Metastable states of hole pairs in semiconductor quantum wells

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Specially made asymmetrical semiconductor heterostructures with two or more quantum wells may contain metastable bound states of hole pairs because holes of some dimensional subbands have negative effective masses. © *1996 American Institute of Physics.*
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A series of relatively narrow lines whose positions can be described by the formula for the hydrogen atom and which converge not on the usual short-wave side of the spectrum but on the long-wave side has been detected in absorption spectra of bismuth iodide at helium temperatures.[1–6] This reversed hydrogen-like series was accounted for in Refs. 1–6 in terms of bound states of pairs of equally charged particles—electrons (bielectron) or holes (bihole)—due to the specific shape of their energy spectra. A reversed hydrogen-like series may be also due to electronic transitions from the valence band or a double-charged impurity center to bound bielectron states formed from two branches of the conductance band in one of which the electron effective mass is negative.[1] A similar effect may take place in the valence band, and a bound state may be formed by two holes. In this case the set of discrete lines is higher than the fundamental absorption edge, hence the bound states are metastable. Conditions under which a reversed hydrogen-like series may be detected in a semiconductor absorption spectrum have been investigated both theoretically and experimentally,[7–10] and it was found that bound states of two quasiparticles with repulsive interaction between them is only possible in materials with rather exotic electronic spectra. The dispersion relation of a semiconductor can rarely be controlled by varying parameters, so the range of materials in which a reversed hydrogen-like series can be observed is very limited.

The progress made in semiconductor nanotechnology allows one to fabricate heterostructures containing quasi-two-dimensional quantum wells, in which the electron and hole spectra are extraordinarily diverse and whose parameters can be varied over wide ranges by either changing the geometry of the quantum wells or by applying an external field. Hole spectra in quantum wells are usually modified more easily because the one-dimensional ($1D$) potential generating the quantum wells lifts the valence band degeneracy at the center of the Brillouin zone and forms a set of two-dimensional ($2D$) subbands in the quantum wells. The hole states corresponding to nonzero $2D$ quasi-momenta are formed from states of both light and heavy holes, so their classification in terms of light and heavy holes makes sense only at zero $2D$ quasimomentum. As a result of this mixing of states of light and heavy holes,[11] the hole effective masses in some subbands may be negative.[12] This, in turn, allows two holes of different subbands to form a bound state, although the interaction between them is repulsive. Since this interaction is Coulomb, even though modified by the field of image charges due to interfaces,[13] it is natural that reversed hydrogen-like series are observed in optical spectra of some quantum wells. Note that the hole spectrum in a quantum well can be engineered by selecting parameters of the heterostructure, moreover, the spectrum can be modified by an external field, e.g., an electric field aligned with the growth axis.[14,15]

The paper describes a theoretical study of the position and shape of the first line in a reversed hydrogen-like series due to the bound state of two holes in an asymmetrical system of quantum wells with a highly nonparabolic spectrum of one hole subband.

1. Let us consider a heterostructure whose hole spectrum is shown in Fig. 1. The numbers 1, 2, and 3 in Fig. 1 denote dimensional subbands in the following order: 1) the lowest heavy-hole subband (HH1); 2) the second heavy-hole subband (HH2); 3) the lowest light-hole subband (LH1). The higher subbands are not shown in the diagram. In what follows, the subbands are labelled by the indices sn, where $s=$HH, LH, and n is the subband number in the respective s band. If the terms up to the fourth order in the $2D$ quasimomentum k are retained, the spectrum of an sn hole subband can be described by the equation

$$E_{sn}(k)=\Delta_{sn}+\frac{\hbar^2k^2}{2m_{sn}}+\frac{\hbar^2}{2m_0}b_{sn}^2k^4, \tag{1}$$

where Δ_{sn} is the energy in the sn subband at zero momentum (the energy is measured so that $\Delta_1=0$), and m_{sn} is its effective mass, which is positive in the HH1 and LH1 subbands and negative in the HH2 subband. The third term on the right-hand side of Eq. (1) describes the subband nonparabolicity and contains the parameter b_{sn}, whose dimension

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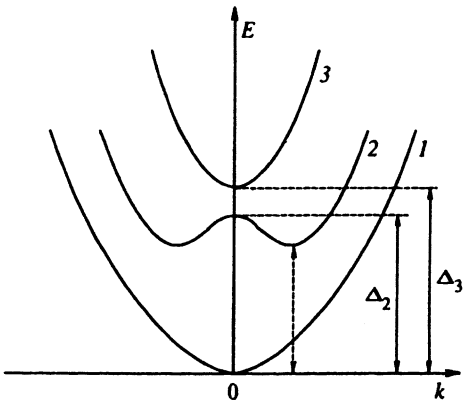

FIG. 1. Energy spectrum of holes in (1) HH1, (2) HH2, and (3) LH1 subbands.

