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## Multi-excitons in self-assembled InAs/GaAs quantum dots: A pseudopotential, many-body approach

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**Abstract.** – We use a many-body, atomistic empirical pseudopotential approach to predict the multi-exciton emission spectrum of a lens-shaped InAs/GaAs self-assembled quantum dot. We discuss the effects of i) the direct Coulomb energies, including the differences of electron and hole wave functions, ii) the exchange Coulomb energies and iii) correlation energies given by a configuration interaction calculation. Emission from the ground state of the  $N$  exciton system to the  $N - 1$  exciton system involving  $e_0 \rightarrow h_0$  and  $e_1 \rightarrow h_1$  recombinations are discussed. A comparison with a simpler single-band, effective mass approach is presented.

High-resolution single-dot spectroscopy [1–5] of InAs/GaAs self-assembled quantum dots shows that as the excitation intensity is increased, thus loading more excitons into the dots, new emission lines appear both to the red and to the blue of the fundamental emission line observed at low excitation power. “State filling” effects, leading to the recombination of high-energy electron-hole pairs, cannot explain the red-shifted emission lines, nor the fact that the number of lines exceeds the number of allowed single-particle transitions. In this letter we present a theory of self-assembled semiconductor quantum-dots, based on a pseudopotential many-body expansion that demonstrates that it is multi-exciton transitions that are responsible for this complex observed spectral structure. We isolate and clarify three distinct physical effects: i) electron-hole wave function asymmetry, leading to a *blue shift* of the fundamental exciton transition as the number of spectator excitons loaded into the dot increases, ii) electron-electron and hole-hole exchange interactions which *red shift* all *even* multi-exciton decays (biexciton, four-exciton) and split the *odd* multi-exciton decays (tri-exciton, five-exciton) into multiple-lines, and iii) correlation effects which *red shift* the biexciton leading to its binding with respect to the monoexciton.

We will first describe the qualitative picture of multi-excitons and then describe a quantitative model. The essential physics of such transitions can be understood by considering what happens to the ground-state recombination of the lowest electron level,  $e_0$ , and the lowest hole level,  $h_0$ , if other electrons and holes are present in the dot as “spectators”. The schematic figures in the center of fig. 1 depict the fundamental  $e_0$ - $h_0$  recombination in the presence of 0 to 5 “spectator” electron-hole pairs (we assume here that all levels are spatially non-degenerate [6]). We distinguish here two exciton series; i) when the initial number of excitons,  $N$  is even, the initial electron configuration is “closed shell”, *e.g.*  $(e_0^2)(h_0^2)$  for  $N = 2$ ,

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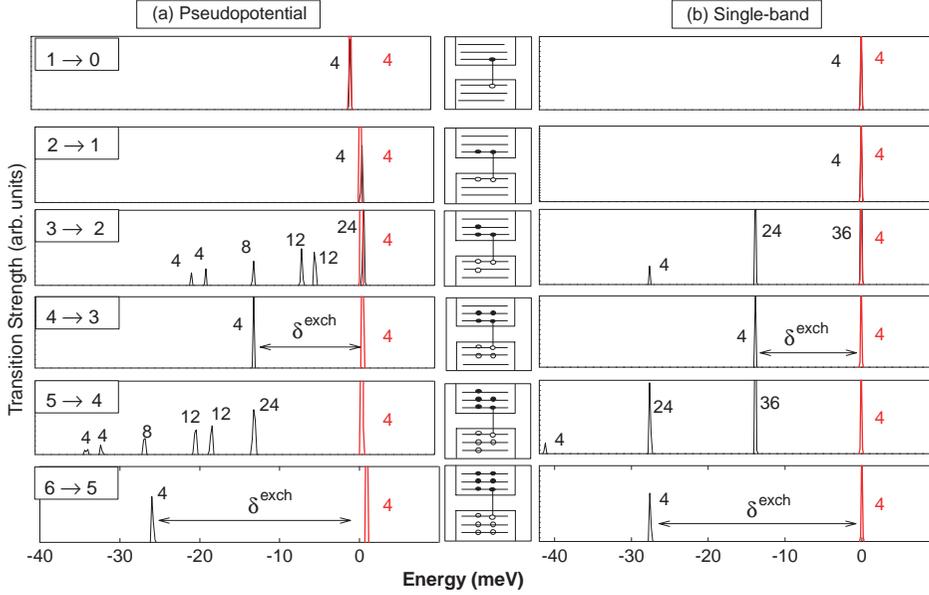


