INFRARED SPECTROSCOPIC AND LASER CHARACTERIZATION OF Tm IN DISORDERED DOUBLE TUNGSTATES

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INFRARED SPECTROSCOPIC AND LASER CHARACTERIZATION OF Tm IN DISORDERED DOUBLE TUNGSTATES

Table of Contents

• Crystallophysics of tetragonal DT

Incorporation of lanthanides (Ln) in DOUBLE TUNGSTATES (DT)

$2\ Ca^{2+}XO_4$ (model)

$M^{+}T^{3+}(XO_4)_2$

ALTERNATIVE

PARTIAL SUBSTITUTION OF BIVALENT CATION

$4\ Ca^{2+}XO_4$

$M^{+}2CaT^{3+}(XO_4)_4 = M_{1/2}Ca_1T_{1/2}(XO_4)_2$

$8\ Ca^{2+}XO_4$

$3M^{+}2Ca^{2+}3T^{3+}(XO_4)_8 = M_{3/4}Ca_{1/2}T_{3/4}(XO_4)_2$

Poster session → $Li_{0.75}Ba_{0.5}Gd_{0.675}Yb_{0.075}(MoO_4)_2$

• Crystallophysics of tetragonal DT

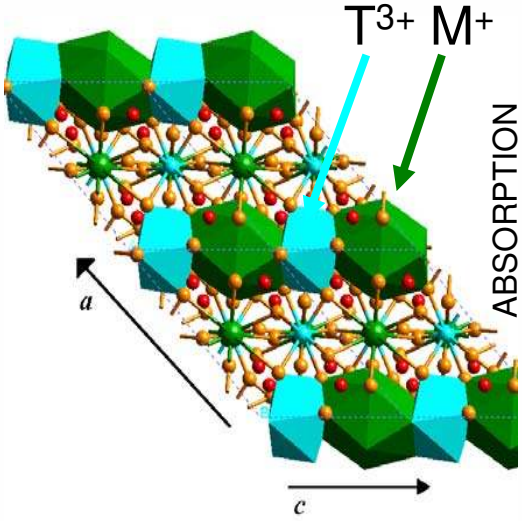
Femtosecond lasers require large optical bandwidths

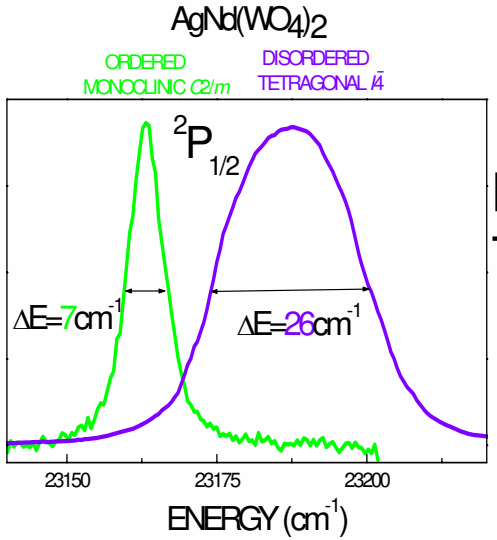
Ions with phonon-coupling (Yb,Tm) and hosts with lattice disorder

