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Biexciton-biexciton interaction in semiconductors

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For a semiconductor with single isotropic conduction and valence bands, the effective biexciton-biexciton interaction is derived starting from the second-quantization representation for the Hamiltonian of the system of interacting excitons and for the biexciton wave function within the framework of the adiabatic approximation. The interaction is found to be an average of the sum of effective interactions between excitons forming interacting biexcitons over the envelope functions of these biexcitons. Depending on the momentum transfer and on the difference between the momenta of interacting biexcitons, the interaction admits an analytical study when the first vanishes. In this case, the interaction has in the **r** space the form of a function of the interbiexciton distance consisting of a strong repulsive and a weak attractive part. At low temperatures the function describes main features in the behavior of the interaction among biexcitons, in which repulsion predominates over attraction. It is shown that while biexcitons remain stable quasiparticles they weakly attract each other for any value of the distance. A quantitative analysis of obtained results for the CuCl crystal shows that the biexciton system in this model substance is a weakly nonideal Bose gas with positive scattering length $a_s \simeq 3a_x$, which closely approaches the ideal one at excitation densities $n \leqslant 10^{18}$ cm^{-3}. [S0163-1829(97)02211-X]

I. INTRODUCTION

Since that time, when Moskalenko[1] and Lampert[2] independently predicted the existence of the biexciton (or excitonic molecule) in semiconductors, the study of this bound complex of two electrons and two holes has rapidly developed. The biexciton has been theoretically[3–7] as well as experimentally[8–14] shown to be the most stable entity of the lowest energy per electron-hole (e-h) pair at low temperatures in many semiconductors.[15] In these semiconductors, the role of the biexciton in the nonlinear optical phenomena in the spectral vicinity of the band edge has long been recognized.[13–25] The electronic excitation at intermediate excitation levels forms a dense gas of charge carriers, excitons and biexcitons.[26] As a quasiparticle, the exciton has bosonlike character when the number of excited e-h pairs is negligibly small compared to the total number of valence electrons. For a finite density, a hypothetical boson space may be imagined, where excitons appear as ideal boson and their deviation from ideal bosons is described by effective interaction between these bosons.[27] It is exactly this exciton-exciton interaction that ultimately determines almost all excitonic optical nonlinearities including the biexciton formation and related phenomena. During the past decades, the interaction between excitons has been a subject of intensive studies within different approaches by many research groups.[5,6,27–33] In the meantime, because of calculational difficulty of the four-exciton problem, very little is known about the interaction between biexcitons which are approximately considered as bound states of two excitons. Brinkman and Rice have been the first and the only so far, to our knowledge, who considered the problem of the interaction between biexcitons.[34] Following closely the similarity between the biexciton and the hydrogen molecule, the authors have calculated the quantum mechanical interbiexciton interaction potential as a function of the interhole distance and pointed out that the biexciton-biexciton interaction is predominantly repulsive. Their result provides the first insight into the biexciton-biexciton interaction strength though it is certainly not sufficient to understand many features in the optical property of the biexciton. Moreover, the bosonlike nature of excitonic particles has been the basis for expecting them to undergo Bose-Einstein condensation (BEC).[6,29,35–37] Except for the special case of the Cu_2O crystal, where a weak (due to nearly equal electron and hole masses) biexciton creation is suppressed by strong e-h exchange interaction [38] and quantum-statistical properties have been displayed in an exciton gas,[39–41] in most other semiconductors BEC is expected to occur inside the biexciton system. Physically, the biexciton condensation can be considered as a result of pair condensation,[42] or coherent pairing[43] in an exciton gas with mutual attraction. The observation of Bose condensates in CuCl was reported many years ago.[10,44] Recently more promising results were obtained by a different experimental approach.[45] Nevertheless, clear evidence for the BEC of biexcitons has not appeared yet. In theoretical respect, the interaction between particles is one of most important aspects of the problem of BEC in a system of weakly interacting bosons. The property of such a system depends critically on the small-momentum scattering amplitudes, among which the only significant one is the amplitude for an ''s-wave'' collision. The determination of this quantity has therefore been a major issue for the study of the possibility of a condensate formation and its properties. Consequently, an elaborate theory of the biexciton-biexciton interaction is of importance not only to the fundamental exciton physics, but as well in connection with experimental efforts to observe BEC in excitonic systems.

The aim of the present paper is to derive the effective biexciton-biexciton interaction in a semiconductor with

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simple band structure; the biexciton we refer to here is the lowest one of the Γ_1 symmetry. Certainly we limit ourselves to excitation densities n satisfying the condition $na_{\mathrm{bx}}^3 \ll 1$, a_{bx} is the biexciton radius, where the biexciton (and the more so the exciton with radius $a_{\mathrm{x}} < a_{\mathrm{bx}}$) remains a stable quasiparticle. Throughout the paper, subscripts x and bx will denote exciton and biexciton, respectively. Most of the biexcitons are assumed to be formed through the interaction among excitons injected into the semiconductor from an incoherent light source with frequency very close to the $1s$-exciton energy level. Under the resonant condition, all of the excitons in the system can be considered ground-state ones. The effective interaction between these excitons, especially that among paraexcitons in the Cu_2O crystal, and the form of the biexciton wave function are discussed in Sec. II. Section III is devoted to the derivation of the effective biexciton-biexciton interaction, the analysis of a particular case allowing an analytical study of the interaction and illustrations on the CuCl crystal. Finally, discussions and conclusions are given in Sec. IV. Throughout the paper we set $\hbar = 1$.

II. EXCITON EFFECTIVE HAMILTONIAN AND BIEXCITON WAVE FUNCTION

We consider a direct cubic semiconductor with isotropic single conduction and valence bands at low temperatures. In such a semiconductor, ground-state $(1s\text{-})$ excitons are of two kinds: the paraexciton Γ_2 with angular momentum $J_x = 0$ and the orthoexciton Γ_5 with $J_x = 1$ which is higher in energy by a value Δ of the e-h exchange interaction. Denoting three basis vectors of the irreducible representation Γ_5 of the crystal symmetry group by $\Gamma_5 X$, $\Gamma_5 Y$, $\Gamma_5 Z$, we write the effective Hamiltonian of the system of interacting $1s$-excitons within the framework of the boson description as follows:

$$
\begin{aligned}
H_{\mathrm{x}} = {} & \sum_{\xi,\mathbf{k}} E_{\mathrm{x}}^{\xi}(k) a_{\xi\mathbf{k}}^{\dagger} a_{\xi\mathbf{k}} + \frac{1}{2} \sum_{\mathbf{q},\mathbf{k}_1,\mathbf{k}_2} \Bigg\{ \Bigg[U^d(\mathbf{q}) \\
& + \frac{1}{2} U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2) \Bigg] \sum_{\xi} a_{\xi\mathbf{k}_1+\mathbf{q}}^{\dagger} a_{\xi\mathbf{k}_2-\mathbf{q}}^{\dagger} a_{\xi\mathbf{k}_2} a_{\xi\mathbf{k}_1} + [U^d(\mathbf{q}) \\
& + U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2)] \\
& \times \sum_{\xi_1 \neq \xi_2} a_{\xi_1\mathbf{k}_1+\mathbf{q}}^{\dagger} a_{\xi_2\mathbf{k}_2-\mathbf{q}}^{\dagger} a_{\xi_2\mathbf{k}_2} a_{\xi_1\mathbf{k}_1} - \frac{1}{2} U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2) \\
& \times \sum_{\xi_1 \neq \xi_2} C_{\xi_1} C_{\xi_2} a_{\xi_1\mathbf{k}_1+\mathbf{q}}^{\dagger} a_{\xi_1\mathbf{k}_2-\mathbf{q}}^{\dagger} a_{\xi_2\mathbf{k}_2} a_{\xi_2\mathbf{k}_1},
\end{aligned}
\tag{1}
$$

where the sums over ξ, ξ_1, ξ_2 go over all possible symmetry states of the $1s$-exciton: Γ_2, $\Gamma_5 X$, $\Gamma_5 Y$, $\Gamma_5 Z$ which for the sake of brevity will be hereby denoted by O, X, Y, Z, respectively. $a_{\xi\mathbf{k}}^{\dagger}$ $(a_{\xi\mathbf{k}})$ is the creation (annihilation) operator for the exciton of the symmetry ξ with momentum $\mathbf{k}$ and energy $E_{\mathrm{x}}^{\xi}(k)$ obeying Bose-Einstein statistics,

$$
[a_{\xi'\mathbf{k}'}, a_{\xi\mathbf{k}}^{\dagger}] = \delta_{\xi'\xi}\delta_{\mathbf{k}'\mathbf{k}},
$$

$$
[a_{\xi'\mathbf{k}'}^{\dagger}, a_{\xi\mathbf{k}}^{\dagger}] = [a_{\xi'\mathbf{k}'}, a_{\xi\mathbf{k}}] = 0. \tag{2}
$$

