

# Bose-Einstein condensation of dipole-active excitons and photons

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The interaction of excitons with each other and with light in a stationary nonequilibrium state is considered with the assumptions that the former interaction exceeds the latter and that the exciton-phonon interaction is negligibly small. The chemical potential, which determines the frequency of the proper vibration in which macroscopic numbers of coherent excitons and photons participate, and the spectrum of the elementary excitations in the presence of the condensate-photon mode are found. Those ranges of variation of the wave vectors of the elementary excitations are found for which there is an absolute instability in the system and the possibility of creating a laser or phaser involving degenerate excitons with  $k=0$ . A spatially nonuniform distribution of coherent excitons participating in the formation of a quantum vortex and in translational motion along the axis of the latter is considered. It is shown that, if the dipole moment of the transition is parallel to the vortex axis, the electromagnetic wave propagating along the axis is almost longitudinal with, in the vicinity of the vortex core, non-zero transverse components of the field intensity which fall off slowly with increasing distance from the axis. This phenomenon is analogous to the formation of light filaments. If the moment is circularly polarized in a plane perpendicular to the vortex axis, the wave is almost transverse and a shadow filament is formed in the center of the light field. If the size of the vortex is restricted, the wave will propagate in the form of a light tube.

## 1. INTRODUCTION AND FORMULATION OF THE PROBLEM

The Bose-Einstein condensation of excitons in crystals has been studied theoretically in many papers<sup>[1-8]</sup>. However, up to now there have been no convincing experimental proofs of its appearance. Partly, this is connected with the fact that the predictions of the theory still do not encompass the optical manifestations of this state to an adequate extent. This should be expected, since in the discussion of the optical properties of crystals in the presence of a Bose-Einstein condensate the light has usually been taken into account only as the cause of quantum transitions and not as influencing the formation of the energy spectrum<sup>[1,3,6,7]</sup>. The papers of the authors<sup>[1b,7,9]</sup> and of Keldysh<sup>[8]</sup> are an exception. In particular, it was shown in<sup>[1b,7]</sup> that a wave of a Bose condensate of dipole-active excitons with wave vector  $\mathbf{k}$  generates a coherent electromagnetic wave with macroscopic amplitude and with the same wave vector  $\mathbf{k}$ . The interaction of these waves leads to the establishment of either of two possible condensate-photon modes. The frequency spectrum of these modes was found and it was shown that application of the Bogolyubov displacement procedure to the exciton operators entails the necessity of applying the displacement procedure to the photon operators also.

In the present work<sup>[9]</sup>, the structure of the condensate-photon modes and the spectrum of the elementary excitations of a uniform condensate-photon system are investigated.

Keldysh<sup>[8]</sup> has considered the coherent states of the exciton-photon system, taking into account the Fermi nature of the electrons and holes forming the excitons. The equations which he obtained for the general case of a non-uniform distribution of excitons in space will be used by us to investigate quantum vortices in a medium of dipole-active excitons and photons.

Of fundamental importance in the study of the proper-

ties of a system of excitons, photons and phonons is the relation between the frequencies  $\nu_{\text{ex-ex}}$ ,  $\nu_{\text{ex-ph}}$  and  $\nu_{\text{ex-latt}}$  of the following processes respectively: 1) collisions of excitons with each other, 2) transformation of light into dipole-active excitons, and 3) scattering of excitons by phonons. We shall assume the relaxation times corresponding to these processes to be much shorter than the exciton lifetime and the time the photons stay in the crystal, the polished end-faces of which can play the role of resonator mirrors. We shall not take into account the effect of phonons on the collective properties of the excitons and photons, assuming that

$$\nu_{\text{ex-ex}}, \nu_{\text{ex-ph}} \gg \nu_{\text{ex-latt}}$$

These inequalities are apparently fulfilled at sufficiently high exciton concentrations and low temperatures in a CdS crystal, in which polariton effects are noticeable. Our results refer to the limiting case (the opposite limiting case is discussed very briefly)

$$\nu_{\text{ex-ex}} > \nu_{\text{ex-ph}} \quad (1)$$

In addition, we assume that the exciton concentration  $n_e$  is not too great, i.e.,  $n_e a_e^3 \ll 1$  ( $a_e$  is the exciton radius), so that they do not break each other up. The kinematic interaction arising because of the Fermi nature of the electrons and holes displaces the exciton level<sup>[4]</sup> in the same way as does the dynamic interaction. Both interactions can be taken into account by renormalizing the exciton interaction constant.

## 2. STRUCTURE AND FREQUENCY SPECTRUM OF THE COHERENT MODE IN A UNIFORM SYSTEM OF EXCITONS AND PHOTONS

For simplicity, we shall consider dipole-active transverse excitons and photons with a definite polarization. The Hamiltonian of the system is chosen to conserve the total number of excitons and photons:

$$H = \sum_{\mathbf{q}} (\Delta + T_{\mathbf{q}} - \mu_k) a_{\mathbf{q}}^+ a_{\mathbf{q}} + \sum_{\mathbf{q}} (\hbar c q - \mu_k) c_{\mathbf{q}}^+ c_{\mathbf{q}} + \sum_{\mathbf{q}} \varphi_{\mathbf{q}} (a_{\mathbf{q}}^+ c_{\mathbf{q}} - c_{\mathbf{q}}^+ a_{\mathbf{q}})$$

$$+ \frac{1}{2V} \sum_{\mathbf{q}, \mathbf{q}', \mathbf{q}_1, \mathbf{q}_2} \nu(\mathbf{q}, -\mathbf{q}') \delta_{\mathbf{K}\Gamma}(\mathbf{q}_1 + \mathbf{q}_2 - \mathbf{q}, -\mathbf{q}_2' - \mathbf{q}_1') a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2}^+ a_{\mathbf{q}_1'} a_{\mathbf{q}_2'}, \quad (2)$$

