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REPORT

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

A QUARTERLY TECHNICAL REPORT

SUBMITTED TO

THE U.S. ARMY RESEARCH OFFICE

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September 19, 1970 to December 18, 1970

REPORTED BY

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13. ABSTRACT This report covers work under contract DA-ARO-D-31-124-70-G50 for the period Sept. 19 to Dec. 18, 1970. Topics discussed include photoconductivity in single GaAs crystal via two-photon absorption of light quanta from either a Q-switched or mode-locked Nd: glass laser. The results obtained with nanosecond light pulses are compared with those obtained with picosecond case. In the latter case, the transient response of the induced charge carrier accounts for the observed effects.			

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Investigation of Two-Photon Conductivity in Gallium Arsenide Using a Nd: Glass Laser:-

Subramana Jayaraman and C. H. Lee

Introduction:

The advent of intense coherent sources of radiation by means of lasers has made it experimentally feasible to observe a number of intensity dependent optical interactions in matter which involve two or more photons. The multiphoton absorption in condensed media with the help of Q-switched or mode-locked lasers can, in principle, be investigated either by photoconductivity measurements or by observing fluorescence. The measurement of photoconductivity is a more sensitive method and so it can be used to study the multiphoton absorption in semiconductors and other crystals even with a very low absorption cross-section. In the present study, we chose to investigate the two photon conductivity in Gallium Arsenide using a Nd: glass laser with a view to examine its suitability to measure picosecond pulse width.

The two photon transition probability for atomic states was calculated by Göppert-Mayer⁽¹⁾ in 1931. Thirty years later in 1961, the first experimental observation of two-photon absorption was reported by Kaiser and Garrett⁽²⁾ in $\text{CaF}_2 : \text{Eu}^{2+}$. Basov⁽³⁾ et al reported the observation at 77°K of laser action in GaAs as a result of two-photon excitation by a Nd: glass laser beam. The two-photon excited photocurrent was studied extensively by Hasegawa and Yoshimura⁽⁴⁾ in anthracene crystals using a ruby laser. S. Wang and

C. C. Chang⁽⁵⁾ reported the emission at room temperature of coherent radiation from ZnS excited by a ruby laser through the two-photon absorption process. The two photon excited emission in CdS was intensively studied by R. Braunstein⁽⁶⁾ et al. V. S. Dneprovskü et al⁽⁷⁾ investigated the two photon photoconductivity of ZnS, CdS and CdS_x CdSe_{1-x} crystals using the ruby laser. B. V. Zubov⁽⁸⁾ et al studied the two-photon absorption in Germanium using a Q-switched CaF₂: Dy²⁺ laser by observing the recombination radiation. B. M. Ashkin- adge⁽⁹⁾ et al observed the two-photon conductivity in CdS excited by Giant pulses from Ruby laser and determined the cross-section for two-quantum absorption. J. M. Ralston and R. K. Chang⁽¹⁰⁾ measured the two photon absorption coefficients of Si, CdSe, CdTe and GaAs using a Nd:YAG Q-switched laser in their experiments investigating optical limiting in semiconductors.

Jick H. Yee⁽¹¹⁾ theoretically calculated the multiphoton conductivity in direct band-gap semiconductors and computed the two photon conductivity in GaAs as a special case. The experimental investigation of two photon conductivity in GaAs has not been made to date. So it was decided to investigate the photo conductivity induced in Gallium Arsenide by two-photon absorption. Such an investigation would throw more light in understanding the mechanism of photo conductivity and the behavior of non-equilibrium charge carriers in GaAs.

Theory

GaAs is a direct band gap semiconductor whose forbidden energy gap is 1.41 eV at room temperature. The photon energy of a Nd: glass laser is 1.17 eV. So it is possible to excite the electrons from the valence band to conduction band by the absorption of two photons simultaneously. In a single photon absorption process, the absorption coefficient is independent of the intensity of light. For the case where the charge carriers are generated by the two photon absorption, the absorption coefficient is linearly dependent on light intensity. For the band structure of GaAs shown in Fig. 1, Basov⁽³⁾ et al derived the two photon transition probability taking into account the transitions from the valence bands $V_{1,2}$ to the conduction band using straight forward 2nd order time dependent perturbation theory. The two photon absorption coefficient

$$k^{(2)} = \beta I \quad \text{where} \quad \beta = 2\hbar \omega A_o I \quad (1)$$

$$A_o = \frac{2^{17/2} \pi e^4}{\epsilon c^2 (\hbar \omega)^6} (2\hbar \omega - E_g)^{3/2} \frac{|\langle \alpha p \rangle|^2}{m^2} (m_{CV_1}^{*1/2} + m_{CV_2}^{*1/2}) \quad (2)$$

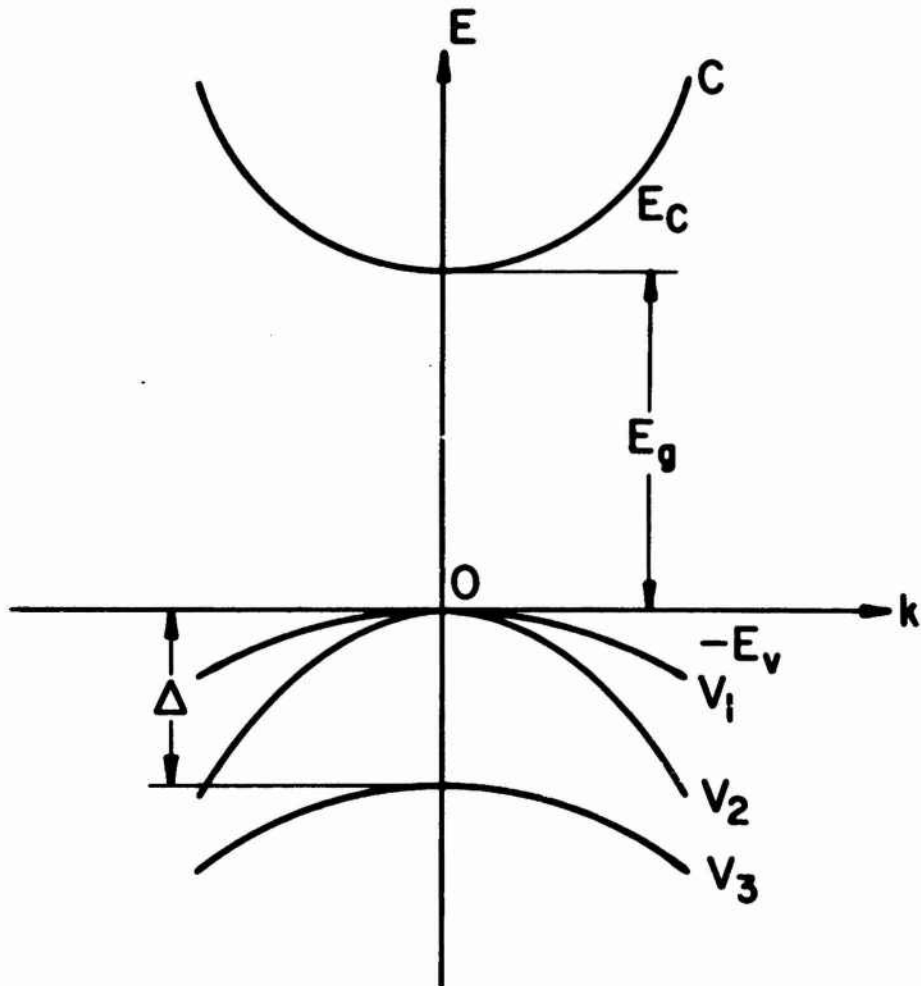
where

I = intensity of light

e = electronic charge

m = rest mass of electron

E_g = forbidden gap



BAND STRUCTURE OF Ga As

C - conduction band ; $V_{1,2,3}$ - valence bands .

E_g - forbidden gap ; Δ - split off band width .

Energy measured w.r.t. the top of valence bands V_1 and V_2 .

$E_g = 1.41 \text{ eV}$, $\Delta = 0.33 \text{ eV}$ (at 300°K) ;

Effective masses m_c^* , $m_{V_1}^*$, $m_{V_2}^*$, $m_{V_3}^*$ = $0.72 m_0$,

$0.68 m_0$, $0.085 m_0$, $0.25 m_0$; m_0 - electron rest mass.