The coefficient C_{ξ} in the last sum on the right-hand side (rhs) of (1) is defined as follows:

$$
C_0 = -1, \quad C_{\xi} = 1 \quad \text{for } \xi = X, Y, Z. \tag{3}
$$

U^d and U^{ex} are, respectively, the direct and the exchange exciton interaction function parametrically depending on the e-h mass ratio $\sigma \equiv (m_e/m_h)$ through $\alpha = [\sigma/(1+\sigma)]$ and $\beta = [1/(1+\sigma)]$. A general formula for U^d and U^{ex} is well known.[5,31–33] The function U^d describes the direct Coulomb interaction between two excitons and depends on the momentum transfer,

$$
U^d(\mathbf{q}) = \frac{1}{V} \int d^3r \exp(i\mathbf{q}\mathbf{r}) \mathcal{U}^d(\mathbf{r}), \tag{4}
$$

where V is the crystal volume and $\mathcal{U}^d(\mathbf{r})$ is a function of the distance between interacting excitons[5]

$$
\begin{aligned}
\mathcal{U}^d(\mathbf{r}) = {} & 2\mathcal{R} \sum_{\tau=\alpha,\beta} \frac{1}{\tau} \exp\left(-\frac{2r}{\tau a_{\mathrm{x}}} \right) \\
& \times \Bigg\{ \left[\frac{6\tau^4}{(2\tau-1)^2} - \frac{4\tau^6}{(2\tau-1)^3} - 1 \right] \frac{\tau a_{\mathrm{x}}}{r} \\
& + \frac{2\tau^4}{(2\tau-1)^2} - \frac{11}{8} - \frac{3}{4}\frac{r}{\tau a_{\mathrm{x}}} - \frac{1}{6}\left(\frac{r}{\tau a_{\mathrm{x}}} \right)^2 \Bigg\},
\end{aligned}
\tag{5}
$$

with $\mathcal{R}$ denoting the exciton binding energy (exciton Rydberg), e the electron charge, and ϵ_0 the static dielectric constant of the semiconductor.

On the contrary, the function U^{ex} describes the effect due to Pauli exclusion principle acting between constituent particles belonging to different interacting excitons. Except for the momentum transfer, U^{ex} depends on the difference between the momenta of interacting excitons and therefore in the $\mathbf{r}$ space cannot be presented as a function of only the distance between these excitons. The nonlocality of $U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2)$ depends on $\alpha\beta$ and on values of vectors $\mathbf{q}$ and $\mathbf{k}_1 - \mathbf{k}_2$. It can be neglected if either the mass of hole is much larger than that of the electron, or all momenta are very small. Here we find it convenient to present the exchange function in the following way:

$$
U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2) = U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}_1-\mathbf{k}_2+\mathbf{q}),
$$

$$
\begin{aligned}
U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}+\mathbf{q}) = {} & \frac{1}{V} \int\int d^3r\, d^3r' \exp[i\mathbf{q}\mathbf{r} + i(\mathbf{k}+\mathbf{q})\mathbf{r}'] \\
& \times \mathcal{U}^{\mathrm{ex}}(\mathbf{r},\mathbf{r}'),
\end{aligned}
\tag{6}
$$

where

$$\mathcal{U}^{\mathrm{ex}}(\mathbf{r},\mathbf{r}')=\frac{2\mathcal{R}}{\alpha^3\beta^3}\int d^3r_1\Biggl\{f(\mathbf{r}_1)f\left(\mathbf{r}_1-\frac{\mathbf{r}}{\alpha}-\frac{\mathbf{r}'}{\beta}\right)\times f\left(\mathbf{r}_1-\frac{\mathbf{r}}{\alpha}\right)f\left(\mathbf{r}_1-\frac{\mathbf{r}'}{\beta}\right)\times\left[\frac{a_x}{\left|\mathbf{r}_1-\frac{\mathbf{r}}{\alpha}-\frac{\mathbf{r}'}{\beta}\right|}-\frac{\alpha a_x}{r}\right]+f(\mathbf{r}_1)\times f\left(\mathbf{r}_1-\frac{\mathbf{r}'}{\alpha}-\frac{\mathbf{r}}{\beta}\right)f\left(\mathbf{r}_1-\frac{\mathbf{r}'}{\alpha}\right)f\left(\mathbf{r}_1-\frac{\mathbf{r}}{\beta}\right)\times\left[\frac{a_x}{\left|\mathbf{r}_1-\frac{\mathbf{r}'}{\alpha}-\frac{\mathbf{r}}{\beta}\right|}-\frac{\beta a_x}{r}\right]\Biggr\},\qquad(7)$$

with f denoting the $1s$ hydrogenlike function,

$$f(\mathbf{r})=\frac{1}{\sqrt{\pi a_{\mathrm{x}}^3}}\exp\left(-\frac{r}{a_{\mathrm{x}}}\right),$$

describing the relative motion of e-h pair in an exciton. As a sum of four-center integrals, $\mathcal{U}^{\mathrm{ex}}$ cannot be calculated analytically. Considering particular cases of Eq. (6),

$$U^{\mathrm{ex}}(\mathbf{k},\mathbf{0})=\frac{1}{V}\int d^3r\,\exp(i\mathbf{kr})\left[\int d^3r'\,\mathcal{U}^{\mathrm{ex}}(\mathbf{r},\mathbf{r}')\right],$$

and

$$U^{\mathrm{ex}}(\mathbf{0},\mathbf{k})=\frac{1}{V}\int d^3r\,\exp(i\mathbf{kr})\left[\int d^3r'\,\mathcal{U}^{\mathrm{ex}}(\mathbf{r}',\mathbf{r})\right],\qquad(8)$$

we find that functions

$$\mathcal{U}_1^{\mathrm{ex}}(\mathbf{r})\equiv\int d^3r'\,\mathcal{U}^{\mathrm{ex}}(\mathbf{r},\mathbf{r}')$$

and

$$\mathcal{U}_2^{\mathrm{ex}}(\mathbf{r})\equiv\int d^3r'\,\mathcal{U}^{\mathrm{ex}}(\mathbf{r}',\mathbf{r})\qquad(9)$$

can be evaluated. As $\mathcal{U}^d(r)$, these functions depend only on the distance between interacting excitons,

$$\mathcal{U}_1^{\mathrm{ex}}(\mathbf{r})=2\mathcal{R}\sum_{\tau=\alpha,\beta}\frac{1}{\tau^3}\left[\left(1+\frac{r}{\tau a_x}\right)\exp\left(-\frac{r}{\tau a_x}\right)-\frac{\tau a_x}{r}S\left(\frac{r}{\tau a_x}\right)\right]S\left(\frac{r}{\tau a_x}\right),$$

$$\mathcal{U}_2^{\mathrm{ex}}(\mathbf{r})=2\mathcal{R}\sum_{\tau=\alpha,\beta}\frac{1}{\tau^3}\Biggl\{\left(1+\frac{r}{\tau a_x}\right)\exp\left(-\frac{r}{\tau a_x}\right)S\left(\frac{r}{\tau a_x}\right)-\left[\frac{5}{8}-\frac{23}{20}\frac{r}{\tau a_x}-\frac{3}{5}\left(\frac{r}{\tau a_x}\right)^2-\frac{1}{15}\left(\frac{r}{\tau a_x}\right)^3\right]\exp\left(-\frac{2r}{\tau a_x}\right)$$
$$-\frac{6}{5}\frac{\tau a_x}{r}\{[\gamma+\ln(r/\tau a_x)]S(r/\tau a_x)^2+S(-r/\tau a_x)^2\mathrm{Ei}(-4r/\tau a_x)-2S(-r/\tau a_x)S(r/\tau a_x)\mathrm{Ei}(-2r/\tau a_x)\}\Biggr\},\qquad(10)$$

where $Ei(z)=\int_z^\infty[\exp(-y)/y]dy$ is the exponent integral, $\gamma\simeq0.577$ the Euler's constant, and

$$S(\lambda r)\equiv\left[1+\lambda r+\frac{1}{3}(\lambda r)^2\right]\exp(-\lambda r).\qquad(11)$$

