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

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OBSERVATION OF TWO PHOTON CONDUCTIVITY IN GaAs WITH NANOSECOND AND PICOSECOND LIGHT PULSES*

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ABSTRACT

The effect of photoconductivity in GaAs single crystals was investigated by using both nanosecond and picosecond light pulses for excitation. Both steady state and transient responses of photoconductivity have been observed.

OBSERVATION OF TWO PHOTON CONDUCTIVITY IN GaAs WITH NANOSECOND AND PICOSECOND LIGHT PULSES

The advent of high power lasers has made it experimentally feasible to observe a number of intensity dependent optical interactions in matters which involve simultaneous absorption of two or more photons. The multi-photon absorption in condensed media can, in principle, be investigated either by photoconductivity effect or by observing fluorescence. In this letter we report the study of two-photon induced conductivity in single crystals of GaAs by using both a nanosecond pulse from a Q-switched Nd:glass laser or a train of picosecond pulses from a mode-locked Nd:glass laser as the excitation light sources. With proper control of the laser output in these two modes of operation one can obtain, in the light pulse, power density ranging from a few kilowatts to tens of gigawatts per square centimeter and pulse duration from a few tens of nanoseconds down to a few picoseconds. These unique features of the laser beam not only allow us to investigate the two-photon conductivity effect over a wide range of power level but also enable us to probe the effect both in steady state and transient responses since the measured lifetime of the induced charge carrier is of the order of one tenth of a nanosecond. GaAs is chosen because it is a direct band gap material whose forbidden energy gap is about 1.4 ev. The Nd:glass laser output has photon energy of 1.17 ev , thus the change of photoconductivity is expected to have significant contribution from two-photon absorption.

Under steady state excitation, Basov et al[1], and Yee[2] have derived an expression for the conductivity change ΔG due to two-photon absorption as

$$\Delta G = (c/a)\,|e|\,(\mu_e + \mu_h) A_o \tau \left[I_o^2 L/(1+KI_oL) \right] \qquad (1)$$

where c/a is a geometric factor of the sample; $|e|$, the magnitude of the electronic charge ; μ_e and μ_h are the electron and hole mobilities respectively; A_o, a constant related to the band parameters; τ, the lifetime of the charge carrier; I_o, the incident laser intensity; L, the thickness of the sample and $K = 2\hbar\omega A_o$ is the two-photon absorption coefficient in cm/MW. In this expression we have neglected the surface recombination effect because the diffusion length is much shorter than the sample thickness and charge carriers are uniformly generated through out the entire volume due to two-photon absorption. If the excitation pulse width is less than the lifetime of the charge carrier, then the effect is transient. The change of conductivity will be reduced. It is appropriate in the transient regime, to replace the lifetime τ in eq. (1) by Δt_p, the duration of the exciting light pulse.

The laser used was a Korad K-1 system with a Nd:glass rod having a Brewster-Brewster configuration. The laser was simultaneously Q-switched and mode-locked by a Kodak 9860 dye solution. The output of the laser consisted of a train of equally spaced picosecond pulses separated by 6 nanoseconds which was equal to the cavity round trip transit time. Such output pulses were used to investigate the transient response of the sample. For steady-state experiments where only the nanosecond "long" pulse was required, a mode-selector was inserted inside the laser cavity to prevent mode-locking. The beam was directed onto a GaAs sample which was connected in series with a 50 ohm load resistor to a battery. The change of conductivity in the GaAs

crystal produced a change of voltage across the load resistor and was monitored by a dual beam oscilloscope. The laser pulse was monitored by a photodiode and displayed on the same oscilloscope. The trigger signal was provided by a beam splitter and another photodiode. The beam intensity could be varied by inserting various neutral density filters in the optical path.

We have measured the photoconductivity in both O_2-doped n-type and Cr-doped semi-insulating GaAs crystals. The former had a thickness of 0.028 centimeters with a concentration of $3 \times 10^{14}/cm^3$ and a resistivity of 2.4 Ω-cm. The latter had a thickness of 0.033 centimeters with a resistivity greater than 10^8 Ω-cm. In both cases, a small rectangular piece was cut and ohmic contacts were alloyed at the two ends of the sample surface. The oscilloscope traces of the photoresponse are shown in Fig. 1. The conductivity change ΔG can be computed from the change in voltage across the load resistor. In Fig. 1a, we can see that the signal due to photoresponse is in time correlation with the laser pulse. It was also observed that whenever the polarity of the bias was reversed so did the polarity of the signal. Furthermore when no bias was applied the signal disappeared. These features showed that the observed signal was indeed due to photoconductive and not photovoltaic effect When the mode-locked pulse train was utilized as the exciting light source, in order to resolve the individual pulses in the train, it was necessary to use Tektronix 519 oscilloscopes for the display of the optical pulses (Fig. 1b) and the corresponding photoconductivity signal (Fig. 1c). The photoconductivity signal clearly shows a similiar periodic structure of the exciting pulse train.

