Energy Transfer Between Multi-Sites in Chromium-Thulium-Doped Yttrium Aluminum Garnet

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Abstract. We consider the energy transfer between different sites in chromium-thulium-doped yttrium aluminum garnet. Subtle changes in the spectroscopic levels allow us to selectively excite different sites, with considerably different dynamic and spectral behavior. A new analytical model is developed to account for the complicated energy transfer mechanisms in this crystal.

I. Introduction

The study of energy transfer in doped solid-state wide-band gap materials is a cross-disciplinary field of research. On one hand, the effect clearly involves the material properties of the host. Dopants are placed in crystals, with each type of dopant preferring particular sites. The host lattice, through the crystal field, affects the energy levels of the optical transitions in these ions. Defects can drain energy from these ions.

On the other hand, the normal methods of causing and of observing energy transfer are optical in nature. The excitation of these ions is by an optical probe; the study of the ions' dynamic behavior is performed by observing their optical luminescence. Thus, to fully understand the phenomenon of energy transfer in solid state materials, one must take into account both the materials properties and the optical properties of the system.

The phenomenon of energy transfer in solid state materials has been known for most of the twentieth century [1]; it was best modeled and described in the pioneering work of Förster [2,3], Dexter [4], Inokuti and Hirayama [5]. In its simplest form, it involves the transfer of energy from one ion to another in glasses, crystals and molecules imbedded in solid-state lattices.

In the last fifteen years, the topic of energy transfer has taken on additional importance. This has been for

several reasons; first of all, neodymium-chromium-doped gadolinium scandium gallium garnet (Nd,Cr:GSGG) and other doubly doped garnets have shown remarkably promising lasing properties [6–9]. It is operated by pumping the chromium ions; the neodymium ions produce the laser emission. The energy from the chromium arrives at the neodymium ions via non-radiative energy transfer. In addition, the phenomena of upconversion [10] (a form of energy transfer whereby an excited electron can be promoted to an even higher state by energy transfer, allowing output radiation to be at a shorter wavelength and higher photon energy than the pump) has allowed a totally new form of laser to be developed.

Since, in doubly-doped solid-state materials, there are many donors (absorbers of pump energy) and acceptors (receivers of energy from the donors and emitters of the desired radiation), there will be a distribution of distances between donors and acceptors and a multitude of possibilities of pairs of donors and acceptors; it is necessary to develop models for the dynamic behavior of such ions. The most famous is based on the paper of Inokuti and Hirayama [5]; their equation for the evolution of the excited state of donors $N_D(t)$ as a function of time t

$$N_D(t) = N_D(0) \exp(-t/\tau_0^D - At^{1/2}) \qquad (1)$$

where τ_0^D is the spontaneous lifetime of the donors and A is a constant based on the details of the energy transfer process, is the standard expression against which experimental data can be evaluated. The $\exp(-At^{1/2})$ dependence is due to an averaging process which occurs over the possible pairs of donors and acceptors with their various distances [11].

Garnets based on rare-earth ions such as Tm^{+3} or Ho^{+3}, codoped by Cr^{+3}, are of great interest for use as solid-state laser materials [12–28]. In particular, yttrium aluminum garnet (YAG), well known as the host of the neodymium YAG laser, has been doped as well with all three ions for use in a sophisticated scheme to achieve $2\ \mu m$ lasing from the holmium ion [29–42]. The Cr^{+3} is flashlamp pumped in its broad absorption bands (the $^4T_2$ and $^4T_1$ states), the chromium ion transfers energy to the $^3H_4$ thulium state, the excited thulium ion cross relaxes with a ground-state neighboring ion to produce two thulium ions in the $^3F_4$ state, and, finally, after energy hopping between thulium states, the thulium transfers energy to the holmium $^5I_7$ state which lases.

Recent spectroscopic studies at liquid-helium temperatures of YAG and other garnets have shown that there may well be several types of sites, each slightly differing in its crystal-field strength, in which chromium ions may be located. Detailed studies of the energy-transfer processes of these chromium ions to similarly situated neodymium ions show that the type of site in which a chromium donor is located can have a strong effect on its temporal behavior and also on the particular type of neodymium ion (which are also distributed at different sites) to which it chooses to transfer [43,44].

We have undertaken the analysis of energy transfer in such sites [45]. We assume [11] that both the donors and acceptors have main sites where the bulk of the donors and acceptors enter. In addition, there are ''subsites'', sites in which a small fraction of the donors or acceptors enter the crystal; these sites have discernibly unique energy levels either due to the nearby presence of defects or other dopants.

Our initial analysis of energy transfer between subsites involved looking at donors and acceptors which were paired to each other; the presence of the neighboring donor/acceptor both shifted the energy levels of the observed dopant and acceptor and noticeably changed the dynamic behavior of these dopants [11,45–50]. In particular, we have developed a method of analysis of their dynamic behavior [11]; we have analyzed the transition from chromium to thulium in chromium-thulium-holmium-doped yttrium aluminum garnet (Cr,Tm,Ho:YAG) [48,49] and from chromium to neodymium in chromium-neodymium-doped yttrium aluminum perovskite (Cr,Nd:YAP) [50].

In this paper, we return to Cr,Tm,Ho:YAG with the following novelties. We will examine the behavior of sub-sites which are caused by natural defects in the crystal. We shall extend our analytical model to the case where the acceptor itself subsequently undergoes a deexciting energy transfer. Finally, for the first time, we will analyze both donor and acceptor behavior in this crystal for the same transitions; we will be able to confirm that the donors and acceptors are indeed interacting with each other as we would expect.

We note for those familiar with the thulium energy migration common in this material that such diffusion occurs in the $^3F_4$ state, as stated above. Before the energy reaches the $^3F_4$ state. We will be studying the emission from the $^3H_4$ state, from which it is much less likely to have energy transfer to other $^3H_4$ states

between thulium ions. (If there is such transfer, it will make all thulium subsites appear identical. The differences observed in the spectroscopy of such sites precludes the likelihood of such transfer.)

