Direct Quantitative Electrical Measurement of Many-Body Interactions in Exciton Complexes in InAs Quantum Dots

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We present capacitance-voltage spectra for the conduction band states of InAs quantum dots obtained under continuous illumination. The illumination leads to the appearance of additional charging peaks that we attribute to the charging of electrons into quantum dots containing a variable number of illumination-induced holes. By this we demonstrate an electrical measurement of excitonic states in quantum dots. Magnetocapacitance-voltage spectroscopy reveals that the electron always tunnels into the lowest electronic state. This allows us to directly extract, from the highly correlated many-body states, the correlation energy. The results are compared quantitatively to state of the art atomistic configuration interaction calculations, showing very good agreement for a lower level of excitations and also limitations of the approach for an increasing number of particles. Our experiments offer a rare benchmark to many-body theoretical calculations.

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Self-assembled quantum dots (QDs) have been intensively studied in the past as model systems for three-dimensional carrier confinement, as well as for applications in novel technologies such as quantum communication [1–7]. In contrast to the situation in atoms, both the carrier-carrier Coulomb interaction and the interlevel energy spacing due to the lateral confinement in QDs are in the same order of magnitude, around 10–30 meV [8,9]. This situation is ideal to study strong electronic correlations, where the ratio between Coulomb energy and kinetic energy is large [10–12]. Correlation effects such as Wigner localization [13] or the violation of the Aufbau principle [9] have been investigated theoretically making successful connection to experiments. However, the evidence for correlations remains indirect in these experiments and a quantitative estimate of the correlation energy is difficult because it is intermingled with quasiparticle noncorrelated quantities. Furthermore, whereas the repulsive interaction between carriers carrying the same charge can be rather directly accessed via capacitance-voltage (C-V) spectroscopy using the Coulomb blockade effect [8,14,15], the attractive interaction between carriers of opposite charges, as given in excitons or charged excitons, is more difficult to determine. Indeed, in optical experiments, these energies are always measured together with intraband (electron-electron or hole-hole) Coulomb interactions [8,16]. In this paper we show that C-V spectroscopy of a single layer of QDs occupied by illumination-induced holes leads to five clearly resolved charging peaks. Our analysis reveals that these peaks correspond to charging events into the lowest electron state in QDs occupied by one to five holes. A theoretical analysis of the results shows that this type of experiment allows us to extract the attractive Coulomb interaction energies as well as the subtle electronic correlation energy [10,11].

It was shown in single QD photoluminescence (PL) experiments [16–19] that in n-type QD heterostructures the QDs can be charged with holes by illumination. The number of holes per QD can be controlled via a gate voltage. From the gate voltage dependence of the PL spectra, Dalgarno and co-workers [17] deduced the electron-hole Coulomb interaction for the neutral exciton X^0 and the negatively charged exciton X^{1-}. Experiments combining illumination to induce holes with n-type C-V spectroscopy have been reported over a decade ago [20,21], however without resolving individual peaks. Our samples show very small inhomogeneous broadening of the charging peaks, so that the addition of a single electron per QD is well resolved.

We studied standard charge-tunable devices [see Fig. 1(a)] with a sequence of the relevant layers as follows: a 300 nm thick highly n-doped back contact is followed by 25 nm GaAs acting as tunneling barrier. Then a single layer of InAs QDs was grown followed by 11 nm GaAs, 41 periods of a 3 nm AlAs/1 nm GaAs short period superlattice (SPS), and a 10 nm GaAs cap layer. Keeping the GaAs layer after the QDs as thin as possible is essential to avoid charge accumulation at the interface to the AlAs/GaAs superlattice, which was observed in [21] and inhibits the controlled charging with illumination-induced holes in the QDs. In this Letter we discuss three different samples: in sample A no InAs at all was introduced in the structure, for sample B only a wetting layer was grown and sample C contains a single layer of QDs. Only sample C shows illumination-related charging peaks (see Fig. 2 and Fig. S1 in the Supplemental

