Calculation of conduction-to-conduction and valence-to-valence transitions between bound states in (In,Ga)As/GaAs quantum dots

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We have calculated the conduction-to-conduction and valence-to-valence absorption spectrum of bound states in (In,Ga)As/GaAs quantum dots charged with up to three electrons or holes. Several features emerge: (i) In pure (non-alloyed) InAs/GaAs dots, the 1S-1P subbands optical transitions are fully in-plane polarized along [110] and [110], respectively, while valence transitions are weakly polarized because the hole P states do not show any in-plane preferential orientation. (ii) In alloyed In_{0.6}Ga_{0.4}As/GaAs dots the [110] and [110] polarization of the corresponding 1S-1P conduction intraband transitions is weakened since the two 1P states are mixed by alloy fluctuations. The polarization of valence intraband transitions is insensitive to changes in alloy fluctuations. (iii) For light polarized along [001], we find a strong valence-to-valence transition that involves a weakly confined hole state with predominant light-hole character. (iv) When charging the dots with a few electrons, the conduction intraband transitions display spectroscopic shifts of \( \sim 1-2 \) meV. These shifts are a result of correlation effects (captured by configuration-interaction) and not well described within the Hartree-Fock approximation. (v) When charging the dots with holes, valence intraband spectra are more complex than the conduction intraband spectra as hole states are strongly affected by spin-orbit coupling, and configuration mixing is more pronounced. Spectroscopic shifts can no longer be identified unambiguously. These predictions could be tested in single-dot spectroscopy of n-doped and p-doped quantum dots.

I. BRIEF BACKGROUND AND SCOPE

Doping quantum dots \( n \)-type (\( p \)-type) followed by infrared-light excitation leads to conduction-to-conduction (valence-to-valence) intraband excitations between confined states. In early experiments, intraband transitions were studied for quantum wells\(^{16} \) where in the case of \( n \)-type doping the selection rules lead to allowed transitions only for light polarized normal to the well\(^{2,3,4} \) whereas \( p \)-type samples afforded transitions allowed in normal incidence. Instead, \( n \)-doped quantum dots offered the possibility of allowed intraband transitions for light incident normal to the plane of the dots. Thus, quantum dots became the focus of much experimental work devoted to study the intraband optical transitions\(^{5,6,7,8,9,10,11,12,13,14,15} \). For example, measurements of optical intraband transitions in (In,Ga)As/GaAs quantum dots charged with electron and holes have been recently performed by Zibik \( et \) \( al. \)^{16}, who measured conduction-to-conduction transitions, and Preisler \( et \) \( al. \)^{17} who have measured valence-to-valence intraband absorption.

On the other hand, calculations of the intraband optical properties are often based on model\(^{18,19,20} \) or \( k \cdot p \)\(^{21} \) approaches and not high-level atomistic approaches. Here, we address this issue by calculating conduction-to-conduction and valence-to-valence intraband optical absorption of (In,Ga)As/GaAs dots charged with up to three electrons or holes. We do not intend to survey the effects of size, shape, and composition on the intraband transitions of the dots. We focus primarily on (i) the effects of alloy fluctuations on the polarization of the intraband transitions, and (ii) the differences between the absorption spectra of \( n \)-doped and \( p \)-doped dots as well as the spectroscopic shifts induced by charging. We use a combined approach to calculate the intraband absorption spectra in which we find the electron and hole single-particle states of the dots with an atomistic pseudopotential method and solve the many-particle states of charged dots within a configuration-interaction approach. The advantage of this approach over simplified methods is that it naturally includes (i) the correct symmetry of the dot; (ii) strain and alloying effects; (iii) multiband and multivalley coupling; and (iv) spin-orbit interaction. Our approach is purely electronic and does not include polaron (electron-phonon coupling) effects.

We illustrate our findings with a prototypical lens-shaped In\(_{0.6}\)Ga\(_{0.4}\)As/GaAs dot with diameter \( b = 252 \) Å and height \( h = 35 \) Å. As a benchmark, for dots charged with a single carrier, we provide results for a pure non-alloyed InAs/GaAs dot with the same size. While so far intraband experiments have focused on ensemble of dots with different degrees of homogeneity, which broadens the observed transitions, we offer predictions that could be probed in single-dot spectroscopy of \( n \)- or \( p \)-doped dots. In addition, as the control of doping carriers is difficult, i.e. in an ensemble of doped dots some have a single electron while other have two or none, we discuss the intraband spectra upon increasing the carriers one at a time.

