

# Interference exciton states in systems of quantum wells

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The exciton spectra of asymmetric semiconductor type-I heterostructures may undergo a radical restructuring as the geometric parameters of the quantum wells and barriers are varied. If the nonparabolicity of the dispersion for one-particle excitations is pronounced, to the point that the effective mass changes sign in certain hole subbands, some of the excitons may have anomalously large binding energies over extremely broad ranges of these parameters. On the other hand, there may be cases in which bound states do not arise at all. © 1995 *American Institute of Physics.*

Elementary excitons in tailored semiconductor heterostructures often exhibit properties of reduced-dimensionality systems quite clearly. In quantum-well heterostructures, for example, exciton resonances are reliably detected in optical spectra all the way up to room temperature.<sup>1</sup> The tendency for the binding energy of a Wannier exciton to increase with decreasing dimensionality of the system is well known. We also know that certain quantum-well heterostructures have some quantum-well subbands of the valence band with an anomalous dispersion at low quasimomenta: a change in the sign of the effective mass of a hole as the result of a quantum interference of states in the subbands of heavy and light holes.<sup>2</sup> It becomes particularly important to incorporate the nonparabolicity of these subbands in an analysis of exciton states.

We have used the method of envelope functions in the effective-mass approximation<sup>3</sup> to study excitons in type-I heterostructures with several quantum wells which are narrow and otherwise of arbitrary width, separated by tunneling-transparent barriers. In asymmetric heterostructures of this sort, it is possible to arrange various versions of the spatial localization of carriers belonging to different subbands, by varying the dimensions of the quantum wells and the barriers. In addition, in certain cases it is possible to cause profound changes in the carrier dispersion in the subbands (particularly the valence subbands).

Figure 1 shows hole dispersion relations calculated for two  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ -GaAs heterostructures with three quantum wells. The concentration  $x$  and the parameters of the wells and the barriers have been chosen in order to demonstrate the qualitative differences in the dispersion in the first light-hole subband (LH1) and the second heavy-hole subband (HH2). To determine the electron and hole envelopes, we use the standard model of a heterostructure with a piecewise-constant potential and boundary conditions which conserve the probability density and the probability-flux density calculated from the envelope functions.<sup>3</sup> As usual, by calculating the dispersion in the hole subbands in the Luttinger formalism we can determine the effective masses of the transverse motion

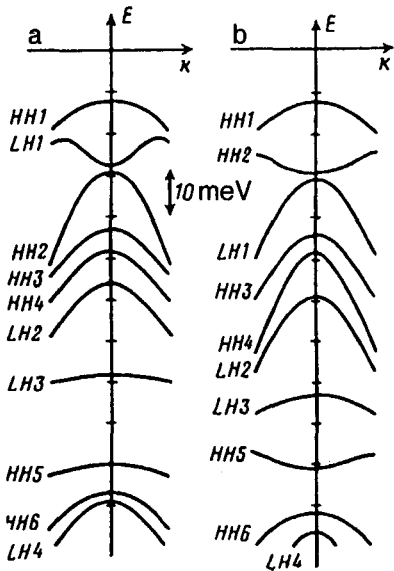


FIG. 1. Dispersion of the hole subbands for an  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ -GaAs heterostructure with three quantum wells with widths of  $13a$ ,  $7a$ , and  $5a$ , separated by barriers with widths of  $5a$  and  $7a$  (a 13-5-7-7-5 structure). a—The concentration is  $x=0.22$ ; b— $x=0.25$ .

(transverse with respect to the axis of the heterostructure) in the subbands, while terms of fourth order in the transverse quasimomentum, i.e., the nonparabolicity of the subbands, are better dealt with in a phenomenological way. In calculating the effective masses we consider both the penetration of the envelopes into the barrier region (and, correspondingly, the difference between the values of the effective masses in the barriers and the quantum wells) and the mixing of states of heavy and light holes. This mixing is extremely important for asymmetric heterostructures, since in this case the parity selection rules of symmetric heterostructures (in particular, of heterostructures with a single quantum well) are violated, and there are also changes in the conditions of the quantum interference of electron and hole states. These effects are reflected in an obvious way in the internal structure of the excitons which arise in such heterostructures.