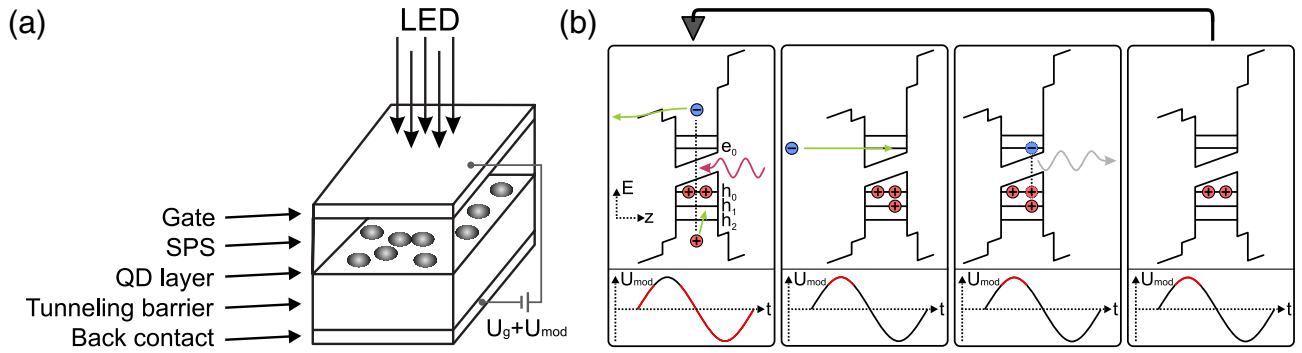

FIG. 1 (color online). (a) We apply an ac voltage U_{mod} to the top gate and measure the capacitance as a function of the dc voltage U_g. Throughout the measurement a constant optical excitation is realized by an LED. (b) Schematic (not to scale) of the illumination-induced hole capture process and C-V spectroscopy ($E \triangleq$energy, $z \triangleq$growth direction). The photons generate electron-hole pairs which separate due to the internal electrical field. The electrons move towards the back contact, while the holes are trapped inside the QDs. In the special case shown, the QD level comes for the given dc bias in resonance with the back contact in the upper part of the ac cycle. The red part of the sine indicates in which part of the ac cycle (U_{mod} as a function of time t) the corresponding QD configuration shown directly above is valid.

Material [22]). Thus, we infer that these peaks are closely connected to the QDs themselves.

For C-V spectroscopy either a standard LCR meter or a lock-in amplifier has been used and all measurements have been performed at 4.2 K. The samples were illuminated through semitransparent Au gates (20 nm thick) by a light emitting diode (LED) mounted closely to the sample surface and driven by a constant current. The results shown here were obtained employing a conventional infrared LED having its radiation maximum at 920 nm at 4.2 K. Further information on the sample structure and the experimental details is given in the supplementary material.

Figure 2 shows the capacitance as a function of the gate voltage for sample C for various illumination intensities. We could resolve up to five peaks in addition to the s_1 and s_2 charging peaks. Whereas it is well known that the peaks labeled s_1 and s_2 correspond to the charging of the twofold degenerated s-like ground-state with the first (s_1) or the second (s_2) electron [8,14], the illumination-induced peaks seem to be related to the charging of QDs occupied by photo-generated holes. As already shown [8,15,16,18,23–25], the equilibrium number of holes stored in the QD can be adjusted through the gate bias. Each gate voltage interval corresponds to a certain equilibrium number of holes, where a larger reverse-bias results in more holes. Other experiments have shown that QDs similar to the ones studied in this work can be charged with up to eight holes [15].

The observed additional peaks in the capacitance occur if the Fermi energy in the back contact comes into resonance with the QD ground state and an electron tunnels into the QD so that an exciton complex is formed. Because of the attractive Coulomb interaction, the resonance condition is fulfilled at lower gate voltages, compared to QDs without holes [16]. For an increasing number of holes, the gate voltage necessary for electron-tunneling decreases. Thus, each additional peak corresponds to the tunneling into a many-body state with a different number of holes. The final states are, accordingly, different positively charged exciton complexes. The formation of the capacitive signal is depicted schematically in Fig. 1 and is discussed in more detail in the Supplemental Material [22].

