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Multi-disciplinary Approaches to Radiation Balanced Lasers:
Rare-Earths and Semiconductors in Disks, Fibers, and Microstructures

Advancing RBLs in rare-earth (RE)-doped solids

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UNM / New Mexico Consortium / LANL

Outline

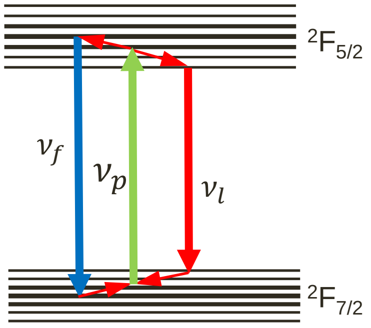
1. Mid-IR lasers in crystals using two-tone RBL
    - Single-dopant two-tone RBLs: Tm^{3+}, Er^{3+}
    - Co-doped two-tone RBLs: (Yb^{3+}, Nd^{3+}) and (Ho^{3+}, Tm^{3+})

2. Advanced approaches to RBL crystals
    - Precursor purification
    - Micro-pulling-down crystal growth
    - Bridgman crystal growth

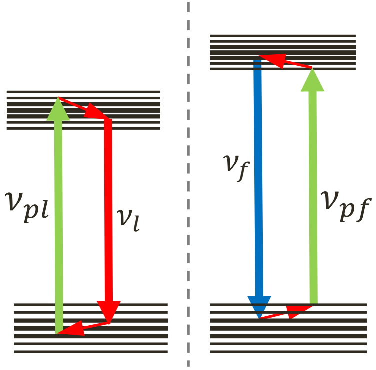
3. Advanced approaches to RBL fibers
    - Materials for RBL glass fibers
    - Micro-structured fibers for RBL
    - Fiber preform synthesis

4. Objectives

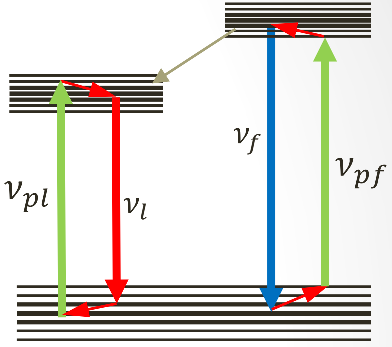
1. Mid-IR lasers in crystals using two-tone RBL



e.g. Yb^{3+}



Type-I (two-ion system)



Type-II (one-ion system)

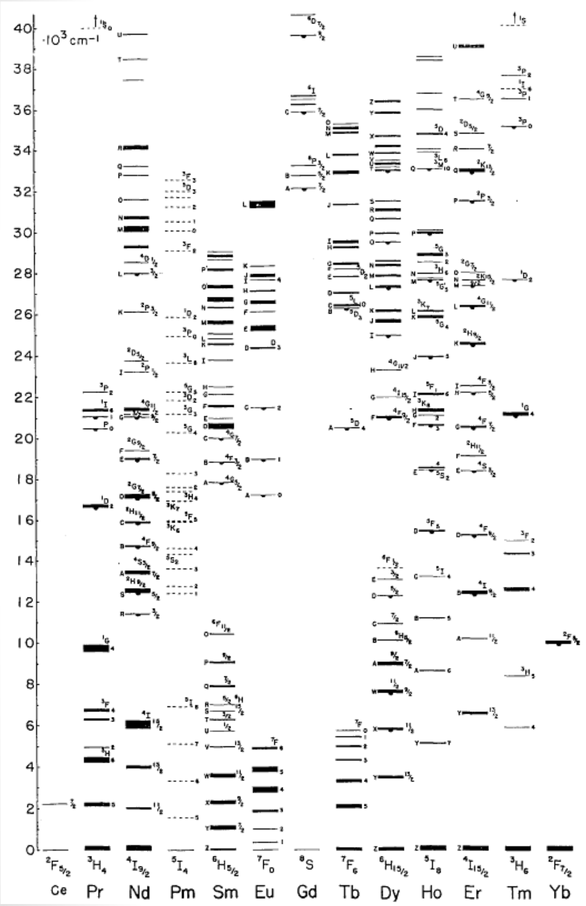
Traditional "single-tone" RBL:

- One pump laser (ν_p)
- Stimulated (lasing, ν_l) and spontaneous (cooling, ν_f) emission on the same transition
- Drawback: Constrained by the crystal-field transitions provided by the ion in the host material

Proposed "two-tone" RBL:

