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THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS: OBSERVATION OF THREE PHOTON CONDUCTIVITY IN CdS WITH MODE-LOCKED Nd: GLASS LASER PULSES

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

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13. ABSTRACT

The photoconductivity in CdS single and polycrystals was investigated by using mode-locked Nd:glass laser pulses for excitation and was found to exhibit a power law $I^{3.0 \pm 0.2}$, where I is the peak excitation intensity. The three-photon absorption coefficient estimated from the photoconductivity measurement agreed well within an order of magnitude with the theoretical values.

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III

OBSERVATION OF THREE PHOTON CONDUCTIVITY IN CdS WITH MODE-LOCKED Nd:GLASS LASER PULSES*

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OBSERVATION OF THREE PHOTON CONDUCTIVITY IN CdS
WITH MODE-LOCKED Nd:GLASS LASER PULSES

The development of powerful sources of optical radiation by means of mode-locked lasers had made it possible to observe a number of higher order optical interactions in solids. In this letter we report the study of three-photon conductivity in single and polycrystals of CdS at room temperature by using a train of picosecond pulses from a mode-locked Nd:glass laser as the excitation light source. B. M. Arykinadze et al[1] were the first to observe three-photon absorption in CdS. They used focussed Q-switched Nd:glass laser to study the recombination radiation in 5200 Å band after three-photon absorption in CdS at 77°K. They observed this emission near the damage threshold. With picosecond pulses, one could get power density upto a few gegawatts / cm^2 without focussing. Further because of the short time duration of the pulse, thermal damage is negligible even at very high power densities. CdS is a direct band gap II-VI semiconductor whose forbidden energy gap is ≈2.42ev. The Nd:glass laser output has a photon energy of 1.17 ev; thus, the change of photoconductivity is expected to be due to three-photon generation of non-equilibrium charge carriers.

Since we used picosecond pulses for excitation, what we measured was transient photoconductivity one could easily write an expression for transient conductivity change ΔG due to three-photon absorption, following Jick yee's calculation.[2]

$$\Delta G = |e| \frac{a}{c} (\mu_c + \mu_h) \frac{\tau}{3\hbar\omega} I_o \left[1 - \frac{1}{(1+2\beta_3 I_o^2 L)^{1/2}} \right] \qquad (1)$$

where $\frac{a}{c}$ is a geometric factor of the sample, $|e|$ is the magnitude of the electronic charge, μ_e and μ_h are the electron and hole mobilities, respectively, $\beta_3 I_o^2$ is the three-photon absorption coefficient in cm^{-1}, τ is the pulsewidth, I is the incident laser intensity, L is the thickness of the sample and β_3 is the three-photon absorption coefficient in cm^3/Gw^2. In this expression, we have assumed that all the recombination times are long compared to the exciting laser pulses.

The laser used was a Korad K-1 system with a Nd:glass rod having a Brewster-Brewster configuration. The laser was mode-locked by a Kodak 9860 dye solution. The output of the laser consisted of a trian of equally spaced picosecond laser pulses separated by 4.5 nsecs, which was equal to the cavity round trip transit time. The laser beam was directed onto a CdS sample which was connected in series with a 127Ω load resistor to a battery. The change of conductivity in the CdS crystals 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. Since both the laser pulse and the photoconductivity were displayed on the dual beam scope with the same resolving time, the photoconductivity peak corresponded to the peak of the laser pulse. The trigger signal was provided by a beam splitter and another photodiode through a 519 scope which monitored the mode-locking of the beam. The beam intensity was varied by inserting calibrated neutral density filters in the optical path.

We have measured the photoconductivity in both polycrystalline and single crystal CdS. The former had a thickness of 0.02 cm with a dark resistance of well over 100 megohms. This is a commercially available photoconductive cell (CL 902) made by Clairex Corporation and has a spectral peak at 5150 Å (2.42 ev). The latter had a thickness of 0.028 cm with a resistivity greater than $10^8 \Omega$-cm. In both cases, ohmic contacts were made at the two ends of the sample surface by alloying with indium. Both the crystals were checked for any photovoltaic effect with the battery short circuited. It was observed that the photovoltaic effect was negligible (less than 0.1%) compared to the photoconductive signal. The conductivity change ΔG was computed from the change in voltage across the load resistance.

