Spin characteristics of excitonic absorption in multiply charged quantum dots

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The excitonic absorption spectra of negatively charged self-assembled quantum dots as a function of resident electron number (N_e=0–5) are theoretically studied using the configuration interaction method. Spin Pauli blockade, Coulomb (exchange and correlation) interactions, and symmetry of dot shape are identified as the three main underlying mechanisms, sensitively depending on N_e, in the spin characteristic spectra. In particular, we predict that polarized absorption in triply charged dots exhibits anomalous spin-dependent features, resulting from the intrinsic correlations in charged exciton X^{-3}.

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I. INTRODUCTION

Spin in semiconductor quantum dots has drawn a great deal of attention for years in both applied and fundamental research. The preparation, storage, manipulation, and detection of electron spin in charged quantum dots (QDs) have been achieved only recently, relying on the spin-selective optical transitions involving both purely electronic states of interacting few electrons and excitonic states of charged excitons.[1–4] For interacting few electrons (neutral excitons) in QDs, Hund's rules (hidden symmetries) have been confirmed as the underlying principle of the electronic (excitonic) ground states.[5,6] Furthermore, recent advances in bias control of self-assembled quantum dots (SAQDs) have made it possible to unambiguously prepare and probe *multiply* charged excitons in individual SAQDs.[7–16] Rich physics has been revealed in the measured emission spectra for negatively charged excitons X^0–X^{-5} and the spin-selective absorption for X^0–X^{-2} in SAQDs.[7,15,16] The experimental results give significant implications for spin-based applications and stimulate great interest in the spin and optical properties of charge-tunable SAQDs.

In this work, we theoretically study the absorption spectra of multiply charged SAQDs as a function of charging electron number N_e=0–5 filling up to the p shell by using the configuration interaction (CI) method.[17] The calculated results account for the recently measured photoluminescence excitation spectra of singly charged SAQDs[16] and predict the spin and optical properties of multiply charged SAQDs. We find that the absorption spectra exhibit remarkable spin characteristics, sensitively depending on the number of charging electrons, and spin Pauli blockade, Coulomb (exchange and correlation) interaction, and symmetry of dot shape are identified as the three main underlying mechanisms.

II. MODEL AND THEORY

For lens-shaped deformed SAQDs, the confining potential can be modeled by a two-dimensional anisotropic parabola, $V_{as}^{\beta}(x,y)=\frac{m_{\beta}^{*}}{2}[(\omega_x^{\beta})^2x^2+(\omega_y^{\beta})^2y^2]$, in terms of the two oscillator frequencies ω_x^{β} and ω_y^{β}, where $\beta=e/h$ denotes the kind of particle (electron or hole) and m_{β}^{*} the effective mass of particle.[18] The asymmetric potential can be decomposed into two parts, $V_{as}=V_s+\delta V$, i.e., the circularly symmetric potential $V_s^{\beta}=\frac{1}{2}m_{\beta}^{*}\bar{\omega}_{\beta}^2(1+\gamma_{\beta}^2)r^2$ characterized by the averaged oscillator frequency $\bar{\omega}_{\beta}\equiv(\omega_x^{\beta}+\omega_y^{\beta})/2$ and the deformation potential given by $\delta V^{\beta}(x,y)=m_{\beta}^{*}\bar{\omega}_{\beta}^2\gamma_{\beta}(x^2-y^2)$, where the parameter $\gamma_{\beta}\equiv(\omega_x^{\beta}-\omega_y^{\beta})/(\omega_x^{\beta}+\omega_y^{\beta})$ is defined to measure the extent of deformation. Throughout this paper, we consider $\gamma_e=\gamma_h=\gamma$. Without deformation (γ=0), the single-particle (SP) spectrum of a symmetric dot is the Fock-Darwin (FD) spectrum, described by $E_{nm\chi}^{\beta}=\hbar\bar{\omega}_{\beta}(n+m+1)$ for zero magnetic field, where $n,m=0,1,2,\ldots$ and $\chi=\uparrow/\downarrow$ denotes particle spin. We approximate the valence hole states of the strained SAQDs to the pure heavy-hole ones with spin projection $j_z=\pm 3/2$.[19] The angular momentum of an electron (a valence hole) in the FD state (n,m) is given by $L_{nm}^{e}=m-n$ ($L_{nm}^{h}=n-m$).

