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TITLE: **BLUE UPCONVERSION THULIUM LASER**

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Blue upconversion thulium laser

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Upconversion has been an active area of research for at least two decades, mainly because of its wide ranging applications from infrared quantum counters,[1,2] visible-emitting phosphors,[3,4] to upconversion lasers.[5-9] The upconversion lasers have recently become attractive with the advent of semiconductor laser diodes as the pump source.[10] In an upconversion laser, the laser active ion is excited by internal upconversion of near-ir or red light via multiphoton excitation or cooperative processes and emits anti-Stokes visible light. Since the laser diode output wavelength can be composition tuned to match the upconversion laser ion absorption lines, a substantial fraction of the ions can be driven into higher energy levels, thus enhancing the upconversion process. These upconversion solid-state lasers offer a potentially simple and compact source of visible coherent light with semiconductor laser diode excitation. We recently reported a novel upconversion thulium laser that emits blue light at 77 K.[11] In this paper additional data on this 77 K upconversion laser as well as preliminary results on the room temperature upconversion laser are presented. In these demonstrations, dye lasers were used instead of diode lasers because they were more readily available than high power semiconductor laser diodes and their wavelengths could be adjusted easily.

The thulium upconversion laser was pumped by a two-step process involving one photon in the near-ir at 781 nm and one in the red at 649 nm. The upconversion laser material was a 1-cm long yttrium lithium fluoride (YLF) crystal doped with nominally 1 at.% of trivalent thulium ion. The laser resonator (Fig. 1) consisted of a total reflector and a 10% output coupler at 450 nm. Both mirrors have a 10-cm radius of curvature. The two pump beams were collinearly directed through the 450 nm total reflector and focused to a spot ~250 μm in diameter inside the crystal. For the first demonstration, the pumping sources were two Nd:YAG-pumped dye lasers. Because the pulse widths of these dye lasers were less than 10 ns, optical damage to the Tm^{3+}:YLF crystal limited the excitation fluences to less than 10 J/cm^2. As a result, the crystal was kept at liquid nitrogen temperature to increase Tm^{3+} peak absorption and stimulated emission cross-sections so that lasing could be achieved at low pump level.

The upconversion excitation for the blue emission laser is shown in Fig. 2. Under π-polarized excitation at 780.78 nm, the Tm^{3+} ion is excited from the $^3H_6$ ground state to the $^3H_4$ intermediate level. There, it is excited to the $^1D_2$ upper laser level by a second red photon, also π-polarized, at 648.77 nm. The fluorescence lifetime of the $^1D_2$ upper laser

level is 59 μs for a 1.5% thulium concentration at 77 K. In the absence of lasing, the excited Tm ions undergo $^1D_2 \rightarrow {}^3F_4$ transitions radiatively with a branching ratio of 49.1% and emit several lines at around 445–455 nm (Fig. 3). These fluorescence lines correspond to the dipole allowed transitions in the S_4 point group assignment made by Jenssen *et al.*[12] We have remeasured the energy levels of Tm^{3+} in YLF and assigned these fluorescence lines as shown in Fig. 4.

With the resonator described above, we achieved laser action as a coherent blue laser beam at 450.20 nm at 77 K. This wavelength corresponds to two transitions, one π-polarized corresponding to a transition from the lowest Γ_2 level of the $^1D_2$ manifold to the second lowest Γ_1 level, and the other σ-polarized corresponding to a transition from the lowest Γ_2 level of the $^1D_2$ manifold to the lowest $\Gamma_{3,4}$ level of the $^3F_4$ manifold (Fig. 4).

The initial performance of the upconversion Tm^{3+}:YLF laser is shown in Fig. 5. The output power increases steadily with input red and near-ir powers until the point where optical damage in the crystal becomes possible. At this point where the pump pulse energy is 10 mJ at 781 nm and 3.5 mJ at 649 nm, we obtained a pulse output energy of 180 μJ, or a 1.3% overall efficiency. In our unoptimized laser resonator, the mode volume is about one-fourth of the pump volume, so the laser should be 4 times more efficient with proper mode matching. The laser threshold fluence for this unoptimized resonator, as measured at the center of the crystal, is 1 J/cm^2 for the 781 nm pump and 4 J/cm^2 for the 649 nm pump. It should be noted that the $^1D_2 \rightarrow {}^3F_4$ is a self-terminating transition, because the lower laser level lifetime of ~12 ms is longer than the upper laser level lifetime (59 μs). This precludes cw operation of this transition and places constraints on the laser pulse repetition frequency. From the buildup time of the laser cavity and the estimated cavity loss, we deduced a gain per pass of 0.4 per cm.[11] Independent kinetic calculations based on rate equations yield a value of the excited population of ~0.02 at. % or 1.37×10^{18} ions/cc. This translates into a peak stimulated emission cross section of 2.9×10^{-19} cm^2.

