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Perturbation Theory and Two-Photon Magnetoabsorption in Semiconductors

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The previous perturbation-theoretical calculations of two-photon magnetointerband transitions in InSb-type semiconductors are found to have underestimated the strengths of the $\Delta n = +1, -3$ transitions by having omitted effective-mass terms. The corrected perturbation theory satisfactorily accounts for new measurements of the two-photon absorption carried out with circularly and linearly polarized radiation.

Two-photon magnetoabsorption in semiconductors has recently been a subject of interest.[1-3] In this Letter we report a correction to the perturbation-theory expressions of two-photon absorption which brings the theory into good agreement with recent experiments.[4] The allowed transitions are $\Delta n = +1$ for left-hand polarization σ_L, $\Delta n = -3$ for right-hand polarization σ_R, and $\Delta n = -1$ for transverse polarization σ.

Two approaches have been used to interpret multiphoton magnetoabsorption in semiconductors: (1) ordinary perturbation theory[5,6] and (2) tunneling theory.[7,8] From the expressions of Ref. 2, the strength of the $\Delta n = -3$ transition predicted by perturbation theory is three orders of magnitude smaller than that of the $\Delta n = 0$ transition predicted by tunneling theory. Yet, according to the tentative assignment[4] of an observed transition as being that of $b^+(0)b^c(0)$, we find that the $\Delta n = -3$ and $\Delta n = 0$ transitions are comparable in intensity. This paradox was removed when a recent report[9] reminded us of the fact that the intraband matrix elements of the momentum between adjacent Landau levels is proportional to m/m^*, the ratio of the free-electron mass to the effective mass. Since this ratio for the conduction band of InSb is $m/m_c^* \approx 60$ and the square of the matrix element occurs, one gains a factor of over 10^3 which is what is needed to make the calculated transition rates agree with the experimental values.

We follow the treatment of Ref. 5 or 6 to calculate the intraband matrix element of the momentum for circular polarization, namely $p_+ = (p_x + ip_y)/\sqrt{2}$ for σ_L and $p_- = (p_x - ip_y)/\sqrt{2}$ for σ_R. Considering the conduction band first, the wave func-

tion for the nth Landau level (leaving out spin indices) is

$$\psi_{c,n} = \varphi_n(k_x)|u_c\rangle + \sum_{j\neq c} \frac{\hbar}{m(E_c - E_j)} \sum_{\alpha} (\varphi_{n-\alpha}, k^{-\alpha}\varphi_n) \times (p_\alpha)_{jc} \varphi_{n-\alpha}(k_x)|u_j\rangle, \quad (1)$$

where u_c and u_j are the cell-periodic functions for bands c and j, and the index α takes the values +, −, 0 corresponding to p_+, p_-, p_z, respectively. $\varphi_n(k_x)$ is the usual harmonic oscillator function.

In computing the intraband matrix element $(\psi_{c,n+1}, p_+\psi_{c,n})$ it is essential to retain terms such as $\langle u_j|\varphi_n(k_x)p_+\varphi_n(k_x)|u_c\rangle$ when the effective algebraic mass $m_c{}^*$ is small.

For a simple band, one then obtains

$$(\psi_{c,n+1}, p_+\psi_{c,n}) = \hbar[s(n+1)]^{1/2} m/m_c{}^*, \quad (2)$$

where $s = |e|H/\hbar c$, H is the applied field, e is the free-electron charge, and c is the velocity of light in the vacuum.