Using the expressions for $\mathcal{U}^d(r)$, $\mathcal{U}_1^{\mathrm{ex}}(r)$, and $\mathcal{U}_2^{\mathrm{ex}}(r)$, the effective interaction potential between excitons of different symmetries at vanishing momentum transfer can be described. To shed some light on the problem of BEC in an exciton gas, we consider a particular case of interaction between paraexcitons which probably are elementary excitations of the lowest energy in the $\mathrm{Cu_2O}$ crystal. From the formula for the interaction potential between two paraexcitons with momenta $\mathbf{k}_1$, $\mathbf{k}_2$ and momentum transfer $\mathbf{q}=\mathbf{0}$,[33]

$$U_{\mathrm{eff}}^{p-p}(\mathbf{k})=\frac{1}{2}\left[U^d(\mathbf{k})+\frac{1}{2}U^{\mathrm{ex}}(\mathbf{0},\mathbf{k})+\frac{1}{2}U^{\mathrm{ex}}(\mathbf{k},\mathbf{0})\right],\qquad(12)$$

$\mathbf{k}\equiv\mathbf{k}_1-\mathbf{k}_2$ and using Eqs. (4), (8), and (9) we find that the function

$$\mathcal{U}_{\mathrm{eff}}^{p-p}(r)=\mathcal{R}\left[\mathcal{U}^d(r)+\frac{1}{2}\mathcal{U}_1^{\mathrm{ex}}(r)+\frac{1}{2}\mathcal{U}_2^{\mathrm{ex}}(r)\right]\qquad(13)$$

describes the potential in the $\mathbf{r}$ space. The variation of this function with the distance is shown in Fig. 1 for a semiconductor with $\sigma=0.7$. One can observe that the repulsion between paraexcitons decreases rapidly at $r>2a_x$ and vanishes at $r\geqslant3a_{\mathrm{x}}$: $\mathcal{U}_{\mathrm{eff}}^{p-p}(3a_{\mathrm{x}})<0.005\mathcal{R}$. Thus the system of paraexcitons in the $\mathrm{Cu_2O}$ crystal ($\sigma=0.7$, $a_x=7$ Å) is a nearly ideal Bose gas even at densities as high as 10^{19} cm^{-3} which has been indicated in experimental works on this crystal.[40,41] Because at low temperatures most particles in such a gas have small momentum $[k\ll(1/a_x)]$,[46] the character of the interaction between paraexcitons in a general case ($\mathbf{q}\neq\mathbf{0}$, $\mathbf{k}_1-\mathbf{k}_2\neq\mathbf{0}$) is expected to be similar to that represented by the curve in Fig. 1. By the use of $\mathcal{U}_{\mathrm{eff}}^{p-p}(r)$, the "$s$-wave" scattering length (or the hard-core radius) of paraexcitons in $\mathrm{Cu_2O}$ is estimated to be approximately $2.2a_x$.[47] The result is in agreement with the experimental upper bound provided by the authors of Ref. 48.

Further we assume for the considered semiconductor the orthopara splitting Δ to be much smaller than the biexciton

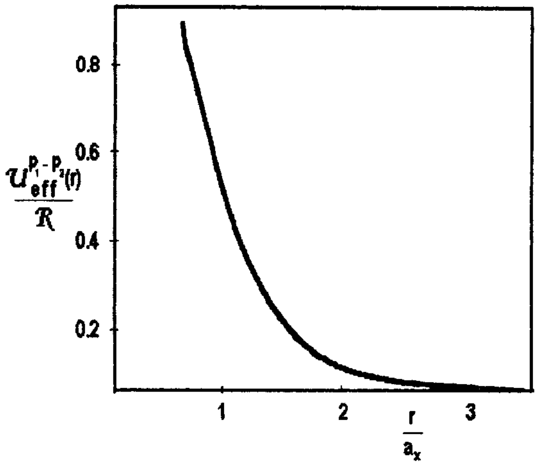

FIG. 1. Variation of the no-momentum-transfer interaction potential between two paraexcitons in Cu_2O with the distance. $\sigma = 0.7$.

binding energy E^b_{bx} (the situation is realized in CuCl where $\Delta = 6.2$ meV and $E^b_{\mathrm{bx}} = 30$ meV[15]). Then, in the formation of a biexciton participate two excitons in a superposition of zero-angular-momentum states which are made up of two ortho- and two paraexcitons, respectively.[15] Since E^b_{bx} is typically an order of magnitude smaller than the corresponding exciton binding energy,[49] the traditionally used adiabatic approximation[5,16,28] for the biexciton wave function is legitimate. In the approximation the biexciton state with momentum **K** can be written in the following form:

$$|\mathbf{K}\rangle = \frac{1}{\sqrt{8V}} \sum_{\mathbf{p}} \phi(\mathbf{p}) \sum_{\xi = O,X,Y,Z} C_\xi a^\dagger_{\xi(\mathbf{K}/2)-\mathbf{p}} a^\dagger_{\xi(\mathbf{K}/2)+\mathbf{p}} |0\rangle \tag{14}$$

where $\phi(\mathbf{p})$ is the Fourier transform of normalized spherically symmetric function $\Phi(\mathbf{r})$ describing the relative motion of two excitons in a biexciton. The equation for $\phi(\mathbf{p})$ can be obtained from the Heisenberg equation of motion for a biexciton with the aid of the Hamiltonian (1). It appears as follows:

$$\begin{aligned} &\left[E_{\mathrm{x}}\left(\frac{\mathbf{K}}{2} - \mathbf{p} \right) + E_{\mathrm{x}}\left(\frac{\mathbf{K}}{2} + \mathbf{p} \right) \right] \phi(\mathbf{p}) \\ &\quad + \sum_{\mathbf{q}} \left[U^d(\mathbf{q}) - U^{\mathrm{ex}}(\mathbf{q}, 2\mathbf{p} - \mathbf{q}) \right] \phi(\mathbf{p} - \mathbf{q}) \\ &= E_{\mathrm{bx}}(K)\, \phi(\mathbf{p}), \end{aligned} \tag{15}$$

where $E_{\mathrm{bx}}(K)$ is the energy of the biexciton with momentum **K**. In Eq. (15) the exciton energy is taken to be the same for both ortho- and paraexcitons,

$$E_{\mathrm{x}}(k) = E_T + \frac{k^2}{2m_{\mathrm{x}}}, \tag{16}$$

with E_T the transverse orthoexciton energy at $k = 0$ and m_{x} the exciton translational mass. Equation (15) admits no rigorous solution even in the case $\sigma = 0$ when it presents the Fourier transform of the Schrödinger equation for the energy and wave function of nuclei relative motion in a hydrogen molecule within the Heitler-London approximation. For any $\sigma \neq 0$, Eq. (15) corresponds to an integro–differential equation for $\Phi(\mathbf{r})$ which can be reduced to a differential one only if the nonlocality of $\mathcal{U}^{\mathrm{ex}}(\mathbf{r}, \mathbf{r}')$ is neglected. However, this equation will not be dealt with in the present paper. The biexciton envelope function is expected to play some averaging role in the problem of biexciton-biexciton interaction and its extension appears to have more influence upon the interaction rather than its form. In our study of the biexciton-biexciton interaction for a particular case, the hydrogenlike form

