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ENERGY TRANSFER IN A RARE EARTH DOPED ACTIVATOR-ACTIVATOR SYSTEM--ETC(U)  
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This report summarizes research carried out to investigate energy transfer in an activator-activator system; that is a system for which energy transfer is reversible between certain metastable levels and for which stimulated emission from each of these levels can be generated. The specific system investigated is Er-Ho:YLF. Laser operation from two different ions in the same solid state host has been demonstrated. The 750nm			

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$(^5S_2 - ^5I_7)$ :Ho) and 850nm  $(^4S_{3/2} - ^4I_{13/2})$ :Er) transitions were observed simultaneously and separately at 77K. Cross transfer between the S levels was also observed and the cross transfer rates measured in crystals of five different compositions. Cross transfer between the S levels is mitigated by other cross relaxation transfer schemes which quench the S level storage time of each ion. Only about 25% of the initial population of one ion is transferred to the other in the optimum concentration, 2%Ho-2%Er, at room temperature. The cross transfer rate is only weakly temperature and concentration dependent; at low temperatures cross transfer is somewhat more effective as the decay rates of each ion are longer.

The stimulated emission cross sections of the 750nm and 850nm laser transitions in Ho and Er:YLF was measured at room temperature and at 77K. The results show that the 750 transition in Ho:YLF might be an efficient laser in lamp pumped operation as the cross section at room temperatures for this four level transition is very high,  $9.7 \times 10^{-19} \text{ cm}^2$  (300K),  $1.6 \times 10^{-18} \text{ cm}^2$  (77K).

Laser operation from both transitions simultaneously or separately was observed in the composition 2%Ho 3.5%Er:YLF at 77K. In this composition the 850nm Er emission dominates at both 300 and 77K, in spite of its lower cross section ( $1.4 \times 10^{-19} \text{ cm}^2$  - 300K,  $7.6 \times 10^{-19} \text{ cm}^2$  - 77K)

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## 1.0 INTRODUCTION

This final report describes the research effort carried out to investigate energy transfer in an "activator-activator" system. This acronym is used to describe a system in which energy transfer between certain metastable levels is reversible and for which stimulated transitions from each of these metastable levels can be generated.

The specific vehicles for this investigation are the S levels of Er and Ho:YLF. Laser operation at 750nm and 850nm has been observed<sup>(1)</sup> from a single rod doped with Er and Ho:YLF. The 750 nm laser line in Ho<sup>3+</sup> is due to the  $^5S_2 - ^5I_7$  transition; the 850nm line in Er<sup>3+</sup> is due to the  $^4S_{3/2} - ^4I_{13/2}$  transition. Transfer between the S level metastables had previously been inferred from fluorescence spectra and studied in detail in this work.

### 1.1 Publications

During this program the following article was published:

"Deep Red Laser Emission in Ho:YLF" by E. P. Chicklis, C. S. Naiman, L. Esterowitz and R. Allen, IEE J. Quantum Electronics QE-13, No 10 1977.

The following will be submitted for publication

"Stimulated Emission Cross Section of the 750nm Laser Transition in Ho<sup>3+</sup>:YLF" by H. P. Jenssen

and

"Two Color Laser Operation in Er-Ho:YLF" by E. P. Chicklis and H. P. Jenssen.

Copies of these papers/abstracts are included as appendices to this report.

## 2.0 PROGRAM RESULTS

Spectroscopic and laser measurements were carried out to characterize the excitation and relaxation modes of the S levels ( $^4S_{3/2}$ :Er,  $^5S_2$ :Ho) in Er-Ho:YLF. The specific tasks included:

- Measurement of fluorescence yields from both ions under selective CW excitation
- Measurement of fluorescence rise and decay times at 300 and 77K under pulsed selective excitation
- Modeling of the energy transfer and quenching mechanisms of the S levels
- Optimization of the Er and Ho concentrations for maximized S level population
- Measurement of the stimulated emission cross sections of both laser transitions at 300 and 77K
- Laser performance measurements at 77K
- Crystal growth.

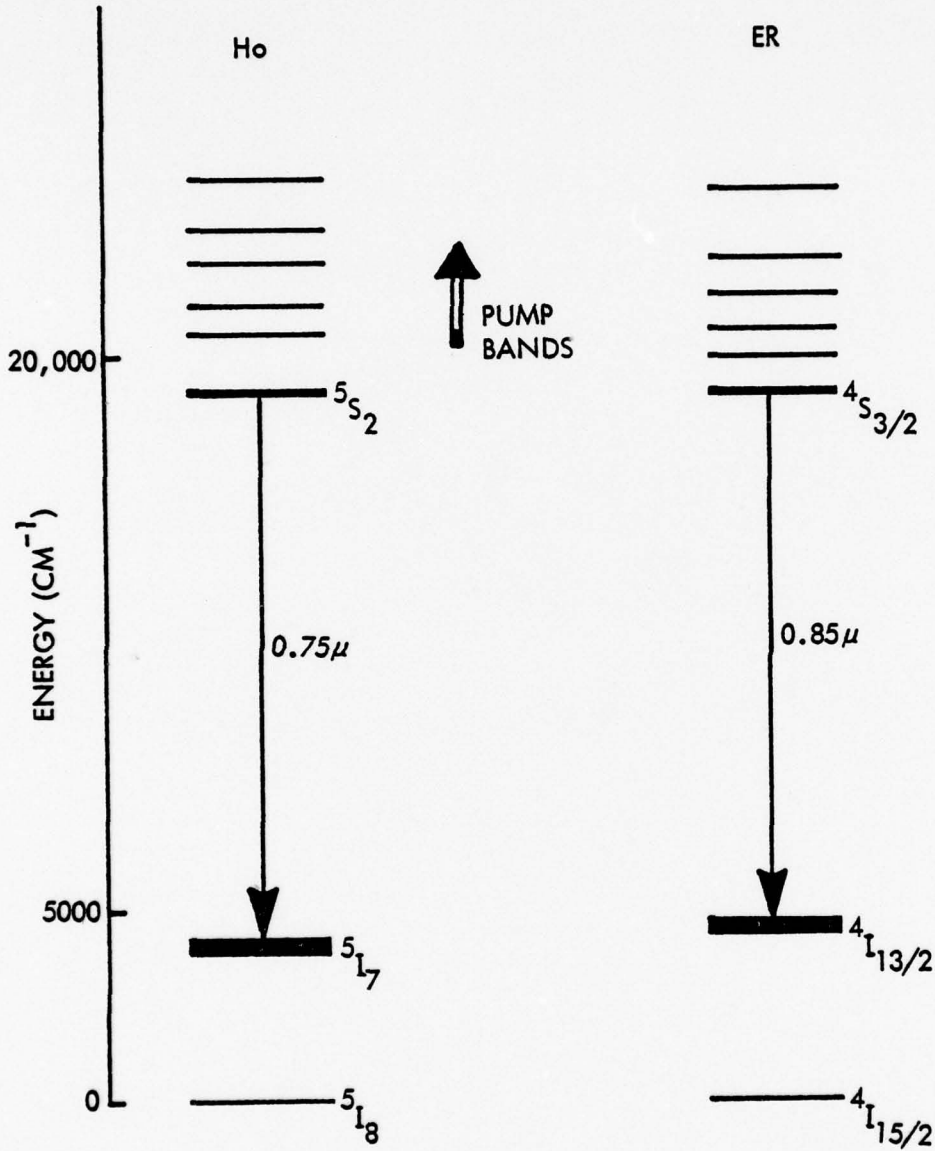
### 2.1 Spectroscopic Measurements - Energy Transfer

#### 2.1.1 Introduction

The  $^4S_{3/2}$  multiplet of  $Er^{3+}$  and the  $^5S_2$  multiplet of  $Ho^{3+}$  are nearly degenerate in energy and when the two ions both are present in a crystal one anticipates efficient resonant energy transfer (see Figure 1). The rate equations governing this transfer can, somewhat simplified, be expressed as:

# TWO COLOR (0.75/0.85) LASER

Ho - Er:YLF



PULSED THRESHOLD LN<sub>2</sub> 0.85: < 4 JOULES  
0.75: 12 JOULES

D-630

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FIGURE 1

$$\begin{aligned} \frac{dN_1}{dt} &= -W_1 N_1 - W_{12} N_1 + W_{21} N_2 + R_1 \quad (2-1) \end{aligned}$$

$$\frac{dN_2}{dt} = -W_2 N_2 - W_{21} N_2 + W_{12} N_1 + R_2$$

where  $N_1$  and  $N_2$  are the S-level populations of Er and Ho respectively,  $W_1$  and  $W_2$  are the normal relaxation rates of the S-levels without transfer and  $W_{12}$  and  $W_{21}$  are the transfer rates between Er and Ho.  $R_1$  and  $R_2$  are the pump rates. The main simplification here is that a simple rate is assured for transfer. This might be a good approximation when high concentrations of both Er and Ho are present in the crystal and every ion of one specie with high probability has an ion of the other specie as a nearest neighbor.

Based on the rate equations, experiments were performed in order to measure the transfer rates. CW excitation measurements and pulsed excitation measurements were carried out and are discussed below.

### 2.1.2 CW selective excitation of one ion only

Under steady state conditions we can solve for  $N_1$  and  $N_2$  directly. If we assume  $W_{12} = W_{21}$  we get, when  $R_1 = R$  and  $R_2 = 0$

$$N_1^1 = \frac{W_{12} + W_2}{W_1 W_2 + W_1 W_{12} + W_2 W_{12}} R \quad (2-2)$$

$$N_2^1 = \frac{W_{12}}{W_1 W_2 + W_1 W_{12} + W_2 W_{12}} R$$

and when  $R_1 = 0$  and  $R_2 = R^1$

$$\begin{aligned} N_1^2 &= \frac{W_{12}}{W_1 W_2 + W_1 W_{12} + W_2 W_{12}} R^1 \\ N_2^2 &= \frac{W_{12} + W_1}{W_1 W_2 + W_1 W_{12} + W_2 W_{12}} R^1 \end{aligned} \quad (2-3)$$

The measurable quantity under CW excitation is the relative fluorescent intensity, or

$$I_i = C_i W_i N_i \quad (2-4)$$

where  $W_i$  is the relaxation rate,  $N_i$  is the excited state population and  $C_i$  is a constant. From 2-2, 3 and 4 we then get:

$$\left( \frac{N_1^1}{N_2^1} \right) \left( \frac{N_1^2}{N_2^2} \right)^{-1} = \left( \frac{I_1^1}{I_2^1} \right) \left( \frac{I_1^2}{I_2^2} \right)^{-1} = \frac{(W_{12} + W_2)(W_{12} + W_1)}{W_{12}^2} \quad (2-5)$$

Figure 2 shown the Er  $^4S_{3/2} \rightarrow ^4I_{13/2}$  and the Ho  $^5S_2 \rightarrow ^5I_7$  emission under the two selective pumping conditions. The crystal had 2% Ho and 5% Er. From the spectra we have

$$\left( \frac{I_1^1}{I_2^1} \right) \left( \frac{I_1^2}{I_2^2} \right)^{-1} = 13.$$

$W_1$  and  $W_2$  determined from pulsed fluorescence measurements are:  $W_1 \approx 2 \times 10^4 \text{ sec}^{-1}$  and  $W_2 \approx 4 \times 10^4 \text{ sec}^{-1}$ . The resulting  $W_{12}$  is then from 5;

$$\frac{(W_{12} + 4 \times 10^4)(W_{12} + 2 \times 10^4)}{W_{12}^2} = 13$$

or

$$W_{12} \cong 2 \times 10^4 \text{ sec}^{-1}$$

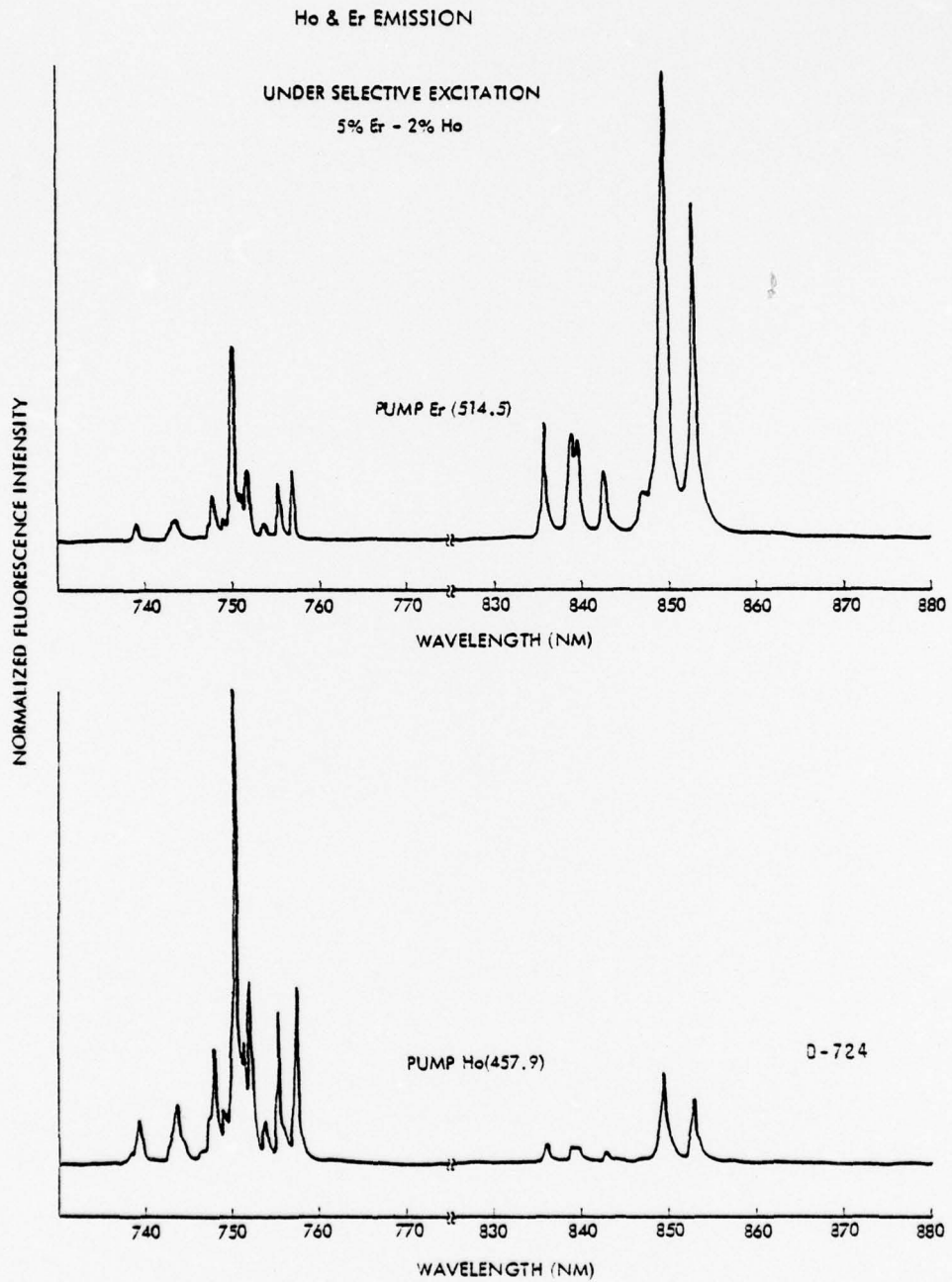


FIGURE 2  
Ho and Er Emission under Selective Excitation

### 2.1.3 Pulsed selective excitation

#### 2.1.3.1 Introduction

The general solutions to the coupled rate equations with  $W_{21}$  assumed equal to  $W_{12}$  after a short pump pulse ( $R = 0$ ) are

