NONLINEAR ABSORPTION AND ASSOCIATED REFRACTION IN SEMICONDUCTORS

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Abstract

Simple parametric scaling rules for the two-photon absorption coefficients β of semiconductors have been experimentally confirmed by measuring β for ten different semiconductors using picosecond pulses. These dependencies agree with recent theory and allow the prediction of β in other materials at different wavelengths. The strong self-defocussing observed in these materials is consistent with nonlinear refraction by the two-photon generated free carriers.

De simples lois paramétriques d'échelle pour les coefficients d'absorption β à deux photons, dans les semiconducteurs, ont été confirmées expérimentalement en mesurant β pour dix semiconducteurs différents avec des pulses-picosecondes. Ces dépendances sont en accord avec la théorie récente et permettent de prédire β pour d'autres matériaux, à différentes longueurs d'onde. La forte auto-focalisation observée dans ces matériaux est compatible avec la réfraction nonlinéaire associée aux porteurs libres générés par un mécanisme à deux photons.

Es wurden einfache Skalengesetze, die nur Materialparameter enthalten, für die Zwei-Photon-Absorptionskoeffizienten β von Halbleitern experimentell bestätigt, indem β für zehn verschiedene Halbleiter unter Verwendung von Picosekunden-Pulsen gemessen wurde. Diese Abhängigkeiten stimmen mit neueren Theorien überein und erlauben die Vorhersage von β in anderen Materialien bei anderen Wellenlängen. Die Beobachtung von starker Selbstfokussierung ist konsistent mit der nichtlinearen Brechung durch freie Ladungsträger die durch 2-Photon-Absorption erzeugt wurden.

The rapidly developing uses of semiconductors in nonlinear optical devices demand careful study of their nonlinear optical properties. One parameter that is important in the design of nonlinear optical devices and that has received considerable attention over the past two decades is β, the two-photon absorption coefficient.

Figure 1 shows experimentally determined values of β reported over the past 19 years for GaAs and CdSe at a wavelength of 1.06 μm. Note the extreme variations (this is a semilogarithmic plot) and the general trend toward smaller values in more recent years. Comparison of theory to these experiments is meaningless unless we understand the reasons for these differences. This work shows that these large discrepancies are primarily experimental and not simply due to extrinsic material properties [1]. We find that our results agree with a simple two-band model where [2,3,4]

$$\beta = (3.1 \pm 0.5) \cdot 10^3 \frac{\sqrt{E_p}\, F_2\, (\frac{2\hbar\omega}{Eg})}{n^2\, Eg^3} \quad . \qquad (1)$$

β is in cm/GW, E_p is approximately 21 eV for the materials studied, n is the refractive index and Eg is the energy gap in eV. The function F_2 is given by $F_2(x) = (x-1)^{3/2}/x^5$.

Table 1 shows the materials studied along with the material parameters, the experimentally determined value of β, and the prediction of eqn. (1). These values of β agree well except for ZnTe where the experimentally determined value of β is considerably higher than expected from eqn. (1). For ZnTe two-photons couple states only 3% above the band edge where the effects of excitons are expected to be the greatest [4,5]. These efects are not included in the simple two-band model.

The constant of proportionality in eqn. (1) was determined by performing a least squares fit of the data, excluding ZnTe to the functional dependence of eqn. (1). This constant is a factor of 1.7 lower than predicted by the simple two-band model [4]. Inclusion of more realistic band structure only changes the functional dependence of F_2 slightly but does give considerably closer agreement in absolute value (on average within 26%) [1,2,3]. This agreement is remarkable in light of previous results and when it is noted that ten different semiconductors were studied and they had different structures.

This is extremely fortuitous since it means that refinements in band structure don't lead to large changes in β nor to changes in scaling. It is also interesting to note that the non-perturbative approach to multi-photon absorption gives the same scaling law as eqn. (1) in the limit of small

Fig. 1. A semilogarithmic plot of the reported two-photon absorption coefficients of GaAs (lower case letters) and CdSe (upper case letters) versus year:

(a) Ref. 16	(b) Ref. 17	(c) Ref. 18
(d) Ref. 19	(e) Ref. 20	(f) Ref. 21
(g) Ref. 21	(h) Ref. 5	(i) Ref. 22
(k) Ref. 23	(l) Ref. 24	(m) Ref. 25
(n) This work	(A) Ref. 16	(B) Ref. 18
(C) Ref. 26	(D) Ref. 9	(E) Ref. 27
(F) This work		

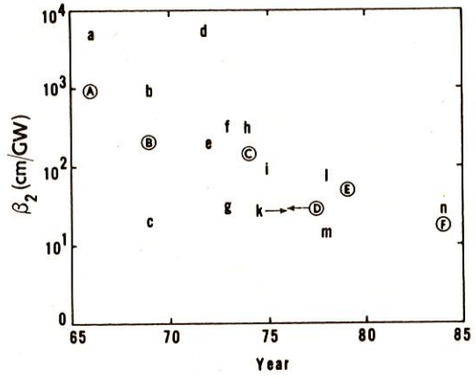

Fig. 2. Vidicon scan through the center of the beam transmitted through the CdSe sample at an irradiance of (a) 1 GW/cm^2, (b) 0.3 GW/cm^2 at a distance of 0.5 m behind the sample. The pulsewidth used was 92 ps FWHM. The beam profiles are normalised to have the same on-axis fluence. The dashed line is experimental and the solid line the theoretical fit.