To support our assignment of the illumination-related peaks given above, we performed magneto-C-V-spectroscopy with a magnetic field perpendicular to the QD layer [14]. We employed constant optical excitation and varied the magnetic field from 0 to 5.5 T. In Fig. 3, one clearly sees the well-known dispersion behavior [8,14] for the s and p-states. The p states shift in energy due to the orbital Zeemann effect [8,14], whereas for the s-like ground state no dispersion is resolved due to the small spin-Zeeman effect. The lack of dispersion in the illumination-induced

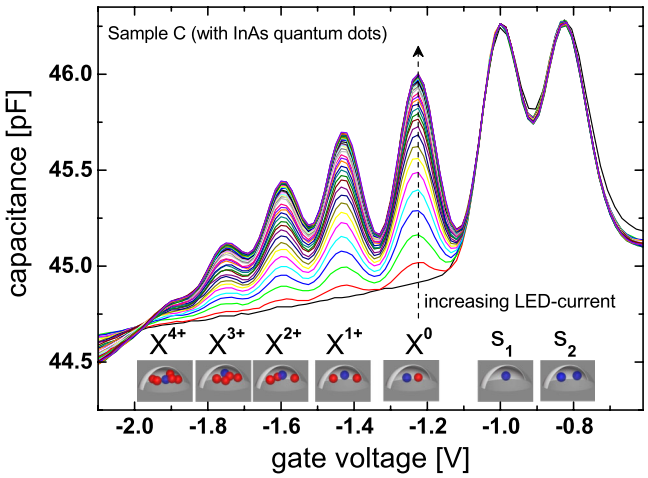

FIG. 2 (color online). Capacitance-voltage measurements under continuous illumination with various intensities on sample C (with QDs). We resolve five additional illumination-induced charging peaks, their occupancy by holes and electrons are indicated schematically by spheres.

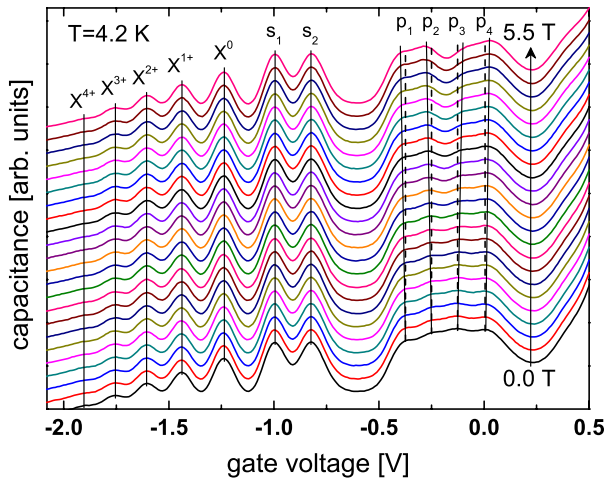

FIG. 3 (color online). Capacitance-voltage measurements under illumination ($I = 72$ W/m^2) with magnetic fields B ranging from 0 to 5.5 T (in steps of 0.5 T) perpendicular to the QD layer. Vertical dashed lines are guides to the eye.

charging peaks proves our assumption that the electron tunnels, for all these peaks, into the s-like QD ground state.

To obtain quantitative information on the interaction energies from the C-V spectra, we extracted the charging energies by converting the gate voltage scale to an energy scale employing a simple lever arm approach [26]. We use the following notation:

$$\begin{aligned}
\Delta E(s_2) &= E(2e,0h) - E(1e,0h)\\
\Delta E(s_1) &= E(1e,0h) - E(0e,0h)\\
\Delta E(X^0) &= E(1e,1h) - E(0e,1h)\\
\Delta E(X^{1+}) &= E(1e,2h) - E(0e,2h)\\
\Delta E(X^{2+}) &= E(1e,3h) - E(0e,3h)\\
\Delta E(X^{3+}) &= E(1e,4h) - E(0e,4h)\\
\Delta E(X^{4+}) &= E(1e,5h) - E(0e,5h),
\end{aligned}$$

where $E(Ne,Mh)$ stands for the many-body total energy of the QD filled with N electrons and M holes. Our situation differs from the case of photoluminescence experiments, where the energy differences involve a change of both, the number of electrons and holes [8,16].

The charging energies ΔE, which correspond to the resonance energy allowing the electron to tunnel, decrease progressively with the number of photo-induced holes. This represents the increase in Coulomb attraction felt by the tunneling electron, with an increased number of resident holes. How this attraction varies as a function of the number of holes is best seen in the addition energies, which are defined as the energy differences between the individual charging peaks and given in Table I.

In Table I we report the addition energies, which describe by how much easier the electron can tunnel, when the number of resident holes has increased by one. This

TABLE I. Comparison between experimental and theoretical addition energies. The experimental values have been extracted from the spectra by converting the differences in charging voltage between consecutive peaks to an energy interval (in meV) by employing a simple lever arm law [26]. Theory 1 and 2 correspond to two different QDs described in the text.