$$N_i = A_i e^{-\alpha t} + B_i e^{-\beta t} \quad (i = 1 \text{ or } 2) \quad (2-6)$$

$$\text{where } \alpha(\beta) = \frac{W_1 + W_2 + 2W_{12}}{2} (\pm) \frac{1}{2} \sqrt{(W_1 - W_2)^2 + 4W_{12}^2} \quad (2-7)$$

and  $A_i$  and  $B_i$  are constants determined by the initial conditions. For the initial conditions  $N_1 = N_0$  and  $N_2 = 0$ , the solution for  $N_2$  will be of the form:

$$N_2(t) = A_2 e^{-\alpha t} - A_2 e^{-\beta t} \quad (2-8)$$

$N_2$  max occurs when  $N_2 = 0$ , or

$$e^{(\alpha-\beta)t_{\max}} = \frac{\alpha}{\beta}$$

or

$$t_{\max} = \frac{\ln \alpha - \ln \beta}{\alpha - \beta}$$

Equation 2-7 and 2-8 provide a prescription for determination of the transfer rate from the observables  $t_{\max}$ ,  $W_1$  and  $W_2$ .  $W_1$  and  $W_2$  are simply the inverse fluorescence lifetimes of Er and Ho respectively. The parameter  $T_{\max}$  is the measured time after a short exciting pulse that the fluorescence peaks when one ion is resonantly pumped and the emission from the other ion monitored.

#### 2.1.3.2 Experimental - 300K

Pulsed excitation data were taken at room and 77K using a PHASE-R dye laser pumping small mixed crystals of Er and Ho:YLF. The experimental setup is diagramed in Figure 3. The PHASE-R was calibrated in wavelength using a Jarrel Ash 0.5 meter monochrometer. Estimated wavelength accuracy of the dye laser output was approximately  $\pm 2\text{\AA}$ .

PULSED EXCITATION EXPERIMENTAL SETUP

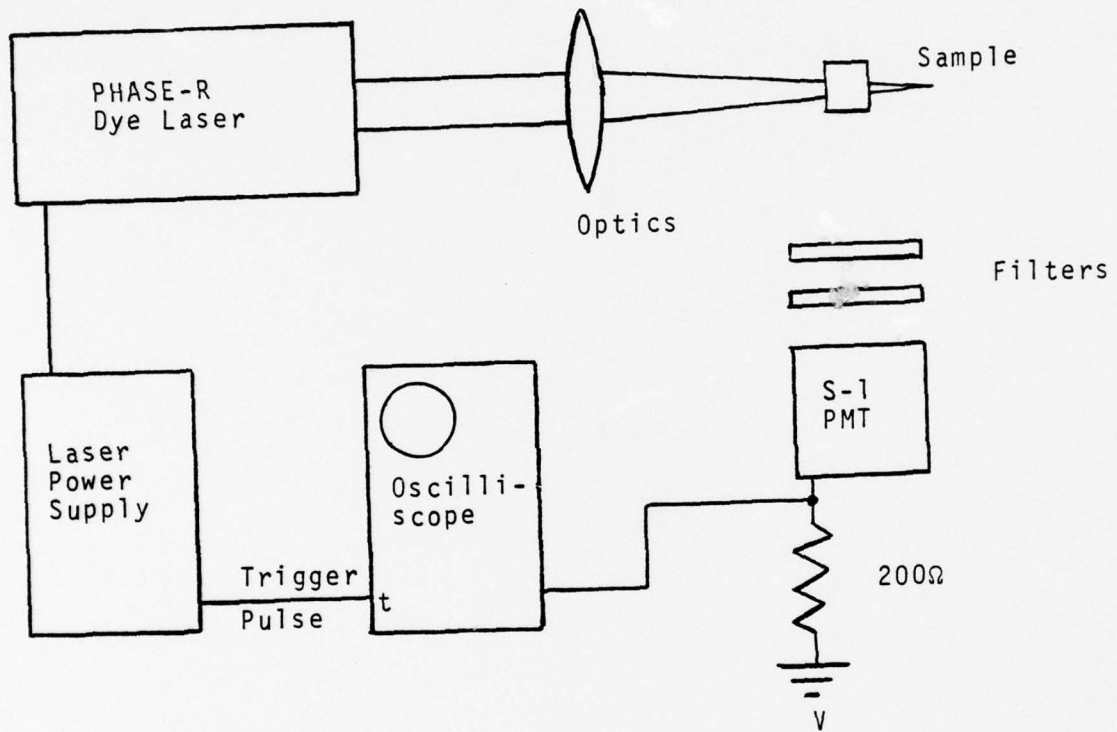


FIGURE 3

The dye laser output consisted of a single pulse of width  $\sim 500$ ns. This together with the large amounts of RFI generated by the high voltage flashlamp discharge limited the fluorescence rise time measurements to about  $1\mu$ s. Measurements of the relative pump band quantum efficiencies were not possible due to the very erratic amplitude variation of the dye laser.

Figure 4 and 5 show the transmission spectra of Ho and Er:YLF in the region of the S level pump bands. Since the object of these measurements was to selectively excite the Ho and Er pump bands separately, it was necessary to identify pump bands for which there was no overlap. At room temperature it was determined that 468nm pumped  $\text{Ho}^{3+}$  and 523nm pumped  $\text{Er}^{3+}$ . At 77K it was possible to pump  $\text{Ho}^{3+}$  at 533nm without overlap with the Er absorption.

Table 1 shows some of the results of these measurements at room temperature in both singly and multiply doped samples. In the singly doped samples the fluorescence rise is due to multiphonon relaxation from the pump band to the S level. For the pump bands investigated in Er and Ho the rate of this process is  $>10^6 \text{ sec}^{-1}$ .

In the multiply doped (Er and Ho) samples the S level fluorescence rise ( $.85\mu$  or  $.75\mu$ ) depends on which ion is pumped. Resonant pumping of a Er or Ho pump band results in rapid fluorescence buildup in the same ion at roughly the same rate as in the singly doped system. However, the fluorescence buildup time of the other ion is by contrast very long. This indicates that the S levels are only weakly coupled at least over the range of concentrations investigated. This is further evidenced by the fact that the measured fluorescence lifetimes of the two ions in the same crystal are very substantially different in contrast to previous data<sup>(2)</sup>: a transfer time much faster than the relaxation time of each ion would result in the same fluorescence lifetimes for both ions.

We can compare the results of the pulsed excitation experiments with the CW measurements in the following way. In section 2.1.2 a transfer rate  $W_{12} = 2 \times 10^4 \text{ sec}^{-1}$  was inferred from the ratio of

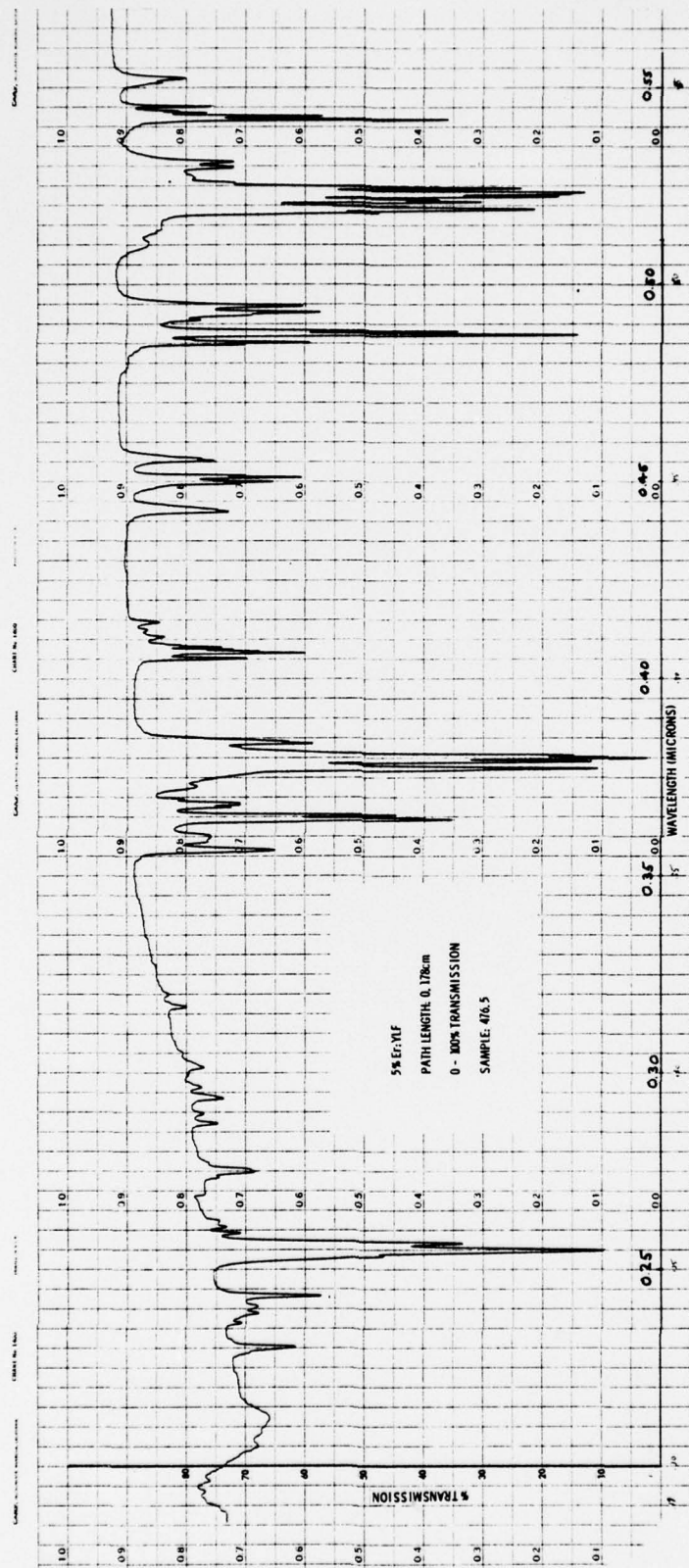
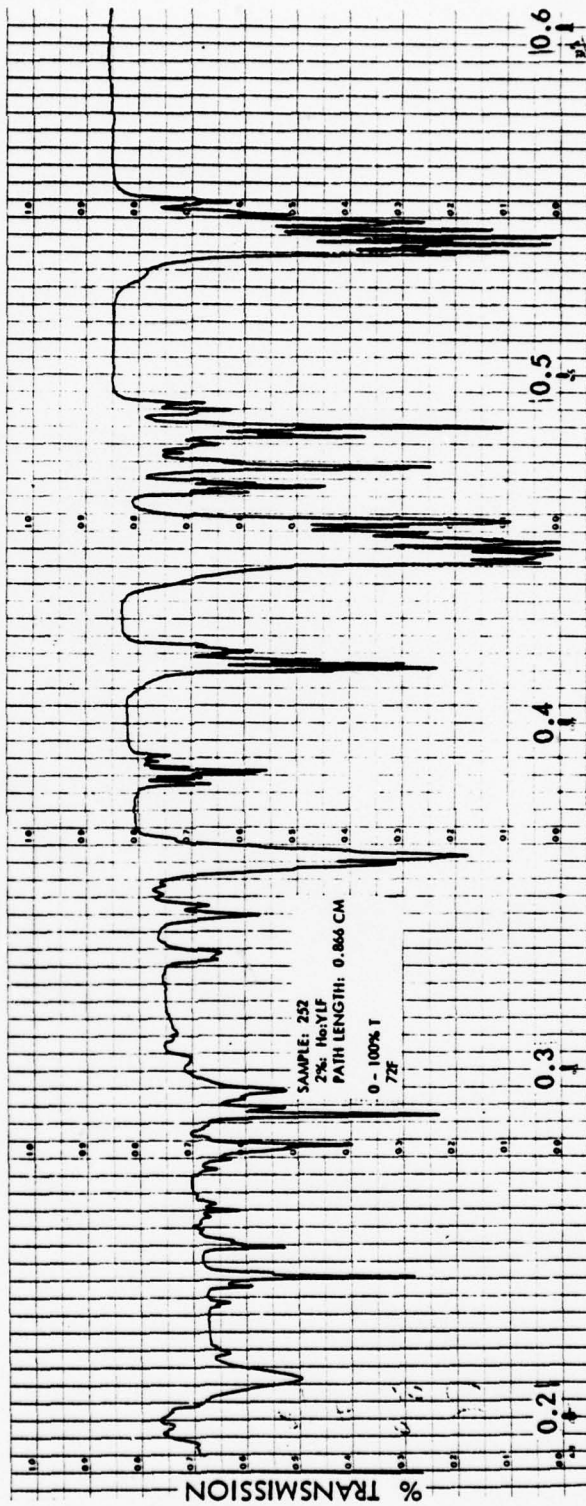


