Many-photon magnetooptic 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 twoband approximation and the solution of this equation is employed to calculate the probability of manyphoton 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.

PACS numbers: 78.20.Ls

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 magnetooptic 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 manyphoton 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 $(\mathbf{k}=0)$. 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 $\mathbf{A} = (0, Hx, 0)$ and $\varphi = -eF(t)z$, we can represent the wave function $\Psi(\mathbf{r}, t)$ in the form

$$\Psi(\mathbf{r},t) = \exp\left\{i\hbar^{-1}\left[p_{z}(t)z + p_{y}y\right]\right\}\psi_{p_{y}}(x,t),$$
(1)

where

$$p_{z}(t) = p_{z0} - e \int F(t) dt.$$
⁽²⁾

The function $\Psi_{py}(x, t)$ satisfies the equation

 $\hat{\mathscr{H}}(t)\psi_{p_y}=i\hbar\partial\psi_{p_y}/\partial t,$

where

$$\hat{\mathscr{H}}(t) = s(\alpha \mathbf{P}) + \gamma^{o} m s^{2}, \qquad (4)$$

$$\mathbf{P} = \left(-i\hbar \frac{\partial}{\partial x}, p_{v} + \frac{e}{s} H' x, p_{z}(t)\right), \quad H' = \frac{s}{c} H; \qquad (5)$$

 α and $\gamma^{\,0}$ are the Dirac matrices in the standard representation. 11

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

$$\psi_{p_{y}} = \hat{U}^{\dagger} \hat{Q} \chi(t) u_{n}(\xi), \qquad (6)$$

(3)

$$\hat{U}^{+} = \exp\left\{-\gamma^{\circ} \operatorname{arctg} \frac{(\boldsymbol{\alpha}\mathbf{P})}{ms^{2} + E_{n}}\right\},\tag{7}$$

$$\hat{Q} = \frac{1}{2\sqrt{2}} \left[(1 + \sigma_{*} + \sigma_{-}) + \frac{1}{(n+1)^{\nu_{h}}} \hat{a}^{*} (\sigma_{*} + \sigma_{+} - 1) \right],$$
(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-

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

$$E_n(t) = [(ms^2)^2 + s^2 p_z^2(t) + 2e\hbar s H'(n+1)]^{th};$$
(9)

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

$$\xi = \frac{1}{a_{H}} (x + a_{H}^{2} \hbar^{-1} p_{v}), \quad a_{H}^{3} = \frac{\hbar s}{eH'} = \frac{\hbar c}{eH},$$

$$a^{+} u_{n} = (n+1)^{*} u_{n+1}, \quad a u_{n} = n^{*} u_{n-1}.$$
(10)

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 $\chi(t)$:

$$i\chi_{1} = \Omega_{n}^{+}(t)\chi_{1} + R(t)\chi_{n}, \quad i\chi_{n} = -\Omega_{n}^{+}(t)\chi_{n} + R^{*}(t)\chi_{n},$$

$$i\chi_{n} = \Omega_{n}^{-}(t)\chi_{2} - R^{*}(t)\chi_{n}, \quad i\chi_{n} = -\Omega_{n}^{-}(t)\chi_{n} - R(t)\chi_{n},$$
(11)

where

$$R(t) = \frac{\hbar \sqrt{2}}{a_{\rm H}} (n+1)^{\frac{1}{2}} f_{s}(t) + i f_{2}(t),$$

$$\Omega_{n}^{\pm}(t) = \frac{1}{\hbar} E_{n}(t) \pm \frac{\hbar \sqrt{2}}{a_{\rm H}} (n+1)^{\frac{1}{2}} f_{1}(t),$$
(12)

and

$$f_{1}(t) = \frac{s^{2}\dot{p}_{s}(t)}{2E_{n}(ms^{2}+E_{n})}, \quad f_{2} = \frac{s(E_{n}+ms^{2})\dot{p}_{s}-sp_{s}\dot{E}_{n}}{2E_{n}(ms^{2}+E_{n})},$$

$$f_{3}(t) = \frac{s\dot{E}_{n}}{2E_{n}(ms^{2}+E_{n})}.$$
(13)

