

THE GREEN'S-FUNCTION METHOD IN THE THEORY OF THE OPTICAL PROPERTIES OF SEMICONDUCTORS

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The topics considered are the application of the Green's-function method to the theory of the dispersion and absorption of electromagnetic radiation in semiconductors, and also to the theory of the shape of resonance lines. Expressions in closed form are derived for the optical constants and the line shape, by finding the energy spectrum of the photons. The method developed is applied to the theory of the line shape of magneto-optical resonance and to the theory of the absorption of radiation by free carriers. It is shown that in a quantizing magnetic field resonance oscillations can occur in scattering both by optical phonons and by acoustical phonons.

IN the treatment of the absorption and dispersion of electromagnetic radiation in matter by the methods of quantum mechanics the usual starting point is the quantum-mechanical expression for the current density, from which one determines the real and imaginary parts of the electric conductivity, after which one connects these quantities with the absorption and dispersion. Here it is often unclear how some complicated mechanisms of absorption and dispersion can be reduced to an electric conductivity. It is more convenient to use the method of Green's functions in the second-quantization representation.^[1,2] The photon spectrum E is determined by the poles of the photon Green's function; the imaginary part Γ of the spectrum gives the damping of the photon—that is, the absorption of the electromagnetic radiation—and the real part E gives the dispersion:

$$\text{Re } E / |\mathbf{k}| = c / n, \tag{1}$$

where \mathbf{k} is the momentum of the photon, c is the speed of light in vacuum, and n is the index of refraction.

In our opinion an advantage of this method is that it allows us by means of a unified scheme to get a simple determination of the line shape in the case of resonance absorption, because the damping of the quasiparticles which absorb the photon is also included in the imaginary part of the spectrum according to a definite rule. The ordinary absorption coefficient per unit length, κ , is connected with the imaginary part of the spectrum by the relation

$$\kappa = \Gamma / v, \tag{2}$$

where v is the speed of light in the substance.

1. When anharmonicity is not taken into account, the Hamiltonian of the system of photons, phonons, and current carriers is of the form

$$H = H_p + H_f + H_e + H_{pe} + H_{fe} + H_{pf}. \tag{3}$$

Here H_p is the Hamiltonian of the photons:

$$H_p = \sum_{\mathbf{k}} \omega_{\mathbf{k}} (\beta_{\mathbf{k}}^+ \beta_{\mathbf{k}} + 1/2), \tag{4}$$

and $\omega_{\mathbf{k}} = |\mathbf{k}|$ is the frequency of the free photon; $\beta_{\mathbf{k}}^+$, $\beta_{\mathbf{k}}$ are photon creation and annihilation operators; $\hbar = c = 1$. The expression (4) corresponds to the Coulomb gauge for the field and to the expansion

$$\mathbf{A}(\mathbf{x}) = \sum_{\mathbf{k}} \{ \mathbf{e}_{\mathbf{k}} F_{\mathbf{k}} \beta_{\mathbf{k}} e^{i\mathbf{k}\mathbf{x}} + \text{Herm. adj.} \}, \tag{5}$$

for the vector potential, where $\mathbf{e}_{\mathbf{k}}$ is the polarization vector of a photon, the volume of the crystal is taken as unity, and $F_{\mathbf{k}} = (2\pi/\omega_{\mathbf{k}})^{1/2}$.

We write the Hamiltonian of the phonons by introducing creation operators $c_{\mathbf{q}}^+$ and annihilation operators $c_{\mathbf{q}}$ of phonons of branch ν with quasimomentum \mathbf{q} and frequency $\Omega_{\mathbf{q}\nu}$:

$$H_f = \sum_{\mathbf{q}\nu} \Omega_{\mathbf{q}\nu} (c_{\mathbf{q}}^+ c_{\mathbf{q}} + 1/2). \tag{6}$$

Introducing creation operators $a_{\mathbf{p}}^+$ and annihilation operators $a_{\mathbf{p}}$ of electrons in the conduction band with quasimomentum \mathbf{p} and energy $\epsilon_{\mathbf{p}}^c$, and also creation operators $b_{\mathbf{p}}^+$ and annihilation operators $b_{\mathbf{p}}$ of holes in the valence band with quasimomentum \mathbf{p} and energy $\epsilon_{\mathbf{p}}^v$, we write the

