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**MULTIPHOTON ABSORPTION COEFFICIENTS IN SOLIDS:  
AN UNIVERSAL CURVE**

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MULTIPHOTON ABSORPTION COEFFICIENTS IN SOLIDS:  
AN UNIVERSAL CURVE \*

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**ABSTRACT.** An universal curve for the frequency dependence of the multiphoton absorption coefficient is proposed based on a "non-perturbative" approach. Specific applications have been made to obtain two, three, four and five photons absorption coefficient in different materials. Properly scaling of the two photon absorption coefficient and the use of the universal curve yields results for the higher order absorption coefficients in good agreement with the experimental data.

**RESUMO.** Utilizamos um método "não-perturbativo" para propor uma curva universal que descreve a dependência com a frequência do coeficiente de absorção de muitos fótons. Aplicamos os resultados para diversos materiais obtendo o coeficiente de absorção para dois, três, quatro e cinco fótons. Uma mudança de escala apropriada para o coeficiente de absorção de dois fótons, juntamente com a utilização da curva universal, leva a resultados para os coeficientes de absorção de ordens mais altas em bom acordo com os resultados experimentais.

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## 1. INTRODUCTION

During the past few years the measurement and the calculation of non-linear optical absorption coefficients in solids have been the subject of extensive work. Although most of the experimental work concern two photon processes (Marh 1975; Liu et al 1978; Bechtel et al 1976; Johnston et al 1980; Miller et al 1979, Catalano and Cingolani 1979, 1982; Catalano et al 1973; Falkenstein 1976; Blau and Penzkoffer 1981; Penzkoffer and Falkenstein 1976; Pidgeon et al 1979, Araújo and Lotem 1977, 1982; Lotem and Araújo 1977; Souza and Araújo (S), Prior and Vogt 1979; Arsenev et al 1969, Bosachi et al 1978; Lee and Fan 1974, Bass et al 1979), absorption of three (Catalano et al 1975a), four and five photons (Catalano et al 1972, 1975b) have been reported in the literature. Theoretically, these highly non-linear processes, have been studied within the framework of perturbation theory (PT) (Vaidyanathan et al 1980a, 1980b, 1981; Pidgeon et al 1979; Weiler 1981; Basov et al 1966; Yee 1971) or by the "non-perturbative" approach suggested by Keldysh (Keldysh 1964) almost two decades ago. However, more recently there has been many criticism concerning the application of the Keldysh formalism to processes involving the absorption of a small number of photons as compared to other theoretical models (Vaidyanathan et al 1980a; Weiler 1981).

Few years ago a "non perturbative" scheme similar to Keldysh's has been applied to calculate multiphoton transition rates in direct-gap crystals for arbitrarily large field

intensities (Jones and Reiss 1977). The fact that simple closed analytical solutions can be obtained for the transition rates, stimulated our interest in applying this type, of scheme to calculate the multiphoton absorption coefficient of several direct gap crystals. In that work the field was chosen circularly polarized; this assumption limitates the comparison with the available experimental data which concerns mainly linearly polarized light. In the present work we use a scheme similar to that of (Jones and Reiss 1977) for a linearly polarized field and show that this model calculation provides a universal curve for the frequency dependence of the N-photon absorption coefficient.

For two photon absorption coefficient,  $\beta^{(2)}$ , experiments have been done in a large range of frequencies involving many different materials. In this case the present approach gives a comprehensive description of both the magnitude and the frequency dependence of the two-photon absorption coefficient of many semiconductors and alkali-halides. Although there is an abundance of both theoretical and experimental results concerning these type of process, the same is not true for higher-order absorption. This is not surprising since, theoretically, high-order PT becomes more complicate and experimentally the situation is difficult due to the smallness of these highly non-linear coefficients. Therefore we are much more restricted when relating our model calculations with different published experimental and theoretical results for three, four and five photon absorption.