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 $\mathbf{F}(t) = F_0 \mathbf{e}_z \cos \omega t$. 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:

$$\chi_{1,t} = v_{1,t} \exp\left\{ \mp i \int_{0}^{t} \Omega_{n}^{+}(t) dt \right\},$$

$$\chi_{2,t} = v_{2,t} \exp\left\{ \mp i \int_{0}^{t} \Omega_{n}^{-}(t) dt \right\}.$$
(14)

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

$$tv_{i} = R(t) \exp\left\{2i \int_{0}^{t} \Omega_{n}^{+}(t) dt\right\} v_{i}, \quad tv_{i} = R^{*}(t) \exp\left\{-2i \int_{0}^{t} \Omega_{n}^{+}(t) dt\right\} v_{i};$$
(15)

 $iv_{s} = -R^{*}(t) \exp\left\{2i \int_{0}^{t} \Omega_{n}^{-}(t) dt\right\} v_{s}, \quad iv_{s} = -R(t) \exp\left\{-2i \int_{0}^{t} \Omega_{n}^{-}(t) dt\right\} v_{s}.$ (16)

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

$$\exp\left\{2i\int_{0}^{t}\Omega_{n}^{+}(t')dt'\right\} = \exp\left\{i\frac{\mathcal{B}_{n}}{\hbar}t\right\}S_{i}(t),$$

$$S_{i}(t) = S_{i}\left(t + \frac{2\pi}{\omega}\right),$$
(17)

where

j

$$\mathscr{E}_{n} = \frac{\hbar\omega}{\pi} \int_{-\pi/\bullet}^{+\pi/\bullet} \Omega_{n}^{\pm}(t) dt = \frac{\omega}{\pi} \int_{-\pi/\bullet}^{+\pi/\bullet} E_{n}(t) dt,$$

$$\int_{-\pi/\bullet}^{+\pi/\bullet} f_{1}(t) dt = 0.$$
(18)

The quantity \mathcal{S}_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

$$R(t)S_{i}(t) = \sum_{l=-\infty}^{+\infty} A_{l}e^{-il\omega t},$$
(19)

$$A_{i} = \frac{\omega}{2\pi} \int_{-\pi/\omega}^{\pi/\omega} e^{i\omega t} R(t) S_{i}(t) dt.$$
⁽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 $\hbar\omega_I \equiv \mathcal{E}_n$ $- l\hbar\omega \ll \hbar\omega$, are described by the system

$$\dot{\overline{v}}_{i} = A_{i} \exp(i\omega_{i}t) \overline{v}_{*}(t), \quad \dot{\overline{v}}_{*} = A_{i}^{*} \exp(i\omega_{i}t) \overline{v}_{i}(t).$$
(21)

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

$$\bar{v}_{i}(t) = \frac{1}{T} \int_{-\pi/2}^{4\pi/2} v_{i}(t) dt.$$
(22)

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

$$\overline{v}_{i}(t) = -\frac{i}{\sqrt{2}} A_{i} \frac{\sin \lambda_{i} t}{\lambda_{i}} \exp\left\{\frac{i}{2} \omega_{i} t\right\},$$

$$\overline{v}_{i}(t) = \frac{1}{\sqrt{2}} \exp\left\{-i\frac{\omega_{i}}{2} t\right\} \left[\cos \lambda_{i}(t) + \frac{i\omega_{i}}{2\lambda_{i}} \sin \lambda_{i} t\right],$$

$$\lambda_{i} = \left(|A_{i}|^{2} + \frac{i}{4} \omega_{i}^{2}\right)^{u}, \quad l = 1, 2, \dots,$$
(23)

