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Two-photon absorption in several direct-gap crystals

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The formulas for describing two-photon absorption in direct-gap crystals, developed by Keldysh, Braunstein, and Basov, are compared. It is shown that the Keldysh formula is closely related to Braunstein's for allowed-allowed transitions, while Basov's proposed formula more closely corresponds to Braunstein's for allowed-forbidden transitions. A number of suggested improvements are introduced in Braunstein's formula and corrections to Basov's are presented. The two-photon absorption coefficients for several direct-gap semiconductors and alkali halides are calculated from these formulas and compared with the available experimental data. The formulas of Keldysh and Braunstein for describing allowed-allowed transitions, where one considers excitonic intermediate states and nonparabolic energy bands, are found to give results in fair agreement with published experimental results. It is noted, in comparison, that Basov's and Braunstein's absorption coefficients for allowed-forbidden transitions are smaller than available experimental data by orders of magnitude.

INTRODUCTION

Nonlinear absorption in crystalline solids has been extensively studied in recent years.[1] The results of different theoretical approaches for calculating multiphoton-absorption coefficients differ from each other and from experimental data by as much as three orders of magnitude. It was found that for direct-gap semiconductors,[2] Braunstein's formula[3] for allowed-allowed transitions underestimated the two-photon-absorption coefficients by an order of magnitude, while the Basov formula[4] overestimated them. This result is somewhat surprising since the Basov formula corresponds to the case for allowed-forbidden transitions. On the other hand, Keldysh's results[5] were close to Braunstein's allowed-allowed results. This observation is in contradiction with the generally held view[4,6-8] that the Keldysh formula is inaccurate when the photon multiplicity is small.

In this paper, we point out the similarity between the Keldysh and Braunstein allowed-allowed formulas and interpret and comment on the numerical results obtained for several direct-gap semiconductors and alkali halides. Several corrections to the Basov formula are suggested and through these modifications improved agreement is achieved. In addition, changes are proposed for Braunstein's formula regarding the oscillator strength and electronic energy-band structures which bring its predictions more in line with the experimental data.

THEORY

A. Braunstein's formulas

Braunstein and Ockman[3] were the first to derive an expression for the probability of electronic transitions from the valence band to the conduction band of a crystalline solid by the simultaneous absorption of two light quanta in terms of the band-structure parameters. They considered three parabolic energy bands: an initial valence band v, a final conduction band c, and an intermediate band n, which was chosen to be a higher conduction band. The minimum energy separation between bands v and c was E_g, and that between v and n was ΔE (Fig. 1). The energy separation between the valence and conduction bands was assumed to be of the form

$$E_{vc}(\vec{k}) = E_g + \frac{\hbar^2 k^2}{2m_{vc}^*}, \tag{1}$$

where m_{vc}^* is the reduced effective mass of the bands v and c. A similar relation can be given for $E_{vn}(\vec{k})$. Depending on the symmetries of the bands, three types of two-photon transitions were possible; allowed-allowed (aa), allowed-forbidden (af), and forbidden-forbidden (ff). For allowed transitions, the interband momentum matrix elements are given by

$$|p_{ij}|^2 = \frac{m\hbar\omega_{ij} f_{ij}}{2}, \tag{2}$$

where the f_{ij}'s are the dimensionless oscillator strengths, while for forbidden transitions,[9]

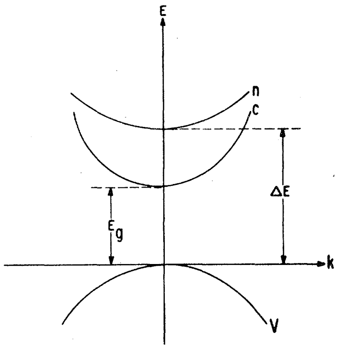

FIG. 1. The energy-band model used in Braunstein's formulation. v, c, and n refer to the initial valence, final conduction, and intermediate conduction bands, respectively. All three energy bands are assumed to be parabolic.

$$|p_{ij}|^2=\left(\frac{m}{m_\dagger}\right)^2\hbar^2(\vec{e}_q\cdot\vec{k}_i)^2\,, \tag{3}$$

where $m_\dagger$ is an "effective mass" for the transition, and $\vec{e}_q$ is a unit vector for the photon polarization.