The envelope function of the  $n$ th quantum-well subband of the  $s$ th bulk band can thus be written

$$\Psi_{s\mathbf{nk}}(\mathbf{r}) = S^{-1/2} \exp(i\mathbf{k}\boldsymbol{\rho}) f_{s\mathbf{nk}}(z), \quad (1)$$

where  $\mathbf{k}$  is the transverse quasimomentum,  $S$  is the normalization area of the heterostructure perpendicular to the  $z$  axis,  $\boldsymbol{\rho}$  is the transverse radius vector, and  $f_{s\mathbf{nk}}(z)$  is the envelope function of the longitudinal motion (along the  $z$  axis). The corresponding dispersion relation is, within terms on the order of  $k^4$ ,

$$E_{s\mathbf{n}}(k) = E_{s\mathbf{n}}^{(0)} + \hbar^2 k^2 / 2m_{s\mathbf{n}} + (\hbar^2 / 2m_0) b_{s\mathbf{n}}^2 k^4. \quad (2)$$

In the isotropic approximation which we are using here (there is isotropy in the plane of the heterostructure), the first term gives the position of the edge of the subband. In the second term, the effective mass  $m_{s\mathbf{n}}$  may be either positive or negative (in different cases). The phenomenological parameter  $b_{s\mathbf{n}}$ , which incorporates the nonparabolicity of

the subband, can be estimated from the position ( $k_0$ ) of the minimum of the hole subband with a negative effective mass:  $b_{sn} = (m_0/2|m_{sn}|)^{1/2}k_0^{-1}$ , where  $m_0$  is the mass of a free electron. We find  $10^{-7}$ – $10^{-6}$  cm.

In calculations of the exciton spectra of asymmetric heterostructures with narrow quantum wells, the standard methods (based on a variational principle<sup>4</sup> or the Luttinger representation for the Hamiltonian of the exciton, with a subsequent expansion of the exciton envelopes in terms of specially selected basis functions<sup>5,6</sup>) are not adequate. It is better to use a 2D  $\mathbf{k}$  representation (in the plane of the heterostructure), dealing with the motion along the heterostructure axis with the help of a set of quantum numbers  $sn$ . For each  $s$ th band, this set takes on a finite number of values corresponding to states of the electron or hole which are localized along the  $z$  direction, along with a quasicontinuum of values corresponding to excitations with higher energies, for which  $n$  actually transforms into the  $z$  component of the quasimomentum. If the quantum wells and the barriers are narrow, it is evidently sufficient to consider only a small number values of  $sn$ , corresponding to the lower quantum-well subbands. By going over to a 2D Wannier exciton representation, one can thus write the equivalent Hamiltonian of the exciton as a linear matrix differential operator which incorporates the Coulomb mixing of excitons corresponding to various pairs of 2D hole and electron subbands (we will denote the composite label  $\{sn; s'n'\}$  of these subbands by the single symbol  $\lambda$ ):

$$\tilde{\mathcal{H}}_{\lambda\lambda_1} = [\Delta_\lambda - (\hbar^2/2\mu_\lambda)\nabla^2 + \hat{V}_\lambda(\rho)]_{\lambda\lambda_1} + U_{\lambda\lambda_1}(\rho). \quad (3)$$

Here  $\Delta_\lambda = E_{sn}^{(0)} - E_{s'n'}^{(0)}$ ,  $\rho$  is the radius vector of the relative transverse motion of the electron-hole pair, the operator  $\hat{V}_\lambda(\rho)$  includes terms proportional to  $k^4$  in the dispersion for the electron and the hole, and the matrix elements of the Coulomb interaction of an electron and a hole are given by

$$U_{\lambda\lambda_1}(\rho) = -e^2 \int dz dz' f_{sn}^*(z) f_{s_1 n_1}(z) G(\rho; z, z') f_{s' n'}(z') f_{s'_1 n'_1}(z'), \quad (4)$$

with the electrostatic Green's function  $G$ , which incorporates the difference between the dielectric constants of the materials of the quantum wells ( $\epsilon_2$ ) and the barriers ( $\epsilon_1$ ). Since the condition

$$\nu = (\epsilon_2 - \epsilon_1)/(\epsilon_2 + \epsilon_1) \ll 1$$

holds in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ – $\text{GaAs}$  and other heterostructures of this type, we can restrict Eq. (4) to terms of no higher than first order in  $\nu$ . The label  $\mathbf{k}$  on the longitudinal envelopes in (4) is set equal to zero and is omitted, since at small values  $k \ll a_{\text{exc}}^{-1}$  (where  $a_{\text{exc}}$  is a length scale of the exciton envelope in the  $xy$  plane) these envelopes are extremely weak functions of  $k$  for both electrons and holes. Furthermore, for the hole subbands the mixing of states of heavy and light holes is taken into account in a calculation of the corresponding effective masses.

If the effective hole mass satisfies  $m_{s'n'} < 0$ , the reduced effective mass of an exciton,

$$\mu_\lambda = m_{sn} m_{s'n'} (m_{sn} + m_{s'n'})^{-1},$$

may be either negative (in which case a bound state of an electron and hole does not arise) or positive (if  $m_{s'n} + m_{s'n'}$  is also less than zero). In the latter case it is particularly important to consider the nonparabolicity of the subbands, i.e., the term with  $\hat{V}_\lambda(\rho)$  in (3).

