



Two-step excitation and direct two-photon absorption in $\text{Tb}^{3+}:\text{LiYF}_4$
by Richard Paul Jones

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in
Physics

Montana State University

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Abstract:

Two-photon absorption is used to study highly excited levels of the Tb^{3+} ion. The known energy levels of Tb^{3+} have thus been extended to much higher energies. This work also makes contributions to the study of these highly excited levels through accurately measured energies and discriminations between those levels which are intrinsic and those which are due to crystal defects.

These experiments involved using either one or two pulsed tunable dye lasers operating in the visible range of the spectrum. The levels studied included the 5K9, 5D2, 5G6, 5K8, a second 5G6, 5K5, and 5K7 of the 4f8 configuration of Tb^{3+} in a host crystal of LiYF_4 . These levels correspond to a total studied energy range of 39,100 cm^{-1} to 42,000 cm^{-1} .

These experimentally measured energies are compared with an advanced computer-fitted free ion and crystal field model showing a high degree of correlation between experiment and theory. The population of these highly excited states was sensitively monitored by observing the anti-Stokes ultraviolet cascade fluorescence from 5D3 to 7F5. Energy levels due to crystal defects were discriminated against the intrinsic energy levels by controlling the laser timing in the two-laser two-step excitation experiments.

These results suggest that the selection rules for pure 4f8 configurations as applied to direct two-photon absorption are not entirely valid and can be broken. This provides further support for a theory of resonant enhancement.

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APPROVAL

of a thesis submitted by

Raymond Paul Jones

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

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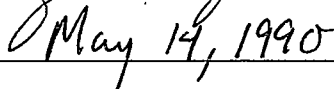
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ABSTRACT

Two-photon absorption is used to study highly excited levels of the Tb^{3+} ion. The known energy levels of Tb^{3+} have thus been extended to much higher energies. This work also makes contributions to the study of these highly excited levels through accurately measured energies and discriminations between those levels which are intrinsic and those which are due to crystal defects.

These experiments involved using either one or two pulsed tunable dye lasers operating in the visible range of the spectrum. The levels studied included the $^5\text{K}_9$, $^5\text{D}_2$, $^5\text{G}_6$, $^5\text{K}_8$, a second $^5\text{G}_6$, $^5\text{K}_5$, and $^5\text{K}_7$ of the $4f^8$ configuration of Tb^{3+} in a host crystal of LiYF_4 . These levels correspond to a total studied energy range of 39,100 cm^{-1} to 42,000 cm^{-1} .

These experimentally measured energies are compared with an advanced computer-fitted free ion and crystal field model showing a high degree of correlation between experiment and theory. The population of these highly excited states was sensitively monitored by observing the anti-Stokes ultraviolet cascade fluorescence from $^5\text{D}_3$ to $^7\text{F}_5$. Energy levels due to crystal defects were discriminated against the intrinsic energy levels by controlling the laser timing in the two-laser two-step excitation experiments.

These results suggest that the selection rules for pure $4f^8$ configurations as applied to direct two-photon absorption are not entirely valid and can be broken. This provides further support for a theory of resonant enhancement.

CHAPTER 1

INTRODUCTION

This thesis contributes to the study of the highly excited states of the Tb^{3+} ion through accurately measured energies and discriminations between those levels which are intrinsic and those which are due to crystal defects. The object of this study is to present new experimental energies for trivalent Terbium in a host crystal of LiYF_4 . This work is particularly interesting in the fact that prior to it, the Tb^{3+} ion was essentially unstudied in any crystal over these energy ranges.

The absorption and emission spectra of rare earth ions in ionic crystals are particularly interesting since the optical linewidths can be as narrow as those in gases. This suggests that these rare earth ions are behaving as if they were almost free ions. In fact, this is somewhat true and is due to a shielding of the 4f electrons (where these transitions take place) by the outer 5s and 5p electrons. The deviations from this free ion model can then be accounted for by applying perturbations to the free ion Hamiltonian and re-diagonalizing to obtain the new energies. This is not as easy as it sounds. The end result is that the calculated energy levels are written in terms of parameters that must be determined by comparison with experimentally measured energy levels.

Two non-linear optical processes were utilized to complete this task, these being Two-Step Excitation (TSE) and Direct Two-Photon Absorption (TPA). In a

TSE process, an atom is excited from its ground state to a real intermediate state by absorbing a single photon and then is excited again to a final state by the absorption of another photon of different energy. In a TPA process the atom is excited from the ground state to the final excited state by the simultaneous absorption of two photons having the same energy. Both of these processes will hereafter be referred to as two-photon transitions.

The advent of tunable dye lasers enabled researchers to observe these sharp parity-allowed $4f^n \rightarrow 4f^n$ two-photon transitions with much more ease. Prior to this development, the first two-photon transition was observed in Eu^{2+} in 1961 by Kaiser and Garrett, capitalizing on a broadband absorption feature that crystal has.¹ What is remarkable is that the basic theory of two-photon transitions was developed thirty years prior to this in 1931 by Göppert-Mayer.² In the time since Kaiser and Garrett's work however, considerable advances have been made in the field of two-photon transitions in rare earths,³⁻⁹ as well as gases.¹⁰

Two-photon transitions have been previously observed in both dilute $\text{Tb}^{3+}:\text{LiYF}_4$ (1% concentration) and concentrated LiTbF_4 by Huang in 1987.¹¹ His experiments concentrated on the $^5\text{G}_6$ level of $\text{Tb}^{3+}:\text{LiYF}_4$ by both TPA and TSE and the $^5\text{K}_8$ level of LiTbF_4 by TSE. These present studies extend his work and are partially based upon work done by Cone at Bell Laboratories.¹² The $\text{Tb}^{3+}:\text{LiYF}_4$ crystal used is the same one used by Huang and Cone, and the energy levels studied were the $^5\text{D}_2$, $^5\text{K}_9$, $^5\text{K}_8$, a second order $^5\text{G}_6$ term, $^5\text{K}_5$, and $^5\text{K}_7$. Huang's results for the $^5\text{G}_6$ level with TPA were also reproduced in this work with a higher level of accuracy by using an opto-galvanic calibration system throughout the experiments. These levels correspond to a total studied energy range extending from $39,100 \text{ cm}^{-1}$ to $42,000 \text{ cm}^{-1}$.

The selection rules (found through group theory considerations) for a single-

step excitation are different than those of a two-step excitation. In other words, each process by itself yields information that the other cannot. Therefore, when both processes are considered together, a maximum amount of information is obtained, and the processes are said to be complementary. In a TPA process the selection rules allow for all possible transitions. This could lead one to believe that this is the ideal technique. However, the transition matrix element for a TPA process is a second order process in the atom-radiation interaction Hamiltonian, and thus its magnitude is very weak when compared to a single-step or two-step excitation. Certain other experimental difficulties, such as the dye laser broadband fluorescence and vibronic absorption, limited the use of TPA processes to only a few levels with the present experimental apparatus. These two-photon processes allowed for the study of energy levels up to $42,000\text{ cm}^{-1}$ with visible light, whereas one would need to use UV spectroscopy to study these same levels with single-step excitations. For these reasons outlined above, TSE was the primary means of obtaining information on these levels.

In this thesis, a brief description of the theory of two-photon transitions as well as a discussion of the selection rules applied to $\text{Tb}^{3+}:\text{LiYF}_4$ will be given in Chapter 2. This is followed by a description of the experimental apparatus and procedures in Chapter 3. A discussion of the experimental results will be given in Chapter 4 followed by some concluding remarks in Chapter 5.

CHAPTER 2

THEORY

The theory of two-photon absorption can be explained by several methods. One method involves expanding the macroscopic polarization in a power series of the optical electric field and taking the imaginary part of the third order nonlinear susceptibility. An alternate method is to treat the optical radiation field as a perturbation to the ionic Hamiltonian and to expand it to second order. Both of these methods are very well developed, so only a brief outline of the perturbation theory approach will be given.

The following derivation of the two-photon transition rate closely follows Gold,¹³ so the reader can consult his work or any of the other references for further details.^{14,15} Following the derivation, a brief description of the energy levels for Tb^{3+} will be given along with an explanation of the group theory approach to the selection rules.

Two-Photon Absorption

Consider a multi-level atomic system with definite eigenstates corresponding to the unperturbed hamiltonian,

$$H_0 | \Psi_n(r,t) \rangle = E_n | \Psi_n(r,t) \rangle, \quad (2.1)$$

where

$$| \Psi_n(r,t) \rangle = | \phi_n(r) \rangle \exp(-i\Omega_n t). \quad (2.2)$$

The two-photon transition process will bring the system to a final excited state $|\Psi_f\rangle$ lying at an energy $E_{fg} = \hbar(\Omega_f - \Omega_g) = \hbar\Omega_{fg}$ above the ground state $|\Psi_g\rangle$. At $t = 0$ a perturbation H' is added so that the new hamiltonian is

$$H = H_0 + \lambda H', \quad (2.3)$$

where H' is an electric dipole interaction described by

$$H' = -er \cdot [E_1 \varepsilon_1 \exp(-i\Omega_1 t) + E_2 \varepsilon_2 \exp(-i\Omega_2 t)]. \quad (2.4)$$

The vectors ε_1 and ε_2 refer to the polarizations of the two respective electric fields E_1 and E_2 , and Ω_1 and Ω_2 are the respective frequencies. At $t > 0$ there will be a superposition of the original eigenstates

$$\Psi = \sum_n a_n(t) \Psi_n(r, t) \quad (2.5)$$

which must obey Schrödinger's time-dependent equation

$$i\hbar \frac{\partial}{\partial t} \Psi = (H_0 + \lambda H') \Psi. \quad (2.6)$$

Upon inserting the superposition state (2.5) above into (2.6), taking the vector product with $\langle k|$, and expanding to second order, the probability density for the population of the final state is found to be

$$|a_n^{(2)}|^2 = \left| \frac{2e^2 E_1 E_2}{\hbar} \right|^2 |R_{fg}|^2 \frac{\sin^2[\frac{1}{2}(\Omega_{fg} - \Omega_1 - \Omega_2)t]}{(\Omega_{fg} - \Omega_1 - \Omega_2)^2} \quad (2.7)$$

where

$$R_{fg} = \sum_k \frac{\langle f|\epsilon_1 \cdot r|k\rangle \langle k|\epsilon_2 \cdot r|g\rangle}{\hbar(\Omega_{kg} - \Omega_2)} + \frac{\langle f|\epsilon_2 \cdot r|k\rangle \langle k|\epsilon_1 \cdot r|g\rangle}{\hbar(\Omega_{kg} - \Omega_1)}. \quad (2.8)$$

In the event that t becomes sufficiently large, the function

$$\frac{\sin^2[\frac{1}{2}(\Omega_{fg} - \Omega_1 - \Omega_2)t]}{(\Omega_{fg} - \Omega_1 - \Omega_2)^2}$$

behaves as a delta function, resulting in a transition rate of

$$\frac{1}{\tau} = \frac{d}{dt} |a^{(2)}_f|^2 = \frac{2\pi}{\hbar^2} e^4 |E_1|^2 |E_2|^2 |R_{fg}|^2 \delta(\Omega_{fg} - \Omega_1 - \Omega_2). \quad (2.9)$$

Here, all of the non-resonant terms have been eliminated by use of the rotating wave approximation. The R_{fg} term is the main item of interest in this last equation. Through it, a detailed investigation can be made by varying the polarization of the incident beams. Since the states $|k\rangle$ in equation 2.8 run over the complete set of eigenstates of the unperturbed hamiltonian H_0 , the function

$$\sum_k \frac{|k\rangle \langle k|}{\hbar(\Omega_{kg} - \Omega_j)} \quad (2.10)$$

transforms as an identity representation. This simplifies equation (2.8) and all of the relevant information can found from the matrix elements

$$\langle f|r_1 r_2|g\rangle, \quad (2.11)$$

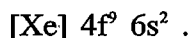
where r_1 and r_2 are the projections of ϵ_1 and ϵ_2 on r and

$$\mathbf{r} = \sum_i \mathbf{r}_i \quad (2.12)$$

is the position operator of the electrons.

Energy Structure of Terbium

The electronic configuration of the terbium atom is that of xenon plus eleven more electrons. These start to fill the 4f and 6s shells resulting in an electronic configuration of



When terbium is placed in a suitable host lattice and becomes triply ionized the two 6s electrons and one 4f electron are removed and its electronic configuration becomes



The 5s and 5p electrons are located spatially further from the nucleus and therefore serve to shield the 4f electrons from the outside environment. This sort of phenomenon gives all rare earths their gas-like spectral properties.

One now might think that there should be no visible allowed transitions since all the transitions will take place within the 4f shell, with no resulting change in the parity. However, configuration mixing due to the crystal field does allow these transitions to take place.

The energy levels are normally labeled in Russell-Saunders term notation. However, both the coulomb interaction and spin-orbit interaction are comparable in magnitude, and an intermediate coupling scheme needs to be applied. Due to the large number of particles with an orbital angular momentum of three in the Tb^{3+} ion, some S and L terms will occur more than once. New additional quantum

numbers need to be taken into account. J still continues to be a good quantum number.

Much work has been done on determining the lower energy levels of Tb^{3+} that will not be discussed in this thesis.¹⁶⁻¹⁹ The references are cited for the interested reader.

Crystal Field

The host crystal for this particular set of experiments was LiYF_4 which crystallizes in a tetragonal scheelite structure. This crystal was doped with 1% Tb^{3+} , with the Tb^{3+} ions taking the place of Y^{3+} ions, resulting in each Tb^{3+} ion having an S_4 site symmetry. The S_4 symmetry operator is the basic element of the group, and it involves a rotation of $\pi/2$ about the symmetry axis followed by a reflection in a plane perpendicular to this axis of rotation. Since this operation does not alter the crystal structure in any way, it must commute with the crystal field hamiltonian.

The crystal field hamiltonian can be expressed as¹⁶

$$H_{\text{cf}} = \sum_{k=1}^6 B_{kq} C_q^{(k)} \quad (2.13)$$

where $C_q^{(k)}$ are spherical harmonics and B_{kq} are empirically determined parameters. The crystal field removes the spherical symmetry of the free ion, and hence, removes some of the $2J+1$ degeneracy of the energy levels.

Group Theory

Since the crystal field hamiltonian and the S_4 symmetry operators commute they must have common eigenfunctions. Therefore, in determining the effect of the crystal field on the free ion hamiltonian one can utilize a group theory approach. A

brief outline of the application of this theory will be given here. For more of the specifics the reader can consult any of the references.²⁰⁻²²

The S_4 symmetry group is composed of four separate symmetry operations. These are E (the identity operation), C_2 (a rotation of π), S_4 (a rotation of $\pi/2$ followed by a reflection), and S_4^{-1} (the inverse of S_4). A multiplication table can then be constructed showing that any combination of these operations is identical to one of the four basic elements. Each of these symmetry operations can be mapped into four separate irreducible matrix representations Γ_1 , Γ_2 , Γ_3 , and Γ_4 , each of which will satisfy the multiplication table. These last two, Γ_3 and Γ_4 , are actually complex conjugates of one another and are therefore degenerate. They will both be referred to as $\Gamma_{3,4}$.