In the Fock-Darwin basis, we write the interacting e-h Hamiltonian for a charged SAQD subject to shape deformation as

$$H=\sum_i E_i^e c_i^{+}c_i+\sum_i E_i^h h_i^{+}h_i-\sum_{ijkl} V_{ijkl}^{e\text{-}h}c_i^{+}h_j^{+}h_k c_l \tag{1}$$

$$+\frac{1}{2}\sum_{ijkl} V_{ijkl}^{e\text{-}e}c_i^{+}c_j^{+}c_k c_l+\frac{1}{2}\sum_{ijkl} V_{ijkl}^{h\text{-}h}h_i^{+}h_j^{+}h_k h_l \tag{2}$$

$$+\sum_{i',i}\Delta_{i',i}^{e}c_{i'}^{+}c_i+\sum_{i',i}\Delta_{i',i}^{h}h_{i'}^{+}h_i, \tag{3}$$

where i,j,k,l are composite indices of FD SP states, c_i^{+} (h_i^{+}) and c_i (h_i) the operators of electron (hole) creation and annihilation operators, respectively, and $V_{ijkl}^{\beta\beta'}$ the Coulomb matrix elements.[17] The last two terms arise from dot shape deformation, with the matrix elements $\Delta_{i',i}^{\beta}\equiv\int d\vec{r}\psi_{i'}^{\beta}(\vec{r})^{*}\delta V(\vec{r})\psi_i^{\beta}(\vec{r})$. Because of the quadratic form of $\delta V(\vec{r})$, $\Delta_{i',i}\neq 0$ only if $L_i-L_{i'}=0,\pm 2$. In this work, we focus on the spin characteristics of the absorption spectrum of self-assembled QDs, typically resolvable at meV energy scale in experiments, as the fingerprint of the spin states of interacting charged excitons. We thus neglect the spin interaction mechanisms in SAQDs with a too weak relevant energy scale ($\ll$meV), such as the spin-orbit and the hyperfine interactions.[2,20–22] The extended states of wetting layers are not taken into account in calculations because the bound state transitions for InGsAs/GaAs SAQDs considered in this

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work are usually below the edge of the ones involving the extended states by some tens of meV.[23,24] The model based on the simplified electron-hole picture has been widely used and successfully accounted for many experiments for the studies of excitons in SAQDs.[25] Throughout this work, we take the following parameters for InAs SAQDs: $m_e=0.05m_0$, $m_h=0.15m_0$, and $\omega_e=3\omega_h=3\pi\mathrm{Ry}^*$, where $\mathrm{Ry}^*\approx 3$ meV is the effective Rydberg for InAs.

In numerical implementations of the CI method, we first generate all possible configurations of N_e electrons or X^{-N_e} charged exciton based on the three lowest shells, classified by total angular momentum L, projection of total electron spin S_z, and projection of hole spin j_z, and then carry out diagonalization for the Hamiltonian matrix.[17] The spectrum of polarized absorption in a dot charged with N_e electrons is evaluated by using Fermi's golden rule,[26] $I_\pm(\omega)=\Sigma_f\Sigma_{i\in GS's}|\langle X^{-N_e};f|P_\pm^+|N_e;i\rangle|^2\delta(\hbar\omega-E_f+E_i)$, where the interband polarization operator P_+^+ (P_-^+) for σ^+ (σ^-) circularly polarized light is defined as $P_\pm^+\equiv\Sigma_{nm}h^+_{nmj_z=\pm 3/2}c^+_{nms_z=\mp 1/2}$, creating a spin exciton with the projection of total spin angular momentum $M_z\equiv s_z+j_z=+1$ ($M_z=-1$) and coupling the charged dot from an N_e electron initial state $|N_e;i\rangle$ to some excited state of charged exciton $|X^{-N_e};f\rangle$. Charged dots are assumed to be stable in the N_e-electron ground states (GSs) prior to photoexcitation. In the following, we set the circular polarization of light to the σ^+ direction and analyze the polarized absorption spectra for different possible spins of the GSs of charged dots.