After taking into account the electron spin, the photon multiplicity, and the correction factor[2] $\epsilon_\infty{}^{-2}$, where ϵ_∞ is the high-frequency dielectric constant of the solid, we obtain the following expressions for the two-photon-absorption coefficients:

$$\beta_{B,p}^{\text{aa}}(\omega)=\frac{2^{5/2}\pi e^4(m_{vc}^*)^{3/2}\Delta E(\Delta E-E_g)f_{vn}f_{nc}}{\epsilon_\infty c^2m^2(\hbar\omega)^3}\times(2\hbar\omega-E_g)^{1/2}[\Delta E-\hbar\omega+(m_{vc}^*/m_{vn}^*)(2\hbar\omega-E_g)]^{-2}\,, \tag{4}$$

$$\beta_{B,p}^{\text{af}}(\omega)=\frac{2^{9/2}\pi e^4(m_{vc}^*)^{5/2}\Delta E f_{vn}(2\hbar\omega-E_g)^{3/2}}{3\epsilon_\infty c^2m(m_\dagger)^2(\hbar\omega)^3}\times[\Delta E-\hbar\omega+m_{vc}^*/m_{vn}^*(2\hbar\omega-E_g)]^{-2}\,. \tag{5}$$

In an attempt to improve the Braunstein formula, we used, in the place of the parabolic energy bands described by Eq. (1), more realistic nonparabolic energy bands of the form[10]

$$E_{vc}(\vec{k})=E_g\left(1+\frac{\hbar^2k^2}{m_{vc}^*E_g}\right)^{1/2}\,, \tag{6}$$

with a similar relation for $E_{vn}(\vec{k})$. The resulting expression for the two-photon-absorption coefficient for allowed-allowed transitions is then

$$\beta_{B,np}^{\text{aa}}(\omega)=\frac{8\pi e^4(m_{vc}^*)^{3/2}\Delta E(\Delta E-E_g)f_{vn}f_{nc}}{\epsilon_\infty c^2m^2(E_g)^{1/2}(\hbar\omega)^2}\times\left(\Delta E\left\{1+\frac{E_g}{\Delta E}\frac{m_{vc}^*}{m_{vn}^*}\left[\left(\frac{2\hbar\omega}{E_g}\right)^2-1\right]\right\}^{1/2}-\hbar\omega\right)^{-2}\times\left[\left(\frac{2\hbar\omega}{E_g}\right)^2-1\right]^{1/2}\,. \tag{7}$$

Later we will calculate the numerical values of the two-photon-absorption coefficients in specific crystals using Eqs. (4), (5), and (7) in order to compare the Braunstein two-photon-absorption edge with the Keldysh result. Since the values of the intermediate band effective masses and the oscillator strengths are unknown, we will assume $m_n=m_c$ and estimate the oscillator strengths from the f sumrule[11] and $\vec{k}\cdot\vec{p}$ approximation.[2,12] Thus

$$f_{vn}=\frac{m}{2m_{vc}^*}\,, \tag{8}$$

$$f_{nc}=\frac{m}{m_c}-1\,. \tag{9}$$

We further assume that $\Delta E=2E_g$, which is justified since the two-photon-absorption coefficient is not sensitive[3] to ΔE as long as $\Delta E>E_g$. Close to the two-photon-absorption edge, $2\hbar\omega\simeq E_g$, both Eqs. (4) and (7) simplify to

$$\beta_B^{\text{aa}}(2\hbar\omega\simeq E_g)\simeq\frac{2^{9/2}\pi e^4(m_{vc}^*)^{1/2}(m/m_c-1)(2\hbar\omega-E_g)^{1/2}}{9\epsilon_\infty c^2m(\hbar\omega)^3}\,. \tag{10}$$

B. Keldysh's formula

Keldysh[5] has calculated the electronic transition rate per unit volume between the valence and conduction bands of a direct-gap crystalline solid, in the presence of an electromagnetic radiation of the form $\vec{E}=\vec{E}_0\cos\omega t$, by assuming that the transitions take place between Stark-shifted energy bands. The electronic wave functions were described by Houston functions,[13] and the following expression was obtained for the probability of the simultaneous absorption of several light quanta:

$$W=\left(\frac{2\omega}{9\pi}\right)\left(\frac{m_{vc}^*\omega}{\hbar}\right)^{3/2}\left(\frac{e^2E_0^2}{16m_{vc}^*\omega^2E_g}\right)^{\langle\tilde{E}_g/\hbar\omega+1\rangle}\times\Phi\left[\left(2\left\langle\frac{\tilde{E}_g}{\hbar\omega}+1\right\rangle-\frac{2\tilde{E}_g}{\hbar\omega}\right)^{1/2}\right]\times\exp\left[2\left\langle\frac{\tilde{E}_g}{\hbar\omega}+1\right\rangle\left(1-\frac{e^2E_0^2}{4m_{vc}^*\omega^2E_g}\right)\right]. \tag{11}$$