The trace of each matrix in a particular representation is called its character (χ_{sk}) where s refers to a particular representation and k refers to a particular symmetry operation. A character table can then be written out as follows.²³

Table 1. Character Table of the S_4 Group

Γ_s	Character (χ_{sk})			
	E	C_2	S_4	S_4^{-1}
Γ_1	+1	+1	+1	+1
Γ_2	+1	+1	-1	-1
Γ_3	+1	-1	-i	+i
Γ_4	+1	-1	+i	-i

The energy levels are characterized by the total angular momentum J, and therefore have a degeneracy of $2J+1$. The effect of the crystal field is to remove

some of this degeneracy, making the previously irreducible representation (Γ_i) of a particular energy level reducible. The number of times each new irreducible representation (Γ_s) is contained in the reducible representation (Γ_i) is given by ^{20,21}

$$p_{sj} = \frac{1}{n} \sum_{k=1}^c h_k \chi_j(\alpha_k) \chi_{sk}^* \quad (2.14)$$

Here, n is the number of operations, h_k is the number of times an element occurs, c is the number of irreducible representations, and $\chi_j(\alpha_k)$ is the character of each operator for a particular reducible representation Γ_i with respect to the group of three dimensional rotations (R_3). These can be calculated from ^{20,21}

$$\chi_j(\alpha_k) = \frac{\sin (2J+1) \alpha/2}{\sin (\alpha/2)} \quad (2.15)$$

Using equations (2.14) and (2.15) along with Table 1 gives the following table of crystal field splittings for the J multiplets.

Table 2. Crystal Field Splittings of J Multiplets

J	Occurrence of each Irreducible Representation		
	Γ_1	Γ_2	$\Gamma_{3,4}$
0	1	0	0
1	1	0	1
2	1	2	1
3	1	2	2
4	3	2	2
5	3	2	3
6	3	4	3
7	3	4	4
8	5	4	4
9	5	4	5

Thus, it has been shown how a particular energy level would have some of its $2J+1$ degeneracy removed by the crystal field. As an example, consider the 7F_6 ground state term of Tb^{3+} . Using table 2 above we can see that it would have 10 sublevels, 3 being of Γ_1 , 4 being of Γ_2 , and 3 being of $\Gamma_{3,4}$. This is represented with the expression $3\Gamma_1 + 4\Gamma_2 + 3\Gamma_{3,4}$.

Selection Rules

The primary mechanism for optical transitions is by electric dipole interactions. Magnetic dipole and electric quadrupole interactions are also allowed, but their relative magnitudes are far less than those of electric dipole transitions. Just as the energy eigenstates transform as certain representations, so do the electric and magnetic dipole operator vector components. The components needed for

determining the selection rules are x , y , and z for the electric dipole operator and L_x , L_y , and L_z for the magnetic dipole operator. The representations of these for a particular symmetry can be found in Tinkham's book.²⁰

For a TSE process the transition is actually two single-photon transitions. Consequently, two applications of the single photon selection rules are necessary. Determining these selection rules involves operating on the initial (or intermediate) state with the electric dipole operator and looking at the resultant representation of this product. This can be done by using the group multiplication tables found in Koster's book.²³ Then, if the final state has the same representation as that of this product, the transition will be allowed. Table 3 can be constructed by applying this procedure to all representations.

Table 3. Selection Rules in S_4 Symmetry with Electric Dipole Transitions

	Polarization with respect to the crystal axis		
	Γ_1	Γ_2	$\Gamma_{3,4}$
Γ_1		π	σ
Γ_2	π		σ
$\Gamma_{3,4}$	σ	σ	π

Here, π and σ refer to the electric field of the laser radiation being parallel or perpendicular to the crystal axis, respectively. In these TSE experiments, the first laser frequency (Ω_1) was π polarized to match the transition from the 7F_6 (Γ_2) ground state to the lowest 5D_4 (Γ_1) intermediate state (see Figure 1). Then, the second laser frequency (Ω_2) was either π or σ polarized to allow for a transition to either a Γ_2 or a $\Gamma_{3,4}$ final state, respectively. The lowest 5D_4 (Γ_1) state is used as the intermediate step to ensure that no other intermediate states are populated.

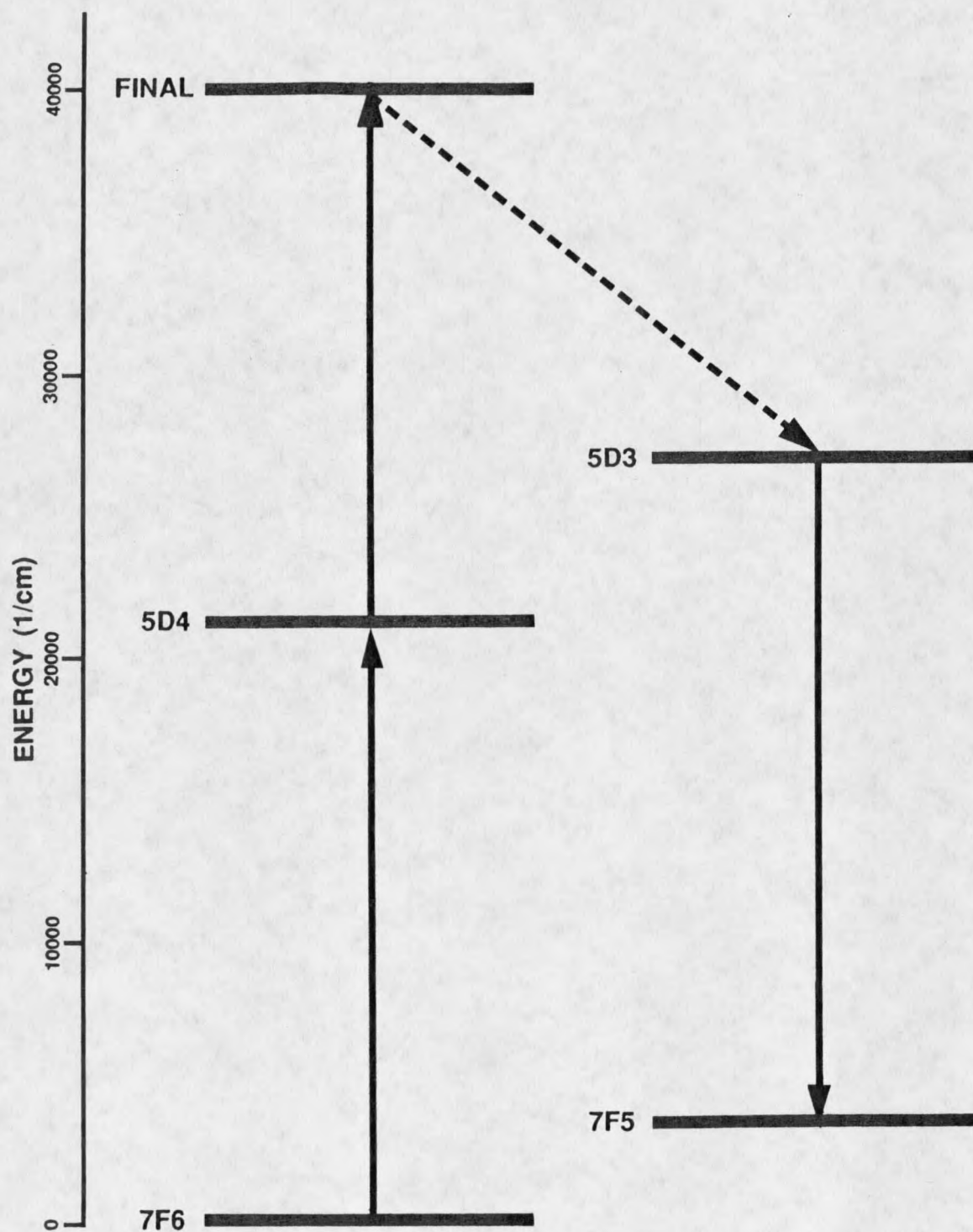


Figure 1. Partial energy level diagram of the Tb^{3+} ion for the two-step excitation experiments.

Since the 5D_4 intermediate state has a representation of Γ_1 , the observation of Γ_1 final states is not allowed by TSE. This is where a complementary aspect of TPA comes in.

The selection rules for a TPA process are determined in much the same way as those of the single photon transitions.^{24,25} In fact, they are identical to those of Raman scattering and are listed in Table 4 for a transition from a Γ_2 initial state.

Table 4. Selection rules for direct two-photon absorption

	Polarization with respect to the crystal axis		
	Γ_1	Γ_2	$\Gamma_{3,4}$
Γ_2	$\sigma\sigma, \pi\sigma, \sigma\pi$	$\pi\pi, \sigma\sigma, \pi\sigma, \sigma\pi$	$\pi\sigma, \sigma\pi$

In these direct two-photon transitions only one laser was used with an energy of $\hbar\Omega$. The transition would take place whenever $2\hbar\Omega$ matched the transition energy (see Figure 2). The labels $\pi\pi$ and $\sigma\sigma$ refer to the electric field of this one laser being parallel or perpendicular to the crystal axis. The labels $\pi\sigma$ and $\sigma\pi$ refer to the electric field being a linear combination of both polarizations. Using this last polarization configuration, the Γ_1 final states, as well as all the others, can be determined. It is a simple matter to classify the crystal field sublevels through a process of elimination.

Since two-photon transitions are very weak, it should be mentioned here how they were detected. Whenever a transition took place, the 410 nm anti-stokes ultraviolet fluorescence cascading from the 5D_3 state to the 7F_5 state could be observed (see Figures 1 and 2). This served as a very sensitive indicator of

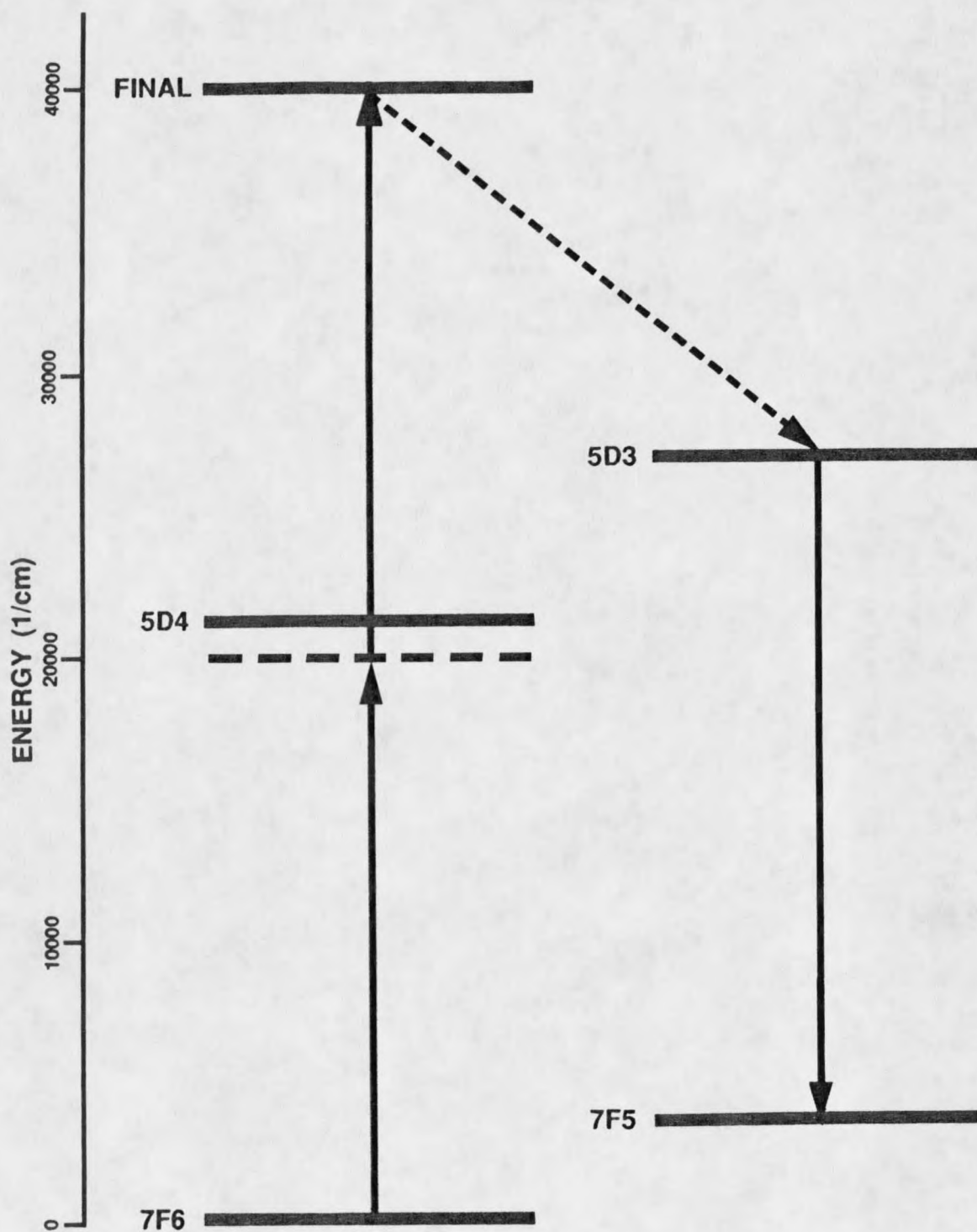


Figure 2. Partial energy level diagram of the Tb^{3+} ion for the direct two-photon absorption experiments.

the presence of a TPA transition. Experimental details will be described in the next chapter.

The selection rules determined via S_4 point group theory are by no means complete. One still has to consider the S, L, and J selection rules for the matrix elements in equation (2.8).

The initial, intermediate, and final states in equation (2.8) result from the odd terms of the crystal field operator, equation (2.13), admixing the original $4f^8$ states with those of the $4f^75d$ and $4f^75g$ configurations. This is the configuration mixing spoken of earlier. Then, through the Wigner-Eckart theorem²¹ and the Judd-Ofelt approximation, we arrive at the selection rules

$$\Delta S = 0; \Delta L \leq 6; \Delta J \leq 6 \quad (2.16)$$

for single-photon transitions. For the direct two-photon transitions the operator within equation (2.11) is a second-rank tensor, giving rise through the Wigner-Eckart theorem to the selection rules for pure configurations as

$$\Delta S = 0; \Delta L \leq 2; \Delta J \leq 2. \quad (2.17)$$

Recent work concerning resonance enhancement may imply that these last symmetry rules could be broken as a result of mixed configurations.⁹ Evidence to support this will be given in Chapter 4.

CHAPTER 3

EXPERIMENTAL ARRANGEMENT

Two experimental methods were utilized to determine the sublevels of the manifolds in question, these being direct two-photon absorption and two-step excitation. Both of these methods are complementary spectroscopic techniques as were described in Chapter 2. However, due to experimental difficulties which will be described in Chapter 4, the two-step excitation experiments were the primary means for gaining information on the majority of the manifolds in question.

In this chapter, these two experimental methods will be described in detail, along with the opto-galvanic calibration system used to calculate the energies and the computer control of the experiments.

Two-Photon Experiments

At the heart of the matter in any laser spectroscopy lab are the lasers themselves. Therefore, these will be described first before moving on to how they were utilized.

Lasers

Either one or two tunable dye lasers were used for these experiments, depending on which type of experiment was being done. For the direct two-photon

absorption experiments only one dye laser was used since the final excited state is created by the simultaneous absorption of two photons of the same energy coming from the same source. In the two-step excitation experiments two dye lasers were used. The first dye laser served to excite the atom from the ground state to a real intermediate state while the second laser was used to excite it from this real intermediate state to the final state.