FIGURE 4



WAVELENGTH (MICROMETERS)

FIGURE 5

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TABLE 1  
PULSED EXCITATION DATA - ROOM TEMPERATURE

CRYSTAL	PUMP $\lambda$ (nm)	DETECTION $\lambda$ (nm)	TIME TO FLUORESCENCE PEAK ( $\mu$ s)	DECAY TIME ( $\mu$ s) $\pm 10\%$
17f-2%Ho	533 (Ho)	750 (Ho)	<1 $\mu$ s	75
467S-3.5%Er 10%Er	523 (Er)	850 (Er)	<1 $\mu$ s	100
	523 (Er)	850 (Er)	<1 $\mu$ s	20
489.3 2%Ho 3.5%Er	468 (Ho)	850 (Er)	20	76
	468 (Ho)	750 (Ho)	<1	40
	523 (Er)	750 (Ho)	20	88
	523 (Er)	850 (Er)	<1	65, 107
433 2%Ho 5%Er	468 (Ho)	850 (Er)	18	~80
	468 (Ho)	750 (Ho)	<1 $\mu$ s	35
	523 (Er)	750 (Ho)	16	70
	523 (Er)	850 (Er)	<1	50, 80
434 0.5%Ho 2%Er	468 (Ho)	850 (Er)	50	not measured
	468 (Ho)	750 (Ho)	<1 $\mu$ s	68
	523 (Er)	750 (Ho)	45	195
	523 (Er)	850 (Er)	<1 $\mu$ s	140, 235
435 0.5%Ho 5% Er	468 (Ho)	850 (Er)	23 $\mu$ s	not measured
	468 (Ho)	750 (Ho)	<1 $\mu$ s	43
	523 (Er)	750 (Ho)	23 $\mu$ s	86
	523 (Er)	850 (Er)	<1 $\mu$ s	60
436 2%Ho 2%Er	523 (Er)	750 (Ho)	25 $\mu$ s	140
	468 (Ho)	750 (Ho)	<1 $\mu$ s	41
	468 (Ho)	850 (Er)	~25 $\mu$ s	120
	523 (Er)	850 (Er)	<1 $\mu$ s	130

fluorescence amplitudes in the composition 2% Ho - 5% Er. In pulsed fluorescence measurements the decay rates for Er and Ho are respectively  $W_1 \cong 2 \times 10^4$  and  $W_2 \cong 4 \times 10^4 \text{ sec}^{-1}$  for a crystal of this composition. The peak of the fluorescence rise when the Ho ions were pumped and the 850nm Er fluorescence detected was  $t_{\text{max}} \sim 18\mu\text{s}$ . From equations 2-6, 2-7 using the measured values of  $W_1$ ,  $W_2$  and the CW value of  $W_{12}$  we calculate

$$t_{\text{max}} \sim 20\mu\text{s}$$

in good agreement with the measured value.

### 2.1.3.3 Experimental - 77K

Pulsed fluorescence measurements using the same setup described above were carried out at 77K. Samples were mounted in thermal contact with the cold finger of a laboratory dewar. The results are shown in Table II. The fluorescence rise times are equal for cross-transfer and the Er lifetime substantially longer than at room temperature. Using  $W_{12} = 2 \times 10^4$ ,  $W_1 = 5 \times 10^3$ ,  $W_2 = 1.4 \times 10^4$  we calculate using 2-6, 2-7

$$t_{\text{max}} = 45\mu\text{s}$$

close to what is observed. This indicates that the cross relaxation rate is only weakly temperature dependent.

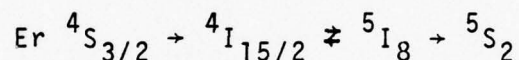
## 2.2 Optimization of the Ho and Er Concentrations

In the practical implementation of any activator-activator system it is necessary that the transfer rate between the two species,  $W_{12}$ , be very much faster than the decay rates of the individual ions. If this condition is not satisfied then the transfer is ineffective for laser operation. This is simply because the population of the meta-stable level is drained away faster than it is fed by energy transfer.

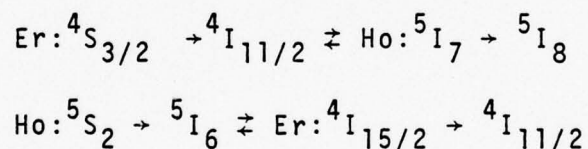
Table III lists the measured decay rates and calculated transfer rates,  $W_{12}$ , as a function of concentration and temperature. The

estimated accuracy of the calculated rates based on the uncertainties in the experimental values of  $t_{\max}$ ,  $W_1$ ,  $W_2$  is about  $\pm 25\%$ . The key conclusion is that the transfer rate is only weakly dependent on temperature and  $\text{Er}^{3+}$  concentration and furthermore, the cross relaxation rate  $W_{12}$  is not substantially faster than the  $\text{Ho}^{3+}$  decay rate for any composition or temperature investigated. In the case of Er at low temperatures or at low concentrations  $W_1 < W_{12}$  and moderate improvements in laser performance at 850nm would be anticipated by codoping with  $\text{Ho}^{3+}$ .

Thus this specific activator-activator system differs from the ideal in that the cross relaxation rate is not very much faster than the metastable decay rate. The difficulty with this system appears to be due to cross relaxation quenching of the S level metastables by lower lying levels of the other ion. Figure 5 shows the possible resonant transitions transfer and quenching of the S levels in Er and Ho:YLF. The transfer mechanism is believed to result from



with the double arrow denoting reversible transfer. The quenching mechanisms associated with cross transfers are believed to be



In addition there are competitive rates of self quenching in Er and Ho which are observed in the singly doped crystals.

Ho & Er:YLF S LEVEL  
 QUENCHING & TRANSFER  
 RESONANCES

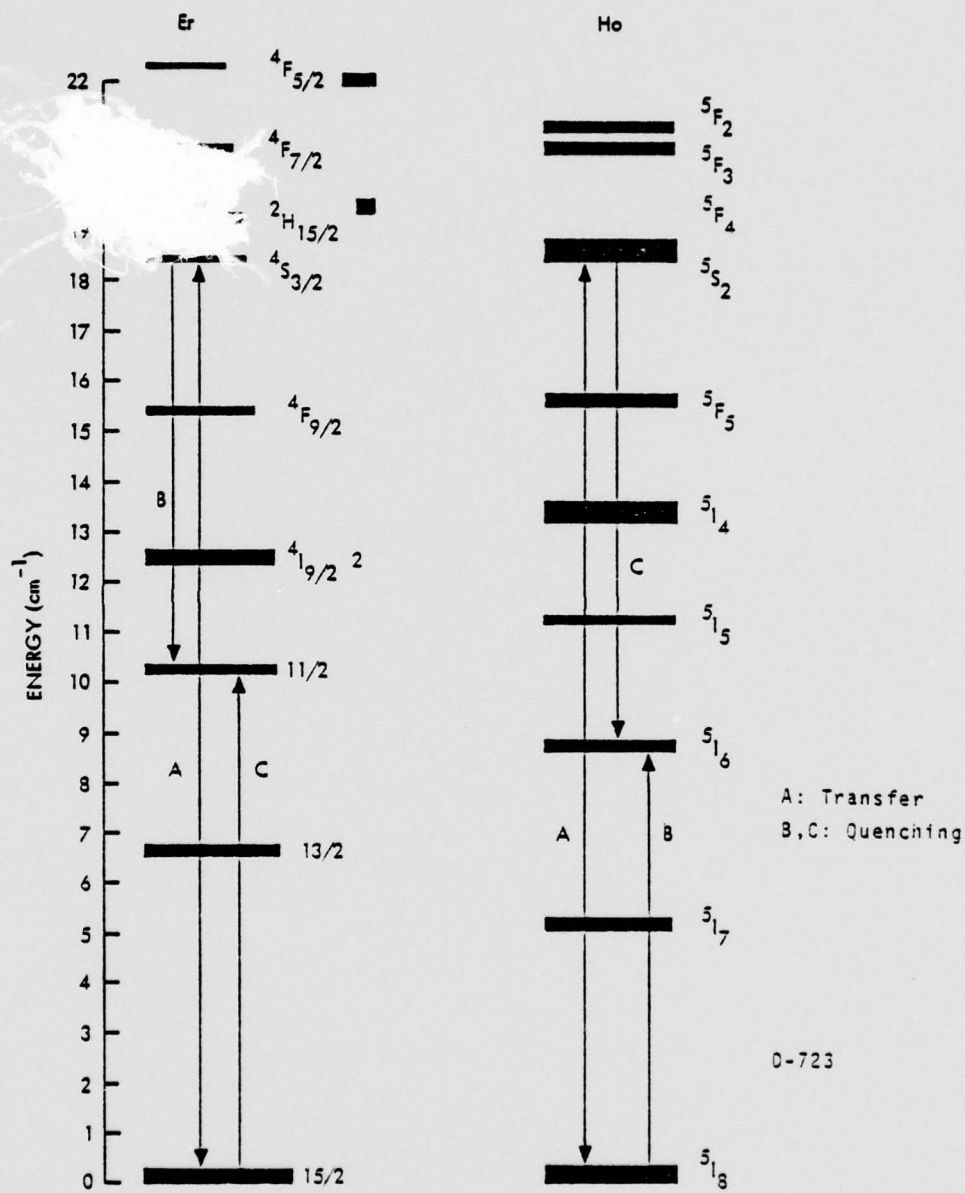


FIGURE 6

PULSED EXCITATION SPECTRA  
77K

CRYSTAL	PUMP $\lambda$ (nm)	DETECTION $\lambda$ (nm)	TIME TO FLUORESCENCE PEAK ( $\mu$ s)	DECAY TIME $\pm 10\%$ ( $\mu$ s)
489.3 2%Ho 3.5%Er	536 (Ho)	850 (Er)	40	200
	536 (Ho)	750 (Ho)	<1	68
	520 (Er)	850 (Er)	<1	200
	520 (Er)	750 (Ho)	40	200

TABLE II

DECAY AND TRANSFER RATES

	$W_1$ ( $\text{sec}^{-1}$ )	$W_2$ ( $\text{sec}^{-1}$ )	$W_{12}$ ( $\text{sec}^{-1}$ )
2%Ho - 2%Er 300K	$7.7 \times 10^3$	$2.4 \times 10^4$	$3-4 \times 10^4$
2%Ho - 3.5%Er 300K	$1.5 \times 10^4$	$2.5 \times 10^4$	$3-4 \times 10^4$
2%Ho - 5%Er 300K	$2 \times 10^4$	$4 \times 10^4$	$3-4 \times 10^4$
0.5%Ho - 2%Er 300K	$7.1 \times 10^3$	$1.47 \times 10^4$	$1.3 \times 10^4$
0.5%Ho - 5%Er 300K	$1.6 \times 10^4$	$2.3 \times 10^4$	$3 \times 10^4$
2%Ho - 3.5% Er 77K	$5 \times 10^3$	$1.47 \times 10^4$	$2 \times 10^4$

TABLE III

To quantify these results we calculate the population of the upper metastable at the fluorescence peak when pumped by cross transfer. Using equations 2-1, 2-6, the measured values of  $W_1$ ,  $W_2$ , the calculated values of  $\alpha$ ,  $\beta$  and  $W_{12}$ , and the initial conditions at  $t = 0$

$$R = 0, N_1 = N_0, N_2 = 0$$

we have

$$A_2 = \frac{W_{12}N_0}{\beta - \alpha} = -B_2$$

substituting into equation 2-6 we calculate  $N_2$  at  $t_{max}$ . Results are tabulated in Table IV below.

TABLE IV

CALCULATED IMERSION MAXIMA DUE TO CROSS TRANSFER

CRYSTAL	$t_{max}$	$N_2(\text{MAX})$
2%Ho - 2%Er	25 $\mu$ s	0.26 $N_0$
2%Ho - 3.5%Er	20 $\mu$ s	0.23 $N_0$
2%Ho - 5%Er	17 $\mu$ s	0.21 $N_0$
0.5%Ho - 2%Er	45 $\mu$ s	0.20 $N_0$
0.5%Ho - 5%Er	23 $\mu$ s	0.12 $N_0$
2%Ho - 3.5%Er 77K	40 $\mu$ s	0.30 $N_0$

$N_2(\text{max})$  corresponds to the effective transfer efficiency when one ion is selectively pumped. It is clear that of the initial population,  $N_0$  in one ion only about 20% is effectively transferred to the other ion. The transfer efficiency would be much higher if  $W_{12} \gg W_1, W_2$  which is not the case for any of the compositions examined at room temperature. The "optimum concentration" for transfer is

~2%Ho - 2%Er at room temperatures.

For broad band pumping however, with Xe flashlamps cross-relaxation quenching requires that very short lamp pulses be utilized to efficiently pump the system. The enhanced pumping efficiency of each ion by cross-transfer may be mitigated by the reduction in the fluorescence lifetime for both Er and Ho. At 77K Ho performance may be improved by cross transfer as the  $^5S_2$  lifetime at 77K in singly doped samples is about the same as in the co-doped crystal.  $Er^{3+}$  singly doped would be more efficient since the  $^4S_{3/2}$  lifetime is very long even in very high concentrations. For example, the fluorescence lifetime in  $LiErF_4$  is about the same as in 3.5%Er - 2%Ho at 77K. The absorption efficiency of the former would be considerably higher<sup>(3)</sup> than the latter.

## 2.3 Stimulated Emission Cross Section

### 2.3.1 Introduction

The key parameters which determine the performance of a laser are the pumping efficiency and the stimulated emission cross section. In the previous section measurements of energy transfer between the S levels were described with the conclusion that the energy transfer rate between the two S levels was not sufficiently fast to compete efficiently with decay rates out of the S levels, particularly in the case of  $\text{Ho}^{3+}$ . As a result in the co-doped systems pumping efficiency higher than that in the single ion system does not appear likely.

In order to understand the results of laser operation of this system the stimulated emission cross sections of the  ${}^5\text{S}_2 \rightarrow {}^5\text{I}_7$  (750nm - Ho) and the  ${}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{13/2}$  (850nm - Er) laser transitions were directly measured. The latter had been measured previously<sup>(3)</sup> at room temperature. On this program both cross sections were measured at 300 and 77K.