ORDERED DT CRYSTALS

like $KY(WO_4)_2$

$C2/c$





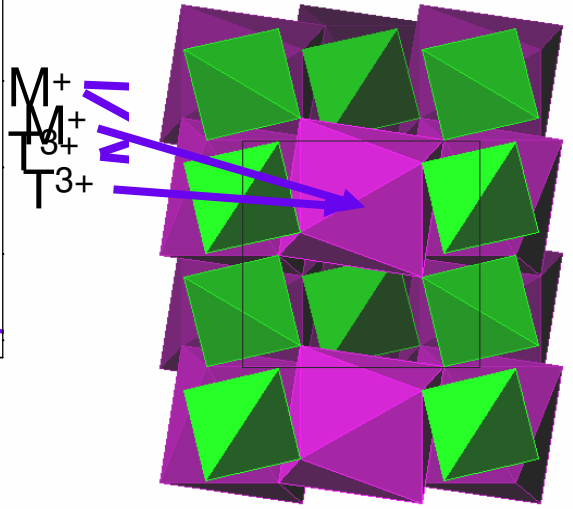
1 site-1 environment

DISORDERED DT CRYSTALS

like $NaY(WO_4)_2$

like $NaY(WO_4)_2$

$I4_1/a$



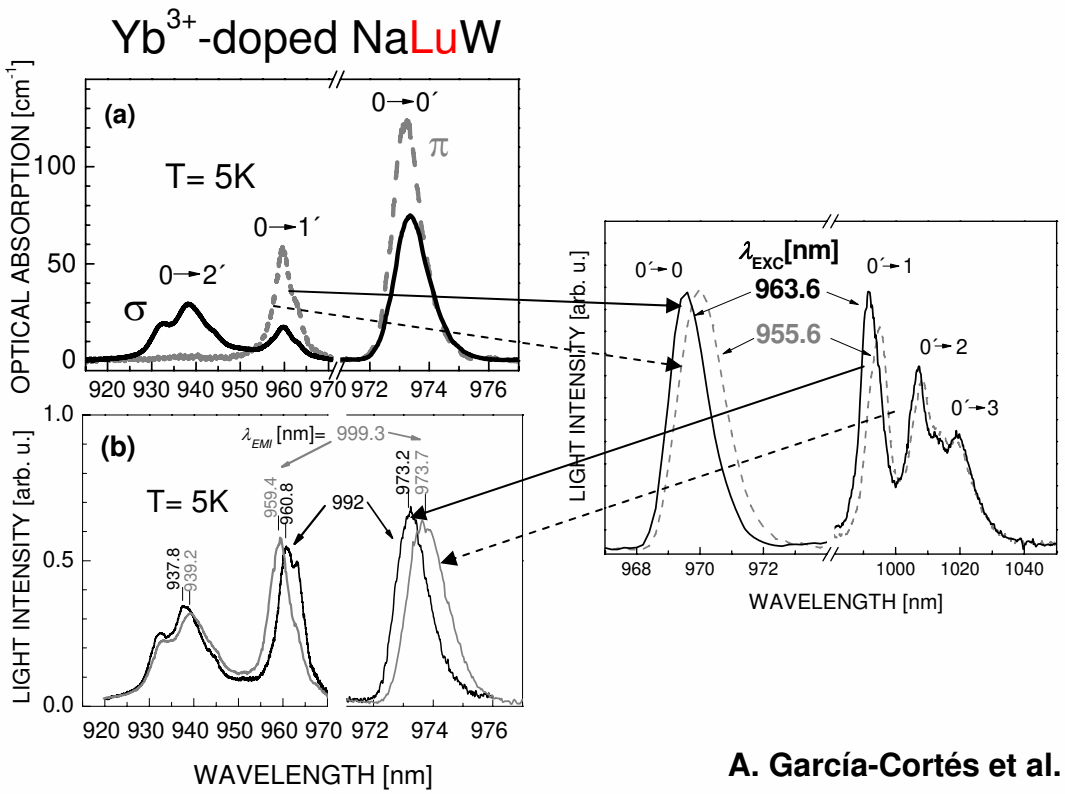
2 sites-each with multiple environments

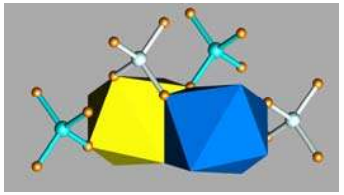
1 site-multiple environments

• Crystallophysics of tetragonal DT

Disordered (tetragonal $I\bar{4}$) $MT(XO_4)_2$ laser hosts

SITES: Spectroscopic evidence of the sites, Yb





	2*d*	2*b*
$^2F_{5/2}(2´)$	10647	10663
(1´)	10423	10408
(0´)	10270	10276
$^2F_{7/2}(3)$	474	475
(2)	378	372
(1)	239	238
(0)	0	0

A. García-Cortés et al. J. Appl. Phys 101, 63110-1-7, 2007

• Crystallophysics of tetragonal DT

Disordered (tetragonal $I\bar{4}$) $MT(XO_4)_2$ laser hosts
Czochralski and TSSG growth methods used

Czochralski method

HOST	UV-EDGE [nm]	
	σ	π
NaYW	297.8	298.5
NaLaW	316	312
NaGdW	299	297

Large sizes
Fast crystallization
Na & W losses

TSSG method

HOST	UV-EDGE [nm]	
	σ	π
NaLuW	288	289

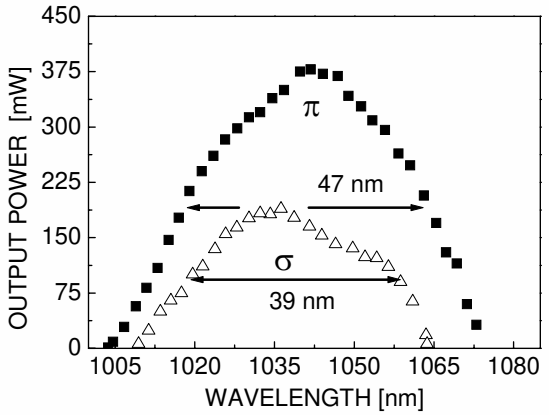
FLUX
$Na_2W_2O_7$
$NaWO_4$

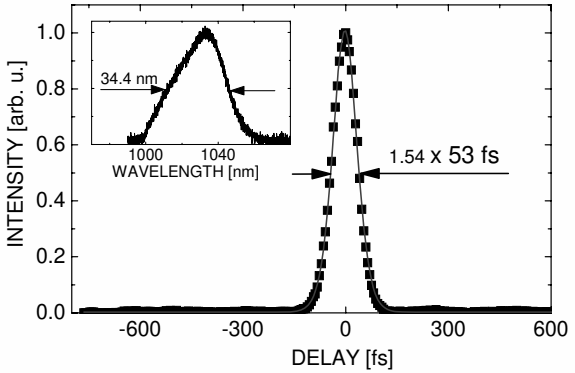
Smaller sizes
Slow crystallization

• Recent laser results based on Yb

Disordered (tetragonal $I\bar{4}$) $MT(XO_4)_2$ laser hosts

	NaYW [1]	NaLuW [2]	NaGdW [3]	NaLaW [4]
Tuning range (nm) for π central λ=1040 nm	70	45	65	40
Slope efficiency (%)	69	61	77	30
Laser threshold (mW)	189	200	400	100
Output (mW), Ti-sa pump				
Single pass	463	465	900	
Double pass		647		
Output (mW),diode pump			>1400	>300
ML-SESAM pulses (fs)	53	90	120	
Average power	91	50	360	



