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Theory of luminescence from highly excited self-assembled quantum dots

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Abstract

The magneto-luminescence from lens-shaped self-assembled quantum dots, containing either many excitons or many electrons is studied. The single-electron and hole energy levels are shown to form degenerate shells in the absence of a magnetic field. In the case of many-exciton dots, the hidden symmetries associated with these degeneracies are responsible for a remarkable dependence of the absorption/emission spectrum on the number of excitons. In the case of many-electron dots, the degeneracies lead to the Hund-like oscillation of the total spin as a function of the number of electrons, which can be probed by photoluminescence. The symmetry-breaking effect of a magnetic field is also demonstrated.

Keywords: Quantum dots; Electronic states; Electron–electron interactions; Excitons; Photoluminescence

1. Introduction

It has been recently demonstrated [1,2] that the self-organization processes can be used to obtain very small, high-quality quantum dots. An epitaxial growth of materials with a large lattice mismatch (e.g. InGaAs on GaAs) is unstable, and at a critical coverage the strain leads to a spontaneous formation of small hills on a narrow wetting layer (WL). The actual shape and size of these quasi-zero-dimensional (quasi-0D) self-assembled dots (SAD) depends on the growth conditions, and different forms have been reported [1–5]. Here we concentrate on a class of lens-shaped InGaAs dots, investigated recently by single-electron capacitance (SECS), photoluminescence (PL), and far-infrared (FIR) spectroscopies [1–3,6–9].

2. Model

The lens-shaped InGaAs SAD [1,2,6–8] is modeled as a part of a sphere with fixed height h and radius at the base s, formed on a narrow WL of width t_w [10,11]. The entire structure is sandwiched between the pair of GaAs barriers. The discontinuities of the conduction and valence band energies at the InGaAs/GaAs interface lead to the confinement of both conduction electrons (e) and valence holes (h) in the quasi-2D InGaAs layer. The carriers are further localized in the area of the dot due to the effectively increased thick-

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ness of the layer, i.e. the effectively decreased energy of the lowest subband confined in the WL.

The detailed calculation [10] showed that within the area of a SAD ($r < s$) the effective lateral potentials acting on quasi-2D carriers within the WL are well approximated by a pair of parabolas: $\frac{1}{2}m_{\rm e}^*\omega_{\rm e}^2 r_{\rm e}^2$ and $\frac{1}{2}m_{\rm h}^*\omega_{\rm h}^2 r_{\rm h}^2$, where r's are the 2D positions, m^*'s denote the effective masses (in-plane mass in the case of h) and ω's measure the confinement strengths. In the absence of a magnetic field, the e and h bound states are these of 2D harmonic wells, labeled by orbital quantum numbers n and m, and spin projection σ: $|nm,\sigma\rangle \sim (a^+)^n(b^+)^m\,|0\;0\rangle\,|\sigma\rangle$. Due to dynamical symmetries of the confining potential the energy levels $\varepsilon_{nm} = \hbar\omega(n+m+1)$ form equidistant degenerate shells (denoted here by s, p, d, f, ...), with the states within a shell labelled by orbital angular momenta: $l_{\rm e} = m_{\rm e} - n_{\rm e}$ and $l_{\rm h} = n_{\rm h} - m_{\rm h}$, respectively. The electronic structure of a SAD is thus characterized by the number of bound e and h shells, and the intershell separations (we assume here $NS_{\rm e} = 5, NS_{\rm h} = 5, \hbar\omega_{\rm e} = 30\,{\rm meV}$, and $\hbar\omega_{\rm h} = 15\,{\rm meV}$, as appropriate for SADs reported by Raymond et al. [5,6]). These parameters are directly connected with the size and composition of a SAD and, to some extent, can be varied independently [10]. The important property of reported SADs is overlapping of e and h orbitals, a result of the cancellation of a difference in the effective mass by a difference in the height of the potential barrier at the interface. In a magnetic field B the degeneracy of shells is removed, and the e and h energy levels form the Fock–Darwin spectra: $\varepsilon_{nm} = \hbar\omega_+(n+\frac{1}{2}) + \hbar\omega_-(m+\frac{1}{2}) + g\mu_{\rm B}B\sigma$. The frequencies ω, degenerate without a field, split here into $\omega_\pm = \frac{1}{2}(\sqrt{\omega_{\rm c}^2 + 4\omega^2} \pm \omega_{\rm c})$, where $\omega_{\rm c} = eB/m^*c$ are the cyclotron frequencies. The Zeeman energies, scaled by the effective g-factors, are typically small compared to the intershell separations and will be neglected.