The method used for measuring the stimulated emission cross sections is the one described by Kushida<sup>(4)</sup> and later by Singh et al.<sup>(5)</sup> The method involves measuring the peak absorption coefficients between the levels of the ground state manifold and the levels of the excited state manifold of  ${}^5\text{S}_2$  (Ho) and  ${}^4\text{S}_{3/2}$  (Er). The absorption cross section is then determined from the relation

$$\sigma_{ij} = \alpha_{ij}/N_i$$

where  $\sigma_{ij}$  is the peak absorption cross section for the line from level i in the groundstate manifold to level j in the upper manifold,  $\alpha_{ij}$  is the peak absorption coefficient for the same line, and  $N_i$  is the population density in level i given by the active ion concentration and Boltzmann statistics.

The stimulated emission cross section for the  $j \rightarrow i$  transition is given by

$$\sigma_{ji} = \frac{g_j}{g_i} \sigma_{ij}$$

where  $g_i$  is the degeneracy of level  $i$ . The stimulated emission cross section for a transition from level  $j$  to a level  $k$  that has no thermal population and therefore can not be observed in absorption, is given by

$$\frac{\sigma_{jk}}{\sigma_{ji}} = \left(\frac{n_{ji}}{n_{ij}}\right)^2 \left(\frac{\lambda_{jk}}{\lambda_{ji}}\right)^5 \left[ \left(\frac{dI}{d\lambda}\right)_{jk} \left(\frac{dI}{d\lambda}\right)_{ji} \right]$$

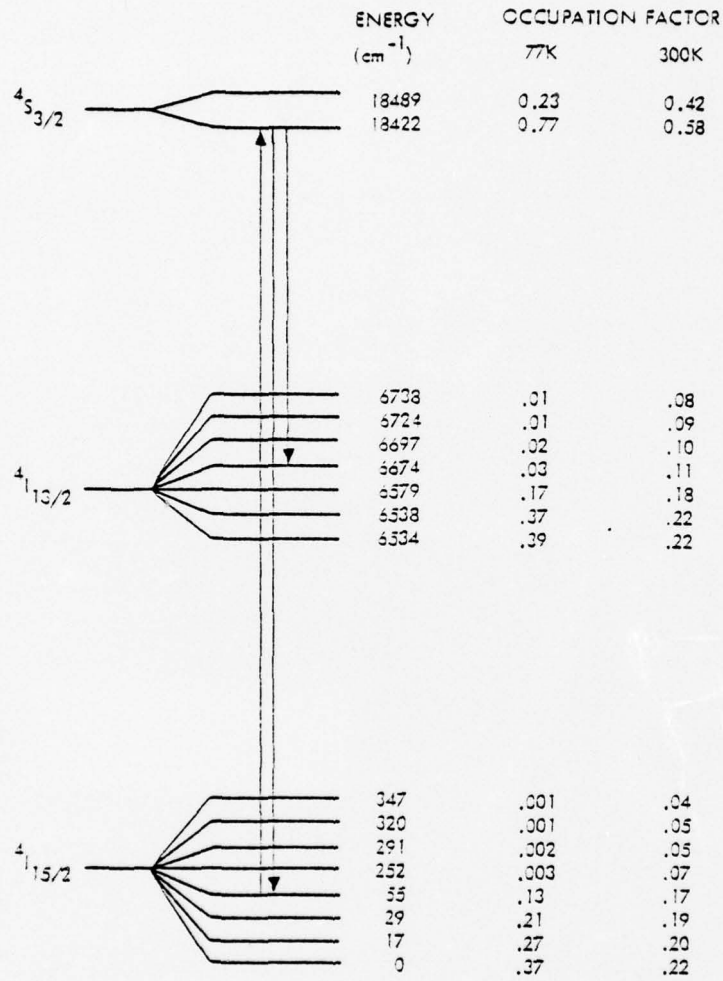
where  $n$  is the refractive index,  $\lambda$  is the vacuum wavelength, and  $\frac{dI}{d\lambda}$  is the peak emission power per unit wavelength interval. Thus, if  $\sigma_{ji}$  is known from absorption,  $\sigma_{jk}$  can be found by measuring the peak fluorescence intensities for the two lines.

### 2.3.2 Er:LiYF<sub>4</sub> (Crystal No. 215f)

The 850 nm laser transition in Er:YLF is from the  $^4S_{3/2}$  multiplet to the  $^4I_{13/2}$  multiplet. The ground state manifold is the  $^4I_{15/2}$  multiplet. The crystal field split levels are listed in Figure 7. The laser transition from  $E_1$  to  $B_4$  is indicated, as is the transition between  $A_4$  and  $E_1$ . The latter transition was used to determine the cross section. The laser line is polarized with the E vector parallel to the c-axis of the YLF crystal ( $\pi$ -polarized) such that all absorption and fluorescence measurements have to be performed with polarized light.

The absorption measurements were performed with the crystal sample mounted in a variable temperature dewar (Janis Super Vari Temp) with the dewar tail fitted into the sample chamber of a Cary 14 double beam recording spectrophotometer. The fluorescence measurements were performed with a 0.3 m McPherson spectrometer and a PMT with S-1 photocathode. The spectra were normalized to yield flat response over the wavelength region of interest by the use of a dedicated mimicomputer. The system response vs wavelength has previously been determined by the help of a calibrated 1,000 W tungsten halogen lamp. Calcite Glan-prisms were used as polarizers, both in absorption and fluorescence measurements.

Er:YLF ENERGY LEVELS



D-720

Figure 8

Er:YLF ENERGY LEVELS

Figure 8 shows the polarized absorption spectra of the  ${}^4I_{15/2} \rightarrow {}^4S_{3/2}$  transitions. As can be seen, the  $A_4 \rightarrow E_1$  transition is free of overlap of other lines. The peak absorption cross section for this line in the  $\pi$ -spectrum is:

$$\sigma_{A_4 \rightarrow E_1} = 0.75 \times 10^{-19} \text{ cm}^2 \text{ (77K)}$$

Figure 9 shows the polarized fluorescence spectra, including both the  $E_1 \rightarrow A_4$  transition and the  $E_1 \rightarrow B_4$  transition. From the absorption cross section  $\sigma_{A_4 \rightarrow E_1}$  and the ratio of the fluorescence intensities, the stimulated emission cross section for the  $E_1 \rightarrow B_4$  transition is determined from Eq. 2-8. The result is:

$$\sigma_{E_1 \rightarrow B_4} = 7.6 \times 10^{-19} \text{ cm}^2 \text{ (77K)}$$

The absorption cross section obtained for the  $A_4 \rightarrow E_1$  transition is critically dependent on the actual  $\text{Er}^{3+}$  concentration in the crystal. It is here assumed that the concentration is the same as in the melt from which the crystal was grown. The linewidth of the laser line at 350.3 nm is  $2.8 \text{ cm}^{-1}$  (FWHM).

At room temperature the absorption cross section for the  $A_4 \rightarrow E_1$  transition in the  $\pi$ -spectrum is  $1.9 \times 10^{-20} \text{ cm}^2$ . The resulting stimulated emission cross section for the  $E_1 \rightarrow B_4$  transition at 300K is:

$$\sigma_{E_1 \rightarrow B_4}(\pi) = 1.4 \times 10^{-19} \text{ cm}^2$$

The room temperature cross section had been previously measured on another program.<sup>(3)</sup> The results obtained here agree to within 10% of the previous measurement.

### 2.3.3 Ho:YLF 750nm Laser Transition

Figure 10 shows the energy levels of the  $\text{Ho}^{3+}$   ${}^5I_8$ ,  ${}^5I_7$  and  ${}^5S_2$ ,  ${}^5F_4$  multiplets in  $\text{LiYF}_4$ . The laser transition and the ground manifold transition used to determine the cross section are indicated. The method used to measure the stimulated emission cross section here is the same as used for Er:YLF.

Er:YLF ABSORPTION

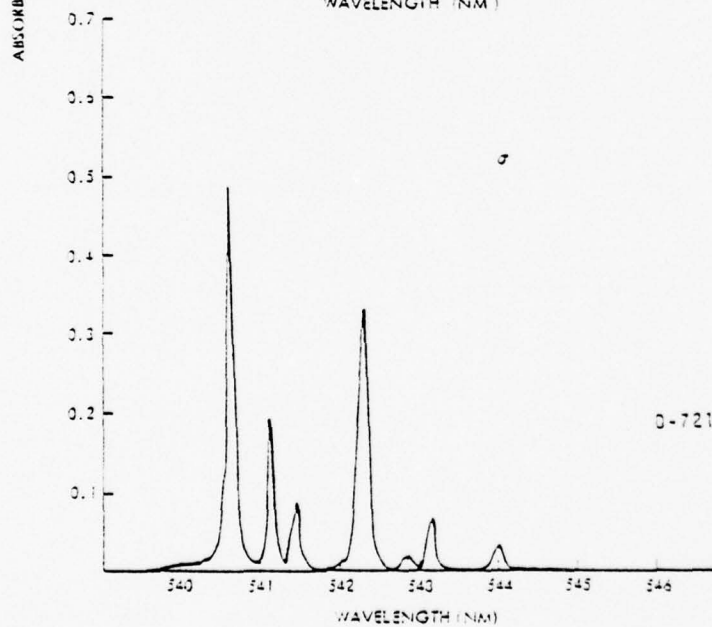
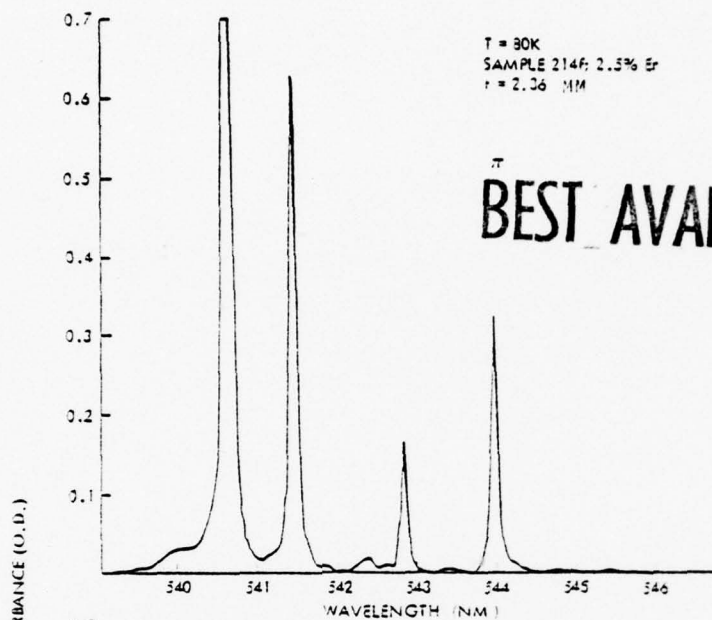
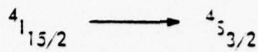
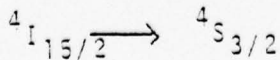


Figure 8

POLARIZED ABSORPTION SPECTRUM, Er:YLF



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Er:YLF EMISSION SPECTRA

77K

SAMPLE 214f; 2.5% Er

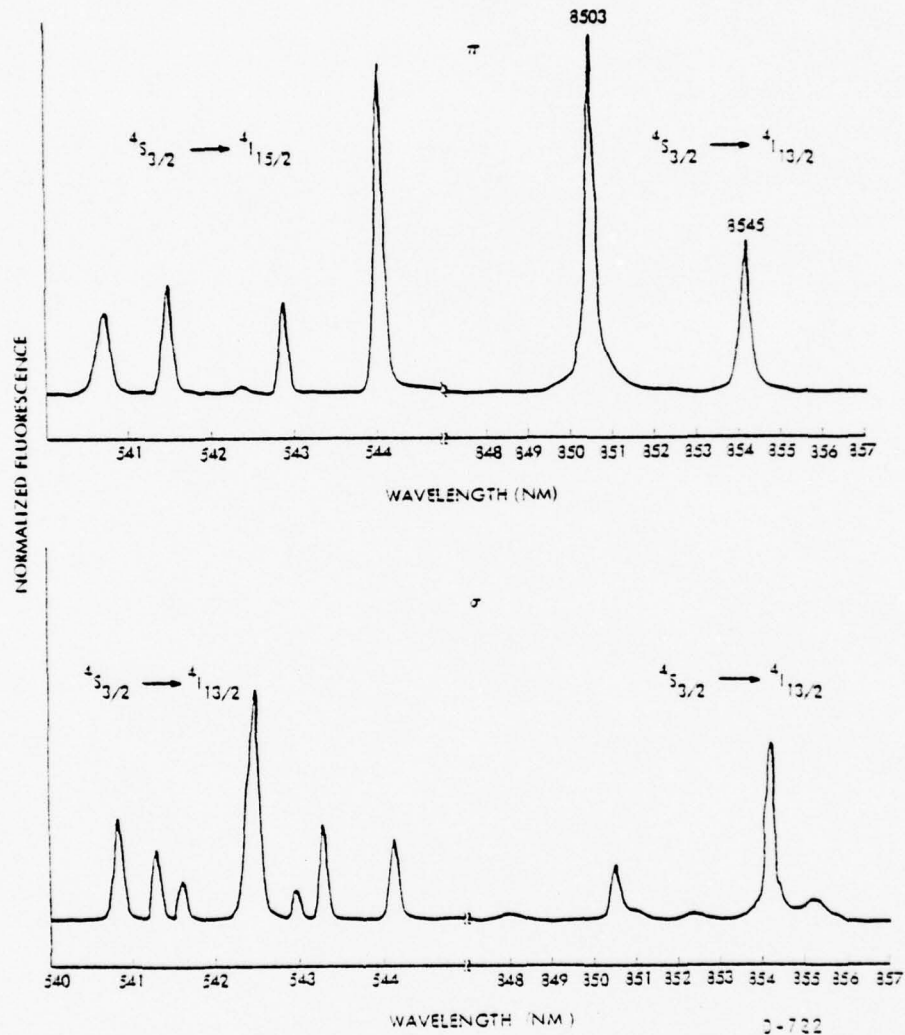


Figure 9

POLARIZED FLUORESCENCE SPECTRUM

Er:YLF,  $4S_{3/2} - 4I_{15/2}$ ,  $4S_{3/2} - 4I_{13/2}$

Ho:YLF ENERGY LEVELS



Figure 10

The peak absorption cross section for the  $A_5(56) \rightarrow E_1(18489)$  transition at 542.4 nm is  $5.3 \times 10^{-20} \text{ cm}^2$  at 300K and  $9.6 \times 10^{-20} \text{ cm}^2$  at 80K. The corresponding stimulated emission cross section for the  $E_1(18489) \rightarrow B_4(5165)$  transition at 750.3 nm is  $1.6 \times 10^{-18} \text{ cm}^2$  at 80K and  $9.7 \times 10^{-19} \text{ cm}^2$  at 300K. The laser transition is  $\pi$ -polarized and the fluorescent linewidth at 80K is  $2.5 \text{ cm}^{-2}$ . At room temperature the linewidth is  $12 \text{ cm}^{-2}$  and the emission line is composed of at least two unresolvable transitions. The peak stimulated emission cross section at room temperature determined did not take this into account and might therefore be in error by an estimated  $\pm 30\%$ . To take into account the overlapping transitions, knowledge of which levels are involved is necessary. Since there is some uncertainty about the exact position of some of the upper levels in all the multiplets, resolving the 750.3 nm emission line into its individual components has not been attempted. Spectra are shown in Figures 11 and 12.

