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Theory of holes in quantum dots

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The single-particle spectrum of holes in parabolic quantum dots in a magnetic field is investigated. The calculated Landau-level structure and allowed transition-energy spectrum for the heavy and light holes is considerably more complicated than that obtained for quantum dots containing electrons. This is because of the valence-band mixing, which produces strong anticrossings of the Landau levels and substantial wave-function mixing. It is noted that the band mixing causes Kohn's theorem to be violated, thus allowing for a richer far-infrared optical response.

With recent progress in nanofabrication technology it has become possible to confine electrons in all three spatial dimensions in semiconductor structures called quantum dots.[1-4] Quantum dots can be viewed as artificial atoms with the atomic Coulomb potential replaced by the confinement potential of the dot. While it was anticipated that quantum dots would exhibit rich optical spectra reflecting a complicated energy-level scheme, as is the case for atoms, the observed far-infrared (FIR) spectra were considerably simpler, being dominated by only two peaks.[1-4] This surprising result was shown to be essentially a consequence of the parabolic form of the confinement of the electrons in quantum dots. For such confinement, the generalized Kohn's theorem[5-8] states that the electron-electron interactions do not influence the long-wavelength response because the dipole field couples only to the center-of-mass motion of the system. Thus, the many-electron system exhibits a single-electron response consisting of two dipole-allowed transitions in finite magnetic field.

This peculiar result raises the question of what quantum-dot systems might exhibit more complicated and interesting spectra. It has been shown that in a two-dimensional (2D) hole gas the strong coupling between heavy- and light-hole (hh and lh) states causes Kohn's theorem to be violated.[9,10] This result applies even if the confining potential is parabolic. In this work, we present a theoretical investigation of a quantum-dot system in which heavy and light holes are confined rather than electrons. Below we develop the formalism to calculate the single-particle energy levels and dipole transition energies of quasi-2D holes in a GaAs-$Al_xGa_{1-x}As$ quantum-well system grown in the [001] direction, which is laterally confined by a parabolic potential. A magnetic field $B=(0,0,B)$ is assumed, with a vector potential, in the symmetric gauge, $\mathbf{A}=B/2(-y,x,0)$.

The Hamiltonian for this problem is given by

$$H=H_L+V(z)+V(\rho)\,, \tag{1}$$

where

$$H_L=\begin{bmatrix} P+Q+\frac{3}{2}\kappa B & S & R & 0 \\ S^\dagger & P-Q+\frac{1}{2}\kappa B & 0 & R \\ R^\dagger & 0 & P-Q-\frac{1}{2}\kappa B & -S \\ 0 & R^\dagger & -S^\dagger & P+Q-\frac{3}{2}\kappa B \end{bmatrix} \tag{2}$$

is the 4×4 Luttinger Hamiltonian in the presence of a magnetic field,[11] $V(z)$ is a quantum well or heterojunction potential, and $V(\rho)=\frac{1}{2}K\rho^2$, $\rho^2=x^2+y^2$, is the lateral confining potential. The diagonal terms

$$P\pm Q=\frac{\hbar^2}{2m_0}(\gamma_1\mp 2\gamma_2)\frac{\partial^2}{\partial z^2}+\frac{1}{2m_0}(\gamma_1\pm\gamma_2)\left[\mathbf{p}+\frac{e}{c}\mathbf{A}\right]^2 \tag{3}$$

together with $V(z)$ give rise to the heavy- and light-hole subband states and corresponding Landau levels. The off-diagonal terms,

$$S=-\sqrt{3}\frac{\hbar^2}{m_0}\gamma_3\hat{k}_z\hat{k}_-,\quad R=-\frac{\sqrt{3}\hbar^2}{2m_0}\bar{\gamma}\hat{k}_-^2+\frac{\sqrt{3}\hbar^2}{2m_0}\mu\hat{k}_+^2\,,$$
$$\hat{k}_i=\frac{1}{i}\frac{\partial}{\partial x_i}+\frac{e}{c}A_i,\ i=x,y,z,\ \hat{k}_\pm=\hat{k}_x\pm i\hat{k}_y,\ \bar{\gamma}=\tfrac{1}{2}(\gamma_2+\gamma_3),\ \mu=\tfrac{1}{2}(\gamma_3-\gamma_2)\,, \tag{4}$$

introduce the mixing between heavy- and light-hole states.

