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DISPERSION OF ELECTROMAGNETIC WAVES IN CRYSTALS

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A microscopic quantum theory of dispersion of electromagnetic waves in molecular crystals is developed. An expression is derived for the index of refraction due to the contribution of the excited states of the electrons. Interaction between exciton states and the field is considered without the use of perturbation theory.

INTRODUCTION

A microscopic quantum theory of dispersion of electromagnetic waves in isotropic, condensed media was developed by Fano.[1] In contrast to earlier work,[2] the oscillator strengths of electromagnetic transitions were not considered to be small in the work of Fano. Certain interest attaches to a generalization of the results obtained by Fano and to an analysis of the dispersion of electromagnetic waves in crystals of arbitrary symmetry.

A microscopic theory of dispersion, within the framework of classical physics, was developed by Born and Ewald.[3]

In the present paper we develop a microscopic quantum theory of the dispersion of electromagnetic waves for a specific model of a molecular crystal. However, the method of consideration can be used for any crystal.

The vibrations of the nucleus were not taken into account in this analysis. Consequently, the problem of the complex index of refraction should be considered separately.

1. TRANSITION TO THE REPRESENTATION OF SECOND QUANTIZATION

In the problem considered here of the interaction of the electromagnetic field with the electrons of a crystal, it is useful to employ a Coulomb gauge for the vector potential and thus assume that the vector potential of the microscopic field satisfies the condition (see reference 3):

$$\operatorname{div}\mathbf{A} = 0. \tag{1}$$

In this case, as is well known, the complete Hamiltonian of the system of the electrons and the field can be written in the form[4]

$$\hat{H} = \hat{H}_1 + \hat{H}_2 + \hat{H}_{\text{int}}, \tag{2}$$

where the Coulomb interaction of all charges making up the crystal are included in $\hat{H}_1$, $\hat{H}_2$ is the Hamiltonian corresponding to the free electromagnetic field of the transverse photons, $\hat{H}_{\text{int}}$ is the operator of interaction between all charges and the transverse field of the photons. In a molecular crystal, the operator $\hat{H}_1$ can best be represented in the following form

$$\hat{H}_1 = \sum_{\mathbf{n}\alpha} \hat{H}_{\mathbf{n}\alpha} + \frac{1}{2} \sum_{\mathbf{n}\alpha,\, \mathbf{n}\beta}' \hat{V}_{\mathbf{n}\alpha,\, \mathbf{m}\beta} \,. \tag{3}$$

In this expression $H_{\mathbf{n}\alpha}$ represents the Hamiltonian of an isolated molecule located at the position $\mathbf{n}\alpha$ ($\mathbf{n}$ is the unit lattice vector and α is the number of the molecule in the cell, $\alpha = 1, 2, \ldots, \sigma$), $\hat{V}_{\mathbf{n}\alpha,\mathbf{m}\beta}$ is the operator of Coulomb interaction between the charges of the molecules $\mathbf{n}\alpha$ and $\mathbf{m}\beta$.

We proceed to the representation of second quantization. For this purpose we choose as the set of basic functions the wave functions describing the stationary states of the isolated molecule.

Let $\varphi^{f}_{\mathbf{n}\alpha}$ be the wave function of the $\mathbf{n}\alpha$ molecule in a state f and not interacting with its surroundings. The value $f = 0$ corresponds to the ground state of the molecule. The function $\varphi^{f}_{\mathbf{n}\alpha}$ satisfies the equation

$$H_{\mathbf{n}\alpha}\varphi^{f}_{\mathbf{n}\alpha}(\tau_{\mathbf{n}\alpha}) = E^{f}_{\mathbf{n}\alpha}\varphi^{f}_{\mathbf{n}\alpha}(\tau_{\mathbf{n}\gamma}), \tag{4}$$

where $\tau_{\mathbf{n}\alpha}$ is the set of coordinates of the electrons of the molecule $\mathbf{n}\alpha$. To abbreviate the notation, we denote the set of indices by the single index $s \equiv (\mathbf{n}\alpha)$.

We now introduce normalized and antisymmetrized products (neglecting the overlap integrals):

$$\Psi_{\ldots N_s(f_s)\ldots}(\tau_1, \ldots, \tau_{N\sigma}) = [(N\sigma S)!]^{-1/2} \sum_{P} (-1)^{P} P \prod_{s,\, f_s'} \varphi_s^{f_s'}(\tau_s), \tag{5}$$

where S is the number of electrons in a molecule. Each occupation number $N_s(f_s)$ is equal to zero or unity and indicates the state of the s-th molecule. Therefore,

$$\sum_{f_s} N_s(f_s) = 1, \qquad \sum_{s,\, f_s} N_s(f_s) = N\sigma. \tag{6}$$

Here N is the number of elementary cells in the basic volume and σ is the number of molecules in the elementary cell. We can represent the wave function, which depends on the variables $\{\tau_s\}$, in the following form:

$$\Psi(\tau_1, \ldots, \tau_{N\sigma}) = \sum_{\ldots N_s(f_s) \ldots} C(\ldots N_s(f_s) \ldots) \times \Psi_{\ldots N_s(f_s) \ldots}(\tau_1, \ldots, \tau_{N\sigma}). \tag{7}$$

Here $C(\ldots N_s(f_s) \ldots)$ is the wave function of the electrons in the second-quantization representation. Let the additive operator

$$\hat{U}_1 = \sum_s \hat{U}_s(\tau_s) \tag{8}$$

and each of the operators $\hat{U}_s$ act only on the variable τ_s. Then, if we neglect the exchange of electrons which belong to different molecules, the matrix element is

$$\{\hat{U}_1\}_{\ldots N_s(f_s) \ldots,\, N'_s(f_s) \ldots} = \sum_{p,\, f_p,\, f'_p} \langle \varphi_{f_p}, \hat{U}_p \varphi_{f'_p} \rangle \{a_{f'_p f_p}\}_{\ldots N_s(f_s) \ldots, \ldots N'_s(f_s) \ldots}, \tag{9}$$

where

$$\{a_{f'_p f_p}\}_{\ldots N_s(f_s) \ldots, \ldots N'_s(f_s) \ldots} = N_p(f_p)\, N'_p(f'_p) \prod_{f_{s_1},\, s_1 \neq p} \delta[N_{s_1}(f_{s_1}) - N'_{s_1}(f_{s_1})].$$

