Stimulated exciton pairing and polarization of biexcitons in a laser radiation field

A. I. Bobrysheva, S. A. Moskalenko, and Hoang Ngok Kam

Institute of Applied Physics, Moldavian Academy of Sciences

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We show that a coherent macroscopic state of excitons with a wave vector $\mathbf{k}_0$ stimulated by resonant laser radiation induces a similar state in biexcitons with a wave vector $2\mathbf{k}_0$. The case of excitation, by linearly polarized light, of a single state from the threefold degenerate level Γ_5 of an exciton in a crystal of the CuCl type is studied. Exciton interaction leads to the formation of extracondensate excitons of three states of the Γ_5 type and an adjacent level Γ_2, which together participate in coherent pairing and formation of biexcitons. Since one type of exciton is specified by pumping conditions, a biexciton proves to be polarized when the detuning from resonance is finite. Polarization is found to lead to a variation in the angular dependence of the probability of a two-photon transition from the ground state of the crystal to a biexcitonic state.

1. INTRODUCTION AND STATEMENT OF THE PROBLEM

Coherent pairing of electrons and holes in semiconductors, which leads to Bose condensation of excitons in the low-density case and to an excitonic insulator in the opposite limit, has been considered by Keldysh and Kozlov,[1] Keldysh and Kopaev,[2] and Comte and Nozieres.[3] Laser radiation stimulates coherent pairing. The laser frequency acts as the chemical potential and the amplitude of the laser radiation as the source determining the amplitude of the coherent macrofilled quasiparticle mode.[4,5] This range of phenomena is known as the optical Stark effect in the excitonic region of the spectrum and has in recent years been under intensive investigation in the papers of Schmitt-Rink, Chemla, and Haug,[6] M. Combescot and R. Combescot,[7] Hanamura,[8] Ivanov, Keldysh, and Panashchenko,[9] and others. The later works (Refs. 7–9) discuss the role of biexcitons in the shifting and splitting of exciton energy levels and in the emergence of quasienergy branches in the spectrum, topics earlier discussed in Ref. 10.

Coherent pairing of fermions has been generalized to the case of coherent pairing of bosons, with excitons acting as bosons. In the latter case there appears a Bose condensate of biexcitons.[11]

The possibility of coherent pairing in a system consisting of four types of exciton with different projections of spins of both electrons and holes was studied by Khadzhi, Moskalenko, and Belkin.[12] These researchers considered two limiting cases, one when the exciton–exciton interaction energy is higher than the ortho-para splitting energy and the other when it is lower. The spin structure of the excitons leads to a situation in which the interaction of an exciton of any of the four types with excitons of all types is, on the average, repulsive, which ensures the stability of the biexciton condensate under collapse.

In this paper we examine the possibility of coherent pairing of excitons in the presence of polarized laser radiation that simulates Bose-Einstein condensation of excitons. We employ a crystal model of the CuCl type, where there are closely spaced exciton levels Γ_5 and Γ_2. The wave function of two excitons participating in formation of a bound biexciton state Γ_1 has the form

$$\psi_{\Gamma_1}(\mathbf{k}_1,\mathbf{k}_2)=\Big[\sum_i \psi_{\Gamma_{5i}}(\mathbf{k}_1)\psi_{\Gamma_{5i}}(\mathbf{k}_2)-\psi_{\Gamma_2}(\mathbf{k}_1)\psi_{\Gamma_2}(\mathbf{k}_2)\Big], \qquad (1)$$

where $\psi_{\Gamma j}(\mathbf{k})$ is the exciton wave function corresponding to row j in the Γ representation, and $\mathbf{k}$ the wave vector of translational motion. As Eq. (1) implies, the weight factors of the four components of $\psi_{\Gamma_1}(\mathbf{k}_1,\mathbf{k}_2)$ are equal in absolute value. The separation between the levels Γ_5 and Γ_2 is assumed small compared to the biexciton dissociation energy.

In the presence of polarized laser radiation with a detuning from resonance, Δ, greater than the interaction energy of the quasiparticles, the numbers of excitons of the four types are different. At exact resonance ($\Delta=0$) the excitons undergo a rapid transformation which removes this difference between the excitons of the four types. In the general case ($\Delta\neq 0$) one should expect the weight factors in (1) to change. This results in polarization of the biexciton and in variation of the angular dependence of the probability of two-photon transition from the ground state of the crystal to a biexcitonic state. Since the biexciton creation and annihilation operators are introduced as the linear combination (1) of products of exciton operators, the Bose condensation of excitons into a state with a wave vector $\mathbf{k}_0$ results in the Bose condensation of biexcitons into a state with the wave vector $2\mathbf{k}_0$. Two Bose condensates, the one-particle-excitonic and the two-particle-biexcitonic, appear and coexist simultaneously due to exciton interaction.

