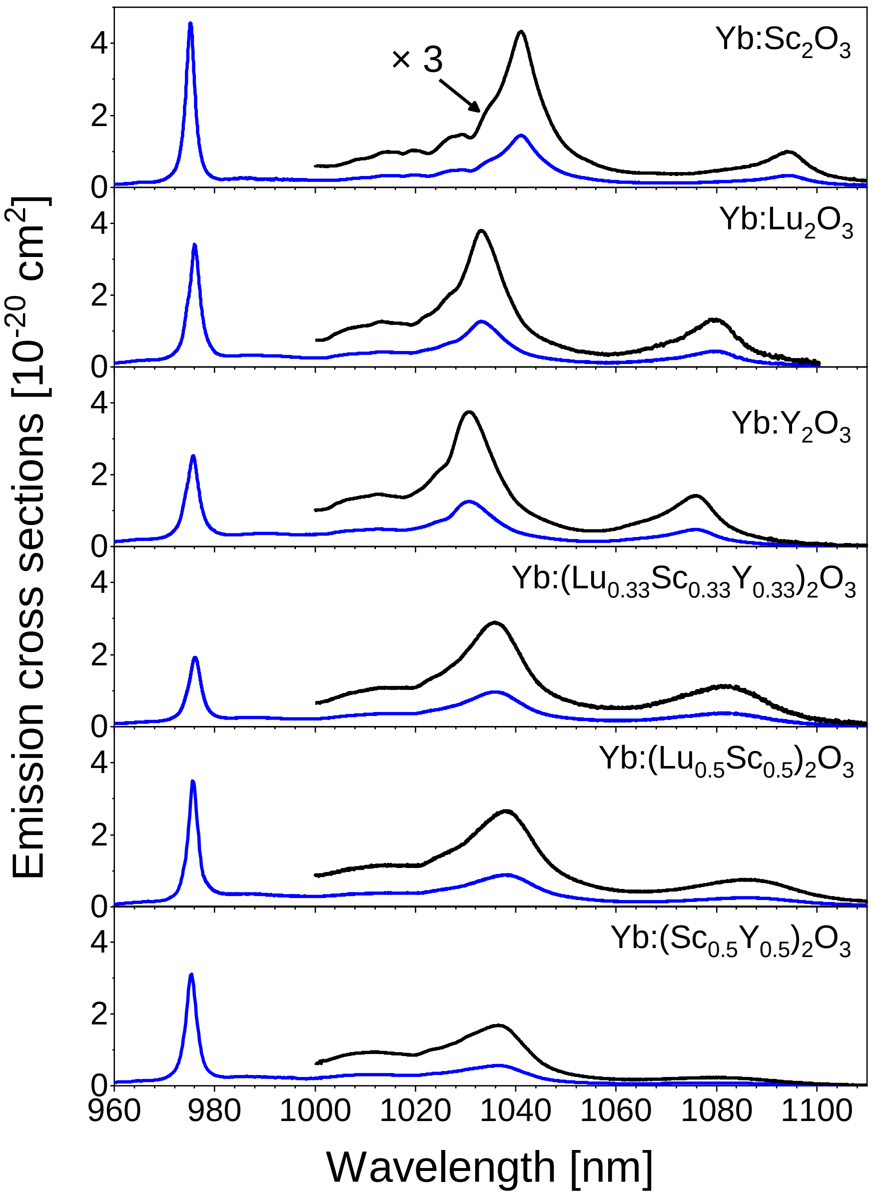


Kränkel *et al.*, Acta Cryst. B, **B77** (4), 550-558 (2021)

Czochralski growth of rare-earth doped $(Y_{0.5}Sc_{0.5})_2O_3$

- $YScO_3$ doped with different rare earth ions successfully grown
- Difficulties in diameter control solved for perovskite-structure rare-earth scandates (Uecker et al., J. Cryst. Growth, 310, 2649 (2008))
- Large ion size difference between Y^{3+} and Sc^{3+} causes strong inhomogeneous broadening
- Tailored spectra by compositional tuning