The photon multiplicity is given by

$$n=\left\langle\frac{\tilde{E}_g}{\hbar\omega}+1\right\rangle\,, \tag{12}$$

where the notation $\langle\cdots\rangle$ means the integer part of the argument and $\tilde{E}_g$ is the effective band gap in the $\vec{E}$ field:

$$\tilde{E}_g=E_g+\frac{e^2E_0^2}{4m_{vc}^*\omega^2}\,, \tag{13}$$

subject to the condition

$$\frac{e^2E_0^2}{4m_{vc}^*\omega^2E_g}\ll 1 \tag{14}$$

in Eq. (11) $\Phi(z)$ is the Dawson integral[14] defined by

$$\Phi(z)=\exp(-z^2)\int_0^z \exp(y^2)dy\,. \tag{15}$$

It is often stated[4,6-8] that in the case of two-photon absorption, Keldysh's formula predicts absorption coefficients that are too small compared with the results of conventional perturbation theory, and that the Keldysh formula is inaccurate or even invalid. Contrary to these beliefs, Mitra *et al.*[2] noted that for several direct-gap semiconductors, the Keldysh two-photon-absorption coefficients were larger than Braunstein's results and in closer agreement with the experimental data. In the following we show that the two-photon-absorption edges resulting from Keldysh's formula are larger than those of Braunstein's formula by a factor whose value is close to 1.7 for semiconductors and 3.4 for alkali halides. Comparison of the calculated two-photon-absorption coefficients, away from the edge, with experimental data will be discussed later in this paper. The applicability of the Keldysh formula for small photon multiplicity is covered in a separate communication.[15]

In view of the condition expressed by Eq. (14), we can assume $\tilde{E}_g = E_g$ and omit the term $e^2E_0^2/4m_{vc}^*\omega^2E_g$ in the exponential in Eq. (11). Close to the two-photon-absorption edge, $2\hbar\omega \simeq E_g$, thus, only the first term in the expansion of the exponential in the Dawson integral need be retained. We then obtain

$$\Phi\left[\left(2\left\langle\frac{E_g}{\hbar\omega}+1\right\rangle-\frac{2E_g}{\hbar\omega}\right)^{1/2}\right]\simeq\exp\left[-\left(2\left\langle\frac{E_g}{\hbar\omega}+1\right\rangle-\frac{2E_g}{\hbar\omega}\right)\right]\times\left(2\left\langle\frac{E_g}{\hbar\omega}+1\right\rangle-\frac{2E_g}{\hbar\omega}\right)^{1/2}. \tag{16}$$

Thus, close to the two-photon-absorption edge

$$W\simeq\left(\frac{2\omega}{9\pi}\right)\left(\frac{m_{vc}^*\omega}{\hbar}\right)^{3/2}\left(\frac{e^2E_0^2}{16m_{vc}^*\omega^2E_g}\right)^2(\exp(4))\times\left[\frac{2}{\hbar\omega}\left(2\hbar\omega-E_g\right)\right]^{1/2}. \tag{17}$$

The two-photon-absorption coefficient is given by

$$\beta=2W(2\hbar\omega/I^2)\,, \tag{18}$$

where I, the intensity of the radiation, is

$$I=\frac{cE_0^2\sqrt{\epsilon_\infty}}{8\pi}. \tag{19}$$

From Eqs. (17)–(19) the Keldysh two-photon-absorption edge is

$$\beta_k(2\hbar\omega\simeq E_g)\simeq\frac{\pi e^4\exp(4)}{9\epsilon_\infty c^2(2m_{vc}^*)^{1/2}}\,\frac{(2\hbar\omega-E_g)^{1/2}}{(\hbar\omega)^3}\,. \tag{20}$$

From Eqs. (10) and (20) we have

$$\frac{\beta_k}{\beta_B^{\mathrm{aa}}}\,(2\hbar\omega\simeq E_g)\simeq\frac{\exp(4)}{32}\left(\frac{m}{m_{vc}^*}\right)\frac{1}{(m/m_c-1)}\,. \tag{21}$$

