

TECHNICAL  
RESEARCH  
REPORT

AD748517

THE STUDY OF THE INTERACTION OF  
INTENSE PICOSECOND LIGHT PULSE  
WITH MATERIALS

A QUARTERLY TECHNICAL REPORT

(TR-9)

SUBMITTED TO

THE U.S. ARMY RESEARCH OFFICE

PERIOD

December 21, 1971 to March 21, 1972

REPORTED BY

CHI H. LEE

and

Young Ho Park

Approved for Public Release  
Distributions Unlimited.

DEPARTMENT OF  
ELECTRICAL ENGINEERING

UNIVERSITY OF MARYLAND

COLLEGE PARK, MARYLAND 20742

Reproduced by  
NATIONAL TECHNICAL  
INFORMATION SERVICE  
U S Department of Commerce



## DOCUMENT CONTROL DATA - R &amp; D

*(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)*

1. ORIGINATING ACTIVITY (Corporate author) Department of Electrical Engineering University of Maryland College Park, Maryland 20742		2a. REPORT SECURITY CLASSIFICATION Unclassified	
3. REPORT TITLE Investigation of Laser Action in GaAs with Two Photon Pump		2b. GROUP	
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) A quarterly technical report, December 22, 1971 to March 21, 1972			
5. AUTHOR(S) (First name, middle initial, last name) Young Ho Park and C. H. Lee			
6. REPORT DATE May 1, 1972 e	7a. TOTAL NO. OF PAGES 64	7b. NO. OF REFS 13	
8a. CONTRACT OR GRANT NO. DA-ARO-D-31-124-71A55	9a. ORIGINATOR'S REPORT NUMBER(S) TR-9		
8b. PROJECT NO. ARPA Order No. 675 Am. 9 c. Program Code No. 9E20	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)		
10. DISTRIBUTION STATEMENT Reproduction in whole or in part is permitted for any purpose of the United State Government.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY ARPA and U. S. Army Research Office	
13. ABSTRACT Laser action in GaAs using a Nd: glass laser as a two photon pump was investigated. External mirrors were employed to form the optical cavity. Laser action in GaAs was observed in the following experiments: Firstly, we observed a narrowing of the emission spectrum. Secondly, we examined the output power dependence upon the input power, and measured the threshold power.  The main advantage of this method of optical excitation is the possibility of obtaining an inverse population throughout a large semiconductor volume. The advantage of using an external mirror resonator is that it may be possible to Q-switch the GaAs laser and to use the adjustable cavity for various other purposes.			

Unclassified

Security Classification

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Two photon absorption Optically pumped semiconductor laser GaAs Laser						

Unclassified

Security Classification

Quarterly Technical Report

for

Period December 21, 1971 to March 21, 1972  
Submitted to the U. S. Army Research Office

ARPA 675, Am 9

Program Code Number: 9E20

Name of Grantee: University of Maryland

Effective Date of Grant: December 22, 1970

Grant Expiration Date: March 21, 1972

Principle Investigator and  
Phone Number: Dr. Chi H. Lee  
(301) 454-2443

Grant Number: DA-ARO-D-31-124-71-55

Project Scientist or  
Research Assistants: S. Jayaraman  
V. Bhanthumnavin and Young Ho Park

Short Title of Work: "The Study of the Interaction of  
Intense Picosecond Light Pulses with  
Materials"

Reported by

Chi-hung Lee

Dr. Chi H. Lee  
Assistant Professor

**INVESTIGATION OF LASER ACTION  
IN GaAs WITH TWO PHOTON PUMP**

by

**Young Ho Park\* and Chi H. Lee**

**\*This is a M.S. Thesis of Mr. Young Ho Park. It is submitted to the Faculty of the Graduate School of the University of Maryland in partial fulfillment of the requirements for the degree of Master of Science, 1972.**

IIA

## ABSRTACT

**Title of Thesis:** Investigation of Laser Action in GaAs with Two Photon Pump

**Name of Candidate:** Young Ho Park

**Thesis directed by:** C.H. Lee, Assistant Professor

Laser action in GaAs using a Nd:glass laser as a two photon pump was investigated. External mirrors were employed to form the optical cavity. Laser action in GaAs was observed in the following experiments: Firstly, we observed a narrowing of the emission spectrum. Secondly, we examined the output power dependence upon the input power, and measured the threshold power.

The main advantage of this method of optical excitation is the possibility of obtaining an inverse population throughout a large semiconductor volume. The advantage of using an external mirror resonator is that it may be possible to Q-switch the GaAs laser and to use the adjustable cavity for various other purposes.

INVESTIGATION OF LASER ACTION IN GaAs  
WITH TWO PHOTON PUMP

by  
Young Ho Park

Thesis submitted to the Faculty of the Graduate School  
of the University of Maryland in partial fulfillment  
of the requirements for the degree of  
Master of Science  
1972

## ACKNOWLEDGEMENTS

It is with pleasure that I now recognize the people whose contributions made this report possible.

I am greatly indebted to my thesis advisor Dr. C.H. Lee. I wish to express my appreciation to him for his guidance, for his constant encouragement and for his support of me during conduct of this project.

I also wish to thank Dr. Siahatgar for his original setting up of the experiment and for his assistance and discussions.

I thank Mr. S. Jayaraman for many of those daily discussions and for having provided me with excellent collection of literature on two photon process.

I also thank Mr. V. Banthumnavin for his assistance during my work.

I am grateful to Mr. V. Rinker and Mr. R. Sumner for their technical assistance.



## TABLE OF CONTENTS

Chapter	Page
I. INTRODUCTION .....	1
II. THE THEORETICAL BACKGROUND .....	7
1. The Transition Probability and the Absorption Coefficient .....	7
2. The Spectral Linewidth Narrowing .....	16
3. The Threshold Pumping Power for Oscilla- tion and Lasing .....	20
III. EXPERIMENT .....	24
1. The Spectral Emission Lines from GaAs ...	24
2. The Dependence of Power Density of Radia- tion from GaAs on the Exciting Light In- tensity .....	30
3. The Power Calibration of the Photodiode..	31
4. The Pumping Source .....	34
IV. THE EXPERIMENTAL RESULTS AND DISCUSSION .....	36
1. The Emission Spectrum from GaAs .....	36
2. The Dependence of the Power Density Radia- ted by GaAs upon the Exciting Light In- tensity .....	38
APPENDIX I. DERIVATION OF $W_i$ .....	42
APPENDIX II. TWO PHOTON ABSORPTION COEFFICIENT .....	47
APPENDIX III. SOLUTION OF DIFFERENTIAL EQUATION .....	49
APPENDIX IV. ADJUSTMENT OF Nd:glass LASER .....	51
APPENDIX V. CIRCUIT DIAGRAM OF DETECTOR .....	53
APPENDIX VI. DEWAR .....	54
REFERENCES .....	55

## CHAPTER I

### INTRODUCTION

The advent of intense coherent optical sources has made it experimentally feasible to observe multiphoton absorption in a number of materials. Two photon absorption in semiconductors can be employed to generate electron-hole pairs resulting in population inversion. Such a two photon pump could be used to observe laser action in bulk semiconductors. The laser action<sup>(1),(2),(3)</sup> in various bulk semiconductors such as GaAs, CdTe, CdSe, CdS, ZnS etc. have been investigated.

In the present experiment, the laser action in GaAs using a Nd:glass laser as a two photon pump was investigated with external mirrors forming the optical cavity. Such a bulk GaAs laser using external mirrors as resonator could be used to produce intense Q-switched optical pulses.

Even though the general theory for two photon absorption was first given by Maria Göppert-Mayer<sup>(4)</sup> in the Göttinger dissertation in 1931, the first observation of two photon absorption was made in 1961 by Kaiser and Garrett<sup>(5)</sup>. Moreover the observation of two photon absorption in semiconductors came later. Braunstein and Ockman<sup>(2)</sup> observed two photon absorption in CdS pumped by a ruby laser. Basov<sup>(1)</sup> et al. investigated two photon absorption

in GaAs pumped by a Nd:glass laser and C. Chang<sup>(3)</sup> investigated two photon absorption for laser action in ZnS pumped by a ruby laser.

Before these observations, it had been thought that no optical transitions were possible which could create electron-hole pairs by absorbing photons possessing less energy than the energy gap. However, if the exciting source has sufficiently high power, many photons can be absorbed simultaneously. The absorption can be detected because the higher order nonlinear terms cannot be neglected.

In the present investigation we used GaAs, which has a band gap of 1.5 eV at 77°K and a Q-switched Nd:glass laser beam as pumping source. The latter has a photon energy of 1.17 eV. So the condition

$$2\hbar\omega > E_g > \hbar\omega$$

is satisfied where  $E_g$  is the energy gap of GaAs,  $\omega$  is angular frequency of the radiation of the laser,  $\hbar = \frac{h}{2\pi}$  and  $h$  is Planck's constant. If the semiconductor were purely intrinsic, only a two photon absorption process would be possible if we were to neglect higher order absorption. But in the experiment we do observe a single photon process when the pumping power is low. This is because of the single photon excitation of impurity levels in the forbidden band. According to the experimental result it is possible to distinguish one and two photon absorption process. Now this is a good

place to compare one and two photon absorption processes.

The transition probability for one and two photon absorption processes was calculated by Basov<sup>(1)</sup>:

$$W^{(n)} \propto (n\hbar\omega - E_g)^{\frac{2n-1}{2}} I^n \quad \text{for } n = 1, 2$$

where  $\hbar\omega$ : photon energy of pumping source beam

$E_g$ : energy gap of the semiconductor

$I$ : intensity of pumping laser beam

$W^{(n)}$ : upward transition probability per unit volume

The absorption coefficient  $K^{(n)}$  can be calculated as follows

$$K^{(n)} = \frac{W^{(n)} n\hbar\omega}{I}$$

In a one-photon process

$$K^{(1)} \propto (\hbar\omega - E_g)^{1/2} = \text{const.},$$

but in a two-photon process

$$K^{(2)} \propto (2\hbar\omega - E_g)^{3/2} I = \text{const} \times I.$$

In a two photon process  $K^{(2)}$  is linearly proportional to the exciting laser intensity. In a one-photon process

$W^{(1)} \propto \text{const} \times I$  and in two photon process  $W^{(2)} \propto \text{const} \times I^2$ .

As the intensities of the pumping source is increased, multi-photon absorption of light (in this case, two photon absorption of radiation in the semiconductors) becomes predominant.

