Effects of electron-electron interactions on excitonic absorption in charged self-assembled quantum dots

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We investigate theoretically the effects of electron-electron interactions on excitonic absorption in charged, lens-shaped, self-assembled quantum dots (SAD). The electronic shells of SAD are filled with N electrons according to generalized Hund's rules. In absorption, an exciton is added to the electronic system. The electronic part of the exciton couples in a nontrivial way to resonant electronic configurations of SAD through direct and exchange interaction while the mobile valence hole contributes through the entanglement of many-electron configurations. The two processes result in an excitonic absorption spectrum that reflects the filling of electronic shells. These processes are illustrated by detailed calculations of absorption spectra for few-electron complexes.

I. INTRODUCTION

The absorption of a photon in a semiconductor creates an electron in the conduction band and a hole in the valence band. An electron and a hole bind to form an exciton and the absorption spectrum is proportional to the density of states of this composite particle. When free carriers are present in the conduction band the absorption is quickly modified.[1] For a localized valence hole the absorption spectrum evolves from excitoniclike to a Fermi edge singularity (FES).[1,2] Much has been understood about the FES because the problem of noninteracting electrons coupled to a localized valence hole is exactly solvable.[2] However, in quasi-two-dimensional semiconductor systems neither electron-electron interactions nor the finite hole mass can be neglected and our understanding of the effect of free carriers on optical absorption is limited.[3–6] We can further our understanding of the effect of electron-electron interaction on excitonic absorption by studying simpler systems with limited Hilbert space where electron-electron and electron-hole interactions can be treated in a systematic way. A quantum dot[7] filled with electrons not only offers such an opportunity but is also an interesting electronic system in its own right. An ability to control, and hence detect, individual charges in self-assembled quantum dots (SAD) is essential for a wide range of applications, from single electron transistors and memories to quantum gates.[8] For these reasons interband optical properties of quantum dots charged with electrons have been investigated experimentally [9–12] and theoretically.[13–17] In this work we focus on excitonic absorption in charged self-assembled quantum dots. Emission spectra and excitons in charged quantum dots have already been investigated using exact diagonalization techniques.[15,17] The emission spectra showed spectral features related to the filling of electronic shells. In particular, partially filled shells led to a splitting of the emission line due to mixing of resonant final-state configurations. The splitting can be traced to spin polarization of half-filled shells. Recent experiments by Schmidt *et al.*[11] confirmed only some of the theoretically predicted features like band-gap renormalization. Other features due to correlations remained obscured by inhomogeneous broadening. In a related recent work, Warburton *et al.*[12] measured absorption spectra of ensembles of charged quantum dots as a function of the number of electrons N in the dot. His experiments nicely demonstrated the Pauli exclusion principle, i.e., that once electronic shells are fully occupied the absorption process into these shells is blocked. Simple mean-field calculations of a few-electron and single-hole configuration reasonably well described the energetic shifts of the inhomogeneously broadened absorption spectrum as a function of the number of carriers.[16] Detailed calculations that would address the role of electron-electron, electron-hole, and finite-hole mass on the absorption spectrum have not been carried out yet. Recent progress in optical spectroscopy of individual quantum dots[18–23] where spectra are no longer obscured by inhomogeneous broadening, warrants such a calculation.

In this work we calculate optical absorption spectra in charged lens-shaped self-assembled quantum dots. The single-particle levels of electrons and of holes form degenerate electronic shells. Electrons fill up the dot according to generalized Hund's rules.[15] We calculate absorption spectra using different approximate schemes and compare with exact calculations. We find that when an exciton is added to a partially filled shell it is resonant with many-electron and a hole configurations with the same energy. The resonant configurations are generated by a valence hole relaxation combined with excitation in the electron system. The interaction with resonant configurations breaks up the exciton and it ceases to be a well-defined quasiparticle. In this case the results of approximate calculations are not trustworthy. When the shell is full the absorption is blocked and only

absorption to higher empty shells is possible. Detailed calculations of the absorption spectra as a fingerprint of the number of carriers N establish interband spectroscopy as a sensitive probe of the number of electrons and the electronic structure of the dot.

II. SINGLE-PARTICLE STATES

Single-particle states in self-assembled quantum dots depend on growth conditions that control facetting, alloy composition, size, and shape. We focus here on a model of lens-shaped quantum dots that is sufficiently general to be applicable to a wide class of zero-dimensional systems. In lens-shaped self-assembled quantum dots the single-particle states $|n,m\rangle$ for electrons in the conduction band and holes in the valence band can be well approximated by those of a pair of harmonic oscillators with quantum numbers n and m.[24] The energy spectrum is given by $\varepsilon_{n,m}=\Omega(n+m+1)$. The single-particle states form degenerate shells with energy separation Ω whenever $n+m=0,1,2,\ldots$. Here $\Omega=\Omega_{e(h)}$ is the electronic shell spacing for electrons and for holes. We identify these shells as $s,p,d,e,f,\ldots$. The angular momentum of single particle states is $r^e=n_e-m_e$ and $r^h=-n_h+m_h$ for electrons and holes, respectively.

In the literature, different values for energies Ω_e and Ω_h for self-assembled dots have been reported. Reference 25 found $\Omega_e=12.4$ meV and $\Omega_h=3.7$ meV as good parameters to model their experiment. On the other hand, authors of Ref. 26 found $\Omega_e\sim 30$ meV and $\Omega_h\sim 15$ meV as being good effective parameters, while Ref. 27 reported $\Omega_e\sim 50$ meV and $\Omega_h\sim 27$ meV. Hence, depending on fabrication method and growth condition quantum dots show ratios of the valence- and conduction-band energy spacing $\Omega_h/\Omega_e\leqslant 1$.

III. MODEL HAMILTONIAN

The Hamiltonian describing the interband optical absorption process in modulation-doped self-assembled quantum dots involves many electrons and a single valence hole. Using composite indices $i=\{n_i,m_i\}$ to describe orbital quantum numbers and σ_i to describe spin, the Hamiltonian in second quantized notation reads

$$\mathcal{H}=\sum_{i\sigma_i}\varepsilon_i^e c_{i\sigma_i}^\dagger c_{i\sigma_i}+\sum_{i\sigma_i}\varepsilon_i^h h_{i\sigma_i}^\dagger h_{i\sigma_i}+\frac{1}{2}\sum_{i,j,k,l,\sigma_i,\sigma_j}\langle ij|v_{ee}|kl\rangle c_{i\sigma_i}^\dagger c_{j\sigma_j}^\dagger c_{k\sigma_j}c_{l\sigma_i}-\sum_{i,j,k,l,\sigma_i,\sigma_j}\langle ij|v_{eh}|kl\rangle c_{i\sigma_i}^\dagger h_{j\sigma_j}^\dagger h_{k\sigma_j}c_{l\sigma_i}. \quad (1)$$

The operators $c_{i\sigma_i}^\dagger(c_{i\sigma_i})$ and $h_{i\sigma_i}^\dagger(h_{i\sigma_i})$ create (anihilate) an electron and valence hole in the state $|i\sigma_i\rangle$ with single particle energy ε_i.