Peak complex	Theory 1	Theory 2	Experiment
$\Delta E(s_2) - \Delta E(s_1)$	19.2	20.2	20.7 ± 0.1
$\Delta E(s_1) - \Delta E(X^0)$	22.6	28.9	27.5 ± 0.3
$\Delta E(X^0) - \Delta E(X^{1+})$	21.3	24.8	24.0 ± 0.2
$\Delta E(X^{1+}) - \Delta E(X^{2+})$	19.4	19.4	20.2 ± 0.2
$\Delta E(X^{2+}) - \Delta E(X^{3+})$	18.7	18.6	19.3 ± 0.5
$\Delta E(X^{3+}) - \Delta E(X^{4+})$	18.6	18.2	19.0 ± 2.0

quantity can be separated into three contributions. One is the direct Coulomb interaction J_{eh} between the electron tunneling into the orbital s-state e_0 (see Fig. 1) and the additional hole residing either in the single particle state h_0 (for X^0, X^{1+}), h_1 (for X^{2+}, X^{3+}) or h_2 (for X^{4+}). Accordingly, this first contribution to the addition energies is given by

$$\begin{aligned}
\Delta E(s_1) - \Delta E(X^0) &\sim -J_{e0h0}\\
\Delta E(X^0) - \Delta E(X^{1+}) &\sim -J_{e0h0}\\
\Delta E(X^{1+}) - \Delta E(X^{2+}) &\sim -J_{e0h1}\\
\Delta E(X^{2+}) - \Delta E(X^{3+}) &\sim -J_{e0h1}\\
\Delta E(X^{3+}) - \Delta E(X^{4+}) &\sim -J_{e0h2}.
\end{aligned}$$

At this level of the theory, the addition energies show degeneracies such as $\Delta E(s_1) - \Delta E(X^0) = \Delta E(X^0) - \Delta E(X^{1+})$ and $\Delta E(X^{1+}) - \Delta E(X^{2+}) = \Delta E(X^{2+}) - \Delta E(X^{3+})$. The second contribution is due to the electron-hole exchange interaction, which is only present if the photogenerated holes are in an open-shell configuration. This energy is typically two orders of magnitude smaller than the addition energy and is, although included in our subsequent calculations, irrelevant in the present discussion. The third contribution is given by the correlation between the carriers. The corresponding correlation energy is generally larger for a larger number of particles. In a schematic picture, this latter effect tends to increasingly delocalize the additional hole wave function, with an increasing number of holes inside the QD. The overlap between the electron and this *effective* additional hole is therefore reduced, as is the magnitude of the many-body Coulomb integral.

To obtain a clear interpretation of the experimental results, we performed theoretical calculations of the exciton complex and extracted from those the addition energies previously defined. The single particle orbitals and energies of the QD are calculated using the atomistic empirical pseudopotential approach [27–29] for a structure with about 3 million atoms, taking strain, band coupling,

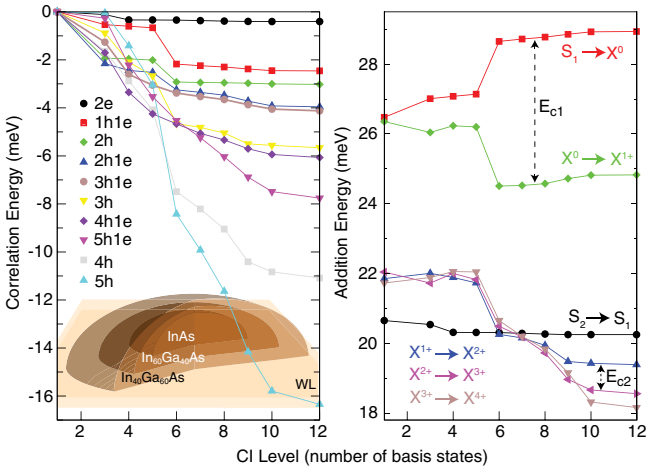

FIG. 4 (color online). Correlation (left) and addition (right) energies calculated with a varying number of basis states in a CI approach, for an InGaAs QD with a graded In composition (see inset in left panel). For five holes and one electron ($5h1e$) using 12 basis states, the number of configurations amounts to 1,020,096, which is our present computational limitation.

coupling between different parts of the Brillouin zone and spin-orbit coupling into account. The Coulomb and exchange integrals are calculated from the atomic wave functions and the correlated excitonic states are calculated by the configuration interaction method (CI) [22,28].