II.   Model Development

The development of an analytical model to describe the dynamic behavior of donors and acceptors has been developed over several years and has been detailed in [11]. A simplified approach there shows that the behavior of the concentration of donors $N_D(5)$ emitting at a particular wavelength at time t is given by

$$N_D(t) = U \exp(-t/\tau_0^{D'}) + V[\exp(-t/\tau_0^D) - (c/c_0)\sqrt{\pi}(t/\tau_0^D)^{1/2}] \qquad (2)$$

This expression needs extensive explanation. The two terms are due to our assumption that, at the given wavelength, there are two types of donors being observed. The regular donor, given by the second term, behaves as normal, transferring to acceptors throughout the crystal. The form of the second term is the standard term given by Inokuti and Hirayama in [5]. τ_0^D is the spontaneous lifetime of the donor, c is the acceptor concentration, and c_0 is the critical acceptor concentration for which the spontaneous decay probability of the donor equals the probability of energy transfer. The first term represents the donors which happen to be paired with a close acceptor. The closely paired donor can transfer in a multitude of ways, both via the exchange interaction or any of the multipole interactions; these rates can be considerably faster than the originally assumed donor-donor interaction. The deactivator of such a donor is very quick with $\tau_0^{D'}$ containing within it both the spontaneous decay rate and the transfer rate to the neighboring acceptor. The ratio of U to V will be very dependent on the particular wavelength we will be pumping and measuring.

If we assume that our acceptor decay time τ_0^A is much larger than $\tau_0^{D'}$, then we can assume that the energy transfer occurs almost instantaneously. In this case, the expression for the acceptor concentration $N_A(t)$ at time t (which undergoes spontaneous decay after excitation) will be

$$N_A(t) = U' \exp(-t/\tau_0^A)$$
$$+ V' \exp(-t/\tau_0^A) \int_0^{t'} \exp(t'/\tau_0^A)$$
$$\times \left[-\frac{dN_D'(t')}{dt'} - \frac{N_D'(t')}{\tau_0^{D'}} \right] dt' \qquad (3)$$

The first term represents the decay of paired acceptors which have received their energy almost instantaneously from the donors ($\tau_0^A \gg \tau_0^{D'}$). The second term is the term for the acceptors which are excited in the standard way by the fraction of unpaired donors labeled here $N_D'(t')$ and represented in Eq. (3) by the term whose initial concentration is V'. U' and V' are once again strongly wavelength dependent.

We must now extend our analysis to the case where the acceptor, when excited, does not merely decay exponentially but can also deactivate via another energy transfer. (In our case of Tm^{+3}, the acceptor when excited in the $^3H_4$ thulium state can relax with a ground-state neighboring ion to produce two thulium ions in the $^3F_4$ state.) At this point, we can assume that the deexcitation for an impulse excitation would be given by a function $\phi_A(t)$ such that

$$N_A(t) = N_A(0)\phi_A(t) \qquad (4)$$

Then the natural extension to Eq. (3) would be

$$N_A(t) = U' \int_0^t \phi_A(t - t')$$
$$\times \left(-\frac{N_D(t')}{\tau_0^D} - \frac{dN_D(t')}{dt'} \right) dt' \qquad (5)$$

For the case of an acceptor undergoing cross-relaxation, $\phi_A(t)$ would be given by

$$\phi_A(t) = \exp\left[(-t/\tau_0^A) - \frac{c}{c_0'} \sqrt{\pi}(t/\tau_0^A)^{1/2} \right] \qquad (6)$$

where c_0' is the critical concentration for the acceptor-acceptor deexcitation process.

We will now make the following simplifying assumptions, appropriate for our system. Assume that the process $\phi_A(t)$ has a mean time of decay $\tau_0^{A'}$. (Note that since $\phi_A(t)$ is not necessarily exponential, the function is not determined by its mean decay time.) Assume also that the unpaired donor has an average deexcitation time of $\tau_0^{D''}$ (including both spontaneous decay and donor acceptor transfer). $\tau_0^{D'}$

and $\tau_0^{D''}$ are thus the average lifetimes for the paired and unpaired donors respectively.

We will assume that

$$\tau_0^{D'} \ll \tau_0^{A'} \tag{7a}$$

and

$$\tau_0^{A'} \ll \tau_0^{D''} \tag{7b}$$

Equation (7a) states that energy transfer from a paired donor to an acceptor is still instantaneous *vis à vis* the deexcitation behavior of the acceptor (even when acceptor-acceptor deexcitation is taken into account). Equation (7b) guarantees that the acceptor will deactivate much faster than the unpaired donor. The unpaired donors thus function as a reservoir of energy for the unpaired acceptors.

If we look at Eq. (5), it is clear that the exact form of $\phi_A(t)$ will be critical for the first term in the equation. However, in the second term, since the overall decay will be determined by the donor concentrations, $\phi_A(t)$ can be replaced by an average decay time with little change to the experimental curves. Thus our simplified Eq. (5) is

$$N_A(t) = U'\phi_A(t) + V'\exp(-t/\tau_0^{A'})$$
$$\times \int_0^{t'} \exp(t'/\tau_0^{A'})\left[-\frac{dN_D'(t')}{dt'} - \frac{dN_D'(t')}{\tau_0^{D'}}\right]\cdot dt' \tag{8}$$

where

$$\phi_A(t) = \exp\left[(-t/\tau_0^A) - \frac{c}{c_0}\sqrt{\pi}(t/\tau_0^A)^{1/2}\right] \tag{9}$$

III.   Experimental Procedure

A single crystal of Cr:Tm:Ho:YAG with a concentration of 1% at Cr^{3+}, 5.76% at Tm^{3+}, and 0.36% at Ho^{3+} was used for this study after extensive polishing of two of its surfaces. The excitation spectra and lifetime measurements were all performed at 8 K using liquid-helium cryogenic system. Excitation spectra were measured using a pulsed Laser Analytical Systems dye laser (10 mJ/pulse, 0.04 cm^{-1} spectral resolution), pumped by a frequency doubled B.M. Industries pulsed Nd:YAG laser (8 nsec pulse duration, 300 mJ/pulse at 532 nm). The

excitation wavelengths were scanned over a wavelength range of 675–695 nm. The thulium emission was followed by monitoring the ($^3F_4 \rightarrow {}^3H_6$) thulium emission at 806 nm and 809 nm. The emission was transmitted through a monochromator (Jobin-Yvon HRS2, 2.4 nm/mm slit resolution), and detected by an R1767 Hamamatsu photomultiplier detecting near 800 nm. The signal was fed into a computerized Stanford Research System SR 250 boxcar averager. The decay of the fluorescence was averaged over 250 pulses and recorded with a 9410 LeCroy oscilloscope coupled with a computerized analyzing system.

