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Effects of band structure and spin in quantum dots

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Band structure and spin can modify the simple two-mode far-infrared spectra of quantum dots. In our investigation of these effects we start from a $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian for the conduction, valence, and spin-orbit split-off bands, with potentials for subband formation and harmonic lateral confinement. Taking into account the nonparabolicity of the conduction band and spin-orbit coupling due to the confining potentials, we calculate the dipole excitation spectra of quantum dots on InSb with one and two electrons. Spin-orbit coupling leads to a level anticrossing in the ω_- mode and nonparabolicity modifies the ω_+ mode demonstrating violation of Kohn's theorem.

Quantum dots are semiconductor nanostructures that confine electrons in all three spatial directions on the lengthscale of the de Broglie wavelength. They are realized by imposing a periodic lateral structure onto the otherwise two-dimensional electron systems of metal-oxide-semiconductor (MOS) devices[1] or heterostructures.[2,3] The observed magneto-dependent far-infrared (FIR) spectra are dominated by only two resonance positions independent of the number of electrons per dot. This can be explained by the assumption of a parabolic potential for the lateral confinement. A generalization of Kohn's theorem[4] then states that the many-particle system exhibits the dipole transitions of a one-particle system, as the dipole operator couples only to the center-of-mass motion.[5–7] Nonparabolicity in the confining potential[8,9] or in the energy-momentum relation[10] prohibits separation into relative and center-of-mass motion and leads to more complex FIR spectra. We investigate the influence of the nonparabolicity of the conduction band and spin-orbit interaction on the dipole excitation spectra of quantum dots with one and two electrons ("quantum dot helium"[11,12]). It is known that for dots on InSb the nonparabolicity leads to a lowering of the ω_+ mode,[1,10,13] whereas for dots on GaAs it is negligible.[2,3] The effects of spin-orbit interaction have not been investigated before.

To describe an electron in an external potential $V(\mathbf{r})$ within the envelope-function approximation, we start from the Schrödinger equation $H_{8\times8}\Psi = E\Psi$ for the multicomponent spinor $\Psi = (\psi_1, \psi_2, ..., \psi_8)$ of the envelope functions for the Γ_6^c conduction band (ψ_1 and ψ_2), the Γ_8^v valence band with heavy (ψ_3 and ψ_6) and light holes (ψ_4 and ψ_5), and the Γ_7^v spin-orbit split-off band (ψ_7 and ψ_8). The $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian $H_{8\times8}$ is determined by the symmetry of the s-like conduction and the p-like valence band:[14,15]

$$H_{8\times8} = \begin{pmatrix}
V & 0 & \frac{-1}{\sqrt{2}}Pk_+ & \sqrt{\frac{2}{3}}Pk_z & \frac{1}{\sqrt{6}}Pk_- & 0 & \frac{-1}{\sqrt{3}}Pk_z & \frac{-1}{\sqrt{3}}Pk_- \\
0 & V & 0 & \frac{-1}{\sqrt{6}}Pk_+ & \sqrt{\frac{2}{3}}Pk_z & \frac{1}{\sqrt{2}}Pk_- & \frac{-1}{\sqrt{3}}Pk_+ & \frac{1}{\sqrt{3}}Pk_z \\
\frac{-1}{\sqrt{2}}Pk_- & 0 & V-E_g & 0 & 0 & 0 & 0 & 0 \\
\sqrt{\frac{2}{3}}Pk_z & \frac{-1}{\sqrt{6}}Pk_- & 0 & V-E_g & 0 & 0 & 0 & 0 \\
\frac{1}{\sqrt{6}}Pk_+ & \sqrt{\frac{2}{3}}Pk_z & 0 & 0 & V-E_g & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{2}}Pk_+ & 0 & 0 & 0 & V-E_g & 0 & 0 \\
\frac{-1}{\sqrt{3}}Pk_z & \frac{-1}{\sqrt{3}}Pk_- & 0 & 0 & 0 & 0 & V-E_g-\Delta_0 & 0 \\
\frac{-1}{\sqrt{3}}Pk_+ & \frac{1}{\sqrt{3}}Pk_z & 0 & 0 & 0 & 0 & 0 & V-E_g-\Delta_0
\end{pmatrix}. \tag{1}$$

