Graduate School of Basic Sciences
"Galileo Galilei"- Physics
XVIII cycle PhD course

Crystal Growth and Spectroscopy of Rare Earth Ions Doped Crystals For Quantum Computing Application

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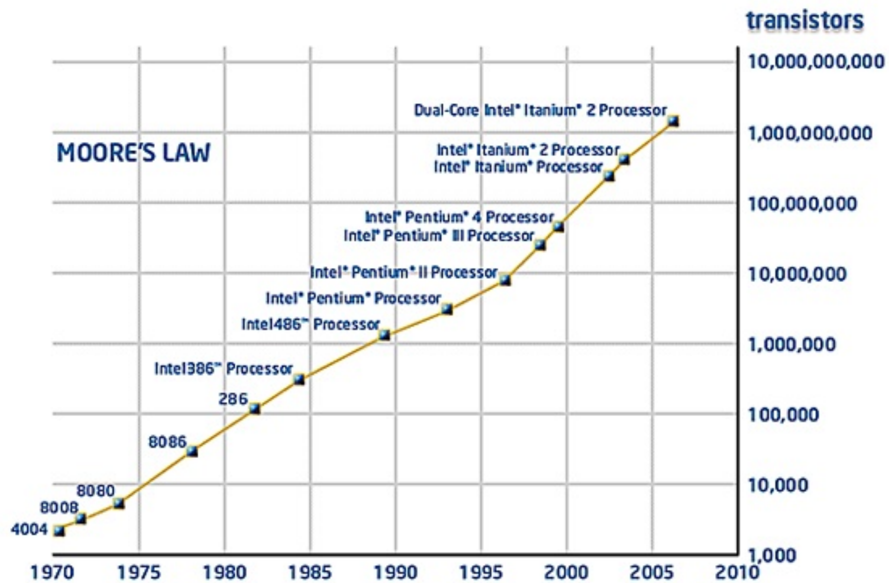
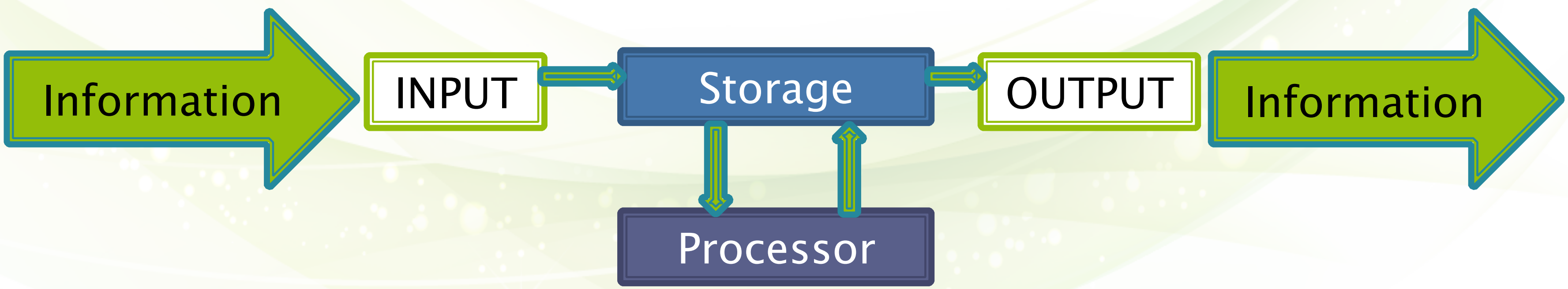
Background: Classical Computing



Units of Processor:
Electronic valve (1911-1946)
Transistor (1947-1958)
Integrated Circuit (1959-1970)
Large Scale IC (1971-today)

Limit:
Thermal dissipation
Quantum tunneling effect



Von Neumann architecture scheme

Background: Quantum Computing

- Classic bits

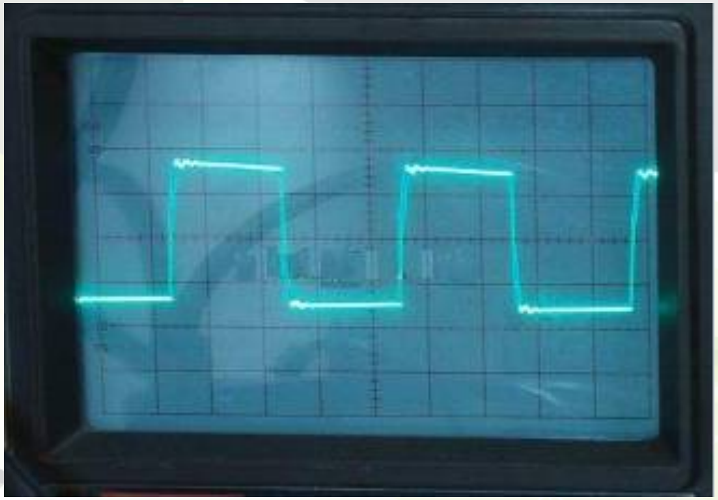
  2 possible voltages encode one bit

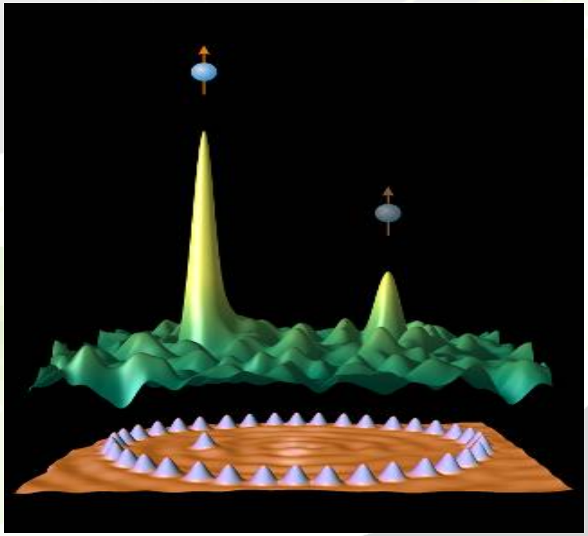
  0 or 1

- Quantum bits

  2 level system encodes one bit

  $|0\rangle$ or $|1\rangle$ or $a|0\rangle+be^{iwt}|1\rangle$





Background: system for quantum computing

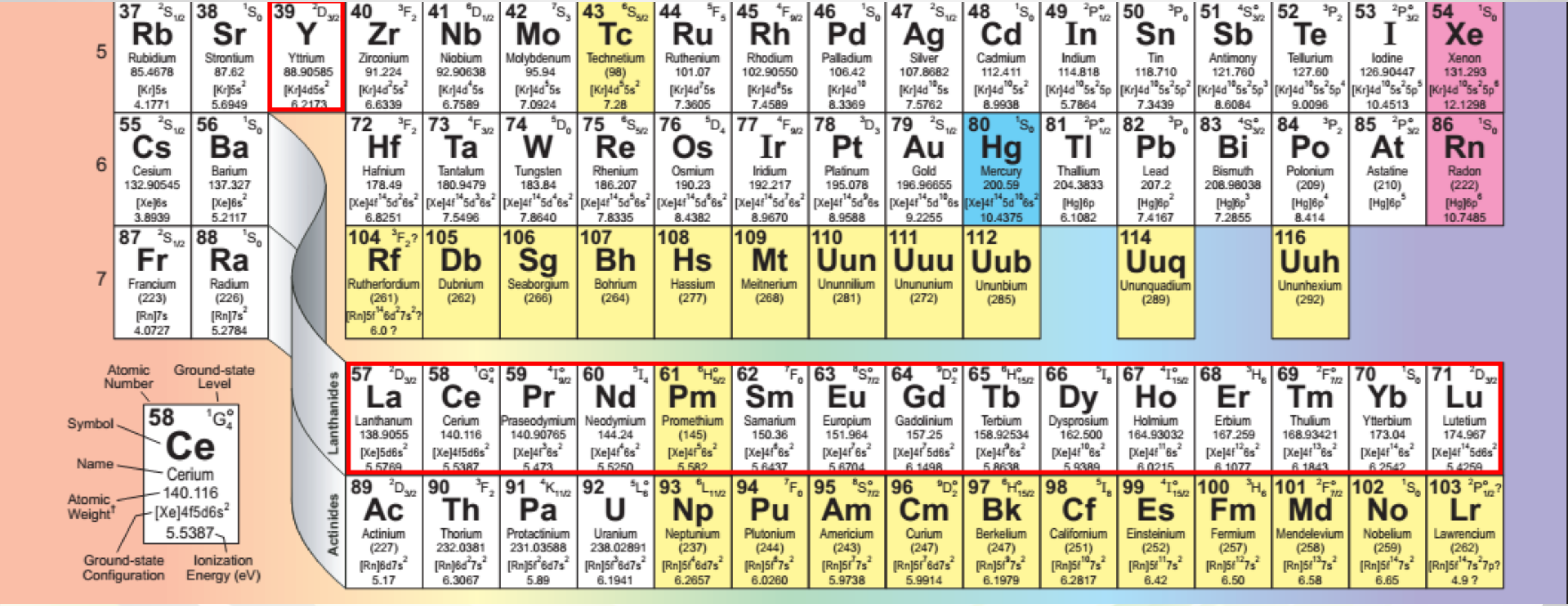
Requirement of a system for quantum computing application:

- The system must have well defined qubits that can be stored for a particular length of time.
- There must be a universal set of gates.
- The system can be easily initialized to a particular state.
- The qubit can be read out and the reading process will not destroy the quantum state of the stored information.

