SPECTROSCOPIC AND 3-MICRON LASING PROPERTIES OF ERBIUM-DOPED YTTRIUM ALUMINUM GARNET AND THE EFFECTS OF HOLMIUM CO-DOPING

by

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Dedication

This dissertation is dedicated to my parents, without whom none of this would have been possible.

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It is very difficult to decide who to leave out of this section. So many people have helped me complete this dissertation, and there isn't room to list everyone. A few of the people who helped me more than any others, though, deserve mention.

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Abstract

The spectroscopy and 3- μ m lasing behavior of Er:YAG are studied. Er:YAG is shown to lase on two distinct lines near 2.94 μ m, specifically 2.9393 and 2.9362 μ m. These two laser lines operate simultaneously. Although both begin in level A2 (the second lowest energy Stark sublevel of the ⁴I_{11/2} level), the 2.9362- μ m line terminates at level Y6 and the 2.9393- μ m line at level Y7 (Y7 and Y6 are the highest and second highest energy Stark sublevels, respectively, of the ⁴I_{13/2} level). These wavelengths imply that the Y6 level, whose energy was previously unknown, has an energy of 6873.8 cm⁻¹.

The effects of adding Ho³⁺ ions to Er:YAG is also reported. Energy is transferred from the ${}^{4}I_{13/2}$ level of Er³⁺ to the ${}^{5}I_{7}$ level of Ho³⁺. As reported in this dissertation, the value of the coefficient describing this energy transfer is W = 9.5±1.0 × 10⁻²⁰ cm³/s. This value suggests that the Er³⁺ \rightarrow Ho³⁺ energy transfer is not strong enough to unblock the Er:YAG 3-µm transition, but is strong enough to unblock the Er:YLF 3-µm transition. The energy transfer also suggests diode-pumping Er³⁺ ions, which then transfer their energy to the Ho³⁺ ions, creating an efficient, diode-pumped 2-µm Ho³⁺ laser.

Multiple wavelength lasing of (30% Er, 1.5% Ho):YAG is described. In addition to the two wavelengths seen in Er:YAG, (Er, Ho):YAG lases at 2.796 and 2.766 μ m during the same pump pulse as the 2.936- and 2.939- μ m lines. Both these transitions are attributed to the Er³⁺ ions. The 3- μ m lasing is shown to have a lower gain than the lasing near 2.8 μ m, but atmospheric losses at 2.8 μ m are sufficient to prevent lasing at these wavelengths. The 2.8- μ m lines are seen in (Er, Ho):YAG after the 3- μ m lines have self-terminated due to excited-state absorption in the Ho³⁺.

Chapter 1—Introduction

1.1 Background

There is considerable interest in 3-µm laser transitions, for industrial,¹ medical,² and military³ applications. The most common 3-µm lasers are HF,⁴ Er:YAG,⁵ and Ho:YAG.⁶ The HF laser is a chemical laser, and will not be considered in this dissertation. This dissertation concentrates on the two common 3-µm solid-state laser transitions, the ⁴I_{11/2}→⁴I_{13/2} in Er:YAG and the ⁵I₆→⁵I₇ in Ho:YAG. Unfortunately, both these transitions have lower level lifetimes longer than the upper level lifetimes,⁷ leading to so-called "blocked transitions." Lasers with blocked transitions are much less efficient than true four-level lasers, as a large population can build up in the lower laser level, requiring very high pump levels to maintain inversion. This population buildup prevents cw lasing unless there is an effect which depopulates the lower level.

In high dopant density Er:YAG, the lower laser level is depopulated by ion-ion energy transfer.⁸ This allows relatively efficient lasing in the pulsed regime,⁴ but not, as yet, true CW operation. In Ho:YAG, the ion-ion interactions are not as strong, leading to lasing which is self-terminated⁵ (*i. e.*, lasing which ends after a particular pulse width even if the pump pulse is lengthened well beyond this time).

The most common method of unblocking laser transitions such as those in Er:YAG and Ho:YAG is co-doping.⁹ This technique involves adding another impurity ion, in addition to the active laser ion, to act as a "deactivator" which relaxes ions from the lower laser level to the ground state. Co-doping as a method of unblocking 3-µm laser transitions has been tried in both Er:YAG¹⁰ and Ho:YAG¹¹ with Nd³⁺ as the deactivator, with limited success. The strong ion-ion interactions among the Nd³⁺ ions and between the Nd³⁺ ions and the active laser ions made modeling difficult.¹² This dissertation is concerned with (Er, Ho):YAG, which has much weaker interactions between the Er³⁺ and

Ho³⁺ ions than either of these ions has with Nd³⁺. The weaker interactions make it possible to model (Er, Ho):YAG with simple rate equations more accurately than (Er, Nd):YAG or (Ho, Nd):YAG, and provides insight into the ion-ion energy transfer processes.

1.2 Organization of the Dissertation

This study of ion-ion energy transfer processes begins with Er:YAG. The most important process in the 3- μ m ⁴I_{11/2} \rightarrow ⁴I_{13/2} laser transition is that in which two ions in the ⁴I_{13/2} level interact. One of these ions transfers its energy to the other, which is then excited into the ⁴I_{9/2} level. This is the Er³⁺ "cross-relaxation" process. Ions in the ⁴I_{9/2} level decay quickly to the ⁴I_{11/2} upper laser level with a branching ratio near unity.¹³ When Ho³⁺ is added to Er:YAG, the Ho^{3+ 5}I₇ level interacts with the Er^{3+ 4}I_{13/2}. This interaction reduces the strength of the cross-relaxation process in Er³⁺ by reducing the population of the ⁴I_{13/2} level.

In the remainder of this chapter, the history of lasers related to those studied, specifically, Er:YAG and Ho:YAG 3- μ m lasers, is discussed. In both Er:YAG and Ho:YAG, dopant ion concentration has some effect on lasing, whether it be an efficiency effect as in Er:YAG⁵ or a wavelength effect as in Ho:YAG.⁶ These effects are reported later in this first chapter. This is an historical chapter, so previously-published co-doping experiments involving Er³⁺, Ho³⁺, and Nd³⁺ ions in YAG are also discussed.

The spectroscopy of Er:YAG and (Er, Ho):YAG formed a major portion of this dissertation.. Absorption and emission vs. wavelength as well as strength and temporal decay characteristics of the dopant energy levels relevant to the 3-µm laser transitions, the various parts of spectroscopy, are described in detail in Chapter 2.

Some of the absorption spectroscopy was unsatisfying. Due to thermal population of many Stark sublevels of the $\text{Er}^{3+4}I_{15/2}$ ground state at room temperature, it was impossible to identify certain energy levels; the absorption features from non-zero sublevels of the ${}^{4}I_{15/2}$ level into upper sublevels of the ${}^{4}I_{13/2}$ and ${}^{4}I_{11/2}$ overlaid the absorption features from low sublevels of the ground state into low sublevels of these two excited states. (In this dissertation, all spectroscopy was done at room temperature, as were the lasing experiments.) Specifically, the Y6 sublevel of the ${}^{4}I_{13/2}$ level (the ${}^{4}I_{13/2}$ is also called the Y level) could not be separated. This was not only true of these experiments, but also of previous work.¹⁴ However, the Y6 sublevel was separated and verified relative to the energy placement of other sublevels by careful measurement of the laser wavelengths. This care in wavelength measurement led to the discovery of simultaneous lasing at 2.936 and 2.939 µm in YAG doped with Er^{3+} .

The lasing properties of Er:YAG and (Er, Ho):YAG were tested in two different systems, one manufactured specifically for the testing of Er:YAG by Quantronix and the other designed for alexandrite by Allied Corporation. The lasing behavior of Er:YAG at extremely long pump pulses lengths was studied by placing an Er:YAG rod in a commercial Nd:YAG laser machining system. All the lasing experiments are described in detail in Chapter 3.

Chapter 4 discusses the experiments described in Chapters 2 and 3. In this chapter, the behavior of singly-doped materials is compared to that of co-doped materials. Chapter 4 also draws conclusions from this information. The energies of all the Stark sublevels of the laser levels and ground states of both Er^{3+} and Ho^{3+} in YAG are specified, and the interactions between the various ions are described. Chapter 5 explains the main result of the experiments, the determination of the coefficient of energy transfer from the $\text{Er}^{3+4}I_{13/2}$ to the $\text{Ho}^{3+5}I_7$ level. Finally, in Chapter 6, recommendations for potentially useful future work are explored.

3

1.3 Solid-State Lasers

The first laser, ruby, was a solid-state laser. As is typical of solid-state lasers, the ruby laser is pumped optically.¹⁵ In an optically-pumped solid-state laser, light from an outside source (usually a flashlamp or laser diode) is absorbed by the active laser ion. This excites the ion into a high-energy state, which rapidly decays to a metastable excited state. This metastable state is the upper laser level. The lower laser level may be the ground state or another excited state. When the density of ions (the "population") in the upper laser level is greater than that in the lower laser level, the laser medium is said to be inverted. In an inverted medium, light at the laser wavelength will be amplified as it passes through the medium. The energy for this amplification comes from the decay of ions which were excited into the upper laser level, into the lower laser level. This emitted light has a characteristic wavelength determined by the energy difference between the upper and lower laser levels, and is the laser wavelength. The random noise present in all materials typically provides enough light to begin the laser action. Some of the amplified light is fed back into the inverted medium by the use of mirrors. In all the laser experiments described herein, two mirrors were used: one whose reflectivity was near 100% (the high reflector or HR), and another which was partially transmitting (the output coupler or OC).

Some light leaves the laser through the OC. Some is also lost through absorption and scattering in the laser medium. If the inverted medium's two-pass amplification is greater than these losses, the medium acts as a laser; the recirculating energy contained by the mirrors increases rapidly until the amplification no longer exceeds the loss. In the laser systems used in these experiments, the excitation (the "pump") was a pulse of light, and the laser output was also a pulse. If a steady-state pump can cause sufficient amplification to overcome the steady-state losses, continuous-wave, or CW, laser output occurs. In most of this dissertation, CW lasers will not be discussed, only pulsed lasers.

One possible laser pumping system¹⁶ is shown in Fig. 1.1. This "three-level" system is typical of ruby. It is called "three-level" because three energy levels of the active ion are involved. The active ion absorbs light in the wavelength band defined by the energy difference from the ground state, level 1, to the "pump band," level 3, which is often fairly extensive. The ion excitation and decay rates appear in Fig. 1.1. The equations describing the excitation of the ions are:

$$\frac{dn_3}{dt} = W_{13}n_1 - \frac{n_3}{\tau_{32}} \tag{1.1}$$

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} - \rho B_{21}(n_2 - n_1)$$
(1.2)

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - W_{13}n_1.$$
(1.3)



Figure 1.1. Energy level diagram and rate equation terms for a three-level laser.

In a typical three-level laser, τ_{32} is much shorter than the pump pulse and τ_{21} is much longer than τ_{32} . All the ions excited into the pump band (at the rate $W_{13}n_1$, governed by the population in the ground state and the power density of the absorbed pump light) decay rapidly into the upper laser level, level 2. Since τ_{21} is long, the ions tend to remain in level 2; it is a metastable state. To give up their energy, the ions may either decay spontaneously or contribute to the lasing process. The spontaneous decay is governed by the lifetime of level 2, τ_{21} ; the lasing process is described by $\rho B_{21}(n_2 - n_1)$, and is dependent on the inversion $(n_2 - n_1)$, the energy density of light at the laser wavelength (ρ), and the Einstein B-coefficient (the coefficient of stimulated emission) for the level 2 to level 1 transition.

In three-level lasers, inversion is reached when at least half of the ions in the ground state (level 1) have been excited through the pump band into the upper laser level. "Four-level" lasers, so called because they use four levels of the active ion, have less stringent requirements. A typical energy level diagram for a four-level laser is shown in Fig. 1.2. An important example of a four-level laser is Nd:YAG. Such a laser absorbs pump light in the wavelength region described by the energy difference between the ground state (now level 0, not level 1) and the pump band. (The pump band usually consists of several levels, but this discussion will refer to the entire band as "level 3.") The ions excited into level 3 rapidly decay into level 2, the upper laser level. These may then decay spontaneously into the lower laser level (level 1), or may lase into level 1. Ions in this excited state then decay into the ground state. The rate equations describing the four-level laser are:

$$\frac{dn_3}{dt} = W_{03}n_0 - \frac{n_3}{\tau_{32}}$$
(1.4)

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} - \rho B_{21}(n_2 - n_1) \tag{1.5}$$

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - \frac{n_1}{\tau_{10}}$$
(1.6)

$$\frac{dn_0}{dt} \quad \frac{n_1}{\tau_{10}} - W_{03}n_0. \tag{1.7}$$



Figure 1.2. Energy levels and rate equation terms for a four-level laser.

In a good four-level laser, τ_{32} will be very short so that all the ions pumped into level 3 (at rate $W_{03}n_0$) will decay rapidly into the upper laser level. This level has a long spontaneous decay lifetime, τ_{21} , so it is a metastable state. The lower laser level lifetime, τ_{10} , is short, so level 1 is virtually free of population. Thus the pump rate $W_{03}n_0$ is nearly

the rate of increase of the inversion. Except for the reduced pumping requirement to reach inversion, caused by the emptiness of the lower laser level, the lasing process of a four-level laser is the same as that of a three-level laser.

In some cases, such as Ho:YAG,⁶ a four-level laser system has a long lower laser level lifetime τ_{10} , longer than τ_{21} . This is the laser with a self-terminated (or "blocked") transition. In such a laser, the lasing ends after a certain amount of time, even if the pump pulse is lengthened. Self-terminated lasers often demonstrate "redshift," a lasing phenomenon in which several output pulses occur during a single pump pulse, each output at a wavelength slightly longer than the previous pulse. This is due to filling of the lowlying Stark sublevels in the lower laser level (see Fig. 1.3). When a low-lying sublevel develops significant population, the inversion relative to this sublevel ends. However, if there is still an inversion relative to a higher-lying sublevel (which has a smaller energy difference from the upper laser level), lasing may occur to this sublevel. Since the energy difference is less, the emission wavelength is longer.



Figure 1.3. Mechanism of redshift in self-terminated lasers.

If a laser has a lower level lifetime longer than the upper level lifetime, it does not need to be self-terminated. One method of unblocking the laser transition is to use nonlinear energy transitions within the active ion (Fig. 1.4). This is the method of lasing seen in high dopant density Er:YAG.⁸ In such a laser, some ion-ion energy transfer depopulates the lower laser level and/or populates the upper laser level. For a cross-relaxation based laser, such as Er:YAG, the rate equations are:

$$\frac{\mathrm{d}n_3}{\mathrm{d}t} = W_{03}n_0 + \frac{1}{2}W_{\mathrm{cr}}n_1^2 - \frac{n_3}{\mathfrak{r}_{32}} \tag{1.8}$$

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} - \rho B_{21}(n_2 - n_1) \tag{1.9}$$

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - W_{\mathrm{cr}} n_1^2 - \frac{n_1}{\tau_{10}}$$
(1.10)

$$\frac{\mathrm{d}n_0}{\mathrm{d}t} = \frac{n_1}{\tau_{10}} + \frac{1}{2} W_{\mathrm{cr}} n_1^2 - W_{03} n_0 \,. \tag{1.11}$$



Figure 1.4. Energy levels and rate equation terms for a cross-relaxation pumped laser.

The cross-relaxation process requires two ions in the lower laser level, level 1. Both ions are removed from level 1 (at a total rate of $W_{cr} n_1^2$); one ion is excited further, into the pump band, while the other is de-excited into the ground state. If this process is sufficiently efficient, it may permit steady-state lasing on a blocked transition. This is the case in Er:YAG.