Hamiltonian of the current carriers in the form

$$H_e = \sum_{\mathbf{p}} (\varepsilon_{\mathbf{p}}^c a_{\mathbf{p}}^+ a_{\mathbf{p}} + \varepsilon_{\mathbf{p}}^v b_{\mathbf{p}}^+ b_{\mathbf{p}}). \quad (7)$$

In the coordinate representation the second-quantized operator of the carriers is

$$\psi(x) = \sum_{\mathbf{p}} \{b_{\mathbf{p}}^+ \varphi_{v\mathbf{p}} + a_{\mathbf{p}} \varphi_{c\mathbf{p}}\}. \quad (8)$$

For the calculation of the matrix elements we use the functions of Luttinger and Kohn^[3]:

$$\varphi_{m\mathbf{p}} = \left[U_{m0} + \sum_{\mathbf{i}} U_{i0} (\mathbf{p}_{mi} \nabla) / \omega_{mi} \right] F_{\mathbf{p}}(x), \quad (9)$$

where U_{i0} are the Bloch functions at the bottom of the bands, and $F_{\mathbf{p}}(x)$ is the solution of the Schrödinger equation with effective mass in an external field. We have

$$H_{pe} = \sum_{\mathbf{p}_1, \mathbf{p}_2, \mathbf{k}} \mathbf{e}_{\mathbf{k}} F_{\mathbf{k}} \{ \mathbf{J}_c^c a_{\mathbf{p}_1}^+ \beta_{\mathbf{k}} a_{\mathbf{p}_2} + \mathbf{J}_c^v b_{\mathbf{p}_1} \beta_{\mathbf{k}} a_{\mathbf{p}_2} + \mathbf{J}_v^c a_{\mathbf{p}_1}^+ \beta_{\mathbf{k}} b_{\mathbf{p}_2}^+ + \mathbf{J}_v^v b_{\mathbf{p}_1} \beta_{\mathbf{k}} b_{\mathbf{p}_2}^+ \} + \text{Herm. adj.}, \quad (10)$$

where

$$\mathbf{J}_m^n(\mathbf{p}_1; \mathbf{p}_2 \mathbf{k}) = \int d^3x \varphi_{m\mathbf{p}_1}^* \hat{\mathbf{J}} e^{i\mathbf{k}\mathbf{x}} \varphi_{n\mathbf{p}_2}, \quad (11)$$

$\hat{\mathbf{J}}$ is the current operator ($m, n = c, v$), and

$$H_{je} = \sum_{\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}} \mathbf{e}_{\mathbf{q}} \mathbf{G}_{\mathbf{q}} \{ i_c^c a_{\mathbf{p}_1}^+ c_{\mathbf{q}} a_{\mathbf{p}_2} + i_c^v b_{\mathbf{p}_1} c_{\mathbf{q}} a_{\mathbf{p}_2} + i_v^c a_{\mathbf{p}_1}^+ c_{\mathbf{q}} b_{\mathbf{p}_2}^+ + i_v^v b_{\mathbf{p}_1} c_{\mathbf{q}} b_{\mathbf{p}_2}^+ \} + \text{Herm. adj.}, \quad (12)$$

where

$$i_m^n(\mathbf{p}_1; \mathbf{p}_2 \mathbf{q}) = \int d^3x \varphi_{m\mathbf{p}_1}^* e^{i\mathbf{q}\mathbf{x}} \varphi_{n\mathbf{p}_2}, \quad (13)$$

$\mathbf{G}_{\mathbf{q}\nu}$ is the interaction constant, and $\mathbf{e}_{\mathbf{q}\nu}$ is the polarization vector of a phonon of branch ν .

