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Polariton Theory of the Scattering of Light Beams in Semiconductors

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A microscopic theory is represented of the scattering of the excitonic polariton on the excitonic polariton in semiconductors with direct band gaps. All possible scattering mechanisms were taken into account. In the calculations the second quantization method was used in an effective manner. The general results were applied to the case of the single exciton level in CdS. The contributions from different scattering mechanisms were compared.

1. INTRODUCTION

In the experiments on the luminescence of highly excited semiconductors with direct band gaps it has been shown that together with the usual free-exciton lines in the luminescence spectra there arise new lines which can be explained as the consequences of the exciton–exciton and exciton–carrier scattering (see, e.g., Refs. [1–4] and literature therein). Within definite approximations the matrix elements of these scattering processes were calculated by many authors [4–8]. The exciton–exciton scattering contributes also to the third-order optical mixing coefficients of the semiconductors, since the photons can be transformed into the excitons and vice versa. This mechanism of the optical mixing becomes a dominant one in the resonance domain where the energies of the photons are nearly equal to those of the excitons. It is well known that in the presence of the exciton–photon transition the eigenstates of the Hamiltonian, i.e., the elementary excitations in the semiconductors, are the appropriate mixtures of the photons and the excitons—the excitonic polaritons or briefly the polaritons. [9–12]. The polariton theory of resonant Raman scattering was developed successfully by many authors [13–16].

In the papers by Anderson [17] the light-by-light interaction in the semiconductors via the polariton–polariton scattering was studied. However, in the derivation of the matrix element of this process only the contribution of the exciton–exciton collision was taken into account. Therefore Anderson's results can be used only in the resonance domain. Moreover, for estimating the exciton–exciton scattering amplitude Anderson made some assumptions on the interaction potential for the excitons as hard spheres.

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In this work we study the scattering of the light by the light as the polariton–polariton scattering, taking into account the microscopic structure of the excitons: excitons are treated as the hydrogen-like two-particle systems; the exciton–exciton scattering and other exciton–photon interaction processes are considered to be the consequences of the Coulomb interaction and the exchange interaction between the charge carriers as well as of their interaction with the photons. For our purpose it is convenient to apply the second quantization method. We shall use the unit system with $\hbar = c = 1$.

2. Polariton–Polariton Scattering in Terms of Elementary Processes

The state vectors of the polaritons are the linear combinations of those of the photons and the excitons. Therefore the polariton–polariton scattering amplitudes are the linear combinations of the amplitudes of the interaction processes involving directly the excitons and the photons, which we call the elementary ones. The following elementary processes contribute to the polariton–polariton scattering:

$$Ex + Ex \rightarrow Ex + Ex, \tag{I}$$

$$\gamma + Ex \rightarrow Ex + Ex, \tag{IIa}$$
$$Ex + Ex \rightarrow \gamma + Ex, \tag{IIb}$$

$$\gamma + Ex \rightarrow \gamma + Ex, \tag{III}$$

$$\gamma + \gamma \rightarrow Ex + Ex, \tag{IVa}$$
$$Ex + Ex \rightarrow \gamma + \gamma, \tag{IVb}$$

$$\gamma + \gamma \rightarrow \gamma + Ex, \tag{Va}$$
$$\gamma + Ex \rightarrow \gamma + \gamma, \tag{Vb}$$

$$\gamma + \gamma \rightarrow \gamma + \gamma. \tag{VI}$$

Here γ denotes the photon, Ex denotes the hydrogenlike two-particle system consisting of an electron and a hole in either their bound state or in the continuous energy spectrum (scattering state) of this system. For simplicity we consider only the photon with a definite polarization and we shall omit the polarization index of the creation or annihilation operators for the photons. Concerning the band structure of the medium we suppose that it is a two-band semiconductor with a direct band gap, the conduction and valence bands are non-degenerate and have their extrema at the center of the Brillouin zone, and the electrical dipole transition between the two bands is allowed.

In the presence of the exciton–photon transition the total Hamiltonian of the system of excitons and photons without the exciton–exciton interaction, i.e., of the system of free polaritons, is of the form

$$\mathscr{H}_0 = \sum_{\mathbf{p}} \Big\{ \omega(\mathbf{p})\, a(\mathbf{p})^+ a(\mathbf{p}) + \sum_{\nu \geqslant 1} E_\nu(\mathbf{p})\, b_\nu(\mathbf{p})^+ b_\nu(\mathbf{p}) + \sum_{\nu \geqslant 1} g_\nu(\mathbf{p}) [a(\mathbf{p})^+ b_\nu(\mathbf{p}) + b_\nu(\mathbf{p})^+ a(\mathbf{p})] \Big\}, \tag{1}$$

where $a(\mathbf{p})$ and $a(\mathbf{p})^+$ are the annihilation and creation operators for the photon with momentum $\mathbf{p}$ and energy $\omega(\mathbf{p})$, $b_\nu(\mathbf{p})$ and $b_\nu(\mathbf{p})^+$ are those for the exciton in the quantum state characterized by the index ν which denotes the set of all its quantum numbers, $E_\nu(\mathbf{p})$ is the energy of the corresponding exciton with momentum $\mathbf{p}$, $g_\nu(\mathbf{p})$ is the effective constant of the exciton–photon transition. For the bound states of the exciton the index ν is discrete while for the free electron–hole pairs with the Coulomb interaction the index ν changes continuously. In Ref. [18] it was shown that by means of the Bogoliubov transformation

$$c_\alpha(\mathbf{p}) = u_\alpha{}^0(\mathbf{p})\, a(\mathbf{p}) + \sum_{\nu \geqslant 1} u_\alpha{}^\nu(\mathbf{p})\, b_\nu(\mathbf{p}) \tag{2}$$

