

## OPTICAL PROPERTIES OF AN EXCITON CONDENSATE IN A SEMICONDUCTOR

V. A. GERGEL', R. F. KAZARINOV, and R. A. SURIS

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A gas of bound electrons and holes (excitons) in a semiconductor is considered. It is demonstrated that such a system is capable of forming a Bose condensate, and the conditions for such a formation are determined. The interaction between light and an exciton condensate is studied. The real and imaginary parts of the dielectric polarizability of a semiconductor near the exciton absorption line are determined. It is shown that negative light absorption should occur at a frequency somewhat lower than that of exciton absorption.

## INTRODUCTION

**P**HENOMENA occurring under conditions of strong injection of nonequilibrium electrons and holes have recently assumed an important role in semiconductor physics. One example of such phenomena is the laser effect in semiconductor diodes. Considerable interest attaches to the question of the occurrence of Bose condensate of bound states of electrons and holes—excitons—in semiconductors under strong carrier injection. Such a possibility was first indicated by Blatt and Boer<sup>[1]</sup> and Moskalenko<sup>[2]</sup>. This question was also investigated by Keldysh and Kozlov<sup>[3]</sup>. Two problems arise in this connection. The first is to ascertain the conditions under which such a condensate can be produced. The second is to determine the observable effects of such a condensate. The question of the possibility of formation of a condensate of excitons is subdivided in turn into two. First, it is necessary to determine more precisely the degree to which the collective properties of the exciton systems are influenced by the fact that the excitons are not, strictly speaking, Bose particles but are bound states of two fermions. Second, since the excitons can be annihilated as a result of electron-hole recombination, it is necessary to determine whether a situation can be realized in which the thermal equilibrium of the excitons and of the lattice is established more rapidly than the exciton annihilation. Clearly, one can speak of a condensate of excitons only if this condition is satisfied.

We shall consider these questions in the first section of the paper, and show that in a number of semiconductors an exciton condensate of low density can be produced. We shall then consider the spectrum of the collective excitations in the exciton condensate and show that, just as in the model of weak non-ideal Bose gas, it has an acoustic character. There is, however, a certain difference, in that in most semiconductors the ground state of the exciton is degenerate. Therefore the condensate itself consists of several components, and the collective excitations correspond to several acoustic branches with different velocities.

Since the excitons determine in essential fashion the optical properties of the semiconductors at frequencies close to the fundamental absorption edge, it is perfectly natural to expect just there the primary manifestation of the presence of the exciton condensate. This question is dealt with in the last section of the paper. We shall

show that in a semiconductor in which the maximum of the valence band and the minimum of the conduction band correspond to the same value of the quasimomentum, the presence of the condensate leads to a negative absorption of light. The frequency corresponding to the negative absorption lies somewhat below the frequency of the exciton transition corresponding to the creation of an exciton in the ground state. This negative absorption of light can lead to the laser effect. It seems to us that the most suitable for the realization of such an effect is the three-layer semiconductor system called the p-i-n structure. This structure consists of a sufficiently pure semiconductor layer into which electrons and holes are injected from the neighboring n- and p-regions.

## BOSE CONDENSATION OF EXCITONS

It is known that "large-radius exciton" or "Mott exciton" is the term used to describe a bound state of an electron and a hole. One can speak of such a state within the framework of the two-band model only if its radius is much larger than the lattice constant. This is precisely the situation realized in most typical semiconductors, in which, owing to the large dielectric constant ( $\epsilon \sim 10-20$ ) and the small reduced mass of the electron and the hole, which is usually of the order of  $1/10$  of the free-electron mass, the radius of the exciton turns out to be of the order of several times ten interatomic distances, and the binding energy is of the order of one-hundredth of an electron volt.

Let us make a few remarks concerning the classification of the exciton states in the semiconductors that are of interest from the experimental point of view. We note first that from the point of view of the band structure, semiconductors are divided into two classes. The first includes semiconductors in which the minimum of the conduction band and the maximum of the valence band are in different points of the Brillouin zone. Typical representatives of this class are germanium and silicon. The second class includes semiconductors in which the minimum of the conduction band and the maximum of the valence band are at the center of the Brillouin zone. This class includes most III-V semiconductors, such as GaAs and InSb.

The conduction band of semiconductors of the first class is characterized by several symmetrically-arranged minima, and the dispersion law near each of

them is quadratic, with an anisotropic effective mass. In semiconductors of the second type, there is one low-lying minimum at the center of the Brillouin zone, and the dispersion law turns out to be quadratic and isotropic. In all these semiconductors, the dispersion law of the holes has two branches, corresponding to "heavy" and "light" holes. These branches coincide, at zero value of the quasimomentum, and have a minimum at this point.

Since the interaction of the electron spin with the hole is exceedingly small, the energy of the excitons corresponding to different spin states of the electrons practically coincide. Owing to the presence of two branches of the hole spectrum, two lower exciton levels are produced. For semiconductors of the GaAs type, each of these levels, with allowance for the electron spin, has four-fold degeneracy. The c.m.s. motion energy of these excitons does not depend on the quantum numbers with respect to which the level is degenerate, and at small values of the quasimomenta  $p$  it takes the form

$$\varepsilon_1 = p^2/2M, \quad \varepsilon_2 = p^2/2M', \quad (1)$$

where  $M$  and  $M'$  are the masses of the "heavy" and "light" excitons. It is clear that the binding energy of the "heavy" exciton exceeds the binding energy of the "light" exciton. In semiconductors such as germanium there is additional degeneracy connected with the presence of several electron-energy minima in the conduction band. This degeneracy is lifted partially when the quasimomentum of the exciton differs from zero.