One other method of unblocking laser transitions consists co-doping laser systems with sensitizer and/or deactivator ions.⁶ A typical laser employing both might have an energy level structure like that in Fig. 1.5. Such a system requires at least two ions; one is the active ion and the other, the deactivator/sensitizer. For ease of discussion a three-ion system is discussed: one ion is the active ion (subscript A), one is the deactivator (subscript D), and the third is the sensitizer ion (subscript S). The rate equations for this system, diagrammed in Fig. 1.5, are:

$$\frac{dn_{S3}}{dt} = W_{03}n_{S0} - \frac{n_{S3}}{\tau_{32}}$$
(1.12)

$$\frac{dn_{S2}}{dt} \approx \frac{n_{S3}}{\tau_{32}} - W_{SA} n_{S2} n_{A0}$$
(1.13)

$$\frac{dn_{S0}}{dt} \approx W_{SA} n_{S2} n_{A0} - W_{03} n_{S0}$$
(1.14)

$$\frac{dn_{A2}}{dt} \quad W_{SA}n_{S2}n_{A0} - \frac{n_{A2}}{\tau_{21}} - \rho B_{21}(n_{A2} - n_{A1}) \tag{1.15}$$

$$\frac{dn_{A1}}{dt} = \frac{n_{A2}}{\tau_{21}} + \rho B_{21}(n_{A2} - n_{A1}) - W_{DA}n_{D0}n_{A1}$$
(1.16)

$$\frac{dn_{A0}}{dt} = W_{DA}n_{D0}n_{A1} - W_{SA}n_{S2}n_{A0}$$
(1.17)

$$\frac{dn_{D1}}{dt} \quad W_{DA} n_{D0} n_{A1} - \frac{n_{D1}}{\tau_{10}}$$
(1.18)

$$\frac{dn_{D0}}{dt} = \frac{n_{D1}}{\tau_{10}} - W_{DA} n_{D0} n_{A1}$$
(1.19)

In this type of laser, pump light is absorbed (at rate $W_{03}n_{s0}$) in the wavelength region defined by the energy difference between the ground state and pump band of the sensi-

tizer, levels S0 and S3. The ions excited into the pump band decay rapidly into the excited state of the sensitizer, level S2. Ions in this state interact with active ions in the ground state. One excited sensitizer ion gives up its excitation energy, decaying to the ground state, S0, and transferring the excitation energy to the active ion. The active ion is then excited into the upper laser level, level A2. From that level, it may decay spontaneously into the lower laser level (A1), or it may lase into this level. Ions in the lower laser level then give up their energy by transferring it to the excited state of the deactivator ion, D1, while decaying to their ions' ground state, A0. The excited deactivator ions then decay rapidly to their ground state, D0.



Figure 1.5. Energy levels and rate equation terms for a laser incorporating sensitizer and deactivator ions.

The net results of sensitizing are (1) to increase the effective absorption of the laser system, as with the (Cr, Nd):YAG system,¹⁷ and (2) to increase the effective upper

laser level lifetime, as with alexandrite laser systems.¹⁸ The net effect of deactivation is to reduce the lower laser level lifetime.¹⁹ Thus, in the case of a blocked transition, where the upper laser level has a longer lifetime than the lower laser level, both sensitization and deactivation may be useful.

1.4 Er: YAG Laser Experiments

The Er:YAG 3- μ m laser has been known since 1974.²⁰ In early papers^{20,21} this transition was reported to be blocked or "self-saturated," although this difficulty is overcome in high dopant density Er:YAG by cross-relaxation.⁸ Er:YAG lasers were also reported¹³ to have many different wavelengths, and a redshift. Ref. 13 reports on lasing in high dopant density Er:YAG at 2.936, 2.830, and 2.795–2.800 μ m. Ref. 21 reports lasing at 2.8302 μ m for 33% Er:YAG, with lasing wavelengths for all Er:YAG crystals ranging from 2.9363 μ m (for 2.5% doping) to 2.9370 μ m (for 100% doping). In contrast, this work describes neither redshifting nor wavelength variation with dopant concentration. The concentrations ranged from 17% to 50%.

The Er:YAG 3- μ m laser is fairly well researched.⁵ After the laser's report in 1974, active Q-switching was reported²² in 1980 and mode-locking²³ in 1982. In both cases, however, the active element used as the loss modulator was LiNbO₃, which is typically fabricated in a way which does not eliminate OH⁻ radicals. These radicals absorb strongly in the 2.5–3.2 μ m region, which greatly reduces the efficiency of 3- μ m lasers with intracavity LiNbO₃ elements. Passive Q-switches were also tested.²⁴ The passive element can be any material with an OH⁻ radical, such as water or ethanol. A unique Q-switch, consisting of a thin graphite sheet vaporized by a Nd:YAG laser, was reported²⁵ in 1986. However, none of these has lead to a commercially-available system.

The first determination of the cross-relaxation coefficients of Er:YAG appeared²⁶ in 1984, with further revisions^{27,28,12} appearing almost immediately. However, model-

ing^{12,27} of the Er: YAG laser is difficult and varying the cross-relaxation coefficients does not always appear to have a large effect on the predicted output. It is therefore difficult to state the values of these coefficients with a great degree of certainty.

1.5 Ho: YAG Laser Experiments

The first report²⁹ of a Ho:YAG laser was in 1965. This was on the 2.1- μ m ⁵I₇ \rightarrow ⁵I₈ transition, however, not the 3- μ m ⁵I₆ \rightarrow ⁵I₇ transition. The first report¹⁴ of lasing on the 3- μ m transition was in 1975. In 1976, more specific data¹⁹ on this lasing transition were reported.

Unlike those of Er:YAG, the relevant laser parameters of Ho:YAG have significant concentration effects.³⁰ The fluorescence decay time of the ${}^{5}I_{6}$ upper laser level has only a slight change in varying the concentration of Ho³⁺ from 0.2% (50 µs) to 100% (42 µs). However, the lower laser level (${}^{5}I_{7}$) exhibits strong concentration quenching, with the lifetime varying from 8.5 ms at 0.2% Ho³⁺ concentration to less than 300 µs for 100% concentration. The decrease in lifetime is roughly linear. Lasing on the 3 µm ${}^{5}I_{6} \rightarrow {}^{5}I_{7}$ transition, without serendipitous energy level resonances like those found in Er:YAG, is self-terminated laser and demonstrates redshifting.

1.6 Co-Doping Experiments

Co-doping to reduce lasing thresholds was reported⁹ as early as 1964. In this work, the authors report energy transfer from the $\text{Er}^{3+4}I_{13/2}$ level to the Ho³⁺⁵I₇ and Tm³⁺ ${}^{3}\text{H}_{4}$ levels in CaMoO₄. These levels are the upper levels of the 2.1-µm holmium laser and the 1.9-µm thulium laser, respectively. In a later work,²⁹ Johnson *et al.* report that 1% Tm:(Y_{0.5}Er_{0.5})₃Al₅O₁₂ fluoresces 40 times as strongly on the Tm³⁺³F₄ \rightarrow ³H₆ 1.9-µm transition as does 1% Tm:Y₃Al₅O₁₂, that is, that replacing 50% of the Y³⁺ ions in Tm:YAG by Er³⁺ ions results in a 40-fold increase in pumping of the Tm³⁺³H₄ level. They also

report that in Ho:YAG, the replacement of 50% of the Y^{3+} ions by Er^{3+} ions results in a 20-fold increase in pumping of the Ho^{3+ 5}I₇ level. In both cases, fluorescence from the $Er^{3+ 4}I_{13/2}$ is strongly quenched as compared to 50% Er:YAG, indicating significant energy transfer. Although these reports deal with other laser transitions than those of interest to this work, they do demonstrate the possibility of using Ho³⁺ or Tm³⁺ co-dopants to reduce the ⁴I_{13/2} lower laser level lifetime in Er:YAG.

Energy transfer from Er^{3+} to Ho^{3+} in YAG was studied thoroughly³¹ in 1971, at temperatures ranging from 77 K to 700 K. This paper reports that, for low concentrations of Er^{3+} and Ho^{3+} ions (2% and 0.2%, respectively), the energy transfer process is more efficient at low temperatures than at high temperatures. However, it also notes that the energy transfer from the $Er^{3+} {}^{4}I_{13/2}$ level to the $Ho^{3+} {}^{5}I_{7}$ is so strong when the Er^{3+} concentration is high (50% in this case) that this transfer has no noticeable temperature dependence. Also, this paper reports that, although the ${}^{4}I_{13/2}$ level of Er^{3+} has a fairly constant lifetime of 12 ms for high concentrations, the ${}^{5}I_{7}$ level of Ho^{3+} , which is also 12 ms at 300 K for all concentrations of Ho^{3+} and Er^{3+} , becomes strongly temperature-dependent when the crystal contains a high concentration of Er^{3+} .

The first suggestion of using Nd³⁺ to deactivate the lower laser levels of the 3µm Er³⁺ and Ho³⁺ transitions appeared¹⁴ in 1975. This work was continued^{10,11} in 1987. In the 1987 reports, reduction of the Er^{3+ 4}I_{11/2} from 107 to 53 µs, the ⁴I_{13/2} from 5,000 to 81 µs, and the Nd^{3+ 4}F_{3/2} from 237 to 8 µs were observed when comparing 17% Er:YAG and 1% Nd:YAG to (15% Er, 1% Nd):YAG. These lifetime changes, especially the factors of 60 in the Er^{3+ 4}I_{13/2} and 30 in the Nd^{3+ 4}F_{3/2} levels, indicate strong energy transfer both from Nd³⁺ to Er³⁺ and from Er³⁺ to Nd³⁺ ions. In (Ho, Nd):YAG the same reduction of the Nd^{3+ 4}F_{3/2} level lifetime, virtually no change in the Ho^{3+ 5}I₆ lifetime, and a factor of 30 reduction in the Ho^{3+ 5}I₇ lifetime (the lower level of the Ho³⁺ 3-µm transition) were reported. In both co-doped cases, lasing at the wavelengths associated with the Er^{3+} and Ho³⁺ 3-µm transitions were observed, isochronic with lasing at wavelengths associated with the Nd³⁺ 1.06-µm and 1.34-µm transitions. However, despite the large relative reductions of the lower laser level lifetimes as compared to the upper laser level lifetimes, there was a loss in lasing efficiency in both (Er, Nd):YAG and (Ho, Nd):YAG compared to Er:YAG and Ho:YAG. This would be expected for the Er^{3+} 3-µm transition, as the $\text{Er}^{3+} \rightarrow \text{Nd}^{3+}$ energy transfer reduces the essential cross-relaxation pumping. However, the loss of efficiency was a surprise in (Ho, Nd):YAG, as Ho:YAG is not a cross-relaxation pumped laser. Reduction in self-termination of the lasing was observed, and a subsequent increase in lasing efficiency would be expected from this as it allows longer laser pulses, approximating the length of the long pump pulses. It is likely that a large part of the loss of efficiency in (Ho, Nd):YAG was due to the low optical quality of the (Ho, Nd):YAG crystals.

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Chapter 2—Spectroscopy

2.1 Introduction

Absorption, emission, and excitation spectroscopy provide much information about laser crystals. Absorption spectroscopy can clearly define the ions' energy levels in this crystalline matrix, which helps in determining efficient pumping wavelengths (usually the work of excitation spectroscopy). Information as to the energy levels of the material is crucial in determining which energy transfer processes will be important and which can be ignored, as well as in predicting the actual lasing wavelengths to be observed. Emission spectroscopy can provide a check on these wavelengths. In addition, the lifetime data discovered during emission spectroscopy (*i.e.*, the ions' fluorescence as a function of time when pumped into a higher-lying level) can suggest existence or non-existence of ion-ion energy transfer processes, both by changes of effective level lifetimes with change in doping concentration of at least one impurity ion and by non-exponential decay of various levels.

In these experiments, both absorption and emission spectroscopy were performed. The thrust of this dissertation, which is to study the energy transfer processes among the Er^{3+} and the Ho³⁺ ions in the YAG crystal, did not require excitation spectroscopy.

2.2 Crystals

For the absorption and emission spectroscopy, pieces of seven boules were used. All seven boules had been grown by the Czochralski technique. The host crystal was YAG ($Y_3Al_5O_{12}$), and the dopants and concentrations were (1) 16.7% Er^{3+} , (2) 30% Er^{3+} , (3) 33.3% Er^{3+} , (4) 40% Er^{3+} , (5) 50% Er^{3+} , (6) 30% Er^{3+} and 1.5% Ho^{3+} , (7) 40% Er^{3+} and 1.5% Ho^{3+} , and (8) 15% Ho^{3+} (all concentrations in at. %). All crystals were of good optical quality and free of defects visible to the naked eye. All emission-spectroscopy crystals were rectangular in cross-section of the output face. The top, bottom, and side of these crystals were polished (in this geometry, the pump light travelled from top to bottom in the crystal and the emission face was the side; see Fig. 2.1).



Figure 2.1. Geometry of crystals used in emission spectroscopy.

The 16.7%, 33.3% and 50% Er:YAG absorption spectroscopy samples were very thin, so as to avoid the difficulties involved in measuring transmission of highly optically dense media. Each of these samples was roughly a disc, 2 cm in diameter and 0.25 mm thick. The (30% Er, 1.5% Ho):YAG sample used in the absorption spectroscopy was roughly triangular, about 15 mm on a side and 3.5 mm thick. The 15% Ho:YAG sample was approximately the same size and shape as the doubly-doped sample. Although the concentrations of Ho³⁺ differed significantly between the singly- and doubly-doped samples, only the absorption density changed significantly, not the spectral location of the peaks.

2.3 Absorption Spectroscopy

2.3.1 Description of the Experiments

Absorption-vs.-wavelength measurements were performed on crystals of 16.7% Er:YAG, 33.3% Er:YAG, 50% Er:YAG, 15% Ho:YAG, and (30% Er, 1% Ho):YAG. These measurements would indicate the presence of unwanted dopants in the crystals, if any, as these contaminants would show up as unexpected absorption features. Comparison with previous work¹ allowed tentative identification of the absorption features with their energy levels.

The absorption of these crystals was measured with a Cary 2300 computer-controlled double-beam spectrophotometer. All absorption spectra were taken at 300 K and used unpolarized light. The Cary 2300 used a tungsten-filament light source for the reported absorption measurements. A photomultiplier measured transmitted light for wavelengths shorter than 600 nm and an uncooled PbS detector measured transmitted light for wavelengths between 600 and 3100 nm.

Although the instrument was a dual-beam spectrophotometer, no undoped YAG samples were used as references. Thus, there is an error in the absorption measurements due to Fresnel losses. There are also measurement errors caused by absorption in the atmosphere and the optics. To remove theses errors, the Cary 2300 took a standard reference using undoped YAG. The computer calibrated the data with this reference by automatically subtracting its effects.

2.3.2 Absorption Spectroscopy Results

The results of the absorption measurements of the 33.3% Er:YAG samples are shown in Fig. 2.2. The absorption spectroscopy of 16.7% and 50% Er:YAG produced identical results with (respectively) 50% less and 50% more absorption. In Fig. 2.3 the absorption measurements of the 15% Ho:YAG are displayed, and in Fig. 2.4 the spectroscopy of the (30% Er, 1.5% Ho):YAG sample appears. In some wavelength regions, the (Er, Ho):YAG and Ho:YAG samples were less than 1% transmissive, exceeding the dynamic range of the spectrophotometers. These regions are spectrally narrow, and their opacity does not affect the measurements outside the opaque regions. In all cases, I considered spectral location of the absorption features to be more important than absolute



Figure 2.2a. Absorptivity of 33% Er:YAG vs. wavelength in the visible region of 350-600 nm.



Figure 2.2b. Absorptivity of 33% Er:YAG vs. wavelength in the visible to near-infrared region of 600-850 nm.


Figure 2.2c. Absorptivity of 33% Er:YAG vs. wavelength in the near-infrared region of 0.85—1.10 µm. (Note: there are no absorption features between 1.10 and 1.40 µm.)





strength of these features. The high absorption did not prevent locating the peaks of the features.

Comparing the figures, it is apparent that the (Er, Ho): YAG absorption spectrum is essentially that which would be obtained by adding the Ho: YAG spectrum to the Er: YAG spectrum.





Figure 2.3b. Absorptivity of 15% Ho:YAG vs. wavelength in the near-infrared region of 650—950 nm. (Note: there are no absorption features between 950 and 1100 nm.)





Figure 2.3d. Absorptivity of 15% Ho:YAG vs. wavelength in the midinfrared region of 1.80–2.20 μ m. (Note: there are no Ho³⁺ absorption features at wavelengths longer than 2.20 μ m.)



Figure 2.4a. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the visible region of 350-650 nm.



Figure 2.4b. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the near-infrared region of 650—950 nm.



Figure 2.4c. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the near-infrared region of 0.95–1.25 µm.



Figure 2.4d. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the infrared region of 1.25–1.55 µm.



Figure 2.4e. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the infrared region of 1.55–1.85 µm.



Figure 2.4f. Absorptivity of (30% Er, 1.5% Ho):YAG vs. wavelength in the mid-infrared region of 1.85–2.15 µm.

2.4 Emission Spectroscopy

2.4.1 Experimental Description

To measure the laser level lifetimes, fluorescence was excited by pumping the crystals with a focused beam from a 532-nm (frequency-doubled), Q-switched Nd:YAG laser operating at a repetition rate of 10 Hz (see Fig. 2.5). The 532-nm output was (temporally) a near-Gaussian, with a full width at half maximum (FWHM) of 25 ns. The energy reaching the crystal was 28 mJ per pulse, and it was focused to a spot 2 mm in diameter with a 50-cm focal length glass lens. Taking into account the Fresnel reflections from the crystal surfaces, the peak intensity of pump light in the crystal was approximately 330 kW/cm², or 8.8×10^{23} photons/cm²s. The pump beam was aligned so that the fluorescing filament was as close as was practical to the exit face of the crystal. This increased the signal at the detector by avoiding reabsorption of fluorescence, but was unlikely to reduce self-trapping effects.