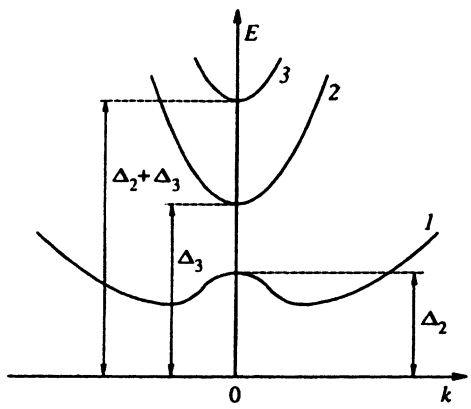

FIG. 2. Energy of two-hole states versus quasimomentum of relative motion. The curves 1, 2, and 3 correspond to the HH1–HH2, HH1–LH1, and HH2–LH1 hole pairs, respectively.

is length and which is introduced phenomenologically, typically 10^{-7}–10^{-6} cm, and m_0 is the free-electron mass.

The kinetic energy of a hole pair from the sn and $s'n'$ subbands at zero center-of-mass quasimomentum can be expressed as

$$E_\lambda(k)=\Delta_\lambda+\frac{\hbar^2k^2}{2m_\lambda}+\frac{\hbar^2}{2m_0}b_\lambda^2k^4, \tag{2}$$

where we have written $\lambda=\{sn,s'n'\}$, $\Delta_\lambda=\Delta_{sn}+\Delta_{s'n'}$, $b_\lambda^2=b_{sn}^2+b_{s'n'}^2$, and the reduced effective mass of the hole pair is defined as usual:

$$m_\lambda^{-1}=m_{sn}^{-1}+m_{s'n'}^{-1}. \tag{3}$$

In Eq. (2) k is the $2D$ quasimomentum of the relative motion in the pair.

It follows from Eq. (3) that if one hole is from the HH2 band, the reduced mass m_λ may be either positive or negative. In the latter case, which will be considered below, two holes can be in a bound state only under the condition

$$|m_{\mathrm{HH2}}|<m_{sn}, \tag{4}$$

where $sn=$HH1 or LH1. We will indicate heterostructures in which the condition given by Eq. (4) holds at $sn=$HH1. In this paper, we assume for definiteness that $m_{\mathrm{HH1}}>|m_{\mathrm{HH2}}|>m_{\mathrm{LH1}}$. In this case, the spectrum of two-hole states without Coulomb interaction has the form given in Fig. 2.

2. The Coulomb interaction between two holes is, naturally, repulsive if their reduced mass is positive. Otherwise their interaction is effectively attractive, which takes place in the case of holes from the HH1 and HH2 subbands.

It is convenient to consider the interaction between holes in the Wannier representation. The effective-mass envelope function of the hole pair without Coulomb interaction, $|\lambda,k,-k\rangle$, can be presented as a product of the envelope functions of their respective subbands[16]:

$$|snk\rangle=\frac{1}{\sqrt{S}}f_{snk}(z)\exp(ik\rho), \tag{5}$$

where S is the heterostructure area, the z-axis is the structure growth axis, ρ is the transverse vector of the relative motion in the pair, and $f_{snk}(z)$ is the $1D$ envelope function of a hole with quasimomentum k in the sn subband, which is the solution of the $1D$ Schrödinger equation with a rectangular quantum-well potential and appropriate boundary conditions on interfaces.[16] The equations are transformed to the Wannier representation, in which the discrete quantum parameter β has a sense of the vector connecting the two holes, through the conventional unitary transformation

$$|\lambda,\beta\rangle=\frac{1}{N}\sum_k \exp(-ik\beta)|\lambda,k,-k\rangle, \tag{6}$$

where N is the number of elementary cells in the heterostructure area.

