Hole levels in InAs self-assembled quantum dots

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We measure the energy levels and charging spectra of holes in self-assembled InAs quantum dots using capacitance-voltage and polarized photoluminescence spectroscopy in high magnetic fields. The pronounced circular polarization of the optical emission, together with the optical selection rules for orbital and spin quantum numbers, allows us to separate the individual electron and hole levels. The magnetic field dependence of the single-particle hole energy levels can be understood by considering a spin-orbit coupled valence band and agrees well with the observed behavior of the charging peaks in the capacitance-voltage spectra.

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Three-dimensional spatial confinement in semiconductor quantum dots (QDs) leads to discrete energy levels that can be filled consecutively by charge carriers.[1] To fully exploit the properties of QDs for advanced applications,[2,3] it is important to understand their energetic shell structure and charging sequence. To date, electron systems were mainly investigated, both in lithographically defined[4] as well as in self-assembled QDs.[5–7] It was found that charging QDs resembles shell filling (s, p, d, etc.) in atoms, while describing the lateral confining QD potential by a two-dimensional (2D) harmonic oscillator and treating the interactions between electrons as a perturbation.[4,6] In this Brief Report, we investigate self-assembled InAs QDs filled with *holes* that exhibit a different behavior.[8–12] The main difference arises from the complicated valence-band structure, which is governed by multiple, spin-orbit coupled bands[12–14] rather than the single band for electrons. Furthermore, for holes the quantization energy is comparable to the Coulomb-interaction energies, which can, therefore, no longer be treated as a perturbation. These aspects make the energy structure and charging sequence of hole systems interesting from a fundamental point of view to explore novel carrier configurations that defy rules of atomic systems.[8,10,11] In addition, knowledge of the hole energy levels is crucial for the optical properties of QDs,[15–20] originating from electron-hole recombination.

We use capacitance-voltage (C-V) and polarized photoluminescence (PL) spectroscopy in high magnetic fields (B) to identify the QD energy levels. Previously, high field energy spectra of electron[4,6] and exciton[6,17,21] systems were analyzed within the Fock-Darwin (FD) model,[22,23] i.e., a 2D harmonic oscillator in a perpendicular magnetic field. Using this description, recent C-V experiments on p-type InAs QDs have suggested an incomplete shell filling, which means that the d shell starts filling before the p shell is full.[8,24] A different, yet similar, anomalous filling due to hole correlation effects was found theoretically by He *et al.*[10,11] using an atomistic pseudopotential approach. An alternative explanation was given by Climente *et al.*[12] in terms of the single-particle hole levels.

Contrary to previous magneto-PL experiments on QDs,[17,20] we explore the profound circular polarization of the PL emission. By using the optical selection rules, we determine the angular momentum of the confined electron and hole energy levels. We find that, unlike the electron levels, the hole levels cannot be described within the FD model, but require a description that takes into account the spin-orbit coupling of the valence band.[12] The resulting hole energy level diagram is consistent with the C-V charging experiments on the same sample, without the need to invoke anomalous filling due to hole correlation effects.

The p-type InAs QD sample was grown by solid source molecular beam epitaxy, and details of the growth can be found elsewhere.[8,9,25] From the samples, optically accessible Schottky diodes were prepared with semitransparant $300 \times 300\ \mu m^2$ indium tin oxide top gates. C-V and polarized PL experiments were performed on the same gate at 4.2 K using a standard capacitance bridge (Andeen-Hagerling AH2500) and a standard PL setup [Ti:sapphire laser excitation (spot size $\sim 10\ \mu m$), single grating spectrometer, liquid-nitrogen-cooled InGaAs detector] respectively.