The parameter of interest when evaluating laser properties is the gain at a total upper manifold population. Thus when comparing lasers such as the 750 nm Ho laser and the 850 nm Er laser, the product of the stimulated emission cross section and the occupation factor of the upper laser level should be used. These numbers are given in Table V together with Nd:YAG as a reference.

Note that the laser threshold is inversely proportional to the  $F\sigma$  product. So for equal pumping efficiencies and fluorescence lifetimes one would expect at room temperature that Ho:YLF would have a lower threshold than Er:YLF and conversely at 77K. The observed laser data suggests that although the S levels are coupled, at least at room temperature, the ions exhibit different pumping efficiencies. Er:YLF exhibits a lower threshold at room and at 77K.

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## POLARIZED ABSORPTION SPECTRUM

0.5% Ho: YLF (266f)

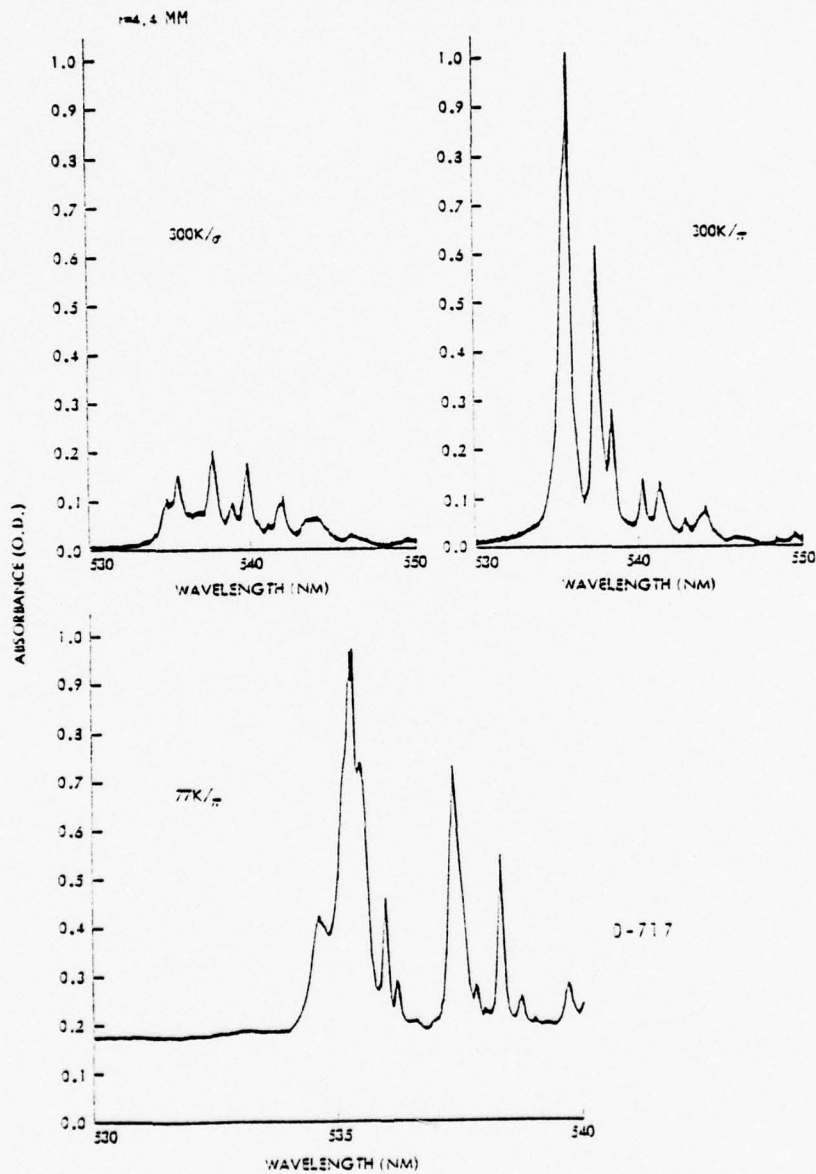


Figure 11

POLARIZED ABSORPTION SPECTRA

Ho: YLF

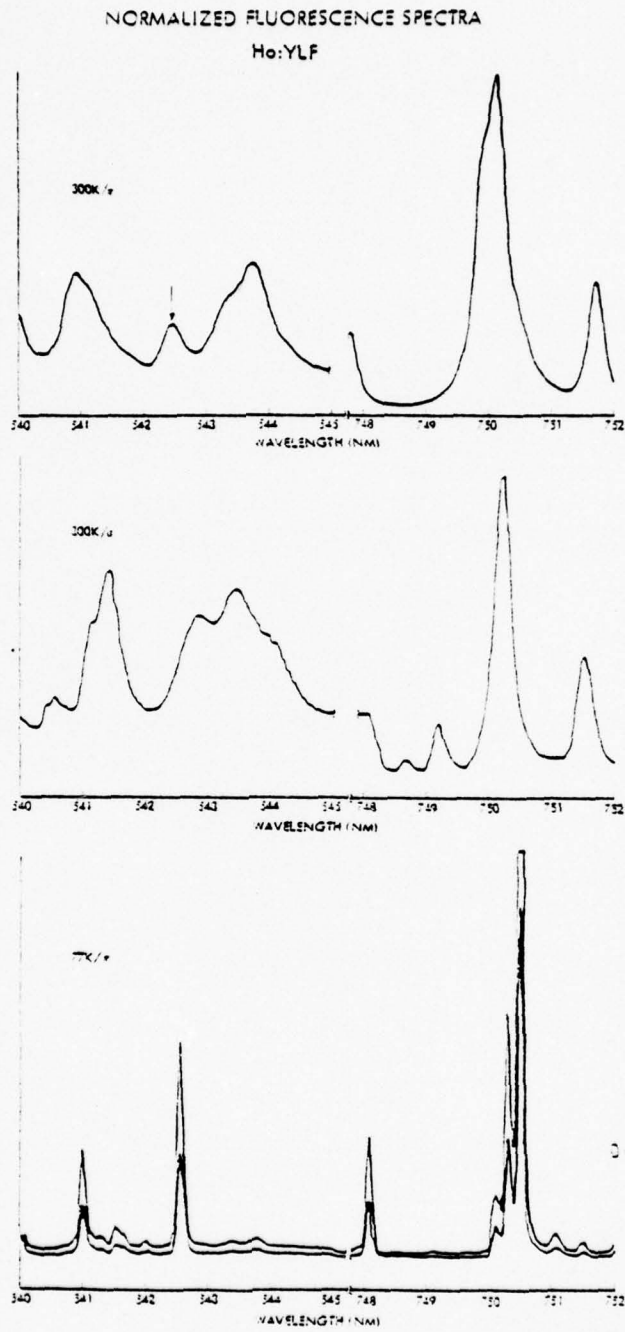


Figure 12

POLARIZED EMISSION SPECTRA

Ho:YLF

TABLE V

## CROSS SECTIONS &amp; OCCUPATION FACTORS

	Temperature	$\sigma$ ( $\times 10^{-19} \text{cm}^2$ )	F	F x $\sigma$
Er:YLF	300	1.4	0.58	0.81
	77	7.6	0.77	5.85
Ho:YLF	300	9.7	0.11	1.07
	77	16	0.24	3.84
Nd:YAG	300	5	0.4	2

$\sigma$  = Stimulated Emission Cross Section

F = Occupation Factor of Upper Laser Level

Laser Gain Proportional to F x  $\sigma$

## 2.4 Laser Measurements

Pulsed laser measurements were carried out using a rod of 3.5%Er-2%Ho of dimensions 5 x 50 mm. The rod was high reflectivity coated at 750-850nm on one end and AR coated on the other. The experimental setup is shown in Figure 13; this pumping system is a modified version of a commercial laser product, Sanders Model 5[7] laser. In this setup the rod is cryogenically cooled with flowing LN<sub>2</sub> and pumped by a linear flashlamp. Gain at each transition was controlled by the coupling mirror reflectivity.

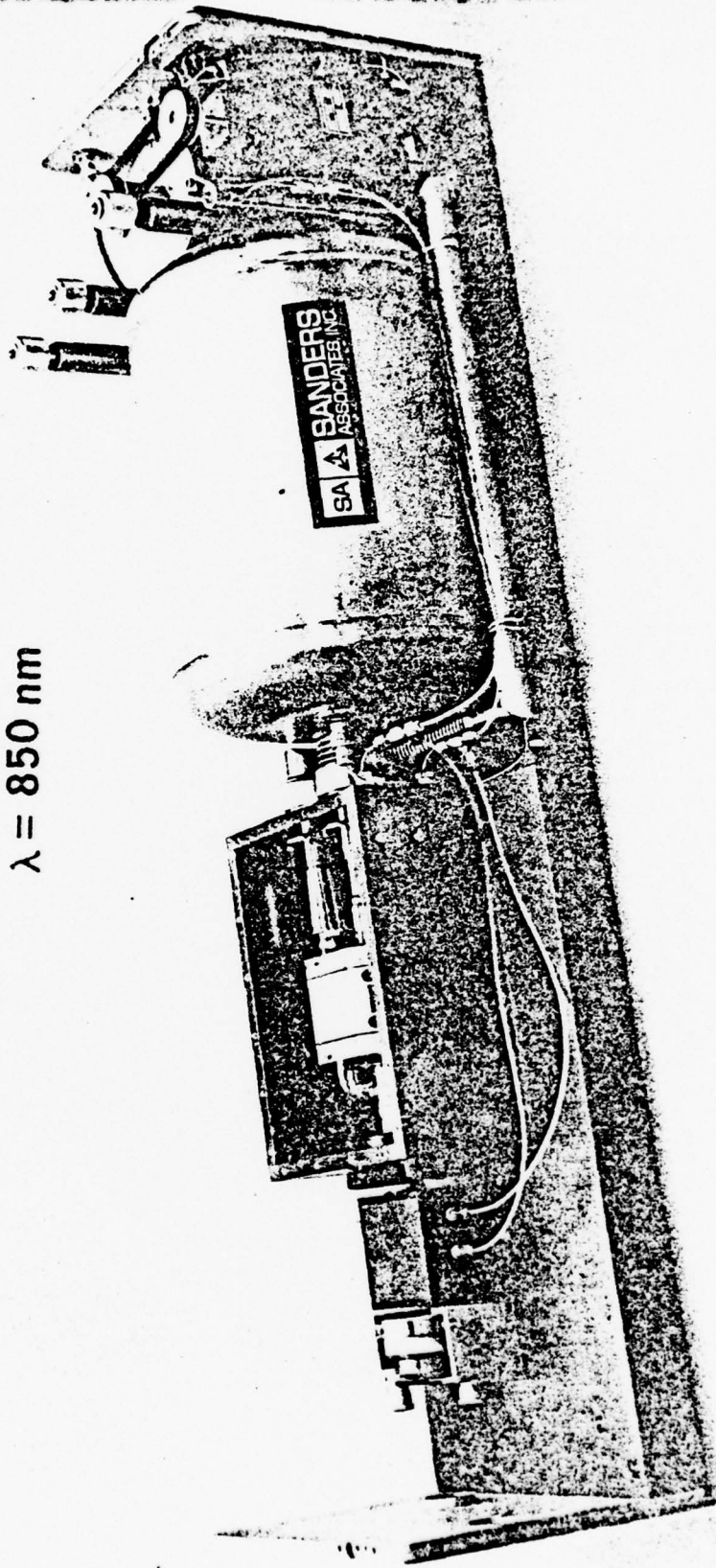
Laser operation of both colors (0.75 and 0.85 $\mu$ m) was demonstrated both simultaneously and separately in this crystal. Laser output power measurements reveal that in this composition (3.5%Er - 2%Ho) the predominant power output is at 0.85 $\mu$ m (Er). Figure 14 shows 77K laser performance data with a 2%Ho:3.5%Er:YLF laser rod operated with different coupling mirror reflectivities. For the same output transmission the .75 threshold is a factor of 5 higher than 0.85 threshold. The three traces of Figure 15 show the 0.85 and 0.75 $\mu$ m spiking oscillations and the flashlamp pulse. All three detectors are triggered the same way. The onset of spiking a 0.75 $\mu$ m threshold suggest that these levels might be decoupled at low temperatures - and that the <sup>4</sup>S<sub>3/2</sub> (Er) lifetime is considerable longer than <sup>5</sup>S<sub>2</sub> (Ho) at 77K. Another possible mechanism is that the 0.75 laser oscillations are quenched (possibly by excited state absorption) as lower lying levels are populated by direct pumping and by relaxation. Specific excited state absorption transitions however, were not identified. A further point of interest is that the Er oscillations are observed up to 100 $\mu$ s after the flashlamp suggesting a strong pumping mechanism (possibly upconversion) separate from multiphonon relaxation from pump bands below the S levels. This is inferred from the fact that none of the levels above the S levels exhibit fluorescence lifetimes of this magnitude. The mechanism for the delayed stimulated emission was not investigated in detail.