Several systems that have been used for quantum computing application.

- Electron spin system
- Nuclear spin system
- Trapped ions system
- Quantum dots
- Optical cavity
- Rare earth doped solids

Background: The Properties of Rare Earth Elements



	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
4d	10	10	10	10	10	10	10	10	10	10	10	10	10	10	10
5p	6	6	6	6	6	6	6	6	6	6	6	6	6	6	6
6s	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
4f		1	3	4	5	6	7	7	9	10	11	2	13	14	14
5d	1	1						1							1

Background: the properties of rare earth elements- energy structures of free ions RE^{3+}

Hamiltonian of free rare earth ions:

$$H = T + V_{en} + V_{ee} + V_{so}$$

$$= -\frac{\hbar^2}{2m}\sum_{i=1}^{N}\nabla_i^2 - \sum_{i=1}^{N}\frac{Ze^2}{r_i} + \sum_{i<j}^{N}\frac{e^2}{r_{ij}} + \sum_{i=1}^{N}\zeta(r_i)\vec{s}$$

The four termers of the formula represent *kinetic energy (T), the interaction between electrons and nucleus (V_{en}), the interaction between electrons (V_{ee}) and spin orbit interaction (V_{so}).*

The non spherically symmetric part of (V_{ee}) and (V_{so}).

Split of the degeneracy to $^{2S+1}L_J$

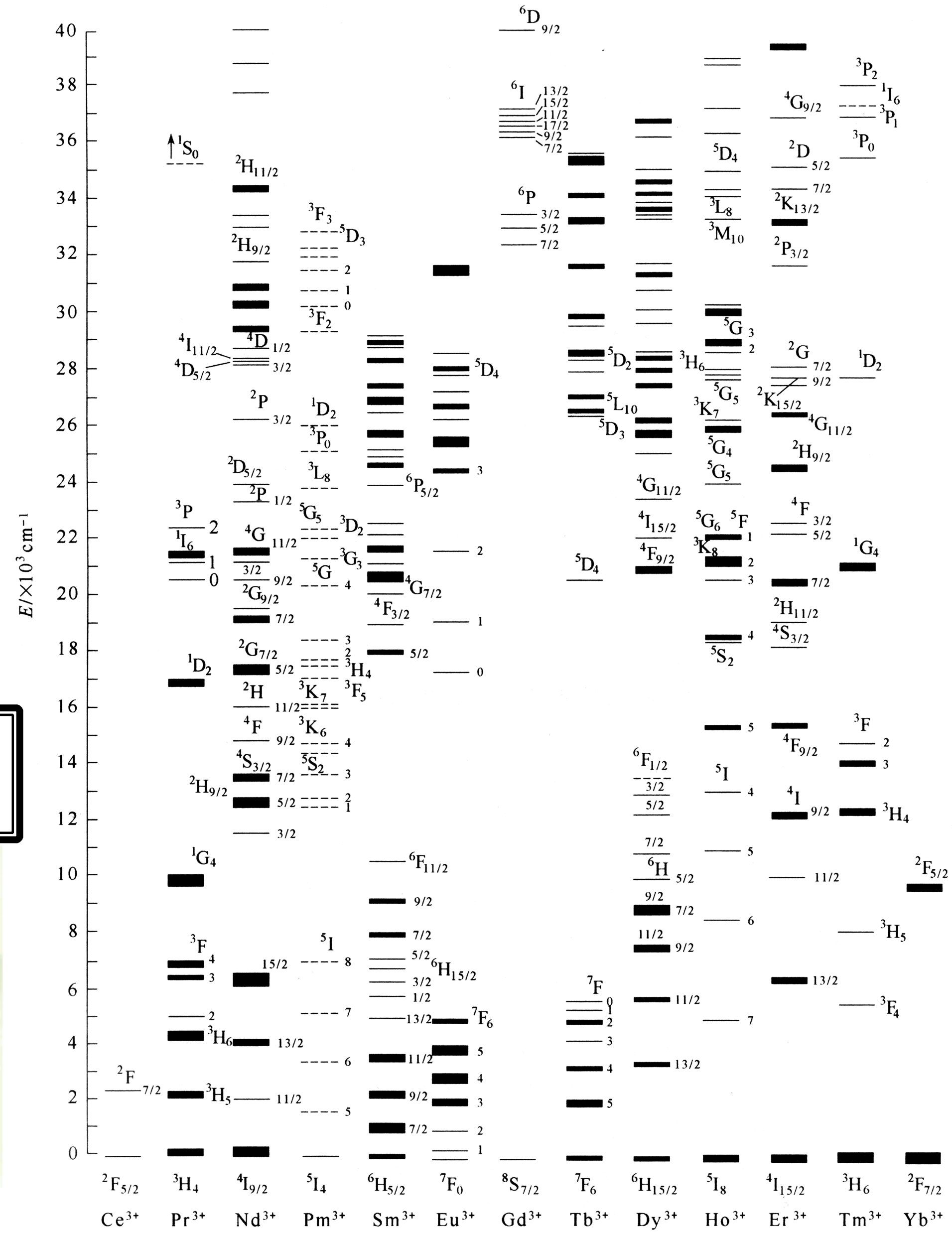


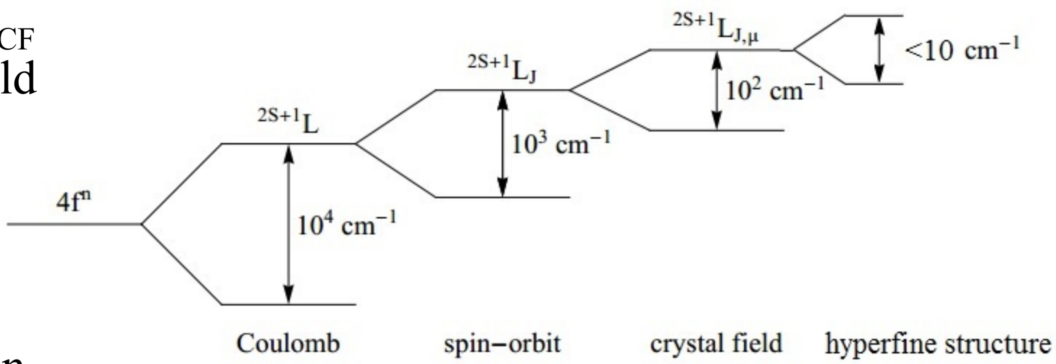
Figure 1. Energy Levels of rare earth ions in $LaCl_3$ (Dieke Diagram)

***Appl. Opt*, 675, 2, 1963**

Background: Properties of rare earth elements-ions doped in crystals

$$H = H_{FI} + H_{CF} + H_{HF} + H_Q + H_z + H_Z$$

The first two terms H_{FI} and H_{CF} are the free ion and crystal field Hamiltonians.
H_{HF}: the hyperfine interaction
H_Q: the nuclear electric quadrupole interaction
H_z: nuclear Zeeman interaction
H_Z: electric Zeeman interaction.