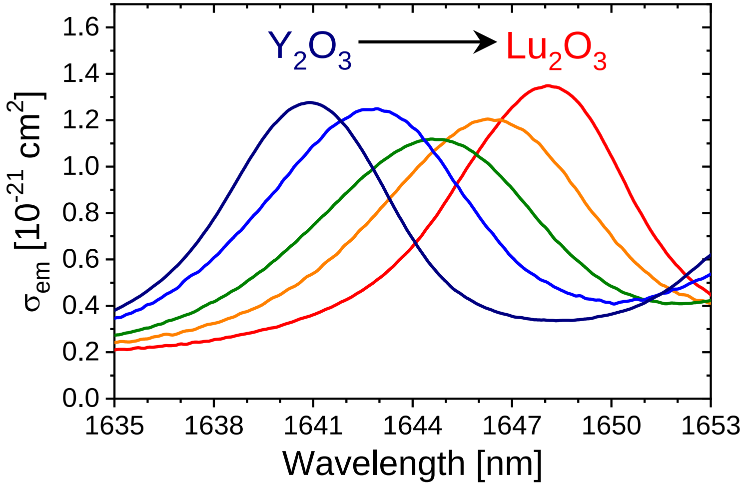


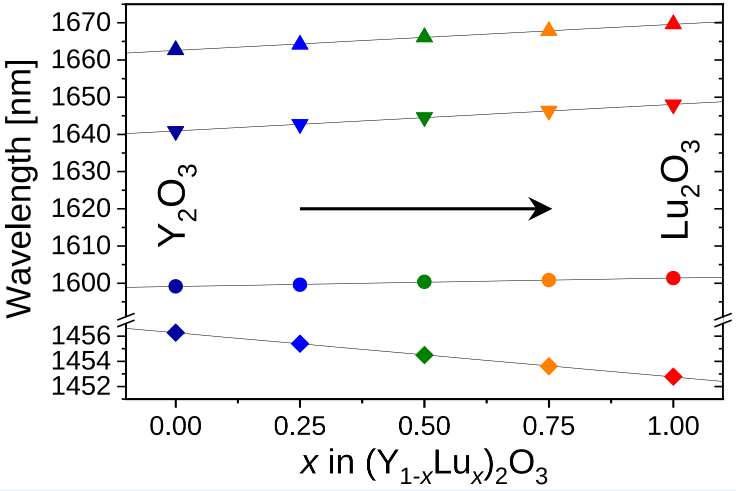
Kränkel *et al.*, Opt. Mat. Express **12** (3), 1074-1091 (2022)

Crystal field tuning

Crystal field tuning in Er:$(Y,Lu)_2O_3$

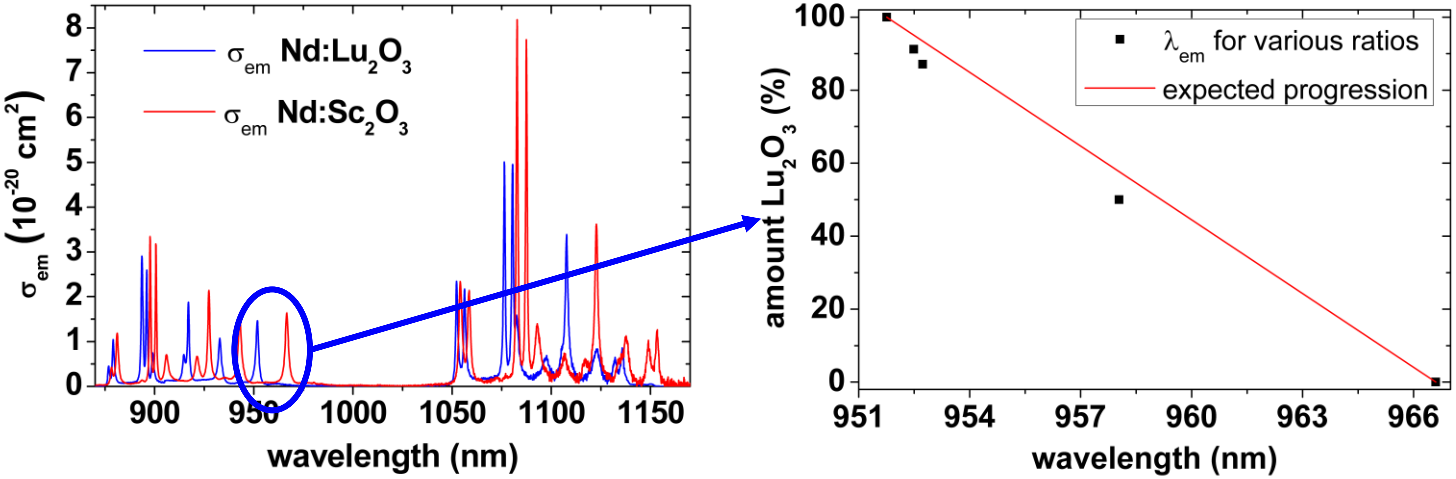
Objective: particular wavelength for LIDAR applications