To evaluate the potential for room temperature laser action, we measured the fluorescence lifetime of the $^1D_2$ upper laser level as a function of temperature. The results are plotted in Fig. 6. After an initial drop in fluorescence lifetime at temperatures below 100 K, the $^1D_2$ fluorescence lifetime remains relatively constant up to 300 K. Assuming a simple thermally activated quenching process, one can relate the fluorescence lifetime to temperature by the following expression,

$$\frac{1}{\tau} = \frac{1}{\tau_o} + \mathrm{Re}^{-\frac{\Delta E}{kT}}$$

A curve fit of the fluorescence lifetime data to the above expression yields a fluorescence lifetime at 0 K, τ_0 of 77.4 μs and an activation energy ΔE of 31.9 cm^{-1}. Since the energy gap between the $^1D_2$ level and the next lower level, $^1G_4$, is more than 6,000 cm^{-1}, several times the effective phonon energy of YLF (400 cm^{-1}),[13] one expects the rate of

multiphonon relaxation to be negligible. The initial rapid drop in fluorescence lifetime can be attributed to a cross relaxation mechanism that involves two thulium ions, one in the $^1D_2$ level and the other in the $^3H_6$ ground manifold. This cross relaxation mechanism can become quite effective in quenching the $^1D_2$ level at concentrations above 3%. A careful study of these excited state dynamics is being prepared for publication elsewhere. Because the $^1D_2$ fluorescence lifetime is relatively constant between 75 and 300 K, the fluorescence quantum efficiency is not significantly degraded. Therefore, it should be possible to obtain room temperature laser action if we can match the pump pulse length to the fluorescence lifetime, which is 53 μs at 300 K. Previous experiments by Baer *et al.* have demonstrated room temperature laser action in Tm^{3+}:YLF with direct excitation using the 353-nm output of an electron-beam pumped XeF laser.[14]

To achieve room temperature upconversion laser action, we used two flashlamp-pumped dye lasers to excite the upconversion laser. The output pulse widths of these dye lasers are approximately 0.8 μs and the spectral bandwidth is about 0.6 nm. The Tm^{3+}:YLF crystal used in this room-temperature upconversion laser demonstration is 2 cm long and has a 1.5% Tm dopant level. The room-temperature upconversion laser resonator is the same as in the 77 K laser, except the output coupling is reduced to 5% (95% reflecting mirror). The dye laser beams were focused to a spot about 1 mm (FWHM) in the crystal. With this setup, we have obtained room temperature laser action in Tm^{3+}:YLF with ~30 mJ excitation from each dye laser. At pump fluences above ~2.6 J/cm^2, a bright coherent blue beam was observed. This blue laser beam was measured to be at 453 nm, corresponding to the same $^1D_2 \rightarrow {}^3F_4$ transition but involves different crystal field levels. As shown in Fig. 4, the room temperature laser transitions are from the $\Gamma_{3,4}$ level of the $^1D_2$ manifold to the highest Γ_1 level and the highest $\Gamma_{3,4}$ level of the $^3F_4$ manifold. The results of 77 K and room temperature upconversion laser action of Tm^{3+}:YLF are summarized in Table I.