Since we shall be interested below in the excited states of a crystal, which correspond to a completely determined — say the f-th — excited state in the isolated molecule, we shall assume that f_s takes on only two values, $f_s = 0$ or f. Then, if $f_p \neq f'_p$,

$$N_p(f_p)\, N'_p(f'_p) \equiv N_p(f_p)\, N'_p(f'_p)\, \delta[N'_p(f'_p) - N_p(f'_p) - 1] \times \delta[N'_p(f_p) - N_p(f_p) + 1],$$

where $\delta(x) = 1$ if $x = 0$, and $\delta(x) = 0$ if $x \neq 0$. Now if $f_p \neq f'_p$,

$$N_p(f_p)\, N'_p(f_p) \equiv N_p(f_p)\, N'_p(f_p) \times \delta[N'_p(f_p) - N_p(f_p)]\, \delta[N'_p(f'_p) - N_p(f'_p)].$$

Therefore, for arbitrary f_p and f'_p,

$$\{a_{f'_p f_p}\}_{\ldots N_s(f_s) \ldots, N'_s(f_s) \ldots} = N_p(f_p)\, N'_p(f'_p) \prod_{s,\, f_s} \delta[N'_s(f_s) - N_s(f_s) + \delta(f_s - f_p) - \delta(f_s - f'_p)].$$

Moreover, since

$$N_p(f_p) = \delta[N_p(f_p) - 1], \qquad N'_p(f'_p) = \delta[N'_p(f'_p) - 1],$$

we conclude that

$$\{a_{f'_p f_p}\}_{\ldots N_s(f_s) \ldots, N'_s(f_s) \ldots} = \delta[N_p(f_p) - 1]\, \delta[N_p(f'_p) - \delta(f_p - f'_p)] \prod_{s,\, f_s} \delta[N'_s(f_s) - N_s(f_s) + \delta(f_s - f_p) - \delta(f_s - f'_p)].$$

Thus the operator $a_{f'_p f_p}$ can be represented as the product of two operators

$$a_{f'_p f_p} = b^+_{f_p} b_{f'_p}, \tag{10}$$

which are defined as follows:

$$b_{f_p} C(\ldots N_s(f_s) \ldots) = \delta[N_p(f_p)]\, C(\ldots N_s(f_s) + \delta(f_s - f_p) \ldots),$$

$$b^+_{f_p} C(\ldots N_s(f_s) \ldots) = \delta[N_p(f_p) - 1]\, C(\ldots N_s(f_s) - \delta(f_s - f_p) \ldots). \tag{11}$$

It is easy to show that

$$a_{f_p f_p} C(\ldots N_s(f_s) \ldots) = N_p(f_p)\, C(\ldots N_s(f_s) \ldots),$$

i.e., that

$$b^+_{f_p} b_{f_p} = N_p(f_p).$$

On the other hand,

$$b_{f_p} b^+_{f_p} = 1 - N_p(f_p).$$

Thus,

$$b_{f_p} b^+_{f_p} + b^+_{f_p} b_{f_p} = 1.$$

Since the operators b_{f_p}, $b^+_{f_p}$ and $b_{f'_p}$, $b^+_{f'_p}$ act on different variables for $f_p \neq f'_p$, the following commutation rules hold:

$$b_{f_p} b_{f'_p} - b_{f'_p} b_{f_p} = 0, \qquad b^+_{f_p} b^+_{f'_p} - b^+_{f'_p} b_{f_p} = 0,$$

$$b_{f_p} b^+_{f'_p} - b^+_{f'_p} b_{f_p} = 0.$$

The operators with different p commute in all combinations.

Thus the operators b_{f_p} are neither Fermi nor Bose operators (they are the so-called Pauli operators). Making use of (9) and (10) we find that

$$U_1 = \sum_{p,\, f_p,\, f'_p} \langle \varphi_{f_p}, \hat{U}_p \varphi_{f'_p} \rangle\, b^+_{f_p} b_{f'_p}. \tag{12}$$

For an operator of the binary type

$$\hat{U}_2 = \sum_{p \neq p_1} U_{pp_1}(\tau_p, \tau_{p_1})$$

we can in similar fashion establish the following expression in the second-quantization representation:

$$\hat{U}_2 = \sum_{\substack{p \neq p_1 \\ (f_p f'_p f_{p_1} f'_{p_1})}} U_{pp_1}(f_p f_{p_1};\ f'_p f'_{p_1}) b^+_{f_p} b^+_{f_{p_1}} b_{f'_p} b_{f'_{p_1}}. \tag{13}$$

Here

$$U_{pp_1}(f_p f_{p_1};\ f'_p f'_{p_1}) = \int \varphi_p^{f_p *} \varphi_{p_1}^{f_{p_1}^*} U_{pp_1} \varphi_p^{f_p} \varphi_{p_1}^{f_{p_1}} d\tau_p d\tau_{p_1}$$

$$- \text{exchange terms} \tag{14}$$

We now write down (in the second-quantization representation) the operator $\hat{H}_1$. Making use of Eqs. (12) and (13), we find that

$$\hat{H}_1 = \sum_{s, f_s} E_s^{f_s} N_s(f_s) + \tfrac{1}{2} \sum_{\substack{s \neq s_1 \\ (f_s, f_{s_1}, f'_s, f'_{s_1})}} V_{ss'}(f_s f_{s_1};\ f'_s f'_{s_1}) b^+_{f_s} b_{f'_s} b^+_{f_{s_1}} b_{f'_{s_1}}$$

$$\equiv \hat{H}_1^{(0)} + \hat{H}_1^{(1)} + \hat{H}_1^{(2)},$$

where

$$\hat{H}_1^{(0)} = \sum_{s, f_s} E_s^{f_s} N_s(f_s) + \tfrac{1}{2} \sum_{\substack{s \neq s_1 \\ (f_s, f_{s_1})}} V_{ss_1}(f_s f_{s_1};\ f_s f_{s_1}) N_s(f_s) N_{s_1}(f_{s_1}),$$

$$\hat{H}_1^{(1)} = \tfrac{1}{2} \sum_{s \neq s_1} V_{ss_1}(0f;\ f0)\,[b^+_{s0} b_{sf} b^+_{s_1 f} b_{s_1 0} + b^+_{s_1 0} b_{s_1 f} b^+_{sf} b_{s0}],$$

$$\hat{H}_1^{(2)} = \tfrac{1}{2} \sum_{s \neq s_1} V_{ss_1}(00;\ ff)\,[b^+_{s0} b^+_{s_1 0} b_{sf} b_{s_1 f} + b^+_{sf} b^+_{s_1 f} b_{s0} b_{s_1 0}]. \tag{15}$$