Crystal field tuning in Nd:$(Sc,Lu)_2O_3$

Objective: tune emission to required pump wavelength

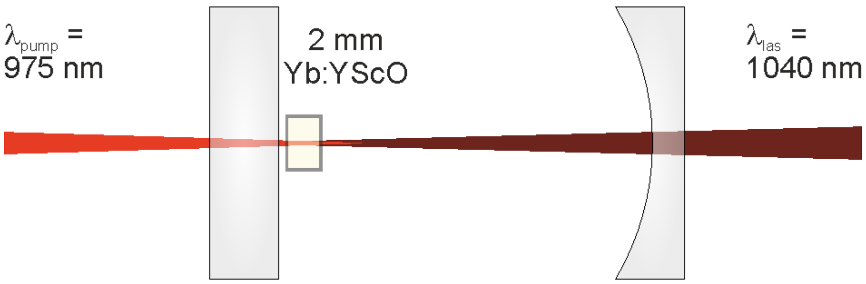


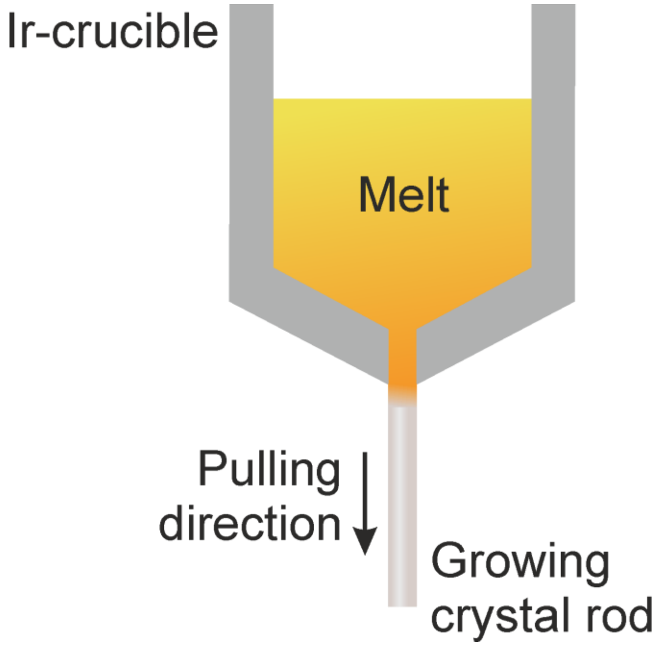
Many host materials enable crystal field tuning, e.g. (Y,Lu)AG, $(Lu,Y)VO_4$ or $(Lu,Y)AlO_3$