where  $a_{\mathbf{q}}^+$ ,  $a_{\mathbf{q}}$  and  $c_{\mathbf{q}}^+$ ,  $c_{\mathbf{q}}$  are the creation and destruction operators for excitons and photons,  $\Delta$  is the energy of formation of an exciton with  $\mathbf{q}=0$ ,  $T_{\mathbf{q}}$  is the exciton kinetic energy,  $\nu(\mathbf{q})$  is the Fourier transform of the exciton-exciton interaction energy,  $V$  is the volume of the system,  $\delta_{\mathbf{K}\Gamma}$  is the Kronecker delta symbol and  $\varphi_{\mathbf{q}} = \frac{1}{2} i (\hbar f c q / \Delta)^{1/2} \hbar \omega_p$ ;  $f$  is the oscillator strength of the dipole transition from the ground state to the exciton state and  $\omega_p$  is the plasma frequency of the background electrons of the crystal. In the Hamiltonian, a single chemical potential  $\mu_{\mathbf{k}}$  has been introduced for the excitons and photons.<sup>1)</sup> It depends on the wave vector  $\mathbf{k}$  of the coherent state.

Antiresonance terms of the type  $a_{\mathbf{k}} c_{-\mathbf{k}}$ ,  $a_{\mathbf{k}}^+ c_{-\mathbf{k}}^+$ , etc., are not taken into account in (2), since they do not conserve the total number of particles. However, it is necessary to take them into account to obtain the correct dependence of the energy spectrum near  $\mathbf{q}=0$ . This question was discussed in detail earlier<sup>[1, 10, 11]</sup>.

The choice of a single-mode resonator with optimum conditions for the existence of an axial light mode with wave vector  $\mathbf{k}$  implies at the same time a choice of wave vector of the Bose condensate of dipole-active excitons that is independent of the method of exciting them. A method of ordering the excitons with respect to their momenta under the action of light with a given wave vector was proposed by one of the authors<sup>[12]</sup>. The creation of a coherent state of excitons and photons with given values of  $\mathbf{k}$  and  $\mu_{\mathbf{k}}$  can also be achieved in the two-photon absorption of two laser beams with frequencies  $\omega_1$  and  $\omega_2$  and wave vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$  satisfying the conditions

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}, \quad \hbar\omega_1 + \hbar\omega_2 = \mu_{\mathbf{k}}.$$

This method was applied successfully to detect polaritons in CuCl crystals<sup>[12]</sup>.

In the case (1) for low temperatures and predominant repulsion, Bose condensation of the mechanical excitons into a state with well-defined wave vector  $\mathbf{k}$  is possible. We shall take this into account by the Bogolyubov displacement procedure on the operators  $a_{\mathbf{q}}$  and  $c_{\mathbf{q}}$ :

$$\begin{aligned} a_{\mathbf{q}} &= N_{\mathbf{k}}^{\hbar} \delta_{\mathbf{K}\Gamma}(\mathbf{q}, \mathbf{k}) \exp(-i\theta_{\mathbf{k}}) + \alpha_{\mathbf{q}}, \\ c_{\mathbf{q}} &= F_{\mathbf{k}}^{\hbar} \delta_{\mathbf{K}\Gamma}(\mathbf{q}, \mathbf{k}) \exp(-i\Phi_{\mathbf{k}}) + \xi_{\mathbf{q}}, \end{aligned} \quad (3)$$

where the amplitudes  $N_{\mathbf{k}}$  and  $F_{\mathbf{k}}$  are proportional to  $V$ . We substitute (3) into (2) and write out the transformed Hamiltonian  $\tilde{H}$  in terms quadratic in the operators  $\alpha_{\mathbf{q}}$  and  $\xi_{\mathbf{q}}$ :

$$\tilde{H} = E_0(\mu_{\mathbf{k}}) + \tilde{H}_2, \quad E_0(\mu_{\mathbf{k}}) = -\frac{1}{2} l_0 N_{\mathbf{k}},$$

$$\begin{aligned} \tilde{H}_2 &= \sum_{\mathbf{q}} (\Delta + T_{\mathbf{q}} + l_0 + l_{\mathbf{q}-\mathbf{k}} - \mu_{\mathbf{k}}) \alpha_{\mathbf{q}}^+ \alpha_{\mathbf{q}} + \sum_{\mathbf{q}} (\hbar c q - \mu_{\mathbf{k}}) \xi_{\mathbf{q}}^+ \xi_{\mathbf{q}} \\ &+ \sum_{\mathbf{q}} \varphi_{\mathbf{q}} (\alpha_{\mathbf{q}}^+ \xi_{\mathbf{q}} - \xi_{\mathbf{q}}^+ \alpha_{\mathbf{q}}) + \frac{1}{2} \sum_{\mathbf{q}} l_{\mathbf{q}-\mathbf{k}} (\alpha_{\mathbf{q}}^+ \alpha_{-\mathbf{q}+2\mathbf{k}} + \alpha_{-\mathbf{q}+2\mathbf{k}} \alpha_{\mathbf{q}}); \\ l_{\mathbf{q}} &= \nu(\mathbf{q}) N_{\mathbf{k}} / V, \quad \hbar\Omega_{1\mathbf{k}} = \mu_{\mathbf{k}} - \Delta - T_{\mathbf{k}} - l_0, \quad \hbar\Omega_{2\mathbf{k}} = \mu_{\mathbf{k}} - \hbar c k. \end{aligned} \quad (4)$$

From the requirement that the coefficients of the terms linear in the operators  $\alpha_{\mathbf{k}}$  and  $\xi_{\mathbf{k}}$  vanish, we find  $\mu_{\mathbf{k}}$ ,  $N_{\mathbf{k}}/F_{\mathbf{k}}$ ,  $\Omega_{1\mathbf{k}}$  and  $\Omega_{2\mathbf{k}}$  and the phase difference  $\Phi_{\mathbf{k}} - \theta_{\mathbf{k}}$ :

$$\hbar^2 \Omega_{1\mathbf{k}} \Omega_{2\mathbf{k}} = |\varphi_{\mathbf{k}}|^2, \quad N_{\mathbf{k}}/F_{\mathbf{k}} = \Omega_{2\mathbf{k}} / \Omega_{1\mathbf{k}}, \quad \Phi_{\mathbf{k}} - \theta_{\mathbf{k}} = \pi/2. \quad (5)$$

$$\mu_{\mathbf{k}} = \frac{1}{2} \{ \Delta + l_0 + T_{\mathbf{k}} + \hbar c k \pm [ (\Delta + l_0 + T_{\mathbf{k}} - \hbar c k)^2 + 4 |\varphi_{\mathbf{k}}|^2 ]^{1/2} \}. \quad (6)$$