The optimized growth procedure resulted in sufficiently small inhomogeneous level broadening to enable the observation of individual charging peaks in C-V and narrow, well-separated PL peaks in ensemble measurements.[25] Figure 1 shows typical C-V traces [Fig. 1(a)] and PL spectra [Fig. 1(b)] recorded for magnetic fields up to 28 T, applied parallel to the growth direction. A linear background was subtracted from each C-V curve to obtain the capacitance signal of the QDs. The signal exhibits six clear charging peaks, each of which corresponds to the charging of one additional hole per QD. With increasing magnetic field, each charging peak displays a characteristic shift. These shifts in voltage ΔV_g are transformed to shifts in energy ΔE by the usual lever arm law: $\Delta E = \lambda^{-1} e \Delta V_g$, where λ^{-1} is the ratio between the thickness of the tunneling barrier (19 nm) and the distance between the back contact and the surface (168 nm).[6,9] This analysis reveals a behavior that was also found in previous C-V measurements on a similar sample.[8] Peaks 1 and 2 exhibit an opposite, but small shift (± 0.02 meV/T). At low fields (<12 T), peaks 3 and 5 (4 and 6) shift to lower

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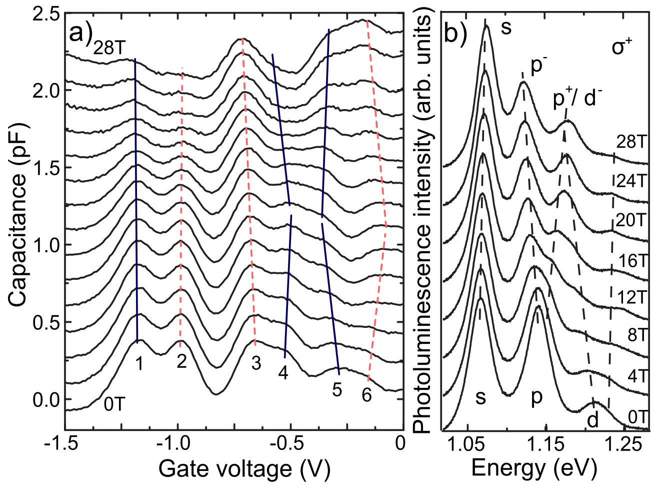

FIG. 1. (Color online) (a) C-V traces at $T=4.2$ K for p-type InAs QDs in magnetic fields from 0 to 28 T (2 T steps). The curves are vertically offset for clarity and the lines are calculated using the model for hole filling outlined in the text. (b) Magnetic field dependent σ^{+}-polarized PL spectra of the QD ensemble ($\lambda_{\mathrm{exc}}=734.8$ nm, $T=4.2$ K, $V_g=0.0$ V, and $P_{\mathrm{exc}}\sim 50$ W cm^{-2}). The dashed lines are a guide for the eyes.

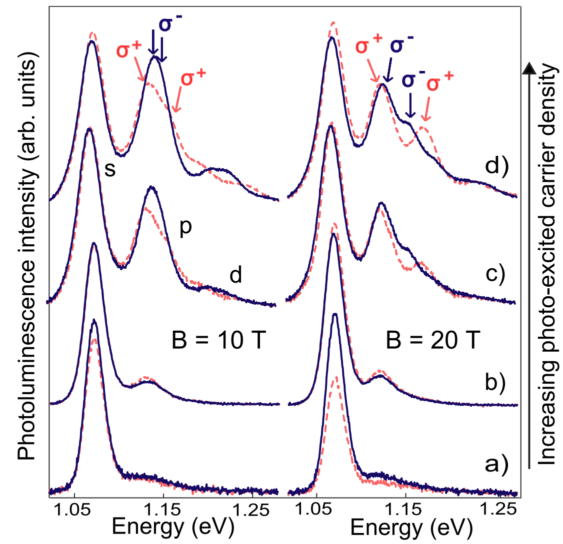

FIG. 2. (Color online) Circular polarized PL spectra of InAs QDs for different photoexcited carrier densities at $B=10$ T and $B=20$ T ($T=4.2$ K and $V_g=0.0$ V). λ_{exc} and P_{exc} are 831.0 nm and $\sim$10 W cm^{-2} for (a), 809.1 nm and $\sim$50 W cm^{-2} for (b), 734.8 nm and $\sim$50 W cm^{-2} for (c), and 734.8 nm and $\sim$80 W cm^{-2} for (d).