III. RESULTS AND ANALYSIS

Figures 1 and 2 show the calculated σ^+ absorption spectra of symmetric charged dots with electrons $N_e=0-5$. Figure 1(a) shows the absorption spectrum of a neutral dot ($N_e=0$), characterized with three absorption lines, i.e., two main lines at $E\sim 0.51t$ and $E\sim 1.62t$ [$t\equiv\hbar(\omega_e+\omega_h)$] corresponding to the bright s-s and p-p transitions, respectively, and a weak line at $E\sim 1.2t$ corresponding to the transition involving the dark configuration $h^+_{d0\uparrow}c^+_{s\downarrow}|0\rangle$, slightly coupled to the bright one $h^+_{p^\pm\uparrow}c^+_{p^\pm\downarrow}|0\rangle$ via e-h Coulomb scatterings.[26]

The polarized absorption starts to exhibit some dependences on the spin states of a dot as it is singly charged ($N_e=1$) [see Fig. 1(b)]. First, due to spin blockade (SB), the σ^+ absorption line of the s-shell transition appears only for the dot charged with a spin-up $\left(s_z=+\frac{1}{2}\right)$ electron at $E\sim 0.46t$.[27] While the spectrum of the p-shell absorption in the dot with $s_z=-\frac{1}{2}$ electron is composed of a sole p-shell line at $E\sim 1.49t$, the spectrum for the dot with $s_z=+\frac{1}{2}$ electron is found to contain three main lines. In the former case, a σ^+ absorption creates two electronic spin triplet (T) X^{-1} configurations, $|X^-;t_a\rangle=h^+_{p^-,\uparrow}c^+_{p^-,\downarrow}c^+_{s,\downarrow}|0\rangle$ and $|X^-;t_b\rangle=h^+_{p^+,\uparrow}c^+_{p^+,\downarrow}c^+_{s,\downarrow}|0\rangle$. Via the e-h interaction $V^{pp,x}_{e\text{-}h}\equiv V^{e\text{-}h}_{p^\pm p^\mp p^\mp p^\pm}$, the two coupled configurations generate a bright trion state of electronic spin triplet $|X^-;T_{-1}\rangle=(|t_a\rangle+|t_b\rangle)/\sqrt{2}$ with eigenenergy $E(X^{-1};T_{-1})=E^{1e}_{GS}+(E^X_p-V^{pp,x}_{e\text{-}h}-V^{sp,x}_{e\text{-}e})$, where $E^{1e}_{GS}=E^e_{00}$ is the $N_e=1$ GS energy and $E^X_p=E^e_p+E^h_p-V^{pp,d}_{e\text{-}h}$ ($V^{pp,d}_{e\text{-}h}\equiv V^{e\text{-}h}_{pppp}$) is denoted as the energy of a bare exciton on a p-shell state that is

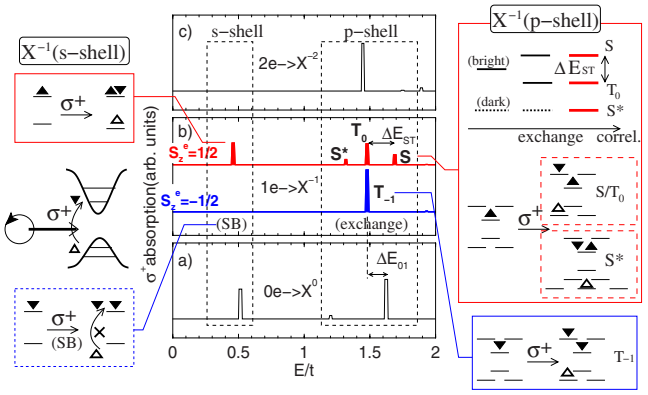

FIG. 1. (Color online) Polarized (σ^+) absorption spectra of symmetric ($\gamma=0$) charged SAQDs with resident electrons $N_e=0-2$ [(a)–(c)] with all possible spin. The main configurations involved in the spin-selective transitions $1e\rightarrow X^{-1}$ and the energy levels of X^{-1} states with $S_z=0$ vs Coulomb interactions are sketched beside the panel. The main underlying mechanisms (spin blockade, exchange, or correlation) in the spin characteristic spectra are denoted in the parentheses. Photon energies in the spectra are rescaled by $t\equiv\hbar(\omega_e+\omega_h)$, the total kinetic energy of an e-h pair in the lowest orbital state ($t\approx 37.7$ meV for the SAQDs considered in this work). $\Delta E_{01}(\approx 7.14$ meV) denotes the redshift of the main p-shell lines as the dot is loaded with electrons from $N_e=0\rightarrow 1$. The energy separation between the S and T lines for X^{-1} is $\Delta E_{ST}(X^{-1})=2V^{sp,x}_{e\text{-}e}=\frac{\sqrt{3}}{8}t\approx 8.16$ meV, in agreement with the measurement in Ref. 16.

further renormalized in the trion state by gaining the energies $V^{sp,x}_{e\text{-}e}=V^{e\text{-}e}_{s^\pm p^\mp s^\mp p^\pm}$ from the exchange interactions with the spin electron on s orbitals and $V^{pp,x}_{e\text{-}h}$ due to configuration intermixing. The renormalization leads to the redshift $\Delta E_{01}=V^{sp,x}_{e\text{-}e}+V^{pp,x}_{e\text{-}h}\left(=\frac{7\sqrt{3}}{64}t\approx 7.14\ \mathrm{meV}\right)$ of the main p-shell lines from $E=1.62t$ to $1.49t$ as the dot is loaded with electrons from $N_e=0\rightarrow 1$, in agreement with the experiment in Ref. 16.