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

It follows from the procedure of deriving the solu-

tions \overline{v}_i and their meaning that the probability of an interband electron transition is given by

$$w_{n}(p_{y}, p_{z0}, t) = |\bar{v}_{1}|^{2} + |\bar{v}_{2}|^{2} = |A_{1}|^{2} \frac{\sin^{2} \lambda_{1} t}{\lambda_{1}^{2}}, \qquad (24)$$

which is obtained bearing in mind that $|\overline{v}_1|^2 = |\overline{v}_2|^2$. As in the case of a two-level system, an interband transition resulting in the creation of an electron-hole pair is a periodic process of frequency λ_i and under resonance conditions $|\omega_i| \ll |A_i|$ the probability of this transition is $\sin^2 |A_i| t$ (Refs. 12 and 9). In the opposite case, it follows from the general results of quantum mechanics, that the transition probability (24) becomes

$$w_n(p_{\nu}, p_{\nu}, t) = 2\pi\hbar t |A_l|^2 \delta(\mathscr{E}_n - l\hbar\omega).$$
⁽²⁵⁾

The result obtained can be presented explicitly if we calculate the coefficient A_i . Since this coefficient is determined by the formulas (20), (17), (12), (13), (9), and (2), we shall introduce the variables $\sin\omega t'=u$, $\sin\omega t = u$, in all of them and represent the expression for A_i at $\omega_i \approx 0$ in the form

$$A_{i}(\omega) = -\frac{es^{s}F_{0}}{4\pi} \oint du \exp\left\{\frac{2i}{\hbar\omega}\int_{0}^{u} \frac{E_{n}(u')du'}{(1-u'^{2})^{\frac{1}{h}}}\right\} \Phi(u), \qquad (26)$$

where

$$\Phi(u) = \exp\left\{-\frac{ip_{B}s^{2}eF_{0}}{\omega}\int_{0}^{u}\frac{du'}{E_{n}(u')[ms^{2}+E_{n}(u')]}\right\} \times \left[\frac{p_{s}(u)(p_{B}-ip_{s}(u))}{E_{n}^{2}(u)[ms^{2}+E_{n}(u)]}+\frac{i}{s^{2}E_{n}(u)}\right].$$
(27)

Integration in Eq. (26) is carried out over a contour which envelops the points $u=\pm 1$. This integral can be calculated by the steepest-descent (saddle) method, assuming that the ratio below is a large parameter:

$$p_{\perp}s/\hbar\omega \gg 1;$$
 (28)

here,

$$p_{\perp}^{2} = (ms)^{2} + p_{H}^{2}, p_{H}^{2} = 2\hbar e H (n+1)/c.$$

As a result we find that the exponential function in Eq. (26) becomes rapidly varying, compared with the function Φ . The main contribution to the integral comes from the first term in the brackets of Eq. (27), which has poles at both saddle points, found from the condition $E_n(u)=0$. The contribution of the second term in Eq. (27) is small compared with the first when considered in relation to the parameter (28). In fact, integration reduces to bypassing singularities in an arc of $(\frac{4}{3})\pi$ size with a vanishingly small radius and the functions free of singularities should be taken at the saddle points allowing for bypassing of the branching points. Details of the calculation of such integrals can be found in Ref. 2 and also in Ref. 13.

The functions in the arguments of the exponential functions in Eqs. (27) and (26) and also the quasienergy & of Eq. (18) can be calculated exactly. In particular, the last two expressions are obtained in Ref. 10 and expressed in terms of elliptic integrals of the first and second kind. However, we shall simplify the results by assuming that

$$\gamma = \omega p_{\perp}/eF_0 \gg 1, \quad p_{z0}/p_{\perp} \ll 1, \tag{29}$$

where the first condition corresponds to the many-photon nature of transitions of interest to us and the second limits the frequencies ω to the most important region adjoining the edges of the Landau bands. Moreover, the argument of the exponential function in Eq. (26) is calculated allowing for the properties of the functions which determine it in the complex plane.⁸ Consequently, the contributions to the integral (26) from both saddle points located in the complex plane acquire respective phase factors which are $\exp(\pm i l\pi/2)$ for $\omega_1 \approx 0$.

