

Many-photon magneto-optic absorption in a narrow-gap semiconductor

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A theoretical investigation is made of the behavior of a semiconductor with a narrow band gap in parallel magnetic and periodic strong electric (for example, laser) fields. The Dirac equation is used in the two-band approximation and the solution of this equation is employed to calculate the probability of many-photon direct allowed interband transitions. This investigation has made it possible to remove a basic discrepancy between the theory and experiment: the theory predicts the appearance of magnetoabsorption maxima only for transitions involving an odd number of photons, whereas the experimental spectra show clearly such maxima in even-photon (specifically two-photon) absorption. A transformation of the Dirac equation, which should be of interest on its own account, is proposed. This transformation is analogous to the Foldy-Wouthuysen transformation and it makes it possible to identify the operators due to the electric field, some of which induce intraband nonstationary states with definite positive and negative quasienergies and can be allowed for exactly, whereas others induce interband many-photon transitions which are allowed for in a resonance approximation. The results are compared with the experimental data.

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A theoretical analysis is made of the behavior of a semiconductor with a narrow band gap subjected to a magnetic field parallel to a strong periodic electric (for example, laser) field. The Dirac equation is used in the two-band approximation^{1,2} and the solution of this equation is employed to calculate the probability of many-photon direct allowed interband transitions. This analysis has made it possible to remove a basic discrepancy between the theory and experiment, which has existed for some time.³⁻⁸ The discrepancy arises because the theory of magneto-optic absorption for parabolic bands,^{6,8} as well as the more general theory allowing for the band nonparabolicity in the absence of a magnetic field,⁹ give very different frequency dependences of the transition probabilities in the case of even and odd numbers of the absorbed photons. In particular, the appearance of magnetoabsorption maxima in the case of allowed interband transitions is predicted only in the case of an odd number of photons, whereas such maxima are clearly observed in two-photon absorption experiments.^{3,5}

In view of this situation we shall investigate many-photon magnetoabsorption in a narrow-gap semiconductor by a consistent analysis of the Dirac equation allowing for parallel magnetic and alternating electric fields. This analysis will be made within the framework of the theory of interband tunneling in an alternating electric field, put forward by Keldysh.¹⁰ Use will be made of a transformation which makes it possible to separate the operators due to the electric field, some of which induce intraband nonstationary states with a definite quasienergy and can be allowed for exactly, whereas others inducing interband many-photon transitions will be included in a resonance approximation. We shall show that in a strong magnetic field and for a sufficiently narrow band gap (of width of the same order as the separation between the Landau levels) the expression for the probability of many-photon transitions may have frequency singularities (maxima) for odd and even numbers of the absorbed photons. This makes it possible to

explain the oscillatory nature of the magnetoabsorption coefficient and of the even-photon photoconductivity in the experiments described in Refs. 3 and 5. A qualitative comparison of the theoretical and experimental results will also be given.

We shall consider the case when the conduction band and the highest valence band are isotropic, orbitally nondegenerate, and have extrema at the same point ($k=0$). The two band equation in the absence of an external field is then the Dirac equation in which the velocity of light is replaced with the parameter $s = (\mathcal{E}_g/2m)^{1/2}$ (\mathcal{E}_g is the band gap and m is the effective mass). In the presence of external fields described by the potentials $A = (0, Hx, 0)$ and $\varphi = -eF(t)z$, we can represent the wave function $\Psi(x, t)$ in the form

$$\Psi(x, t) = \exp\{i\hbar^{-1}[p_x(t)x + p_y y]\} \psi_{p_x}(x, t), \quad (1)$$

where

$$p_x(t) = p_{x0} - e \int_0^t F(t) dt. \quad (2)$$

The function $\psi_{p_x}(x, t)$ satisfies the equation

$$\hat{\mathcal{H}}(t) \psi_{p_x} = i\hbar \partial \psi_{p_x} / \partial t, \quad (3)$$

where

$$\hat{\mathcal{H}}(t) = s(\alpha P) + \gamma^0 m s^2, \quad (4)$$

$$P = \left(-i\hbar \frac{\partial}{\partial x}, p_y + \frac{e}{s} H' x, p_x(t) \right), \quad H' = \frac{s}{c} H; \quad (5)$$

