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Four-Body Scattering of Light as a Method of Detection of Bose-Einstein Condensate of Excitons

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Abstract: Process of four-particle light scattering in molecular crystals with participation of the exciton Bose-condensate is considered. The intensities and tensor of scattering are found for this effect. The frequency and polarization characteristics of this process are studies. It is shown that the investigation of the spectrum shapes of scattered radiation allows one to find and prove the existence of the Bose-Einstein condensate of excitons.

Keywords: Molecular Crystal, Four-Particle Light Scattering, Exciton, Polaritons, Bose-Einstein Condensate

1. Introduction

Effect of condensation of Bose particles in pulse space was predicted by Einstein in 1924-1925 [1, 2]. For the first time, the effect was experimentally detected in 1995 [3-5] in atomic Bose gases. As a result of this outstanding experiment, a branch emerged that dealt with studying of Bose-Einstein condensation (BEC) in gases. As a consequence, interesting effects were found: stimulated reinforcement ("atomic laser"), vortex structures [3-5] etc. The existence of three types of the systems associated with possible BEC was predicted [6]. The first type included the systems where BEC resulted in macroscopic filling of the lowest single energy state. The systems of the second type were characterized by discrete continuum and a finite number of the lowest states. In this case, BEC proceeds in two stages. First, at some temperature, all the states of the system were filled. At the further temperature drop, BEC took place in the lowest state. The third type of the systems was also characterized by two-stage BEC. At the beginning, BEC occurred in the lowest continuum with the infinite number of states that were characterized by macroscopic filling. Studying of BEC gases was complicated because very low temperatures had to be reached (~ 100 nK), being determined by a relatively large weight of gas particles.

This demerit can be compensated by the study of BEC of other Bose particles, e.g. excitons. The mass of quasi-particles is four-order smaller, so BEC can be registered at much higher temperature. Powerful sources of coherent radiation provide a possibility of formation of the exciton density about $10^{17} - 10^{18}$ cm^{-3} that makes the collective properties substantial. As exciton excitations of a crystal can be treated as bosons with a reasonable degree of accuracy, Bose-Einstein condensation (BEC) should be detected at low temperatures and repulsive exciton-exciton interaction when the values of the wave-vector $\vec{k}$ correspond to the energy minimum [7]. Later a number of papers was published that were aimed at detection and testing of BEC excitons. In the pioneer works, emergence of a narrow luminescence peak was the main evidence of detection of the condensate. But the application of this method to three-dimensional crystals did not resulted in reliable identification of BEC because of short lifetime of excitons. This fact did not allow accumulation of sufficiently large amount of quasi-particles in the lowest state. The main factors reducing the lifetime were various non-radiative relaxation processes. In order to extend the lifetime of quasiparticles, attempts of forming

BEC within quantum confinements or in composite sandwich-like materials were made. For most experiments to date, exciton-polariton condensates are produced at cryogenic temperatures in the vicinity of ~ 10 K using materials such as GaAs and CdTe. However, using other materials such as GaN, ZnO, and organic semiconductors, polariton condensates at higher temperatures including room temperature have been also realized [8-11]. Theoretical works dealing with the problem of detection of exciton BEC were mostly concerned with the spectrum calculations and the following conclusion about the position of the luminescence line and the shape of the tails [12, 13]. In [12], incorrect application of Belyaev method was made [14], that was valid only in the case when the condensate was the ground state of the system and BEC of excitons was an excited state. The list of possible candidates able to demonstrate BEC was recently supplemented with a new type of quasiparticles [15]. In particular, it was shown [16] that in the case of a two-dimensional system of spin-less electrons under quantum Hall effect, so-called composite particles (CP) could emerge. These objects are point vortices fixed to a bare particle. At certain conditions, CPs obey Bose statistics and BEC can be realized. Review [17] was focused upon several physical phenomena exhibited by excitonpolariton condensates. In particular, the topics related to the difference between a polariton BEC, a polariton laser, and a photon laser, as well as physical phenomena such as superfluidity, vortex formation, BKT (Berezinskii-Kosterlitz-Thouless) and BCS (Bardeen-Cooper-Schrieffer) physics were examined. The physics and application of engineered polariton structures was also discussed. The results of studying of BEC excitons form the basis of application of the effect. In particular, a possibility of the existence of an exciton dielectric [18] and the related double-wire superconducting line [19] is predicted

Having regard to the above, the existence proof of BEC requires additional methods beside luminescence. The related opportunity exists because BEC of excitons interacts with other subsystems, e.g. phonon and photon ones. As a result, the spectrum of the elementary excitations is modified. It is demonstrated in [15] that the presence of BEC excitons makes possible the appearance of additional polariton states. In [20], acoustic methods of testing are suggested to be applied. In particular, it is shown that unusual behavior of the absorption coefficient of surface acoustic waves is determined by BEC exciton gas. Besides, various non-linear optical effects can be used [21-23]. They provide BEC detection at the state of $k = 0$. These states cannot irradiate because the use of luminescence implies $k \neq 0$. But an exciton can be drawn out of this state by means of various non-linear processes. In particular, in [23, 24], a method of double-photon recombination is developed. The main idea is radiation of two excitons characterized by the opposite impulses of the same moduli. In the present work, one on numerous non-linear processes is suggested to be applied to the testing and identification of BEC excitons. BEC excitons associated with both zero and non-zero impulse can participate in the process.

2. Theoretical Background

Consider a cubic molecular crystal with one molecule in the unit cell. Assume that a Bose – Einstein condensate (BEC) of excitons has been created in the crystal on branch σ for $k = 0$ and at the temperature of *0K*. The energy per one BEC quasiparticle [20] is equal to the chemical potential μ.