The band structure near the Γ point is characterized by the fundamental gap E_g, the spin-orbit splitting Δ_0, and Kane's matrix element P. A magnetic field $\mathbf{B} = \nabla \times \mathbf{A}$ can be included by defining $\mathbf{k} = -i\nabla + e/\hbar\,\mathbf{A}$, $k_\pm = k_x \pm ik_y$ with commutator relations $\mathbf{k}\times\mathbf{k} = -i\,e/\hbar\,\mathbf{B}$. By eliminating the components ψ_3 to ψ_8 from the Schrödinger equation for the spinor Ψ we obtain[16]

$$\left(\left[\frac{1}{E_g+\Delta_0+E-V}+\frac{2}{E_g+E-V}\right]\frac{P^2}{3\hbar^2}(\mathbf{p}+e\mathbf{A})^2+V(\mathbf{r})-E+\left[\frac{1}{E_g+\Delta_0+E-V}-\frac{1}{E_g+E-V}\right]\frac{P^2}{3\hbar}e\,\boldsymbol{\sigma}\cdot\mathbf{B}\right.$$
$$+\left[\frac{1}{(E_g+\Delta_0+E-V)^2}-\frac{1}{(E_g+E-V)^2}\right]\frac{P^2}{3\hbar}\boldsymbol{\sigma}\cdot[\boldsymbol{\nabla}V\times(\mathbf{p}+e\mathbf{A})]$$
$$\left.+\left[\frac{1}{(E_g+\Delta_0+E-V)^2}+\frac{2}{(E_g+E-V)^2}\right]\frac{P^2}{3\hbar}(-i\boldsymbol{\nabla}V)\cdot(\mathbf{p}+e\mathbf{A})\right)\cdot\begin{pmatrix}\psi_1(\mathbf{r})\\ \psi_2(\mathbf{r})\end{pmatrix}=0. \quad (2)$$

This Hamiltonian for the conduction band consists of a kinetic-energy term, the external potential $V(\mathbf{r})$, the Zeeman term $\sim\boldsymbol{\sigma}\cdot\mathbf{B}$, the spin-orbit term $\sim\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\mathbf{p})$, and the Darwin term $\sim\boldsymbol{\nabla}V\cdot\mathbf{p}$. The form of this Hamiltonian is known from the Pauli equation in relativistic quantum mechanics. This similarity[17] arises from the fact that the Pauli equation is derived from the 4×4 Hamiltonian of the Dirac equation in a similar way to the derivation of (2) from $H_{8\times8}$.

We describe the confinement of electrons in quantum dots by a separable potential of the form

$$V(\mathbf{r})=v_{xy}(\varrho)+v_z(z)=\tfrac{1}{2}m^*\omega_0{}^2\varrho^2+v_z(z), \quad (3)$$

where v_{xy} is the parabolic confinement in the xy plane and v_z the potential in the z direction that leads to subband formation. We assume that the subband separation is much bigger than the level separation due to v_{xy} and that only the lowest z subband is occupied. Then $E-V(\mathbf{r})$ is equal to $\langle E_k\rangle_z+E-v_{xy}(\varrho)$ and we approximate it by $\langle E_k\rangle_z$ in all terms of (2) but the kinetic energy. $\langle E_k\rangle_z$ stands for the expectation value of the kinetic energy for the lowest subband and E is now the eigenenergy for the problem in the xy plane. By introducing the effective mass for the bottom of the lowest subband

$$\frac{1}{2m^*}=\left[\frac{1}{E_g+\Delta_0+\langle E_k\rangle_z}+\frac{2}{E_g+\langle E_k\rangle_z}\right]\frac{P^2}{3\hbar^2} \quad (4)$$

we get from (2) a correction $H_K(\varrho)$ to the kinetic energy in effective-mass approximation. It is an axially symmetric function and depends on the eigenenergy E.