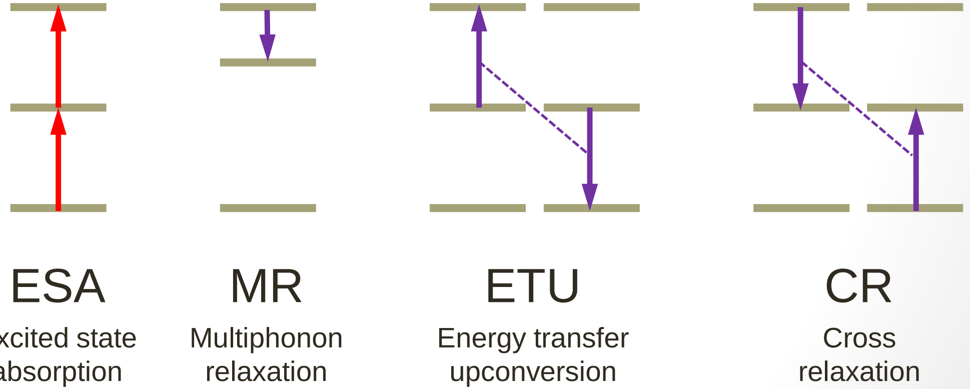
- Separate pump lasers for lasing (ν_{pl}) and cooling (ν_{pf})
- Stimulated (lasing, ν_l) and spontaneous (cooling, ν_f) emission on different transitions
- Two types: two-ion or one-ion systems
- Potential advantage: lasing and cooling partially decoupled → greater design freedom

1. Mid-IR lasers in crystals using two-tone RBL



Two-tone RBL adds complexity

- Yb^{3+} (and Ce^{3+}) are special: only one excited state!
- All other RE^{3+} ions have a large number of excited states
- Competing radiative and non-radiative processes may become active
- We will perform a comprehensive analysis of potential RE ions to identify promising two-tone RBL candidates



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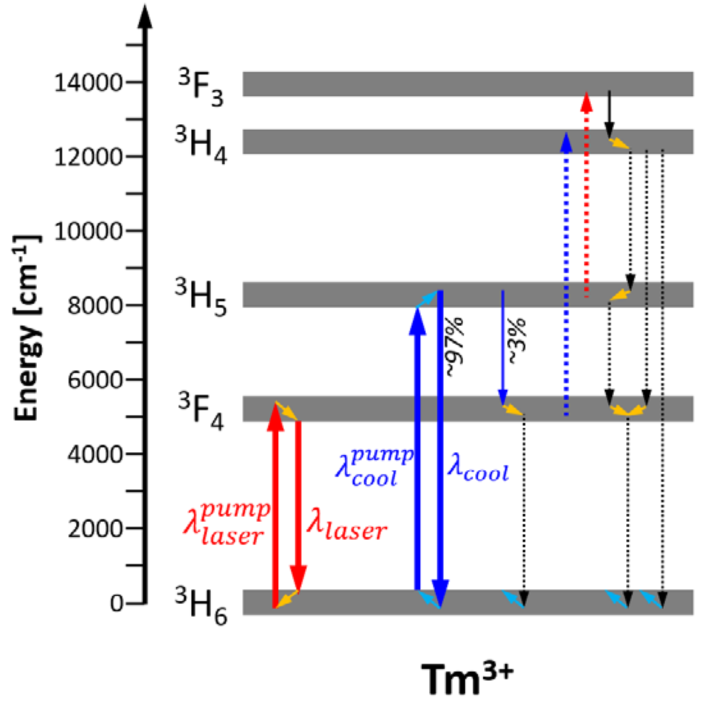
1. Mid-IR lasers in crystals using two-tone RBL

Type-II Candidate: Tm^{3+} two-tone RBL



- Cooling and lasing can be implemented using the $^3H_6 \leftrightarrow {}^3H_5$ (~1.2 μm) and $^3F_4 \leftrightarrow {}^3H_6$ (~2 μm) transitions, respectively
- The cooling / lasing transitions could also be reversed. Cooling on $^3F_4 \leftrightarrow {}^3H_6$ has been demonstrated by UNM
- Multiphonon relaxation of $^3H_5$ must be negligible → low-energy phonon material (e.g. fluoride)
- $^3H_5$ has a high branching ratio to the ground state
- Potential for relatively high 300 K cooling efficiency ($\sim 2.5kT/\Delta E$): ~6% for $^3H_5$ and ~10% for $^3F_4$
- Competing processes are ESA and MR (heating)