## 2. THEORY

The present scheme is based on the S matrix formalism

where

$$S = -\frac{i}{\hbar} \int dt d^3x \psi_f^{(-)*}(\vec{x}, t) H_1 \psi_i(\vec{x}, t)$$

and in the dipole approximation  $H_1 = -\frac{e}{mc} \vec{A} \cdot \vec{p}$ , with

$\vec{A} = A_0 \hat{z} \cos \omega t \equiv E_0 / \omega \hat{z} \cos \omega t$ . The spatially homogeneous term  $\frac{e^2}{2mc^2} A^2(t)$  has been removed through the contact transformation  $e^{-\frac{i}{2mc^2\hbar} \int_0^t A^2(\tau) d\tau}$

Following Jones and Reiss 1977 we approximate the final state, related to the full Hamiltonian by a Volkov type of solution (Volkov 1935)

$$\psi_f(\vec{x}, t) = u_{\vec{k}}^c(\vec{x}) e^{i(\vec{k} \cdot \vec{x} - \frac{1}{\hbar} \int_0^t E_c[\vec{k}(\tau)] d\tau)}$$

(2.a)

$$E_c[\vec{k}(\tau)] = E_g + \frac{\hbar^2 k^2}{2m_1} - \frac{e}{m_1 c} \vec{k} \cdot \vec{A}(\tau)$$

Where  $m_1$  is the effective mass of the conduct on band and  $E_g$  the energy gap.

Taking Eqs. (2.a) and (2.b) into Eq. (1) we get

$$S = -\frac{i}{\hbar} \frac{eA_0}{m} P_{vc} \delta_{\vec{k}, \vec{k}'} \int_{-\infty}^{\infty} dt e^{i\left(\frac{\hbar^2 k^2}{2m_1} + E_g - \frac{\hbar^2 k^2}{2m_0}\right)t} \cos \omega t x$$

(3)

$$e^{\frac{i}{\hbar} \left( \frac{e\hbar \vec{k} \cdot \vec{A}_0}{m_1 \omega c} \sin \omega t \right)}$$

where

$$p_{vc} = \int d^3x e^{-i\vec{k}\cdot\vec{x}} u_{\vec{k}}^{\xi}(\vec{x}) \hat{z} \cdot \vec{p} u_{\vec{k}}^{\nu}(\vec{x}) e^{i\vec{k}\cdot\vec{x}} .$$

To evaluate the time integral in Eq.(3) we use the fact that

$$\cos \omega t e^{i\alpha \sin \omega t} = \frac{1}{\alpha} \sum_{n=-\infty}^{\infty} n J_n(\alpha) e^{+in\omega t}$$

to obtain

$$S = - \frac{2\pi i m_1 \omega}{\vec{k} \cdot \hat{z}} \frac{p_{vc}}{m} \sum_N N J_N \left( \frac{e\vec{k} \cdot \hat{z} A_0}{m_1 \omega c} \right) \delta \left( \frac{\hbar^2 k^2}{2m^*} + E_g - N\hbar\omega \right) \quad (4)$$

In Eq.(4) we have defined the reduced effective mass

$$(m^*)^{-1} = (m_1)^{-1} + (|m_0|)^{-1} .$$

From energy conservation we easily obtain the transition rate (per unit of volume) associated to N photon processes.

$$W^{(N)} = \int \frac{d^3k}{(2\pi)^3} W^{(N)}(\vec{k}) \quad (5)$$

where

$$W^{(N)}(\vec{k}) = \left( \frac{2\pi m_1 \omega}{m} \right) \frac{p_{vc}^2}{(\vec{k} \cdot \hat{z})^2} N^2 J_N^2 \left( \frac{e\vec{k} \cdot \hat{z} A_0}{m_1 \omega c} \right) \frac{\delta \left( \frac{\hbar^2 k^2}{2m^*} + E_g - N\hbar\omega \right)}{2\pi \hbar} \quad (6)$$

Using the properties of Bessel's functions, the weak field limit for the multiphoton transition rate is given by

$$W^{(N)} = \frac{1}{2\pi \hbar} \left[ \frac{m_1 \omega p_{vc}}{m(N-1)!} \right]^2 \left( \frac{eA_0}{2m_1 \omega c} \right)^{2N} \left( \frac{2m^*}{\hbar^2} \right)^{\frac{2N+1}{2}} \frac{(N\hbar\omega - E_g)^{\frac{2N-1}{2}}}{(2N-1)!} \quad (7)$$