Using a composite index $i = [n,m]$ the Hamiltonian of the interacting e–h system may be written in a compact form:

$$H = \sum_{i\sigma} \varepsilon_i^{\rm e} c_{i\sigma}^+ c_{i\sigma} + \sum_{i\sigma} \varepsilon_i^{\rm h} h_{i\sigma}^+ h_{i\sigma} - \sum_{ijkl\sigma\sigma'} \langle ij|v_{\rm eh}|kl\rangle\, c_{i\sigma}^+ h_{j\sigma'}^+ h_{k\sigma'} c_{l\sigma} + \frac{1}{2}\sum_{ijkl\sigma\sigma'} \langle ij|v_{\rm ee}|kl\rangle\, c_{i\sigma}^+ c_{j\sigma'}^+ c_{k\sigma'} c_{l\sigma} + \frac{1}{2}\sum_{ijkl\sigma\sigma'} \langle ij|v_{\rm hh}|kl\rangle\, h_{i\sigma}^+ h_{j\sigma'}^+ h_{k\sigma'} h_{l\sigma}. \quad (1)$$

Operators $c_{i\sigma}^+$ ($c_{i\sigma}$) and $h_{i\sigma}^+$ ($h_{i\sigma}$) create (annihilate) an electron and hole with the spin projection σ on the orbital i with the single-particle (SP) energy ε_i, and $\langle ij|v|kl\rangle$ are the two-body Coulomb matrix elements [12,13].

The Hilbert space of e–h states is spanned by products of SP states: $|i_{\rm e1}\sigma_{\rm e1}, i_{\rm e2}\sigma_{\rm e2}, \ldots; i_{\rm h1}\sigma_{\rm h1}, i_{\rm h2}\sigma_{\rm h2}, \ldots\rangle = (c_{i_{\rm e1}\sigma_{\rm e1}}^+, c_{i_{\rm e2}\sigma_{\rm e2}}^+, \ldots)(h_{i_{\rm h1}\sigma_{\rm h1}}^+, h_{i_{\rm h2}\sigma_{\rm h2}}^+, \ldots)\,|{\rm vac}\rangle$. Each state is classified by a pair of good quantum numbers: total angular momentum R and total spin projection S_z. The total spin S^2 is not resolved in this basis. The eigenstates are obtained through the diagonalization of the Hamiltonian (1), in the eigen-subspaces (R, S_z).

The crucial property of small SADs is the strong quantization of SP motion compared to the strength of interparticle Coulomb interaction. This allows for the approximate description of correlated many-particle states in terms of occupation of SP shells. The intershell Coulomb scattering is weak, and in the ground state (GS) e and h fill degenerate shells according to the Pauli exclusion principle, applied independently to each type of carriers. The correlation effects appear only within a partially filled degenerate shell (containing a defined number of particles), and the underlying completely filled shells form a rigid core.

An important property of the electron–hole system is the hidden symmetry [13] associated both with degeneracies of electronic shells and with the equal strength of e–e, e–h, and h–h interactions due to the exact overlapping of e and h orbitals.