The expression for  $\mu_{\mathbf{k}}$  resembles the polariton spec-

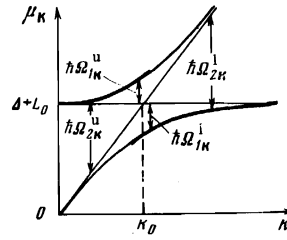


FIG. 1. Frequency spectrum  $\mu_{\mathbf{k}}$  of the condensate-photon modes with neglect of the antiresonance terms. For the upper branch,  $\Omega_{1\mathbf{k}}^u > 0$ ; for the lower branch,  $\Omega_{1\mathbf{k}}^l < 0$ . Along the thickened lines (before the intersection with the vertical dashed line),  $N_{\mathbf{k}} \geq F_{\mathbf{k}}$ .  $k_0$  is the wave vector at which the spectra of the photons and of the mechanical excitons intersect.

trum and determines the vibrational eigenfrequencies of the two possible condensate-photon modes. Precisely which mode is realized experimentally depends on the external conditions, e.g., on the resonator, pumping, etc. Macroscopic numbers of excitons and photons take part in the vibration selected. The structure of this self-consistent, strongly excited vibration depends on  $\mu_{\mathbf{k}}$  and is determined by the ratio  $\Omega_{2\mathbf{k}}/\Omega_{1\mathbf{k}}$ . The possible values of  $\mu_{\mathbf{k}}$  for the various  $\mathbf{k}$  lie along the upper and lower polariton-like branches (see Fig. 1).  $\Omega_{1\mathbf{k}}$  and  $\Omega_{2\mathbf{k}}$  are equal to the distances, at the same  $\mathbf{k}$ , from the given branch of the chemical potential to the mechanical-exciton branch and photon branch respectively.

The ground-state energy  $E_0(\mu_{\mathbf{k}})$  for a given  $\mathbf{k}$  depends only on  $N_{\mathbf{k}}$ . For a given  $\mathbf{k}$ ,  $N_{\mathbf{k}} > F_{\mathbf{k}}$  along one branch of the chemical potential, and  $N_{\mathbf{k}} < F_{\mathbf{k}}$  along the other, with the condition that  $N_{\mathbf{k}} + F_{\mathbf{k}} = \text{const}$ . For a constant level of pumping,  $E_0(\mu_{\mathbf{k}})$  is a minimum along the portions where  $N_{\mathbf{k}} > F_{\mathbf{k}}$ . In Fig. 1 these portions are drawn with thicker lines. The coherent vibrations at the frequencies corresponding to these are predominantly mechanical. Only at one point ( $\hbar c k_0 = \Delta + l_0$ ) do we have  $N_{\mathbf{k}_0} = F_{\mathbf{k}_0}$  for both values of  $\mu_{\mathbf{k}_0}$ . We note that we are confining ourselves to the case  $T=0$ , and so the free energy coincides with the ground-state energy.

We shall take into account the exciton-level damping  $\gamma$  due to the finite exciton lifetime  $\tau_e = (2\gamma)^{-1}$ , the photon damping  $\kappa$  due to the finite time that the light stays in the resonator, and the pumping  $gV$  of the excitons in the condensate. In this case, the equations of motion for the operators  $a_{\mathbf{k}}$  and  $c_{\mathbf{k}}$  have the form

$$\begin{aligned} i\hbar \frac{da_{\mathbf{k}}}{dt} &= \left( \varepsilon_{\mathbf{k}} - i\hbar\gamma + \frac{i\hbar gV}{2N_{\mathbf{k}}} \right) a_{\mathbf{k}} + \varphi_{\mathbf{k}} c_{\mathbf{k}}, \\ i\hbar \frac{dc_{\mathbf{k}}}{dt} &= (\omega_{\mathbf{k}} - i\hbar\kappa) c_{\mathbf{k}} - \varphi_{\mathbf{k}} a_{\mathbf{k}}. \end{aligned} \quad (7)$$

Substituting

$$a_{\mathbf{k}} = N_{\mathbf{k}}^{1/2} \exp(-i\theta_{\mathbf{k}} - i(\varepsilon_{\mathbf{k}} + \Omega_{1\mathbf{k}})t), \quad c_{\mathbf{k}} = F_{\mathbf{k}}^{1/2} \exp(-i\Phi_{\mathbf{k}} - i(\omega_{\mathbf{k}} + \Omega_{2\mathbf{k}})t),$$

$$\varepsilon_{\mathbf{k}} + \Omega_{1\mathbf{k}} = \omega_{\mathbf{k}} + \Omega_{2\mathbf{k}} = \mu_{\mathbf{k}},$$

into (7), we obtain

$$\begin{aligned} \frac{\Omega_{1\mathbf{k}}}{\Omega_{2\mathbf{k}}} &= \frac{|\varphi_{\mathbf{k}}|^2}{\hbar^2 (\Omega_{2\mathbf{k}}^2 + \kappa^2)}, \quad N_{\mathbf{k}} = \frac{gV}{2\gamma + 2\kappa \Omega_{1\mathbf{k}} / \Omega_{2\mathbf{k}}}, \\ \frac{N_{\mathbf{k}}}{F_{\mathbf{k}}} &= \frac{\Omega_{2\mathbf{k}}}{\Omega_{1\mathbf{k}}}, \quad \Phi_{\mathbf{k}} - \theta_{\mathbf{k}} = \frac{\pi}{2} + \text{arctg} \frac{\kappa}{\Omega_{2\mathbf{k}}}. \end{aligned} \quad (8)$$

In the paper by Elesin and Kopaev<sup>[13]</sup>,<sup>2)</sup> the problem of the formation of a Bose condensate is solved under the condition that the position of the exciton chemical potential is determined by the frequency of the laser radiation, while the exciton concentration is a parameter to be found. The frequency dependence of the ex-

citon concentration at constant amplitude of the field and the dependence on the amplitude at a constant laser frequency were found. It is necessary to take into account, however, that an exciton level will be at resonance with a given laser frequency only for a certain pumping power. To maintain the resonance on further increase of the power, it is necessary to have a laser whose frequency is self-consistently tuned with the change in position of the chemical potential, i.e., in such a resonance situation the frequency of the electromagnetic field will depend on the pumping power in a similar way to the exciton chemical potential.