Let us investigate a process of nonlinear scattering when a polariton of energy $\hbar\omega_1 \equiv E_{\rho_1}(\vec{k}_1)$ and a condensate exciton are annihilated, while a scattered polariton $\hbar\omega_2 \equiv E_{\rho_2}(\vec{k}_2)$ and a supracondensate exciton are born at branch ν. The process is represented in Figure 1, where the wavy lines stand for polaritons, while the dotted and solid lines correspond to the condensate and supracondensate excitons.
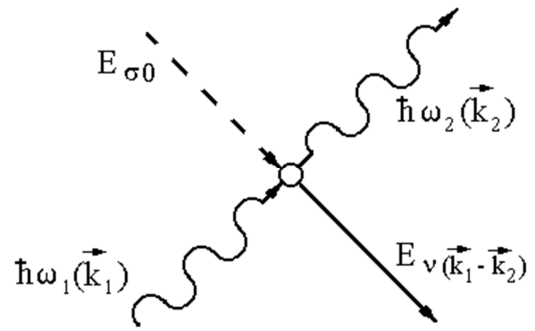

Figure 1. *The scheme of the four-body process. The wavy lines stand for polaritons, while the dotted and solid lines correspond to the condensate and supracondensate excitons.*

The problem could be solved within the framework of exciton – polariton Hamiltonian which takes into account the summands of the third and fourth order terms in operators of quasi – particle generation and annihilation. However, the use of interaction constants shown in [25, 26] gives a very bulky scattering tensor. To simplify the tensor, the method of acting field was applied [27, 28].

As known [29], in molecular crystals, the electronic molecular escitations obey the Pauli statistics. But after the transition from the Pauli operators to Bose operators with using the procedure developed in [30], the Hamiltonian of the system composed by the crystal and the field of transverse photons can be represented as

$$H = \frac{1}{2}\sum\left\{\left(\vec{p}_{nf} - \sqrt{\frac{v_0}{4\pi}}\frac{\omega_0}{c}\vec{A}_n\right)^2 + \omega_f^2 q_{nf}^2\right\} + V_c +$$
$$+ \int dV\left(\frac{E^2 + H^2}{8\pi}\right),$$

(1)

where $\vec{p}_{nf}, \vec{q}_{nf}$ are generalized momentum and the coordinate of molecular cell of number n, which are expressed through Bose operators

$$\vec{q}_{nf} = \sqrt{\frac{\hbar}{2m\omega_f}}\left(b_n + b_n^+\right)\vec{e}_{nf},$$

$$\vec{p}_{nf} = i\sqrt{\frac{m\hbar\omega_f}{2}}\left(b_n^+ - b_n\right)\vec{e}_{nf},$$

here $\vec{e}_{nf}$ is the unit vector; v_0 is the volume of a crystal unit cell; ω_0 is the plasma frequency, ω_f is the oscillation frequency of number f ; V_c is the Coulomb interaction between the molecules in the dipole – dipole approximation [28]; $\vec{A}_n$ is the vector potential at site n; $\vec{E}, \vec{H}$ are the densities of electrical and magnetic field of the external electromagnetic wave, respectively. It should be noted that in the Hamiltonian (1), the kinematic interaction [29] is not taken into consideration due to non – Bose elementary excitations. Such an approximation is justified for the case of four – particle processes only for specially taken resonance conditions that will be discussed further. If (1) is subjected to canonical transformation of the form [31]

$$H' = UHU^{-1}, \tag{2}$$

where

$$U = \exp\left(\frac{i}{\hbar c}\sum_n \vec{d}_n \vec{A}_n\right) \tag{3}$$

and

$$\vec{d}_n = \omega_0 \sqrt{\frac{v_0}{4\pi}}\sum_f \vec{q}_{nf} \tag{4}$$

is the dipole moment of site *n*, the Hamiltonian of a cubic crystal can be represented as [32]

$$H = \frac{1}{2}\sum_{nf}\left(\tilde{p}_{nf}^2 + \omega_f^2 q_{nf}^2\right) +$$

$$+ \int dV \left\{\frac{\left(\vec{D} - 4\pi\vec{P}_L\right)^2 + \vec{H}^2}{8\pi} - \frac{2\pi}{3}P_L^2\right\} - \sum_n \vec{d}_n^{NL}\vec{E}_n^{ef}. \tag{5}$$

In (5) the following designations are introduced:

$$\tilde{\vec{p}}_{nf} = \vec{p}_{nf} - \frac{\omega_0}{c}\sqrt{\frac{v_0}{4\pi}}\vec{A}_n \tag{6}$$

is a new generalized momentum; $\vec{D}$ is the vector of electrical induction; $\vec{P}_L$ is the linear part of crystal polarization shown by the expression

$$\vec{P}_L(\vec{r}) = \sum_n \vec{d}_n \delta(\vec{r} - \vec{r}_n), \tag{7}$$

$\vec{d}_n^{NL}$ is the nonlinear part of the dipole moment);

$$E_{ni}^{ef} = E_{ni} - \frac{4\pi}{v_0}\sum_m Q_{nm}^{ij}d_m^j \tag{8}$$

is a field acting on the molecule, $\vec{E}_n$ is the external field density at site *n*;

$$Q_{nm}^{ij} = N^{-1}\sum_k \exp\left[i\vec{k}(\vec{r}_n - \vec{r}_m)\right]\times$$

$$\times\left\{\frac{1}{3}\delta_{ij} - \sum_g\left[\frac{(\vec{k}+\vec{g})_i(\vec{k}+\vec{g})_j}{(\vec{k}+\vec{g})^2} - \frac{1}{3}\delta_{ij}\right]\right\} \tag{9}$$

is the coefficient of the internal field [28]; indices *i, j* run independent of *x, y, z* values; the summation over $\vec{k}$ and $\vec{g}$ in (9) is done for the first Brillouin zone and in other zones, respectively.

