Few-particle interactions in charged $In_xGa_{1-x}As/GaAs$ quantum dots

F. Guffarth,* R. Heitz, A. Schliwa, O. Stier, M. Geller, C. M. A. Kapteyn, R. Sellin, and D. Bimberg
Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstrasse 36, 10623 Berlin, Germany
(Received 26 July 2002; revised manuscript received 23 January 2003; published 9 June 2003)

The impact of few-particle interactions on excited states of excitons localized in self-organized $In_xGa_{1-x}As/GaAs$ quantum dots (QD's), charged either with electrons or holes, is investigated. Excited-state absorption is probed size selectively by photoluminescence excitation spectroscopy improving the achieved resolution beyond the inhomogeneous broadening. Charging QD's embedded in suitable diode structures leads to nonlinear changes of the absorption characteristics for the individual excited-state transitions, enabling their unambiguous identification. Few-particle interactions lead to a renormalization of the excited-state transition energies, which ranges from a pronounced red shift to a blue shift depending on the excited-state transition and the type of spectator charge. The most pronounced effects occur charging the ground state of the QD's. The results are supported by eight-band $\mathbf{k}\cdot\mathbf{p}$ model calculations using a configuration-interaction scheme to account for the Coulomb interaction in the few-particle states.

DOI: 10.1103/PhysRevB.67.235304 PACS number(s): 78.67.Hc, 78.55.Cr, 73.21.La, 71.15.Qe

I. INTRODUCTION

In recent years, fundamental properties of quantum dots (QD's), which confine electrons and holes in all three dimensions on a nanometer scale, have been studied extensively.[1] In particular, self-organized QD's have attracted much interest due to their defect-free, coherent nature, which is a precondition for the investigation of intrinsic electronic properties. The possibility to grow complex nanostructures on unpatterned substrates in a way compatible with conventional heterostructures and the high luminescence yield have led to the successful development of injection lasers,[2,3] as well as the demonstration of detectors[4,5] and memory devices.[6–8]

The localization of charges in the small volume of a QD strongly enhances the Coulomb interaction between the localized carriers. However, the kinetic confinement energy is predicted to reduce the impact of correlation and exchange in the strong confinement limit, leaving state filling as the dominant nonlinear effect.[9] In this limit, the Coulomb interaction alters the transition energies, but the change of the wave functions due to correlation/exchange is negligible. For self-organized QD's, the confinement is in the intermediate regime and the Coulomb interaction is expected to noticeably change the wave functions. Indeed, calculations predict a renormalization of the ground-state exciton transition energy by the presence of spectator charges as well as of spectator excitons.[10] For self-organized QD's, the structural details and, in particular, the resulting inhomogeneous strain distribution lead to a complex potential landscape, which is distinctively different for electrons and holes.[11] Correspondingly, the effect of the Coulomb interaction on localized few-particle states is expected to depend strongly on the actual structural details of the QD's.[12]

In recent years, investigations on single QD's have provided experimental information on the renormalization of the ground-state exciton in the presence of spectator charges or excitons.[13–18] The spectral resolution in such experiments is limited ideally only by the homogeneous linewidth of the transitions and allows resolving the fine structure as well as renormalization on a μeV scale. Both binding and antibinding renormalizations have been observed for exciton and biexciton complexes in QD's charged with electrons[13–16] and holes.[17,18] However, such single-QD PL (photoluminescence) experiments concentrate on the ground state of the multiparticle configurations. In order to gain information on the excited-state transitions, one has to probe the absorption spectrum of the charged QD's. Previous investigations of near-infrared exciton[19] and mid-infrared electron[20] absorption of negatively charged QD's probed the full ensemble and, thus, were limited by inhomogeneous broadening, which obscures the information expected from such experiments.

Here we investigate the excited-state spectra of subensembles of charged self-organized $In_xGa_{1-x}As/GaAs$ QD's defined by the ground-state transition energy. Incorporating the QD's in the space-charge region of p^+n- and n^+p-diodes allows controlled charging of the QD's with electrons and holes, respectively. To overcome the inhomogeneous broadening PL excitation spectroscopy (PLE) is used, which probes size selectively the absorption spectrum of QD's with the selected ground-state transition energy.[21] The bias-dependent charge state of the QD's is determined from capacitance-voltage (C-V) investigations (Sec. III A), showing that the present QD's can be filled with up to approximately eight electrons and seven holes, respectively. A characteristic saturation as well as a renormalization of the transition energies of the individual absorptions with increasing charging help in identifying the excited-state transitions and to understand the few-particle effects (Secs. III B and III C). The experimental results are supported by eight-band $\mathbf{k}\cdot\mathbf{p}$ calculations (Sec. III D) accounting for the Coulomb interaction including correlation and exchange in a configuration-interaction scheme.[22]

II. EXPERIMENT

The investigated samples were grown by metalorganic chemical vapor deposition (MOCVD) on GaAs(001) substrates, whereby the QD layer was incorporated in p^+n- and n^+p-diode structures to allow for controlled charging. The QD layer surrounded by $\sim$10 nm semi-insulating GaAs is

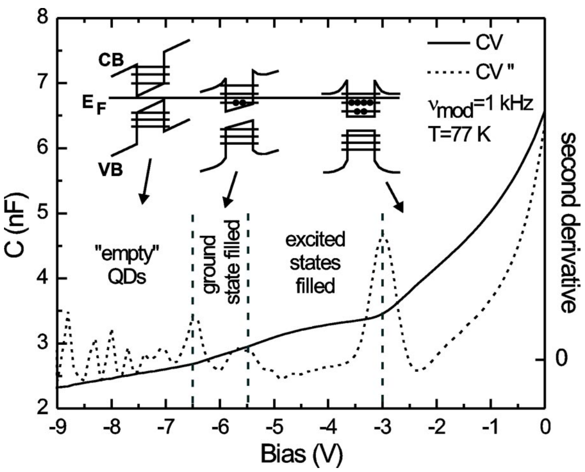

FIG. 1. *C-V* plot of sample *E*1 allowing to identify the bias-dependent average charge state of the QD's. Below −6.5 V the QD's are empty; between −6.5 and −5.5 V, the ground state is filled; and between −5.5 and −3 V, excited states are filled with spectator electrons. Above −3 V, localization of further spectator electrons in the QD's is prevented by Coulomb charging. The insets depict schematically vertical cuts through the QD potential for different charge states.

embedded in the low-doped ($\sim 10^{16}$ cm^{-3}) side of the diodes. The QD's are outside the space-charge region without bias and, thus, completely charged in a conductive surrounding. Increasing the reverse bias, the QD's are successively depopulated and finally become neutral. The process is illustrated schematically by the insets (from right to left) of Fig. 1 for electrons.