Spectra measurement at room temperature

Spectra measuremenat at liquid helium temperature

Spectra analysis with high resolution at liquid temperature with strong magnetic field

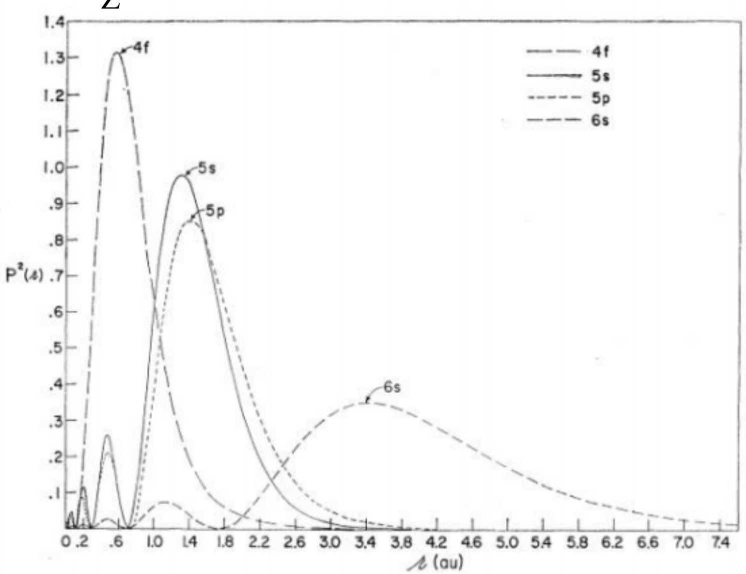


Figure 2, the electron density of the 4f, 5s, 5p and 6s orbitals in Gd atom

Background: Energy transfer in rare earth doped crystals

Excited States

- The optical lifetime T_1.

$$T_{1a} = \frac{1}{\sum_b W_{ab}^R + \sum_b W_{ab}^{NR}}$$

W_{ab}^R the radiative decay rate

W_{ab}^{NR} *non-radiative decay rate*

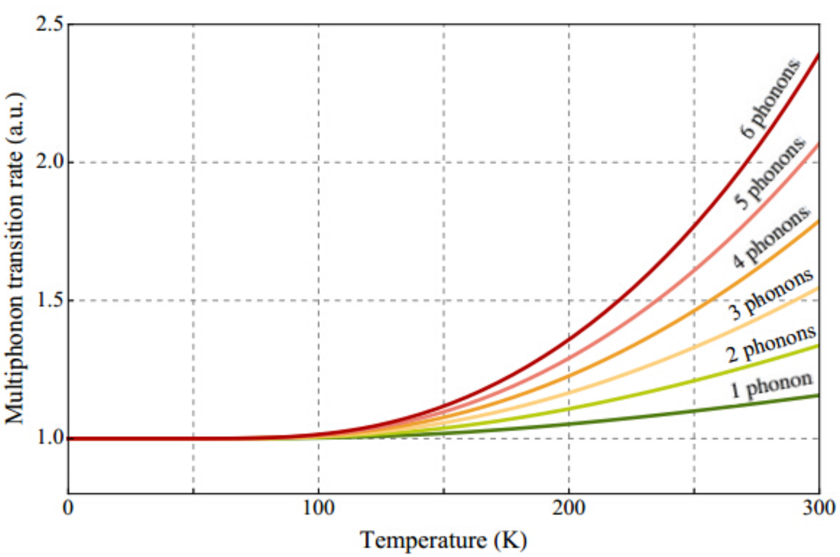


Figure 3. Decay rate via temperature for single and multiphonon relaxation.

Hyperfine structure

- The coherence lifetime T_2.

$$\Gamma_h = \frac{1}{\pi T_2} = \frac{1}{2\pi T_1} + \Gamma_\phi$$

- Homogeneous bandwidth Γ_h
- Inhomogeneous bandwidth Γ_{ih}

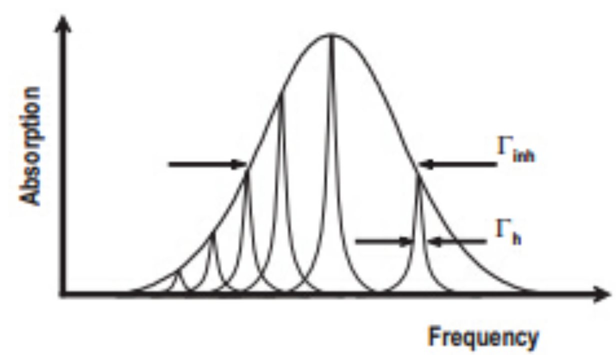


Figure 4. Inhomogeneous linewidth for a resonant optical material and homogeneous linewidth for individual groups of ions.

Background: photon echo techniques for quantum memory application

- Photon echo (two level system):

$$E_{echo}(t) \propto \int E_{write}(w) E_{data}(w) E_{read}(w) e^{iwt} dw$$

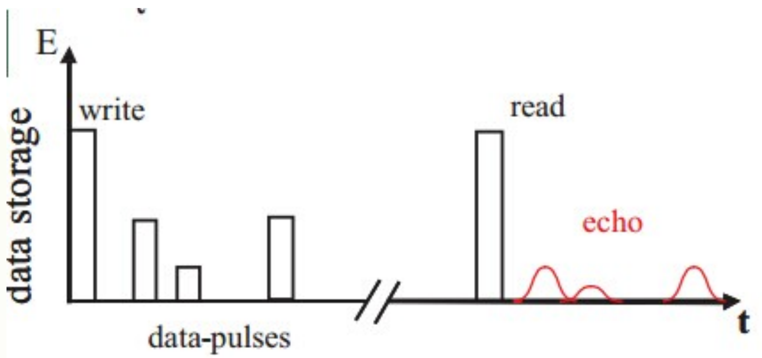


Figure 5. scheme for photon echo data storage.
***Optics Communications* 247 (2005) 393–403**
***Laser & Photon. Rev*. 4, (2010) 244-267**

- Controlled reversible inhomogeneous broadening, CRIB:

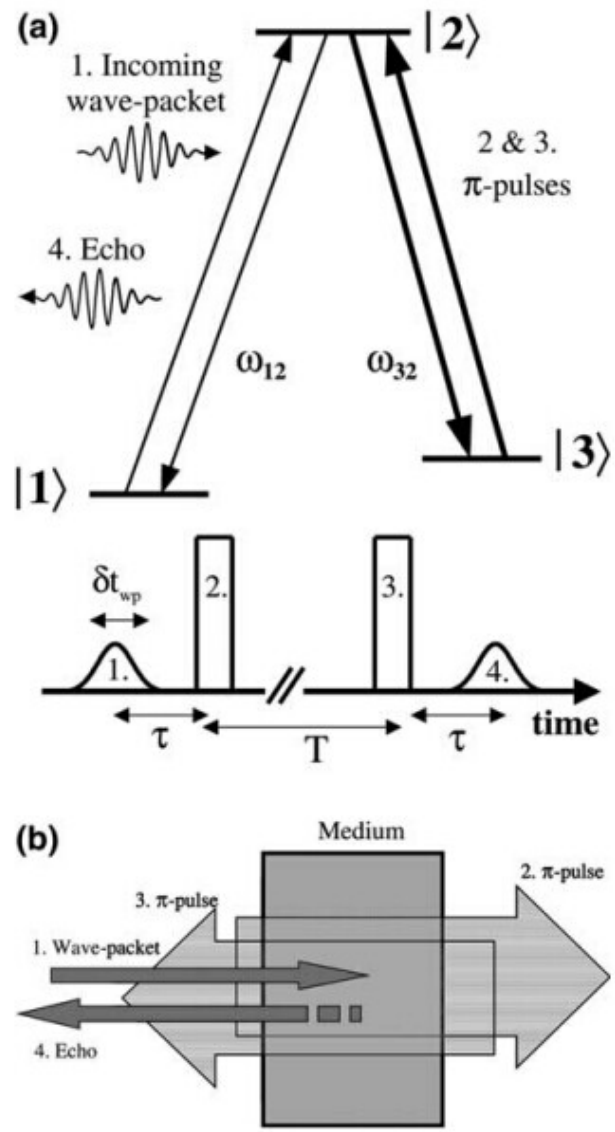


Figure 6. the Lambda type energy levels structure used in CRIB.
***Optics Communications* 247 (2005) 393–403**

