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 $2K_{e_0e_1}$, whereas the $X_a^{4-}-X_b^{4-}=D$ splitting is $2K_{e_0e_2}$. On the other hand, the $0\rightarrow(\pm 1)\rightarrow$ (± 2) shifts reflect direct Coulomb energy differences. The $X^0-X^-=A^-$ shift is $J_{e_0e_0}-J_{e_0h_0}$, the $X^0-X^+=A^+$ shift is $J_{h_0h_0}-J_{e_0h_0}$, and the $X^--X_a^{2-}=B^-$ shift is $J_{e_0e_1}-J_{e_1h_0}$. 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 $B+$

state e_0 or h_0