The first two terms on the right-hand side of Eq. (1) describe the kinetic energy of electrons and a single hole. The third term accounts for Coulomb interactions among electrons and the fourth term describes the interaction of electrons with the valence hole. The interactions are given by the two-particle Coulomb matrix elements $\langle i\sigma_i,j\sigma_j|v_{ee}|k\sigma_j,l\sigma_i\rangle$ for electron-electron and $\langle i\sigma_i,j\sigma_j|v_{eh}|k\sigma_j,l\sigma_i\rangle$ for electron-hole scattering.[28] These quantities are measured in units of $V_0=\sqrt{\pi}(a_B/l_{eff})Ry^*=\sqrt{\pi}(a_B\sqrt{2\Omega_e m_e^*})Ry^*$, with a_B the effective Bohr radius, Ry^* the effective Rydberg, and $l_{eff}=1/\sqrt{2\Omega_e m_e^*}$ the characteristic length scale of the SAD. Although the electron-electron and electron-hole Coulomb matrix elements depend on the detailed form of the single-particle states and the form of the interaction, the best starting point is the case of symmetric interactions: $\langle i,j|v_{ee}|k,l\rangle=\langle i,j|v_{hh}|k,l\rangle$ and $\langle i,j|v_{eh}|k,l\rangle=\langle i,k|v_{ee}|j,l\rangle$. As discussed in Ref. 29 one can choose the symmetric interactions characterized by a single value of V_0. Therefore, we are left with the smallest possible number of parameters in the theory, the ratio of Coulomb to electron kinetic energy V_0/Ω_e and the ratio of the hole kinetic energy to the kinetic energy of the electron Ω_h/Ω_e. Capacitance and photoluminescence experiments in InAs/GaAs quantum dots give the charging energy of the ground state as $E_C\approx V_0\approx 20$ meV (Ref. 27) and kinetic energy ratios $\Omega_h/\Omega_e\approx 0.5$.

IV. MANY-PARTICLE STATES AND HUND'S RULES IN QUANTUM DOTS

In the initial ground state the total spin of a partially filled shell is maximized while the total angular momentum is minimized in accordance with Hund's rule generalized to SAD. The generalized Hund's rule has been predicted and verified numerically in Ref. 15. We calculate the electron number dependence of the absorption spectra starting from a single configuration consistent with Hund's rules. The applicability of this approximation and breakdown of Hund's rule are tested by a detailed calculation of the absorption spectrum to a correlated $N=4$ ground state as a function of the strength of Coulomb interactions.

The interacting, final many-particle states $|\nu_f\rangle$ are expanded in the basis of the noninteracting configurations of $N+1$ electrons and a single valence hole: $|j;i_1,\ldots,i_{N+1}\rangle=h_j^\dagger c_{i_1}^\dagger\ldots c_{i_{N+1}}^\dagger t|\text{vac}\rangle$, where $|\text{vac}\rangle$ denotes vacuum. These configurations can be also written in a more physically transparent way by emphasizing the optically created electron-hole pair $h_i^\dagger c_i^\dagger$ and enumerating different many-particle processes:[14]

$$|\nu_f\rangle=\sum_i A_i^f h_i^\dagger c_i^\dagger|\nu_G\rangle+\sum_{j,k,l}B_{j,k,l}^f h_{j+k}^\dagger c_j^\dagger c_{l+k}^\dagger c_l|\nu_G\rangle+\cdots. \quad (2)$$

The first set of processes corresponds to injecting an optically active electron-hole pair to empty states $|i\rangle$ above the ground state with amplitude A_i^f. These processes correspond to absorption in the presence of a frozen Fermi sea (frozen ground state). The second series of processes corresponds to indirect electron-hole pairs $h_{j+k}^\dagger c_j^\dagger$ with momentum $-k$ coupled to what appears to be a one pair excitation $c_{l+k}^\dagger c_l$ with momentum $+k$. This one pair excitation is, however, coupled to an additional photoexcited electron. The photoexcited electron can be turned into a well-defined excitation of the $N+1$ electron system by adding an extra electron at the Fermi level $|F\rangle$. Redefining our ground state $|\nu_G'\rangle_{N+1}$

$=c_F^\dagger|v_G\rangle$ allows us to write our final states in terms of excitations of the $N+1$ electron state:

$$|v_f\rangle=\sum_i A_i^f h_i^\dagger(c_i^\dagger c_F)|v_G'\rangle_{N+1}$$

$$+\sum_{j,k,l} B_{j,k,l}^f h_{j+k}^\dagger(c_j^\dagger c_{l+k}^\dagger c_l c_F)|v_G\rangle+\cdots. \quad (3)$$

This way of defining our final states[14] emphasizes the fact that the photoexcited electron is an indistinguishable particle in the final $N+1$ electronic system. It enters as part of an excitation of the system. The first and the second contributions can now be viewed on the same footing as one pair and two pair excitations of the $N+1$ electron systems. The valence hole clearly entangles these excitations. Obviously there are many other excitations besides the ones listed here. In exact diagonalization of small systems all excited states are taken into account. The states are labeled by the total angular momentum R, the z component of the total spin of electrons S_z^i, and the hole's spin σ_h. The Hamiltonian (1) is diagonalized numerically in each eigensubspace (R,S_z^i,σ_h). Equation (3) and exact diagonalization helps us to identify important processes that play a role in extended systems.

V. OPTICAL ABSORPTION SPECTRUM

The absorption spectrum $A(\omega)$ of a photon with frequency ω due to transitions from an exact ground state $|v_G\rangle$ of a charged SAD to all final states $|v_f\rangle$ is given by the Fermi's golden rule:

$$A(\omega)=\sum_f |\langle v_f|\mathcal{P}^\dagger|v_G\rangle|^2\delta(\omega-[\mathcal{E}_f(N+1)-\mathcal{E}_G(N)]). \quad (4)$$

The summation is over many-body $N+1$ electron and one hole final states $|v_f\rangle$ with energy $\mathcal{E}_f(N+1)$. The ground state of the charged dot before absorption is $|v_G\rangle$ and its energy is $\mathcal{E}_G(N)$. The initial ground and final states are coupled through the interband polarization operator $\mathcal{P}^\dagger=\Sigma_{j\sigma}c_{j\sigma}^\dagger h_{j-\sigma}^\dagger$. The magnitude of this coupling is the oscillator strength of the optical transition between the ground and the final state. This operator adds an electron hole pair into the ground state of the system. It can be decomposed into two operators $\mathcal{P}_{+(-)}^\dagger=\Sigma_j c_{j\uparrow(\downarrow)}^\dagger h_{j\downarrow(\uparrow)}^\dagger$, with definite circular polarization of light: σ_+ or σ_-.