IV.   Experimental Results

Figures 1 and 2 are excitation spectra obtained from our Cr,Tm,Ho:YAG crystal for two thulium emission wavelengths 809 nm and 806 nm. The two largest peaks for exciting the 806 nm emission are at chromium absorption peaks at 686.66 nm and 685.778 nm; thus these are the two excitation wavelengths we will choose to study. Figures 3 and 4 show the decay from the chromium donors at 705 nm when excited at 686.66 and 685.778 nm respectively using Eq. (2) with $U = 0$. The fit is good for times greater than 1 msec (Table 1); less than that, the donors show an enhanced decay, much faster than the predicted model. (Since we are looking at a log scale, the deviation is significant.) The natural time decay of the Cr^{+3} ions are site dependent: 77.96 and 49.91 msec for the 686.66 and 685.778 nm sites, respectively.

Theoretically, it should be possible to analyze the deviation of excited donors from the standard behavior, as given by the first term in Eq. (2). However, its exact value is difficult to measure, both because the resolution of the system as a function of time is uncertain and because the decay is very fast in the initial decay. It is *much* easier to see the fraction of energy which has been transferred to the acceptors and is stored there. Therefore, let us immediately examine the acceptor decay.

Figure 5 shows the Tm^{+3} acceptor decay obtained at 806 nm when pumped at 865.778 nm. The bottom curve represents the response expected from the standard acceptor decay in Eq. (3) with U' equal to 0. While the fit is good for times after 600 μsec, it is poor for times less than this. Moreover, no value of U' in Eq. (3) will help; the decay from 20 msec and onward is decidedly non exponential (which would have
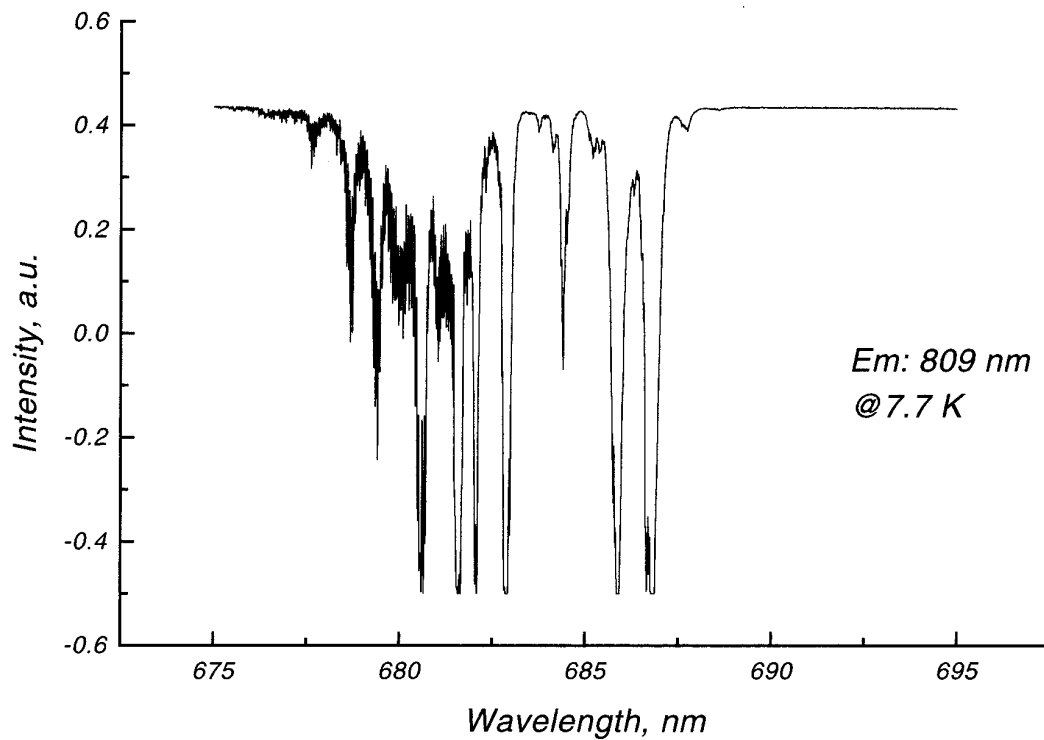
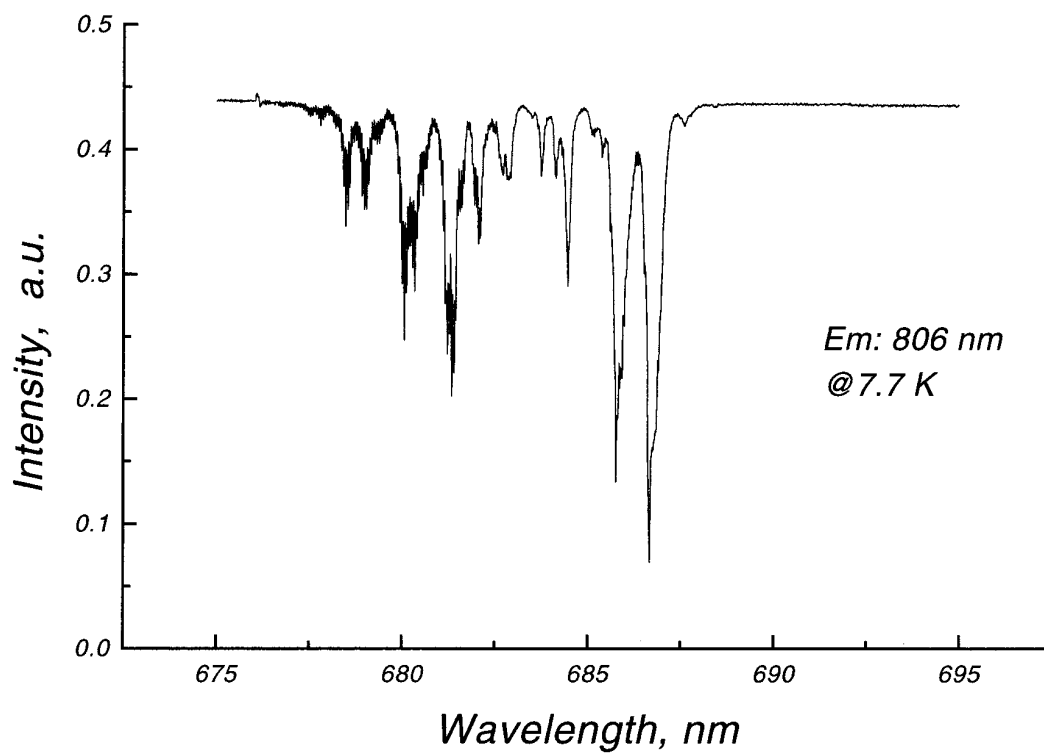*Fig. 1.* Excitation spectrum of Tm^{+3} emission at 809 nm.