FIGURE 1: BAND STRUCTURE OF GaAs

$\hbar\omega$ = 1.17 eV for Nd: glass laser

ϵ = permittivity of GaAs

c = velocity of light

$\langle\alpha p\rangle$ = matrix element

$$(m_{CV_i})^{-1} = m_C^{-1} + (m_{V_i})^{-1} \quad i = 1, 2$$

m_C = electron effective mass

m_{V_i} = hole effective mass

Basov derived for the intensity of light through a thickness "x" cm of the crystal for two photon absorption as

$$I(x) = I_0 (1 + 2\hbar\omega A_0 I_0 x)^{-1} \quad (3)$$

Where I_0 is the intensity at the surface of the crystal. The generation rate of charge carriers in this type of crystal as a result of two photon absorption can then be written as

$$F(x) = A_0 I(x)^2$$

The concentration of generated carriers obey the following differential equation for steady state

$$D_p \frac{\partial^2 p}{\partial x^2} - \frac{p}{\tau} = -F(x) \quad (4)$$

where τ is the steady state life time of carriers

D_p is the Diffusion Constant.

Jick H. Yee⁽¹¹⁾ solved this equation by the method of variation of parameters and used the solution to calculate the photoconductivity ΔG as

$$\Delta G = \int_0^L \int_0^{c/a} q (\mu_p + \mu_e) p(x) dy dx \quad (5)$$

The final formula for steady state two photon conductivity is as follows:

$$\Delta G = \frac{\alpha I_o^2 L A_o}{D \lambda^2 (1 + \beta I_o L)} - \frac{2 V_s I_o^2 A_o e^{\frac{-\lambda L}{2}}}{D \lambda^2 [(\lambda + V_s) - (\lambda - V_s) e^{-\lambda L}]} \times \int_0^L \frac{\cosh \lambda (\frac{L}{2} - x) dx}{(1 + \beta I_o x)^2} \quad (6)$$

$$\alpha = \frac{c}{a} q (\mu_e + \mu_p)$$

$\mu_{e,p}$ = electron, hole mobility

L = thickness of crystal

$c \times a$ = area of crystal

λ = inverse diffusion length $(D\tau)^{-1/2}$

V_s = surface recombination $(c_m^{-1}) \approx \frac{V_s^1}{D_p}$

V_s^1 = surface recombination velocity

For GaAs, whose inverse diffusion length is small, the second term in ΔG is negligible for not too high intensities, the steady state photo conductivity due to two photon absorption ΔG is given by

$$\Delta G = \frac{C}{a} q (\mu_e + \mu_p) \frac{\beta L I_o^2}{1 + \beta L I_o} \frac{\tau}{2\hbar\omega} \quad (7)$$

In the case of Q-switched laser pulses, the pulse duration is longer compared to the life time of carriers in GaAs, and so the experimental two photon conductivity could be compared to the theoretically computed values.

Experimental Set Up:-

The particular crystal used in the present investigation was an n-type GaAs of thickness 0.028 cms doped with oxygen (concentration $3 \times 10^{14}/\text{cm}^3$) and having a resistivity $2.4\Omega \text{ cm}$. A small rectangular piece was cut and ohmic contacts were alloyed at the two ends of the surface with Gold-Germanium alloy at 400°C . The crystal was exposed to Q-switched Nd: glass laser. The experimental set up is shown in Fig. 2. A dye Q-switched Nd: glass laser was used. The Q-switched laser pulse entered a pair of beam splitters through a flash lamp filter, an aperture and calibrated neutral density filters (to vary the intensity of radiation). Two photo diodes were used, one to trigger and the other to measure the intensity falling on the crystal. The GaAs crystal was connected in series with a 50Ω resistance to a battery. The change in voltage across the 50Ω resistance was fed to a dual beam oscilloscope along with the laser pulse from the photo diode. A polaroid camera was used to photograph the dual beam traces. The

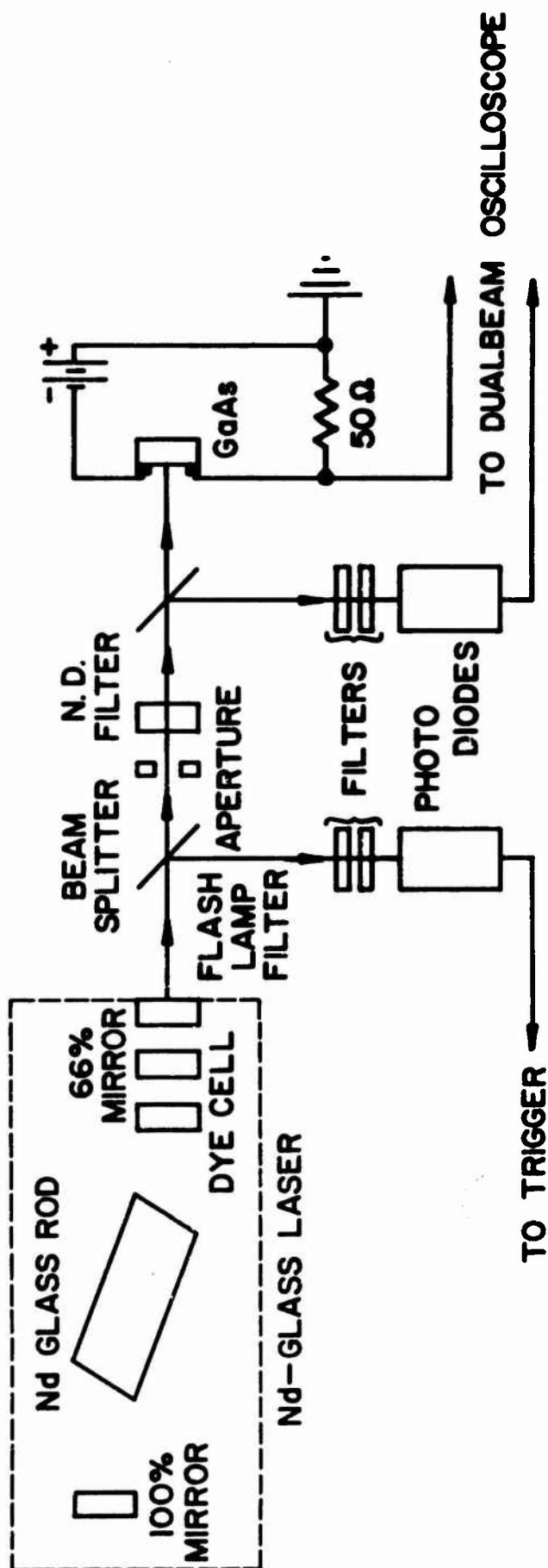


FIGURE 2: EXPERIMENTAL SET UP

photo conductivity was computed from the change in voltage across the 50Ω resistance.

Photo conductivity of GaAs due to Q-switched pulses:-

The measured photo conductivity ΔG (milli mhos) against laser intensity (MW/cm^2) is shown in Fig. 3 in a log-log graph. The maximum laser intensity was approximately $10 \text{ MW}/\text{cm}^2$ with a pulse duration of 60-80 n secs. The photo conductivity was measured over three decades of laser intensity .01 to $10 \text{ MW}/\text{cm}^2$. The result of two independent runs gave concordant results.

Figure 3 displays a break-in-slope between 1 and $2 \text{ MW}/\text{cm}^2$ and this agrees with the measured absorption coefficients (single and two photon) in earlier transmission measurements. The small signal single photon absorption coefficient for the crystal used in the experiment at 1.06μ was approximately $3-4 \text{ cm}^{-1}$ and the measured two photon absorption coefficient was $\approx 5.6 \text{ cm}/\text{MW}$ which agrees favorably with Basov's ⁽¹²⁾ experimental and theoretical values. Below $1 \text{ MW}/\text{cm}^2$, the single photon process predominates and above $1 \text{ MW}/\text{cm}^2$, the two photon absorption takes over.

Below $1 \text{ MW}/\text{cm}^2$, the slope of the curve is less than unity and is approximately equal to 0.6 thus indicating the nature of single photon absorption, probably due to impurity levels. According to A. Rose ⁽¹³⁾, the slope of the photo conductivity curve between 1/2 and 1 can possibly be explained by a continuous spectrum of level in the forbidden band