At higher intensities  $K^{(2)}$  becomes much larger than  $K^{(1)}$ . One of the most important features of the semiconductor laser using two photon absorption is that the intensity distribution inside the lasing crystal is more homogeneous than that due to single photon absorption. This is obvious conclusion of equations (2-1-13) and (2-1-14). Basov et al.<sup>(1)</sup> determined the intensity of light  $I(X)$  at penetration depth  $X$  into the medium from the equation

$$\frac{dI}{dX} = -2\hbar\omega W^{(2)} - KI = -2\hbar\omega a I^2 - KI = -\beta I^2 - KI$$

where  $K=K^{(1)}$ ; single photon absorption coefficient .

$$\beta = K^{(2)}/I = 2\hbar\omega a$$

$$W^{(2)} = a I^2$$

$a$ ; constant given in equation (2-1-11)

From now on we will use  $W$  instead of  $W^{(2)}$  since we are mainly interested in two photon process. The derivation of  $I(X)$  is shown in appendix III but the result is as follows

$$I(X) = \frac{I_0 \exp(-KX)}{1 + \frac{\beta I_0}{K} [1 - \exp(-KX)]}$$

where  $I_0 = I_S (1 - r)$

$I_S$ : total intensity of the exciting radiation incident on the surface of the medium

$r$ : the reflection coefficient .

If two photon absorption is dominant, so  $KX \ll 1$ , then

$$I(X) = \frac{I_0}{\beta XI_0 + 1}$$

This equation indicates that a large volume of the semi-conducting medium has participated in the lasing action.

This gives rise to a much larger power output and better coherence of radiation than from p-n junction lasers.

We have seen that electrons can be excited into the conduction band by absorbing two photons, thus creating electron-hole pairs. These electrons and holes can recombine to emit optical radiation commonly called recombination radiation.

If we introduce resonant reflectors to this semiconductor medium, we can get stimulated emission as well as spontaneous emission. The probabilities for spontaneous emission and stimulated emission were derived by Lasher and Stern<sup>(6)</sup>, followed by a theoretical analysis by Dumke<sup>(7)</sup>. In order to get the laser action in the semiconductor, it has to satisfy the population inversion condition and threshold condition.

Even though we know it is possible to get laser action, it is not easy to experimentally verify whether it is truly laser action. But the group in Levedev Physics Institute<sup>(1)</sup>

and others<sup>(8)</sup> proved it with several experimental results. One of method is to determine the narrowing of the width of emission spectrum. Farfield pattern would also help to determine laser action. Accurate bandwidth measurement can be performed to make sure of the laser action.

We examined the narrowing of the linewidth of the emission spectrum. As we increased the power density of the pumping laser above threshold, the linewidth of the emission spectrum narrowed abruptly. This can be a criterion for lasing in the GaAs quantum generator. But even though the power density was below the threshold, the narrowing of the linewidth was observed. This can be explained as the super-radiance. The details are discussed in the next chapter.

## CHAPTER II

### THE THEORETICAL BACKGROUND

The transition probability and absorption coefficient of two photon process are derived. The threshold power density of the exciting radiation is calculated. Then the narrowing of the linewidth of emission spectra is discussed.

#### §1. The Transition Probability and the Absorption Coefficient

In order to derive the absorption coefficient, it is necessary to calculate transition probability using time dependent perturbation theory. In the present investigation, the transition probability has to be calculated taking into account the band structure of GaAs. GaAs is a direct band gap semiconductor having a zinc sulfide structure. The band structure of such semiconductors have been extensively investigated by E.O. Kane<sup>(9)</sup>. In the following we assume the band structure of GaAs as given by T.S. Moss<sup>(10)</sup> and calculate the transition probability and two photon absorption coefficient in GaAs.

First the Hamiltonian of the electron moving in the presence of an electromagnetic field can be written as

$$H = \left( \mathbf{P} + \frac{e}{c} \mathbf{A} \right)^2 / 2m + V$$



$$\begin{aligned}
 H &= \frac{1}{2m} \left( \bar{P} + \frac{e}{c} \bar{A} \right) \cdot \left( \bar{P} + \frac{e}{c} \bar{A} \right) + V \\
 &= \frac{p^2}{2m} + V + \frac{e}{2mc} \left( \bar{P} \cdot \bar{A} + \bar{A} \cdot \bar{P} \right) + \frac{e^2}{2mc^2} A^2
 \end{aligned}$$

Since  $\bar{P}$  and  $\bar{A}$  commute if we choose the gauge such that  $\nabla \cdot \bar{A} = 0$ ,

$$H = \frac{p^2}{2m} + V + \frac{e}{mc} \bar{P} \cdot \bar{A} + \frac{e^2}{2mc^2} A^2 \quad (2-1-1)$$

where  $\bar{P}$ : momentum operator  
 $\bar{A}$ : vector potential  
 $V$ : potential energy  
 $m$ : mass of free electron

Let  $H_0 = \frac{p^2}{2m} + V$  be the unperturbed Hamiltonian.

Then the term  $\frac{e}{mc} \bar{P} \cdot \bar{A} + \frac{e^2}{2mc^2} A^2$  can be regarded as a

perturbing term. But for one or two photon processes, the term  $\frac{e^2}{2mc^2} A^2$  is much smaller than  $\frac{e}{mc} \bar{P} \cdot \bar{A}$ . Figure

2-1-1 shows the band structure of GaAs. Gallium Arsenide has one conduction band and three valence bands. However in this experiment the transition probability from a valence band  $V_3$  to conduction band C is small compared to transition probability from  $V_1$  or  $V_2$  to C (Figure 2-1-1). Thus the transition probability from  $V_3$  to C can be neglected. In calculating the two photon transition probability, only the transition from  $V_1$  and  $V_2$  to C will be considered.

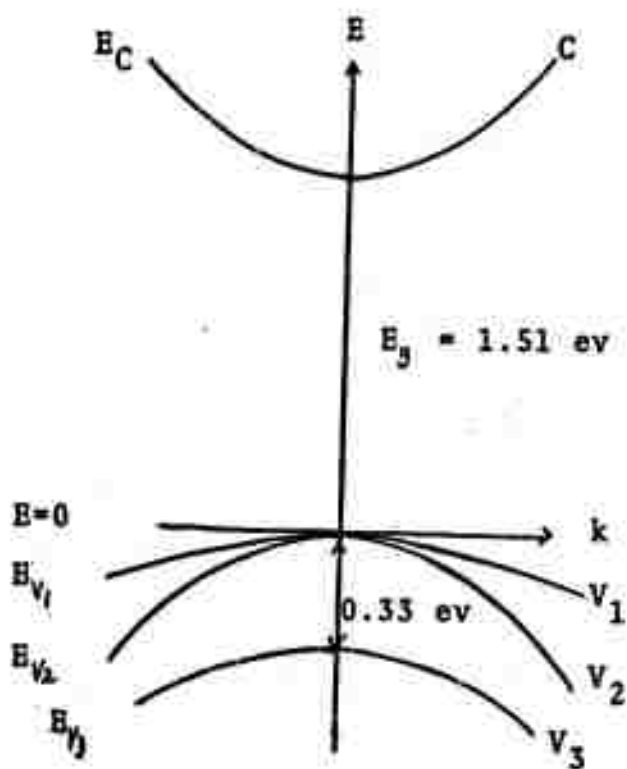


Figure 2-1-1. GaAs Band Structure<sup>(10)</sup>

- C** : Conduction band  
**V<sub>1</sub>** : heavy hole band  
**V<sub>2</sub>** : high hole band  
**V<sub>3</sub>** : split-off band
 } valence bands
- m** : electron rest mass
- m<sub>C</sub><sup>\*</sup>** : effective mass in band C      **m<sub>C</sub><sup>\*</sup>** = 0.072 m  
**m<sub>V<sub>1</sub></sub><sup>\*</sup>** : effective mass in band V<sub>1</sub>      **m<sub>V<sub>1</sub></sub><sup>\*</sup>** = 0.68 m  
**m<sub>V<sub>2</sub></sub><sup>\*</sup>** : effective mass in band V<sub>2</sub>      **m<sub>V<sub>2</sub></sub><sup>\*</sup>** = 0.085 m  
**m<sub>V<sub>3</sub></sub><sup>\*</sup>** : effective mass in band V<sub>3</sub>      **m<sub>V<sub>3</sub></sub><sup>\*</sup>** = 0.25 m
- E<sub>α</sub>** : the energy of electron in' the α band  
 where α = C, V<sub>1</sub>, V<sub>2</sub>, V<sub>3</sub>.

Transition probability  $W$  via two photon absorption can be calculated using second order perturbation theory. Let total transition probability be  $W = W_1 + W_2$  where  $W_i$  is individual transition probability from  $i$ th valence band to conduction band. We need to calculate  $W_i$  for  $i = 1, 2$ .

$$W_i = \frac{2\pi}{\hbar} \iint d^3k_f d^3k_i \left( \sum_{\nu} \frac{\langle i | H' | \nu \rangle \langle \nu | H' | f \rangle}{E_{\nu} - E_i - \hbar\omega} \right)^2 \times \delta(E_f - E_i - 2\hbar\omega) \quad (2-1-2)$$

where  $H' = \frac{e}{mc} \bar{P} \cdot \bar{A}$ ; perturbing Hamiltonian

$|i\rangle$  denotes initial state

$|f\rangle$  denotes final state

$|\nu\rangle$  denotes intermediate state

$\hbar\omega$ ; single photon energy of incident radiation

The initial state is one of the valence bands and the final state is the conduction band. There are two photons which participate simultaneously in this process. The two photon absorption process can be considered as follows. An electron is excited from an initial state  $i$  (valence band) to a final state  $f$  (conduction band) through a virtual intermediate state  $\nu$  by absorbing two photons. Basov<sup>(1)</sup> considered both the conduction band and the valence band as intermediate states. If we take one intermediate state as conduction band (the final state) and the other as a valence band (the initial state), and neglect the contribution from other intermediate states, then  $W_i$  becomes as follows.

$$W_i = \frac{2\pi}{\hbar} \iint d^3k_C d^3k_{V_i} \left( \frac{H'_{V_i C} H'_{CC}}{E_C - E_{V_i} - \hbar\omega} + \frac{H'_{V_i V_i} H'_{V_i C}}{E_{V_i} - E_{V_i} - \hbar\omega} \right)^2 \times \delta(E_C - E_{V_i} - 2\hbar\omega) \quad (2-1-3)$$

where  $H'_{CC} = \langle \psi_C | H' | \psi_C \rangle$

$H'_{CV_i} = \langle \psi_C | H' | \psi_{V_i} \rangle$

... etc.

$H'_{CV_i} = (H'_{V_i C})^\dagger$

$\psi_C, \psi_{V_i}$  are wave function in band C and  $V_i$

Two photons can be absorbed only when  $E_C - E_{V_i} = 2\hbar\omega$ .