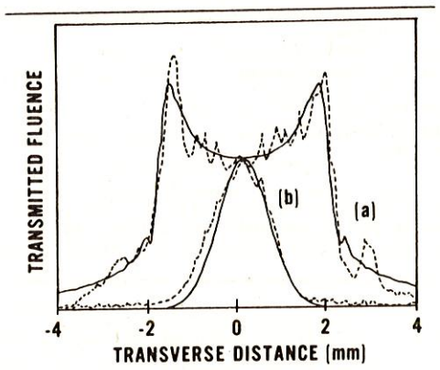

Table 1 Materials Studied, Materials Parameters, and Two Photon Absorption coefficients

Material	Form[a]	E_g(ev)	n	E_p(ev)[b]	β^{exp}(cm/GW)	β^{Theor}(cm/GW)
		λ = 1.06μm		$2\hbar\omega$ = 2.34ev		
GaAs[c]	Z	1.42[i]	3.43[i]	25.7	23	19.7
CdTe[d]	Z	1.44[e]	2.84[e]	20.7	22	25.1
CdTe[d]	Z_p	1.44[e]	2.84[e]	20.7	15	25.1
CdSe[e]	W	1.74[j]	2.56[e]	21	18	18.6
$CdS_{.25}Se_{.75}$[e]	W	1.78[k]	2.51[k]	21	15	17.7
$CdS_{.5}Se_{.5}$[e]	W	1.93[k]	2.45[k]	21	10	12.1
ZnTe[e]	Z	2.26[i]	2.79[e]	19.1	4.5	0.89
		λ = 0.53μm		$2\hbar\omega$ = 4.68ev		
CdS[e]	W	2.42[e]	2.60[i]	21	5.5	4.87
ZnSe[f]	Z_p	2.67[i]	2.70[i]	24.2	5.5	4.27
ZnO[g]	W	3.2[m]	2.05[i]	21	5.0	4.77
ZnS[h]	Z_p(clear)	3.66[i]	2.40[i]	20.4	2.0	2.10
ZnS[h]	Z_p(yellow)	3.66[i]	2.40[i]	20.4	3.5	2.10

(a) Z = zincblende; W = wurtzite; p = polycrystalline.
(b) values taken from Ref. 28. For values not listed in this reference, the value of 21 eV was assumed.
(c) Morgan Semiconductors, Garland, Texas.
(d) II-VI Inc., Saxonburg, Pennsylvania.
(e) Cleveland Crystals, Euclid, Ohio.
(f) Raytheon Co., Bedford, Mass.
(g) Atomergic Chemetals, Plainview, N.Y.
(h) CVD Inc., Woburn, Mass.
(i) Ref. 12.
(j) Ref. 13.
(k) These values were obtained by extrapolation as a function of composition between the known values for CdS and CdSe, see Ref. 14.
(m) Ref. 15.

two-photon absorption [6]. Actually, it has been shown that this must be true for any theory of two-photon absorption by Wherrett using, among other methods, a dimensional analysis approach [2].

The reasons for the extreme spread in previous results as exemplified by the data of Fig. 1 are primarily experimental. The downward trend shows the improvement in experimental method and interpretation. Early work was plagued by multimode lasers in both space and frequency which resulted in large variations in irradiance which will always yield an underestimate of the irradiance when not resolved. This in turn leads to an overestimate of β. An additional problem is that most researchers have used pulses of several nanoseconds duration where the two-photon generated carrier absorption can actually dominate the overall absorption. This again will result in an overestimate of β. The use of much shorter pulses which allows higher irradiance with lower energy, reduces the relative contribution of photogenerated carrier absorption with respect to two-photon absorption to a negligible ratio.

Another possible experimental problem is the large self-defocussing observed in these samples. Even when the absorption of the photogenerated carries is kept small by using picosecond pulses the change in refractive index caused by them can be large [1,7]. Additionally, even if the defocussing within the sample is negligible, free-space propagation to the detector can change the energy distribution on the detector and even result in energy missing the detector. We show in Fig. 2 the transmitted spatial fluence distribution as displayed on a vidicon used in a region of linear response (equivalent to a pinhole scan) for two different input irradiance levels. The theoretical fit shown assumes that the nonlinearity is completely accounted for by an index change proportional to the carrier density produced by the previously determined value of β [1]. This represents a single parameter fit for the index change per carrier. The result gives an index change of ~ 3.5 ± 1 times the Drude contribution, which may be explained by band blocking from interband transitions [8].
Given the above experimental considerations, we used well characterised collimated TEM_{00}, nearly transform limited picosecond pulses, thin samples (2-5 mm), and large area uniform response detectors. The transmission detector was in turn placed directly behind the sample. We note that the work of Bechtel and Smith in 1976 which used basically the same technique (see Fig. 1) included four of the ten materials studied here. These values agree closely with our results [9].

In conclusion we have experimentally verified the scaling rules predicted by theory for two-photon absorption in semiconductors giving us a

predictive capability. For example, if we extend our result to two-photon absorption in InSb at 10 μm at 300°K, we obtain a value of $\beta = 6.8$ cm/MW. This value is in excellent agreement with the measured value of 8 cm/MW where photogenerated carriers were carefully accounted for [10]. We have also explained the mechanism responsible for the observed self-defocussing, namely the photogenerated carriers. Combinations of two-photon absorption and self-refraction have uses for nonlinear optical elements such as optical power limiters [11].

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