Fig. 2. Excitation spectrum of Tm^{+3} emission at 806 nm.

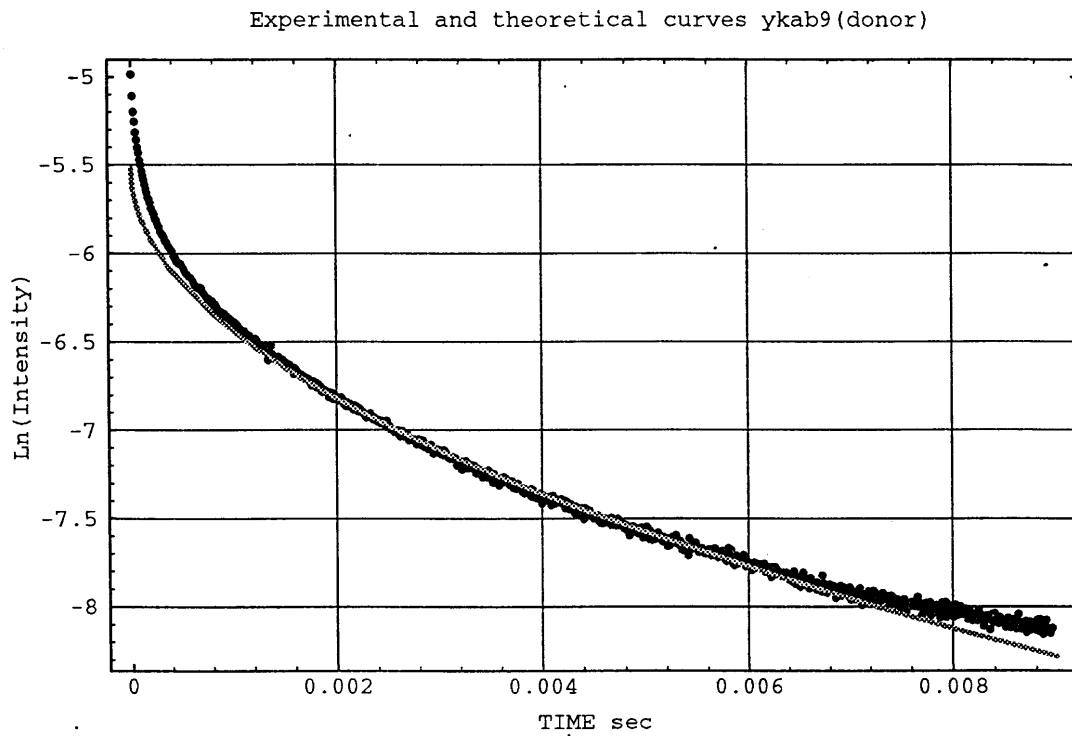Experimental and theoretical curves ykab9(donor)

Fig. 3. Cr^{+3} decay at 705 nm when excited at 686.660 nm. In this figure and the following one, the data are given by dots; the light solid line is the modeled fit.
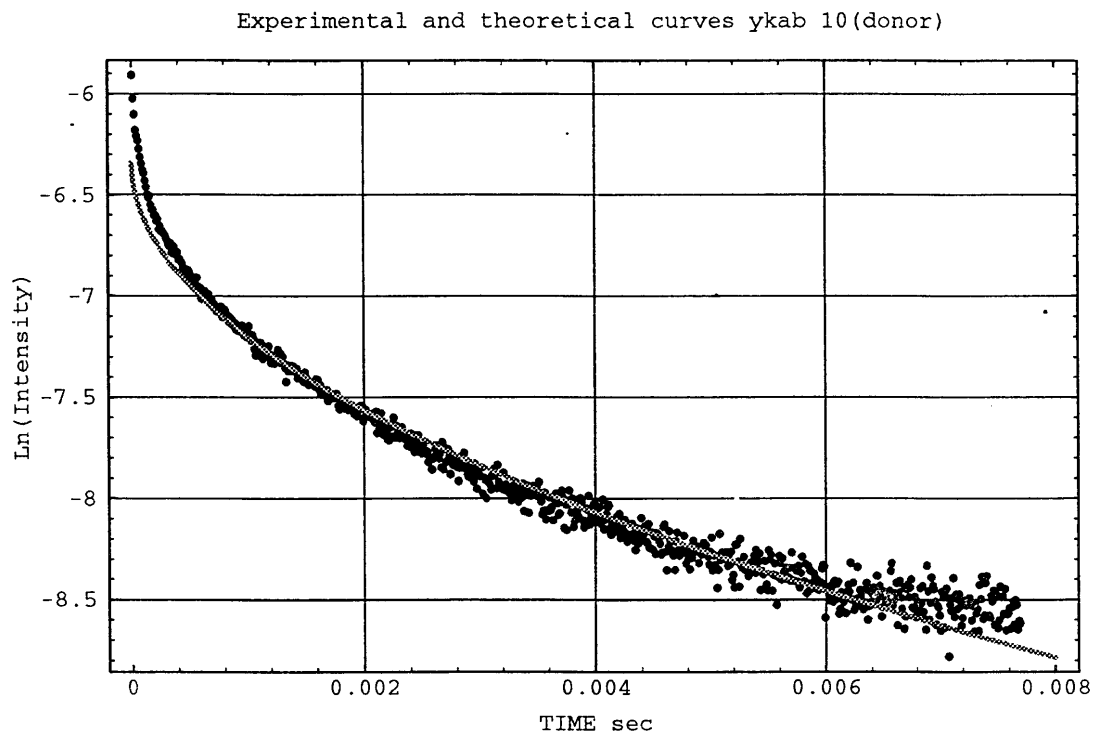
Experimental and theoretical curves ykab 10(donor)

Fig. 4. Cr^{+3} decay at 705 nm when excited at 685.778 nm.