As already stated in the Introduction, an important factor for the feasibility of Bose condensation is the relation between the exciton lifetime and the time necessary to establish thermal equilibrium between the excitons and the lattice. We shall discuss first the lifetime of the exciton in a semiconductor such as germanium, brought about by recombination in which the impurities do not take part. The concentration of the impurities may be so low that their role in the recombination is negligible. In such semiconductors, the exciton is made up of electrons and holes from essentially different regions of quasimomentum space. The difference between the wave vectors of the electron and of the hole is a quantity on the order of the reciprocal-lattice vector. Since the wave vector of the light radiated during recombination is much smaller than this difference, recombination is possible only with participation of a short-wave phonon. Simple calculations show that at low temperatures, in interaction with an acoustic phonon, the exciton lifetime  $\tau$  is given by the expression

$$\frac{1}{\tau} = \left(\frac{g}{\Delta}\right)^2 \frac{\hbar k_0}{\rho s r_e^3 \tau_0}. \quad (2)$$

Here  $g$  is the constant of the deformation potential of the electron, with an approximate value 4 eV,  $\Delta$  is the difference between the electron energy at the center of the Brillouin zone and at the minimum, which is located at the point  $k_0$ ,  $\rho$  is the crystal density,  $s$  is the speed of sound,  $r_e$  is the radius of the exciton, and  $\tau_0$  is the characteristic atomic time, which is inversely proportional to the interband dipole moment of the transition and to the cube of the wave vector of the emitted light;  $\tau_0$  is a quantity on the order of  $10^{-8}$  sec. Substituting in

(2) the values of the parameters corresponding to germanium, we find that the lifetime of the exciton  $\tau \sim 10^{-4}$  sec. The lifetime in silicon is of the same order.

An estimate of the lifetime in semiconductors of the GaAs type is somewhat more complicated. Since the exciton is made up here of an electron and a hole with quasimomenta close to zero, direct conversion of the exciton into light is possible. All that is necessary for this purpose is that the quasimomentum of the center of gravity of the exciton be equal to the momentum of the light. Since the inverse process is also possible, one cannot speak of the exciton and of the light separately near the point of intersection of the optical and exciton branches. The region  $\Delta k$  of the wave vectors near the point of intersection, in which this intermingling is significant, is of the order of  $\Omega k/\omega_0$ , where  $\omega_0$  and  $k$  are the frequency and the wave vector at the point of intersection, and  $\hbar\Omega = 4\pi|d|^2 r_e^{-3}$  is the characteristic frequency which determines the interaction between the excitons and the light<sup>[4]</sup> (here  $d$  is the dipole moment of the transition between the valence band and the conduction band). Inasmuch as  $\Omega$  is a quantity on the order of  $10^{10}$  sec<sup>-1</sup> and  $\omega_0 \sim 10^{15}$  sec<sup>-1</sup>, it follows that the interval  $\Delta k$  is very narrow compared with  $k$ . It is clear that the vanishing of the exciton as a result of its conversion into light and the emergence of the latter from the crystal are possible only in this narrow interval of wave vectors. Simple calculations show, however, that this process is insignificant. The point is that, owing to the smallness of the phase volume  $\Gamma \sim k^2 \Delta k$  in which such a process is possible, the number of excitons passing into this region per unit time is small. The main process determining the lifetime of the excitons is their recombination accompanied by emission or absorption of a long-wave phonon. This process is possible for an exciton with any quasimomentum of the c.m. This is precisely why this process determines the average lifetime of the exciton, in spite of the additional smallness introduced by the participation of the phonon into the recombination probability. The expression for the recombination probability per unit time differs from (2) in that  $\Delta$  is replaced by a quantity on the order of the average thermal energy of the exciton, and  $k_0$  is replaced by its thermal momentum. The exciton lifetime in GaAs at liquid-helium temperature, estimated from this formula, is on the order of  $10^{-6}$  sec.

We now discuss the establishment of thermal equilibrium of the excitons. This occurs in two stages: the binding of the electron with the hole into an exciton, and the cooling of the resultant exciton to the lattice temperature. The cross section for the binding of an electron and a hole to form an exciton was calculated by Lipnik<sup>[5]</sup> and is approximately  $10^{-13}$  cm<sup>2</sup> at helium temperatures for crystals such as GaAs or Ge. At electron and hole concentrations on the order of  $10^{15}$  cm<sup>-3</sup>, the time of binding of an electron and a hole to form an exciton is of the order of  $10^{-9}$  sec.

As shown by Lipnik<sup>[5]</sup>, the kinetic energy of the exciton produced as a result of binding of an electron with a hole is close to the exciton binding energy. Therefore the resultant excitons are "hot" and it is necessary to consider the process of their "cooling" with transfer of energy to the lattice, that is, by emission of phonons

from the exciton. It can be shown that the exciton cooling time due to this process is given by the relation

$$\frac{1}{\tau'} = \frac{4}{3\pi} \frac{(g_1 + g_2)^2 M^2}{\rho \hbar^3 r_e},$$

where  $g_1$  and  $g_2$  are the constants of the deformation potential for the electrons and holes respectively. Substituting parameters that are characteristics of the crystals under consideration, we obtain  $\tau \sim 10^{-9} - 10^{-10}$  sec. We note incidentally that the time of collisions between the excitons, at an exciton concentration  $10^{15} \text{ cm}^{-3}$  and at helium temperature, is of the same order of magnitude.