For a singly charged dot with spin $s_z=+\frac{1}{2}$ electron, a σ^+ p-shell absorption creates the two coupled configurations with spins $(S_z=0,j_z=+3/2)$, $|X^{-1};1\rangle=h^+_{p^-,\uparrow}c^+_{p^-,\downarrow}c^+_{s,\uparrow}|0\rangle$ and $|X^{-1};2\rangle=h^+_{p^+,\uparrow}c^+_{p^+,\downarrow}c^+_{s,\uparrow}|0\rangle$. Furthermore, the two bright configurations are resonantly coupled to two other dark ones via

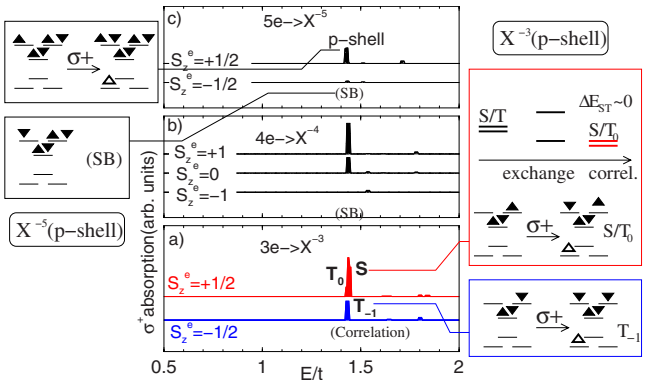

FIG. 2. (Color online) Same as Fig. 1 but for $N_e=3-5$. Note that (unlike X^{-1}) the S and T states of X^{-3} are nearly degenerate [$\Delta E_{ST}(X^{-3})\sim 0$].

e-e exchange interaction $V_{e\text{-}e}^{sp,x}$, $|X^{-1};3\rangle=h_{p^-,\uparrow}^{+}c_{p^-,\uparrow}^{+}c_{s,\downarrow}^{+}|0\rangle$ and $|X^{-1};4\rangle=h_{p^+,\uparrow}^{+}c_{p^+,\uparrow}^{+}c_{s,\downarrow}^{+}|0\rangle$. In the basis of the even-parity configurations, $|X^{-1};a+\rangle=(|X^{-1};1\rangle+|X^{-1};2\rangle)/\sqrt{2}$ and $|X^{-1};b+\rangle=(|X^{-1};3\rangle+|X^{-1};4\rangle)/\sqrt{2}$, we have the 2×2 Hamiltonian matrix

$$\begin{bmatrix} E_p^X+E_s^e-V_{e\text{-}h}^{pp,x} & -V_{e\text{-}e}^{sp,x} \\ -V_{e\text{-}e}^{sp,x} & E_p^X+E_s^e-V_{e\text{-}h}^{pp,x} \end{bmatrix}, \tag{4}$$

and the two trion eigenstates, both of which are optically active, i.e., a singlet (S) state $|X^{-1};S\rangle=(|X^{-1};a+\rangle-|X^{-1};b+\rangle)/\sqrt{2}$ and a T one $|X^{-1};T_0;+\rangle=(|X^{-1};a+\rangle+|X^{-1};b+\rangle)/\sqrt{2}$, possessing the eigenenergies $E(X^{-1};S)=E_{GS}^{1e}+(E_p^X-V_{e\text{-}h}^{pp,x}+V_{e\text{-}e}^{sp,x})$ and $E(X^{-1};T_0)=E_{GS}^{1e}+(E_p^X-V_{e\text{-}h}^{pp,x}-V_{e\text{-}e}^{sp,x})$, respectively. The two main lines shown in Fig. 1(b) at $E\sim1.69t$ and $E\sim1.49t$, respectively, correspond to the bright S and T states, separated in energy by $2V^{sp,x}\left(=\frac{\sqrt{3}}{8}t\approx8.16\text{ meV}\right)$. In addition, the bright S state $|X^{-1};S\rangle$ is nonresonantly coupled to the dark S configuration $|X^{-1};S^*\rangle=h_{d0,\uparrow}^{+}c_{s,\downarrow}^{+}c_{s,\uparrow}^{+}|0\rangle$ with the same total orbital and spin angular momenta via Coulombic e-h scattering. That induces the weak line at $E\sim1.32t$, denoted by S^* in Fig. 1(b). As a result, the σ^+ p-shell absorption pattern for $N_e=1$ dots with $S_z=+\frac{1}{2}$ is composed of three main lines, in agreement with the experimental observation in Ref. 16.