The tunable dye lasers were based on a design conceived by T.W. Hänsch in 1972.²⁶ Each of these tunable dye lasers was pumped by its own pulsed nitrogen gas laser which operated at a wavelength of 337.1 nm, a pulse rate of 6 Hz and a pulse width of 10 nanoseconds. The timing between the two nitrogen gas lasers could be varied manually with a timing stabilization unit and was measured with a Tektronix 7912AD Programmable Digitizer. This was useful in later experiments when trying to determine the origins of certain ambiguous transitions, which will be discussed in detail in the next chapter. Both the tuneable dye lasers and the pulsed nitrogen gas lasers were constructed by Dr. R.L. Cone and his previous graduate students.

Typically, the nitrogen gas lasers were producing a power of 200-300 kW. Bearing in mind the pulse width of 10 nanoseconds, one realizes that the actual amount of energy in one pulse is on the order of only several millijoules. The beam, upon emerging from the nitrogen gas laser, is spatially rectangular with dimensions of roughly 1 cm in height and 2.5 cm in width. The bottom half of this beam was split off and sent to the oscillator section of the tunable dye laser while the top half of the beam continued on to the amplifier section of the dye laser. This pumping configuration was essentially the same for both dye lasers (see Figure 3).

Both dye lasers had an oscillator cavity defined by a partially reflecting

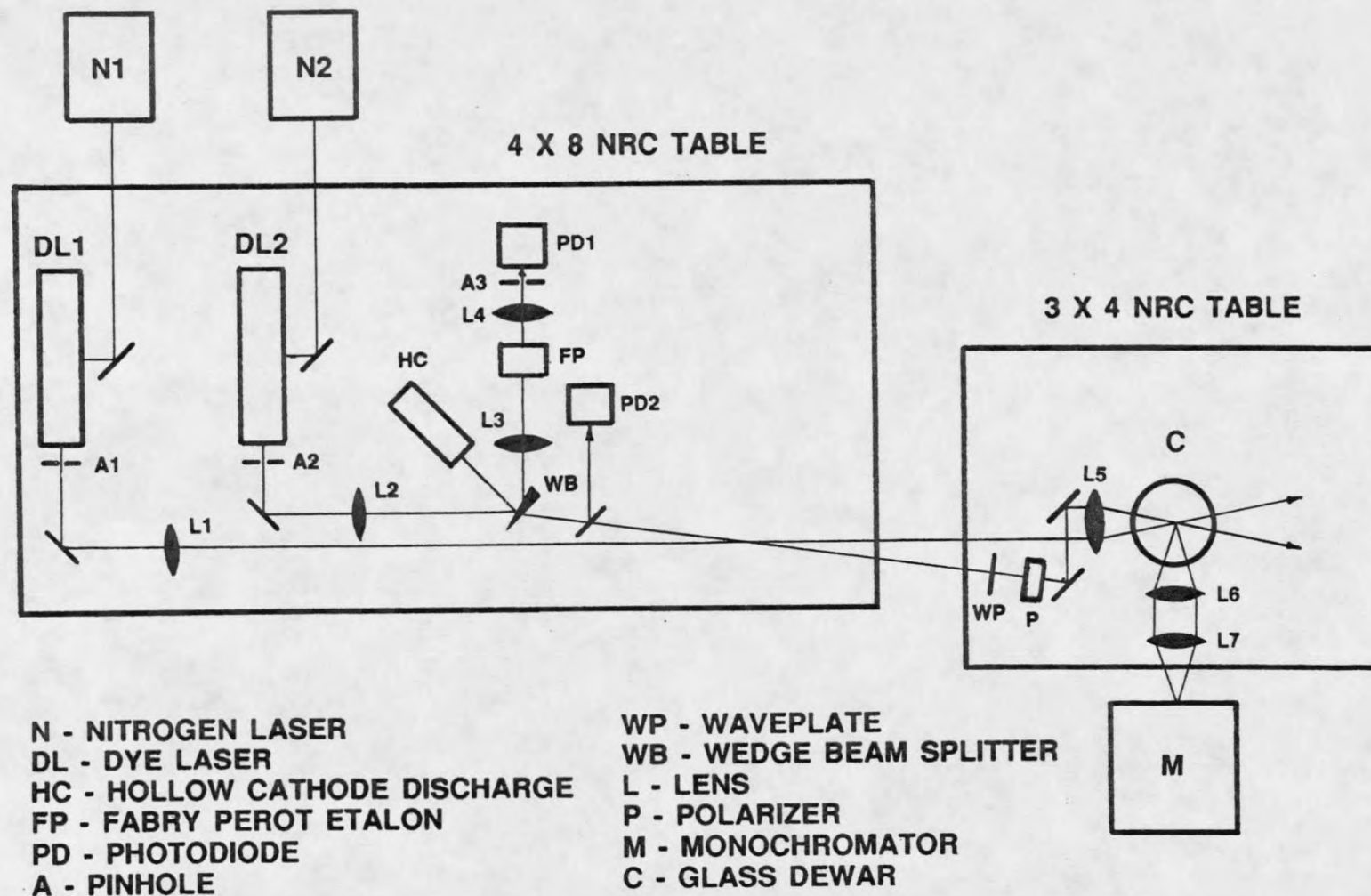


Figure 3. Experimental setup for the two-photon experiments

mirror at the front end and a 632 lines per millimeter reflection diffraction grating at the rear end. To fully illuminate the diffraction grating, a telescopic beam expander was inserted between the dye cell and the diffraction grating. The diffraction grating itself was mounted on a gimbal-type mount with micrometer adjustments for movement in both the vertical and horizontal planes. Motion in the horizontal plane would change the angle of the incident beam upon the grating, therefore changing the wavelength at which the dye laser would oscillate (within the constraints defined by the gain curve of the dye solution), while motion in the vertical plane would fine tune the lasing itself. It should be mentioned here that if the horizontal plane of rotation was not exactly parallel to the surface of the optics table the laser power would diminish considerably during a scan if not monitored. A spurious reflection from the main beam was monitored with a photodiode and oscilloscope, and periodically a vertical adjustment was made to the diffraction grating mount to maximize the laser power.

The dye solution was contained in cuvette-type dye cells. Motor driven magnetic stirrers maintained thermal homogeneity of the dye solution throughout the experiments. The dyes used in these experiments were Exciton 7D4TMC in a 10^{-2} molar solution of p-dioxane and Exciton Coumarin 500 in a 10^{-2} molar solution of ethanol. Both dye lasers were mounted on a Newport Research Corporation 4' x 8' optical table.

Direct Two-Photon Absorption Experiments

As was stated before, the direct two-photon absorption experiments required the use of only one tunable dye laser, specifically dye laser DL2 in Figure 3. The horizontal micrometer adjustment on the diffraction grating mount was attached to a stepper motor which was computer controlled. This way, the wavelength of the dye

laser could be changed at a continuous rate by moving the grating through a prescribed angle determined by the number of revolutions of the stepper motor. The output power of this laser was typically between 15-20 kW.

The beam, upon exiting the dye laser, continued on to a wedge beam splitter (WB) where two beams were split off to the calibration system (which will be described in a later section). This wedge beam splitter also refracted the beam so as to throw it slightly off the optic axis. The main part of the beam continued on to another beam splitter where a small part of the beam was monitored with a sample and hold photodiode (PD2) for recording the relative laser power. This laser power was later used for normalizing the fluorescence signal from the sample. The beam continued on to a $\lambda/2$ waveplate (WP) and a polarizer (P) so that the polarization of the incident radiation could be selected to be parallel ($\pi\pi$) or perpendicular ($\sigma\sigma$) to the crystal axis as well as a linear superposition of both these polarizations ($\pi\sigma/\sigma\pi$). A pair of 90° prism reflectors were used to bring the beam back on the optic axis and the beam was then focused onto the crystal with a 250 mm focal length lens, giving a minimum waist diameter of approximately 0.06 mm at the location of the crystal.

The crystal sat inside a glass cryostat (C) designed and built by Pope Scientific, Inc. which had optically transparent windows for the incoming laser beam as well as for observing the resulting UV fluorescence. The crystal was continuously kept at pumped liquid helium temperatures ($T \approx 1.3$ K) for all of these experiments.

The fluorescence signal was collected by a 75 mm lens (L6) and focused onto the entrance slit of a MacPherson Model 218 f/5.3 monochromator (M) by a 200 mm focal length lens (L7). This monochromator was tilted 90° so that its entrance slit was parallel with the long path of the laser beam through the sample. This way, the maximum amount of fluorescence could be collected. A CS 5-58

glass bandpass filter was placed at the entrance slit of the monochromator to help filter out any reflected laser radiation and lower energy fluorescence from the 5D_4 level.

The UV fluorescence due to a direct two-photon absorption process was quite weak so a gated photon counting system was utilized for observing this signal (Figure 4) in order to eliminate the dark counts. An RCA C31034 photomultiplier tube (PMT) was mounted at the exit slit of the monochromator and was contained in a thermoelectric and water cooled housing manufactured by Pacific Precision Instruments. The PMT was operated at -1480 volts for the TPA experiments. The signal output from the PMT was fed into a preamplifier and then into a homemade discriminator and TTL pulse generator. These pulses were then gated to ensure that only those pulses coming from a fluorescence signal were passed. This gate was 3 milliseconds in duration with a delay of 5 microseconds. The remaining pulses were then counted by a Northern Scientific Model 575 Multichannel Analyzer. At the completion of each experiment these data were transferred to a PDP-11/03 microcomputer and later to a VAX 11/780 computer or VAX 8850 cluster for data analysis.

A brief word on alignment should be mentioned here before moving on. When the 5D_4 (Γ_1) state at 20553.5 cm^{-1} is populated, the resulting yellow fluorescence (18358.5 cm^{-1}) from this state to the ground state term 7F_5 is very strong and plainly visible with the naked eye from several meters away. The image of this fluorescence was easily placed on the monochromator entrance slit by the collection optics. The monochromator was then tuned to observe this yellow fluorescence and the signal from the PMT was fed directly into a Keithley Picoammeter. The needle deflection on the picoammeter thus gave a very accurate means of fine tuning the alignment of the beam and collection optics. Fortunately,

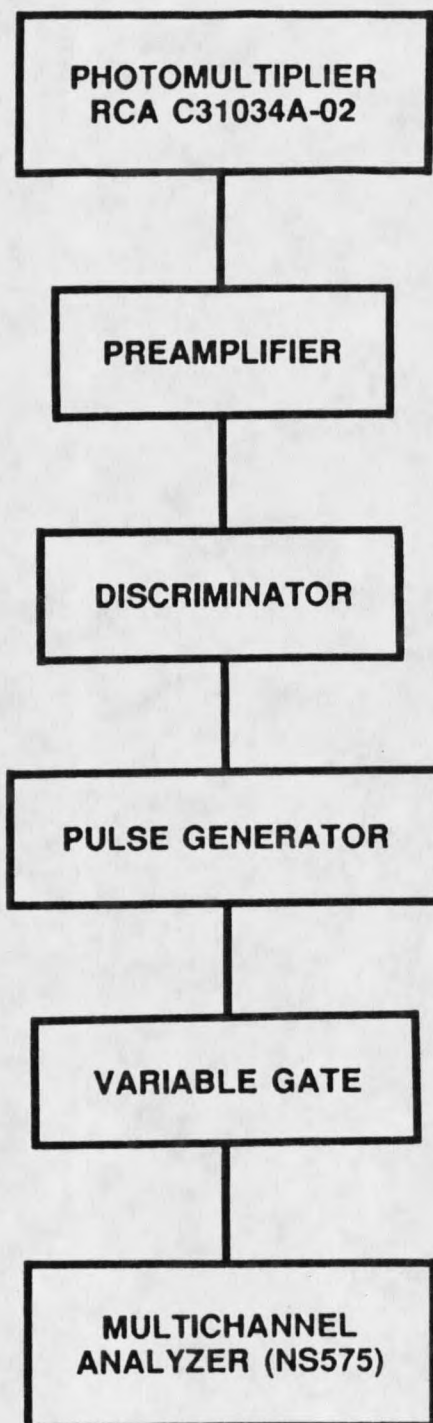


Figure 4.

Photon counting system employed in the direct two-photon absorption experiments

the two laser dyes used both lased at 20553.5 cm^{-1} and this process of alignment could always be used. After the alignment was completed the monochromator was tuned back to observe the UV fluorescence, the CS 5-58 filter was reinserted, the laser was tuned to the starting energy of the scan, and the experiment was started.

Two-Step Excitation Experiments

These experiments required the use of both dye lasers DL1 and DL2 in Figure 3. Laser DL1, which will hereby be referred to as Ω_L , was tuned to match the the real intermediate 5D_4 state at 20553.5 cm^{-1} . This energy was held constant throughout these experiments. Laser DL2, which will hereby be referred to as Ω_H , was then scanned through an energy region in question. The UV anti-Stokes cascade fluorescence would then take place whenever $\Omega_L + \Omega_H$ resulted in a two-step transition.

The mechanics of this setup were almost identical to that of the direct two-photon absorption experiments, the only difference being the introduction of an additional dye laser. As stated before, this dye laser was pumped by its own pulsed nitrogen gas laser and the timing between the two lasers could be varied. The first set of two-step excitation experiments had Ω_H arriving after Ω_L on a time scale of the order of a microsecond, while the second set of experiments had Ω_H firing 0 to 5 nanoseconds after Ω_L . If Ω_H was set to fire before Ω_L the fluorescence signal would vanish. This served as a good test to see if the fluorescence signal was due to a two-step excitation process.

The laser powers were typically 5 to 10 kW for Ω_L and 15 to 20 kW for Ω_H . The polarization of Ω_L was left parallel to the crystal axis and the polarization of Ω_H was set either parallel ($\pi\pi$) or perpendicular ($\pi\sigma$) to the crystal axis.

The last remaining problem in the setup for the two-step excitation experiments was aligning the two beams in the sample. The location of the two beams could be roughly estimated by observing the sample through a telescope focused at infinity along with a 15 cm focal length lens and appropriate filters (to protect the eye). Based on previous experiments, the approximate energies of some strong two-step transitions were known.¹² The frequency Ω_H was then tuned so that $\Omega_L + \Omega_H$ matched this energy and the resulting fluorescence signal was again viewed with the picoammeter while the positions of the beams were fine tuned. Then, as before, Ω_H was tuned to the starting energy of a scan, and the experiment was started.

In the first set of two-step excitation experiments the original photon counting system was employed. In later experiments, a different system was employed since the fluorescence signal was quite strong and sensitivity was not a major problem. In these later experiments the image of the crystal was focused onto the entrance slit of the same MacPherson Monochromator. A Hamamatsu R928 photomultiplier tube, which was operated at -1200 volts, was placed at the exit slit and the resulting signal was fed into a 1 M Ω input of a Princeton Applied Research Corporation model 162 Boxcar Integrator where the signal was gated and integrated. The gate was set at 5 milliseconds in length with a 5 microsecond delay. The output from the boxcar integrator was then monitored by the PDP-11/03 computer, that was controlling the experiment.

Calibration System

In both sets of two-photon experiments, two beams from laser DL2 were sent to a calibration system consisting of a hollow cathode discharge and a Fabry-Perot etalon. The optogalvanic effect was then used,²⁷⁻²⁹ along with central spot

scanning,³⁰ in calculating the transition energies. These two techniques are described below.