with the coefficients

$$u_\alpha{}^0(\mathbf{p}) = \Big\{ 1 + \sum_{\nu \geqslant 1} \frac{g_\nu(\mathbf{p})^2}{[\Omega_\alpha(\mathbf{p}) - E_\nu(\mathbf{p})]^2} \Big\}^{-1/2},$$
$$u_\alpha{}^\nu(\mathbf{p}) = \frac{g_\nu(\mathbf{p})}{\Omega_\alpha(\mathbf{p}) - E_\nu(\mathbf{p})}\, u_\nu{}^0(\mathbf{p}), \qquad \nu \geqslant 1, \tag{3}$$

we can diagonalize the Hamiltonian (1) and rewrite it in the following form

$$\mathscr{H}_0 = \sum_{\alpha,\mathbf{p}} \Omega_\alpha(\mathbf{p})\, c_\alpha(\mathbf{p})^+ c_\alpha(\mathbf{p}), \tag{4}$$

where the polariton energy $\Omega_\alpha(\mathbf{p})$ is determined by the algebraic equation

$$\frac{\omega(\mathbf{p})}{\Omega_\alpha(\mathbf{p})} = 1 + \frac{1}{\Omega_\alpha(\mathbf{p})} \sum_{\nu \geqslant 1} \frac{g_\nu(\mathbf{p})^2}{E_\nu(\mathbf{p}) - \Omega_\alpha(\mathbf{p})}. \tag{5}$$

In the case of a single exciton level this equation can be solved exactly, and we have the precise expression for $\Omega_\alpha(\mathbf{p})$

$$\Omega_\alpha(\mathbf{p}) = \tfrac{1}{2}\{E(\mathbf{p}) + \omega(\mathbf{p}) \mp ([E(\mathbf{p}) - \omega(\mathbf{p})]^2 + 4g(\mathbf{p})^2)^{1/2}\}, \tag{6}$$

where $E(\mathbf{p})$ is the energy of this unique exciton and $g(\mathbf{p})$ is the effective constant of the transition between this exciton and the photon $\omega(\mathbf{p})$.

The operators $C_\alpha(\mathbf{p})$ and $C_\alpha(\mathbf{p})^+$ are the annihilation and creation operators for the polariton in the branch characterized by the index α. From the expression (2) of $C_\alpha(\mathbf{p})$, we obtain the polariton–polariton scattering amplitude in the form

$$M_{\alpha_1\alpha_2\alpha_3\alpha_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4) = \sum_{\nu_i \geqslant 0} u_{\alpha_1}^{\nu_1}(\mathbf{p}_1)\, u_{\alpha_2}^{\nu_2}(\mathbf{p}_2)\, u_{\alpha_3}^{\nu_3}(\mathbf{p}_3)\, u_{\alpha_4}^{\nu_4}(\mathbf{p}_4) \cdot T_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4), \tag{7}$$

where $u_{\nu_i}^{\alpha_i}(\mathbf{p}_i)$, $\nu_i \geqslant 0$, are determined by Eqs. (3) and $T_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4)$ are the matrix elements of the elementary processes (I)–(VI), which will be studied in detail below.

3. Photon–Photon Scattering

Now we begin to calculate the matrix elements of the elementary interaction processes involving directly photons (with indices $\nu_i = 0$) and excitons (with indices $\nu_i \geqslant 1$) First we consider the photon–photon scattering (process (VI)):

$$\gamma + \gamma \to \gamma + \gamma.$$

The corresponding Feynman diagrams are represented in Fig. 1. For simplicity we use the abbreviated notations $(\omega(\mathbf{p}_1), \mathbf{p}_1) \equiv 1$, $(\omega(\mathbf{p}_2), \mathbf{p}_2) \equiv 2, \ldots$, etc.; $R(-1, 2, 3, 4)$, e.g., will denote the function obtained from $R(1, 2, 3, 4)$ by the substitutions $\mathbf{p}_1 \to -\mathbf{p}_1$, $\omega(\mathbf{p}_1) \to -\omega(-\mathbf{p}_1)$ while the other variables being unchanged. It can be shown that the matrix element of process (VI) has the form

$$T_{0000}(1, 2, 3, 4) = S(1, 2, 3, 4) + S(1, 2, 4, 3) + S(2, 1, 3, 4) + S(2, 1, 4, 3), \tag{8}$$

$$\begin{aligned} S(1, 2, 3, 4) = {} & R(1, 2, 3, 4) + R(1, -3, -2, 4) + R(1, -3, 4, -2) \\ & + R(-3, 1, 4, -2) + R(-3, 1, -2, 4) \\ & + R(-3, -4, -1, -2), \end{aligned} \tag{9}$$

$$R(1, 2, 3, 4) = X(1, 2, 3, 4) + Y(1, 2, 3, 4) + Z(1, 2, 3, 4). \tag{10}$$

where the Feynman diagrams corresponding to the matrix elements $X(1, 2, 3, 4)$, $Y(1, 2, 3, 4)$ and $Z(1, 2, 3, 4)$ are given in Figs. 1a, b and c, respectively. Introduce the notations:

$\boldsymbol{\xi}(\mathbf{p})$—the unit polarization vector of the photon

$\mathscr{P}_{cv}$—the matrix element of the allowed electromagnetic transition between the conduction and valence bands.