This indicates that the conductivity change responded rapidly to the excitation of an individual pulse in the train. In other words, the lifetime of the induced charge carriers is much less than the separation between two adjacent pulses. The theoretical estimate for the lower limit of the lifetime for the sample is of the order of one tenth of a nanosecond. Since the width of an individual pulse was a few picoseconds, the effect shown in Fig. 1c should be of transient nature. In Fig. 1c one notices that the photconductivity does not fall to zero between two adjacent pulses in the train. This was mainly due to the fact that time response of the measuring circuit was not fast enough.

The measured photoconductivity ΔG against laser intensity with long pulse excitation is shown in Fig. 2 in a log-log graph. The maximum laser intensity was approximately 10MW/cm^2 with a pulse duration of 60 nanoseconds. The photoconductivity was measured over three decades of laser intensity. Fig. 2 displays a break in slope between 1 and 2 MW/cm^2. This agrees with the one and two-photon absorption coefficients which we measured in this sample. The small signal one-photon absorption coefficient for the crystal used in the experiment at 1.06 μ was approximately 3-4 cm^{-1} and the measured two-photon absorption coefficient was $\approx$ 5.6 cm/MW which agrees favorably with Basov's experimental and theoretical values.[3] Below 1MW/cm^2, the one-photon process predominates and above it the two-photon absorption takes over. In Fig. 2 one also notices that, below 1MW/cm^2, the slope of the curve is in fact less than unity and equal to 0.6. This can be interpreted as an indication of one-photon absorption via impurity levels. According to A. Rose[4], the slope of the photoconductivity curve between 1/2 and 1 can possibly be

explained by a continuous spectrum of levels in the forbidden band with an exponential energy dependence of the level density. Nearly the same slope has been observed by L. M. Blinov et al.[5] and R. H. Bube.[6]

Above 1MW/cm^2, the slope of the curve is more than doubled indicating the two-photon nature of the effect. If the single photon conductivity line is extrapolated and subtracted from the curve above 1MW/cm^2, the resulting curve will represent the true two-photon effect. The result is shown in Fig. 3. Line B in Fig. 3 represents a least square fit and has a slope of 1.85. To effect a comparison of the experimental data with the theoretical prediction one requires the values of the mobility and the lifetime of carriers. The mobility of the sample used is 7000 cm^2/v-sec. The life time τ is estimated from the measured value of the single photon conductivity ΔG_s,

$$\Delta G_s = \frac{c}{a} |e| (\mu_e + \mu_h) I_0 \alpha L \tau \qquad (2)$$

where α is the single photon absorption coefficient and was determined to be $\sim 3\text{cm}^{-1}$ from small signal measurement using a Cary Spectrometer. At $I_0 = 1\text{MW/cm}^2$, two photon contribution is negligible, $\Delta G = 0.02$ MHOS, $L = 0.028$ cm, $\frac{c}{a} = 1/3$. τ is found to be one tenth of a nanosecond. This value is consistent with the observed temporal structure of the photoresponse with mode-locked pulse train excitation as shown in Fig. 1c. Line A in Fig. 3 is the theoretical counterpart of line B. The slopes of both lines are nearly equal thus indicating the two-photon nature of the excitation. Although the magnitude of the observed photoconductivity agrees within an order of magnitude with the theoretical one, nevertheless, the latter is slightly

greater than the former. This discrepancy may be attributed to an over estimate of the lifetime and/or the non-uniformity of the beam distribution.