Sample *E*1 is a p^+n structure with $In_xGa_{1-x}As/GaAs$ QD's embedded in the *n* region. The $In_xGa_{1-x}As$ QD's were grown at 500 °C depositing ∼7 ML $In_{0.7}Ga_{0.3}As$ on top of a QD "seed" layer formed by the deposition of ∼3.5 ML $In_{0.7}Ga_{0.3}As$ QD's, where ML stands for monolayer. The GaAs spacer of only 3 nm thickness ensures vertical correlation, which allows to fix the QD density in favor of a larger size of the upper $In_xGa_{1-x}As$ QD layer.[23] Transmission electron microscopy investigations of similar samples[24] yield an area density of $\sim 5\times 10^{10}$ cm^{-2} and a truncated shape with an average base width of ∼20 nm and height of ∼4 nm. X-ray diffraction[25] and cross-sectional scanning tunnel microscopy data[26] suggest that the indium content of the QD's is indeed higher than the nominal one. The ground-state transition energy is ∼1.0 eV at He temperatures. Note that the smaller $In_xGa_{1-x}As$ seed QD's have only a minor effect on the electronic and optical properties of the $In_xGa_{1-x}As$ QD's due to the large mismatch of the ground-state transition energies.[27]

In order to compare the impact of negative and positive charging of QD's with electrons and holes, respectively, two additional structures, dubbed *E*2 and *H*2, have been investigated. The samples contain a single $In_xGa_{1-x}As/GaAs$ QD layer formed by the deposition of ∼3 ML $In_{0.8}Ga_{0.2}As$ at 500 °C in a p^+n (E2) and a n^+p (H2) structure. The QD's are less dense ($\sim 3\times 10^{10}$ cm^{-2}) and smaller than in sample *E*1,[28] resulting in a larger ground-state transition energy of ∼1.12 eV.

The bias-dependent charging of the QD's in the diode structures was investigated in C-V experiments using a precision *LCR* meter as described in Ref. 29. For the PL and PLE experiments, the samples were mounted in a continuous-flow He cryostat at temperatures between 7 K and 220 K. A tungsten lamp dispersed by a 0.27 m double-grating monochromator served as a low-density, tunable light source. The luminescence was spectrally dispersed by a 0.3 m double-grating monochromator and detected with a cooled Ge-diode using lock-in techniques.

III. EXPERIMENTAL RESULTS

A. Bias-dependent charge state

Incorporating QD's in *pn*-diode structures allows us to tune the Fermi level in the vicinity of the QD's by applying an appropriate bias voltage, which is commonly exploited, e.g., for deep level transient capacitance spectroscopy investigations.[30] In the present context, *C-V* spectra reveal the biases needed to charge the different QD levels in the investigated samples, whereby a low modulation frequency (1 kHz at 77 K) ensures compatibility to the quasistatic situation in the optical measurements. Figure 1 depicts the *C-V* spectrum (solid line) and its second derivative (dashed line) for sample *E*1. The low-bias onsets of the plateaus, i.e., the peaks in the second derivative mark the beginning population of a QD shell.[31] Three different regimes can be identified: Below a bias of −6.5 V, the electron levels of the QD's are completely above the Fermi level and therefore no spectator electrons are in the QD's (left inset of Fig. 1). Between −6.5 and −5.5 V, the ground state is populated with two spectator electrons (middle inset). Above −5.5 V, excited states are populated with electrons, whereby above −3 V the QD's leave the space-charge region being completely filled (right inset).

Note that the Coulomb charging limits the equilibrium population of the QD's well before all localized levels are filled. The localized charges lead to a local band bending around the QD's lifting the higher excited states above the Fermi level (see the right inset of Fig. 1). Such excited states are separated only by a potential barrier from the surrounding barrier continuum and often dubbed "resonant states." Even though the carriers in such resonant states could principally tunnel to the surrounding matrix, they can be optically investigated as shown below.

The *C-V* results allow us to estimate the number of carriers accumulated in the QD layer.[32] The independent knowledge of the QD density is required to estimate the average population of the completely charged QD's. For sample *E*1, this yields approximately eight electrons per QD. The large substate splitting of the electron levels allows us to distinguish population of the ground and excited QD states, resulting in an additional plateau in the *C-V* curve.[29] The evaluation of the *C-V* data in this bias region yields two carriers per QD, as expected for a spin-degenerated ground state.

C-V measurements of sample *E*2 reveal charging of the ground state above −1.1 V and of the excited states above

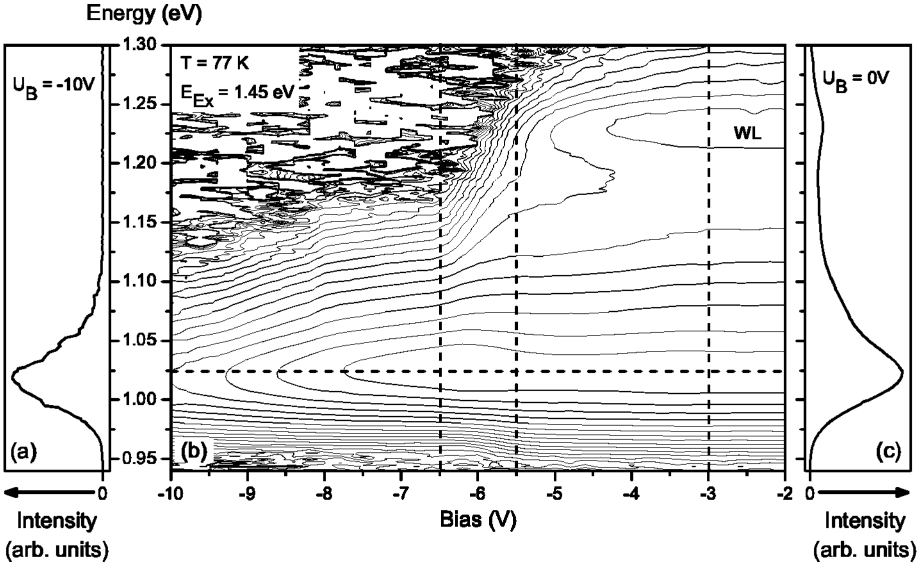

FIG. 2. Bias-dependent PL spectra at 77 K on a logarithmic scale (b), showing the ground-state shift to be negligible above -6.5 V. Panels (a) and (c) show PL spectra for the limiting cases of empty and completely charged QD's, respectively, on a linear scale.