$$\mathscr{P}_{cv} = \langle c \mid -i\nabla \mid v \rangle$$

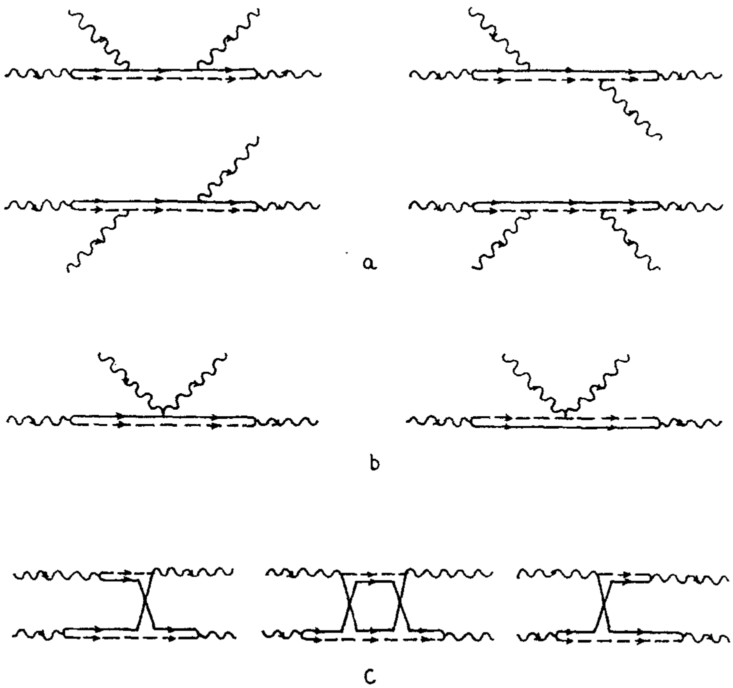

FIG. 1. Feynman diagrams of process (VI). Wavy line: photon, solid line: electron, dashed line: hole, double line consisting a solid line and a dashed one: exciton.

$F_\nu(\mathbf{r})$—hydrogen-like wave functions of the relative motion of the electron–hole pair in the quantum state with the index ν,

$f_\nu(\mathbf{p})$—the Fourier transforms of $F_\nu(\mathbf{r})$.

m_e^* and m_h^*—the effective masses of the electron and the hole in the corresponding energy band

$$\alpha = \frac{m_e^*}{m_e^* + m_h^*}, \qquad \beta = \frac{m_h^*}{m_e^* + m_h^*}, \tag{11}$$

ϵ_0—the background dielectric constant,

V—the volume of the crystal,

e—the charge of the electron,

m—the mass of the free electron.

With these notations the coupling constant $g_\nu(\mathbf{p})$ in formula (1) equals

$$g_\nu(\mathbf{p}) = \frac{e}{(2\epsilon_0\omega(\mathbf{p}))^{1/2}} \cdot \frac{\xi(\mathbf{p}) \cdot \mathscr{P}_{CV}}{m} \cdot F_\nu(0). \tag{12}$$

We can prove that the matrix elements X, Y, Z are determined by the following formulae:

$$X(1,2,3,4) = \sum_{\nu_1}\sum_{\nu_2}\sum_{\nu_3}\int d\mathbf{q} \cdot \frac{g_{\nu_1}(\mathbf{p}_1)\,\Gamma_{\nu_1\nu_2}(\mathbf{p}_2,\mathbf{p}_1;\mathbf{q})\,\Gamma_{\nu_3\nu_2}(\mathbf{p}_3,\mathbf{p}_4;\mathbf{q})^*\,g_{\nu_3}(\mathbf{p}_4)}{[\omega(\mathbf{p}_1)-E_{\nu_1}(\mathbf{p}_1)][\omega(\mathbf{p}_1)+\omega(\mathbf{p}_2)-E_{\nu_2}(\mathbf{q})][\omega(\mathbf{p}_4)-E_{\nu_3}(\mathbf{p}_4)]}, \tag{13}$$

$$Y(1,2,3,4) = \sum_{\nu_1}\sum_{\nu_2}\frac{g_{\nu_1}(\mathbf{p}_1)\,\Gamma_{\nu_1\nu_2}(\mathbf{p}_2,\mathbf{p}_1;\mathbf{p}_3,\mathbf{p}_4)\,g_{\nu_2}(\mathbf{p}_4)}{[\omega(\mathbf{p}_1)-E_{\nu_1}(\mathbf{p}_1)][\omega(\mathbf{p}_4)-E_{\nu_2}(\mathbf{p}_4)]}, \tag{14}$$

$$Z(1,2,3,4) = \sum_{\nu_1}\sum_{\nu_2}\sum_{\nu_3}\sum_{\nu_4}\int d\mathbf{q}_1\int d\mathbf{q}_2 \cdot \frac{g_{\nu_1}(\mathbf{p}_1)\,\Gamma_{\nu_1\nu_2\nu_3}(\mathbf{p}_2,\mathbf{p}_1;\mathbf{q}_1,\mathbf{q}_2)\,\Gamma_{\nu_4\nu_3\nu_2}(\mathbf{p}_3,\mathbf{p}_4;\mathbf{q}_1,\mathbf{q}_2)^*\,g_{\nu_4}(\mathbf{p}_4)}{[\omega(\mathbf{p}_1)-E_{\nu_1}(\mathbf{p}_1)][\omega(\mathbf{p}_1)+\omega(\mathbf{p}_2)-E_{\nu_2}(\mathbf{q}_1)-E_{\nu_3}(\mathbf{q}_2)][\omega(\mathbf{p}_4)-E_{\nu_4}(\mathbf{p}_4)]}, \tag{15}$$