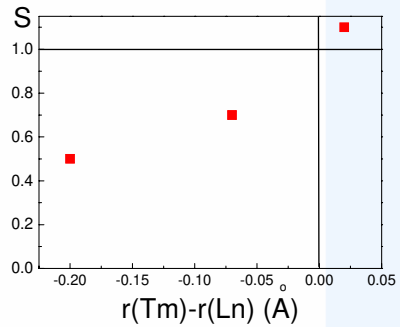


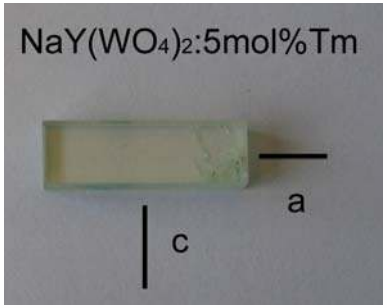
1 A. García-Cortés et al. IEEE J.l Quant. Electr.
2 A. García-Cortés et al. J. Appl. Phys 101, 63110-1-7, 2007
3 C. Cascales et al. Phys. Rev B 74, 174114-1-15, 2006
4 J. Liu phys status solidi a 202, R29, 2005

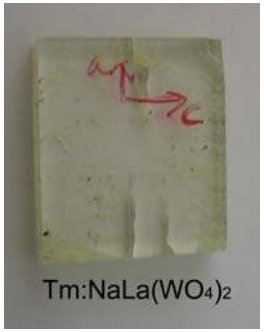
• Tm-doped DT

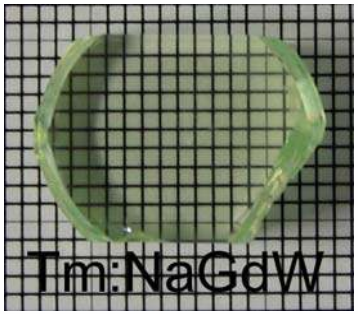
Growth of **Tm-doped** disordered (tetragonal $I\bar{4}$) DT laser crystals

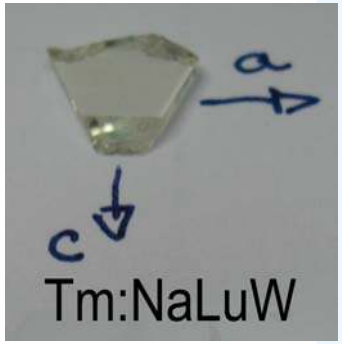
		$NaLnTm(WO_4)_2$	S	[Tm] (10^{20} cm^{-3})
Cz	5at%Tm:NaYW		--	----
Cz	5at%Tm:NaLaW	$Na_{0.918}La_{0.92}Tm_{0.025}$	0.5	1.505
Cz	5at%Tm:NaGdW	$Na_{1-\delta}Gd_{1.07}Tm_{0.037}$	0.7	2.34
TSSG	5at%Tm:NaLuW			3.92
TSSG	10at%Tm:NaLuW	$Na_{0.973}Lu_{0.947}Tm_{0.117}$	1.1	7.847