Fig. 1 – Energy of  $e_0 \rightarrow h_0$  recombinations in the presence of 0 to 5 spectator excitons. Energies calculated in the  $(\epsilon + J)$  (red) and  $(\epsilon + J + K)$  (black) approximations are shown. (a) shows our pseudopotential calculations and (b) shows calculations with the assumptions from refs. [1, 7, 8]. The multiplicity of each line is labelled.

whereas ii) when  $N$  is odd, the initial configuration has an open shell both in the electron and in the hole manifold, *e.g.*  $(e_0^2)e_1^1(h_0^2)h_1^1$  for  $N = 3$  (parentheses are used for the closed shell orbitals). The distinction between the “closed shell” and “open shell” multi-exciton is important, since in the initial state of the  $N$ =even, “closed shell” series the spectator levels can be occupied in only a single unique manner by the spectator spins, while in the  $N$ =odd, “open shell” series, many spin arrangements are possible in the initial and final states. This will lead to a large number of exciton lines.

To understand qualitatively the effects of spectator electrons and holes, let us consider the Hartree-Fock (HF) energy of a single configuration of electrons and holes. We denote the direct Coulomb interaction between carriers in levels  $i$  and  $j$  by  $J_{ij}$  and the exchange interactions as  $K_{ij}$ . The recombination energy of the monoexciton is given by

$$E_{e_0h_0}^{1 \rightarrow 0} = (\epsilon_{e_0} - \epsilon_{h_0}) - J_{e_0,h_0}, \quad (1)$$

where  $\epsilon_{e_0}$  and  $\epsilon_{h_0}$  are the single-particle levels and  $E_{e_0h_0}^{1 \rightarrow 0}$  denotes the energy associated with decaying from a single exciton to the ground state. Neglecting the electron-hole exchange (N.B.: it is included in the calculation) the  $e_0$ - $h_0$  recombination energy in the presence of  $N_s$  electrons and holes is

$$E_{e_0h_0}^{N \rightarrow N-1} = E_{e_0h_0}^{1/0} + \left[ \sum_{e_s}^{N_s} (J_{e_0e_s} - J_{e_s h_0}) + \sum_{h_s}^{N_s} (J_{h_0h_s} - J_{e_0h_s}) \right] - \left[ \sum_{e_s}^{N_s} K_{e_0e_s} + \sum_{h_s}^{N_s} K_{h_0h_s} \right], \quad (2)$$

where  $e_s$  and  $h_s$  are spectator electrons and holes, such that  $\sum_{e_s}^{N_s} = \sum_{h_s}^{N_s} = N - 1$ . We see from eq. (2) that the  $e_0$ - $h_0$  recombination energy is shifted with respect to the fundamental exciton. This shift has two sources indicated by the two bracketed terms in eq. (2). First, the Coulomb shift, which vanishes if the electrons and holes,  $e_0$  and  $h_0$  have the same wave functions, *i.e.* if  $J_{e_0e_s} = J_{e_s h_0}$  and  $J_{h_0h_s} = J_{e_0h_s}$ . This “Coulomb shift”,  $\delta_{N \rightarrow N-1}^{\text{Coul}}$  thus reflects

the difference in the electron and hole wave functions. It vanishes artificially in single-band effective-mass calculations that use an infinite well depth, such as those in refs. [1,2,7]. Second, there is an exchange shift,  $\delta_{N \rightarrow N-1}^{\text{exch}}$ , given by the second term in brackets in eq. (2). This exchange shift is familiar from theories of band gap renormalization [9] where the existence of high carrier densities during high power photoexcitation acts to reduce the band gap. In addition, since the exchange interaction depends on the spin orientation of the carriers, the exchange contribution (second term in eq. (2)) can split the excitonic transitions.