The frequency and amplitude hystereses obtained by Elesin and Kopayev<sup>[13]</sup> is, from a physical point of view, a consequence of the existence of the two branches of the chemical potential, which are real for the Bose-Einstein condensation of excitons (cf. Fig. 1). Indeed, frequency hysteresis at a constant level of pumping is easily obtained from our results, if in Fig. 1 we make a series of sections parallel to the  $\mathbf{k}$ -axis (different values of  $\mu_{\mathbf{k}}$ , or different  $\epsilon$  from<sup>[13]</sup>). Some of the lines intersect only the lower or only the upper thickened portions of the chemical potential, and some intersect both portions. Here it should be borne in mind that, along portions where  $N_{\mathbf{k}} \gg F_{\mathbf{k}}$ , the value of  $N_{\mathbf{k}}$  is smaller along the upper branch than along the lower. The latter statement is also valid for  $\mathbf{k} \approx 0$ , if the antiresonance terms are taken into account in the Hamiltonian. Amplitude hysteresis can be obtained qualitatively if, in Fig. 1, we draw a family of curves of  $\mu_{\mathbf{k}}$  for different pumping levels and examine the points of intersection of these curves with one straight line parallel to the  $\mathbf{k}$ -axis (this straight line selects the same value of the chemical potential for different pumping levels).

### 3. SPECTRUM OF THE ELEMENTARY EXCITATIONS

The problem of this section has much in common with the determination of the energy spectrum of a semiconductor with a saturated interband transition. Saturation of the exciton band leads to Bose degeneracy, i.e., to the separation of the Bose condensate. The level of the chemical potential  $\mu_{\mathbf{k}}$  plays the role of the lowest energy level for an individual particle. To transfer the particle from this level to a mechanical-exciton state with energy

$$\Delta + l_0 + (T_q^2 + 2T_q l_q)^{1/2}$$

or a photon state with energy  $\hbar c q$ , work must be done. This work determines the spectrum of the elementary excitations. It has nothing in common with the polariton spectrum and is found by the standard method of diagonalizing the Hamiltonian  $\hat{H}_2$ .

To determine the elementary-excitation spectrum for arbitrary  $\mathbf{k}$  and  $\mathbf{q}$  in the general case, it is necessary to solve a fourth-order algebraic equation. The problem is simplified considerably for the particular case  $\mathbf{q} \perp \mathbf{k}$ , when  $|\mathbf{k} + \mathbf{q}| = |\mathbf{k} - \mathbf{q}|$ . The equation becomes biquadratic and the spectrum has the form

$$E_{\mathbf{q}\pm} = (1/2)(A_{\mathbf{k}+\mathbf{q}}^2 - l_q^2 + B_{\mathbf{k}+\mathbf{q}}^2 + 2|\varphi_{\mathbf{k}+\mathbf{q}}|^2) \pm 1/2[(A_{\mathbf{k}+\mathbf{q}}^2 - l_q^2 - B_{\mathbf{k}+\mathbf{q}}^2)^2 + 4|\varphi_{\mathbf{k}+\mathbf{q}}|^2((A_{\mathbf{k}+\mathbf{q}} + B_{\mathbf{k}+\mathbf{q}})^2 - l_q^2)]^{1/2} \quad (9)$$

$$A_{\mathbf{k}+\mathbf{q}} = \Delta + l_0 + l_q + T_{\mathbf{k}+\mathbf{q}} - \mu_{\mathbf{k}}, \quad B_{\mathbf{k}+\mathbf{q}} = \hbar c |\mathbf{k} + \mathbf{q}| - \mu_{\mathbf{k}} \quad (10)$$

The spectrum of the elementary excitations in the case  $\mathbf{k} = 0$ , when  $\mu_{\mathbf{k}} \equiv \mu_0 = \Delta + l_0$ , is shown in Fig. 2. The dashed lines show the functions

$$[T_q^2 + 2T_q l_q]^{1/2}, \quad |\hbar c q - \mu_0|$$

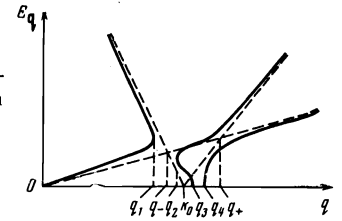


FIG. 2. Spectrum of the elementary excitations in the Bose-Einstein condensation of excitons into a state with wave vector  $\mathbf{k} = 0$ .

which are the branches of the spectrum when the exciton-photon interaction is neglected. These branches intersect at the points  $q_{\pm}$ , where

$$\hbar c q_{\pm} = \mu_0 \pm [T_{q_{\pm}}^2 + 2T_{q_{\pm}} l_{q_{\pm}}]^{1/2}.$$

In the small region  $q_1 < q < q_2$  containing the point  $q_{-}$ , where  $q_1$  and  $q_2$  are the values of the wave vector for which  $E_{\mathbf{q}}^{+} = E_{\mathbf{q}}^{-}$ , the elementary-excitation spectrum does not exist, i.e.,  $E_{\mathbf{q}}^{\pm}$  are complex. This is to be expected, since for  $\hbar c q < \mu_0$  the group velocities of the two non-interacting waves are opposite in sign and, in a saturated system, there should be an instability<sup>[11,14]</sup>. In this region, excitons of the condensate with  $\mathbf{k} = 0$  can be transformed into photons with wave vector  $\mathbf{q}$ , emitting sound-vibration quanta (hydrons<sup>[1b]</sup>) with wave vector  $-\mathbf{q}$ . For such a process, the energy and momentum conservation laws are simultaneously obeyed in the region  $q_1 < q < q_2$ . An instability of this type is called an absolute or non-convective instability<sup>[11,15]</sup>. Thus, a system of degenerate excitons can simultaneously generate both light and sound with macroscopic amplitudes, i.e., it can operate as a laser-phaser.