Using the fact that  $A_0^2 = 8\pi c I / n\omega^2$ , where  $I$  is the intensity of the radiation and  $n$  the index of refraction of the crystal, we may easily obtain the multiphoton absorption coefficient  $\beta^{(N)}$  from Eq. (7) in terms of an universal curve  $f_N(\hbar\omega/E_g)$  for the frequency dependence of  $\beta^{(N)}$

$$\beta^{(N)} = 2 \left( \frac{N\hbar\omega}{I} \right)^N W^{(N)} = 2 \frac{2^{2N+1/2} \pi^{N-1}}{[(N-1)!]^2 (2N-1)} \left( \frac{e^2}{\hbar c} \right)^N \hbar^{3N-4} S_N f_N \left( \frac{\hbar\omega}{E_g} \right)$$

where

(8)

$$f_N(\zeta) \equiv \frac{(N-1)^2}{\zeta^{4N-3}}, \quad \text{and} \quad S_N \equiv \left( \frac{p_{vc}^2}{m^2} \right) \frac{(m^*)^2}{m_1^{2N-2} n^N E_g^{3N-5/2}}$$

Notice that the scaling for different materials is given by the factor  $S_N$  in Eq. (8).

### 3. RESULTS AND DISCUSSIONS

As a first application of the present scheme we calculate the two photon absorption coefficient for several crystals, and compare with available experimental results. From Eq. (8) we have

$$\beta^{(2)} = \frac{2^{11/2} \pi e^4}{3 c^2} \left\{ \frac{p_{vc}^2 (m^*)^{5/2}}{m^2 m_1^2 n^2 E_g^{7/2}} \right\} f_2(\zeta) \quad (9)$$

Numerical calculations for  $\beta^{(2)}$  may be performed with the knowledge of the crystal parameters contained in  $S_2$ . In most cases informations concerning the parameters  $E_g$ ,  $m^*$ ,  $m_1$

and  $n$  may be obtained from the literature, while the knowledge of  $p_{VC}$  usually relies on band structure model calculations. In the present work we used the value obtained by (Ashkinadze et al 1969)  $p_{VC}^2/m^2 \approx 3E_g/4m^*$ , based in a Hartree-Fock calculation. This value has been derived using a better approximation for the band structure than the value obtained within the  $k \cdot p$  approximation ( $\frac{p_{VC}}{m} = \frac{E_g}{4m^*}$ ) which has been used by (Vaidyanathan 1980a, 1980b).

Other approximations may lead to results for the momentum matrix element which are higher by an order of magnitude (Vaidyanathan et al 1981), therefore the results shown in table I must be examined regarding these considerations. In Table I we present the results for the two photon absorption coefficient, given by Eq.(9), for several semiconductors. In this table we also present the theoretical results of (Weiler 1981) and (Vaidyanathan et al 1980a, 1980b, 1981). There is a close agreement between the modified Basov model as quoted by (Vaidyanathan et al 1981) and the present results; the difference with their empirical pseudo-potential calculations is mainly due to their more precise evaluation of  $p_{VC}$ . A comparison of the present results with Weiler's shows that our values lies between his parabolic model calculation and his more elaborated results including exciton effects.

In Fig.1(a) we show the universal curve for  $\beta^{(2)}$  referred specifically to CdS; the theoretical points have been scaled arbitrarily by a factor 6 in order to better account for the experimental data (Araújo and Lotem 1978, Lotem and Araújo 1977). This could be justified by the possibility of



different choices of  $p_{vc}$ . A similar universal curve for  $\beta^{(2)}$  has been obtained by Weiler, modifying the original work of (Pidgeon et al 1979); the difference between Weiler's and the results herein are due to different scaling factor..

To construct Table II we use the curve presented in Fig.1(a), properly scaled for each crystal by a factor  $S_2(\text{crystal})/S_2(\text{CdS})$ . In the case of alkali-halides, due to the lack of information in the literature, we assumed  $m^* = m_1$ ; and have considered the different existing assignments for the energy gap since calculations are particularly sensitive to this parameter.

The results of Tables I and II show that, although very simple, the present scheme leads to results comparable with more elaborated theoretical methods involving two photon absorption. Of course the complexity of those methods based on PT increases with the multiplicity of the process, becoming quite difficult for high order absorption. This is not the case of the present "non-perturbative" scheme for which closed analytical results are obtained for a general process involving N photon absorption.

As a second application of this method we calculated the absorption coefficient for three, four and five photons in different crystals.