The ΔG versus laser intensity with picosecond pulse train excitation is shown in Fig. 4. It differs from the result of nanosecond pulse excitation in two aspects. First, the curves start with slope near 2 and falls off to a value less than unity. Second, the magnitude of ΔG in the slope 2 region is smaller by a factor of one hundred to one thousand. To explain the first feature we simply point out that the picosecond pulses have higher peak power than the previously utilized nanosecond pulse. The exact power could not be determined due to the uncertainty in their pulse width measurement. However, the lowest intensity in Fig. 4 should be between one to ten megawatts per square centimeter. The overall reduction in magnitude of ΔG comes from the fact that in eq. (1) the pulse duration Δt_p instead of the lifetime τ should be used because of transient response. The saturation of ΔG at higher laser intensity is partially due to sample thickness and intensity dependent denominator in eq. (1) and partially due to other form of nonlinear processes that are not quite understood at present. However, preliminary calculations indicate that the nonlinearity is most likely due to stimulated intraband hole absorption between valence bands.[7]

In conclusion, we have reported the observation of the two-photon induced conductivity in GaAs with either nanosecond or picosecond pulse excitation. The experimental data presented here indicated that it is now possible to study the fast relaxation process and transport phenomena in semiconductors with picosecond pulse excitation. By switching out a single

picosecond pulse from the train as the exciting source, one should be able to obtain more quantitative data.

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FIGURE CAPTIONS

Fig. 1 (a) Oscilloscope trace of the Q-switched laser pulse (upper trace) and the corresponding photoconductivity signal (lower trace).

(b) Oscillogram of the mode-locked pulse train

(c) The photoconductivity signal excited by the light pulse train shown in part (b).

Fig. 2 The measured photoconductivity ΔG versus laser intensity.

Fig. 3 Two-photon conductivity ΔG versus laser intensity. The data points in this figure were obtained by substracting the extrapolated one-photon contribution to the conductivity change for laser intensity above 1 MW/cm^2 as shown in Fig. 2. See text for detailed explanation.

Fig. 4 Photoconductivity of two samples versus relative laser intensity with mode-locked pulse excitation.