$$\Phi(\mathbf{r}) = \frac{1}{\sqrt{\pi a^3_{\mathrm{bx}}}} \exp\left(-\frac{r}{a_{\mathrm{bx}}} \right) \tag{17}$$

will be used for the biexciton envelope function. The extent of the biexciton then is defined by the radius a_{bx}, which is connected with the binding energy E^b_{bx} by the following relation:

$$\begin{aligned} E^b_{\mathrm{bx}} = \frac{1}{V\pi a^3_{\mathrm{bx}}} \Bigg[&\int\int d^3r\, d^3r' \exp\left(-\frac{|\mathbf{r} + \mathbf{r}'|}{a_{\mathrm{bx}}} - \frac{|\mathbf{r} - \mathbf{r}'|}{a_{\mathrm{bx}}} \right) \mathcal{U}^{\mathrm{ex}}(\mathbf{r}, \mathbf{r}') - \int d^3r \\ &\times \exp\left(-\frac{2r}{a_{\mathrm{bx}}} \right) \mathcal{U}^d(r) \Bigg], \end{aligned} \tag{18}$$

resulting from Eqs. (15) and (17). In (18) the biexciton binding energy E^b_{bx} is meant to be greater than the dissociation energy $D = 2E_T - E_{\mathrm{bx}}(0)$ by the energy of the so called zero-point vibration.[50] By an acceptable approximation for $\mathcal{U}^{\mathrm{ex}}(\mathbf{r}, \mathbf{r}')$ from (18) a formula relating a function of the biexciton-exciton radius ratio $a \equiv a_{\mathrm{bx}}/a_{\mathrm{x}}$ to a value of $E \equiv E^b_{\mathrm{bx}}/\mathcal{R}$ taken from experiment can be derived. In this way the biexciton radius may be estimated. The two quantities a and E are the most important individual characteristics of the biexciton. Along with σ, they determine the picture of the biexciton-biexciton interaction as one can see in the next section.

III. EFFECTIVE BIEXCITON-BIEXCITON INTERACTION

Since in the adiabatic approximation excitons are assumed to remain themselves binding into biexcitons, the effective interaction between biexcitons has as the origin the interaction between their component excitons. Then, we may expect the interaction between two biexcitons to be the sum of the effective interactions between constituent excitons averaged over wave functions of their relative motions. Introducing the creation operator $b_{\mathbf{K}}^{\dagger}$ for the biexciton with momentum $\mathbf{K}$, we write the biexciton wave function as follows:

$$|\mathbf{K}\rangle = b_{\mathbf{K}}^{\dagger}|\mathbf{0}\rangle. \tag{19}$$

Comparing (19) with (14), we get biexciton operators expressed in terms of exciton ones

$$b_{\mathbf{K}}^{\dagger} = \frac{1}{\sqrt{8V}}\sum_{\mathbf{p}} \phi(\mathbf{p}) \sum_{\xi=O,X,Y,Z} C_{\xi} a_{\xi(\mathbf{K}/2)-\mathbf{p}}^{\dagger} a_{\xi(\mathbf{K}/2)+\mathbf{p}}^{\dagger}. \tag{20}$$

From the bosonlike character of exciton operators and the normalization of the biexciton envelope function, we obtain the following commutation relation:

$$[b_{\mathbf{K}'}, b_{\mathbf{K}}^{\dagger}] = \delta_{\mathbf{K}'\mathbf{K}} + \frac{1}{2V}\sum_{\mathbf{p}} \phi(\mathbf{p})\phi\left(\mathbf{p} + \frac{\mathbf{K}'-\mathbf{K}}{2}\right) \times \sum_{\xi=O,X,Y,Z} a_{\xi(\mathbf{K}/2)+\mathbf{p}}^{\dagger} a_{\xi\mathbf{K}'-(\mathbf{K}/2)-\mathbf{p}}, \tag{21}$$

with $b_{\mathbf{K}}$ the annihilation operator for the momentum $\mathbf{K}$ biexciton. Equation (21) indicates that strictly speaking the biexciton is not a boson. However, it is easy to see that matrix elements of the last term on the rhs of (21) are of order na_{bx}^{3}. So, in the considered interval of excitation densities, the biexciton may be treated as a boson. Then two-biexciton wave function can be constructed as a direct product of single-biexciton wave functions for each biexciton,

$$|\mathbf{K}_1\mathbf{K}_2\rangle = N_{\mathbf{K}_1\mathbf{K}_2} b_{\mathbf{K}_1}^{\dagger} b_{\mathbf{K}_2}^{\dagger}|\mathbf{0}\rangle, \tag{22}$$

where $\mathbf{K}_1$ and $\mathbf{K}_2$ are the momenta of two biexcitons, and

$$N_{\mathbf{K}_1\mathbf{K}_2} = 1 + \left(\frac{1}{\sqrt{2}} - 1\right)\delta_{\mathbf{K}_1\mathbf{K}_2} \tag{23}$$

ensures the orthonormalization of two-biexciton wave function (22),

$$\langle\mathbf{K}_1'\mathbf{K}_2'|\mathbf{K}_1\mathbf{K}_2\rangle = \delta_{\mathbf{K}_1'\mathbf{K}_1}\delta_{\mathbf{K}_2'\mathbf{K}_2} + \delta_{\mathbf{K}_1'\mathbf{K}_2}\delta_{\mathbf{K}_1\mathbf{K}_2'} - \delta_{\mathbf{K}_1\mathbf{K}_2}\delta_{\mathbf{K}_1'\mathbf{K}_2'}\delta_{\mathbf{K}_1'\mathbf{K}_1}. \tag{24}$$

By the use of (20), (22) can be written as follows:

$$|\mathbf{K}_1\mathbf{K}_2\rangle = N_{\mathbf{K}_1\mathbf{K}_2}\frac{1}{8V}\sum_{\mathbf{p}_1}\phi(\mathbf{p}_1) \times \sum_{\xi_1=O,X,Y,Z} C_{\xi_1} a_{\xi_1(\mathbf{K}_1/2)-\mathbf{p}_1}^{\dagger} a_{\xi_1(\mathbf{K}_1/2)+\mathbf{p}_1}^{\dagger} \times \sum_{\mathbf{p}_2}\phi(\mathbf{p}_2) \times \sum_{\xi_2=O,X,Y,Z} C_{\xi_2} a_{\xi_2(\mathbf{K}_2/2)-\mathbf{p}_2}^{\dagger} a_{\xi_2(\mathbf{K}_2/2)+\mathbf{p}_2}^{\dagger}|\mathbf{0}\rangle. \tag{25}$$

Putting the Hamiltonian (1) of the exciton system between two-biexciton wave functions in the form (25), we arrive after intricate calculations with the use of Eqs. (2), (15), (24) at the following result:

$$\langle\mathbf{K}_2'\mathbf{K}_1'|H_{\mathrm{x}}|\mathbf{K}_1\mathbf{K}_2\rangle = [E_{\mathrm{bx}}(K_1) + E_{\mathrm{bx}}(K_2)]\langle\mathbf{K}_2'\mathbf{K}_1'|\mathbf{K}_1\mathbf{K}_2\rangle + \delta_{\mathbf{K}_1'+\mathbf{K}_2',\mathbf{K}_1+\mathbf{K}_2} \times \mathcal{P}(\mathbf{K}_1'-\mathbf{K}_1, \mathbf{K}_1'-\mathbf{K}_2), \tag{26}$$

where $\mathcal{P}(\mathbf{K}_1'-\mathbf{K}_1,\ \mathbf{K}_1'-\mathbf{K}_2)$ is the effective interaction potential between two biexcitons with momenta $\mathbf{K}_1$, $\mathbf{K}_2$ and with momentum transfer $\mathbf{Q}\equiv\mathbf{K}_1'-\mathbf{K}_1$. The potential is found to depend on $\mathbf{Q}$ and on the difference $\mathbf{K}=\mathbf{K}_1-\mathbf{K}_2$ between the momenta of two interacting biexcitons. It can be written as follows:

$$\mathcal{P}(\mathbf{Q},\mathbf{K}+\mathbf{Q}) = \frac{1}{2}[W(\mathbf{Q},\mathbf{K}+\mathbf{Q}) + W(\mathbf{K}+\mathbf{Q},\mathbf{Q})], \tag{27}$$

where W is the effective biexciton-biexciton interaction first introduced by Hanamura for the boson description of a biexciton gas by means of the model Hamiltonian,

$$H_{\mathrm{bx}} = \sum_{\mathbf{K}} E_{\mathrm{bx}}(K) b_{\mathbf{K}}^{\dagger} b_{\mathbf{K}} + \frac{1}{2}\sum_{\mathbf{Q},\mathbf{K}_1,\mathbf{K}_2} W(\mathbf{Q},\mathbf{K}_1,\mathbf{K}_2) b_{\mathbf{K}_1+\mathbf{Q}}^{\dagger} b_{\mathbf{K}_2-\mathbf{Q}}^{\dagger} b_{\mathbf{K}_2} b_{\mathbf{K}_1}. \tag{28}$$