To obtain  $\beta^{(3)}$  we use Eq.(8), and the results for several materials are shown in Fig.1(b) and Table III. Fig.1(b) has been obtained using the same arbitrary scaling factor (attributed to  $p_{vc}$ ) as in the calculation of  $\beta^{(2)}$  for CdS. In Table III we compare our results with the third order PT of

(Mitra et al 1981) and the agreement is in general quite good, except for ZnSe. We cannot explain this discrepancy. We also present in this same table the value for the scaled  $\beta^{(3)}$ , which is in general agreement with the experimental data.

To avoid the uncertainties in the calculations of  $p_{vc}$ , we also determined the ratio between the coefficients involving different multiplicities for the same crystal. This ratio is independent of  $p_{vc}$ . We present calculations for materials where experiments have been reported in the literature. For CdS the theoretical ratio  $\beta^{(3)}/\beta^{(2)}$  is  $0.57 \times 10^{-3} \text{ cm}^2/\text{GW}$  to be compared with  $0.53 \times 10^{-3} \text{ cm}^2/\text{GW}$  obtained from (Araújo and Lotem 1978; Lotem and Araújo 1977; Catalano and Cingolani 1979).

The experimental value of  $\beta^{(3)}$  from (Catalano and Cingolani 1979) was obtained in terms of  $\beta^{(2)}$  (Araújo and Lotem 1978). This value has been corrected to take into account the proper frequency and resistivity dependence, using an empirical relation connecting  $\beta^{(2)}$  to the resistivity (Souza and Araújo(S)). Without this correction the reported results are  $0.15 \times 10^{-3} \text{ cm}^2/\text{GW}$  (4a,17) and  $0.69 \times 10^{-3} \text{ cm}^2/\text{GW}$  (Catalano et al 1973; Araújo and Lotem 1978). This same ratio calculated for GaP yields  $0.15 \text{ cm}^2/\text{MW}$  which agrees with the experimental ratio  $0.13 \text{ cm}^2/\text{MW}$  (Catalano et al 1975a; Araújo and Lotem 1978) within 15%.

Experiments concerning the absorption of more than three photons are very few, and due to the smallness of the absorption coefficients, the use of high power lasers is necessary. This may cause damage, F-center formation and excess of free-carriers in the samples, which make this type of experiment very difficult (Liu et al 1978; Araújo and Lotem 1982;

Prior and Voigt 1979). We briefly compare our results with the experimental data available for four photon absorption in ZnS (Catalano et al 1975b) and KI and five photons in NaCl (Catalano et al 1972). The ratio  $\beta^{(4)}/\beta^{(3)}$  in ZnS agrees with the data within an order of magnitude. This is also the case for  $\beta^{(5)}/\beta^{(3)}$  in NaCl. Concerning  $\beta^{(4)}/\beta^{(2)}$  in KI the theoretical result is higher by five orders of magnitude compared with the experimental data (Catalano et al 1972). We should mention that if one uses the experimental value reported in (Dneprovkii 1966) the discrepancy reduces to two orders of magnitude.

#### 4. CONCLUSIONS

In conclusion we may say that there is close agreement between our results and those obtained from different theoretical approaches. Furthermore the simplicity of the present method in treating high multiplicity processes makes it very convenient in comparison to the perturbative treatments. The universal curve of Eq.(8) may greatly simplify any future comparison between theory and experiment. The present approach could eventually be modified to include effects of non parabolicity of the bands, at least, in the weak field limit. This could be of interest when studying a particular material in a very large range of frequencies.

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FIGURE CAPTIONS

Fig.1 (a) Frequency dependence of the two photon absorption coefficient.

Experimental points from (Araújo and Lotem 1978, Lotem and Araújo 1977)

(b) Frequency dependence of the three photon absorption coefficient. Experimental points from Catalano and Cingolani 1979; Catalano et al 1973.

Table I: Two photon absorption coefficient  $\beta^{(2)}$  [cm/MW].