Figure 2.5. Layout of fluorescence study system.

The fluorescence from the sample was captured by a 50-mm focal length, 65-mm diameter lens which imaged it (with unit magnification) onto the entrance slit of a Jar-rell-Ash 0.27-m monochromator. Since this lens was fabricated out of glass, only wave-lengths shorter than 2.5 µm were transmitted; various filters were used to further restrict the wavelength region when the lifetimes were measured. Due to the large solid angle subtended by the lens (approximately 0.1 sr), most of the emitted light was collected, resulting in visible intensities at the detector without causing optical damage to the crystal.

Oscilloscope triggering was accomplished by placing a fast silicon photodiode where it would intercept some of the pump light scattered from the surface of the sample. This allowed the fluorescence measurements to be synchronized to the pump with great accuracy.

The monochromator used had a focal length of approximately 27 cm. It had three available gratings, of which only one was used. This grating had 300 grooves/mm and was blazed at 2 μ m. The monochromator was calibrated with a low-pressure mercury lamp using five visible emission lines (408.0, 435.7, 546.1, 577.0, and 579.0 nm) in four orders each. In addition, it was calibrated in place with two laser lines from HeNe lasers: 632.8 and 594.1 nm. The accuracy of the monochromator could thus be calibrated to the instrumental limit of approximately 0.5 nm when the narrowest (10 μ m wide) slits were used. The repeatability of measurement was also excellent. When there was sufficient intensity, averaged multiple readings of the monochromator output could allow accuracies of approximately 0.5 nm. However, in most cases, the fluorescence measurements did not achieve sufficient intensity to allow use of narrow slits; 250 μ m was the normal width. This degraded resolution somewhat, but individual emission peaks within fluorescence levels were still easy to distinguish.

An InSb detector, cooled to 77 K by liquid nitrogen, was used to convert the emission to electrical signals. This detector operated with a matched transimpedance amplifier whose bandwidth was 500 kHz. The detector itself had a rise time shorter than 1 μ s, although this was achieved at the expense of some sensitivity. The InSb detector measured emission from 0.4 to 5.5 μ m, but the imaging lens used to collect the fluorescence limited the long-wavelength response to less than 2.5 μ m.

The amplifier's output was then sampled by a digital processing oscilloscope (DPO) which was controlled by a microcomputer. This combination made it possible to average a large number of fluorescence signals, greatly improving the signal-to-noise ratio (SNR). SNR improves as the square root of the number of signals averaged. In theory, virtually any number of signals could have been averaged, achieving an infinite SNR. Reality, however, was limited by time constraints. The pump laser only produced stable pulses for approximately one hour, limiting the maximum number of averaged signals to about 400, for an SNR improvement ratio of 20.

The DPO sampled the signal at 512 evenly-spaced points across its temporal view. Each point was digitized with 10 bit resolution of detector voltage. The digitizing resolution was based on the ratio of the signal amplitude to the maximum viewable on the screen. It was thus independent of signal voltage. However, noise level was also independent of signal voltage, so very small signals had low SNR's.

The nature of the data acquisition process required that many precautions be taken to ensure data accuracy. For example, all data signals acquired a dc offset due to the amplifier. This offset was measured by repeating each fluorescence data acquisition run with the monochromator slit blocked. The offset was then automatically subtracted from the measured signal, point by point. The offset, unfortunately, could be so large compared to the signal that a dc offset amplifier was required. Although this amplifier introduced another source of measurement drift, it was a better choice than using ac coupling to measure the signal, as the latter method caused severe distortions at long (>1 ms) measurement times.

The system response was checked carefully to be sure that no systematic errors were present. This check was confirmed by measuring the fluorescence decay of a standard 1% Nd:YAG sample which had a known time constant. Such care was necessary as signal offsets appeared in these measurements as non-exponential fluorescence decays. Since these non-exponential decays were specifically sought in the experiments, it was necessary to be certain that the measured non-exponential decays were not instrument artifacts. Thus, the accuracy required forced the extra effort and time spent ensuring the accuracy of these measurements.

Fluorescence decay of the $Er^{3+4}I_{11/2}$ and ${}^{4}I_{13/2}$ energy levels was measured in 30% Er:YAG, 40% Er:YAG, (30% Er, 1.5% Ho):YAG, and (40% Er, 1.5% Ho):YAG. Fluorescence decay of the Ho³⁺⁵I₆ and ${}^{5}I_7$ energy levels were also measured in the two doubly-doped crystals. There was very little fluorescence from the higher energy levels, and the decay times were too short to measure given the weak signals. The 532-nm pump excited the crystals into the $Er^{3+4}S_{3/2}$ and ${}^{2}H_{11/2}$ levels; in the doubly-doped crystals, some Ho³⁺ ions were also excited into the ${}^{5}F_4$ and ${}^{5}S_2$ energy levels. The fast risetime of the fluorescence in the $Er^{3+4}I_{11/2}$ and Ho³⁺⁵I₆ energy levels indicates that there were few ion-ion interactions involving levels above these, which was also indicated by the weakness of the fluorescence from these higher levels.

The decay lifetime of the $\text{Er}^{3+4}I_{11/2}$ energy level was measured by setting the monochromator to 1.02 μ m, corresponding to its decay to the ground-state ${}^{4}I_{15/2}$ level. Likewise, the decay of the ${}^{4}I_{13/2}$ energy level was monitored by setting the monochromator to 1.54 μ m, corresponding to its ground-state decay. Fluorescence of the Ho³⁺⁵I_c

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level was measured at 1.21 μ m and the ${}^{5}I_{\gamma}$ at 2.06 μ m, corresponding to their decays to the Ho^{3+ 5}I₈ ground level. As an added precaution, in the doubly-doped samples, the fluorescence was filtered through a piece of germanium (with a long-pass cutoff of 1.8 μ m) when measuring the Ho^{3+ 5}I₇. Similarly, a piece of colored glass (with a short-pass cutoff of 1.1 μ m) was used when measuring the Er^{3+ 4}I_{11/2} level. This was to avoid confusing the second order of the ⁴I_{11/2}, which extended from about 1.92—2.05 μ m, with the first order of the ⁵I₇, which extended from about 2.01—2.10 μ m.

After all the data were collected, the signals were plotted on semilog graphs. Thus, a purely exponential decay would appear as a straight line. The computer also performed a least-squares linear fit to the data to provide a reference; this line defines the equivalent exponential lifetime of the level. When the ${}^{4}F_{3/2}$ energy level of Nd:YAG was measured, its decay was found to be exponential at $237 \pm 10 \,\mu$ s, which agrees with other well-documented experimental values and resulted in a validation of this measurement system.

2.4.2 Results of Experiments

Each decay signal was measured between 40 and 400 times during the data acquisition and averaging process. In many cases the noise on the measurements was as large as 10% of the signal, but much of this is random and can be ignored. In some cases, the strongly non-exponential nature of the fluorescence decay made interpretation of the results difficult; it is unreasonable to assign an exponential decay lifetime to data that are strongly non-exponential. However, the decays often seem to approach exponentiality after sufficient time. In these cases it is possible to assign an asymptotic value for the exponential decay lifetime. Decays of the $\text{Er}^{3+4}I_{11/2}$ level are shown in Fig. 2.6. In the 30% and 40% Er:YAG samples, the lifetime of this level is approximately 120 µs. The decay is non-exponential, lengthening due to upconversion from the ${}^{4}I_{13/2}$. In the (Er, Ho) materials, this decay is very close to exponential, being about 59 µs in (30% Er, 1.5% Ho):YAG and about 51 µs in (40% Er, 1.5% Ho):YAG.

The $\mathrm{Er}^{3+4}I_{13/2}$ fluorescence is seen in Fig. 2.7. These decays are exceptionally non-exponential in the singly-doped Er: YAG samples, with asymptotic lifetimes approximating 4.2 ms, and are much more nearly exponential in the doubly-doped samples, at 5.5 ms for the (30% Er, 1.5% Ho): YAG sample and 4.4 ms for the (40% Er, 1.5% Ho): YAG sample.

The Ho^{3+ 5}I₆ and 5 I₇ fluorescence signals are shown in Figs. 2.8 and 2.9. I did not have any low-concentration Ho:YAG samples to compare with; the (30% Er, 1.5%



Figure 2.6a. Fluorescence from the ${}^{4}I_{_{III2}}$ level of Er^{3+} in 30% Er:YAG. Straight line is a least-squares fit to the measured initial decay time of 120 μ s.



Figure 2.6b. Fluorescence from the ${}^{4}I_{_{III2}}$ level of Er^{3+} in 40% Er:YAG. Straight line is a least-squares fit to the measured initial decay time of 120 µs.



Figure 2.6c. Fluorescence from the ${}^{4}I_{\mu\nu}$ level of Er^{3+} in (30% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 59 µs.



Figure 2.6d. Fluorescence from the ${}^{4}I_{1112}$ level of Er^{3+} in (40% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 51 µs.



Figure 2.7a. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in 30% Er:YAG. Straight line is a least-squares fit to the measured late decay time of 4.2 ms.



Figure 2.7b. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in 40% Er:YAG. Straight line is a least-squares fit to the measured late decay time of 4.2 ms.



Figure 2.7c. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in (30% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 5.5 ms.



Figure 2.7d. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in (40% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 4.4 ms.



Figure 2.8a. Fluorescence from the ${}^{5}I_{6}$ level of Ho³⁺ in (30% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 69 μ s.



Figure 2.8b. Fluorescence from the ${}^{5}I_{6}$ level of Ho³⁺ in (40% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 67 µs.



Figure 2.9a. Fluorescence from the ${}^{5}I_{7}$ level of Ho $^{3+}$ in (30% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 5.5 ms.



Figure 2.9b. Fluorescence from the ${}^{5}I_{7}$ level of Ho $^{3+}$ in (40% Er, 1.5% Ho):YAG. Straight line is a least-squares fit to the measured decay time of 4.9 ms.

Ho): YAG and (40% Er, 1.5% Ho): YAG samples are compared to 15% Ho: YAG². The Ho³⁺ lifetimes are slightly longer in the doubly-doped samples, but the more significant fact is that the Ho³⁺ lifetimes approximate the lifetimes of the nearest Er^{3+} energy levels in the doubly-doped materials (*i.e.*, the Ho^{3+ 5}I₇ lifetime is within 10% of the Er^{3+} ⁴I_{13/2} and the Ho^{3+ 5}I₆ is within 30% of the Er^{3+} ⁴I_{11/2}). This indicates energy transfer between the unlike ions. In the (30% Er, 1.5% Ho): YAG sample the different ions' lifetimes are closer than in the (40% Er, 1.5% Ho): YAG sample. Also, in the doubly-doped samples the Er^{3+} ⁴I_{11/2} lifetimes are shorter than in the singly-doped samples, while the ⁴I_{13/2} lifetimes are longer. These comparisons are easily made in Table 2.1.

Dopant	⁴ I _{11/2} lifetime	⁵ I ₆ lifetime	⁴ I _{13/2} lifetime	⁵ I ₇ lifetime
30% Er	120 µs	A20.4200	4.2 ms	
30% Er, 1.5% Ho	59 µs	69 µs	5.5 ms	5.5 ms
40% Er	120 µs		4.2 ms	Status Para
40% Er, 1.5% Ho	51 μs	67 µs	4.4 ms	4.9 ms
15% Ho		47 µs	THE OF YORK	4.6 ms

Table 2.1. Lifetimes of the studied energy levels of Er^{3+} and Ho^{3+} in Er:YAG, Ho:YAG, and (Er, Ho):YAG.

References—Chapter 2

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- 2. J. Machan, Spectroscopic and 3 μm Lasing Properties of Ho:Y₃Al₅O₁₂ incorporating Nd³⁺ Deactivator and Yb³⁺ Sensitizer Ions, Ph.D. Dissertation, USC (1989).

Chapter 3—Lasing Tests

3.1 Introduction

The true test of a laser medium occurs when the laser itself is tested. In order to understand the dynamics of the (Er, Ho):YAG laser system, its lasing performance and its spectroscopic parameters were tested. The results of the lasing tests were then compared to those of related laser media, especially Er:YAG. In this way, it was possible to study the effects of co-doping Er:YAG lasers with Ho³⁺ ions and to contrast this with other co-doped lasers.

For this test, laser rods were fabricated from 17% Er:YAG, 33% Er:YAG, 50% Er:YAG, 15% Ho:YAG, (15% Er, 1% Nd):YAG, (10% Ho, 1% Nd):YAG, and (30% Er, 1.5% Ho):YAG. The "effective" length of these rods (defined as the length of the rods which were exposed to flashlamp light in the pump cavity) varied from 72 mm [in the case of (Er, Nd):YAG] to 100 mm (in the case of Er:YAG). This length was also dependent on the pump cavity used; the MALLET and Apollo 1610 laser test beds accepted effective rod lengths of up to 100 mm, while the Quantronix laser test bed allowed a maximum effective length of only 75 mm. The rods themselves all had plane parallel ends with no anti-reflection coatings.

Another parameter that varied among the tested laser rods was optical quality of the laser crystal. The worst optical quality was found in the (Ho, Nd):YAG crystal, which had several striations visible along its length, indicating possible locations for scatter of the recirculating laser light as well as susceptibility to thermal stress and nonuniform thermal lensing. The Er:YAG laser rods, on the other hand, were the product of more mature growth technology. None of the tested Er:YAG crystals had any defects visible to the naked eye. The best quality Er:YAG laser rod produced a distortion of only $\lambda/2$ at 594.1 nm in transmission and induced no birefringence effects; the worst had a distortion of approximately 5λ in transmission and added an unpumped polarization distortion of nearly π radians. The optical quality variation appeared to be the most significant cause of laser performance variation among the Er:YAG laser rods, making a larger difference in lasing efficiency than doping level.

In lasing experiments involving the MALLET and Quantronix test beds, the laser rod was pumped with xenon-filled flashlamps. Krypton-filled lamps were tested in the Apollo 1610 system to compare their pumping capabilities with xenon-filled lamps. The xenon-filled lamps were more efficient as they have a better spectral match to the Er^{3+} pump bands than do the krypton-filled lamps. The main pump bands of Er^{3+} occur in the blue and green, where the xenon-filled lamps are brighter, and there are no Er^{3+} absorption features in the 810-nm range where the krypton-filled lamps have bright emission.

All laser rods were tested in the MALLET test bed. The Er:YAG, (Er, Ho):YAG, Ho:YAG, and (Ho, Nd):YAG rods were also tested in the Quantronix system, and the 33% Er:YAG rod in the Apollo 1610 cavity. All three test systems consisted of pump cavities with water cooling, a high reflector (HR) with at least 99% reflectivity at 2.94 µm (the Er:YAG laser wavelength), and several output couplers (OC's). Each system is described in greater detail below.

3.2 MALLET Laser Test Bed

The MALLET laser test bed was originally designed for use in an alexandrite laser system. The pump cavity was a double ellipse with the rod occupying the joint focus of the ellipses. One flashlamp was at each of the other two foci. Each flashlamp and the rod had its own Pyrex flow tube which contained the cooling water and prevented pump wavelengths shorter than about 350 nm from reaching the laser rod. The flashlamp reflector was silver-backed Pyrex. This system allowed pumping of up to 100 mm of the rod length. The pulse-forming network (PFN) provided up to 500 J per pulse to the flashlamps, with a temporal shape similar to a rectified sinusoid.

The maximum power available to the PFN was 2000 W, which limited the pump repetition frequency (PRF) to four Hz at maximum input. The maximum PRF used in the experiments in this cavity was seven Hz at lower input energies. The pump pulse length was 175 μ s full width at half maximum (FWHM).

The HR and OC were separated by 41 cm, and both were flat. Thermal lensing in the laser medium was sufficient to allow stability. The measured reflectivity of the HR was 99.7% between 2.94 and 3.02 μ m, >98.5% between 1.0 and 1.4 μ m, and 98.8% at 2.80 μ m. [These wavelengths were chosen for mirror reflectivity measurement because Er:YAG lased at 2.94 μ m, while Ho:YAG lased between 2.94 and 3.02 μ m, (Er, Ho):YAG at 2.94 and 2.8 μ m, (Er, Nd):YAG at 2.94 and 1.01---1.15 μ m, and (Ho, Nd):YAG at 2.94, 3.01, 1.06, and 1.34 μ m.] The OC's had reflectivities ranging from 70.3% to 98.9%. The reflectivity of each OC was measured at each laser wavelength at which it was used.