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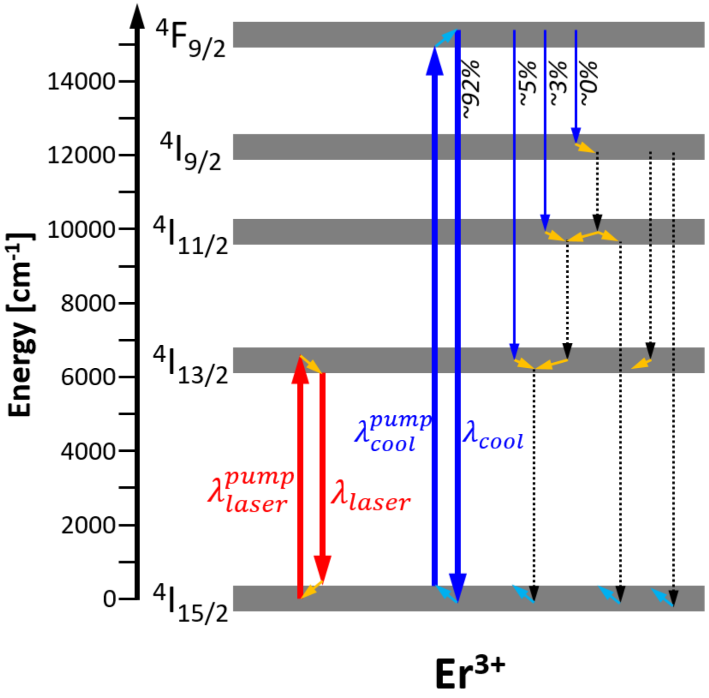
1. Mid-IR lasers in crystals using two-tone RBL

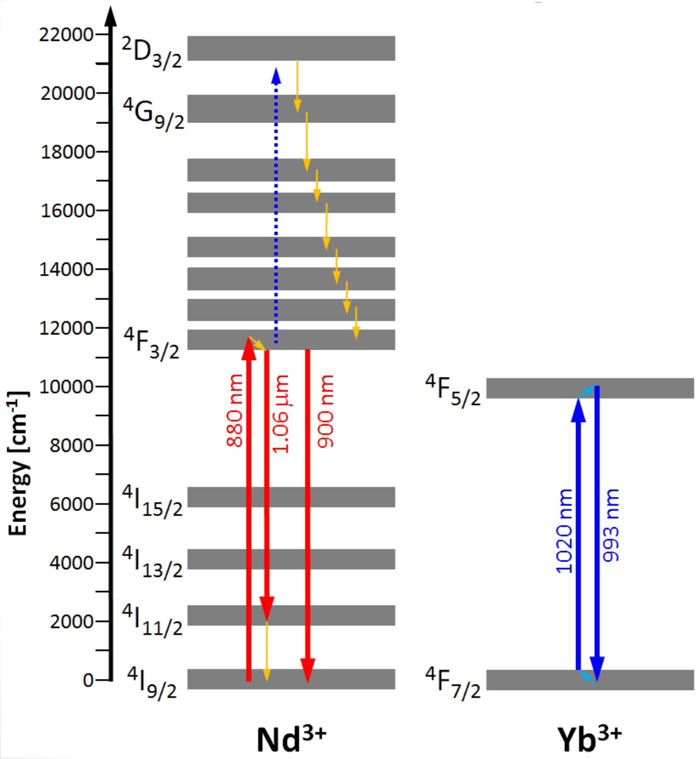
Type-II Candidate: Er^{3+} two-tone RBL



- Lasing possible on $^4I_{13/2} \rightarrow {}^4I_{15/2}$ (~1.55 μm) with pumping at ~1.48 μm
- Cooling on the $^4I_{13/2} \leftrightarrow {}^4I_{9/2}$ (~865 nm) transition has been demonstrated, but it has a low cross section (10× lower than Yb^{3+}) and a low branching ratio to the ground state (~85%)
- Alternatively, the $^4I_{13/2} \leftrightarrow {}^4F_{9/2}$ (~650 nm) transition has a cross section comparable to Yb^{3+} and a ~92% branching ratio to the ground state. However, relatively low cooling efficiency (~3.3% at 300 K) .
- Cooling and lasing transitions could be reversed to achieve higher cooling efficiency
- Competing processes are ESA and MR (heating)

1. Mid-IR lasers in crystals using two-tone RBL

Type-I Candidate : Nd^{3+}, Yb^{3+} two-tone RBL



- $Yb^{3+} \leftrightarrow Nd^{3+}$ energy transfer (and associated heating) expected to be relatively inefficient
- Cooling on Yb^{3+} well established
- Lasing on the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ (~900 nm) transition with pumping at ~880 nm
- Potential for ESA of the pump laser on Nd^{3+} causing heating via subsequent MR. Depends on specific material.