In the region of  $q_{+}$ , the group velocities of the waves have the same sign, the condensate excitons cannot be transformed spontaneously into photons, and the system is stable. The elementary-excitation spectrum consists of two simple waves.

At the points  $q_3$  and  $q_4$  defined by the conditions

$$\hbar c q_3 = \mu_0 + \frac{|\varphi_{q_3}|^2}{(T_{q_3} + 2l_{q_3})}, \quad \hbar c q_4 = \mu_0 + \frac{|\varphi_{q_4}|^2}{T_{q_4}},$$

the lower branch  $E_{\mathbf{q}}^{-} = 0$ , and in the region  $q_3 < q < q_4$  this branch does not exist. This instability is associated with the possibility of transforming condensate excitons into photons with wave vector  $\mathbf{q}$  with simultaneous creation of an elementary excitation with momentum  $-\mathbf{q}$ , the energy of which is negligibly small for  $\varphi_{\mathbf{q}} \rightarrow 0$ . It should be noted that, depending on the relation between  $\varphi_{\mathbf{q}}$  and  $l_{\mathbf{q}}$ , the wave vector  $q_{+}$  will satisfy one or other of the following inequalities:

$$k_0 < q_{+} < q_3, \quad q_3 < q_{+} < q_4 \quad \text{OR} \quad q_{+} > q_4.$$

In the limiting case  $q \ll k$ , by expanding the coefficients in a series in  $q$ , we find from (9) that there are two branches of elementary excitations, the dispersion laws of which can be written in the form

$$E_{\mathbf{q}}^{+} = \epsilon_0(\mathbf{k}) + \hbar^2 q^2 / 2M(\mathbf{k}), \quad E_{\mathbf{q}}^{-} = \hbar u(\mathbf{k}) q. \quad (11)$$

The lower branch is a sound branch. The expressions for  $\epsilon_0(\mathbf{k})$ ,  $u(\mathbf{k})$  and  $M(\mathbf{k})$  are real and positive for values of  $\mu_{\mathbf{k}}$  lying on the thickened portions in Fig. 1. For  $\mathbf{k} \ll \mathbf{k}_0$  and  $\mathbf{k} \gg \mathbf{k}_0$ , we find  $\epsilon_0(\mathbf{k}) \approx |\hbar \Omega 2\mathbf{k}|$  and  $u(\mathbf{k}) \approx (l_0/m_e)^{1/2}$ . In this case, the upper branch of the spectrum is of no interest, and the lower branch is the same as in the Bose condensation of excitons in the absence of photons.

The case  $\mathbf{k} = \mathbf{k}_0$ , when both branches of the chemical

potential are present, is interesting. For the  $\mu_{\mathbf{k}_0}$  lying on the lower branch, we have

$$\varepsilon(\mathbf{k}_0) = (2l_0 |\varphi_{\mathbf{k}_0}|)^{1/2}, \quad u(\mathbf{k}_0) = (c |\Omega_{2\mathbf{k}_0}| / 2k_0)^{1/2}. \quad (12)$$

Putting  $\Delta = 2 \text{ eV}$ ,  $l_0 = 10^{-3} \text{ eV}$  and  $|\varphi_{\mathbf{k}_0}| = \hbar \Omega_{2\mathbf{k}_0} = 10^{-4} \text{ eV}$ , we obtain  $\varepsilon_0 \approx 5 \times 10^{-4} \text{ eV}$  and  $u \approx 10^8 \text{ cm/sec}$ . This type of structure of the energy spectrum is obtained in the theory of the laser state which arises in a system of two-level molecules placed in a resonator<sup>[16]</sup>. When the total number of protons and excited molecules, determined by the external conditions, exceeds the threshold value, Bose condensation of photons with wave vector  $\mathbf{k} \neq 0$  appears under stationary quasi-equilibrium conditions. This leads to a sharp narrowing of the spectral and angular characteristics of the radiation. It is clear that these indications can be used to detect the state that we are studying.

Self-focusing processes are another possible manifestation of a Bose-condensed state of excitons and photons. They are investigated in the following section.

#### 4. QUANTUM VORTICES IN A MEDIUM OF DIPOLE-ACTIVE EXCITONS AND PHOTONS

Coherent states of excitons and photons may be spatially nonuniform, varying slowly over distances of the order of the exciton mean free path. Well-known examples of coherent non-uniform states are the quantum vortices in liquid helium, type-II superconductivity<sup>[17,18]</sup>, and the phenomena of self-focusing and defocusing of laser radiation<sup>[19,20]</sup>. Using the example of excitons, we trace the inherent connection between the phenomena of the formation of quantum vortices and of the self-focusing and defocusing of light. A similar attempt, unrelated to excitons, was undertaken in the paper by Grob and Wagner<sup>[21]</sup>, in which, however, it was assumed that the density of the substance and the intensity of the electromagnetic field were largest in the same region of space.