For semiconductors $m_c \ll m$ and $m_{vc}^* \simeq m_c$. Equation (21) then gives

$$\beta_k \simeq 1.7\beta_B^{\mathrm{aa}}\,,$$

while for alkali halides $m_c \simeq 0.5m$, $m_{vc}^* \simeq m_c$, and we obtain

$$\beta_k \simeq 3.4\beta_B^{\mathrm{aa}}\,.$$

C. Basov's formula

Basov's formula corresponds to the special case of Braunstein's allowed-forbidden transitions where the intermediate states are taken to be the initial valence and final conduction bands themselves. This amounts to setting $\Delta E = E_g$ in the Braunstein result, expressed by Eq. (5). However, the final expression for the two-photon-absorption coefficient, obtained by Basov *et al.*,[4] differs considerably from the corresponding expression obtained from Eq. (5) when ΔE is assumed to be equal to E_g, both in numerical coefficients and in the effective-mass term. More surprisingly, Mitra *et al.*[2] noted that for several semiconductors the Basov formula predicted two-photon-absorption coefficients that were larger than the results of Braunstein's formula for allowed-allowed transitions, by about two orders of magnitude. These anomalous results arise from several apparent inaccuracies in Basov's formula.

Basov *et al.* used the following relations for the interaction Hamiltonian and vector potential:

$$H=\frac{e}{mc}\vec{A}\cdot\vec{p}\,, \tag{22}$$

$$A_0^2=\frac{8\pi cI}{(\epsilon_\infty)^{1/2}\omega^2}\,. \tag{23}$$

The proper expression for A_0^2 consistent with Eq. (22) is[16]

$$A_0^2=\frac{2\pi cI}{(\epsilon_\infty)^{1/2}\omega^2}\,. \tag{24}$$

Basov *et al.* also assumed the intraconduction-band Hamiltonian matrix elements to be

$$H_{cc}=\frac{eA}{m_cc}\hbar k\,, \tag{25}$$

with a similar expression for the valence-band matrix elements. The appropriate quantum-mechanical expression for these forbidden transitions is[9,11]

TABLE I. Values of parameters used for alkali halides. The effective masses appropriate for the valence bands are not available in the literature and were taken to be infinitely large.

Crystal	ϵ_∞ at 0.266 μm (Ref. 18)	E_g (eV)	ΔE (eV) (Ref. 18)	Conduction-band effective mass (in units of free electron mass) (Ref. 19)	Reduced effective mass (in units of free-electron mass) (Ref. 19)
NaCl	2.68	8.97 (Ref. 20)	6.36	0.6	⋯
KCl	2.468	8.9 (Ref. 19)	6.52	0.496	⋯
RbCl	2.484	8.5 (Ref. 19)	7.29	0.66	⋯
KBr	3.1	7.3 (Ref. 21)	6.0	0.48	⋯
KI	3.756 (3.11 at 0.355 μm)	6.3 (Ref. 21)	5.06	⋯	0.21
RbI	3.675 (2.99 at 0.355 μm)	6.37 (Ref. 21)	5.04	⋯	0.32

$$H_{vv} = H_{cc} = \left(\frac{eA}{mc}\right)\hbar k\,, \tag{26}$$

where m is the free electron mass. Finally, a correction factor of $\frac{1}{3}$ should be introduced in the Basov formula due to the averaging of the matrix elements, over all directions in $\vec{k}$ space. (Lee and Fan[17] earlier stated without explanation that a correction factor of $\frac{1}{48}$ should be applied to the Basov formula.) This factor obviously arises from the averaging of the matrix elements and the correction to the vector potential. When all these corrections are made, Basov's expression for the two-photon-absorption coefficient takes the form

$$\beta_{\text{Basov}}(\omega) = \frac{2^{13/2}\pi e^4 (m_{cv}^*)^{5/2} E_g f_{vc}}{3\epsilon_\infty c^2 m^3 (\hbar\omega)^5}(2\hbar\omega - E_g)^{3/2}\,, \tag{27}$$

with f_{vc} as given by Eq. (8).