Thus

$$W_i = \frac{2\pi}{\hbar} \frac{1}{(\hbar\omega)^2} \iint d^3k_C d^3k_{V_i} \left( H'_{CC} H'_{CV_i} + H'_{CV_i} H'_{V_i V_i} \right)^2 \times \delta(E_C - E_i - 2\hbar\omega) \quad (2-1-4)$$

In order to derive matrix elements, we have to know the wave functions. The wave function of the electrons in the respective bands can be written as the normalized Bloch functions.

$$\psi_C(\vec{k}) = \frac{1}{(2\pi)^{3/2}} U_C(\vec{r}) \exp(i\vec{k}_C \cdot \vec{r}) \quad (2-1-5)$$

$$\psi_{V_i}(\vec{k}_{V_i}) = \frac{1}{(2\pi)^{3/2}} U_{V_i}(\vec{r}) \exp(i\vec{k}_{V_i} \cdot \vec{r})$$

where  $\psi_c$  and  $\psi_{V_i}$  are normalized, and  $U_c(\bar{r})$  and  $U_{V_i}(\bar{r})$  are functions of the position vector  $\bar{r}$  over the dimensions of a unit cell.

Thus

$$\begin{aligned} H'_{cV_i} &= \frac{e}{mC} \langle \psi_c(\bar{k}_c) | \hat{P} \cdot \bar{A} | \psi_{V_i}(\bar{k}_{V_i}) \rangle \\ &= \frac{eA}{mC} (\hat{P} \cdot \bar{\alpha})_{cV_i} \delta(\bar{k}_c - \bar{k}_{V_i}) \end{aligned} \quad (2-1-6)$$

where  $(\hat{P} \cdot \bar{\alpha})_{cV_i} = \langle U_c(\bar{r}) | \hat{P} \cdot \bar{\alpha} | U_{V_i}(\bar{r}) \rangle$

Note that photon wave vector is small compared with crystal momentum  $k_c$  or  $k_{V_i}$ . Thus  $\delta(\bar{k}_c - \bar{k}_{V_i})$  implies a vertical transition since  $H'_{cV_i}$  is not zero only when  $\bar{k}_c = \bar{k}_{V_i}$ .

Note that

$$\hat{P} |\psi\rangle = \frac{m}{\hbar} \frac{\partial E}{\partial \bar{k}} |\psi\rangle \quad (2-1-7)$$

where  $\hat{P}$  is the operator of momentum  $\bar{P}$ .

Let  $\bar{A} = A\bar{\alpha}$

where  $\bar{\alpha}$  is the polarization vector.

Using the relation (2-1-7), we can easily get

$$H'_{cc} = \frac{eA}{m_c^* C} (\hbar k_c \alpha) \delta(\bar{k}_c - \bar{k}'_c) \quad (2-1-8)$$

$$H'_{V_i V_i} = \frac{-eA}{m_{V_i}^* c} (\hbar k_{V_i} \cdot \alpha) \delta(\hbar k_{V_i} - \hbar k'_{V_i}) \quad (2-1-9)$$

where  $m_c^*$  and  $m_{V_i}^*$  are the effective masses of the electron in the conduction band and the hole in the valence band.

Substituting these values in equation (2-1-4) we get  $W_i$  (see Appendix I).

$$W_i = \frac{2 \frac{17}{2} \pi e^4}{\epsilon c^2 (\hbar \omega)^6} \frac{|(\bar{\alpha} \cdot \hat{p})_{cV_i}|^2}{m^2} m_{cV_i}^* (2\hbar\omega - E_g)^{3/2} I^2 \quad (2-1-10)$$

where  $m$ : free electron mass

$$m_{cV_i}^* = \frac{m_c^* m_{V_i}^*}{m_c^* + m_{V_i}^*}$$

$\epsilon$ ; dielectric constant

$E_g$ ; Energy gap

and  $I$  is the intensity of radiation in the medium

$$I = v N \hbar \omega = \frac{v \epsilon E^2}{8\pi} = \frac{v \omega^2 \epsilon A^2}{8\pi c^2}$$

Since  $\vec{E} = -\frac{1}{c} \frac{\partial \vec{A}}{\partial t}$

$$A \sim e^{i\omega t}$$

$$E = \frac{\omega}{c} A$$

$$A = |\vec{A}|$$

$$E = |\vec{E}|$$

Note that  $v = \frac{c}{\epsilon^{1/2}}$

$N$  = photon density

$\bar{E}$  = electric field intensity

$\bar{A}$  = the vector potential

Thus the transition probability per unit volume  $W_i$  is

$$W_i = \frac{W_i}{V} \frac{2 \frac{17}{2} \pi e^4}{\epsilon c^2 (\hbar \omega)^6} \frac{|(\hat{p} \cdot \bar{\alpha})_{cV_i}|^2}{m^2} m_{cV_i}^{*1/2} (2\hbar\omega - E_g)^{3/2} I^2$$

$$W = W_1 + W_2 = a I^2 \quad (2-1-11)$$

Absorption coefficient  $K_i$  is defined as follows

$$K_i = - \frac{1}{I} \frac{dI}{dX} = \frac{2\hbar \omega W_i}{I}$$

where  $X$  is the penetration depth of radiation from the surface of the crystal

$$K_i = \frac{2 \frac{19}{2} \pi e^4}{\epsilon c^2 (\hbar \omega)^5} \frac{|(\hat{p} \cdot \bar{\alpha})_{cV_i}|^2}{m^2} m_{cV_i}^{*1/2} (2\hbar\omega - E_g)^{3/2} I$$

Total two photon absorption coefficient is sum of  $K_i$ 's for  $i=1,2$ .

$$K^{(2)} = K_1 + K_2$$

The calculation of  $K^{(2)}$  of GaAs is shown in Appendix II.

The intensity dependence on the depth of penetration into the crystal can be calculated from the following equation.

$$\frac{dI}{dX} = -2\hbar\omega W - KI = -\beta I^2 - KI \quad (2-1-12)$$

where  $W = W_1 + W_2$

X: depth of penetration

$K^{(2)}$ : two photon absorption coefficient

K: one photon absorption coefficient

$$W = \alpha I^2, \quad \beta = 2\hbar\omega\alpha = K^{(2)}/I$$

If we solve the equation (2-1-12) (see Appendix III),

$$I = \frac{I_0 \exp(-KX)}{1 + \frac{\beta I_0}{K} [1 - \exp(-KX)]}$$

where  $I_0$  is initial intensity of exciting radiation. This expression can be approximated as

$$I(X) = \frac{I_0}{\beta X I_0 + 1} \quad (2-1-13)$$

if  $KX \ll 1$ .

This is the case when two photon absorption is dominant.

If the absorption is mostly due to a one photon process,

$$I(X) = I_0 e^{-KX} \quad (2-1-14)$$



These calculations can be applied to Nd:glass laser pulse as exciting source.

## 52. The Spectral Linewidth Narrowing

As we will see from the experimental results, the spectral width above the threshold becomes very narrow but finite. Even though the power level of the exciting beam is below the threshold, the spectral width becomes narrower as the power increases. This spectral width narrowing occurs in the emission spectrum from the crystal even without resonant mirrors. This can be explained in terms of the superradiance.

### a. The Spectral Width of the Laser Output

The finite spectral width of the output of a laser oscillator is due to the presence of random phase and amplitude fluctuations in its output field. These random fluctuations are caused by the radiative spontaneous transitions from the upper laser level.

A laser oscillator can be considered as a regenerative amplifier with a continuous input of spontaneous emission noise. Thus by using the laser equivalent circuit, the expression for the spectral width of the laser output was derived by Gordon<sup>(11)</sup>, Schawlow and Townes<sup>(12)</sup>. The result, taking into account only amplitude fluctuations, is as follows.

$$\Delta\nu = \frac{2\pi h \nu_0 (\Delta\nu_c)^2}{P} \frac{N_2}{\left[ N_2 - N_1 \frac{g_2}{g_1} \right]_{th}} \quad (2-2-1)$$

where  $\Delta\nu$  : the spectral width of the laser output  
 $\nu_0$  : resonant frequency  
 $\Delta\nu_c$  : the width of passive cavity response curve  
 or cavity bandwidth  
 $P$  : the total power leaving the cavity  
 $N_1$  : the population density of the lower level  
 $N_2$  : the population density of the upper level  
 $g_1, g_2$  : the level degeneracies

$\left[ N_2 - N_1 \frac{g_2}{g_1} \right]_{th}$  : the value of the threshold

As we shall see, the foregoing arguments justify the finite linewidth observed in the laser emission spectrum.

#### b. The Spectral Width Narrowing Below the Threshold

This type of narrowing is known as the super-radiant narrowing in fluorescence radiation. The spontaneous recombination radiation emitted with a lineshape  $g(\nu)$  is amplified in passing through the medium by a factor  $e^{ag(\nu)}$  where  $a$  is a constant which is proportional to the degree of inversion. A Yariv<sup>(13)</sup> studied the superradiance. The gain constant  $\gamma(\nu)$  is given as follows.

$$\gamma(\nu) = a g(\nu) = \frac{c^2 \left( N_2 - N_1 \frac{g_2}{g_1} \right) g(\nu)}{8\pi\nu^2 t \text{ spont}} \quad (2-2-2)$$

where  $\gamma(\nu)$ : the gain of the optical system  
 $c$  : the velocity of light  
 $N_1, N_2, g_1, g_2$ ; same as in the previous section  
 $g(\nu)$ : the line shape of spontaneous recombination radiation.

Thus the intensity of the radiation at the position  $\chi$  from the surface is

$$I(X) = I_0 e^{\gamma(\nu)X} = I_0 e^{a g(\nu)X} \quad (2-2-3)$$

where  $I_0 = I(0)$

But since the line shape narrows as the light propagates,  $g(\nu)$  actually depends also on  $\chi$ . Thus we have to modify above equation.

$$I(X) = I_0 e^{a \int_0^X g(X, \nu) dX} = I_0 e^{a F(\nu)} \quad (2-2-4)$$

where  $F(\nu) = \int_0^X g(X, \nu) dX$

Yarrvet al. showed that the line shape of the spontaneous emission is

$$F(\nu) = g(\nu) \frac{e^{[\gamma(\nu) - \alpha]L} - 1}{\gamma(\nu) - \alpha} \quad (2-2-5)$$

where  $\alpha$  : a distributed loss constant  
 $l$  : the length of crystal  
 $\nu_0$  : the frequency at which the line maximum occurs.

An immediate consequence of the equation (2-2-5) is that for  $\gamma(\nu_0)l \ll 1$ ,  $F(\nu) = \text{const } g(\nu)$  and no change in the spectrum is expected. The more interesting condition is that if  $\gamma(\nu_0)l \gg 1$  but  $\alpha \gg \gamma(\nu_0)$ , no narrowing occurs since  $F(\nu) = \text{const } g(\nu)$ . If  $\gamma(\nu_0)$  is increased to a point where it becomes comparable yet smaller than  $\alpha$ ,

$$F(\nu) = \text{const } \left\{ \frac{g(\nu)}{\gamma(\nu) - \alpha} \right\}$$

which is, because of the frequency-dependent denominator, narrower than  $g(\nu)$ .