The final states $|v_f\rangle$ are written in the basis of the product of the $N+1$ electron states $|v_l\rangle$ with total angular momentum R_l^e and a single valence-hole state $h_s^\dagger|\text{vac}\rangle$ with angular momentum r_s^h. Addition of an electron-hole pair does not change the total angular momentum. Hence the angular momentum of the final state $R_l^e+r_s^h$ is equal to the angular momentum of the initial state R_G^e. Then, we can write $|v_f\rangle=\Sigma_{l,s;R_l^e+r_s^h=R_G^e}B_{l,s}^f h_{s\sigma_s}^\dagger|\text{vac}\rangle|v_l\rangle_{N+1}$. Amplitudes $B_{l,s}^f$ are determined by e-h scattering between these many-particle electron hole states. The optical absorption spectrum can now be expressed in terms of only electronic degrees of freedom as

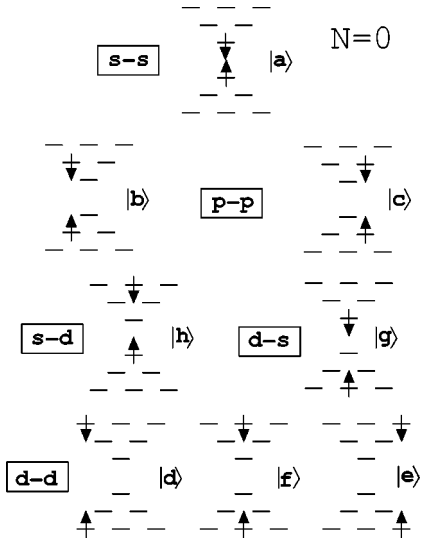

FIG. 1. All possible configurations of an electron-hole pair with total angular momentum $R=0$ on three lowest shells in the conduction and valence band.

$$A(\omega)=\sum_f\left|\sum_{ls} B_{ls}^f({}_{N+1}\langle v_l|c_{s\sigma_s}^\dagger|v_G\rangle_N)\right|^2$$
$$\times\delta(\omega-[\mathcal{E}_f(N+1)-\mathcal{E}_G(N)]). \quad (5)$$

We see that the oscillator strength of the optical transition has contributions coming from electronic configurations belonging to different electronic Hilbert spaces. This kind of interference effect is responsible for the Fermi edge singularity in the optical absorption of two-dimensional electron gases, even in the absence of e-e interactions.[5] In this work, we focus on the effect of electron-electron interactions on the absorption spectrum.

VI. RESULTS AND DISCUSSION

We now turn to the results of calculations of the absorption spectrum as a function of the number of carriers.

A. Excitonic absorption in undoped SAD

To understand the absorption spectra of charged SAD's, we must first understand the excitonic $(N=0)$ absorption case.[24] There are two classes of electron-hole pairs: bright and dark. To introduce these classes it is enough to consider only the three lowest s, p, and d shells with a total of six orbitals, as shown in Fig. 1. All calculations will be carried out in this simple model of SAD. In the optical absorption process only direct transitions are allowed, creating electron-hole pairs with zero angular momentum. However, an exciton is composed of all pairs with total angular momentum zero. In addition to the six optically active configurations there are two other configurations. They involve a hole (electron) in the zero angular momentum state $|00\rangle$ of the s shell and a hole (electron) in the zero angular momentum state $|11\rangle$ of the d shell.

Hence an exciton is composed of eight states with zero total angular momentum:

$$|a\rangle = h^{\dagger}_{00\uparrow} c^{\dagger}_{00\downarrow} |\mathrm{vac}\rangle,$$

$$|b\rangle = h^{\dagger}_{01\uparrow} c^{\dagger}_{01\downarrow} |\mathrm{vac}\rangle, \quad |c\rangle = h^{\dagger}_{10\uparrow} c^{\dagger}_{10\downarrow} |\mathrm{vac}\rangle,$$

$$|d\rangle = h^{\dagger}_{02\uparrow} c^{\dagger}_{02\downarrow} |\mathrm{vac}\rangle, \quad |e\rangle = h^{\dagger}_{20\uparrow} c^{\dagger}_{20\downarrow} |\mathrm{vac}\rangle, \tag{6}$$

$$|f\rangle = h^{\dagger}_{11\uparrow} c^{\dagger}_{11\downarrow} |\mathrm{vac}\rangle,$$

$$|g\rangle = h^{\dagger}_{11\uparrow} c^{\dagger}_{00\downarrow} |\mathrm{vac}\rangle, \quad |h\rangle = h^{\dagger}_{00\uparrow} c^{\dagger}_{11\downarrow} |\mathrm{vac}\rangle.$$

Figure 1 shows both the six optically active and two dark electron-hole configurations. In optically active configurations charge distribution of an electron and a hole is identical and they do not interact with other charges. Dark *s*-*d* and *d*-*s* configurations involve different orbitals of an electron and a hole. This leads to a spatial imbalance in the charge distribution of the electron and the hole. The repulsion of an electron by other electrons is not compensated by the attraction of the hole and these pairs interact with charges. In this sense we can call them charged electron-hole pairs. The energy of each dark configuration appears to be very different from any of the bright configurations even in the absence of *e*-*h* interaction. For example, the *p*-*p* configuration has kinetic energy $2\Omega_e + 2\Omega_h$ while the energy of the *s*-*d* configuration is $\Omega_e + 3\Omega_h$. However, when we form a new state $|H\rangle$ as a linear superposition of *s*-*d* and *d*-*s* configurations $|H\rangle = (1/\sqrt{2})(|g\rangle + |h\rangle)$ we generate a state with energy $\frac{1}{2}(\Omega_e + 3\Omega_h) + \frac{1}{2}(\Omega_h + 3\Omega_e) = 2\Omega_e + 2\Omega_h$. Even more surprisingly these configurations are degenerate when *e*-*e* and *e*-*h* interactions are switched on. This rather counterintuitive result implies that irrespective of the quantization of electron and hole kinetic energy the dark exciton is always degenerate with optically active transition involving electrons and holes on *p* orbitals. The interaction between these configurations will make both these states optically active and leads to a splitting of the exciton transition into the *p* shell without the breaking of geometrical symmetry.