In the  $N$ = even, “closed shell” series, the initial state contains no open shells while the final state contains one open shell. This results in one- and four-fold degeneracies for the initial and final states, and hence 4 transitions. These 4 transitions are split by the small electron-hole exchange interaction. In contrast, for the  $N$ = odd transitions, the initial and final states contain one and two open shells, respectively, resulting in four- and 16-fold degeneracies and hence a total of 64 possible transitions. Different alignments of the spins produce splittings of the different transitions resulting from electron-electron and hole-hole exchange interactions. In summary, within the HF approximation the presence of “spectator excitons” will shift the  $e_0$ - $h_0$  transition in the  $N$ = even series and shift and split the  $e_0$ - $h_0$  transition in the  $N$ = odd series.

The above treatment neglects the effects of correlation. These can be conveniently introduced by considering configuration-interaction (CI) effects [10]. In other words, instead of evaluating the energy (eq. (1)) of the  $e_0$ - $h_0$  monoexciton by considering only the 4 spin arrangements consistent with a *single* orbital configuration,  $e_0^1 h_0^1$ , we allow the presence of other orbital configurations such as  $e_1^1 h_0^1$ ,  $e_0^1 h_1^1$  and  $e_2^1 h_0^1$ . This configuration interaction approach can shift the Hartree-Fock transitions of eqs. (1) by a “correlation shift”,  $\delta_{e_0 h_0}^{\text{CI}}$ , which includes both self-consistent adjustments of the single-particle orbitals and correlation. This will, for example, provide additional binding to the biexciton with respect to the monoexciton:

$$E_{e_0 h_0}^{2 \rightarrow 1} - E_{e_0 h_0}^{1 \rightarrow 0} = [J_{e_0, e_0}^{\text{ee}} + J_{h_0, h_0}^{\text{hh}} - 2J_{e_0, h_0}^{\text{eh}}] - \delta_{e_0 h_0}^{\text{CI}}. \quad (3)$$

Correlation effects can also add new transitions to the spectrum, due to the mixing of new configurations into the Hartree-Fock configuration. Thus, CI will, in general, also alter the intensity of the HF transitions.

The purpose of this letter is to make a realistic prediction of the “Coulomb shift”,  $\delta^{\text{Coul}}$ , the “exchange-splitting”,  $\delta^{\text{exch}}$ , and “correlation shift/splitting”,  $\delta^{\text{CI}}$ , due to the presence of spectator excitons in a self-assembled semiconductor quantum dot. We adopt the experimentally determined lens-shaped dot with a base of 250 Å and a height of 35 Å and a PL peak at 1.1 eV. The optical and electronic properties of this dot geometry have been extensively studied, see refs. [11,12] and references therein. Recently, alternative dot geometries for *uncapped* dots have been proposed, based on {136} facets [13]. However, there is strong evidence of Ga in-diffusion during the capping process which acts to produce a lens-shaped geometry. The single-particle bound states are calculated using an empirical pseudopotential Hamiltonian [6]. Recent developments in these pseudopotentials enable the full inclusion of the effects of strain, multi-band couplings, band non-parabolicity and spin-orbit coupling in the single-particle Schrödinger equation. In ref. [6] we demonstrate that by accurately fitting the *bulk* band structure, effective masses and deformation potentials, we are able to use these pseudopotentials to obtain excellent agreement with a wide range of optical and electronic properties for the lens-shaped dots discussed in this paper. These comparisons were not possible earlier when the shape of the dot and its composition profile were unknown. In ref. [6], we model the shape and composition profile of the dot and then compare calculated and measured single-exciton energies, inter-band energy spacings, electron and hole binding energies and wetting layer energies, finding excellent agreement. For completeness, we treat here the

same lens-shape, alloyed dot with a 1.1 eV PL peak. Unfortunately, although numerous experiments were conducted on this dot, no multi-exciton spectra were taken since the lowest PL is outside the range of conventional CCD detection equipment, so quantitative comparison awaits a future measurement.