If this mixing is neglected ($R=S=0$), the corresponding eigenstates become distinct. It is then convenient to include the lateral confinement, $V(\rho)$, by introducing independent sets of "hybrid oscillators"[12] for heavy holes and light holes, respectively. The single-particle spectra for the two species of holes consists of two contributions: The z confinement gives rise to a series of subbands whose splittings are determined essentially by the corresponding mass along the z direction, $(\gamma_1 \mp 2\gamma_2)^{-1}$; the lateral confinement produces a series of Landau ladders with splittings determined by the in-plane masses, $(\gamma_1 \pm \gamma_2)^{-1}$ in the high-field limit (see Fig. 1). The full single-particle spectrum is just the superposition of these two spectra. For vanishing magnetic field, the degeneracy of the eigenstates is characteristic of the SU(2) dynamical symmetry of the 2D harmonic oscillator.[13]

The coupling between heavy and light holes produced by the off-diagonal terms in the Luttinger Hamiltonian leads to a much more complicated spectrum. Its evaluation, using the two sets of hybrid oscillators mentioned above, requires the calculation of matrix elements between heavy- and light-hole oscillator states. While we have performed this calculation, we present here an alternative approach. We consider only a single hybrid oscillator defined by

$$H_0=\frac{\gamma_1}{2m_0}\left(\mathbf{p}+\frac{e}{c}\mathbf{A}\right)^2+\tfrac{1}{2}K\rho^2=\hbar\Omega(1+\delta)(a_-^\dagger a_-+\tfrac{1}{2})+\hbar\Omega(1-\delta)(a_+^\dagger a_++\tfrac{1}{2}),$$
$$\Omega=\left[\frac{\gamma_1 K}{m_0}+(\gamma_1\omega_L)^2\right]^{1/2},\quad \omega_L=\tfrac{1}{2}\omega_c=\frac{eB}{2m_0c},\quad \delta=\frac{\gamma_1\omega_L}{\Omega}. \tag{5}$$

Here, the parameter δ provides a measure of the strength of the cyclotron energy produced by the magnetic field to that produced by the lateral confinement, while $a_\pm$ and $a_\pm^\dagger$ define the annihilation and creation operators. Using a representation in terms of 4×4 angular momentum matrices, J_z, $J_\pm=(1/\sqrt{2})(J_x\pm iJ_y)$ for $J=\frac{3}{2}$ the full Hamiltonian, in the axial approximation ($\mu=0$),[9] reads

$$\begin{aligned}H_a^{\rm IV}=&-\frac{\hbar^2}{2m_0}[\gamma_1-2\gamma_2(J_z^2-\tfrac{1}{3}J^2)]\frac{\partial^2}{\partial z^2}+V(z)+H_0+\hbar\omega_c\kappa J_z\\
&+\frac{\hbar\Omega}{2}\frac{\gamma_2}{\gamma_1}(J_z^2-\tfrac{1}{3}J^2)\{(1+\delta)^2(a_-^\dagger a_-+\tfrac{1}{2})+(1-\delta)^2(a_+^\dagger a_++\tfrac{1}{2})-(1-\delta^2)(a_-^\dagger a_+^\dagger+a_-a_+)\}\\
&+\frac{\hbar\Omega}{\gamma_1}\left\{\frac{\sqrt{2}\gamma_3}{\beta}\frac{\partial}{\partial z}(\{J_zJ_+\}[(1+\delta)a_--(1-\delta)a_+^\dagger]-\{J_zJ_-\}[(1+\delta)a_+^\dagger-(1-\delta)a_+])\right.\\
&\left.+\frac{\bar\gamma}{2}[(1+\delta)^2(J_+^2a_-^2+J_-^2a_-^{\dagger2})+(1-\delta)^2(J_+^2a_+^{\dagger2}+J_-^2a_+^2)-2(1-\delta^2)(J_+^2a_-a_+^\dagger+J_-^2a_-^\dagger a_+)]\right\},\end{aligned} \tag{6}$$

with $\beta=(m_0\Omega/\hbar\gamma_1)^{1/2}$. The first two lines of Eq. (6) correspond to the hole Hamiltonian in the absence of band mixing. The next term, proportional to γ_3, corresponds to the coupling term, S of Eqs. (2) and (4), while that proportional to $\bar\gamma$ corresponds to the coupling term, R. This Hamiltonian commutes with the total angular momentum operator,