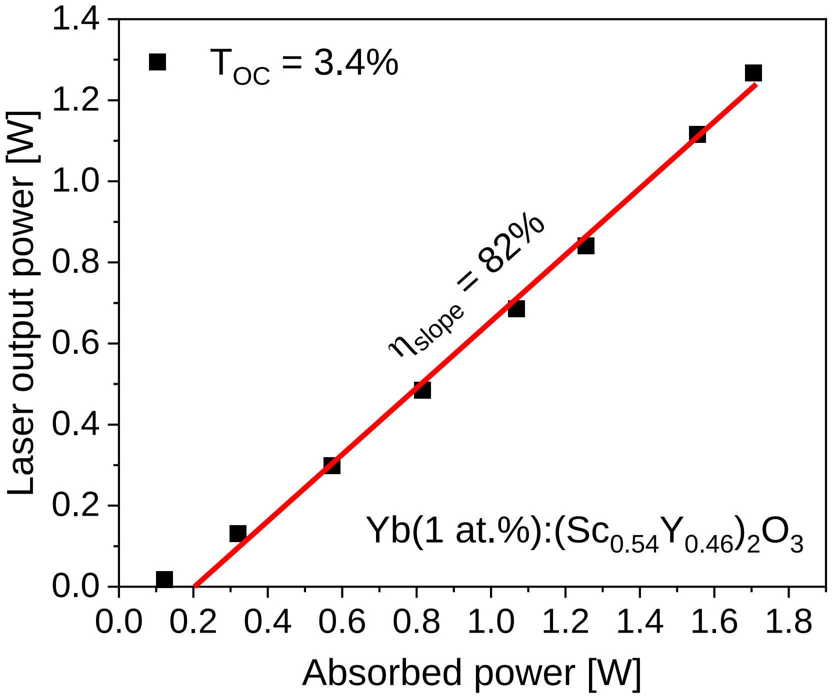
Laser operation of RE-doped mixed sesquioxide crystals

1 µm laser operation of Yb(1.0 at.%):$(Sc_{0.54}Y_{0.46})_2O_3$

- Micro-pulling down grown crystal
- OPSL-pumped at 975 nm
- Laser emission at 1040 nm
- 82% slope efficiency at T_{OC} = 3.4%
- 74% optical-to-optical efficiency