Having obtained the single-particle levels ( $e_0, e_1, \dots, h_0, h_1, \dots$ ) from the pseudopotential method, we then calculate numerically the screened Coulomb and exchange integrals

$$J_{ijkl} = \iint \frac{\psi_i^*(\mathbf{r}_1)\psi_j(\mathbf{r}_2)\psi_k^*(\mathbf{r}_1)\psi_l(\mathbf{r}_2)}{\bar{\epsilon}(\mathbf{r}_1 - \mathbf{r}_2)|\mathbf{r}_1 - \mathbf{r}_2|} d\mathbf{r}_1 d\mathbf{r}_2, \quad K_{ijkl} = \iint \frac{\psi_i^*(\mathbf{r}_1)\psi_j(\mathbf{r}_2)\psi_k^*(\mathbf{r}_2)\psi_l(\mathbf{r}_1)}{\bar{\epsilon}(\mathbf{r}_1 - \mathbf{r}_2)|\mathbf{r}_1 - \mathbf{r}_2|} d\mathbf{r}_1 d\mathbf{r}_2, \quad (4)$$

where  $\bar{\epsilon}$  is a size-dependent, phenomenological, screened dielectric function [14]. Our exchange automatically includes both short- and long-range components [15]. In the configuration-interaction approach, we expand the wave function of the  $N$ -exciton,  $\Psi$ , as a linear combination of Slater determinants,  $\Phi$ , obtained by exciting  $N$  electrons from the valence band to the conduction band. For example, in the  $N = 2$  (biexciton) case we have

$$\Psi(\mathbf{r}_1, \dots, \mathbf{r}_M) = \sum_{vv', cc'} A_{vv', cc'} \Phi_{vv', cc'}(\mathbf{r}_1, \dots, \mathbf{r}_M). \quad (5)$$

Here  $M$  is the total number of electrons in the quantum dot,  $v, v'$  denote the unoccupied valence band single-particle states (holes) and  $cc'$  denote the conduction band states occupied by the  $N$  excited electrons. To obtain our many-body states, we diagonalize the many-body Hamiltonian in the basis of  $\Phi$ .

To clarify and isolate the physical factors contributing to multi-exciton effects, we solve the problem in a series of steps. First, we neglect configuration interaction effects, treating only single configurations and set all the exchange integrals to zero, ( $\delta^{\text{exch}} = 0$ ). In this ( $\epsilon + J$ ) approximation we see the effect of the Coulomb ‘‘chemical shift’’,  $\delta^{\text{Coul}}$ . Second, we will still use a single configuration, but include exchange integrals. In this ( $\epsilon + J + K$ ) approximation we will see the added effects of carrier-carrier exchange,  $\delta^{\text{exch}}$ . Finally, the effects of correlation are included in the CI calculation.

Figure 1(a) shows our calculated energies associated with the fundamental recombination of an electron and hole,  $e_0 \rightarrow h_0$  in the presence of 0 to 5 spectator excitons. These are denoted as the  $1 \rightarrow 0$  to  $6 \rightarrow 5$  transitions. The  $N$  excitons that form the initial state of each transition are assumed to occupy the ground-state configuration as predicted by the Aufbau principle. A schematic energy level scheme in the center of each figure shows this initial occupation, where the vertical solid line marks the recombination taking place. The spectrum shown in each panel is obtained from a sum of Gaussians (0.1 meV width) whose means represent the transition energies and heights are proportional to the calculated dipole transition element. The zero of energy is taken as the fundamental exciton,  $\epsilon_{e_0 - \epsilon_{h_0} - J_{e_0 h_0}}$ . The multiplicities of each individual group of transitions in the ( $\epsilon + J$ ) and ( $\epsilon + J + K$ ) approximations are marked in black and red.