We see thus that the excitons enter into thermal equilibrium with the lattice within a time much shorter than their lifetime.

As already stated in the Introduction, it is questionable whether the low-density exciton system can be regarded as a weakly non-ideal Bose gas. In particular, is their Bose-Einstein condensation possible? The point is that the exciton creation and annihilation operators obey the Bose commutation relations with accuracy to terms of the order of  $n r_e^3$ , where  $n$  is the exciton density. On the other hand, to consider the collective properties of the an exciton system it is necessary to take their interaction into account. Since the amplitude for the scattering of the excitons by one another is of the order of  $r_e$ , the energy of their interaction per exciton is proportional to  $n r_e^3$  in the lowest order in the concentration. Thus, it must be taken into account in the consideration of the interaction that the excitons are not bosons but consist of Fermi particles.

It can be shown, nonetheless, that, accurate to second order in the concentration inclusive, the system of excitons behaves like a gas of low-density Bose particles. The "non-Bose nature" of the excitons becomes manifest only in the fact that the scattering amplitude that enters into the theory of a weakly non-ideal Bose gas<sup>[6]</sup> is defined here as the total amplitude for the forward scattering of two excitons, determined with allowance for their internal structure. This amplitude is obtained by solving the Schrödinger equation for two electrons and two holes with boundary conditions corresponding to two excitons at infinity.

However, the situation becomes somewhat more complicated because the ground state of the exciton is degenerate and is characterized by several quantum numbers  $\sigma$ . Therefore the condensate consists of several components, and the interaction is determined by the aggregate of the amplitudes for the scattering of each of the components by the other. The exciton energy<sup>1)</sup> at zero temperature, in an approximation quadratic in the concentration of the condensates, takes the form

$$\frac{U}{V} = \xi \sum_{\sigma} n_{\sigma} + \frac{2\pi\hbar^2}{M} \sum_{\sigma\sigma'} f_{\sigma\sigma'} n_{\sigma} n_{\sigma'}, \quad (3)$$

where  $V$  is the volume of the system,  $n_{\sigma} \equiv |\varphi_{\sigma}|^2$  is the concentration of the  $\sigma$ -component of the condensate,  $\varphi_{\sigma}$

is the condensate "wave function," and  $f_{\sigma\sigma'}$  is the amplitude for scattering from the state  $\sigma$  into  $\sigma'$  at zero quasimomentum.

The quantity  $\xi$  is defined by the relation  $\xi = E_0 - \mu_1 + \mu_2$ , where  $E_0$  is the exciton excitation energy, which is equal to the width of the forbidden band  $E_g$  less the exciton binding energy  $E_e$ , while  $\mu_1$  and  $\mu_2$  are the Fermi quasilevels for the electrons and holes respectively.

Expression (3) has a simple physical meaning. The first term is the energy of formation of immobile excitons from electrons and holes, and the second is the energy of their interaction. This expression does not contain excited exciton states, and is therefore valid if the interaction energy is small compared with the energy of transition to the excited state. Since the energy of the transition is of the order of the binding energy and the scattering amplitudes are of the order of  $r_e$ , this condition coincides with the low-density condition, that is, with the condition that allows us to confine ourselves to terms quadratic in the concentration in the expression for the system energy.

In order for the system to be stable, the second term of (3) must be positive definite, that is, the matrix  $f_{\sigma\sigma'}$  must have positive eigenvalues. In the opposite case, the excitons form a liquid.

It is clear that if the matrix  $f_{\sigma\sigma'}$  is positive definite, then the condensate appears only if  $\xi < 0$ . This means that the work expended in bringing the electron and hole into the system from the reservoirs that determine the chemical potentials of the electrons and holes is smaller than the energy gained during the formation of the exciton. In the case of a p-i-n structure the role of the reservoirs is played by the p and n regions. For condensate to occur it is necessary that the voltage applied to it, which is equal to  $(\mu_1 - \mu_2)/e$ , exceed  $E_0/e$ , that is,  $\xi < 0$ .

Minimizing expression (3) with respect to  $\varphi_{\sigma}$ , we obtain an expression for the concentration of the components of the condensate:

$$\varphi_{\sigma} \left( \xi + \sum_{\sigma'} \frac{4\pi\hbar^2}{M} f_{\sigma\sigma'} n_{\sigma'} \right) = 0. \quad (4)$$

This equation must be solved with allowance for the natural condition  $n_{\sigma} \geq 0$ . It has two solutions:

$$\varphi_{\sigma} = 0, \quad n_{\sigma} = |\varphi_{\sigma}|^2 = -\xi \frac{M}{4\pi\hbar^2} \sum_{\sigma'} (f^{-1})_{\sigma\sigma'}.$$

If any  $n_{\sigma_0}$ , defined by the last formula, turn out to be negative when  $\xi < 0$ , they must be set equal to zero, and it is necessary to minimize the energy (3), in which there are no terms with  $\sigma = \sigma_0$ . As a result we get

$$n_{\sigma} = -\xi \frac{M}{4\pi\hbar^2} \sum_{\sigma'} (f^{-1})_{\sigma\sigma'}, \quad (5)$$

where  $\tilde{f}_{\sigma\sigma'}$  is the matrix from which the elements with one of the indices  $\sigma$  or  $\sigma'$  coinciding with  $\sigma_0$  have been discarded.