Opto-galvanic Effect

A portion of the beam was directed coaxially into the hollow uranium cathode of a neon discharge lamp (HC). This lamp was powered by a constant current source operating between 20 to 25 milliamps. The voltage across the discharge was then measured via a coupling capacitor by a sample and hold circuit, and the subsequent signal was recorded by the computer.

The opto-galvanic effect works on the principle that when atoms in a discharge absorb electromagnetic radiation, they become more or less easily ionized. This results in an impedance change in the discharge and manifests itself as a change in voltage when the lamp is powered by a constant current supply. If the absorbed radiation results in a transition to a higher energy state, whereby ionization due to electron collisions can proceed more easily, the impedance of the discharge decreases. Alternatively, the absorbed radiation can cause a depletion of a long lived meta-stable state resulting in the atoms being less easily ionized. This results in an increase in the impedance of the discharge.

By far, however, the majority of the transitions resulted in the atoms being more easily ionized. This produced a decreasing voltage and a subsequent negative signal. For ease of viewing and data analysis these data were made positive by multiplying by a constant negative factor.

Central Spot Scanning

The second portion of the beam from DL2 was expanded (L3) and projected into a Fabry-Perot etalon (FP). The resulting interference pattern was focused (L4) onto a screen with a pinhole at the central spot (A3). The central spot intensity

was then monitored by a sample and hold photodiode (PD1) and recorded by the computer.

The interference condition for a Fabry-Perot etalon is $m\lambda = 2nd\cos\theta$. With central spot scanning this interference condition simply becomes $m\lambda = 2nd$, which is a constant governed strictly by the construction of the etalon. Based on comparisons with the hollow cathode discharge data, this constant was calculated to be 6.00 mm, corresponding to an etalon thickness of 3.00 mm. The energy difference between consecutive interference conditions is then

$$\Delta E = \frac{1}{\lambda_m} - \frac{1}{\lambda_{m-1}} = \frac{m}{2nd} - \frac{m-1}{2nd} = \frac{1}{2nd} = 1.667 \text{ cm}^{-1}.$$

The hollow cathode discharge, along with the central spot scanning, thus provided an excellent means of calculating the transition energies observed since all the data were taken simultaneously. A visual representation of this is given in Figure 5. The hollow cathode discharge data are tabulated,³¹ so all one has to do to determine the laser's energy is count the number of interference conditions (b) between a uranium or neon transition (a) and a two-photon transition (c). A linear fit between interference conditions was assumed, and the energy of any particular two-photon transition was calculated from several uranium or neon transitions. This way, an idea of the relative error could be obtained.

Computer Control

At the heart of the control of these experiments was a Digital Equipment Corporation PDP-11/03 (LSI-11/2) microcomputer. This computer was interfaced to the experimental apparatus as shown in Figure 6.

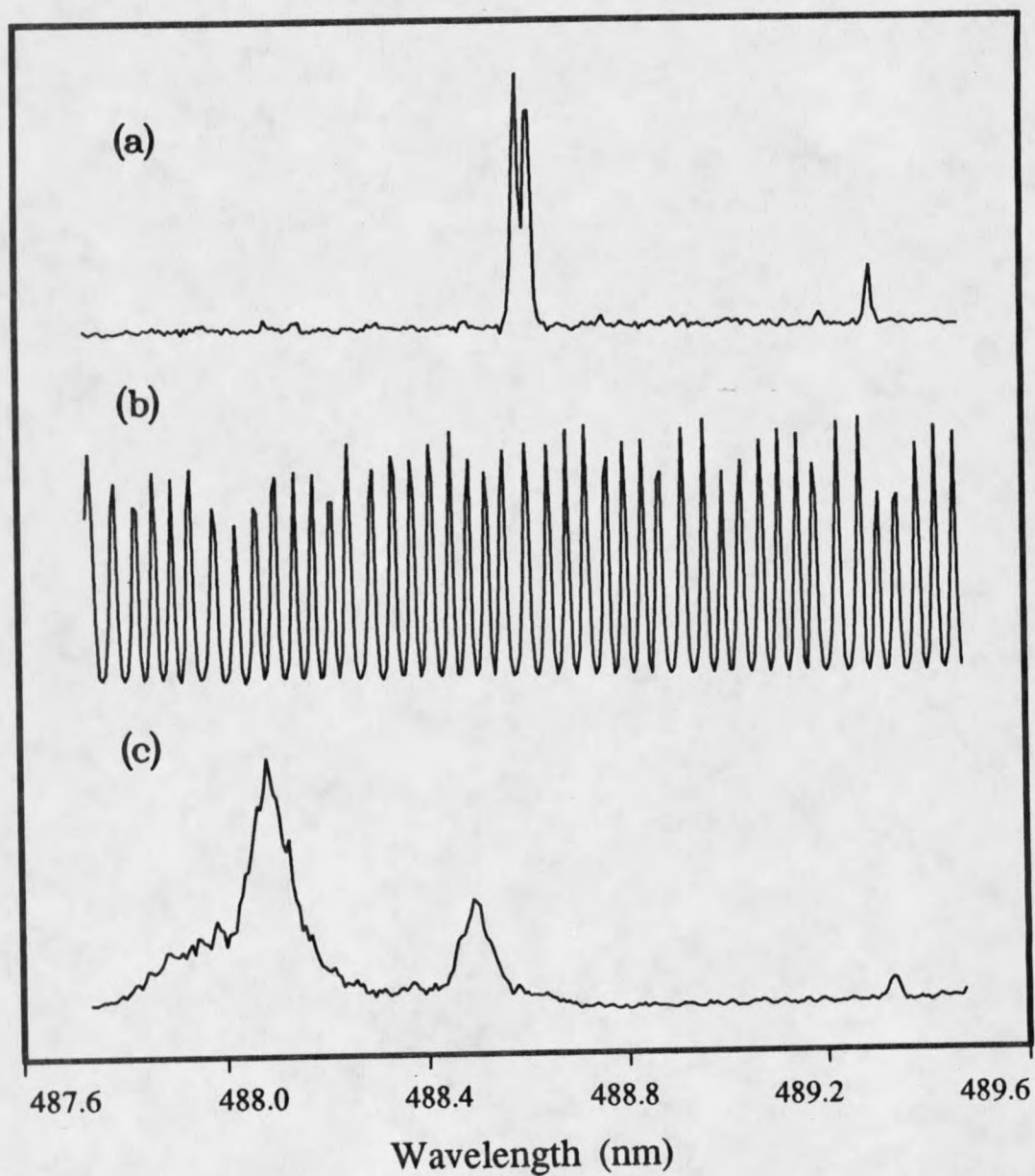


Figure 5. Spectra of the calibration system. (a) Hollow cathode discharge impedance. (b) Fabry-Perot etalon central spot intensity. (c) Two-photon transition spectra.

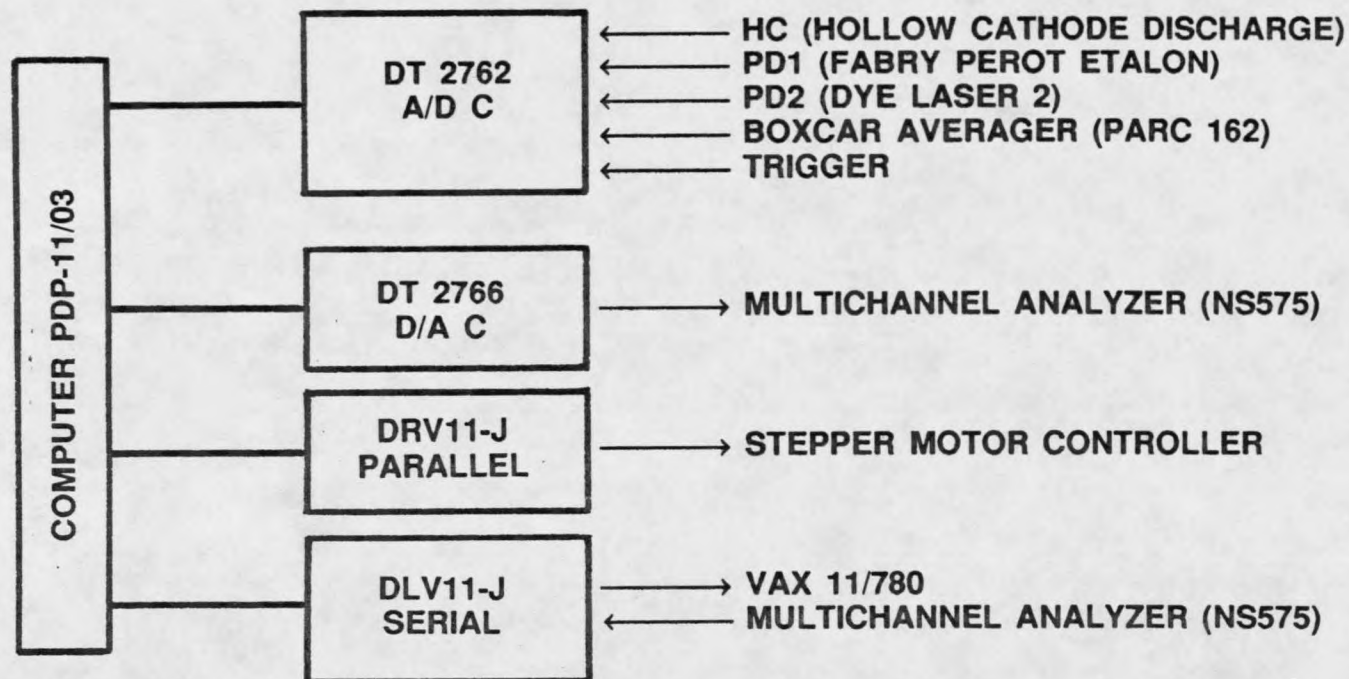


Figure 6. Computer control of the experiments.

In the direct TPA experiments, several variables were monitored with an analog to digital converter. These variables were the laser power of DL2, the hollow cathode discharge impedance, the Fabry-Perot central spot intensity, and a trigger pulse from the laser. In the TSE experiments the signal from the PARC 162 Boxcar Integrator was also observed. Two separate signals were sent out to trigger the NS575 Multichannel Analyzer in the TPA experiments by a digital to analog converter. In both the TPA and TSE experiments the computer was interfaced to a stepper motor controller via the parallel port. This stepper motor was used to tune laser DL2 in Figure 3. Finally, the PDP-11/03 computer could accept information which was temporarily stored in the NS575 Multichannel Analyzer and transfer data out to a VAX 11/780 Microcomputer for storage and data analysis. This was done via serial ports.

The software to control the experiments was written in the C computer language on a Whitesmiths C compiler. Both programs allowed the user to vary the resolution, speed, and range of each scan. The TPA program plotted both the hollow cathode discharge impedance and the Fabry-Perot central spot intensity, but it could not plot the TPA signal since those particular data were stored in the multichannel analyzer during the experiments. However, the TSE program plotted all of this information since the TSE signal was directly monitored with the computer. Both programs are listed in the appendix.

CHAPTER 4

EXPERIMENTAL RESULTS

As stated in the Introduction, TSE was the primary means of obtaining information on the energy levels in question. The reason for this is that the Ω_1 laser energy in the TPA experiments had to be kept below the 5D_4 (Γ_1) resonance. If Ω_1 was tuned higher, vibronic absorption led to a population of this state. This resulted in a TSE process taking place which gave such a strong signal that the TPA signal could not be discriminated. This left only the 5K_9 , 5D_2 and 5G_6 levels to be studied by TPA.

In this chapter, the results for the 5G_6 by TPA will be given first, followed by the results of all the subsequent experiments. The measured energy values are compared with a recent computer fitted model constructed by Liu at Argonne National Laboratory which used these experimental energies as a basis for determining the various crystal field and free ion parameters.

5G_6 by TPA

As stated earlier, the 5G_6 level was studied by Huang in 1987.¹¹ The reasons for studying this level again were to check reproducibility, obtain improved accuracy, and to test the efficiency of the experimental apparatus. The energy

region scanned was from 40263 cm^{-1} to 40410 cm^{-1} , so that Ω_1 was tuned from 20131.5 cm^{-1} to 20205.0 cm^{-1} . This is well enough below the $^5\text{D}_4$ (Γ_1) state so that no signal observed was from a TSE process.

The data is presented in Table 5 and Figure 7 for all three polarization configurations. The data corresponds well to that of Huangs, with only minor differences in energy values ($\pm 1.7 \text{ cm}^{-1}$ max).

Table 5. Comparison of Observed Sublevels for the $^5\text{G}_6$ Energy Level by TPA

Representation	E_{Exp} (cm^{-1})	E_{Com} (cm^{-1})	ΔE (cm^{-1})
Γ_1	40341.7	40348.0	-6.3
	40345.9	40350.5	-4.6
	40403.7	40392.2	11.5
Γ_2	40272.7	—	—
	40274.0	40276.8	-2.8
	40323.5	40318.4	5.1
	40388.2	40387.5	0.7
	—	40484.3	—
$\Gamma_{3,4}$	40319.3	40323.2	-3.9
	40352.6	40354.4	-1.8
	—	40446.6	—

Just as in Huang's experiments, only two of the three possible $\Gamma_{3,4}$ sublevels, and all the Γ_1 and Γ_2 sublevels, were identified. Based upon Liu's computer fitted model, there may be a discrepancy in the classification of the two lowest energy Γ_2

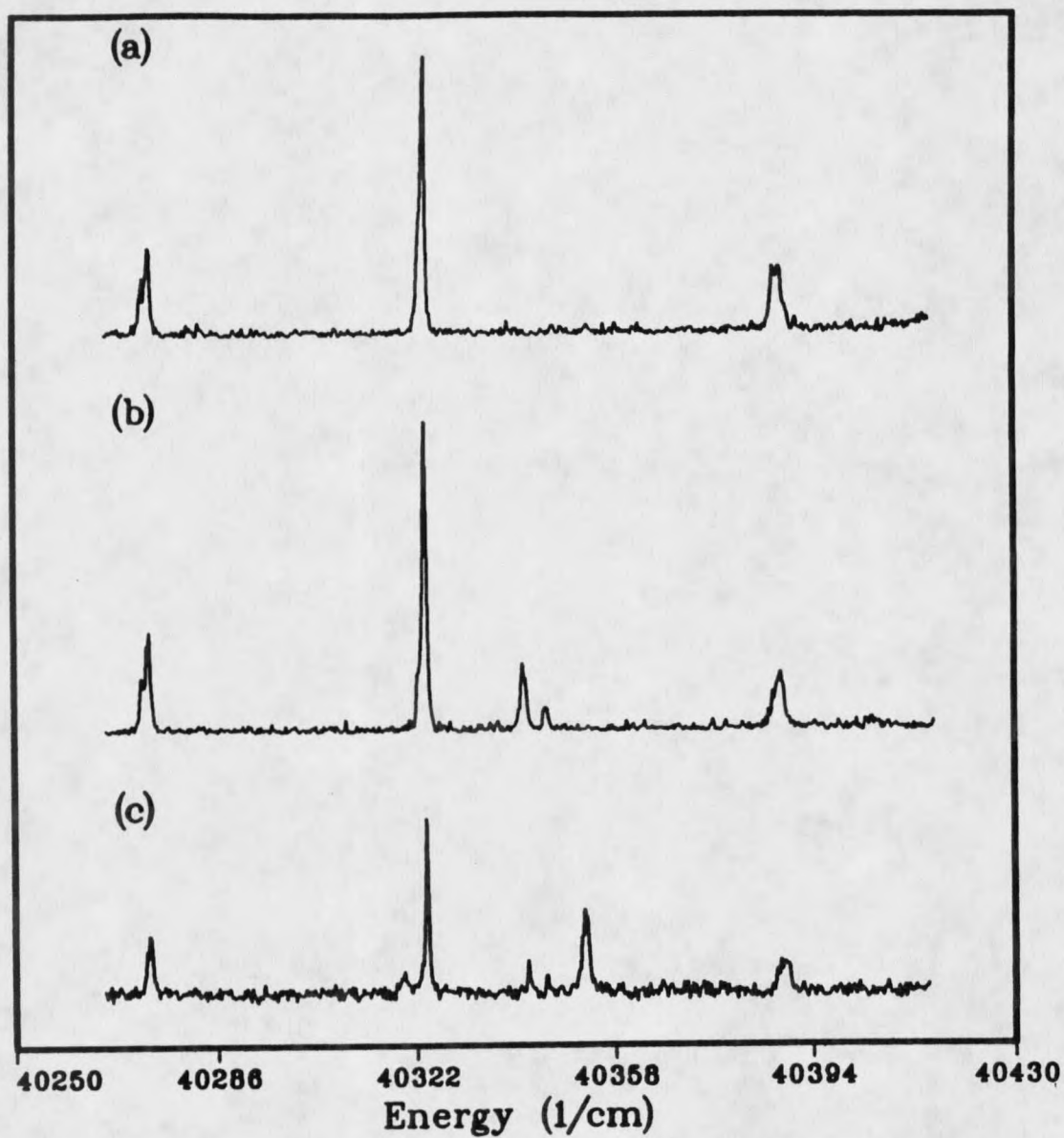


Figure 7. Direct two-photon absorption spectra of the Tb^{3+} ion from ground state $^7\text{F}_6$ (Γ_2) to $^5\text{G}_6$. (a) $\pi\pi$ polarized. (b) $\sigma\sigma$ polarized. (c) $\pi\sigma/\sigma\pi$ polarized.

