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Two-photon absorption in parallel electric and magnetic fields

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Abstract. The coefficient of two-photon absorption in the presence of a static external electric field $\boldsymbol{E}$ and a static external magnetic field $\boldsymbol{H}$ when applied parallel to each other is theoretically investigated. It is shown that in the case of direct-gap semiconductors the two-photon electroabsorption exhibits an oscillatory behaviour around the van Hove singularity M_0. This is due to discrete Stark levels at high electric fields ($E \geqslant 1000\ \mathrm{V\ cm^{-1}}$) and low magnetic fields ($H \leqslant 100\ \mathrm{kG}$).

1. Introduction

The problem of Bloch electrons in the presence of external electric and magnetic fields when applied perpendicular to each other has been theoretically investigated (Aronov 1963). It has been shown that, taking into account the $\boldsymbol{k}\cdot\boldsymbol{p}$ interaction between two energy bands, one obtains solutions with discrete eigenvalues for small E/H-ratios and a continuous spectrum for large E/H-ratios. In other words a sufficiently strong transverse electric field destroys the Landau orbital quantization. This has been confirmed experimentally (Reine *et al* 1967).

The theory of two-photon absorption (TPA) in crossed electric and magnetic fields has been theoretically investigated (Hassan and Moussa 1977). The energy levels and wavefunctions are obtained by solving the Schrödinger equation for small values of the ratio E/H (Zawadzki and Kawalski 1971). The general behaviour of the TPA is of magnetic type with a square-root dependence on the two-photon frequencies $\omega_1 + \omega_2$.

The presence of a magnetic field opens a real gap between the conduction and hole bands equal to $\mathscr{E}_g = \frac{1}{2}(\hbar\omega_c^c + \hbar\omega_c^h)$, both bands being quantized into Landau levels. When a weak transverse electric field is applied, the conduction Landau levels are shifted downwards by the amount $\frac{1}{2}m_c^* c^2E^2/H^2$. The hole Landau levels are shifted upwards because of the electric field by the amount $\frac{1}{2}m_h^* c^2E^2/H^2$. For high electric fields, Landau quantization is suppressed; the energy levels become closer to each other to form a continuum and the crossed-field configuration is no longer valid.

In the case of one-photon absorption the effect of parallel electric and magnetic fields has been investigated for the case of the direct electronic interband transitions (Ciobanu, 1965). Subsequently, the cases of both transverse and parallel electric and magnetic fields have been considered (Reine *et al* 1967, Weiler *et al* 1967).

Katana (1983, 1986) calculated multi-photon absorption in bulk semiconductors submitted to external electric and magnetic fields applied parallel to each other. Katana obtained agreement regarding the dependence of the two-photon absorption coefficient on

(i) the two-photon frequencies ω_1 and ω_2 and
(ii) the two-photon configurations $A_1 \| A_2$, $A_1 \perp A_2$ and $k_1 \| k_2$, $k_1 \perp k_2$.

It is shown that the TPA near each Landau level has an oscillatory behaviour because of the presence of discrete Stark levels in the presence of a strong electric field.

The purpose of the present paper is to develop the theory of TPA in parallel electric and magnetic fields in solids. The TPA has been numerically calculated in the case of GaP crystals. These results show an oscillatory behaviour owing to a subtle electric field. Landau quantization no longer occurs in this field limit where the absorption coefficient gives an Airy function profile.

In bulk semiconductors subjected to external parallel electric and magnetic fields, the Landau quantization rule could not easily be obtained by means of a computer.

2. Method of calculation

From second-order time-dependence perturbation theory the transition rate for an electron to perform a vertical transition from a valence state v to a conduction state c through an intermediate state l is (Hassan 1970)

$$\omega_{\mathrm{cv}}^{(2)} = \frac{2}{8\pi^3}\left(\frac{e}{mc}\right)^4 \int \mathrm{d}k \, \frac{1}{T}\left|\left(\frac{-\mathrm{i}}{\hbar}\right)\int_0^T \mathrm{d}t' \, M_{\mathrm{cl}}(t') \int_0^{t'} \mathrm{d}t'' \, M_{\mathrm{lv}}(t'')\right|^2 \tag{1}$$

in which M_{cl} and M_{lv} are the kinetic momentum matrix elements due to electric dipole interaction:

$$\begin{aligned} \mathbf{M} &= A_1 \cdot P + A_2 \cdot P + eE \cdot r \\ P &= p + (e/c)A \\ A_j &= \varepsilon_j A_{0j} \exp(\mathrm{i}k \cdot r - \mathrm{i}\omega_j t) \\ A &= \tfrac{1}{2}(r \times H) \end{aligned} \tag{2}$$

the magnetic vector potential and ε_j is a vector due to polarization of the photon.