It is clear from the above that spectral narrowing, unlike laser action, is a gradual process and no exact narrowing condition can be given. At the same time we can summarize the observations made in previous part by giving the following approximate condition for "starting" of narrowing

$$\gamma(\nu_0)l \sim 1$$

(2-2-6)

$$\gamma(\nu_0) \sim \alpha$$

The physical interpretation of this result is as follows. Condition (2-2-6) is equivalent to say that a photon emitted at  $\nu_0$ , on the average, stimulates the emission of

order of 1 photon in a distance  $\ell$ . Photon emitted at the wings of  $g(\nu)$  have, according to equation (2-2-4), a much smaller probability of inducing emissions so that the condition (2-2-6) marks the point at which the photon distribution is beginning to vary appreciably from  $g(\nu)$ . If we limit ourselves to cases where

$$\begin{aligned} \gamma(\nu_0)\ell &\gg 1 \\ \gamma(\nu_0) &> \alpha \end{aligned} \tag{2-2-7}$$

and define  $\Delta\nu$  as the width at half-maximum of  $F(\nu)$  and  $(\Delta\nu)_g$  as the width at half-maximum of  $g(\nu)$ , Gordon derived the amount of narrowing  $\Delta\nu/(\Delta\nu)_g$  at a given value of  $\gamma(\nu_0)\ell$ . The result is

$$\frac{\Delta\nu}{(\Delta\nu)_g} = [\gamma(\nu_0)\ell]^{-1/2} \text{ for a Gaussian } g(\nu) \tag{2-2-8}$$

$$\frac{\Delta\nu}{(\Delta\nu)_g} = 0.832 [\gamma(\nu_0)\ell]^{-1/2} \text{ for a Lorentzian } g(\nu) \tag{2-2-9}$$

### §3. The Threshold Pumping Power for Oscillation and Lasing

First let's discuss about the oscillation condition.

The rate equation for the electrons in conduction band is as follows.

$$\frac{dn_c}{dt} = W - \frac{n_c}{\tau_S} \tag{2-3-1}$$

where  $n_c$ ; electron concentration in conduction band  
 $W$ ; transition probability  
 $\tau_s$ ; spontaneous lifetime of electron

The rate of increase of electrons in conduction band is the difference between stimulated upward transition and downward transition due to spontaneous emission. The threshold condition can be determined by letting

$$\frac{dn_c}{dt} = 0$$

Then

$$W = \frac{n_c}{\tau_s} \quad (2-3-2)$$

As we derived before  $W$  is a function of  $I^2$ . Let  $W = aI^2$ .

Thus

$$aI_{os}^2 = \frac{n_c}{\tau_s} \quad \text{where } I_{os} \text{ denotes threshold pumping power density for oscillation}$$

This is the threshold for stimulated emission, not for laser action.

$$I_{os} = \sqrt{\frac{n_c}{a\tau_s}} \quad (2-3-3)$$

This is the threshold pumping power density for oscillation. But the threshold for lasing would be higher than this due to the losses. The losses are the sum of reflection losses at the mirrors of the optical cavity and the diffraction losses. Let  $K_{\text{loss}}$  be the losses. If  $\alpha$  is the gain in the laser medium, at the threshold for lasing, then the following relation is satisfied.

$$\sqrt{R_1 R_2} e^{(\alpha - K_{\text{loss}})L} = 1 \quad (2-3-4)$$

where  $R_1, R_2$ : the reflection coefficient of the resonator mirrors,  
 $L$  ; effective optical length of cavity,  
 $K_{\text{loss}}$ ; loss  
 $\alpha$  ; gain

The gain is the function of pumping power density  $I$ . This is given in the Basov's paper<sup>(1)</sup>. Then from (2-3-4)

$$\alpha(I) = -\frac{1}{2L} \ln R_1 R_2 + K_{\text{loss}} \quad (2-3-5)$$

$$\alpha(I) = K^{(i)} \frac{1}{4kT} (\mu - \hbar\omega_g) \left[ 1 + \frac{K^{(i)} I}{4kT B \hbar\omega_g} \right]^{-1} \quad (2-3-6)$$

- where
- $\kappa^{(i)}$  : absorption coefficient where  $i = 1, 2$
  - $\mu$  : Fermi quasi level of the electrons ( $\mu_e$ )  
and the holes ( $\mu_n$ )
  - $\omega_g$  : generation frequency
  - $B$  : the derivative of recombination rate with  
respect to the sum of quasi levels  $\mu$  at  
the point corresponding to the oscillation  
threshold  $\mu_{OS}$ .
  - $T$  : carrier temperature
  - $k$  : Boltzman const.

From (2-3-5) and (2-3-6), the threshold pumping power density  $I_{OS}$  for lasing can be derived.



## CHAPTER III

### EXPERIMENT

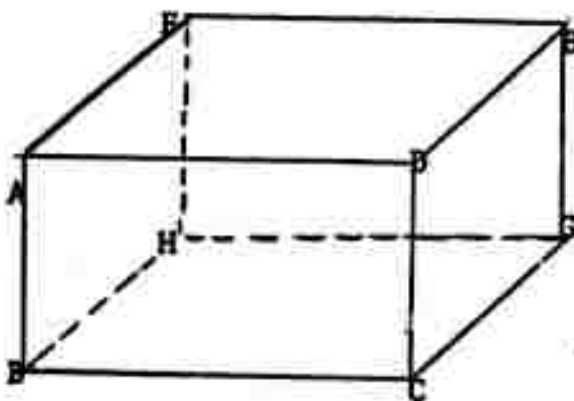
First, the emission spectra of GaAs were observed at different levels of pump power density. Secondly, the dependence of output power from the GaAs crystal was studied. To carry out these experiments the power calibration of the photodiode was required. The arrangement of the pumping laser system was important but it is described elsewhere. (See Appendix IV.)

In this chapter mainly describes the design of the experiments. The results are discussed in the next chapter.

#### §1. The Spectral Emission Lines from GaAs

First of all we require the pumping source in order to get spontaneous and stimulated recombination radiation. The Nd:glass laser was used as the pump. The maximum power density was approximately 50 MW per unit area ( $\text{cm}^2$ ) at the front surface of the GaAs crystal. The description of exciting pulse is shown in §4 of this chapter.

The GaAs crystal with dimensions of 4 mm x 5 mm x 10 mm, has a carrier concentration, i.e., impurity density, of about  $2 \times 10^{17}/\text{cm}^3$ . The mobility of the crystal is approximately  $4000 \text{ cm}^2/\text{v-sec}$  at room temperature.



$$AD = 10 \text{ mm}$$

$$EG = 4 \text{ mm}$$

$$CG = 5 \text{ mm}$$

Figure 3-1-1. Dimension of the GaAs Crystal

The Figure 3-1-1 is given for convenience to identify directions of surfaces which will be referred later.

The bulk crystal was prepared such that the surfaces of bulk coincided with the specific planes of GaAs crystal-line structure. GaAs has the cubic zinc sulfide structure. The positions of the Ga atoms in the unit cell are at  $0,0,0$ ;  $0, \frac{1}{2}, \frac{1}{2}$ ;  $\frac{1}{2}, 0, \frac{1}{2}$ ;  $\frac{1}{2}, \frac{1}{2}, 0$  in the Cartesian coordinate whose unit length is lattice constant. The position of the  $A_s$  atoms in the unit cell are at  $\frac{1}{4}, \frac{1}{4}, \frac{1}{4}$ ;  $\frac{1}{4}, \frac{3}{4}, \frac{3}{4}$ ;  $\frac{3}{4}, \frac{1}{4}, \frac{3}{4}$ ;  $\frac{3}{4}, \frac{3}{4}, \frac{1}{4}$ . The space lattice type is fcc. The surfaces  $\square ABCD$  and  $\square EGHG$  are  $\{1\ 1\ 0\}$  planes and  $\square ABHF$  and  $\square DCGE$  are  $\{\bar{1}\ 1\ 0\}$  and  $\square ADEF$  and  $\square BCGH$  are  $\{0\ 0\ 1\}$  planes. The

orientation of the plane (hkl) is expressed in Miller indices. Optical polishing was done on all faces, with particular emphasis on the two 4mm x 5mm end faces. Broad band anti-reflection coating was done on two 4mm x 5mm end faces and on one of the (1 1 0) faces (i.e., □ABCD face shown in the Figure 3-1-1; the wavelength ranges from 8200 Å to 1.1 μ for reflectivity less than 1%). The face □ ABCD was placed perpendicular to the exciting neodymium-glass laser beam. The crystal was attached to a cold finger placed in liquid nitrogen (77°K). The diagram of dewar which contained the cold finger and crystal in vacuum is shown in Appendix VI. The direction of the Nd:glass laser beam was changed by 180° using a prism and a mirror mounted on a vernier. This was done because another experiment required the same Nd:glass laser. The prism (P) could be moved in and out of the path of the Nd:glass laser beam. All these arrangements are shown in Figure 3-1-2 in block diagram. The polarizer was used to be sure that the beam was polarized in one direction. In this case it was horizontally polarized. The half wave plate was used for changing the polarization to the vertical direction. Calibrated neutral density filters were used to vary the pump intensity incident on the crystal. The diameter of the spot of the beam was larger than the width of the crystal surface on which exciting radiation was incident. We also used a set