-0.45 V. At $+0.4$ V, the QD's leave the space-charge region. The actual biases differ for each sample depending on the position of the QD's with respect to the space-charge region and the doping levels. Sample $H2$ is populated with holes between -9 V and -6.8 V, whereby the smaller quantization of the hole levels does not allow to distinguish population of the ground and excited states. The smaller size of the QD's in samples $E2$ and $H2$ results in a smaller localization energy and a larger substate splitting. Therefore, we expect less carriers per completely charged QD compared to sample $E1$. Evaluating the C-V data yields approximately five electrons per QD for sample $E1$ and approximately seven holes per QD for sample $H2$.

B. Negatively charged QD's

Charge-dependent PL spectra of the p^+n structure $E1$ with the seeded InGaAs QD's are summarized in Fig. 2. The spectra have been recorded at 77 K exciting at 1.45 eV with a density below 5 $\mathrm{mW/cm^2}$ in order to ensure that the carrier distribution reaches and maintains its bias-dependent equilibrium under optical excitation. Figure 2(b) shows a contour plot of the logarithmic PL intensity in dependence on the detection energy and the applied bias. Figures 2(a) and 2(c) depict, respectively, the PL spectra for the two limiting cases of empty ($U_{Bias}=-10$ V) and completely filled ($U_{Bias}=0$ V) QD's. At zero bias, PL from localized wetting layer (WL) states[33] ($\sim$1.23 eV) is seen in addition to the QD PL peak at 1.024 eV. Applying a reverse bias leads to an electric field across the QD layer, which ultimately decreases the luminescence yield. The threshold for the PL quenching depends on the exciton localization, starting with the WL (~-5 V) and finally also affects the QD (~-8 V) luminescence. The probability of carrier escape by tunneling across the approximately triangular barrier depends on the localization energy and the local electric field. Interestingly, the inhomogeneously broadened PL peak does not change its energy position when depopulating the QD's between -3 V and -6.5 V. In single-QD investigations, a renormalization of the ground-state exciton recombination energy of up to $\sim$8 meV has been observed for negatively charged $In_xGa_{1-x}As/GaAs$ QD's,[16] which is clearly not the case for the present sample. A possible explanation might be the dependence of the renormalization on the details of the confining potential, being obviously quite different for the investigated $In_xGa_{1-x}As$ QD's ($E_G\sim1$ eV) and QD's with ground-state energies >1.2 eV, typically probed in single-QD experiments. The minor red shift ($\sim$5 meV) decreasing the bias voltage from -6.5 V to -10 V (the QD's are neutral in this bias range) is attributed to the Stark effect[34] and the dependence of the carrier escape rate on the localization energy.

Figure 3 compares normalized PL spectra of sample $E1$ for various biases, i.e., as a function of the charging of the QD's. At 7 K [Fig. 3(a)], the spectator electrons do not affect the PL spectrum even when the electrons populate excited states, suggesting that independent of the charge state, recombination of electrons and holes in the respective ground-state dominates. Obviously, the relaxation of the photogenerated holes to the ground state is faster than recombination, and "allowed" transitions dominate. Note that the broad luminescence between 1.1 eV and 1.3 eV [Fig. 2(c)] originates from a broad distribution of localized WL states[33] and not from transitions involving the recombination of electrons in excited states of the charged QD's.

At 220 K [Fig. 3(b)], thermal redistribution within the QD ensemble leads to a narrowing of the PL spectrum, and excited-state transitions become visible in the charged QD's. The latter is attributed to the thermal population of excited hole levels, which enables allowed recombination with electrons in excited states. Obviously, allowed transitions with $\Delta n=0$ dominate in the spectra of the investigated flat, truncated QD's, though strict selection rules would be expected only for idealized QD's with, e.g., a two-dimensional harmonic-oscillator potential. Theoretical studies indicate that the reduction of the piezoelectric potential when truncating QD's partly recovers the selection rules.[10] In addition,

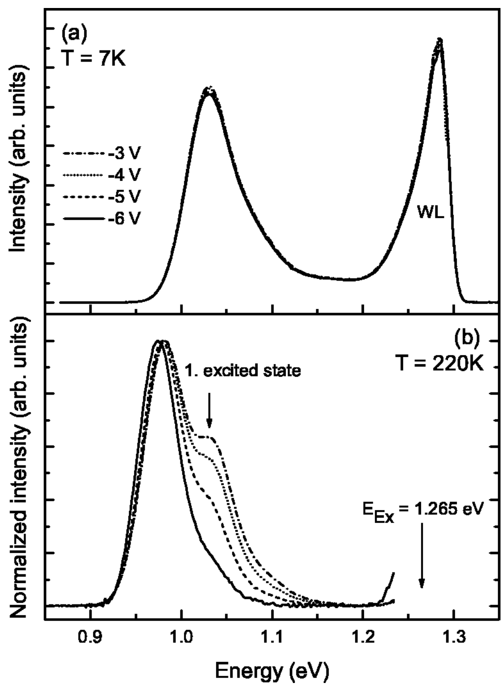

FIG. 3. PL spectra of sample $E1$ for different charging conditions at 7 K (a) and 220 K (b). At low temperature, the hole relaxes to the ground-state before recombination and no excited-state transitions can be seen. At 220 K, the holes are thermally activated into excited states, and excited-state recombination is observed in PL. The results indicate that allowed transitions dominate the near-infrared optical spectra in truncated QD's.

the spectator charges in the QD's lead to a screening of the piezoelectric potential further reducing the lateral asymmetry.