Substituting the obtained expression for $|A_i|^2$ in Eq. (25) and summing over the quantum numbers n, p_y , and p_{z0} subject to the explicit form of the quasienergy (18) under the conditions of Eq. (29), which is

$$\mathscr{S}_{n}=2p_{\perp}s\left(1+\frac{1}{4\gamma^{2}}+\frac{1}{2}\frac{p_{z0}^{2}}{p_{\perp}^{2}}\right),$$
(30)

we obtain the final result for the total probability of an interband *l*-photon transition per unit time and per unit volume:

$$W^{(1)} = \frac{\omega^{2} \exp(2l)}{36\pi^{3} (ms)^{2} a_{H}^{2}} \sum_{n} \left(\frac{p_{\perp}}{s}\right)^{\frac{1}{2}} \frac{p_{\perp} + p_{H}}{p_{\perp} - p_{H}} (16\gamma^{2})^{-l} \\ \times \left[p_{H}^{2} \cos^{2} G(\Delta) + p_{\perp}^{2} \sin^{2} G(\Delta)\right] \Delta^{-\frac{1}{2}}, \\ G(\Delta) = l \frac{\pi}{2} + (2\gamma^{2} - \gamma) \left(\frac{\Delta}{p_{\perp} s}\right)^{\frac{1}{2}}, \quad \Delta = l\hbar\omega - 2p_{\perp} s \left(1 + \frac{1}{4\gamma^{2}}\right) \right\}$$
(31)
$$l = 1, 2, \dots$$

Equation (31) is valid if the inequalities (28) and (29) $(\gamma \gg 1, \Delta \ll p_{\perp}s)$ are satisfied.

According to Eq. (31), the many-photon magnetooptic absorption spectrum has singularities $\sim \Delta^{-1/2}$ as well as step-like dependences $\sim \Delta^{1/2}$ at frequencies ω satisfying the condition $\Delta = 0$. The positions and intensities of the absorption peaks and steps depend strongly on the parity of the number of the absorbed photons *l*. For odd *l* the absorption singularities are due to the term $\sim p_{\perp}^2$ and the steps due to the term $\sim p_{H}^2$ in Eq. (31), whereas in the case of even values of *l* the situation is reversed so that singularities are due to the term $\sim p_{H}^2$ and the steps are due to $\sim p_{\perp}^2$.

The presence of singularities in the even-photon absorption is a circumstance characteristic of narrowgap semiconductors, which appears when these semiconductors are described by the Dirac equation in the two-band approximation. The singularities are due to the term $\sim p_H$ in the brackets of Eq. (27). In the case of wide-gap semiconductors, which satisfy in particular $p_H \ll p_{\perp}$, this term is ignored so that we have to assume $p_H=0$ in Eq. (31) and the absorption singularities appear only for the odd-photon transitions.^{6,8}

We can generalize Eq. (31) to optically anisotropic crystals by making consistent substitutions²

$$H' \to \left[\sum_{i=1}^{a} H_{i}'^{2} \frac{m_{i}}{m}\right]^{V_{a}}, \quad F \to \left[\sum_{i=1}^{a} F_{i}^{2} \frac{m}{m_{i}}\right]^{V_{a}}, \quad m \to (m_{x}m_{y}m_{z})^{V_{a}}, \quad (32)$$

where m_i and H'_i are the components of the effective mass and of the reduced magnetic field along the principal axes.

If the frequency ω is assumed to be fixed, the condition $\Delta = 0$ defines a discrete series of magnetic fields *H*

	2ħw1		ħω₁ +ħω	2	27102		2ħ@1	<i>ħ</i> ω₁+ <i>ħ</i> ∞₂	2ħw2
	<i>n</i> =0					n=1			
$\frac{H_{\text{theor}}, \text{kOe}}{H_{\text{exp}}, \text{kOe}}$	39 34	l	46 44		54 53	H _{theor} ,kOe H _{exp} ,kOe	19 	23 23	27 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 magnetooptic 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 9.6 μ .

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 crytal 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 $2\hbar\omega$ $-\hbar\omega_1 + \hbar\omega_2$ becomes possible because of the similarity of these frequencies in the experiments described in Refs. 3 and 5, and also because when the condition γ $\gg 1$ 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 corresponding 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 magnetooptic 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 .

The authors are grateful to Yu.N. Demkov for a valuable discussion of the results.

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Translated by A. Tybulewicz