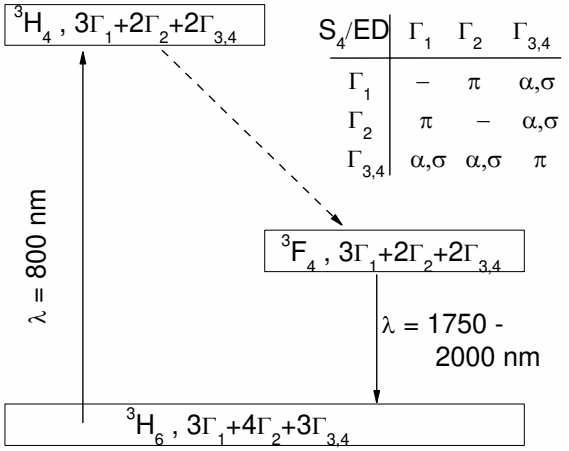
• Tm-doped DT. 5 K Spectroscopy

Tm^{3+} energy levels related to 2 μm emission

$$\sigma_{ABS} = \alpha/[Tm] \quad \text{300 K optical absorption}$$

$$\sigma_{EMI} = \sigma_{ABS} \frac{Z_l}{Z_u} e^{(E_{zl} - h\nu)/k_B T}$$

$$Z = \sum_k \left(-E_k / k_B T\right)$$



First objective is a complete Tm^{3+} energy level characterization

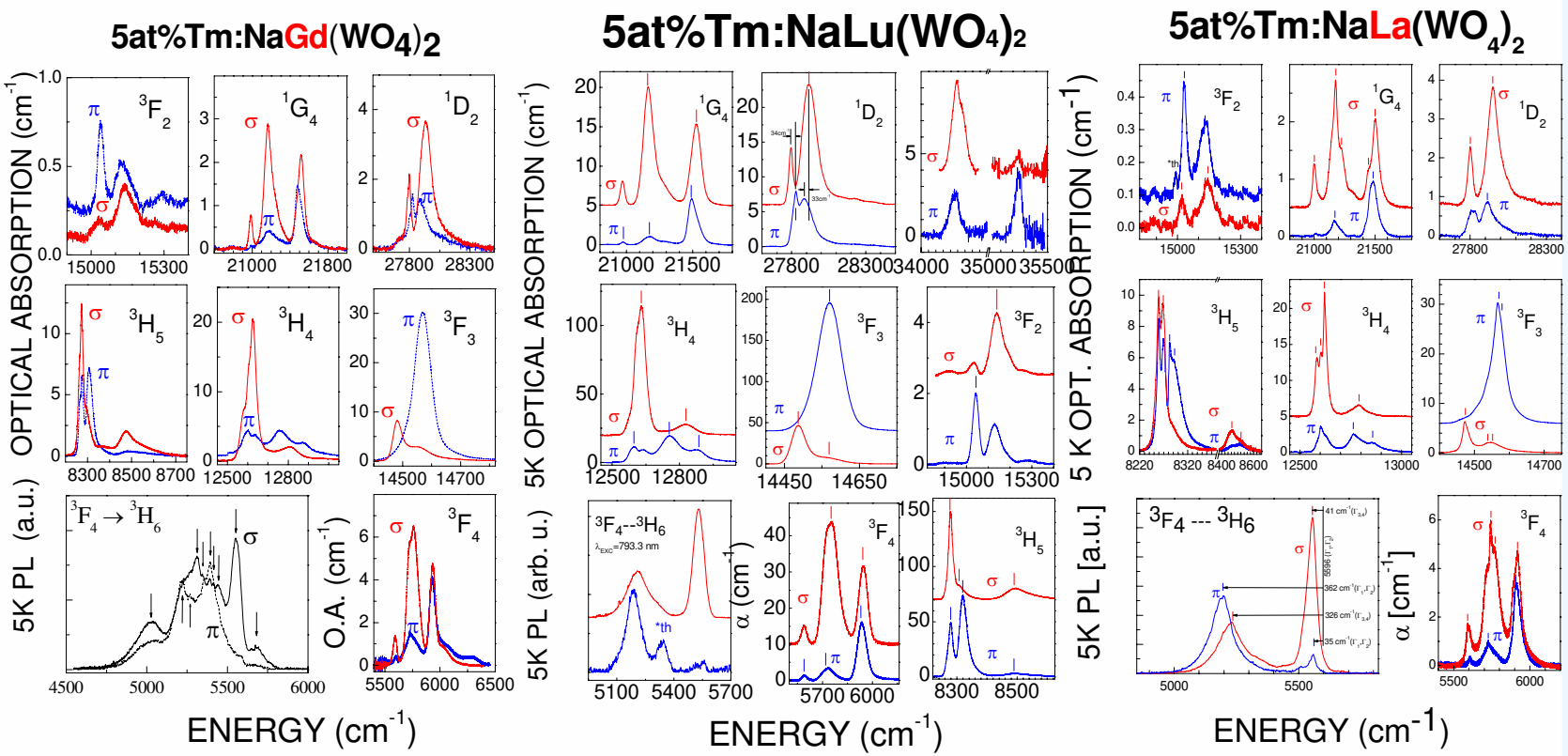
5 K optical absorption and photoluminescence

$$\sigma_{GAIN}(\lambda) = \beta\sigma_{EMI}(\lambda) - (1-\beta)\sigma_{ABS}(\lambda)$$

• Tm-doped DT. 5 K Spectroscopy

Extensive research of Tm spectroscopic properties in disordered (tetragonal $I\bar{4}$) DT

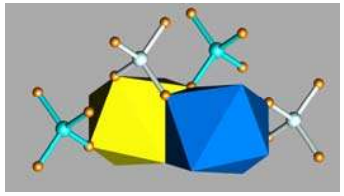
ENERGY LEVELS OBTAINED FROM 5K MEASUREMENTS



• Tm-doped DT. 5 K Spectroscopy

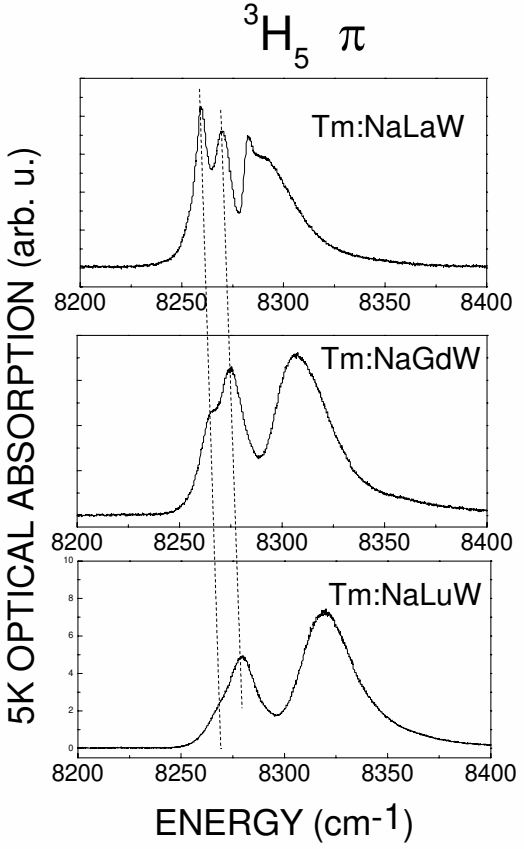
Extensive research of Tm spectroscopic properties in disordered (tetragonal $I\bar{4}$) DT