We shall consider in what follows such excited states of the crystal which, for $V_{ss_1} \to 0$, correspond to a number of excited molecules which is small in comparison with the number N. Therefore, we shall neglect in the Hamiltonian $\hat{H}_1^{(0)}$ the components that are quadratic relative to $N_s(f)$.

In this approximation (neglecting the interaction of the excitons),

$$\hat{H}_1^{(0)} = E_0 + \sum_s N_s(f) \Big\{ (E_s^f - E_s^0)$$

$$+ \sum_{s_1}{}' [V_{ss_1}(0f,\ 0f) - V_{ss_1}(00,\ 00)] \Big\},$$

where

$$E_0 = \sum_s E_s^0 + \tfrac{1}{2} \sum_{s \neq s_1} V_{ss_1}(00,\ 00).$$

We now introduce the operators

$$B_{sf} = b^+_{s0} b^+_{sf}, \qquad B_{sf} = b^+_{sf} b_{s0}. \tag{16}$$

Then

$$B^+_{sf} B_{sf} = b^+_{s0} b_{sf} b^+_{sf} b_{s0} = N_s(f)\,[1 - N_s(0)]$$

$$= N_s(f) - N_s(0) N_s(f) = N_s(f),$$

since

$$N_s(0) + N_s(f) = 1.$$

Further,

$$B_{sf} B^+_{sf} - B^+_{sf} B_{sf} = N_s(0)\,[1 - N_s(f)]$$

$$- N_s(f)\,[1 - N_s(0)] = 1 - 2N_s(f).$$

In the states under examination, the mean value is $\overline{N_s(f)} \ll 1$. Therefore, we can assume approximately that

$$B_{sf} B^+_{sf} - B^+_{sf} B_{sf} = 1. \tag{17}$$

Since, for $s \neq s_1$, the operators B_{sf}, B^+_{sf} and $B_{s_1 f}$, $B^+_{s_1 f}$ commute with one another, we conclude that these operators are approximately operators of the Bose type. Thus, using (15) and (16), we obtain the following expression for the operator $\hat{H}_1$:

$$\hat{H}_1 = E_0 + \sum_s \Big\{ (E_s^f - E_s^0) + \sum_{s_1}{}' [V_{ss_1}(0f,\ 0f)$$

$$- V_{ss_1}(00,\ 00)] \Big\} B^+_{sf} B_{sf} + \sum_{s \neq s_1} V_{ss_1}(0f,\ 0f)\, B^+_{sf} B_{s_1 f}$$

$$+ \tfrac{1}{2} \sum_{s \neq s_1} V_{ss_1}(00,\ ff)\,[B_{sf} B_{s_1 f} + B^+_{sf} B^+_{s_1 f}]. \tag{18}$$

To determine the elementary excitations corresponding to the Hamiltonian $\hat{H}_1$, it is necessary to diagonalize the quadratic form (18). As Tyablikov has shown, a quadratic form of the type (18) is diagonalized as the result of a transformation to the new Bose-operators B_ρ and B^+_ρ (the method of Tyablikov is given in detail in the book of Bogolyubov[5]):

$$B_{sf} = \sum_\rho (B_\rho u_{s\rho} + B^+_\rho v^*_{s\rho}), \qquad B^+_{sf} = \sum_\rho (B^+_\rho u^*_{s\rho} + B_\rho v_{s\rho}), \tag{19}$$

where the quantities $u_{s\rho}$ and $v_{s\rho}$ entering into Eq. (19) are determined as the solutions of the equations

$$Eu_s = \sum_{s_1} [\delta_{ss_1}\Delta + (1 - \delta_{ss_1}) V_{ss_1}(0f,\ 0f)]\, u_{s_1}$$

$$+ \sum_s (1 - \delta_{ss_1}) V_{ss_1}(00,\ ff)\, v_{s_1},$$

$$-Ev_s = \sum_{s_1} [\delta_{ss_1}\Delta + (1 - \delta_{ss_1}) V_{ss_1}(0f,\ 0f)]\, v_{s_1}$$

$$+ \sum_{s_1} (1 - \delta_{ss_1}) V_{ss_1}(00,\ ff)\, u_{s_1}. \tag{20}$$

The possible values of E are determined from the condition of the existence of nontrivial solutions for the set of equations (20), where

$$\Delta \equiv (E_s^f - E_s^0) + \sum_{s_1}{}' [V_{ss_1}(0f, 0f) - V_{ss_1}(00, 00)].$$

The solutions of Eq. (20) should be chosen to satisfy the following normalization conditions (see reference 5):

$$\sum_s (|u_s|^2 - |v_s|^2) = 1. \qquad (21)$$

Recalling now that s = $(\mathbf{n}\alpha)$, we shall seek solutions of the system (20) in the following form:

$$u_{\mathbf{n}\alpha} = N^{-1/2} u_\alpha e^{i\mathbf{k}\mathbf{r}_{\mathbf{n}\alpha}}, \qquad v_{\mathbf{n}\alpha} = N^{-1/2} v_\alpha e^{i\mathbf{k}\mathbf{r}_{\mathbf{n}\alpha}}. \qquad (22)$$