Table 1. Parameters for Donor Decay

Figure	Excitation (nm)	Emission (nm)	τ_0^D (ms)	$\dfrac{c}{c_0}\dfrac{1}{\sqrt{\tau_0^D}}$ (ms$^{-1/2}$)	U/V
3	686.660	705	77.96	29.1	—
4	685.778	705	49.91	27.3	—

Table 2. Parameters for Acceptor Decay

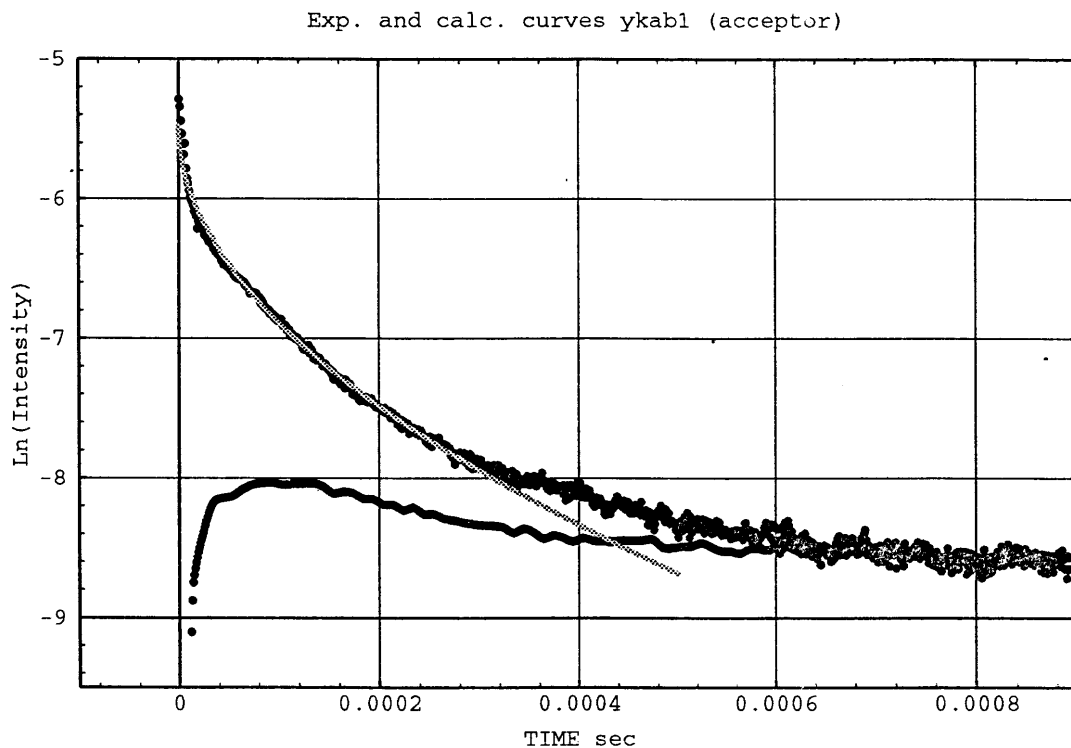| Figure	Excitation (nm)	Emission (nm)	τ_0^A (ms)	$\tau_0^{A'}$ (ms)	$\dfrac{c}{c_0}\dfrac{1}{\sqrt{\tau_0^A}}$ (ms$^{-1/2}$)	U/V
5	685.778	806	13	0.1	137.7	12.98
6	685.778	809	13	0.1	143.2	22.69
7	685.778	824	13	0.1	154.8	38.17
8	685.778	795	13	0.1	139.5	16.22
9	686.660	806	13	0.1	148.6	22.25
10	686.660	809	13	0.1	134.3	42.49
11	686.660	824	13	0.1	147.9	80.0
12	686.660	795	13	0.1	157.1	41.86

Exp. and calc. curves ykab1 (acceptor)

Fig. 5. Tm^{+3} decay at 806 nm when Cr^{+3} is excited at 685.778 nm. In this and the following figures, (through Fig. 12), the solid light line is the modeled decay of paired thulium, the solid dark line is the modeled decay of unpaired thulium and the dots are experimental data.
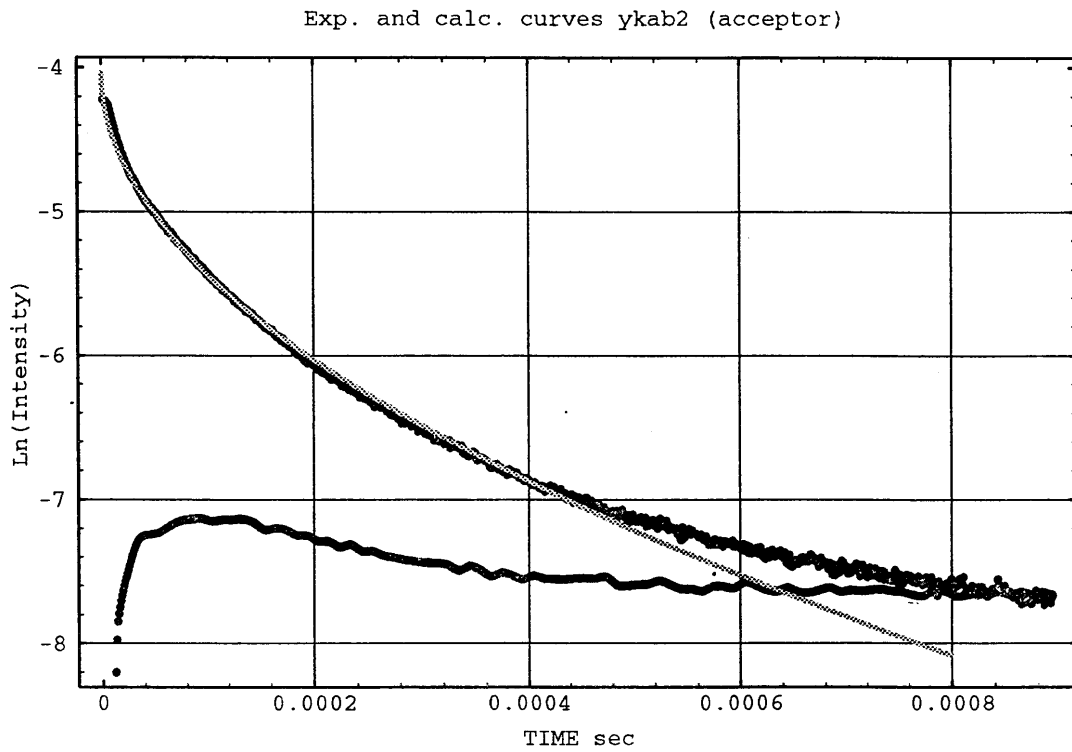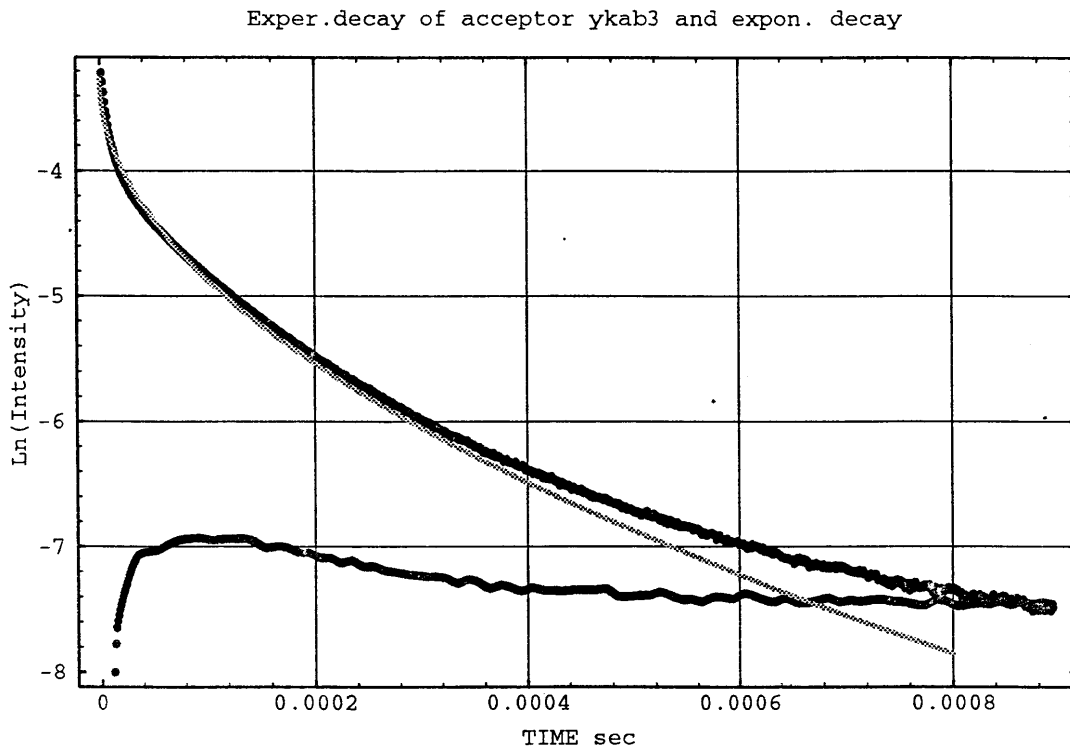
Exp. and calc. curves ykab2 (acceptor)