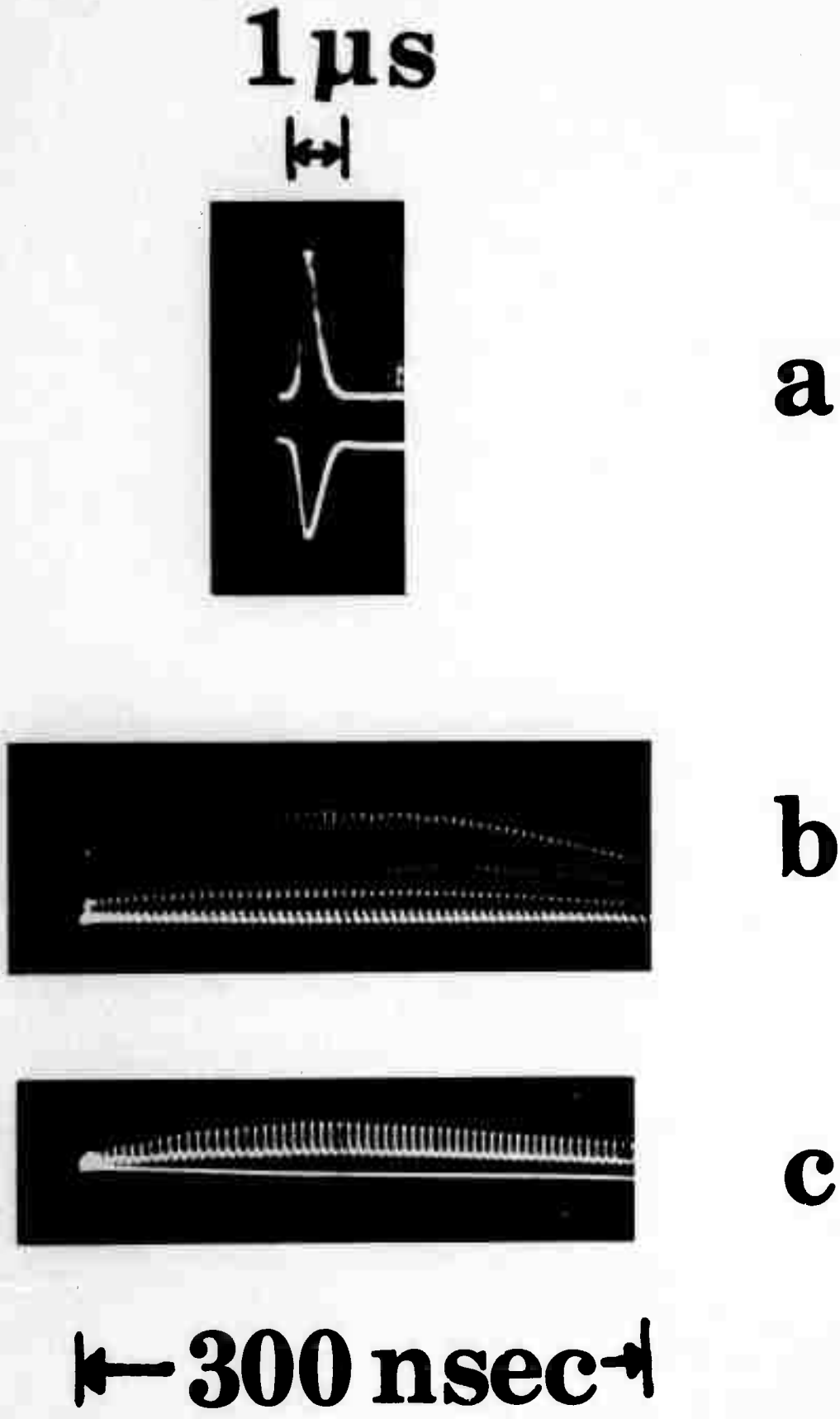
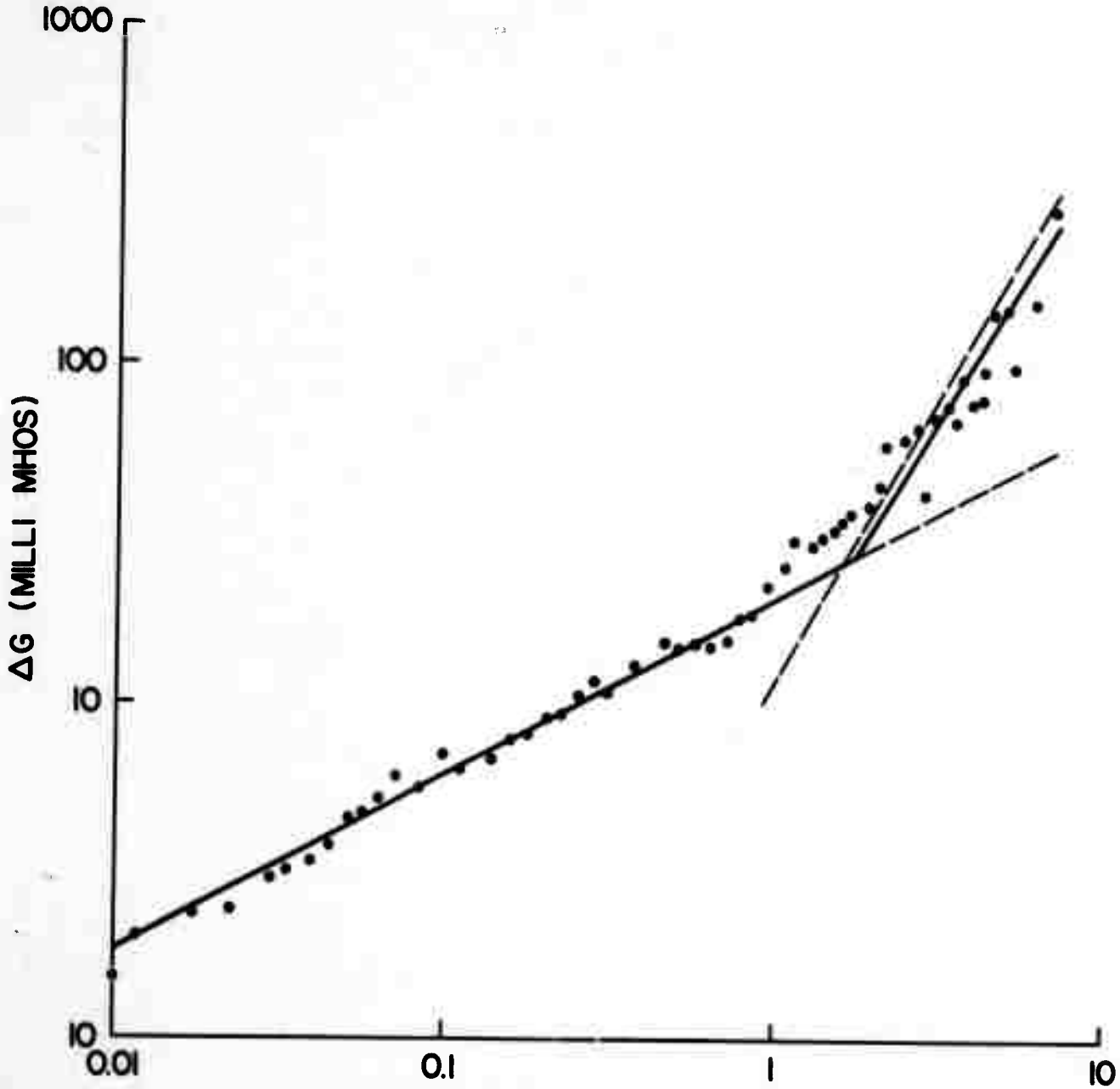