Substituting (22) in (20), we conclude that u_α and v_α satisfy a set of 2σ equations

$$(E - \Delta)\, u_\alpha = \sum_\beta \Gamma^{(1)}_{\alpha\beta}(\mathbf{k})\, u_\beta + \sum_\beta \Gamma^{(2)}_{\alpha\beta}(\mathbf{k})\, v_\beta,$$

$$-(E + \Delta)\, v_\alpha = \sum_\beta \Gamma^{(1)}_{\alpha\beta}(\mathbf{k})\, v_\beta + \sum_\beta \Gamma^{(2)}_{\alpha\beta}(\mathbf{k})\, u_\beta, \qquad (23)$$

where

$$\Gamma^{(1)}_{\alpha\beta}(\mathbf{k}) = \sum_{\mathbf{m}} V_{\mathbf{n}\alpha,\, \mathbf{m}\beta}(0f,\ 0f) \exp\{i\mathbf{k}(\mathbf{r}_{\mathbf{m}\beta} - \mathbf{r}_{\mathbf{n}\alpha})\},$$

$$\Gamma^{(2)}_{\alpha\beta}(\mathbf{k}) = \sum_{\mathbf{m}} V_{\mathbf{n}\alpha,\, \mathbf{m}\beta}(00,\ ff) \exp\{i\mathbf{k}(\mathbf{r}_{\mathbf{m}\beta} - \mathbf{r}_{\mathbf{n}\alpha})\}. \qquad (23a)$$

Equating the determinant of the set of equations (22) to zero, we obtain an equation for E. In the general case, for arbitrary **k**, an equation of order σ is obtained for the quantity E^2. This equation determines σ positive roots of $E_\mu(\mathbf{k})$ $(\mu = 1, 2, \dots, \sigma)$.

We consider, as an example, a crystal with a single molecule in the elementary cell $(\sigma = 1)$. In this case, we find that

$$E(\mathbf{k}) = \{[\Delta + \Gamma^{(1)}(\mathbf{k})]^2 - [\Gamma^{(2)}(\mathbf{k})]^2\}^{1/2}.$$

In those cases in which $\Delta \gg \Gamma^{(2)}(\mathbf{k})$,

$$E(\mathbf{k}) \approx \Delta + \Gamma^{(1)}(\mathbf{k}) - [\Gamma^{(2)}(\mathbf{k})]^2/2\Delta. \qquad (24)$$

In the theory, where the Heitler-London method is used (which is equivalent to carrying out the diagonalization of the quadratic form (18) under the additional condition

$$\sum_s B^+_{sf} B_{sf} = 1,$$

see reference 6), the last term in (24) is absent. The value of the energy (24), as expected, turns out to be smaller than the value obtained with the use of the additional condition. However, this decrease is unimportant, since the ratio $|\Gamma^{(2)}(\mathbf{k})|/\Delta$ in molecular crystals, where Δ is of the order of several electron volts, is at most less than 0.1.* The operator $\hat{H}_1$ is represented by the new Bose-operators in the following form (see reference 5):

$$\hat{H}_1 = E_0 - \sum_{\alpha,\, k\mu} E_\mu(\mathbf{k})\, |v_\alpha(\mathbf{k},\ \mu)|^2 + \sum_{k\mu} E_\mu(\mathbf{k})\, B^+_{k\mu} B_{k\mu}. \qquad (25)$$

Thus $E_\mu(\mathbf{k})$ is the energy of the elementary excitation $(\mu, \mathbf{k})$. The second term in (25), which is absent when the Heitler-London method is used, is readily shown to be a small correction to the energy $E_0 \sim [\Gamma^{(2)}]^2/\Delta$.

2. DISPERSION OF ELECTROMAGNETIC WAVES IN MOLECULAR CRYSTALS

The Hamiltonian of transverse photons[4] has the form

$$\hat{H}_2 = \sum_{(qj)} \hbar qc\, a^+_{\mathbf{q}j} a_{\mathbf{q}j}. \qquad (26)$$

In this expression, **q** is the wave vector, j (= 1 or 2) is the number of the transverse polarization, $a^+_{\mathbf{q}j}$ and $a_{\mathbf{q}j}$ are the Bose operators of annihilation and creation of transverse photons $(\mathbf{q}j)$. The interaction operator in the nonrelativistic approximation can be represented in the following form:

$$\hat{H}_{\text{int}} = -\sum_{(\mathbf{n}\alpha)} \frac{e}{mc} \hat{\mathbf{A}}(\mathbf{r}_{\mathbf{n}\alpha})\, \hat{\mathbf{J}}_{\mathbf{n}\alpha}$$

$$+ \frac{e^2 S}{2mc^2} \sum_{(\mathbf{n}\alpha)} \hat{\mathbf{A}}^2(\mathbf{r}_{\mathbf{n}\alpha}) \equiv H^{\text{I}}_{\text{int}} + H^{\text{II}}_{\text{int}}, \qquad (27)$$

where $\hat{\mathbf{J}}_{\mathbf{n}\alpha}$ is the operator of the total momentum of the electrons of the molecule $(\mathbf{n}\alpha)$.

According to reference 4, the vector potential operator is

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{(\mathbf{q}j)} \sqrt{2\pi c^2 \hbar / Vqc}\, \mathbf{l}_{\mathbf{q}j} (a_{\mathbf{q}j} e^{i\mathbf{q}\mathbf{r}} + a^+_{\mathbf{q}j} e^{-i\mathbf{q}\mathbf{r}}). \qquad (28)$$

We choose the wave functions of the isolated molecules to be real and assume that the isolated molecules are not in any of the stationary states and do not possess dipole moments.

Substituting Eq. (28) in (27), we get

$$\hat{H}^{\text{I}}_{\text{int}} = -\sum_{\mathbf{q}j} \frac{e}{m} \sqrt{2\pi\hbar / Vqc} \left\{ a_{\mathbf{q}j} \left[\sum_{\mathbf{n}\alpha} \mathbf{l}^{i\mathbf{q}\mathbf{r}_{\mathbf{n}\alpha}} (\mathbf{l}_{qj} \hat{\mathbf{J}}_{\mathbf{n}\alpha}) \right] \right.$$

$$\left. + a^+_{\mathbf{q}j} \left[\sum_{\mathbf{n}\alpha} e^{-i\mathbf{q}\mathbf{r}_{\mathbf{n}\alpha}} (\mathbf{l}_{\mathbf{q}j} \hat{\mathbf{J}}_{\mathbf{n}\alpha}) \right] \right\}. \qquad (29)$$