FIGURE 13

**2 COLOR LASER TEST CAVITY**

$\lambda = 750 \text{ nm}$

$\lambda = 850 \text{ nm}$



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## 0.75/0.35 MICRON LASER PERFORMANCE

2% Ho - 3.5% Er

77K

A: 0.35 $\mu$  ONLY, 32%R,  $E_T = 5J$

B. SIMULTANEOUS: 35%R AT 0.75,  $E_T = 12J$

45%R AT 0.35,  $E_T = 9J$

C. SIMULTANEOUS: 95%R AT 0.75,  $E_T = 10J$

95%R AT 0.35,  $E_T = 3J$

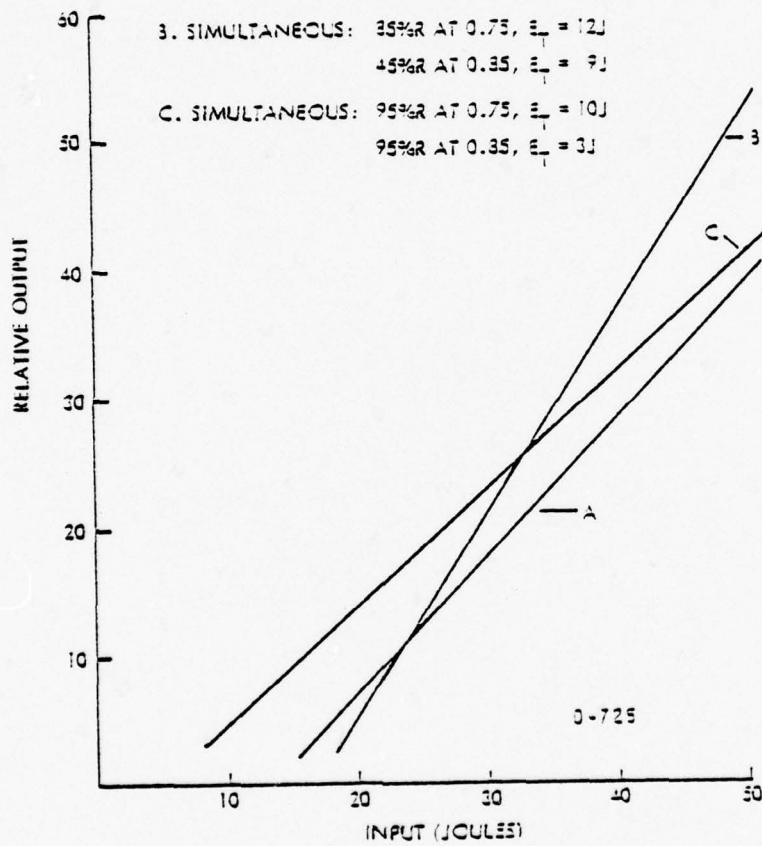


Figure 14

TWO COLOR LASER PERFORMANCE (U)

SIMULTANEOUS .75/.85 LASER OSCILLATIONS

Output Coupling; 95% at 0.75

94% at 0.85

ROD 489.2: 2% Ho, 3.5% Er:YLF

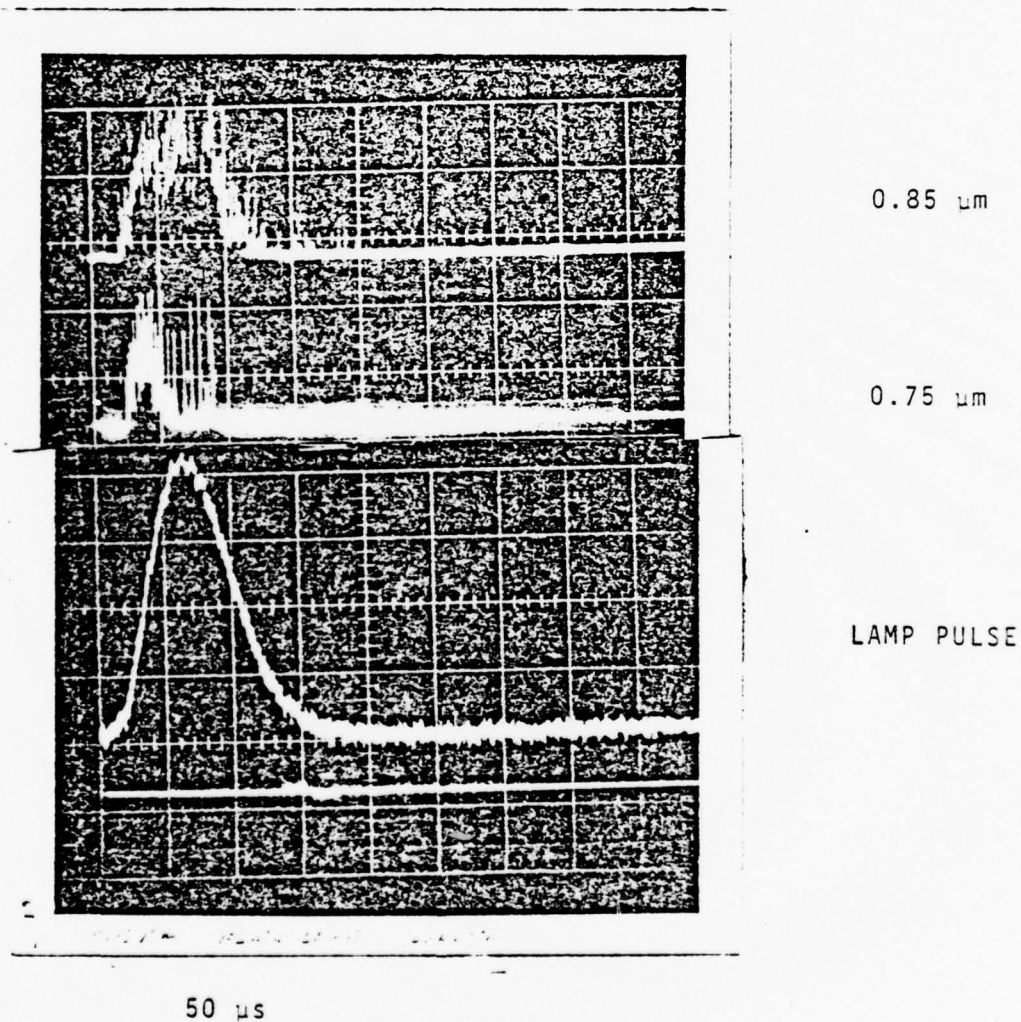


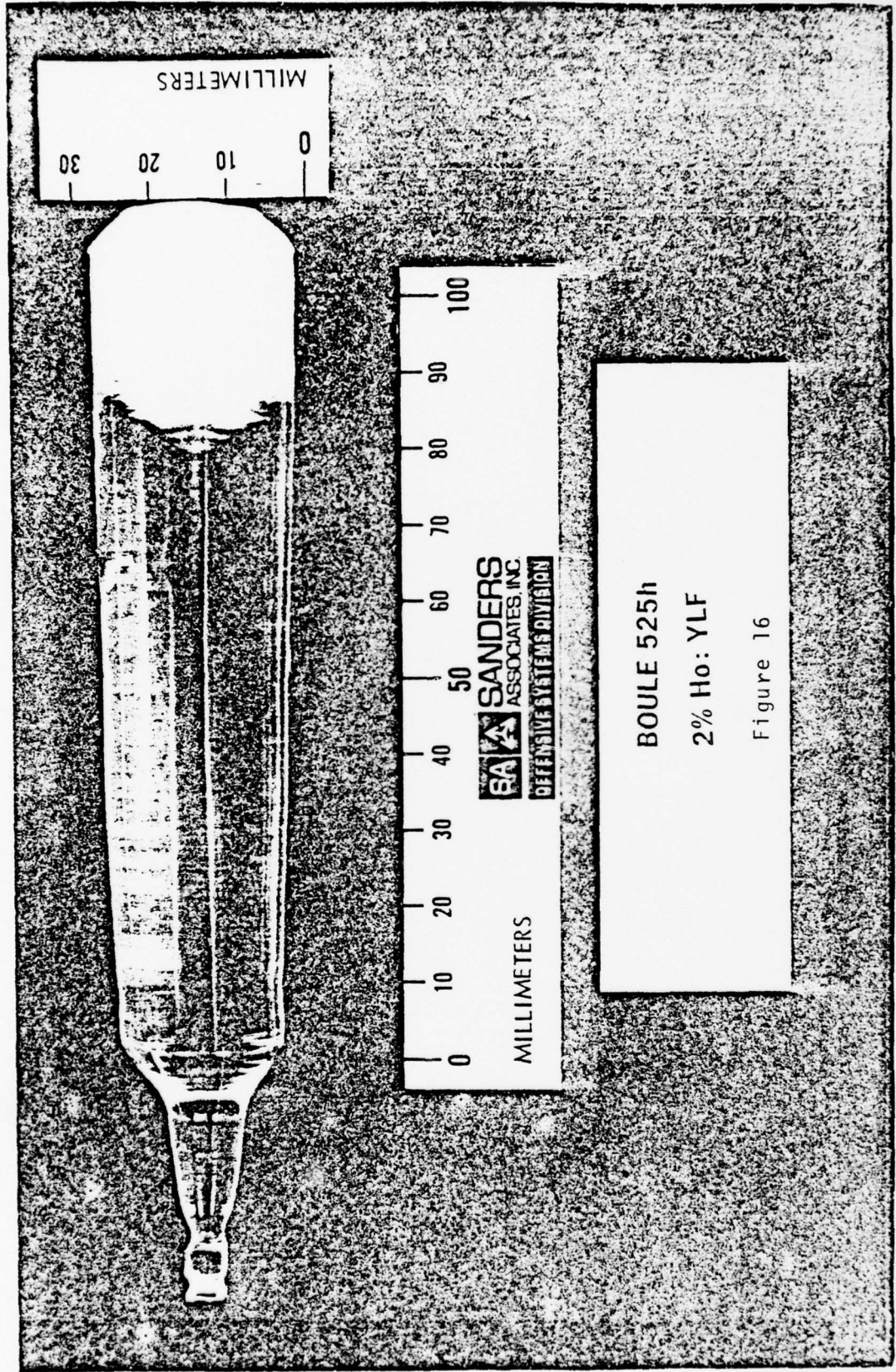
FIGURE 15

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Laser measurements at room temperature were carried out with the same crystal but in a different (more efficient) pump cavity. In these tests only .85 $\mu$ m oscillations were observed - no .75 $\mu$ m oscillations were detected at up to 3 x the .85 $\mu$ m threshold. The poorer .75 $\mu$ m performance may have been due to experimental conditions. The flashlamp pulse width in all these experiments was considerably longer than the  $^5S_2$  lifetime which at room temperature is about 1/2 the Er ( $^4S_{3/2}$ ) lifetime in this composition.

In order to more fully characterize this system it was determined that more laser crystals were required of different compositions. It was decided to grow three boules of composition 2%Ho, 4%Ho - 2%Er, 10%Ho - 2%Er. Figure 16 shows the 2%Ho:YLF boule grown on this program. Growth difficulties prevented growth of the other compositions before the completion of this program.

2% Ho: YLF



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APPENDIX I

DEEP RED LASER EMISSION IN Ho:YLF

APPENDIX II

STIMULATED EMISSION CROSS SECTION  
of the  
750nm LASER TRANSITION IN Ho<sup>3+</sup>:YLF

APPENDIX III

TWO COLOR LASER OPERATION IN Er-Ho:YLF

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E. P. CHICKLIS, C. S. NAIMAN, L. ESTEROWITZ, AND R. ALLEN

**Abstract**—Laser operation of the  $^3S_2 - ^3F_4$  transition in 2 percent Ho:YLF at room temperature is reported. Oscillations at  $\lambda = 750$  nm were obtained in flashlamp and dye laser pumped experiments. Thresholds of 4 J/cm and  $3 \times 10^{-3}$  J/cm were observed in flashlamp and laser pumped operation, respectively. The 750-nm transition in the Ho:YLF is a four-level laser with a simulated emission cross section of  $\sigma = 9.7 \times 10^{-19}$  cm<sup>2</sup>.

LASER emission from trivalent Ho is normally associated with the  $^3F_4 - ^3I_6$  transition in the 2- $\mu$ m region. This transition has operated in a variety of host materials with the most efficient room temperature operation reported in Er-Tm sensitized Ho:YLF [1], [2]. Morozov *et al.* [3] and Podkozinia *et al.* [4] report the generation of many other laser transitions from higher excited levels in LiHoF<sub>4</sub> and 2 percent Ho:YLF at 90 K including  $^3S_2 - ^3F_4$  transition. We report laser operation of the  $^3S_2 - ^3F_4$  transition at room temperature in 2 percent Ho:YLF at  $\lambda = 750$  nm. Laser operation was obtained with a Xe flashlamp (threshold 4 J/cm) and with a 0.48- $\mu$ m dye laser pump (threshold  $0.3 \times 10^{-3}$  J/cm).

The energy levels of Ho:YLF are reported in [5]. The measured fluorescence lifetimes of the upper and lower 750-nm laser levels in 2 percent Ho:YLF at room temperature are

$$^3S_2: 90 \pm 10 \mu\text{s}$$

$$^3F_4: 15 \pm 1 \text{ ms.}$$

We see that the lifetime of the terminal manifold is much longer than that of the upper manifold. A similar situation exists for the 850-nm laser transition ( $^4S_{3/2} - ^4I_{13/2}$ ) in Er:YLF [6]. Performance limitations imposed by the long lifetime of the lower manifold will be discussed below.

Fig. 1 shows an unpolarized absorption spectrum at room temperature of 2 percent Ho:YLF crystal beginning with the upper laser level. In low concentrations most of the pump bands in the 0.2-0.55- $\mu$ m region are optically thin even through a 0.9-cm path. Higher absorption coefficients at higher Ho concentrations (material quality and ease of growth are essentially independent of Ho<sup>3+</sup> concentration) would improve the pumping efficiency but probably reduce the fluorescence lifetime of the upper laser level. For example, the measured room temperature lifetimes of the  $^3S_2$  fluorescence are 105  $\mu$ s at 0.5 percent and 90  $\mu$ s at 2 percent with an accuracy of  $\pm 10$  percent.