Within the secondary quantization, the first two summands of expression (5) are reduced to diagonal type by transition to the states of mechanical excitons [29] and then to polaritons. In this case, the last summand of (5), which in the approximation of secondary quantization is cubic over the operators of quasi – particles, takes the form

$$H^{III} = N^{-\frac{3}{2}}\sum_{\substack{\vec{k}_1\vec{k}_2\vec{k}_3 \\ \alpha_1\alpha_2\alpha_3}}\left\{R^{(1)}_{\alpha_1\alpha_2\alpha_3}(\vec{k}_1,\vec{k}_2,\vec{k}_3)A^+_{\alpha_1\vec{k}_1}A_{\alpha_2\vec{k}_2}A_{\alpha_3\vec{k}_3} + \right.$$

$$\left. + R^{(2)}_{\alpha_1\alpha_2\alpha_3}(\vec{k}_1,\vec{k}_2,\vec{k}_3)A_{\alpha_1\vec{k}_1}A_{\alpha_2\vec{k}_2}A_{\alpha_3\vec{k}_3} + э.c.\right\} \tag{10}$$

The indices of α run all values corresponding to branches of polaritons $A_{\vec{k}\rho}$ and mechanical excitons $A_{\vec{k}s}$. Later on, from the whole set of the constants $R^{(1)}$ and $R^{(2)}$, only $R^{(1)}_{s_1s_2\rho}$, $R^{(1)}_{\rho_1\rho_2s_1}$ and $R^{(1)}_{s_1\rho_1\rho_2}$ are required, which are written down in the following form:

$$R^{(1)}_{s_1s_2\rho}(\vec{k}_1,\vec{k}_2,\vec{k}_3) = \left(4\pi v_0\right)^3 \sum_{in} e^{i(\vec{k}_2+\vec{k}_3-\vec{k}_1)\vec{r}_n}d^i_{s_1s_2}\varepsilon^i_{\vec{k}_3\rho}, \tag{11}$$

$$R^{(1)}_{\rho_1\rho_2s}(\vec{k}_1,\vec{k}_2,\vec{k}_3) = \left(4\pi v_0\right)^3 \hbar^{-1}\sum_{ins_1j} e^{i(\vec{k}_2+\vec{k}_3-\vec{k}_1)\vec{r}_n}d^j_{0s_1}d^i_{s_1s}{}^*$$

$$*\left[\frac{\varepsilon^i_{\vec{k}_1\rho_1}\varepsilon^j_{\vec{k}_2\rho_2}}{\omega_{s_1} - \omega_{\vec{k}_2\rho_2}} + \frac{\varepsilon^i_{\vec{k}_2\rho_2}\varepsilon^j_{\vec{k}_1\rho_1}}{\omega_{s_1} + \omega_{\vec{k}_1\rho_1}}\right], \tag{12}$$

$$R^{(1)}_{s\rho_1\rho_2}(\vec{k}_1,\vec{k}_2,\vec{k}_3) = \left(4\pi v_0\right)^3 \hbar^{-1}\sum_{ins_1j} e^{i(\vec{k}_2+\vec{k}_3-\vec{k}_1)\vec{r}_n}d^i_{ss_1}d^j_{s_10}{}^*$$

$$*\left[\frac{\varepsilon^i_{\vec{k}_2\rho_2}\varepsilon^j_{\vec{k}_1\rho_1}}{\omega_{s_1} - \omega_{\vec{k}_2\rho_2}}\right], \tag{13}$$

where $d_{s_1s_2}$ is a matrix element of the operator of dipole moment over the states of mechanical excitons; $\vec{\varepsilon}_{\vec{k}\rho}$ is the vector of the electric intensity for a polarition wave in the crystal [29].

As the effect is a four – body one, then the summands which are cubic over the operators of quasi – particles will contribute to the effect of the second order of the perturbation theory, while the fourth – order summands are associated with in the first-order one.

The process is schematically shown in Figure 2. Here the dotted and the straight lines show the condensate and supracondensate excitons, while the wavy line expresses the polaritons. On the supposition that one of the following resonance conditions is satisfied:

$$
\begin{aligned}
&1)\quad E_s \sim \hbar\omega_1 + E_{\sigma 0}, \\
&2)\quad E_\rho(\vec{k}_1) \sim \hbar\omega_1 + E_{\sigma 0}, \\
&3)\quad \hbar\omega_2 \sim E_{\sigma 0} - E_s, \\
&4)\quad \hbar\omega_2 \sim E_{\sigma 0} - E_\rho(-\vec{k}_2)
\end{aligned}
\qquad (14)
$$

In this case the dominating contribution to nonlinear scattering will be formed by the cubic summands of the Hamiltonian (10) in the second order of the perturbation theory, while the contribution of the fourth – order summands may be neglected, a part of which is determined by the kinematic interaction.
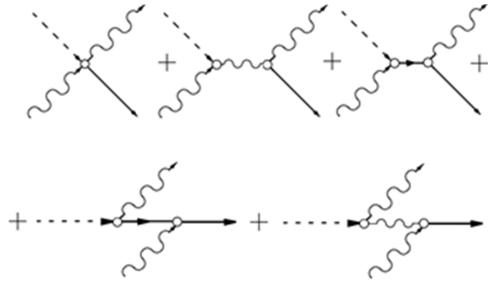

Figure 2. *The scheme of the four-body process sin the first and second order of perturbation theory. The wavy lines stand for polaritons, while the dotted and solid lines correspond to the condensate and supracondensate excitons.*

Now the energy flow of scattered radiation S should be found. Two different situations are considered.