For estimating purposes, let us consider the simplest case, when the lowest level of the exciton is not degenerate, and the condensate consists of one component. Then

$$n \sim M |\xi| / 4\pi\hbar^2 f.$$

Recognizing that the binding energy of the electron and the hole in the exciton is

<sup>1)</sup>We shall consider henceforth, for simplicity, only zero temperature. However, the results can be generalized to the case of nonzero temperatures in a manner similar to that used in the model of low-density Bose gas. It is clear that the results obtained below remain qualitatively in force at all temperatures that are not too close to the Bose-condensate temperature.

$$|E_0| \approx \hbar^2/2\mu r_e^2,$$

where  $\mu$  is their reduced mass, we get

$$n \sim |\xi| M/4\pi r_e^2 \mu |E_0|.$$

If the excitons do not experience resonant scattering, then  $f \sim r_e$ . Therefore the condition of small density denotes that  $|\xi| \ll |E_0| \mu/M$ .

We have assumed above that the amplitude for the scattering of two excitons, as the function of their energy, has no poles on the real axis. This denotes that there is no bound state—"molecule"—of two excitons. If such a "molecule" is produced, then it is not difficult to generalize the theory to include this case. The difference is that the condensate is made up not of excitons but of molecules, and the interaction energy is determined by their forward scattering amplitude.

### COLLECTIVE EXCITATIONS

It is perfectly clear that the spectrum of the long-wave single-exciton excitations should have an acoustic character, in analogy with the non-ideal Bose gas<sup>[6,7]</sup>.

The Hamiltonian of the system of electrons and holes is of the form

$$H = H_0 + H_{int}.$$

$$H_0 = \sum_{\alpha=1}^2 \int dx \psi_{\alpha}^+(x) [\epsilon_{\alpha}(\hat{\mathbf{p}}) - \mu_{\alpha}] \psi_{\alpha}(x),$$

$$H_{int} = \frac{1}{2} \int dx dx' \sum_{\alpha=1}^2 \psi_{\alpha}^+(x) \psi_{\alpha}^+(x') \frac{e^2}{\epsilon|r-r'|} \psi_{\alpha}(x') \psi_{\alpha}(x) - \int dx dx' \psi_1^+(x) \psi_1(x) \frac{e^2}{\epsilon|r-r'|} \psi_2^+(x') \psi_2(x'). \quad (6)$$

Here  $\psi_1^{\dagger}(\mathbf{x})$  and  $\psi_2^{\dagger}(\mathbf{x})$  are the operators for the creation of an electron and a hole, respectively, and  $\epsilon_1(\hat{\mathbf{p}})$ ,  $\epsilon_2(\hat{\mathbf{p}})$  and  $\mu_1$ ,  $\mu_2$  are their kinetic energies and chemical potentials. The variable  $\mathbf{x}$  is the aggregate of the spatial coordinate  $\mathbf{r}$  and the variables  $s$  which characterize the internal states of the electron and the hole.

The spectrum of the single-exciton excitations is determined by the poles of the Green's function<sup>2)</sup>

$$G_{\omega}(x_1, x_2 | x_1', x_2') \equiv i \int dt e^{i\omega t} \langle T \{ \psi_1(x_1, t) \psi_2(x_2, t) \psi_2^+(x_2') \psi_1^+(x_1') \} \rangle, \quad (7)$$

$$\psi(x, t) = e^{iHt} \psi(x) e^{-iHt}.$$

In the case when there is no condensate and the temperature is equal to zero, this function takes the form<sup>[4]</sup>

$$g_{\omega}(x_1, x_2 | x_1', x_2') = \sum_{\lambda k} g_{\lambda}(k) \chi_{\lambda k}(x_1, x_2) \chi_{\lambda k}(x_1', x_2'), \quad (8)$$

$$g_{\lambda}(k) = [\omega - E_g + \mu_1 - \mu_2 - E_{\lambda} - \epsilon_{\lambda}(k)]^{-1}.$$

Here  $E_{\lambda} + \epsilon_{\lambda}(k)$  and  $\chi_{\lambda k}(x, x')$  are the self-energies and the eigenfunctions, normalized to unity, of an exciton whose c.m. has a wave vector  $\mathbf{k}$ , including an electron and hole in the scattering state.

As already mentioned in the preceding section, in the case of semiconductors of the GaAs type, the lowest exciton branch has a quadratic dispersion law. Let us see now how the dispersion law is changed by the appearance of the condensate.