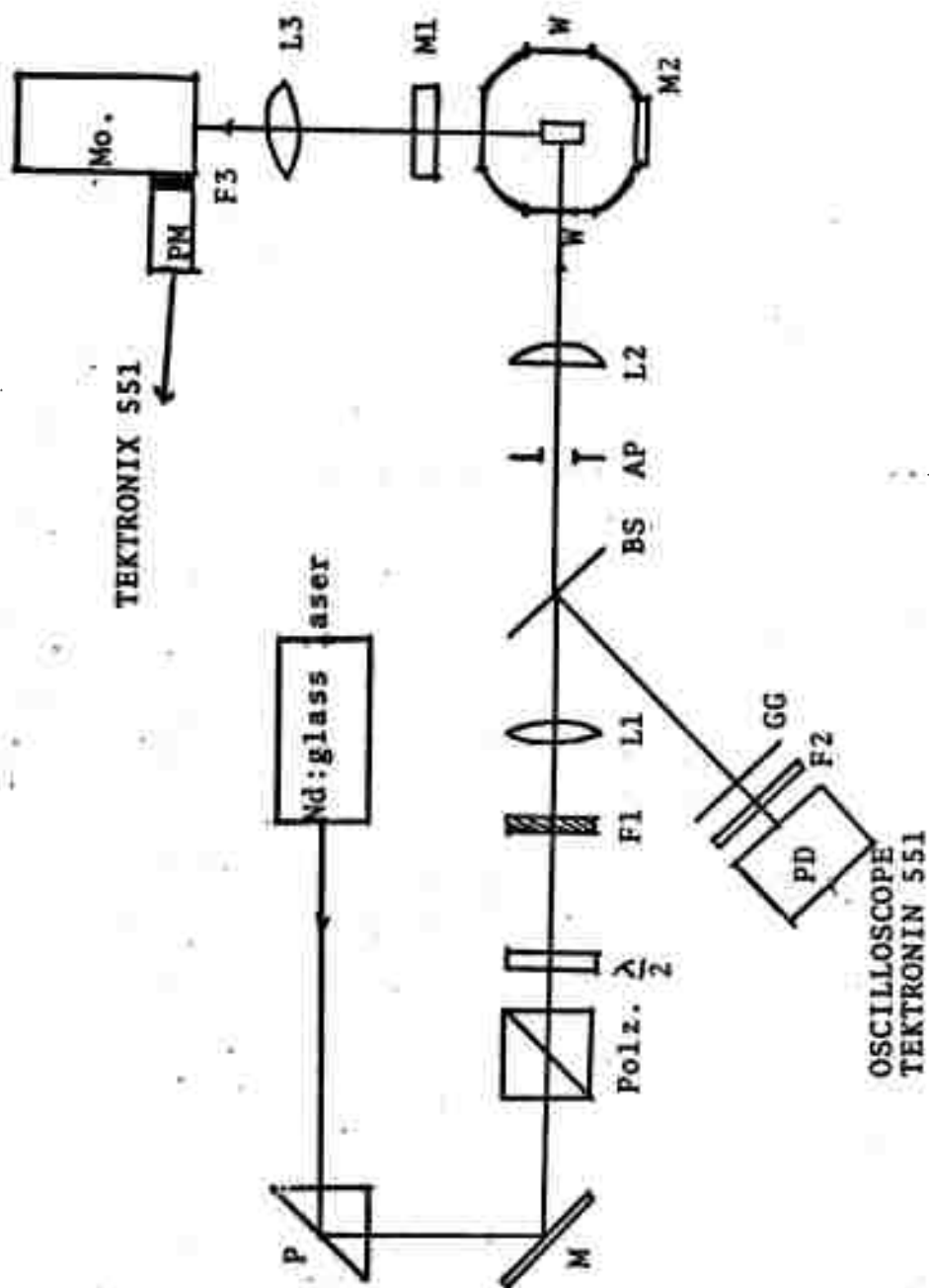


Figure 3-1-2.

## NOTATIONS in Figure 3-1-2

P : Prism  
M : Mirror  
Polz: Polarizer  
 $\frac{\lambda}{2}$  : Half wave plate  
F1 : Neutral density filter  
F2 : Neutral density filter  
F3 : Filter for filtering flash light (No. 2-58 Corning  
glass color filter)  
L1 : Lens  
L2 : Cylindrical lens  
AP : Aperture  
M1 : Mirror  
M2 : Mirror  
D : Dewar  
S : Sample  
W : Window  
Mo : Monochromator (Jarrel Ash 82-020)  
PM : Photomultiplier (RCA 7102)  
PD : Photodiode (RCA 917)

of lenses L1 and L2. As the crystal was shorter in height than in length, i.e.,  $\overline{AB} < \overline{AD}$ , we used the cylindrical lens (L2) to narrow beam spot along the AB direction. The aperture (AP) was used as the reference point to check correctness of the direction of exciting beam. As the Nd:glass laser pulses were not always of same power level even for the same operating conditions, i.e., for the same dye concentration and the same operating voltage, it was necessary to monitor the exciting laser pulse. If we a put beam splitter (BS) after lens (L1), then part of the beam enters photodiode (PD) through the ground glass (GG) and neutral density filter (F2). The ground glass and neutral density filter prevent saturation of the photodiode. The details of the photodiode circuit diagram is presented in Appendix V. The GaAs crystal was attached to the cold finger in the dewar (D) which was evacuated by the air pumping. The cold finger was mounted on the container in which we supplied liquid nitrogen. The crystal and cold finger remained at liquid nitrogen temperature, 77°K. There were three windows on the cylindrical wall of dewar. In one side of the crystal one mirror (M2) which had 99% reflectivity was placed. The other mirror (M1) was mounted on a vernier mount. These M1 and M2 constitute the laser cavity. The generated radiation from the GaAs laser was focused by the lens (L3) and sent into the entrance slit of the monochromator.

The signals from the photodiode (PD) and from the photomultiplier (PM) were connected to the upper and the lower input terminals of a dual beam oscilloscope, respectively. As we will see in next section, we can calibrate the photodiode such that the peak value of the pulse in the photographs corresponds to the power of the laser pulse. For a fixed power level, we took photographs of the exciting and emitted radiation pulses as we varied the wavelength dial of the monochromator. Figure 4-1-1 shows the emission spectrum of GaAs crystal.

## §2. The Dependence of the Power Density Radiated by GaAs upon the Exciting Light Intensity

It was necessary to calibrate the photodiode in order to relate the power and the peak value of the pulse photographed from the oscilloscope.

The arrangement of the experimental equipment and the result of the experiment will be presented later.

First, the output power dependence will be discussed. The experimental setup is the same as in previous experiment. Here we fixed the wavelength dial of the monochromator at the value corresponding to peak value in emission spectrum, i.e., 8386 Å. Then we introduced neutral density filters in F1 in Figure 3-1-2. The exciting pulse was monitored using the photodiode (PD) and sent to upper trace terminal

of the Tektronix 551 oscilloscope; the radiation from the GaAs crystal was collected through monochromator, amplified by photomultiplier (PM) and sent to lower trace terminal of the oscilloscope. The two signals were then simultaneously photographed. The result of this experiment after analyzing the data is given in Chapter IV.

### §3. The Power Calibration of the Photodiode

This was necessary to estimate the radiated power. In this experiment the power of the emission spectrum had to be estimated because we had to find the power level where the narrowing of the width in emission spectrum occurred. In the second experiment it was more necessary because we had to know the power or intensity of the exciting beam. Figure 2-1-5 shows the arrangement of experimental equipment for calibration of photodiode. The neutral density filters (F1) and (F), the lens (L1), beam splitter (BS) and ground glass (GG) were used for same reasons which were discussed previously. The lens (L2) was used to focus the beam into the hole of the thermopile. The thermopile was calibrated as following.

$$[\mu\text{V reading}/198.2] = \text{The energy [joules]}$$

If we can measure the pulse width, then we can calculate power as follows.



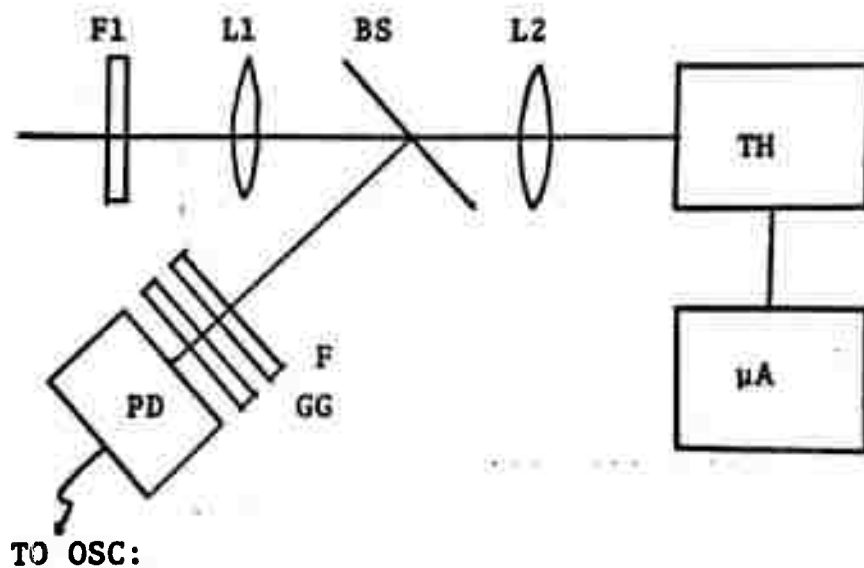
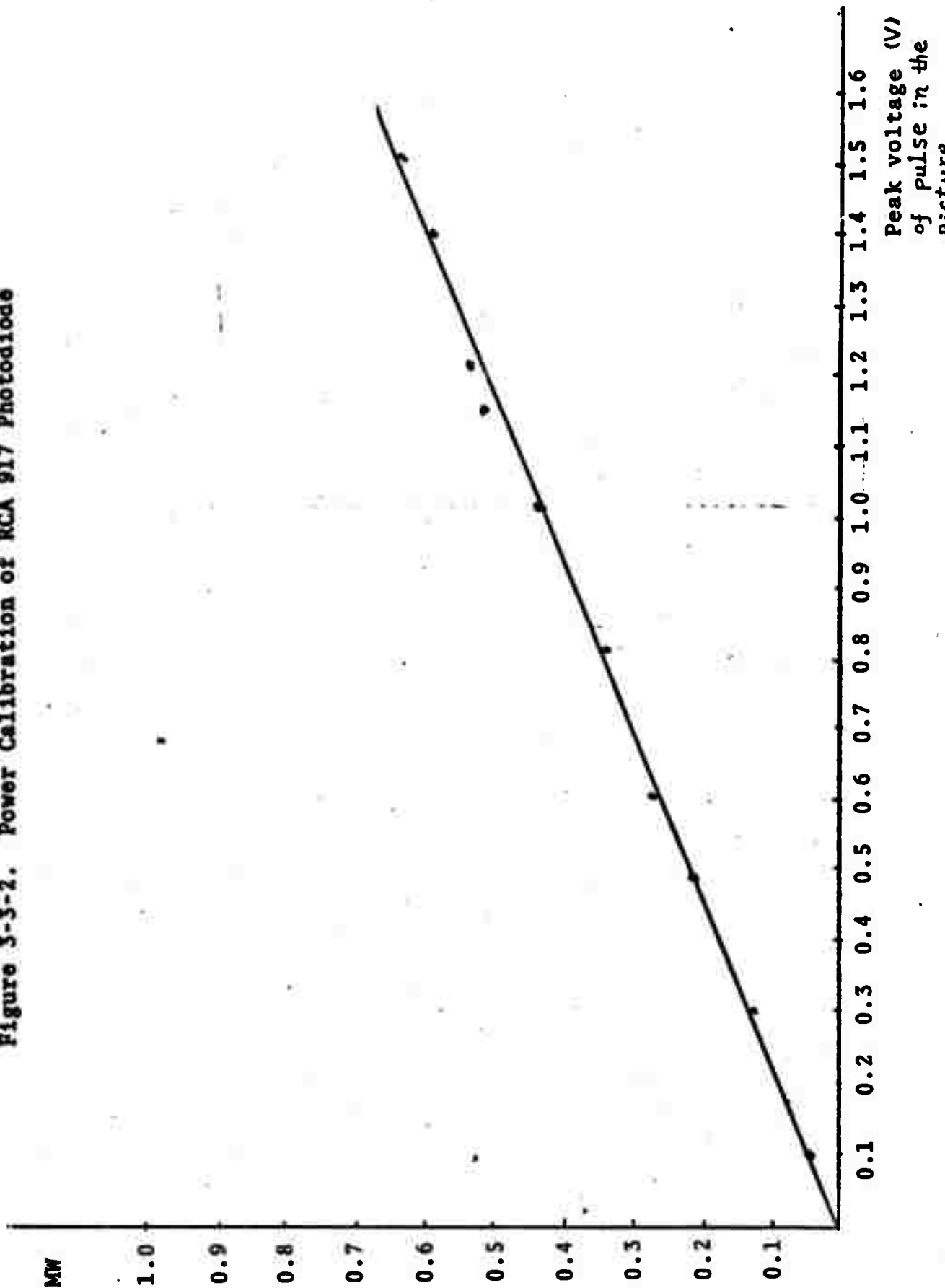


Figure 3-3-1.