We obtain here the expression for W in the following form:

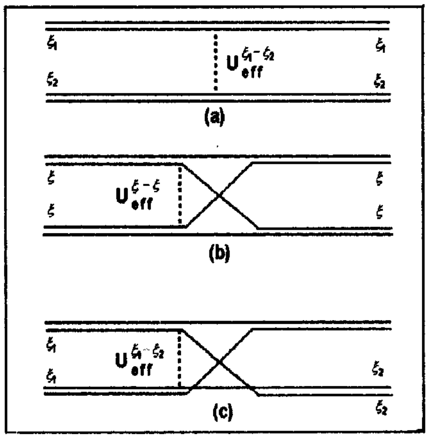

FIG. 2. Diagrams showing the nature of the biexciton-biexciton interaction. (a) the direct and (b) and (c) the exchange processes. Single solid lines denote excitons. Two close parallel exciton lines denote a biexciton. A crossing of lines denotes an exchange of excitons belonging to different biexcitons. Dashed lines represent effective interactions between excitons depending on the momentum transfer, the momenta difference of interacting excitons and on their symmetry ξ , ξ_1, or ξ_2.

$$
\begin{aligned}
W(\mathbf{Q},\mathbf{K}+\mathbf{Q}) = {} & \frac{1}{V^2}\sum_{\mathbf{p}_1,\mathbf{p}_2}\left[4U^d(\mathbf{Q})+\frac{7}{2}U^{\mathrm{ex}}\left(\mathbf{Q},\mathbf{p}_2-\mathbf{p}_1+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\right]\phi(\mathbf{p}_1)\phi\left(\mathbf{p}_1-\frac{\mathbf{Q}}{2}\right)\phi(\mathbf{p}_2)\phi\left(\mathbf{p}_2+\frac{\mathbf{Q}}{2}\right) \\
& + \frac{1}{V^2}\sum_{\mathbf{p}_1,\mathbf{p}_2}\left[U^d(\mathbf{p}_1)+\frac{1}{2}U^{\mathrm{ex}}(\mathbf{p}_1,\mathbf{Q})\right]\phi(\mathbf{p}_2)\phi\left(\mathbf{p}_2+\frac{\mathbf{Q}}{2}\right)\phi\left(\mathbf{p}_2-\mathbf{p}_1+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\phi\left(\mathbf{p}_2-\mathbf{p}_1+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right) \\
& + \frac{1}{V^2}\sum_{\mathbf{p},\mathbf{q}}\left[U^d(\mathbf{q})-U^{\mathrm{ex}}(\mathbf{q},2\mathbf{p}+\mathbf{Q}-\mathbf{q})\right]\phi\left(\mathbf{p}-\mathbf{q}+\frac{\mathbf{Q}}{2}\right)\phi(\mathbf{p})\phi\left(\mathbf{p}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\phi\left(\mathbf{p}+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right). \qquad (29)
\end{aligned}
$$

The first term on the rhs of (29) is the contribution of the biexciton interaction process of the direct character, when each of interacting biexcitons after the interaction consists of the same excitons that form the biexciton before the interaction [see Fig. 2(a)]. In contrast, the two last terms on the rhs of (29) represent exchange processes when interacting biexcitons exchange with each other one of the constituent exciton particles [see Figs. 2(b),(c)]. As expected, the effective biexciton-biexciton interaction is found to be the sum of effective interactions $U_{\mathrm{eff}}^{\xi_1-\xi_2}$ between component excitons averaged over biexciton envelope functions. Besides their dependence on the momentum transfer and the momenta difference, the functions $U_{\mathrm{eff}}^{\xi_1-\xi_2}$ depend on the symmetry ξ_1, ξ_2 of interacting excitons. Thus, in the process of Fig. 2(b) as well as of Figs. 2(a) and 2(c) for the case $\xi_1=\xi_2$ we have $U_{\mathrm{eff}}^{\xi-\xi}=U^d(\mathbf{q})+\frac{1}{2}U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}+\mathbf{q})$, while for $\xi_1\neq\xi_2$, $U_{\mathrm{eff}}^{\xi_1-\xi_2}=U^d(\mathbf{q})+U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}+\mathbf{q})$ in the process of Fig. 2(a) and $U_{\mathrm{eff}}^{\xi_1-\xi_2}=-\frac{1}{2}U^{\mathrm{ex}}(\mathbf{q},\mathbf{k}+\mathbf{q})$ in that of Fig. 2(c). It is interesting to note that the direct biexciton-biexciton interaction schematically shown in Fig. 2(a) in reality is the interaction between two excitons belonging to different interacting biexcitons and therefore includes both the direct and the exchange exciton interactions. Thus not only the exchange biexciton-biexciton interaction which certainly is nonlocal, but the direct one contains a nonlocal term as well. For this reason, it is convenient to present the biexciton-biexciton interaction as a sum of a local and a nonlocal part,

$$W(\mathbf{Q},\mathbf{K}+\mathbf{Q})=W^l(\mathbf{Q})+W^{\mathrm{nl}}(\mathbf{Q},\mathbf{K}+\mathbf{Q}), \qquad (30)$$

where the local one consists in the net direct interaction, when not only excitons in biexcitons but also the electrons and holes in those excitons interact directly. W^l depends only on the momentum transfer,

$$W^l(\mathbf{Q})=4U^d(\mathbf{Q})\left[\frac{1}{V}\sum_{\mathbf{p}}\phi(\mathbf{p})\phi\left(\mathbf{p}-\frac{\mathbf{Q}}{2}\right)\right]^2, \qquad (31)$$

whereas the whole dependence of the biexciton-biexciton interaction on the difference between momenta of interacting biexcitons enters the nonlocal part W^{nl},

$$
\begin{aligned}
W^{\mathrm{nl}}(\mathbf{Q},\mathbf{K}+\mathbf{Q}) = {} & \frac{1}{2V^2}\sum_{\mathbf{p}_1,\mathbf{p}_2}\left\{7U^{\mathrm{ex}}\left(\mathbf{Q},\mathbf{p}_1+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\phi(\mathbf{p}_2-\mathbf{p}_1)\phi\left(\mathbf{p}_2-\mathbf{p}_1-\frac{\mathbf{Q}}{2}\right)+\left[2U^d(\mathbf{p}_1)+U^{\mathrm{ex}}(\mathbf{p}_1,\mathbf{Q})\right]\right. \\
& \left.\times\phi\left(\mathbf{p}_2-\mathbf{p}_1+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\phi\left(\mathbf{p}_2-\mathbf{p}_1+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\right\}\phi\left(\mathbf{p}_2+\frac{\mathbf{Q}}{2}\right)\phi(\mathbf{p}_2)-E_{\mathrm{bx}}^b\frac{1}{V^2}\sum_{\mathbf{p}}\phi(\mathbf{p})\phi\left(\mathbf{p}+\frac{\mathbf{Q}}{2}\right) \\
& \times\phi\left(\mathbf{p}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right)\phi\left(\mathbf{p}+\frac{\mathbf{Q}}{2}+\frac{\mathbf{Q}+\mathbf{K}}{2}\right), \qquad (32)
\end{aligned}
$$

the last term of which is written applying Eq. (15) to the sum over $\mathbf{q}$ in the last term on the rhs of Eq. (29).