To gain further insight into the structure of an exciton let us make a transformation into a basis of coherent exciton states for each shell.[30] The coherent exciton states are equivalent to Jacobi coordinates for two and three particles:

$$|A\rangle = |a\rangle, \tag{7}$$

$$|B\rangle = \frac{1}{\sqrt{2}}(|b\rangle + |c\rangle),$$

$$|C\rangle = \frac{1}{\sqrt{2}}(|b\rangle - |c\rangle), \tag{8}$$

$$|D\rangle = \frac{1}{\sqrt{3}}(|d\rangle + |e\rangle + |f\rangle),$$

$$|E\rangle = \frac{1}{\sqrt{2}}(|d\rangle - |e\rangle),$$

$$|F\rangle = \frac{1}{\sqrt{6}}(|d\rangle + |e\rangle - 2|f\rangle), \tag{9}$$

$$|H\rangle = \frac{1}{\sqrt{2}}(|g\rangle + |h\rangle),$$

$$|G\rangle = \frac{1}{\sqrt{2}}(|g\rangle - |h\rangle). \tag{10}$$

Only states $|A\rangle$, $|B\rangle$, and $|D\rangle$ have finite oscillator strength. This is seen by expanding the interband polarization operator in this new basis: $\mathcal{P}^{\dagger}|\mathrm{vac}\rangle = |a\rangle + |b\rangle + |c\rangle + |d\rangle + |e\rangle + |f\rangle = |A\rangle + \sqrt{2}|B\rangle + \sqrt{3}|D\rangle$. The three states can be identified with the three available shells within our model. State $|A\rangle$ corresponds to the *s* shell, state $|B\rangle$ to the *p* shell, and state $|D\rangle$ to the *d* shell. One can show that in Jacobi coordinates the exciton Hamiltonian separates into two blocks: one 5×5 block and one 3×3 block. The five coupled states involve the three optically active states: $|A\rangle$, $|B\rangle$, $|D\rangle$, and two dark states: $|F\rangle$ and $|H\rangle$. Denoting the kinetic energy of the *e*-*h* pair as $t = \Omega_e + \Omega_h$, the matrix representation of $\mathcal{H}_f$ in the subspace $\{|A\rangle, |B\rangle, |H\rangle, |D\rangle, |F\rangle\}$ is

$$\begin{pmatrix}
t - V_0 & -\sqrt{2}(0.25V_0) & +\sqrt{2}(0.25V_0) & \sqrt{2}(0.375V_0) & \sqrt{\frac{2}{3}}(0.0938V_0) \\
-\sqrt{2}(0.25V_0) & 2t - 0.875V_0 & -0.125V_0 & -\sqrt{\frac{2}{3}}(0.4688V_0) & -\frac{1}{\sqrt{3}}(0.1406V_0) \\
+\sqrt{2}(0.25V_0) & -0.125V_0 & 2t - 0.875V_0 & \sqrt{\frac{2}{3}}(0.0938V_0) & -\frac{1}{\sqrt{3}}(0.2344V_0) \\
\frac{1}{\sqrt{3}}(0.375V_0) & -\sqrt{\frac{2}{3}}(0.4688V_0) & \sqrt{\frac{2}{3}}(0.0938V_0) & 3t - 0.7969V_0 & \sqrt{2}(0.0117V_0) \\
\sqrt{\frac{2}{3}}(0.0938V_0) & -\frac{1}{\sqrt{3}}(0.1406V_0) & -\frac{1}{\sqrt{3}}(0.2344V_0) & \sqrt{2}(0.0117V_0) & 3t - 0.7715V_0
\end{pmatrix} \tag{11}$$

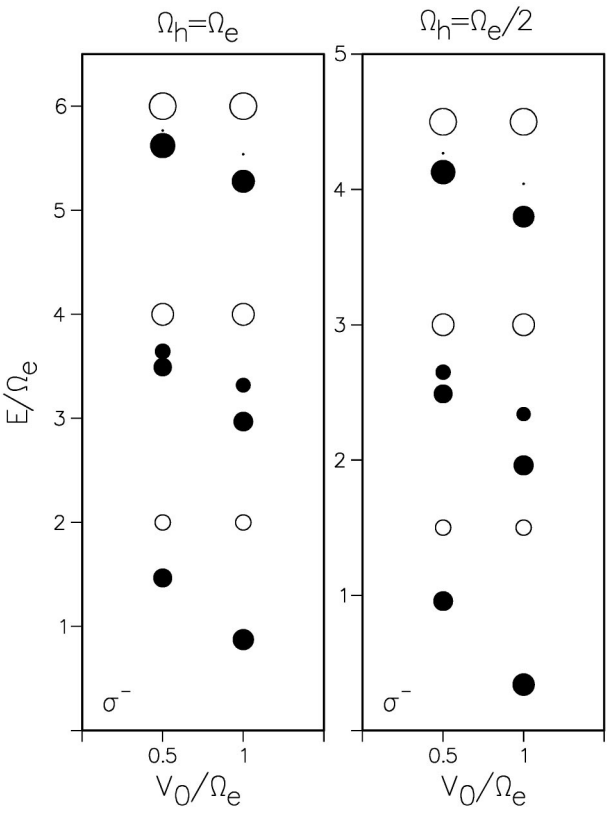

FIG. 2. Excitonic optical absorption in the SAD for different e-h interaction strength, and quantization of electron and hole energy.

We see immediately that there are two relevant states with kinetic energy $2t$ for a p shell and two relevant states with kinetic energy $3t$ for a d shell. The coupling of states in a p shell is an order of magnitude stronger than the coupling of states in a d shell. The d shell states do not include dark excitons. The five final optically active excitonic states $|\nu_f\rangle$ are written as a linear combination of the basis states: $|\nu_f\rangle = C_A^f|A\rangle + C_B^f|B\rangle + C_D^f|D\rangle + C_F^f|F\rangle + C_H^f|H\rangle$. Amplitudes C_i^f are the eigenstates of the exciton Hamiltonian (11). All five states contribute to the absorption spectrum $A(\omega) = \Sigma_f|C_A^f + \sqrt{2}C_B^f + \sqrt{3}C_C^f|^2\delta(\omega - E_f)$. The absorption spectrum $A(\omega)$ is shown in Fig. 2. Empty circles correspond to a noninteracting and dark circles to an interacting exciton. The different spectra were calculated for different SAD parameters: $\Omega_h/\Omega_e = \{1, 0.5\}$ and different e-h interaction strengths $V_0 = \{1, 0.5\}\Omega_e$. In the absence of interactions the transition energies are equally spaced in units of $\Omega_h + \Omega_e = 2\Omega_e$. The oscillator strength of the spectral lines increases linearly with the photon energy, reproducing the joint density of states of an empty dot. The inclusion of the e-h scattering changes the absorption spectrum. The attractive electron-hole interaction redshifts transition energies and leads to additional structures in the spectrum. The oscillator strength in the p-shell absorption is almost equally distributed between the p-p-like transition $|B\rangle$ and the s-d-like transition $|H\rangle$. We wish to emphasize that the splitting of the p-shell transition is an exciton feature not related to the splitting of single particle levels but to the presence of higher-energy shells.