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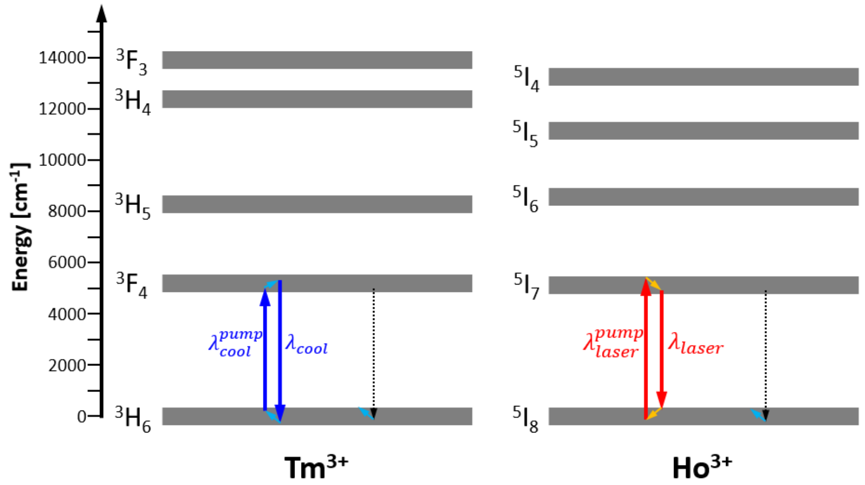
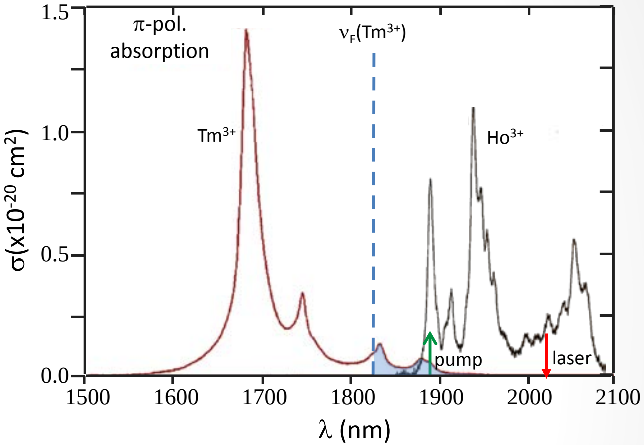
1. Mid-IR lasers in crystals using two-tone RBL

Type-I Candidate : Tm^{3+}, Ho^{3+} two(?)-tone RBL





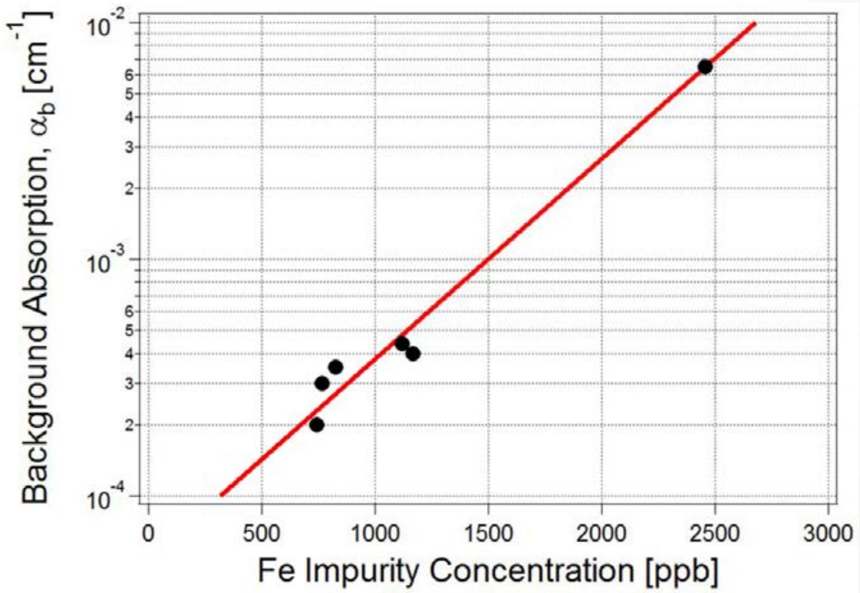
- Interesting case: lasing on Ho^{3+} and cooling on Tm^{3+} might each be driven by a single pump laser at ~1890 nm
- The balance between lasing and cooling could be tuned by varying the pump laser wavelength around ~1890 nm
- Potential for high cooling efficiency on Tm^{3+} (~10% at 300 K)