The measured photoconductivity ΔG against laser intensity with mode-locked pulse excitation is shown in Fig. 1 and 2 in a log-log graph. The maximum laser intensity was a few gegawatts/cm^2 and the length of the mode-locked pulse train was 200-400 nsecs. The energy of the pulse train was measured with a calibrated thermopile detector and the pulse width was measured with a TPF cell containing 10^{-3} molar solution of Rhodhamine 6G in ethanol in the usual collapsing geometry.[3] The pulse width was found to be 5-9 psecs without measuring contrast ratio. Both figures 1 and 2 display a slope of (3.0 ± 0.2) indicating the three-photon nature of the excitation. When $\beta_3 I_o^2 L << 1$, we see from eq. (1), ΔG is proportional to I_o^3. This explains the slope 3 observed in the figures 1 and 2. Using eq. (1) we could estimate the three-photon absorption coefficient from the measured photoconductivity. The peak power density I_o was measured. a/c was ≈ 2.0 in the present experiment. Since the crystals were of compensated high resistivity type, the mobility could not be measured accurately and so the normal mobility of 200 cm^2/volt-sec[4] was assumed. The order of magnitude estimate of β_3 gave approximately 0.04 cm^3/Gw^2 for polycrystal and 0.013 cm^3/Gw^2 for single crystal CdS. Recently Jick yee[5] calculated the three-photon absorption coefficient in CdS and found β_3 to be $\sim$ 0.2 cm^3/Gw^2. Arykinadrye et al[1] reported a value of 2.5 cm^3/Gw^2. Their experiment was done with a Q-switched laser pulse and their intensity dependence of recombination radiation was $I_o^{3.4}$. Arsenev et al[6] in their three-photon photoluminescence experiment estimated β_3 using mode locked pulses and they got a value of 0.02 cm^3/Gw^2. Our experimental arrangement was similar to Arsenev's and the order of magnitude agreement with the calculated value and Arsenev's experimental value gives one more evidence to three photon generation process in CdS. The lower values got in the present experiment in comparison to the theory may be attributed to the uncertainty used in the estimation and partly due to the inhomogeneity of the beam distribution. No attempt was made to detect the changes in the three photon absorption coefficient due to anisotropy of CdS as predicted by Jick yee[5]. All the measurements were done with a general random orientation of the crystal. The discrepancy in the values of β_3 for poly and single crystals is too small to ascertain a physical cuase in the difference in response.

The photoconductivity decay of single crystals exhibited three distinct regions, an initial fast decay $\sim$ 1 µsec associated with free electron recombination and trap filling, then an intermediate region of 35µsecs time constant when traps released electrons and finally a very slow decay (of the order of several seconds) associated with emptying of trapping levels close to the equilibrium Fermi level. This agreed with the observations made by Nicholas and Wood.[7] The photoconductor regained its dark resistance only after 2-3 minutes. For this reason, successive laser shots were fixed at 2-3 minutes interval. The very slow decay was conspicuously absent in polycrystal CdS and this could be explained by invoking low trap densities compared to single crystal CdS as observed and explained by J. S. Skarman.[8]

In conclusion, we have reported the observation of the three photon conductivity in single and polycrystalline CdS using mode-locked pulse excitation from a Nd:glass laser. When Q-switched pulses of the same envelope density as that of the mode-locked pulse train was used, the photoconductivity signal was barely observable. The foregoing investigation illustrates the advantage of using picosecond pulses in studying the higher order optical nonlinearities.