The need to redefine biexciton operators in the presence of laser radiation was first pointed out by Ivanov, Keldysh, and Panashchenko.[9]

2. COHERENT PAIRING AND THE STRUCTURE OF A BIEXCITON

The Hamiltonian of the excitons of four types $i=1,2,3,4$ (these number the states Γ_{5x}, Γ_{5y}, Γ_{5z}, and Γ_2, respectively), which intereact with each other and with the laser radiation, has the form

$$H=H_0+H_1+H_2, \qquad (2)$$

where

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$$H_0=\sum_{\mathbf{k}}\sum_{j=1}^{3}E_{\Gamma_5}(\mathbf{k})a_{j\mathbf{k}}^+a_{j\mathbf{k}}+\sum_{\mathbf{k}}E_{\Gamma_2}(\mathbf{k})a_{4\mathbf{k}}^+a_{4\mathbf{k}} \quad (3)$$

$$H_1=-\mu_{ex}F_{\mathbf{k}_0}^{1/2}[\exp(-i\omega_L t)a_{1\mathbf{k}_0}^+ +\exp(i\omega_L t)a_{1\mathbf{k}_0}], \quad (4)$$

$$H_2=\frac{1}{2V}\sum_{\mathbf{k}_1\mathbf{k}_2\mathbf{q}}\Big[\frac{1}{4}(3\nu_{ss}+\nu_{aa})\sum_{i=1}^{4}a_{i\mathbf{k}_1}^+a_{i\mathbf{k}_2}^+a_{i\mathbf{q}}a_{i\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}}$$
$$+\sum_{i,j=1;\, i\neq j}^{4}\nu_{ss}a_{i\mathbf{k}_1}^+a_{j\mathbf{k}_2}^+a_{j\mathbf{q}}a_{i\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}}$$
$$+\sum_{i,j=1;\, i\neq j}^{3}\frac{1}{4}(\nu_{aa}-\nu_{ss})a_{i\mathbf{k}_1}^+a_{i\mathbf{k}_2}^+a_{j\mathbf{q}}a_{j\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}}$$
$$+\sum_{j=1}^{3}\frac{1}{4}(\nu_{ss}-\nu_{aa})(a_{j\mathbf{k}_1}^+a_{j\mathbf{k}_2}^+a_{4\mathbf{q}}a_{4\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}}$$
$$+a_{4\mathbf{k}_1}^+a_{4\mathbf{k}_2}^+a_{j\mathbf{q}}a_{j\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}})\Big], \quad (5)$$

with μ_{ex} the coupling constant proportional to the matrix element of the dipole transition from the ground state of the crystal to an excitonic state, ω_L the laser radiation frequency, and $\nu_{ss}(\mathbf{k}_1,\mathbf{k}_2;\ \mathbf{q},\ \mathbf{k}_1+\mathbf{k}_2-\mathbf{q})$ and $\nu_{aa}(\mathbf{k}_1,\mathbf{k}_2;\ \mathbf{q},\ \mathbf{k}_1+\mathbf{k}_2-\mathbf{q})$ the Fourier transforms of the exciton interaction energy, which are constructed from the wave functions of two excitons, symmetric or antisymmetric, under simultaneous permutations of the projections of the spins of the two electrons and the projections of the angular momenta of the two holes. For small exciton momenta,

$$\nu_{aa}(\mathbf{k}_1,\ \mathbf{k}_2;\ \mathbf{q},\ \mathbf{k}_1+\mathbf{k}_2-\mathbf{q})\approx-\nu_{ss}(\mathbf{k}_1,\ \mathbf{k}_2;\ \mathbf{q},\ \mathbf{k}_1+\mathbf{k}_2-\mathbf{q}), \quad (6a)$$

and

$$\nu_{ss}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{q}.\mathbf{k}_1+\mathbf{k}_2-\mathbf{q})=\nu_{ss}(0)=\frac{26\pi}{3}R_y^{ex}a_{ex}^3, \quad (6b)$$

where R_y^{ex} and a_{ex} denote the ionization potential and the exciton radius, respectively.

We will assume that the laser radiation is polarized in the direction $\mathbf{e}_1\|x$ and excites only one excitonic mode with the wave vector $\mathbf{k}_0$. The polariton effect is not taken into account since it has no influence on the formation of biexcitons.