For an $N_e=2$ dot with closed s shell, the absorption spectrum is simply composed of a sole peak corresponding to the p-shell transition, as shown in Fig. 1(c). The s-shell absorption, absent in Fig. 1(c), is actually forbidden for all dots with $N_e\geqslant2$ because of Pauli blockade.

So far, we have seen two underlying physics in the spin characteristic absorption for dots with $N_e\leqslant2$, i.e., the spin Pauli blockade (SB) and Coulomb exchange interaction. In our studies, we confirm that SB determines the spin characteristics of polarized absorption for the dots filled with electrons on a shell equal to or more than the shell orbital degeneracy, e.g., the s-shell absorption for $N_e=1$ [see Fig. 1(b)] and the p-shell absorption for $N_e=4,5$ [see Figs. 2(b) and 2(c)]. In contrast, as a dot is filled with electrons less than the shell orbital degeneracy, e.g., the p-shell absorption for dots with $N_e=1$ and $N_e=3$ [Figs. 1(b) and 2(a)], there exists no SB, and Coulomb interactions may play a key role.

Now, let us proceed to study the case of $N_e=3$. Following Hund's rules, a GS of $N_e=3$ dot consists of a pair of spin antiparallel electrons on the s orbital and a p-shell electron with up or down spin. Like $N_e=1$ dots, a triply charged dot provides a net spin $S_z=\pm\frac{1}{2}$ to interact with spin exciton created by polarized absorption. Thus, one might expect some similar features of the absorption spectra for both $N_e=1$ and $N_e=3$ cases.

For an $N_e=3$ dot with $S_z=-\frac{1}{2}$, a σ^+ p-shell excitation pumps the dot in one of the doubly degenerate T X^{-3} states, $h_{p^-,\uparrow}^{+}c_{p^+,\downarrow}^{+}c_{p^-,\downarrow}^{+}|2e;\text{GS}\rangle$ or $h_{p^+,\uparrow}^{+}c_{p^+,\downarrow}^{+}c_{p^-,\downarrow}^{+}|2e;\text{GS}\rangle$, with the energy $E=E_{GS}^{3e}+E_p^X-V^{sp,x}-V^{pp,x}$. As shown in Fig. 2(a), its absorption spectrum does show a similarity to that for $N_e=1$ dot with the same spin $S_z=-\frac{1}{2}$ [see Fig. 1(b)], containing only one main p-shell line (T_{-1}) corresponding to the T state.

Such a similarity however, is not seen in the absorption spectra for the $N_e=1$ and $N_e=3$ dots with up spin $S_z=+\frac{1}{2}$. In both cases, σ^+-excited dots could be in the states of X^{-1} or X^{-3} with electronic S or T spins. As discussed previously, the S and T X^{-1} states are energetically separated by e-e exchange interaction $2V^{sp,x}$. Surprisingly, the S and T states of the X^{-3} complex in a σ^+-excited dot are found to be nearly degenerate. Consequently, one sees only a single main line, a doublet for the S and T states, at $E\sim1.43t$ in the spectrum of Fig. 2(a). For more illustration, we perform the following analysis.

Taking the three coupled low lying X^{-3} configurations into account, $|X^{-3};1\rangle=h_{p^-\uparrow}^{+}c_{p^+\uparrow}^{+}c_{p^-\downarrow}^{+}|2e;\text{GS}\rangle$, $|X^{-3};2\rangle=h_{p^-\uparrow}^{+}c_{p^+\downarrow}^{+}c_{p^-\uparrow}^{+}|2e;\text{GS}\rangle$, and $|X^{-3};3\rangle=h_{p^+\uparrow}^{+}c_{p^+\downarrow}^{+}c_{p^+\uparrow}^{+}|2e;\text{GS}\rangle$, we have a T eigenstate given by $|X^{-3};T\rangle=(|X^{-3};1\rangle+|X^{-3};2\rangle)/\sqrt{2}$ and two S states, obtained by diagonalizing the Hamiltonian matrix in the basis of S configuration, $(|X^{-3};1\rangle-|X^{-3};2\rangle)/\sqrt{2}$ and $|X^{-3};3\rangle$,