- F1: Neutral density filter
- L1: lens
- L2: lens
- BS: beam splitter
- F : ND 2.0 neutral density filter
- GG: Ground glass
- TH: TRG 100 thermopile
- $\mu$ A: Microammeter

Figure 3-3-2. Power Calibration of RCA 917 Photodiode



$$\text{Power in MW} = \frac{\mu\text{V}}{198.2} \times \frac{10^9}{W} \times \frac{1}{10^6}$$

where  $\mu\text{V}$ ; microvoltmeter reading in [ $\mu\text{V}$ ]

$W$  ; width of the pulse in [n sec] .

If we divide power by the area of the spot of the beam on the surface of the sample, we can calculate power density  $I$ .

$$I = \frac{\text{MW}}{S}$$

where  $\text{MW}$  is power in [MW]

$S$  is area of beam spot in [ $\text{cm}^2$ ]

$I$  is power density in  $\left[ \frac{\text{MW}}{\text{cm}^2} \right]$

Figure 3-3-2 shows the power vs. peak of the pulse which was taken on the photograph.

#### §4. The Pumping Source

As we mentioned before, Nd:glass laser was used as the pumping source. The Nd:glass laser rod was of dimensions, 0.5" diameter and 8.31" length. The ends of the rod were cut at Brewster angles to avoid reflection losses. The rod was placed inside an open resonator consisting of two mirrors (99.9% reflectivity and 65% reflectivity at 1.06  $\mu$ ). The Nd:glass laser was Q-switched with a dye

cell inside the cavity. Eastman Kodak dye solution (No. 9860) was used. The Q-switched pulse had a maximum power of about  $50 \text{ MW/cm}^2$  with a pulse width of about 150 n sec. To insure that only the Q-switched nanosecond pulse was generated we inserted a mode selector inside the laser cavity to prevent mode-locking.

## CHAPTER IV

### THE EXPERIMENTAL RESULTS AND DISCUSSION

#### §1. The Emission Spectrum from GaAs

Figure 4-1-1 shows the emission spectrum of GaAs crystal. The ratio of output power density of the emitted radiation with respect to the power density of exciting beam was plotted on the y-axis and the wavelength of emission spectrum on the x-axis. The curve 1 in Figure 4-1-1 is the emission spectrum at the exciting power density, approximately  $2\text{MW}/\text{cm}^2$ . The curve 2 is that at  $5\text{MW}/\text{cm}^2$ . The curve 3 is that at  $10\text{MW}/\text{cm}^2$ . We increased the level of exciting power density by reducing the value of the neutral density filter (F1). In Figure 4-1-1, we can see that the width of the emission spectrum of curve 1 was about  $390 \text{ \AA}$  and it was narrowed to about  $200 \text{ \AA}$  at curve 2 and then it was narrowed abruptly down to  $16 \text{ \AA}$ . We expected these narrowings of the widths of the emission spectrums as discussed in Chapter II. We saw that there were mainly two different reasons for narrowing of the width of the emission spectrum.

First, let's discuss about the linewidth narrowing due to the laser action. The linewidth of the emission spectrum in curve 2 was about  $200 \text{ \AA}$ . It was narrowed abruptly to about  $16 \text{ \AA}$  in curve 3. This indicates there occurs laser action. But because of the limited resolution of the

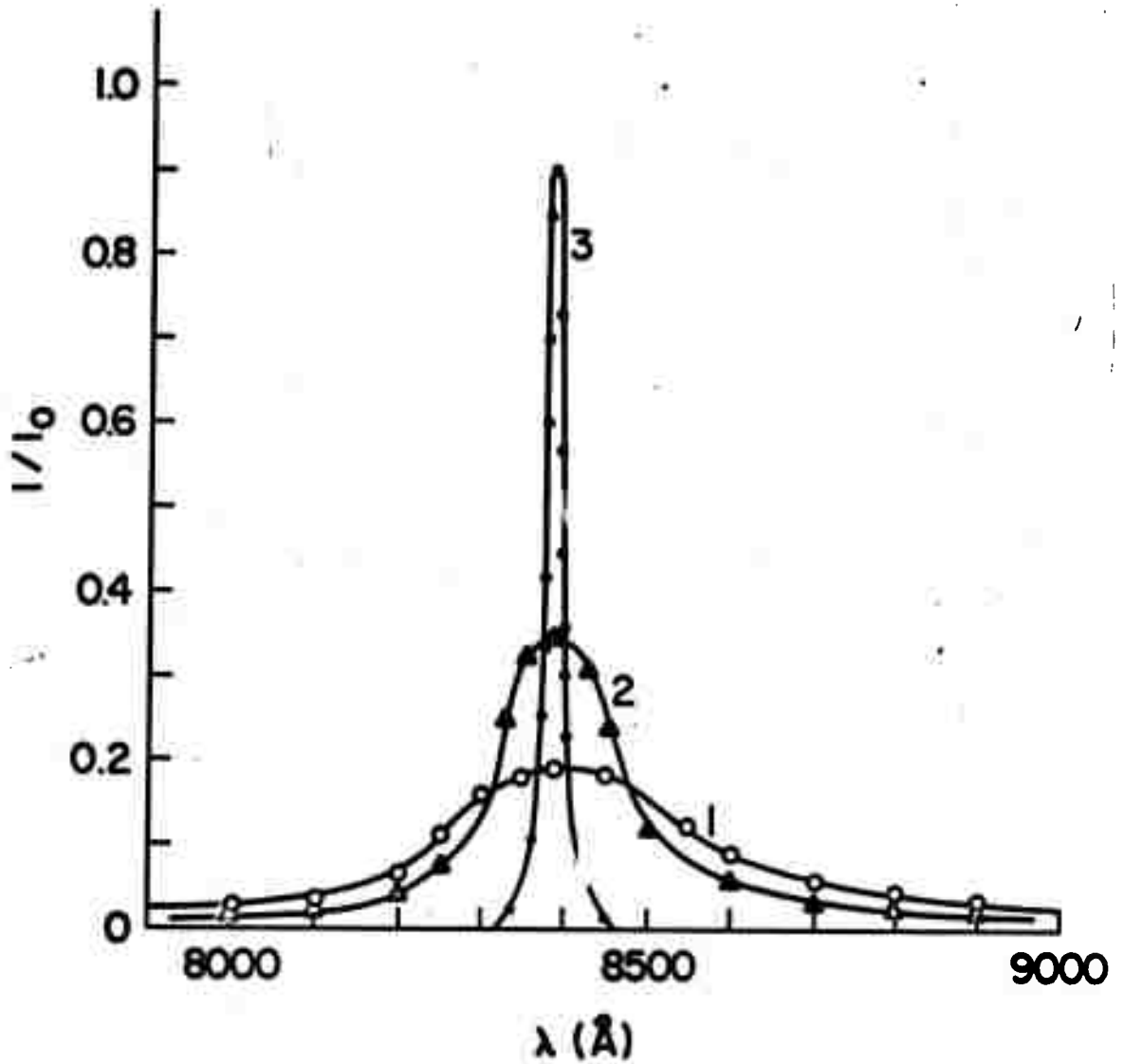


FIG4-1-1 THE VERTICAL SCALE OF CURVE 1,2 IS  
MAGNIFIED 8 TIMES.

monochromator, it was hard to detect a spectral line narrower than the order of  $10 \text{ \AA}$ . Next note that the width of  $390 \text{ \AA}$  of curve 1 narrowed to  $200 \text{ \AA}$  of curve 2.

The power density of exciting beam was about  $5 \text{ MW/cm}^2$  for curve 2. As we can see in next section it was below the threshold. This is the narrowing due to the super-radiance. If we increase the power density of the exciting beam then according to the equation (2-2-2),  $\gamma(\nu_0)$  increases. Then in equation (2-2-8)

$$\frac{\Delta\nu}{(\Delta\nu)_g} = [\gamma(\nu_0)\ell]^{-1/2},$$

we can see  $\Delta\nu$  decreases as  $\gamma(\nu_0)\ell$  increases. This justifies the line narrowing below threshold.