Equations describing the spatially non-uniform coherent states of excitons and photons in the general case were derived by Keldysh<sup>[8]</sup> with allowance for the non-Bose nature of the excitons. This section is, in essence, a continuation of his work. The system of equations consists of a material equation, of the Ginzburg-Pitaevskii type, for the quantum macroscopic wavefunction  $a(\mathbf{r}, t)$ , and the Maxwell equations:

$$i\hbar \frac{\partial a(\mathbf{r}, t)}{\partial t} = \left( U - \frac{\hbar^2}{2m_0} \Delta \right) a(\mathbf{r}, t) + \frac{g}{V} |a(\mathbf{r}, t)|^2 a(\mathbf{r}, t) - \mathbf{d} \cdot \left( \mathbf{E} + \frac{4\pi}{3} \mathbf{P} \right), \quad (13)$$

$$\begin{aligned} (\nabla \mathbf{D}) = 0, \quad (\nabla \mathbf{H}) = 0, \quad [\mathbf{VE}] = -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t}, \quad [\nabla \mathbf{H}] = \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, \\ \mathbf{P} = \mathbf{d} a(\mathbf{r}, t) / v_0, \quad \mathbf{D} = \mathbf{E} + 4\pi \mathbf{P}, \end{aligned} \quad (14)^*$$

where  $U$  is the energy of formation of the excitons,  $g$  is the exciton-exciton interaction constant,  $\mathbf{d}$  is the dipole moment of the transition from the ground state to the exciton state of the crystal, and  $v_0$  is the volume of a unit cell of the crystal. In the first order of perturbation theory,  $g = \nu(0) > 0$ . The quantities  $\mathbf{E}$ ,  $\mathbf{P}$ ,  $\mathbf{D}$ ,  $\mathbf{H}$  and  $a$  are macroscopically large ( $\sim V^{1/2}$ ), and so can be regarded not as operators but as functions of  $\mathbf{r}$  and  $t$ . For  $\omega > 0$ , they depend on  $t$  like  $e^{-i\omega t}$ . Having appeared in the crystal, the exciton wave creates transition dipole moments at each lattice site. The effective field created by all the dipoles (except the one occupying the site at which we are finding the field) equals  $\mathbf{E} + 4\pi \mathbf{P}/3$ .

In the case of a spatially uniform distribution of excitons, when the solutions have the form  $e^{i\mathbf{k} \cdot \mathbf{r}}$ , Eqs. (13) and (14) go over into Eqs. (5.27) of the paper<sup>[1b]</sup> of one of the authors, if in the latter we neglect the antiresonance terms. Equation (13) without the last term was investigated by Pitaevskii<sup>[17]</sup>. It was a solution of the type  $e^{i\mathbf{l}\varphi}$  describing a vortex filament with quantum number  $l$ , where  $\varphi$  is the angle in the cylindrical coordinate system. In the other limiting case, Eqs. (13) and (14) go over into the equations of the theory of light-excitons. Discarding the nonlinear term in (13) and going over from the second-quantization representation to the Schrödinger equation, we can regard  $a(\mathbf{r}, t)$  as the wavefunction of an individual exciton in the continuum model.

We shall consider the formation of a vortex filament in a medium of dipole-active excitons. In Eqs. (13) and (14) we change to cylindrical coordinates, with the  $z$ -axis along the direction of the vortex axis. We introduce the radius  $r_0$  of the vortex core and the dimensionless variable  $\xi$ :

$$r_0^2 = \hbar^2 / 2m_0 g n_0, \quad \xi = r / r_0. \quad (15)$$

The quantity  $r_0$ , which plays the role of the coherence length in the theory of superconductivity, should be greater than the exciton mean free path  $l_{e-e}$ . However, the exciton concentration  $n_0$  should be less than  $a_e^{-3}$ . In these conditions,  $r_0 \sim n_0^{-1/2}$  may turn out to be less than  $l_{e-e}$ . The theory described here is valid in the region  $r > l_{e-e}$ ; but its continuation into the region  $0 < r \leq r_0$  is formal in character.

In the theory of superconductivity there is also a parameter  $\lambda = mc^2 / 4\pi e^2 n_0$ , where  $n_0$  is the concentration of Cooper pairs; this parameter characterizes the depth of penetration of the field into the metal. When  $\lambda$  is greater than the coherence length, Abrikosov filaments, which are the analogs of the quantum vortices in He II, appear in type-II superconductors. In the case of excitons and He II, the charge  $e = 0$  and  $\lambda \rightarrow \infty$ . Therefore, we have the case  $\lambda > r_0$  and the formation of quantum vortices is possible. A vortex in an exciton medium can probably be excited by a non-uniform distribution of excitons in space. The excitons can perform spiral motion about the  $z$ -axis, and this motion can be separated into translational motion along the vortex axis and rotational motion in a plane perpendicular to the axis.

We shall choose the dipole moment  $\mathbf{d}$ , in (13) to be either parallel to the  $z$ -axis or rotating in a circle in a plane perpendicular to the vortex axis, i.e.,

$$1) \quad d_x = d \neq 0, \quad d_x = d_y = 0; \quad 2) \quad d_x = 0, \quad d_x = i\eta d_y, \quad \eta = \pm 1. \quad (16)$$

Circular polarization obtains for the  $\sigma$ -components of dipole-active exciton levels in an external magnetic field  $H_0 \parallel z$ . For example, in a CdS crystal ( $z \parallel C_6$ ), the dipole-active excitons have either transverse (the level  $\Gamma_6$ ) or longitudinal (the level  $\Gamma_1$ ) polarization.

Let an electromagnetic wave (like the exciton wave) propagate along the  $z$ -axis. We shall seek the stationary solutions for the excitons and the field in the form

$$a(\mathbf{r}, t) = N_0^{1/2} \Psi(\xi, \varphi) \exp(-i\omega t + ik_z z), \quad (17)$$

$$\mathbf{E}(\mathbf{r}, t) = N_0^{1/2} \mathbf{e}(\xi, \varphi) \exp(-i\omega t + ik_z z),$$

assuming that the transverse components of the field are also circularly polarized

$$e_x = i\eta e_y. \quad (18)$$

Then the system of equations (13) and (14) is brought to the form

$$L(\xi, \varphi) \Psi + \frac{\hbar \Omega}{n_0 g} \Psi - |\Psi|^2 \Psi + 2 \frac{d_y^* e_y}{n_0 g} + \frac{d_x^* e_x}{n_0 g} = 0, \quad (19)$$

$$L(\xi, \varphi) e_y + 2\kappa^2 r_0^2 e_y + 8\pi \frac{d_y}{v_0} \left( \frac{\omega r_0}{c} \right)^2 \Psi - k_z r_0 e^{-i\varphi} \left( \eta \frac{\partial}{\partial \xi} + \frac{i}{\xi} \frac{\partial}{\partial \varphi} \right) e_x = 0, \quad (20)$$