The time dependence in (4) can be eliminated by applying the unitary transformation $\hat{u}=(\exp)(-i\omega_L t\hat{N})$, where $\hat{N}=\Sigma_{iq}a_{iq}^+a_{iq}$. Instead of the operator $\hat{H}$ we will consider the operator $\mathscr{H}=\hat{u}^+\hat{H}\hat{u}-\hbar\omega_L\hat{N}$.

We introduce the two Bose condensates, of excitons and of biexcitons, into the Hamiltonian via the unitary transformation

$$D=D_1([N_{x\mathbf{k}_0}^{ex}]^{1/2})D_2([N_{x2\mathbf{k}_0}^{biex}]^{1/2}), \quad (7)$$

where D_1 and D_2 are the displacement operators in the exciton and biexciton operators, with their macroscopic values $N_{x\mathbf{k}_0}^{ex}\sim V$ and $N_{x2\mathbf{k}_0}^{biex}\sim V$ obeying the following relation: The operator D_1 has the form

$$D_1([N_{x\mathbf{k}_0}^{ex}]^{1/2})=\exp\{[N_{x\mathbf{k}_0}^{ex}]^{1/2}(a_{1\mathbf{k}_0}^+-a_{1\mathbf{k}_0})\}, \quad (8)$$

and D_2 is determined in a similar manner in terms of the biexciton creation and annihilation operators $\hat{\psi}_{2\mathbf{k}_0}^+$ and $\hat{\psi}_{2\mathbf{k}_0}$. The latter have the form

$$\hat{\psi}_{2\mathbf{k}_0}^+=\frac{1}{V^{1/2}}\sum_{i=1}^{4}\sum_{\mathbf{q}}\Phi(\mathbf{q})C_i a_{i,\mathbf{k}_0+\mathbf{q}}^+a_{i,\mathbf{k}_0-\mathbf{q}}^+, \quad (9)$$

where the function of the relative motion of a biexciton, $\Phi(q)$, and the coefficients C_i obey the normalization conditions

$$\frac{1}{V}\sum_{\mathbf{q}}\Phi^2(\mathbf{q})=1;\qquad 2\sum_i|C_i|^2=1. \quad (10)$$

The displacement operation D_2 is equivalent to the (u_i, v_i) transformations of the exciton operators:

$$D_2a_{i,\mathbf{k}_0+\mathbf{q}}D_2^+=u_i(\mathbf{q})a_{i,\mathbf{k}_0+\mathbf{q}}-v_i(\mathbf{q})a_{i,\mathbf{k}_0-\mathbf{q}}^+, \quad (11)$$

where

$$u_i(\mathbf{q})=\cosh(2C_i[n_{2\mathbf{k}_0}^{biex}]^{1/2}\Phi(\mathbf{q})),$$
$$v_i(\mathbf{q})=\sinh(2C_i[n_{2\mathbf{k}_0}^{biex}]^{1/2}\Phi(\mathbf{q})), \quad (12)$$
$$u_i^2(\mathbf{q})-v_i^2(\mathbf{q})=1;\qquad n_{2\mathbf{k}_0}^{biex}=N_{2\mathbf{k}_0}^{biex}/V. \quad (13)$$

In the limit of low densities $n_{2\mathbf{k}_0}^{biex}a_b^3\ll 1$, where a_b is the biexciton radius, we have

$$v_i(\mathbf{q})\approx 2C_i[n_{2\mathbf{k}_0}^{biex}]^{1/2}\Phi(\mathbf{q}).$$

The conditions for the stability of the new ground state of the system described by the Hamiltonian $\hat{D}\hat{H}\hat{D}^+$ make it possible to conclude that $C_2=C_3=-C_4$ and to find equations that together with the normalization conditions (10) make it possible to determing the coefficients C_1 and C_2 and the concentrations $n_{\mathbf{k}_0}^{ex}=N_{\mathbf{k}_0}^{ex}/V$ and $n_{2\mathbf{k}_0}^{biex}$. The exciton concentration in the condensate is

$$n_{x\mathbf{k}_0}^{ex}=\frac{|\mu_{ex}|^2n_{x\mathbf{k}_0}^{ph}}{[E_{\Gamma_5}(0)-\hbar\omega_L+L_0/2+\mathscr{L}(\mathbf{k}_0)+M(\mathbf{k}_0)]^2+\gamma_{ex}^2} \quad (14)$$