In the PL process, the emission of a photon by a dot is accompanied by the annihilation of an e–h pair. The measured PL spectrum represents an average over a large number of photons emitted from dots containing different numbers of carriers and being in different initial states. We shall assume here that an average number of electrons and holes is bound in each dot, controlled by intensity of illumination. Assuming further the initial equilibrium, we can derive the approximate PL intensity $I(\omega)$ as a function of the photon energy $\hbar\omega$ from the Fermi golden rule:

$$I_{\phi_0}(\omega) = \sum_{\rm f} |\langle\phi_{\rm f}|\mathscr{P}|\phi_0\rangle|^2 \delta(\mathscr{E}_0 - \mathscr{E}_{\rm f} - \hbar\omega), \quad (2)$$

where ϕ_0 and ϕ_f are the initial and final states of the system, with corresponding energies $\mathscr{E}_0$ and $\mathscr{E}_f$. The interband polarization operator $\mathscr{P}$ removing an e–h pair from the system can be decomposed into a pair of operators with definite circular polarizations of light: $\mathscr{P}=\mathscr{P}_+ + \mathscr{P}_-$, which simplify to $\mathscr{P}_+ = \sum_i c_{i\downarrow} h_{i\uparrow}$ and $\mathscr{P}_- = \sum_i c_{i\uparrow} h_{i\downarrow}$ due to the e–h orbital symmetry [14]. In order to account for the emission from excited initial states one can assume that the initial states are populated according to the Boltzmann distribution with an effective temperature T, and express the quasi-equilibrium PL spectrum as $I(T,\omega) \sim \sum_i \exp(-\mathscr{E}_i/T) I_{\phi_i}(\omega)$, where the summations goes over all possible initial eigenstates of the system ϕ_i [14].

3. Many-exciton SAD

Let us begin with the description of PL from a SAD containing many (N) excitons [13]. Thus, the upper edge of the PL spectrum coincides with the exciton addition/subtraction energy (exciton chemical potential), defined as the difference between the ground-state energies (GSE) of N and $N-1$ bound excitons: $\mu(N) = \mathrm{GSE}(N) - \mathrm{GSE}(N-1)$. The addition energy μ (circles), shown in Fig. 1, as a function of the number of excitons N, forms plateaus separated by discontinuous jumps. This resembles the behavior of a noninteracting e–h system (squares), where consecutive electron–hole pairs are added to the SP states according to the Pauli exclusion principle. Due to the degeneracy of shells, $\mu(N)$ is constant within a shell and increases by $\hbar\omega_e + \hbar\omega_h$ whenever a new shell begins to fill ($N = 1, 3, 7, 13, \ldots$). A result of the plateaus in $\mu(N)$ is a simple PL spectrum of a many-exciton SAD (see the top-left inset for $N=13$), containing few peaks corresponding to the recombination from (four) occupied shells. A similar, simple PL spectrum, with a comparable spacing between the peaks, might be recorded in a low-excitation experiment (one exciton per dot, dashed line), where the recombination from higher shells could be due to a low relaxation rate. However, the situation where the recombination occurs from many-exciton SADs can be identified by studying the power dependence of the PL spectrum [6,7].

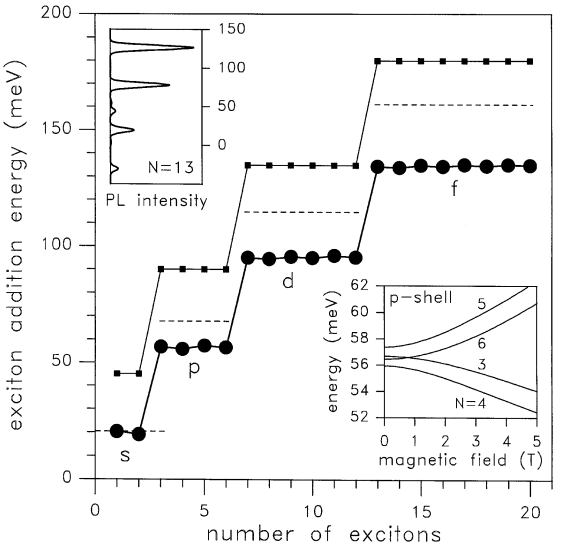

Fig. 1. Dependence of the exciton addition energy μ on the number of excitons N in a SAD. Circles – exact calculation as described in the text; squares – SP energy of the Nth added e–h pair; dashed line – a single interacting exciton added to isolated shells. Insets: PL spectrum for $N=13$ (top-left) and the magnetic-field dependence of $\mu(N)$ for $N=3$–6 (bottom-right).