sublevels which are nearly overlapping. The computer model suggests there may be only one Γ_2 sublevel here while the last one is at an energy of 40484.3 cm^{-1} . The computer fitted model also suggests that the third $\Gamma_{3,4}$ is at an energy of 40446.6 cm^{-1} . Neither of these last two energies were scanned by Huang or myself.

Effect of Laser Timing on TSE Results

The initial TSE experiments had the first laser firing before the second on the order of a microsecond. Since the lifetime of the 5D_4 state is on the order of milliseconds, the TSE signal could be clearly distinguished, and was observed to vanish when either beam was blocked. However, the experimental results showed many more lines than could be accounted for.

The lifetime of this intermediate state allows plenty of time for a non-radiative (phonon) transfer to a slightly lower energy crystal defect level.^{32,33} Therefore, other nearby intermediate states could be populated along with the intrinsic 5D_4 (Γ_1). This would imply that the second step would proceed from more than one intermediate state, resulting in the presence of extra lines. In later experiments, this delay in laser timing was reduced to a few nanoseconds to test this hypothesis. The results, shown in Figure 8 for a representative level, clearly show that some lines dramatically diminished and even disappeared, while others remained the same relative intensity. The $\pi\sigma$ polarization configuration, used in obtaining the data for the 5K_8 level in Figure 8, should result in only four observed sublevels. Figure 8 clearly shows how the laser timing isolated these four sublevels. All of the data presented in this thesis was taken with the laser timing set on the order of a few nanoseconds.

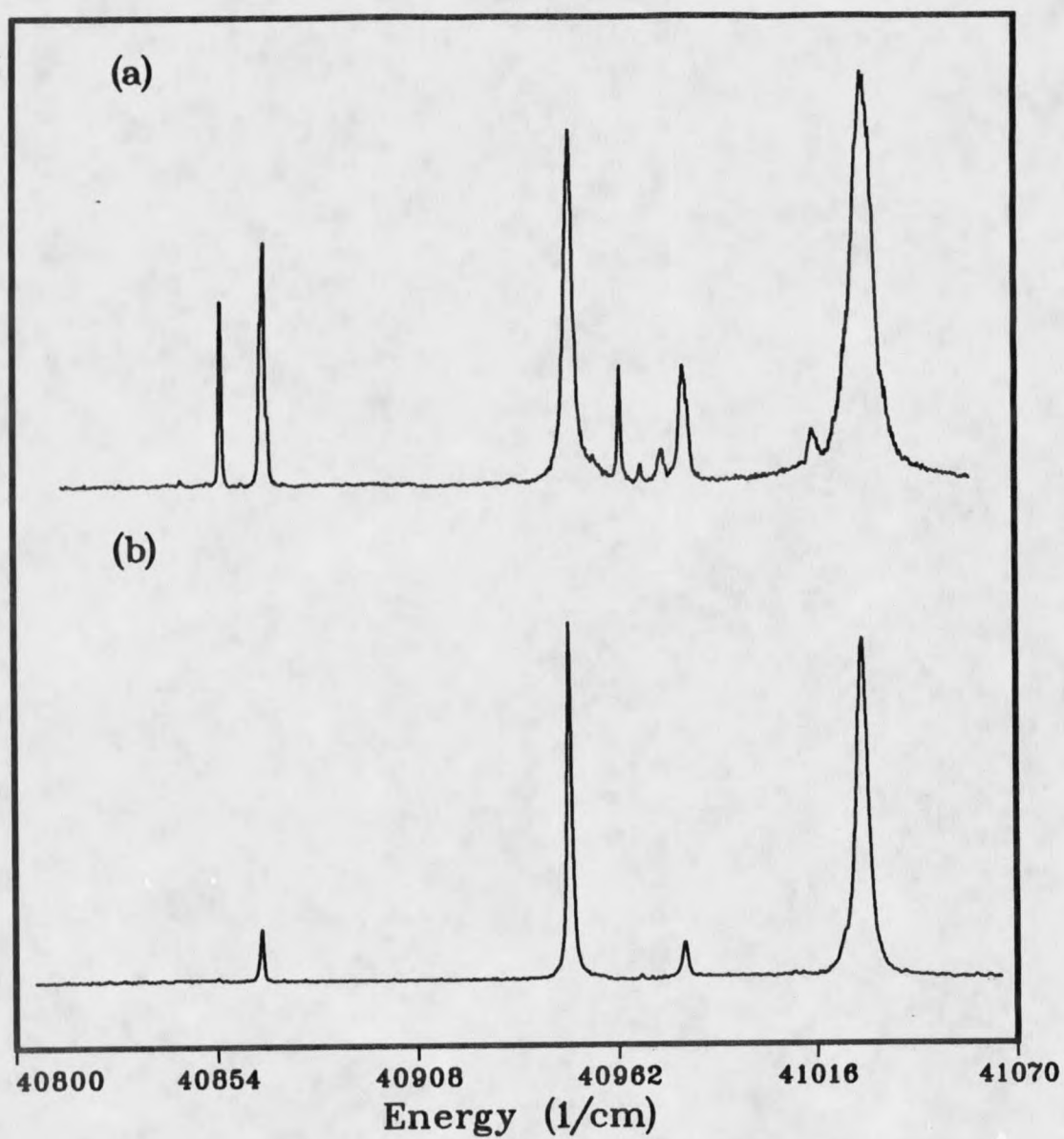


Figure 8. Effect of laser timing on the two-step excitation spectra for the $\pi\sigma$ transition ${}^7F_6 \rightarrow {}^5D_4 \rightarrow {}^5K_8$. (a) 1 μ s delay. (b) 2 ns delay.

5D_2 by TPA and TSE

The 5D_2 level was studied by both TPA and TSE. The selection rules (2.17) imply that this level should be forbidden by TPA, but, as stated previously, Huang's resonant enhancement results⁹ imply that the intermediate states making contributions to this transition may include not only the traditionally considered $4f^75d$ levels far from resonance, but also the $4f^8$ states. This implies that the ΔJ selection rules could go as high as 12 for TPA transitions.

The data is presented in Figure 9 for the TPA results and Figure 10 for the TSE results. The numerical results in Table 6 show that the data agree quite well with the computer fitted model. The one $\Gamma_{3,4}$ sublevel was not discernible in the TPA results, but was identified in the TSE results. Therefore, all the sublevels of the 5D_2 manifold were identified. The difference ΔE is computed from the TSE results when available since these scans had a higher resolution than the TPA scans.

Table 6. Comparison of Observed Sublevels for the 5D_2 Energy Level by TPA and TSE

Representation	E_{TPA} (cm^{-1})	E_{TSE} (cm^{-1})	E_{Com} (cm^{-1})	ΔE (cm^{-1})
Γ_1	39509.6	—	39508.9	0.7
Γ_2	39538.5	39539.6	39540.5	-0.9
	39546.1	39546.6	39547.2	-0.6
$\Gamma_{3,4}$	—	39506.0	39505.6	-0.4

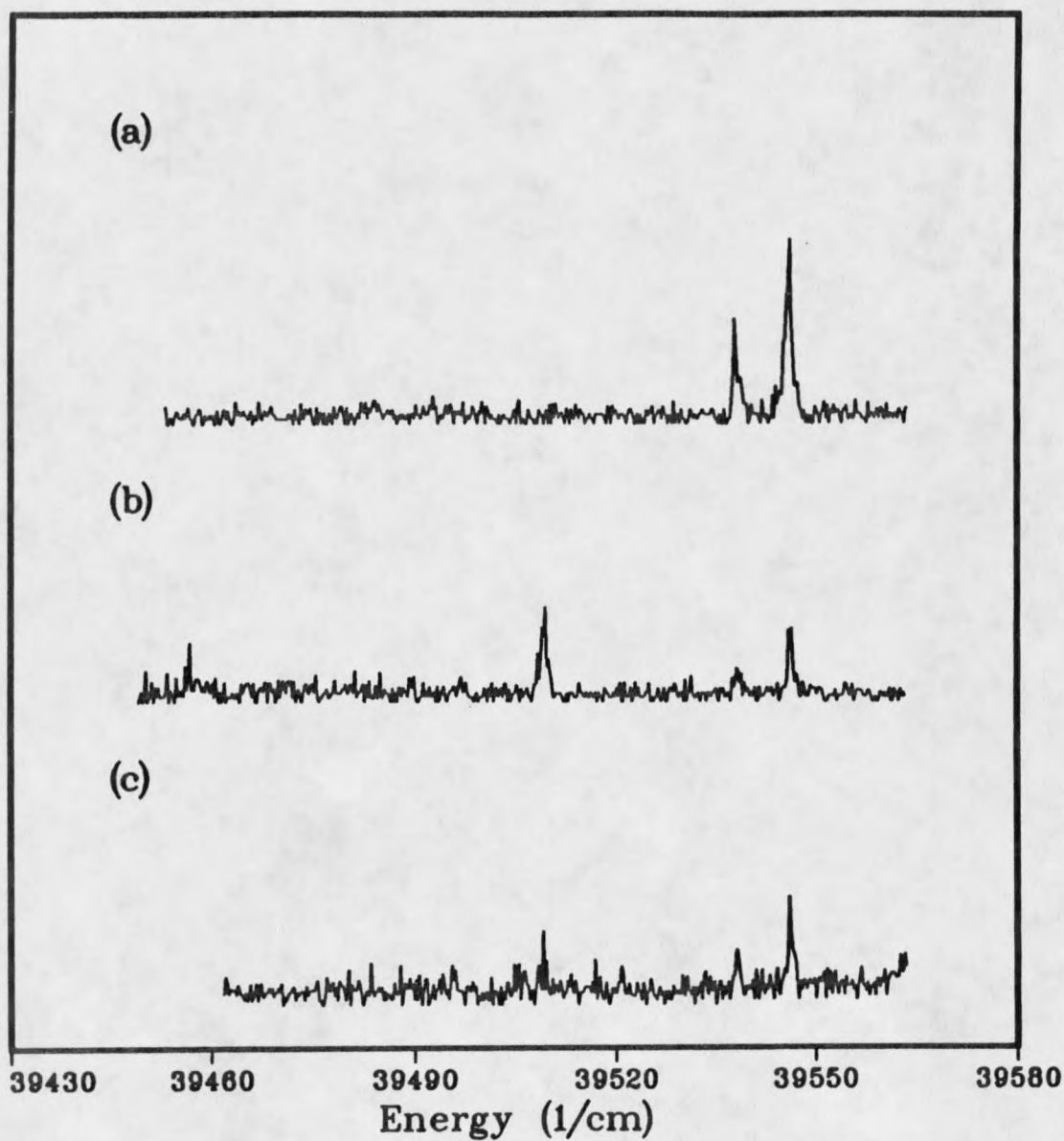


Figure 9. Direct two-photon absorption spectra of the Tb^{3+} ion from ground state $^7\text{F}_6$ (Γ_2) to $^5\text{D}_2$. (a) $\pi\pi$ polarized. (b) $\sigma\sigma$ polarized. (c) $\pi\sigma/\sigma\pi$ polarized.

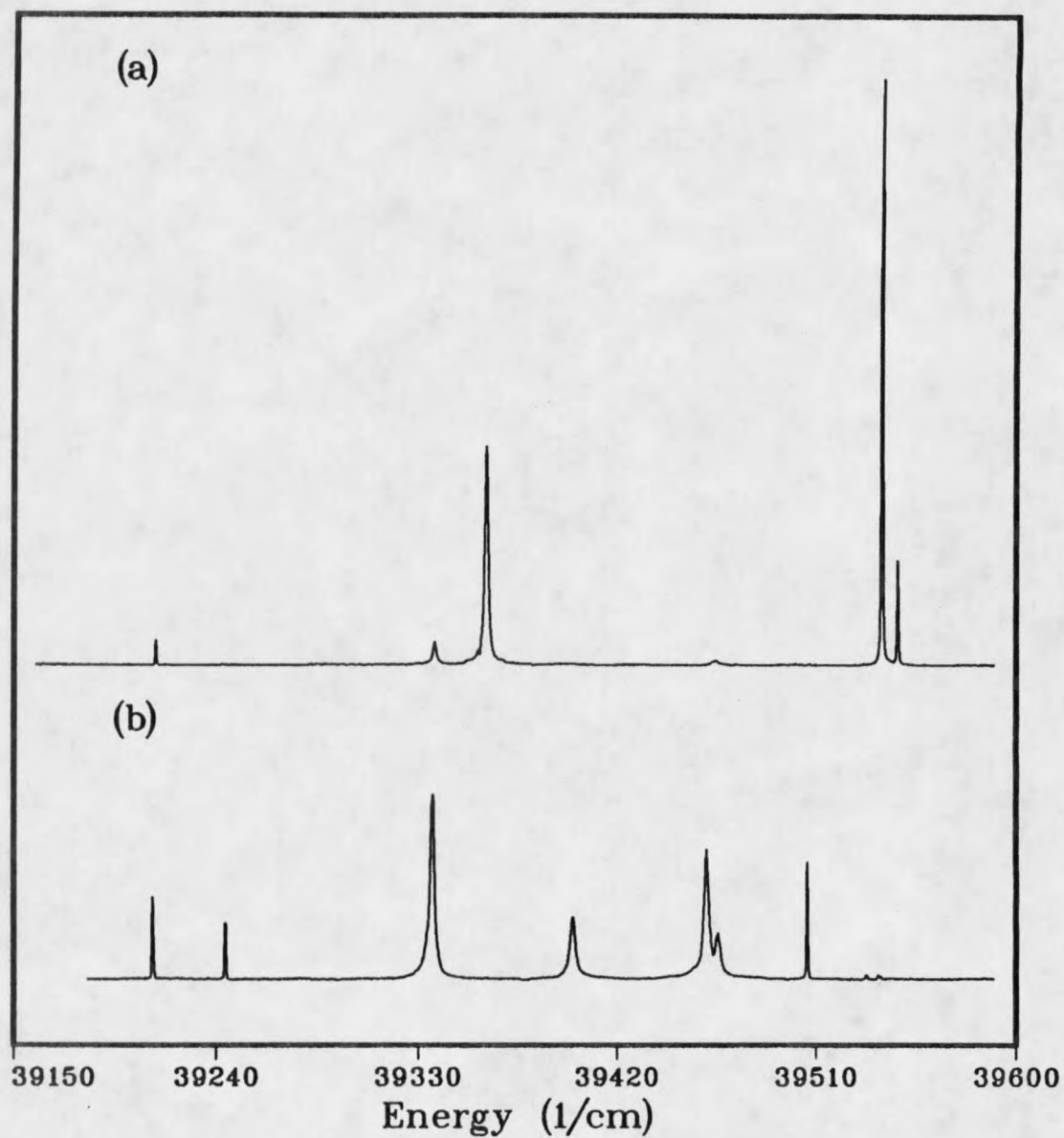


Figure 10. Two-step excitation spectra of the Tb³⁺ ion from ground state ⁷F₆ (Γ₂) to ⁵K₆ and ⁵D₂. (a) ππ polarized. (b) πσ polarized.