where

$$L_0=n_{x\mathbf{k}_0}^{ex}\nu_{ss}(0),$$

$$\mathscr{L}(\mathbf{k}_0)=\frac{1}{V}\sum_{\mathbf{q}}\sum_{i=1}^{4}\nu_{ss}(\mathbf{k}_0,\mathbf{q};\mathbf{k}_0,\mathbf{q})v_i^2(2\mathbf{k}_0-\mathbf{q}),$$

$$M(\mathbf{k}_0)=\frac{1}{2V}\sum_{\mathbf{q}}\nu_{ss}(\mathbf{k}_0,\mathbf{k}_0;2\mathbf{k}_0-\mathbf{q},\mathbf{q})$$
$$\times\Big[\sum_{i=1}^{4}u_i(\mathbf{q})v_i(\mathbf{q})-2u_1(\mathbf{q})v_1(\mathbf{q})\Big] \quad (15)$$

and γ_{ex} has been introduced phenomenologically.

The Schrödinger equations for the functions $v_i(k)$ are

$$\left[2(E_{\Gamma_5}(0)-\hbar\omega_L+L_0+\mathscr{L}(\mathbf{k}_0))+\frac{\hbar^2\mathbf{k}^2}{m_{ex}}\right]u_1(\mathbf{k})v_1(\mathbf{k})$$

$$+\frac{1}{V}\sum_{\mathbf{q}}\nu_{aa}(\mathbf{k},2\mathbf{k}_0-\mathbf{k};\mathbf{q},2\mathbf{k}_0-\mathbf{q})\frac{1}{2}(3u_2(\mathbf{q})v_2(\mathbf{q})$$

$$-u_1(\mathbf{q})v_1(\mathbf{q}))$$

$$\times(u_1(\mathbf{k})u_1(2\mathbf{k}_0-\mathbf{k})+v_1(\mathbf{k})v_1(2\mathbf{k}_0-\mathbf{k}))$$

$$=\frac{L_0}{2}(u_1(\mathbf{k})u_1(2\mathbf{k}_0-\mathbf{k})+v_1(\mathbf{k})v_1(2\mathbf{k}_0-\mathbf{k})),\qquad(16)$$

$$\left[2(E_{\Gamma_5}(0)-\hbar\omega_L+L_0+\mathscr{L}(\mathbf{k}_0))+\frac{\hbar^2k^2}{m_{ex}}\right]u_2(\mathbf{k})v_2(\mathbf{k})$$

$$+\frac{1}{V}\sum_{\mathbf{q}}\nu_{aa}(\mathbf{k},2\mathbf{k}_0-\mathbf{k};\mathbf{q},2\mathbf{k}_0-\mathbf{q})\frac{1}{2}(u_2(\mathbf{q})v_2(\mathbf{q})$$

$$+u_1(\mathbf{q})v_1(\mathbf{q}))'$$

$$\times(u_2(\mathbf{k})u_2(2\mathbf{k}_0-\mathbf{k})+v_2(\mathbf{k})v_2(2\mathbf{k}_0-\mathbf{k}))$$

$$=-\frac{L_0}{2}(u_2(\mathbf{k})u_2(2\mathbf{k}_0-\mathbf{k})+v_2(\mathbf{k})v_2(2\mathbf{k}_0-\mathbf{k})).\qquad(17)$$

In the absence of laser radiation, $\Phi(\mathbf{q})$ satisfies the Schrödinger equation for relative biexciton motion:

$$\frac{1}{V}\sum_{\mathbf{k}}\frac{\hbar^2k^2}{m_{ex}}\Phi^2(\mathbf{k})$$

$$+\frac{1}{V^2}\sum_{\mathbf{k},\mathbf{q}}\nu_{aa}(\mathbf{k},2\mathbf{k}_0-\mathbf{k};\mathbf{q},2\mathbf{k}_0-\mathbf{q})\Phi(\mathbf{q})\Phi(\mathbf{k})$$

$$=-I_b=\langle T\rangle+\langle V\rangle,\qquad(18)$$

where $E_{biex}(0)=2E_{\Gamma_5}(0)-I_b$, with I_b the biexciton dissociation energy, and $\langle T\rangle$ and $\langle V\rangle$ the average kinetic and potential energies of internal motion. In the zeroth approximation in the biexciton concentration we arrive at the following expressions of the C_i coefficients:

$$C_1=\frac{(|\langle V\rangle|-\Delta_{biex})}{\sqrt{2}[3(|\langle V\rangle|+\Delta_{biex})^2+(|\langle V\rangle|-\Delta_{biex})^2]^{1/2}},\qquad(19)$$

$$C_2=\frac{(|\langle V\rangle|+\Delta_{biex})}{\sqrt{2}[3(|\langle V\rangle|+\Delta_{biex})^2+(|\langle V\rangle|-\Delta_{biex})^2]^{1/2}}\qquad(20)$$

and, as a corollary,

$$C_2-C_1=\frac{2^{1/2}\Delta_{biex}}{[3(|\langle V\rangle|+\Delta_{biex}]^2+(|\langle V\rangle|-\Delta_{biex})^2]^{1/2}}.\qquad(21)$$