1) Let in the final state, alongside with the polariton, a mechanical exciton on branch ν emerges. The energy of the exciton does not depend on the wave vector [28]. Then, for the energy flow of the scattered radiation to the unit solid angle $d\Omega$, the following expression is written:

$$
\frac{\partial S}{\partial \Omega} = IVn_0 A(\omega_1,\omega_2)\sum_{n=1}^{l_\varpi}\sum_{m=1}^{l_\nu}\left|\sum_{ij} a_{ij}^{nm}(\omega_1,\omega_2)\varepsilon_{\vec{k}_1\rho_1}^i \varepsilon_{\vec{k}_2\rho_2}^j\right|^2, \quad (15)
$$

Where I is the intensity of monochromatic pumping of frequency ω_1; n_0 is the exciton density in the condensate; l_σ, l_ν is the degeneracy multiplicity for the initial (condensate) and final (supracondnsate) excitonic states, respectively; ω_2 is the scattered radiation frequency which satisfies the law of momentum conservation

$$
\hbar\omega_1(\vec{k}_1) + E_{\sigma 0} = \hbar\omega_2(\vec{k}_2) + E_{\nu(\vec{k}_1 - \vec{k}_2)}; \quad (16)
$$

$$
A(\omega_1,\omega_2) = \frac{4\omega_2^3 v_0^2 n_2^2}{\hbar^2 \omega_1 v_1 v_2 c^2}, \qquad (17)
$$

n_i, v_i $(i=1,2)$ is the index of refraction and group velocity of the wave at frequency ω_i; a_{ij}^{nm} is the scattering tensor; indices n and m characterize the initial and final states, respectively.

2) The both quasi – particles emerged at the final state are polaritons. Similar to parametric luminescence or generation of the second harmonic [25], the flow of energy of scattered radiation will highly depend on the scattering angle. Thus, the phase synchronism conditions should be taken into consideration.

Let an isotropic crystal be cut out in the form of a parallel-sided plate of small thickness along OZ axis. Vector $\vec{k}_1 - \vec{k}_2 = \vec{k}_3$ is directed along the same axis. Then, the expression for the energy flow of scattered radiation to the unit solid angle $d\Omega$ in the spectral interval from ω_2 to $\omega_2 + d\omega_2$ is written as follows

$$
\frac{\partial^2 S}{\partial \omega_2 \partial \Omega} = \frac{In_0 L_x L_y A(\omega_1,\omega_2)}{2\pi v_{\rho_3}}\sum_{n=1}^{l_\sigma}\left|\sum_{ij} a_{ij}^{nm}(\omega_1,\omega_2)\varepsilon_{\vec{k}_1}^i \varepsilon_{\vec{k}_2}^j\right| *
$$
$$
*\frac{\sin^2\left(\dfrac{\Delta k_z L_z}{2}\right)}{(\Delta k_z)^2}, \qquad (18)
$$

where L_x, L_y, L_z are linear dimensions of the crystal along axes OX, OY, OZ, respectively; v_{ρ_3} is the group velocity of the polariton associated with wave vector $\vec{k}_3$; $\Delta\vec{k} = \vec{k}_1 - \vec{k}_2 - \vec{k}_3$. For the first resonant situation, tensor a_{ij}^{nm} can be represented as

$$
a_{ij}^{nm}(1) = \sum_s \frac{d_{ns}^i d_{sm}^j}{E_s - \hbar\omega_1 - E_{\sigma 0}}. \qquad (19)
$$

For other resonant conditions we have

$$
a_{ij}^{nm}(2) = \frac{4}{\hbar^2}\sum_{s_1 s_2 l\rho}\frac{d_{ns_1}^i d_{s_1 0}^l d_{0s_2}^l d_{s_2 m}^j \left(\varepsilon_{\vec{k}_1\rho}^l\right)^2}{E_s - \hbar\omega_1 - E_{\sigma 0}}\frac{\left(\omega_{s_1}^2 + \omega_{\vec{k}_1\rho}^2\right)\left(\omega_{s_2}^2 + \omega_{\vec{k}_1\rho}^2\right)}{\left(\omega_{s_1}^2 - \omega_{\vec{k}_1\rho}^2\right)\left(\omega_{s_2}^2 - \omega_{\vec{k}_1\rho}^2\right)}; \quad (20)
$$

$$
a_{ij}^{nm}(3) = \sum_s \frac{d_{ns}^j d_{sm}^i}{E_s - E_{\sigma 0} + \hbar\omega_2}; \qquad (21)
$$

$$
a_{ij}^{nm}(4) = 4\hbar^{-2}\sum_{s_1 s_2 \rho l}\frac{d_{ns_1}^j d_{s_1 0}^l d_{0s_2}^l d_{s_2 m}^i \left(\varepsilon_{-\vec{k}_2\rho}^l\right)^2}{E_\rho(-\vec{k}_2) - E_{\sigma 0} + \hbar\omega_2} *
$$
$$
*\frac{1}{(\omega_{s_1} - \omega_{-\vec{k}_2\rho})(\omega_{s_2} - \omega_{-\vec{k}_2\rho})} \qquad (22)
$$

Let us investigate the tensor of nonlinear scattering. Now

the theoretical group analysis of tensor a_{ij}^{nm} is performed. Suppose that there is the first resonant situation described above. Then $a_{ij}^{nm}(1)$ can be represented in the form

$$a_{ij}^{nm}(1) = < n \,|\, d^i \Lambda \left(\hbar \omega_1 + E_{\varpi 0} \right) d^j \,|\, m > \qquad (23)$$

where

$$\Lambda(\hbar \omega) = \sum_s \frac{|\, s > < s \,|}{E_s - \hbar \omega}. \qquad (24)$$

In the limit $\vec{k} \to 0$, operator Λ is invariant with respect to the point group of the crystal. This group is a quotient group of the space group of the crystal with respect to the translation subgroup. Thus, it is convenient to represent tensor $a_{ij}^{nm}(1)$ in the form

$$a_{ij}^{nm}(1) \sim \int \psi_n^{\sigma} a_{ij} \psi_m^{\nu} d\tau \qquad (25)$$

where a_{ij} is a tensor of rank two; $\psi_n^{\sigma}, \psi_m^{\nu}$ are the n-th and m-th basic wave functions of representations Γ_{σ} and Γ_{ν}, respectively. Non-zero expression (25) is realized only if tensor a_{ij} contains a function transformed in the same way as product $\psi_n^{\sigma} \psi_m^{\nu}$. As the systems of functions $\psi_1^{\sigma}, \psi_2^{\sigma}, ..., \psi_{l_{\sigma}}^{\sigma}$, and $\psi_1^{\nu}, \psi_2^{\nu}, ..., \psi_{l_{\nu}}^{\nu}$, are the basic functions of irreducible representations Γ_{σ} and Γ_{ν} respectively, then their products are the basis in the general case of a reducible representation of dimension $l_{\sigma} l_{\nu}$ These products are irreducible when at least one of l_{σ} or l_{ν} equals unity. In this case, products of the type $\psi_n^{\sigma} \psi_m^{\nu}$ are basic functions of an irreducible representation Γ_{α} determined by the relation

$$\Gamma_{\sigma} \times \Gamma_{\nu} = \Gamma_{\alpha} \qquad (26)$$

Projecting tensor a_{ij} onto the basic function of representation Γ_{α}, one can determine nonzero components of the first summand in expression for $a_{ij}^{nm}(1)$.