Experiment Apparatus: crystal growth by Czocharalski method

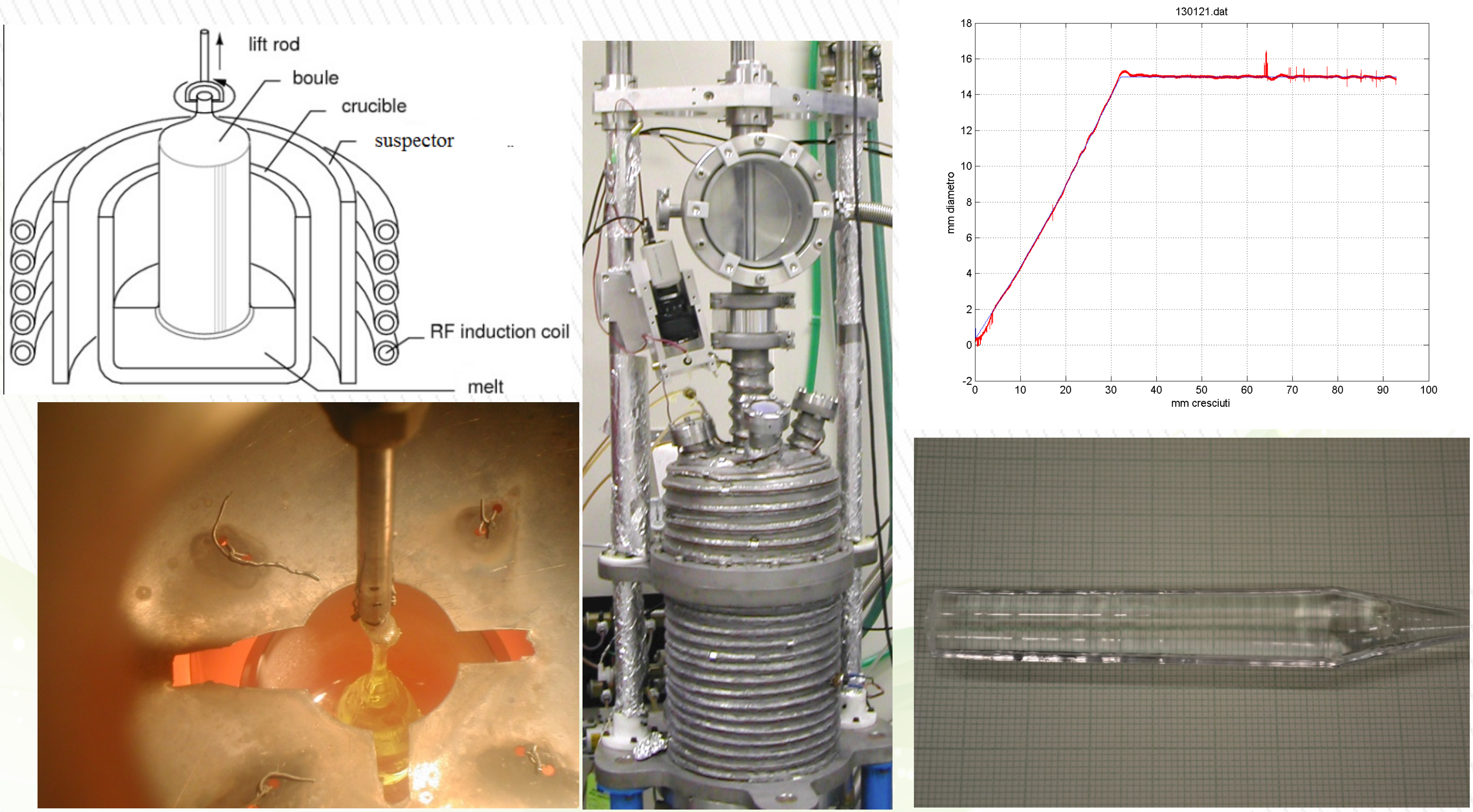
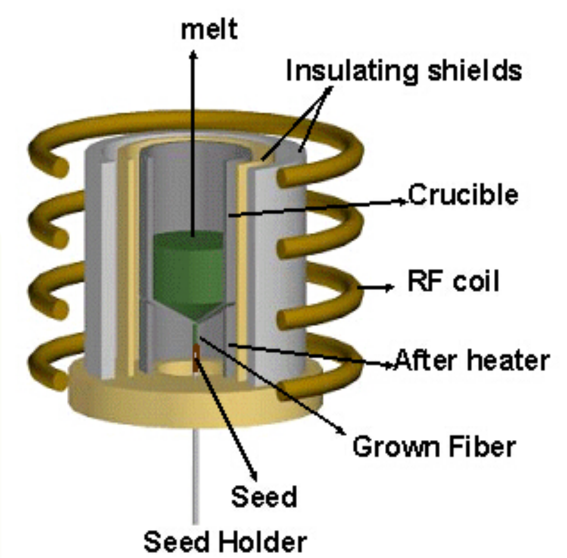
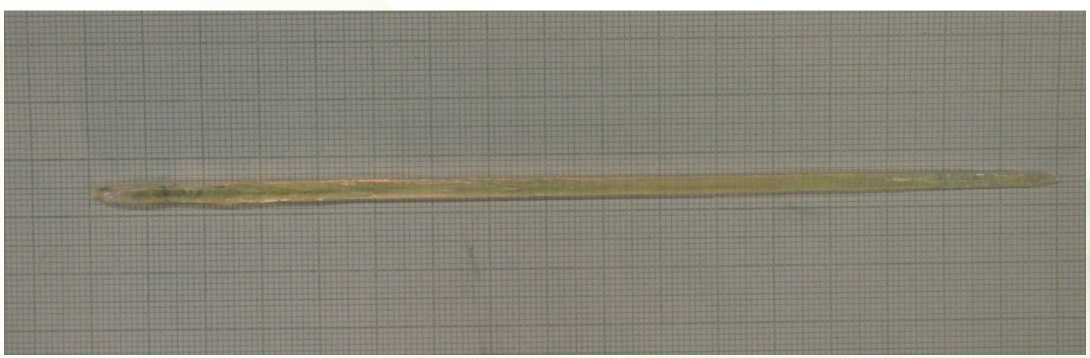


Figure 7. a typical crystal growth process using CZ method. The crystal shown in the picture is Yb^{3+} doped $LiYF_4$.

Experiment Apparatus: crystal growth by Micro Pulling Down Method





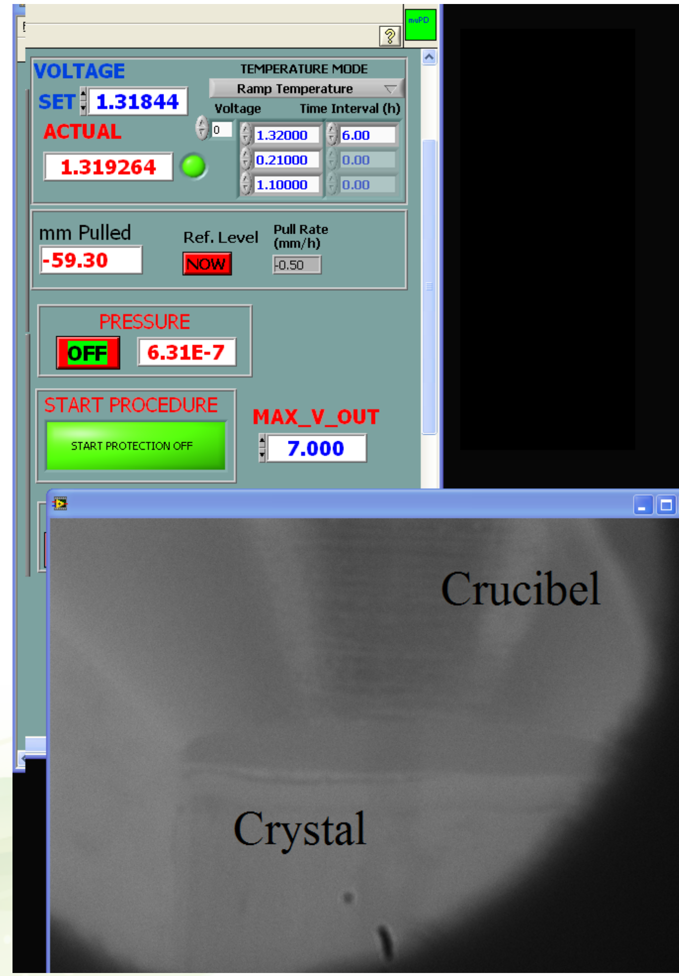


Figure 8. a typical crystal growth process using CZ method. The crystal shown in the picture is Pr^{3+} doped $SrAl_{12}O_{19}$.

Experiment Apparatus: spectra measurement

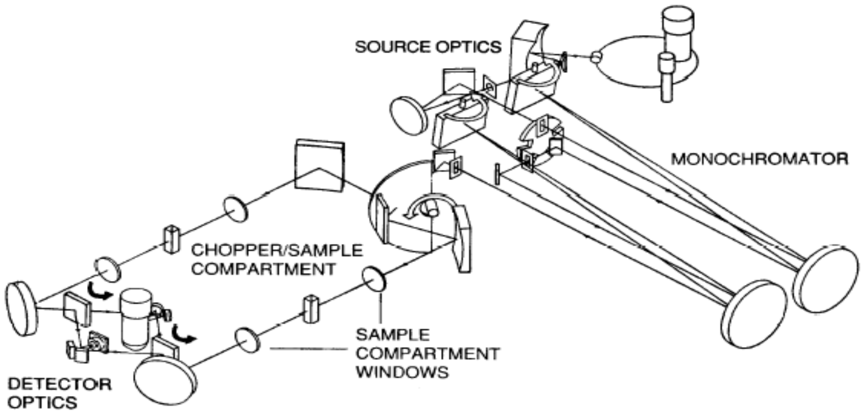


Figure 9. The set up of the VARIAN CARY 500 spectrophotometer.