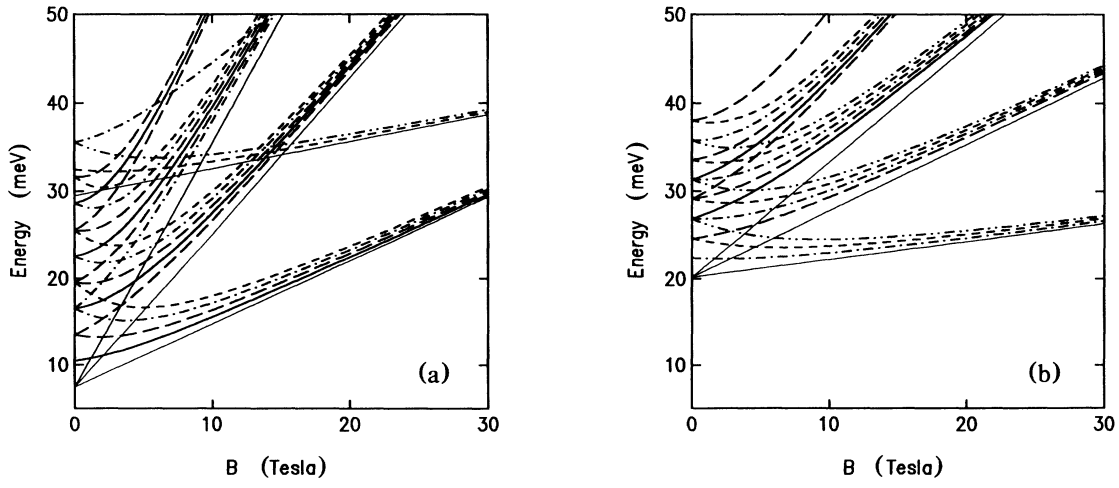

FIG. 1. Energy of a few of the lowest hole states without band mixing deriving from the lowest subbands due to confinement in the z direction vs magnetic field. (a) Heavy holes; (b) light holes. Thin solid lines indicate levels without lateral confinement.

$F_z = a_-^\dagger a_- - a_+^\dagger a_+ + J_z$. Thus, a proper basis is

$$\{|j,v,n_-,n_+\rangle,\ j=H,L,\ v=1,2,\ldots,\ n_\pm=0,1,\ldots n_+ - n_- \text{ is fixed}\}, \tag{7}$$

where $H(L)$ designates heavy (light) holes, v ranks the subbands, and n_+ (n_-) is the eigenvalue of $a_+^\dagger a_+$ ($a_-^\dagger a_-$). For sufficiently large B or vanishing lateral confinement, $\delta \to 1$ and H_a^{cv} without $V(z)$ becomes identical with the Hamiltonian of hole Landau levels under uniaxial strain.[14,15] This formulation has the advantage that the oscillator part can be evaluated throughout by using the algebra of $a_\pm$, $a_\pm^\dagger$, however, the confinement potential $V(\rho)$ is not diagonal in this basis.

Since no experimental data are currently available on quantum-dot systems containing holes, we consider a hypothetical but reasonable case of a GaAs quantum dot, where the z confinement is produced by a GaAs-$Al_xGa_{1-x}As$ quantum well of thickness, L. The potential energy then has the form $V(z) = \Delta E_v \Theta(|z| - L/2)$, where ΔE_v is the valence-band discontinuity, and increasing hole energy is measured as positive. We choose the following set of parameters:

$$L = 100\ \text{Å},\ \Delta E_v = 130\ \text{meV}\ (x = 0.3),$$

$$K = 1.32 \times 10^{-4}\ \text{meV/Å}^2,$$

and Luttinger parameters: $\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$, and $\kappa = 1.2$. These parameters give uncoupled heavy- and light-hole oscillator energies of 3 and 2.2 meV, respectively.