$$\begin{bmatrix} E_{GS}^{3e}+E_p^X-V_{e\text{-}e}^{sp,x}+V_{e\text{-}e}^{pp,x} & -\sqrt{2}V_{e\text{-}h}^{pp,x} \\ -\sqrt{2}V_{e\text{-}h}^{pp,x} & E_{GS}^{3e}+E_p^X-V_{e\text{-}e}^{sp,x} \end{bmatrix}. \tag{5}$$

The e-h interactions in the off-diagonal parts lead to the intermixing of the S configurations and shifts the energy of the lower state down to $E_{S_1}=E_{GS}^{3e}+E_p^X-V^{sp,x}-V_{e\text{-}h}^{pp,x}$. Thus, the low S state becomes degenerate to the T GS if the dot possesses a *hidden symmetry* (leading to $V_{e\text{-}e}^{pp,x}=V_{e\text{-}h}^{pp,s}$), which has been previously evidenced in photoluminescence spectroscopy studies for some SAQDs.[6] Here, we see that the hidden symmetry results in the single S and T doublet line of polarized absorption and removes the spin dependence of absorption spectra for triply charged dots.

Nevertheless, slight deformations in SAQDs create additional spin dependences of the absorption spectra.[18] Figure 3 shows the σ^+ absorption spectra for asymmetric $N_e=1$ dots and $N_e=3$ dots with spin $S_z=\pm\frac{1}{2}$ and $\gamma=0,0.01,\ldots,0.1$. The deformation $\delta V(\vec{r})$ in the quadratic form opens the channels of particle transferring between the orbitals differing in L by $0,\pm2$ (e.g., between the p^+ and p^- orbitals). For electronic S states of X^{-3}, the bright main configurations $h_{p^-\uparrow}^{+}c_{p^-\downarrow}^{+}c_{p^-\uparrow}^{+}|2e;\text{GS}\rangle$ for the line labeled by S in Fig. 3(c) become coupled to other dark S ones with $|\Delta L|=2$ (e.g., $h_{p^+\uparrow}^{+}c_{p^-\downarrow}^{+}c_{p^-\uparrow}^{+}|2e;\text{GS}\rangle$, $h_{p^-\uparrow}^{+}c_{p^+\downarrow}^{+}c_{p^-\uparrow}^{+}|2e;\text{GS}\rangle,\ldots$). The deformation-induced configuration intermixing lowers the bright S GSs and separates it from the T_0 state. Moreover, it gives rise to the line involving those dark S states, labeled by S^* in Fig. 3(a). Likewise, the p-shell absorption lines for the S and T X^{-1} trions in which the p shell is also singly occupied by an electron or a hole is split into two with finite γ [see Figs. 3(a) and 3(b)].

In contrast, a weak deformation does not give rise to any new absorption line for T X^{-3} states. This is because the T X^{-3} states must have the total electronic angular momentum $L^e=0$ and deformation breaks the conservation of angular momentum. We see that there exists only one absorption line for the T X^{-3} states in Figs. 3(b) and 3(c) as $\gamma<0.05$. As a result, the polarized absorption spectra for $N_e=3$ dots subject