Since $U^d(\mathbf{0})=0$ the local part of the biexciton-biexciton interaction vanishes when no momentum is transferred between biexcitons. Estimations show that for $\mathbf{Q}\neq\mathbf{0}$, W^l is as well negligible for all values of σ and possible forms of ϕ. So the biexciton-biexciton interaction is mostly nonlocal and, in general, cannot be evaluated analytically. To see this more clearly, we go to the $\mathbf{r}$ space by the following transformations:

$$W^l(\mathbf{Q})=\frac{1}{V}\int d^3r\,\exp\left(i\frac{\mathbf{Q}}{2}\mathbf{r}\right)\mathcal{W}^l(\mathbf{r}) \qquad (33)$$

$$
\begin{aligned}
W^{\mathrm{nl}}(\mathbf{Q},\mathbf{Q}') = {} & \frac{1}{V}\int\int d^3r\,d^3r' \\
& \times\exp\left(i\frac{\mathbf{Q}}{2}\mathbf{r}+i\frac{\mathbf{Q}'}{2}\mathbf{r}'\right)\mathcal{W}^{\mathrm{nl}}(\mathbf{r},\mathbf{r}'), \qquad (34)
\end{aligned}
$$

where $\mathcal{W}^l(\mathbf{r})$ and $\mathcal{W}^{\mathrm{nl}}(\mathbf{r},\mathbf{r}')$ are obtained from (31), (32), taking into account (4) and (6),

$$\mathcal{W}^l(\mathbf{r})=4\int d^3r_1\,\mathcal{U}^d(\mathbf{r}_1)\int d^3r_2\,\Phi(\mathbf{r}_2)^2\Phi(2\mathbf{r}_1+\mathbf{r}_2-\mathbf{r})^2, \qquad (35)$$

$$\begin{aligned}\mathcal{W}^{\rm nl}(\mathbf{r},\mathbf{r}')=\int d^3r_1\Phi(\mathbf{r}_1)\Phi(\mathbf{r}_1-\mathbf{r}')\Bigg\{&\Phi(\mathbf{r}_1-\mathbf{r}-\mathbf{r}')\\ &\times\Phi(\mathbf{r}_1-\mathbf{r})[\mathcal{U}^d(\mathbf{r}')-E^b_{\rm bx}]\\ &+\frac{1}{2}\int d^3r_2\Phi(\mathbf{r}_1+2\mathbf{r}_2-\mathbf{r}-\mathbf{r}')\Phi(\mathbf{r}_1+2\mathbf{r}_2-\mathbf{r})\\ &\times[\mathcal{U}^{\rm ex}(\mathbf{r}',\mathbf{r}_2)+7\mathcal{U}^{\rm ex}(\mathbf{r}_2,\mathbf{r}')]\Bigg\}.\end{aligned}\tag{36}$$

It can be seen from (35), (36) that while $\mathcal{W}^l(\mathbf{r})$ can be evaluated for any possible form of Φ using bipolar variables, multicenter integrals in the expression for $\mathcal{W}^{\rm nl}(\mathbf{r},\mathbf{r}')$ cannot be calculated. The essential problem resides as well in the fact that full knowledge of $\mathcal{U}^{\rm ex}(\mathbf{r},\mathbf{r}')$ is lacking. For this reason, even in particular cases $\mathbf{K}=\mathbf{0}$ and $\mathbf{K}+\mathbf{Q}=\mathbf{0}$, Eq. (36) remains hard to deal with and has no practical value. An exception is the case $\mathbf{Q}=\mathbf{0}$ when in place of $\mathcal{U}^{\rm ex}$ appears $\mathcal{U}^{\rm ex}_1$ and $\mathcal{U}^{\rm ex}_2$ and multicenter integrals on the rhs of (36) turn to products of two- and single-center ones. From now in this section we focus our attention on this case to extract from it general features in the behavior of the biexciton-biexciton interaction.

Putting $\mathbf{Q}=\mathbf{0}$ in Eqs. (30), (31), (34) and using (36), we find the no-momentum-transfer biexciton-biexciton interaction in the form

$$W(\mathbf{0},\mathbf{K})=\frac{1}{V}\int d^3r\exp\left(i\frac{\mathbf{K}}{2}\mathbf{r}\right)\mathcal{W}_0(r),\tag{37}$$

with $\mathcal{W}_0(r)$ being a product of the sum of no-momentum-transfer effective interactions between excitons which form interacting biexcitons and squared overlap integral of envelope functions of these biexcitons,

$$\begin{aligned}\mathcal{W}_0(r)&=\int d^3r'\mathcal{W}^{\rm nl}(\mathbf{r},\mathbf{r}')\\ &=\left[\mathcal{U}^d(r)+\frac{1}{2}\mathcal{U}^{\rm ex}_1(r)+\frac{7}{2}\mathcal{U}^{\rm ex}_2(r)-E^b_{\rm bx}\right]\\ &\quad\times\left[\int d^3r'\Phi(\mathbf{r}')\Phi(\mathbf{r}'-\mathbf{r})\right]^2.\end{aligned}\tag{38}$$

The overlap integral takes the unit value at $\mathbf{r}=\mathbf{0}$ due to the normalization of Φ and decreases with the increase of the distance r at sufficiently large r as Φ does. For Φ of the hydrogenlike form (17) the integral is equal to $S(r/a_{\rm bx})$ [see Eq. (11)]. Thus the binding of excitons into biexcitons somewhat decreases the magnitude of the effective interaction between them and the influence depends on the binding degree, namely, on the relative size of the biexciton and the exciton.

The sum of effective interactions between constituent excitons consists of a linear combination of exciton interaction functions and a small constant inverse in sign to the biexciton binding energy. With the smooth weighting factor $S(r/a_{\rm bx})^2$, the first

$$\left[\mathcal{U}^d(r)+\frac{1}{2}\mathcal{U}^{\rm ex}_1(r)+\frac{7}{2}\mathcal{U}^{\rm ex}_2(r)\right]S\left(\frac{r}{a_{\rm bx}}\right)^2\tag{39}$$

describes the main strong repulsive part of the biexciton-biexciton interaction. Due to the exchange exciton-exciton interaction, this part varies drastically near $r=0$, then takes very large positive values giving the biexciton an incompressible ‘‘core.’’ From distances of few a_x the behavior of (39) is characterized by factors $\exp(-2r/\alpha a_{\rm x})S(r/a_{\rm bx})^2$ and $\exp(-2r/\beta a_{\rm x})S(r/a_{\rm bx})^2$ which drop with the increase of r much more rapidly than $S(r/a_{\rm bx})^2$ does. Then, as the distance increases, there comes a turning point r_0 where the biexciton-biexciton interaction reverses its sign, after which the weak attractive part described by the term

$$-E^b_{\rm bx}S\left(\frac{r}{a_{\rm bx}}\right)^2\tag{40}$$

becomes dominant. The position of r_0 is determined by the e-h mass and biexciton-exciton binding energy ratios σ and E. In the theory of intermolecular interaction, r_0 is called the effective diameter of the molecule: when the distance between molecules becomes greater than this diameter, they begin to attract each other. We see in the case of biexciton-biexciton interaction that the attraction is due to exchange interaction between excitons of the same symmetry resulting in the biexciton interaction process of Fig. 2(c). It has nothing in common with the van der Waals attraction between neutral complex particles which exists at distances a few times larger than the biexciton diameter. That attraction is important only at low densities and is not under consideration in this paper.