In the presence of static external parallel electric and magnetic fields the eigenvalues $\mathscr{E}_n$ and eigenfunctions ψ_n are (Zak and Zawadzki 1966, Zawadzki and Kowalski 1971)

$$\mathscr{E}_n = \mathscr{E}_0 \pm \hbar^2 k_z^2/2m^* \pm (n + \tfrac{1}{2})\hbar\omega_{\mathrm{c}} - eEL_M^2 k_x^2 \mp \frac{2\pi eE\nu}{k_0}. \tag{3}$$

Here $L_{\mathrm{M}} = \sqrt{\hbar c/eH}$ is the magnetic radius and $\omega_{\mathrm{c}} = eH/m^*c$ is the Larmor frequency.

$$\psi_n(r) = F_n(r) u_{n0}(r) \tag{4}$$

in which u_{n0} is the periodic part of the Bloch states near the extremum of the energy band. Here n is a set of quantum numbers which characterize the states of the conduction electron and $F_n(r)$ is an envelope function (Zawadzki and Kowalski 1971):

$$F_n(r) = \left(\frac{eH}{\pi\hbar c}\right)^{1/4} \frac{1}{\sqrt{2^n n!}} \exp(\mathrm{i}(x \cdot k_x + z \cdot k_z) - \frac{eH}{2\hbar c}(y - y_0))$$

$$\times H_n\left(\sqrt{\frac{eH}{\hbar c}}(y - y_0)\right) \exp\left(-\frac{\mathrm{i}}{\hbar}\int_0^t \mathscr{E}_n(t')\,\mathrm{d}t'\right). \tag{5}$$

H_n is a Hermite polynomial and y_0 is the centre of oscillation:

$$y_0 = k_x L_M^2 - \frac{eE}{\hbar\omega_c} L_M^2.$$

From equations (2)–(5), equation (1) becomes

$$\omega_{\mathrm{cv}}^{(2)} = \frac{2}{8\pi^3}\left(\frac{e}{mc}\right)^4 |A_{01}A_{02}|^2 |\varepsilon_1 p_{\mathrm{cl}} \cdot \varepsilon_2 p_{\mathrm{lv}}|^2 \int \frac{\mathrm{d}k}{T}(1 + \rho_{12})$$

$$\times \left| \int_0^T \mathrm{d}t' \exp\left(\frac{\mathrm{i}}{\hbar}\int_0^{t'} \mathrm{d}t_1 (\mathscr{E}_c - \mathscr{E}_l - \hbar\omega_1\right)\right.$$

$$\left. \times \int_0^{t'} \mathrm{d}t'' \exp\left(-\frac{\mathrm{i}}{\hbar}\int_0^{t''} \mathrm{d}t_2 (\mathscr{E}_l - \mathscr{E}_v - \hbar\omega_2)\right)\right|^2$$

$$\times \left| H_n^*\left(\sqrt{\frac{eH}{\hbar c}}(y - y_0) H_{n'}\left(\sqrt{\frac{eH}{\hbar c}}(y - y_0')\right)\right|^2. \tag{6}$$

The permutation operator ρ_{12} changes ω_1 into ω_2 and vice versa because they are not geometrical and

$$|A_{0i}|^2 = 2\pi\hbar c^2 N_i/\omega_i n_i^2. \tag{7}$$

Here N_i is the photon density and n_i is the refractive index.