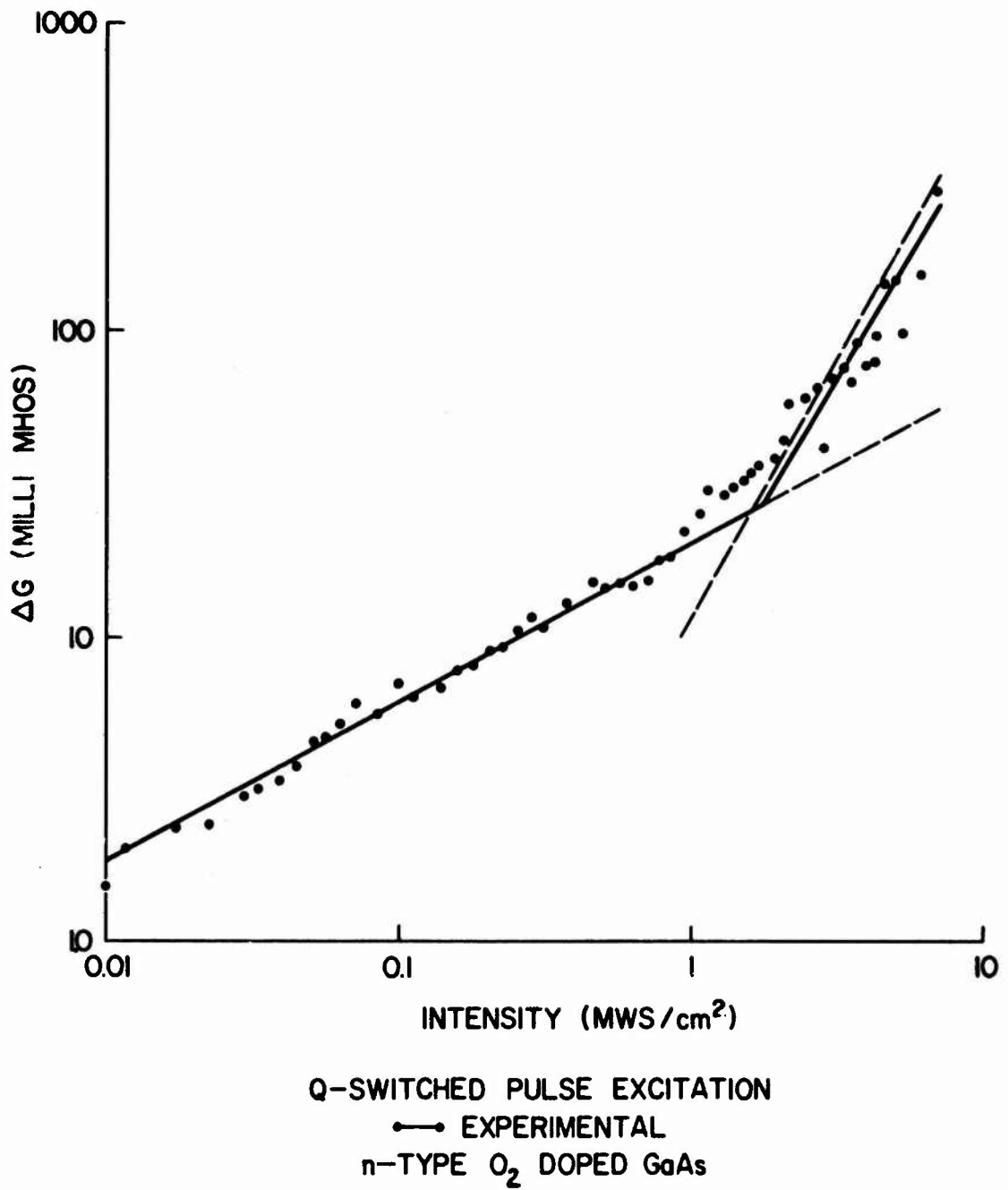


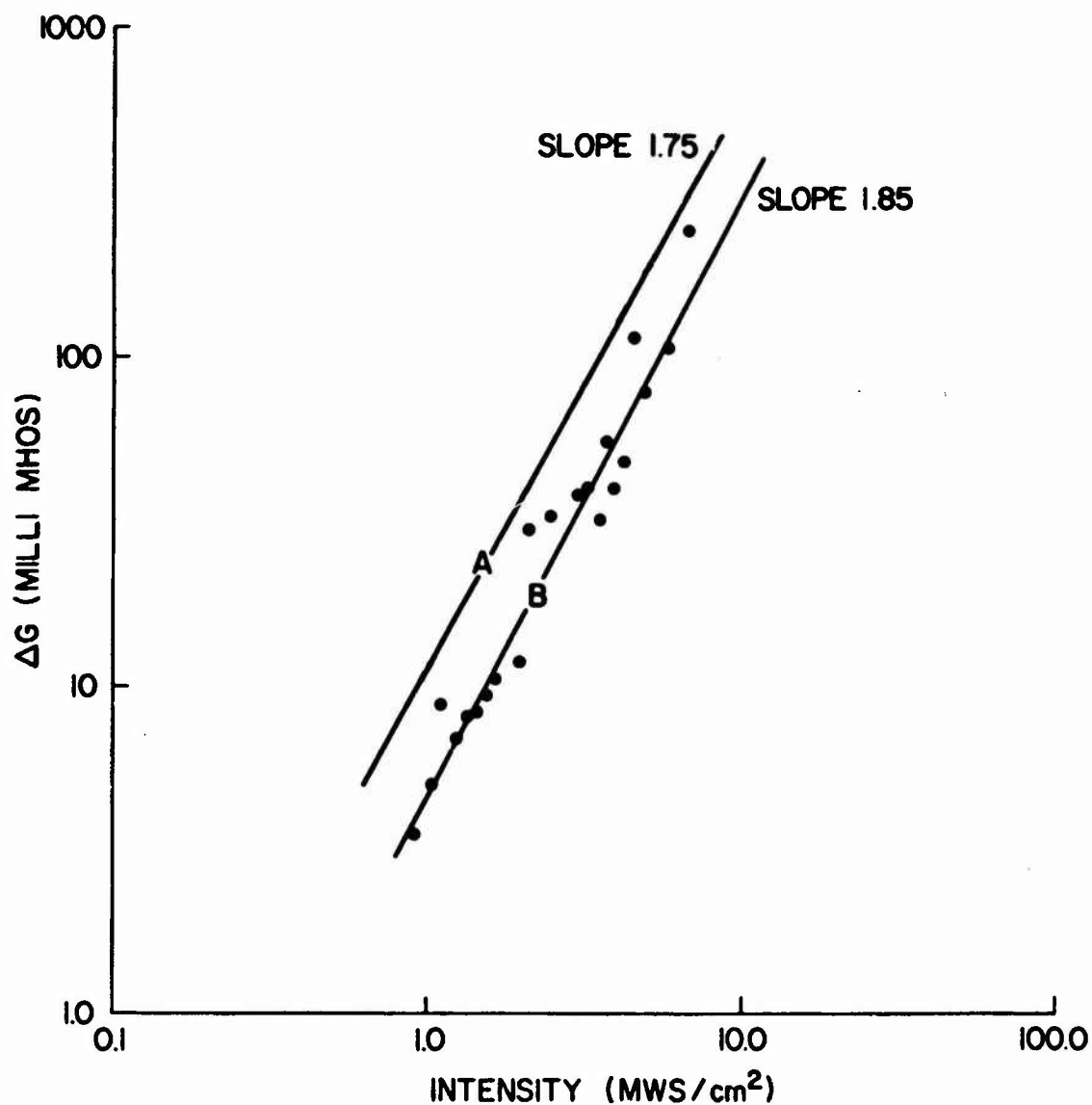
FIGURE 3: PHOTO CONDUCTIVITY VERSUS INTENSITY - Q-SWITCHED EXCITATION

with an exponential energy dependence of the level density. Such a model gives an intensity dependent life time. Nearly the same slope has been observed by L. M. Blinov et al⁽¹⁴⁾ and R. H. Bube⁽¹⁵⁾ in their photo conductivity (single photon) experiments with GaAs.

Above 1 MW/cm^2 , the slope of the curve is more than doubled indicating the two photon nature of the photo conductivity. The impurity (single photon) conductivity line below 1 MW/cm^2 is extrapolated and is subtracted from the curve above 1 MW/cm^2 . The resulting photo conductivity is due to true two photon effect and the log-log plot against intensity is shown in Fig. 4. A least square fit straight line to these points gave a slope of 1.85.

To effect a comparison of the experimentally observed two photon conductivity with Jick Yee's⁽¹¹⁾ theoretically calculated values, one requires the correct values of the mobility and the life time of carriers. The mobility of carriers is fairly well known for a given concentration. The life time τ for the calculation of steady state two photon conductivity is estimated as follows:

The high intensity of the laser pulse excitation results in reaching a steady state value of the photo conductivity. The photo conductivity decay curve gives an idea of the response time and the steady state life time of the carriers could be observed only if the laser excitation pulse width is smaller than the carrier life time. At lower intensities below 1 MW/cm^2 , the carrier life time depends on the laser intensity (continuously decreasing with increase of intensity according to A. Rose⁽¹³⁾).



Q-SWITCHED PULSE EXCITATION
 A - JICK YEE'S CURVE
 B - EXPERIMENTAL CURVE
 n-TYPE O₂ DOPED GaAs
 CRYSTAL THICKNESS = 0.028cm; MOBILITY = 7000cm²/v-sec
 LIFE TIME ≈ 10⁻¹⁰secs

FIGURE 4: TWO PHOTON CONDUCTIVITY

However at high intensities, where two photon effects are observable, the carrier density increases rapidly resulting in moving the Fermi level towards the conduction band very rapidly, and the crystal essentially behaves like a trap free material and so the life time remains constant at higher intensities of light. To calculate the two photon conductivity, one requires this constant life time and as an approximation the following estimation is done at 1 MW/cm^2 where two photon effects are just observable. This estimation gives a slightly higher value of the life time resulting in increase in the calculated value of the two photon conductivity.