In the following, excited-state absorption in charged QD's is investigated by PLE spectroscopy. As has been demonstrated recently, in an ensemble of noninteracting QD's the PLE spectra reveal absorption spectra of a subensemble defined by the ground-state transition energy.[21] Varying the detection energy allows, e.g., to probe the size dependence of the quantization. Here, a fixed detection energy is used to select always the same sub-ensemble in the charge-dependent experiments. The renormalization of the ground-state transition energy is negligable as was shown in Fig. 2(b). Figure 4(b) shows the PL intensity on a logarithmic scale in dependence of the excitation energy and the bias voltage. Figures 4(a) and 4(c) show the PLE spectra of empty ($U_{Bias}=10$ V) and completely charged ($U_{Bias}=0$ V) QD's, respectively. As has been demonstrated previously,[21] the dominating line *groups* can be labeled with respect to a two-dimensional harmonic-oscillator model ($n=0,1,2$), which might serve as a first approximation, since for electrons the quantization is significantly larger than the impact of the structure-dependent symmetry lowering. This notation captures only the main contributions, neglecting the fine structure resulting from the actual structural properties of the QD's.[11,21] For the neutral QD's, the first excitation resonance ~ 16 meV above the detection energy has previously been attributed to a transition between an excited hole state and the electron ground state.[21] The resonance at ~ 36 meV is attributed to LO-phonon-assisted absorption of the exciton ground state. Both peaks involve the electron ground state and are, therefore, marked $n=0$. For the first excited ($n=1$) electron state, three peaks are resolved around 53 meV, 67 meV, and 77 meV, involving two nearly degenerate electron states and several hole states. Finally, the components of the second excited ($n=2$) state lead to absorption ~ 140 meV above the ground state transition energy.

Charging the QD's with electrons, the PLE spectra, i.e., the absorption spectra, change drastically. For each excitation resonance, the transition energy and intensity are, respectively, renormalized and quenched in a characteristic way, see Fig. 5. The quenching of the PLE peaks is attributed in first order to saturation resulting from successive filling of the electron levels, providing additional information on the origin of the various absorptions. The intensity of the excitation resonances is shown in Fig. 5(a) as a function of the

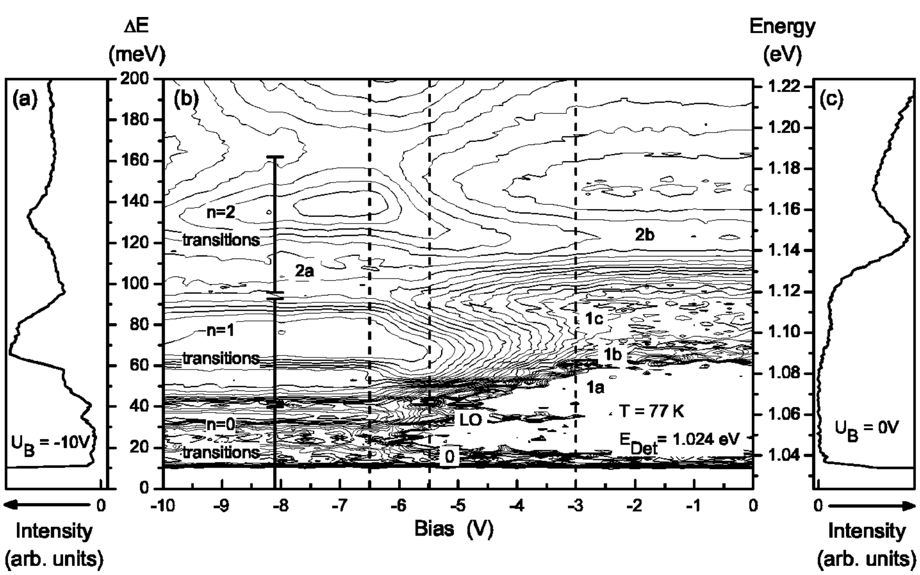

FIG. 4. Contour plot of the PL intensity at 1.024 eV on a logarithmic scale in dependence on the bias and the excitation energy for sample $E1$, panel (b). Two $n=0$ transitions, involving an excited hole state (marked 0) and LO-phonon-assisted ground-state absorption (LO), are shown. Substructures of the $n=1$ transitions ($1a$–$1c$) and $n=2$ transitions ($2a$,$2b$) can be identified. Panels (a) and (c) depict PLE spectra for the limiting cases of empty and completely charged QD's, respectively, on a linear scale.

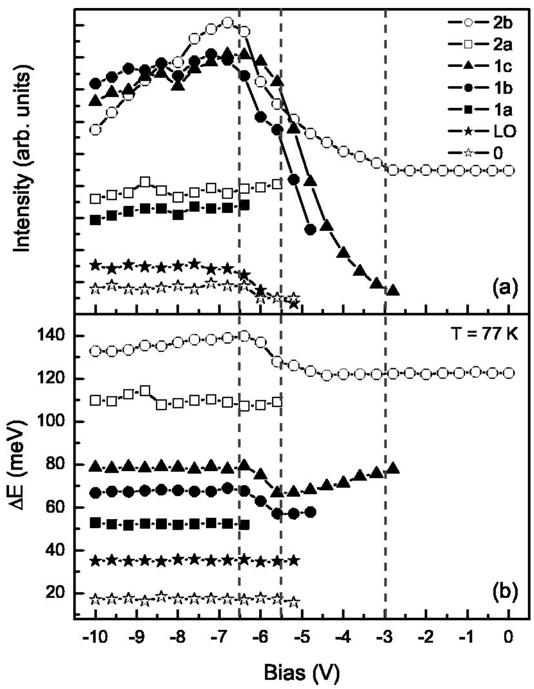

FIG. 5. Evaluation of Fig. 3, (a) bias-dependent intensity and (b) bias dependent transition energy of the excited-state transitions for sample $E1$.