II. METHOD: PSEUDOPOTENTIAL APPROACH AND CONFIGURATION-INTERACTION

We calculate the single-particle electron and hole energy levels of the self-assembled dot within an atomistic, pseudopotential-based method\(^{22} \). The wave func-
where $v_\alpha$ is the screened pseudopotential for atom of type $\alpha$ (In,Ga,As) with position $R_{i,\alpha}$ within the dot or barrier, and $V_{SO}$ is a non-local pseudopotential that accounts for the spin-orbit interaction. These pseudopotentials are carefully fitted to bulk GaAs, InAs, and (In,Ga)As alloys, thus removing the local-density-approximation (LDA) errors. The basis in which we expand $\psi_j$ is a non-local pseudopotential that accounts for underlying strain in the dot and barrier.

The prototypical alloyed dot we consider here presents a pure (non-alloyed) InAs/GaAs dot (a) than in the alloyed In$_{0.6}$Ga$_{0.4}$As/GaAs dot (b). For holes, we show the first 40 confined states. Black and grey arrows indicate, respectively, the lowest intraband conduction transition for polarization along [110] and [100]; while numbers show the transition energy in meV.

FIG. 1: Electron (top) and hole (bottom) energy levels in lens-shaped (In,Ga)As/GaAs dots with base diameter $b = 252 \, \text{Å}$ and height $h = 35 \, \text{Å}$ relative to the GaAs barrier conduction band minimum $E_c$(GaAs) = $-4.093 \, \text{eV}$ (w.r.t. vacuum) and its valence band maximum $E_v$(GaAs) = $-5.620 \, \text{eV}$ (w.r.t. vacuum). The energy levels are more confined in the pure (non-alloyed) InAs/GaAs dot (a) than in the alloyed In$_{0.6}$Ga$_{0.4}$As/GaAs dot (b). For holes, we show the first 40 confined states. Black and grey arrows indicate, respectively, the lowest intraband conduction transition for polarization along [110] and [100]; while numbers show the transition energy in meV.

III. RESULTS FOR SINGLE-PARTICLE ELECTRON AND HOLE LEVELS

A. Electron levels

Bound states can be labeled by their leading orbital character and approximately arranged into shells: $\{1S; 1P_1, 1P_2; 1D_1, 1D_2, 2S; \ldots\}$. For the dot size considered in our calculation, a pure (non-alloyed) dot confines 15 states arranged in five shells [Fig. 1(a)].
more shells than in its alloyed counterpart which binds 10 states [Fig. 1(b)]. The larger number of confined levels in a pure dot is due to a larger strain-modified conduction band offset than that in an alloyed dot. In the pure InAs/GaAs dot [Fig. 1(a)] the 1S state is located 306 meV below the conduction band minimum (CBM) of the GaAs barrier, while in the alloyed dots [Fig. 1(b)] is $\sim 118$ meV below the GaAs CBM. (This value changes by a few meVs depending on the random alloy fluctuations in the dot.) These energies of the 1S state relative to the GaAs CBM set the cutoff for conduction-to-valence intraband transitions between bound states. The P-shell consists of two non-degenerate states $1P_1$ and $1P_2$. The origin of this splitting is atomic-like and a consequence of the underlying $C_{2v}$ symmetry of the (pure, nonalloyed) dots, which is lower than $C_{\infty v}$ symmetry of the macroscopic (lens) shape, which is normally assumed in continuum effective-mass models. In our calculation we consider perfectly cylindrical dots; yet, the $P$-$P$ splitting is as large as 2 meV in pure dots. Piezoelectricity and non-cylindrical shapes further contribute to this $P$-$P$ splitting. In addition, each of these $P$ states presents a nearly equal mixture of $L_z = \pm 1$ components contrary to the axially-symmetric case in which each state has a well defined $L_z$ component. In the pure (non-alloyed) dot $1P_1$ is oriented along [110] and $1P_2$ is oriented along [110] [Fig. 2(a)]. In alloyed dots the symmetry of the dots is lower than $C_{2v}$ due to random alloy fluctuations. In this case, not only [110] and [110] are mixed and different disorder realizations (fluctuations) change the magnitude of the $P$-$P$ splitting by 1-3 meV but, more remarkably, alloy fluctuations affect the in-plane orientation (polarization) of the $P$ states, as is shown in Fig. 2. Or, equivalently, alloy fluctuations change the relative phase $\phi_2$ of the $L_z = \pm 1$ components in the $1P_1$ and $1P_2$ states, which results in different in-plane orientations (polarizations) of these states. For instance, in dot A we have $\phi_+ \simeq 0$ and $\phi_- \simeq \pi/2$, while for dot C we have $\phi_+ \simeq \phi_- \simeq 0$. In turn, the $D$-shell consists of non-degenerate $1D_1$, $1D_2$, and $2S$ states. States $1D_1$ and $1D_2$ show a nearly even mixture of $L_z = \pm 2$ components. Depending on alloy fluctuations, state $2S$ can also have sizeable $L_z = \pm 2$ components, thus making it not possible in those cases to assign a leading orbital character to these $D$-shell states.