We make first a number of remarks. The Hamiltonian (6) is invariant to the transformation

$$\psi_1 \rightarrow \psi_1 e^{i\varphi_1}, \quad \psi_2 \rightarrow \psi_2 e^{i\varphi_2}$$

with arbitrary phases  $\varphi_1$  and  $\varphi_2$ . The indicated invariance leads to identical vanishing of the mean values  $\langle \psi_1^{\dagger} \psi_2^{\dagger} \rangle$  and  $\langle \psi_2 \psi_1 \rangle$ . The occurrence of the condensate corresponds to violation of this symmetry, and the mean value of the operator  $\psi_1^{\dagger} \psi_2^{\dagger}$ , which determines, in particular, the creation of an exciton, turns out to be different from zero in the ground state. A similar situation arises also in Bose-gas condensation<sup>[8]</sup>.

The function  $\langle \psi_1(x_1) \psi_2(x_2) \rangle \equiv \Phi(x_1, x_2)$  can be represented in the form of an expansion in the eigenfunctions of the exciton:

$$\Phi(x_1, x_2) = \sum_{\lambda k} \Phi_{\lambda}(k) \chi_{\lambda k}(x_1, x_2). \quad (9)$$

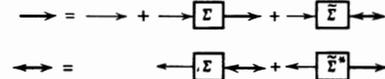
The appearance of a low-density exciton condensate corresponds to a macroscopically large filling of the ground exciton level, that is, of the state with  $\mathbf{k} = 0$  and  $\lambda = \sigma$ . Therefore

$$\Phi_{\sigma}(0) = \sqrt{V} \varphi_{\sigma},$$

where  $\varphi_{\sigma}$  is the amplitude of the  $\sigma$ -component of the condensate, and its concentration  $n_{\sigma} \equiv |\varphi_{\sigma}|^2$ . The connection between  $n_{\sigma}$  and the difference between the Fermi quasilevels of the electrons and holes was established in the preceding section.

In the presence of the condensate, the coefficients of the expansion of the Green's function in the eigenfunctions of the exciton  $G_{\lambda\lambda'}(k)$  differ from the coefficients  $g_{\lambda}(k)$  that arise in the case of a free exciton and are given by relation (8).

The functions  $G_{\lambda\lambda'}(k)$  satisfy equations that can be represented graphically in the form shown in the figure.



The heavy line with one arrow corresponds here to  $G_{\lambda\lambda'}(k)$ , the heavy line with two arrows corresponds to the anomalous Green's function  $F_{\lambda\lambda'}(k)$  which is used in the theory of low-density Bose gas<sup>[6]</sup>, and the thin line corresponds to  $g_{\lambda}(k) \delta_{\lambda\lambda'}$ . The vertex  $\Sigma$  describes the scattering of an exciton by excitons of the condensate, and the vertices  $\tilde{\Sigma}^*$  and  $\Sigma$  describe the creation of two excitons with opposite momenta from the condensate, and, respectively, their departure to the condensate.

A significant change takes place only in the spectrum of those excitons which make up the condensate, that is, the spectrum of the lowest exciton branch with  $\lambda = \sigma$ . The change in the spectrum of the remaining exciton branches, in the case of low condensate density under consideration, reduces to a trivial energy shift, due to the interaction with the condensate. In the equations for  $G_{\lambda\lambda'}$  (see the figure) we can discard the terms with  $\lambda \neq \sigma$  since the Green's functions  $G_{\lambda\lambda'}(k)$  that enter there do not contain resonant denominators.

The vertices that enter into the equations for  $G_{\sigma\sigma}$ , have in the lowest order in the condensate concentration the following form

$$\Sigma_{\sigma\sigma} = \delta_{\sigma\sigma'} \sum_{\sigma''} T_{\sigma\sigma''} n_{\sigma''} + \varphi_{\sigma} T_{\sigma\sigma'} \varphi_{\sigma'}^*, \quad (10)$$

$$\tilde{\Sigma}_{\sigma\sigma'} = \varphi_{\sigma} T_{\sigma\sigma'} \varphi_{\sigma'},$$

where  $T_{\sigma\sigma'} \equiv 4\pi f_{\sigma\sigma'} \hbar^2/M$ . The first term in the expression for  $\Sigma_{\sigma\sigma'}$  describes the direct interaction of the

<sup>2)</sup>We put  $\hbar = 1$  throughout.

exciton, characterized by a quantum number  $\sigma$ , with the condensate, whereas the second term describes the exchange interaction.

Thus, the equations for  $G_{\sigma\sigma'}$  and  $F_{\sigma\sigma'}$  take the form

$$\left(\omega - \frac{k^2}{2M}\right)G_{\sigma\sigma'} - \varphi_{\sigma} \sum_{\sigma''} T_{\sigma\sigma''\varphi_{\sigma''}G_{\sigma''\sigma'} - \varphi_{\sigma'} \sum_{\sigma''} T_{\sigma\sigma''\varphi_{\sigma''}F_{\sigma''\sigma'} = \delta_{\sigma\sigma'}, \quad (11)$$

$$\varphi_{\sigma}^* \sum_{\sigma''} T_{\sigma\sigma''\varphi_{\sigma''}^*G_{\sigma''\sigma'} + \left(-\omega - \frac{k^2}{2M}\right)F_{\sigma\sigma'} - \varphi_{\sigma'}^* \sum_{\sigma''} T_{\sigma\sigma''\varphi_{\sigma''}F_{\sigma''\sigma'} = 0.$$

We have used here the equality (4).

These equations are similar to those obtained by Belyaev<sup>[6]</sup> for a weakly non-ideal Bose gas. The only difference is that in our case the condensate consists of several components.