In forming the Hamiltonian for the interaction of the photons with the phonons we use the following arguments. The vibrations of the atoms at the lattice sites give rise to a current (ionic polarization current in ionic crystals, polarization current of the electron shells in homopolar crystals). This current interacts with the electromagnetic field. For example, for the displacement vector in an ionic crystal we have from^[1,2] the expression

$$\mathbf{u}(x) = \sum_{\mathbf{q}\nu} [2d\Omega_{\mathbf{q}\nu}]^{-1/2} \{ \mathbf{e}_{\mathbf{q}\nu} c_{\mathbf{q}}(t) e^{i\mathbf{q}\mathbf{x}} + \text{Herm. adj.} \}. \quad (14)$$

Using the fact that

$$\partial_t c_{\mathbf{q}}(t) = -i\Omega_{\mathbf{q}\nu} c_{\mathbf{q}}(t),$$

we get for the current of the lattice vibrations

$$\mathbf{J}_{\mathbf{p}} = Ze\dot{\mathbf{u}} = \sum_{\mathbf{q}\nu} \mathbf{e}_{\mathbf{q}\nu} g_{\mathbf{q}\nu} c_{\mathbf{q}}(t) e^{i\mathbf{q}\mathbf{x}} + \text{Herm. adj.} \quad (15)$$

The form of $g_{\mathbf{q}\nu}$ depends on the nature of the crystal.

The Hamiltonian for the photon-phonon interaction will be of the form

$$H_{pt} = \int d^3x \mathbf{J}_{\mathbf{p}} \mathbf{A} = \sum_{\mathbf{k}\mathbf{q}\nu} (\mathbf{e}_{\mathbf{k}} \mathbf{e}_{\mathbf{q}\nu}) F_{\mathbf{k}} g_{\mathbf{q}\nu}^* \beta_{\mathbf{k}} c_{\mathbf{q}}^+ \delta(\mathbf{k} - \mathbf{q}) + \text{Herm. adj.} \quad (16)$$

2. For the determination of the energy spectrum we introduce the retarded Green's function of the photon:

$$D_{\mathbf{k}\mathbf{k}'}(t) = \theta(t) \text{Sp} \{ e^{-\beta H} \beta_{\mathbf{k}'}^+(t) \beta_{\mathbf{k}}(0) \} \equiv \theta(t) \langle \beta_{\mathbf{k}'}^+ | \beta_{\mathbf{k}} \rangle, \quad \beta_{\mathbf{k}'}^+(t) = e^{iHt} \beta_{\mathbf{k}'}^+(0) e^{-iHt}, \quad \theta(t) = \begin{cases} 1, & t > 0 \\ 0, & t < 0 \end{cases} \quad (17)$$

and its Fourier transform with respect to time:

$$D_{\mathbf{k}\mathbf{k}'}(E) = \int_{-\infty}^{+\infty} dt e^{iEt} D_{\mathbf{k}\mathbf{k}'}(t). \quad (18)$$

The poles of $D(E)$ in the lower half-plane of the variable E determine the spectrum of the photons.

The equation for $D(t)$ can be obtained by differentiating (17) with respect to time:

$$i \frac{\partial}{\partial t} D_{\mathbf{k}\mathbf{k}'}(t) = i\delta(t) \langle \beta_{\mathbf{k}'}^+ | \beta_{\mathbf{k}} \rangle + \theta(t) \langle [\beta_{\mathbf{k}'}^+ H] | \beta_{\mathbf{k}} \rangle. \quad (19)$$

The right member of Eq. (19) contains new Green's functions which involve operators for phonons, electrons, and holes. By writing analogous equations for these functions, we get an infinite system of coupled equations. To the lowest order in the interaction constants we can truncate this system, using the approximations

$$\langle a_{\mathbf{p}_1}^+ \beta_{\mathbf{k}'}^+ a_{\mathbf{p}_2} | \beta_{\mathbf{k}'} \rangle = \delta_{\mathbf{p}_1, \mathbf{p}_2} n_{\mathbf{p}_1}^c \langle \beta_{\mathbf{k}'}^+ | \beta_{\mathbf{k}'} \rangle \quad (20)$$

and so on, where $n_{\mathbf{p}}^c$ for the electrons and $n_{\mathbf{p}}^v$ for the holes are given by the Boltzmann distribution.