The reason for the regular dependence of the exciton addition energy μ (i.e. the PL spectrum) of a SAD on the number of excitons N (i.e. the excitation power) are the hidden symmetries associated with (i) the degeneracy and large spacing of SP shells and (ii) the e and h orbital symmetry (interaction symmetry). As shown in Ref. [13] these symmetries lead to the condensation of excitons within an isolated shell t (in analogy to the condensation of magneto-excitons in the lowest Landau level [15–17]), with the addition energy $\mu(N) = E_X^t$ independent of N. For simplicity we shall ignore here the e and h spin (see Ref. [13] for the complete discussion). The correlated N-exciton states are constructed with the polarization creation operator $P^+ \sim \sum_i c_i^+ h_i^+$, where the summation over orbitals i is limited to the given shell t. The creation operator P^+, the annihilation operator P^-, and the projection $P_z = \sum_i (c_i^+ c_i + h_i^+ h_i - 1)$ form the polarization algebra $\boldsymbol{P}$, with the usual angular-momentum-like commutation relations. The approximate commutation relations between $\boldsymbol{P}$ and the total Hamiltonian (1) are the following: $[H, P_z] = 0$ and $[H, P^+] = E_X^t P^+$. Thus, the N-exciton eigenstates of P_z

and $P^2=\frac{1}{2}(P^+P^-+P^-P^+)+P_z^2\equiv P(P+1)$, constructed by acting N times with P^+ on vacuum, are at the same time the eigenstates of H (GSs at a given N), with the energy linear in N. These states correspond to: $P=\frac{1}{2}L=\text{const.}$ and $P_z=N-\frac{1}{2}L$, where L is the shell degeneracy ($N=1,\ldots,L$), and hence the addition/subtraction of excitons to/from a shell can be viewed as the rotation of the polarization vector $\boldsymbol{P}$ without changing its length P. The empty (completely filled) shell is represented by $\boldsymbol{P}$ pointing down (up): $P_z=-(+)P$.

The effects due to spin and intershell Coulomb scattering are weak [13] and, despite strong correlations, the consecutive excitons are added to the shell of a SAD with the same energy. A strong effect, however, is the (exchange) interaction between the excitons from a partially filled shell with the excitons filling the underlying shells, leading to a considerable renormalization of the chemical potential (band-gap renormalization). Summarizing, the GSE of a correlated N-exciton complex in a SAD depends on N in such a way, as if excitons from the same shell did not interact, while excitons from different shells attracted each other. The GSE(N), and thus also μ, can be estimated based on (i) the exciton binding energies for each shell E_X^{t}, and (ii) the exciton–exciton attraction energy for each pair of shells $E_{XX}^{\mathrm{t,t'}}$.

In a magnetic field B the degeneracy of SP shells is removed, and the plateaus in the exciton addition energy $\mu(N)$ are destroyed. The splitting of $\mu(N)$ for $N=3$–6 (four excitons added to the degenerate p-shell at $B=0$) is shown in the bottom-right inset in Fig. 1. The energy differences: $\mu(4)<\mu(3)$ and $\mu(6)<\mu(5)$, for excitons added with opposite spins to the same orbital, show the weak bi-exciton binding.

4. Many-electron SAD

We now turn to the PL from a SAD containing many electrons and a single hole ($N\mathrm{e}+\mathrm{h}$) [14]. To a good approximation, in the initial GS the h occupies the lowest-energy level $|0\ 0\rangle$, while the e's fill a number of lowest shells. In the PL process the h annihilates an e in the lowest (s) shell, i.e. creates a vacancy in the Ne initial system, which can be viewed as an excitation in the $(N-1)$e final system. The PL spectrum measures the spectral function of this vacancy. A crucial problem is the dependence of this spectral function on N, which determines whether PL can monitor N in charging experiments [9].