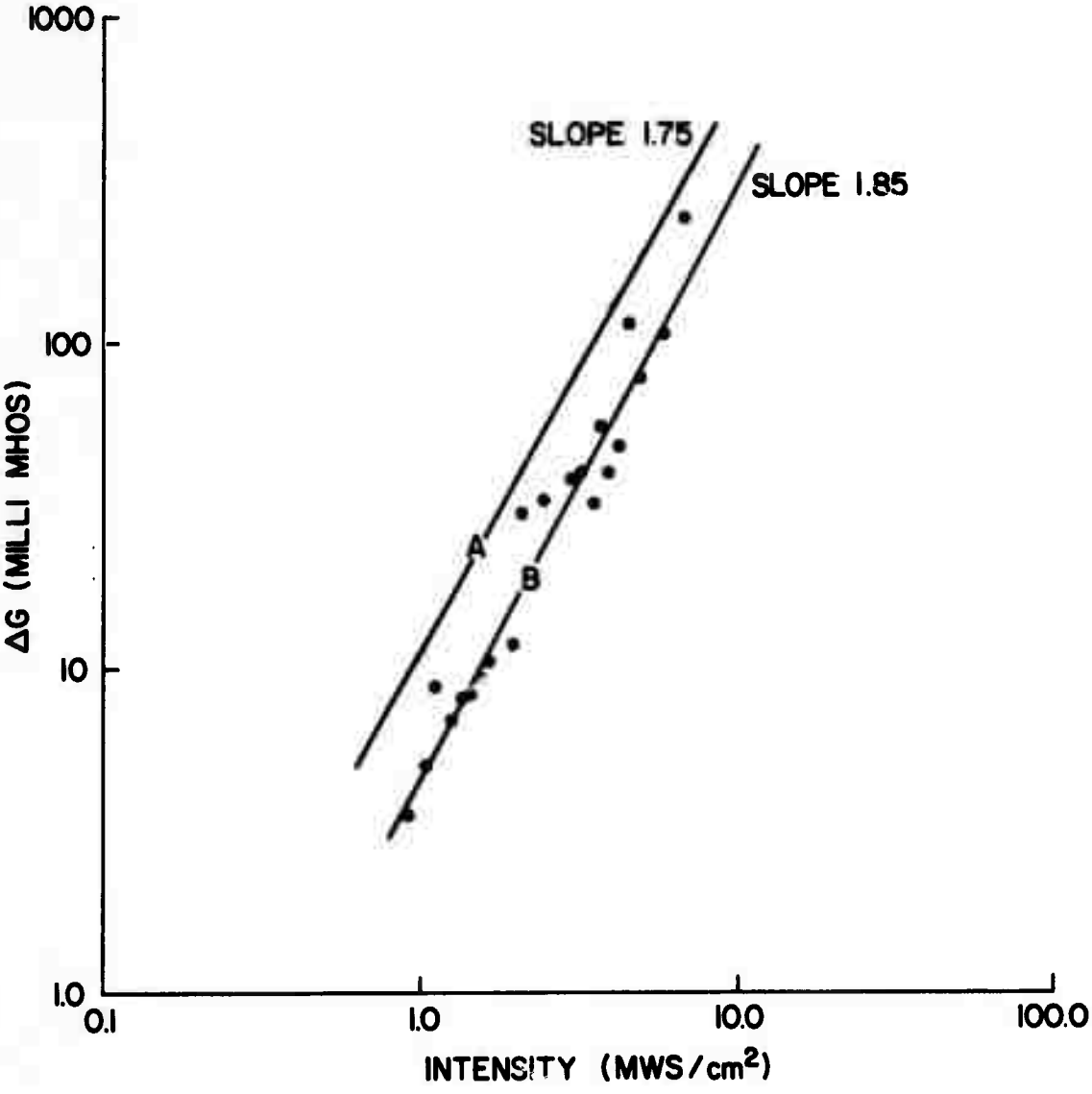