For a magnetic field $\mathbf{B}=(0,0,B)$ the Zeeman term is $\frac{1}{2}g^*\mu_B B\sigma_z$ with Bohr's magneton $\mu_B=e\hbar/(2m_e)$ and the effective g factor for the lowest subband

$$g^*=\frac{2e}{\mu_B}\left[\frac{1}{E_g+\Delta_0+\langle E_k\rangle_z}-\frac{1}{E_g+\langle E_k\rangle_z}\right]\frac{P^2}{3\hbar}. \quad (5)$$

In the spin-orbit term we replace $v_z'(z)$ and p_z by their expectation values $\langle v_z'\rangle$ and $\langle p_z\rangle=0$ for the lowest subband. In the symmetric gauge $\mathbf{A}=B/2(-y,x,0)$ and with the angular momentum $l_z=xp_y-yp_x$ this leads to

$$H_{\rm SO}=\left[\frac{1}{(E_g+\Delta_0+\langle E_k\rangle_z)^2}-\frac{1}{(E_g+\langle E_k\rangle_z)^2}\right]\frac{P^2}{3\hbar}$$
$$\times\left\{m^*\omega_0{}^2[l_z+\tfrac{1}{2}eB(x^2+y^2)]\sigma_z\right.$$
$$\left.-\frac{P^2}{3\hbar}\langle v_z'\rangle[(p_y+\tfrac{1}{2}eBx)\sigma_x-(p_x-\tfrac{1}{2}eBy)\sigma_y]\right\}. \quad (6)$$

For a parabolic potential in the xy plane the Darwin term is equal to a small and constant energy shift and can be neglected, but we consider its contribution due to v_z.

After this approximate separation we get a Schrödinger equation for the xy plane that, without H_K and $H_{\rm SO}$, can be solved analytically.[18] The eigenenergies are

$$E(n,m)=(2n+|m|+1)\hbar\omega+\frac{\hbar\omega_c}{2}m \quad (7)$$

with an additional Zeeman splitting of $g^*\mu_B Bm_s$. Here $\omega_c=eB/m^*$ stands for the cyclotron frequency and $\omega=\sqrt{(\omega_c/2)^2+\omega_0{}^2}$. The eigenstates $|n,m,m_s\rangle$ are characterized by the radial quantum number $n=0,1,2,...$, angular momentum $m=0,\pm1,\pm2,...$, and spin $m_s=\pm1/2$. The system exhibits two dipole transitions from the groundstate $|0,0,+1/2\rangle$ into $|0,\pm1,+1/2\rangle$ which fulfill the selection rule $\Delta m=\pm1$. The transition energies and oscillator strengths are

$$\Delta E_\pm=\hbar(\omega\pm\omega_c/2)=\hbar\omega_\pm, \qquad f_\pm=\frac{\omega_\pm}{2\omega}. \quad (8)$$

For zero magnetic field this gives $\hbar\omega_\pm=\hbar\omega_0$ and $f_\pm=1/2$.

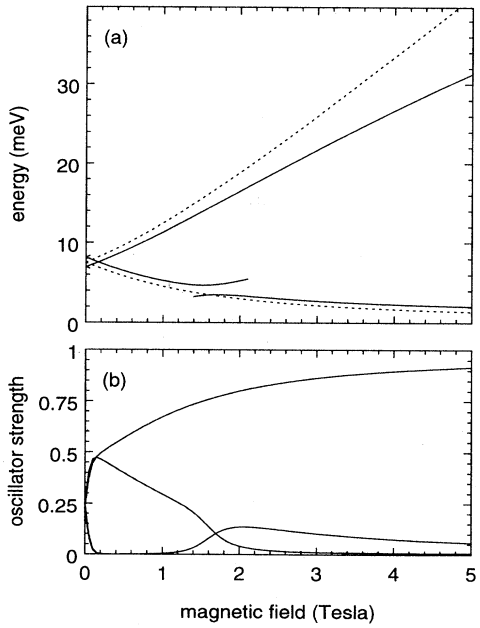

FIG. 1. (a) One-particle dipole transition energies for quantum dots on InSb in comparison with the results from effective-mass approximation (dotted lines), (b) corresponding oscillator strengths.

With increasing field ω_+ approaches ω_c and f_+ tends to one, while ω_- and f_- go to zero.

To include H_K and H_{SO} we diagonalize the corresponding matrix. H_K is diagonal in m and m_s and thus couples only states that are separated by an energy of at least $2\hbar\omega$. It does not induce a remarkable mixing of states with new dipole transitions, but it lowers all levels by amounts that are bigger for higher energies and therefore shifts the ω_+ mode to lower frequencies. The spin-orbit term (6) consists of a small first part $\sim \sigma_z$ that can be considered analytically and a second part $\sim \langle v_z' \rangle$ that leads to a coupling of states with same total angular momentum $m_j = m + m_s$ and the new selection rule for dipole transitions $\Delta m_j = \pm 1$.