Near the critical point of type M_0, one can expand equation (3) for isotropic energy bands into the following form:

$$\begin{aligned} \mathscr{E}_v &= \hbar^2 k_z^2/2m_v^* - (n+\tfrac{1}{2})\hbar\omega_{\mathrm{cv}} - eEL_M k_x + 2\pi e E_\nu/k_0 \\ \mathscr{E}_c &= \mathscr{E}_{0g} + \hbar^2 k_z^2/2m_c^* + (n'+\tfrac{1}{2})\hbar\omega_{\mathrm{cc}} - eEL_M k_x - 2\pi e \bar{E}_\nu'/k_0 \\ \mathscr{E}_l &= \mathscr{E}_{0l} + \hbar^2 k_z^2/2m_l^* + (n''+\tfrac{1}{2})\hbar\omega_{\mathrm{cl}} - eEL_M k_x - 2\pi e \bar{E}_\nu''/k_0. \end{aligned} \tag{8}$$

$\mathscr{E}_{0g}$ and $\mathscr{E}_{0l}$ are the differences between the energies of the extremum of the bands and the top of the valence band. n, n', n'' are three different sets of quantum numbers due to Landau quantization, and ν, ν', ν'' are another three different sets of quantum numbers which characterize the Stark levels because of the presence of the electric field (Girlanda 1971, Callaway 1963).

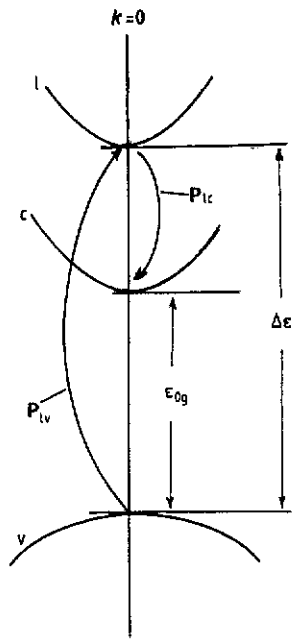

Figure 1. Schematic diagram for a three-band model containing a valence band v, a conduction band c and an intermediate band l, P_{cl} and P_{lv} indicate the coupling matrix elements.

In figure 1 we trace a three-band model containing a valence band v and a conduction band c with an intermediate band l. k_0 is the width of the Brillouin zone in the z direction.

3. Absorption coefficient

In bulk semiconductors subjected to static external electric and magnetic fields applied parallel to each other the TPA coefficient $K_{cv}^{(2)}(\omega_1, \omega_2, \boldsymbol{E}\|\boldsymbol{H})$ for the first photon ω_1 in the presence of the second ω_2 is given by

$$K_{cv}^{(2)}(\omega_1, \omega_2, \boldsymbol{E}\|\boldsymbol{H}) = [(\omega_1 + \omega_2)/\omega_1](n_1/cN_1)\omega_{cv}^{(2)}. \tag{9}$$

For electric dipole transitions between parabolic energy bands of opposite parities (allowed transitions), P_{cl}^2 and P_{lv}^2, the squares of the kinetic momentum matrix elements, are constant near the band edge $\mathscr{E}_g$. For transitions between bands that have the same parity—forbidden transitions, the squares of the kinetic momentum matrix elements can be assumed to be proportional to k^2 (Braunstein 1962). There are three different kinds of electric dipole transition possible depending upon the parities of the three spherical isotropic non-degenerate energy bands. This can be designated as

(i) allowed–allowed indirect two-photon transitions,
(ii) forbidden–allowed indirect two-photon transitions and
(iii) forbidden–forbidden indirect two-photon transitions.

For case (i) (allowed–allowed direct two-photon transitions) the two-photon kinetic momentum matrix elements that appear in equation (6) can be approximated as

$$\boldsymbol{\varepsilon} P_{ij} = \boldsymbol{\varepsilon}_1 p_{ik} \times \boldsymbol{\varepsilon}_2 p_{kj} \int_{\text{crystal volume}} \mathrm{d}r\, F^*_{n'}(r) F_n(r) \tag{6'}$$

in which

$$\boldsymbol{\varepsilon} p_{12} = \int_{\text{unit cell}} \mathrm{d}r\, u_{10}(r) \boldsymbol{\varepsilon} p u_{20}(r).$$