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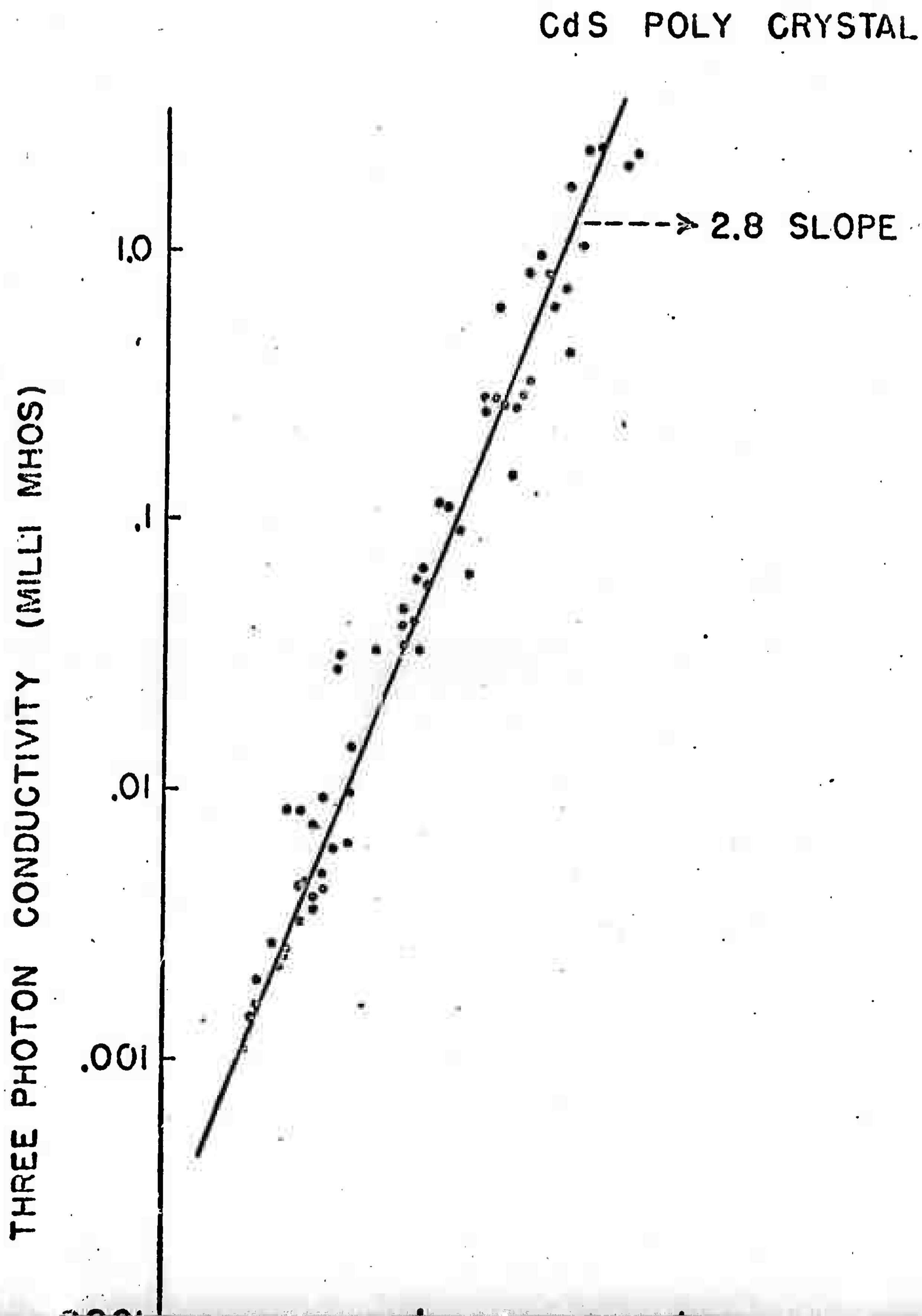
Figure Captions:

FIG. 1. Three - photon conductivity versus relative laser intensity with mode-locked pulse excitation of polycrystal CdS (0.02 cm thick). Data points indicate experimental results. The continuous line represents the least square fit.

Fig. 2. Three-photon conductivity versus relative laser intensity with mode-locked pulse excitation of single crystal CdS (0.028 cm thick, compensated high resistivity type). Data points indicate experimental results. The continuous line represents the least square fit.