Tm SITE CONTRIBUTIONS



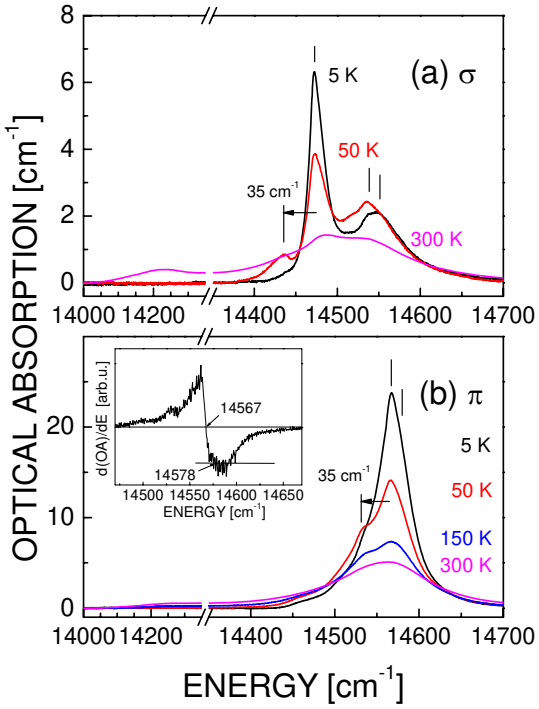
2*b* AND 2*d* OCUPANCY FACTORS

		NaYW	NaLaW	NaGdW	NaLuW
Na(I)/T(I)	*2d*	0.67(1)/0.33(1)	0.35(3)/0.65(3)	0.55(2)/ 0.45(2)	0.57(1)/0.43(1)
Na(II)/T(II)	*2b*	0.26(1)/0.74(1)	0.63(3)/0.37(3)	0.41(2)/ 0.59(2)	0.42(1)/0.58(1)



• Tm-doped DT. 5 K Spectroscopy

Tm^{3+} energy levels. Energy level partition functions



$^3H_6(0)$ belongs to Γ_2 (based on Tm-doped $CaWO_4$)
Absorption transitions $\Gamma_2 \rightarrow \Gamma_2$ not observed
5K OA alone can not provide full energy level list

$^{2S+1}L_J$	Energy [cm^{-1}]	Γ_1 , $\Gamma_{3,4}$
$^3H_6$	π 35 , 362 – σ 0, 326	
$^3F_4(^1G_4)$	σ 5596, 5742, 5769, 5892, 5918 – π 5726, 5912	3, 2
$^3H_5$	σ 8259, 8268, 8463, 8519 – π 8283, 8292	3, 3
$^3H_4(^3F_4)$	σ 12584, 12603, 12621, 12790 – π 12763, 12863	3, 2
$^3F_3$	σ 14472, 14538, 14551 – π 14567, 14578	1, 2
$^3F_2(^1D_2)$	σ 15019, 15145 – π 15031	1, 1
$^1G_4(^3H_4)$	σ 21000, 21177, 21223, 21448, 21502 – π 21172, 21486	3, 2
$^1D_2$	σ 27801, 27951 – π 27914	1, 1

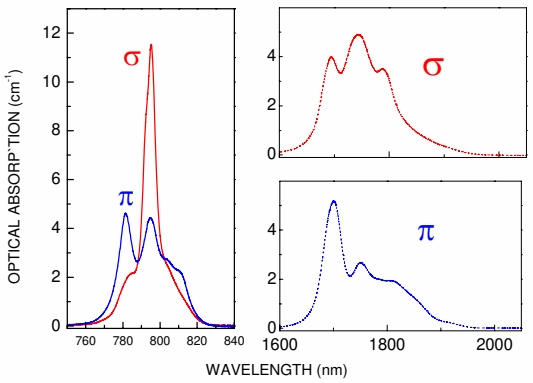
E_{zl}= 5596 cm^{-1} Z_l/Z_u= 1.26

• Tm-doped DT. 300 K Spectroscopy

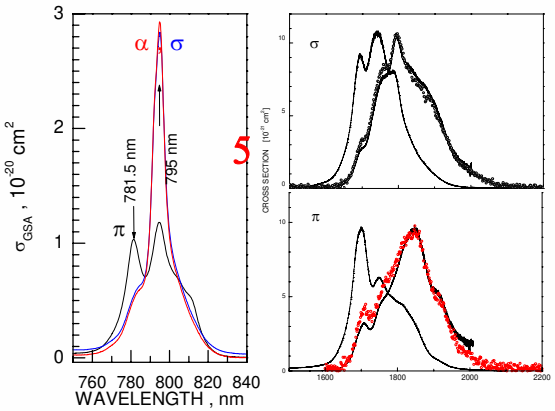
Absorption and emission (reciprocity) cross sections.