Crystal	$E_g$ (a)	$n$ (b)	$\zeta$	$m^*$ (c)	$m_1$ (c)	Present work	Weiler (d)	Modified Basov (c)	(E.P.M.) (c)
CdS	2.42	2.41	0.74	0.192	0.20	0.012	-	-	-
ZnSe	2.58	2.65	0.69	0.132	0.17	0.008	0.009	0.011	0.081
GaAs	1.44	3.42	0.81	0.063	0.07	0.034	0.024	0.039	2.179
InP	1.28	3.3	0.91	0.06	0.073	0.035	0.030	0.056	0.351
CdSe	1.74	2.5	0.67	0.12	0.13	0.031	-	-	-
CdTe	1.44	2.6	0.81	0.08	0.11	0.034	0.034	0.037	0.201
InSb	0.18	4.2	0.67	0.007	0.019	3.4	-	-	-

(a) Strehlow and Cook 1973

(b) Driscoll and Vaughan 1978

(c) Vaidyanathan et al 1980b, 1981

(d) Weiler 1981

Table II: Scaled two photon absorption coefficient  $\bar{\beta}^{(2)}$  : ((cm/MW).

Crystal	$\zeta$	Scaling factor	$\bar{\beta}^{(2)}$	Experimental results
ZnSe	0.69	0.618	0.049	0.04 - 0.08 <sup>(a)</sup>
GaAs	0.81	3.02	0.20	0.02 - 5.6 <sup>(b)</sup>
InP	0.91	4.2	0.223	0.21 <sup>(c)</sup>
CdSe	0.67	2.36	0.184	0.03 - 0.95 <sup>(d)</sup>
CdTe	0.81	2.66	0.176	0.13 - 0.17 <sup>(e)</sup>
InSb	0.67	262.8	20.5	0.2 - 16 <sup>(f)</sup>
NaCl	0.52	0.047	$0.7 \times 10^{-3}$	$3.5 \times 10^{-3}$ (g)
KCl	0.55	0.03	$1.05 \times 10^{-3}$	$1.7 \times 10^{-3}$ (g)
RbCl	0.56	0.06	$7.4 \times 10^{-3}$	$1.02 \times 10^{-3}$ (g)
KBr	0.62	0.071	$4.8 \times 10^{-3}$	$2 \times 10^{-3}$ (g)
KI	0.57	0.117	$5.6 \times 10^{-3}$	$7.29 \times 10^{-3}$ (g)
RbI	0.60	0.138	$8.3 \times 10^{-3}$	$5.08 \times 10^{-3}$ (g)

(a) Araújo and Lotem 1978; Catalano and Cingolani 1982, Arsenev et al 1969

(b) Bosacchi et al 1978

(c) Lee and Fan 1974

(d) Bechtel and Smith 1976; Vaidyanathan et al 1980a,1980b,1981

(e) Catalano and Cingolani 1982; Arsenev et al 1969

(f) Johnston et al 1980; Vaidyanathan et al 1980a

(g) Liu et al 1978



Table III: Three photon absorption coefficient  $\beta^{(3)}$  and scaled  $\bar{\beta}^{(3)}$ , [ $\text{cm}^3/(\text{GW})^2$ ].

Crystal	$E_g$	n	$\zeta$	$m^*$	$m_1$	MITRA et al [Ref. 8c] (Parabolic Model)	Present work	Scaling Factor	$\bar{\beta}^{(3)}$	Experimental results
CdS	2.42	2.32	0.48	0.192	0.20	$8.2 \times 10^{-3}$	$8.1 \times 10^{-3}$	1	$49 \times 10^{-3}$	$11 \times 10^{-3} - 55 \times 10^{-3}$ (a)
GeAs	1.44	3.34	0.42	0.063	0.07	$4.27 \times 10^{-1}$	$1.77 \times 10^{-1}$	24.4	$10.7 \times 10^{-1}$	-
ZnS	3.8	2.34	0.47	0.28	0.3	-	$0.35 \times 10^{-3}$	$0.43 \times 10^{-1}$	$2.12 \times 10^{-3}$	$1.7 \times 10^{-3}$ (b)
ZnSe	2.58	2.65	0.34	0.132	0.17	1.0	$0.29 \times 10^{-3}$	3.56	$1.73 \times 10^{-3}$	-
InSb	0.18	3.85	0.39	0.007	0.015	$7.76 \times 10^4$	$1.4 \times 10^4$	$28.6 \times 10^5$	$7.87 \times 10^4$	-
GaP	2.78	3.15	0.42	0.187	0.187	-	$1.69 \times 10^{-3}$	0.23	$10 \times 10^{-3}$	$33 \times 10^{-3}$ (c)
NaCl	8.97	1.6	0.4	0.6	0.6	-	$2.87 \times 10^{-6}$	$0.47 \times 10^{-3}$	$16.5 \times 10^{-6}$	$600 \times 10^{-6}$ (d)
KCl	8.5	1.5	0.353	0.496	0.496	$4.57 \times 10^{-6}$	$0.91 \times 10^{-6}$	$1.06 \times 10^{-3}$	$5.5 \times 10^{-6}$	-