It can be seen directly from (4) that the diagonal matrix elements of the Coulomb interaction are generally much larger than the off-diagonal ones, since in the former case the integrand is positive, while in the case  $\lambda_1 \neq \lambda$  (first) the integrand is of variable sign, and (second) the electron and hole envelopes are localized for the most part in different wells, if the heterostructure is asymmetric. We can accordingly treat the off-diagonal elements as perturbations and use the standard classification of excitons with the help of the label  $\lambda$  introduced above: HH1-C1, LH1-C1, etc. Here HH $n$  and LH $n$  mean the  $n$ th subband of the heavy- and light-hole bands, and C $n$  means the  $n$ th subband of the conduction band. In a first approximation, we can thus ignore effects of the Coulomb mixing of excitons. Hamiltonian (3) then leads to independent equations for the exciton envelope functions for each  $\lambda$ . We can use the method of Ref. 7 to solve these equations. That method involves determining an effective dielectric constant of the heterostructure for each exciton state from the condition that the correction to the exciton energy vanish in first-order perturbation theory in the deviation of  $U_{\lambda\lambda}(\rho)$  from some 2D Coulomb potential which contains a factor  $\chi$  to be determined. Specifically, we write the Hamiltonian  $\hat{\mathcal{H}}_{\lambda\lambda} \equiv \hat{\mathcal{H}}_\lambda$  in the form

$$\hat{\mathcal{H}}_\lambda = \hat{\mathcal{H}}_\lambda^{(0)} + \hat{W}_\lambda(\chi), \quad (5)$$

where

$$\hat{\mathcal{H}}_\lambda^{(0)} = \Delta_\lambda - (\hbar^2/2\mu_\lambda)\nabla^2 - (\chi e^2)/\rho, \quad (6)$$

and

$$\hat{W}_\lambda(\chi) = (\chi e^2)/\rho + U_\lambda(\rho) + \hat{V}_\lambda(\rho). \quad (7)$$

The envelope wave function  $|\lambda\rangle$  of the ground state of a  $\lambda$  exciton is a solution of the Schrödinger equation with Hamiltonian (6):

$$|\lambda\rangle = (2\pi)^{-1/2} (4\chi/a_\lambda) \exp(-2\chi\rho/a_\lambda), \quad (8)$$

where  $a_\lambda = \hbar^2/\mu_\lambda e^2$ . In the zeroth approximation, the binding energy of the  $\lambda$  exciton is

$$E_\lambda^{(0)} = 4\chi^2 R y^{(\lambda)}, \quad (9)$$

where  $R y^{(\lambda)} = \mu_\lambda e^4/2\hbar^2$ . The condition which determines the parameter  $\chi$ , i.e.,

$$\langle \lambda | \hat{W}_\lambda(\chi) | \lambda \rangle = 0, \quad (10)$$

can easily be transformed to

$$4\chi^2 e^2/a_\lambda + (\hbar^2 b_\lambda^2/8m_0)(4\chi/a_\lambda)^4 [A - \ln\chi] + \langle \lambda | U_\lambda | \lambda \rangle = 0. \quad (11)$$

Here  $A = \ln(\pi a_\lambda/2a) - 3/4$ , and  $a$  is the lattice constant. The second term in (11) is related to the particular form of the operator  $\hat{V}_\lambda$ . The divergence which arises in the calculation of  $\langle \lambda | \hat{V}_\lambda | \lambda \rangle$  can be eliminated in the usual way by cutting off the integral in quasimomentum space at an upper limit  $k_m = \pi/a$ . Since the electrostatic Green's function in (4)

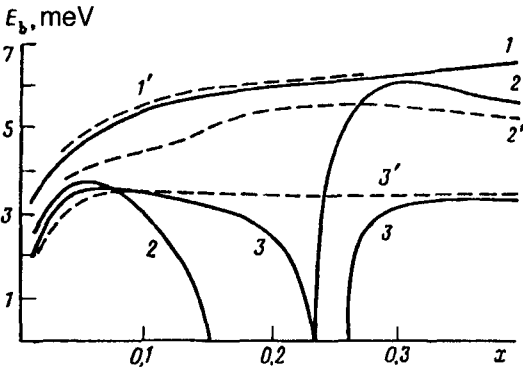


FIG. 2. 1, 2, 3—Binding energies of HH1-C1, LH1-C1, and HH2-C1 excitons versus the concentration  $x$  in an  $\text{Al}_x\text{Ga}_{1-x}\text{As}-\text{GaAs}$  heterostructure (13-5-7-7-5); 1', 2', 3'—the same, for a (14-5-7-7-5) structure. These calculations were carried out with  $b_\lambda = 10^{-6}$  cm.