Fig. 6. Tm^{+3} decay at 809 nm when Cr^{+3} is excited at 685.778 nm.

Exper.decay of acceptor ykab3 and expon. decay

Fig. 7. Tm^{+3} decay at 824 nm when Cr^{+3} is excited at 685.778 nm.

Exp. and calc. curves ykab4 (acceptor)

Fig. 8. Tm^{+3} decay at 795 nm when Cr^{+3} is excited at 685.778 nm.

Experimental and theoretical curves ykab5(acceptor)

Fig. 9. Tm^{+3} decay at 806 nm when Cr^{+3} is excited at 686.660 nm.

Exp. and calc. curves ykab6 (acceptor)

Fig. 10. Tm^{+3} decay at 809 nm when Cr^{+3} is excited at 686.660 nm.

Exp. and calc. curves ykab7 (acceptor)

Fig. 11. Tm^{+3} decay at 824 nm when Cr^{+3} is excited at 686.660 nm.

Experimental and theoretical curves ykab8(acceptor)
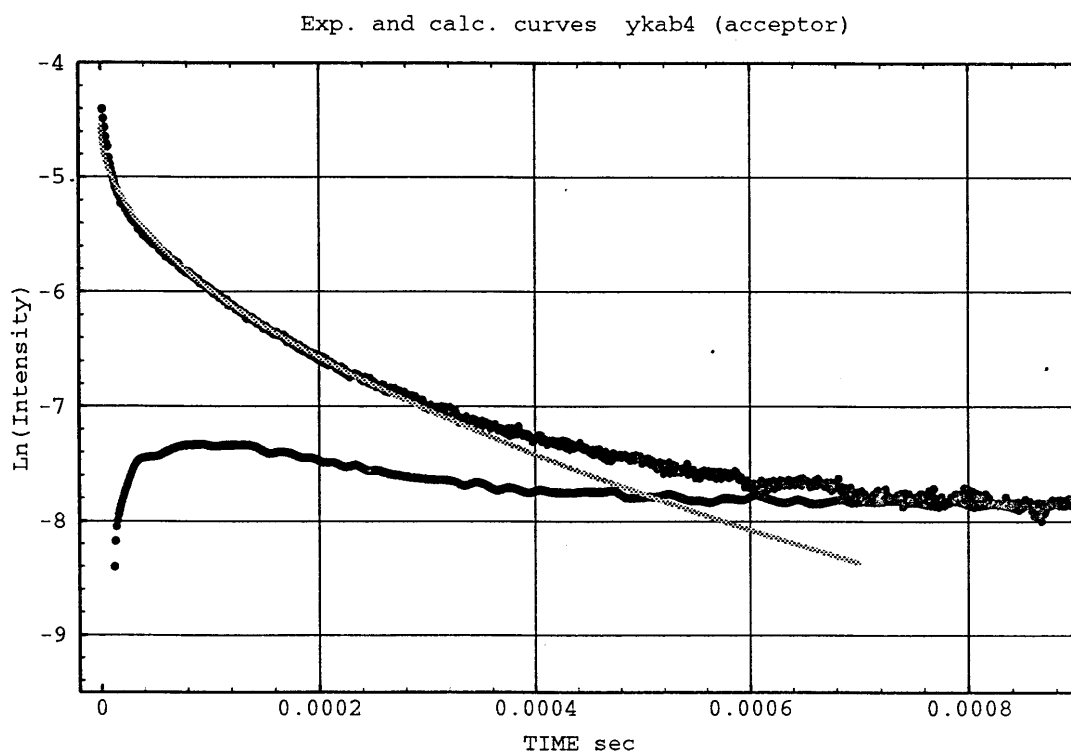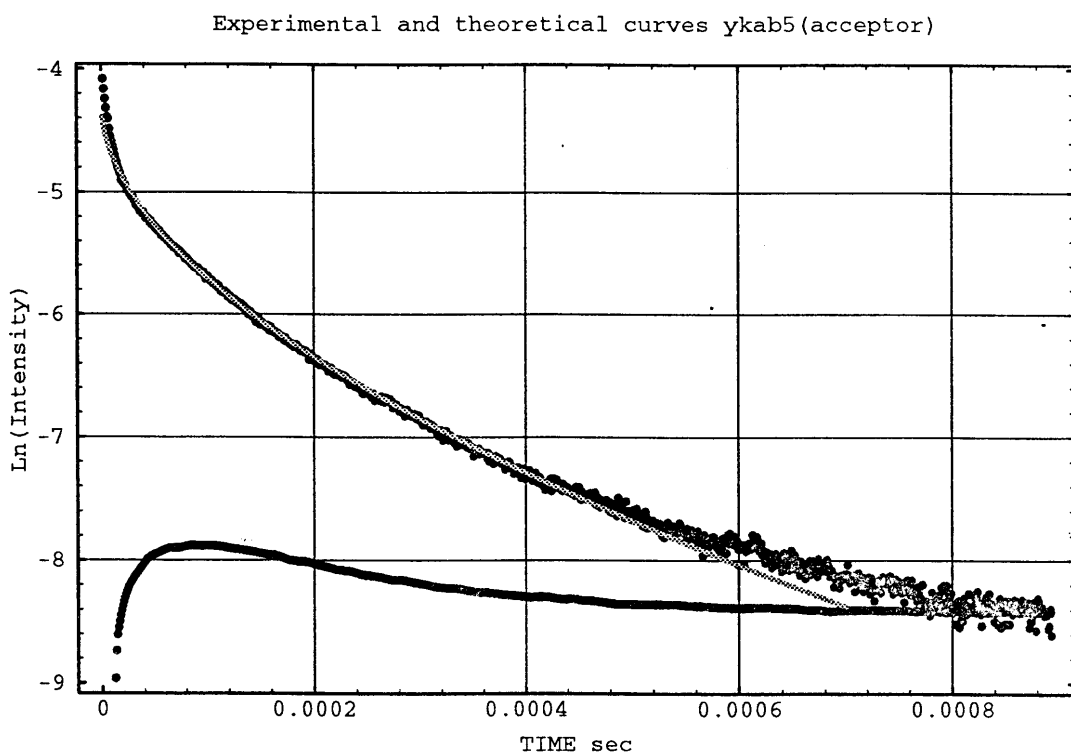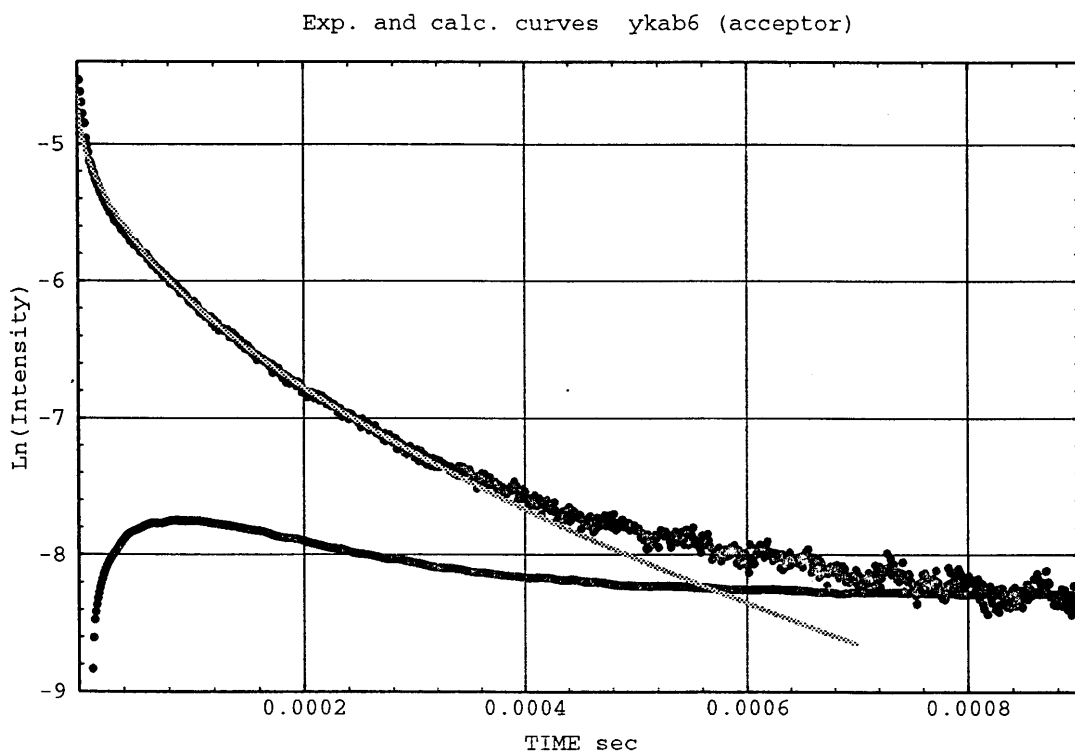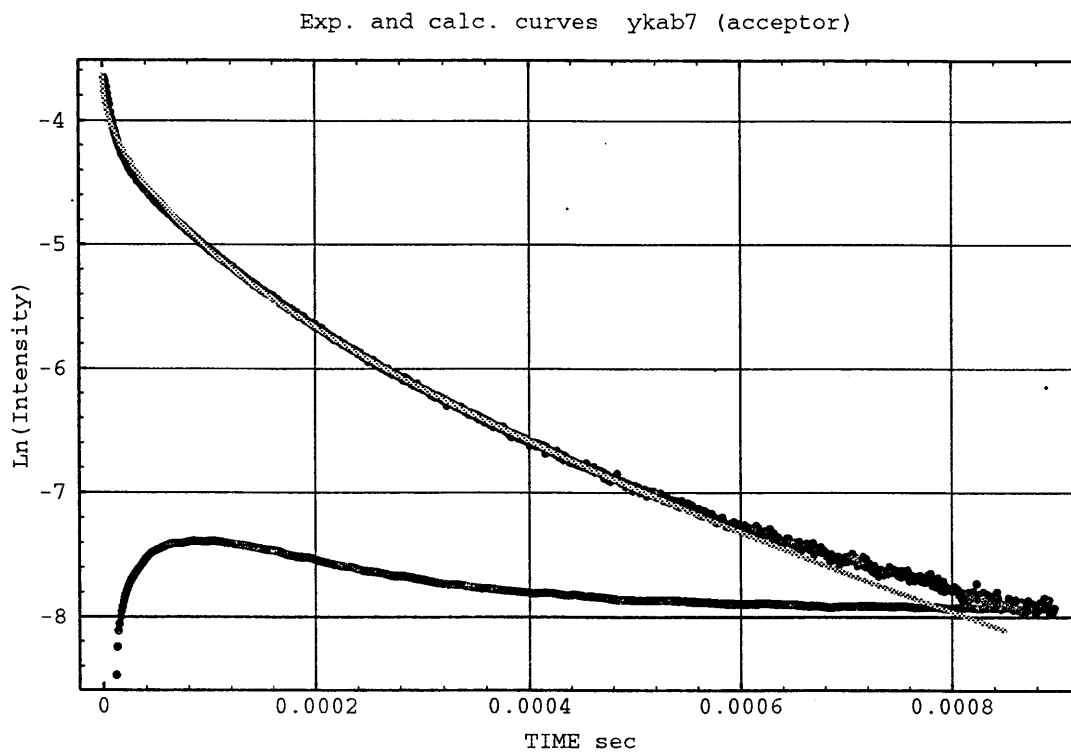
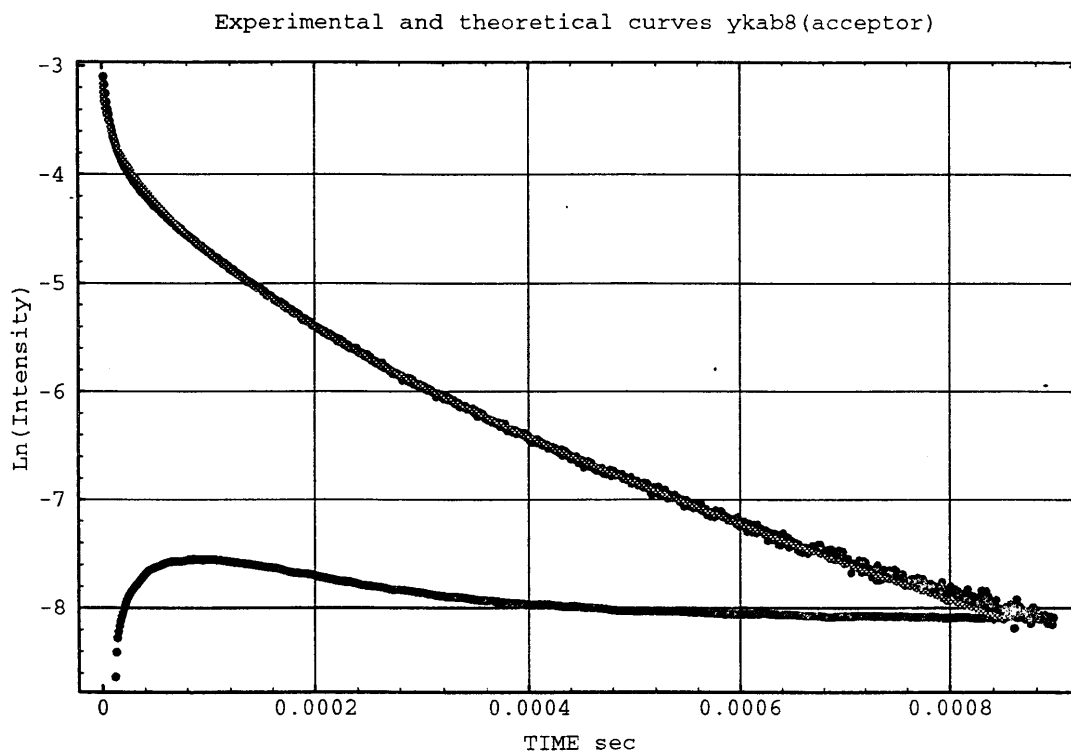*Fig. 12.* Tm^{+3} decay at 795 nm when Cr^{+3} is excited at 686.660 nm.