*The effect of direct Coulomb interactions.* – The red lines in fig. 1(a) show the transition energies in the ( $\epsilon + J$ ) approximation which include only single-particle and direct Coulomb energies. Inspection of these lines shows that:

i) All the observed Coulomb shifts are relatively small ( $\delta^{\text{Coul}} \sim 2$  meV) and result in a blue shift of the transition with respect to the fundamental transition. For example, at this level of approximation, the biexciton is ‘‘unbound’’ with respect to two single excitons, *i.e.*  $E^{2 \rightarrow 1} > E^{1 \rightarrow 0}$ .

ii) As all the transitions involve the same single-particle  $e_0 \rightarrow h_0$  recombination, they all have the same oscillator strength.

iii) All the transitions show only a single degenerate line as neither the initial or the final state exhibit any exchange splittings in this approximation.

*The effect of exchange interactions.* – The black lines in fig. 1(a) show the transition energies in the  $(\epsilon + J + K)$  approximation which includes single-particle, direct and exchange Coulomb energies. Inspection of these lines shows that:

i) The  $1 \rightarrow 0$  and  $2 \rightarrow 1$  transitions contain only one or zero spectator excitons and hence no electron-electron or hole-hole exchange takes place. Therefore, only the small electron-hole exchange fine splitting is observed. We therefore classify these transitions as essentially 4-fold multiplets. Due to the lack of correlation, the biexciton is still unbound with respect to two single excitons as in the  $(\epsilon + J)$  approximation.

ii) For *even*  $\rightarrow$  *odd* there are 4 possible transitions. In each case,  $2 \rightarrow 1$ ,  $4 \rightarrow 3$  and  $6 \rightarrow 5$  we observe only one 4-fold multiplet. The fine splitting within this group of transitions arises from electron-hole exchange splittings of the 4 final states. The initial states contain no unpaired electrons and holes and hence exhibit no exchange splittings. As the number of spectator excitons increases, there is a red shift of these transitions due to exchange interactions. The exchange energy shifts from eq. (2) are  $\delta_{4 \rightarrow 3}^{\text{exch}} = [K_{e_0 e_1} + K_{h_0 h_1}]$  and  $\delta_{6 \rightarrow 5}^{\text{exch}} = [K_{e_0 e_2} + K_{e_0 e_1} + K_{h_0 h_2} + K_{h_0 h_1}]$ .

iii) For *odd*  $\rightarrow$  *even* with  $N \geq 3$  there are 64 possible transitions. In both cases:  $3 \rightarrow 2$  and  $5 \rightarrow 4$  we see 6 groups of transitions with multiplicities of 4:4:8:12:12:24. The splitting between the 6 groups arises from electron-electron and hole-hole exchange splittings between the final states which contain two unpaired electrons and holes. The 6 groups of transitions in  $3 \rightarrow 2$  and  $5 \rightarrow 4$  span 23 and 22 meV. This energy span reflects the span of the 16 eigenvalues of the  $16 \times 16$  matrix generated by the different spin occupations of the final states.

iv) The exchange interaction alters the oscillator strength of the transitions so that they are not all identical as in the  $(\epsilon + J)$  approximation.