From equations (6)–(8), equation (9) becomes

$$\begin{aligned} K^{(2)}_{\mathrm{cv}}(\omega_1, \omega_2, \boldsymbol{E} \| \boldsymbol{H}) &= \frac{\omega_1 + \omega_2}{\omega_1} \frac{n_1}{CN_1} \left(\frac{e}{mc}\right)^4 \frac{4\pi\hbar^3 c^4 N_1 N_2}{n_2^2 \omega_2 n_1^2 \omega_1} |\boldsymbol{\varepsilon}_1 p_{\mathrm{cl}} \times \boldsymbol{\varepsilon}_2 p_{\mathrm{lv}}|^2 \\ &\times \left| H_n\left(\sqrt{\frac{eH}{\hbar c}}(y - y_0)\right) H_n\left(\sqrt{\frac{eH}{\hbar c}}(y - y_0)\right)\right|^2 \\ &\times \sum_{n,n'} \int_0^{k_z} \mathrm{d}k_z \left| \mathrm{Ai}\left(\frac{\mathscr{E}_{0\mathrm{g}} - \mathscr{E}_{0\mathrm{l}} - \hbar\omega_1 + (n + \frac{1}{2})(\hbar\omega_{\mathrm{cc}} - \hbar\omega_{\mathrm{cl}}) + (\alpha_{\mathrm{c}} - \alpha_{\mathrm{l}})k_z^2}{(eE)^{2/3} \alpha_{\mathrm{cl}}^{-1/3}}\right)\right. \\ &\times \left. \mathrm{Ai}\left(\frac{\mathscr{E}_{0\mathrm{l}} - \hbar\omega_2 + (n' + \frac{1}{2})(\hbar\omega_{\mathrm{cl}} + \hbar\omega_{\mathrm{cv}}) + (\alpha_{\mathrm{l}} + \alpha_{\mathrm{v}})k_z^2}{(eE)^{2/3} \alpha_{\mathrm{lv}}^{-1/3}}\right)\right|^2 \end{aligned} \tag{10}$$

in which $\alpha_i = \hbar^2/2m_i^*$ and $\mathrm{Ai}(x)$ denotes the Airy function integral (Jeffreys and Jeffreys 1972) given by

$$\mathrm{Ai}(x) = \int \frac{1}{2\pi \mathrm{i}} \exp(xt - \tfrac{1}{3}t^3)\, \mathrm{d}t.$$

In bulk semiconductors subjected to static parallel electric and magnetic fields, the dominant contribution to the direct TPA coefficient is due to a subtle electric field. We propose that the general behaviour of the absorption spectrum becomes of an electric type with the Airy function profile.

4. General discussion and conclusions

Equation (10) was numerically computed using a computer at Ain Shams University for the special case of GaP crystals. The direct energy gap $\mathscr{E}_{\mathrm{g}} = 2.22$ eV; we choose the electric field $E = 3.0 \times 10^4\ \mathrm{V\,cm^{-1}}$ and the magnetic field $H = 50.0$ kG (figure 2). We observe oscillatory behaviour when $\hbar\omega_1 + \hbar\omega_2 > \mathscr{E}_{\mathrm{g}}$, but before $\hbar\omega_1 + \hbar\omega_2 < \mathscr{E}_{\mathrm{g}}$ a tendency towards the exponential tail of the Franz–Keldysh effect (Franz 1958, Keldysh 1958) is seen.

In the present case of parallel electric and magnetic fields, we cannot easily trace the Landau levels because of the high electric field strength and low magnetic field.

The direct TPA near each Landau level has an oscillatory behaviour because of the presence of discrete Stark levels with energy separation $\Delta\mathscr{E} = 2\pi eE/k_0$ from which we can experimentally obtain the width k_0 of the Brillouin zone in the direction of the electric field, in the case of parallel polarization $\boldsymbol{A}_1 \| \boldsymbol{A}_2$. For the transverse polarization $\boldsymbol{A}_1 \perp \boldsymbol{A}_2$, additional absorption lines must occur (Katana 1983, 1986).