2. Advanced approaches to RBL crystals

Precursor purification

- Any RBL material has to first be a “cooling-grade” material
- Requires host material with (1) low intrinsic multiphonon relaxation and (2) high purity to minimize background absorption
- Primary impurities are believed to be transition metals such as Fe, Cu, Co, Ni, etc. present in commercial starting materials *and* introduced during sample preparation and growth
- Impurities absorb pump laser and decay non-radiatively (heating)
- Purification of precursors prior to growth is necessary in order to maximize the cooling efficiency

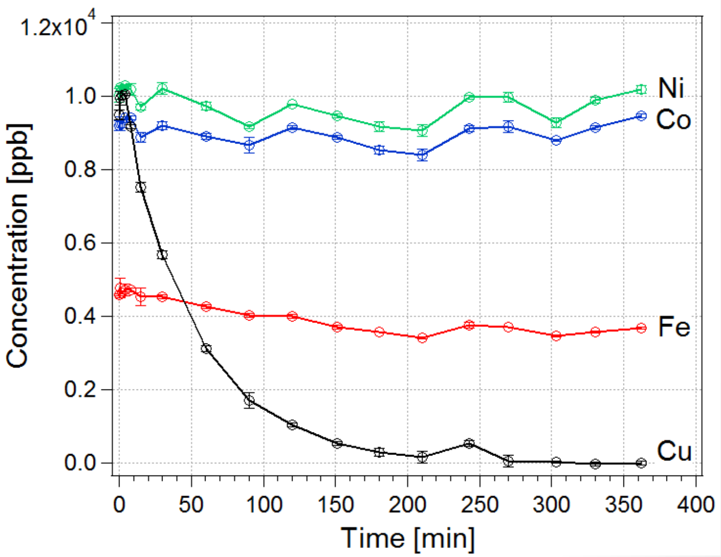
Correlation of background absorption and Fe impurity concentration in YLF:Yb samples (Melgaard et al, UNM)



2. Advanced approaches to RBL crystals

Precursor purification

- Preliminary successes with electroplating of impurities:
  - o Demonstrated electrochemical reduction of copper impurities in a concentrated YCl_3 aqueous solution (LANL)
  - o Electrochemical processing of fluoride melts (ZBLAN) shown to improve laser cooling performance (Fajardo et al)
- These purification methods are particularly suited for small batches (~10 g)
- Use in conjunction with small-scale micro-pulling-down and Bridgman crystal growth methods
- Further development is needed to identify a suited process

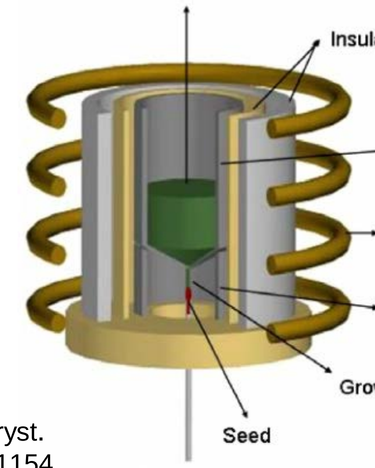


Concentration of transition metal impurities in a solution of YCl_3 as a function of electroplating time (LANL).

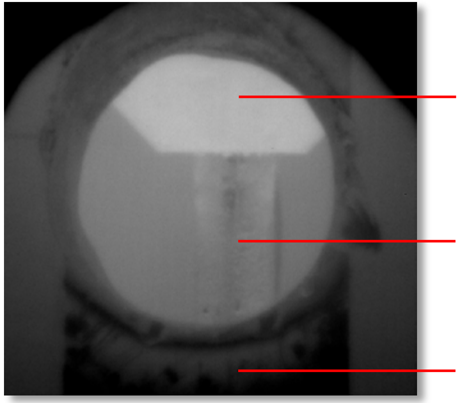
2. Advanced approaches to RBL crystals

Micro-pulling-down (μPD) crystal growth

- Crystal growth *downward* from the melt, which flows through a small hole at the bottom of the stationary crucible.
- μPD growth can start from relatively small (1-5 g) quantities of starting material (compare to >100 g for traditional Czochralski growth)
- Small starting material quantities are (1) ideal for material screening and (2) advantageous to maintaining high purity



from Alshourbagy et al, Cryst. Res. Technol., 41 (2006) 1154.