	77 K operation	300 K operation
Lasing Wavelength	450 nm	453 nm
Dopant Level	1%	1.5%
Crystal Length	1 cm	2 cm
Pump Dye Lasers	Nd:YAG-pumped	Flashlamp-pumped
Pump Pulse Length	<10 ns	0.8 μs
Pump Bandwidth	1 cm^{-1}	~10 cm^{-1}
Output Coupling	10%	5%
Threshold Fluence	1 J/cm^2	~2.6 J/cm^2
Efficiency	1-2%	Not measured
Optical Damage Threshold	7-10 J/cm^2	Expected 60-90 J/cm^2

Table I

Summary of 77 K and 300 K Upconversion Tm^{3+}:YLF Laser

In conclusion, we have demonstrated blue-emission upconversion laser action in Tm^{3+}:YLF both at 77 K and room temperature. The output wavelengths are 450 and 453 nm for upconversion laser action at 77 K and 300 K, respectively. With excitation pulse length comparable to the fluorescence lifetime of thulium, the optical damage theshold of YLF is expected to be much higher than the lasing threshold fluence, thus reliable upconversion laser performance can be expected. To our best knowledge, this is the first room-temperature upconversion laser ever achieved with emission at higher energy than the pump wavelength. Future work is directed toward improving the pump geometry to obtain better efficiency and higher output energy.