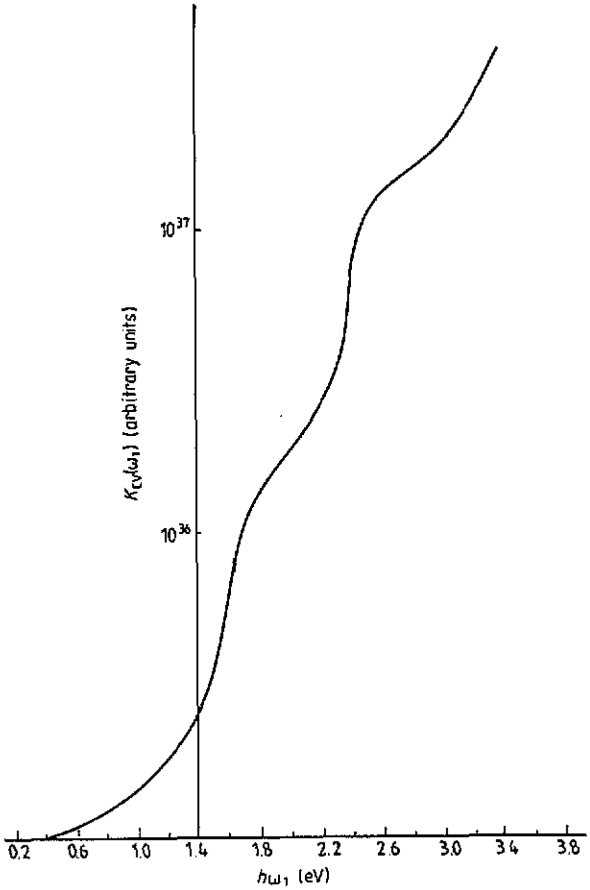

Figure 2. Absorption coefficient $K_{cv}(\omega_1)$ due to two-photon transitions versus $\hbar\omega_1$ for an electric field $E = 3.0 \times 10^4$ V cm^{-1} and a magnetic field $H = 50.0$ kG for GaP (equation (10)).

Figure 2 shows that direct TPA in a strong electric field exhibits an oscillatory behaviour in the presence of a weak magnetic field in agreement with one-photon experimental results (Kovalerskaya *et al* 1976).

We present the asymptotic expression for equation (10) obtained with the help of the asymptotic formula for the Airy function in the absence of a magnetic field, i.e. $H = 0$ (Fok 1946):

$$\mathrm{Ai}(x) = \begin{cases} \exp(-\frac{2}{3}x^{3/2}) \\ x^{3/2}[1 - \cos(-\frac{4}{3}x^{3/2})] \end{cases} \qquad \text{for} \begin{cases} x > 0 \\ x < 0. \end{cases} \tag{11}$$

Using equation (11), one can find the profile of the two-photon electroabsorption after the direct energy gap:

$$\hbar\omega_1 + \hbar\omega_2 \gtrless E_{0g}.$$

Thus knowing that, at $x > 0$, $\mathrm{Ai}(x) \simeq \exp(-\frac{4}{3}x^{3/2})$ we derive the two-photon Franz–Keldysh effect.

Finally, at $E = 0$ and $H = 0$ we obtain the well known result of Braunstein (1962) for the direct allowed–allowed two-photon transitions:

$$K_{\mathrm{cv}}^{(2)}(\omega_1, \omega_2) = \frac{(2)^{7/2}\pi n_1 e^4 N_2 |\boldsymbol{\varepsilon}_1 p_{\mathrm{cl}} \cdot \boldsymbol{\varepsilon}_2 p_{\mathrm{lv}}|^2}{cm^{3/2}(\alpha_{\mathrm{c}} + \alpha_{\mathrm{v}})^{3/2}\hbar^2\omega_1\omega_2} \times \left[\!\left[\frac{(\hbar\omega_1 + \hbar\omega_2 - \mathscr{E}_{\mathrm{g}})^{1/2}}{\{\Delta\mathscr{E} + [(\alpha_1 + \alpha_{\mathrm{v}})/(\alpha_{\mathrm{c}} + \alpha_{\mathrm{v}})](\hbar\omega_1 + \hbar\omega_2 - \mathscr{E}_{\mathrm{g}}) - \hbar\omega_1\}^2} + \frac{(\hbar\omega_1 + \hbar\omega_2 - \mathscr{E}_{\mathrm{g}})^{1/2}}{\{\Delta\mathscr{E} + [(\alpha_1 + \alpha_{\mathrm{v}})/(\alpha_{\mathrm{c}} + \alpha_{\mathrm{v}})](\hbar\omega_1 + \hbar\omega_2 - \mathscr{E}_{\mathrm{g}}) - \hbar\omega_2\}^2} \right]\!\right]. \tag{12}$$

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