Let us consider the operator

$$\hat{R}_\pm = \sum_{\mathbf{n}\alpha} e^{\pm i\mathbf{q}\mathbf{r}_{n\alpha}} (\mathbf{l}_{\mathbf{q}j} \hat{\mathbf{P}}_{\mathbf{n}\alpha}),$$

where $\hat{\mathbf{P}}_{\mathbf{n}\alpha}$ is the dipole-moment operator of the molecule $(\mathbf{n}\alpha)$. In the second-quantization representation,

$$\hat{R}_\pm = \sum_{\mathbf{n}\alpha} e^{\pm i\mathbf{q}\mathbf{r}_{n\alpha}} (\mathbf{l}_{qj} \mathbf{P}^{0f}_{\mathbf{n}\alpha})(b^+_{\mathbf{n}\alpha,0} b_{\mathbf{n}\alpha,f} + b_{\mathbf{n}\alpha,0} b^+_{\mathbf{n}\alpha,f}),$$

or, transforming to the Bose operators $B_{\mathbf{q}\mu}$, and neglecting transfer processes, we conclude that the operator $\hat{R}_\pm$ is given by

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***The difference between the expression E(k) and the corresponding value obtained by the Heitler-London method can become substantial when the width of the forbidden zone is of the same order as that of the allowed zone.**

$$\hat{R}_{\pm}=\sqrt{N}\sum_{\mu}\Big\{\sum_{\alpha}(\mathbf{l}_{\mathbf{q}j}\mathbf{P}_{\alpha}^{0f})\,[u_{\alpha}(\mathbf{q}\mu)+v_{\alpha}(\mathbf{q}\mu)]\Big\}(B_{\mp\mathbf{q}\mu}+B^{+}_{\pm\mathbf{q}\mu}).$$

We introduce the operator

$$\hat{\mathbf{J}}_{\pm}=\sum_{\mathbf{n}\alpha}e^{\pm i\mathbf{q}\mathbf{r}_{\mathbf{n}\alpha}}(\mathbf{l}_{\mathbf{q}j}\mathbf{J}_{\mathbf{n}\alpha}).$$

This operator is defined in the following way:

$$\hat{J}_{\pm}=(im/\hbar e)(\hat{H}_1\hat{R}_{\pm}-\hat{R}_{\pm}\hat{H}_1).$$

Therefore, making use of Eq. (25), we find that

$$\hat{\mathbf{J}}_{\pm}=\frac{im}{\hbar e}\sum_{\mu}E_{\mu}(\mathbf{q})\Big\{\sum_{\alpha}(\mathbf{l}_{\mathbf{q}j}\mathbf{P}_{\alpha}^{0f})\,[u_{\alpha}(\mathbf{q}\mu)+v_{\alpha}(\mathbf{q}\mu)]\Big\}(B_{\mp\mathbf{q}\mu}-B^{+}_{\pm\mathbf{q}\mu}).$$

Thus, we finally obtain the following expression for the operator $H^{\mathrm{I}}_{\mathrm{int}}$:

$$H^{\mathrm{I}}_{\mathrm{int}}=\sum_{\mu,\mathbf{q},j}T(j,\mathbf{q},\mu)\,[a_{\mathbf{q}j}(B_{-\mathbf{q},\mu}-B^{+}_{\mathbf{q},\mu})+a^{+}_{\mathbf{q}j}(B_{\mathbf{q}\mu}-B^{+}_{-\mathbf{q}\mu})],\tag{30}$$

where

$$T(j,\mathbf{q},\mu)=i\sqrt{2\pi N/qcV\hbar}\sum_{\alpha}(l_{\mathbf{q},j}\mathbf{P}^{0f}_{\alpha})\times[u_{\alpha}(\mathbf{q},\mu)+v_{\alpha}(\mathbf{q},\mu)]\,E_{\mu}(\mathbf{q}).\tag{30a}$$

Equations (30) and (30a) were obtained for crystals possessing a center of inversion. In such crystals the values of u_{α} and v_{α} can be chosen such that the conditions

$$u_{\alpha}(\mathbf{q},\mu)=u_{\alpha}(-\mathbf{q},\mu)=u^{*}_{\alpha}(\mathbf{q},\mu),$$
$$v_{\alpha}(\mathbf{q},\mu)=v_{\alpha}(-\mathbf{q},\mu)=v^{*}_{\alpha}(\mathbf{q},\mu).$$

are satisfied. In a similar way, we find that, with accuracy to within an unimportant constant term:

$$H^{\mathrm{II}}_{\mathrm{int}}=\frac{\hbar\omega_0^2}{4}\sum_{\mathbf{q}j}\frac{1}{qc}(2a^{+}_{\mathbf{q}j}a_{\mathbf{q}j}+a_{\mathbf{q}j}a_{-\mathbf{q}j}+a_{\mathbf{q}j}+a^{+}_{\mathbf{q}j}a^{+}_{-\mathbf{q}j}).\tag{31}$$

Here ω_0 is the frequency of the plasma oscillations

$$\omega_0^2=4\pi e^2N_1\sigma S/m,$$

where $N_1 = N/V$.

Combining Eqs. (30), (31), (26), and (25), and returning to the Hamiltonian of the entire system (2), we conclude that this Hamiltonian is given by

$$\begin{aligned}\hat{H}=E_0&-\sum_{\alpha,\mu,\mathbf{q}}E_{\mu}(\mathbf{q},\mu)\,|v_{\alpha}(\mathbf{q},\mu)|^2+\sum_{\mathbf{q},\mu}E_{\mu}(\mathbf{q})\,B^{+}_{\mathbf{q}\mu}B_{\mathbf{q}\mu}\\&+\sum_{\mathbf{q}j}\hbar qc\,a^{+}_{\mathbf{q}j}a_{\mathbf{q}j}-\sum_{\mu,q,j}T(j,\mathbf{q},\mu)\,[a_{\mathbf{q}j}(B_{-\mathbf{q}\mu}-B^{+}_{\mathbf{q}\mu})\\&+a^{+}_{\mathbf{q}j}(B_{\mathbf{q}\mu}-B^{+}_{-\mathbf{q}\mu})]\\&+\frac{\hbar\omega_0^2}{4}\sum_{\mathbf{q}j}\frac{1}{qc}(2a^{+}_{\mathbf{q}j}a_{\mathbf{q}j}+a_{\mathbf{q}j}a_{-\mathbf{q}j}+a^{+}_{\mathbf{q}j}a^{+}_{-\mathbf{q}j}).\end{aligned}\tag{32}$$