To solve the system, we represent the amplitudes of the condensate in the form

$$\varphi_{\sigma} = \sqrt{n_{\sigma}} e^{i\gamma_{\sigma}}, \quad (12)$$

where  $\gamma_{\sigma}$  is the phase of the corresponding component of the condensate, and introduce the auxiliary Green's functions

$$\begin{aligned} \bar{G}_{\sigma\sigma'} &= \exp(-i\gamma_{\sigma})G_{\sigma\sigma'}\exp(i\gamma_{\sigma'}), \\ \bar{F}_{\sigma\sigma'} &= \exp(i\gamma_{\sigma})F_{\sigma\sigma'}\exp(i\gamma_{\sigma'}). \end{aligned} \quad (13)$$

Then the system (11) takes the form

$$\left(\omega - \frac{k^2}{2M}\right)\bar{G}_{\sigma\sigma'} - \sqrt{n_{\sigma}} \sum_{\sigma''} T_{\sigma\sigma''}\sqrt{n_{\sigma''}}\bar{G}_{\sigma''\sigma'} - \sqrt{n_{\sigma'}} \sum_{\sigma''} T_{\sigma\sigma''}\sqrt{n_{\sigma''}}\bar{F}_{\sigma''\sigma'} = \delta_{\sigma\sigma'}, \quad (11a)$$

$$-\sqrt{n_{\sigma}} \sum_{\sigma''} T_{\sigma\sigma''}\sqrt{n_{\sigma''}}\bar{G}_{\sigma''\sigma'} + \left(-\omega - \frac{k^2}{2M}\right)\bar{F}_{\sigma\sigma'} - \sqrt{n_{\sigma'}} \sum_{\sigma''} T_{\sigma\sigma''}\sqrt{n_{\sigma''}}\bar{F}_{\sigma''\sigma'} = 0.$$

We proceed now to a representation in which the Hermitian operator  $\sqrt{n_{\sigma}}T_{\sigma\sigma'}\sqrt{n_{\sigma'}}$  is diagonal. Its eigenfunctions  $\xi_{\sigma}^m$  and eigenvalues  $4\pi f_m n/M$  ( $n = \sum_{\sigma} n_{\sigma}$ , and  $f_m$  has the meaning of the effective scattering amplitude) are determined by the equation

$$\sum_{\sigma} \sqrt{n_{\sigma}} T_{\sigma\sigma'} \sqrt{n_{\sigma'}} \xi_{\sigma}^m = \frac{4\pi f_m n}{M} \xi_{\sigma}^m. \quad (14)$$

The solution of the system (11a) in this representation is of the form

$$\begin{aligned} G_m(\omega, \mathbf{k}) &= \frac{N_{\mathbf{k}}^m + 1}{\omega - \omega_m(\mathbf{k}) + i\delta} - \frac{N_{\mathbf{k}}^m}{\omega + \omega_m(\mathbf{k}) - i\delta}, \\ F(\omega, \mathbf{k}) &= \frac{4\pi f_m n/M}{\omega^2 - \omega_m^2(\mathbf{k}) + i\delta}, \end{aligned} \quad (15)$$

where

$$\omega_m = \left(\frac{k^4}{4M^2} + \frac{4\pi f_m n}{M^2} k^2\right)^{1/2},$$

and the distribution function with respect to  $\mathbf{k}$  of the supercondensate excitons  $N_{\mathbf{k}}^m$  is equal to

$$N_{\mathbf{k}}^m = 8\pi^2 f_m^2 n^2 / M \omega_m(\mathbf{k}) \left[ \omega_m(\mathbf{k}) + \frac{k^2}{2M} + \frac{4\pi f_m n}{M} \right]. \quad (16)$$

We have thus arrived at the acoustic dispersion law which is the usual one for weakly non-ideal Bose gas at small momenta. The only difference is that the presence of many components in the condensate gives rise to several acoustic branches with velocities

$$v_m = \sqrt{4\pi f_m n / M^2}. \quad (17)$$

As to the matrix elements of the Green's functions  $G_{\lambda\lambda'}(\mathbf{k})$  pertaining to the excited states of the excitons ( $\lambda, \lambda' = \sigma$ ), they do not differ from their values in the absence of the condensate  $\delta_{\lambda\lambda'} g_{\lambda}(\mathbf{k}, \omega)$ , apart from an insignificant energy shift proportional to  $\xi$ . As a result we arrive at an expression for the Green's function of an exciton interacting with the condensate:

$$\begin{aligned} G_{\omega}(x_1, x_2 | x_1', x_2') &= \sum_{m, \mathbf{k}} \chi_{m\mathbf{k}}(x_1, x_2) \chi_{m\mathbf{k}}(x_1', x_2') G_m(\omega, \mathbf{k}) \\ &+ \sum_{\mathbf{k}, \lambda \neq \sigma} \chi_{\lambda\mathbf{k}}(x_1, x_2) \chi_{\lambda\mathbf{k}}(x_1', x_2') g_{\lambda}(\omega, \mathbf{k}), \end{aligned} \quad (18)$$

where

$$\chi_{m\mathbf{k}}(x, x') = \sum_{\sigma} \xi_{\sigma}^m e^{i\gamma_{\sigma}} \chi_{\sigma\mathbf{k}}(x, x')$$

are wave functions that are orthogonal and are normalized to unity, and  $G_m(\omega, \mathbf{k})$  and  $g_{\lambda}(\omega, \mathbf{k})$  have been defined in (15) and (8).