Growth of small-diameter (~1 mm) LiSAF crystal

2. Advanced approaches to RBL crystals

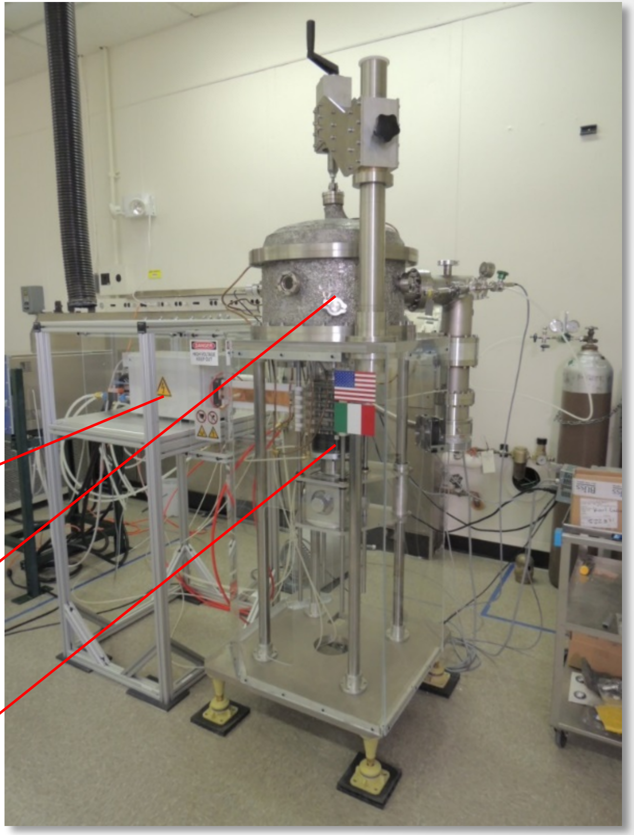
Micro-pulling-down (μPD) crystal growth

- LANL collaboration with Prof. Mauro Tonelli (University of Pisa, Italy) has established a μPD crystal growth facility
- This infrastructure is available to the MARBLE project for the growth of RE-doped RBL materials
- Prof. Tonelli is a MARBLE project collaborator and will continue to contribute to the refinement of the system
- System offers flexibility in growing a wide range of materials under inert atmosphere

RF generator

Growth chamber

Translation stage with overpressure capability



2. Advanced approaches to RBL crystals

Micro-pulling-down (μPD) crystal growth

- μPD growth is ideally used for systems that melt congruently, e.g. $LiLuF_4$.
- The crystal diameter / shape is determined by the design of the crucible:
  - Circular hole for the growth of small-diameter crystal fibers (0.3 – 2.0 mm). Maximum diameter limited by surface tension and viscosity of melt.
  - Addition of die enables growth of larger diameters (~5 mm) and non-circular shapes (plates, tubes, etc.)
  - Need to design glassy carbon crucible with die for the μPD bulk growth of RE-doped RBL crystals