α and γ^0 are the Dirac matrices in the standard representation.¹¹

We shall seek the solution of Eq. (2) in the form

$$\psi_{p_x} = \hat{U}^+ \hat{Q} \chi(t) u_n(\xi), \quad (6)$$

$$\hat{U}^+ = \exp \left\{ -\gamma^0 \arctg \frac{(\alpha P)}{m s^2 + E_n} \right\}, \quad (7)$$

$$\hat{Q} = \frac{1}{2\sqrt{2}} \left[(1 + \sigma_x + \sigma_y) + \frac{1}{(n+1)^{1/4}} \hat{a}^+ (\sigma_x + \sigma_y - 1) \right], \quad (8)$$

where $\sigma_{\pm} = \sigma_x \pm i\sigma_y$; σ_x , σ_y , and σ_z are the four-row Pauli matrices; $\chi(t)$ is a four-component function which de-

depends only on time; $E_n^2(t)$ are the eigenvalues of the operator $\hat{\mathcal{H}}^2(t)$:

$$E_n(t) = [(ms^2)^2 + s^2 p_z^2(t) + 2ehsH'(n+1)]^{1/2}; \quad (9)$$

$u_n(\xi)$ are the normalized functions of a harmonic oscillator;

$$\xi = \frac{1}{a_H}(x + a_H^2 \hbar^{-1} p_x), \quad a_H^2 = \frac{\hbar s}{eH'} = \frac{\hbar c}{eH}, \quad (10)$$

$$a^+ u_n = (n+1)^{1/2} u_{n+1}, \quad a u_n = n^{1/2} u_{n-1}.$$

The above transformation is analogous to the Foldy-Wouthuysen transformation and it makes it possible to separate the positive and negative states (this separation is exact for $F=0$).

The substitution of Eqs. (6)–(8) in Eq. (2) gives the following system of equations for the components of the function $\chi(t)$:

$$i\dot{\chi}_1 = \Omega_n^+(t)\chi_1 + R(t)\chi_2, \quad i\dot{\chi}_2 = -\Omega_n^+(t)\chi_1 + R^*(t)\chi_2, \quad (11)$$

$$i\dot{\chi}_3 = \Omega_n^-(t)\chi_3 - R^*(t)\chi_4, \quad i\dot{\chi}_4 = -\Omega_n^-(t)\chi_3 - R(t)\chi_4,$$

where

$$R(t) = \frac{\hbar\sqrt{2}}{a_H}(n+1)^{1/2} f_1(t) + i f_2(t), \quad (12)$$

$$\Omega_n^\pm(t) = \frac{1}{\hbar} E_n(t) \pm \frac{\hbar\sqrt{2}}{a_H}(n+1)^{1/2} f_1(t),$$

and

$$f_1(t) = \frac{s^2 p_z^2(t)}{2E_n(ms^2 + E_n)}, \quad f_2 = \frac{s(E_n + ms^2) \dot{p}_z - s p_z \dot{E}_n}{2E_n(ms^2 + E_n)}, \quad (13)$$

$$f_3(t) = \frac{s \dot{E}_n}{2E_n(ms^2 + E_n)}.$$

It is clear from the above formulas that, in the absence of an electric field, the system (11) splits into separate equations whose solutions correspond, in a static magnetic field, to stationary states of energy $E_n > 0$ (an electron in the conduction band) or of energy $-E_n < 0$ (an electron in the valence band). Application of an alternating electric field modifies the diagonal coefficients of the system $\Omega_n^\pm(t)$, describing the intraband motion and also gives rise to nondiagonal terms $\sim R(t)$, which mix the states from different bands and give rise to many-photon interband transitions.

We shall now consider a periodic electric field $F(t) = F_0 e_z \cos \omega t$. The field can induce transitions between such quasienergy band states which transform, in the limit $F_0 \rightarrow 0$, to stationary states of energies equal in magnitude and opposite in sign.