Absorption into the d shell is also split into two lines, one strong and one weak. The ratio of the oscillator strength of this transitions is due to the weak coupling between the coherent exciton state $|D\rangle$ and state $|F\rangle$. If a higher energy shell e was included the d shell transitions would be further split due to dark p-e transitions. To conclude this section, the optical excitonic absorption of an empty SAD is strongly modified by the combined effect of the shell structure of the conduction and valence band, and the interaction between excitonic configurations.

B. Absorption in charged SAD-frozen ground-state approximation

We now move to the optical absorption spectrum as a function of the number of electrons N. We focus mainly on σ_- polarization of light, the σ_+ calculations can be done following the same procedure. The σ_- polarization is chosen to inject electrons with spin down, parallel to the arbitrarily chosen spin-down polarization of the ground state. Warburton *et al.*[16] calculated absorption spectra by assuming that the initial state is given by a single electronic configuration. The final state consisted also of a single $N+1$ electron and a valence-hole configuration. The next level of approximation often used in the study of absorption in semiconductors is to include the electron-hole scattering of the photoexcited electron-hole pair in the presence of a frozen Fermi sea. The electrons in the Fermi sea block the states available to the photoexcited pair. This results in a bound state below the Fermi level, the so-called ''Mahan exciton.'' We, therefore, start our investigation of the carrier dependence of the absorption process by approximating the ground state by a single configuration consistent with the generalized Hund's rules. The final states consist of configurations corresponding to a photoinjected electron-hole pair and a frozen ground state. This frozen ground-state approximation (FGSA) is tested against exact diagonalization studies in the last section.

We start with just one electron $N=1$ in the SAD. The ground state $|\nu_G\rangle = c_{00,\downarrow}^\dagger|\text{vac}\rangle$ corresponds to an electron in the lowest single-particle level $|00\rangle$, with spin down. The energy, the total spin projection, and the angular momentum of the ground state are $\mathcal{E}_G(1) = \Omega_e$, $S_z = -1/2$, and $R=0$, respectively. In the σ_- polarization we inject an electron with spin parallel to the spin of the electron in the ground state. We construct final states of two electrons and a hole by keeping one of the electrons in the ground state $|00\rangle$. To make contact with low-lying bright and dark exciton states described previously and with the notation used in the expansion of the final state wave function, let us write our wave functions as

$$|b\rangle = h_{01,\downarrow}^\dagger c_{01,\uparrow}^\dagger|\nu_G\rangle, \quad |c\rangle = h_{10,\downarrow}^\dagger c_{10,\uparrow}^\dagger|\nu_G\rangle,$$
$$|d\rangle = h_{02\uparrow}^\dagger c_{02\downarrow}^\dagger|\nu_G\rangle, \quad |e\rangle = h_{20\uparrow}^\dagger c_{20\downarrow}^\dagger|\nu_G\rangle,$$
$$|f\rangle = h_{11\uparrow}^\dagger c_{11\downarrow}^\dagger|\nu_G\rangle, \tag{12}$$
$$|h\rangle = h_{00,\downarrow}^\dagger c_{11,\uparrow}^\dagger|\nu_G\rangle = h_{00,\downarrow}^\dagger c_{00,\uparrow}^\dagger(c_{11,\uparrow}^\dagger c_{00,\uparrow})|\nu_G\rangle.$$

A comparison with states of an exciton shows that the state $|a\rangle$ is missing due to the Pauli exclusion principle and absorption into the s shell is blocked. The two transitions $|b\rangle$

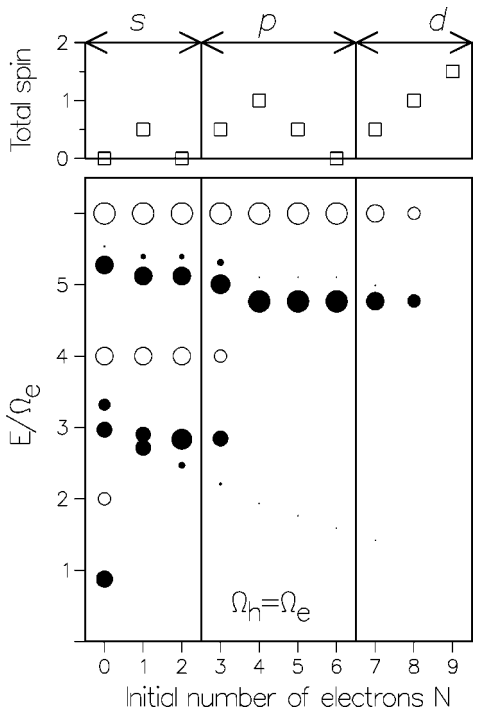

FIG. 3. Optical absorption spectra of the self-assembled quantum dot calculated within the frozen ground-state approximation, as a function of the number of electrons in the dot. The total spin of the ground state as a function of the number of electrons is indicated. σ_- polarization.

and $|c\rangle$ into the p shell, and transitions $|d\rangle$, $|e\rangle$, and $|f\rangle$ into the d shell are present. In addition, there is a dark configuration $|h\rangle$, but no configuration $|g\rangle$, which is now identical to configuration $|e\rangle$. Configuration $|h\rangle$ can be also viewed as excitation of electron-hole pairs with angular momentum compensated by the excitation from the ground state. This configuration can no longer be combined with configuration $|g\rangle$ to form a resonant configuration $|H\rangle$, and its energy and oscillator strength are sensitive to the presence of an electron. The calculation proceeds in a similar fashion to the calculation of an exciton. We define coherent exciton coordinates and diagonalize the Hamiltonian in this basis. The p-shell configurations $|B\rangle$ and $|H\rangle$ mix and the p-shell absorption peak splits into two peaks. The $N=1$ absorption spectrum is shown in Fig. 3. The oscillator strength of the two peaks depends on the ratio of Ω_h/Ω_e.

Let us now increase the number of electrons to $N=2$. The ground state $|\nu_G\rangle$ is a doubly occupied $|00\rangle$ orbital $|\nu_G\rangle = c^\dagger_{00,\uparrow}c^\dagger_{00,\downarrow}|\text{vac}\rangle$. The total angular momentum is $R=0$ and the total spin projection is $S_z=0$. The final state configurations are the same as in the previous case $N=1$. The only difference is the charge of the ground state. Because the dark configuration $|h\rangle$ contains an electron and a hole in different orbitals, its energy is sensitive to the charge in the dot. The bright configuration $|B\rangle$ is not sensitive to the charge so the energies of two configurations become different, and the two configurations are no longer in resonance. The oscillator strength of optical transitions in the p shell is much stronger than the dark one, as shown in Fig. 3. Absorption in the d shell resembles the case of $N=1$.