$$L(\xi, \varphi) e_x + \kappa^2 r_0^2 e_x + \frac{4\pi}{v_0} d_x \kappa^2 r_0^2 \Psi - \frac{4\pi}{v_0} d_y k_z r_0 e^{-i\varphi} \left( \eta \frac{\partial}{\partial \xi} - \frac{i}{\xi} \frac{\partial}{\partial \varphi} \right) \Psi = 0, \quad (21)$$

where we have introduced the following notation:

$$L(\xi, \varphi) = \frac{\partial^2}{\partial \xi^2} + \frac{1}{\xi} \frac{\partial}{\partial \xi} + \frac{1}{\xi^2} \frac{\partial^2}{\partial \varphi^2}, \quad \kappa^2 = \frac{\omega^2}{c^2} - k_z^2, \quad (22)$$

$$\hbar \Omega = \hbar \omega - U + 4\pi |d|^2 / 3v_0 - \hbar^2 k_z^2 / 2m_e.$$

In Eqs. (20) and (21) there are terms proportional to  $k_z r_0$ , which take into account the non-uniformity of the field of the wave at distances of the order of  $r_0$ . At large distances ( $\xi \rightarrow \infty$ ) we can neglect the derivatives and terms containing inverse powers of  $\xi$ . Then the equations become algebraic and permit us to determine the dependence of  $\omega$  on  $k_z$ :

$$\hbar \omega - U - g n_0 - \frac{\hbar^2 k_z^2}{2m_e} = \frac{4\pi}{3v_0} \left\{ 2|d_x|^2 + (|d_x|^2 + |d_y|^2) \frac{2 + c^2 k_z^2 / \omega^2}{1 - c^2 k_z^2 / \omega^2} \right\}. \quad (23)$$

The frequencies  $\hbar \omega_{\parallel}$  and  $\hbar \omega_{\perp}$  of the longitudinal and transverse excitons are found to be the same as in the theory of polaritons, with  $\hbar \omega_{\perp}$  coinciding with  $\mu_{\mathbf{k}}$  (Sec. 2). Since it makes sense to consider only the thickened portions in Fig. 1, the frequencies turn out to be bounded from below; this resembles the condition for the propagation of waves in a wave-guide<sup>[21,22]</sup>.

We shall take the existence of vortices into account by choosing functions of the form

$$\Psi(\xi, \varphi) = \Psi(\xi) e^{i\varphi}, \quad e_x(\xi, \varphi) = e_x(\xi) e^{i\varphi}, \quad e_y(\xi, \varphi) = e_y(\xi) e^{i(\varphi + \eta\pi)} \quad (24)$$

for the case  $d_z = d \neq 0$ ,  $d_x = d_y = 0$ . Then, in the region  $\xi \gg 1$ , the solutions of Eqs. (19)–(21) are obtained in the form

$$\Psi(\xi) = 1 - \frac{1}{2} \left( 1 + \frac{4\pi d^2}{g n_0 v_0 \kappa^2 r_0^2} \right) \frac{1}{\xi^2}, \quad (25)$$

$$e_y(\xi) = \frac{2\pi d k_z}{v_0 \kappa^2 r_0} \frac{1}{\xi},$$

$$e_x(\xi) = -\frac{4\pi d}{v_0} \left\{ 1 - \left( \frac{1}{2} + \frac{2\pi d^2}{g n_0 v_0 \kappa^2 r_0^2} - \frac{1}{\kappa^2 r_0^2} \right) \frac{1}{\xi^2} \right\}$$

Extending the solutions into the region  $\xi < 1$  gives

$$\Psi(\xi) = p\xi + q\xi^3, \quad e_x(\xi) = s\xi + t\xi^3, \quad (26)$$

$$e_y(\xi) = k_z r_0 \begin{cases} \alpha + \beta\xi^2 & \text{if } \eta = -1 \\ \gamma\xi^2 & \text{if } \eta = 1. \end{cases}$$

On the vortex axis,  $\Psi$  and  $e_z$  vanish, and  $e_y \neq 0$  if  $\eta = -1$  (Fig. 3). As  $\xi \rightarrow \infty$ , the functions  $\Psi$  and  $e_z$  tend to values corresponding to a uniform distribution of excitons, and  $e_y$  falls off slowly ( $\sim 1/\xi$ ) in the same way as the azimuthal velocity of a quantum vortex in He II.

Thus, for a non-uniform distribution of longitudinal excitons there is an almost longitudinal coherent wave, for which the dependence  $\hbar \omega_{\parallel}(k_z)$  is the same as for uniformly distributed longitudinal excitons. The situation in which a longitudinal exciton can be observed optically has already been encountered in CdS<sup>[23]</sup>. Since the transverse components of the field take their maximum value in the region of the vortex core, we shall perceive a light flux passing principally through the central region of the volume. We emphasize that the

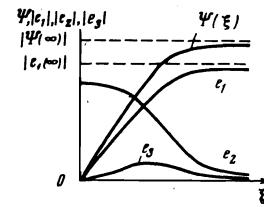


FIG. 3. Qualitative dependence on  $\xi$  of the exciton function and of the components of the electric-field intensity. For longitudinal excitons,  $e_1 = e_z(\xi)$ ,  $e_2 = e_y(\xi)$  for  $\eta = -1$ , and  $e_3 = e_y(\xi)$  for  $\eta = 1$ ; for transverse excitons,  $e_1 = e_y(\xi)$ ,  $e_2 = e_z(\xi)$  for  $\eta = 1$ , and  $e_3 = e_z(\xi)$  for  $\eta = -1$ .

energy, per unit length of the vortex, of the transverse components of the electromagnetic field is proportional to  $\ln \xi$ , like the mechanical energy per unit length of a vortex in He II. This phenomenon is analogous to self-focusing and to the formation of fine light filaments.