$$\Gamma_{\nu_1\nu_2}(\mathbf{q},\mathbf{p}_1;\mathbf{p}_2) = -\frac{1}{V^{1/2}}\cdot\frac{e}{(2\epsilon_0\omega(\mathbf{q}))^{1/2}}\,\delta(\mathbf{q}+\mathbf{p}_1-\mathbf{p}_2)\cdot\boldsymbol{\xi}(\mathbf{q}) \cdot\left\{\frac{\mathscr{T}_{\nu_1\nu_2}(\beta\mathbf{p}_1,\beta\mathbf{p}_2)}{m_e^*}+\frac{\mathscr{T}_{\nu_1\nu_2}(\beta\mathbf{p}_1,\mathbf{p}_1-\alpha\mathbf{p}_2)}{m_h^*}\right\}, \tag{16}$$

$$\Gamma_{\nu_1\nu_2}(\mathbf{q}_1,\mathbf{p}_1;\mathbf{q}_2,\mathbf{p}_2) = \frac{1}{4V}\cdot\frac{e^2}{m\epsilon_0}\cdot\frac{\boldsymbol{\xi}(\mathbf{q}_1)\cdot\boldsymbol{\xi}(\mathbf{q}_2)^*}{(\omega(\mathbf{q}_1)\,\omega(\mathbf{q}_2))^{1/2}}\cdot\delta(\mathbf{q}_1+\mathbf{p}_1-\mathbf{q}_2-\mathbf{p}_2) \cdot\{I_{\nu_1\nu_2}(\beta\mathbf{p}_1,\beta\mathbf{p}_2)+I_{\nu_1\nu_2}(\beta\mathbf{p}_1,\mathbf{p}_1-\alpha\mathbf{p}_2)\}, \tag{17}$$

$$\Gamma_{\nu_1\nu_2\nu_3}(\mathbf{q},\mathbf{p}_1;\mathbf{p}_2,\mathbf{p}_3) = \frac{1}{V^{1/2}}\cdot\frac{e}{(2\epsilon_0\omega(\mathbf{q}))^{1/2}}\cdot\frac{\boldsymbol{\xi}(\mathbf{q})\cdot\mathscr{P}_{CV}}{m} \cdot\left\{\frac{1}{(2V)^{1/2}}\,G_{\nu_1\nu_2\nu_3}(\beta\mathbf{p}_1,\mathbf{p}_1-\alpha\mathbf{p}_2,\beta\mathbf{p}_3)\,\delta(\mathbf{q}+\mathbf{p}_1-\mathbf{p}_2-\mathbf{p}_3) - V^{1/2}F_{\nu_2}(0)^*\,\delta_{\nu_1\nu_3}\delta(\mathbf{q}-\mathbf{p}_2)\,\delta(\mathbf{p}_1-\mathbf{p}_3)\right\}, \tag{18}$$

where $\mathscr{T}_{\nu_1\nu_2}(\mathbf{p}, \mathbf{q})$, $I_{\nu_1\nu_2}(\mathbf{p}, \mathbf{q})$ and $G_{\nu_1\nu_2\nu_3}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3)$ denote the integrals

$$\mathscr{T}_{\nu_1\nu_2}(\mathbf{p}, \mathbf{q}) = \frac{1}{(2\pi)^3} \cdot \int d\mathbf{k} f_{\nu_1}(\mathbf{k} - \mathbf{p}) f_{\nu_2}(\mathbf{k} - \mathbf{q})^* \mathbf{k}, \tag{19}$$

$$I_{\nu_1\nu_2}(\mathbf{p}, \mathbf{q}) = \frac{1}{(2\pi)^3} \cdot \int d\mathbf{k} f_{\nu_1}(\mathbf{k} - \mathbf{p}) f_{\nu_2}(\mathbf{k} - \mathbf{q})^*, \tag{20}$$

$$G_{\nu_1\nu_2\nu_3}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3) = \frac{1}{(2\pi)^3} \cdot \int d\mathbf{k} f_{\nu_1}(\mathbf{k} - \mathbf{p}_1) f_{\nu_2}(\mathbf{k} - \mathbf{p}_2)^* f_{\nu_3}(\mathbf{k} - \mathbf{p}_3)^*, \tag{21}$$

In the Eqs. (14)–(16) the notation $\sum_{\nu_i}$ means the sums over discrete indices and the integrals over the continuous ones. It is easy to verify that the formulae (13)–(21) determine the most general photon–photon scattering amplitude in the semiconductor, when the Coulomb interaction of the electron and the hole in the intermediate state is also taken into account. In special cases they reduce to the results obtained in many earlier works [19–23].

4. Exciton–Exciton Scattering

Now we study the exciton–exciton scattering (process (I)) as the consequence of the electromagnetic interaction between the electrons and the holes. The corresponding Feynman diagrams are given in Fig. 2. Its matrix element can be written in the form ($\nu_i \geqslant 1$)

$$T_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = S_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) + S_{\nu_1\nu_2\nu_4\nu_3}(1, 2, 4, 3) + S_{\nu_2\nu_1\nu_3\nu_4}(2, 1, 3, 4) + S_{\nu_2\nu_1\nu_4\nu_3}(2, 1, 4, 3), \tag{22}$$

$$S_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \sum_{i=1}^{7} V^{(i)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4), \tag{23}$$

$$V^{(1)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{e^2}{V\epsilon_0 \mid \mathbf{p}_1 - \mathbf{p}_3 \mid^2} \cdot I_{\nu_1\nu_3}(\beta\mathbf{p}_1, \mathbf{p}_1 - \alpha\mathbf{p}_3) I_{\nu_2\nu_4}(\beta\mathbf{p}_2, \mathbf{p}_2 - \alpha\mathbf{p}_4), \tag{24}$$