Charging the dot with $N=3$ electrons leads to the population of the p shell with one electron. We can chose the ground state to be $|\nu_G\rangle = c^\dagger_{10,\downarrow}c^\dagger_{00,\uparrow}c^\dagger_{00,\downarrow}|\text{vac}\rangle$, with total angular momentum and total spin projection: $R=1$ and $S_z=-1/2$. The one state of the p shell is occupied and the photoexcited electron has only one state to go into. There is no mixing of final state configurations by e-h scattering. In the absence of an electron, the mixing lowered the energy of the p-shell exciton. The lack of e-h scattering is, however, compensated by the exchange of the photoexcited electron with the electron already present in a partially filled shell. The compensation leads to the energy of the absorption line being insensitive to the filling of the shell, in contrast to mean-field results.[16] The second line due to the dark s-d exciton configuration has a very small oscillator strength because the dark exciton energy is far from resonance with the bright exciton configuration. The same principles operate for a larger number of electrons. Figure 3 shows the total spin of the ground state as a function of the number of electrons and calculated absorption spectra in the frozen ground state approximation for up to $N=12$ electrons. These calculations illustrate the blocking of phase space available to transitions, the renormalization of energy due to direct and exchange interactions, and the cancellation of excitonic and band-gap renormalization effects due to phase space filling in partially filled shells.

C. Correlation effects in the optical absorption spectra

Optical absorption spectra obtained so far were calculated in the FGSA, which assumes that an exciton remains a well-defined quasiparticle in the presence of extra electrons. The results capture some of the effects like energy renormalization, phase space blocking, and phase space filling, and appear interesting and convincing. Unfortunately, we will show that the concept of an exciton as a well-defined quasiparticle fails in the interacting electron system. In this section we discuss the effect of electron-electron and electron-hole interactions, and the entangling of electronic configurations by the mobile valence hole beyond the FGSA. We first study the regime of strong confinement where the ground state is very well approximated by the single configuration consistent with Hund's rules. In the second part we study the breakdown of Hund's rules and its manifestation in the absorption spectra.

1. Breakdown of FGSA

We start to relax the requirement of having an electron in the frozen ground state in the simplest case of $N=1$ electron. The ground state $|\nu_G\rangle = c^\dagger_{00,\downarrow}|\text{vac}\rangle$ corresponds to an electron in the lowest single-particle level $|00\rangle$, with spin down. The angular momentum of the ground state is $R=0$. We construct the two electron and one hole states of a charged exciton as products of electron and hole occupied states with total angular momentum $R=0$. There is a total of 16 states. We list them and their kinetic energies measured from the noninteracting s-s transition in Table I.

All states separate into groups corresponding to main optical transitions with energies $n\Omega_e+n\Omega_h$ plus transitions to excited electronic states $n\Omega_e+n\Omega_h+2\Omega_e$, which form replicas of main transitions. Because the s shell is occupied there are no transitions to the s shell. For $\Omega_e=\Omega_h$ transitions

TABLE I. Configurations of two electrons and one hole.

R_e	R_h	2 Electron states	1 Hole states	Energy	Energy for $\Omega_e=\Omega_h$
0	0	$\|11,00\rangle$	$\|00\rangle$	$2\Omega_e$	2
0	0	$\|10,01\rangle$	$\|00\rangle$	$2\Omega_e$	2
0	0	$\|20,02\rangle$	$\|00\rangle$	$4\Omega_e$	4
0	0	$\|11,00\rangle$	$\|11\rangle$	$2\Omega_e+2\Omega_h$	4
0	0	$\|10,01\rangle$	$\|11\rangle$	$2\Omega_e+2\Omega_h$	4
0	0	$\|20,02\rangle$	$\|11\rangle$	$4\Omega_e+2\Omega_h$	6
1	−1	$\|10,00\rangle$	$\|10\rangle$	$1\Omega_e+1\Omega_h$	2
1	−1	$\|10,11\rangle$	$\|10\rangle$	$3\Omega_e+1\Omega_h$	4
1	−1	$\|01,20\rangle$	$\|10\rangle$	$3\Omega_e+1\Omega_h$	4
−1	1	$\|01,00\rangle$	$\|01\rangle$	$1\Omega_e+1\Omega_h$	2
−1	1	$\|01,11\rangle$	$\|01\rangle$	$3\Omega_e+1\Omega_h$	4
−1	1	$\|10,02\rangle$	$\|01\rangle$	$3\Omega_e+1\Omega_h$	4
2	−2	$\|20,00\rangle$	$\|20\rangle$	$2\Omega_e+2\Omega_h$	4
2	−2	$\|20,11\rangle$	$\|20\rangle$	$4\Omega_e+2\Omega_h$	6
−2	2	$\|02,00\rangle$	$\|02\rangle$	$2\Omega_e+2\Omega_h$	4
−2	2	$\|02,11\rangle$	$\|02\rangle$	$4\Omega_e+2\Omega_h$	6

to the p shell with energy $2\Omega_e$ are fourfold degenerate. To make contact with low-lying states described previously in the FGSA we write the fourfold degenerate subspace as

$$|b\rangle=h^{\dagger}_{01,\uparrow}c^{\dagger}_{01,\downarrow}|v_G\rangle,\quad |c\rangle=h^{\dagger}_{10,\uparrow}c^{\dagger}_{10,\downarrow}|v_G\rangle,$$
$$|h\rangle=h^{\dagger}_{00,\uparrow}c^{\dagger}_{00,\downarrow}(c^{\dagger}_{11,\uparrow}c_{00,\uparrow})|v_G\rangle, \tag{13}$$
$$|i\rangle=h^{\dagger}_{00,\uparrow}c^{\dagger}_{10,\downarrow}(c^{\dagger}_{01,\uparrow}c_{00,\uparrow})|v_G\rangle$$

These configurations are shown in Fig. 4. A comparison with states of an exciton shows that the state $|a\rangle$ is missing due to the Pauli exclusion principle and absorption into the s shell is blocked. The two transitions $|b\rangle$ and $|c\rangle$ into the p shell are present. There is a dark configuration $|h\rangle$ that has an electron frozen in the ground state. However, a new configuration $|i\rangle$ appears. This new configuration does not have an electron in the initial ground state and was not included in the FGSA. The configurations $|h\rangle$ and $|i\rangle$ can be viewed as indirect excitation of electron-hole pairs with angular momentum compensated by the excitation from the ground state. Neither configuration $|h\rangle$ nor $|i\rangle$ is optically active. However, through e-e and e-h interactions the optically active configuration $|B\rangle=(1/\sqrt{2})(|b\rangle+|c\rangle)$ acquires a spectral weight at energies corresponding to all three configurations. For $\Omega_e=\Omega_h$ the oscillator strength is divided among only two configurations and the absorption peak is split into two main peaks. The spectrum, shown in Fig. 5, looks similar to the spectrum of an exciton and to the spectrum in FGSA. However, its dependence on the ratio of kinetic energies and on interaction strength is quite different. For example, the oscillator strength of each of the three transitions is independent of the strength of Coulomb interactions but does depend on the ratio of Ω_e/Ω_h.