We retain the lowest two heavy-hole and lowest two light-hole subbands in our calculation. The inversion symmetry with respect to z requires that the subband functions entering each envelope function component have definite parity. For this case, R (S) couples heavy- and light-hole states of the same (opposite) parity.[16] Since the angular momentum, F_z, is conserved, we define the quantum number, $l = n_+ - n_- + M_J - \frac{3}{2}$, and coupling occurs only between states deriving from Landau levels with the same l. For each l, the envelope function components connected with hole spin M_J (eigenvalue of J_z) are expanded in a basis of oscillator functions with different n_- and n_+, but fixed $n_+ - n_- = l - M_J + \frac{3}{2}$. Up to fifteen oscillator functions are required for each component in order to achieve convergent results for the lowest five states with fixed l.

Figure 2(a) displays a few of the lowest hole Landau levels obtained by diagonalizing H_a^{cv} as a function of magnetic field B, for several different values of the total angular momentum l. It is apparent that the coupling between heavy and light holes produces a dramatic difference in the spectrum. The terms, R and S, break the dynamical SU(2) symmetry and cause a partial lifting of the $B = 0$ degeneracies observed in Fig. 1. They also cause a strong mixing of the basis states, which in general does not allow a strict classification as heavy- or light-hole states and leads to anticrossings between levels corresponding to states of the same angular momentum quantum number l. This is illustrated in Fig. 2(b), which displays the lowest few Landau levels for the case $l = 0$, in the range $B = 0$ to 20 T with (solid lines) and without (dashed lines) band mixing. The strength of the level repulsion is reflected by the considerably weaker field dependence and strongly compressed level spacings of the solid lines.

We consider now the effect of applying a microwave electric field, $\mathbf{E} = \mathbf{E}_0 e^{-i\omega t}$, where $\mathbf{E}_0 = E_x\hat{x} + E_y\hat{y}$. The corresponding vector potential is given by $\mathbf{A}' = -(ic/\omega)\mathbf{E}$. Following the general formulation developed by Trebin, Rössler, and Ranvaud[15] to determine the selection rules for electric dipole transitions between Landau levels in zinc-blende semiconductors, it is straightforward to generate the perturbation matrix, H', which is proportional to $\mathbf{E}\cdot\mathbf{v}$ where $\mathbf{v} = \partial H/\partial \hbar\mathbf{k}$ is the velocity operator derived from the Hamiltonian of Eq. (1). Therefore, $\mathbf{v}$ is determined by the Luttinger Hamiltonian, H_L [Eq. (2)], and has matrix character, i.e., the valence-band mixing modifies the selection rules.