(higher) fields, while the slopes of peaks 5 and 6 ($\pm$0.38 meV/T) are twice those of peaks 3 and 4 ($\pm$0.20 meV/T). Above 12 T, the shifting direction of peaks 4–6 is reversed. Within the FD model, these shifts are directly related to the orbital angular momentum quantum number l, which would indicate that peaks 1 and 2 are s-like ($l=0$), 3 and 4 are p-like ($l=1$), and 5 and 6 are d-like ($l=2$). These results suggest that the d shell starts filling before the p shell is completely full, in contrast to the rules of sequential filling in atomic physics.

In PL we observe three peaks, which are related to s, p, and d electron-hole transitions. To further characterize the relevant QD energy levels, we follow these peaks as a function of magnetic field for two circular polarizations ($\sigma^{\pm}$) and several photoexcited carrier densities [Figs. 1(b) and 2]. In σ^{+} polarization, the s peak shifts slightly to higher energies and the p peak splits into two separate peaks, one of which crosses a d peak that shifts to lower energies [Fig. 1(b)]. The s, p, and d levels can be consecutively populated with increasing carrier density, realized via variation of both laser excitation power P_{exc} and wavelength λ_{exc}. When the excitation energy is lower than the GaAs energy gap [Fig. 2(a)], carriers are only excited in the QDs and wetting layer, leading to a relatively low carrier density and only s-level recombination. With an excitation energy higher than the GaAs band gap, carriers are excited throughout the GaAs barrier layers and are subsequently captured by the QDs, leading to emission from p and d states as well [Figs. 2(b)–2(d)]. Within the resolution of our ensemble measurements, the PL peak positions were found to be independent of the photoexcited carrier density.[16] Furthermore, our experiments are insensitive to hole-charging effects, since we did not observe a change in the relative energy distances between PL transitions with gate voltage (the number of holes per dot), despite small shifts of the spectra as a whole due to charge accumulation in the structure. A possible reason is the slow hole tunneling through the 19 nm barrier ($\sim$10 μs, estimated from frequency dependent C-V measurements) compared to the carrier relaxation and recombination times (approximately nanoseconds), which means that an average over different charge states is measured on top of the average over different dot dimensions.

Important information is revealed by the pronounced circular polarization of the PL peaks, visible in the spectra at high carrier densities [Figs. 2(c) and 2(d)]. The s peak splits up into two, oppositely polarized, closely spaced peaks. The p transition splits up into four lines with a characteristic sequence of, with increasing energy, $\sigma^{+},\sigma^{-},\sigma^{-},\sigma^{+}$ peaks, indicated by arrows in Fig. 2(d). The PL spectra were fitted by Gaussian peaks of 26 meV width (full width half maximum), resulting in the fan diagram shown in Fig. 3(a). PL series taken at different carrier densities and gate voltages resulted in similar diagrams. The fan diagram resembles the behavior of excitons within the FD model,[17,21] which assumes FD spectra for both carrier types. However, the splitting of the p levels at $B=0$ T and the sequence of polarized peaks already indicate that this is an oversimplification, since the FD model predicts no splitting and a sequence of alternating polarized peaks. To obtain the underlying single-particle levels, we disentangle the electron and hole contributions to the PL energy. For electrons, we take the FD energies:[22,23]

$$E_{l,m_z}=\hbar(l+1)\sqrt{\omega^2+\left(\frac{\omega_c}{2}\right)^2}+\frac{1}{2}\hbar m_z\omega_c+gS_z\mu_B B, \quad (1)$$

where $\omega_c=eB/m^{*}$ is the cyclotron frequency, e is the electron charge, m^{*} is the electron effective mass, g is the Landé factor, and μ_B is the Bohr magneton. ω defines the energy scale due to the lateral confinement and $S_z=\pm 1/2$ is the z component of the electron spin. The energy levels are further