For a hypothetical substance having a simple and full valence band v, and a simple conduction band c, the two-photon transition from $|v,n\rangle$ to $|c,n+1\rangle$ would go via two intermediate states:

$$\text{first } |v,n\rangle \rightarrow |c,n\rangle, \text{ then } |c,n\rangle \rightarrow |c,n+1\rangle, \quad (3a)$$

$$\text{first } |v,n+1\rangle \rightarrow |c,n+1\rangle, \text{ then } |v,n\rangle \rightarrow |v,n+1\rangle. \quad (3b)$$

The energy denominator for the transition in (3b) (two-electron process[10]) is opposite to that of the one-electron sequence:

$$|v,n\rangle \rightarrow |v,n+1\rangle, \text{ then } |v,n+1\rangle \rightarrow |c,n+1\rangle. \quad (3c)$$

However, the final state of (3b) differs from that of (3a) by the permutation of two electrons. One must then change again the sign of the contribution of process (3b) to the second-order matrix element. The net result is that one may just add the contributions of (3a) and (3c) calculated as one-electron transitions.[11] The energy denominators for (3a) and (3c) are, respectively, $-\hbar\omega + \hbar\omega_c$ and $\hbar\omega + \hbar|\omega_v|$, where ω_c and $|\omega_v|$ are the cyclotron frequencies in bands c and v.

In InSb, the valence band is not simple[12] and also there are three intermediate states instead of two. These are $b^c(n)$, $b^+(n+1)$, and $b^-(n+1)$ for the transition $b^+(n)$ to $b^c(n+1)$ in σ_L. For the transitions reported below, the second-order matrix elements of $p_\pm$ (for circular polarization) and $p_\perp$ (for transverse polarization) denoted by $p^{(2)}$ obtained by neglecting $(\hbar\omega_c/\hbar\omega)$ and $(\hbar\omega_v/\hbar\omega)$ in the energy denominators, are given in Table I. E_G is the energy gap, Δ the spin-orbit splitting, and P the interband momentum matrix element. For completeness, we give the transition rate $W_\pm$ when a left or right circularly polarized field $E_0(\hat{e}_x \cos\omega t \pm \hat{e}_y \sin\omega t)$ is incident on a unit

Table I. Second-order matrix elements $p^{(2)}$ for allowed two-photon magnetointerband transitions.

1 $\sigma_R(\Delta n=-3)$ Transitions	2 $\sigma_L(\Delta n=+1)$ Transitions	3 $\sigma(\Delta n=\pm 1,-3)$ Transitions	4 $p_+^{(2)}, p_-^{(2)}$ or $p_\perp^{(2)}$
$b^-(n)b^c(n-3)$		$b^-(n)b^c(n-3)$	$\frac{P}{\omega}\left[\frac{s(n-2)n}{4n-1}\right]^{\frac{1}{2}}\left[\frac{5}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
$a^-(n)a^c(n-3)$		$a^-(n)a^c(n-3)$	$\frac{P}{\omega}\left[\frac{s(n-2)n}{4n-3}\right]^{\frac{1}{2}}\left[\frac{7}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
		$b^+(n+1)b^c(n)$	$\frac{P}{\omega}\left[\frac{s}{3(4n+3)}\right]^{\frac{1}{2}}\left[\frac{38n+27}{3}\frac{P^2}{mE_G} + \frac{8n+6}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
		$a^+(n+1)a^c(n)$	$\frac{P}{\omega}\left[\frac{s}{3(4n+3)}\right]^{\frac{1}{2}}\left[\frac{38n+11}{3}\frac{P^2}{mE_G} + \frac{8n+2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
	$b^+(n)b^c(n+1)$ $n\geq 1$	$b^+(n)b^c(n+1)$ $n\geq 1$	$\frac{P}{\omega}\left[\frac{3s(n+1)n}{4n-1}\right]^{\frac{1}{2}}\left[\frac{7}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
	$b^+(0)b^c(1)$	$b^+(0)b^c(1)$	$\frac{P}{\omega}s^{\frac{1}{2}}\left[\frac{7}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
	$a^+(n)a^c(n+1)$ $n\geq 1$	$a^+(n)a^c(n+1)$ $n\geq 1$	$\frac{P}{\omega}\left[\frac{s(n+1)n}{3(4n-3)}\right]^{\frac{1}{2}}\left[\frac{5}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$
	$a^+(0)a^c(1)$	$a^+(0)a^c(1)$	$\frac{P}{\omega}\left[\frac{s}{3}\right]^{\frac{1}{2}}\left[\frac{5}{3}\frac{P^2}{mE_G} + \frac{2}{3}\frac{P^2}{m(E_G+\Delta)}\right]$