For laser wavelength determination, the laser output was directed into a 0.27-m focal length monochromator with 250-nm slits. The wavelength resolution of this system was approximately ± 3 nm. Before use this monochromator was calibrated with mercury lamps, using at least five mercury emission lines in at least four orders. While in place, the calibration was checked with lasers at 594.1 and 632.8 nm, also in at least four monochromator orders. All three concentrations of Er:YAG, the 17% Er^{3+} , 33% Er^{3+} , and 50% Er^{3+} , lased at the same apparent wavelength of 2936.2 nm, determined by making several measurements and averaging them. Due to the resolution of the monochromator, however, if there were any other laser lines emitting between 2933 and 2940 nm, they would not have been observed.

In order to force single-mode lasing, apertures were placed within the laser cavity. Although it was possible to achieve lasing on the lowest-order mode by this method, lasing efficiency also dropped by a factor of 10. Because Er:YAG is optically dense, much of the gain appears near the outside of the rod, which contributes to higher-order modes and greatly reduces the efficiency of lower-order modes.



Figure 3.1. Input vs. output for three dopant concentrations of flashlamp pumped Er:YAG laser. Only the best results from each concentration are presented.

Free-running lasing, with no Q-switching or mode control, provided the greatest efficiency in Er:YAG. The best results for each concentration are presented in Fig. 3.1. For the 33% and 50% doping levels, the best performance was with an OC reflectivity of 73.1%. For the 17% doping level, the best results were obtained with an OC reflectivity of 82.8%. The very best results obtained with this cavity were 8.0 J output for 500 J input at 1.1 Hz, at an overall efficiency of 1.6% and a slope efficiency of 1.8% with an

OC reflectivity of 73.1%. At 3.3 Hz, the best power output was obtained, being 5.3 J per pulse, or 17.5 W, for 1650 W in, or 1.1% overall efficiency (and 1.2% slope efficiency in this case) with an OC reflectivity of 82.8%. In these cases, the rod was doped with 50% Er^{3+} ions, and was seven mm in diameter and 120 mm long.



Figure 3.2. Input vs. output for two dopant concentrations of flashlamp pumped Er:YAG laser. Early comparison with 6.35-mm diameter rods.

In all tests, the 33% and 50% Er:YAG laser rods performed better than the 17% Er:YAG rods. In all tests except those described in the above paragraph, the rods were 6.35 mm in diameter and 120 mm long. Tests of these rods demonstrated that optical quality of the laser rod was a better predictor of lasing efficiency than dopant level, at least when 33% and 50% dopant levels were compared. As Fig. 3.2 demonstrates, early experiments with 6.35-mm diameter rods showed the 33% Er:YAG laser as somewhat more efficient than the 50% Er:YAG laser (in this case, the OC reflectivity was 84.5%). However, close examination of the rods (using crossed polarizers to locate stress lines in

the crystal) demonstrated that the optical quality of the tested 33% Er:YAG laser rod was noticeably better than that of the 50% Er:YAG laser rod. This difference is probably the cause of the lack in efficiency shown by the 6.35-mm diameter 50% Er:YAG laser rod. The optical quality of the seven-mm diameter 50% Er:YAG laser rod was the highest of any crystal tested, and it produced the best results, so it is fairly certain that optical quality plays a major role in efficiency of Er:YAG lasers.

Thermal lensing of laser rods is an important parameter. It can be used to determine the best mirror curvatures and pump levels for a laser. Thermal lensing of Er:YAG laser rods was measured by shining a 594.1-nm laser through the cavity. This wavelength was selected because it is visible, there is a convenient HeNe laser line at that wavelength, and Er:YAG is transparent at 594.1 nm (as it is not at 632.8 nm, the most common HeNe laser wavelength). The beam diameter was 10 nm, and it passed through two holes just before reaching the laser rod. These holes were 0.5 nm in diameter and the point halfway between them was centered on the rod axis. The centers of the two holes were separated by two, three, or five mm to allow thermal lensing measurements in different portion of the laser rod. Effectively, two parallel 594.1-nm laser beams, each 0.5 mm in diameter, passed through the laser rod at equal distances from its axis. These two beams were projected onto a screen 3.7 m from the laser rod. The maximum separation of the two spots on the screen was then used to determine the thermal focal length of the laser rod.

In all thermal lensing measurements, the effective lensing was linear with input power. For 33% and 50% Er:YAG thermal lensing occurred at a rate of 1.4 diopters/kW to the flashlamps at 2.5 mm from the center of the rod, dropping to 1.2 diopters/kW at 1.5 mm and 1.1 diopters/kW at 1.0 mm. For 17% Er:YAG, thermal lensing was between 1.0 and 1.1 diopters/kW across the entire radius. This indicates that 17% Er:YAG is not as dense (optically) as are 33% and 50%, which show a marked increase in thermal lensing as radius (from the rod center) increases. Also, since 1% Nd:YAG lenses thermally at a rate of approximately 0.7 diopters/kW in this measurement system, it appears that Er:YAG with dopant concentrations of at least 33% absorb approximately twice as much energy from the flashlamp as does 1% Nd:YAG.

3.3 Quantronix Laser Test Bed

The Quantronix laser test bed was similar to the MALLET system in many ways. For example, it also used silver-backed Pyrex as its pump reflector. It was different from the MALLET in that it was a single ellipse, with a single flashlamp at one focus and the laser rod at the other. There were no flow tubes in the cavity, which was flooded with water. The maximum pumped length allowed by this system was 75 mm. The pump pulse length was approximately $300 \ \mu$ s, and it was a "flat-topped" pulse with rise time approximately $20 \ \mu$ s and fall time about $50 \ \mu$ s. The PFN provided up to 245 J per pulse to the flashlamp.

The maximum power available to the PFN was 820 W, which limited the PRF to 3 Hz. All of the lasing experiments were performed at 2 Hz to avoid overheating the cavity reflector and degrading its performance.

The HR and OC were separated by 36 cm in these experiments. The same mirrors were used in the Quantronix system as in the MALLET. The rods tested in this cavity were 17%, 33%, and 50% Er:YAG, (30% Er, 1.5% Ho):YAG, 15% Ho:YAG, and (10% Ho, 1% Nd):YAG.

One of the most interesting discoveries made in this laser cavity was that Er:YAG actually lases on two lines near 2.94 μ m simultaneously. These lines were separated by directing the laser output into a 0.27-m focal length monochromator with 10 nm slits. With these slits in place, the accuracy of the monochromator was ± 0.5 nm. The laser lines appeared at 2936.2 nm and 2938.6 nm. When the accuracy of the monochromator

is taken into account, and the precision with which these measurements were made, the total error can be determined to be ± 0.8 nm. The two laser lines, although they appeared simultaneously, showed consistently different waveforms (see Fig. 3.8) and had slightly different lasing thresholds. Both lines appeared in all three dopant concentrations of Er: YAG and (Er, Ho): YAG with no discernable wavelength shift.

In addition to these two laser lines, (Er, Ho):YAG lased at 2795.6 and 2765.5 nm, again with possible errors up to ± 0.8 nm. A more thorough description of the lasing of (Er, Ho):YAG appears in Section 3.10.

The lasing at 2.8 μ m seen in (Er, Ho):YAG appears to come from Er³⁺ ions. In (Er, Ho):YAG, the gain seems to be higher at 2.8 μ m than at 2.94 μ m, but the loss due to atmospheric absorption is also much greater at 2.8 μ m. In an attempt to lase Er:YAG at 2.8 μ m by reducing the loss at this wavelength, a box was constructed around the laser cavity and this box was flooded with a positive pressure of cold, dry nitrogen (taken from a liquid nitrogen tank). This reduced the temperature inside the box to approximately -20° C, as well as driving out some of the carbon dioxide (which also absorbs at 2.8 but not 2.94 μ m). This reduced the intracavity water vapor by about 95% and the carbon dioxide by half. Even with this reduction in intracavity absorption, Er:YAG lased only at 2.94 μ m. The efficiency, however, increased by approximately 15% when the box was flooded with nitrogen.

3.4 Apollo 1610 Laser Test Bed

The Apollo 1610 was designed as an Nd: YAG machining/welding laser. Like the MALLET, its cavity was a double ellipse, using two flashlamps to pump the rod. There were Pyrex flow tubes containing the cooling water around each flashlamp and the rod. The reflector was gold, not silver-backed Pyrex as in the MALLET and Quantronix cav-

ities, which reduced the lasing efficiency because gold is not an efficient reflector in the blue and green where Er:YAG has its strongest pump bands.

The pump pulse length of this cavity was adjustable, from 660 μ s FWHM (a rectified sinusoid) to 6.6 ms flat-topped in steps of approximately 660 μ s. In this cavity, the HR was a copper mirror with a radius of curvature of 5 m, and a reflectivity of about 97% at 2.94 μ m. The OC was a dielectric-coated sapphire mirror with reflectivity of 93.2% at 2.94 μ m. The spacing between these mirrors was 47 cm and the effective rod length was 100 mm.

The maximum power available to the PFN was 3 kW, which allowed lasing studies at relatively high PRF's. The highest PRF at which lasing was observed was 22.8 Hz, with the 660 μ s pulse length. The output at this frequency was 10 mW for 2.7 kW in, for an overall efficiency of 3.7×10^{-4} %.

Even when the pump pulse was lengthened to 6.6 ms, the Er: YAG laser still lased in the "quasi-Cw" mode. Fig. 3.3 shows lasing waveforms above threshold for three different pump pulse lengths, 660 µs, 2.0 ms, 3.9 ms, and 6.6 ms. Fig. 3.3d is especially interesting, as it shows two distinct laser pulses within one 6.6-ms pump pulse. A hint of this behavior is seen in Fig. 3.3c, during a 3.9-ms pump pulse. As Fig. 3.3e demonstrates, however, even a 6.6-ms pump pulse generates several ms of "quasi-Cw" lasing when the Er: YAG laser is pumped well above threshold.

When the pump pulse length was varied in this cavity, the effective loss changed. Fig. 3.4 shows a graph of laser threshold power vs. loss due to mirrors. If the Er:YAG laser had no temporal effects, like Nd:YAG, the slopes and intercepts of the lines would have been independent of pump pulse length. The slope of the line should be proportional to the stimulated emission cross section of Er^{3+} in YAG and the y-axis intercept should be proportional to the nonproductive losses in the laser cavity.¹ In this test, shorter



a. Pulse length = 660 µs. Horizontal scale is 400 µs/div, signal scale is 2.5 kW/div, pump scale is 40 kW/div.



b. Pulse length = 2.0 ms. Horizontal scale is 1 ms/div, signal scale is 5 kW/div, pump scale is 40 kW/div.



c. Pulse length = 3.8 ms. Horizontal scale is 1 ms/div, signal scale is 15 kW/div, pump scale is 40 kW/div.



d. Pulse length = 6.6 ms. Horizontal scale is 2 ms/div, signal scale is 2 kW/div, pump scale is 30 kW/div.



e. Pulse length = 6.6 ms. Horizontal scale is 2 ms/div, signal scale is 25 kW/div, pump scale is 40 kW/div.

Figure 3.3. Lasing of 33% Er:YAG at flashlamp pulse lengths from 0.66 to 6.6 ms. Upper trace is pump; lower trace is laser output signal in each oscillograph. Signal scale is on the left axis and pump scale is on the right axis.



Figure 3.4. Laser threshold power of 33% Er:YAG vs. negative logarithm of roundtrip mirror reflectivity for four pump pulse lengths. Apollo 1610 cavity was used.

pulses are seen to have lower absolute intercept values than longer ones (suggesting a time-increasing nonproductive loss). The slopes remain constant with pulse length. Ref. 1 derived the relationship between this slope and the cross section for CW lasers. If lasing occurred in the short pulse mode (*i.e.*, if the laser pulses were short compared to the upper level lifetime) the slope-cross section relationship would hold for a graph of threshold energy, rather than power. The fact that this relationship appears in a power-*vs*.-loss graph indicates that these lasers are running in true CW mode. It may be concluded, then, that what is referred to as "quasi-CW" lasing is, in fact, true CW lasing.

Nonetheless, the lasing threshold energy changed with pulse length (in fact, threshold energy was proportional to pulse length). If both mirrors were perfect reflectors, the threshold for overcoming internal loss would be 100 J for 1.3 ms, increasing to 250 J for 3.3 ms (see Fig. 3.5).



Figure 3.5. Oscillation threshold energy of 33% Er:YAG laser in Apollo 1610 cavity as a function of pump pulse length. Mirrors are assumed to be perfect reflectors.

3.5 Q-Switching Experiment

The peak power of the free-running Er:YAG laser (typically less than 50 kW) is insufficient for many applications. To increase this peak power, Q-switching of the Er:YAG laser was attempted. Both electro-optical and acousto-optical methods were ruled out by the fact that the substrates for these types of Q-switch are typically opaque at 2.94 μ m, and would completely prevent lasing if inserted intracavity.

The Q-switch used in this experiment was soap film. The first step of the experiment was to get the laser free-running well above threshold. Then the loss element was inserted. This element was a film of soap, made from a mixture of water and dishwashing liquid, suspended inside a loop made of thin wire. When the film became sufficiently thin, absorption of the 2.94-µm fluorescence within the cavity would cause it to evaporate rapidly, suddenly reducing the intracavity loss to permit lasing. This soap film then acted as a Q-switch.

It was a slow Q-switch. Assuming the film evaporated explosively at the speed of sound, the time required to change from 100% loss to 0% was nearly 10 μ s. There was also little control on when the film would evaporate. Nonetheless, the Q-switching results were interesting.

An example of the comparison between Q-switched and non-Q-switched lasing waveforms is shown in Fig. 3.6. This figure shows that the Q-switched waveform has several peaks ranging in power from one to eight times that of the free-running system, and reaching a peak power of nearly 300 kW. Better Q-switches should result in higher peak powers and more control over the timing.



Figure 3.6. Lasing of 33% Er:YAG in MALLET cavity. Upper trace is freerunning and lower trace is Q-switched. Horizontal scale is 100 µs/div; vertical scale is 75 kW/div.

Because the soap film evaporated at an unknown rate, it would be anywhere from 30 to 55 seconds between setting it in place and Q-switching. Also, the evaporation tended to be violent, and made a snapping noise. This was accompanied by soap film droplets flying throughout the room, landing on all the optical elements including the

laser rod. Any droplets landing on the rod led to surface damage during subsequent laser pulses. Therefore, this does not seem to be a useful Q-switch for the Er:YAG laser.

3.6 Lasing Behavior of Er:YAG

In a true four-level laser, the lower laser level lifetime is very short. In fact, it is usually modeled as being approximately zero, so no population can ever collect in it. In contrast to this, the lower level lifetime of Er:YAG is long, approximately 100 times as long as the upper laser level lifetime. Er:YAG has an energy level structure like that shown in Fig. 1.4. If a sufficiently large population builds up in the lower laser level (in this case the ${}^{4}I_{13/2}$) the cross-relaxation process dominates the depopulation of that level. Cross-relaxation also populates the upper laser level. In fact, in Er:YAG, the cross-relaxation process is such that two ions in the ${}^{4}I_{13/2}$ level interact, and both are removed from this level. One of the ions drops into the ground state, and the other is excited into the pump band (typically the ${}^{4}S_{3/2}$), from which it decays rapidly into the upper laser level (the ${}^{4}I_{11/2}$). Thus, this cross-relaxation between two ions produces a net increase of three in the inversion; its efficiency is 150%.

One interesting effect of this type of pumping is that the lasing tends to begin after the peak of the pump pulse and continue until near the end of the pulse (Fig. 3.7). This effect is caused by the cross-relaxation pumping system of Er:YAG. Because the inversion is caused by the cross-relaxation process (equations 1.8—11), which is dependent on the square of the population density in the lower laser level (n_1), there is no inversion, and therefore no gain, until a large population has accumulated in this state. The pre-lasing delay will depend on the rapidity with which the ions decay into the ${}^{4}I_{13/2}$ state. This delay is then a function of pump rate, lifetimes of all the levels between the pump band and the ${}^{4}I_{13/2}$, and any nonlinear processes which may affect the decay. (For a more thorough study of these processes, see Reference 1.12.)



Figure 3.7. Lasing behavior of 33% Er:YAG in the MALLET cavity. Upper trace is signal and lower trace is pump. Horizontal scale is 100 µs/div, signal scale (on left) is 10 kW/div, pump scale (on right) is 50 kW/div.

Another interesting feature of Er: YAG lasing also apparent from Fig. 3.7 is that the lasing power rapidly reaches a steady-state value, then remains approximately at this value until it stops completely. This effect is referred to in this dissertation as "quasicw" lasing although, as seen above, it really is true Cw lasing. It is another side effect of Er: YAG's being a cross-relaxation pumped laser. As lasing occurs, the population in the lower laser level remains large (since this is where the lasing ions collect), so the crossrelaxation remains strong, which keeps the laser running. (For a more thorough study of this phenomenon, see Reference 1.27.)