From the finite system of equations obtained in this way we find the real part

$$\begin{aligned} \text{Re } E = & -\omega_k + \sum_{\nu} \frac{(\mathbf{e}_{\mathbf{k}} \mathbf{e}_{\mathbf{k}\nu}) |g_{\mathbf{k}\nu}|^2 |F_{\mathbf{k}}|^2}{\Omega_{\mathbf{k}\nu} - \omega_k} \\ & + \sum_{\mathbf{p}_1, \mathbf{p}_2} |F_{\mathbf{k}}|^2 \left\{ \frac{|\mathbf{e}_{\mathbf{k}} \mathbf{J}_c^c|^2 (n_{\mathbf{p}_2}^c - n_{\mathbf{p}_1}^c)}{\varepsilon_{\mathbf{p}_1}^c - \varepsilon_{\mathbf{p}_2}^c - \omega_k} \right. \\ & + \frac{|\mathbf{e}_{\mathbf{k}} \mathbf{J}_c^v|^2 (1 - n_{\mathbf{p}_2}^v - n_{\mathbf{p}_1}^c)}{\varepsilon_{\mathbf{p}_1}^c + \varepsilon_{\mathbf{p}_2}^v - \omega_k} + \frac{|\mathbf{e}_{\mathbf{k}} \mathbf{J}_v^c|^2 (1 - n_{\mathbf{p}_1}^v - n_{\mathbf{p}_2}^c)}{\varepsilon_{\mathbf{p}_1}^v + \varepsilon_{\mathbf{p}_2}^c + \omega_k} \\ & \left. + \frac{|\mathbf{e}_{\mathbf{k}} \mathbf{J}_v^v|^2 (n_{\mathbf{p}_1}^v - n_{\mathbf{p}_2}^v)}{\varepsilon_{\mathbf{p}_2}^v - \varepsilon_{\mathbf{p}_1}^v - \omega_k} \right\} \quad (21) \end{aligned}$$

and the imaginary part

$$\begin{aligned} \Gamma = & \pi \sum_{\nu} (\mathbf{e}_{\mathbf{k}} \mathbf{e}_{\mathbf{k}\nu})^2 |g_{\mathbf{k}\nu}|^2 |F_{\mathbf{k}}|^2 \delta(\Omega_{\mathbf{k}\nu} - \omega_k) \\ & + \pi \sum_{\mathbf{p}_1, \mathbf{p}_2} |F_{\mathbf{k}}|^2 \{ |\mathbf{e}_{\mathbf{k}} \mathbf{J}_c^c|^2 (n_{\mathbf{p}_2}^c - n_{\mathbf{p}_1}^c) \delta(\varepsilon_{\mathbf{p}_1}^c - \varepsilon_{\mathbf{p}_2}^c - \omega_k) \\ & + |\mathbf{e}_{\mathbf{k}} \mathbf{J}_c^v|^2 (1 - n_{\mathbf{p}_2}^v - n_{\mathbf{p}_1}^c) \delta(\varepsilon_{\mathbf{p}_1}^c + \varepsilon_{\mathbf{p}_2}^v - \omega_k) \\ & + |\mathbf{e}_{\mathbf{k}} \mathbf{J}_v^c|^2 (n_{\mathbf{p}_1}^v - n_{\mathbf{p}_2}^v) \delta(\varepsilon_{\mathbf{p}_2}^v - \varepsilon_{\mathbf{p}_1}^v - \omega_k) \} \quad (22) \end{aligned}$$

of the spectrum.

The first sums in the expressions (21) and (22) describe the dispersion and resonance absorption caused by the interaction of photon and phonon. The case of absorption has been considered earlier,^[4] and there the expression for the line width caused by anharmonicity was also derived. The first and last terms in the second sums describe the interaction of photons with free carriers. In the absence of external fields these terms do not lead to absorption owing to the impossibility of simultaneously satisfying the conservation laws for energy and momentum. When there is an external magnetic field these terms describe the absorption associated with the cyclotron resonance. The remaining terms describe interband transitions. With an external magnetic field present they lead to expressions for the Faraday and Voigt dispersion magneto-optical effects,^[5] and also for magnetooscillatory effects.^[6,7]