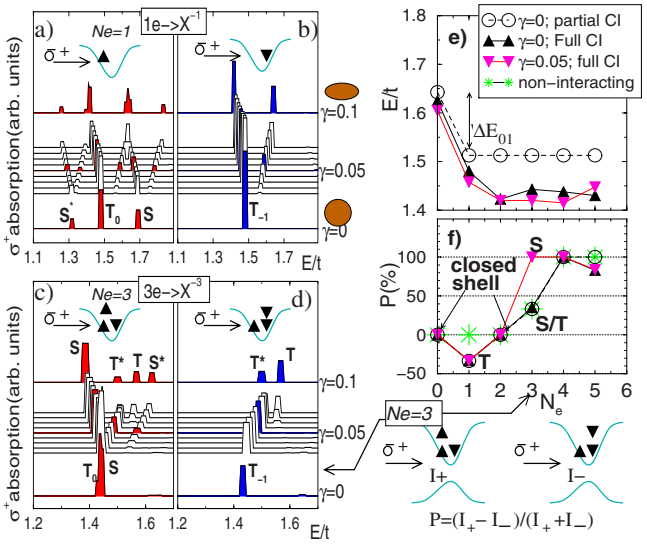

FIG. 3. (Color online) Spin-dependent polarized σ^+ absorption spectra of singly charged SAQDs with spins (a) $S_z=\frac{1}{2}$ and (b) $S_z=-\frac{1}{2}$ and triply charged dots with (c) $S_z=\frac{1}{2}$ and (d) $S_z=-\frac{1}{2}$ subject to different shape deformations ($\gamma=0,0.01,\ldots,0.1$). (e) Energies of the p-shell absorption lines at the highest intensity of charged dots as a function of N_e. (f) Degree of circular polarization P of the absorptions at the energies shown in (e) for N_e charged dots. Open circles: results calculated by using partial CI method, in which only the resonantly coupled configurations with the lowest total kinetic energies are taken into account. Up (down) filled triangles: results calculated by using full CI method based on the three lowest electronic shells for interacting symmetric (asymmetric) charged dots.

to a slight deformation exhibit remarkable spin dependence, in contrast to the spin insensitivity of the spectra of symmetric $N_e=3$ dots.

Figure 3(e) shows the energies of the p-shell absorption lines of charged dots at the highest intensities as a function of N_e and Fig. 3(f) shows the corresponding degree of circular polarization, defined as $P\equiv(I_+-I_-)/(I_++I_-)$, to the main lines. For noninteracting dots, we derive $P=0,33.3\%,100\%$ for $N_e=\{0,1,2\},\{3\},\{4,5\}$, respectively. Turning on Coulomb interactions, the degree of polarization for $N_e=1$ is changed from 0 to -33.3%. The negative finite $P=-33.3\%$ for X^{-1} results from the Coulomb exchange that splits the T and S states and decreases the intensity of the main line T_0 to half of the line T_{-1} [see Fig. 3(b)]. The P for $N_e=3$, however, remains unchanged because of the degeneracy of the S and T X^{-3} GSs, resulting from the intrinsic correlation in X^{-3}. With deformation, the P for $N_e=3$ abruptly increases from 33.3% to almost 100%, indicating the well resolved spin characteristic absorption pattern for X^{-3}.

IV. SUMMARY

In summary, we present theoretical studies of polarized absorption in charge-tunable SAQDs with resident electrons filling up to the p shell. Spin Pauli blockade, Coulomb exchange and correlation, and symmetry of dot shape are identified as the three main underlying mechanisms in the spin characteristic absorption. We point out that triply charged excitons in SAQDs possess intrinsic correlated nature and exhibit anomalous spin-related features of polarized absorption.

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[1] S. Cortez, O. Krebs, S. Laurent, M. Senes, X. Marie, P. Voisin, R. Ferreira, G. Bastard, J. M. Gerard, and T. Amand, Phys. Rev. Lett. **89**, 207401 (2002).
[2] V. Cerletti, W. A. Coish, O. Gywat, and D. Loss, Nanotechnology **16**, R27 (2005).
[3] A. Imamoglu, D. D. Awschalom, G. Burkard, D. P. DiVincenzo, D. Loss, M. Sherwin, and A. Small, Phys. Rev. Lett. **83**, 4204 (1999).
[4] X. Li, Y. Wu, D. Steel, D. Gammon, T. H. Stievater, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Science **301**, 809 (2003); M. Atature, J. Dreiser, A. Badolato, A. Hogele, K. Karrai, and A. Imamoglu, *ibid.* **312**, 551 (2006); J. Berezovsky, M. H. Mikkelsen, O. Gywat, N. G. Stoltz, L. A. Coldren, and D. D. Awschalom, *ibid.* **314**, 1916 (2006); M. Kroutvar, Y. Ducommun, D. Heiss, M. Bichler, D. Schuh, G. Abstreiter, and J. J. Finley, Nature (London) **432**, 81 (2004).
[5] S. M. Reimann and M. Manninen, Rev. Mod. Phys. **74**, 1283 (2002).
[6] M. Bayer, O. Stern, P. Hawrylak, S. Fafard, and A. Forchel, Nature (London) **405**, 923 (2000), and references therein.
[7] R. J. Warburton, C. Schaflein, D. Haft, F. Bickel, A. Lorke, K. Karrai, J. M. Garcia, W. Schoenfeld, and P. M. Petroff, Nature (London) **405**, 926 (2000).
[8] K. Karrai, R. J. Warburton, C. Schulhauser, A. Hogele, B. Urbaszek, E. J. McGhee, A. O. Govorov, J. M. Garcia, B. D. Gerardot, and P. M. Petroff, Nature (London) **427**, 135 (2004).
[9] E. A. Stinaff, M. Scheibner, A. S. Bracker, I. V. Ponomarev, V. L. Korenev, M. E. Ware, M. F. Doty, T. L. Reinecke, and D. Gammon, Science **311**, 636 (2006).
[10] F. Findeis, M. Baier, A. Zrenner, M. Bichler, G. Abstreiter, U. Hohenester, and E. Molinari, Phys. Rev. B **63**, 121309(R) (2001).
[11] L. Besombes, J. J. Baumberg, and J. Motohisa, Phys. Rev. Lett. **90**, 257402 (2003).
[12] F. Guffarth, R. Heitz, A. Schliwa, O. Stier, M. Geller, C. M. A. Kapteyn, R. Sellin, and D. Bimberg, Phys. Rev. B **67**, 235304 (2003).
[13] B. Urbaszek, R. J. Warburton, K. Karrai, B. D. Gerardot, P. M. Petroff, and J. M. Garcia, Phys. Rev. Lett. **90**, 247403 (2003).
[14] A. S. Bracker, E. A. Stinaff, D. Gammon, M. E. Ware, J. G.