A surprising phenomenon seen in Er:YAG lasers is the double laser line. The 2.94-µm laser is actually lasing on two lines simultaneously, one at 2936 nm and the other at 2939 nm. These two lines have different time signatures (see Fig. 3.8). The laser line near 2936 nm corresponds to the transition $A_2 \rightarrow Y_6$, predicted to be 2936.4 nm, and the line near 2939 nm corresponds to $A_2 \rightarrow Y_7$, predicted to be 2938.8 nm. (The ${}^4I_{11/2}$ is also known as the A level, and the ${}^4I_{13/2}$ as Y. Thus, the A_2 , for example, is the sublevel of ${}^4I_{11/2}$ with the second lowest energy.) It is possible that these two lines have different

polarizations, although neither appeared to vary its power when a sapphire Brewster plate mounted in the output beam was rotated through 90°.



Figure 3.8. Lasing of 50% Er:YAG on two distinct lines near 2.94 µm in Quantronix cavity. Lower trace is laser output signal and upper trace is flashlamp pump. Horizontal scale is 100 µs/div, signal scale is 3 kW/div, pump scale is 10 kW/div.

The equations describing the Er: YAG system are 1.8—11. According to Reference 1.12, a good value for the cross-relaxation parameter W_{cr} is 4.8×10^{-17} cm³/s (note that my definition for W_{cr} is twice their definition for W_1). Rewriting equations 1.8—11 to involve only the lasing levels, assuming that τ_{q_2} is very short,

$$\frac{dn_2}{dt} = R + \frac{1}{2} W_{cr} n_1^2 - \frac{n_2}{\tau_{21}} - \rho B_{21} (n_2 - n_1)$$
(3.1)

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - \frac{n_1}{\tau_{10}} - W_{cr} n_1^2, \qquad (3.2)$$

where $R \equiv W_{03}n_0$ is the rate per unit volume at which pump photons are absorbed (considered to be a constant in this analysis). Comparing equations 3.1—2 to equations 1.5—6, it is tempting to define effective time constants for this laser as

$$\tau_1^{\text{eff}} = \frac{\tau_{10}}{1 + \beta(t)} \tag{3.3a}$$

$$\beta(t) \equiv W_{cr} \tau_{10} n_i(t) \tag{3.3b}$$

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and

$$\tau_2^{\text{eff}} \equiv \frac{\tau_{21}}{1 - \gamma(t)} \tag{3.4a}$$

$$\gamma(t) \equiv W_{cr} \tau_{21} \frac{n_1^2(t)}{n_2(t)}$$
 (3.5a)

It is particularly interesting to note the differences in the time evolution of $\beta(t)$ and $\gamma(t)$ for the continuously pumped (lasing) and delta-pumped (emission spectroscopy) cases. The time evolution in the delta-pumped case is shown in Fig. 3.9. This case does not approach steady state conditions, and β is much less important than γ . β reaches approximately 6.5 for a 28-mJ pump (the value used in the emission spectroscopy), implying a decrease of about 85% in the lifetime of the ${}^{4}I_{13/2}$ level, and it seems to be returning to zero as time increases. On the other hand, γ seems to be approaching one as time increases. These points indicate that, in the delta-pumped case, the effective ${}^{4}I_{11/2}$ lifetime increases to near infinity while the ${}^{4}I_{13/2}$ lifetime changes significantly less.



Figure 3.9. Time evolution of the lifetime functions $\beta(t)$ and $\gamma(t)$ after excitation by a 28-mJ frequency doubled, Q-switched Nd:YAG laser pulse.

The continuously pumped case is quite different. In this case, the equations rapidly approach steady state. Then

$$n_2 \approx \frac{1}{2} W_{cr} \tau_{21}^2$$
 (3.6)

and

$$n_{1} \approx \frac{\sqrt{1 + 2W_{cr} \tau_{10}^{2} R} - 1}{W_{cr} \tau_{10}},$$
(3.7)

which lead to

$$\beta(t) \approx \sqrt{1 + 2W_{cr}\tau_{10}^2R} - 1$$
 (3.8a)

$$\approx \tau_{10} \sqrt{2W_{cr}R} \tag{3.8b}$$

for strong pumping. β can get very large with strong pumping. At inversion threshold (see Appendix A), for example, $R \approx 10^{24}$, so $\beta \approx 70$. On the other hand,

$$\gamma(t) \approx \frac{1}{2} \,. \tag{3.9}$$

In the continuously pumped case, the main effect of the cross-relaxation is to reduce the effective ${}^{4}I_{13/2}$ lifetime to near zero, while only increasing the effective ${}^{4}I_{11/2}$ lifetime by a factor of two. It is the shortening of the effective ${}^{4}I_{13/2}$ level lifetime that makes Er:YAG act as a CW laser at the pulse lengths reported herein.

3.7 Lasing Behavior of Ho: YAG

Unlike Er: YAG, Ho: YAG does not have a strong cross-relaxation process to depopulate its lower laser level. It is not a particularly efficient laser at 3 μ m. The 15% Ho: YAG laser emitted two lines, 2.940 μ m and 3.012 μ m. The maximum output from this laser was 14 mJ (for 400 J input), lasing only at 3.012 μ m in the MALLET laser test bed. It only lased at 3.012 μ m in the MALLET system, but emitted both lines in the Quantronix test bed. This is most likely due to the OC's reflectivity. The OC used in the MALLET experiments had a reflectivity of about 95% at 3.012 μ m and 90% at 2.940 μ m. Using the same mirror on the Quantronix system produced lasing only at 3.012 μ m.

Because there is no outlet for the ions collecting in the ${}^{5}I_{7}$ lower laser level, the 15% Ho:YAG laser self-terminates. With a 170- μ s (FWHM) pump pulse, 3.012- μ m lasing takes place in the first 80 μ s, terminating before the peak of the pump pulse. Shortening the pump pulse increases the laser's efficiency as this eliminates pumping after the laser has terminated.

In the Quantronix cavity, the Ho:YAG laser demonstrated a redshift. During the first 60 μ s of the pump pulse, lasing would begin and terminate at 2.940 μ m. Then lasing would begin at 3.012 μ m, terminating within 20 μ s. This redshift is most likely due to filling of the lower Stark sublevels in the ⁵I₇ level (see Fig. 1.3).

3.8 Lasing Behavior of (Er, Nd): YAG

When Nd^{3+} is added to Er:YAG, efficiency of lasing at 2.94 µm drops significantly. The threshold is about the same, but the output is lower. This is probably due to the interactions between the $Nd^{3+} I_{15/2}$ level and the $Er^{3+} I_{13/2}$, and between the ${}^{4}F_{3/2}$ of Nd^{3+} and the ${}^{4}I_{11/2}$ of Er^{3+} . Since the upper laser level of the Er^{3+} laser may be depopulated by the Nd^{3+} ions, the inversion will be reduced. And, since the lower laser level may also be depopulated, the cross-relaxation which leads to the inversion will also be reduced. Comparisons of the lifetimes of 17% Er:YAG with those of (15% Er, 1% Nd):YAG shows a decrease of 50% in the $Er^{3+} I_{11/2}$ and of over 98% in the $Er^{3+} I_{13/2}$, indicating strong interactions between the Er^{3+} and Nd^{3+} ions that tend to depopulate these crucial levels of the Er:YAG laser.

Co-doping Er^{3+} and Nd^{3+} in YAG has even more significant effects on the 1-µm lasing of the Nd^{3+} ions. Nd:YAG lasers are efficient, narrow band lasers emitting at 1.064 µm. The (15% Er, 1% Nd):YAG laser emitted in a broad band between 1.01 and 1.15

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 μ m with a significant peak at 1.06 μ m. When 3- μ m and 1- μ m lasing occurred simultaneously, they had similar thresholds, and the efficiency of the 1- μ m lasing was reduced by 90% over that of a 1% Nd:YAG rod in the same laser test bed.

3.9 Lasing Behavior of (Ho, Nd): YAG

Adding Nd³⁺ to Ho:YAG produced even more significant results. Lasing of a (10% Ho, 1% Nd):YAG rod occurred at 2.940 and 3.012 μ m, the same wavelengths as those emitted by 15% Ho:YAG. The output and efficiency of the 3- μ m lasing of (Ho, Nd):YAG were a factor of two greater than those of Ho:YAG.

The interactions between the Nd³⁺ and Ho³⁺ ions mainly consisted of energy transfer from the Nd^{3+ 4}F_{3/2} level to the ⁵I₅ of Ho³⁺ and from the Ho^{3+ 5}I₇ to the Nd^{3+ 4}I_{13/2}. These energy transfers are seen by the shortening of the Nd^{3+ 4}F_{3/2} and Ho^{3+ 5}I₇ lifetimes by 97% each. Lifetimes of the other levels were not significantly altered by the co-doping.

A more surprising effect concerned lasing near 1 μ m. Ho:YAG has some absorption at 1.06 μ m, the usual lasing wavelength of Nd³⁺ ions. This added to the loss at 1.06 μ m, sometimes allowing lasing at the secondary Nd:YAG laser line of 1.34 μ m. In fact, when the cavity used mirrors with approximately equal reflectivities at 1.06 μ m and 1.34 μ m, both Nd³⁺ lines lased within the same pump pulse. It was not true simultaneous lasing, as seen in Er:YAG at 2.936 and 2.939 μ m; careful examination of the laser waveforms indicated that 1.06- μ m lasing began and ended the output, 1.34- μ m lasing dominated in the middle, and 1.06- μ m and 1.34- μ m lasing occurred on alternate relaxation oscillation spikes during the transition from one regime to the other.

3.10 Lasing Behavior of (Er, Ho): YAG

The lasing behavior of (Er, Ho): YAG indicates some self-termination. Measured lasing of the (30% Er, 1.5% Ho): YAG laser is shown in Fig. 3.10. Near threshold, the

lasing occurs at 2.94 μ m and is only seen near the beginning of the pump pulse. Above threshold, lasing occurs at 2.94 μ m during the beginning of the pump pulse, self-terminates, and then lasing occurs at 2.8 μ m. Lasing at 2.8 μ m does not appear to be self-terminated.

Due to the normal process of self-termination, most self-terminated lasers have a lasing redshift as the lower Stark sublevels fill. (Er, Ho):YAG has a lasing blueshift, indicating that a different process is at work. Since the first lasing takes place at 2.94 μ m, a wavelength at which Ho³⁺ has a ${}^{5}I_{7} \rightarrow {}^{5}I_{6}$ excited state absorption (ESA), then at 2.8 μ m, at which wavelength Ho³⁺ does not have an absorption, the most likely cause of the wavelength blueshift is ESA in the holmium.

There appears to be energy transfer from the ${}^{5}I_{6}$ level of Ho³⁺ to the ${}^{4}I_{11/2}$ level of Er³⁺, as well as bidirectional energy transfer between the Ho^{3+ 5}I₇ and Er^{3+ 4}I_{13/2} levels. This is indicated by the reduction of the ${}^{4}I_{11/2}$ lifetime, increase of the ${}^{5}I_{6}$ lifetime, and decrease of both the ${}^{4}I_{13/2}$ and ${}^{5}I_{7}$ lifetimes.

The four wavelengths at which (30% Er, 1.5% Ho):YAG lased were measured to be 2938.6, 2936.2, 2795.6, and 2765.5 nm. These wavelengths all correlate to transitions in Er^{3+} ; none corresponds to a transition in Ho³⁺. The 2938.6 nm transition appears to be $A_2 \rightarrow Y_7$ (predicted to be 2938.8 nm), the 2936.2 nm to $A_2 \rightarrow Y_6$ (predicted at 2936.4 nm), the 2795.6 nm transition to $A_3 \rightarrow Y_4$ (predicted to be 2794.9 nm), and the 2765.5 nm to $A_6 \rightarrow Y_5$ (predicted wavelength of 2766.6 nm). It is interesting to note that the terminal sublevels of the 2.8-µm lasing are higher in the Stark structure of the ⁴I_{13/2} level, which would be expected of a redshift. However, the slope efficiency of the 2.8-µm lasing is greater than that of the 2.94-µm lasing, indicating that the 2.8 µm transitions are stronger than those at 2.94 µm) is all that prevents these lines from lasing in Er:YAG. In that

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case, the Ho^{3+} ESA, preventing lasing at 2.94 μ m, allows the Er^{3+} ions to lase at their preferred wavelengths.

References—Chapter 3

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Chapter 4—Results and Conclusions

4.1 Absorption Spectroscopy

4.1.1 Singly-Doped Materials

All the absorption features of Er; YAG and Ho: YAG are identified (see Figs. 2.2 and 2.3) and correlated to known energy levels of these crystals. The absorption features are very strong, due not only to the innate strengths of the absorption transitions, but also to the high concentrations of Er^{3+} and Ho^{3+} ions present. Most absorption spectra are taken at low concentrations to avoid potential difficulty in separating features, which may occur if absorption features saturate the measurement device. The measurements reported herein do not rely on saturated measuring devices; all the absorption features were narrow enough to be resolved.

4.1.2 Multiply-Doped Materials

The absorption features of the multiply-doped materials approximate those which would be seen if the absorption graph of its component singly-doped materials were overlaid. Comparing Figs. 2.2, 2.3, and 2.4, it is apparent that the absorptivity of (30% Er, 1.5% Ho):YAG is what would be seen if Fig. 2.3, the absorptivity of 15% Ho:YAG, were corrected for the Ho³⁺ concentration difference and overlaid on Fig. 2.2, the absorptivity of 30% Er:YAG. Co-doping Er^{3+} or Ho³⁺ ions in a YAG crystal does not result in a sufficient change to the crystal structure or crystal field to cause a noticeable change in the absorption spectrum.

4.2 Emission Spectroscopy

In Er: YAG, the laser level lifetimes are strongly non-exponential. This validates the claim that like ion cross-relaxation is very strong in highly-doped Er: YAG. The inter-

actions among the ions may be described in either of two ways: by defining a crossrelaxation parameter or by defining an "effective lifetime" for each level. Both methods are discussed in Section 3.6. It is actually easiest to describe the "effective lifetime" in terms of the cross-relaxation parameter.

If the cross-relaxation is described in terms of the "effective lifetime," the parameters $\beta(t)$ and $\gamma(t)$ appear in this term. The model for these parameters (see Section 3.6), assuming a 28 mJ pump pulse, indicates that an exponential fit to the ⁴I_{13/2} level decay time would vary over a factor of approximately five in the first few hundred μ s, then change only slowly, while the fit to the ⁴I_{11/2} level decay time would have a large change between 100 and 500 μ s, then stay fairly constant. This is, in fact, what is seen in the fluorescence experiment (Figs. 2.6 and 2.7). It should be noted, however, that this modeling of "effective lifetimes" assumes that the emitting ions never reach steady state conditions, as they do during lasing.

In Ho:YAG, the laser level lifetimes are also non-exponential. Since the emission wavelengths are identical with strong absorption wavelengths in this crystal, it is possible that this is caused by self-trapping, which increases the apparent lifetimes at long observation times by reabsorbing and reemitting the fluorescence several times. This does not seem the likeliest explanation, however, as the experiments were designed to minimize self-trapping in all cases. The prediction of self-trapping due to reabsorption is also valid for Nd:YAG and Er:YAG, yet the measured decay of Nd:YAG is purely exponential, and that of Er:YAG is fully explained by accepted cross-relaxation theory. It is more likely that the non-exponential decay of Ho:YAG is caused by interactions amongst the Ho³⁺ ions.

The lifetimes of (Er, Nd):YAG and (Ho, Nd):YAG indicate very strong interactions amongst the unlike ions. In both cases the lifetime of the Nd^{3+ 4} $F_{3/2}$ level was greatly

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reduced, as was the lower laser level lifetime in the other ion. The huge reductions in the Nd^{3+ 4}F_{3/2} level lifetime without corresponding reductions in the $Er^{3+} I_{11/2}$ or Ho³⁺ ${}^{5}I_{6}$ lifetimes indicates that energy is being transferred from the Nd³⁺ to the Er^{3+} and Ho³⁺ ions. This is also apparent from the decrease in efficiency when the 1.06-µm lasing of (Er, Nd): YAG is compared to that of Nd: YAG. However, there is no increase in the efficiency of the 3-µm lasing in (Er, Nd): YAG, indicating that the energy transfer to the Er^{3+} ${}^{4}I_{11/2}$ does not overcome the loss of cross-relaxation pumping caused by the decrease in the lifetime of the ${}^{4}I_{13/2}$ level.