Here the detuning from resonance is defined as

$$\Delta_{biex}=2(E_{\Gamma_5}(0)-\hbar\omega_L+L_0+\mathscr{L}(\mathbf{k}_0))-I_b=E_{biex}(0)-2\hbar\omega_L+2(L_0+\mathscr{L}(\mathbf{k}_0)).\qquad(22)$$

In this approximation the biexciton concentration in the condensate is

$$n_{2\mathbf{k}_0}^{biex}=\frac{(n_{x\mathbf{k}_0}^{ex}\nu_{ss}(0))^2}{\pi a_b^3[4(C_1-C_2)^2(\Delta_{biex}+2|\langle V\rangle|)^2+\gamma_{biex}^2]}.\qquad(23)$$

Here we have also introduced a phenomenological decay constant for biexcitons, γ_{biex}.

3. POLARIZATION OF QUANTUM TRANSITIONS

Let us consider the probability of a transition from the ground state of the crystal to a biexcitonic state. The biexciton wave function in the zeroth approximation is

$$|\psi_{2\mathbf{k}}^{biex}\rangle=\hat{\psi}_{2\mathbf{k}}^{+}|0\rangle=\frac{1}{V^{1/2}}\sum_{\mathbf{q}}\Phi(\mathbf{q})[C_1a_{1,\mathbf{k}+\mathbf{q}}^{+}a_{1,\mathbf{k}-\mathbf{q}}^{+}+C_2(a_{2,\mathbf{k}+\mathbf{q}}^{+}a_{2,\mathbf{k}-\mathbf{q}}^{+}+a_{3,\mathbf{k}+\mathbf{q}}^{+}a_{3,\mathbf{k}-\mathbf{q}}^{+}-a_{4,\mathbf{k}+\mathbf{q}}^{+}a_{4,\mathbf{k}-\mathbf{q}}^{+})]|0\rangle,\qquad(24)$$

where C_1 and C_2 are determined by (19) and (20), and $|0\rangle$ is the crystals ground state.

If we assume that the polarizations of the two probing photons are both equal to $\bar{\mathbf{e}}_t$ the transition probability P is found to depend on the orientation of $\bar{\mathbf{e}}_t$ with respect to the x axis along which the pump radiation is polarized. The fractional value of the probability, P/P_0, is given by the following relation:

$$P/P_0=\begin{cases}8|C_1|^2, & \mathbf{e}_t\|x\\ 8|C_2|^2, & \mathbf{e}_t\perp x\end{cases},\qquad(25)$$

where P_0 is the probability of transition in the absence of biexciton transformations, and $|C_1^0|^2=|C_2^0|^2=1/8$. The dependence of P/P_0 on the detuning Δ_{biex} in units of $|\langle V\rangle|$ is depicted in Fig. 1.

It has proved more convenient experimentally to use in the two-photon transition one photon from the pump laser proper. Then the second probing photon must have a polarization $(\mathbf{e}_t\bar{\mathbf{e}}_x)^2\neq0$ and an energy $\hbar\omega_L+\Delta_{biex}$.

To conclude, we note that as the detuning from resonance grows and the deviation of the curves *1* and *2* from

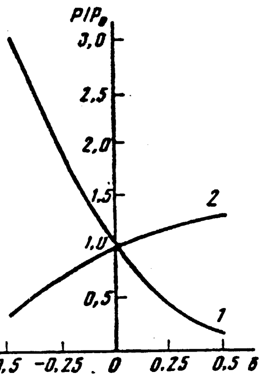

FIG. 1. The relative probabilities of two-photon transitions for two different orientations of the polarization vector of the probing photons: curve *1*, $\mathbf{e}_t\|x$; curve *2*, $\mathbf{e}_t\perp x$; $\delta\Delta_{biex}/V\rangle|$.

unity increases, the biexciton binding energy decreases, apparently, and the biexciton radius increases. These factors are not taken into account in this paper, and for this reason our results are valid only in a small region δ. But even in this case the effect is significant and can be observed in experiments.

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