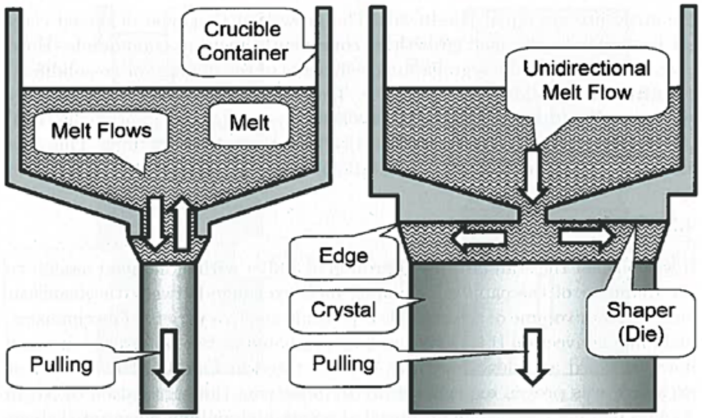


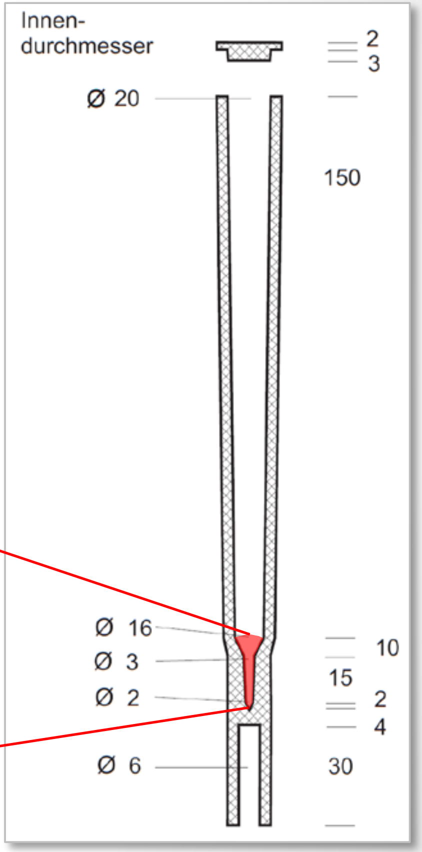
Fig. 4.5. *μ*-PD system for fiber (*left*) and bulk (*right*) crystal growth. Note ratio of crystal to capillary diameters

from *Shaped Crystals*, Fukuda, Chani (Eds.), Springer 2007

2. Advanced approaches to RBL crystals

μPD system used for Bridgman crystal growth

- Crystal growth *upward* in a crucible that is translated downward in a stationary temperature gradient.
- Bridgman growth is suited for systems that melt incongruently ($YLiF_4$ is borderline incongruent) and/or suffer from evaporative losses (e.g. LiF evaporation from LiSAF melts)
- The LANL μPD system allows translation of a Bridgman crucible through the temperature gradient established by the RF coil
- Single crystals of LiSAF have been grown successfully this way. Additional process development is necessary (crucible shape, temperature gradient)



Bridgman growth of LiSAF crystal from 1 gram of starting material using the LANL μPD system

3. Advanced approaches to RBL fibers

Glassy materials for fiber-based RBL

- "Single-tone" RBL
  - One material
    e.g. Yb^{3+}
    - Traditional single-mode optical fiber
    - $\Delta E \approx 9800$ cm^{-1}
      $\hbar\omega_{max} < 850$ cm^{-1}
- "Two-tone" RBL
  - Potentially two materials
    e.g. Yb^{3+} (cooling) and Nd^{3+} (lasing)
    - Micro-structured optical fiber
    - $\Delta E < 5000–6000$ cm^{-1}
      $\hbar\omega_{max} < 420 - 500$ cm^{-1}

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3. Advanced approaches to RBL fibers

Materials for RBL glass fibers: Heavy-metal oxides for “single-tone” RBL

- **Oxide glasses**:
  - o Excellent properties for optical fiber fabrication
  - o SiO_2-based glass has high phonon energies (~1100 cm^{-1}) → significant multiphonon relaxation → reduced quantum efficiency → reduced laser-cooling efficiency
  - o Some oxide glasses have lower phonon energies: e.g. TeO_2-based glass (790 cm^{-1}) → may offer sufficiently low multiphonon relaxation to enable efficient “single-tone” RBL in Yb^{3+}

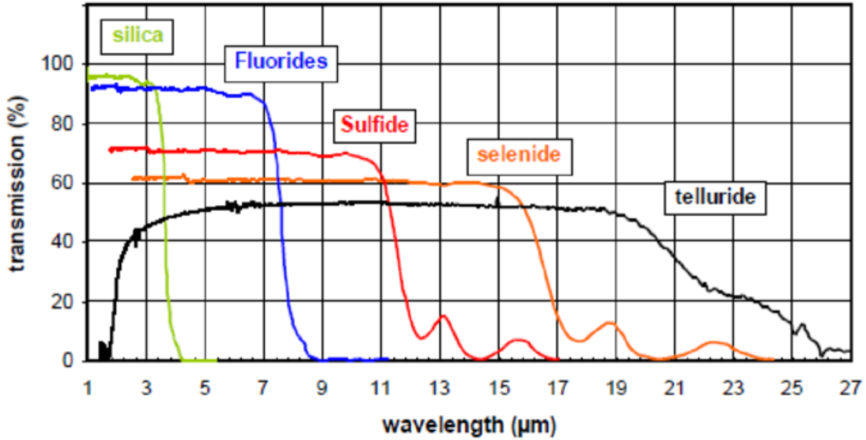
- **Fluorides glasses** (e.g. ZrF_4 based, such as ZBLAN):
  - o Moderate phonon energies (350–600 cm^{-1})
  - o Prone to crystallization due to small T_x-T_g range
  - o Prone to oxidation
  - o Drawing of low-loss fibers is challenging

- **Heavy halide glasses** (Cl, Br, I):
  - o Low phonon energies (100–260 cm^{-1})
  - o Extremely hygroscopic
  - o Only chlorides form glasses in very limited compositional ranges
  - o Chloride glasses have very low T_g