In the Wannier representation, the kinetic energy of the holes in Eq. (2) is diagonal in both subband index and β if the quasimomentum k is replaced with $-i\nabla$, where ∇ is the gradient operator with respect to β. The potential energy can be expressed using the approximation $f_{snk}(z)\approx f_{sn0}(z)\equiv f_{sn}(z)$,[16] i.e., the mixing of light and heavy holes is ignored in the envelope function, although it is taken into account in the kinetic energy [Eq. (2)]. This approximation is justified because the mixing vanishes as $k\to 0$ and the typical dimension a^* of bound states of hole pairs is considerably larger than the lattice constant: $a^*\gg a$. Therefore, the envelope function is constructed largely from the states whose quasimomentum satisfies the condition $ka^*\ll 1$. Hence the Coulomb matrix elements connecting the HH and LH subbands, which are proportional to a/a^*, may be equated to zero, and the spinor nature of the hole envelope functions defined by Eq. (5) may be ignored. Another consequence of this approximation is that the Coulomb interaction is diagonal with respect to β in the Wannier representation:[17]

$$\langle\lambda\beta|U|\lambda_1\beta\rangle=\int dz\,dz' f_{sn}(z)f_{s_1n_1}(z)G(\beta;z,z') \times f_{s'n'}(z')f_{s_1'n_1'}(z'). \tag{7}$$

Here we have written $\lambda_1\equiv\{s_1n_1,s_1'n_1'\}$, $G(\beta;z,z')$ is the electrostatic Green's function for the heterostructure, and purely real $1D$ envelopes $f_{sn}(z)$ can be selected in all cases.

Usually the difference between the dielectric constants of quantum-well and quantum-barrier materials is small, so the effect of image charges can be neglected in our qualitative analysis of hole states, although it may be significant and the electrostatic Green's function can be expressed as

$$G(\beta;z,z')=\frac{1}{\varepsilon}\frac{1}{\sqrt{\beta^2+(z-z')^2}}, \tag{8}$$

where ε is the average dielectric constant of the heterostructure.

Thus, the effective Hamiltonian of the hole pair can be written in the form

$$H_{\lambda\lambda_1}=E_\lambda(-i\nabla)\delta_{\lambda\lambda_1}+U_{\lambda\lambda_1}(\beta), \tag{9}$$

where $U_{\lambda\lambda_1}(\beta)$ is determined by Eq. (7). We will denote the diagonal elements $U_{\lambda\lambda}$ of this operator by U_λ, where λ only includes the indices of the heavy-hole subbands. It is obvious that nondiagonal elements $U_{\lambda\lambda_1}$ are considerably smaller than diagonal ones because the envelope function of the HH1 subband has no nodes, whereas the HH2 envelope has one node, and hence the integrand in Eq. (7) changes sign.

3. Let us determine the density of states $g(E)$ in the usual way through the Green's operator $G(E)$ of the Hamiltonian defined by Eq. (9) written in the form

$$H=H^{(0)}+U,$$

where $H^{(0)}$ is the operator of the hole pair kinetic energy and U is the operator of the pair potential energy. We have[18]

$$g(E)=\frac{1}{\pi}\lim_{\gamma\to+0}\operatorname{Im}\operatorname{Tr}G(E-i\gamma), \tag{10}$$

where $G(E)=(E-H)^{-1}$ satisfies the Dyson equation

$$G=G^{(0)}+G^{(0)}UG, \tag{11}$$

and $G^{(0)}(E)=(E-H^{(0)})^{-1}$ is the Green's operator of the Hamiltonian $H^{(0)}$. Instead of expressing G in the form of an infinite series in powers of U, let us use a different technique for calculating the density of states in the region of expected resonance. Suppose that the eigenfunctions of H are known:

$$H|\alpha nm\rangle=E_{\alpha nm}|\alpha nm\rangle. \tag{12}$$

Here α is a quantum number that determines the type of the hole pair, which is, generally speaking, a mixture of states with different λ, and n and m are quantum numbers characterizing the relative motion of the holes. Then the Dyson equation can be easily solved in the $\{\alpha nm\}$-representation, and the diagonal element of the Green's operator, $\langle\alpha nm|G|\alpha nm\rangle\equiv G_{\alpha nm}$, can be written in closed form, i.e.,

$$G_{\alpha nm}=\frac{\langle\alpha nm|G^{(0)}|\alpha nm\rangle}{1-\langle\alpha nm|G^{(0)}U|\alpha nm\rangle}. \tag{13}$$

We expand the functions $|\alpha nm\rangle$ in the basis formed by the eigenfunctions of the operator $H^{(0)}$:

$$|\alpha nm\rangle=\sum_{\lambda k}|\lambda k\rangle\langle\lambda k|\alpha nm\rangle, \tag{14}$$

where

$$|\lambda k\rangle\equiv|\lambda,k,-k\rangle=\frac{1}{\sqrt{S}}f_{sn}(z)f_{s'n'}(z')\exp(ik\rho). \tag{15}$$