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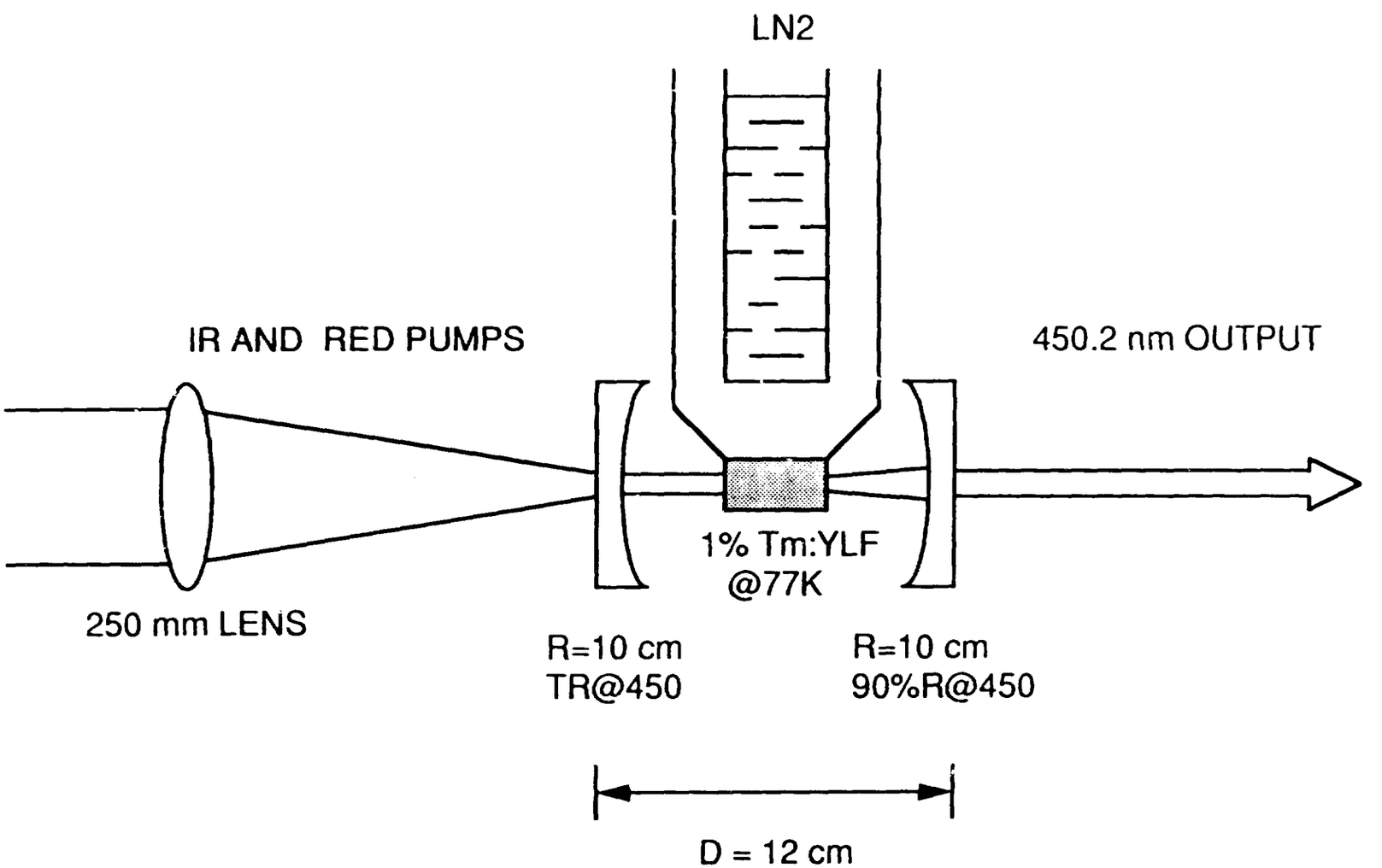
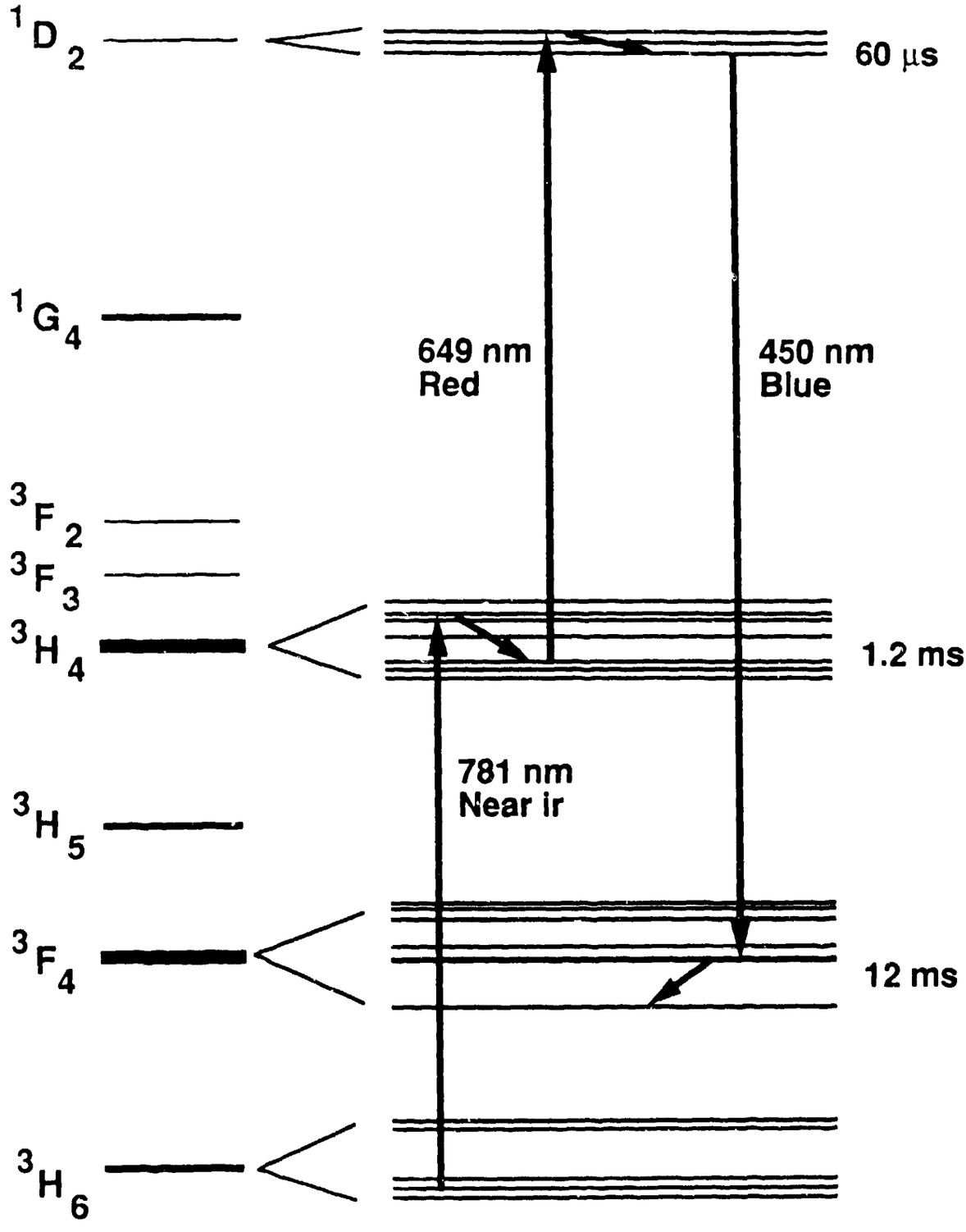

LN2
IR AND RED PUMPS
450.2 nm OUTPUT
1% Tm:YLF
@77K
250 mm LENS
R=10 cm
TR@450
R=10 cm
90%R@450
D = 12 cm