The absorption spectra can be measured from 180nm to 3200nm. A cryostat can be applied and the temperature of the sample can reach about 10K.

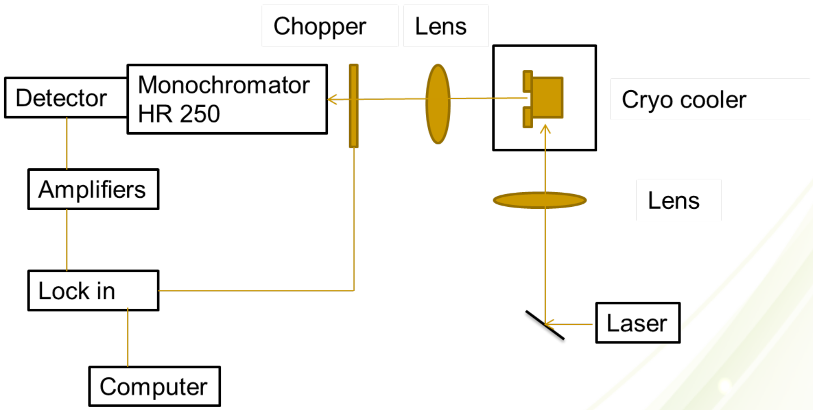
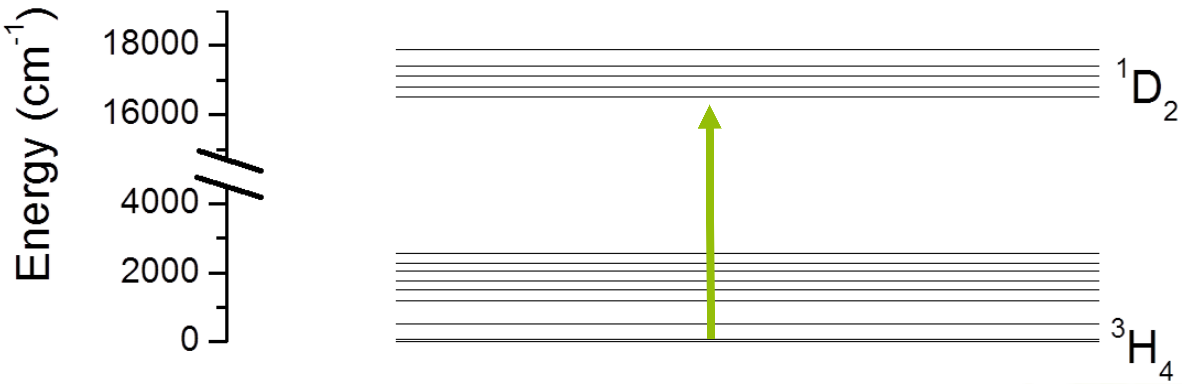


Figure 10, the set-up of fluorescence measurement. The lase here can be diode lasers or cw Ti:Al_2O_3 laser.

A cryostat can also be applied for fluorescence measurement. Various detectors are available covering a spectrum range from UV to IR.

Previous Work: Spectra measurement of Pr^{3+} doped $LiYF_4$, $LiLuF_4$ and BaY_2F_8

Zero phonon transition between $^3H_4$ and $^1D_2$ levels **of Pr^{3+} ion.**



crystal	*$LiLuF_4$:Pr^{3+} 1.25%*	*$LiYF_4$:Pr^{3+} 1%*	*BaY_2F_8:Pr^{3+} 1.25%*
growth (melt composition)	*congruent (LiF 50%, LuF_3 50%)*	*incongruent (LiF 53%, YF_3 47%)*	*congruent BaY_2F_8*
T_m (°C)	*830*	*850*	*960*
ionic radii (Å)	*R_{Lu}=1.12*	*R_Y=1.16*	*R_{Gd}=1.12*
ionic radius (Å) R_{Pr}=1.27			
k_{eff}	*0.1-0.2*	*0.22*	*0.3*
Pr^{3+}density (10^{19} cm^{-3})	*18.11*	*13.85*	*16.75*
Site symmetry Pr^{3+}	*S_4*	*S_4*	*S_2*

Previous Work: Spectra measurement of Pr^{3+} doped $LiYF_4$, $LiLuF_4$ and BaY_2F_8 -room temperature spectra

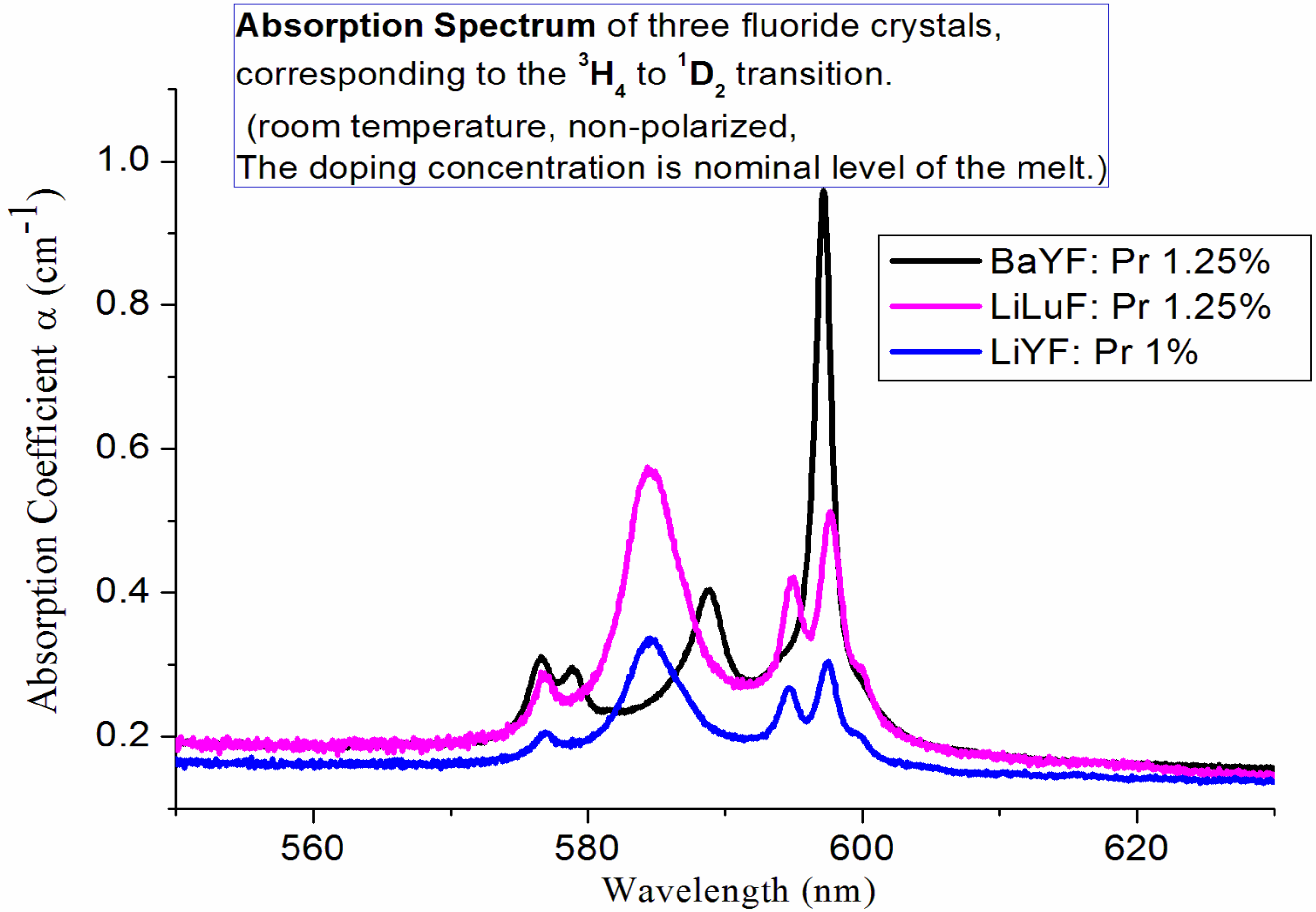
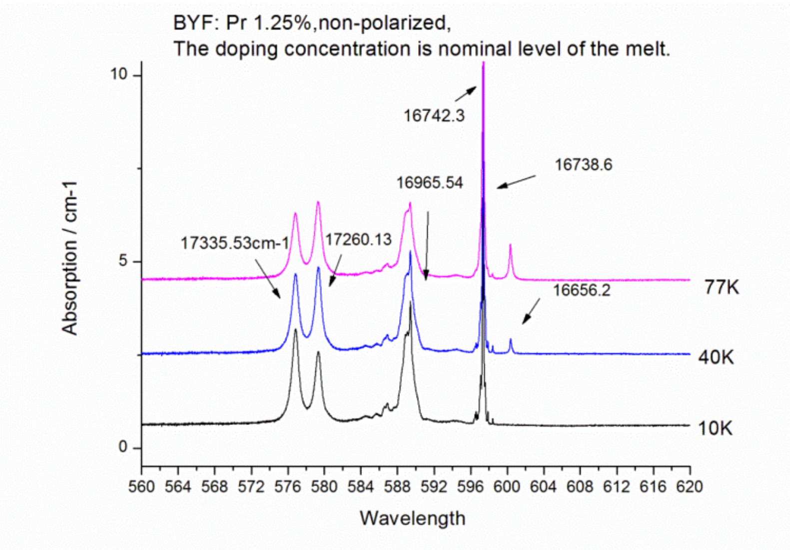
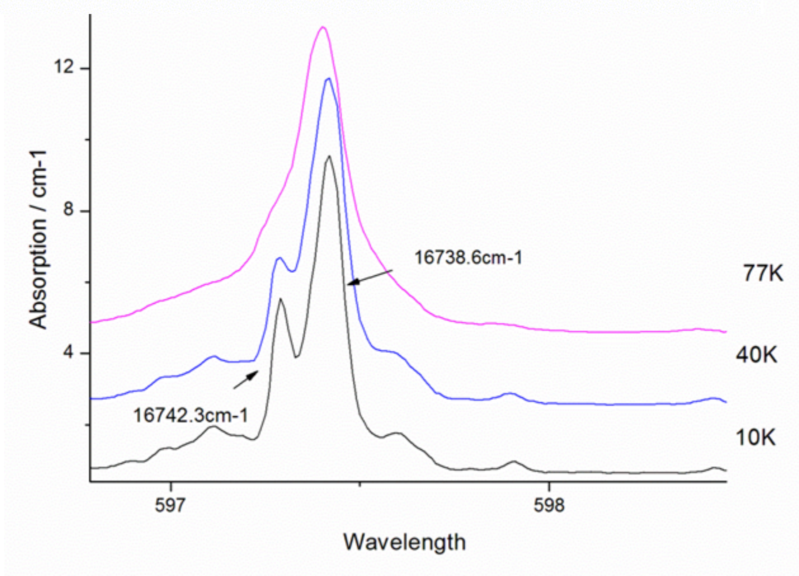