applied bias. Both $n=0$ transitions are quenched between -6.5 and -5.5 V indicating saturation of the QD ground state with electrons in good agreement with the C-V results discussed above, see Fig. 1. The results support the previous assignment to a ''forbidden'' transition between the electron ground state and an excited hole state.[21] The three resolved components of the $n=1$ excited-state transition group quench between -5.5 V and -3 V. The energy splitting of the components of the $n=1$ state is small compared to the main quantization, being consistent with the occurrence of line groups in PLE as well as the observation of a single inhomogeneously broadened peak in high-density PL experiments.[35] Finally, also the $n=2$ excitation resonance quenches by about 50%. Note that the absorption of the $n=2$ excited state decreases already populating the ground and the $n=1$ excited states. This behavior suggests a reduction of the corresponding oscillator strength when charging the QD's. Due to the Coulomb charging, the $n=2$ excited state is expected to become a resonant state above the GaAs band edge. The concomitant change of the wave functions could lower the oscillator strength and thus mimic a saturation of the absorption. By assuming a two-dimensional harmonic-oscillator model, a maximum charging with approximately eight to nine spectator electrons is estimated when the QD's are outside the space-charge region, being in good agreement with the estimate based on the C-V data (Sec. III A). Note that in a conducting environment, the maximum charge state is insensitive to the actual doping level, which however affects the width of the tunnel barriers and, thus, the properties of the resonant states and the time needed to reach an equilibrium charge distribution. For the investigated samples and excitation energies above ~ 1.23 eV, the PLE spectra show almost no dependence on the charge state of the QD's. Apparently, holes relax to the ground state recombining with a ground state electron, and electron relaxation dominates the tunnel escape for the resonant states.

Figure 5(b) summarizes the transition energies, which are given with respect to the ground-state transition, of the various excitation resonances as a function of the applied bias. The QD's are empty below -6.5 V and experience an increasing local electric field with decreasing bias. The field-induced tunnel escape and the Stark effect reduce the excitation yield and lower the energy of the highest excited state $2b$, see Fig. 5. More interestingly, the transition energies renormalize when the QD's are charged with electrons above -6.5 V. The largest effect on the excited-state transition energies have the first two charges occupying the ground state, which result in red shifts of the $n=1$ and $n=2$ transitions of up to ~ 17 meV. With increasing charging, few-particle effects lead to a renormalization of the transition energies. The observed red shift implies that the excited-state transitions are more sensitive to correlation/exchange than the ground-state transition and that the direct Coulomb interaction is less important than exchange and correlation. A possible explanation for the large effect of the first two charge carriers might be the efficient screening of the piezoelectric potential, a nonlinear effect that is well known for $In_xGa_{1-x}N$ structures.[36,37] Note that band bending due to Coulomb charging and the finite electric field in the growth direction [as long as the QD's are in the depletion region ($U_{Bias} < -3$ V)] affects the wave functions and transition energies, too.

The combination of optical data and typical properties of $In_xGa_{1-x}As$ QD's allows another estimation of the number of electrons in sample $E1$. The estimate is based on the level splitting of ~ 70 meV extracted from Fig. 5(b), a Coulomb charging energy per carrier of ~ 20 meV,[31,38,39] and an electron localization energy of typically 280 meV (Ref. 10) for a ground-state transition energy of 1.024 eV. Therewith, about seven spectator electrons will push the $n=2$ excited states above the bulk GaAs band edge due to Coulomb charging, i.e., they will become ''resonant states.'' The estimate agrees well with approximately eight electrons per QD extracted from C-V (Sec. III A) and the optical data discussed above.

C. Positively charged QD's

Samples $E2$ and $H2$ allow us to compare the impact of spectator electrons and holes on the excited states in nominally identical $In_xGa_{1-x}As$ QD's. Figure 6 compares the results of PLE experiments on samples $E2$ (left-hand side) and $H2$ (right-hand side). The PLE spectra are shifted vertically for clarity. The $In_xGa_{1-x}As/GaAs$ QD's are smaller than the seeded $In_xGa_{1-x}As/GaAs$ QD's in sample $E1$, resulting in a higher ground-state transition energy (~ 1.12 eV) and slightly larger substate splittings. A consequence of the weaker electron localization is a lower maximum charge state. The PLE spectra support a lower maximum charging showing still some absorption due to the $n=1$ excited-state