While the range of the repulsive core of the biexciton-biexciton interaction scales in the exciton radius, that of the attractive part is defined by the radius of the biexciton. The attractive part is therefore relatively extended. We suppose, however, that it does not lead to pair bound states and the biexciton remains the fundamental unit in the considered semiconductor. Then, in the considered range of excitation densities ($na^3_{\rm bx}\ll1$), the biexciton system can be treated as a weakly nonideal Bose gas,[51] with most particles having small momentum k, $ka_{\rm bx}\ll1$ at low temperatures. In this connection, it is worthwhile to calculate the small-momentum value of the biexciton-biexciton interaction by which the interaction is often replaced in practical estimations.[6,32] From (37), (38) we have

$$\begin{aligned}W(\mathbf{0},\mathbf{0})=&\int d^3r\left[\mathcal{U}^d(r)+\frac{1}{2}\mathcal{U}^{\rm ex}_1(r)+\frac{7}{2}\mathcal{U}^{\rm ex}_2(r)\right]S\left(\frac{r}{a_{\rm bx}}\right)^2\\ &-E^b_{\rm bx}\int d^3r\,S\left(\frac{r}{a_{\rm bx}}\right)^2.\end{aligned}\tag{41}$$

A reasonable estimation may be made for the first integral of $W(\mathbf{0},\mathbf{0})$: Since $S(r/a_{\rm bx})^2$ is a very smooth function in comparison with exciton interaction functions, it can be approximated by one over the interval where the last noticeably differ from zero. Taking into account Eqs. (4), (8), (9) and the fact $U^d(\mathbf{0})=0$, we find this integral equal to

$$4U^{\mathrm{ex}}(\mathbf{0},\mathbf{0})=4\frac{26\pi}{3}\mathcal{R}a_{\mathrm{x}}^{3}.$$

The last integral of $W(\mathbf{0},\mathbf{0})$ in (41) can be easily calculated. Its value is proportional to the effective volume of the biexciton. As a result,

$$W(\mathbf{0},\mathbf{0})\simeq\frac{104\pi}{3}\mathcal{R}a_{\mathrm{x}}^{3}-\frac{33\pi}{2}E_{\mathrm{bx}}^{b}a_{\mathrm{bx}}^{3}. \tag{42}$$

As in practice $E_{\mathrm{bx}}^{b}a_{\mathrm{bx}}^{3}$ is usually of the order of $\mathcal{R}a_{\mathrm{x}}^{3}$, we have $W(\mathbf{0},\mathbf{0})\approx 2U^{\mathrm{ex}}(\mathbf{0},\mathbf{0})$.

The first term of $W(\mathbf{0},\mathbf{0})$ in (41) can be obtained from the result (4.11) of Ref. 34 by performing the Fourier transformation and taking into account the energy and length units used there. The last one has not been considered in Ref. 34. As a matter of fact, in the sum of effective interactions between excitons [see Eq. (38)], the negative term at first sight may be neglected because E_{bx}^{b} is typically a tenth of $\mathcal{R}$, while the average value of the exchange interaction functions in the range of their maximum ($0<r<a_{\mathrm{x}}$) are of order of tens $\mathcal{R}$. However, this small negative term gives rise to a fairly extended attractive part of the biexciton-biexciton interaction which not only considerably decreases the small-momentum value but also essentially changes the picture of the interaction.

For a thorough evaluation of the first integral of $W(\mathbf{0},\mathbf{0})$ in (41) we write it as follows:

$$\int d^{3}r\left[\mathcal{U}^{d}(r)+\frac{1}{2}\mathcal{U}_{1}^{\mathrm{ex}}(r)+\frac{7}{2}\mathcal{U}_{2}^{\mathrm{ex}}(r)\right]S\left(\frac{r}{a_{\mathrm{bx}}}\right)^{2} = 32\pi I_{\sigma}(a)\mathcal{R}a_{\mathrm{x}}^{3}. \tag{43}$$

From the above estimation we know that for $a_{\mathrm{bx}}\gg a_{\mathrm{x}}$, the parametric dependence of I on σ vanishes, $I_{\sigma}(\infty)=\frac{13}{12}$. By the use of Eqs. (5) and (10) we get $I_{\sigma}(a)$ in the following form:

$$\begin{aligned}I_{\sigma}(a)=&\int dt\frac{t^{2}}{4}\exp(-2t)\sum_{\tau=\alpha,\beta}\tau^{2}S\left(\frac{\tau}{a}t\right)^{2}\left\{\left[\frac{6\tau^{4}}{(2\tau-1)^{2}}\right.\right.\\&\left.\left.-\frac{4\tau^{6}}{(2\tau-1)^{3}}-1\right]\frac{1}{t}+\frac{2\tau^{4}}{(2\tau-1)^{2}}-\frac{11}{8}-\frac{3}{4}t-\frac{1}{6}t^{2}\right\}\\&+\int dt\frac{t}{8}\left\{\left(\frac{29}{8}+\frac{481}{20}t+\frac{223}{15}t^{2}+\frac{47}{15}t^{3}\right)t\right.\\&\times\exp(-2t)-\frac{42}{5}\left[\left(\frac{5}{42}+\gamma+\ln t\right)S(t)^{2}\right.\\&\left.\left.+S(-t)^{2}\mathrm{Ei}(-4t)-2S(t)S(-t)\mathrm{Ei}(-2t)\right]\right\}\\&\times\sum_{\tau=\alpha,\beta}S\left(\frac{\tau}{a}t\right)^{2}.\end{aligned} \tag{44}$$

From explicit expressions for S and for the exponent integral we can see that $I_{\sigma}(a)$ can be calculated analytically. However, as the result is found to be very awkward and the dependence of I on a and σ is too intricate to follow, we shall not present it here. We wish to note only that $I_{\sigma}(a)$ slightly decreases as a decreases. For practical estimation, it is much more simple to compute (44) for definite σ and a, which we shall do for the CuCl crystal afterwards.

With the aid of (43), we have

$$W(\mathbf{0},\mathbf{0})=\left[32\pi I_{\sigma}(a)-\frac{33\pi}{2}Ea^{3}\right]\mathcal{R}a_{\mathrm{x}}^{3}. \tag{45}$$

It is certain that a proper calculation of $I_{\sigma}(a)$ which gives for the integral on the lhs of (43) a result a little smaller than $4U^{\mathrm{ex}}(\mathbf{0},\mathbf{0})$ does not change the relation between the contributions of two parts of the biexciton-biexciton interaction to $W(\mathbf{0},\mathbf{0})$. The last is a positive quantity: the attraction is predominated over by the repulsion which is extremely strong at small distances. The repulsion that comes from a quantum-statistical principle working between identical component particles of interacting biexcitons (both between excitons and between electrons and holes of the excitons) makes the biexcitons impenetrable to each other. Thus each biexciton like any molecule may be imagined as a hard sphere of diameter r_{0} surrounded by an attractive potential of the range r_{w} (of order of few a_{bx}) and maximum depth ε_{w} (of order of tenth of E_{bx}^{b}). It is well known from quantum mechanics that the relationship between r_{w}, ε_{w} and the particle mass determines the possibility of a bound state formation by the attractive potential.[52] We shall return to this question while considering the example of CuCl crystal later in this section. Here no bound state is assumed. Then the biexciton-biexciton ''s-wave'' scattering length a_{s} is positive,[53] and as it was mentioned earlier, the biexciton system is a weakly nonideal Bose gas in the excitation range $na_{\mathrm{bx}}^{3}\ll 1$. At low temperatures the last condition means that $ka_{\mathrm{bx}}\ll 1$ too. Hence the no-momentum-transfer biexciton-biexciton interaction (38), although not exact, possesses not only main qualitative but as well quantitative features of the interaction among biexcitons. It can therefore be used for the computation of the scattering length a_{s} and in this way an important problem of the theory of biexciton Bose condensates becomes solvable: According to the standard procedure of describing a weakly nonideal Bose gas with positive scattering length — the Bogoliubov approximation — properties of the ground state of such a gas (the condensate) are described in terms of first two powers of the small parameter $(na_{s}^{3})^{1/2}$.[46]

To interpret the results discussed above , we consider the CuCl crystal, a prototypical material for the study of biexcitons on which most extensive experimental work has been carried out. The following parameters of CuCl are used: $\sigma=0.25$, $E_{\mathrm{bx}}^{b}=30$ meV, $\mathcal{R}=200$ meV, $a_{\mathrm{x}}=7$ Å. According to Eq. (18), the values of σ and E correspond to $a\simeq 1.7$ if we neglect the terms proportional to the second and higher powers of α in the expansion of $\mathcal{U}^{\mathrm{ex}}$ in terms of $\mathcal{U}_{1}^{\mathrm{ex}}$ and $\mathcal{U}_{2}^{\mathrm{ex}}$. The computation of the integral (44) gives $I_{0.25}(1.7)=0.9234$ and from (45) the corresponding value of $W(\mathbf{0},\mathbf{0})$ is $17.4\pi\mathcal{R}a_{\mathrm{x}}^{3}$. So, for CuCl the assumption $W(\mathbf{0},\mathbf{0})\simeq 2U^{\mathrm{ex}}(\mathbf{0},\mathbf{0})$ used in Ref. 6 is in a very close agreement with our calculation.