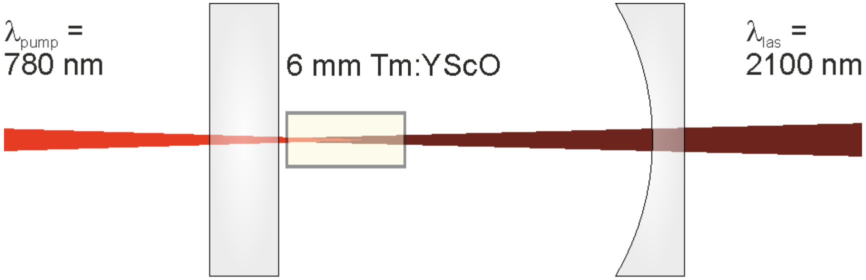


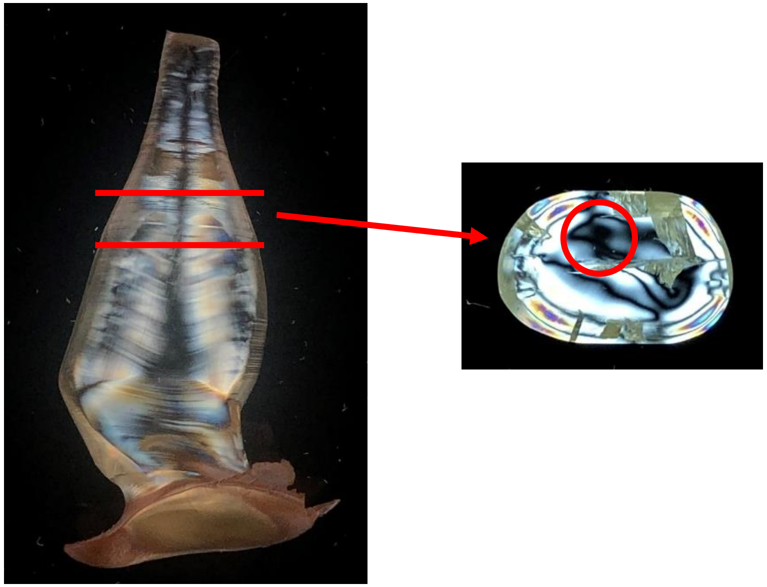


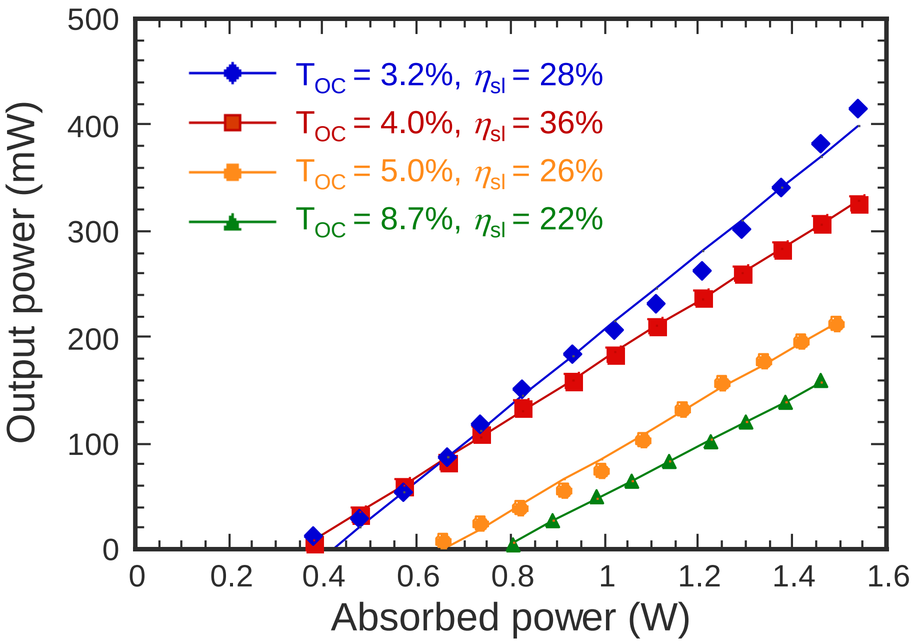


2 µm laser operation of Tm(2.2 at.%):$(Sc_{0.5}Y_{0.5})_2O_3$

- Czochralski-grown crystal
- Strong tensions in the laser sample
  → improve quality by reduced heat gradients
- Diode-pumped at 780 nm
- Laser emission at 2100 nm
- 36% slope efficiency at T_{OC} = 4.0%