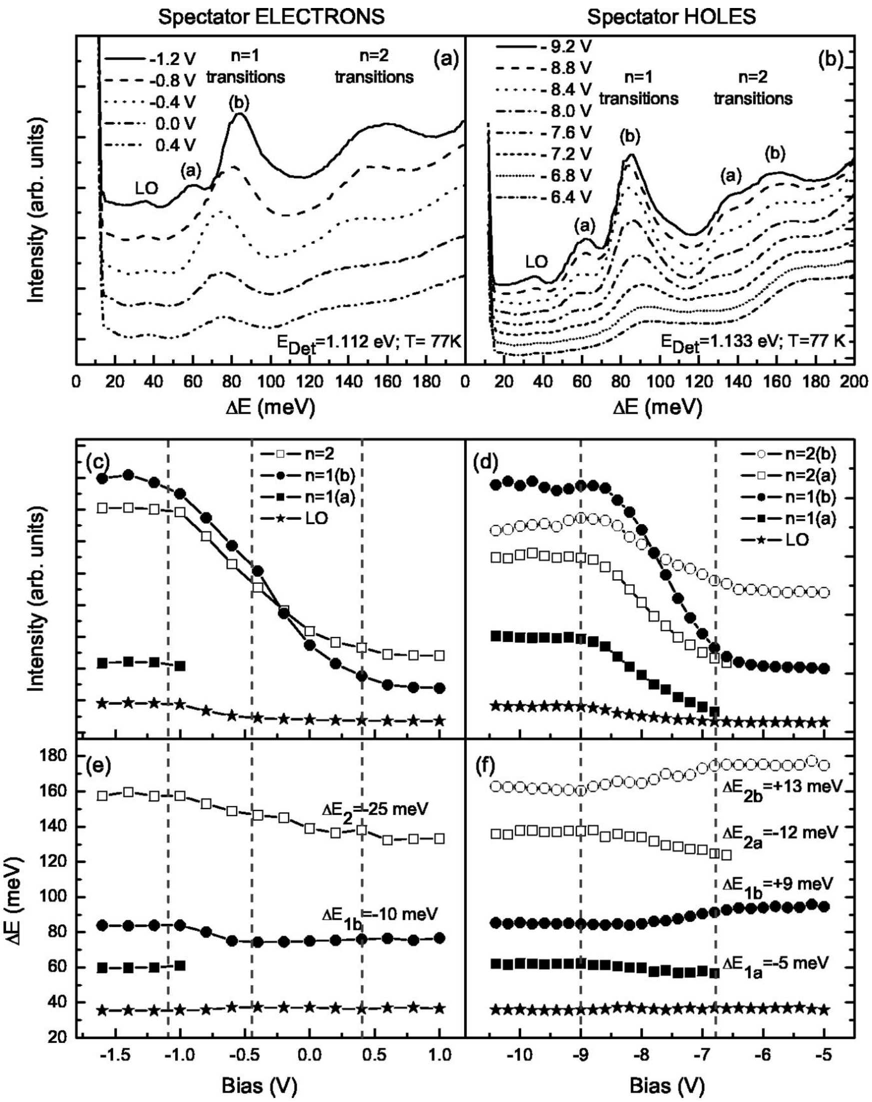

FIG. 6. Comparative PLE study of negatively (sample $E2$) and positively (sample $H2$) charged $In_xGa_{1-x}As$ QD's. Panels (a) and (b) show the PLE spectra in the energy range of the excited state transitions for different charging conditions. The evaluation of the intensity and transition energy is shown for both samples in panels (c) and (e) for spectator electrons and panels (d) and (f) for spectator holes. Additional information is extracted from C-V measurements and plotted as vertical-dashed lines indicating the plateaus where the QD's become charged/discharged.

transitions for sample $E2$ under completely charged conditions ($U_{Bias}>0.4$ V). The occupation of completely charged QD's with approximately five electrons extracted from C-V measurements (Sec. III A) is consistent with the optical results. Populating the electron ground state leads again to a pronounced renormalization of the $n=1$ transitions by about 10 meV. However, for the $n=2$ excited-state transition the red shift continues until the QD's are "completely" charged, reaching 25 meV. This behavior is clearly different from that of sample $E1$, Fig. 4(b), which might be attributed to different structural properties of the QD's in both samples.

The right-hand side of Fig. 6 shows the influence of spectator holes as observed for sample $H2$. The spectra of positively and negatively charged QD's are quite different with respect to both the saturation of absorption and the renormalization of the transition energies, as depicted in Figs. 6(d) and 6(f), respectively. The additional holes affect similarly the excitation resonances $1a$ and $2a$ as well as $1b$ and $2b$. This becomes particularly obvious from the renormalization of the transition energies leading, respectively, to a red shift and blue shift, Fig. 6(f). Additionally, the quenching is more pronounced for the *a* components than for the *b* components. The results suggest that the subpeaks *a* and *b* involve different hole levels. The only partly quenched subpeaks *a* and *b* support the estimate of approximately seven holes per completely charged QD as extracted from the C-V data (Sec. III A).

D. Predicted absorption spectra of charged QD's

The renormalization of the excited-state transition energies in charged QD's is a consequence of the Coulomb interaction between the localized carriers and the altered barrier potential due to band bending caused by the Coulomb charging. The latter leads to a transition from a localized to a resonant character of the higher excited states, which are lifted above the GaAs band edge. Experiments, as well as calculations considering realistic structural properties for the QD's, have already demonstrated that the properties of the ground state of few-particle complexes depends sensitively on the structural properties of the QD's, leading to either blue or red shifts for one and the same few-particle state in differently shaped QD's.[12] The reason is that the direct Coulomb energies depend sensitively on the difference between

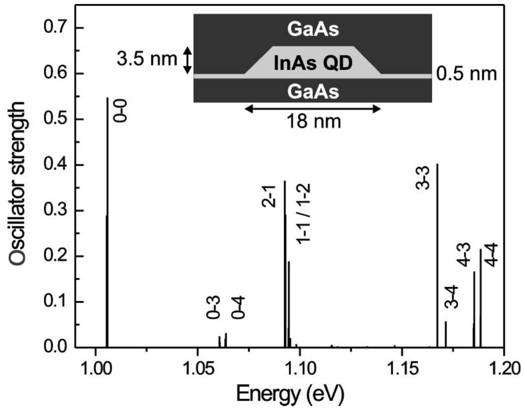

FIG. 7. Predicted absorption spectrum of the neutral QD. The transitions are labeled with the dominant participating electron and hole levels. The inset shows a schematic of the calculated InAs/GaAs QD.

the electron and hole charge distributions, and that correlation and renormalization depend inversely on the quantization, i.e., the kinetic confinement energy and the sublevel splitting. It is shown in the following that the behavior of the excited states can be qualitatively understood by a comparison to absorption spectra predicted for positively and negatively charged QD's.

For the calculations, a truncated pyramidal InAs/GaAs QD with a baselength (height) of 18 nm (3.5 nm) was assumed guided by the QD's in the investigated samples.[40] The structure is shown schematically in the inset of Fig. 7. The calculations were performed based on the eight-band $\mathbf{k}\cdot\mathbf{p}$ model used previously to successfully predict the properties of pyramidal InAs/GaAs QD's.[11,41] The few-particle states are calculated by evaluating the Coulomb interaction in a configuration-interaction scheme including correlation and exchange.[11] The exciton absorption spectrum of the neutral QD is shown in Fig. 7, whereby the height of the bars represents the oscillator strength. The transitions are labeled according to the dominating single-particle components. The flat, truncated shape reduces the amplitude of the piezoelectric potential and the strain-induced vertical modulation of the confinement potential, compared to the situation in pyramidal QD's, such that allowed transitions arranged in line groups dominate the spectrum.[10]

Figure 8 compares the absorption spectra of negatively [panel (a)] and positively [panel (b)] charged QD's with up to three spectator charges. The transitions have been artificially broadened using Gaussians with a full width at half maximum of 10 meV in order to sum up contributions of subcomponents that are not resolved in the experiments. The calculated spectra show clearly the quenching of the ground-state absorption observed in the experiments and the shift of the transition energies. Charging the QD with up to three carriers reduces also the oscillator strength of the $n=2$ excited-state transitions, as experimentally observed. The calculated spectra show red shifts up to $\sim$9 meV for the excited-state transitions in case of negatively charged QD's, whereby the population of the ground state already leads to a pronounced renormalization. The renormalization is much smaller for positively charged QD's and recovers for the $n=1$ state transition of the triply charged QD.