Fig 1

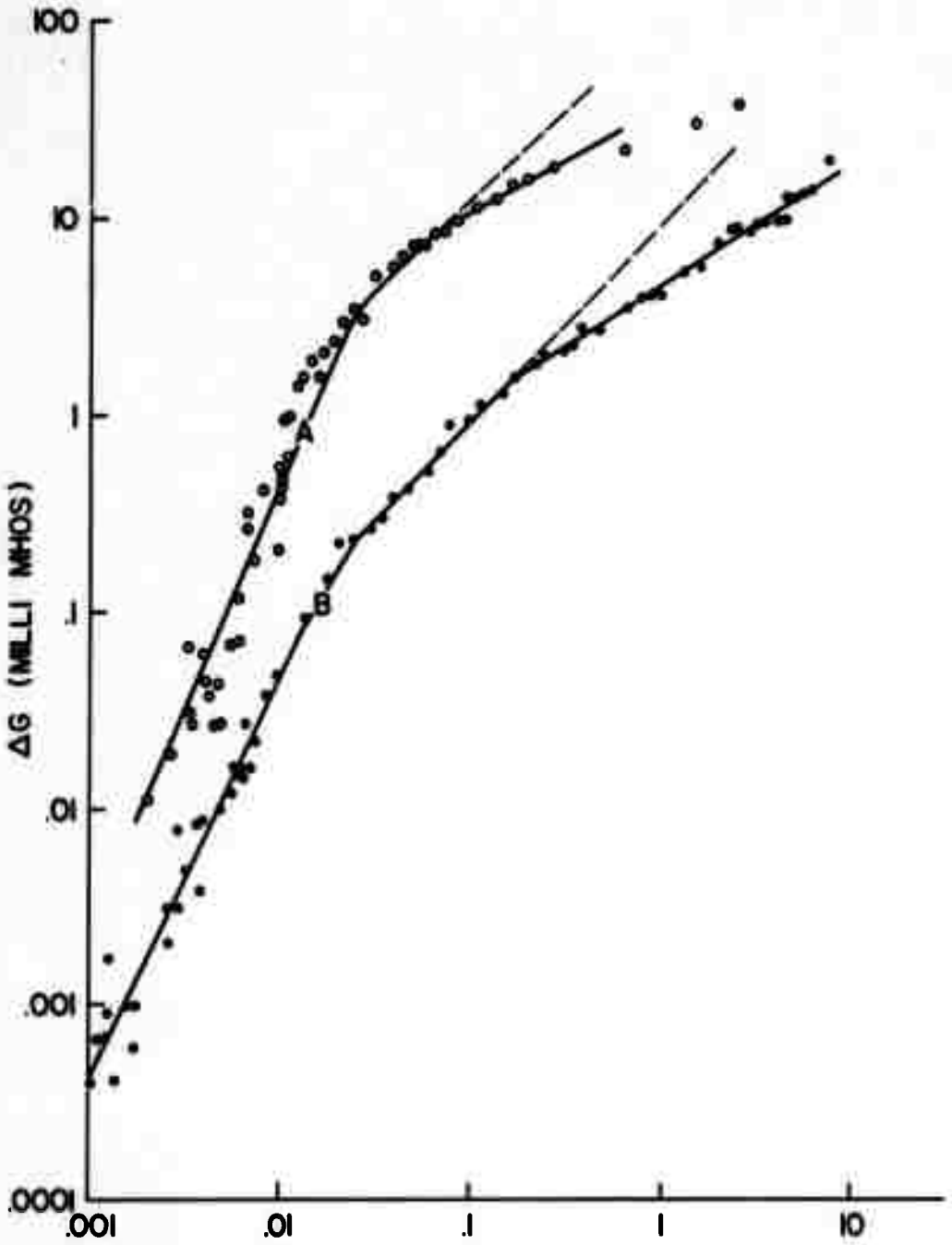


Q-SWITCHED PULSE EXCITATION
•—• EXPERIMENTAL
n-TYPE O^2 DOPED GaAs



Q-SWITCHED PULSE EXCITATION
A – JICK YEE'S CURVE
B – EXPERIMENTAL CURVE
n-TYPE O_2 DOPED GaAs
CRYSTAL THICKNESS = 0.028cm; MOBILITY = 7000cm²/v-sec
LIFE TIME ≈ 10^{-10}secs

Fig 3



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 $\tau >> t_1$ (pulse width) $\approx$ 1 p sec

Fig. 4