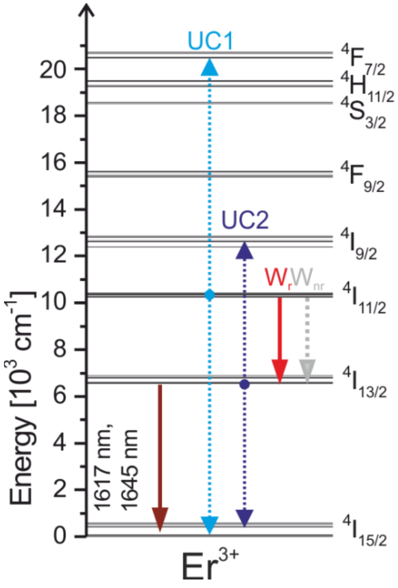


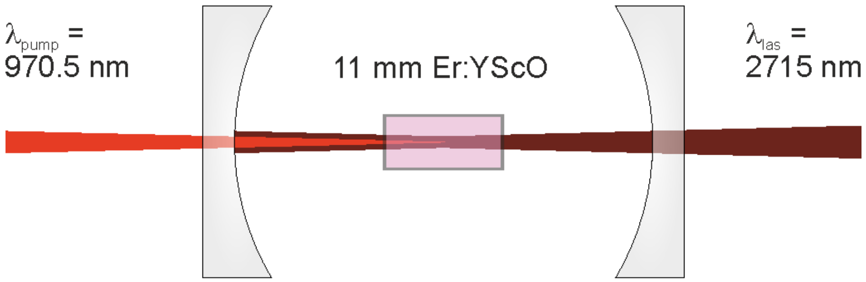


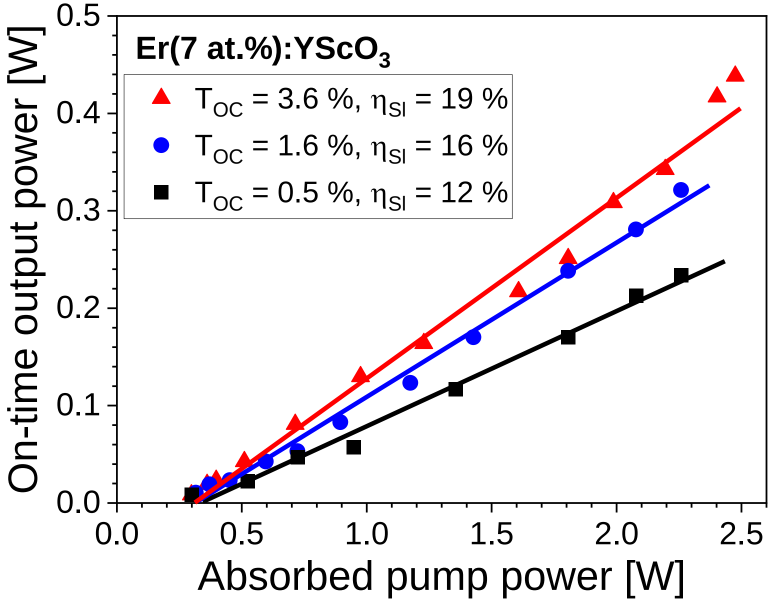
Laser operation of RE-doped mixed sesquioxide crystals

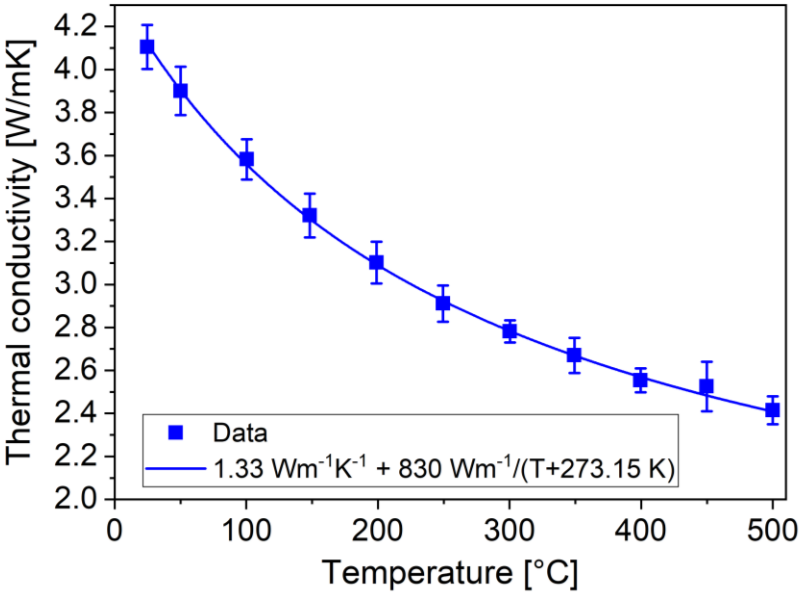
3 µm laser operation of $Er(7\ at.\%):(Sc_{0.54}Y_{0.46})_2O_3$

- OPSL-pumping at 970.5 nm
- Laser operation at 2.7 µm
- cw pumping instable,
  1:1 chopped at 480 Hz:
  19% slope efficiency
- Disordered lattice
  reduces thermal
  conductivity