5K_9 by TPA and TSE

No definitive results were obtained for the 5K_9 level by TPA. However, a signal was detected at 39242.7 cm^{-1} for the $\pi\pi$ and $\pi\sigma/\sigma\pi$ scans while nothing was observed for the $\sigma\sigma$ scan (see Figure 11). This is contradictory to the selection rules which imply that if a signal is seen in the $\pi\sigma/\sigma\pi$ scan it should also be seen in both the $\pi\pi$ and $\sigma\sigma$ scans. A signal at the same energy was also observed in a later TSE scan with a 1 microsecond laser timing delay. When this delay was shortened to 2 nanoseconds this signal was no longer observed. The origin of the transition is not yet determined, but the TSE scans seem to suggest that it is not an intrinsic level of the 5K_9 .

All of the four allowed Γ_2 sublevels were identified in the $\pi\pi$ scan by TSE. The selection rules predict the presence of five $\Gamma_{3,4}$ sublevels, but the $\pi\sigma$ scan revealed six lines. The data and classifications are presented in Table 7 along with comparisons with the computer fitted model. Both the 5D_2 and 5K_9 levels were scanned in the same experiment and the experimental results are shown in Figure 10.

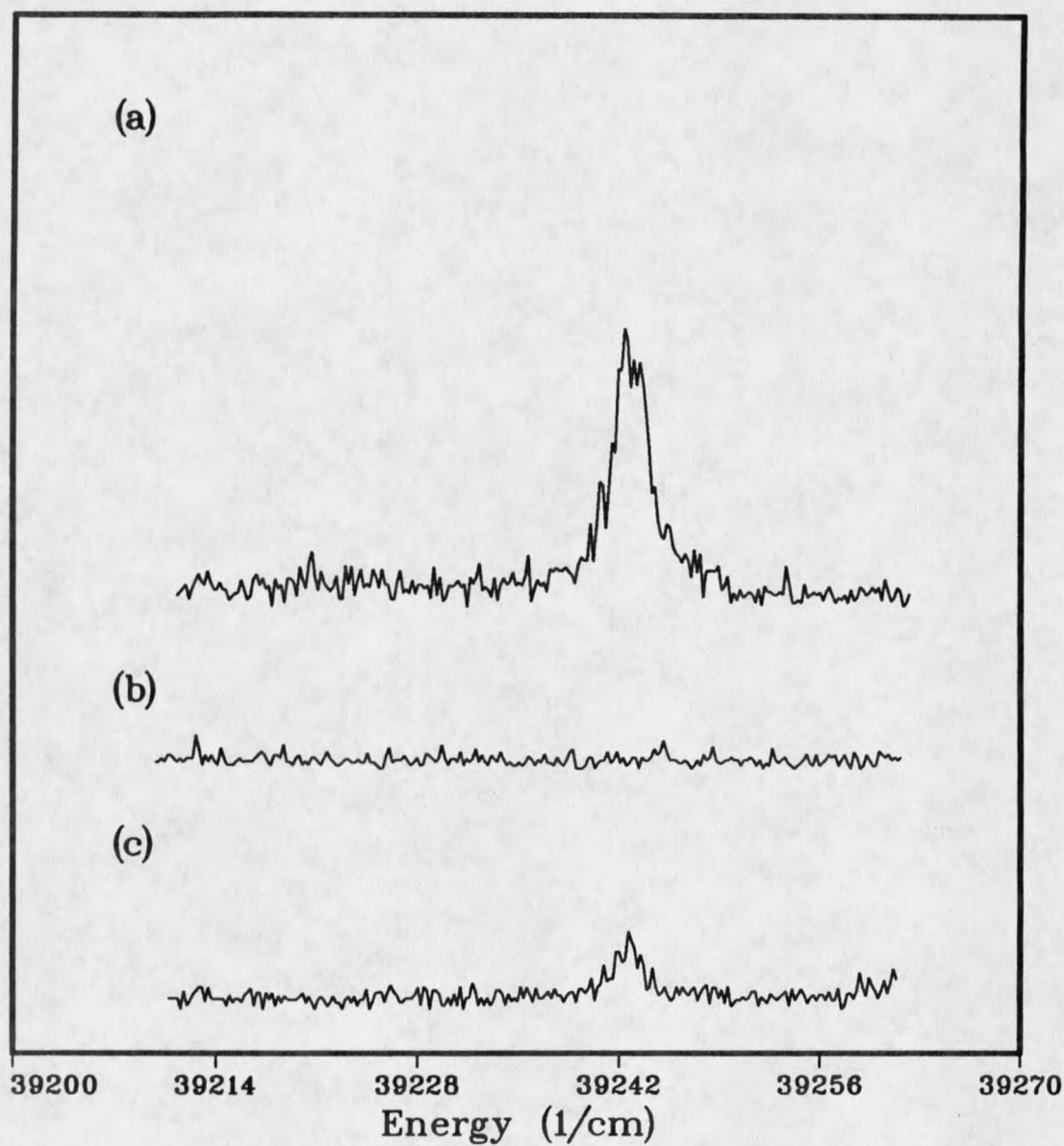


Figure 11. Direct two-photon absorption spectra of the Tb^{3+} ion from ground state ${}^7\text{F}_6 (\Gamma_2)$ to ${}^5\text{K}_9$. (a) $\pi\pi$ polarized. (b) $\sigma\sigma$ polarized. (c) $\pi\sigma/\sigma\pi$ polarized.

Table 7. Comparison of Observed Sublevels for the 5K_8 Energy Level by TSE

Representation	E_{Exp} (cm^{-1})	E_{Com} (cm^{-1})	ΔE (cm^{-1})
Γ_2	39221.1	39218.7	2.4
	39338.4	39358.1	-19.7
	39360.9	39363.2	-2.3
	39463.2	39452.4	10.8
$\Gamma_{3,4}$	39217.8	39217.3	0.5
	39248.5	39250.2	-1.7
	39336.7	39339.3	-2.6
	39399.3	39389.2	10.1
	39460.3	39455.9	4.4
	39465.3	—	—

 5K_8 by TSE

The $\pi\pi$ scan for the 5K_8 level revealed five sublevels whereas the selection rules imply the existence of only four. By making comparisons with the computer fitted model, the four sublevels were identified. The four possible $\Gamma_{3,4}$ sublevels were observed in the $\pi\sigma$ scan with no ambiguity as to their classifications. The data and classifications are presented in Table 8 and the experimental results are shown in Figure 12.

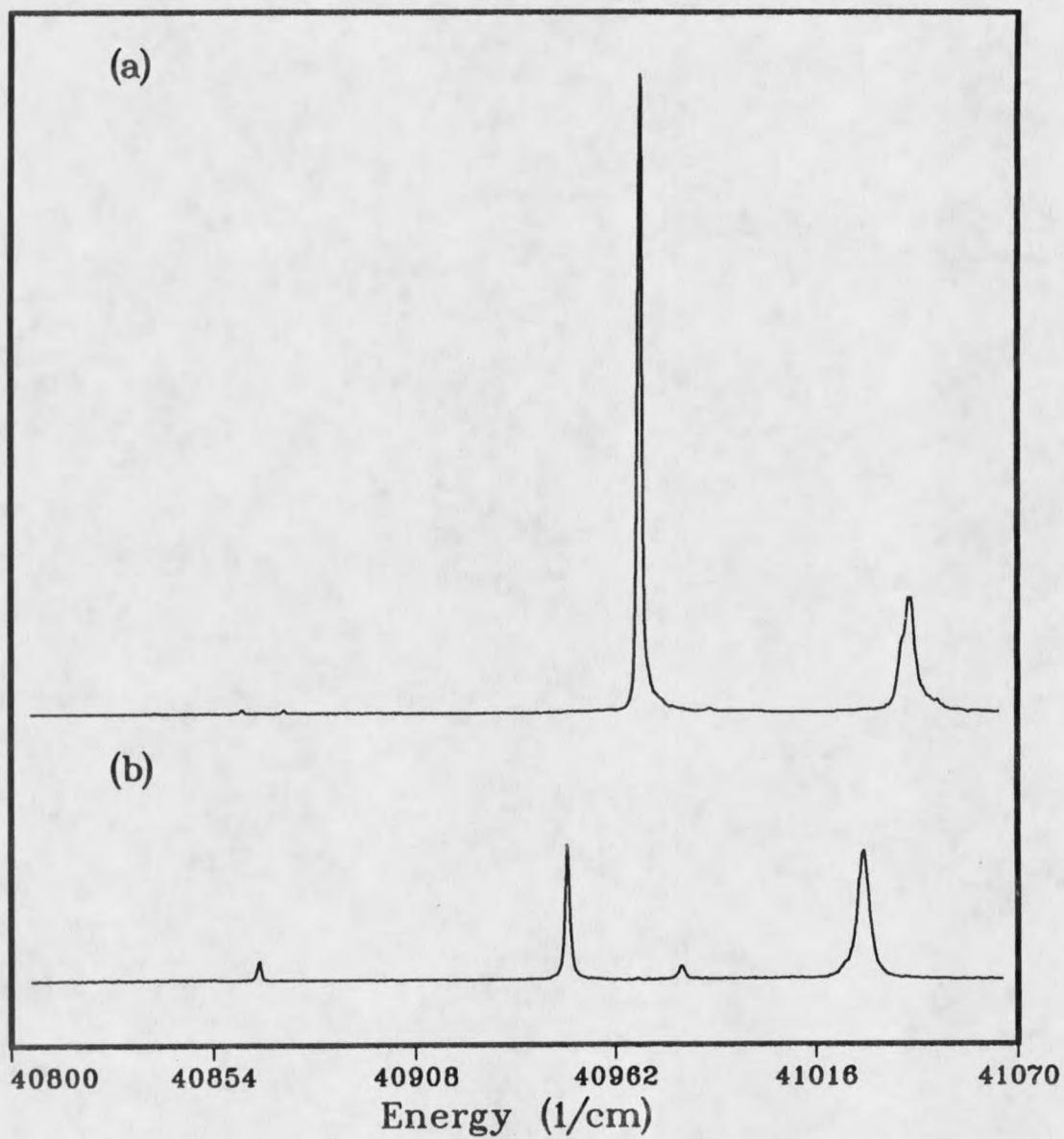


Figure 12. Two-step excitation spectra of the Tb^{3+} ion from ground state ${}^7\text{F}_6$ (Γ_2) to ${}^5\text{K}_8$. (a) $\pi\pi$ polarized. (b) $\pi\sigma$ polarized.

Table 8. Comparison of Observed Sublevels for the 5K_8 Energy Level by TSE

Representation	E_{Exp} (cm^{-1})	E_{Com} (cm^{-1})	ΔE (cm^{-1})
Γ_2	40861.6	—	—
	40872.3	40872.2	0.1
	40968.9	40966.0	2.9
	40987.7	40989.3	-1.6
	41041.4	41037.0	4.4
$\Gamma_{3,4}$	40867.3	40868.7	-1.4
	40948.9	40952.7	-3.7
	40980.1	40976.7	3.4
	41028.7	41026.2	2.5

 5G_6 and 5K_5 by TSE

Since the centers of gravity of these two levels are nearly overlapping, the exact term classification of each sublevel cannot be determined. Therefore, both levels are presented together.

A total of seven Γ_2 sublevels were observed in the $\pi\pi$ scan, but only six are predicted by the selection rules. All of the six possible $\Gamma_{3,4}$ sublevels were positively identified in the $\pi\sigma$ scan as can be seen in Figure 13. Comparisons with the computer fitted model suggest the classifications given in Table 9.

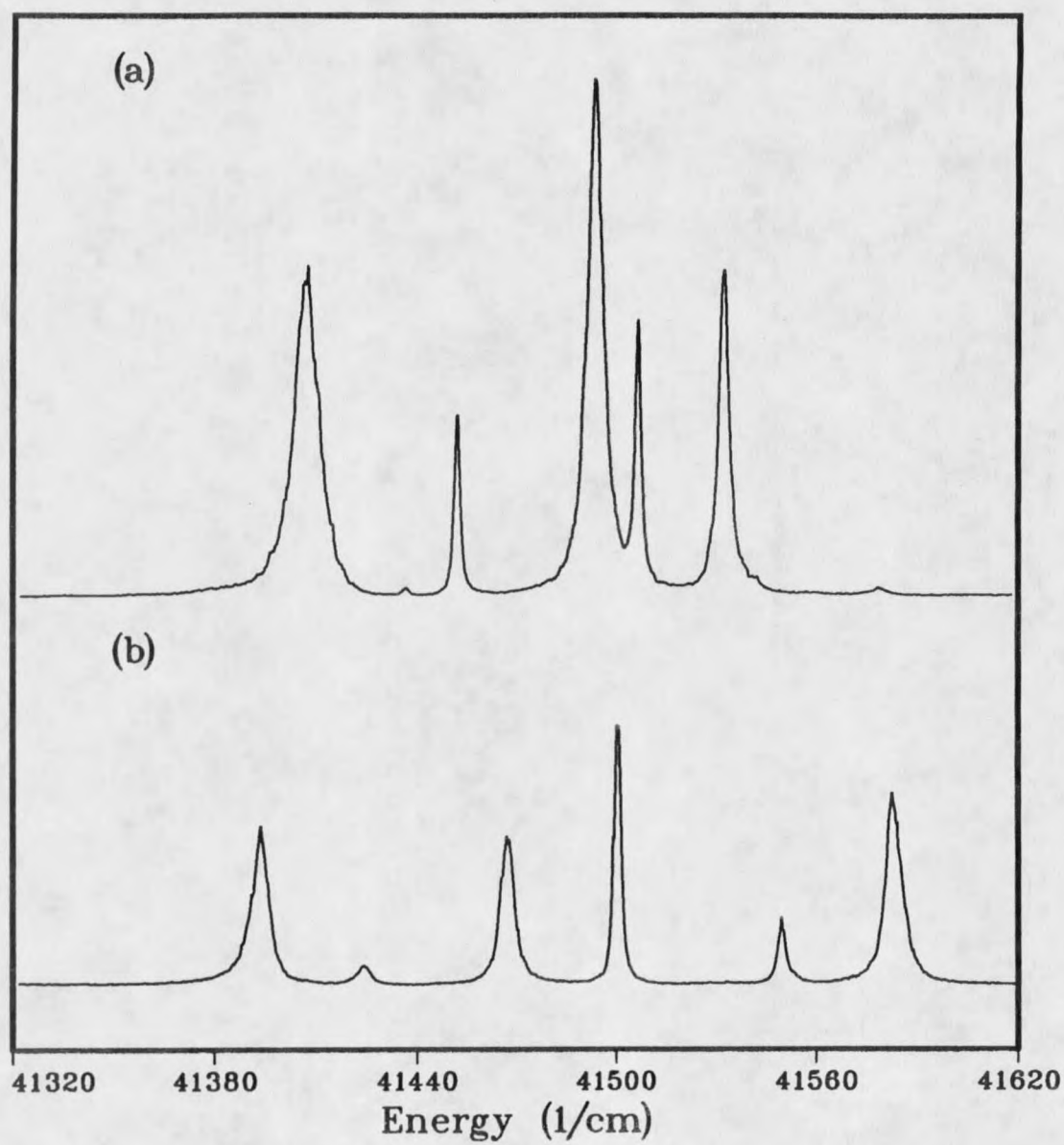


Figure 13. Two-step excitation spectra of the Tb^{3+} ion from ground state ${}^7\text{F}_6$ (Γ_2) to ${}^5\text{K}_5$ and ${}^5\text{G}_6$. (a) $\pi\pi$ polarized. (b) $\pi\sigma$ polarized.