Figure 11, the absorption spectra for these three crystals at room temperature.

Previous Work: Spectra measurement of Pr^{3+} doped BaY_2F_8-low temperature spectra





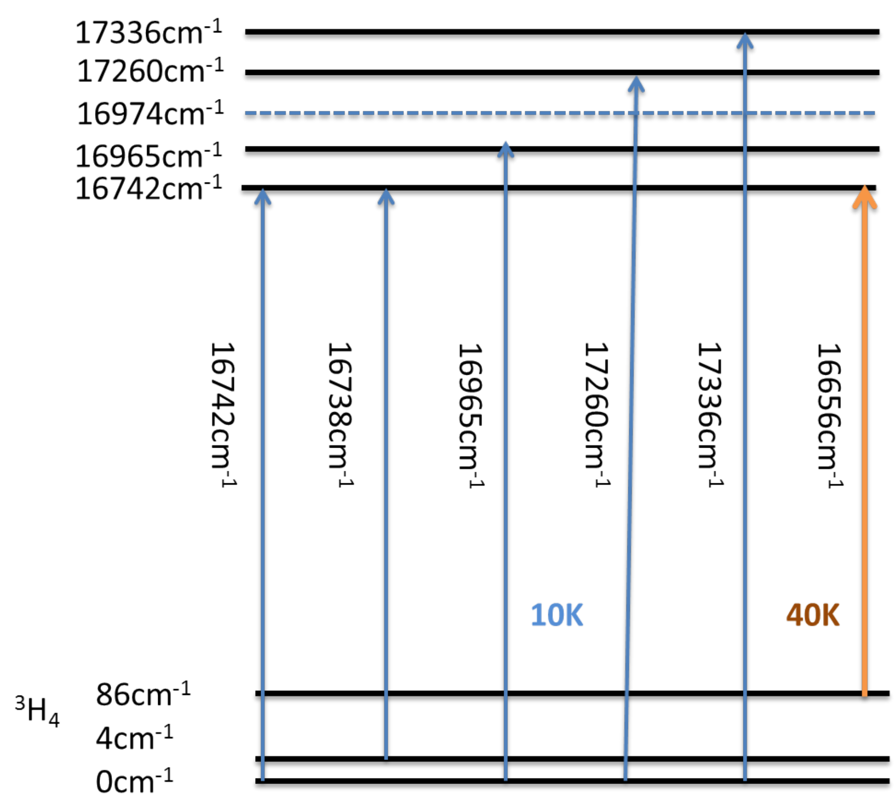
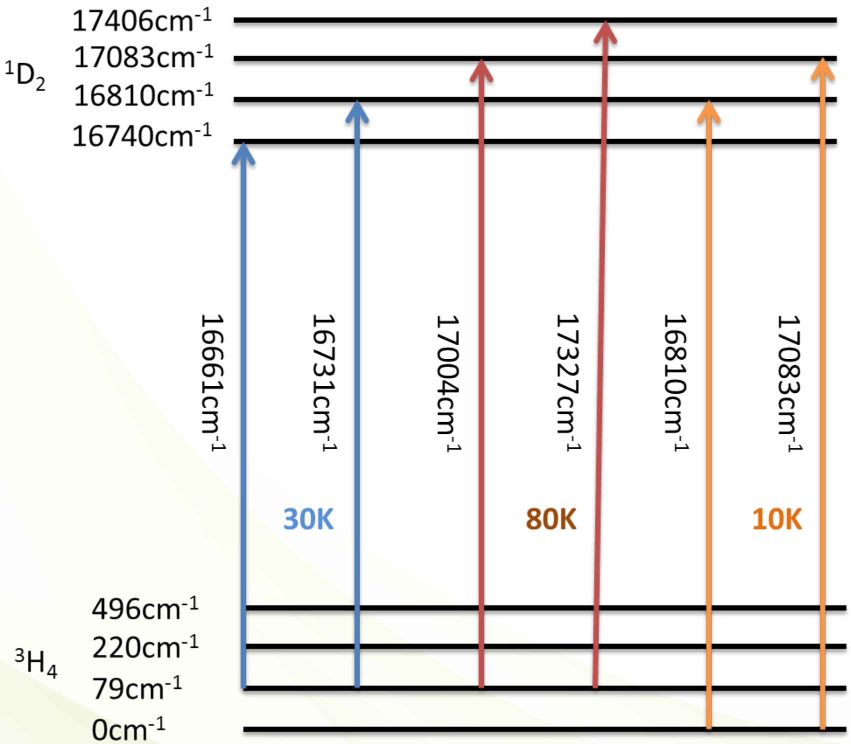
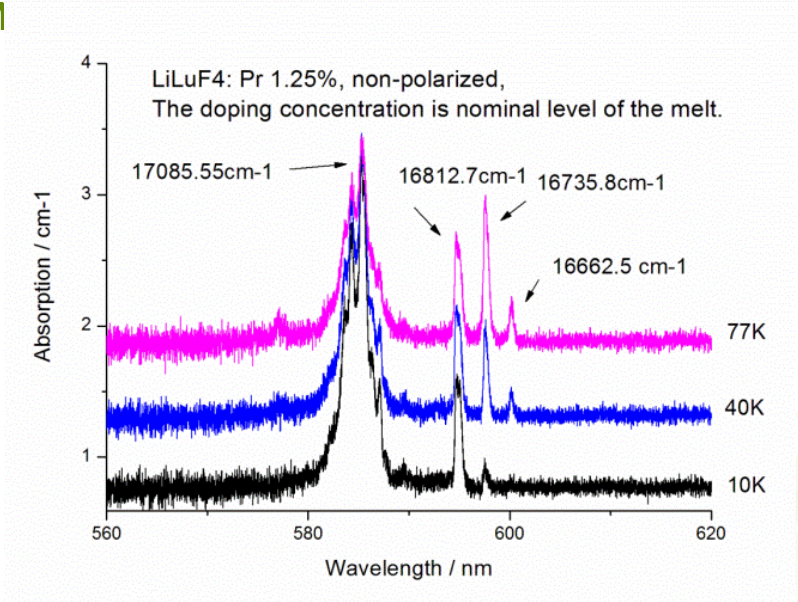


Figure 12, the absorption spectrum of Pr^{3+} doped BaY_2F_8 crystal at low temperature. Analysis of transitions was shown.