To characterize a density of the biexciton gas in CuCl, following the authors of Ref. 34, we use the radius r of the sphere of volume n^{-1} as the mean distance between quasiparticles. The dependence of the biexciton-biexciton interac-

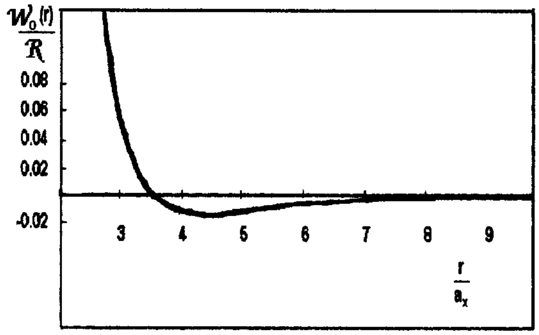

FIG. 3. No-momentum-transfer effective biexciton-biexciton interaction in CuCl as a function of the distance. $\sigma=0.25$, $E=0.15$, and $a=1.7$.

tion $\mathcal{W}_0(r)$ on this distance is plotted in Fig. 3. We see that the biexciton system in CuCl may be considered as a nearly ideal Bose gas at densities $n \lesssim 10^{18}$ cm^{-3}, when attraction among particles almost vanishes: $|\mathcal{W}_0(r)| < 0.01 E_{\rm bx}^b$ for $r > 8a_{\rm x}$. As the density increases, the attraction between biexcitons rises and its magnitude mounts up to about eighth of $E_{\rm bx}^b$ in the interval $r \simeq 4a_{\rm x}$–$4.5a_{\rm x}$. With further increase of the density, the biexciton-biexciton interaction reaches the turning point at $n \simeq 1.5\times 10^{19}$ cm^{-3} ($r_0 \simeq 3.6a_{\rm x}$), beyond which it goes over into the region of a rapidly increasing repulsion. For $n \gtrsim 2\times 10^{19}$ cm^{-3} the magnitude of the repulsion becomes comparable with $E_{\rm bx}^b$ and causes the biexciton to break up. Strictly speaking, the excitation densities corresponding to $r < r_0$ is out of the range of validity of the model used in this paper. At such densities the biexciton is no longer a stable quasiparticle. In this way, in the range of their stability ($n \lesssim 1.5\times 10^{19}$ cm^{-3} for CuCl), biexcitons attract each other at any distance. The question of whether the attraction leads to the formation of biexciton pair bound state becomes essential. To answer to it, we approximate the biexciton-biexciton interaction in CuCl by the following Morse's formula:

$$\mathcal{W}_0(r) \simeq \varepsilon_w \{\exp[-2\delta(r-r_1)] - 2\exp[-\delta(r-r_1)]\}, \tag{46}$$

with $\varepsilon_w = E_{\rm bx}^b/8$, $r_1 = 4.36a_{\rm x}$, and $\delta = \ln 2/0.76a_{\rm x}$. Equation (46) does not represent the behavior of $\mathcal{W}_0(r)$ at $r<r_0$ exactly, but describes very well main features of the shallow potential well of Fig. 3. According to a result of Ref. 52 (see problem 4), if

$$\frac{\sqrt{2m_{\rm x}\varepsilon_w}}{\delta} < \frac{1}{2} \tag{47}$$

bound state cannot be formed in the system with pair interaction (46). Using the relation between $m_{\rm x}$ and m and between m and $\mathcal{R}$ and $a_{\rm x}$, we gain (47) in the form

$$\frac{0.38(1+\sigma)}{\ln 2}\sqrt{\frac{E}{2\sigma}} < \frac{1}{2}$$

which is satisfied for CuCl with $\sigma=0.25$ and $E=0.15$. Definitely, the biexciton system in CuCl is a weakly nonideal Bose gas with positive scattering length. By the variational method,[53] a_s is estimated to be approximately $3a_{\rm x} \approx 2$ nm, smaller than the experimental value $a_s \approx 3$ nm provided by Hasuo *et al.* recently.[45]

At last, it is interesting to note that the expression for the biexciton-biexciton interaction in the form (38) places an upper limit on possible values of the biexciton radius. Indeed, for a given semiconductor with definite σ and E, the range r_0 of the repulsive core of the biexciton-biexciton interaction is determinate and $r_0 \gtrsim 2a_{\rm bx}$. For CuCl, we see from foregoing analysis that $a_{\rm bx} \lesssim 1.8a_{\rm x}$. This result may partially remove the existing discrepancy in the estimation of the value of the biexciton radius in the model crystal.

IV. DISCUSSION AND CONCLUSION

For simplicity, the effective biexciton-biexciton interaction in the present paper is derived for a simple model of isotropic single bands. In principle, the derivation can be performed for any direct cubic two-band semiconductor. The effective bosonic Hamiltonian for an exciton system can be written by the use of general formula from Ref. 33, and the biexciton wave function constructed by group-theoretical methods. Qualitative features of the biexciton-biexciton interaction in the general case of a direct cubic semiconductor may be expected to be the same as obtained for the simple model. Being an intricate function of the momentum transfer and of the difference between momenta of interacting biexcitons, the interaction allows an analytical study when the first vanishes. In this case the biexciton-biexciton interaction in the **r**-space depends only on the interbiexciton distance and consists of a short-range strong repulsive core and a short-range weak attractive part (which at far distances may join up with the very weak long-range van der Waals tail). As stable quasiparticles, biexcitons attract each other at any distance. If the attraction of exchange nature does not lead to pair bound state formation, biexciton system can be considered as a weakly nonideal Bose gas with positive "s-wave" scattering length. The last can be estimated using the expression obtained for no-momentum-transfer biexciton-biexciton interaction.

The behavior of the biexciton-biexciton interaction versus distance is characteristic of the intermolecular interaction. In this respect, the biexciton closely resembles the hydrogen molecule. The result obtained in the present paper in the limit $\sigma=0$ describes the interaction between H_2 molecules within the Heitler-London approximation. For $\sigma=0$, the computation of the interaction can be carried out for general case of $\mathbf{Q}\neq\mathbf{0}$ since the nonlocality in $\mathcal{U}^{\rm ex}$ vanishes and the exchange interaction function in the **r** space can be analytically evaluated. The attraction between H_2 is certainly more strong than that among biexcitons in CuCl since the parameter E of H_2 is more than twice greater, $E \simeq 0.35$ for $\sigma=0$. In a molecular system, attractive pair interaction leads to an abrupt transformation from gaseous to a liquid state at low temperatures. It was shown by Keldysh that because of the smallness of the mass of excitonic particles, such a gas-liquid transition is not expected neither in a biexciton, nor in an exciton gas.[54] An N-body bound state in semiconductors is formed at very high densities, where not only biexcitons but excitons as well are destroyed and the system of charge carriers exists in the form of a dense nonequilibrium e-h liquid. Such densities which are above the Mott threshold are not considered in this paper. Hence, despite the resemblance to H_2, the biexciton system remains a gas down to absolute zero. From the interparticle interaction point of view, the formation of a stable Bose condensate with a sufficiently high condensation fraction (owing to relative smallness of a_s) is possible at low temperatures. In this way the biexciton system in semiconductors is the prime candidate for the observation of such an important phase transition as BEC.

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