At approximately 1 MW/cm^2 , where two photon conductivity is negligible, the measured value of the single photon conductivity is used to estimate the steady state life time of the carriers. Neglecting surface recombination, we get for single photon conductivity

$$\Delta G = \frac{c}{a} q (\mu_e + \mu_p) I_o \alpha L \tau_{St} \quad (8)$$

where α = single photon absorption coefficient

At $I_o = 1 \text{ MW/cm}^2$, $\Delta G = 0.02 \text{ MHOS}$

$$L = 0.028, \frac{c}{a} = \frac{1}{3}, \alpha L \approx 0.1$$

$$\mu_e + \mu_p \approx 7000 \text{ cm}^2/\text{v-sec}.$$

τ_{St} , the steady state life time of carriers is estimated to give approximately 10^{-10} secs which agrees favourably with Blinov's value⁽¹⁴⁾ ($\approx 5 \times 10^{-11}$ secs).

Using this value of τ , the two photon conductivity of GaAs is calculated using Jick yee's⁽¹¹⁾ expression

$$\Delta G = \frac{c}{a} q (\mu_e + \mu_p) \frac{\beta L I_0^2}{1 + \beta L I_0} \frac{\tau}{2\hbar\omega} \quad (9)$$

$$\beta = 5 \text{ cm/MW}, (\mu_e + \mu_p) = 7000 \text{ cm}^2/\text{v-sec}$$

$$\hbar\omega = 1.17 \text{ eV}, \frac{c}{a} = \frac{1}{3}$$

The theoretically calculated two photon conductivity is also shown in Fig. 4. The slopes of the experimental and theoretical curves are nearly equal thus indicating the two photon nature of the excitation. The magnitude of the observed photo conductivity agrees well within an order of magnitude with the theoretically computed values. The discrepancy may be attributed to the inaccuracy involved in the estimation of life time and also to the nonuniformity of beam distribution.

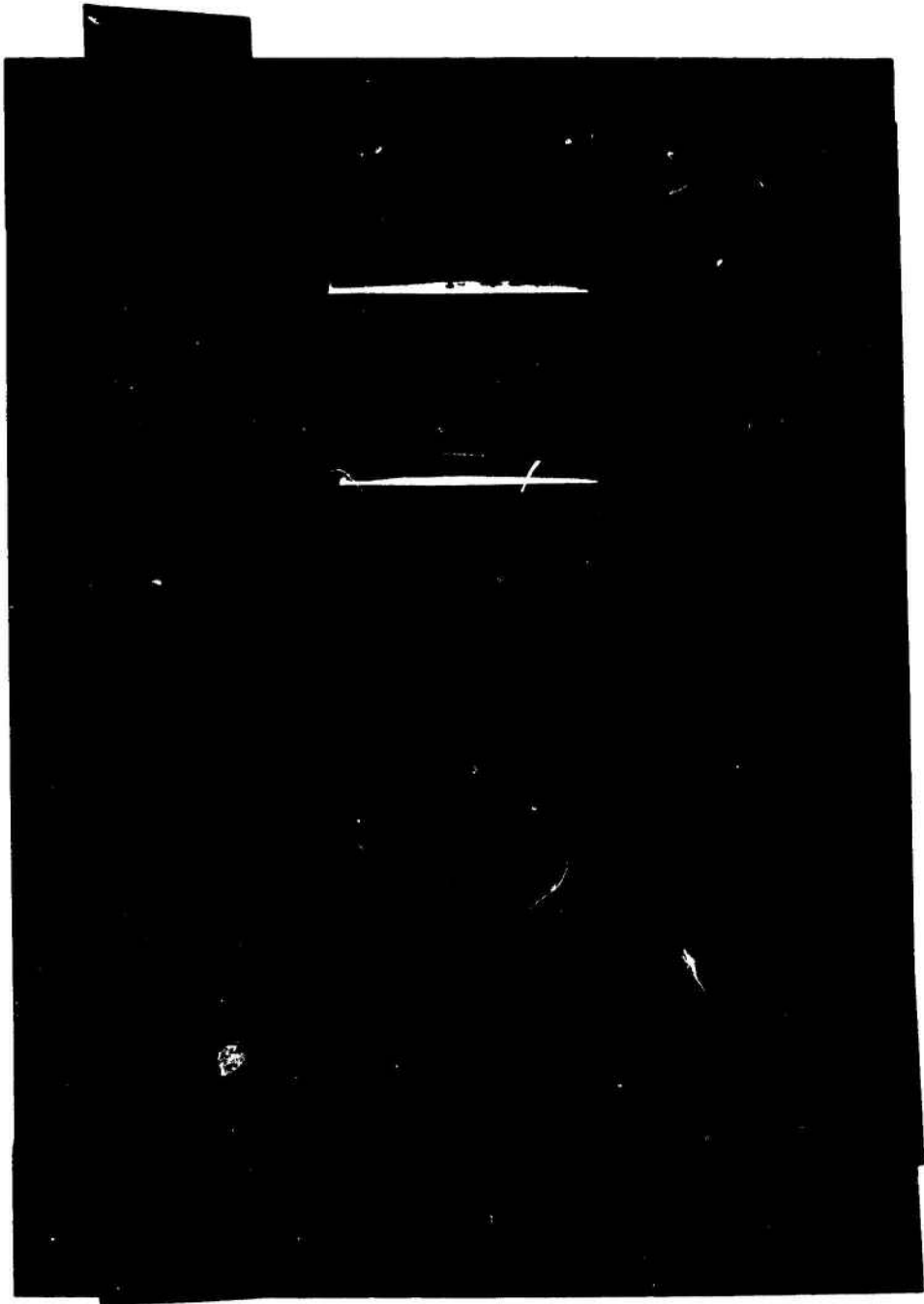
In an actual experiment, where we may use this two photon excitation of conductivity for picosecond pulse width measurement, the crystal has to be exposed to mode locked laser pulses with higher individual pulse intensity. Therefore the photoconductivity with a mode locked laser was investigated using two different types of GaAs crystal.

1. O_2 -doped n-type GaAs (same as in Q-switched investigation)
2. Cr-doped high resistivity GaAs.

Mode-locked excitation of photo conductivity:-

The mode locked pulse train from Nd: glass laser is shown in Fig. 5(A, B).

FIGURE 5: MODE-LOCKED PULSES AND STRUCTURE
OF PHOTO CONDUCTIVITY



A

B

C

D

E

The photo conductivity signal from Cr-doped GaAs was observed on a 519 oscilloscope with mode locked pulse train incident on the crystal. Fig. 5 (C, D, E) displays the p.c signals when the bias voltage is normal, reversed and shorted. The excitation of photo conductivity is due to each individual pulse in the pulse train as shown by the structure in photo conductivity response. The life time of the carriers produced is of the order of 10^{-10} secs and so the excitation pulse (width of the order of p sec) causes a transient build up of charge carriers by two photon absorption. The time separation between two subnanosecond pulses is of the order of 5 n secs ($2L/C$ where L is cavity length and C velocity of light) and since the life time is smaller than the pulse separation. The carriers decay in between the pulses. This explains the observation of the structure visible on the photo conductivity wave form. In the figure, however the photo conductivity does not fall to the zero value and this may possibly be due to the increased time constant in the measuring circuit. We concluded from this that the observed photo conductivity was in the form of spikes each spike corresponding to single subnanosecond pulse of the mode locked pulse train. It is evident that such a short pulse excitation produces non-equilibrium charge carriers dependent only on the intensity of the short pulse and on the time width of the pulse. So the photo conductivity must be independent of surface recombination, trapping, etc. Since the individual pulse intensity is of the order of GW/cm^2 , the two photon absorption and the effect of higher order processes, stimulated carrier absorption on the two photon

absorption could be studied at higher intensities of light.

The photo conductivity signal along with the laser pulse in the dual beam scope is shown in Fig. 6. In the Q-switched case, Fig. 6(A) the photo conductivity peak is near the end of the laser pulse while in Fig. 6 (B, C), the mode-locked pulse, the two pulses follow each other. This also shows that in the case of mode-locked excitation, the crystal behaves like a trap free material exhibiting no response time.