Previous Work: Spectra measurem





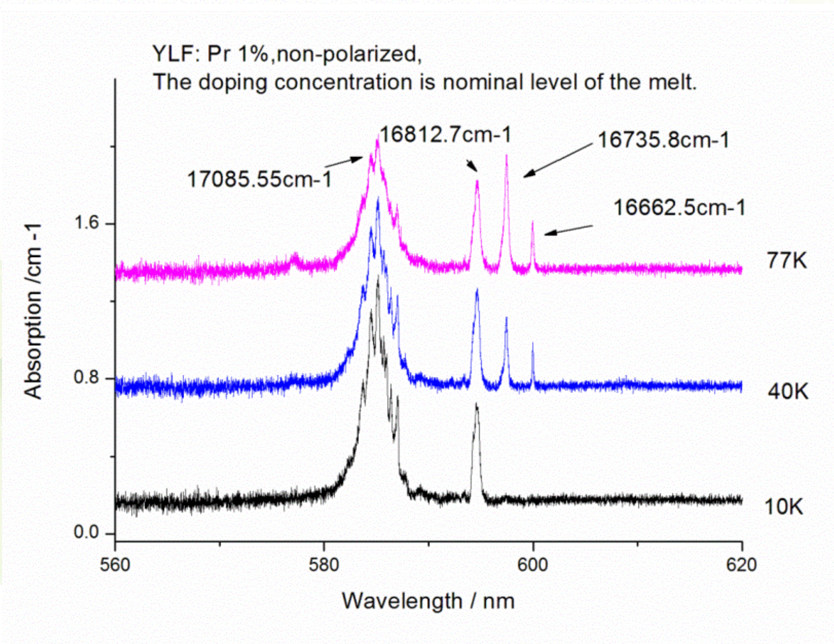


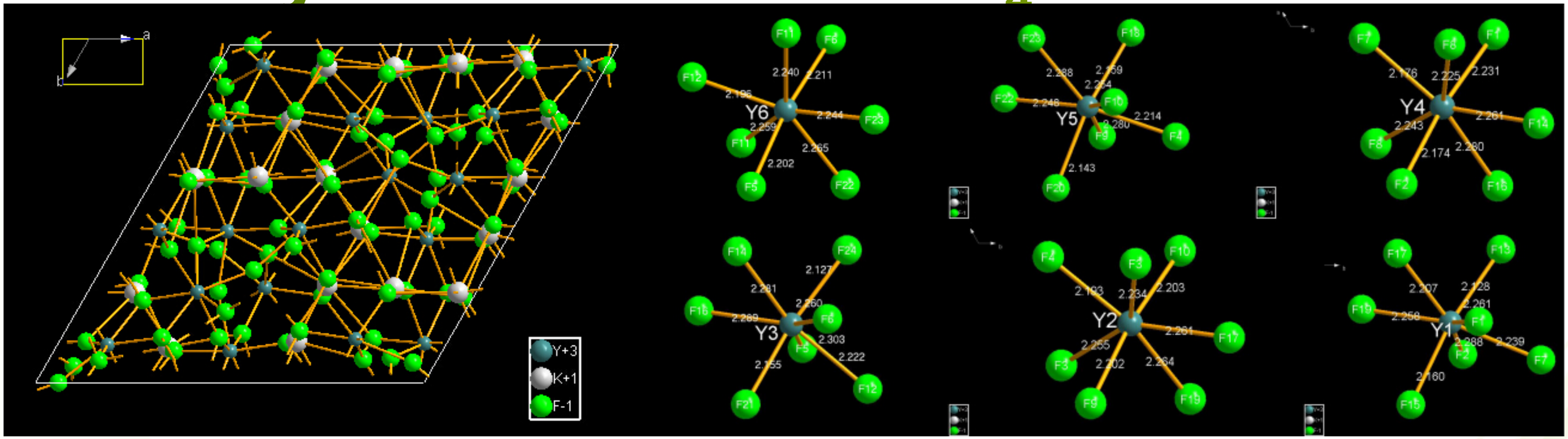
Figure 13, the absorption spectra of Pr^{3+} doped $LiYF_4$ and $LiLuF_4$. Analysis of transitions was shown.

Summary of Spectra measurement of Pr^{3+} doped $LiYF_4$, $LiLuF_4$ and BaY_2F_8

- $LiYF_4$ and $LiLuF_4$: The zero to zero transition is forbidden by select rule of S_4 symmetry.

- BaY_2F_8: The zero to zero transition is strong. However, there is a transition very close to it that comes from higher crystal field level in the $^3H_4$ state of Pr^{3+}.

4.2 Crystal Growth of KYF_4: Er^{3+}0.02%



Parameters of KYF4	
Crystal lattice Type	Tragonal
Space Group	P_{31}
a	14.06
c	10.10
Z	18
Difference of Y^{3+}	6

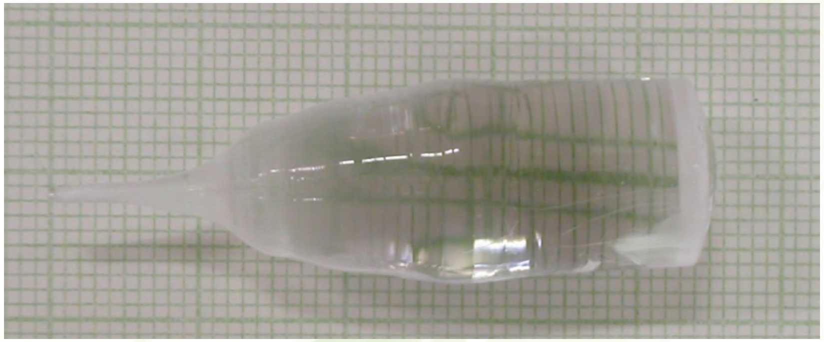
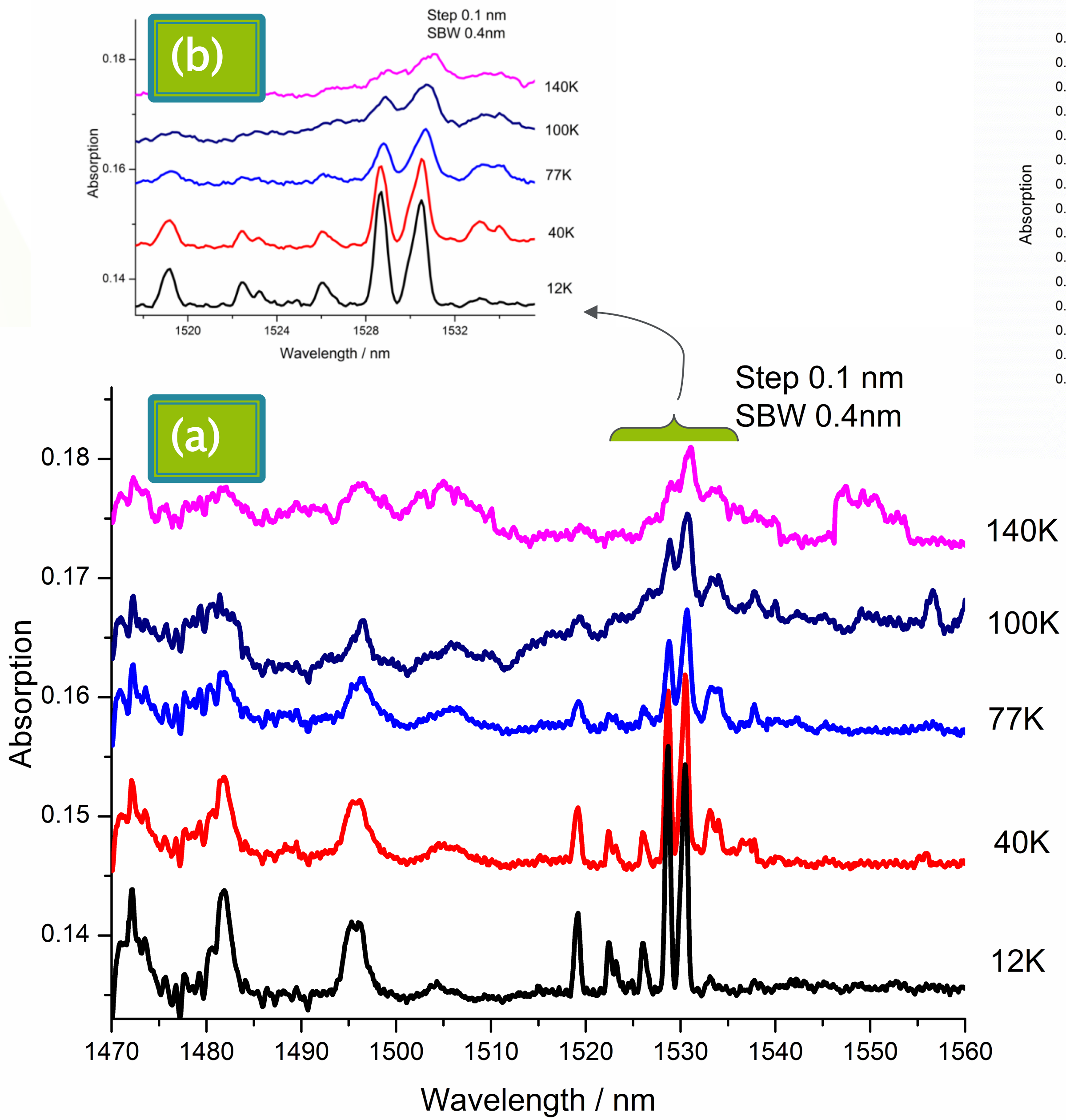
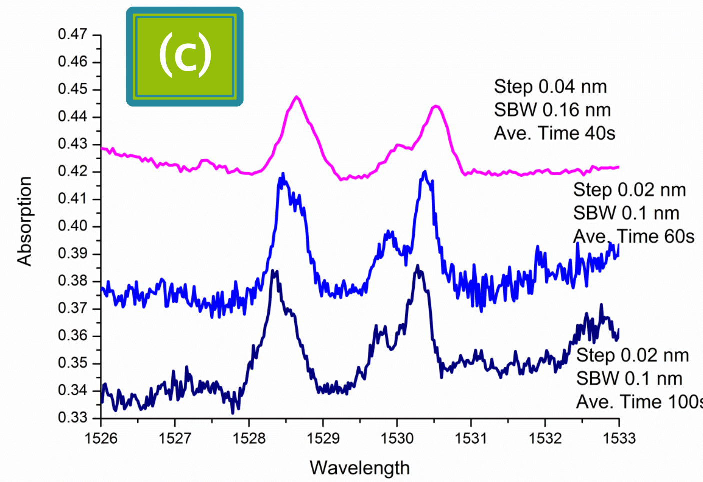