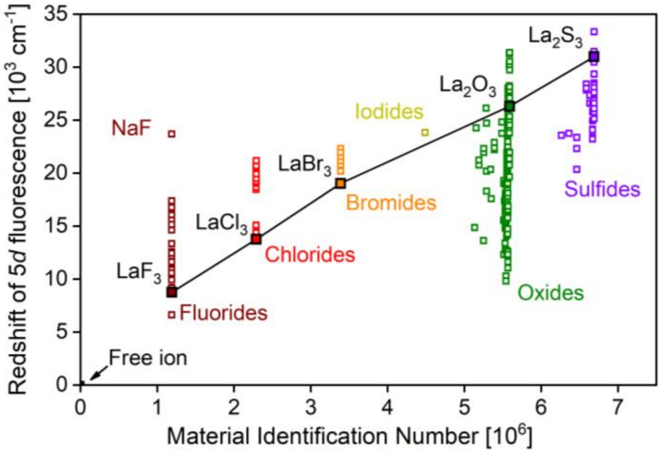


Conclusion & Outlook

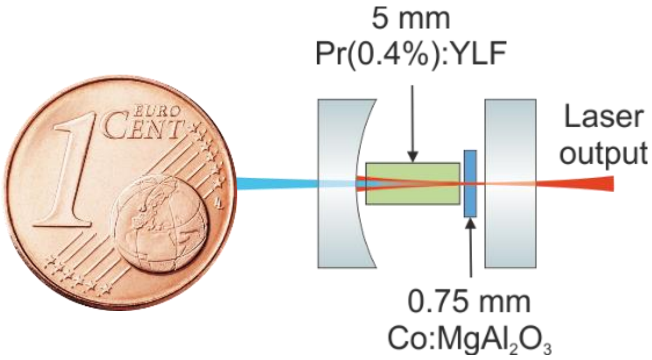
Tailored gain materials enable efficient lasing from the visible to the mid-IR range

Fluoride host materials for visible lasers

- High band-gap energies and low crystal field strengths suppress *4f*-*5d* ESA even at high photon energies
- Reduced thermal conductivity and mechanical strength impede power scaling
- Future aim: find suitable oxide materials



- Pr^{3+} is a mature laser ion which allows for highly efficient visible laser operation at several transitions
- Q-switched Pr^{3+}-lasers enable kW-peak power ns-pulses



- Tb^{3+} for blue pumped yellow lasers
  - Diode pumping is feasible
  - UV pumping enables use of thin gain media
  - Compatible with low-beam-quality UV diode modules

Watt-level UV-diode pumped Tb^{3+} DPSSLs within reach

Conclusion & Outlook

Tailored gain materials enable efficient lasing from the visible to the mid-IR range

Mixed sesquioxide host materials for infrared lasers

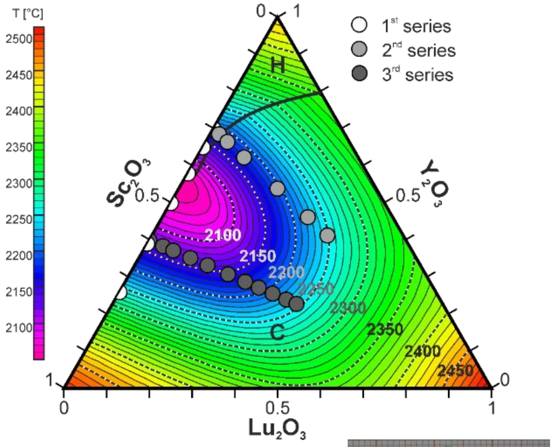
- Reduced melting points of mixed sesquioxides enable novel growth methods and future commercial availability:
  - Czochralski growth from iridium crucibles
  - µPD growth from iridium crucibles

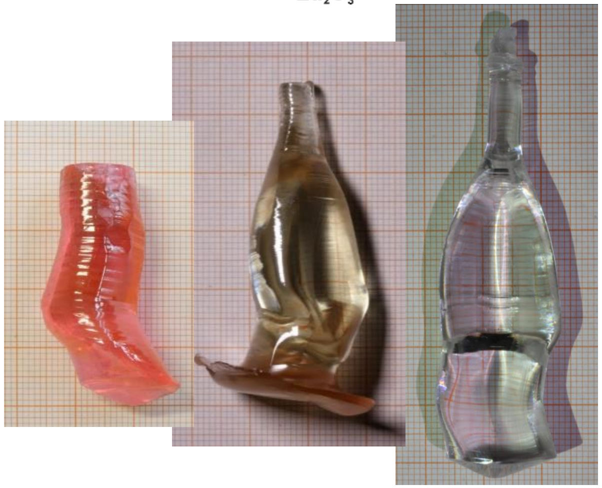
- Reduce stress in crystal by optimized isolation setup