### 3. ELECTROMAGNETIC PROPERTIES OF EXCITON CONDENSATE

Let us consider first the interaction of an exciton condensate with an external electromagnetic field whose frequency  $\omega$  is close to  $\omega_0 \equiv E_g - E_e$ , that is, to the frequency of the exciton transition.

The mean value of the dipole moment, produced under the influence of an external field  $\mathcal{E}(t, \mathbf{r})$ , is in the linear approximation

$$\langle P^{\alpha}(\mathbf{r}, t) \rangle = i \int_{-\infty}^t dt' \int_{-\infty}^{\infty} d\mathbf{r}' \langle [P^{\alpha}(\mathbf{r}, t), P^{\beta}(\mathbf{r}', t')] \rangle \mathcal{E}^{\beta}(t', \mathbf{r}'). \quad (19)$$

Here  $P(\mathbf{r}, t)$  is the operator of the dipole-moment density in the Heisenberg representation:

$$P(\mathbf{r}, t) = \sum_{s_1, s_2} \mathbf{d}_{s_1 s_2} \{ \tilde{\Psi}_1^{\dagger}(\mathbf{r}, s_1, t) \tilde{\Psi}_2^{\dagger}(\mathbf{r}, s_2, t) + \tilde{\Psi}_2(\mathbf{r}, s_2, t) \tilde{\Psi}_1(\mathbf{r}, s_1, t) \}, \quad (20)$$

where  $\mathbf{d}_{s_1 s_2}$  is the dipole moment of the transition between the top of the valence band and the bottom of the conduction band. We assume that these transitions are allowed in the crystal. The entire theory can be generalized in obvious fashion to the case of forbidden dipole transitions ( $\mathbf{d}_{s_1 s_2} = 0$ ), but allowed quadrupole transitions.

The operators  $\tilde{\Psi}_1(t)$  and  $\tilde{\Psi}_2(t)$  differ from the operators introduced in (7) by the factors  $\exp(-i\mu_1 t)$  and  $\exp(-i\mu_2 t)$ , respectively. These factors are connected with the fact that so far we have disregarded the interband transitions and therefore could introduce different reference points from which to reckon the energy for the electrons and the holes. In problems where electron hole pairs can be produced under the influence of light, their energy must be reckoned from one level.

Going over in (2) to the earlier operators, we get

$$\begin{aligned} P(\mathbf{r}, t) &= \sum_{s_1, s_2} \{ \mathbf{d}_{s_1 s_2} \Psi_1^{\dagger}(\mathbf{r}, s_1, t) \Psi_2^{\dagger}(\mathbf{r}, s_2, t) e^{i\mu_1 t} \\ &+ \mathbf{d}_{s_1 s_2}^* \Psi_2(\mathbf{r}, s_2, t) \Psi_1(\mathbf{r}, s_1, t) e^{-i\mu_2 t} \}. \end{aligned} \quad (21)$$

Here  $\mu_{12} \equiv \mu_1 - \mu_2 = \omega - \xi$ . Substituting (21) in (19), we get an expression for the dielectric polarizability:

$$\chi^{\alpha\beta}(\omega, \mathbf{k}) = \sum_{s_1, s_2} \int d\mathbf{r} d\mathbf{r}' \mathbf{d}_{s_1 s_2}^{\alpha} (\mathbf{d}_{s_1 s_2}^{\beta})^* e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')} [G_{\omega-\mu_{12}}^R + G_{\omega-\mu_{12}}^R]. \quad (22)$$

The retarded Green's functions which enters in this equation

$$G_{\omega}^R(x, x') \equiv -i \int_0^{\infty} dt e^{-i\omega t} \langle [\psi_2(x, t) \psi_1(x, t), \psi_1^+(x') \psi_2^+(x')] \rangle$$

is connected when  $T = 0$  with the Causal function (18) by the relation<sup>[9]</sup>

$$\text{Im } G_{\omega}^R = \frac{\omega}{|\omega|} \text{Im } G_{\omega}, \quad \text{Re } G_{\omega}^R = \text{Re } G_{\omega}.$$

We are interested in frequencies such that  $|\omega - \omega_0| \ll E_e$ . We can then retain in (22) only the terms that pertain to the ground state, and neglect the nonresonant terms. In this case we have

$$\chi^{\alpha\beta}(\omega, \mathbf{k}) = - \sum_m \frac{d_m^{\alpha}(d_m^{\beta})^*}{r_0^3} \left[ \frac{N_{\mathbf{k}}^m + 1}{\omega - \mu_{12} - \omega_m(\mathbf{k}) + i\delta} - \frac{N_{\mathbf{k}}^m}{\omega - \mu_{12} + \omega_m(\mathbf{k}) + i\delta} \right] \quad (23)$$

where

$$\mathbf{d}_m = r_0^{3/2} \sum_{s_1, s_2} \mathbf{d}_{s_1 s_2} \int d\mathbf{r} \chi_m(\mathbf{r} s_1; \mathbf{r} s_2)$$

is the effective dipole moment of the transition, and  $\omega$  and  $\mathbf{k}$  are the frequency and wave vector of the light in the semiconductor. As seen from (16), in the absence of condensate we have  $N_{\mathbf{k}}^m = 0$ ,  $\omega_m(\mathbf{k}) = k^2/2M$ , and the expression (23) takes on the usual form of the polarizability of an ideal gas of atoms with a dipole moment squared equal to  $\sum_m d_m^{\alpha}(d_m^{\beta})^*$ , a resonant frequency  $\mu_{12}$ , and a concentration  $r_0^{-3}$ <sup>[4]</sup>. In this case, as always, there is absorption of light at the resonant frequency.