We shall replace the functions $\chi_i(t)$ with $v_i(t)$, defining them as follows:

$$\chi_{1,3} = v_{1,3} \exp\left\{ \mp i \int_0^t \Omega_n^+(t) dt \right\}, \quad (14)$$

$$\chi_{2,4} = v_{2,4} \exp\left\{ \mp i \int_0^t \Omega_n^-(t) dt \right\}.$$

Then, instead of the system (11), we have

$$i\dot{v}_1 = R(t) \exp\left\{ 2i \int_0^t \Omega_n^+(t) dt \right\} v_2, \quad i\dot{v}_2 = R^*(t) \exp\left\{ -2i \int_0^t \Omega_n^+(t) dt \right\} v_1; \quad (15)$$

$$i\dot{v}_3 = -R^*(t) \exp\left\{ 2i \int_0^t \Omega_n^-(t) dt \right\} v_4, \quad i\dot{v}_4 = -R(t) \exp\left\{ -2i \int_0^t \Omega_n^-(t) dt \right\} v_3. \quad (16)$$

In the case of a periodic field we can separate the periodic part from the exponential factors in Eq. (15):

$$\exp\left\{ 2i \int_0^t \Omega_n^+(t') dt' \right\} = \exp\left\{ i \frac{\mathcal{E}_n}{\hbar} t \right\} S_1(t), \quad (17)$$

$$S_1(t) = S_1\left(t + \frac{2\pi}{\omega}\right),$$

where

$$\mathcal{E}_n = \frac{\hbar\omega}{\pi} \int_{-\pi/\omega}^{+\pi/\omega} \Omega_n^\pm(t) dt = \frac{\omega}{\pi} \int_{-\pi/\omega}^{+\pi/\omega} E_n(t) dt, \quad (18)$$

$$\int_{-\pi/\omega}^{+\pi/\omega} f_1(t) dt = 0.$$

The quantity \mathcal{E}_n which appears in Eq. (17) represents the change in the electron quasienergy as a result of an interband transition or, which is equivalent, a change in the quasienergy of a newly created electron-hole pair.

Since $R(t)$ varies periodically at the same period as the field, we have

$$R(t) S_1(t) = \sum_{l=-\infty}^{+\infty} A_l e^{-il\omega t}, \quad (19)$$

$$A_l = \frac{\omega}{2\pi} \int_{-\pi/\omega}^{+\pi/\omega} e^{il\omega t} R(t) S_1(t) dt. \quad (20)$$

We shall now substitute Eq. (19) into Eq. (16) and average out the coefficients of the resultant equations over the field period $T = 2\pi/\omega$. We then find that the functions $v_1(t)$ and $v_4(t)$ corresponding to states with a quantum number n , satisfying the condition $\hbar\omega_l \equiv \mathcal{E}_n - l\hbar\omega \ll \hbar\omega$, are described by the system

$$i\dot{\bar{v}}_1 = A_l \exp(i\omega_l t) \bar{v}_4(t), \quad i\dot{\bar{v}}_4 = A_l^* \exp(i\omega_l t) \bar{v}_1(t). \quad (21)$$

Here, $\bar{v}_1(t)$ and $\bar{v}_4(t)$ are the functions $v_1(t)$ and $v_4(t)$ averaged at a moment t over one period of an external field:

$$\bar{v}_i(t) = \frac{1}{T} \int_{t-\pi/\omega}^{t+\pi/\omega} v_i(t) dt. \quad (22)$$