Lifetimes of the laser levels of (Er, Ho): YAG indicate moderate interactions amongst the unlike ions. There are no huge changes in level lifetimes, as in the Nd³⁺ co-doping experiments, but all decays from the laser levels of (Er, Ho): YAG can be described by single exponentials.

The lifetime of the $\text{Er}^{3+4}I_{11/2}$ is reduced, indicating that energy is being transferred from this level into the Ho³⁺ ions. Likewise, the lifetime of the Ho³⁺⁵I₆ level is increased to approximate that of the co-doped ${}^{4}I_{11/2}$, indicating that the ${}^{5}I_{6}$ is the level to which the ${}^{4}I_{11/2}$ is transferring its energy.

The $\operatorname{Er}^{3+4}I_{13/2}$ level decay, strongly non-exponential in Er:YAG, has been made exponential in (Er, Ho):YAG. This indicates that there is significant energy transfer from this level to the ${}^{5}I_{7}$ level of the Ho³⁺ ions. There is, however, no increase in the lifetime of the ${}^{5}I_{7}$ level, perhaps because the lifetimes of the two levels are fairly close in singlydoped materials. The fact that cross-relaxation is completely suspended by the addition of such a small concentration (1.5%) of Ho³⁺ ions to Er:YAG may seem to indicate that the $\operatorname{Er}^{3+4}I_{13/2} \rightarrow \operatorname{Ho}^{3+5}I_{7}$ energy transfer is strongly preferred over the ${}^{4}I_{13/2}$ ion crossrelaxation. On the other hand, energy transfer probabilities are proportional to the product of populations in two levels: for the cross-relaxation, the product is the square of the population in the ${}^{4}I_{13/2}$ level, while for the inter-ion energy transfer, it is the product of this level with that in the ground level of the Ho³⁺ ions. Since the ground-state population of 1.5% Ho³⁺ ions in YAG will be approximately 2.1×10^{20} cm⁻³, while the population of the ${}^{4}I_{13/2}$ level of Er³⁺ was approximately 2.3×10^{19} cm⁻³ in these experiments, the unlike ion energy transfer does not need to be stronger than, and may even be weaker than, the cross-relaxation. In fact, because the initial population in the final state (the Ho^{3+ 5}I₇) is zero, the unlike ion energy transfer parameter may be much lower than the cross-relaxation parameter.

4.3 Lasing Experiments

4.3.1 Er:YAG

Er: YAG is an efficient solid-state laser operating at 3 μ m. Its slope efficiency of 1.8% and overall efficiency of 1.6% compare favorably with other solid-state lasers such as ruby and alexandrite. When the energy defect (the ratio between the energy of a laser photon and a pump photon) is taken into account, its overall efficiency of 0.14% in single lowest order mode is not too far below that of alexandrite and Nd: YAG (both around 1%, or 0.26% corrected for photon energy).

Cross-relaxation is the pumping method which allows Er: YAG to lase even though its lower level lifetime is longer than its upper level lifetime. If Er: YAG did not have the strong cross-relaxation from its lower laser level, it would be an inefficient, self-terminated laser like Ho: YAG. In high-concentration Er: YAG, a large population may build up in the lower laser level because of its long lifetime. Since the cross-relaxation probability is proportional to the square of the population in the lower laser level, this allows efficient cross-relaxation pumping. This method of pumping allows Er: YAG to be such an efficient laser. Development of semiconductor lasers at 0.98 μ m suggests the possibility of using these lasers to pump Er: YAG laser. Fig. 2.1c indicates that the ${}^{4}I_{11/2}$ level absorbs from 0.96—1.0 μ m. Since this is the upper laser level for the 3- μ m transition, such a pumping scheme could be very efficient.

4.3.2 Ho:YAG

Ho:YAG is not an efficient 3-µm laser. It fits the classical model of a laser with a blocked transition due to a lower laser level lifetime longer than that of the upper level. Without some way of depopulating the lower level, Ho:YAG cannot be made into an efficient laser on this transition.

To become useful, it is necessary to co-dope Ho:YAG. The (Ho, Nd):YAG experiments indicate that it is possible to increase the 3- μ m lasing efficiency and unblock the laser transition by using Nd³⁺ as a deactivator.

4.3.3 (Er, Nd):YAG

Co-doping Er:YAG with Nd³⁺ to improve the efficiency of 3- μ m lasing was not successful. The lifetime of the lower laser level of the Er³⁺ ions was reduced drastically, but not to the point of being shorter than the upper level lifetime. The reduction in the lifetime of the ⁴I_{13/2} level resulted mainly in a reduction in cross-relaxation pumping of the 3- μ m laser transition. This reduced the efficiency of 3- μ m lasing considerably.

4.3.4 (Ho, Nd):YAG

Co-doping Ho:YAG with Nd^{3+} to improve the efficiency of 3-µm lasing met with limited success. The lower level lifetime was shorter, which increased the lasing efficiency on the Ho³⁺ 3-µm transition.

The 3- μ m lasing efficiency of (Ho, Nd): YAG is higher than that of Ho: YAG. This is due not only to the energy transfer to the ${}^{5}I_{6}$ level of the Ho³⁺ ion, but also to the reduction in the ${}^{5}I_{7}$ level lifetime (from 5.5 ms to 170 μ s). Unlike Er: YAG, Ho: YAG is not a cross-relaxation pumped laser. The long lifetime of the ${}^{5}I_{7}$ level decreases the efficiency of the Ho: YAG laser by causing self-termination of lasing. This self-termination appears to be reduced, if not eliminated, in (Ho, Nd): YAG.

The (Ho, Nd): YAG laser exhibits interesting 1- μ m lasing behavior. Although not along the main line of study of this dissertation, which is the 3 μ m lasing transitions, the 1- μ m lasing behavior of (Ho, Nd): YAG deserves mention. In YAG, Nd³⁺ lases preferentially at 1.06 μ m. If care is taken to prevent lasing at this wavelength, Nd: YAG will lase at approximately 1.34 μ m. It seems that the residual absorption of Ho³⁺ ions at 1.06 μ m is sufficient to prevent 1.06- μ m lasing in (Ho, Nd): YAG, allowing lasing at 1.34 μ m. This is only true for fairly low pump levels. If the pump level is increased, and the mirrors used allow it, lasing occurs at both 1.06 and 1.34 μ m within the same pump pulse. This is not truly simultaneous lasing, however. For a rectified sinusoid pump pulse, the lasing begins at 1.34 μ m, then changes to 1.06 μ m, and changes back to 1.34 μ m as the pump rate decreases on the falling edge of the pump pulse. During the transitions between the two lasing wavelengths, relaxation oscillation spikes of both are seen, but not quite simultaneously.

4.3.5 (Er, Ho):YAG

The lasing behavior of (Er, Ho): YAG was unique. This was not a particularly efficient 3- μ m laser; its efficiency was midway between those of Er: YAG and Ho: YAG. However, it lased at four wavelengths during a single pump pulse; at 2.939 and 2.936 μ m simultaneously near the beginning of the pump, then at 2.796 and 2.766 μ m simultaneously later in the pulse.

The 2.94- μ m lasing of (Er, Ho): YAG showed definite self-termination. Unlike other self-terminated lasers, such as Ho: YAG, (Er, Ho): YAG showed a lasing blueshift. The 2.939 and 2.936 μ m laser lines appeared first, terminated, then the 2.796 and 2.766 μ m laser lines appeared. These latter two lines did not appear to be self-terminated, indicating that they were pumped by cross-relaxation. The self-termination of the first two laser lines can be attributed to ESA from the ⁵I₇ to the ⁵I₆ level of Ho³⁺, which has a transition at 2.94 μ m. This ESA confirms that there is energy transfer from the ⁴I_{13/2} level of Er³⁺ to the ⁵I₇ level of Ho³⁺; this is how the ⁵I₇ level acquires sufficient population for the ESA.

4.4 Determination of Er³⁺ Energy Levels

The exact energies of the Stark sublevels of laser levels can be important in determining at what wavelengths the laser is useful. These energies are also useful for predicting which levels of any ion will react with other levels, of the same or different ions, and how strong the interactions will be.

The energies reported here are determined by starting from the work of Agladze *et al.*¹ With this basis, it was easy to identify all the Stark sublevels of the ${}^{4}I_{11/2}$ level, and six of the seven of the ${}^{4}I_{13/2}$ level, by comparing their results with the absorption spectroscopy of this dissertation. All the level energies they reported corresponded with detected absorption peaks.

The only level whose energy was in debate was the Y6, the second-highest sublevel of the ${}^{4}I_{13/2}$ level. Agladze *et al.* did not report a value for this level, and it appeared that absorption features into other ${}^{4}I_{13/2}$ sublevels overlaid all the absorptions into the Y6.

The data from lasing wavelengths was also taken into account. Calculations showed that a transition between the A2 and Y6 sublevels would be at 2936.4 μ m if the Y6 level were at 6873.8 cm⁻¹, at which all absorption features into the Y6 would be hidden by other features. It is reasonable to assume, then, that the Y6 sublevel has an energy of 6873.8 cm⁻¹ over the ground state.

The energies of each sublevel, as determined by absorption and laser spectroscopy, are shown in Table 4.1.

sublevel	1	2	3	4	5	6	7	8
⁴ I _{15/2} (X)	0.0	18.5	58.6	75.3	410.3	423.2	520.8	565.5
⁴ I _{13/2} (Y)	6543.5	6593.5	6600.5	6777.2	6796.4	6873.8	6876.6	
⁴ I _{11/2} (A)	10249.2	10279.3	10355.1	10364.5	10406.1	10410.9		

Table 4.1. Energies (in cm^{-1}) of the Stark sublevels of Er:YAG.

References—Chapter 4

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Chapter 5—Determination of the Er→Ho Energy Transfer Coefficient

5.1 Introduction

Since the first report in 1974,¹ Er: YAG has come to be the dominant 3-µm solid-state laser. Er: YAG can have a slope efficiency as high as 1.8%.² Unfortunately, its lasing threshold pump energy is typically 50 times greater than that of Nd: YAG. Since this pump energy must be supplied for each pulse, Er: YAG is inefficient at high pulse repetition frequencies and at low output energies.

The high lasing threshold of the Er:YAG 3-µm laser is the result of the laser level lifetimes. The ratio of the lifetime of the upper laser level to that of the lower laser level, which is much greater than 1 for efficient four-level lasers such as Nd:YAG, is typically between 0.01 and 0.02 in Er:YAG. The low value of this ratio prompted the search for a co-dopant which would increase it.³

As part of the study of co-dopants aimed at improving the lifetime ratio of Er^{3+} in YAG, YAG doped with 40 at. % Er^{3+} and 1.5 at. % Ho³⁺ ions was studied. The lifetime ratio was not significantly improved, however, because the energy transfer out of the Er^{3+} lower laser level was relatively weak (the Er—Ho transfer coefficient, W_{EH} , was 500 times less than the equivalent Er—Er cross-relaxation coefficient). Nonetheless, calculations show that this energy transfer is strong enough to allow a room-temperature (Er, Ho): YAG 2-µm laser. Additionally, it is likely that this energy transfer is strong enough in other hosts, such as YLF, to improve the efficiency of the Er^{3+} 3-µm laser.

5.2 Background

Because the lifetime of the lower level is longer than that of the upper level in the Er:YAG 3-µm laser transition, one might expect this transition to be self-terminated. (In

a self-terminated laser, lasing ends because of filling of the lower laser level, and increasing the length of the pump pulse does not increase the length of the output pulse.) High dopant density Er:YAG, however, does not self-terminate. The self-termination is defeated by cross-relaxation, in which two ions in the long-lived lower laser level (the ${}^{4}I_{13/2}$) interact (Fig. 5.1). One ion drops to the ground state (the ${}^{4}I_{15/2}$), transferring its energy to the other, which is excited into the pump band (the ${}^{4}I_{9/2}$). Cross-relaxation is especially powerful in Er:YAG because it is resonant. That is, the energy given up by one ion in the ${}^{4}I_{13/2}$ state in dropping to the ${}^{4}I_{15/2}$ ground state is exactly that needed to excite another ion from the ${}^{4}I_{13/2}$ state into the ${}^{4}I_{9/2}$ state in the pump band. Such a transfer does not require emission or absorption of a phonon, as it would if it were non-resonant.



Figure 5.1. Energy levels and cross-relaxation process of Er^{3+} ions in Er:YAG. Arrows indicate resonant energy transfer processes.

Another reason this process is so efficient in Er:YAG is that the cross-relaxation process causes a net inversion gain of three ions. Two ions in the lower laser level interact, removing one and pumping the other. The pumped ion then decays quickly into the upper laser level. Since two ions leave the lower laser level and one reaches the upper laser level, the net gain in the inversion caused by the cross-relaxation process is three.

It is possible to describe the Er:YAG 3-µm laser with rate equations. These equations are similar to those used to model ordinary four-level lasers. The difference between the Er:YAG rate equation model and that of a typical four-level laser is the inclusion of cross-relaxation. Ignoring pump terms,

$$\frac{dn_3}{dt} = \frac{1}{2} W_{CR} n_1^2 - \frac{n_3}{\tau_3}$$
(5.1)

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_2}$$
(5.2)

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = \frac{n_2}{\tau_{21}} - \frac{n_1}{\tau_{10}} - W_{\mathrm{CR}} n_1^2$$
(5.3)

$$\frac{\mathrm{d}n_0}{\mathrm{d}t} = \frac{n_1}{\tau_{10}} + \frac{1}{2} W_{\mathrm{CR}} n_1^2 \,. \tag{5.4}$$

In these equations, n_i represents the population density (cm⁻³) of ions in the "i" Er³⁺ state. The population in the ground state, the ${}^{4}I_{15/2}$, is n_0 ; that in the ${}^{4}I_{13/2}$ is n_1 ; n_2 is the population of the ${}^{4}I_{11/2}$ level; and n_3 is the population in the ${}^{4}I_{9/2}$. The τ_i include radiative and non-radiative decay. Since each Er³⁺ level is strongly coupled to the next level below it in energy, ${}^{4}\tau_3$ (eq. 5.1) may be replaced by τ_{32} and τ_2 (eq. 5.2) by τ_{21} as a reasonable experimental approximation.

For doping levels around 40%, the concentration used in these experiments, the lifetimes of the relevant Er^{3+} levels are

$$\tau_{32} \ll 1 \ \mu s$$
 (5.5)

$$\tau_{21} \approx 120 \ \mu s$$
 (5.6)

$$\tau_{10} \approx 7.1 \text{ ms.}$$
 (5.7)

Values of τ_{21} and τ_{10} were measured directly; the value quoted for τ_{32} was obtained from Ref. 5.4. The measured values were obtained from the crystal used in the experiments reported herein. The lifetime of the ${}^{4}I_{13/2}$ level could be increased by self-trapping of the fluorescent radiation. This effect does not appreciably affect the conclusions deduced from these measurements.

The cross-relaxation constant may be calculated using Ref. 5.5:

$$W_{CR} \neq 4.8 \times 10^{-17} \text{ cm}^3/\text{s.}$$
 (5.8)

 $W_{CR}n_1$ is thus the rate of cross-relaxation out of the ${}^4I_{13/2}$ level. $W_{CR}n_1^2$ is the number of ions per unit time per unit volume at which this level is being depopulated by cross-relaxation.

Two assumptions are made in the rate equations (eqs. 5.1–5.4). First, it is assumed that stimulated emission can be neglected. This is justified because of the low level of excitation and the lack of the resonant structure required by lasing. Second, the rate equations are assumed to be valid only after the pump is over, with its effects seen as initial conditions on n_1 , n_2 , and n_3 . This second assumption is valid because these experiments used a short (~20 ns) pump pulse, which ended before the start of fluorescence data acquisition.

After the pump pulse, a large population density collects in level 1 (the ${}^{4}I_{13/2}$) because it has by far the longest lifetime of any relevant level. The ions in level 1 leave it by two main processes: decay (radiative and non-radiative, rate $1/\tau_{10}$) and cross-relaxation (rate $W_{CR}n_{1}$). If the cross-relaxation rate is at appreciable compared to the rate of decay, the fluorescence of the ${}^{4}I_{13/2}$ level is non-exponential. In addition, the cross-relaxation pumps level 2 (the ${}^{4}I_{11/2}$) through level 3; this pumping makes the fluorescence from the ${}^{4}I_{11/2}$ level also non-exponential. Non-exponential fluorescence may be described by effective lifetimes, derived from eqs. 5.2 and 5.3:

$$\tau_1^{\text{eff}} = \frac{\tau_{10}}{1 + \beta(t)}$$
(5.9a)

$$\beta(t) \equiv W_{cr} \tau_{10} n_1(t)$$
(5.9b)

$$\tau_2^{\rm eff} = \frac{\tau_{21}}{1 - \gamma(t)}$$
(5.10a)

$$\gamma(t) = \frac{1}{2} W_{cr} \tau_{21} \frac{n_1^2(t)}{n_2(t)} \,. \tag{5.10b}$$

Calculated values of $\beta(t)$ and $\gamma(t)$, using the constants from eqs. 5.5–5.8, are shown in Fig. 5.2.