Introducing the notation $G_\lambda^{(0)}(k,E)=\langle\lambda k|G^{(0)}|\lambda k\rangle$, we have

$$G_{\alpha nm}(E)=\frac{\Sigma_{\lambda'k'}G^{(0)}_{\lambda'}(k',E)|\langle\alpha nm|\lambda'k'\rangle|^2}{1-\Sigma_{\lambda k}G^{(0)}_\lambda(k,E)\langle\alpha nm|\lambda k\rangle\langle\lambda k|U|\alpha nm\rangle}. \tag{16}$$

4. It is obvious that the elementary excitations of hole pairs are determined by the poles of the function (16), and the positions of the peaks in the density of states corresponding to metastable states can be found by calculating the roots of the denominator on the right-hand side of Eq. (16). The functions $|\alpha nm\rangle$ can be approximately calculated using the technique[19] based on the Hamiltonian in Eq. (9) presented in the form

$$H_{\lambda\lambda_1}=H^{(C)}_\lambda(\chi)\delta_{\lambda\lambda_1}+W_{\lambda\lambda_1}(\chi), \tag{17}$$

where

$$H^{(C)}_\lambda(\chi)=\Delta_\lambda-\frac{\hbar^2}{2m_\lambda}\nabla^2+\frac{\chi e^2}{\rho}, \tag{18}$$

and the operator $W_{\lambda\lambda_1}(\chi)$ is added to the Hamiltonian in Eq. (18) to obtain the full Hamiltonian in Eq. (17). The parameter χ is derived from the condition that the first-order energy correction due to the operator

$$W_{\lambda\lambda}(\chi)=\frac{\hbar^2}{2m_0}b^2_\lambda(\nabla^2)^2+U_{\lambda\lambda}(\rho)-\frac{\chi e^2}{\rho} \tag{19}$$

should be zero, then the nondiagonal elements $W_{\lambda\lambda_1}=U_{\lambda\lambda_1}$ are calculated using perturbation theory, as a result of which hole states are mixed through the Coulomb interaction. In what follows, we will ignore this mixing because, as was stated above, the nondiagonal elements are relatively small. In this approximation, the quantum parameters α and λ are, naturally, equivalent.

5. If $m_\lambda<0$ holds, the operator in Eq. (18) has both discrete spectrum and continuum eigenvalues. The discrete states form, naturally, a Coulomb set of levels with the energies

$$E^{(C)}_{\lambda n}=\Delta_\lambda+\frac{\chi^2\mathrm{Ry}^{(\lambda)}}{(n+1/2)^2}, \tag{20}$$

where $n=0,1,2,\ldots$, and $\mathrm{Ry}^{(\lambda)}=|m_\lambda|e^4/2\hbar^2$. The parameter χ in Eq. (20) is derived from the equation

$$\langle\lambda nm|W_{\lambda\lambda}(\chi)|\lambda nm\rangle=0, \tag{21}$$

in which the eigenvalues of the Hamiltonian in Eq. (18) are the usual wave functions of a $2D$ hydrogen atom. Specifically, the "ground" state has the wave function

$$|\lambda 00\rangle=\frac{1}{\sqrt{2\pi}}\frac{4\chi}{a_\lambda}\exp\left(-\frac{2\chi\rho}{a_\lambda}\right)f_{sn}(z)f_{s'n'}(z'), \tag{22}$$

where $a_\lambda=\hbar^2/|m_\lambda|e^2$. In calculating $\langle\lambda 00|\lambda k\rangle$, we obtain the well-known Gegenbauer integral, which yields

$$\langle \lambda 00|\lambda k\rangle = \sqrt{\frac{2\pi}{S}}\left(\frac{2\chi}{a_\lambda}\right)^2 \frac{2}{[(2\chi/a_\lambda)^2+k^2]^{3/2}}, \tag{23}$$

and, taking into account that $G_\lambda^{(0)}(k,E)=(E-E_\lambda(k))^{-1}$, we obtain

$$\sum_k G_\lambda^{(0)}(k,E)|\langle \lambda 00|\lambda k\rangle|^2 = -\left(\frac{2m_0 b_\lambda^2}{\hbar^2}\right)\left(\frac{2\chi b_\lambda}{a_\lambda}\right)^4 \frac{1}{x_E(x_E+x_\lambda)^3}\ln\left|\frac{x_E+m_0/2m_\lambda}{x_E-m_0/2m_\lambda}\right|. \tag{24}$$