## §2. The Dependence of Power Density of Radiation from GaAs on the Exciting Light Intensity

Using the data we got in the experiment of §2, Chapter III, we can draw a graph shown in Figure 4-2-1. As the photodiode was calibrated in Figure 3-3-2, we can find absolute power density of exciting signal. Figure 4-2-1 shows the graph in x-axis correspond to the intensity of the exciting beam and y-axis correspond to the output power in arbitrary scale. It can be easily seen that at  $5\text{-}6 \text{ (MW/cm}^2\text{)}$  power level the power of emitted radiation

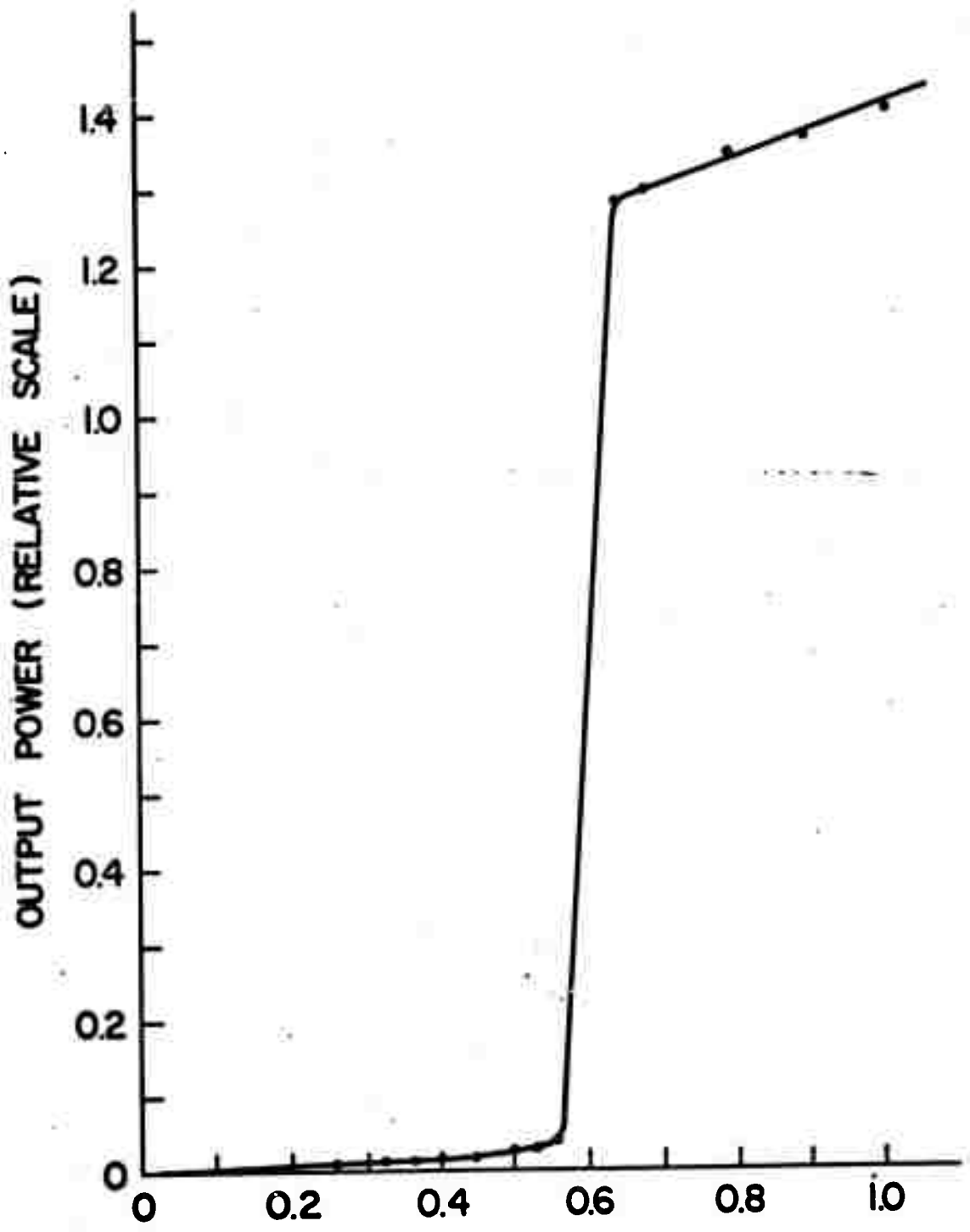


FIG 4-2-1 INPUT POWER ( $\times 10$  MW)



jumps up to a higher level. So we can say this value corresponds to the threshold for lasing. Of course, this data was taken by varying the value of the neutral density filter. It was better to start at a low power level, because at high power levels the surface of the crystal may be damaged. According to the experiment approximately  $50 \text{ MW/cm}^2$  was the maximum intensity tolerated by the crystal. If the power density of incident beam is higher than this, it might give damage on the surface of the sample. According to the Figure 4-2-1 it is obvious that the part of the graph below threshold is due to spontaneous emission, and the part of the graph above threshold is because of the stimulated emission. The output power when the GaAs is lasing, is 30~50 times that of the fluorescence below the threshold. In the threshold region, the ratio is around 30.

The threshold power density for oscillation can be calculated using the equation (2-3-3)

$$I_{OS} = \sqrt{\frac{n_c}{a\tau_s}}$$

The threshold concentration of electrons in the conduction band can be calculated using the population inversion condition. The iteration method is presented in C. Chang's<sup>(8)</sup> paper. Dumke<sup>(7)</sup> calculated the radiative lifetime of an electron in the conduction band of semiconductor GaAs. Here it is only necessary to estimate the level of threshold intensity

According to above papers,

$$n_c \sim 10^{19} \text{ electrons/cm}^3$$

$$\tau_c \sim 10^{-7} \text{ sec}$$

From appendix II  $a = 0.15 \left[ \frac{\text{cm} \cdot \text{sec}}{\text{erg}^2} \right]$

Since  $a I_{OS}^2 = 10^2$  and we want  $I_{OS}$  in  $\left[ \frac{\text{MW}}{\text{cm}^2} \right]$ ,

$$0.15 I_{OS}^2 \times (10^7)^2 \times (10^6)^2 = 10^{26}$$

$$I_{OS}^2 \sim 6.7$$

$$I_{OS} \sim 2.6 \text{ [MW/cm}^2\text{]}$$

This is the threshold pumping power density for oscillation. The threshold for lasing would be higher than this. The experimental value was about 5 MW/sec. If we recall that approximate values of electron density and spontaneous lifetime were used, and if we admit experimental deviation, the order of the experimental result can be considered good enough.

# APPENDIX I

## DERIVATION OF $W_i$

$$W_i = \frac{2\pi}{\hbar} \left( \frac{1}{\hbar\omega} \right)^2 \iint d^3k_c d^3k_{V_i} (H'_{cc} H'_{cV_i} + H'_{cV_i} H'_{V_i V_i})^2 \times \delta(E_c - E_{V_i} - 2\hbar\omega) \quad (1)$$

$$H'_{cV_i} = \frac{eA}{m_c c} (\mathbf{P} \cdot \bar{\alpha})_{cV_i} \delta(\bar{k}_c - \bar{k}_{V_i}) \quad (2)$$

$$H'_{cc} = \frac{eA}{m_c c} (\hbar k_c \alpha) \delta(\bar{k}_c - \bar{k}'_c) \quad (3)$$

$$H'_{V_i V_i} = \frac{-eA}{m_{V_i} c} (\hbar k_{V_i} \alpha) \delta(\bar{k}_{V_i} - \bar{k}'_{V_i}) \quad (4)$$

Substitute equations (2), (3) and (4) in (1). Note that

$$\int \psi_c^* \psi_{V_i} d^3r = \delta(\bar{k}_c - \bar{k}_{V_i})$$

$$\frac{V}{(2\pi)^3} \int U_c^* U_{V_i} d^3r = \langle U_c | U_{V_i} \rangle = \delta_{cV_i}$$

Thus

$$\delta^2(\bar{K}) = (2\pi)^{-3} V \delta(\bar{K})$$

After operating  $\delta$  functions,

$$\bar{K}_c = \bar{K}_{V_i}$$

so let  $\bar{K}_c = \bar{K}_{V_i} = \bar{K}$

Then

$$W_i = \frac{W_i}{V} = \frac{2\pi}{\hbar} \left(\frac{1}{2\pi}\right)^3 \left(\frac{1}{\hbar\omega}\right)^2 \frac{e^4 A^4}{m^2 c^4} \hbar^2 \alpha^2 |(\bar{P} \cdot \bar{\alpha})_{cV_i}|^2 \left(\frac{1}{m_c^*} + \frac{1}{m_{V_i}^*}\right) \\ \times \int d^3k k^2 \delta(E_c - E_{V_i} - 2\hbar\omega)$$

$$\text{Let } m_{cV_i}^* = \frac{m_c^* m_{V_i}^*}{m_c^* + m_{V_i}^*}$$

Note that

$$E_c - E_{V_i} = E_g + \left( \frac{\hbar^2 k_c^2}{2m_c^*} + \frac{\hbar^2 k_{V_i}^2}{2m_{V_i}^*} \right) \\ = \frac{\hbar^2 k^2}{2m_{cV_i}^*} + E_g$$

$$\delta(f(X)) = \frac{d(X-X_0)}{\left| \frac{df(X)}{dX} \right|} \quad \text{where } f(X_0) = 0$$

Thus

$$d(E_c - E_{V_i} - 2\hbar\omega) = \delta\left(\frac{\hbar^2 k^2}{2m_{cV_i}} + E_g\right)$$

$$\frac{\delta(k-k')}{\frac{\hbar^2 k'}{m_{cV_i}}} = \frac{m_{cV_i}}{\hbar^2 k'} \delta(k-k')$$

Let

$$I = \int d^3k k^2 \delta(E_c - E_{V_i} - 2\hbar\omega)$$

$$I = \int d^3k k^2 \frac{m_{cV_i}}{\hbar^2 k'} \delta(k-k')$$

$$= \int d^3k' k' \frac{m_{cV_i}}{\hbar^2}$$

$$= \frac{m_{cV_i}}{\hbar^2} \int k' d^3k'$$

Since  $d^3k' = 4\pi k'^2 dk'$

$$I = \frac{m_{cV_i}^* 4\pi}{\hbar^2} \int dk' k'^3 \delta(k-k')$$

$$= \frac{m_{cV_i}^*}{\hbar^2} 4\pi k^3$$

$$\frac{\hbar^2 k^2}{2m_{cV_i}^*} + E_g = E_c - E_{V_i} = 2\hbar\omega$$

$$k^2 = \frac{2m_{cV_i}^*}{\hbar^2} (2\hbar\omega - E_g)$$

$$k^3 = \frac{2^{\frac{3}{2}} m_{cV_i}^* \frac{3}{2} (2\hbar\omega - E_g)^{\frac{3}{2}}}{\hbar^3}$$

Thus

$$I = \frac{2^{\frac{7}{2}} \pi m_{cV_i}^* \frac{5}{2}}{\hbar^5} (2\hbar\omega - E_g)^{\frac{3}{2}}$$

Thus

$$W_i = \frac{2\pi}{\hbar} \left(\frac{1}{2\pi}\right)^3 \left(\frac{1}{\hbar\omega}\right)^2 \frac{e^4 A^4}{m^2 c^4} \hbar^2 \alpha^2 |(\hat{P} \cdot \bar{\alpha})_{cV_i}|^2 \frac{1}{m_{cV_i}^*}$$

$$\times \frac{2^{\frac{7}{2}} \pi m_{cV_i}^* \frac{5}{2}}{\hbar^5} (2\hbar\omega - E_g)^{\frac{3}{2}}$$