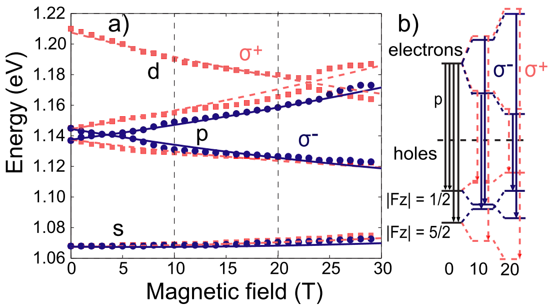

FIG. 3. (Color online) (a) Exciton recombination energies (symbols) in magnetic fields up to 29 T (T=4.2 K, V_g=0.0 V, λ_{exc}=734.8 nm, and P_{exc}=50 W cm^{-2}). The symbol size corresponds to the uncertainty in the energy position (2 meV). The lines are fits of the exciton energies, as outlined in the text. (b) Electron and hole p levels at B=0, 10, and 20 T. The arrows indicate the optically allowed polarized transitions. The polarization of the peaks ($\sigma^+,\sigma^-,\sigma^-,\sigma^+$, in order of increasing energy) is in agreement with the observations.

labeled by their quantum numbers, the orbital quantum number l (l=0,1,2 for the s,p,d shell, respectively) and its z projection $m_z=-l,-l+2,\ldots,l-2,l$. We use $\hbar\omega$=55 meV, $m^*=0.077m_0$ (m_0 is the free electron mass), and g=1.2, which are reasonable values considering previous experiments.[6,15,26–28]

The shifts of the PL lines with field are mainly governed by the FD spectrum of electrons. However, since the electron Zeeman splitting is small, the PL polarization is predominantly determined by the hole levels, as is explained in the following. Due to spin-orbit coupling within the valence band, the effect of which is enhanced due to the lateral confinement,[13,14] m_z and S_z are no longer proper quantum numbers for holes. It is convenient to define the quantum number $F_z=J_z+m_z$, the sum of the z projection of the Bloch angular momentum J_z ($J_z=\pm3/2$ and $\pm1/2$ for heavy and light holes, respectively) and m_z.[12] For each F_z, different combinations of J_z and m_z are possible. As a result of the particular mixture of these combinations, the magnetic field dependence of the energy levels differs strongly from that of a FD model for holes. The effect on the PL polarization is most clearly seen in the behavior of the p states, schematically shown in Fig. 3(b) for B=0, 10, and 20 T. The p-like hole levels, labeled by $|F_z|=1/2$ and 5/2, with a small zero-field splitting due to the coupled valence bands, split up in a field, leading to a characteristic emission pattern as a result of the optical selection rules: $\Delta m_z=0$ and spin conservation $S_z+J_z=\pm1$. To determine the allowed transitions, we use only the dominant m_z contribution of each F_z level. For example, the main contribution to the $F_z=1/2$ (5/2) p-like levels comes from $J_z=3/2$; $m_z=-1$ (+1), which both recombine in σ^+ polarization. In contrast, the $F_z=-1/2$ ($-5/2$) p levels [$J_z=-3/2$; $m_z=+1(-1)$] give σ^- emission. The resulting polarization series with increasing energy is indeed $\sigma^+,\sigma^-,\sigma^-,\sigma^+$ in the 5–25 T field range, as observed experimentally (Fig. 3). In fact, using this model, we are able to quantitatively describe the field dependent exciton energies [lines in Fig. 3(a)]. An additional energy E=1.0125 eV was