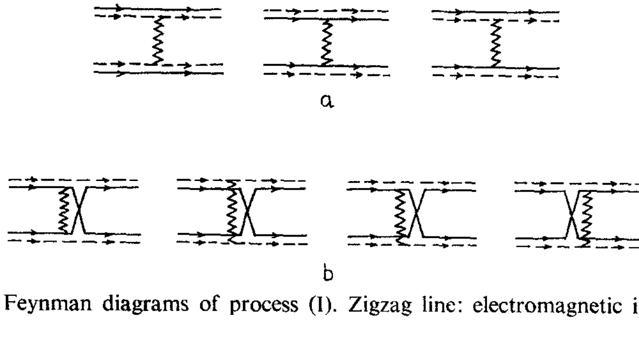

FIG. 2. Feynman diagrams of process (I). Zigzag line: electromagnetic interaction.

$$V^{(2)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{e^2}{V\epsilon_0 \mid \mathbf{p}_1 - \mathbf{p}_3 \mid^2} I_{\nu_1\nu_3}(\beta\mathbf{p}_1, \beta\mathbf{p}_3)\, I_{\nu_2\nu_4}(\beta\mathbf{p}_2, \beta\mathbf{p}_4), \tag{25}$$

$$V^{(3)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{-2e^2}{V\epsilon_0 \mid \mathbf{p}_1 - \mathbf{p}_3 \mid^2} \cdot I_{\nu_1\nu_3}(\beta\mathbf{p}_1, \mathbf{p}_1 - \alpha\mathbf{p}_3)\, I_{\nu_2\nu_4}(\beta\mathbf{p}_2, \beta\mathbf{p}_4), \tag{26}$$

$$V^{(4)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{-e^2\rho^2}{V\epsilon_0} L_{\nu_1\nu_2\nu_3\nu_4}(\beta\mathbf{p}_1, \beta\mathbf{p}_2, \beta\mathbf{p}_3, \beta\mathbf{p}_4; \mathbf{p}_1 - \mathbf{p}_4), \tag{27}$$

$$V^{(5)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{-e^2\rho^2}{V\epsilon_0} L_{\nu_1\nu_2\nu_3\nu_4}(\beta\mathbf{p}_1, \beta\mathbf{p}_2, \mathbf{p}_1 - \alpha\mathbf{p}_4, \mathbf{p}_2 - \alpha\mathbf{p}_3; \mathbf{p}_3 - \mathbf{p}_2), \tag{28}$$

$$V^{(6)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{2e^2\rho^2}{V\epsilon_0} N_{\nu_1\nu_2\nu_3\nu_4}(\beta\mathbf{p}_1, \mathbf{p}_3 - \alpha\mathbf{p}_2, \beta\mathbf{p}_3, \beta\mathbf{p}_4; \mathbf{p}_3 - \mathbf{p}_2), \tag{29}$$

$$V^{(7)}_{\nu_1\nu_2\nu_3\nu_4}(1, 2, 3, 4) = \frac{2e^2\rho^2}{V\epsilon_0} N_{\nu_1\nu_2\nu_3\nu_4}(\beta\mathbf{p}_3, \mathbf{p}_1 - \alpha\mathbf{p}_4, \beta\mathbf{p}_1, \beta\mathbf{p}_2; \mathbf{p}_1 - \mathbf{p}_4). \tag{30}$$

The integrals $I_{\nu_1\nu_2}(\mathbf{p}_1, \mathbf{p}_2)$ are determined by Eq. (20) while for the integrals $L_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4; \mathbf{q})$ and $N_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3 . \mathbf{p}_4; \mathbf{q})$ we have following definitions

$$L_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4; \mathbf{q}) = \frac{1}{(2\pi)^6 \rho^2} \int d\mathbf{k} \int d\mathbf{p} \cdot \frac{f_{\nu_1}(\mathbf{p} - \mathbf{p}_1) f_{\nu_2}(\mathbf{k} - \mathbf{p}_2) f_{\nu_3}(\mathbf{p} - \mathbf{p}_3)^* f_{\nu_4}(\mathbf{k} - \mathbf{p}_4)^*}{\mid \mathbf{k} - \mathbf{p} + \mathbf{q} \mid^2}, \tag{31}$$

$$N_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4; \mathbf{q}) = \frac{1}{(2\pi)^6 \rho^2} \int d\mathbf{k} \int d\mathbf{p} \cdot \frac{f_{\nu_1}(\mathbf{p} - \mathbf{p}_1) f_{\nu_2}(\mathbf{p} - \mathbf{p}_2) f_{\nu_3}(\mathbf{p} - \mathbf{p}_3)^* f_{\nu_4}(\mathbf{k} - \mathbf{p}_4)^*}{\mid \mathbf{k} - \mathbf{p} + \mathbf{q} \mid^2}. \tag{32}$$

For convenience we have introduced into Eqs. (27)–(32) the parameter ρ which is the radius of the exciton

$$\rho = \frac{\epsilon_0}{e^2} \cdot \frac{1}{\alpha m_{\mathrm{h}}^*}. \tag{33}$$

Since the wave functions $f_\nu(\mathbf{p})$ are orthogonal and normalized, the integral $I_{\nu_1\nu_2}(\mathbf{p}_1, \mathbf{p}_2)$ has the following property

$$I_{\nu_1\nu_2}(\mathbf{p}, \mathbf{p}) = \delta_{\nu_1\nu_2}.$$

Using this condition we can prove that in the limit $\mathbf{p}_j = 0; j = 1, 2, 3, 4$, we have

$$\lim_{\mathbf{p}_j \to 0} \sum_{i=1}^{3} V^{(i)}_{\nu_1\nu_2\nu_3\nu_4}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4) = 0 \tag{34}$$

We conclude that at small values of the exciton momenta $\mathbf{p}_j$ the contribution of the Coulomb interaction between the charge carriers consisting of different excitons (diagrams in Fig. 2a) to the scattering amplitude is negligible in comparison with that of the exchange interaction (diagrams in Fig. 2b). We note that a similar result was obtained in the paper by Buttner [5], in which some diagrams (but not all diagrams) take into account the exchange interaction.