COMPARISON OF THEORETICAL AND EXPERIMENTAL TWO-PHOTON-ABSORPTION COEFFICIENTS

Numerical calculations were performed for several direct-gap semiconductors and alkali halides using the formulas of Keldysh, Braunstein, and Basov. In using Braunstein's formulas for alkali halides, the intermediate states were taken to be the excitonic levels reported by Liu *et al.*,[18] instead of the higher conduction band as in the original Braunstein formulation. The intermediate levels in the forbidden gap play a more important role than the higher conduction band in determining the two-photon-absorption coefficients, due to their smaller energy denominators in Eqs. (4) and (5). In accordance with Eq. (26), the free-electron mass was used for $m_\dagger$ in the Braunstein formula for allowed-forbidden transitions. The values of the parameters used for alkali halides are listed in Table I. The parameters used for semiconductors are the same as those reported in Ref. 2. The calculated two-photon-absorption coefficients are listed along with a sampling of the available experimental data in Tables II and III. The experimental two-photon-absorption coefficients for semiconductors were collected from diverse sources (see Ref. 2 for information regarding the experiments), and they differ considerably from each other. In the case of alkali halides, even though all the data were obtained

TABLE II. Comparison of theoretical and experimental two-photon absorption coefficients (in units of 10^{-3} cm°MW) in several direct-gap semiconductors.

		Calculated					
		Allowed-allowed			Allowed-forbidden		
		Keldysh	Braunstein		Braunstein	Basov	Published experimental results (Ref. 2)
Crystal	Wavelength (μm)	(nonparabolic)	(parabolic)	(nonparabolic)	(parabolic)	(parabolic)	
CdS	0.694	6.1	2.19	3.26	0.04	0.608	30, 70, 660
ZnSe	0.694	6.4	2.03	2.96	0.018	0.297	40
GaAs	1.064	18.7	6.65	11.56	0.014	0.214	200, 300, 800, 5600, 9000
	1.318	22.6	10.08	13.99	0.01	0.221	33
InP	1.064	25.8	7.63	14.77	0.018	0.267	260, 180–260
CdTe	1.064[a]	23	7.37	12.21	0.023	0.459	200–300
InSb	10.64	2100	1185	1223	0.0006	0.127	256, 15 000, 16 000

[a] In Ref. 2 the wavelength was incorrectly reported as 1.318 μm.

TABLE III. Comparison of theoretical and experimental two-photon absorption coefficients (in units of 10^{-3} cm/MW) in several direct-gap alkali halides.

		Calculated					
		Allowed-allowed			Allowed-forbidden		Published
	Wavelength	Keldysh	Braunstein		Braunstein	Basov	experimental
Crystal	(μm)	(nonparabolic)	(parabolic)	(nonparabolic)	(parabolic)	(parabolic)	results (Ref. 18)
NaCl	0.266	0.226	0.216	0.227	0.035	0.038	3.5 ±25%
KCl	0.266	0.291	0.269	0.286	0.031	0.04	1.7 ±20% 2.7 ±30%
RbCl	0.266	0.344	0.054	0.061	0.063	0.16	1.02±15% 1.26±30%
KBr	0.266	0.502	0.114	0.160	0.105	0.265	2.0 ±30%
KI	0.266	0.802	0.205	0.362	0.037	0.1	3.75±20%
	0.355	1.02	0.646	.744	0.027	0.055	7.29±20%
RbI	0.266	0.655	0.163	0.284	0.073	0.187	2.49±20%
	0.355	0.848	0.514	.582	0.048	0.092	5.08±15%

from the same experiment, there are still noticeable variations in the results and large percentage errors. This makes the comparison between theory and experiment difficult. Though one cannot draw precise conclusions, one can note the following general trends: In the case of semiconductors, the Keldysh formula gives the best estimates for the two-photon-absorption coefficients. Braunstein's formula for allowed-allowed transitions, with nonparabolic bands gives results that are smaller than Keldysh's by a factor whose value is approximately two. On the other hand, the results of allowed-forbidden transitions are smaller by orders of magnitude. For alkali halides, the Braunstein's allowed-allowed formula results are in fair agreement with those of Keldysh's, and are also in closer agreement with the experimental data. Furthermore, results of Braunstein's formula for allowed-forbidden transitions are in better agreement with the predictions of Basov's formula. Both of these results are due to our use of the excitonic states for the intermediate states in Braunstein's formulas.

Thus, the Keldysh and the Braunstein's formulas for allowed-allowed transitions through excitonic states with nonparabolic energy bands give the best results. However, these results are still smaller than the cited experimental data, in many cases by as much as an order of magnitude. This is probably due to the simplifying assumptions made with regard to the electronic energy-band structures and oscillator strengths. Closer agreement between theory and experiment could probably be achieved by replacing the approximate saddle-point integration in the Keldysh derivation by an exact integration. It would also be instructive to modify the Braunstein formulas by considering the electronic transitions between perturbed eigenstates, whose wave functions are described by the Houston functions.

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