Fig. 2 shows the  $^3S_2 - ^3F_4$  emission in  $\pi$  and  $\sigma$  polarization. The laser line corresponds to the sharp  $\pi$  transition (linewidth 12 cm<sup>-1</sup>) centered at 750.3 nm. Measurement [7] of the simulated emission cross section is complicated by the many overlapping levels of the ground manifold. The room temperature cross section for the  $\pi$  polarized laser line is  $9.7 \times 10^{-19}$  cm<sup>2</sup> compared to  $4.6 \times 10^{-19}$  cm<sup>2</sup> in Nd:YAG. The high cross section is mitigated somewhat by the low occupation factor of the upper laser level, approximately 10 percent. Gain is proportional to the product of the cross section and occupation factor.

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L. Esterowitz and R. Allen are with the Naval Research Laboratory, Washington, DC 20375.

Deep Red Laser Emission in Ho:YLF

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Flashpumped laser measurements were made with a 3 X 22-mm rod mounted in a close coupled polished Al pump cavity. The rod was pumped with a 3 X 20-mm, 3000-torr Xe lamp with a clear fused quartz envelope. Flashlamp pulse width was 40  $\mu$ s. With a 9 percent transmitting coupling mirror in a plane parallel resonator the pulsed threshold was 12 J. The laser output was detected with a spike-filtered Si photodiode; above threshold relaxation oscillations began within about 10  $\mu$ s of the flashlamp initiation and persisted for the duration of the lamp pulse. The wavelength of the oscillation was measured directly with a 0.5-m grating monochromator and found to be 750.5  $\pm$  0.5 nm. The output was also visually detected off of a diffuse surface in a well-lighted room and exhibited a deep red hue.

Under these pumping conditions the lamp current density was approximately 3000 A/cm<sup>2</sup>, which resulted in lamp emission predominantly in the blue-ultraviolet region, with particularly intense lines between 0.22 and 0.26  $\mu$ m. High lamp current densities are in general required to efficiently pump this material for a given lamp pulse energy since all the pump bands lie above 0.55  $\mu$ m and the fluorescence lifetime is short. YLF is particularly suitable as the host for this transition since it is transparent to 150 nm and thus all the excited levels are accessible. Furthermore, the ultraviolet damage resistance of YLF grown with purified feed in an inert atmosphere [8] is extremely high.

Laser operation using the same rod was also obtained with a flash-pumped dye laser (modified Synergetics Model 1050), tuned by a 1200 groove/mm grating. The dye laser output was set to pump the multiplet centered at 480 nm. The rod was pumped transversely by expanding the beam to 20 mm and then focusing with a cylindrical lens along the rod length. The pump beam cross section at the rod was 0.5 X 20 mm. The resonator consisted of a 1-m radius high-reflectivity mirror and a flat output mirror with 1 percent transmission.

With a dye laser output pulsewidth of 220 ns, 750-nm oscillations were detected with a dye laser output of only 0.5 X 10<sup>-3</sup> J. The delay between the exciting pulse and the 750-nm laser output was less than 100 ns, indicating very fast relaxation from the 4800-Å multiplet to <sup>3</sup>S<sub>2</sub>. Above threshold multiple spiking was observed but its duration did not exceed 1  $\mu$ s. Energy conversion efficiencies (dye laser input/750-nm output) greater than 50 percent were measured without optimization of the active ion concentration or resonator conditions. The maximum possible conversion efficiency, limited by the photon decrement, is 64 percent for a 480-nm pump.

In spite of the long lifetime of the <sup>3</sup>I<sub>1</sub> manifold it can be shown that good energy extraction can be obtained under certain conditions in flash-pumped Q-switched operation. In the following calculations it is assumed that no appreciable amount of excitation in the upper laser level has decayed by spontaneous emission before the laser is Q switched. Also it is assumed that thermal equilibrium is maintained among the levels of the terminal manifold even during the laser pulse, which is reasonable given the rapid manifold relaxation times in solid hosts.

For single-shot operation, since the occupation factor of the lower laser level in the terminal laser manifold is ~10 percent, operation well above threshold lasing continues until the upper laser level population is reduced to ~10 percent of its initial value. Thus, up to ~90 percent of the initial inversion can be extracted.

In repetitively pulsed operation the terminal manifold population decays between pulses by a factor of

$$e = \exp(-T/\tau)$$

where T is the interpulse period and  $\tau$  is the <sup>3</sup>I<sub>1</sub> lifetime.

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If we denote the population density of the terminal manifold due to the induced transitions for each pulse by  $p$ , it is easy to show that the asymptotic population density of the terminal manifold is given by

$$\bar{N} = \frac{ep}{1-e}$$

For an output of 0.2 J from a 6.2 x 76-mm rod, operating at 20 Hz,

$$p = 3.2 \times 10^{17} \text{ cm}^{-3}$$

and

$$\bar{N} = 6.4 \times 10^{13} \text{ cm}^{-3}$$

Since the occupation factor of the terminal laser level is ~10 percent the equilibrium population  $\bar{N}_l$  of that level is only

$$\bar{N}_l = 6 \times 10^{14} \text{ cm}^{-3}$$

To see the significance of this population density we calculate the threshold population density of the upper level  $N_u$ :

$$N_u = \frac{-\ln R}{2L\sigma} \frac{s}{a}$$

where

$R$  mirror reflectivity, 0.7

$s$  scattering loss, 0.005  $\text{cm}^{-1}$

$\sigma$  stimulated emission coefficient,  $\sim 10^{-18} \text{ cm}^2$

$L$  rod length, 7.6 cm.

We have

$$N_u = 3 \times 10^{16} \text{ cm}^{-3}$$

Clearly the equilibrium population of the terminal manifold is negligible for a 2.4-cm<sup>3</sup> 4-W laser at 20 Hz.

At sufficiently high repetition rates or power loadings the terminal level population will become significant, and limit power output. To overcome this, several ions might be added as codopants in the crystal to effectively deplete the <sup>5</sup>I<sub>1</sub> terminal manifold (Tb<sup>3+</sup>, Nd<sup>3+</sup>, Dy<sup>3+</sup>, Pr<sup>3+</sup>). However, the depleting ion must be chosen with care so that upper level quenching or absorption at the laser line is avoided.

The <sup>5</sup>S<sub>2</sub> - <sup>5</sup>I<sub>1</sub>, Ho:YLF laser may provide a practical high-pak power pulsed source in the gap between ruby (694 nm) and Er:YLF (850 nm). It operates as a four-level laser with a high specific gain and could operate at high efficiencies at room temperature in flash-pumped operation. Optimum operation may be achieved with dye laser flashlamps pumping high Ho concentration crystals. A 750-nm laser is of practical interest because of the availability of highly sensitive optical imaging receivers and photographic film in this region. Further, such a laser may be extremely useful for pumping infrared organic dye lasers for a variety of applications requiring tunable output in the 0.75-1- $\mu\text{m}$  region at moderate pulse repetition rates.

#### ACKNOWLEDGMENT

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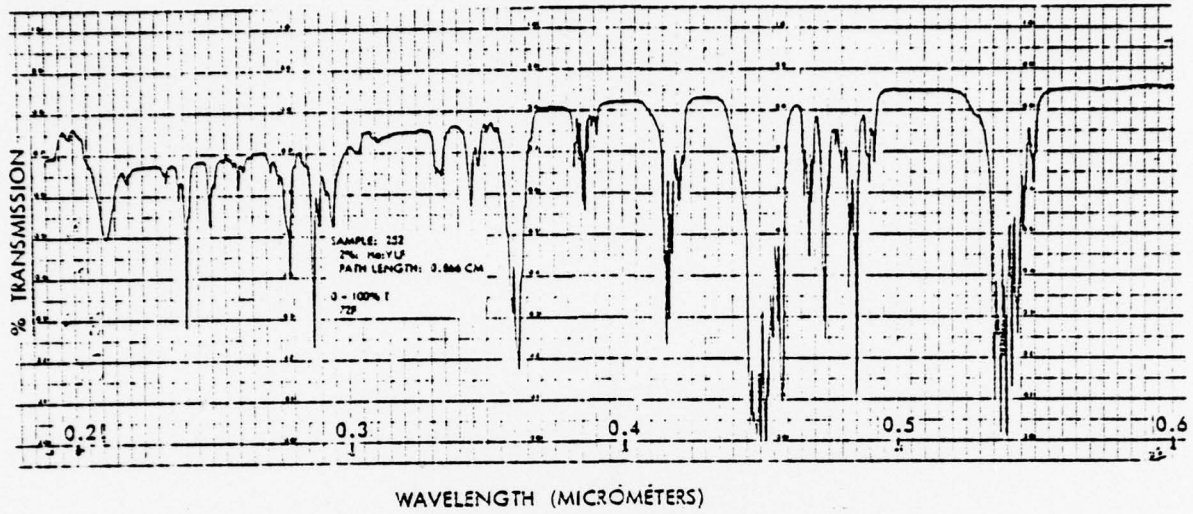
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Fig. 1. Room temperature, unpolarized transmission spectrum of 2 percent Ho:YLF above 600 nm.

Fig. 2. Normalized fluorescence spectrum of the  $^5S_2 - ^5I_7$  transition in  $\pi$  and  $\sigma$  polarization at room temperature. Laser transition corresponds to the peak centered at 750.3 nm.

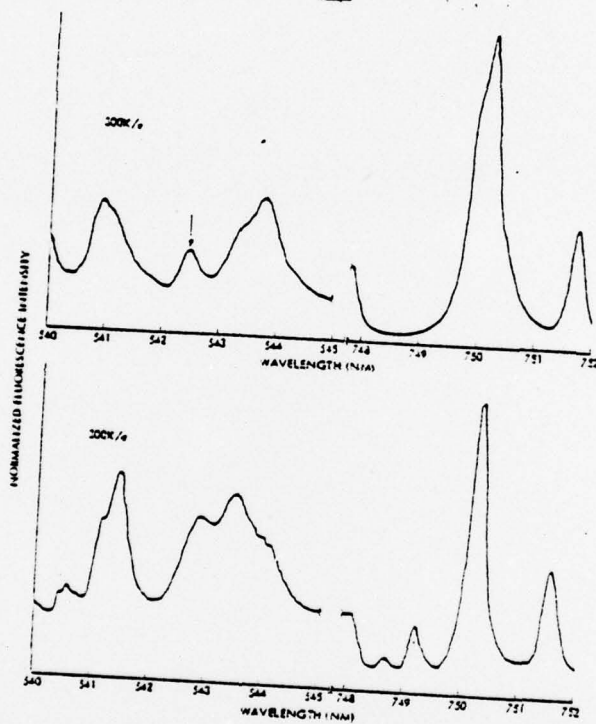
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101 OF ISSUE ADV. KR FIG 1

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JRL CF ISSUE NOV KB FIG 2  
44 % OF ORIG \_\_\_\_\_ DENSITOMETER

STIMULATED EMISSION CROSS SECTION  
of the  
750nm LASER TRANSITION IN  $\text{Ho}^{3+}:\text{YLF}^*$

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\* Work supported under AFOSR contract F44620-76-C-0111

+ Consultant to Sanders Associates, Inc. 95 Canal Street, Nashua,  
N.H. 03061

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INTRODUCTION

The 750nm laser transition in Ho:YLF is only one of several observed laser transitions in this material.<sup>(1-4)</sup> It is however, the shortest wavelength transition and is of interest as it is a four level transition which may provide an alternative to ruby laser systems with the capability of higher repetition rates.<sup>(4)</sup>

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The stimulated emission cross section is one of the parameters that is needed in order to both evaluate the potential laser material and to design practical laser systems. The gain of the laser transition and the amount of energy that can be stored in the material for Q-switching depends directly on the cross section. Both of these properties involve the product of the upper level occupation  $\beta$ , and the cross section  $\sigma$ . When the upper level is one of several levels of a multiplet, the total population of the multiplet determines the stored energy. Only the population in the upper laser level determines the instantaneous gain. Thus both  $\sigma$  and the product  $\beta\sigma$  are parameters.

The stimulated emission cross section for the Ho:YLF 750nm laser transition as well as an accurate energy level determination of the relevant multiplets are reported here. The peak stimulated emission cross section was found to be  $\sigma = 9.7 \times 10^{-19} \text{ cm}^2$  for the linearly polarized laser transition at 750.3 nm, and the occupation factor  $\beta = 0.1$  at room temperature.

#### SPECTROSCOPY

The 750nm laser transition Ho:YLF is between levels of the  $^5S_2 \rightarrow ^5I_7$  multiplets. In order to determine the stimulated emission cross section it is necessary to know what particular levels of the two multiplets are involved in the transition. The positions of the crystal field split levels of  $\text{Ho}^{3+}$  in YLF have previously been reported,<sup>(5)</sup> however, the accuracy was not high enough to unambiguously identify the laser transition. Because of the large number of levels in all the multiplets involved

and their close spacing, most transitions observed could be between any of several pairs of levels when an uncertainty of a few  $\text{cm}^{-1}$  has to be included.

Figure 1 shows the energy levels of  $\text{Ho}^{3+}$  in YLF. Only the  $^5\text{I}_8$ ,  $^5\text{I}_7$  and  $^5\text{S}_2$ ,  $^5\text{F}_4$  multiplets are included since these are the ones used to determine the stimulated emission cross section for the laser line. In YLF the site symmetry of the rare earth site is  $S_4$  and the level designations in the figure are irreducible representations in this symmetry group,  $\Gamma_1$ , and  $\Gamma_2$  are singly degenerate levels whereas  $\Gamma_{3,4}$  is a Cramer's degenerate pair. The selection rules for electric dipole transitions in  $S_4$  symmetry help in identifying the energy levels, however a great many transitions that are allowed are not observed in the case of  $\text{Ho}^{3+}$ . This is also the case for other rare earths in YLF indicating that the  $S_4$  symmetry here is only a slight perturbation of  $D_2$  symmetry.<sup>(6)</sup> The use of only absorption and fluorescence spectra was therefore insufficient information to determine the position of most of the energy levels. The energy level scheme determined theoretically in reference <sup>(5)</sup> was based on a few transitions between levels of designation that had been determined with high certainty from low temperature spectroscopy. The resulting relative positions of other levels were then confirmed spectroscopically.