The photo conductivity of the two samples was investigated with mode-locked pulses and a log-log plot of this versus relative intensity is shown in Fig. 7. Both curves display a slope of 2 changing to unity slope at higher intensities. One could calculate the transient two photon conductivity theoretically as follows:

According to Basov⁽³⁾, the intensity absorbed in a crystal of thickness L due to two photon absorption is given by

$$I = \frac{\beta I_o^2 L}{1 + \beta I_o L} \quad (10)$$

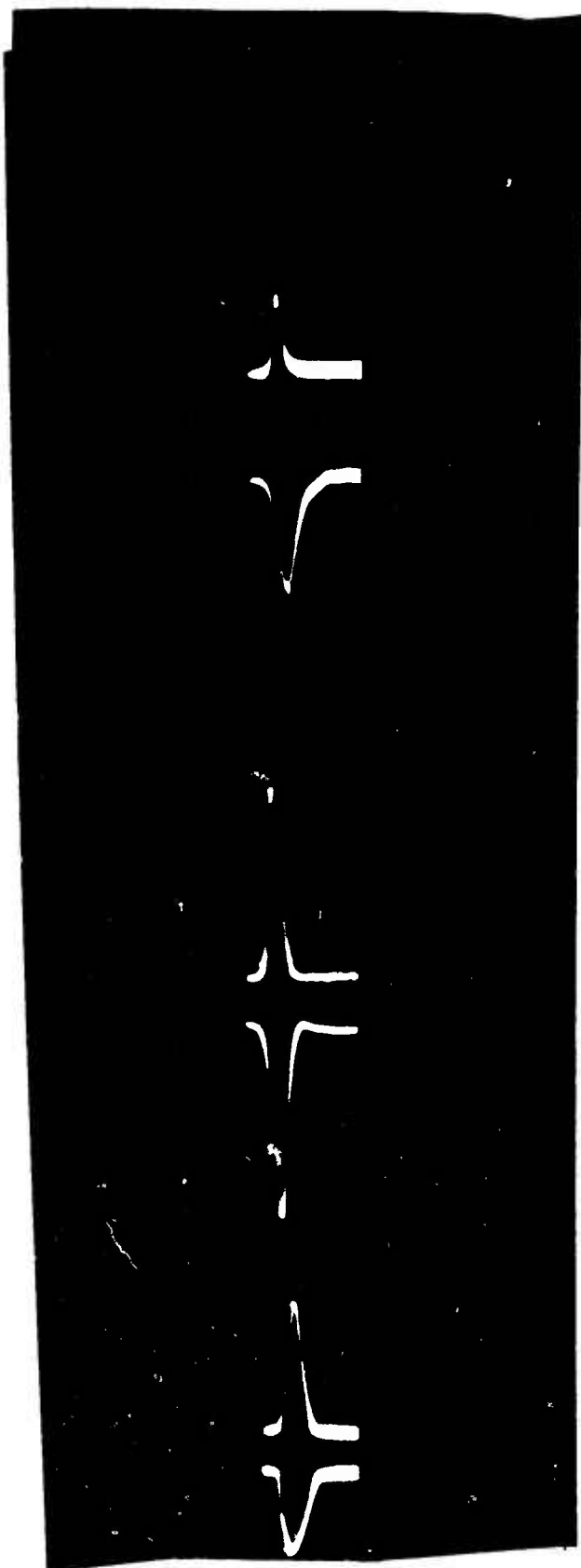
$$\text{Average no. of carriers produced/cm}^2 = \frac{\beta I_o^2 L}{1 + \beta I_o L} \times \frac{t_i}{2\hbar\omega} \quad (11)$$

Where t_i is the pulse width.

$$\Delta G = (C/a) q (\mu_e + \mu_p) \frac{\beta I_o^2 L}{1 + \beta I_o L} \times \frac{t_i}{2\hbar\omega} \quad (12)$$

This formula agrees with Jick Yee's steady state conductivity derived earlier when we replace τ by t_i . According to Ryukin⁽¹⁶⁾, if the pulse

FIGURE 6: PHOTO CONDUCTIVITY VERSUS LASER PULSE



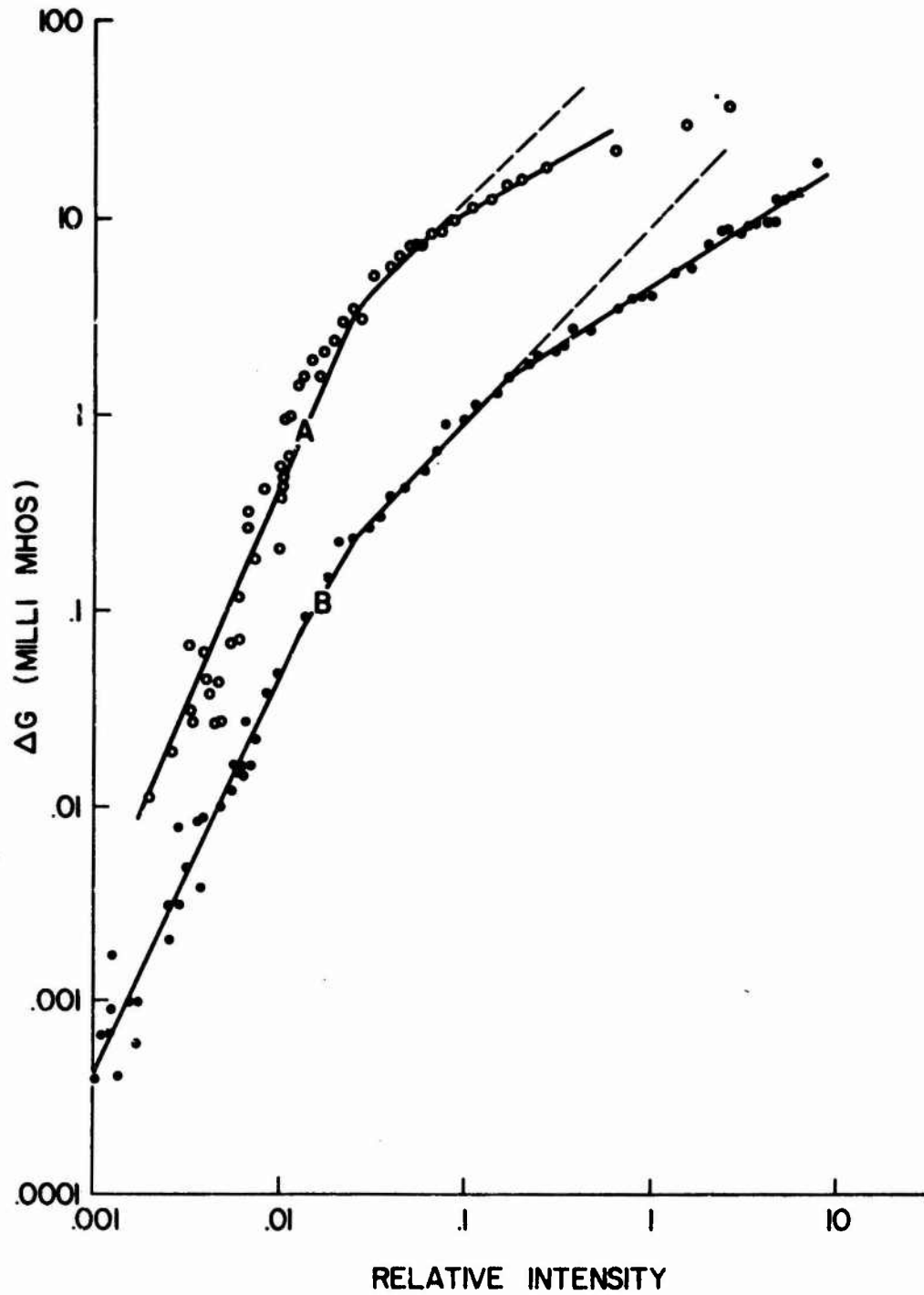
A

B

C

18A

6



MODE LOCKED PULSE EXCITATION
EXPERIMENTAL PHOTO CONDUCTIVITY

A n-TYPE O_2 DOPED GaAs (.028cm THICK)
B Cr-DOPED SEMI INSULATING GaAs
(.033cm THICK)

LIFE TIME $\tau \gg t_1$ (pulse width) ≈ 1 p sec

FIGURE 7: PHOTO CONDUCTIVITY OF THE TWO SAMPLES VERSUS RELATIVE INTENSITY

width is smaller than the life time, then

$$\Delta G = \Delta G_{St} (1 - e^{-\frac{t_i}{\tau}}) \approx \Delta G_{St} \frac{t_i}{\tau} \quad \text{if } \frac{t_i}{\tau} \ll 1.$$

This justifies the use of this formula for computing two photon conductivity.