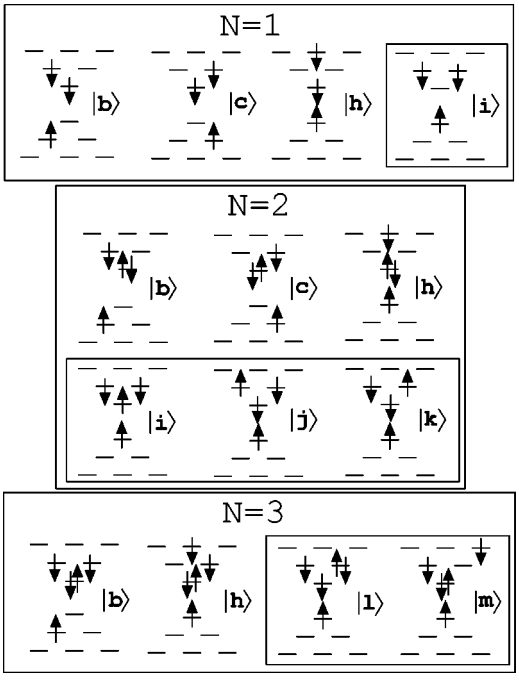

FIG. 4. Final-state configurations for $N=1,2,3$ electrons and a photoexcited electron and valence hole. Configurations shown in boxes are beyond the frozen ground-state approximation. σ_- polarization.

We follow the same procedure for $N=2,3,4$. The relevant low-lying configurations for $N=1-3$ are shown in Fig. 4. In each case, new configurations clearly marked with a box in Fig. 4, appear. These configurations involve electrons excited from the initial state, and were not included in FGSA. A closer inspection of these new configurations reveals a simple principle. We excite an optically active electron-hole pair, e.g., p-p. We next search for all possible states that have the same energy and total momentum as the optically excited one. These pairs can be generated by relaxing the valence hole to the lower-energy state while simultaneuosly exciting an electron that absorbs both the energy and momentum. Due to many electronic degrees of freedom there can be many states that satisfy this condition. For $N=2$ we find a band of $n=4$ additional states, with hole relaxed to the top of the valence band, as shown in Fig. 4. The states $|b\rangle$, $|c\rangle$, $|h\rangle$, and $|i\rangle$ are familiar from the $N=1$ case. The new states

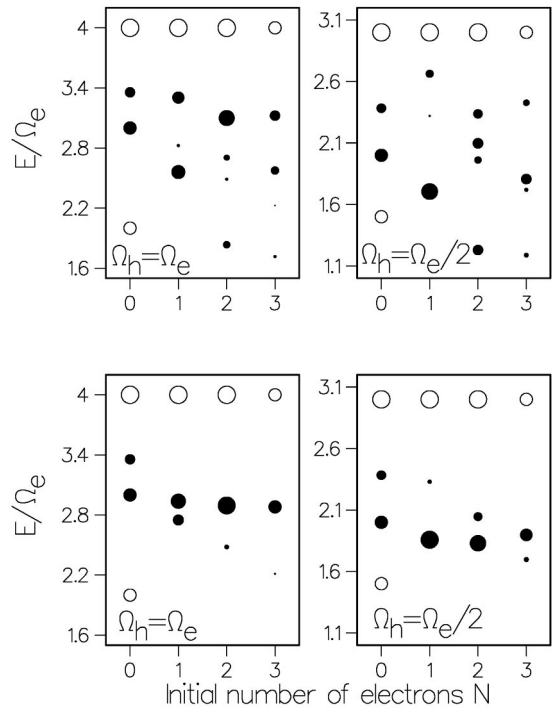

FIG. 5. (a) Exact and (b) FGSA absorption spectrum for SAD with $N=1,2,3$ electrons for different values of kinetic energy for holes and for electrons. σ_- polarization.

$|j\rangle$ and $|k\rangle$ are obtained by rotating the minority spin electron in state $|i\rangle$ among the three orbitals.[17] The single optically active configuration interacts strongly with the band of states and the absorption spectrum breaks into a band of peaks. The interaction of this band of dark states with bright ones, and the absorption spectrum, depends strongly on the ratio Ω_e/Ω_h but is independent of the strength of Coulomb interactions. The calculated absorption spectra are shown in Fig. 5 for two representative values of Ω_h. The same analysis follows for $N=3$ for which resonant configurations are shown in Fig. 4 and absorption spectra in Fig. 5. The same principles apply to higher number of electrons. The $N=4$ spectra are discussed in the next section.

The conclusion from these calculations is that neither the mean field nor the FGSA works in the description of the absorption spectra. This is because the photoexcited electron-hole pair is resonant with many electronic configurations and an exciton is no longer a well-defined quasiparticle.

2. Correlations in the initial state

Here we study the effect of correlations in the initial state and the effect of the breakdown of Hund's rules on the absorption spectra. The ground state of the N electron system $|v_G\rangle$ is a linear combination of many-body states belonging to the same Hilbert space. The Hilbert space is determined by the total angular momentum $R^e_{N,H}$ of the lowest kinetic energy configuration compatible with Hund's rules. This leads to: $|v_G\rangle=\Sigma_{i;R^e_i=R^e_{N,H}}A^G_i|v_i\rangle_N$. As we can see, all the excited configurations have finite amplitudes A^G_i determined by the *e-e* interaction.

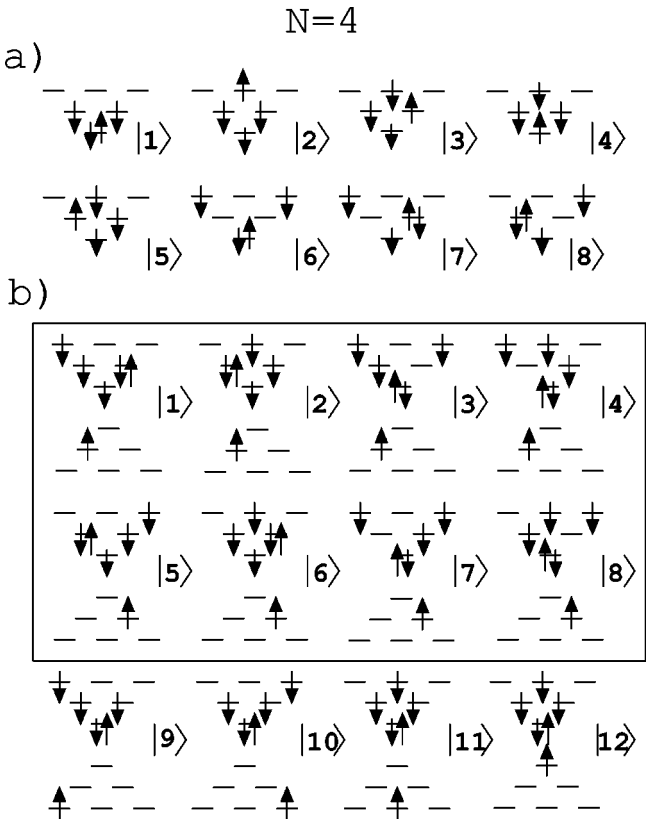

FIG. 6. (a) Electronic configurations for $N=4$ electrons in the lowest kinetic energy state compatible with Hund's rule and low-lying excitations. (b) Low-lying states for $N+1=5$ electrons plus a valence-hole.