The system (21) is analogous to the corresponding equations of the two-level problem¹² and its solution subject to the initial conditions $\bar{v}_1(0) = 0$ and $\bar{v}_4(0) = 1/\sqrt{2}$ gives

$$\bar{v}_1(t) = -\frac{i}{\sqrt{2}} A_l \frac{\sin \lambda_l t}{\lambda_l} \exp\left\{ \frac{i}{2} \omega_l t \right\},$$

$$\bar{v}_4(t) = \frac{1}{\sqrt{2}} \exp\left\{ -i \frac{\omega_l}{2} t \right\} \left[\cos \lambda_l t + \frac{i\omega_l}{2\lambda_l} \sin \lambda_l t \right], \quad (23)$$

$$\lambda_l = (|A_l|^2 + 1/4\omega_l^2)^{1/2}, \quad l = 1, 2, \dots,$$

where the values of the quasienergy of a pair \mathcal{E}_n and of the coefficient A_l are given by Eqs. (18) and (20). Exactly the same procedure gives the functions $\bar{v}_2(t)$ and $\bar{v}_3(t)$. The expressions obtained for the functions $\bar{v}_i(t)$ are valid if $\lambda \ll \omega$ (resonance approximation).

It follows from the procedure of deriving the solu-

TABLE I.

	$2\hbar\omega_1$	$\hbar\omega_1+\hbar\omega_2$	$2\hbar\omega_2$		$2\hbar\omega_1$	$\hbar\omega_1+\hbar\omega_2$	$2\hbar\omega_2$
	$n=0$				$n=1$		
$H_{\text{theor}}^{\text{kOe}}$	39	46	54	$H_{\text{theor}}^{\text{kOe}}$	19	23	27
$H_{\text{exp}}^{\text{kOe}}$	34	44	53	$H_{\text{exp}}^{\text{kOe}}$	—	23	28

corresponding to various values of n at which peaks and steps appear in the optical absorption spectrum.

The results obtained make it possible to explain qualitatively the appearance of peaks in the experimental studies of the two-photon magneto-optical absorption by narrow-gap semiconductors InSb and PbTe (Refs. 3 and 5). In these experiments a crystal was subjected to a magnetic field and laser radiation of 1–3 kW power of fixed wavelengths $\lambda = 10.6$ and 9.6μ .

If the amplitude of the electric field in the laser wave does not exceed $F_0 \leq 10^5$ V/cm, the parameter γ^2 for a PbTe crystal in fields $H \approx 10^5$ Oe satisfies the condition $\gamma^2 > 15$. It should be noted that under these conditions the amplitudes of the two-photon absorption peaks exceed by over an order of magnitude the three-photon absorption peaks. At certain values of the magnitude field there are photoconductivity peaks interpreted as the maxima of the interband transition probability. Thus, these peaks can be regarded as induced by a magnetic field $H \propto p_H^2$ and corresponding to the absorption of the energies $2\hbar\omega_1$, $2\hbar\omega_2$, and $\hbar\omega_1 + \hbar\omega_2$. In the case of a PbTe crystal we can also provide a quantitative description based on Eq. (31) derived from the Dirac equation because the electron and hole bands of this crystal are simple and have extrema located at the point L on the c_3 axis in the Brillouin zone; moreover, these bands are characterized by approximately equal transverse and longitudinal masses.

Generalization of the expression for Δ in Eq. (31) to the case of various frequencies by the substitution $2\hbar\omega \rightarrow \hbar\omega_1 + \hbar\omega_2$ becomes possible because of the similarity of these frequencies in the experiments described in Refs. 3 and 5, and also because when the condition $\gamma \gg 1$ is satisfied, the quasienergies \mathcal{E}_n are practically independent of the frequency.

When a magnetic field H is directed along the c_4 axis,¹¹ the spectra are practically identical for the longitudinal and transverse polarizations of the laser radiation. We allowed for the experimental geometry and estimated, for three values of the absorbed energy, the magnetic fields H for various values of n corre-

sponding to the condition for a maximum $\Delta=0$ and we ignored the term $(2\gamma)^{-2}$ compared with unity. In these calculations we assumed $m_{\parallel} = 0.25m_0$, $m_{\perp} = 0.028m_0$, and $\mathcal{E}_g \approx 0.19$ eV, deduced from magneto-optical measurements.¹⁴

The results of a comparison of the calculated values of H and those observed experimentally⁹ is made in Table I, which shows that the agreement between the theory and experiment can be regarded as quite satisfactory.

The absence in the experimental spectra of a peak corresponding to the lowest magnetic field is probably associated with the low intensity of this peak, which—according to Eq. (31)—is of the order of H^2 .

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