Here

$$x_E^2=\left(\frac{m_0}{2m_\lambda}\right)^2+\frac{2m_0 b_\lambda^2(E-\Delta_\lambda)}{\hbar^2}, \quad x_\lambda=\left(\frac{2\chi b_\lambda}{a_\lambda}\right)^2-\frac{m_0}{2m_\lambda}. \tag{25}$$

6. Given the envelope functions $f_{sn}(z)$ and $f_{s'n'}(z')$, we can calculate the matrix element $\langle \lambda k|U|\lambda 00\rangle$ numerically. Since the electrostatic Green's function in Eq. (8) can be approximated by the formula $G(\beta;z,z')\sim 1/\varepsilon\beta$ in the limit $\beta \gg d$, where d is the typical dimension of the heterostructure along the z-axis, we use this approximation to demonstrate some properties of Eq. (16) for $n=m=0$. In this case the matrix element can be calculated through a Gegenbauer integrals and is expressed as

$$\langle \lambda k|U|\lambda 00\rangle \approx \sqrt{\frac{2\pi}{S}}\left(\frac{4\chi e^2}{\varepsilon a_\lambda}\right)\frac{1}{\sqrt{(2\chi/a_\lambda)^2+k^2}}. \tag{26}$$

After introducing the notation $E_i=E-\Delta_\lambda$ and dimensionless variables

$$\alpha=\frac{m_0}{2|m_\lambda|}\left(\frac{a_\lambda}{2\varepsilon\chi b_\lambda}\right)^2, \quad x=\sqrt{1+\frac{2}{\alpha}\frac{E_i}{4\varepsilon^2\chi^2 \mathrm{Ry}^{(\lambda)}}}, \tag{27}$$

we can express the denominator on the right-hand side of Eq. (16) as

$$D(x)=1+\frac{2}{\varepsilon\chi}\frac{1}{x}\left\{\frac{1}{1+\alpha(1-x)}\left[1+\frac{\ln|\alpha(x-1)|}{1+\alpha(1-x)}\right]-\frac{1}{1+\alpha(1+x)}\left[1+\frac{\ln|\alpha(x+1)|}{1+\alpha(1+x)}\right]\right\}. \tag{28}$$

The binding energy E_i and parameter x are complex variables with vanishingly small imaginary parts, according to the definition given by Eq. (10). The real part of Eq. (28) allows us to determine the positions of the peaks in the density of states corresponding to metastable states from the condition $\mathrm{Re}\, D(x)=0$. It is natural that Eq. (28) has a unique solution corresponding to the hole pair state with the quantum numbers $n=m=0$. The imaginary part of Eq. (28) can be easily determined by calculating the sum with respect to k in Eq. (16) in the limit $\gamma\to+0$, and it determines the width of the peak in the density of states. It is easier, however, to calculate the line shape numerically in accordance with Eq. (10) by separating the real and imaginary parts of the denominator on the right-hand side of Eq. (16). For this reason, we do not give the results of the simple, but rather lengthy calculations.

7. The approximate expression, Eq. (2), for the spectrum of the hole subbands at small k may be used if the curves do not cross at $ka^*\leqslant 1$ and corresponding states of different subbands are not mixed. Therefore, we limit our model by the condition $E_2(k_m)>E_1(k_m)$, where k_m is the momentum of the HH2 subband minimum (the radius of the minimum energy ring in the quasimomentum space). Besides, we can approximate the HH1 spectrum for simplicity by a quadratic function, i.e., $b_{\mathrm{HH1}}=0$. Then the condition on the effective masses in the HH1 and HH2 subbands in our model takes the form

$$|m_2|>\frac{m_1}{\sqrt{(2m_1x_0/m_0)^2+1}-1}, \tag{29}$$

where $x_0^2=2m_0b_2^2\Delta/\hbar^2$. The necessary condition for the existence of bound states of the HH1 and HH2 holes expressed by Eq. (4) is compatible with Eq. (29) only if the denominator on the right-hand side of Eq. (29) is larger than unity. Therefore the range of admissible parameters of our model is defined by the condition

$$b_2^2>3\left(\frac{m_0}{2m_1}\right)^2\frac{\hbar^2}{2m_0\Delta}. \tag{30}$$

For example, in a nanometer heterostructure with $\Delta\geqslant 40$ meV and $m_1\geqslant 0.1m_0$, the lower limit of the nonparabolicity parameter is about $5\cdot 10^{-7}$ cm.