The problem of the determination of the elementary excitations that characterize the entire system of electrons and the field is equivalent to the problem of the diagonalization of the Hamiltonian (32). From these considerations it follows that the only Bose operators involved are those which correspond to the wave vectors $\mathbf{q}$ and $-\mathbf{q}$. Therefore, for the diagonalization of Eq. (32), it is sufficient to examine the quadratic form

$$\begin{aligned}H_q=&\sum_{\mu}E_{\mu}(\mathbf{q})(B^{+}_{\mathbf{q}\mu}B_{\mathbf{q}\mu}+B^{+}_{-\mathbf{q}\mu}B_{-\mathbf{q}\mu})\\&+\sum_{j}\hbar qc\,(a^{+}_{\mathbf{q}j}a_{\mathbf{q}j}+a^{+}_{-\mathbf{q}j}a_{-\mathbf{q}j})\\&\times\Big(1+\frac{\omega_0^2}{2q^2c^2}\Big)+\frac{\hbar\omega_0^2}{2}\sum_{j}\frac{1}{qc}(a_{\mathbf{q}j}a_{-\mathbf{q}j}+a^{+}_{\mathbf{q}j}a_{-\mathbf{q}j})\\&-\sum_{\mu j}T(j,\mathbf{q},\mu)\,[(a_{\mathbf{q}j}+a^{+}_{-\mathbf{q}j})(B_{-\mathbf{q}\mu}-B^{+}_{\mathbf{q}\mu})\\&+(a_{-\mathbf{q}j}+a^{+}_{\mathbf{q}j})(B_{\mathbf{q}\mu}-B^{+}_{-\mathbf{q}\mu})].\end{aligned}\tag{33}$$

The quadratic form (33) can be diagonalized with the aid of the method of Tyablikov, already used in Sec. 1. Following this method, we transform to new Bose operators ξ_{ρ} and ξ^{+}_{ρ}:

$$B_{\mathbf{q}\mu}=\sum_{\rho}[\xi_{\rho}u_{\mathbf{q}\mu}(\rho)+\xi^{+}_{\rho}v^{*}_{\mathbf{q}\mu}(\rho)],$$
$$a_{\mathbf{q}j}=\sum_{\rho}[\xi_{\rho}u_{\mathbf{q}j}(\rho)+\xi^{+}_{\rho}v^{*}_{\mathbf{q}j}(\rho)],\tag{34}$$

where the quantities u and v satisfy the following set of equations:

$$\begin{aligned}&[E_{\mu}(\mathbf{q})-\mathscr{E}]\,u_{\mathbf{q}\mu}+\sum_{j}T(j,\mathbf{q},\mu)(u_{\mathbf{q}j}+v_{-\mathbf{q}j})=0,\\&[E_{\mu}(\mathbf{q})+\mathscr{E}]\,v_{-\mathbf{q}\mu}-\sum_{j}T(j,\mathbf{q},\mu)(u_{\mathbf{q}j}+v_{-\mathbf{q}j})=0,\end{aligned}\tag{35}$$

$$\begin{aligned}&(\hbar qc-\mathscr{E})\,u_{\mathbf{q}j}-\sum_{\mu}T(j,\mathbf{q},\mu)(u_{\mathbf{q}\mu}-v_{-\mathbf{q}\mu})\\&\quad+\frac{\hbar\omega_0^2}{2qc}(u_{\mathbf{q}j}+v_{-\mathbf{q}j})=0,\\&(\hbar qc+\mathscr{E})\,v_{-\mathbf{q}j}-\sum_{\mu}T(j,\mathbf{q},\mu)(u_{\mathbf{q}\mu}-v_{-\mathbf{q}\mu})\\&\quad+\frac{\hbar\omega_0^2}{2qc}(u_{\mathbf{q}j}+v_{-\mathbf{q}j})=0.\end{aligned}$$

Comparing the first equation in this set with the second, and the third with the fourth, we find that

$$v_{-\mathbf{q}\mu}=-\frac{E_{\mu}(\mathbf{q})-\mathscr{E}}{E_{\mu}(\mathbf{q})+\mathscr{E}}u_{\mathbf{q}\mu},\qquad v_{-\mathbf{q}j}=\frac{\hbar qc-\mathscr{E}}{\hbar qc+\mathscr{E}}u_{\mathbf{q}j}.$$

Up to now we have considered only one of the excited states of the molecules. However, it is possible to take all the excited states into consideration. For this purpose, it suffices to consider the index μ as a compound index characterizing not only the number of exciton zones, but also the excitation level of the molecule corresponding to this zone. In this approximation, u and v are determined as before from the set of equations (35). However, the index μ now runs over an infinite set of values.

Such consideration of all the excited states of the molecules of the crystal is carried out in the spirit of the correspondence principle, according to which, in the dipole approximation, the optical behavior of the molecule is equivalent to the behavior of a corresponding set of oscillators. It should be noted that an account of all the excited states of the molecule is necessary only for obtaining the correct asymptotic behavior of $n^2(\omega)$ when $\omega \rightarrow 0$ and $\omega \rightarrow \infty$. No such account is necessary if the study is limited to the dependence of $n^2(\omega)$ in the region of frequencies close to one of the absorption bands in the molecule.