Table 9. Comparison of Observed Sublevels for the 5G_6 and 5K_5 Energy Levels by TSE

Representation	$E_{\text{Exp}} \text{ (cm}^{-1}\text{)}$	$E_{\text{Com}} \text{ (cm}^{-1}\text{)}$	$\Delta E \text{ (cm}^{-1}\text{)}$
Γ_2	41407.6	41395.6	12.0
	41437.2	—	—
	41452.5	41460.4	-7.9
	41494.1	41493.8	0.3
	41507.1	41520.1	-13.0
	41532.8	41530.8	2.0
	41576.4	41589.1	-12.7
$\Gamma_{3,4}$	41393.6	41378.9	14.7
	41424.6	41406.5	18.1
	41466.3	41456.3	10.0
	41499.5	41505.3	-5.8
	41548.9	41551.8	-2.3
	41582.3	41593.9	-11.6

 5K_7 by TSE

All of the possible Γ_2 and $\Gamma_{3,4}$ sublevels were positively identified as can be seen in Figure 14. The energies and classifications are shown in Table 10. During the $\pi\pi$ scan of this experiment the timing between the lasers reversed itself because of equipment malfunctions. This was quickly and easily corrected, but not quick

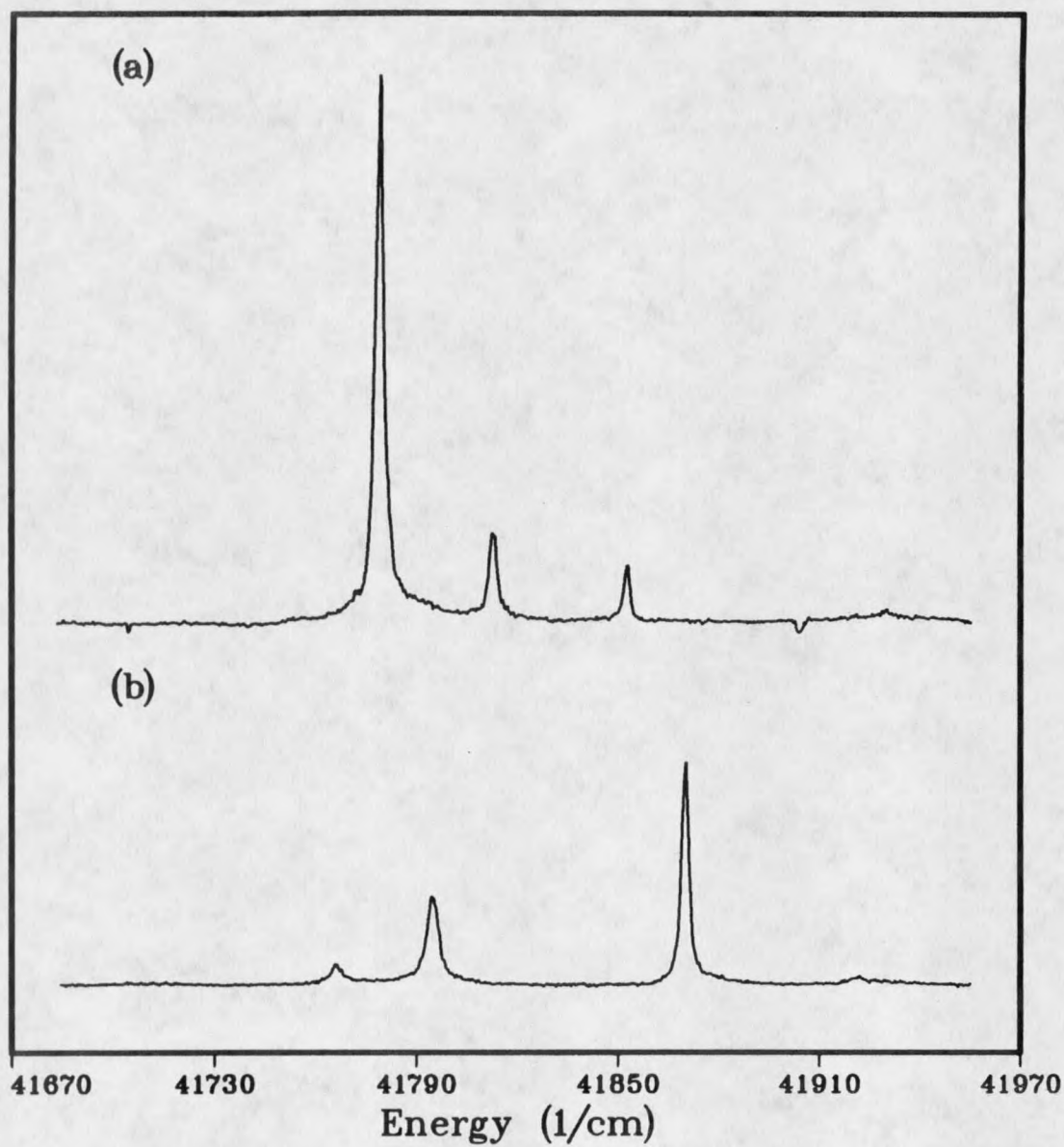


Figure 14. Two-step excitation spectra of the Tb^{3+} ion from ground state ${}^7\text{F}_6$ (Γ_2) to ${}^5\text{K}_7$. (a) $\pi\pi$ polarized. (b) $\pi\sigma$ polarized.

enough to avoid two small dips in the spectra around 41700 cm^{-1} and 41900 cm^{-1} . These, however, give an indication of the contribution to the TSE signal from the laser broadband fluorescence.

Table 10. Comparison of Observed Sublevels for the 5K_7 Energy Level by TSE

Representation	$E_{\text{Exp}}\text{ (cm}^{-1}\text{)}$	$E_{\text{Com}}\text{ (cm}^{-1}\text{)}$	$\Delta E\text{ (cm}^{-1}\text{)}$
Γ_2	41780.4	41783.0	-2.6
	41814.5	41818.4	-3.9
	41853.7	41855.7	-2.0
	41932.0	41923.4	8.6
$\Gamma_{3,4}$	41766.5	41774.8	-8.3
	41794.9	41797.7	-2.8
	41870.5	41867.5	3.0
	41923.1	41913.3	9.8

CHAPTER 5

CONCLUSIONS

Two-photon spectra have been observed by monitoring the anti-Stokes ultraviolet cascade fluorescence from 5D_3 to 7F_5 for the Tb^{3+} ion in the $Tb^{3+}:LiYF_4$ crystal. The two-photon transitions were specifically direct two-photon absorption and piecewise two-step excitation. The energy region scanned by these techniques ranged from $39,100\text{ cm}^{-1}$ to $42,000\text{ cm}^{-1}$.

Some energy levels could not be studied by direct two-photon absorption with the experimental apparatus used. This was because a phonon assisted energy transfer populated a real intermediate level, resulting in a huge two-step excitation background signal from which the direct two-photon absorption signal could not be discriminated. Consequently, the direct two-photon absorption experiments could only be applied to those energy levels which were lower than twice the energy of the intermediate 5D_4 level. Specifically, these energy levels were the 5K_9 , 5D_2 and 5G_6 . Two-step excitation experiments were also carried out for the first two of these three energy levels as well as for the 5K_8 , 5K_5 , a higher order 5G_6 , and 5K_7 energy levels. All of the experimental energies were compared with a computer fitted model, which used these energies as a basis for determining the various free ion and crystal field parameters, showing good agreement between experiment and theory.

The study of the 5G_6 energy level, which was also studied by Huang, yielded some interesting results when compared with the computer fitted model. Based on the relative accuracy of this model thus far, it suggests that there are two undetected energy sublevels between $40,400\text{ cm}^{-1}$ and $40,500\text{ cm}^{-1}$.

The study of the 5D_2 energy level yielded all of the four possible sublevels through both the direct two-photon absorption and two-step excitation experiments. In addition, the presence of direct two-photon absorption transitions suggests a breaking of the selection rules for pure configurations as applied to these transitions. This implies further evidence to support a theory of resonant enhancement, where energy levels within the same configuration and close to resonance can make significant contributions.

The study of the 5K_9 energy level yielded some interesting but unexplainable results. A signal was seen in the direct two-photon absorption scan which was later shown to be a crystal defect energy level in a two-step excitation scan. The exact origin of this ambiguous transition is undetermined at this time. The two-step excitation scans, when compared with the computer fitted model, yielded the classification of all the possible Γ_2 and $\Gamma_{3,4}$ sublevels. Since no useable data were obtained from the direct two-photon absorption scans, the Γ_1 sublevels were undetermined.

The study of the four remaining energy levels, the 5K_8 , 5K_5 , 5G_6 and 5K_7 , yielded all the classifications of the possible Γ_2 and $\Gamma_{3,4}$ sublevels when compared with the computer fitted model. Since these levels were not studied by direct two-photon absorption, the Γ_1 sublevels are undetermined.

Further work in the study of these excited energy states, using the same experimental apparatus, might include extending the energy region studied for the lower 5G_6 up to $40,500\text{ cm}^{-1}$ to determine if there are more sublevels present. This

need only be done by two-step excitation since the Γ_1 sublevels were uniquely determined by direct two-photon absorption. The study of energies above 42,000 cm^{-1} could be done by simply using laser dyes that would extend the laser energy into the near ultraviolet.

Further work might also include absorption and fluorescence studies of the energy region starting at the $^5\text{D}_3$ level at approximately 26,000 cm^{-1} and extending up to 39,100 cm^{-1} . This way, the energy map of the Tb^{3+} ion could be extended leading to further refinements in the free ion and crystal field parameters in the computer fitted model. Such experiments are currently in progress.

REFERENCES CITED

REFERENCES CITED

1. W. Kaiser and C.G.B. Garrett, Phys. Rev. Lett. 7, 229 (1961).
2. M. Göppert-Mayer, Ann. Phys. 9, 273 (1931).
3. U. Fritzler and G. Schaak, J. Phys. C9, L23 (1976).
4. U. Fritzler, Z. Phys. B27, 289 (1977).
5. M. Dagenais, M. Downer, R. Neumann, and N. Bloembergen, Phys. Rev. Lett. 46, 561 (1981).
6. M.C. Downer and A. Bivas, Phys. Rev. B28, 3677 (1983).
7. M.C. Downer, A. Bivas, and N. Bloembergen, Opt. Comm. 41, 335 (1982).
8. C.D. Cordero-Montalvo and N. Bloembergen, Phys. Rev. B30, 438 (1984).
9. J. Huang, G. Liu, and R.L. Cone, Phys. Rev. B39, 6348 (1989).
10. J.E. Bjorkholm and P.F. Liao, Phys. Rev. Lett. 33, 128 (1974).
11. Jin Huang, Ph.D. Thesis at Montana State University (1987) (unpublished).
12. R.L. Cone, T.W. Scott, J.M. Friedman, H.J. Guggenheim, H. Crosswhite, J. Huang, and G. Liu, "Excited State Absorption Spectra of Tb^{3+} from 5D_4 Manifold to 46,000 cm^{-1} in 1% $Tb^{3+}:LiYF_4$ " (to be submitted to Phys.Rev.).
13. A. Gold, "Two-Photon Spectroscopy", in: Quantum Optics, ed. R.J. Glauber (Academic Press, New York, 1969) pp. 397-420.
14. R. Loudon, The Quantum Theory of Light (Clarendon Press, Oxford, 1973) pp. 335-346, pp. 268-329.
15. A. Yariv, Quantum Electronics (John Wiley and Sons, Inc., 1989) pp. 64-66.
16. S. Hufner, Optical Spectra of Transparent Rare Earth Compounds (Academic Press, New York, 1978).
17. B.G. Wybourne, Spectroscopic Properties of Rare Earths (John Wiley and Sons, Inc., 1965).
18. B.R. Judd, Operator Techniques in Atomic Spectroscopy (McGraw-Hill Book Company, Inc., 1963).
19. H.P. Christensen, Phys. Rev. B17, 4060 (1978).

20. M. Tinkham, Group Theory and Quantum Mechanics (McGraw-Hill Book Company, Inc., 1964).
21. M. Weissbluth, Atoms and Molecules (Academic Press, New York, 1978) pp. 60-88.
22. P.H.E. Meier and E. Bauer, Group Theory - The Application to Quantum Mechanics (John Wiley and Sons, Inc., 1962)
23. G.F. Koster, J.O. Dimmock, R.J. Wheeler, and H. Statz, Properties of the 32 Point Groups (MIT Press, Cambridge, 1963) pp. 40-42.
24. W. Hayes and R. Loudon, Scattering of Light by Crystals (John Wiley and Sons, Inc., 1978) pp. 43-49.
25. A. Kiel and S.P.S. Porto, J. Mol. Spect. 32, 458 (1969).
26. T.W. Hänsch, Appl. Opt. 11, 895 (1972).
27. K. Arnett, R. Anderson, and R. Alexander, Am. J. Phys. 49, 767 (1981).
28. D.S. King, P.K. Schenk, K.C. Smyth, and J.C. Travis, Appl. Opt. 16, 2617 (1977).
29. C.J. Sansonetti and K.H. Wheber, J. Opt. Soc. Am. B 1, 361 (1984).
30. E. Hecht and A. Zajac, Optics (Addison-Wesley Publishing Company, 1979) pp. 307-311.
31. B.A. Palmer, R.A. Keller, and R. Engleman, Jr., "An Atlas of Uranium Emission Intensities in a Hollow Cathode Discharge", LASL Rep. LA-8251-MS (Los Alamos Scientific Laboratory, Los Alamos, NM, 1980).
32. Private discussions with R.L. Cone, May 1989.
33. R.L. Cone and R.S. Meltzer "Ion-Ion Interactions and Exciton Effects in Rare Rare Earth Insulators", in: Spectroscopy of Solids Containing Rare Earth Ions, ed. A.A. Kaplyanski and R.M. MacFarlane (North Holland, Amsterdam, 1987).