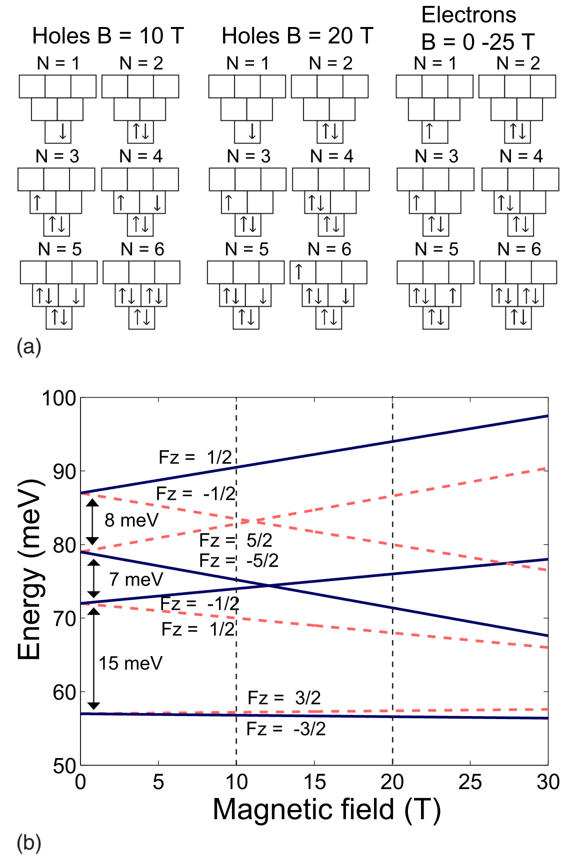

FIG. 4. (Color online) (a) Filling sequence for electrons and holes up to N=6. The up (down) arrows correspond to J_z=3/2 (J_z=−3/2) for holes and S_z=1/2 (S_z=−1/2) for electrons. (b) Single-particle hole energy levels in magnetic field as obtained from the PL spectra. The shifts (in order of increasing energy) are ±0.02, ±0.20, ±0.38, and ±0.35 meV/T.

used to account for the energy gap of InAs, the z-confinement energy, and the exciton binding energy.[29] From this fit we find hole levels that shift with magnetic field in an approximately linear fashion [Fig. 4(b)].[12] The difference in shift of the F_z=1/2 and F_z=5/2 p-like levels is due to the specific mixture of m_z contributions to these levels.

Remarkably, the field dependence of the single-particle hole levels determined by PL agrees very well with the C-V data [see lines in Fig. 1(a)]. Adding a constant Coulomb blockade energy of 18 meV to the zero-field splitting results in the proper spacing between the different charging peaks. The field induced shifts in energy of the C-V charging peaks are nicely reproduced. Finally, the cusps in the shifting direction of peaks 4–6 around 12 T naturally follow from crossings of hole levels. For example, peaks 4 and 5 change shifting direction when the F_z=−1/2 and F_z=−5/2 p-like levels cross. Below (above) this field, the fourth hole occupies the F_z=−1/2 (F_z=−5/2) p-like state [Fig. 4(a)]. Likewise, peak 6 changes slope when the F_z=5/2 p-like level crosses the F_z=−1/2 d-like level.

Our final level diagram, as shown in Fig. 4(b), qualitatively agrees with recent band structure calculations of hole levels in InAs QDs.[12] In particular, hole-charging effects do not have to be invoked for a satisfactory explanation of the magnetic field dispersion of energy levels for both a noninteraction system as measured with PL as well as for the charging spectra deduced from *C*-*V* spectroscopy. However, other recent theoretical calculations, using an atomistic pseudopotential description, propose a different filling sequence of the hole levels.[10,11] Because of the competition between Coulomb repulsion and level spacing, the *d*-like shell starts filling before the *p*-like shell is fully occupied. In terms of the hole levels in Fig. 4(b), this implies that the *d*-like $|F_z|=1/2$ levels energetically move below the *p*-like $|F_z|=5/2$ levels at zero magnetic field. Since the magnetic field dispersion of these levels is very similar, such a reversed level sequence is still consistent with our experimental results for an interacting system as measured by *C*-*V* spectroscopy.

In conclusion, by combining optical and *C*-*V* spectroscopy experiments in high magnetic fields, we determine the single-particle levels and charging energies of holes in self-assembled InAs QDs. Our analysis of the PL polarization shows that, unlike electrons, holes do not follow the Fock-Darwin spectrum and underlines the crucial role of the complex valence-band structure for the properties of semiconductor QDs. The resulting model for the single-particle hole levels is consistent with the charging spectra obtained from *C*-*V* spectroscopy.

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