Compositional and size-dependent spectroscopic shifts in charged self-assembled $In_xGa_{1-x}As/GaAs$ quantum dots

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Atomistic pseudopotential many-body calculations of excitonic (X) recombination in charged, self-assembled $In_xGa_{1-x}As/GaAs$ dots predict and explain remarkable trends. (i) The redshift of the exciton energy upon negative charging is rapidly reduced with increasing the In content and increasing the dot height. The opposite behavior is observed upon positive charging. (ii) The recombination peak energies of different charge states show intriguing symmetries and alignments, e.g., X^- aligns with X^{2-} and X^{3-} aligns with X^{4-}. (iii) The X^{3-} spectrum shows that a triplet initial state is lower in energy for flat dots (yielding two spectral lines),

one monolayer thick InGaAs wetting layer. Figure 2 shows the relative shifts of the main X^0, X^-

Coulomb and like-particle exchange terms, neglecting the very small electron-hole exchange terms $K_{e_i h_j}$ (included in Figs. 1 and 2). We can now interpret the main splittings in terms of specific interactions: The two main peaks for Q = even reflect like-carrier exchange interactions: the $X_a^{2-} - X_b^{2-} = C^-$ splitting is $2K_{e_0 e_1}$, whereas the $X_a^{4-} - X_b^{4-} = D$ splitting is $2K_{e_0 e_2}$. On the other hand, the $0 \rightarrow (\pm 1) \rightarrow (\pm 2)$ shifts reflect direct Coulomb energy differences. The $X^0 - X^- = A^-$ shift is $J_{e_0 e_0} - J_{e_0 h_0}$, the $X^0 - X^+ = A^+$ shift is $J_{h_0 h_0} - J_{e_0 h_0}$, and the $X^- - X_a^{2-} = B^-$ shift is $J_{e_0 e_1} - J_{e_1 h_0}$. Thus, whereas the splitting of the Q = even peaks reflect absolute exchange energies, the shifts A^-, A^+, B^+, B^- reflect relative Coulomb energies that vanish at zero order. Indeed, if the hole and electron wave functions were the same (as is assumed in single-band effective mass models with infinite wells), then $A^- = A^+ = B^+ = B^- = 0$.

While Fig. 3 neglects the effect of correlations, these are taken into account in Figs. 1 and 2. To understand the effect of correlations we compare in Fig. 1 the full CI results (black peaks) with the spectra calculated without correlations (dashed lines). The effect of the exchange and scattering terms can be seen by comparing the dashed and the solid black lines (neglecting correlations and exchange integrals). Whereas the direct Coulomb energies merely shifts the PL peaks, the exchange interaction splits (X^{even}) and shifts ($X^{\pm 3}$, $X^{\pm 4}$) peaks. Correlation effects tend to shift peaks to the red by as much as 2 meV in the present dots. In fact, neglecting the effect of correlations would result in a downward shift by about 2 meV of the A^+ and A^- curves in Fig. 2 and leading to the wrong conclusion that In rich dots exhibit a blueshift (redshift) of the X^- (X^+), since A^- and A^+ would be negative. The B^+ and B^- curves are nearly unaffected by correlations.

Our foregoing analysis of the origins of the spectra allows us to comment on the spectroscopic observations (i)–(iii) made above.

(i) *Trends in $X^0 \rightarrow X^-$ and $X^0 \rightarrow X^+$.* We saw that the shifts A^+ and A^- in Fig. 3 reflect the balance between like-particle ($e_0 - e_0$ or $h_0 - h_0$) and different-particle ($e_0 - h_0$) Coulomb interactions. In the lower panels of Fig. 4 the difference between the electron e_0 and hole h_0 densities ρ_{diff} is plotted for a pure InAs and for an $In_{60}Ga_{40}As$ dot. It shows, for both dots, that h_0 is more *localized* in the growth direction than e_0 since ρ_{diff} has a negative value (characteristic for h_0) in the center of the dot. However, in the pure InAs dot [panel (b)] h_0 is more *delocalized* in the (001) plane than e_0. In contrast, for the $In_{0.6}Ga_{0.4}As$ dot, e_0 and h_0 have equivalent localization in this plane. This effect can be appreciated by the percentage of the charge density inside the physical dimension of the dot. This yields 90.1% (88.4%) for the first hole state and 82.4% (82.8%) for the first electron state in the pure (onion) dot. For the onion dot, the stronger hole localization contributes to a negative value for A^- (since $J_{e_0 h_0}$ is larger than $J_{e_0 e_0}$) and a positive value for A^+ (since $J_{h_0 h_0}$ is larger than $J_{e_0 h_0}$). The physics underlying the observed trends is therefore related to the degree of localization of the wave functions and can be understood as follows. (1) The reduction of size (reducing the width of the potential well) increases the confinement energy of both electrons and holes, i.e., their single particle levels move up and down, respectively. Thereby, their wave functions become more delocalized. This effect is more pronounced for the electron than for the hole state: The electrons tend to be more delocalized than the holes when the size is reduced. (2) The reduction of the In content lowers the band offsets between the material in the dot and the surrounding GaAs. This reduction delocalizes electrons more strongly than holes: The electrons tend to be more delocalized than the holes when the In concentration is reduced.

(ii) *Alignment of peaks in different charged states.* The alignments evident in the spectrum of the negatively charged dot (Fig. 1) can be understood from the different integral contributions shown in Fig. 3: The X^-, X_a^{2-}, X^{3-}, and X_a^{4-} peaks are predicted to be shifted from the fundamental X^0 transition by A^-, $(A^- + B^-)$, $(A^- + 2B^- - K_{e_0 e_1})$, and $(A^- + 2B^- - K_{e_0 e_1} + J_{e_0 e_2} - J_{e_2 h_0})$, respectively. For certain heights and compositions, $B^- = J_{e_0 e_1} - J_{e_1 h_0}$ is close to zero which results in the alignment of the of X_a^{2-} and X^- peaks. For B^- to vanish, the states e_0 and h_0 do not necessarily need to be identical. Unlike the shifts A^+ and A^- that involved integrals over S and

state e_0 or h_0