It follows from the system of equations (35) that the only states that interact with the transverse electromagnetic field are those for which $T\cdot(j, \mathbf{q}, \mu) \neq 0$. Expressing the quantity $u_{\mathbf{q}\mu}$ for these states in terms of $u_{\mathbf{q}j}$ with the help of the first of Eqs. (35), and substituting the resultant expression in the third of Eqs. (35), we get a system of two linear equations for the quantities $u_{\mathbf{q}j}$. Setting the determinant of this equation equal to zero, we obtain an involved equation for the energy of the elementary excitation $\mathscr{E}$ in the system of electron plus field. This equation has the following form:

$$(\hbar^2q^2c^2 + \hbar^2\omega_0^2 - \mathscr{E}^2)^2 - (\hbar^2q^2c^2 + \hbar^2\omega_0^2 - \mathscr{E}^2)(\Sigma_{11} + \Sigma_{22}) + \Sigma_{11}\Sigma_{22} - \Sigma_{12}\Sigma_{21} = 0, \tag{36}$$

where

$$\Sigma_{ij} = \Sigma_{ji} = \sum_{\mu} T(i, \mathbf{q}, \mu)\, T(j, \mathbf{q}, \mu) \frac{4\hbar qcE_\mu(\mathbf{q})}{\mathscr{E}^2 - E_\mu^2(\mathbf{q})}. \tag{37}$$

Equation (36) does not determine the energy of elementary excitations without dipole moments [for which $\mathscr{E} = E_\mu(\mathbf{q})$]. Moreover, Eq. (36) does not determine the longitudinal elementary excitations. For consideration of the latter, it is necessary to set the determinant of the system of equations for the quantities $u_{\mathbf{q}\mu}$ and $u_{\mathbf{q}j}$ equal to zero. This set of equations is obtained from (32) after elimination of the quantities $v_{-\mathbf{q}\mu}$ and $v_{-\mathbf{q}j}$.

The index of refraction of light waves $n^2 \equiv \hbar^2q^2c^2/\mathscr{E}^2$ can be obtained from Eq. (33):

$$n_{12}^2(\mathbf{s}, \omega) = 1 - \frac{\omega_0^2}{\omega^2} + \frac{1}{2\mathscr{E}^2}(\Sigma_{11} + \Sigma_{22}) \pm \frac{1}{2}\left[\left(\frac{\Sigma_{11} - \Sigma_{22}}{\mathscr{E}^2}\right)^2 + 4\left(\frac{\Sigma_{12}}{\mathscr{E}^2}\right)^2\right]^{1/2}. \tag{38}$$

In this equation and in what follows, the notation

$$\omega \equiv \mathscr{E}/\hbar, \qquad \mathbf{s} \equiv \mathbf{q}/q$$

is employed.

Equation (38) appears to have a pole at $\omega = 0$. We shall show that the components $\sim \mathscr{E}^{-2}$ in Eq. (38) vanish identically. Actually, let us consider the two operators

$$\hat{\mathbf{\Pi}} = \sum_{n\alpha} \hat{\mathbf{P}}_{n\alpha}, \qquad \hat{\dot{\mathbf{\Pi}}} = \sum_{n\alpha} \hat{\dot{\mathbf{P}}}_{n\alpha},$$

where $\hat{\mathbf{P}}_{\mathbf{n}\alpha}$ is the operator of the dipole moment of the $\mathbf{n}\alpha$-th molecule. We introduce $\mathbf{x}$ and $\mathbf{y}$ as two orthogonal coordinates. The operators $\dot{\Pi}_x$ and Π_y satisfy the following commutation relations:

$$\dot{\Pi}_x\Pi_y - \Pi_y\dot{\Pi}_x = -i\hbar \frac{e^2}{m} N\sigma S\delta_{xy}. \tag{39}$$

In the second-quantization representation

$$\Pi_y = \sum_{n\alpha} P_{\alpha y}^{0f}(\overset{+}{b}_{n\alpha,0} b_{n\alpha,f} + b_{n\alpha,0}\overset{+}{b}_{n\alpha,f})$$

or, transforming to the Bose operators $B_{\mathbf{q}\mu}$ ($B_{\mathbf{q}\mu}|_{q=0} \equiv B_{\mathbf{s}\mu}$),

$$\Pi_y = \sqrt{N}\sum_{\mu\alpha}\left\{P_{\alpha y}^{0f}[u_\alpha(\mathbf{s}\mu) + v_\alpha(\mathbf{s}\mu)]\right\}(B_{\mathbf{s}\mu} + \overset{+}{B}_{\mathbf{s}\mu}). \tag{40}$$

The operator $\dot{\Pi}_x$ is determined in the following fashion:

$$-i\hbar\dot{\Pi}_x = \hat{H}_1\Pi_x - \Pi_x\hat{H}_1.$$

Inserting Eq. (25) in this relation, we find that

$$\dot{\Pi}_x = \frac{i\sqrt{N}}{\hbar}\sum_{\mu} E_\mu(\mathbf{s}) \times \left\{\sum_{\alpha} P_{\alpha y}^{0f}[u_\alpha(\mathbf{s}\mu) + v_\alpha(\mathbf{s}\mu)]\right\}(B_{\mathbf{s}\mu} - \overset{+}{B}_{\mathbf{s}\mu}). \tag{41}$$

Making use of Eqs. (39), (40), and (41) we obtain the desired summation rule:

$$2\sum_{\mu} E_\mu(\mathbf{s})\left\{\sum_{\alpha} P_{\alpha x}^{0f}[u_\alpha(\mathbf{s}\mu) + v_\alpha(\mathbf{s}\mu)]\right\} \times \left\{\sum_{\beta} P_{\beta y}^{0f}[u_\beta(\mathbf{s}\mu) + v_\beta(\mathbf{s}\mu)]\right\} = \hbar^2\frac{e^2}{m}\sigma S\delta_{xy}. \tag{42}$$

If we introduce the notation

$$\mathbf{P}_{0\mu}(\mathbf{s}) \equiv \sum_{\alpha} P_\alpha^{0f}[u_\alpha(\mathbf{s}\mu) + v_\alpha(\mathbf{s}\mu)],$$

we can rewrite Eq. (42) somewhat differently:

$$2\sum_{\mu} E_\mu(\mathbf{s})\,|\mathbf{P}_{0\mu}(\mathbf{s})|^2 \cos\varphi_y(\mu\mathbf{s})\cos\varphi_x(\mu\mathbf{s}) = \hbar^2\frac{e^2}{m}\sigma S\delta_{xy}. \tag{43}$$