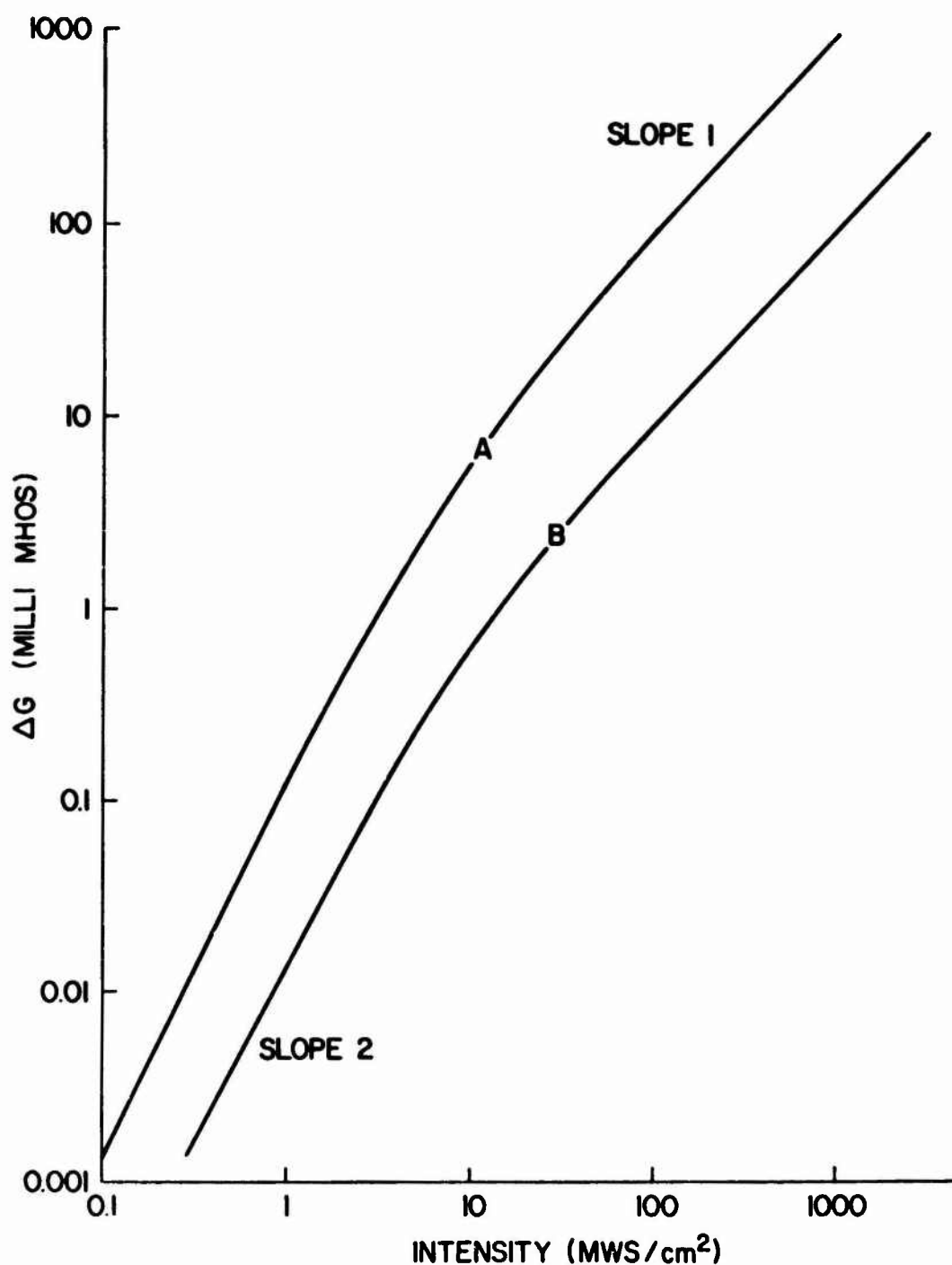
The theoretical photo conductivity versus intensity is displayed on Fig. 8 in a log-log graph. Here also the slope is 2 at lower intensities and changing to unity at higher intensities.

$$\Delta G \propto \frac{\beta L I_o^2}{1 + \beta L I_o} \quad (13)$$

At lower intensities, $\beta L I_o \ll 1$, $\Delta G \propto I_o^2$

At higher intensities, $\beta L I_o \gg 1$, $\Delta G \propto I_o$

Thus the experimental photo conductivity displays the two photon nature of the excitation. In the unity slope region, $\Delta G \propto \mu I_o$, the ratio of the conductivities in this region at a particular intensity gives the mobility ratio of the two semiconductors. The mobility ratio was found to be 30. The mobility for GaAs (O_2 -doped) is $7000 \text{ cm}^2/\text{v-sec}$. This gives the mobility for Cr-doped GaAs $\approx 230 \text{ cm}^2/\text{v-sec}$. This value of the mobility was used for calculation. In the region of slope 2, the conductivity ratio is proportional to the thickness ratio, β ratio and the mobility ratio. Using the mobility ratio determined earlier, and the thickness ratio (.028/.033), the ratio of two photon absorption coefficients in two different GaAs samples was found to be approximately unity.



CALCULATED TWO PHOTON CONDUCTIVITY

A n-TYPE O₂ DOPED GaAs
(.028 cm THICK, $\mu_e + \mu_r = 7000 \text{ cm}^2/\text{v-sec}$)

B Cr-DOPED SEMI INSULATING GaAs
(.033 cm THICK, $\mu_e + \mu_r \approx 235 \text{ cm}^2/\text{v-sec}$)

LIFE TIME $\tau \gg t_i$ (pulse width) $\approx 1 \text{ p sec}$

FIGURE 8: COMPUTED TWO PHOTON CONDUCTIVITY

This gives a check on the measured β values for the two samples (≈ 5 cm/MW). Such a low mobility in Cr-doped GaAs can be explained by the compensated impurities in the high resistivity crystal as observed by Cronin and Haisty⁽¹⁷⁾. Since the intensity was not measured absolutely, the magnitudes of the photo conductivity could not be compared exactly.

Cr-doped GaAs crystal had a high dark resistance and so we could measure the photo conductivity at very high intensities quite accurately. At high intensities of light, the photo conductivity curve showed a sub-linear dependence as shown in Fig. 9. If the conductivity is due to two photon excitation alone, we should observe a slope of 1 at very high intensities as shown by the extrapolated linear dependence. The observed conductivity is less than the ideal extrapolated curve of slope unity.

The non linearity is definitely not due to the surface recombination since the excitation is due to short pulse whose width is much less than the recombination times. Since conductivity is directly proportional to mobility, it was first suspected that mobility may decrease at high excitation levels because of electron-hole scattering. This decrease in mobility at high excitation levels in Si (4.2°K) was observed by A. A. Patsin⁽¹⁸⁾ et al. The following discussion rules out this possibility in the present experiment.

At room temperature, the mobility in GaAs is determined by screened polar scattering and impurity scattering⁽¹⁹⁾. The room tem-

