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PICOSECOND PROCESS OF NONLINEAR ABSORPTION IN HIGHLY EXCITED CdS SINGLE CRYSTAL

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Résumé - Il est trouvé que l’absorption nonlinéaire dans le monocristal de CdS dépend du temps par la mesure de transmission avec la technique de corrélation d’impulsion égale de PS. Nous résolvons la variation de l’absorption en fonction du temps en deux composantes : un processus "rapide" et celui "lent" qui sont attribués à l’interaction d’exciton-exciton et à celle d’exciton-électron respectivement.

Abstract - The nonlinear absorption in CdS single crystal is found to be time dependent by measurement of transmission with PS equal-pulse correlation technique. The variation of nonlinear absorption with time can be divided into two components: a 'fast' process and a 'slow' one, which are ascribed to exciton-exciton interaction and exciton-electron interaction, respectively.

I - INTRODUCTION

Many workers have studied the nonlinear absorption phenomenon in CdS at low temperature (4.2K) at exciton energy or less. Several explanations have been given out:

(a) Two-photon absorption due to formation of excitonic molecule /1/.
(b) The inverse process of exciton scattering-induced absorption (e,hs;hν−ex;ex)/2/.
(c) The Mott transition from an exciton gas to an electron-hole plasma/3,4/.
(d) Exciton-exciton interaction/5/.

Experimentally, we studied the nonlinear transmission characteristics of a thin sample of CdS. Using equal-pulse correlation technique/6/, we measured the excited-state relaxation in the picosecond scale, which revealed time dependent light transmission. We find a 'fast' and a 'slow' processes and ascribe them to exciton-exciton interaction and exciton-electron interaction respectively.

II - EXPERIMENT AND DISCUSSION

Mode-locked Ar* laser is used as pump source. Laser wave length λː=5145 Å (2.41eV). Pulse width is 160 PS. Repeating rate is 82 MHz. The pulses is splitted into two beams of orthogonal polarizations and recombined collinearly with one beam delayed with respect to the other by a time interval (160PS,270PS). Since the two pulses are orthogonally polarized there is no coherent effect. Laser spot is focused to ø 0.1 mm. the thickness of CdS single crystal plate is about 100 μm. The experiment arrangement is shown in Fig. 1.
At room temperature band gap of CdS $E_g=2.423\text{eV}$, exciton binding energy $E_x=28\text{meV}$, so that under given condition the absorption is due to the formation of exciton.

When a single beam is used in excitation, the dependence of transmitted light $I$ on the incident light $E$ is shown in Fig. 2. In the case of $E < 15\text{KW/CM}^2$, $I$ increases with $E$ and absorption coefficient $\alpha = \text{const}$. In the case of $E > 15\text{KW/CM}^2$, $I$ will decrease with $E$. Obviously, nonlinear absorption happens in CdS when excitation intensity is greater than $15\text{KW/CM}^2$.

Here, the nonlinearity of absorption process is not due to two-photon absorption. For two photon transition, the selection rule $\Delta l=0,2$ is not satisfied. In our case, excited dense excitons play an important role.
The relation between $T$ and the relative transmission of laser. $\eta$. 

$$\eta = \frac{I(260\text{PS}) - I(\tau)}{I(260\text{PS})}$$

Measure the transmission of the total power of two beams $I$ at different delay time $\tau$. In the case of $E > 15\text{KW/CM}^2$, there exist absorption peaks in about 150PS, when $\tau \geq 167\text{PS}$, $I$ does not change with $\tau$; In the case of $E < 15\text{KW/CM}^2$, $I = \text{const.}$ (Fig. 3). The appearance of deficiency of transmitted light is due to the existence of a 'fast' absorption process.

Next, we set the delay time $\tau$ at 167PS, where 'fast' process has already finished. Then, $I$ does not change with $\tau$. If each beam of incident light varies from a value 15KW/CM$^2$(that is in the case of linear transmission), We find the nonlinearity of transmission happens so long as the total incident intensity is greater than 23KW/CM$^2$(Fig. 4). Let's suppose $\tau$ can be set long enough that the effect of the first pulse on sample completely disappears. Then, no nonlinear absorption should appear, because each beam itself gives only linear transmission. Now, we can make the conclusion: After 167PS, the excited state formed by first pulse interacts with the second pulse still. The nonlinear absorption arises from a slow relaxation process.

Fig. 3 The relation between $T$ and the relative transmission of laser. Cds $\lambda_i = 5145 \text{ Å}$

Fig. 4 The relation between $I$ and $E$, when $\tau = 167\text{PS}$ and $\tau = 0$ under two beams condition.
Experiment shows a broad emission band of CdS centered at 5300 Å with long-wave length tail at low energy side. This peak was observed in cathodoluminescence and photoluminescence by various authors/7,8/ and was ascribed to stimulated emission of exciton-exciton scattering (x-x) process. While according to Koch/9/ and Klingshirn/10/ exciton-electron scattering (x-e) process is more important than (x-x) process at room temperature and has lower threshold of exciton density than x-x process.

Since excitons are neutral particles, the interaction between them is short ranged. It is strong within an exciton Bohr radius and will decay quickly after several Bohr radii/11/. It must happen under highly excited condition. In our case, if 80% photons are absorbed by CdS plate, under the excitation level $E=15 \text{KW/CM}^2$, we have 1 photon/(100Å)².

![Figure 5](image)

Fig. 5 — Absorption peak (experiment) $E=38 \text{KW/CM}^2$
- - Convolution of autocorrelation of the laser pulse. (calculated)
  • $\Delta T(\tau)$ with $b=0.8$ (PS)$^2$, (calculated)

There are sufficiently large number of exciton to produce the interaction between the excitons transiently. Hanamura/12/ pointed out theoretically that when the specific photon density $N/V=10^9 \text{cm}^{-3}$ the nonlinear absorption due to the formation of excitonic molecules is greatly enhanced due to giant oscillation. There are no excitonic molecules at room-temperature, but the excitons may interact each other and result in appearance of additional absorption i.e. the 'fast' process in nonlinear absorption. Since x-x interaction decays with time,

$$\frac{dn_x}{dt} = -\frac{n_x^2}{(1+bt)^2}$$

for bimolecular reaction, the absorption peak must be wider than that of the autoconvolution of the pulses. Let x-x interaction decay according to $n_x^2/(1+bt)^2$ then, the change of transmission power with $\tau$ is

$$\Delta T(\tau) \propto \int_0^\infty C(\tau-t) \frac{1}{(1+bt)^2} \, dt$$

Here $C(\tau)$ is the convolution of the pulse. We find calculation is well coincided with the experimental absorption peak when $b=0.8$ (PS)$^2$(Fig.5). After 'fast' process, exciton density decreases, large number of free carriers may be produced due to x-x scattering or exciton decomposing. Since the life time of exciton in CdS is 0.5ns /10/ and the life time of electron is about $\mu$s, the x-e interaction may exist after the 'fast' process. We ascribe the 'slow' process to x-e process.
CONCLUSION

Time dependent transmission or nonlinear absorption in CdS gives us information about relaxation of excited system. Under highly excited condition, there are various kinds of interactions between quasi-particles with definite relaxation time. We find, there are 'fast' and 'slow' components in non-linear absorption process. Here we ascribe the 'fast' and 'slow' absorption process to x-x and x-e interaction respectively.

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