$$\sigma_{EMI} = \sigma_{ABS} \frac{Z_l}{Z_u} e^{(E_{zl} - h\nu)/k_B T}$$

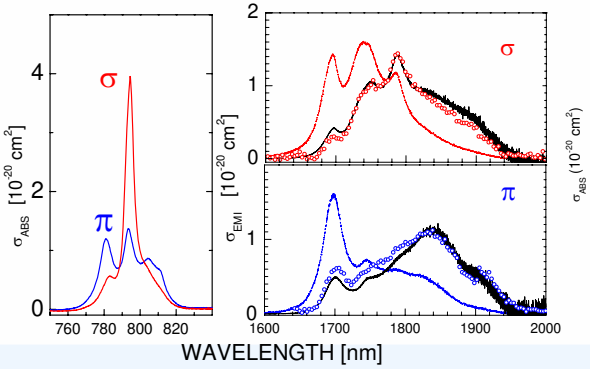
Tm:NaYW



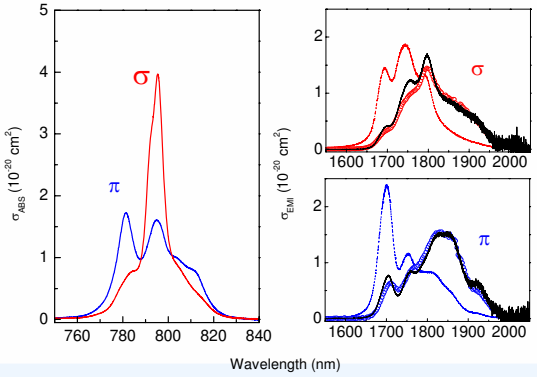
5at%Tm:NaLaW



Tm:NaGdW



Tm:NaLuW



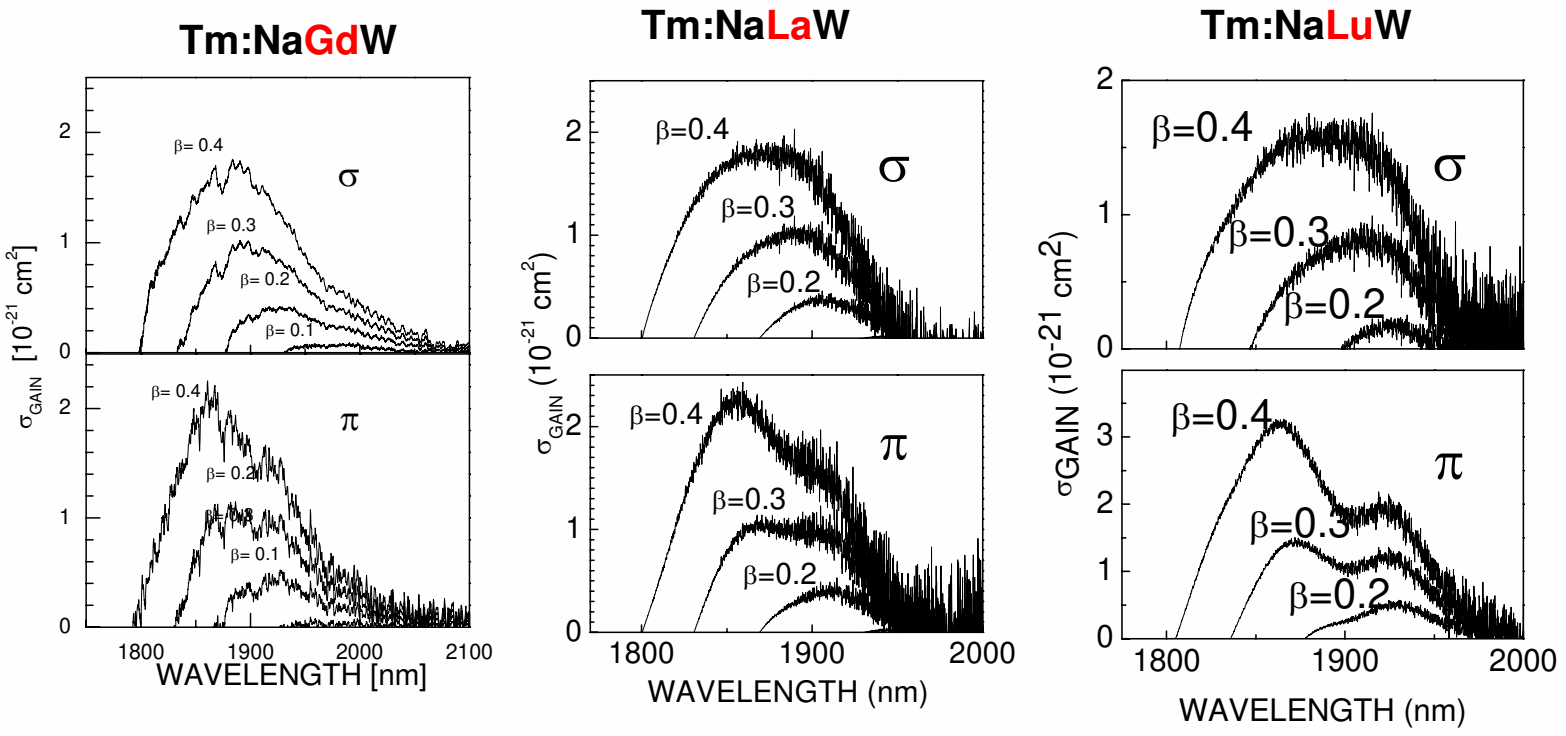
High pumping anisotropy

	NaLaW	NaGdW	NaLuW
$\sigma^{\pi}_{ABS}(10^{-20}cm^2)$	1.37	1.18	1.6
λ(nm)	794.5	795	795.5
$\sigma^{\sigma}_{ABS}(10^{-20}cm^2)$	3.94	2.9	3.96
λ(nm)	794.5	795	795.5
$\sigma^{\pi}_{EMI}(10^{-20}cm^2)$	1.13	0.95	1.47
λ(nm)	1840	1847	1848
$\sigma^{\sigma}_{EMI}(10^{-20}cm^2)$	1.43	1.06	1.66
λ(nm)	1787	1796	1796

• Tm-doped DT. 300 K Spectroscopy

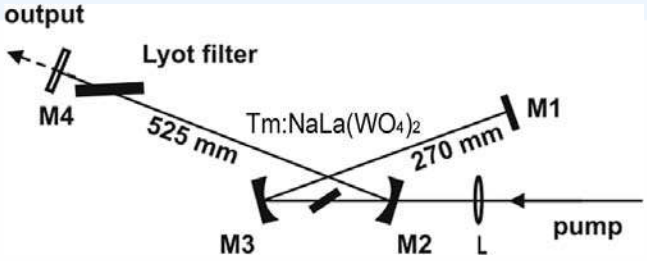
Gain cross sections

$$\sigma_{GAIN}(\lambda)=\beta\sigma_{EMI}(\lambda)-(1-\beta)\sigma_{ABS}(\lambda)$$