CdS POLY CRYSTAL
2.8 SLOPE
1.0
.1
.01
.001
THREE PHOTON CONDUCTIVITY (MILLI MHOS)

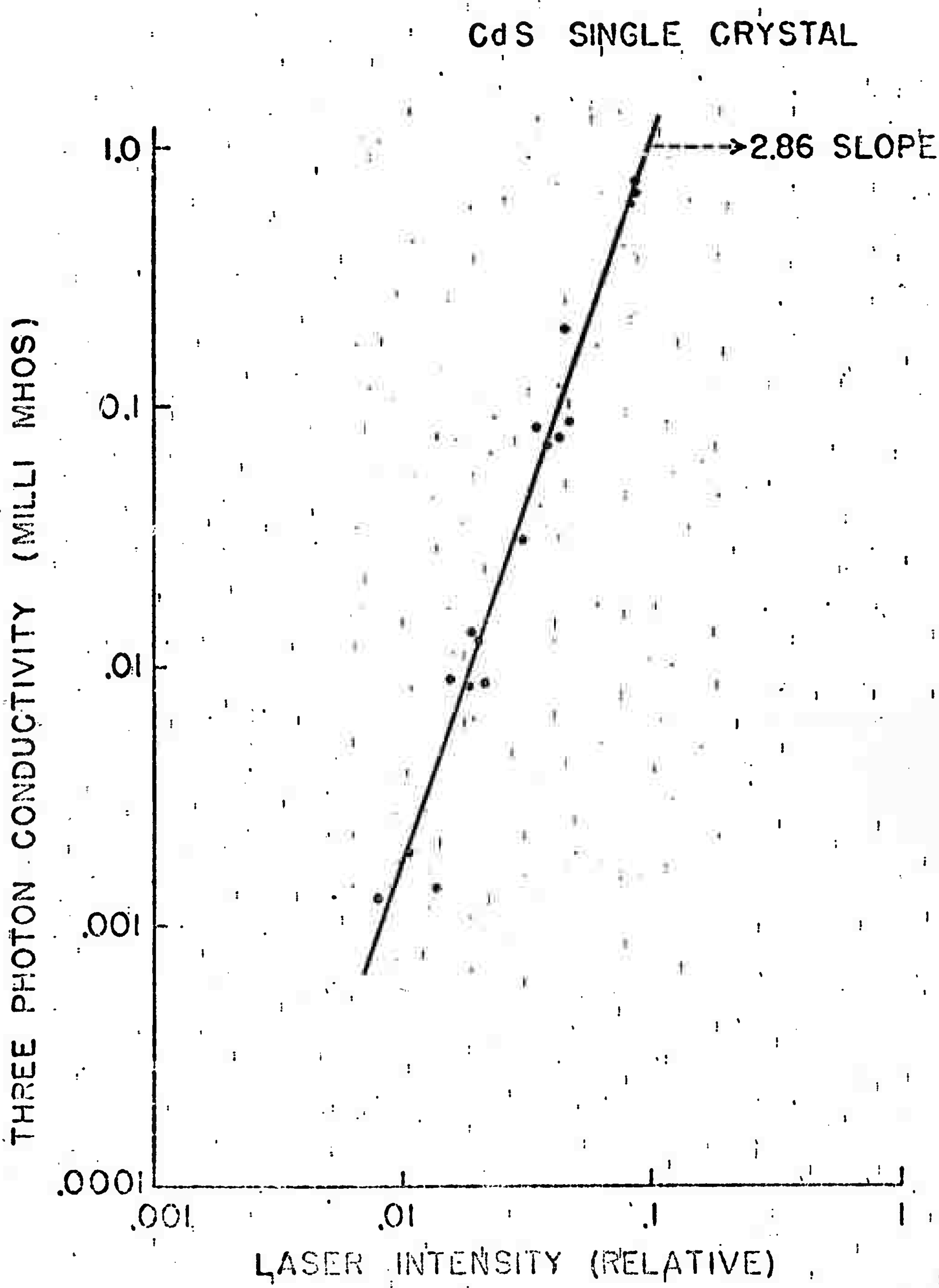

CdS SINGLE CRYSTAL
2.86 SLOPE
1.0
0.1
.01
.001
.0001
THREE PHOTON CONDUCTIVITY (MILLI MHOS)
.001
.01
.1
1
LASER INTENSITY (RELATIVE)

Fig 2