*Comparison with previous calculations.* – Previous calculations of multi-excitons in quantum dots include the works of Hu [16], Takagahara [17], Barenco and Dupertuis [7], Dekel *et al.* [1,2], Landin *et al.* [3] and Hawrylak [8]. References [1,2,7,16,17] and [8] adopt single-band effective-mass models with either an infinite potential barrier [1,2,7,16,17] or a parabolic potential [8], both of which artificially force the electron and hole wave functions to be identical. Therefore, in all these calculations the Coulomb shift,  $\delta^{\text{Coul}}$ , is zero and the electron-hole exchange vanishes,  $K_{e_i h_j} = 0$ . To obtain realistic single-particle energy spacings in refs. [1,2,7], an unrealistic cuboidal shape had to be assumed and the size of the dots was treated as adjustable parameters. By choosing different lengths for all three dimensions both the measured *s-p* and *p-p* splittings can be reproduced. The two-dimensional parabolic potential adopted in ref. [8] can also be adjusted to reproduce the correct *s-p* splitting, but will always produce degenerate *p* states. It has recently been shown [2] that only models which can split this *p* level degeneracy can provide a realistic interpretation of experimental results. Such a splitting can be introduced either by adopting an atomistic model such as the one described here, where the inequivalence of the [110] and  $[1\bar{1}0]$  directions provide the splitting, or by adding an additional potential to the Hamiltonian such as a piezo-electric potential. In addition to approximating the single-particle states, refs. [1,2,7,8,16,17] neglect the effects of strain which can shift electron and hole single-particle levels by several 100 meV and the spin-orbit interaction, which introduces additional splittings in the calculated emission spectra (see fig. 1).

To calculate the exchange and correlation contribution to the excitonic energies, ref. [16] uses path integral quantum Monte Carlo techniques which provide an exact (to within statistical error) determination of the exchange and correlation energy. However, quantum Monte Carlo methods are currently restricted to single-band Hamiltonians and cannot therefore predict the Coulomb shift,  $\delta^{\text{Coul}}$ , discussed above. Reference [8] adopts a limited basis CI to estimate correlation energies. References [1–3,7,17] use only a single-configuration approach which does not include correlation effects.

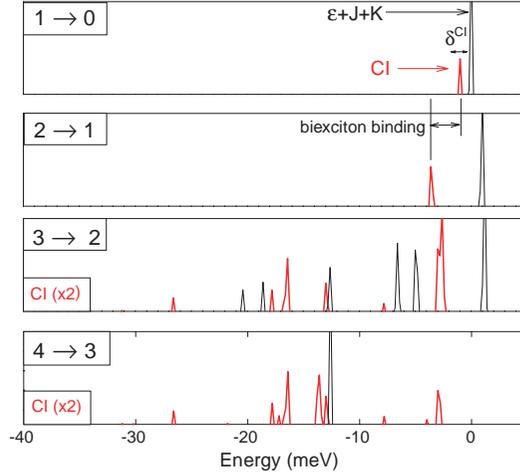


Fig. 2 – Energy of  $e_0 \rightarrow h_0$  recombinations in the presence of 0 to 3 spectator excitons. The black and red lines show energies calculated in the  $(\epsilon + J + K)$  and CI approximations.

To assess the above approximations used in [1, 2, 7, 8, 16], we show in fig. 1(b) a repeat of our calculations for the transition energies within the  $(\epsilon + J)$  and  $(\epsilon + J + K)$  approximations applying the assumptions adopted in refs. [1, 2, 7, 8], namely  $J_{e_i e_j} = J_{h_i h_j} = J_{e_i h_j}$ ,  $K_{e_i e_j} = K_{h_i h_j}$ ,  $K_{e_i h_j} = 0$  and  $\Delta^{\text{SO}} = 0$ . We observe that:

i) Within the  $(\epsilon + J)$  approximation, the chemical shift,  $\delta^{\text{Coul}}$ , is zero by definition, so all the red lines lie on the zero of energy. ii) Within the  $(\epsilon + J + K)$  approximation for *even*  $\rightarrow$  *odd*, there are 4 possible transitions that are exactly degenerate as  $K_{e_i h_j} = 0$ . iii) For *odd*  $\rightarrow$  *even*, there are 64 possible transitions, split into 4:24:36 multiplets, compared to the 4:4:8:12:12:24 multiplets obtained in the pseudopotential calculations. The reduction in the number of multiplets arises from the assumptions  $\psi_{e_i} = \psi_{h_i}$  and  $\Delta^{\text{SO}} = 0$ . Other than these changes, we find that many of the qualitative features noted in the calculations of Dekel *et al.* [1, 2] are retained in the pseudopotential description.