volume of material:

$$W_{\pm}=\left[\frac{8\pi}{c^2\epsilon}\left(\frac{e}{m\omega}\right)^4\frac{(2\mu)^{1/2}s}{\hbar^2}\times\sum_{n',n}\frac{|p_{\pm,n',n}^{(2)}|^2}{[2\hbar\omega-(E_{n',c}-E_{n,v})]^{1/2}}\right]I^2, \quad (4)$$

where $\mu^{-1}=m_c^{*-1}+|m_v^*|^{-1}$ and $I=c\epsilon^{1/2}E_0^2/8\pi$, where ϵ is the dielectric permittivity. For transverse polarization, a factor $\frac{1}{4}$ must be included in the square bracket (for a given I) and the matrix element of $p_{\perp}^{(2)}$ must be used. The term in the square bracket, hereafter called $A_{\pm}$ or $A_{\perp}$, can be evaluated using the matrix element $p_{\pm}^{(2)}$ or $p_{\perp}^{(2)}$ for the various allowed transitions given in Table I. The theoretical value of $A_{\pm}$ or $A_{\perp}$, will now be compared with A, the experimental value deduced from the study[4] of the absorption of CO_2 laser radiation by two-photon-excited free holes[13] in InSb. The absorption α_ν of high-intensity light at frequency ν in the sample of thickness d is given,[14] in terms of the same quantity A, by

$$\alpha_\nu=1-\frac{I(d)}{T^2I(0)}=1-[1+2q\tau_p AT^2I^2(0)d]^{-1/2}. \quad (5)$$

Here $I(d)$ is the transmitted light intensity, $I(0)$ is the light intensity at the surface of the sample, T is the power transmission coefficient for each surface of the sample, q is the cross section of free-hole absorption, and τ_p is the lifetime of excited holes. The absorption peaks of α_ν correspond to the resonances of A. Details of the measurements of α_ν for several frequencies were described elsewhere.[4] For the present purpose, in a transverse field configuration ($E\perp H$), Fig. 1(a) shows for n-type InSb and for a typical intensity of $I(0)=1.5\times10^{12}$ erg cm^{-2} sec^{-1} and $d=0.2$ cm, the absorption of CO_2 laser radiation as a function of the magnetic field for σ_R and σ_L and for incident frequency $\nu=1046.8$ cm^{-1}. The theoretical assignment of absorption peaks was obtained by using the quasi-Ge model with the band parameters given by Pidgeon and Groves.[15] Precise values of the magnetic field H at the absorption maxima are given in columns 1, 3, and 5 of Table II; columns 2, 4, and 6 list the identifications of the transitions. For circular polarization, with one exception at 50.9 kG (see Ref. c, Table II), all peaks of absorption occur for $\Delta n=-3$ in σ_R and $\Delta n=+1$ in σ_L. This is in agreement with our stated selection rules. In column