If the excitons are circularly polarized ( $d_z = 0$ ,  $d_x$ ,  $d_y \neq 0$ ), the solutions of Eqs. (19)–(21) can be sought in the form

$$e_x(\xi, \varphi) = e_x(\xi) e^{i\varphi}, \quad e_z(\xi, \varphi) = e_z(\xi) e^{i(1-\eta)\varphi}, \quad \Psi(\xi, \varphi) = \Psi(\xi) e^{i\varphi}, \quad (27)$$

where for  $\xi \gg 1$  we have

$$\Psi = 1 + \frac{1}{2} \left( 1 + \frac{d}{g n_0 \kappa^2 r_0^2} \right) \frac{1}{\xi^2}, \quad e_x = \frac{4\pi d k_z}{v_0 \kappa^2 r_0} \frac{1}{\xi},$$

$$e_y = -\frac{4\pi d}{v_0 [1 - (c k_z / \omega)^2]} - \frac{1}{2\kappa^2 r_0^2 \xi^2} \left[ 1 - \frac{4\pi d}{v_0} \left( \frac{\omega r_0}{c} \right)^2 \left( 1 + \frac{d}{g n_0 \kappa^2 r_0^2} \right) \right] \quad (28)$$

and for  $\xi \ll 1$  we have

$$\Psi(\xi) = p\xi + q\xi^3 + \dots, \quad e_x(\xi) = s\xi + t\xi^3 + \dots,$$

$$e_z(\xi) = k_z r_0 \begin{cases} \alpha + \beta\xi^2 & \text{for } \eta = 1, \\ \gamma\xi^2 & \text{for } \eta = -1. \end{cases} \quad (29)$$

The qualitative dependence of the functions on  $\xi$  is shown in Fig. 3. The transverse component of the field, together with the exciton function, goes to zero along the vortex axis. In this region a non-zero longitudinal component of the field appears, which falls off slowly at infinity ( $\sim 1/\xi$ ). The wave is almost transverse. Observing the light emerging from the crystal, we notice a weakening of the light field along the vortex axis, i.e., a shadow filament.

If several parallel quantum vortices are formed in the crystal and the region occupied by each of them is bounded, tubular light fluxes will be observed. It is possible to propose an exciton or polariton mechanism for the self-focusing of the light. It could explain the existence of the fine filaments by the appearance of quantum vortices in the exciton medium. The lifetime of a filament would be determined by the exciton lifetime, and its thickness would depend on the value of  $r_0$ . The maximum value of the field in the filament is limited by the ionization potential of the excitons and by their maximum concentrations. Analogous results can be obtained if we eliminate the first of Eqs. (13) and replace the polarization vector  $\mathbf{p}$  by the expression  $\kappa \mathbf{E} + \chi |\mathbf{E}|^2 \mathbf{E} / V$ , where  $\chi < 0$ .

## 5. INTERACTION OF EXCITONS WITH RESONANCE LASER RADIATION

If  $\nu_{\text{ex-ph}} > \nu_{\text{ex-ex}} > \nu_{\text{ex-latt}}$  and the duration  $T$  of the light pulse satisfies the inequalities  $\nu_{\text{ex-ph}} > T^{-1} > \nu_{\text{ex-ex}}$ , then a coherent polariton wave with  $\mathbf{k} \neq 0$  is formed under the action of the laser. It exists at least until exciton-exciton scattering processes come into

play. This state has still not been discussed in the literature. It can be created experimentally at the present time. Scattering of excitons can destroy the coherent polariton state. If the latter is preserved over a period of time longer than the exciton relaxation time, and if the number of polaritons can be assumed to be given, then Bose-Einstein condensation of polaritons with  $k \neq 0$  is realized. Thus, a more general case than that considered by Knox<sup>[23]</sup> can occur. In this case, it is appropriate to eliminate the exciton-photon interaction from the beginning and go over to polariton creation and destruction operators and treat the interaction of the polaritons. Such an approach has been used in studying non-linear optical properties of solids<sup>[24]</sup>. The polariton variant of the formation of coherent states is also important because polariton concepts are used in the theory of the interaction of resonance radiation with a gas of two-level molecules<sup>[10,11]</sup>. If the duration of the laser pulse is shorter than all the relaxation times in the system, it is possible to observe an exciton echo<sup>[1b,25]</sup>.

In the case (1), by the methods of two-photon spectroscopy we create considerable concentrations of excitons in the bulk of the crystal and create the conditions necessary for degeneracy of the excitons. The switching-on of a laser source whose frequency is in resonance with a renormalized exciton level can stimulate the appearance of an exciton Bose condensate<sup>[1a,13,26]</sup>. The results of this paper describe processes of this type, provided that we can achieve conservation of the total number of excitons and photons and the concept of the chemical potential is applicable.

The formation of an exciton Bose condensate can be manifested as lightening of the sample due to suppression of scattering processes<sup>[1b,8]</sup>, or can lead to the appearance of light filaments or shadow filaments at the exit surface of the crystal, etc.

Under the action of resonance laser radiation, excitons whose distribution in the band is not a quasi-equilibrium distribution can also be formed. However, in this case too, the elementary-excitation spectrum will have regions of instability leading to the generation or amplification of waves. We call attention to these phenomena, since at the present time the technical possibilities for investigating them experimentally already exist<sup>[27]</sup>.

The well known analogy between the theory of the laser and the theory of second-order phase transitions<sup>[28,29]</sup>, and between the coherent states in quantum optics and in solids<sup>[30,31]</sup>, will facilitate the further investigation of the Bose condensation of excitons. In addition, analogous effects with non-equilibrium optical phonons<sup>[32]</sup>, vacancies<sup>[33]</sup>, and other elementary excitations in solids and in biological systems<sup>[34]</sup> have recently been investigated.

In conclusion, the authors consider it their pleasant duty to express their deep gratitude to L. V. Keldysh for many fruitful discussions of the questions touched upon and for acquainting them with his paper<sup>[8]</sup> before it was published.

\* $[\nabla E] \equiv \nabla \times E$

<sup>1)</sup>This possibility was pointed out to us by L. V. Keldysh.

<sup>2)</sup>The paper by Elesin and Kopaev<sup>[13]</sup> was published while our article was being revised.

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