In the presence of the condensate, the imaginary part of  $\chi^{\alpha\beta}$ , which determines the absorption, is

$$\chi''^{\alpha\beta}(\omega, \mathbf{k}) = \pi \sum_m \frac{d_m^{\alpha}(d_m^{\beta})^*}{r_0^3} [(N_{\mathbf{k}}^m + 1) \delta(\omega - \mu_{12} - \omega_m(\mathbf{k})) - N_{\mathbf{k}} \delta(\omega - \mu_{12} + \omega_m(\mathbf{k}))]. \quad (24)$$

We see that besides increasing the absorption at the frequencies  $\mu_{12} + \omega_m(\mathbf{k})$ , amplification of the light at the frequency  $\mu_{12} - \omega_m(\mathbf{k})$  takes place. The absorption is connected with the ordinary process, in which the energy of the absorbed quantum of light goes into the formation of an exciton with a c.m.s. momentum equal to the moment of the light quantum. In our case, however, the produced exciton interacts with the condensate, and the energy of its center of gravity is renormalized and, as seen from the formula for  $\omega_m(\mathbf{k})$ , it turns out to be much larger when  $k \ll Mv_m$  than the recoil energy of the free exciton  $k^2/2M$ . The increase in the absorption is connected with the fact that the interaction causes pairs of excitons with opposite momenta to be "pushed out" from the condensate. This leads to a nonzero distribution function of the excitons over the momenta  $N_{\mathbf{k}}^m$  at zero temperature. We have already seen that a low-density exciton gas behaves like a Bose-particle gas. It is therefore natural that the exciton production probability is proportional to  $N_{\mathbf{k}}^m + 1$ .

The amplification of the light is due to stimulated annihilation of the excitons. Owing to the momentum conservation law, the only excitons that can be transformed into light are those having a momentum equal to the momentum of light. The interaction causes two excitons with momenta  $\mathbf{k}$  and  $-\mathbf{k}$  to be "expelled" from the condensate. One of them subsequently annihilates

and emits a quantum of light, while the other excites a condensate acoustic wave with frequency  $\omega_m(\mathbf{k})$ . Since the energy of the exciton at rest is  $\mu_{12}$ , the frequency of the emitted light is  $\mu_{12} - \omega_m(\mathbf{k})$ . It is clear that the probability of this process is proportional to  $N_{\mathbf{k}}^m$ .

We see that the difference in the frequencies of the absorption and amplification of the light is equal to  $2\omega_m(\mathbf{k})$  and increases with increasing concentration of the condensate. At concentrations such that  $f_{mn} \gtrsim k^2$ ,  $N_{\mathbf{k}}^m$  becomes larger than or of the order of unity and the amplification coefficient turns out to be of the order of the absorption coefficient. This condition is attained in GaAs at exciton concentrations on the order of  $10^{16} \text{ cm}^{-3}$ .

Generally speaking, to find the connection between the frequency and the momentum of light in matter it is necessary to take into account the contribution of the excitons to the real part of the polarizability of the crystal, which is determined by expression (23). It follows from this expression that the polarizability of the exciton condensate has a resonance at frequencies corresponding to absorption and amplification of light. Let us ascertain the degree to which this addition influences the dispersion of light. It is necessary to take into account here that the exciton line width  $\delta$  is finite and is determined essentially by the scattering of the excitons by the impurities. At charge-impurity concentrations on the order of  $10^{15} \text{ cm}^{-3}$ , the mean free path time  $\delta^{-1}$  of an exciton with a momentum equal to the momentum of the absorbed or emitted light is of the order of  $10^{-10}$  sec in GaAs. Since the characteristic frequency  $\Omega = 4\pi d^2/r_0^3 \hbar$  ( $d$ —dipole moment of the transition between bands), which determines the interaction of the excitons with the light, is of the order of  $10^{10} \text{ sec}^{-1}$  in GaAs, the addition to the dielectric constant of the crystal, due to the excitons, will be  $\Omega/\delta \sim 1$  at resonance.

Inasmuch as the dielectric constant of the crystal is  $\epsilon \approx 10$ , we can neglect the contribution of the excitons. This means that, owing to the scattering of the excitons and their weak coupling with the light, no effective intermixing of the exciton and optical branches takes place near their intersection. Consequently the dispersion of the velocity of light near the amplification and absorption exciton lines can be neglected, and the amplification of light over the wavelength is  $4\pi\chi''/\epsilon$ . Thus, the amplification coefficient at the center of the line is of the order of

$$\Omega N_{\mathbf{k}}^m / \epsilon \delta \sim N_{\mathbf{k}}^m / \epsilon.$$

We recall that at exciton concentrations on the order of  $10^{16} \text{ cm}^{-3}$  we have  $N_{\mathbf{k}}^m \sim 1$ .

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