actually has the same dependence on the transverse radius vector in each order in the parameter  $\nu$ , we write the last term in (11) in terms of the universal function

$$F(\zeta) = \zeta \{ (\pi/2) [H_1(\zeta) - Y_1(\zeta)] - 1 \}, \quad (12)$$

where  $Y_1(\zeta)$  and  $H_1(\zeta)$  are Bessel and Struve functions, respectively. Their argument, in zeroth order in  $\nu$ , for example, is  $\zeta = (4\chi/a_\lambda)(z - z')$ . The further integration over  $z$  and  $z'$  in (4) is carried out by numerical methods. By using the function in (12), which allows an elementary tabulation at any required accuracy, one can effectively solve Eq. (11) numerically for any heterostructure with a finite number of quantum wells. Figure 2 shows the results of a numerical solution of Eq. (11) for a heterostructure with three wells. We see that an extremely small structural difference among these wells (there is one additional monolayer in one of the wells) may (in certain intervals of the concentration  $x$ ) lead to a radical restructuring of the exciton spectra. As comparison with Fig. 1 shows, this restructuring is due primarily to the nature of the hole excitations, in particular, the formation of subbands with negative effective masses. By varying the concentration (and thus the depth of the quantum wells for electrons and holes), one can make the spacing between hole subbands (e.g., LH1 and HH2) extremely small for certain dimensions of the quantum wells and the barriers. In this case the interference of the states of heavy and light holes,<sup>2</sup> which vanishes in the limit  $k \rightarrow 0$  and which does not alter the values of  $E_{sn}^{(0)}$  in (2), leads to an effective repulsion of the quantum-well levels in the case  $k \neq 0$ . If the sign of the effective mass (LH1 in Fig. 1a or HH2 in Fig. 1b) changes in one of the subbands (with the smaller excitation energy), then a loop of extrema arises<sup>8,9</sup> according to (2). This loop corresponds to an energy smaller than  $E_{sn}^{(0)}$ . Near this energy the density of states has the shape typical of 1D systems. By further varying the concentration for a heterostructure with more than one well, one can change (for a certain  $x = x_0$ ) the order of certain hole subbands (e.g., HH2 and LH1; Fig. 1b), with a corresponding change in the density of states from 2D to 1D near the energy corresponding to the loop of extrema which has just arisen. There evidently exist concentration values  $x_1$  and  $x_2$  such that in the interval  $x_1 < x_0 < x_2$  and for all  $x$  in the interval  $x_1 < x < x_0$  the reduced mass of an LH1-C1 exciton is negative, while for all  $x$  in the interval  $x_0 < x < x_2$  the reduced mass of an HH2 exciton is negative. The concentration intervals in Fig. 2, in which there are no LH1-C1 or HH2-C1 excitons, evidently also correspond to the case of a negative reduced mass of an exciton. The sharp increase<sup>10</sup> in the binding

energies of these excitons (particularly at small values of  $b_\lambda$ ) corresponds to large values of the reduced mass, with  $m_h < 0$  but  $|m_h| \geq m_e$ . The reason why the binding energy of an HH2-C1 exciton is small in comparison with that of an LH1-C1 exciton is that in the former case the hole and the electron are localized primarily in different wells, while the hole in the LH1 subband and the electron in the C1 subband are localized in a common (wider) quantum well.

The value of  $\chi^{-1}$  found through a solution of Eq. (11) can be interpreted as the effective dielectric constant of a heterostructure which characterizes the interaction of an electron and a hole bound in a  $\lambda$  exciton. As the reduced mass of the exciton increases, the parameter  $\chi$  decreases; the binding energy in (9) is thus a nonmonotonic function of the reduced mass, reaching a maximum at a certain value of this mass. Accordingly, incorporating  $\hat{V}_\lambda$  in the Hamiltonian of the exciton leads to a natural limitation on the binding energy as the reduced mass is increased substantially, so one can use the Wannier scheme to describe the excitons.

In calculating the exciton energies we considered all the quantum-well subbands which exist at the given concentration  $x$ , but we ignored the continuum states. We should accordingly point out that in the limit  $x \rightarrow 0$ , i.e., as the depth of the quantum well decreases, the accuracy of the calculations based on perturbation theory worsens. For excitons formed by deep subbands of electrons and holes, the contribution of continuum states becomes unimportant at concentrations  $x \geq 0.05$ .

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