APPENDIX

COMPUTER PROGRAMS

Figure 15. Program to perform direct two-photon absorption experiments.

```

/*      A PROGRAM TO DO DIRECT TWO-PHOTON ABSORPTION.
        SCANS LASER VIA A STEPPER MOTOR CONTROL AND
        TRIGGERS NS575 MULTICHANNEL DIGITAL SIGNAL
        ANALYZER.
        PROGRAMMER: RAYMOND JONES
        DATE:      JULY 8, 1988
        MODIFIED:  NOVEMBER 13, 1988
*/

#include <std.h>
#include <rt11.h>
#include "ad.c"
#include "da.c"
#include "stp.c"
#include "mini.h"

main()
{
    FIO    afio,bfio,cfio;
    char   opga,fape,lapo;
    long   hc,fp,hct,fpt,d;
    long   lp,lpt;
    int    j,i,k,shots;
    int    z[640];
    int    zz[640];
    int    zlp[640];
    char   c[2];
    int    nodd,step,steps,m,loop;
    char   start,fhk,set,end;
    int    rtn;
    long   INITT(),DRWABS(),MOVABS();
    int    x0 = 30;
    int    y0 = 150;
    int    x,y,xx,yy;
    int    baud = 2400;
    int    xmax = 660;
    int    ymax = 730;
    int    ys,yy;
    float  xscale,yscale,yyscale;
    char   bell[2];
    bell[0]=07;
    bell[1]=0;

    getinit();
    putinit();

```

Figure 15. Continued

```

/* --- check the computer interfaces --- */

putfmt("Please check the inputs on the ADC:\n");
putfmt("channel 0: Hollow cathode lamp\n");
putfmt("channel 1: Fabry Perot Etalon\n");
putfmt("channel 2: Laser power diode\n");
putfmt("\n");
putfmt("Outputs on the DAC:\n");
putfmt("B: Ext. trigger of NS575\n");
putfmt("C: MCS scanner of NS575\n");
putfmt("\n");

/* --- set the initial conditions --- */

set:   putfmt("how many data points do you want?\n");
       getfmt("%i\n",&nodd);
       putfmt("how many steps between each data point?\n");
       putfmt("(200 steps/revolution)\n");
       getfmt("%i\n",&step);
       steps=step*nodd;
       putfmt("How many shots per data point?\n");
       getfmt("%i\n",&shots);
       xscale = ((float)600/nodd);

start: putfmt("Old number for HC scale: %i\n",ys);
       putfmt("Input new number:\n");
       getfmt("%i\n",&ys);
       putfmt("\n");
       putfmt("Old number for FPscale: %i\n",yys);
       putfmt("Input new number:\n");
       getfmt("%i\n",&yys);
       yscale = ((float)500/ys);
       yyscale = ((float)500/yys);
       x = 30;
       xx = 30;
       y = 150;
       yy = 150;

/* --- Opening the data files --- */

       putfmt("Hollow cathode file name?\n");
       getfmt("%p\n",opga);
       if(!fcreate(&afio,opga,1))
       {
           putfmt("ERROR: can't open %p\n",opga);
           return;
       }
       putfmt("Fabry-Perot file name?\n");

```

Figure 15. Continued

```

    getfmt("%p\n",fape);
    if(!fcreate(&bfio,fape,1))
    {
        putfmt("ERROR: can't open %p\n",fape);
        return;
    }
    putfmt("Laser power file name?\n");
    getfmt("%p\n",lapo);
    if(!fcreate(&cfio,lapo,1))
    {
        putfmt("ERROR: can't open %p\n",lapo);
        return;
    }

/* --- Taking zero readings --- */

    putfmt("Are you ready to get zero?\n");
    getfmt("\n");
    gettrg();
    getzero(0),getzero(1),getzero(2);
    for(k=0;k<=nodd+10;k++)
    {
        z[k] = 0;
        zz[k] = 0;
        zlp[k] = 0;
    }
    putfmt("Zero has been taken.\n");
    putfmt("Hit return when you are ready to continue\n");
    getfmt("\n");

/* --- starting the scan --- */

    putfmt("Hit return to start the scan\n");
    putfmt("%p\n\n",bell);
    getfmt("\n");
    m=0;
    putvolt(2,2047);
    putvolt(1,2047);

/* --- initialize plot and draw axes --- */

    fcall (INITT,1,&baud);
    fcall (MOVABS,2,&x0,&ymax);
    fcall (DRWABS,2,&x0,&y0);
    fcall (MOVABS,2,&xmax,&y0);
    fcall (DRWABS,2,&x0,&y0);

```

Figure 15. Continued

```

/* --- Trigger NS575 by external --- */

    putvolt(1,1023);
    for(k=1;k<=100;k++)
    {
        d=0;
    }
    putvolt(1,2047);

/* --- starting actual loop for doing experiment --- */

    for(i=1;i<=nodd;i++)
    {
        hct=0;
        fpt=0;
        lpt=0;

/* --- TTL signal to MCS scanner --- */

        putvolt(2,1023);
        for(k=1;k<=100;k++)
        {
            d=0;
        }
        putvolt(2,2047);

        for(j=1;j<=shots;j++)
        {
            gettrg();
            hc=getvolt(0);
            fp=getvolt(1);
            lp=getvolt(2);
            hct += hc;
            fpt += fp;
            lpt += lp;
            for(k=1;k<=100;k++)
            {
                d=0;
            }

            z[i] = hct/50;
            zz[i] = fpt/50;
            zlp[i] = lpt/50;
            getcw(step);
            m+=step;

```

Figure 15. Continued

```

/* --- plotting the data --- */

    fcall(MOVABS,2,&x,&y);
    x=(int)((float)(i*xscale+30));
    y=(int)((float)(hct*yscale+150));
    fcall(DRWABS,2,&x,&y);
    fcall(MOVABS,2,&xx,&yy);
    xx=x;
    yy=(int)((float)(fpt*yyscale+150));
    fcall(DRWABS,2,&xx,&yy);
}
putfmt("%p\n\n",bell);
putfmt("%p\n\n",bell);
putfmt("%p\n\n",bell);

/* --- End of scan --- */

    putvolt(2,1023);
    for(k=1;k<=100;k++)
    {
        d=0;
    }
    putvolt(2,2047);
    i=1;
    putfmt("No. of data = %i\n",i);
    putfmt("Scan is finished. Total data points = %i\n",nodd);
    putfmt("Total scan is %i steps.\n",m);

/* --- sending data to files --- */

    putf(&afio,"%n");
    for(k=0;k<=(i/8);k++)
    {
        putf(&afio,"%4i  %6i  %6i  %6i  %6i  %6i  %6i  %6i\n",
(int)(8*k),z[8*k+0],z[8*k+1],z[8*k+2],z[8*k+3],z[8*k+4],z[8*k+5],z[8*k+6],z[8*k+7]);
    }
    fclose(&afio);
    putf(&bfio,"%n");
    for(k=0;k<=(i/8);k++)
    {
        putf(&bfio,"%4i  %6i  %6i  %6i  %6i  %6i  %6i  %6i\n",
(int)(8*k),zz[8*k+0],zz[8*k+1],zz[8*k+2],zz[8*k+3],zz[8*k+4],zz[8*k+5],zz[8*k+6],zz[8*k+7]);
    }
    fclose(&bfio);
    putf(&cfio,"%n");
    for(k=0;k<=(i/8);k++)

```

Figure 15. Continued

```

        {
            putf(&cfio,"%4i %6i %6i %6i %6i %6i %6i %6i
%6i\n",(int)(8*k),zlp[8*k+0],zlp[8*k+1],zlp[8*k+2],zlp[8*k+3],zlp[8*k+4],zlp[8*k+5],z
lp[8*k+6],zlp[8*k+7]);
        }
    fclose(&cfio);

    putfmt("%p\n\n",bell);
    putfmt("How many steps do you want to go back?\n");
    getfmt("%i\n",&rtn);
    if(rtn == 0)
    {
        goto fhk;
    }
    else
    {
        rtn+=100;
        getccw(rtn);
        getcw(100);
    }
fhk:  c[0]='0';
      putfmt("%p\n\n",bell);
      putfmt("Do you wish to run again with same settings?\n");
      putfmt("hit return to end\n");
      getfmt("%p\n",c);
      if(c[0] == 'Y' || c[0] == 'y') goto start;
      if(c[0] == 'N' || c[0] == 'n') goto set;
end:  putfmt("GOOD LUCK\n");
}

```

Figure 16. Program to perform two-step excitation experiments.

```

/*          A PROGRAM TO DO TWO-STEP EXCITATION.
           SCANS LASER VIA A STEPPER MOTOR CONTROL AND
           MONITORS FOUR ANALOG INPUTS.
           PROGRAMMER: RAYMOND JONES
           DATE:      AUG 21,1989

*/

#include    <std.h>
#include    <rt11.h>
#include    "ad.c"
#include    "stp.c"
#include    "mini.h"

main()
{
    FIO    afio;
    char    flnm;
    long    hc,fp,hct,fpt,d;
    long    lp,lpt,tp,tpt;
    int     j,i,k,shots;
    int     z[640];
    int     zz[640];
    int     zlp[640];
    int     ztp[640];
    char    c[2];
    int     nodd,step,steps,m,loop;
    char    start,fhk,set,end;
    int     rtn;
    long    INITT(),DRWABS(),MOVABS();
    int     x0 = 30;
    int     y0 = 150;
    int     x,xold,y1,y2,y3;
    int     baud = 2400;
    int     xmax = 660;
    int     ymax = 730;
    int     y1s,y2s,y3s;
    float    xscale,y1scale,y2scale,y3scale;
    char    bell[2];
    bell[0]=07;
    bell[1]=0;

    getinit();

    /* --- check the computer interfaces --- */

    putfmt("Please check the inputs on the ADC:\n");
    putfmt("channel 0: Hollow cathode lamp\n");
    putfmt("channel 1: Fabry Perot Etalon\n");

```

Figure 16. Continued

```

putfmt("channel 2: Boxcar signal\n");
putfmt("channel 3: Laser power diode\n");
putfmt("\n");

/* --- set the initial conditions --- */

set:   putfmt("how many data points do you want?\n");
       getfmt("%i\n",&nodd);
       putfmt("how many steps between each data point?\n");
       putfmt("(200 steps/revolution)\n");
       getfmt("%i\n",&step);
       steps=step*nodd;
       putfmt("How many shots per data point?\n");
       getfmt("%i\n",&shots);
       xscale = ((float)600/nodd);

start: putfmt("Old number for HC scale: %i\n",y1s);
       putfmt("Input new number:\n");
       getfmt("%i\n",&y1s);
       putfmt("\n");
       putfmt("Old number for FPscale: %i\n",y2s);
       putfmt("Input new number:\n");
       getfmt("%i\n",&y2s);
       putfmt("\n");
       putfmt("Old number for signal scale: %i\n",y3s);
       putfmt("Input new number:\n");
       getfmt("%i\n",&y3s);
       y1scale = ((float).003*y1s);
       y2scale = ((float).003*y2s);
       y3scale = ((float).003*y3s);
       x = 30;
       y1 = 150;
       y2 = 150;
       y3 = 150;

/* --- Opening the data files --- */

       putfmt("File name?\n");
       getfmt("%p\n",flnm);
       if(!fcreate(&afio,flnm,1))
       {
           putfmt("ERROR: can't open %p\n",flnm);
           return;
       }

/* --- Taking zero readings --- */

       putfmt("Are you ready to get zero?\n");
       getfmt("\n");
       gettrg();

```

Figure 16. Continued

```

    getzero(0),getzero(1),getzero(2),getzero(3);
    for(k=0;k<=nodd;k++)
    {
        z[k] = 0;
        zz[k] = 0;
        zlp[k] = 0;
        ztp[k] = 0;
    }
    putfmt("Zero has been taken.\n");
    putfmt("Hit return when you are ready to continue\n");
    getfmt("\n");

/* --- starting the scan --- */

    putfmt("Hit return to start the scan\n");
    putfmt("%p\n\n",bell);
    getfmt("\n");
    m=0;

/* --- initialize plot and draw axes --- */

    fcall (INITT,1,&baud);
    fcall (MOVABS,2,&x0,&ymax);
    fcall (DRWABS,2,&x0,&y0);
    fcall (MOVABS,2,&xmax,&y0);
    fcall (DRWABS,2,&x0,&y0);

/* --- starting actual loop for doing experiment --- */

    for(i=1;i<=nodd;i++)
    {
        hct=0;
        fpt=0;
        lpt=0;
        tpt=0;

        for(j=1;j<=shots;j++)
        {
            gettrg();
            hc=getvolt(0);
            fp=getvolt(1);
            tp=getvolt(2);
            lp=getvolt(3);
            hct += hc;
            fpt += fp;
            lpt += lp;
            tpt += tp;
        }
    }

```

Figure 16. Continued

```

        for(k=1;k<=100;k++)
        {
            d=0;
        }
    }
    hct = -1*hct;
    tpt = -1*tpt;
    z[i] = hct/50;
    zz[i] = fpt/50;
    zlp[i] = lpt/50;
    ztp[i] = tpt/50;
    getcw(step);
    m+=step;

/* --- plotting the data --- */

    xold=x;
    x=(int)((float)(i*xscale+30));
    fcall(MOVABS,2,&xold,&y1);
    y1=(int)((float)(hct*y1scale+150));
    fcall(DRWABS,2,&x,&y1);
    fcall(MOVABS,2,&xold,&y2);
    y2=(int)((float)(fpt*y2scale+150));
    fcall(DRWABS,2,&x,&y2);
    fcall(MOVABS,2,&xold,&y3);
    y3=(int)((float)(tpt*y3scale+150));
    fcall(DRWABS,2,&x,&y3);
    }
    putfmt("%p\n\n",bell);
    putfmt("%p\n\n",bell);
    putfmt("%p\n\n",bell);

/* --- End of scan --- */

    i-=1;
    putfmt("No. of data = %i\n",i);
    putfmt("Scan is finished. Total data points = %i\n",nodd);
    putfmt("Total scan is %i steps.\n",m);

/* --- sending data to file --- */

    putf(&afio,"%\n");
    for(k=1;k<=nodd;k++)
    {
        putf(&afio," %4i %6i %6i %6i %6i\n",k,z[k],zz[k],ztp[k],zlp[k],(int)(1000*((float)ztp[k]/zlp[k])));
    }
    fclose(&afio);

    putfmt("%p\n\n",bell);

```

Figure 16. Continued

```

    putfmt("How many steps do you want to go back?\n");
    getfmt("%i\n",&rtn);
    if(rtn == 0)
    {
        goto fhk;
    }
    else
    {
        rtn+=100;
        getccw(rtn);
        getcw(100);
    }
fhk:  c[0]='0';
    putfmt("%p\n\n",bell);
    putfmt("Do you wish to run again with same settings?\n");
    putfmt("hit return to end\n");
    getfmt("%p\n",c);
    if(c[0] == 'Y' || c[0] == 'y') goto start;
    if(c[0] == 'N' || c[0] == 'n') goto set;
end:  putfmt("GOOD LUCK!\n");
}

```

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