In this work the accurate positions of the levels were determined based on the theoretical relative positioning of the levels and careful absorption and emission spectroscopy at 4.2K and intermediate temperatures up to 77K. The wavelength calibration involved use of mercury and neon spectral arc lamps. The frequency of the transitions was obtained from the air wavelengths by use of tables.<sup>(7)</sup> The resulting

energy level positions in Figure 1 should be accurate to within  $\pm 1\text{cm}^{-1}$  except for the upper levels in each multiplet that can't be accurately determined due to overlapping vibronic transitions.

### STIMULATED EMISSION CROSS SECTIONS

The method used for measuring the stimulated emission cross sections is the one described by Kushida<sup>( )</sup> and later by Singh et al.<sup>( )</sup> The method involves measuring the peak absorption coefficients between the levels of the ground state manifold and the levels of the excited state manifold of  $^5S_2$  (Ho). The absorption cross section is then determined from the relation

$$\sigma_{ij} = \alpha_{ij}/N_i$$

where  $\sigma_{ij}$  is the peak absorption cross section for the line from level  $i$  in the groundstate manifold to level  $j$  in the upper manifold,  $\alpha_{ij}$  is the peak absorption coefficient for the same line, and  $N_i$  is the population density in level  $i$  given by the active ion concentration and Boltzmann statistics.

The stimulated emission cross section for the  $j \rightarrow i$  transition is given by

$$\sigma_{ji} = \frac{g_j}{g_i} \sigma_{ij}$$

where  $g_i$  is the degeneracy of level  $i$ . The stimulated emission cross section for a transition from level  $j$  to a level  $k$  that has no thermal population and therefore cannot be observed in absorption, is given by

$$\frac{\sigma_{jk}}{\sigma_{ji}} = \left(\frac{n_{ji}}{n_{ij}}\right)^2 \left(\frac{\lambda_{jk}}{\lambda_{ji}}\right)^5 \left[ \left(\frac{dI}{d\lambda}\right)_{jk} \left(\frac{dI}{d\lambda}\right)_{ji} \right]$$

where  $n$  is the refractive index,  $\lambda$  is the vacuum wavelength, and  $\frac{dI}{d\lambda}$  is the peak emission power per unit wavelength interval. Thus if  $\sigma_{ji}$  is known from absorption,  $\sigma_{jk}$  can be found by measuring the peak fluorescence intensities for the two lines.

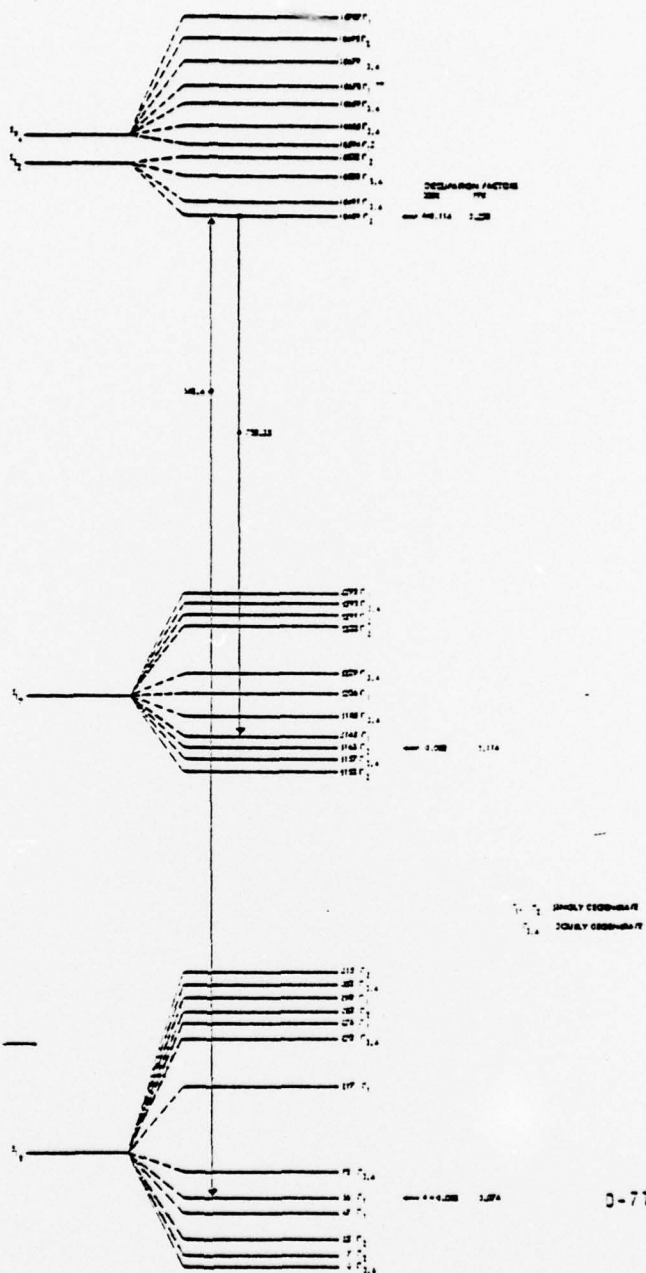
The 750.3nm laser transition is indicated in Figure 1 as being from the bottom of the  $^5S_2$  level ( $E_1$ ) to the fourth level of  $^5I_7$  ( $B_4$ ). Since the transition is a  $\Gamma_2 \rightarrow \Gamma_1$  transition the selection rules demand linearly polarization with the E-vector parallel to the C-axis of the YLF crystal ( $\pi$ -polarized). It is not necessary that the transition used to measure the absorption cross for the upper laser level has the same polarization as the laser transition. However, since the ratio of the fluorescence intensities of the two lines has to be determined, accurate measurements are easier obtained when only one polarization is involved. For this reason the fourth level of  $^5I_8$  ( $A_4$ ) which is a  $\Gamma_1$  level was chosen. The  $A_4 \rightarrow E_1$  transition is also relatively free of overlap from other transitions. Figure 2 shows the  $\pi$ -polarized absorption spectra Ho  $^5I_8 \rightarrow ^5S_2$ ,  $^5F_4$  at 80K. The 542.4nm  $A_4 \rightarrow E_1$  transition is well separated from any other lines. The peak absorption cross section calculated from the absorption coefficient, the Ho concentration and the occupation factor of the  $A_4$  level is  $9.6 \times 10^{-20} \text{ cm}^2$ . Figure 3 shows the partial 77K  $\pi$ -polarized  $^5S_2 \rightarrow ^5I_8$  and  $^5S_2 \rightarrow ^5I_7$  emission spectra. From the intensity ratio of the 542.4nm line and the 750.3nm line, and the absorption cross

section for the laser line is  $1.6 \times 10^{-18} \text{ cm}^2$ .

At room temperature neither the absorption line nor the emission lines are free from overlap. The main contribution to the lines seems to be due to the original transitions however. With this assumption the stimulated emission cross section at 300K is found to be  $9.7 \times 10^{-19} \text{ cm}^2$ . The accuracy of this value is estimated to be  $\pm 30\%$  due to the overlap problem. Room temperature spectra are shown in Figure 3.

It is interesting to compare the gain per unit inversion for Nd:YAG and Ho:YLF. In the case of Nd:YAG, the stimulated emission cross section is  $5 \times 10^{-19} \text{ cm}^2$  and the occupation factor of the upper level is 0.4 giving a gain of  $2 \times 10^{-19} \text{ cm}^{-1}$ . For Ho:YLF the occupation factor is 0.11 and the gain  $1 \times 10^{-19} \text{ cm}^{-1}$ . With nearly twice the cross section the gain per unit inversion in Ho:YLF is only one half that of Nd:YAG. It should also be pointed out that the allowable energy storage depends on the inverse  $\beta\sigma$  product. So despite the higher  $\sigma$  in Ho<sup>3+</sup>:YLF, the Q-switched energy storage at the fluorescence amplification limit should be about twice that of Nd:YAG.

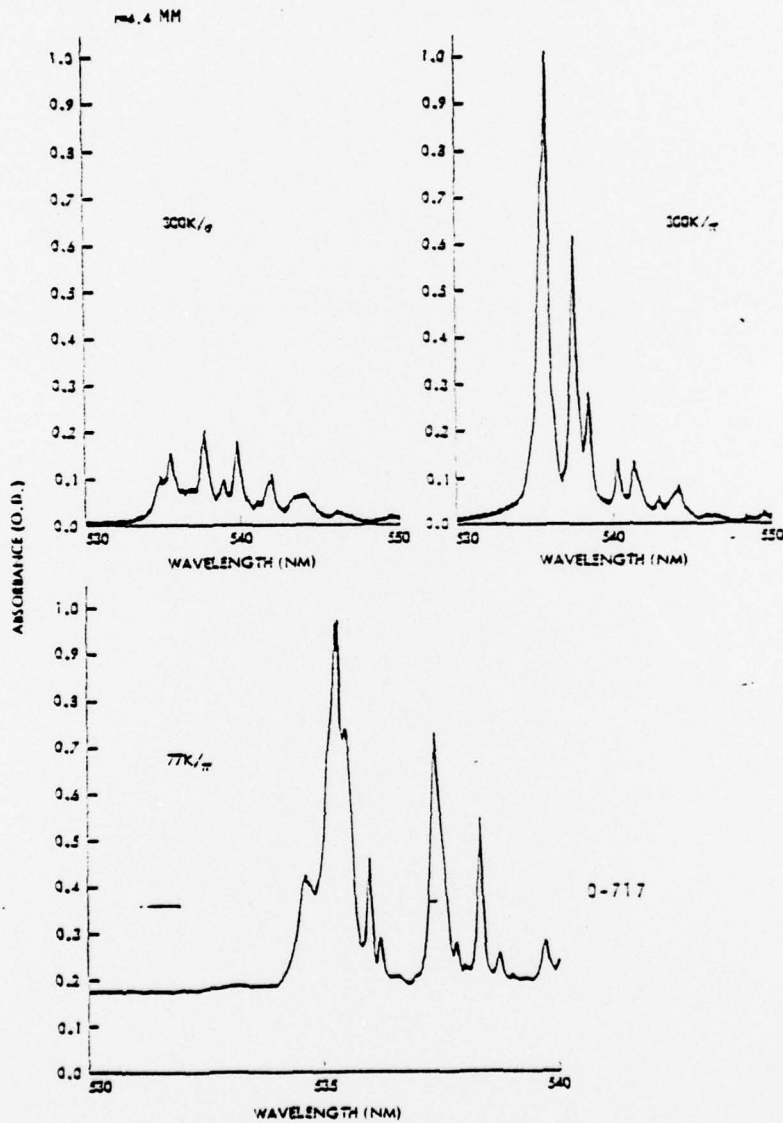
# Ho:YLF ENERGY LEVELS



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POLARIZED ABSORPTION SPECTRUM

0.5% Ho: YLF (266F)

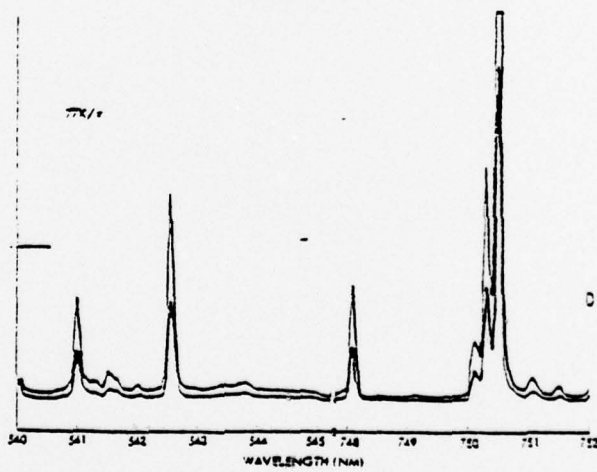
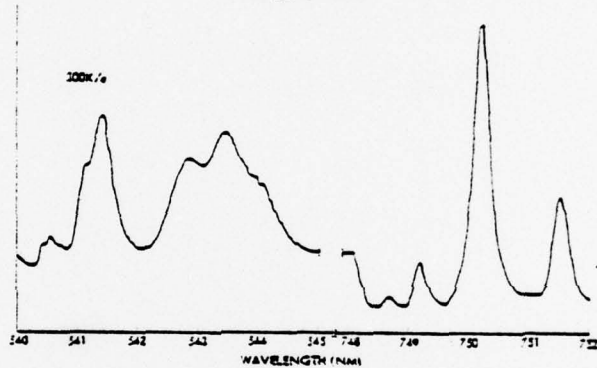
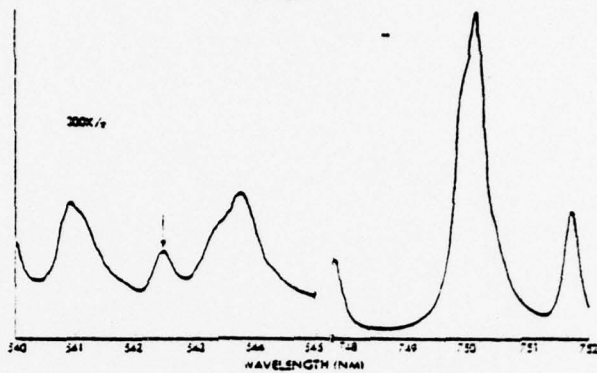


POLARIZED ABSORPTION SPECTRA

Ho:YLF

NORMALIZED FLUORESCENCE SPECTRA

Ho:YLF



POLARIZED EMISSION SPECTRA

Ho:YLF

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TWO COLOR LASER OPERATION IN Er-Ho:YLF

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ABSTRACT

Laser operation from two different, interacting ions in the same host has been observed. Stimulated emission at 750nm (Ho) and 850 (Er) at 77K is reported. The crystal Er-Ho:YLF whose room temperature stimulated emission cross sections are  $9.7 \times 10^{-19} \text{ cm}^2$  and  $1.4 \times 10^{-19} \text{ cm}^2$  respectively. Measurement of reversible energy transfer between the upper laser levels is reported as a function of Er and Ho concentration and temperature.