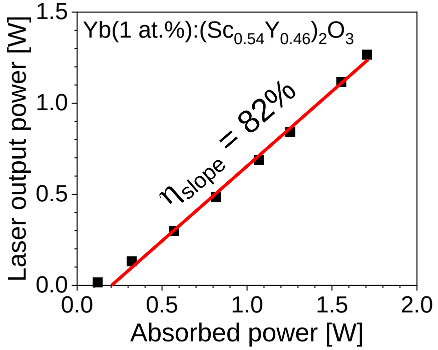
- Prospects for crystal field tuning, broad wavelength tuning and/or ultrashort pulse generation with Tm^{3+} and Ho^{3+}-doped mixed sesquioxides

- Er-doped mixed sesquioxides should enable wavelength tunable 3 µm laser operation

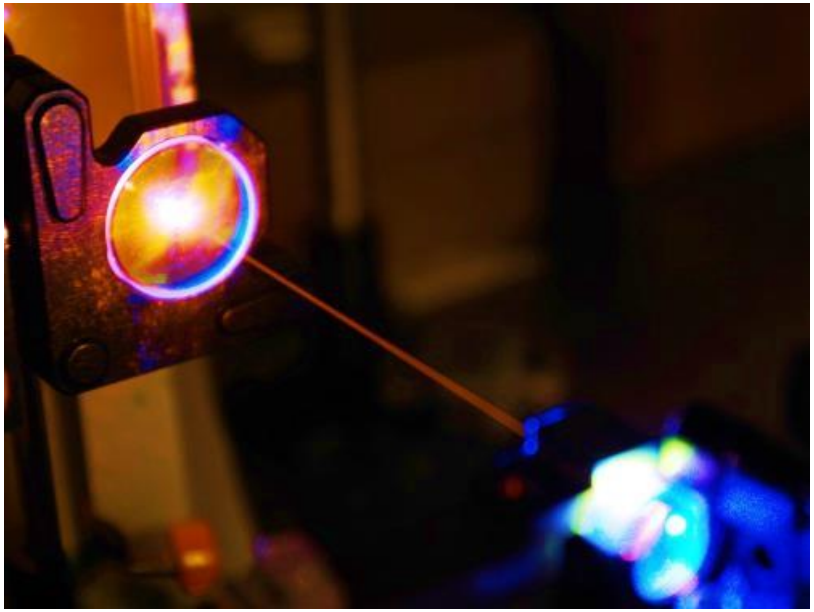
Mixed sesquioxides are highly promising host materials for ultrafast infrared lasers

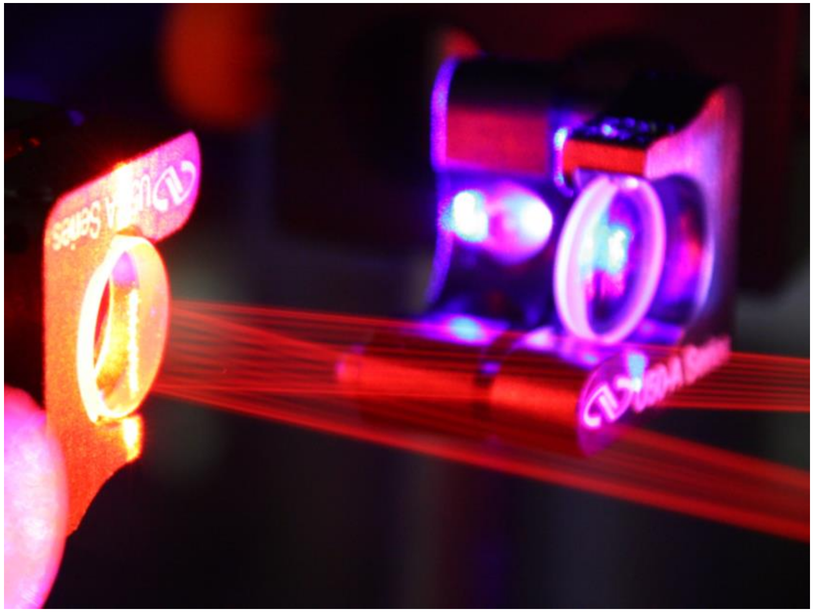






Thanks for your attention!





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