The absorption spectrum [Eq. (5)] is modified due to the entanglement of the electronic configurations in the intial state, namely,

$$A(\omega)=\sum_f\left|\sum_{lsi}B^f_{ls}A^G_i\ {}_{N+1}\langle l|c^\dagger_{s\sigma_s}|i\rangle_N\right|^2 \times\delta(\omega-[\mathcal{E}_f(N+1)-\mathcal{E}_G(N)]). \quad (14)$$

The oscillator strength of the optical transitions has contributions coming from all the possible many-electron configurations in the initial state. Hence many of the final states, which do not couple to the single configuration given by the Hund's rules, now have a finite overlap with the correlated electron state. To the interference effects due to *e-h* scattering in the transition matrix element we need to add the interference effects coming from the *e-e* entanglement of configurations in the initial state.

The first application of Hund's rules takes place for $N=4$. We, therefore, illustrate the effect of correlations in the initial state by calculating the absorption spectrum for a partially polarized p shell $N=4$. The lowest kinetic energy state with energy $t=6\Omega_e$, and the first excited states, having an excess of kinetic energy equal to $\Delta=2\Omega_e$, are both shown in Fig. 6(a). Excited $N=4$ electronic states separate into two groups: (i) four configurations $2-5$ with singly occupied orbitals, and (ii) three configurations $6-8$ with doubly occupied orbitals. Configurations in group (i) differ only by the position of the spin-up electron. Alternatively, we can think

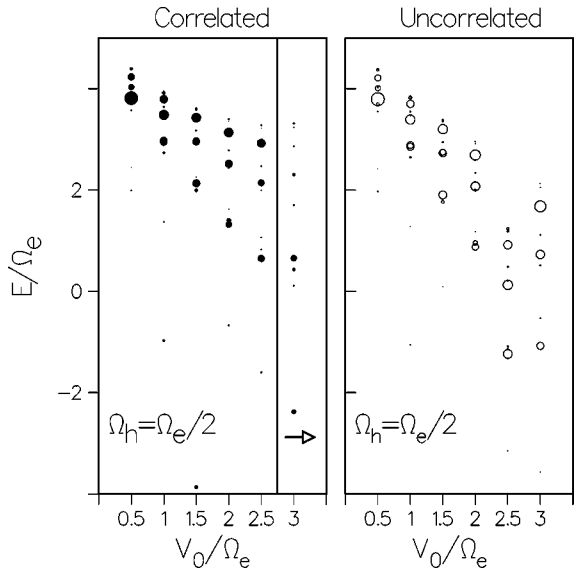

FIG. 7. Optical absorption for $N=4$ electrons in the SAD with σ_- polarized light. Left: correlated initial state. Right: ground state given by a single configuration. σ_- polarization.

of these states as generated by the hopping of the spin-up electron on a four orbital plaquette.

The Hamiltonian of $N=4$ interacting electrons was diagonalized within the Hilbert space shown in Fig. 6 for different ratios of Coulomb to kinetic energy V_0/Ω_e. The ground state for $V_0/\Omega_e=0.5$ is very well approximated by the lowest kinetic energy configuration $|1\rangle$ with $|A_1^G|^2=0.985$. Increasing the strength of Coulomb interactions to $V_0/\Omega_e=1$ gives $|A_1^G|^2=0.895$. Configurations $|2\rangle$, $|7\rangle$, and $|8\rangle$ have a small contribution to the ground state: $|A_2^G|^2=0.011$, $|A_7^G|^2=0.042$, and $|A_8^G|^2=0.042$. Increasing the value of Coulomb interactions penalizes doubly occupied configurations and favors singly occupied configurations. The quadruplet configuration composed of configurations $|2\rangle$–$|5\rangle$ becomes the lowest energy configuration for $V_0>2.5\Omega_e$.

We now turn to the absorption spectrum. In Fig. 6(b) we show all final-state configurations built from configurations shown in Fig. 6(a) by the addition of an electron-hole pair. These configurations can be viewed as excitations of the ground state in the presence of the photoexcited electron-hole pair. They are allowed because valence hole recoils and absorbs energy and momentum. In Fig. 6 state $|1\rangle$ can be obtained from the optically created configuration $|9\rangle$, exciting a spin-up electron from state $|00\rangle$ to state $|10\rangle$. This new configuration corresponds to an electron-hole pair excitation of the ground state coupled with the relaxation of valence hole. All other states can be regarded as one electron-hole pair excitations: $|3\rangle$ being an excitation of state $|9\rangle$; $|2\rangle$, $|4\rangle$, and $|6\rangle$ are excited states of configuration $|11\rangle$; $|5\rangle$, $|7\rangle$, and $|8\rangle$ are excited states of $|10\rangle$.

Figure 7 shows the optical absorption spectrum obtained for correlated ground state (left panel) for different values of the ratio V_0/Ω_e. For comparison, the optical absorption neglecting correlations are also shown in the right panel. The absorption spectrum for a correlated ground state shows a band of peaks corresponding to initial and final states in Fig. 6. New optical transitions appear due to the interference effects in the optical matrix element. These transitions slightly broaden main peaks. Up to $V_0/\Omega_e \leqslant 2.5$, the effects of correlations is small. The ground state is dominated by configuration $|1\rangle$ consistent with Hund's rules and neglecting correlations in the ground state does not lead to significant changes in the absorption spectrum.

For $V_0/\Omega_e>2.5$ the ground state is no longer described by Hund's rules and is a quadruplet. The absorption spectrum changes drastically at this point to reflect the changes in the ground state.

VII. SUMMARY

We investigated the effects of electron-electron interactions on excitonic absorption in charged, lens-shaped, SAD. The electronic shells of SAD were filled with N electrons according to generalized Hund's rules. In absorption, an exciton was added to the electronic system. The electronic part of the exciton coupled in a nontrivial way to resonant electronic configurations through direct and exchange interaction. The mobile valence hole contributed through the entanglement of many-electron configurations. For partially filled shells the valence hole relaxation coupled with spin related excitations of the electronic system led to a breakdown of exciton as a meaningful quasiparticle.

The resulting complex absorption spectra are specific to the number of electrons and their interaction in a quantum dot.

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