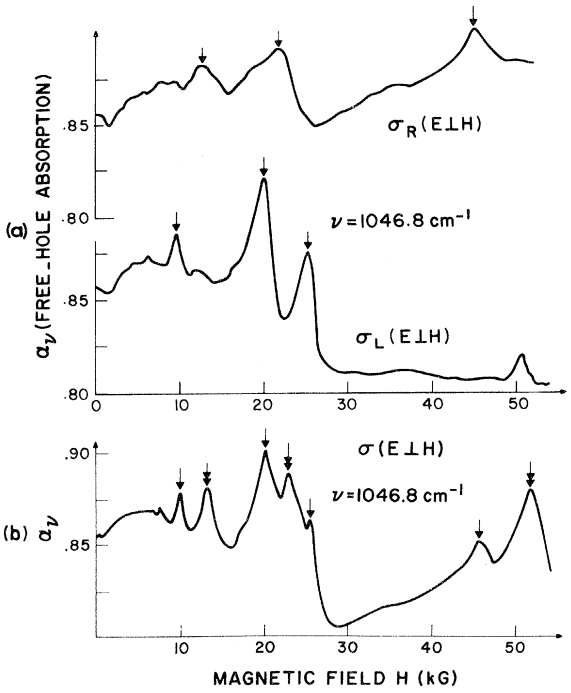

FIG. 1. (a), (b) Typical magnetic field dependence in n-type InSb of the absorption of CO_2 laser radiation by two-photon–excited free holes for right-hand polarization σ_R, left-hand polarization σ_L and for transverse polarization σ in a $E\perp H$ configuration for incident frequency $\nu=1046$ cm^{-1} (single arrows show the $\Delta n=+1, -3$ transitions and double arrows show the $\Delta n=-1$ transition).

5 we have listed only the σ transitions which are not observed in either σ_L or σ_R polarizations. These additional transitions in σ shown with double arrows in Fig. 1(b) are identified as $\Delta n=-1$ transitions.

To calculate $A_{\pm}$, $A_{\perp}$, we have assumed, for each resonant transition, a Lorentzian spread[16] in the energy of width $\hbar/\tau$ determined by the collision time τ. We have used the value $\tau=2\times10^{-13}$ sec used by Boswarva[17] in low-field experiments. We have also taken $P^2=0.39$ a.u. In calculating A from Eq. (5) we have used the values $q=3.2\times10^{-15}$ cm^2 (see Ref. 13) and $\tau_p=5.10^{-9}$ sec.[18] The predicted intensities of all the transitions is in good agreement with the experimental values which depend of course on the assumed values of the parameter q, τ_p, and τ.

In summary, it is thus possible to explain the entire spectrum, with the exception of the line at 50.9 kG in σ_L, with perturbation theory alone.

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Table II. Free-hole absorption maxima with their theoretical identification. Theoretical and experimental values of the parameter A of Eq. (5) are also given for each absorption peak.

1	2	3	4	5	6	7	8
$\sigma_R(E\perp H)$		$\sigma_L(E\perp H)$		$\sigma(E\perp H)$		A_+, A_- or $A\perp$	A_+, A_- or $A\perp$
Abs. Maxi (KG)[a]	Transition[b]	Abs. Maxi (KG)[a]	Transition[b]	Abs. Maxi (KG)[a]	Transition[b]	Theor (cm sec erg^{-2})	Exp. (cm sec erg^{-2})
		10.1	$b^+(1)b^c(2)$ $\Delta n = +1$			1.55	1.46
12.1	$a^-(5)a^c(2)$ $\Delta n = -3$					1.01	1.24
				12.8	$a^+(2)a^c(1)$ $\Delta n = -1$	2.11	1.62
		19.2	$b^+(0)b^c(1)$ $\Delta n = +1$			2.80	4.00
20.9	$a^-(4)a^c(1)$ $\Delta n = -3$					2.04	1.24
				22	$b^+(1)b^c(0)$ $\Delta n = -1$	1.85	1.88
		24.2	$a^+(0)a^c(1)$ $\Delta n = +1$			0.90	1.12
44.6	$b^-(3)b^c(0)$ $\Delta n = -3$					2.32	1.80
		50.9	c	50.9	$a^+(1)a^c(0)$ $\Delta n = -1$	2.10	1.62

[a]Reproducibility of these measurements is ±1 %. Each of the magnetic field values is the average of values found for the two directions of field sweep.

[b]For notation, see Ref. 12.

[c]We have no explanation for this absorption peak; it might be due to a residual polarization.

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