In Table I, we show results for an $In_{0.7}Ga_{0.3}As/GaAs$ QD with homogeneous composition, a circular base and an overall lens shape with a diameter of 25 nm and a height of 3.5 nm (Theory 1) and for an InGaAs QD consisting of three nested lenses: a pure InAs core with 12 nm diameter and 2 nm height, a 60% In intermediate region with 23 nm diameter and 3.1 nm height and a 40% In outer shell with 31 nm diameter and 11 nm height (Theory 2). Such a composition gradient is inspired by results of cross-sectional scanning tunneling microscopy [30,31].

Table I shows that the theoretical results for the QD with a graded In composition profile (Theory 2) agrees very well with the experimental results: the electron-electron repulsion (first row of Table I) is weaker than the electron hole attraction (second row), which is in agreement with previous experimental findings [16,18]. Addition energies progressively reduce with increasing number of holes in agreement with the experimental results. One of the unique features of our experiment is the direct measurement of the correlation energy. This energy is entirely determined, as we have argued above, by the energy differences between $\Delta E(X^0) - \Delta E(s_1)$ and $\Delta E(X^{1+}) - \Delta E(X^0)$ and between $\Delta E(X^{2+}) - \Delta E(X^{1+})$ and $\Delta E(X^{3+}) - \Delta E(X^{2+})$. To show that these differences are indeed purely given by correlation effects, we plot in Fig. 4 the correlation energy (left) and the addition energy (right) as a function of the number of basis states used in the CI expansion. At the CI level 1, correlations are exactly ignored while CI level 12 represents the largest CI level available to us. We see that the correlation energy for the most highly charged states can amount to 16 meV, comparable to the 20 meV of direct Coulomb interaction between electrons. These states are highly correlated states. The right panel of Fig. 4 shows that the addition energies $[\Delta E(X^0) - \Delta E(s_1)]$ and $[\Delta E(X^{1+}) - \Delta E(X^0)]$ are equal at the uncorrelated level (CI level = 1) and progress to their converged values, given in Table I, for an increased number of basis states. The splitting between these addition energies (E_{c1} in Fig. 4) is therefore purely a correlation effect. This splitting can directly be accessed by our experiment and amounts to 3.5 meV. Our theoretical result of 4.1 meV (Theory 2) is in good agreement with this value. The two electron, the exciton (1h1e) and the two hole cases are well converged at the CI level 12. The other addition energy splitting, labeled E_{c2}, which is purely given by correlation as well, amounts to 0.8 meV theoretically, in good agreement with the measured 0.9 meV. However, this splitting involves the energy of the 4h many-body state and the left panel of Fig. 4 shows that this quantity is not converged. Indeed, even at our largest (full) CI calculation involving over one million configurations, the magnitude of the correlation energy still increases with increasing CI level. This comparison demonstrates that state of the art many-body approaches, such as configuration interaction using a truncated basis set [28] are well suited for a small number of carriers, such as X^{1+} with two holes and one electron, but should be regarded critically for more particles, such as in X^{3+} with four holes and one electron. A stochastic sampling of the Slater determinant space [32], allowing for calculations at a significantly larger CI level, may be a worthwhile avenue to explore in the future.

In summary, C-V spectroscopy under illumination represents a powerful tool to study the interaction of a single electron with a variable and controllable (until five in our experiment) number of holes in a nanostructure. The measured variations in the addition energy as a function of the number of holes residing in the QD, gives information about the electron-hole pair Coulomb interaction, and most interestingly, enables a rare direct measurement of the many-body correlation energy. We show that many-body effects modify the addition energy by up to 20%, which is due to the highly correlated nature of the many-body states where the direct Coulomb interaction (20 meV for electron-electron interaction) is in the same order of magnitude as the correlation energy (16 meV for 5h). The experimental principle of combining optical carrier generation with electrical measurements can also be applied to other QD systems, for example GaAs QDs prepared by droplet epitaxy [33], InGaN QDs, or even indirect systems such as Ge QDs in Si [34,35].

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