The e's filling a shell maximize their total spin S^i (in order to maximize the total exchange energy) according to the Hund rule. Since for $S^i\neq 0$ the GS is degenerate with respect to the projection S_z^i, we assume that a weak magnetic field removes this degeneracy and picks a non-degenerate GS with a specified $S_z^i=S^i$. The field has, however, negligible effect on the SP energy levels, and allows for the thermal depolarization of the h spin. The polarization of total e spin results in the polarization of the PL spectrum, and we shall analyze the two circular components separately.

The σ_- spectrum corresponds to the removal of a spin-minority ($\uparrow$) e, and thus to an increase of the total e spin projection $S_z^f=S_z^i+\frac{1}{2}$. The optically created final state $\mathscr{P}_-\,|\phi_0\rangle$ is the eigenstate of e spin with $S^f=S_z^f$. The calculated σ_--PL spectra for $N=1$–9 are shown in the left-hand column of Fig. 2. Two features are visible: (i) plateaus in the dependence of the main-peak energy on N, at $N=2$–4 and $N=6$–9, and (ii) emergence of the lower-energy peak for $N=5$.

The plateaus (i) can be explained in the single-configuration approximation (SCA), where $c_i^+c_i\equiv 0$ or 1, and the initial state minimizes the SP energy and is consistent with the Hund rule. The emission energy $\hbar\omega$ can be divided into: (1) SP energy of the recombining e–h pair, (2) interaction within the pair, and (3) interaction of the pair with the remaining $(N-1)$e. The first two terms are independent of N, and the third one (mainly e–e exchange, since e and h almost exactly screen each other) counts the number $N_\uparrow$ of remaining e spins parallel to the removed e spin (notation: $N_\sigma=\langle\phi_{\mathrm{f}}|c_\sigma^+c_\sigma|\phi_{\mathrm{f}}\rangle$). When N increases, until a shell is half-filled $N_\downarrow$ increases and $N_\uparrow$ is constant – plateau in $\omega(N)$. Conversely, when N passes the half-filling, $N_\downarrow$ remains constant and $N_\uparrow$ increases – $\omega(N)$ changes. As shows the exact numerical calculation (cf. Fig. 2), the Coulomb mixing between different configurations does not destroy this effect, and hence the shell structure of a SAD should be observed in the PL.

The splitting (ii) occurs when $N_\uparrow>0$, i.e. the final-state optical configuration $\mathscr{P}_-\,|\phi_0\rangle$ can be mixed via the Coulomb scattering with other, degenerate SP configurations. These Auger-like processes are