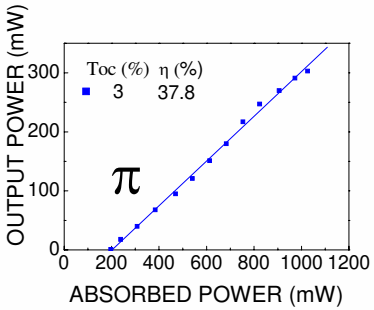


• Tm-doped DT. 300 K laser results

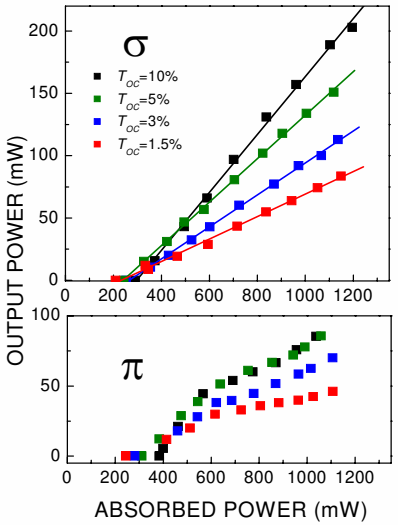
Free running cw laser output operation.
Ti-sapphire pump.



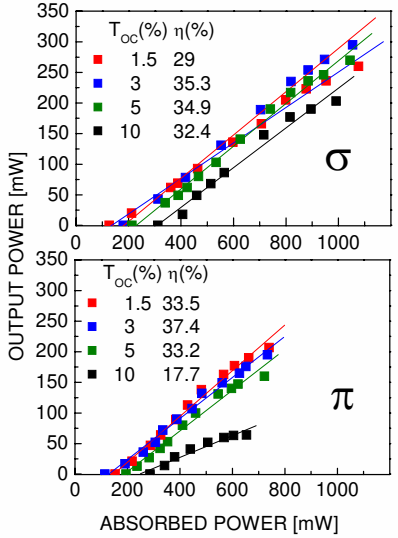
5at%Tm:NaYW



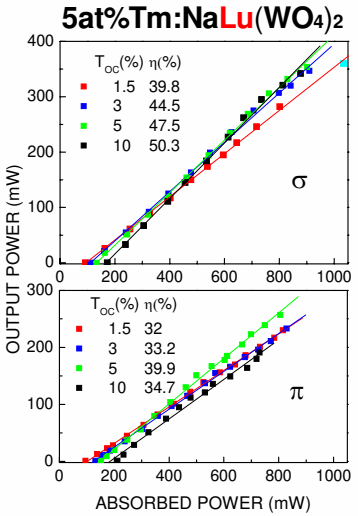
5at%Tm:NaLaW



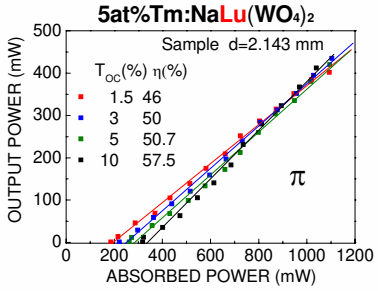
5at%Tm:NaGdW



5at%Tm:NaLu(WO4)2

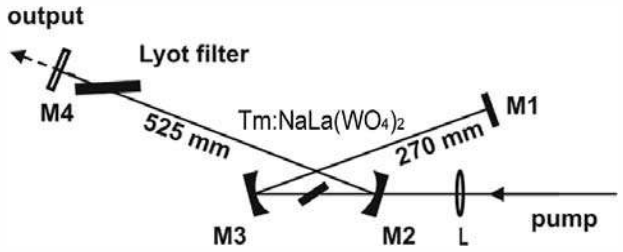


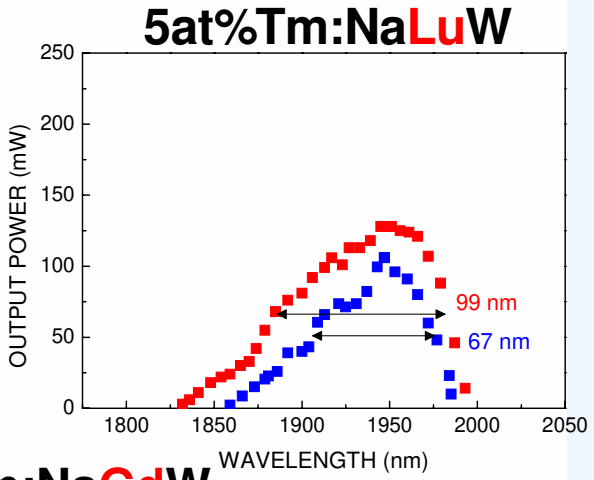
5at%Tm:NaLu(WO4)2

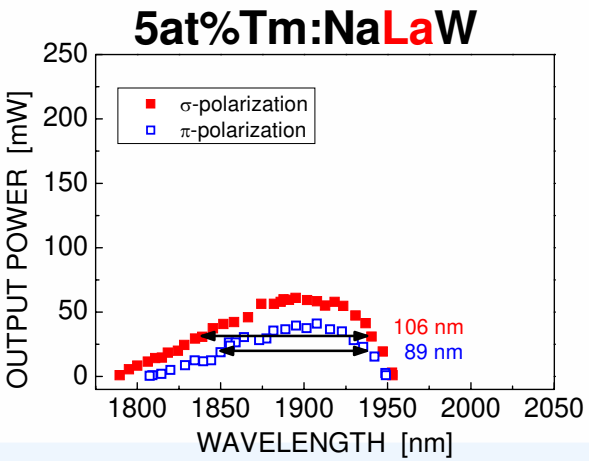


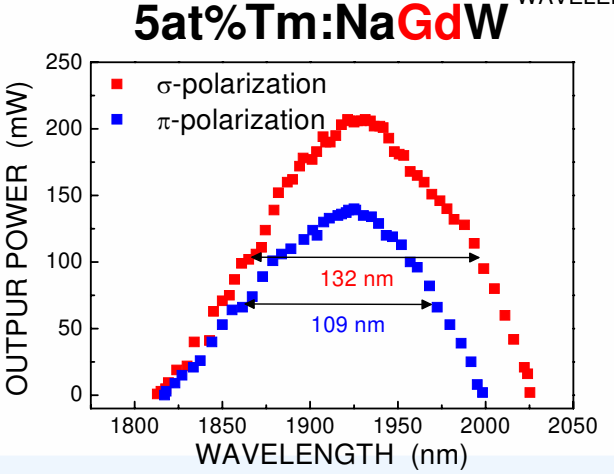
• Tm-doped DT. 300 K laser results

Laser Tunability.
Ti-sapphire pump.



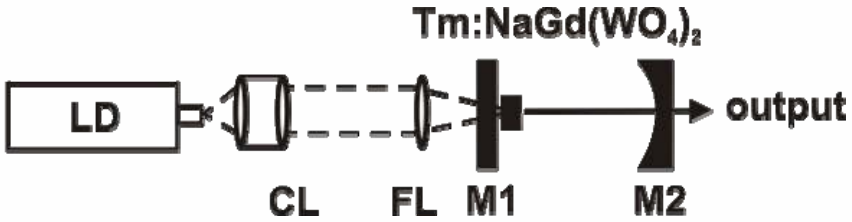




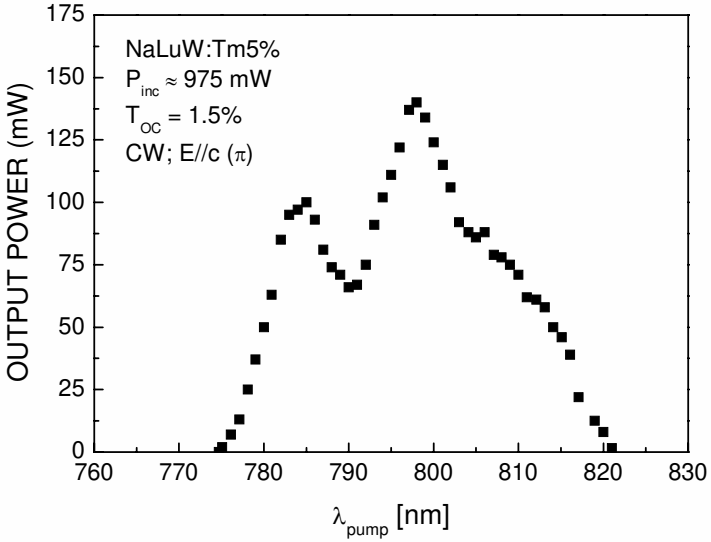


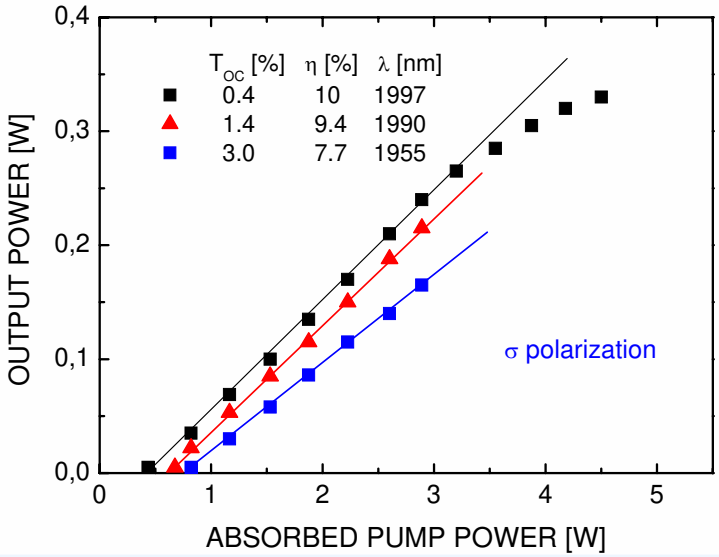
• Tm-doped DT. 300 K laser results

Laser operation under diode pump



BROAD EXCITATION!





INFRARED SPECTROSCOPIC AND LASER CHARACTERIZATION OF Tm IN DISORDERED DOUBLE TUNGSTATES

CONCLUSIONS

- Optical linewidths of Tm^{3+} in tetragonal $I\bar{4}$ DT are further broaded by the thullium occupancy of two different crystallographic sites and by the disordered environments associated to the random occupancy of Na and host lanthanide in these same sites.

- Tm-doped NaLnW single crystals without special cooling nor sample coatings showed in continuos wave operation output laser efficiency above η=50%, output laser intensity above 400 mW and smooth tuning range of up to 200 nm around 1930 nm.

- The spectroscopic measurements predict little laser performance differences in these hosts, but in practice the present NaLaW crystal quality limits its laser performance.

- The broad nature of the $^3H_4$ optical absorption allows non critically wavelength pump with diode laser. Laser operation under DL was shown in Tm-doped NaGdW.

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