5. Exciton–Photon Interaction Processes

The interaction processes involving directly both photons and excitons in the initial and final states can be considered in a similar manner. For example, the matrix element of the process (IIb) and the complex conjugate of that of the process (IIa) equal

$$T_{\nu_1\nu_2\nu_3 0}(1, 2, 3, 4) = R_{\nu_1\nu_2\nu_3}(1, 2, 3, 4) + R_{\nu_2\nu_1\nu_3}(2, 1, 3, 4), \tag{35}$$

$$R_{\nu_1\nu_2\nu_3}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{q}) = \frac{1}{(2V)} \cdot \frac{e}{(\epsilon_0\omega(\mathbf{q}))^{1/2}} \cdot \frac{\xi(\mathbf{q})^* \cdot \mathscr{P}_{CV}}{m} \cdot G_{\nu_1\nu_2\nu_3}(\beta\mathbf{p}_1, \mathbf{p}_1 - \alpha\mathbf{p}_2, \beta\mathbf{p}_3)^* \, \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{p}_3 - \mathbf{q}), \tag{36}$$

where the function $G_{\nu_1\nu_2\nu_3}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3)$ is determined by Eq. (21). The Feynman diagram of this process is given in Fig. 3. We observe again, by looking at this diagram, the important role of the exchange interaction.

For the photon–exciton scattering process (III) we have the following matrix element

$$T_{0\nu_2 0\nu_4}(1, 2, 3, 4) = R_{\nu_2\nu_4}(1, 2, 3, 4) + R_{\nu_2\nu_4}(-3, 2, -1, 4), \tag{37}$$

$$R_{\nu_2\nu_4}(1, 2, 3, 4) = X_{\nu_2\nu_4}(1, 2, 3, 4) + Y_{\nu_2\nu_4}(1, 2, 3, 4) + Z_{\nu_2\nu_4}(1, 2, 3, 4), \tag{38}$$

$$X_{\nu_2\nu_4}(1, 2, 3, 4) = \sum_{\nu} \int d\mathbf{q} \, \frac{\Gamma_{\nu_2\nu}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{q}) \, \Gamma_{\nu_4\nu}(\mathbf{p}_3, \mathbf{p}_4; \mathbf{q})^*}{\omega(\mathbf{p}_1) + E_{\nu_2}(\mathbf{p}_2) - E_{\nu}(\mathbf{q})}, \tag{39}$$

$$Y_{\nu_2\nu_4}(1, 2, 3, 4) = \Gamma_{\nu_2\nu_4}(1, 2; 3, 4), \tag{40}$$

$$Z_{\nu_2\nu_4}(1, 2, 3, 4) = \sum_{\nu} \sum_{\nu'} \int d\mathbf{q} \int d\mathbf{q}' \cdot \frac{\Gamma_{\nu_2\nu\nu'}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{q}, \mathbf{q}') \, \Gamma_{\nu_4\nu\nu'}(\mathbf{p}_3, \mathbf{p}_4; \mathbf{q}, \mathbf{q}')^*}{\omega(\mathbf{p}_1) + E_{\nu_2}(\mathbf{p}_2) - E_{\nu}(\mathbf{q}) - E_{\nu'}(\mathbf{q}')}, \tag{41}$$

where the vertices $\Gamma_{\nu_2\nu}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{q})$, $\Gamma_{\nu_2\nu_4}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{p}_3, \mathbf{p}_4)$ and $\Gamma_{\nu_2\nu'}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{q}, \mathbf{q}')$ are determined by Eqs. (16)–(21). The Feynman diagrams corresponding to the matrix elements $X_{\nu_2\nu_4}(1, 2, 3, 4)$, $Y_{\nu_2\nu_4}(1, 2, 3, 4)$ and $Z_{\nu_2\nu_4}(1, 2, 3, 4)$ are represented in Figs. 4a, b, and c, respectively.

FIG. 3. Feynman diagram of process (IIb).

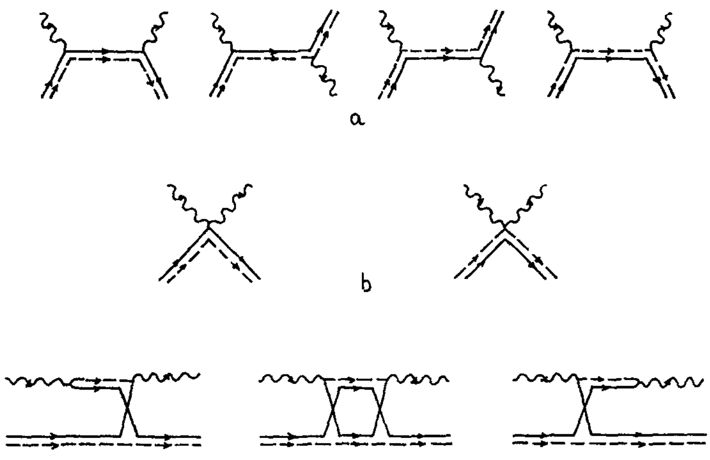

FIG. 4. Feynman diagrams of process (III).