Now, let both representations Γ_{σ} and Γ_{ν} be other than one–dimensional. In this case, the set $\psi_n^{\sigma} \psi_m^{\nu}$ should be expanded in basic functions of irreducible representations according to formula [33].

$$\psi_n^{\sigma} \psi_m^{\nu} = \sum_{\alpha, s} \psi_s^{\alpha} \qquad (27)$$

where

$$\psi_s^{\alpha} = \frac{l_{\alpha}}{g} \sum_G G_{ss}^{(\alpha)*} \hat{G} \left(\psi_n^{\sigma} \psi_m^{\nu} \right), \qquad (28)$$

g is a number of group elements; $\hat{G}$ is a symmetry operation; $G_{ss}^{(\alpha)}$ are diagonal elements of transformation matrix. Next, as follows from (25) and (26), it is necessary to project a_{ij} on every function ψ_s^{α} (27) and to add the produced matrices together.

Tensor $a_{ij}(3)$ (21) is analyzed in the same way.

Now it is assumed that only the second resonant situation is correct. In this case, tensor $a_{ij}^{nm}(2)$ can be represented as

$$a_{ij}^{nm}(2) \sim \sum_{l\rho} \frac{< n \,|\, d^i d^l \,|\, 0 > < 0 \,|\, d^l d^j \,|\, m > \left(\varepsilon_{\vec{k}_1 \rho}^l \right)^2}{E_{\rho}(\vec{k}_1) - \hbar \omega_1 - E_{\sigma 0}} \qquad (29)$$

As follows from (29), $a_{ij}^{nm}(2)$ is a convolution of two second-rank tensors projected on the irreducible representation Γ_{σ} and Γ_{ν}, respectively. The appropriate tables are given in [34]. Analysis of $a_{ij}^{nm}(4)$ should be done in the same way.

As an example, the scattering in crystal of symmetry T_h is considered. Let $|\, n >$ and $|\, m >$ be transformed as the X - th component of the axial vector. Then, product $|\, n > |\, m >$ is also transformed as $|\, n >$, and tensor $a_{ij}^{nm}(1)$ will have the form [27]

$$\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & a \\ 0 & a & 0 \end{pmatrix} \qquad (30)$$

To establish the type of $a_{ij}^{nm}(3)$, it is necessary to find the square of matrix (30). Thus,

$$\begin{pmatrix} 0 & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & b \end{pmatrix} \qquad (31)$$

As seen from (30) and (31), the form of the scattering tensor is varied under different resonant situations.

3. Some Corollaries of Laws of Conservation for a Four – Body Process

As follows from expression (18), in the case when condensate and supracondensate quasi – particles are at the upper polariton branch, the energy flow of scattered radiation will be maximal if the laws of conservation operate simultaneously.

$$\begin{cases} \hbar\omega_1(\vec{k}_1) + E_{\sigma 0} = \hbar\omega_2(\vec{k}_2) + E_{\sigma\vec{k}_3} \\ \vec{k}_1 = \vec{k}_2 + \vec{k}_3 \end{cases} \quad (32)$$

Taking into account [29] that $|\vec{k}_i| = \dfrac{\omega_i}{c} n_i$ (where $n_i = n(\omega_i)$ is the index of refraction at the appropriate frequency), from system (32), scattering angle θ is found, which satisfies the conditions of phase synchronism

$$\sin\frac{\theta}{2} = \frac{(\omega_1 - \omega_2 + \omega_\sigma)^2 n_\sigma^2 - (\omega_1 n_1 - \omega_2 n_2)^2}{4 n_1 n_2 \omega_1 \omega_2} \quad (33)$$

In (33), n_σ corresponds to frequency $\omega_\sigma = \hbar^{-1} E_{\sigma\vec{k}_3}$. $E_{\sigma\vec{k}_3} > E_{\sigma 0}$ then in direction of the maximal intensity of scattered radiation, the scattering spectrum is similar to that represented in Figure 3, where I is an intensity of scattered radiation; lines of frequencies $(\omega_1 - E_\nu \hbar^{-1})$ and ω_2 respond to ordinary combination scattering and condensate – aided scattering, respectively. Moreover, due to the δ - shaped function of the condensate exciton distribution over the quasi – impulses, there is a coincidence in the spectral forms of incident and scattered radiation, in the case of narrow band pumping.
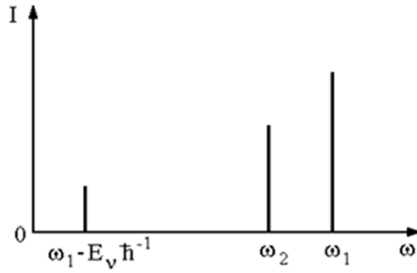

Figure 3. *Frequency spectrum of the process. I is the intensity of the scattered radiation; the lines at the frequencies of $(\omega_1 - E_\nu \hbar^{-1})$ and ω_2 are associated with conventional combination scattering and scattering with the condensate involved.*

Now the case is considered when an exciton originates in the final state. In the crystal, the energy of real excitons depends on the wave vector. Therefore it is necessary to take into account the presence of synchronism and to determine directions of the largest energy transformation by using the system (32). To be definite, it is assumed that the law of exciton dispersion is parabolic, i.e.