Figure 5.2. Modeled time evolution of the lifetime functions $\beta(t)$ and $\gamma(t)$. Pump is 28 mJ in 20 ns at 532 nm.

Even if self-termination of a laser transition is not defeated by cross-relaxation, there are methods of making the transition more efficient. One such method is to remove the self-termination by co-doping. A co-dopant ion can receive energy from the laser ion, which reduces the laser ion's effective lifetime. This method was used in (Er, Nd):YAG, in which the ⁴I_{13/2} level of Er³⁺ transferred energy to the ⁴I_{15/2} level of Nd³⁺ (Fig. 5.3). The Er³⁺—Nd³⁺ interaction reduced the ⁴I_{13/2} level lifetime of 15% Er:YAG by a factor of nearly 100 in (15% Er, 1% Nd):YAG, although other interactions between the Er³⁺ and Nd³⁺ ions reduced the Er³⁺ ⁴I_{11/2} lifetime enough that it was shorter than the ⁴I_{13/2} lifetime,³ like it was in Er:YAG.





Figure 5.3. Energy levels and energy transfer processes between Er^{3+} and Nd^{3+} ions in (Er, Nd):YAG. Arrows represent resonant energy transfer.

5.3 Experimental Description

In order to determine the energy transfer coefficients, fluorescence was studied from crystals of (40% Er):YAG and (40% Er, 1.5% Ho):YAG. These crystals were roughly wedge-shaped, 15 mm on a side. As shown in Fig. 5.4, sample crystals were excited with a repetitively pulsed, Q-switched Nd:YAG laser frequency-doubled to 532 nm. The laser pulse length was much shorter than the lifetime of any relevant level. The laser was focused to a 2-mm diameter spot on the surface of the sample crystal with the beam position adjusted so that the filament pumped was along the exit face of the crystal. This increased the signal at the detector, but did not eliminate self-trapping. Fluorescence from the crystal was captured by a lens, and relayed to the entrance slit of a 0.27-m focal length monochromator. The monochromator selected the wavelength observed by a LN_2 -



Figure 5.4. Layout of fluorescence study system.

cooled InSb detector. The output of the detector was digitized by a digital processing oscilloscope (DPO). Many signals were averaged by the computer to increase the signal-to-noise ratio (SNR). Up to 400 separate signals could be averaged, resulting in an SNR improvement of a factor of 20. The computer plotted this averaged signal on semilog axes, so that an exponential fluorescence decay appeared as a straight line.

The limiting factor in taking these measurements was the equipment used. The DPO could not digitize a fluorescence signal in a single pass, but required up to 10 input signals for a single stored signal. The pump laser had a problem with power drift; it maintained roughly the same energy per pulse for 20 minutes, but then began to change. Finally, the computer used was relatively primitive, and did not always download the data correctly from the DPO. These factors often limited the number of signals available to average, and the precision of these signals, which is reflected in the estimated errors of the measured lifetimes.

The computer also provided a linear least-squares fit to whatever portion of the data desired, which was used to determine effective lifetimes for the different levels. The fit was usually chosen near the end of the ${}^{4}I_{13/2}$ level decay and near the beginning of the ${}^{4}I_{11/2}$ level decay. Since the cross-relaxation was from the ${}^{4}I_{13/2}$ into the ${}^{4}I_{11/2}$ level, these fits correspond to measuring the lifetime of the ${}^{4}I_{13/2}$ level after cross-relaxation has stopped reducing it and measuring the lifetime of the ${}^{4}I_{11/2}$ level before cross-relaxation has started increasing it.

For the experiments reported in this dissertation, the laser was run at a pulse repetition rate of 10 Hz. Its pulse length was approximately 20 ns and the energy per pulse was 30 mJ. Fresnel reflections at the surface of the Er:YAG and (Er, Ho):YAG crystals were about 7%, so the total energy entering the crystal was 28 mJ. The absorptivity of either the (40% Er):YAG or the (40% Er, 1.5% Ho):YAG was about 9.5 cm⁻¹ at 532 nm, and the effective pumped volume was roughly 3.3×10^{-3} cm³. The pumped energy density was 8.5 J/cm³, corresponding to 2.3×10^{19} absorbed photons per cm³.

The value of 2.3×10^{19} cm⁻³ was used as an initial condition for n₂ in a computer model. (This implies that virtually all the ions excited by the pump laser decay rapidly into the ⁴I_{11/2} level.) The computer model was based on simple rate equations, with a linear integration system. For example, for Er:YAG the model used the rate equations (5.1–5.4). The integration routine applied

$$n_i(t' + \Delta t) = n_i(t') + \Delta t \cdot \frac{dn_i}{dt}\Big|_{t = t'}.$$
(5.11)

Although this integration routine may not have been as accurate as the Runge-Kutta-Verner method used by Shi *et al.*⁵ it was sufficient for these experiments.

5.4 Results and Discussion

Fluorescence is reported from the ${}^{4}I_{11/2}$ and ${}^{4}I_{13/2}$ levels of Er^{3+} in Er:YAG, and from the ${}^{4}I_{13/2}$ level of Er^{3+} in (Er, Ho):YAG. The fluorescence from these levels is compared with that predicted by our simple rate equation model in Figs. 5.5–5.7. Figs. 5.5 and 5.6, showing calculated and measured fluorescence from the ${}^{4}I_{13/2}$ and ${}^{4}I_{11/2}$ levels of Er^{3+} in Er:YAG, demonstrate the validity of the model. Fig. 5.7 compares fluorescence from the Er^{3+} ${}^{4}I_{13/2}$ level in (Er, Ho):YAG with that predicted by the computer model using $W_{EH} = 9.5 \times 10^{-20} \text{ cm}^{3}/\text{s}$.

For Er:YAG the model used the rate equations from eqs. 5.1–5.4; for (Er, Ho):YAG the rate equations used in the model were:

$$\frac{dn_3}{dt} = \frac{1}{2} W_{CR} n_1^2 - \frac{n_3}{\tau_{32}}$$
(5.12)

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}}$$
(5.13)

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_{21}} - \frac{n_1}{\tau_{10}} - W_{CR} n_1^2 - W_{EH} n_1 n_a$$
(5.14)

$$\frac{dn_0}{dt} = \frac{n_1}{\tau_{10}} + \frac{1}{2} W_{CR} n_1^2 + W_{EH} n_1 n_a$$
(5.15)

$$\frac{\mathrm{d}\mathbf{n}_{b}}{\mathrm{d}t} = \mathbf{W}_{\mathrm{EH}}\mathbf{n}_{1}\mathbf{n}_{a} - \frac{\mathbf{n}_{b}}{\tau_{b}} \tag{5.16}$$

$$\frac{\mathrm{d}n_{a}}{\mathrm{d}t} = \frac{n_{b}}{\tau_{b}} - W_{\mathrm{EH}}n_{1}n_{a} \,. \tag{5.17}$$

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Figure 5.5. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in 40% Er:YAG. Solid curve is measured, dashed curve is modeled, and straight line is least squares fit to late decay.



Figure 5.6. Fluorescence from the ${}^{4}I_{_{III}}$ level of Er^{3+} in 40% Er:YAG. Solid curve is measured, dashed curve is modeled, and straight line is least squares fit to early decay.



Figure 5.7. Fluorescence from the ${}^{4}I_{13/2}$ level of Er^{3+} in (40% Er, 1.5% Ho):YAG. Solid curve is measured; dashed curve is modeled with $W_{EH} = 9.5 \times 10^{-20} \text{ cm}^{3}/\text{s}$.

In these equations, the numbered subscripts indicate Er^{3+} levels and the lettered subscripts indicate Ho³⁺ levels. The Ho³⁺ levels used in the model with population densities n_a and n_b were the ⁵I₈ and ⁵I₇ levels, respectively. The ⁵I₈ is the ground level and the ⁵I₇ is the first excited state. W_{EH} is the coefficient of energy transfer from the ⁴I_{13/2} level of Er³⁺ to the ⁵I₇ level of Ho³⁺ (see Fig. 5.8).

The lifetime of the Ho^{3+ 5}I₇ level has been measured⁶ at $\tau_b \approx 4.8$ ms for dopant densities near the 1.5% used in these experiments. The initial population density in the ground level is equal to 1.5% of the density of Y³⁺ ions in undoped YAG, or 2.07 × 10²⁰ cm³. During our experiment the value of n_a did not change significantly; 28 mJ of pump at 532 nm was insufficient to bleach the crystal.

Eq. 5.14 suggests that $W_{EH}n_a$ is the energy transfer rate (the inverse of the characteristic energy transfer time) from the $Er^{3+4}I_{13/2}$ to the Ho³⁺⁵I₇ level. Since n_a remains approximately constant, this rate will also be constant. One can obtain an approximate



Figure 5.8. Lower energy levels and energy transfer from Er^{3+} to Ho^{3+} ions in (Er, Ho):YAG. Arrows indicate non-resonant energy transfer process; curved line represents generation of a phonon for energy match.

value for W_{EH} by assuming that it is the only effect changing the effective lifetime of the ${}^{4}I_{13/2}$ level. Ignoring cross-relaxation and pumping, the decay of the $Er^{3+} {}^{4}I_{13/2}$ level in (Er, Ho): YAG is described by the equation

$$\frac{dn_1}{dt} = -\frac{n_1}{\tau_{10}} - W_{\rm EH} n_1 n_a \,. \tag{5.18}$$

In (40% Er): YAG, where $n_a = 0$, the exponential decay time of this level is 7.1 ms. With the addition of 1.5% Ho, this time becomes 5.5 ms. The effective decay time can be calculated from eq. 18 as

$$\tau_1^{\text{eff}} = \frac{\tau_{10}}{1 + W_{\text{EH}} n_a \tau_{10}},$$
(5.19)

which can be inverted to yield

$$W_{\rm EH} = \frac{\tau_{10} - \tau_1^{\rm eff}}{n_{\rm a} \tau_{10} \tau_1^{\rm eff}} \,. \tag{5.20}$$

Using the measured values of $\tau_{10} = 7.1$ ms and $\tau_1^{\text{eff}} = 5.5$ ms in eq. 5.20 one obtains the value of W_{pH} as 2.0×10^{-19} cm³/s.

When the value $W_{EH} = 2.0 \times 10^{-19} \text{ cm}^3/\text{s}$ is used in the computer model, which includes cross-relaxation, the predicted fluorescence decays significantly faster than the measured fluorescence. The cross-relaxation also depopulates the ${}^{4}I_{13/2}$ level, increasing the decay speed. Because the computer model includes cross-relaxation, it provides a more accurate comparison. Fig. 5.7 shows the accuracy of the computer model using the value $W_{EH} = 9.5 \times 10^{-20} \text{ cm}^3/\text{s}$. This value for W_{EH} provided the best overall match to the measured decay curve. When we increased value of W_{EH} in our computer model by 10^{-20} cm³/s, the predicted decay was noticeably faster than that measured; if W_{EH} was decreased by the same amount, the shape of the curve no longer matched the measured decay. We therefore conclude that the Er^{3+ 4}I_{13/2} to Ho^{3+ 5}I₇ energy transfer coefficient is

$$W_{\rm EH} = 9.5 \pm 1.0 \times 10^{-20} \,\rm cm^3/s.$$
 (5.21)

Using the above value for W_{EH} , the energy transfer efficiency as defined by Rubin *et al.*⁷ may be calculated as

$$\eta_{\tau_1} \equiv 1 - \frac{\tau_1^{\text{eff}}}{\tau_{10}} = 23\%.$$
 (5.22)

Rubin's efficiency is related to the energy transfer coefficient by

$$\eta_{\tau_1} = \frac{W_{EH} n_a \tau_{10}}{1 + W_{EH} n_a \tau_{10}},$$
(5.23)

which is independent of Er^{3+} concentration, or, alternatively, by

$$W_{\rm EH} = \frac{\eta_{\tau_1}}{(1 - \eta_{\tau_1}) n_{\rm a} \tau_{10}} \,. \tag{5.24}$$

However, these equations still ignore cross-relaxation; eqs. 5.23 and 5.24 again suggest

the value of 2.0×10^{-19} cm³/s for W_{EH}. Eq. 5.23 provides an energy transfer efficiency of 12%, implying that, in these experiments, approximately half the energy transferred out of the ${}^{4}I_{13/2}$ level of the Er³⁺ ions went to the ${}^{5}I_{7}$ of the Ho³⁺ ions, while the other half went to cross-relaxation.

In addition to helping predict the efficiency of $\text{Er}^{3+} \rightarrow \text{Ho}^{3+}$ energy transfer, the measured value of W_{EH} allows definition of requirements for improved laser efficiency. First, looking at the Er^{3+} 3-µm laser, we see that to reduce the lifetime of the ${}^{4}I_{13/2}$ level to less than that of the ${}^{4}I_{11/2}$, we must have

$$\frac{1}{\tau_{21}} < W_{\rm EH} n_{\rm a} \,.$$
 (5.25)

Given the values of 120 µs for τ_{21} and 9.5×10^{-20} cm³/s for W_{EH}, eq. 5.25 implies

$$n_a > \frac{1}{\tau_{21} W_{EH}} = 8.8 \times 10^{22} \,\mathrm{cm}^3$$
. (5.26)

Since this density is greater than that found for 100% doping, it is impossible to use Ho^{3+} to completely deactivate the lower level of the Er:YAG 3-µm laser.

This conclusion may not hold for other Er^{3+} lasers, such as Er:YLF. The work of Rubin *et al.*⁷ indicates that the value of W_{EH} is 75 times greater in YLF than YAG. They report on energy transfer from the ${}^{4}I_{13/2}$ level of Er^{3+} to the ${}^{5}I_{7}$ level of Ho³⁺ in YLF using the transfer efficiency in eq. 5.24. Their results, and calculations based on these results, are listed in Table 5.1.

	(50% Er, 0.5% Ho):YLF	(50% Er, 2% Ho):YLF	(50% Er, 5% Ho):YLF
η _m	75%	93%	97%
n	$7.00 imes 10^{19} \mathrm{cm}^{-3}$	$2.80 \times 10^{20} \mathrm{~cm^{-3}}$	$7.00 imes 10^{20} \mathrm{cm}^{-3}$
W _{EH}	$7.15\pm0.39\times10^{-18}$ cm ³ /s	$7.91\pm0.71 \times 10^{-18} \text{ cm}^3/\text{s}$	$5.72\pm2.0\times10^{-18}\mathrm{cm}^3/\mathrm{s}$

Table 5.1. Energy transfer coefficients for (Er, Ho):YLF.

The error ranges listed are the results of applying eq. 5.24 to Rubin's values with the published precision. The error-weighted average of the calculated values for W_{EH} in YLF suggest a value of

$$W_{_{\rm FH}} = 7.23 \pm 0.67 \times 10^{-18} \,{\rm cm}^3/{\rm s}$$
 (5.27)

for (Er, Ho):YLF. Plugging this value, and the reported value⁷ of 3 ms for $\tau_{21}^{}$, into eq. 5.25, it appears that the effective lifetime of the ${}^{4}I_{13/2}^{}$ level of Er:YLF can be reduced to less than that of the singly-doped upper (${}^{4}I_{11/2}^{}$) level with addition of 4.6×10^{19} cm⁻³ of Ho³⁺, corresponding to 0.33% doping. This addition of Ho³⁺, however, also reduces the lifetime of the upper level. Nonetheless, it is apparent that the strong Er³⁺ \rightarrow Ho³⁺ energy transfer provides a four-level Er:YLF laser. Rubin's work shows that the lower level has a shorter lifetime than the upper level for Ho³⁺ doping concentrations greater than 0.5%.

Co-doping YAG with Er^{3+} and Ho^{3+} ions also affects the Ho^{3+} 2-µm laser. To determine this effect the sensitization of the ${}^{5}\text{I}_{7}$ level of Ho^{3+} is calculated. The $\text{Er}^{3+} \rightarrow \text{Ho}^{3+}$ energy transfer pump rate is $W_{\text{EH}}n_{1}$, so the maximum density of ions in the ${}^{5}\text{I}_{7}$ level is $n_{b} = W_{\text{EH}}n_{1}n_{a}\tau_{b} = 4.6 \times 10^{-22}n_{1}n_{a} \text{ cm}^{-3}.$ (5.28)

For the laser to achieve inversion at room temperature between the lowest Stark sublevel of the ${}^{5}I_{7}$ level and the highest Stark sublevel of the ${}^{5}I_{8}$ level, n_{b}/n_{*} must be at least 10%, which requires

$$n_1 > 2.2 \times 10^{20} \text{ cm}^{-3}$$
. (5.29)

This population density is achievable with flashlamp pumping of an (Er, Ho): YAG laser rod; it is only a factor of 15 higher than the density reached during the fluorescence experiments described in this Chapter. The $\text{Er}^{3+} \rightarrow \text{Ho}^{3+}$ energy transfer may therefore have applications to an (Er, Ho): YAG 2-µm laser.