The two-exciton recombination process (IVb) and the exciton pair creation process (IVa) have complex conjugate matrix elements. The latter is decribed by the Feynman diagram in Fig. 5. Its amplitude equals

$$T_{00\nu_3\nu_4}(1, 2, 3, 4) = R_{\nu_3\nu_4}(1, 2, 3, 4) + R_{\nu_3\nu_4}(2, 1, 3, 4) + R_{\nu_4\nu_3}(1, 2, 4, 3) + R_{\nu_4\nu_3}(2, 1, 4, 3), \tag{42}$$

$$R_{\nu_3\nu_4}(1, 2, 3, 4) = \sum_{\nu} \frac{g_\nu(\mathbf{p}_1)\, \Gamma_{\nu\nu_3\nu_4}(\mathbf{p}_2, \mathbf{p}_1; \mathbf{p}_3, \mathbf{p}_4)}{\omega(\mathbf{p}_1) - E_\nu(\mathbf{p}_1)}, \tag{43}$$

where $\Gamma_{\nu\nu_3\nu_4}(\mathbf{p}_2, \mathbf{p}_1; \mathbf{p}_3, \mathbf{p}_4)$ is given by Eqs. (18) and (21).

Finally, for the matrix element of the process (Va) and the complex conjugate of that of the process (Vb) we have the following expressions

$$T_{000\nu_4}(1, 2, 3, 4) = S_{\nu_4}(1, 2, 3, 4) + S_{\nu_4}(2, 1, 3, 4), \tag{44}$$

$$S_{\nu_4}(1, 2, 3, 4) = R_{\nu_4}(1, 2, 3, 4) + R_{\nu_4}(1, -3, -2, 4) + R_{\nu_4}(-3, 2, -1, 4), \tag{45}$$

$$R_{\nu_4}(1, 2, 3, 4) = X_{\nu_4}(1, 2, 3, 4) + Y_{\nu_4}(1, 2, 3, 4) + Z_{\nu_4}(1, 2, 3, 4), \tag{46}$$

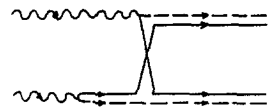

FIG. 5. Feynman diagram of process (IVa).

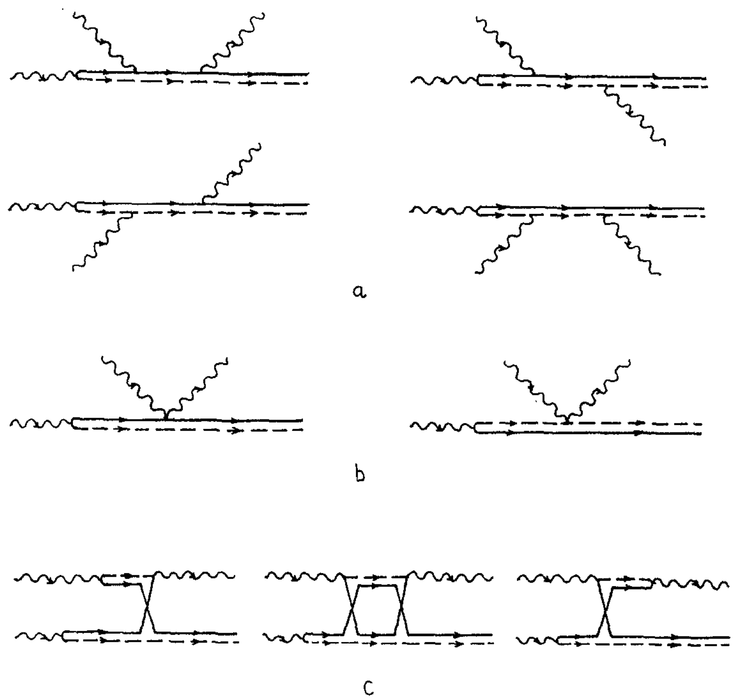

FIG. 6. Feynman diagrams of process (Vb).

where $X_{\nu_4}(1, 2, 3, 4)$, $Y_{\nu_4}(1, 2, 3, 4)$ and $Z_{\nu_4}(1, 2, 3, 4)$ are the matrix elements of the Feynman diagrams in Figs. 6a, b, and c, respectively.

$$X_{\nu_4}(1, 2, 3, 4) = \sum_{\nu_1}\sum_{\nu_2}\int d\mathbf{q}\, \frac{g_{\nu_1}(\mathbf{p}_1)\, \Gamma_{\nu_1\nu_2}(\mathbf{p}_2, \mathbf{p}_1; \mathbf{q})\, \Gamma_{\nu_4\nu_2}(\mathbf{p}_3, \mathbf{p}_4; \mathbf{q})^*}{[\omega(\mathbf{p}_1) - E_{\nu_1}(\mathbf{p}_1)][\omega(\mathbf{p}_1) + \omega(\mathbf{p}_2) - E_{\nu_2}(\mathbf{q})]}, \tag{47}$$

$$Y_{\nu_4}(1, 2, 3, 4) = \sum_{\nu} \frac{g_{\nu}(\mathbf{p}_1)\, \Gamma_{\nu\nu_4}(\mathbf{p}_2, \mathbf{p}_1; \mathbf{p}_3, \mathbf{p}_4)}{\omega(\mathbf{p}_1) - E_{\nu}(\mathbf{p}_1)}, \tag{48}$$

$$Z_{\nu_4}(1, 2, 3, 4) = \sum_{\nu_1}\sum_{\nu_2}\sum_{\nu_3}\int d\mathbf{q}\int d\mathbf{q}'$$

$$\cdot \frac{g_{\nu_1}(\mathbf{p}_1)\, \Gamma_{\nu_1\nu_2\nu_3}(\mathbf{p}_2, \mathbf{p}_1; \mathbf{q}, \mathbf{q}')\, \Gamma_{\nu_4\nu_2\nu_3}(\mathbf{p}_3, \mathbf{p}_4; \mathbf{q}, \mathbf{q}')^*}{[\omega(\mathbf{p}_1) - E_{\nu_1}(\mathbf{p}_1)][\omega(\mathbf{p}_1) + \omega(\mathbf{p}_2) - E_{\nu_2}(\mathbf{q}) - E_{\nu_3}(\mathbf{q}')]}. \tag{49}$$

The vertices $\Gamma_{\nu_1\nu_2}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{p}_3)$, $\Gamma_{\nu_1\nu_2}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{p}_3, \mathbf{p}_4)$ and $\Gamma_{\nu_1\nu_2\nu_3}(\mathbf{p}_1, \mathbf{p}_2; \mathbf{p}_3, \mathbf{p}_4)$ in these formulae are determined by Eqs. (16)–(21).