$$E(\vec{q}) = E(0) + \frac{\hbar^2 q^2}{2m}, \quad (34)$$

where m is effective mass. For polaritons, the relation $\omega = |\vec{k}| c n^{-1}$ is still used
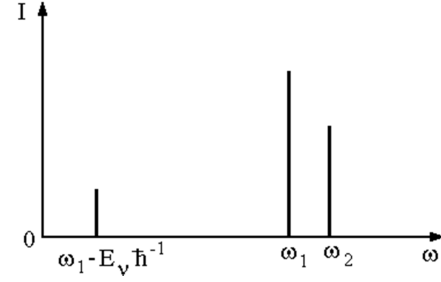

Figure 4. *Spectrum in the case of $E_{\nu\vec{q}} < E_{\sigma 0}$.*

Let us study some cases.
1. Condensate and supracondensate excitons belong to different branches σ and ν. According to the law of energy conservation, for $E_{\nu\vec{q}} > E_{\sigma 0}$, the scattering spectrum will look like that of Fiure 3. For $E_{\nu\vec{q}} < E_{\sigma 0}$, the spectrum is shown in Figure 4.

Similar to the effect of second – harmonic generation [35], the cases of collinear and vector phase synchronisms are probable. Let us first consider the collinear phase synchronism.

1. Forward scattering. In this case, the wave vectors of pumping and of scattered radiation are collinear and unidirectional. Then, by elimination of one equation from system (32) with condition of (34) taken into account, we have

$$\left[\omega_1 n_1(\theta, \phi) - \omega_2 n_2(\theta, \phi)\right]^2 = \frac{2mc^2}{\hbar^2}\left[\hbar\omega_1 - \hbar\omega_2 + E_{\sigma 0} - E_{\nu 0}\right] \quad (35)$$

where θ and ϕ are polar angles characterizing the direction of wave vectors. In formula (35), the left – hand part is a function of θ and ϕ, while the right –hand one is not. As, in the general case, $n(\theta, \phi)$ is a rather complex surface, it is clear that (35) will be satisfied only in preferred directions of $\vec{k}$ that should be calculated for each crystal with using parameters determining the shape of the surface of indices. In a cubic crystal, the surface $n(\theta, \phi)$ is spherical and equation (35) can only be exceptionally satisfied for specially selected values of $\omega_1, \omega_2, E_{\sigma 0}$.

2. Back scattering. Wave vectors $\vec{k}_1$ and $\vec{k}_2$ are collinear as well but their direction is opposite. From (32) after simple transformations, we have

$$\left[\omega_1 n_1(\theta, \phi) + \omega_2 n_2(\pi - \theta, \pi - \phi)\right]^2 = \frac{2mc^2}{\hbar^2}\left[\hbar\omega_1 - \hbar\omega_2 + E_{\sigma 0} - E_{\nu 0}\right] \quad (36)$$

(36) and (35) are satisfied only in certain directions. Hence, in cases of collinear arrangement of wave vectors $\vec{k}_1$ and $\vec{k}_2$, the intensity of scattered radiation will be essentially dependent on the angles between $\vec{k}_1$ and optical axes.

It should be noted that in the case when the energy of a "supracondensate "exciton is independent of the wave vector, i.e. $m = \infty$, the system of equations (32) has got the solution for any directions of incident and scattered radiation.

3. Vector synchronism. In this case, the solution of system (32) is of the form

$$\sin^2\frac{\theta}{2} = \frac{2mc^2\hbar^{-1}}{\hbar}\left[\frac{(\omega_1 + \omega_\sigma - \omega_2 - \hbar^{-1}E_{\nu 0}) - (\omega_1 n_1 - \omega_2 n_2)^2}{4n_1 n_2 \omega_1 \omega_2}\right] \tag{37}$$

To study the nonlinear effect in cubic crystals, equation (37) was used for finding the scattering angles. Let the frequency of incident radiation be $\omega_1 = 3.66 \cdot 10^{15}\, sm^{-1}$. Then, for crystals of *ZnTe* type (where $n=3.23$; $m = 2.83 \cdot 10^{-28}\, g$; $\omega_\sigma = 3.61 \cdot 10^{15}\, sm^{-1}$), the following results are obtained (see Table 1).

Table 1. *The dependence of the scattering angle of the frequency vector in synchronism.*

θ (degree)	$\omega_2 \cdot 10^{15}\, sm^{-1}$	θ (degree)	$\omega_2 \cdot 10^{15}\, sm^{-1}$
24,08	3.66215	102.04	3.6615
34,4	3.66210	154.8	3.6611
49,3	3.6620	169.7	3.66105

As it can be seen from the table, the scattering angle is highly dependent on frequency ω_2, i.e. the scattered radiation is very much dispersed.

II. Condensate and supracondensate excitons are at identical branches.

The case of $m = \infty$ is excluded. If this requirement is satisfied, then, as it follows from the first equation in (32), $\omega_1 = \omega_2$ and the line with the participation of condensate would coincide with the Rayleigh one. Suppose that $\partial E_\nu / \partial q > 0$. Then the scattering spectrum will be similar to that in Figure 3. Analogously to it. 1, system (32) not always has got the solution in this case. The angles associated with the collinear phase synchronism with forward and backward scattering are determined from equations (35) and (36). In the right - hand parts of those equations, it is assumed that $\sigma = \nu$. When $\partial E_\nu / \partial q < 0$, the spectrum is similar to the represented in Figure 4.

Let the case of "easy" excitons be considered. Then lines of ω_1 and ω_2 (Figure 3 and 4) will be very close. They could only be distinguished in the case when the frequency shift $(\omega_1 - \omega_2)$ is higher than the instrument resolution $\Delta\nu$, i.e. when the inequality

$$\hbar(2\pi mc)^{-1}(n_1^2\omega_1^2 + n_2^2\omega_2^2 - 2n_1 n_2 \omega_1 \omega_2 \cos\theta) > \Delta\nu \tag{38}$$

is satisfied. Here θ is the scattering angle. From (38) it follows that the most favorable conditions for the effect of nonlinear scattering with the participation of condensate are the backward geometry and low effective masses of a supracondensate exciton.