Tischler, A. Shabaev, Al. L. Efros, D. Park, D. Gershoni, V. L. Korenev, and I. A. Merkulov, Phys. Rev. Lett. **94**, 047402 (2005).

[15] S. Laurent, B. Eble, O. Krebs, A. Lemaitre, B. Urbaszek, X. Marie, T. Amand, and P. Voisin, Phys. Rev. Lett. **94**, 147401 (2005).

[16] M. E. Ware, E. A. Stinaff, D. Gammon, M. F. Doty, A. S. Bracker, D. Gershoni, V. L. Korenev, S. C. Badescu, Y. Lyanda-Geller, and T. L. Reinecke, Phys. Rev. Lett. **95**, 177403 (2005); M. E. Ware, A. S. Bracker, E. Stinaff, D. Gammon, M. F. Doty, D. Gershoni, and V. L. Korenev, Physica E (Amsterdam) **26**, 55 (2005).

[17] S. J. Cheng and P. Hawrylak, Phys. Rev. B **73**, 035326 (2006).

[18] D. Chithrani, M. Korkusinski, S. J. Cheng, P. Hawrylak, R. L. Williams, J. Lefebvre, P. J. Poole, and G. C. Aers, Physica E (Amsterdam) **26**, 322 (2005).

[19] W. Sheng, S. J. Cheng, and P. Hawrylak, Phys. Rev. B **71**, 035316 (2005).

[20] The spin-orbit and Hyperfine interactions are negligible to the optical spectroscopy discussed here because the energy scale ($\ll 10^{-1}$ meV) of the interactions (Ref. 2) is much smaller than the resolution of the conventional spectroscopy ($>10^{-1}$ meV). Nevertheless, the spin interactions might significantly affect the coherence and dynamical properties of excitons in self-assembled quantum dots (Ref. 22).

[21] M. Florescu and P. Hawrylak, Phys. Rev. B **73**, 045304 (2006).

[22] R. Oulton, A. Greilich, S. Yu. Verbin, R. V. Cherbunin, T. Auer, D. R. Yakovlev, M. Bayer, I. A. Merkulov, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. Lett. **98**, 107401 (2007).

[23] G. A. Narvaez and A. Zunger, Phys. Rev. B **74**, 045316 (2006).

[24] A. Vasanelli, R. Ferreira, and G. Bastard, Phys. Rev. Lett. **89**, 216804 (2002).

[25] P. Hawrylak and M. Korkusinski, in *Single Quantum Dots: Fundamentals, Applications, and New Concepts*, edited by P. Michler, Topics in Applied Physics Vol. 90 (Springer-Verlag, Berlin, 2003), pp. 25–92.

[26] P. Hawrylak, G. A. Narvaez, M. Bayer, and A. Forchel, Phys. Rev. Lett. **85**, 389 (2000).

[27] A. Hogele, M. Kroner, S. Seidl, K. Karrai, M. Atatüre, J. Dreiser, A. Imamoglu, R. J. Warburton, A. Badolato, B. D. Gerardot, and P. M. Petroff, Appl. Phys. Lett. **86**, 221905 (2005).