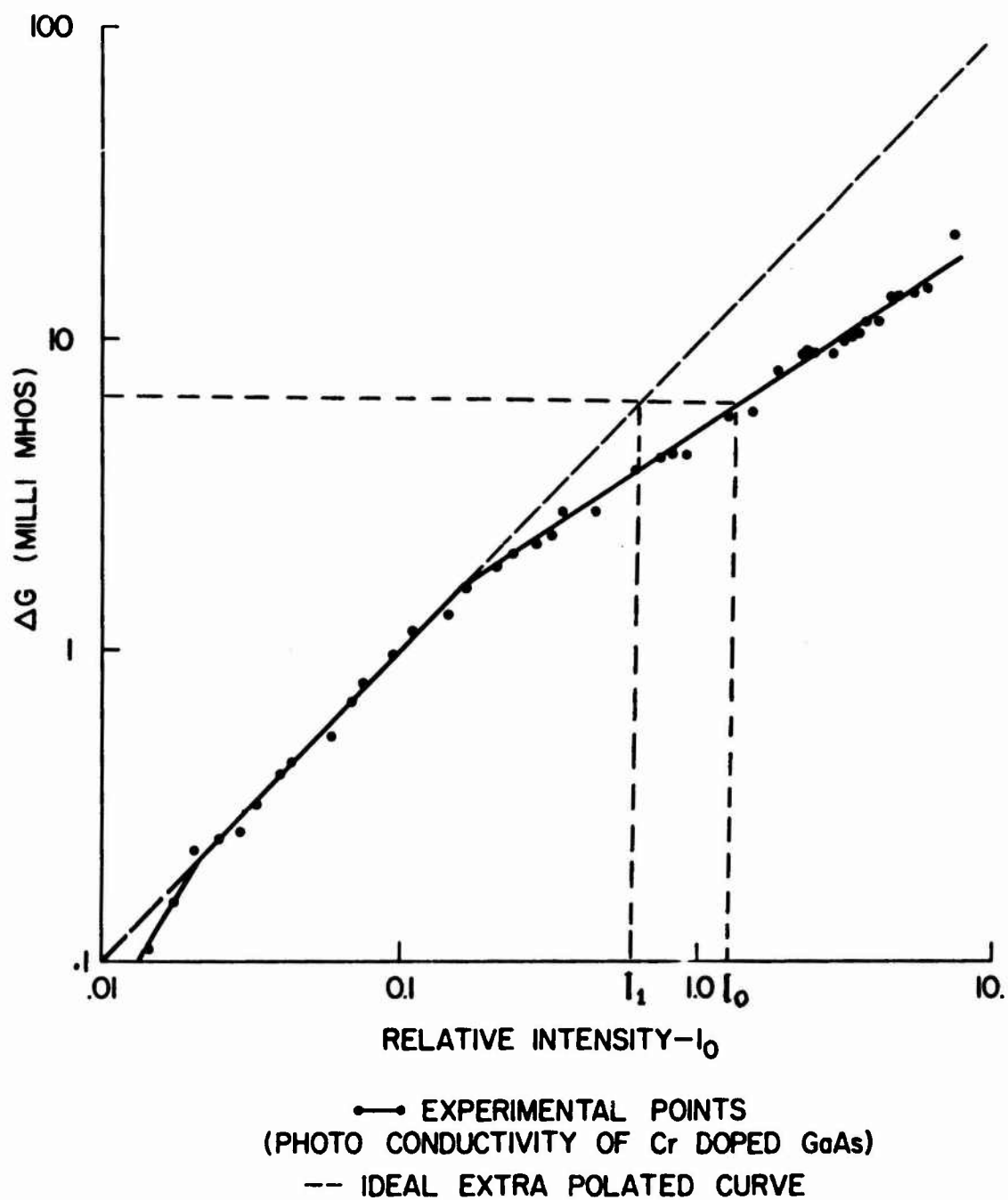


FIGURE 9: NON-LINEARITY IN THE PHOTO CONDUCTIVITY RESPONSE

perature mobility $\approx 230 \text{ cm}^2/\text{v-sec}$ was estimated in the earlier section.

When the crystal is excited by high intensity laser pulses, the density of non-equilibrium charge carriers increases. The contribution of mobility due to electron-hole scattering was calculated using the Brooks-Herring Formula⁽²⁰⁾

$$\mu_{e-h} = \frac{2^{7/2} \epsilon^2 (kT)^{3/2} (m_e + m_h)^{1/2}}{\pi^{3/2} e^3 (m_e m_h)^{1/2} (n_e n_h)^{1/2} \left[\ln(1+B) - \frac{B}{1+B} \right]}$$

where $B = \frac{6 \epsilon (kT)^2 m_e m_h}{\pi \hbar^2 e^2 (m_e + m_h) (n_e n_h)^{1/2}}$ (14)

ϵ = dielectric constant

m_e = electron effective mass

m_h = hole effective mass

e = electronic charge

k = Boltzmann's constant

T = Absolute temperature

$\hbar$ = Planck's constant/ 2π

$$\epsilon = 11.8, \quad m_e = 0.072 m_0$$

$$m_h \approx m_{01}^* = 0.68 m_0$$

At $T = 300^\circ \text{K}$, the computed value of μ_{e-h} at high carrier densities of the order of $10^{18}/\text{cc}$ gave a value of $10^4 \text{ cm}^2/\text{v-sec}$. Mobilities combine approximately as $\frac{1}{\mu_{\text{eff}}} = \frac{1}{\mu_{\text{normal}}} + \frac{1}{\mu_{e-h}}$. The normal mobility

is low ($\approx 230 \text{ cm}^2/\text{v-sec}$) and so will not be affected by the high electron-hole scattering mobility. Therefore the observed non-linearity is not due to decrease of mobility due to electron-hole scattering.

The second possibility may be due to the stimulated free carrier absorption (free electron absorption). For III-V Semiconductors, the absorption by free holes can be much larger than the electron absorption cross section⁽²²⁾. This is due to the presence of transitions between sub-bands v_1 and v_2 of the valence band. The free electron absorption in GaAs⁽²¹⁾ for a concentration of $10^{17}/\text{cm}^3$ is approximately $.03 \text{ cm}^{-1}$ at 1μ wavelength. So the stimulated free carrier absorption is low and the observed non-linearity is not due to this process.

The more likely possibility is that due to stimulated intraband absorption. The intra-valence band absorption in GaAs with various p-type doping densities had been extensively studied by Braunstein⁽²⁸⁾ and Braunstein and Kane⁽²⁴⁾. The hole absorption coefficient at 1μ for the crystal doped with $10^{17}/\text{cm}^3$ is about 3cm^{-1} . This absorption coefficient is due to the transitions between the light hole, heavy hole and split off valence bands. The band structure is shown in Fig. 1.

In the present case, at higher intensities of light, the non equilibrium charge carriers are produced proportional to the light intensity and the associated stimulated absorption by the non equilibrium holes (intra-valence band transitions) should also increase linearly with the light intensity. Referring to Fig. 9, from the nonlinear region, the absorption coefficient due to intra valence band transitions was calculated as follows:

For a particular value of ΔG , the intensities I_1 and I_0 at the ideal slope 1 curve and the experimental photo conductivity curve are determined.

$$I_1 = I_0 e^{-\alpha L}$$

$$\alpha = \frac{1}{L} \ln \left(\frac{I_0}{I_1} \right)$$

L = thickness of crystal

α = absorption coefficient due to intraband absorption.

The carrier density is proportional to I_1 and so α is plotted against I_1 in Figure 10 in a log-log graph. Again at lower intensities, α increases linearly with light intensity (carrier density) thereby showing that the non-linearity in the two photon conductivity is due to stimulated intra valence band absorption.

However at very high light intensities, α increases rather slowly. This means that part of the light intensity is being utilized in the generation of non-equilibrium carriers (contributing to conductivity). The difference in absorption coefficient between the linear dependence and the sublinear one in the α versus I , graph (Fig. 10) is plotted against relative intensity I_0 in a log-log graph Fig. 11. The slope of ≈ 2 in Fig. 11 suggest that there is a generation mechanism whose absorption coefficient increases as the square of the light intensity. For three photon absorption, the absorption coefficient must be proportional to the square of the intensity of radiation. Hence we conclude that we have observed the three photon generation of non-equilibrium charge

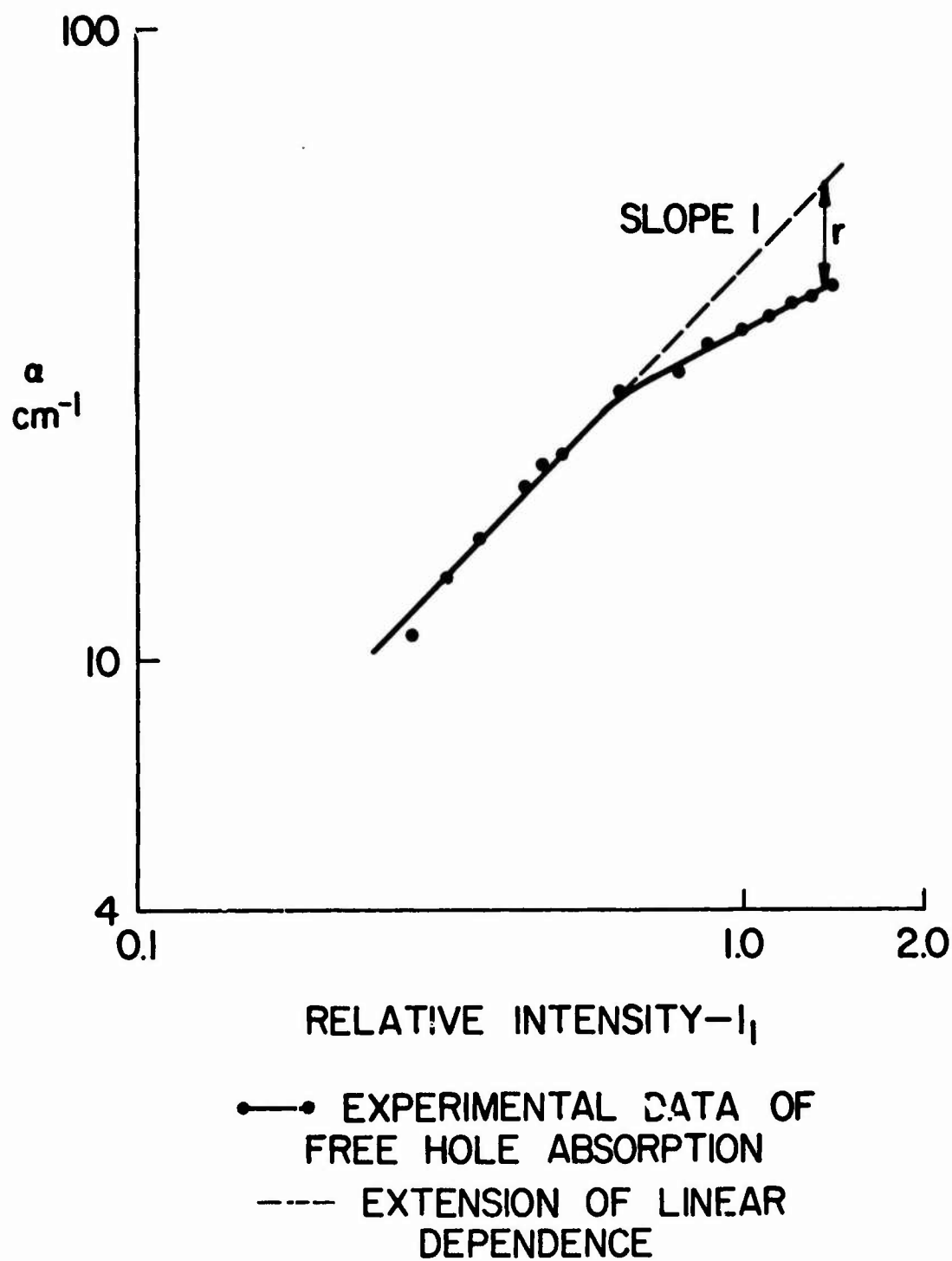


FIGURE 10: HOLE ABSORPTION

carriers in GaAs at very high intensities of light.