IV. CONCLUSION

Excited-state transitions in positively and negatively charged $In_xGa_{1-x}As/GaAs$ QD's have been investigated by size-selective PLE spectroscopy and eight-band $\mathbf{k}\cdot\mathbf{p}$ model calculations. The results clearly identify few-particle effects

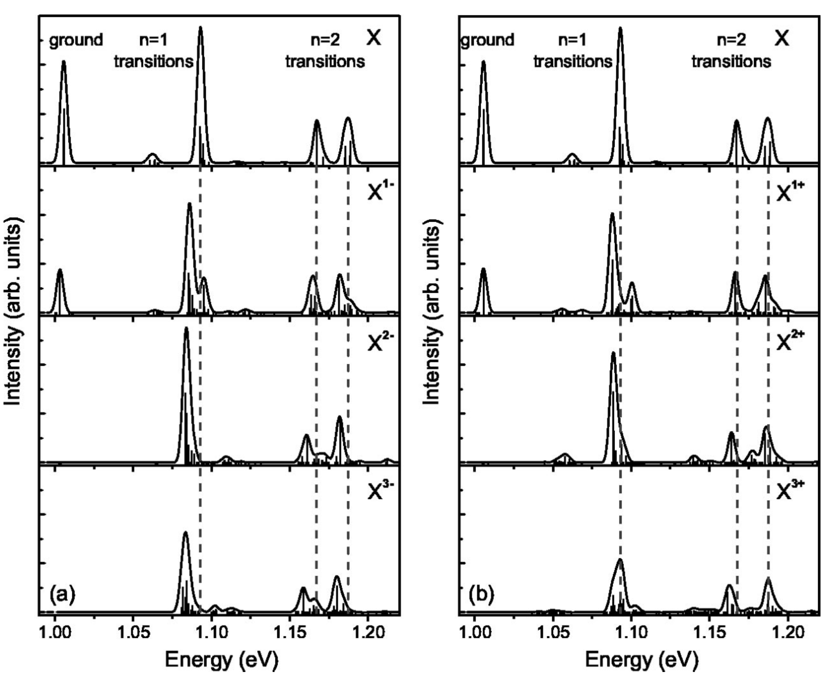

FIG. 8. Absorption spectra calculated in an eight-band $\mathbf{k}\cdot\mathbf{p}$ model for a neutral QD and a QD charged by up to three spectator electrons [panel (a)] or spectator holes [panel (b)]. In addition to the δ-type absorption lines, the spectra were broadened by 10 meV to have a linewidth comparable to the experimental results.

for the excited states, which are strongest when filling the ground state with spectator charges. Successive quenching and renormalization of PLE resonances allow to identify transitions corresponding to specific electron and hole levels. The band bending in the vicinity of the QD's due to Coulomb charging alters the confinement due to the formation of a "triangular" barrier, which reduces the confinement of higher excited states. The Coulomb charging limits the number of localized states and therewith the maximum charging of the QD's. However, the resonant excited states above the GaAs barrier are still observed in PLE, indicating carrier relaxation to be faster than tunnel escape.

ACKNOWLEDGMENTS

We thank Lutz Müller-Kirsch for growing samples $E2$ and $H2$. Parts of this work were supported by Deutsche Forschungsgemeinschaft in the framework of Grant No. SFB 296, NanOp Center of Competence, INTAS Project No. 2001-774, and the Nanomat project of the European Commission's Growth Programme, Contract No. G5RD-CT-2001-00545. Parts of the electronic structure calculations were performed on the Cray T3E computer of the Konrad-Zuse-Zentrum für Informationstechnik Berlin within Project No. Bvpt13.

*Electronic address: florian@sol.physik.tu-berlin.de

[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, Chichester, 1999).

[2] M. Grundmann, Physica E (Amsterdam) **5**, 167 (2000), and references therein.

[3] D. Bimberg, M. Grundmann, N.N. Ledentsov, M.H. Mao, Ch. Ribbat, R. Sellin, V.M. Ustinov, A.E. Zhukov, Zh.I. Alferov, and J.A. Lott, Phys. Status Solidi B **224**, 787 (2001).

[4] J.C. Campbell, D.L. Huffaker, H. Deng, and D.G. Deppe, Electron. Lett. **33**, 1337 (1997).

[5] H.C. Liu, M. Gao, J. McCaffrey, Z.R. Wasilewski, and S. Fafard, Appl. Phys. Lett. **78**, 79 (2001).

[6] G. Yusa and H. Sakaki, Appl. Phys. Lett. **70**, 345 (1997).

[7] H. Pettersson, L. Baath, N. Carlsson, W. Seifert, and L. Samuelson, Appl. Phys. Lett. **79**, 78 (2001).

[8] J.J. Finley, M. Skalitz, M. Arzberger, A. Zrenner, G. Böhm, and G. Abstreiter, Appl. Phys. Lett. **73**, 2618 (1998).

[9] S. Schmitt-Rink, D.A.B. Miller, and D.S. Chemla, Phys. Rev. B **35**, 8113 (1987).

[10] O. Stier, R. Heitz, A. Schliwa, and D. Bimberg, Phys. Status Solidi A **190**, 477 (2002).

[11] O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999).

[12] O. Stier, A. Schliwa, R. Heitz, M. Grundmann, and D. Bimberg, Phys. Status Solidi B **224**, 115 (2001).

[13] F. Findeis, M. Baier, A. Zrenner, M. Bichler, G. Abstreiter, U. Hohenester, and E. Molinari, Phys. Rev. B **63**, 121309 (2001).