(a) Catalano and Cingolani;

(b) Catalano et al 1975 b

(c) Catalano et al 1975 a

(d) Catalano et al 1972

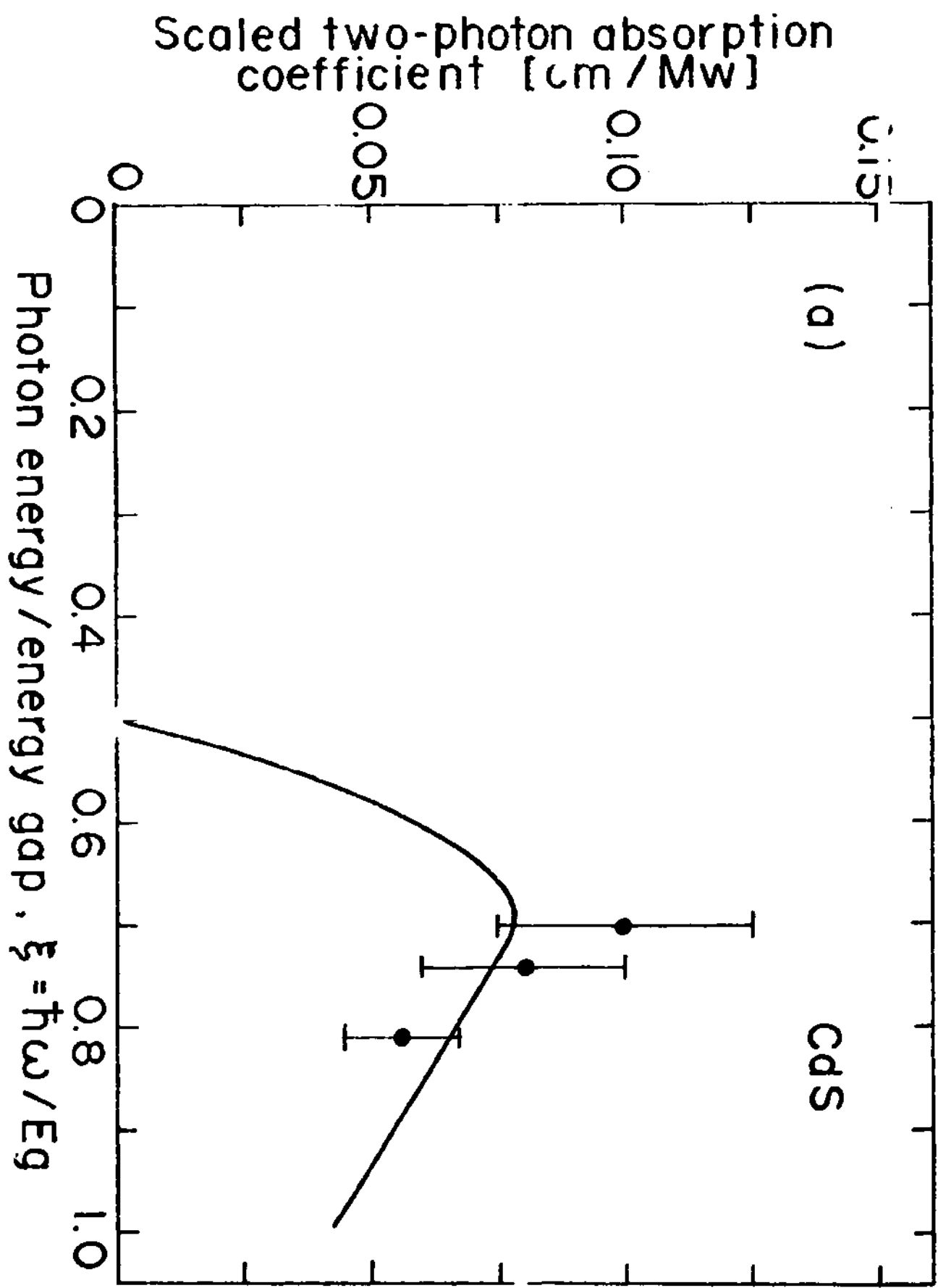


Figure 1(a)