Referring to Fig. 11, the maximum intensity was approximately determined to be 10 GW/cm^2 and the corresponding observed 3 photon absorption coefficient was 20 cm^{-1} . Using the formula derived by A. I. Bohryskava⁽²⁵⁾ et al for three photon band to band transitions in semiconductors and applying it to GaAs (transitions from $V_{1,2}$ to C) we get for 10 GW/cm^2 of light intensity, a three photon absorption coefficient as 14 cm^{-1} which agrees approximately with the experimentally determined values.

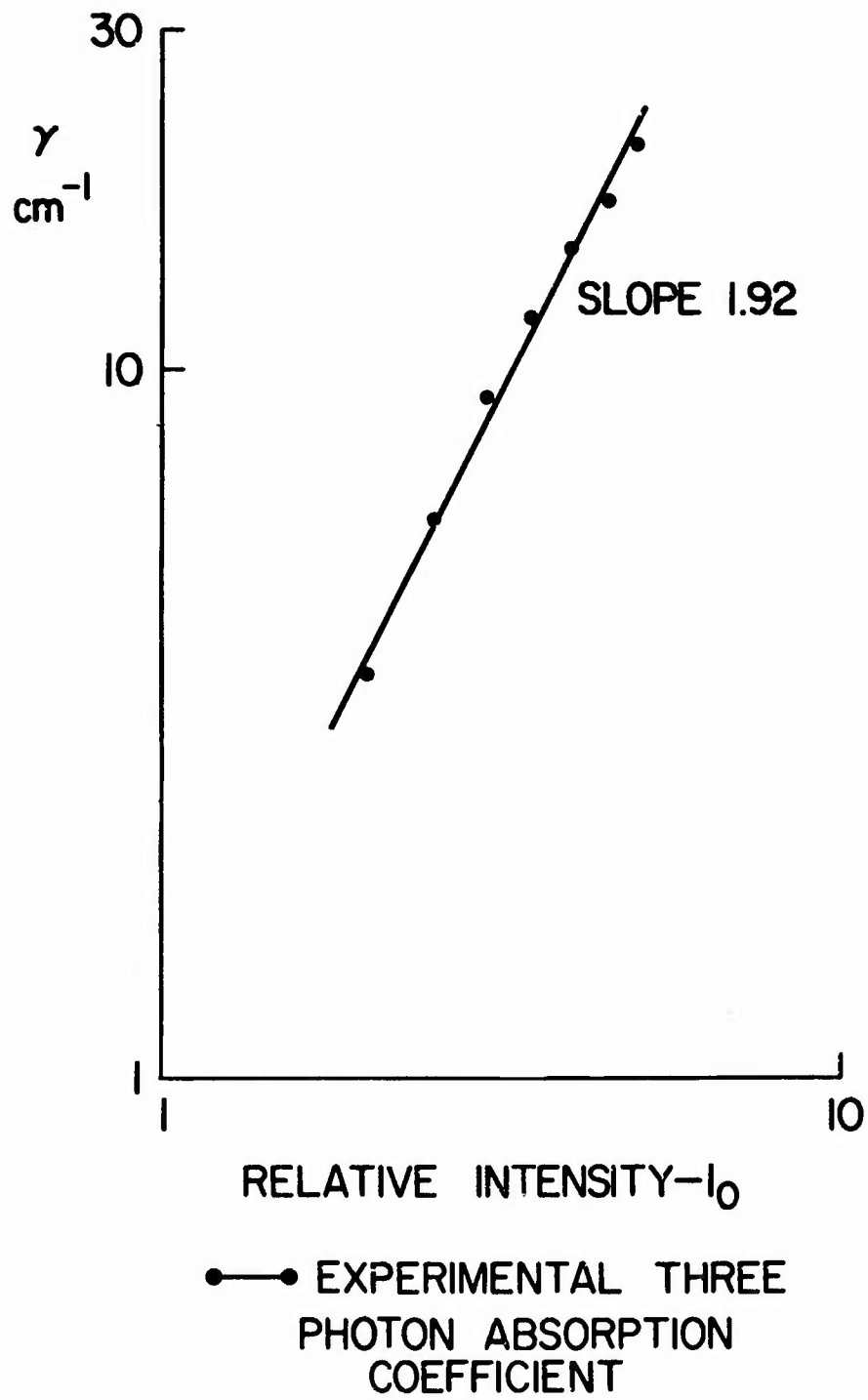


FIGURE 11: ESTIMATED THREE PHOTON ABSORPTION

Conclusion:-

The two photon conductivity in GaAs was investigated with Q-switched and mode-locked Nd: glass laser pulses. In the Q-switched pulse excitation, photo conductivity of O_2 -doped GaAs was measured in the intensity range 0.01 to 10 MW/cm^2 . In the low intensity region below 1 MW/cm^2 , single photon process was dominant and the slope of the $\log \Delta G - \log I$ line was ≈ 0.6 which could be understood by a continuous distribution of levels in the forbidden gap as proposed by A. Rose. Above 1 MW/cm^2 , the two photon process dominated and the observed two photon conductivity was in agreement with Jick yee's theoretical calculations (slopes ≈ 1.8). Mode locked excitation of the photo conductivity displayed the two photon nature over a wide range of intensities because of the short pulse excitation (picosecond pulses) with a higher intensity. The photo conductivity curve in the case of both O_2 doped GaAs and Cr-doped semi-insulating GaAs displayed a slope of 2 at lower intensities changing to unity slope at higher intensities. This slope change was expected since $\Delta G \propto \frac{\beta I_o^2 L}{1 + \beta I_o L}$, at lower intensities, $\Delta G \propto \beta I_o^2 L$ since $\beta I_o L \ll 1$ and at higher intensities, $\Delta G \propto I_o$ since $\beta I_o L \gg 1$. At very high intensities, the photo conductivity curve displayed a non-linear region which was proved to be due to stimulated intraband absorption of holes in GaAs. The magnitude of the absorption coefficient varied from 10 to 30 cm^{-1} linearly with intensity. This agreed approximately with the values reported in the references^(23,24). The effect of stimulated

intraband (valence) absorption on two photon conductivity in I_{Sb} at 90°K using a Q-switched CO_2 laser was studied by A. M. Danishevskiy et. al. (27).

Towards the end of the high intensity region, absorption which led to the generation of carriers was observed. This absorption coefficient was found to be proportional to the square of the light intensity indicating the generation of non-equilibrium charge carriers in GaAs due to three photon absorption. The calculated three photon absorption coefficient agrees approximately with the experimentally observed value.

Future experiments with thinner crystals (are suggested to extend the quadratic dependence of photo conductivity to higher intensity regions.

Finally, we conclude that two photon conductivity in semiconductors need not necessarily depend on I^2 as discussed by Jick H. Yee (11) and by Paul Kelly et. al. (28). Because of absorption of laser beam in the sample and surface recombination, this dependence is modified. Picosecond pulse excitation in the present experiment excluded surface recombination and because of absorption of laser beam in the sample, the two photon conductivity is found to be proportional to first power of Intensity. Near to damage threshold, stimulated intraband absorption and higher order processes modify the exponent in the present experiment. So care must be exercised in detecting two photon absorption in semiconductors.

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Figure Captions:-

- 1) Band structure of GaAs
- 2) Experimental set up
- 3) Photo conductivity versus intensity - Q-switched excitation
- 4) Two photon conductivity
- 5) Mode-locked pulses and structure of photo conductivity
- 6) Photo conductivity versus laser pulse
- 7) Photo conductivity of the two samples versus relative intensity
- 8) Computed two photon conductivity
- 9) Non-linearity in the photo conductivity response
- 10) Hole absorption
- 11) Estimated three photon absorption