6. Comparison of the Contributions from Different Scattering Mechanisms

In this concluding section we apply the above results to calculate the polariton–polariton scattering cross section near the resonance in the crystal CdS. We use the following values of the parameters [24]:

$$m_e^* = 0.2\ \text{m}, \qquad m_h^* = 0.65\ \text{m}, \qquad E_g = 2.58\ \text{eV}$$

$$\epsilon_0 = 10, \qquad \frac{|\mathscr{P}_{CV}|}{m} = 4.95 \times 10^{-3}.$$

Here E_g is the band gap.

For simplicity we use the approximation with a single undamped dispersionless exciton level. The energy of exciton is taken to be

$$E = E_{1S}(0) = 2.55\ \text{eV}.$$

The effective photon–exciton transition coupling constant is derived from Eq. (12) and equals

$$g = g_{1S}(0) = 51.85\ \text{meV}$$

From this values of the constant g we obtain for the longitudinal–transverse spliting energy

$$E_{lt} = \frac{2g^2}{E} \approx 2.1\ \text{meV},$$

while the experimental data give 2 meV [13].

We consider in details the special kinematic configuration of the scattering process in which the momenta satisfy the conditions

$$\mathbf{p}_1 + \mathbf{p}_2 = \mathbf{p}_3 + \mathbf{p}_4 = 0,$$
$$\mathbf{p}_1 \perp \mathbf{p}_3, \qquad p = |\mathbf{p}_1| = |\mathbf{p}_3|,$$

while the polariton energies are smaller than that of the exciton in the ground state

$$\Omega_{\alpha_i} \leqslant E_{1S}(0).$$

In this particular case we have only the transition between the polaritons in the same lowest branch, and we can omit the branch indices α_i. The scattering cross section at the given kinematics equals

$$\frac{d\sigma}{d\Omega} = \frac{p^2}{16\pi^2} \cdot \frac{|VM(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4)|^2}{v(\mathbf{p})^2}, \tag{50}$$

where $v(\mathbf{p})$ is the group velocity of the polariton at the momentum $\mathbf{p}$. From the above expressions for the scattering amplitude it is easy to check that the product $VM(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4)$ does not depend on the volume V of the crystal. We shall take $V = 1$, substite $M(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4)$ for $VM(\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4)$, and denote by M^n the contribution of the scattering mechanism involving directly n excitons in the initial and final states all together:

$n = 0$—the contribution from the mechanism without external excitons (process (VI)).

$n = 1$—the contribution from the mechanism with one initial or final exciton (two processes: (Va) and (Vb)).

$n = 2$—the contribution from the mechanism involving two external excitons (three processes: (III), (IVa) and (IVb)).

$n = 3$—the contribution from the processes (IIa) and (IIb).

$n = 4$—the exciton–exciton scattering mechanism.

In Fig. 7 we represent the squared values of the contributions from different scattering mechanisms to the scattering amplitude as the functions of the ratio

$$\kappa = \frac{\Omega}{E}$$

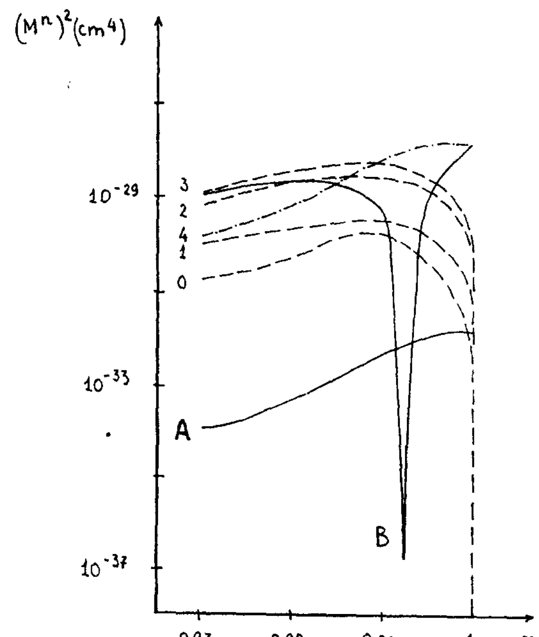

FIG. 7. Squared values of the contributions from different scattering mechanisms to the scattering amplitude (curves with the indices $n = 0, 1, 2, 3, 4$) and squared value of their sum (curve B—this work, curve A—Anderson [17]).

at the polariton energies below the ground-state level of the exciton. For comparison we plot in the same figure the squared sum of all contributions M^n (curve B) and also the same quantity calculated in the crude approximation by Anderson (curve A). We remark that due to the cancelation between the contributions from different scattering mechanisms there exists an antiresonance at the value of κ near 1.

The numerical calculation was done by the computers of the Laboratory of Computer Technique and Automatization of the Joint Institute for Nuclear Research, Dubna, USSR.

In conclusion we note that throughout this work we have ignored the interaction between the excitons. It is well known that due to this interaction two excitons can exist in their bound state—the biexciton or the excitonic molecule. The biexciton effect in the polariton–polariton scattering will be investigated in a subsequent work.

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