*The effect of CI interactions.* – In fig. 2 we contrast the transition energies from our pseudopotential calculations within the  $(\epsilon + J + K)$  approximation (black lines) with those from a CI calculation (red lines) which includes single-particle, direct and exchange Coulomb and “correlation” effects.

In the CI calculations we expand the many-body wave function in a basis of Slater determinants constructed from all possible orbital and spin occupations of the lowest 10 (including spin degeneracy) electron and lowest 10 hole single-particle levels. For example, the biexciton basis contains  ${}^{10}C_2 \cdot {}^{10}C_2 = 2025$  Slater determinants, and the 3-exciton basis contains  ${}^{10}C_3 \cdot {}^{10}C_3 = 14400$  Slater determinants. This basis neglects the contributions for higher-lying bound states and continuum states. To investigate the effects of adopting this limited CI basis [18], we have compared the results of diffusion quantum Monte Carlo calculations (DMC) and CI calculations for the exciton correlation energy in a model, single-band system with equivalent size, band offsets and number of bound states. We find that our CI calculations retrieve approximately 50-60% of the 5-6 meV of correlation energy obtained in the DMC calculations and can therefore be used as a *lower bound* for the effects of correlation. Using these calculations we find that:

i) Correlation effects lower the energy of both the initial and final state of a transition. Our calculated correlation varies from 2-3 meV for a single exciton to 10 meV for multiple

excitons. For all the transitions shown here, the correlation shift,  $\delta^{\text{CI}}$ , for the initial state with  $N$  excitons is greater than that for the final  $N - 1$  exciton state. Therefore, all the CI *transition* peaks are red shifted with respect to those from the  $(\epsilon + J + K)$  approximation. ii) This ( $\delta^{\text{CI}}$ ) red shift is larger for the  $2 \rightarrow 1$  transition than for the  $1 \rightarrow 0$  transition and is able to overcome the Coulomb blue shift and “bind” the biexciton. Our calculated binding energy is  $\sim 3$  meV, however, our model QMC calculations suggest that a fully converged value would be closer to 5 meV. iii) As the number of spectator excitons increases, the *difference* in the red shift for the initial and final states decreases, so that the red shift of the *transition* energy decreases. For  $3 \rightarrow 2$  and  $4 \rightarrow 3$  transitions the red shifts of the transitions rapidly decrease. iv) The mixing of configurations within the CI results in both additional peaks in the CI spectra and changes in the relative magnitude of the peaks, *e.g.* for  $4 \rightarrow 3$  the single  $(\epsilon + J + K)$  peak is split into two equally strong multiplets of peaks in the CI spectra. The additional peaks result from the mixing of excited states within the CI calculation. For example, the additional CI peaks blue shifted from the main peaks in the  $4 \rightarrow 3$  spectra result from excited-states configurations mixed into the  $N = 3$  exciton.

In conclusion, we present results of the first pseudopotential, many-body calculation of multi-exciton states within InAs/GaAs quantum dots. We are able to isolate the effects of the direct and exchange Coulomb interactions and correlation on the energies of  $N \rightarrow N - 1$  excitonic transitions. We find that direct Coulomb energies introduce small blue shifts. Electron-electron and hole-hole exchange splittings, which are responsible for the majority of the observed structure, introduce both red shifts and splittings. Correlation effects red shift all transitions and change the relative energies of transitions (*e.g.*, bind the biexciton).

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