Since

$$E_\mu^2(\mathbf{q})/\mathscr{E}^2(\mathscr{E}^2 - E_\mu^2(\mathbf{q})) \equiv 1/[\mathscr{E}^2 - E_\mu^2(\mathbf{q})] - 1/\mathscr{E}^2,$$

$$\frac{1}{\mathscr{E}^2}\Sigma_{jj_1} = -\frac{8\pi N_1}{\mathscr{E}^2}\sum_{\mu} E_\mu(\mathbf{q})\,|P_{0\mu}(\mathbf{q})|^2\cos\varphi_j(\mu\mathbf{q})\cos\varphi_{j_1}(\mu\mathbf{q}) - 8\pi N_1\sum_{\mu}\frac{E_\mu(\mathbf{q})\,|\mathbf{P}_{0\mu}(\mathbf{q})|^2\cos\varphi_j(\mu\mathbf{q})\cos\varphi_{j_1}(\mu\mathbf{q})}{\mathscr{E}^2 - E_\mu^2(\mathbf{q})}.$$

Here j, $j_1 = 1, 2$, and $\varphi_j(\mu\mathbf{q})$ is the angle between the vectors $\mathbf{P}_{0\mu}(\mathbf{q})$ and $l_{\mathbf{q}j}$.

Making use of the summation rule (43), we find that the substitution of the resultant expression for Σ_{jj_1} in Eq. (38) eliminates the terms proportional to $\mathscr{E}^{-2}$. We can write the following as the final equation for the quantities $n_{1,2}(\omega)$, with accuracy to small terms $\sim (d/\lambda)^2$:

$$n_{1,2}^2(\omega, \mathbf{s}) = 1 - \frac{1}{2}\sum_{\mu}\frac{\omega_0^2 F_\mu(\mathbf{s})\sin^2\varphi(\mu\mathbf{s})}{\omega^2 - \Omega_\mu^2(\mathbf{q})}$$
$$\pm \frac{1}{2}\left\{\left[\sum_{\mu}\frac{\omega_0^2 F_\mu(\mathbf{s})(\cos^2\varphi_1(\mu\mathbf{q}) - \cos^2\varphi_2(\mu\mathbf{q}))}{\omega^2 - \Omega_\mu^2(\mathbf{q})}\right]^2\right.$$
$$\left. + 4\left[\sum_{\mu}\frac{\omega_0^2 F_\mu(\mathbf{s})\cos\varphi_1(\mu\mathbf{q})\cos\varphi_2(\mu\mathbf{q})}{\omega^2 - \Omega_\mu^2(\mathbf{q})}\right]^2\right\}^{1/2}, \qquad (44)$$

where

$$F_\mu(\mathbf{s}) \equiv \frac{2m}{\sigma S e^2 \hbar^2}|\mathbf{P}_{0\mu}(\mathbf{s})|^2 E_\mu(\mathbf{s}), \quad \Omega_\mu(\mathbf{q}) \equiv E_\mu(\mathbf{q})/\hbar,$$

F_μ is the oscillator strength corresponding to the level μ. This quantity is essentially independent of $\mathbf{s}$.

It is easy to show that as $\omega \to \infty$,

$$n_{1,2}^2(\omega, \mathbf{s}) \to 1 - \omega_0^2/\omega^2.$$

If $\omega \approx \Omega_f$, i.e., if the frequency is close to one of the absorption bands in the isolated molecule, one can separate in the sums over μ the resonance components that play the dominant role.

Let us consider some special cases.

1. The elementary cell of the crystal contains a single molecule ($\sigma = 1$). In this case,

$$n_1^2(\omega, \mathbf{s}) = \varepsilon(\omega, \mathbf{s}) - \frac{\omega_0^2 F(\mathbf{s})}{\omega^2 - \Omega^2(\mathbf{q})}\sin^2\varphi(\mathbf{s}),$$
$$n_2^2(\omega, \mathbf{s}) = \varepsilon(\omega, \mathbf{s}),$$

where the function $\epsilon(\omega, \mathbf{s})$ depends weakly on the frequency ω and the direction $\mathbf{s}$.

2. The crystal possesses cubic symmetry. In this case, the f-th molecular term corresponds to a twofold degenerate exciton band of transverse waves ($\mu = 1, 2$; $\mathbf{P}_{0\mu}(\mathbf{s}) \perp \mathbf{s}$, $|\mathbf{P}_{01}| = |\mathbf{P}_{02}|$, $\mathbf{P}_{01} \perp \mathbf{P}_{02}$) and to an exciton band of longitudinal waves, $\mu = 3$, $\mathbf{P}_{03} \parallel \mathbf{s}$. In this case, E_μ depends only on the absolute value q of the vector $\mathbf{q}$ and, consequently,

$$n_1^2(\omega, \mathbf{s}) = n_2^2(\omega, \mathbf{s}) = 1 - \tfrac{1}{2}\sum_{\mu}[\omega_0^2 F_\mu/(\omega^2 - \Omega_\mu^2(q))].$$

The factor ½ stands before the sum because each component appears twice in the sum over μ owing to the twofold degeneracy of the terms. Close to one of the resonances,

$$n_1^2 = n_2^2 = \varepsilon(\omega) - \omega_0^2 F_2/[\omega^2 - \Omega^2(q)],$$

where the function $\epsilon(\omega)$ dependes weakly on the frequency.

3. Let us now consider a crystal containing two molecules in the elementary cell. Molecular crystals of this group are naphthalene, anthracene and a number of others. In such crystals, each allowed molecular term corresponds to two allowed crystalline terms ($\mu = 1, 2$), for which the vectors $\mathbf{P}_{0\mu}$ are orthogonal (see reference 5). If the wave vector $\mathbf{q}$ is orthogonal to the plane of the vectors $\mathbf{P}_{0\mu}$, the general expression for the indices of refraction is materially simplified in the frequency region close to the frequencies of the doublet.

In this case we find from the general expression (44) that

$$n_1^2(\omega) = \varepsilon(\omega) - \omega_0^2 F_1/[\omega^2 - \Omega_1^2(\mathbf{q})],$$

$$n_2^2(\omega) = \varepsilon(\omega) - \omega_0^2 F_2/[\omega^2 - \Omega_2^2(\mathbf{q})].$$

Account of the spatial dispersion in the expressions obtained above can be carried out similarly to what was done in references 7 – 9. However, it must be noted that in inactive crystals the role of new effects, which arise upon consideration of spatial dispersion, is evidently small (in spite of the estimates given in reference 8).

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