3. Let us now consider the line width of the magneto-optical absorption owing to damping of the carrier states because of scattering by phonons. For this we must include in the chain of equations Green's functions containing operators of the electron-hole and phonon fields, and make the truncation in the next order of perturbation theory. For example,

$$\langle a_{\mathbf{p}_1}^+ c_{\mathbf{q}_1}^+ c_{\mathbf{q}_2} b_{\mathbf{p}_2}^+ | \beta_{\mathbf{k}'} \rangle = \delta_{\mathbf{q}_1 \mathbf{q}_2} N_{\mathbf{q}_1} \langle a_{\mathbf{p}_1}^+ b_{\mathbf{p}_2}^+ | \beta_{\mathbf{k}'} \rangle \quad (23)$$

and so on, where

$$N_{\mathbf{q}} = [e^{\beta \Omega_{\mathbf{q}}} + 1]^{-1}.$$

When we find the Green's function that appears in the right member of (23) and substitute it in (19), the result is that the third sum in (21) is replaced by

$$\Gamma = \sum_{\mathbf{p}_1 \mathbf{p}_2} \frac{|F_{\mathbf{k}}|^2 |\mathbf{e}_{\mathbf{k}} J_c^v|^2 (1 - n_{\mathbf{p}_2}^v - n_{\mathbf{p}_1}^c) \gamma}{(\varepsilon_{\mathbf{p}_1}^c + \varepsilon_{\mathbf{p}_2}^v - \omega_{\mathbf{k}})^2 + \gamma^2}, \quad (24)$$

where the line width at the resonance is the quantity

$$\begin{aligned} \gamma = \pi \sum_{\mathbf{p} \mathbf{q}} & |\mathbf{e}_{\mathbf{q}v} G_{\mathbf{q}v}|^2 \{ |i_c^c|^2 [N_{\mathbf{q}} \delta(\varepsilon_{\mathbf{p}}^c - \varepsilon_{\mathbf{p}-\mathbf{q}}^c - \Omega_{\mathbf{q}}) \\ & + (1 + N_{\mathbf{q}}) \delta(\varepsilon_{\mathbf{p}}^c - \varepsilon_{\mathbf{p}}^c + \Omega_{\mathbf{q}})] \\ & + |i_v^v|^2 [(1 + N_{\mathbf{q}}) \delta(\varepsilon_{\mathbf{p}}^v - \varepsilon_{\mathbf{p}+\mathbf{q}}^v - \Omega_{\mathbf{q}}) \\ & + N_{\mathbf{q}} \delta(\varepsilon_{\mathbf{p}}^v - \varepsilon_{\mathbf{p}}^v + \Omega_{\mathbf{q}})] \}. \end{aligned} \quad (25)$$

Let us consider the case of scattering by acoustical phonons at high temperatures. When we include the scattering of electrons by longitudinal phonons, we have

$$|G_{\mathbf{q}v}|^2 = \frac{E_0^2 q}{2Ms},$$

$$i_c^c = \delta(p_{1x} - p_x + q_x) \delta(p_{1z} - p_z + q_z) I_{n_1 n_2} \left(\frac{q_{\perp}^2}{2\gamma_0^2} \right),$$

where $I_{n_1 n_2}$ is the generalized Laguerre polynomial with a weight factor, E_0 is the constant in the deformation potential, s is the speed of sound, M is the mass of a cell, and $\gamma_0^2 = eH$. We consider the case in which

$$\varepsilon_{\mathbf{p}}^c = \omega_0(n + 1/2) + p_z^2 / 2m^c, \quad \Omega_{\mathbf{q}} = s|\mathbf{q}|, \quad \omega_0 = eH / m^c$$

(m^c is the effective mass of the carrier). Assuming that $\Omega_{\mathbf{q}} \ll \varepsilon_{\mathbf{p}}^c$, we get

$$\gamma = E_0^2 \gamma_0 (\beta M s^2 8\pi)^{-1} \sum_{n; i=c, v} m_i [p_{iz}^2 - 2\gamma_0(n_i - n)]^{-1/2}. \quad (26)$$

It can be seen from the expression for γ that resonance oscillations are possible, as in the case of scattering in a strong magnetic field.^[8] For scattering by optical phonons the line width will contain an oscillating expression analogous to the magnetophonon resonance.