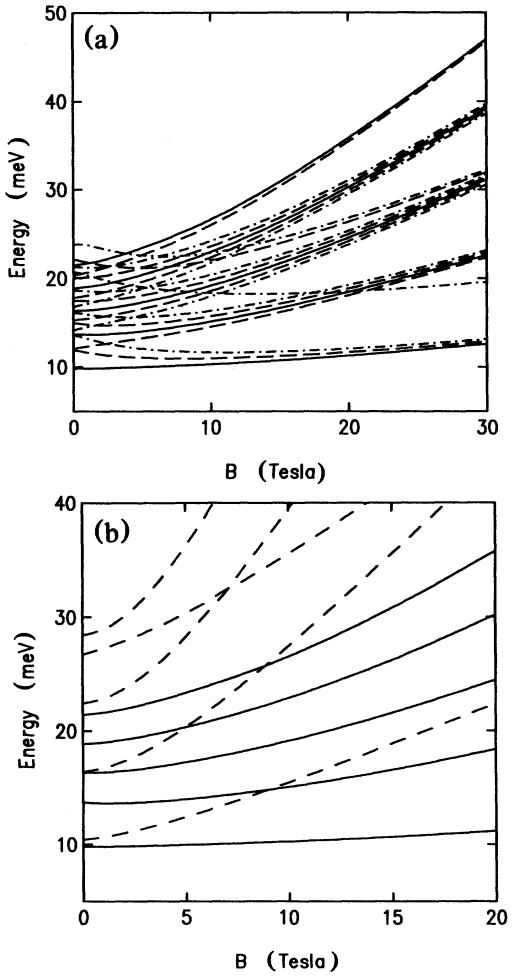

FIG 2. (a) Energy of hole states as in Fig. 1, but with band mixing included. For this case, the heavy- and light-hole states are no longer distinct and exhibit strong anticrossings that cause the levels to have a weaker field dependence. (b) Coupled (solid lines) and uncoupled (dashed lines) hole levels for $l = 0$. Strong anticrossings cause the coupled levels to be more closely spaced and to exhibit a weaker field dependence.

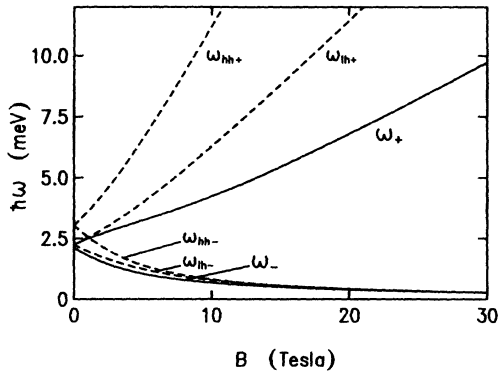

FIG. 3. Energy of dipole transitions vs magnetic field. Dashed lines are obtained without band mixing. Solid lines are the lowest transition energies for $\Delta l = \pm 1$ including band mixing.

Neglecting the band mixing produced by the off-diagonal terms in H and H', the selection rules lead to only four allowed transition energies, two for each species of hole: $\omega_{\mathrm{hh}\pm}$ and $\omega_{\mathrm{lh}\pm}$. Figure 3 shows these transition energies as a function of B (dashed lines). For zero field, the + and − frequencies for hh and lh coincide while for fields sufficiently large that the cyclotron frequency is much larger than the oscillator frequency, the 2D spectrum is recovered with the − frequencies tending to zero and the + frequencies approaching the hh and lh cyclotron transition energies.

Taking into account the valence-band mixing we obtain for the lowest transition energies the solid lines shown in Fig. 3. The remarkable reduction in energy of the upper transition compared to the case where mixing is neglected is a consequence of the substantially weaker field dependence of the corresponding hole Landau levels (compare Figs. 1 and 2) and their correspondingly smaller energy spacings. While these transitions are obtained by considering only the diagonal terms of the dipole interaction matrix, H', the off-diagonal terms will allow for otherwise forbidden transitions to become allowed and further modify the uncoupled spectrum. These characteristics should provide the most noticeable differences between the optical properties of quantum dots containing holes and those containing electrons.

The matrix structure of the Hamiltonian, Eq. (1), and the dipole interaction H' is expected to have consequences if the Coulomb interaction between holes is considered. Following the result for electron systems[6–8] it is straightforward to show that when the Coulomb interaction is included but the band mixing neglected, the radiation dipole operator only connects the many-electron eigenstates whose energies differ by $\hbar\omega_{\mathrm{hh}\pm}$ and $\hbar\omega_{\mathrm{lh}\pm}$, and thus the single-particle spectrum is obtained. Including the band mixing, which can be understood as a coupling between the hole spin with the orbital motion in the confinement potential, the dipole operator takes a more complex form which leads to a violation of Kohn's theorem. It is therefore anticipated that the FIR response of quantum dots containing holes will be much richer that for dots containing electrons.

In summary, we have investigated the single-particle properties of holes in quantum dots. We have shown that the strong mixing between heavy and light holes leads to dramatic changes in the Landau-level spectrum and in the allowed optical transitions compared to the case of electrons in quantum dots.

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[16]It should be pointed out that in many quantum dot systems, the z confinement is produced by a heterojunction potential. The lack of inversion symmetry in this case causes all s_{ij} and κ_{ij} to be nonvanishing, and leads to additional couplings not presented here.