[14] V. Türck, Elektronische Eigenschaften Einzelner Halbleiterquantenpunkte (Mensch und Buch Verlag, Berlin, 2001).

[15] R.J. Warburton, C. Schäflein, D. Haft, F. Bickel, A. Lorke, K. Karrai, J.M. Garcia, W. Schoenfeld, and P.M. Petroff, Nature (London) **405**, 926 (2000).

[16] J.J. Finley, P.W. Fry, A.D. Ashmore, A. Lemaitre, A.I. Tartakovskii, R. Oulton, D.J. Mowbray, M.S. Skolnick, M. Hopkinson, P.D. Buckle, and P.A. Maksym, Phys. Rev. B **63**, 161305 (2001).

[17] D.V. Regelman, E. Dekel, D. Gershoni, E. Ehrenfreund, A.J. Williamson, J. Shumway, A. Zunger, W.V. Schoenfeld, and P.M. Petroff, Phys. Rev. B **64**, 165301 (2001).

[18] J.J. Finley, A.D. Ashmore, A. Lemaitre, D.J. Mowbray, M.S. Skolnick, I.E. Itskevich, P.A. Maksym, M. Hopkinson, and T.F. Krauss, Phys. Rev. B **63**, 073307 (2001).

[19] R.J. Warburton, C.S. Dürr, K. Karrai, J.P. Kotthaus, G. Medeiros-Ribeiro, and P.M. Petroff, Phys. Rev. Lett. **79**, 5282 (1997).

[20] K. Goede, A. Weber, F. Guffarth, C.M.A. Kapteyn, F. Heinrichsdorff, R. Heitz, D. Bimberg, and M. Grundmann, Phys. Rev. B **64**, 245317 (2001).

[21] R. Heitz, O. Stier, I. Mukhametzhanov, A. Madhukar, and D. Bimberg, Phys. Rev. B **62**, 11 017 (2000).

[22] O. Stier, *Electronic and Optical Properties of Quantum Dots and Wires* (Wissenschaft und Technik Verlag, Berlin, 2001).

[23] I. Mukhametzhanov, R. Heitz, J. Zeng, P. Chen, and A. Madhukar, Appl. Phys. Lett. **73**, 1841 (1998).

[24] F. Heinrichsdorff, *MOCVD Growth and Laser Applications of In(Ga)As/GaAs Quantum Dots* (Mensch und Buch Verlag, Berlin, 1998).

[25] A. Krost, J. Blasing, F. Heinrichsdorff, and D. Bimberg, Appl. Phys. Lett. **75**, 2957 (1999).

[26] H. Eisele, O. Flebbe, T. Kalka, F. Heinrichsdorff, A. Krost, D. Bimberg, and M. Dähne-Prietsch, Phys. Status Solidi B **215**, 865 (1999).

[27] R. Heitz, I. Mukhametzhanov, P. Chen, and A. Madhukar, Phys. Rev. B **58**, 10 151 (1998).

[28] R. Sellin, N.N. Ledentsov, D. Bimberg, V.M. Ustinov, and Z.I. Alferov, in *Proceedings of the Sixth International Symposium in Advanced Physics of Fields*, edited by N. Koguchi (National Research Institute for Metals, Tsukuba, 2001), p. 49.

[29] R. Wetzler, A. Wacker, E. Schöll, C.M.A. Kapteyn, R. Heitz, and D. Bimberg, Appl. Phys. Lett. **77**, 1671 (2000).

[30] C.M.A. Kapteyn, M. Lion, R. Heitz, D. Bimberg, P.N. Brunkov, B.V. Volovik, S.G. Konnikov, A.R. Kovsh, and V.M. Ustinov, Appl. Phys. Lett. **76**, 1573 (2000).

[31] W.-H. Chang, T.M. Hsu, N.T. Yeh, T.E. Nee, and J.-I. Chyi, Phys. Rev. B **62**, 13 040 (2000).

[32] The number of carries can by extracted from the C-V data using $n=(e\rho A)^{-1}\int_{U1}^{U2}CdU$, with e being the elementery charge, ρ the QD density, and A the sample area.

[33] V. Türck, F. Heinrichsdorff, M. Veit, R. Heitz, M. Grundmann, A. Krost, and D. Bimberg, Appl. Surf. Sci. **123/124**, 352 (1998).

[34] P.W. Fry, I.E. Itskevich, D.J. Mowbray, M.S. Skolnick, J.J. Finley, J.A. Barker, E.P. O'Reilly, L.R. Wilson, I.A. Larkin, P.A. Maksym, M. Hopkinson, M. Al-Khafaji, J.P.R. David, A.G. Cullis, G. Hill, and J.C. Clark, Phys. Rev. Lett. **84**, 733 (2000).

[35] R. Heitz, F. Guffarth, I. Mukhametzhanov, M. Grundmann, A. Madhukar, and D. Bimberg, Phys. Rev. B **62**, 16 881 (2000).

[36] H. Sakai, T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Amano, and I. Akasaki, J. Cryst. Growth **189–190**, 831 (1998).

[37] O. Ambacher, R. Dimitrov, M. Stutzmann, B.E. Foutz, M.J. Murphy, J.A. Smart, J.R. Shealy, N.G. Weimann, K. Chu, M. Chumbes, B. Green, A.J. Sierakowski, W.J. Schaff, and L.F.

Eastman, Phys. Status Solidi B **216**, 381 (1999), and references therein.

[38] B.T. Miller, W. Hansen, S. Manus, R.J. Luyken, A. Lorke, J.P. Kotthaus, S. Huant, G. Medeiros-Ribeiro, and P.M. Petroff, Phys. Rev. B **56**, 6764 (1997).

[39] G. Medeiros-Ribeiro, D. Leonard, and P.M. Petroff, Appl. Phys. Lett. **66**, 1767 (1995).

[40] The limited knowledge of the actual structural properties of the QD's in the samples does not allow to reasonably exploit the differences in the investigated samples in the calculation. The calculations provide a qualitative explanation comparing the different impact of spectator electrons and holes.

[41] M. Grundmann, O. Stier, and D. Bimberg, Phys. Rev. B **52**, 11 969 (1995).