3. Let us study the influence of a quantizing magnetic field on the absorption by carriers within the bands, with phonons participating. To find the absorption coefficient it is necessary to find the quantity $\langle a_{\mathbf{p}_1} a_{\mathbf{p}_2} | \beta_{\mathbf{k}'} \rangle$ which appears in the right member of (19), making use of the arguments that have been given. When we make an approximation of the type

$$\langle a_{\mathbf{p}}^+ c_{\mathbf{q}}^+ c_{\mathbf{q}_1} | \beta_{\mathbf{k}'} \rangle = \delta_{\mathbf{q} \mathbf{q}_1} \delta_{\mathbf{p} \mathbf{p}_1} n_{\mathbf{p}}^c N_{\mathbf{q}} \langle \beta_{\mathbf{k}'} | \beta_{\mathbf{k}'} \rangle \quad (27)$$

and use the fact that

$$J_c^c = em^{-1} [e_z \delta_{n_1 n_2} p_{1z} + (ie_y + e_x) \gamma_0 \sqrt{n_1 / 2} \delta_{n_1, n_2+1} + (e_x - ie_y) \gamma_0 \sqrt{(n_1 + 1) / 2} \delta_{n_1, n_2-1}] \delta_{p_{1x} p_{2x}} \delta_{p_{1z} p_{2z}},$$

after substituting these expressions in (19) and averaging over the polarizations of the photons and phonons we get for the main term

$$\begin{aligned} \Gamma = \frac{\pi |F_{\mathbf{k}}|^2}{3\omega_{\mathbf{k}}^2} \sum_{\substack{n_1 n_2 \\ \mathbf{q} \mathbf{p}_z}} & |G_{\mathbf{q}v}|^2 q_z^2 I_{n_1 n_2} \{ [n_{\mathbf{p}} (1 + N_{\mathbf{q}}) - n_{\mathbf{p}-\mathbf{q}} N_{\mathbf{q}}] \\ & \times \delta(\omega_{\mathbf{k}} + \varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}-\mathbf{q}} - \Omega_{\mathbf{q}}) + [n_{\mathbf{p}} N_{\mathbf{q}} - n_{\mathbf{p}+\mathbf{q}} (1 + N_{\mathbf{q}})] \\ & \times \delta[\omega_{\mathbf{k}} + \varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}+\mathbf{q}} + \Omega_{\mathbf{q}}], \quad \mathbf{p} = (p_x, p_z, n). \end{aligned} \quad (28)$$

When for the optical phonons we substitute

$$|G_{\mathbf{q}v}|^2 = \frac{2\pi \Omega_0 e^2}{q^2 V_0} \left(\frac{1}{\varepsilon_{\infty}} - \frac{1}{\varepsilon_0} \right)$$

(V_0 is the volume of the unit cell, ε_0 is the static dielectric constant, $\varepsilon_{\infty} = n^2$, and Ω_0 is the limit-

ing frequency of the optical phonons), we get:

$$\Gamma = \frac{|F_k|^2 \Omega_0 e^3 \gamma_0^2}{16 \omega_k^2 V_0} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \times \sum_{n_1, n_2} \int d p_z \frac{n_p (1 + N) - n_{p-q} N}{[p_z^2 - 2\gamma_0^2 (n_1 - n_2) - 2m^c (\omega_k \pm \Omega_0)]^{1/2}}. \quad (29)$$

It can be seen from the expression for Γ that for

$$\omega_k = \gamma_0^2 (n_2 - n_1) / m^c \pm \Omega_0$$

there will be resonance oscillations of the type treated by V. Gurevich and Firsov^[6] for $p_z = 0$. In addition to this, and in contrast with the results of L. Gurevich and Uritskiĭ,^[6] for the scattering by acoustical phonons there will also be a resonance absorption at $\omega_k = \gamma_0^2 / m^c (n_2 - n_1)$. It is easy to see that the integration over p_z in (29) leads to a logarithmic divergence; that is, it does not remove the resonance character of the absorption. Another important point is that the effect can occur with the emission of an optical phonon, unlike the magnetophonon oscillations in the electric conductivity.

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