The intensity of radiation in the medium

$$I = v N \hbar \omega = v \epsilon \frac{E^2}{8\pi} = \frac{v \omega^2 \epsilon A^2}{8\pi c^2}$$

since  $E = -\frac{1}{c} \frac{\partial A}{\partial t}$

$$A \sim e^{i\omega t}$$

$$E = \frac{\omega}{c} A$$

If we take account spin degeneracy, and  $\alpha^2 = 1$ ,

$$W_i = \frac{2^{\frac{17}{2}} \pi e^4}{\epsilon c^2 (\hbar\omega)^6} \frac{|(\hat{P} \cdot \bar{\alpha})_{cV_i}|^2}{m^2} m_{cV_i}^* \frac{1}{2} (2\hbar\omega - E_g)^{\frac{3}{2}} I^2$$

## APPENDIX II

### CALCULATION OF TWO PHOTON ABSORPTION COEFFICIENT

$$W = \alpha I^2 \left[ \frac{1}{\text{sec} \cdot \text{cm}^2} \right]$$

where

$$\alpha = \sum_{i=1,2} \frac{2^{\frac{17}{2}} \pi e^4}{\epsilon c^2 (\hbar\omega)^6} (2\hbar\omega - E_g)^{\frac{3}{2}} \frac{|(\hat{P} \cdot \vec{\alpha})_{cV_i}|^2}{m} m_{cV_i}^* \frac{1}{2}$$

$$e : 4.8 \times 10^{-10} \text{ [stat coulomb]}$$

$$\epsilon : 11.8$$

$$\hbar\omega : 1.17 \text{ [ev]} = 1.17 \times 1.6 \times 10^{-19} \times 10^7 \text{ [erg]}$$

$$m_c^* : 0.072 m$$

$$m_{V_1}^* : 0.68 m$$

$$m_{V_2}^* : 0.085 m$$

$$m : 9.1 \times 10^{-28} \text{ [g]}$$

$$m_{cV_i}^* : \frac{m_c^* m_{V_i}^*}{m_c^* + m_{V_i}^*}$$

$$\frac{|(\hat{P} \cdot \vec{\alpha})_{cV_i}|}{m^2} = \frac{3E_g}{2m_c^*}$$

$$E_g = 1.51 \text{ [ev]} = 1.51 \times 1.6 \times 10^{-19} \times 10^7 \text{ [erg]} \text{ at } 77^\circ\text{K}$$



Substitute these values

$$\text{Then } \alpha = 0.15 \left[ \frac{\text{cm} \cdot \text{sec}}{\text{erg}^2} \right]$$

Two photon absorption coefficient  $K^{(2)}$  is  $2\hbar\omega\alpha I$

$$K^{(2)} = 2 \times 1.17 \times 1.6 \times 10^{-19} \times 10^7 \times 0.15 \times I \text{ where } I \text{ is in } \left[ \frac{\text{erg}}{\text{cm}^2 \cdot \text{sec}} \right]$$

Thus

$$K^{(2)} = 2 \times 1.17 \times 1.6 \times 10^{-12} \times 0.15 \times I \times 10^7 \times 10^6$$

$$\text{where } I \text{ is in } \left[ \frac{\text{MW}}{\text{cm}^2} \right].$$

$$\text{If we assume } I = 1 \left[ \frac{\text{MW}}{\text{cm}^2} \right],$$

$$K^{(2)} = 5.6 \left[ \frac{1}{\text{cm}} \right]$$

## APPENDIX III

### SOLUTION OF DIFFERENTIAL EQUATION

$$\frac{dI}{dX} = - 2\hbar\omega W - KI$$

Note that  $W \equiv W^{(2)}$  and  $K \equiv K^{(1)}$ .

$$\frac{dI}{dX} = - 2\hbar\omega W - KI = - 2\hbar\omega a I^2 - KI$$

Let  $2\hbar\omega a = \beta$

$$\frac{dI}{dX} = - \beta I^2 - I$$

$$\frac{dI}{dX} = - \beta \left( I^2 + \frac{K}{\beta} I \right)$$

$$\frac{dI}{I \left( I + \frac{K}{\beta} \right)} = - \beta dX$$

$$\int_{I_0}^I \frac{dI}{I \left( I + \frac{K}{\beta} \right)} = - \int_0^X dX$$

$$\frac{\beta}{K} \ln \left. \frac{I}{I + \frac{K}{\beta}} \right|_{I_0}^I = - \beta X$$

$$\ln \left( \frac{I}{I + \frac{K}{\beta}} / \frac{I_0}{I_0 + \frac{K}{\beta}} \right) = - KX$$

$$\frac{I}{1 + \frac{K}{\beta}} = \frac{I_0}{1_0 + \frac{K}{\beta}} e^{-KX}$$

$$\frac{I}{\frac{K}{\beta}} = \frac{I_0 e^{-KX}}{I_0(1 - e^{-KX}) + \frac{K}{\beta}}$$

$$I = \frac{I_0 e^{-KX}}{1 + \frac{\beta}{K} I_0(1 - e^{-KX})}$$

when  $KX \ll 1$

$$I(X) = \frac{I_0}{2\hbar\omega a I_0 X + 1}$$

Hence,

$$I = \frac{I_0 \exp(-KX)}{1 + \frac{\beta}{K} I_0 [1 - \exp(KX)]}$$

$$\approx \frac{I_0}{1 + \frac{\beta}{K} I_0 [1 - (1 - KX)]}$$

$$= \frac{I_0}{1 + \beta I_0 X}$$

## APPENDIX IV

### ADJUSTMENT OF Nd:glass LASER

There are important factors for lasing of Nd:glass mode locked laser.

At first, even though all components of the laser were arranged according to estimation and calculation, it was not easy to get good laser pulse. In order to improve lasing, a great deal of attention should be paid to the following factors,

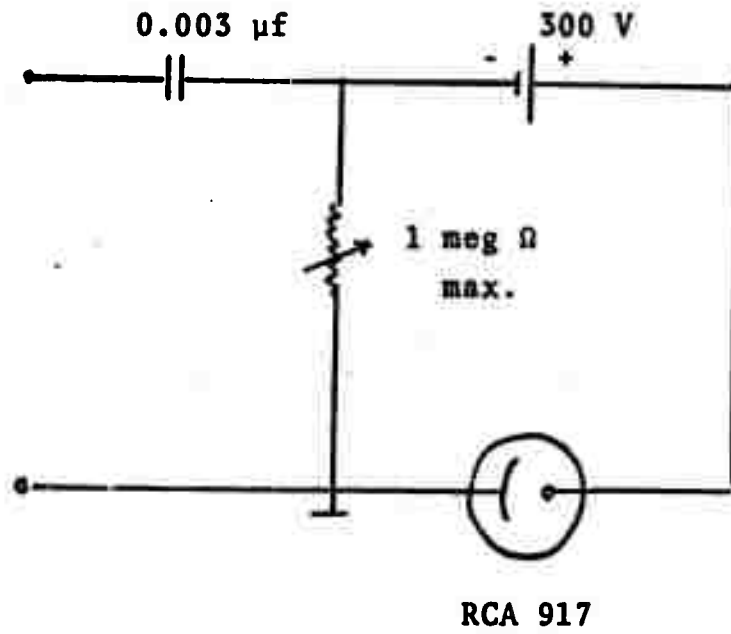
1. Microscale alignment of the equipments
2. Operating voltage of pumping flash lamp
3. Concentration of dye.

According to the experience, it was not difficult to estimate the operating level of voltage and the concentration of dye. To accomplish part 1., first it was helpful to take burn pattern on dark photographic picture. That is, we could examine the spatial coherence of the laser beam. When the burn pattern was not good, we tried same thing after adjusting venier of mirror mount. Repeating those procedures it was possible to get good shape of pulses. In the experiment, it was necessary to have single envelope of pulse. If not, it was difficult to estimate the power. To get single pulse, the operating voltage should be just above the threshold.

Difficulty occurred when the threshold came down. This was because the light from flash lamp might deteriorate the function of bleachable dye. In that case, it was needed to adjust the operating voltage. For the experiment of spectral response of GaAs, it was required to keep power level constant, so instead of adjusting operating voltage, we added dye a little bit to keep threshold constant.

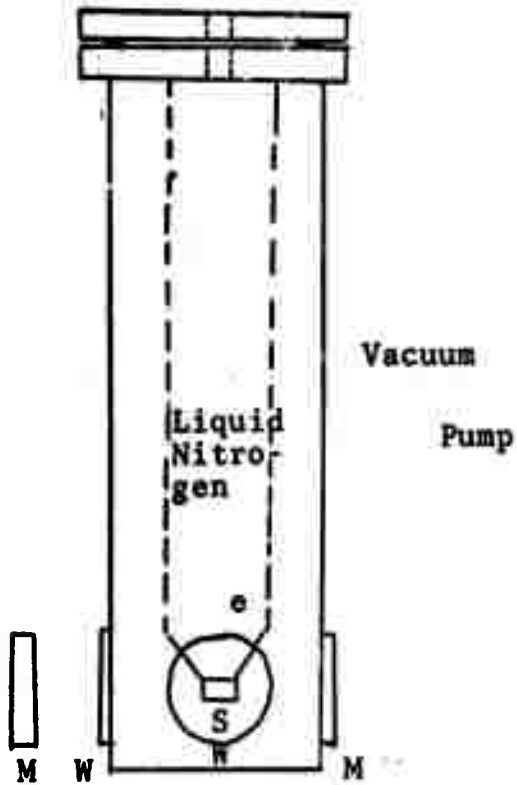
APPENDIX V

CIRCUIT DIAGRAM OF DETECTOR



# APPENDIX VI

## DEWER



**M:** Mirror

**W:** Window

**C:** Cold finger

**S:** Sample (GaAs)

## REFERENCES

- (1) N.G. Basov et al., JETP 23, 366 (1966).
- (2) R. Braunstein and N. Ockman, Phys. Rev., 134, A499 (1964).
- (3) S. Wang and C. Chang, APL 12, 193 (1968).
- (4) M. Göppert-Mayer, Ann. Physik. 9, 273 (1931).
- (5) W. Kaiser and C.G.B. Garrett, Phys. Rev. Letters, 7, 229 (1961).
- (6) G.J. Lasher and F. Stern, Phys. Rev., 133, A553 (1964).
- (7) W.P. Dumke, Phys. Rev., 132, 1998 (1963).
- (8) C. Chang, Dissertation 09-14, 853 University Microfilms, Inc., Ann Arbor, Michigan.
- (9) E.O. Kane, J. Phys. Chem. Solids, 1, 249, (1957).
- (10) T.S. Moss, J. Appl. Phys. Supplement to vol. 32, 2136 (1961).
- (11) E.I. Gordon, Bell Syst. Tech. J., Jan., 507 (1964).
- (12) A.L. Schawlow and C.H. Townes, 112, 1940 (1958).
- (13) A. Yariv and R.C.C. Leite, J. of Appl. Phys., 34, 3410 (1963).