3. Advanced approaches to RBL fibers

Materials for RBL glass fibers : Chalcogenides for two-tone RBL

- Relatively low phonon energies (300-400 cm^{-1}); lower than fluorides
- Band edge at ~500 nm (sulfides) and ~800 nm (selenides) enables active systems in the near IR
- High refractive index: ~2.35 (sulfides) and ~2.72 (selenides) can be both detrimental (low escape efficiency) and beneficial (tailoring of temperature gradient)
- Candidate material Ga_2S_3-La_2S_3 (~320 cm^{-1})
- Can be drawn into solid and micro-structured fibers of high optical quality
- Laser cooling has not yet been investigated in these systems
- Fabrication must mitigate OH, H_2S, H_2Se, H_2O, and C-H impurities



Infrared transmittance of different glasses (Cui et al, Molecules, 2013)

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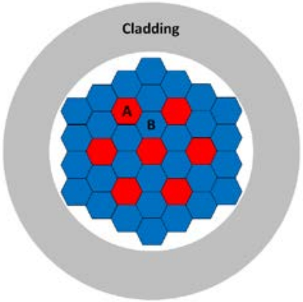
3. Advanced approaches to RBL fibers

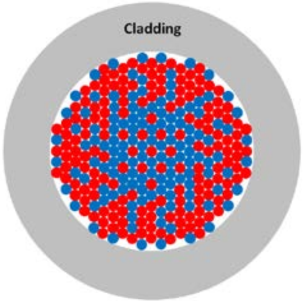
Micro-structured fibers for two-tone RBL

- **Approach:** Create an optical fiber with a micro-structured core which consists of a laser-cooling material and a lasing material to create an RBL overall

- This allows for:
  - o separate optimization of the laser-cooling and lasing materials
  - o spatial tailoring of the transverse temperature profile in the fiber

- The microstructure is chosen to be:
  - o sub-wavelength such as to appear homogeneous for the optical mode
  - o large compared to typical RE-RE energy transfer distances (<100 nm), allowing the two materials to exist side by side without much interaction

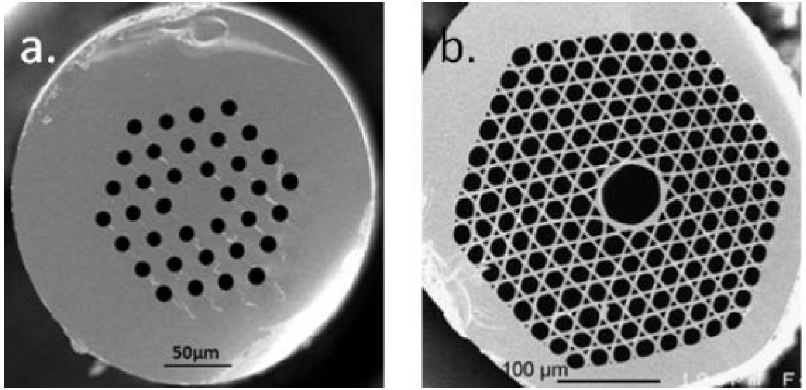




3. Advanced approaches to RBL fibers

Fiber preform synthesis

- Fabrication of micro-structured fibers by the “stack-and-draw” method:
    1. Fabrication of glass rods (composition, cross-sectional shape) e.g. by extrusion
    2. Manual assembly of rods to create a preform
    3. Drawing of preform to the final fiber diameter
- Suitable for solid or holey fibers
- Sufficient overlap of the T_x-T_g ranges must exist when using different materials in the preform
- Extrusion method may enable control of the composition in the longitudinal direction



Selenide holey fibers produced by the stack-and-draw method (Cui et al, 2013)

4. Objectives

Project Year 1:

- Grow Tm and Er doped fluoride crystals (single-dopant 2-tone RBL)
- Explore electrochemical purification methods
- Fabricate and characterize RE-doped chalcogenide and heavy-metal oxide glasses
- Procure load frame for the extrusion of glass preforms

Project Year 2:

- Grow YLF:Yb,Nd and YLF:Tm,Ho crystals (co-doped 2-tone RBL)
- Refine electrochemical purification methods
- Fabricate Yb-doped heavy-metal oxide glass preform
- Develop chalcogenide glass extrusion process

Project Year 3:

- Explore large-diameter μPD growth from purified starting materials
- Synthesize and characterize target chalcogenide glass
- Fabricate micro-structured chalcogenide glass preform

Thank you!

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