$^1D_2$
60 μs
$^1G_4$
649 nm
Red
450 nm
Blue
$^3F_2$
$^3F_3$
$^3H_4$
1.2 ms
781 nm
Near ir
$^3H_5$
$^3F_4$
12 ms
$^3H_6$

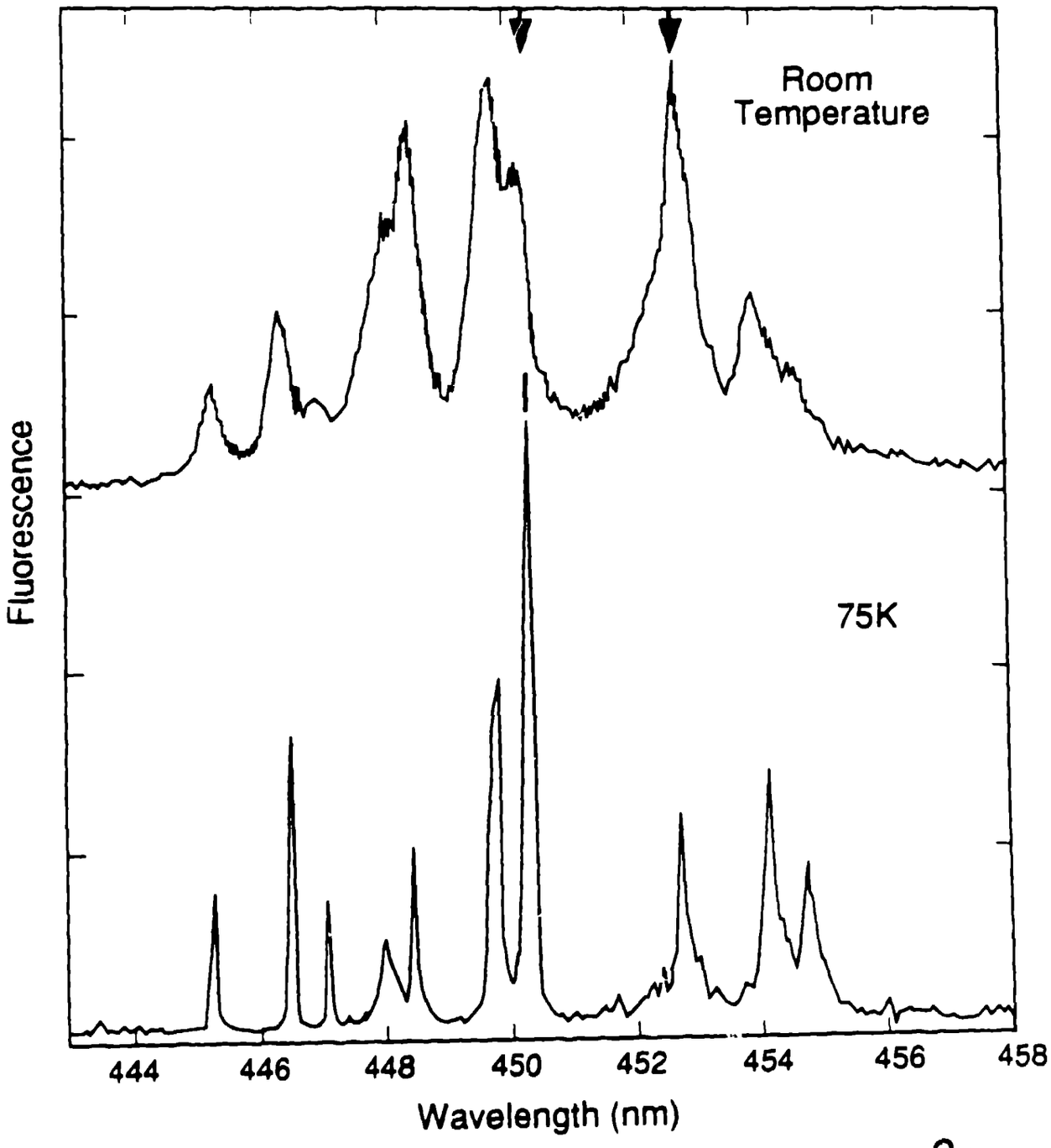
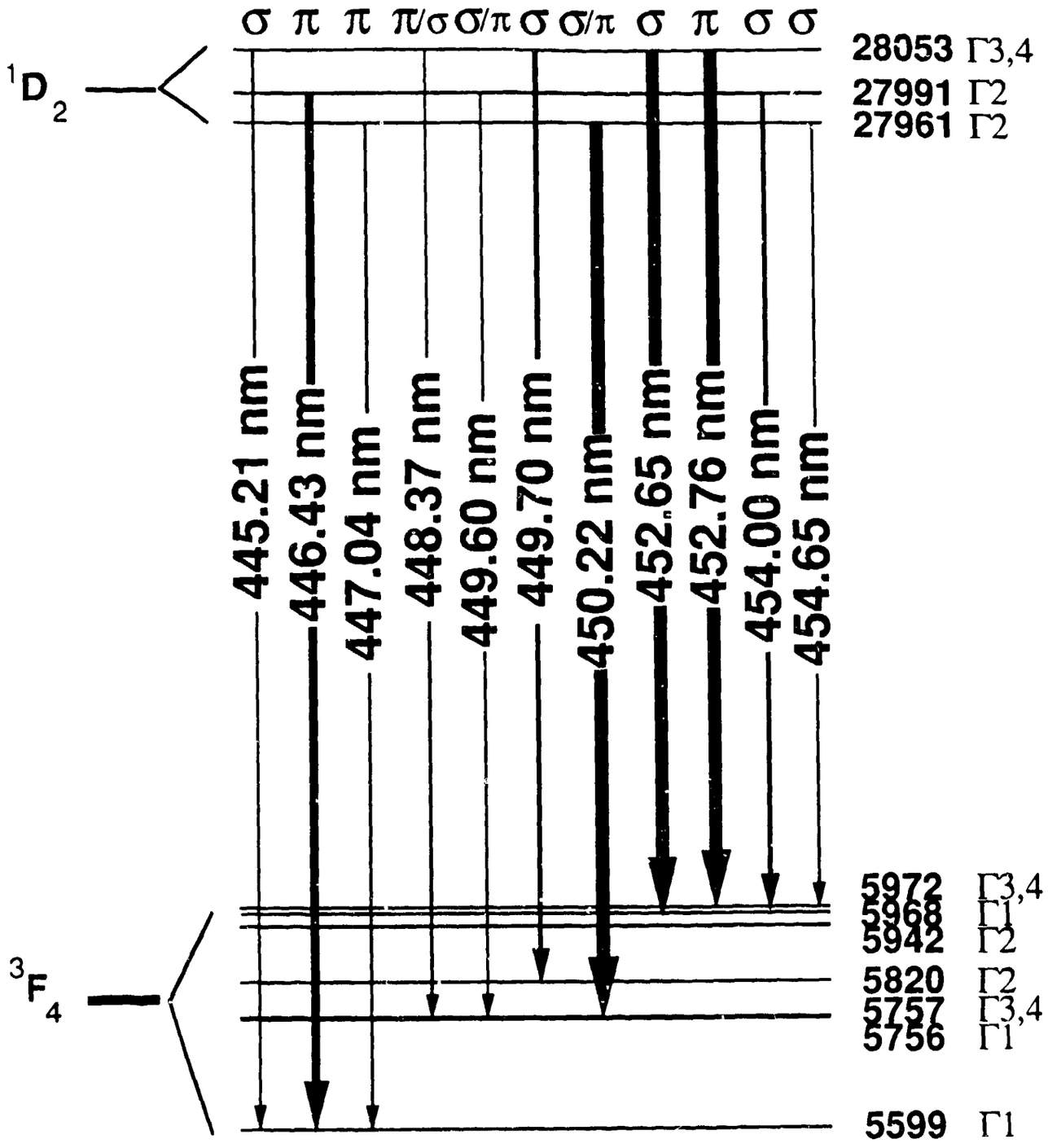
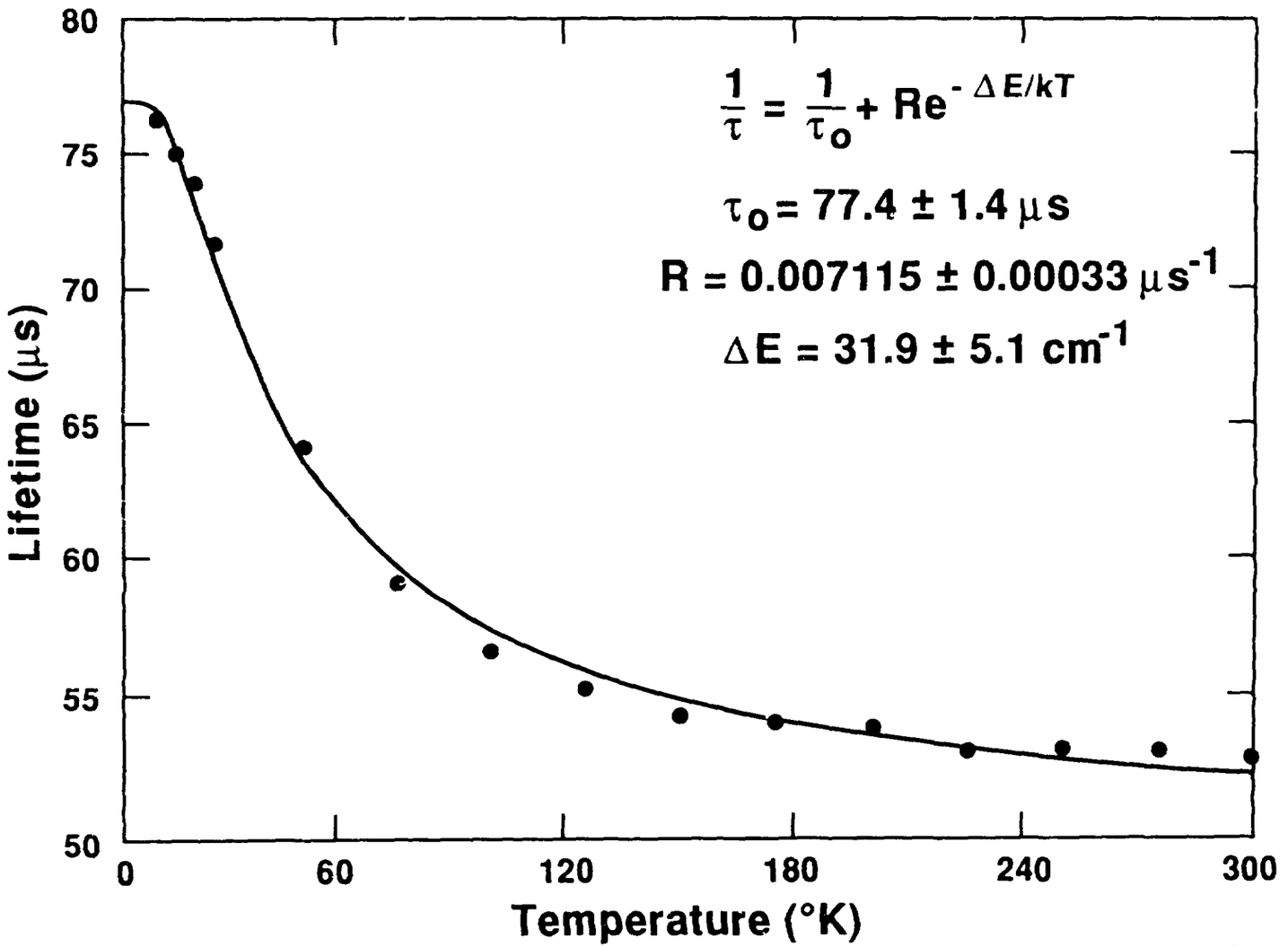


Figure 4. Fluorescence Spectra of Tm^{3+}:YLF at 75K and Room Temperature.


σ π π π/σ σ/π σ σ/π σ π σ σ
$^1D_2$
28053 Γ3,4
27991 Γ2
27961 Γ2
445.21 nm
446.43 nm
447.04 nm
448.37 nm
449.60 nm
449.70 nm
450.22 nm
452.65 nm
452.76 nm
454.00 nm
454.65 nm
$^3F_4$
5972 Γ3,4
5968 Γ1
5942 Γ2
5820 Γ2
5757 Γ3,4
5756 Γ1
5599 Γ1

FLUORESCENCE LIFETIME OF 1.5% Tm^{+3}:$LiYF_4$ $^1D_2$ LEVEL



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