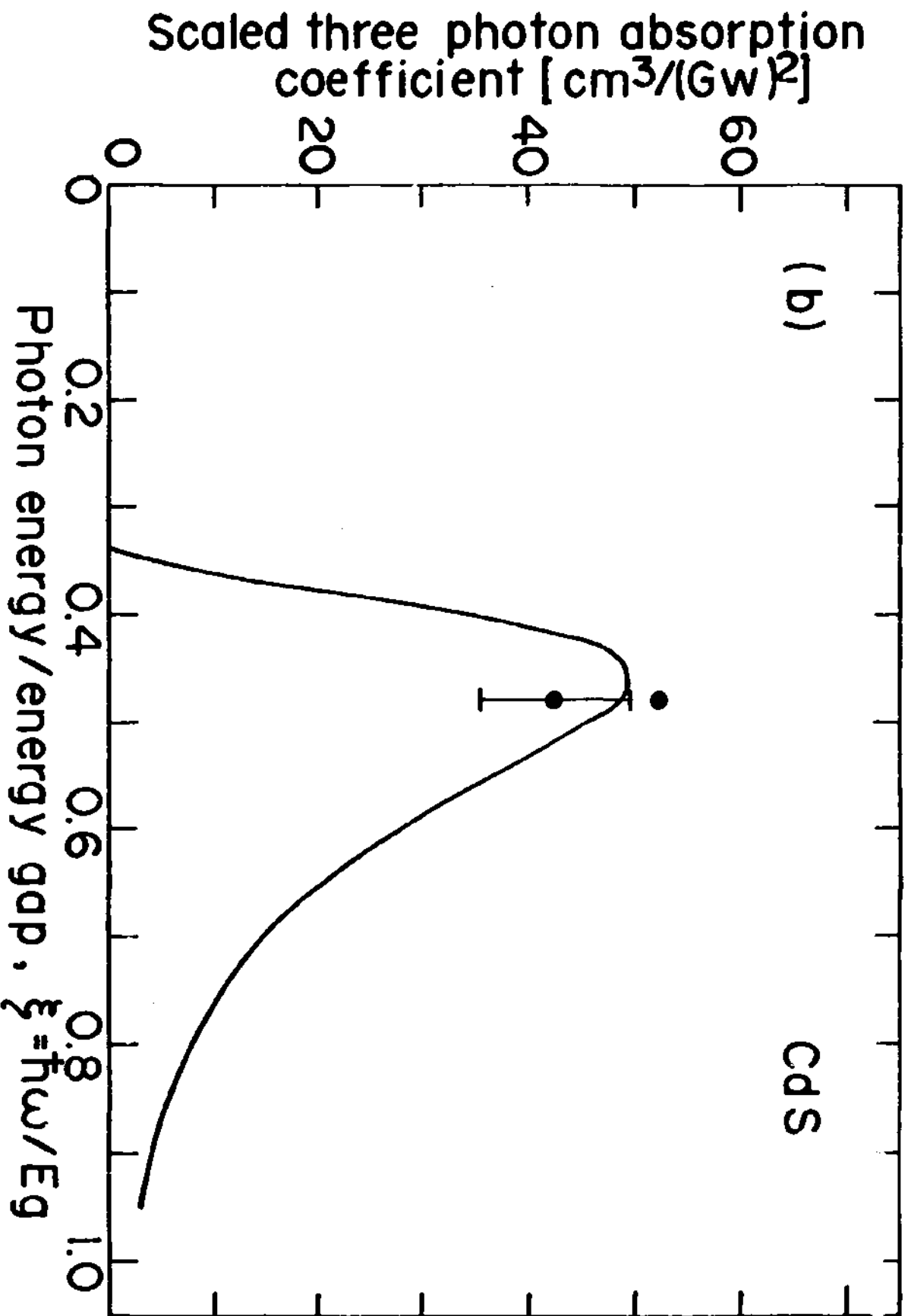


Figure 1 (b)