5.5 Summary and Conclusions

The value of the $Er^{3+4}I_{13/2} \rightarrow Ho^{3+5}I_7$ energy transfer coefficient, W_{EH} , was measured at $9.5 \pm 1.0 \times 10^{-20} \text{ cm}^3$ /s. This coefficient was determined by a process using computer modeling, comparing the fluorescence of the $Er^{3+4}I_{13/2}$ level in (40% Er):YAG to fluorescence from the same level in (40% Er, 1.5% Ho):YAG. The reported value for W_{EH} implies an $Er^{3+} \rightarrow Ho^{3+}$ energy transfer efficiency of about 12% for (40% Er, 1.5% Ho):YAG. Because this value comes from an equation independent of Er^{3+} concentration, the energy transfer efficiency will be about 12% for any concentration near 40% (as long as the Ho³⁺ concentration is 1.5%). Energy transfer, while not sufficiently strong to improve the efficiency of the $Er^{3+} 3$ -µm laser in (Er, Ho):YAG, may be able to improve the $Er^{3+} 3$ -µm laser in (Er, Ho):YLF. It does appear that this transfer is strong enough in YAG to improve the Ho³⁺ 2-µm transition. Calculations indicate that it may be possible to develop an efficient room-temperature 2-µm (Er, Ho):YAG laser as well as an efficient, room-temperature, four-level 3-µm (Er, Ho):YLF laser. The 2-µm laser is especially promising as Er^{3+} has convenient diode-pumpable absorption bands.

References—Chapter 5

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Chapter 6-Recommendations for Future Study

6.1 cw Solid-State Lasers at 3 µm

This dissertation is concerned with the transfer of energy between ions in a laser. One reason for studying this energy transfer is the attempt to unblock some of the 3-µm transitions so that there are commercially-available solid state CW lasers in this wavelength region, presently the domain of the HF chemical laser. (The Er:YLF laser can run CW, but only with laser pumping.)

Selection of crystalline host is critical in developing energy transfer lasers. The precise interaction between the ion and the crystal field can make a major difference in the energy transfer efficiency. For example, Cr^{3+} ions do not transfer energy to Nd³⁺ ions in YAG, but they do in GSGG. The first point in determining which crystal to use is choosing a size match with the active ion. For Er^{3+} and Ho³⁺ the best ion to replace is Y^{3+} . This discussion is therefore limited to crystals based on yttrium.

One of the easiest yttrium-based crystals to grow is lithium yttrium fluoride, or YLF. It is also a crystal in which Er^{3+} has a strong tendency to cross-relax. The interion energy transfer parameters are very large as well. Er^{3+} has longer lifetimes of both the ${}^{4}\text{I}_{13/2}$ and ${}^{4}\text{I}_{11/2}$ levels in YLF than in YAG, and the two levels' lifetimes are also much closer together $[\tau({}^{4}\text{I}_{13/2}) \approx 20 \text{ ms} \text{ and } \tau({}^{4}\text{I}_{11/2}) \approx 10 \text{ ms}]$. It is possible to relax the lower laser level more than the upper laser level by adding Ho³⁺. It should also be possible to relax the lower nultiple-wavelength laser) or Tb³⁺ (which may be more efficient).

Another possibility is to dope Er^{3+} into a crystal with a large crystal field, such as YAlO₃. The larger field allows higher phonon energies and may lead to greater Stark splitting, making an energy match easier to find. A good possibility is to put a large con-

centration of Er^{3+} ions and a small concentration of a deactivator, possibly Nd³⁺, into such a crystal. A good choice for this might be (30% Er, 0.2% Nd):YAlO₃.

There is a chance that Tb^{3+} could relax the ${}^{4}I_{13/2}$ level sufficiently to allow the Er^{3+} ions to lase CW at 3 μ m. Tb^{3+} has no direct absorption at 2.94 μ m, the main Er^{3+} line, and has a tremendous series of closely-packed levels that could easily deactivate the lower level. There is also a good size match between Tb^{3+} and Y^{3+} , which it would replace in (Er, Tb):YAG or (Er, Tb):YLF. It may be, however, that there is sufficient energy level overlap for one of the Tb^{3+} levels to relax the ${}^{4}I_{11/2}$ into the ${}^{4}I_{13/2}$, which would prevent lasing. Nonetheless, the crystal (30% Er, 1% Tb):YAG is probably worth investigating.

It may be that there is a possible CW 3- μ m laser based on Ho³⁺. The work with (Ho, Nd):YAG indicates that it is not difficult to relax the Ho^{3+ 5}I₇ level. One crystal which should be looked into here is one with lower concentrations of Nd³⁺, such as (10% Ho, 0.2% Nd):YAG. It would also be interesting to co-dope Ho³⁺ and Nd³⁺ into a crystal which has Ho^{3+ 5}I₇ and ⁵I₆ lifetimes nearer each other, such as yttrium scandium gallium garnet (YSGG) or YLF.

6.2 Q-Switchable Lasers at 3 µm

In general, a four-level laser makes a good Q-switchable laser. The usual requirements for Q-switching are a strong transition and a lower level lifetime significantly less than the upper level lifetime. All the suggestions made in Section 6.1 about CW laser apply equally well here. One additional point is that the 3 μ m Er³⁺ transitions in YAlO₃ have slight wavelength variations depending on polarization. This implies that it would be possible to tune the laser slightly by using a different Q-switch orientation.

There is a problem in getting Q-switch materials for 3- μ m lasers. Most Q-switches are based on LiNbO₃ in some manner, and it is nearly impossible to obtain this material free of O-H bonds, which absorb in this wavelength region. A good electro-optical Q-

switch could be fabricated from some other material, such as CdTe, but it is difficult to find a material that is transparent at 3 μ m, has a large electro-optic effect, and will accept anti-reflection coatings for 3 μ m.

Another possibility is an acousto-optical Q-switch. Many materials, such as quartz and germanium, are transparent at 3 μ m and are often made into Q-switches. It would be possible to cut the faces of the Q-switch at Brewster's angle to reduce insertion loss. This is probably the line to pursue for developing 3- μ m Q-switches.

6.3 Ho^{3+} Lasers at 2 μm

According to the measurements presented in Chapter 5, moderate pumping of the Er^{3+} ions in (Er, Ho): YAG should be sufficient to create an inversion on the 2-µm Ho³⁺ ${}^{5}I_{7} \rightarrow {}^{5}I_{8}$ transition. Since Er^{3+} has considerable absorption in the spectral region pumped by flashlamps, its addition to Ho: YAG would improve the pumping of this transition. In addition, Er^{3+} has absorption features near the spectral peaks of several commercially available diode lasers. The (Er, Ho): YAG 2-µm laser could be exceptionally efficient when diode pumped.

6.4 Suggested Materials for Energy Transfer Studies

It would be useful to study energy transfer in low-concentration materials. In YAG doped with < 1% Er³⁺, for example, there would be much less cross-relaxation. Then it would be possible to study lifetime variations due solely to co-doping, without the added distraction of cross-relaxation.

Crystals doped with 1% Er^{3+} and 0.5% Ho^{3+} , for example, should allow a better characterization of the Er \leftrightarrow Ho energy transfers, including a more accurate value for the energy transfer parameters.
The most useful experiments for studying energy transfer would require excitation spectroscopy. By pumping lightly-doped Er:YAG into, for example, the ${}^{4}I_{11/2}$ with a 0.98-µm laser, then watching the fluorescence of the Ho³⁺ ions would reveal much more about the energy transfer processes than the experiments reported here. Such a study would provide not only the excitation spectrum of the Er³⁺ ions, but also the Er—Ho cross-excitation spectrum.

Appendix A-Determination of the Er:YAG Inversion Threshold

To determine the inversion threshold of an Er:YAG laser, we make use of equations 1.8—11:

...

$$\frac{dn_3}{dt} \approx W_{03}n_0 + \frac{1}{2}W_{cr}n_1^2 - \frac{n_3}{\tau_{32}}$$
(1.8)

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} \approx \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} - \rho B_{21}(n_2 - n_1) \tag{1.9}$$

$$\frac{dn_1}{dt} \approx \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - W_{cr} n_1^2 - \frac{n_1}{\tau_{10}}$$
(1.10)

$$\frac{\mathrm{d}n_0}{\mathrm{d}t} \approx \frac{n_1}{\tau_{10}} + \frac{1}{2} W_{\mathrm{cr}} n_1^2 - W_{03} n_0 \,. \tag{1.11}$$

We rewrite these to include only 1.9 and 1.10 by assuming that τ_{32} is so fast that no population collects in level 3. If we also define a value

$$R \equiv \frac{P_{abs}}{V_{abs}h\nu_{abs}} = W_{03}n_0 \tag{A.1}$$

as the rate of photon absorption per unit volume, and assume this is a constant, these equations become

$$\frac{dn_2}{dt} = R + \frac{1}{2}W_{cr}n_1^2 - \frac{n_2}{\tau_{21}} - \rho B_{21}(n_2 - n_1)$$
(3.1)

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_{21}} + \rho B_{21}(n_2 - n_1) - \frac{n_1}{\tau_{10}} - W_{cr} n_1^2.$$
(3.2)

Since Er:YAG lases in a quasi-cw mode, it is reasonable to model the laser as having reached stead-state conditions. In addition, since we are looking for inversion threshold pumping, we can set p = 0. Then equations 3.1 and 3.2 may be combined and solved, to give

$$n_{1} = \frac{\sqrt{1 + 2W_{cr}R\tau_{10}^{2} - 1}}{W_{cr}\tau_{10}} \approx \sqrt{\frac{2R}{W_{cr}}}$$
(A.2)

$$n_2 = n_1 \frac{\tau_{21}}{\tau_{10}} + \frac{1}{2} W_{cr} n_1^2 \tau_{21} = \frac{1}{2} W_{cr} n_1^2 \tau_{21} .$$
 (A.3)

Since $n_1 = n_2$ at inversion threshold, equations A.2 and A.3 may be combined to find

$$R_{\rm th} \approx \frac{1}{2W_{\rm cr}\tau_{21}^2} \approx 10^{24} \, \frac{\rm photons}{\rm cm^3 \, s} \,. \tag{A.4}$$

If the pump is absorbed by the ${}^{4}S_{3/2}$ level at 550 nm, this implies an inversion threshold power density of nearly 400 kW/cm³. The actual lasing threshold will be higher.

Appendix B—Published Papers

Following is a list of all publications, whether or not in reviewed journals, I have written that are relevant to this dissertation. Where possible, I include the abstract of the publication as well as the author list.

B.1 Journal Articles

"Simultaneous, multiple wavelength lasing of (Er, Nd): $Y_3AI_5O_{12}$," by W. Q. Shi, R. Kurtz, J. Machan, M. Bass, M. Birnbaum, and M. Kokta, *Appl. Phys. Lett.* **51**, 1218–20. Simultaneous lasing of both Er^{3+} and Nd^{3+} ions in yttrium aluminium garnet is reported. The crystal was doped with 15% Er^{3+} and 1% Nd^{3+} ions. The Er^{3+} ions lased at 2.94 µm and the Nd^{3+} ions in a broadband from 1.01 to 1.15 µm with a strong peak at 1.064 µm. Significant ion-ion interaction is suggested by the drastically altered fluorescent lifetimes and unusual lasing properties.

"Simultaneous, multiple wavelength lasing of (Ho, Nd):Y₃Al₅O₁₂," by J. Machan, R. Kurtz, M. Bass, M. Birnbaum, and M. Kokta, *Appl. Phys. Lett.* **51**, 1313–5. Simultaneous lasing of both Ho³⁺ and Nd³⁺ in the same crystal of yttrium aluminum garnet (YAG) is reported. The crystal was doped with 10% Ho³⁺ and 1% Nd³⁺ ions. Lasing occurred at 2.940 and 3.011 μ m due to Ho³⁺ ion transitions and at 1.064 μ m due to a Nd³⁺ transition. Appropriate mirrors produced simultaneous lasing at 1.064 and 1.339 μ m due to Nd³⁺ ion transitions. The fluorescent lifetimes of both the Nd^{3+ 4}F_{3/2} and Ho^{3+ 5}I₇ states were significantly lower in the doubly doped material than in Nd:YAG and Ho:YAG. This indicates very strong ion-ion interactions in the (Ho, Nd):YAG crystal.

B.2 Conference Proceedings

"Operation of the high dopant density Er: YAG at 2.94 µm," by M. Bass, W. Q. Shi, R. Kurtz, M. Kokta, and H. Deigl, CLEO 1986 and OSA *Topical Meeting on Tun-able Solid State Lasers*, 1986. Free running, pulsed, flashlamp excited operation of 50

and 33% Er doped YAG lasers is reported at 2.94 μ m. This laser was described by researchers in the Soviet Union as early as 1975. Since then there have been a number of further reports concerning this material all published by Soviet scientists. This paper represents, to our knowledge, the first publication outside of the Soviet Union about high dopant density Er:YAG laser operation. In addition to confirming some of the performance properties described earlier, this paper presents, for the first time, the unusual temporal waveforms of the Er:YAG, 2.94 μ m laser. An outline is given of possible pumping and relaxation processes which may contribute to the laser's operation.

"Simultaneous, multiple wavelength lasing of Er^{3+} and Nd^{3+} ions in $\text{Y}_{3}\text{Al}_{5}\text{O}_{12}$," by W. Q. Shi, R. Kurtz, J. Machan, M. Bass, and M. Birnbaum, OSA *Topical Meeting* on *Tunable Solid State Lasers*, 1987.

"The (holmium, neodymium):yttrium aluminum garnet multiple wavelength solid state laser," by J. Machan, R. Kurtz, M. Bass, M. Birnbaum, and M. Kokta, OSA *Topical Meeting on Tunable Solid State Lasers*, 1987. Simultaneous lasing of both Ho³⁺ and Nd³⁺ in the same crystal of yttrium aluminum garnet (YAG) is reported. The crystal was doped with 10% Ho³⁺ and 1% Nd³⁺ ions. Lasing occurred at 2.940 and 3.011 µm due to Ho³⁺ ion transitions and at 1.064 µm due to a Nd³⁺ transition. Appropriate mirrors produced simultaneous lasing at 1.064 and 1.339 µm due to Nd³⁺ ion transitions. The fluorescent lifetimes of both the Nd^{3+ 4}F_{3/2} and Ho^{3+ 5}I₇ states were significantly lower in the doubly doped material than in Nd: YAG and Ho:YAG. This indicates very strong ionion interactions in the (Ho, Nd):YAG crystal.

"Multiple-wavelength lasing of (erbium, holmium):yttrium aluminum garnet," by Russell Kurtz, Laurie Fathe, Jason Machan, Michael Bass, and Milton Birnbaum, OSA *Topical Meeting on Tunable Solid-State Lasers*, 1989. We tested a solid state laser material, YAG doped with 30% (at.) Er³⁺ ions and 1.5% (at.) Ho³⁺ ions. The laser levels in both Er^{3+} and Ho^{3+} demonstrated altered lifetimes when compared to equivalentlydoped Er:YAG and Ho:YAG, indicating moderate interactions between the Er^{3+} and Ho^{3+} ions. When we lased (Er, Ho):YAG, we observed output at three wavelengths: approximately 2.939, 2.936, and 2.796 µm. The first two of these lased simultaneously, while the third appeared later in the same pump pulse. This lasing blueshift may be explained by excited-state absorption (ESA) in the Ho^{3+} ions.

"New laser lines of erbium in yttrium aluminum garnet," by Russell Kurtz, Laurie Fathe, and Milton Birnbaum, OSA *Topical Meeting on Advanced Solid-State Lasers*, 1990. We studied the lasing and spectroscopic properties of erbium in yttrium aluminum garnet, both as a single impurity and when codoped with neodymium or holmium. In all cases, we observed lasing at 2.936 and 2.939 μ m; when erbium was codoped with holmium, we also observed lasing at 2.796 and 2.766 μ m. [This is in contrast to (Er,Nd):YAlO₃, which lased only on one line, 2.73 μ m.] By determining the energy–level splitting implied by the four observed laser lines, and combining this with transmission spectroscopy, we were able to assign unambiguous values to the Stark sublevels of the three lowest energy levels of Er³⁺ in YAG at room temperature.