4. Shape of a Scattered Radiation Line

Above, only the frequency dependence of scattering

tensors was considered. However, near the bands of exciton absorption, the question of the spectral shape of scattered radiation line becomes very important.

As it is known [36], within the interaction representation, time dependence of probability amplitudes in state $|n>$ is determined by the formula

$$i\hbar b_n(t) = \sum_m V_{nm} b_m(t) e^{i(E_n - E_m)\hbar^{-1}t} \tag{39}$$

where V_{nm} is a matrix element of perturbation operator; t is time. Suppose that the first resonance situation is valid. Then the effect goes as follows: first, condensate exciton $E_{\sigma 0}$ and polariton $\hbar\omega_1$ are annihilated. Because of this fact, a mechanical exciton E_{s_1} is created. After that it is decomposed to polariton $\hbar\omega_2$ and exciton E_s. In this case, the energy $\hbar\omega_1 + E_{\sigma 0}$ coincides with that of mechanical exciton E_{s_1}. To solve (39), we use a Fourier transform

$$b_n(t) = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} dE\, G_{n0}(E)\exp\left\{i(E_n - E)\hbar^{-1}t\right\} \tag{40}$$

For convenience, the multiplier G_{00} is separated, then

$$G_{n0}(E) = u_{n0}(E)G_{00}(E)\varsigma(E - E_n) \quad (n \neq 0) \tag{41}$$

where

$$\varsigma(E) = \frac{P}{E} - i\pi\delta(E) \tag{42}$$

P means that the corresponding expression is taken in the sense of the principal value. For function u_{n0}, the relations resulting from substitution of (41) into (40) are valid:

$$u_{s_1,\sigma\rho_1} = H_{s_1,\sigma\rho_1} + \sum_{s\rho_2} H_{s_1,s\rho_2} u_{s\rho_2 0}(E)\varsigma(E - E_{s\rho_2}), \tag{43}$$

$$u_{s\rho_2,\sigma\rho_1} = H_{s\rho_2,s_1} u_{s_1,\sigma\rho_1}(E)\varsigma(E - E_{s_1}) \tag{44}$$

Substituting (44) into (43), we find

$$u_{s\rho_2,\sigma\rho_1}(E) = H_{s_1,\sigma\rho_1} - \frac{i\hbar\gamma}{2} u_{s_1,\sigma\rho_1}(E)\varsigma(E - E_{s_1}), \tag{45}$$

where

$$\frac{\hbar}{2}\gamma(E) = i\sum_{s\rho_2}\left|H_{s\rho_2,s_1}\right|^2 \varsigma(E - E_{s\rho_2}). \tag{46}$$

Multiplying (45) by $E - E_{s_1}$,

$$u_{s_1,\sigma\rho_1}(E) = \frac{(E - E_{s_1})H_{s_1,\sigma\rho_1}}{(E - E_{s_1}) + \dfrac{i\hbar}{2}\gamma(E)}, \qquad (47)$$

and from (44)

$$u_{s_1,\sigma\rho_1}(E) = \frac{H_{s\rho_2,s_1}H_{s_1,\sigma\rho_1}}{(E - E_{s_1}) + \dfrac{i\hbar}{2}\gamma(E)}, \qquad (48)$$

Hence, for $t \to \infty$

$$b_{s_1}(\infty) = \frac{u_{s_1,\sigma\rho_1}(E_{s_1})}{E_{s_1} - E_{\sigma\rho_1} + \dfrac{i}{2}\hbar\Gamma(E_{s_1})} \qquad (49)$$

$$b_{s\rho_2}(\infty) = \frac{u_{s\rho_2,\sigma\rho_1}(E_{s\rho_2})}{E_{s\rho_2} - E_{\sigma\rho_1} + \dfrac{i}{2}\hbar\Gamma(E_{s\rho_2})} \qquad (50)$$

Where

$$E_{s\rho_2} = E_s + \hbar\omega_1, \qquad E_{\sigma\rho_1} = E_{0\sigma} + \hbar\omega_1,$$

$$\frac{\hbar}{2}\Gamma(E) = i\sum_{s_1\rho_1}\frac{\left|H_{s_1,\sigma\rho_1}\right|^2}{E - E_{s_1} + \dfrac{i}{2}\hbar\gamma(E)} \qquad (51)$$

With (50) squared, the scattering probability is

$$\left|b_{s\rho_2}(\infty)\right|^2 = \frac{\left|H_{s\rho_2,s_1}\right|^2\left|H_{s_1,\sigma\rho_1}\right|^2}{\left[E_{s\rho_2} - E_{\sigma\rho_1} + \dfrac{i\hbar}{2}\Gamma(E_{s\rho_2})\right]^2\left[E_{s\rho_2} - E_{s_1} + \dfrac{i\hbar}{2}\Gamma(E_{s\rho_2})\right]^2} \qquad (52)$$

As it follows from (43), for $E \sim E_{s_1}$, magnitude γ is, in fact, a probability of decomposition of exciton E_{s_1} into polariton $\hbar\omega_2$ and exciton E_s. It determines the radiation broadening of excitonic levels s_1. Magnitude $\Gamma(E)$ (51) is a broadening of $E_{\sigma\rho_1}$ state due to its finite lifetime for $E = E_{\sigma\rho_1}$. It follows that the real and imaginary parts of Γ and γ are responsible for the broadening of the related levels and the shifts, respectively. The latter values are not of interest. Now, we replace γ and Γ with their real parts and denote them as γ and Γ once again. Then

$$\gamma(E) = \frac{2\pi}{\hbar}\sum_{s\rho_2}\left|H_{s\rho_2,s_1}\right|^2\delta(E - E_{s\rho_2}) = \frac{2\pi}{\hbar}\int d\Omega_{\rho_2}d\Omega_s\left|H_{s\rho_2,s_1}\right|^2\rho(E_s)\rho(\hbar\omega_2),$$

$$\Gamma(E) = \gamma\sum_{s,\rho_1}\frac{\left|H_{s_1,\sigma\rho_1}\right|^2}{(E - E_{s_1})^2 + \dfrac{\hbar^2\gamma^2}{4}}. \qquad (53)$$

Here Ω_n is a solid angle with the emitted quantum E_n; $\rho(E_n)$ is the density of states.