We determined the expectation values of the lowest subband $\langle E_k \rangle_z = 20$ meV and $\langle v_z' \rangle = -10^{-3}$ eV/Å by the variational method for a Stern-Howard trial function.[19] For the MOS structure[1] the potential v_z arises from the interface between InSb and SiO_2. As the electronic properties of the oxide are not known, there is an uncertainty in the estimation of $\langle v_z' \rangle$.

Figure 1 shows the calculated energies and oscillator strengths of the one-particle dipole transitions for quantum dots on InSb with $\hbar\omega_0 = 7.5$ meV.[1] In comparison with $\hbar\omega_\pm$ given by (8), the lowering of the ω_+ mode due to the nonparabolicity of the conduction band and a splitting of the ω_- mode at a magnetic field of $B \approx 1.7$ T can be seen. This splitting results from a spin-orbit-induced level anticrossing of the states $|0,0,-1/2\rangle$ and $|0,-1,+1/2\rangle$ which is the final state of the ω_- transition. Because of the uncertainty in the determination of $\langle v_z' \rangle$ the size of this splitting cannot be given with absolute accuracy.

Finite-temperature effects have been considered by including dipole transitions from higher states with oscillator strengths multiplied by Boltzmann factors. For zero magnetic field the ground state is degenerate in m_s and we find four transitions with oscillator strength 0.25. With increasing field the Zeeman term lifts the degeneracy and the oscillator strengths for the transitions from the higher state $|0,0,-1/2\rangle$ decrease rapidly due to the Boltzmann factors, while those for the transitions from the ground state $|0,0,+1/2\rangle$ approach f_+ and f_- given by (8). This behavior can be seen in Fig. 1(b), where we used a temperature of 1 K. Furthermore f_- is modified by the anticrossing in the ω_- mode. In Fig. 1 only transitions with an oscillator strength exceeding 0.03 are drawn.

In the effective-mass-approximation the corresponding problem for two electrons can be separated into two-dimensional center-of-mass [$\mathbf{R} = \frac{1}{2}(\boldsymbol{\varrho}_1 + \boldsymbol{\varrho}_2)$, $\mathbf{P} = \mathbf{p}_1 + \mathbf{p}_2$, $M = 2m^*$] and relative coordinates [$\mathbf{r} = \boldsymbol{\varrho}_1 - \boldsymbol{\varrho}_2$, $\mathbf{p} = \frac{1}{2}(\mathbf{p}_1 - \mathbf{p}_2)$, $\mu = \frac{1}{2}m^*$]. The center-of-mass Hamiltonian is that of a single particle with eigenvalues $E(n_c, m_c)$ as given in (7). Without Coulomb interaction the relative motion also leads to eigenenergies $E(n_r, m_r)$. To include the interaction we diagonalize the Coulomb matrix whose elements can be calculated analytically.[20] It is diagonal in m_r with elements depending only on $|m_r|$. The Coulomb interaction raises all eigenenergies by an amount that is larger for small $|m_r|$ and the ground state changes its quantum number m_r from 0 to $-1,-2,...$ with increasing magnetic field.[11,12] Features of the relative motion cannot be seen in the dipole spectrum, as the dipole operator couples only to the center-of-mass motion and thus induces transitions which conserve the state of the relative motion.[5–7]

The spin state of the two-electron system can be an antisymmetric singlet with $S=0$, $M_S=0$ or a symmetric triplet with $S=1$, $M_S=-1,0,1$. As the symmetry of the orbital wave functions is given by m_r, states with odd m_r are spin triplets and states with even m_r are spin singlets. For small magnetic fields the ground state is $|n_c, m_c; n_r, m_r\rangle \otimes |S, M_S\rangle = |0,0;0,0\rangle \otimes |0,0\rangle$ which is not affected by the Zeeman term $g^*\mu_B B M_S$ and spin-orbit coupling, but states with odd m_r are lowered by the Zeeman term and for $B > 1$ T (InSb parameters) the ground state is $|0,0;0,-1\rangle \otimes |1,1\rangle$.