Another limitation on the model parameters follows from the general condition of the Wannier scheme validity, namely, the effective Bohr radius of the hole bound state ($a^*\sim\varepsilon a_\lambda$) should be much larger than the interatomic distance a. Thus the admissible reduced hole mass in our model has both upper and lower limits:

$$-\frac{\varepsilon a_B}{a}<\frac{m_\lambda}{m_0}<-\frac{m_1}{m_0}\frac{1}{\sqrt{(2m_1x_0/m_0)^2+1}-2}, \tag{31}$$

where $a_B=\hbar^2/m_0e^2$. One can verify that these inequalities define a wide range of realistic model parameters, so our results can be applied to real structures.

8. The parameter χ^{-1} derived from Eq. (21) can be interpreted as an effective dielectric constant of the heterostructure characterizing interaction between the holes in the state with $n=m=0$. Within the framework of our simple model, Eq. (21) can be rewritten in the form[17]

$$\chi-\frac{1}{\varepsilon}+\frac{8|m_\lambda|}{m_0}\left(\frac{b_\lambda}{a_\lambda}\right)^2\chi^3(A-\ln\chi)=0, \tag{32}$$

where $A=\ln(\pi a_\lambda/2a)-3/4$. Therefore, within the range of model parameters defined by Eqs. (29)–(31) there is a unique solution of Eq. (32), which defines χ (note that the condition $\varepsilon\chi<1$ holds), so that the parameter α introduced by Eq. (27) is determined. Figure 3 shows the lines of zero denominator $D(x)$ of the Green's function in Eq. (16) plotted on the plane defined by the two parameters $\varepsilon\chi$ and α, at different values of $\varepsilon^2E_i/4\mathrm{Ry}^{(\lambda)}$ (these values are shown on the corresponding curves). The diagram demonstrates that the binding energy E_i is always smaller than $4\,\mathrm{Ry}^{(\lambda)}/\varepsilon^2$, which is the upper limit of the binding energy due to the potential $\propto\rho^{-1}$ in two dimensions.[20]

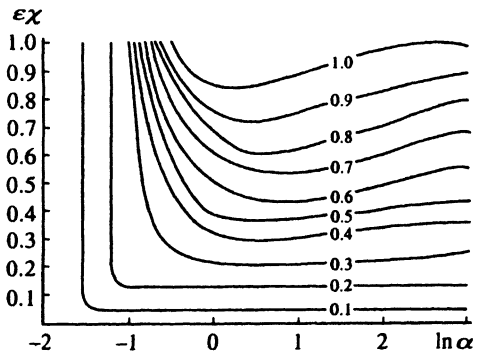

FIG. 3. Solutions of the equation $D(x)=0$. The numbers on the curves denote the binding energy E_i divided by $4\mathrm{Ry}^{(\lambda)}/\varepsilon^2$.

The line shape corresponding to the discussed metastable state can be easily derived by numerical methods from Eq. (16). It is very close to a Lorentzian curve

$$\delta g(E)=\frac{1}{\pi}\frac{\Gamma}{(E-E_i)^2+\Gamma^2}, \tag{33}$$

and for admissible model parameters the line width Γ is usually much smaller than E_i. For example, with $b_\lambda \approx 5\cdot 10^{-7}$ cm and $|m_\lambda|/m_0 \approx 0.1$ we have $\Gamma \sim 0.1(4\mathrm{Ry}^{(\lambda)}/\varepsilon^2)$.

A numerical calculation for an $Al_{0.3}Ga_{0.7}As$–GaAs heterostructure with quantum wells of widths $9a$ and $5a$ separated by a quantum barrier of a width $4a$ indicates that the hole spectrum in HH1, HH2, and LH1 subbands is similar to that given in Fig. 1, and the conditions on the model parameters described above are satisfied. The reduced mass of the pair of holes from the HH1 and HH2 subbands is $m_\lambda=-0.27m_0$ ($m_1=0.123m_0$ and $m_2=-0.085m_0$), and the energy gap between edges of the HH1 and HH2 subbands is $\Delta_2=25.6$ meV ($\Delta_{\mathrm{HH1}}=22.4$ meV and $\Delta_{\mathrm{HH2}}=48.0$ meV, energies are measured with respect to the GaAs valence band top). These parameters are compatible with the limitations on the parameter b_2 for the HH2 subband: $6.3\cdot 10^{-7}$ cm $\leqslant b_2 \leqslant 10^{-6}$ cm.

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