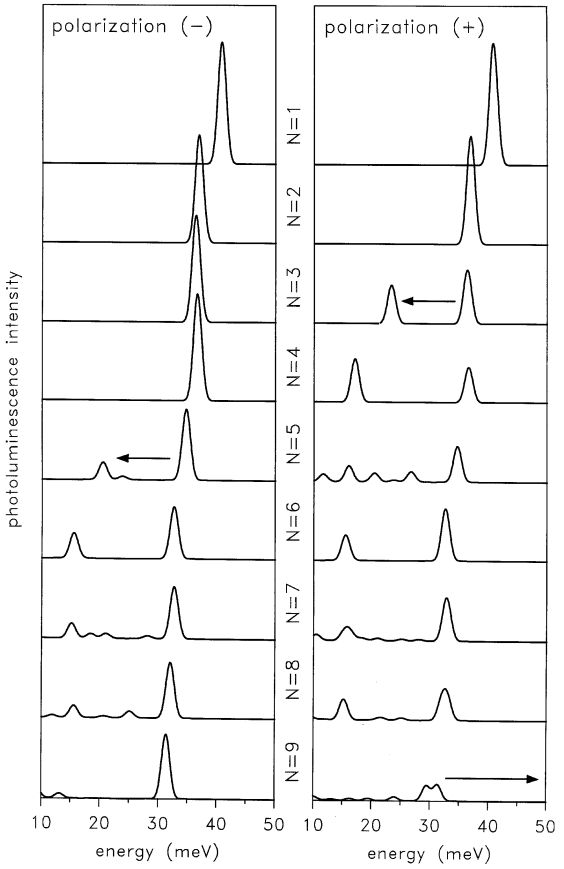

Fig. 2. Dependence of the polarized photoluminescence spectra $I_-(\omega)$ (left) and $I_+(\omega)$ (right) on the number of electrons N in a SAD. Arrows show the splittings; the additional peak for $N=9$ and polarization σ_+ appears at $\hbar\omega\sim 96$ meV and is not shown.

responsible for the occurrence of additional PL peaks, which take a fraction of intensity from the main peak.

The σ_+ spectrum corresponds to the removal of a spin-majority ($\downarrow$) e, and thus to a decrease of the total e spin projection $S_z^f = S_z^i - \frac{1}{2}$. The optically created final state $\mathscr{P}_+ |\phi_0\rangle$ has finite projections on a pair of spin subspaces: $S^f = S_z^f$ and $S^f = S_z^f + 1$. Consequently, the σ_+-PL spectrum can be decomposed into a pair of sub-spectra $I_\pm(\omega)$ corresponding to $S^f = S^i \pm \frac{1}{2}$. The sub-spectrum I_+ repeats the σ_- spectrum (with a reduced intensity), and I_- is unique for the σ_+ polarization. The calculated overall σ_+-PL spectra for $N=1$–9 are shown in the right-hand column of Fig. 2. The I_+ spectrum has been already discussed, and the two new features appearing in the I_- spectrum are: (i) emergence of the lower-energy peak for $N=3$, and (ii) emergence of the higher-energy peak for $N=9$.

The splitting (i) occurs whenever there is a partially filled shell in the initial state ($N=3$–$5, 7$–$11, \ldots$), i.e. when both final-state spins $S^f = S^i \pm \frac{1}{2}$ can be achieved by optically removing the s-shell e, with its spin parallel to the total spin of the partially filled shell.

The splitting (ii) signals the filling of a next zero-angular-momentum SP e state: $|1\ 1\rangle$, i.e. the possible Coulomb mixing between the optically created final configuration $\mathscr{P}_+ |\phi_0\rangle$ and the lower-energy configuration with the e from $|1\ 1\rangle$ moved to $|0\ 0\rangle$ state [8]. In other words, for $N>9$ the Coulomb relaxation $|1\ 1\rangle \rightarrow |0\ 0\rangle$ can accompany the recombination process.

5. Conclusion

We have studied the interacting e–h complexes confined in a SAD. The e and h SP energy levels in a SAD form degenerate shells, with the intershell spacing exceeding characteristic interaction energy. In the GS the confined carriers fill the consecutive shells so as to minimize the SP energy. In a many-exciton SAD the hidden symmetry associated with the equal strengths of e–e, e–h, and h–h interactions leads to the condensation of excitons within shells, and thus to the plateaus in the exciton addition energy μ as a function of N. As a result, the high-excitation PL spectrum of a SAD consists of few peaks, corresponding to the recombination of e–h from matching shells. In a many-electron SAD the e–e exchange interaction induces the spin-polarization of partially filled shells (Hund rule), which also leads to the plateaus in $\omega(N)$, and to the strong dependence of the PL spectrum on the polarization and N. In particular, the splittings in the PL spectrum, which can be identified based on the position and polarization, signal reaching critical N's (e.g. in a charging experiment monitored with PL). Both in many-exciton and many-electron SADs the magnetic field removes the degeneracy of SP shells and destroys the plateaus in $\omega(N)$, leading to more complicated PL spectra.

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