Spin-orbit coupling for the two-electron system can be treated in analogy to LS coupling by writing

$$\boldsymbol{\Sigma} \cdot \sum_{i=1,2} \boldsymbol{\nabla} V(\mathbf{r}_i) \times [\mathbf{p}_i + e\mathbf{A}(\mathbf{r}_i)], \tag{9}$$

where $\boldsymbol{\Sigma}$ are the 3×3 spin matrices for the triplet. In analogy to (6) this gives a term consisting of a small first part diagonal in M_S which can be considered analytically and a second part that couples spin to the center-of-mass-motion for states with same $M_S + m_c$.

The correction to the kinetic energy $H_K(\varrho_1) + H_K(\varrho_2)$ can approximately be written in center-of-mass and relative coordinates as

$$\frac{H_K(R)}{2} + \frac{H_K(r)}{2} - \sqrt{H_K(R)H_K(r)} - \frac{(\mathbf{P}\cdot\mathbf{p})^2}{2E_g\mu M}. \tag{10}$$

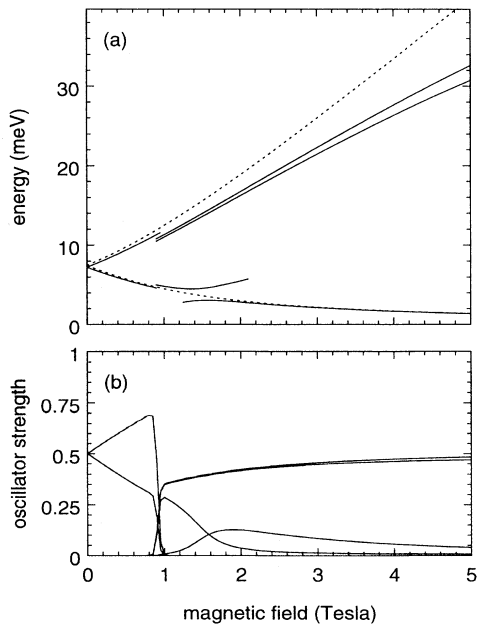

FIG. 2. (a) Dipole transition energies for quantum dot helium with InSb parameters in comparison with the results from effective-mass approximation (dotted lines), (b) corresponding oscillator strengths.

The last term (as the whole expression) is exact if H_K is just a fourth-order term in the momentum. It couples relative and center-of-mass motion for states with $\Delta m_c = -\Delta m_r = \pm 2$, but same total orbital angular momentum $m_c + m_r$. Important for dipole transitions is the coupling between the otherwise degenerate states $|0,1;0,-1\rangle \otimes |1,1\rangle$ and $|0,-1;0,1\rangle \otimes |1,1\rangle$. The ω_+ mode is split for magnetic fields where $|0,0;0,-1\rangle \otimes |1,1\rangle$ is the ground state.

Figure 2 shows the calculated dipole transitions and the corresponding oscillator strengths for quantum dot helium with the same InSb parameters as used for the one-particle system. For $B < 1$ T the spectrum is nearly identical with the results from (8). For $B > 1$ T the ground state is $|0,0;0,-1\rangle \otimes |1,1\rangle$ and the splitting of the ω_+ mode due to the correction of the kinetic energy can be seen in addition to the lowering of this mode. A level anticrossing between the states $|0,-1;0,-1\rangle \otimes |1,1\rangle$ and $|0,0;0,-1\rangle \otimes |1,0\rangle$ due to spin-orbit coupling becomes visible in the ω_- mode.

In summary we have calculated the deviations from the dipole spectrum of quantum dots in the effective-mass approximation due to band structure and spin. For InSb the nonparabolicity of the conduction band leads to a lowering of the ω_+ mode in good agreement with experimental data.[1] For quantum dot helium coupling of relative and center-of-mass motion leads to an additional splitting of the ω_+ mode. Nonparabolicity in the confinement potential couples in a similar way and induces a comparable modification of the dipole spectrum.[9] However, the magnetic-field dependence is different: corrections to the kinetic energy grow with increasing field, while the influence of the potential decreases.[10] With spin-orbit coupling included, the selection rules for dipole transitions have to be formulated for the total angular momentum and we predict an anticrossing in the ω_- mode that is basically similar for the one- and two-electron system. We repeated our computations with the band parameters of GaAs. In this case all deviations from the effective-mass approximation are negligible.

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