appeared as a straight line on our semi-log graph). Instead, our fit is based on Eq. (8) where the parameters are given in Table 2. Similar results are shown in Figs. 6–12 for the other pairings of the two excitation wavelengths 685.778 and 686.66 nm and the four emission wavelengths (806, 809, 824 and 795 nm).

V. Discussion

From these data, a set of questions can be answered concerning the behavior of the multi-sites based on our measurements.

There is a notable difference in the decay behavior of donors (Cr^{+3}) absorbing at 686.660 nm vs. those absorbing at 685.778 nm. The natural decay constant is 78 msec at 686.66 nm and 50 msec at 685.778 nm. Such a change in decay time is not unexpected. The differences in crystal field, causing the shift in the absorption lines, would be expected to cause changes in the transition rate and hence in the decay times.

There is also a notable difference in the ratio of paired acceptors (Tm^{+3}) excited (U') compared to unpaired acceptors excited (V'). The ratio for each of the four acceptor emission lines of U'/V' is approximately two times larger when exciting at 686.66 nm than at 685.778 nm. The sites absorbing at 686.66 nm are twice as likely to have acceptor ions paired to them as those absorbing at 685.778 nm.

Certain things do not change. The acceptors which get their energy from the donors interactively deexcite independent of whether they got their energy from a donor at 685.778 nm or 685.66 nm; this is determined by the fairly constant value (within experimental error) of $c/(c_0\sqrt{\tau_0^A})$. No changes were observed in τ_0^A or $\tau_0^{A'}$ as a function of donor excitation wavelength. Once the energy got to any acceptor, it acted as did all acceptors.

The proportion of acceptor emission due to the paired acceptors compared to the unpaired acceptors for a fixed donor excitation was highly dependent on the observed acceptor wavelength. The 824 nm emission was the most associated with the paired acceptors; the 795 nm emission was the most related

to the unpaired acceptors. Each emission was a combination of paired and unpaired emissions. Further studies on other lines will enable us to see if such patterns persist.

This model could be potentially advantageous in understanding the exact mechanisms of energy transfer. No longer will our understanding be based on the average donor and acceptor behavior. We can now see which donor sub-sites transfer to which acceptor sub-sites, and which do not transfer at all; one paper on chromium, neodymium:GSGG shows that the main chromium site does not transfer to neodymium [52]. Moreover, we should be able to see if backtransfer (transfer from acceptor to donors) affects particular ions in codoped laser materials. A new field for research has opened.

VI.   Conclusions

We have developed a model to enable the analysis of multi-site energy transfer in solid-state wide-bandgap laser materials. This model has even been extended to cases where a subsequent non-radiative deexcitation of the acceptor occurs. A good fit to the data is obtained. A systematic pattern of change appears when one varies the excitation and/or emission wavelength.

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