Figure 14, the picture of KYF_4: 0.02%Er^{3+}.

Growth Parameters:
Weight: 12g
Pulling Rate:0.5mm/h
Temperature: 823-819 ℃
Rotation Speed: 5 rpm.
Grown along –a axis.

Absorption Spectrum of Er: KYF_4 0.02%



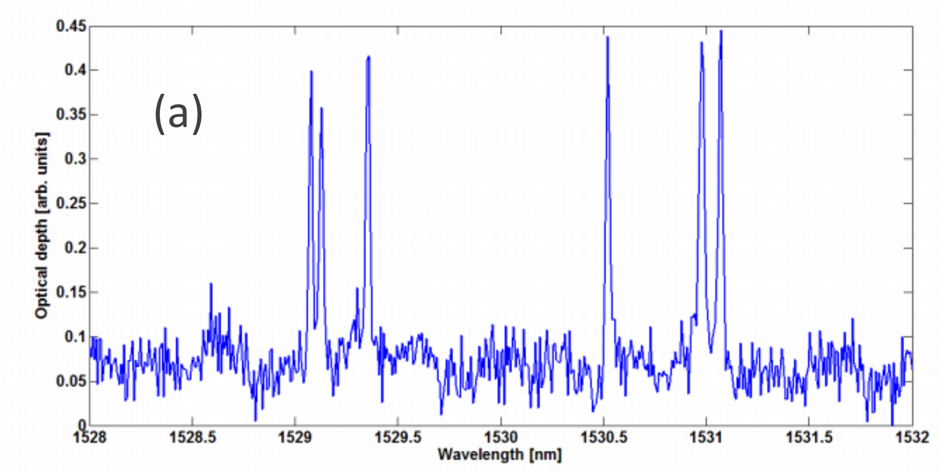


(a). Absorption Spectrum at the range 1470 nm to 1560 nm at different temperatures.

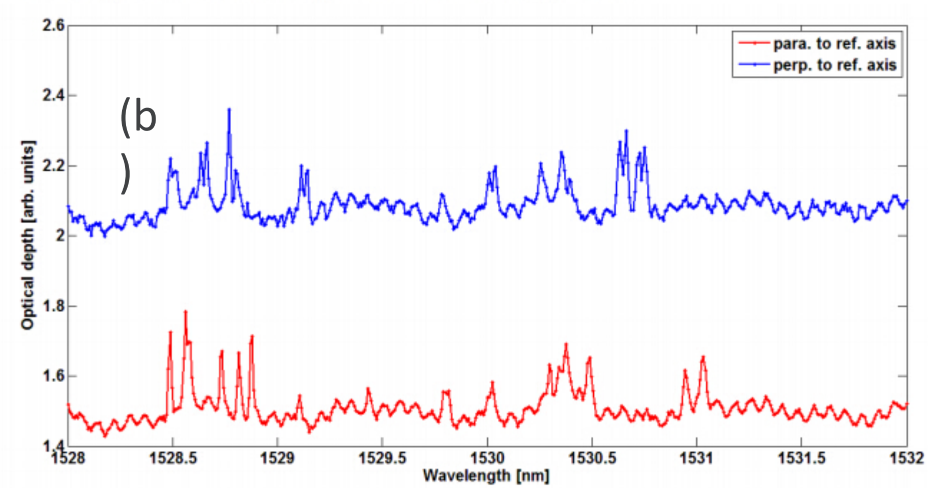
(b). Details within the range 1520 nm to 1532 nm.

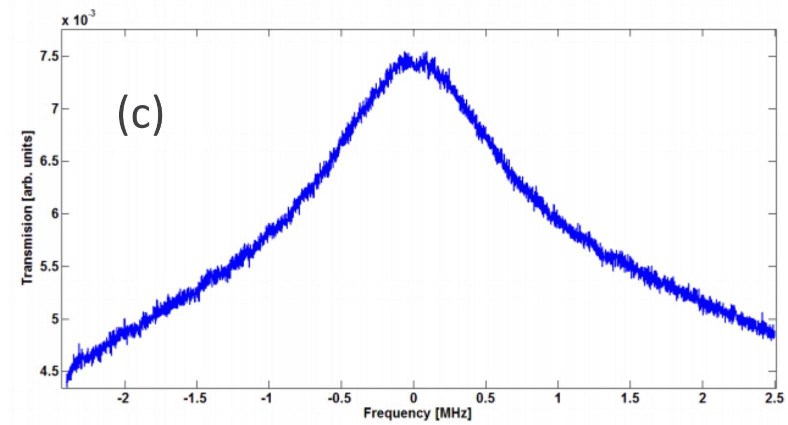
(c). Absorption Spectrum from 1528 nm to 1532 nm measured with different SBW settings.

Absorption Spectrum of Er: KYF_4 0.02% measured with high resolution.



Inhomogeneous broadening of 3 GHz for the largest lines





Optical pumping not very efficient, optical depth of the hole about 0,003

(a). The Absorption of the zero to zero transition at 2K.

(b). The absorption of the zero to zero transition at 2K with 3T 's magnetic field.

(c). The result of the spectrum hole burning at one of the zero to zero transition.

Summary of the experiments with Er: KYF_4 0.02%

- Crystals with high quality were grown.
- The wavelength of zero phonon transition was measured.
- High resolution spectra analysis implied that there are six separated zero phonon transitions.

Future Plan

- Investigate some more coherent measurement on the sample of Er^{3+} doped KYF_4 to learn the prospect of this crystal in quantum information application.
- Grow some other fluoride crystals (OR oxide) (KYF_4 or $LiCaAlF_6$ or YAG) using CZ method or Micro Pulling Down method and investigate the spectra of these crystals.
- Spectra measurement will be carried on the new crystals. Absorption and fluorescence spectra will help us to detect the 0 to 0 transitions. Photon echo measurement will tell the coherence time of particular state.

Thank You For Your Attention.