Now two cases are studied.

1. A crystal is exposed to broad – band radiation of intensity I which is the constant in the region of radiation broadening of level s. As I is constant, $\displaystyle\sum_{\rho_1}$ can be replaced by the integral. In the limit of $\gamma \to 0$, with only one level$s$ taken into account, we have

$$\Gamma = \frac{V}{(2\pi)^2}\int d^3k_1\left|H_{s_1,\sigma\rho_1}\right|^2\delta\left(E_{\sigma\rho_1} - E_{s_1}\right) = \frac{2\pi IVn_0}{\hbar^2\omega_1 v_1}\left|H_{s_1,\sigma\rho_1}\right|^2\rho(\omega_1), \qquad (54)$$

where $\rho(\omega)$ is density of states; I is primary intensity; $\hbar\omega_1 = E_{s_1} - E_{\sigma 0}$; v_1 is the group velocity of polariton $\hbar\omega_1$. Expression (54) performs the total probability of polariton $\hbar\omega_1$ and condensate exciton $E_{\sigma 0}$ absorption per unit time. So, Γ could be considered to be small enough. As follows from (52), for small Γ, the first multiplier of denominator is

a δ - function. Thus, the difference $(E_{s\rho_2} - E_{\sigma\rho_1})$ is different from zero only by $\hbar\Gamma$, i.e. the law of energy conservation is valid for scattering.

Consider now the shapes of absorption and scattering lines. For probability of emission, it follows from (52) and (54):

$$\sum_{\rho_1} \left| b_{s\rho_1}(\infty) \right|^2 = \frac{\left| H_{s\rho_2,s_1} \right|^2}{(E_{s\rho_2} - E_{s_1})^2 + \dfrac{\hbar^2\gamma^2}{4}} \qquad (55)$$

The expression coincides with the shape of line for spontaneous irradiation. For absorption probability we have

$$\sum_{\rho_2} \left| b_{s\rho_2}(\infty) \right|^2 = \frac{\gamma}{\Gamma} \frac{\left| H_{s\rho_2,s_1} \right|^2}{(E_{s\rho_2} - E_{s_1})^2 + \dfrac{\hbar^2\gamma^2}{4}} \qquad (56)$$

As seen from (55), (56) the shapes of absorption and emission lines coincide. It can be concluded that when primary intensity is a broad – band and constant in the region of radiation broadening of the resonance level E_{s_1} , the process behaves in the same way as in the case of two independent processes, i.e. absorption and emission (as compared to the shape of the absorbed and emitted lines). However, the formula (52) for probability of the process does not coincide with the product of emission and absorption probabilities, since it does not connect energies $E_{\sigma\rho_1}$ and E_{s_1} by a δ - function.

2. A crystal is exposed to narrow – band radiation, the linewidth being small as compared to radiation broadening level s_1 . Then it can be supposed that the primary intensity is nonzero only at frequency ω_1 . The intensity of emitted line is obtained by making the summation (52) over ρ_1 . In denominator, the first multiplier is a δ - function. Hence, $E_{s\rho_2} = E_{\sigma\rho_1}$. The integration gives

$$\sum_{\rho_1} \left| b_{s\rho_2}(\infty) \right|^2 = \frac{2\pi I(\omega_1) n_\sigma \left| H_{s\rho_2,s_1} \right|^2 \left| H_{s_1,\sigma\rho_1} \right|^2 \rho(\omega)}{\hbar^2 \Gamma v_1 \left[\left(E_{s_1} - E_{\sigma\rho_1} \right)^2 + \dfrac{\hbar^2\gamma^2}{4} \right]} \qquad (57)$$

It follows from (57) that as the denominator is constant in the area where $I(\omega)$ is nonzero, the shape of emission line is defined by the primary intensity. The emitted line should be of the same width as pumping. Thus, it can be concluded that during the excitation by a narrow spectral line, the scattering process should be considered as a single act, not two consecutive independent acts.

Some other resonant situations could be considered in this way. But in the case of conditions №2 and №4, in the intermediate state, a polariton wave originates and this fact should be taken into account. In those cases, the corresponding small denominator is provided by the choice of both the pumping frequency and the direction of radiation propagation. The value similar to γ (46) will characterize the line shape for a wave originating in the intermediate state.

The calculations similar to the present ones confirm that if a narrow – band radiation is falling (incident) on the crystal and none of frequencies satisfies the conditions of synchronism, the process should be treated as a single one. If one of the frequencies satisfies the conditions of synchronism and pumping is broad – band and of constant intensity in the region of wave broadening in the intermediate state, the process should be treated as a cascade one.

5. Conclusions

After obtaining of exciton polariton condensates in the scientific community, an intense studies both theoretical and experimental aspects of this phenomenon are developed [38-43]. Experimental observation of the considered four-body process in the crystal requires BEC of excitons to be formed. At the same time, conditions of collinear and phase synchronisms should be taken into account. As a consequence of theoretical group analysis of the scattering tensor, both dipole excitons that are able to interact with the light (polaritons) and dipole-less excitons can participate in afour-body process. Besides, detection of BEC in the state of $k=0$ becomes possible, contrary to the case of luminescence application. It should be noted that non-linear effects can be also applied to the existence proof of excitons BEC in one-dimensional non-ideal array of coupled microcavities containingquantum dots [44] where the minima of the polaritons energy in the states of $k \neq 0$ are identified.

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