B. Hole levels

Both non-alloyed and alloyed dots confine a large ($M_b > 20$) number of single-particle levels. Due to the multi-band nature of these hole states and for flat dots like the one we consider here, only low-lying states present shell structure that is less pronounced, i.e. larger $P$-$P$ and $D$-$D$ splittings, than in the electron case. For these states, one can still use their leading $S$, $P$, $D$ orbital character to identify them. Higher lying states show heavy mixing of orbital character. For pure and alloyed dots the 1S hole state is located, respectively, 211 meV and 186 meV above the valence band maximum of the GaAs barrier; see Fig. 1. These values are the cutoff for valence-to-valence intraband transitions between bound states. In addition, the $P$ states are not oriented along a preferential in-plane direction [Fig. 2].

![Fig. 2: (Color online) Isosurface representation of the electron (top) and hole (bottom) wavefunctions of states $1P_1$ and $1P_2$ in a pure non-alloyed InAs/GaAs dot (a), and three In$_{0.6}$Ga$_{0.4}$As/GaAs dots (b) with different alloy fluctuations. Dots are lens-shaped and have the same size: $b = 252 \, \text{Å}$ and $h = 35 \, \text{Å}$.

IV. INTRA-CONDUCTION AND INTRA-VALENCE TRANSITIONS IN DOTS WITH A SINGLE CARRIER

Assuming that only the 1S state is occupied by doping, for in-plane polarized light we expect intra-conduction transitions between bound states that satisfy $\Delta L_z = \pm 1$ when the dot is occupied by a single electron. The lowest-energy transitions correspond to $1S$-$1P_1$ and $1S$-$1P_2$ (indicated by arrows in Fig. 1). The orientation (polarization) of the $1P$ states determines the polarization properties of these transitions:

(i) For pure (non-alloyed) InAs/GaAs dots, we expect that for polarization $\hat{e} \parallel [110]$ only transition $1S$-$1P_1$ to be allowed, while for $\hat{e} \parallel [110]$ only $1S$-$1P_2$.

(ii) For alloyed dots, alloy fluctuations dictates the orientation of states $1P_1$ and $1P_2$ [Fig. 2(b)]. Thus, we expect a strong dot-to-dot dependence of the polarization properties of the two conduction intraband transitions. In particular, for the in-plane polarization $\hat{e} \parallel [110]$ we expect both transitions $1S$-$1P_1$ and $1S$-$1P_2$ to be allowed, with intensities
In a pure (non-alloyed) InAs/GaAs charged with one electron the lowest transition 1S-1P1 appears at 56 meV and is fully polarized along [110], while the second transition 1S-1P2 is located at 58 meV and is fully polarized along [110]. In contrast, in alloyed dots [Figs. 3(a) and 3(b)] we only find partial in-plane polarization: For dot A we find a transition at 46 and another at 48 meV corresponding; the lowest-energy transition 1S-1P1 is partially (∼75%) polarized along \( \hat{e} = [100] \); while the higher-energy 1S-1P2 is partially (∼75%) polarized along \( \hat{e} = [110] \).

The valence intraband transitions have a smaller oscillator strength than those of the conduction transitions. For holes, as in the case of electrons, we find two strong transitions corresponding to the 1Sh-1P0 transitions. However, these transitions are weakly in-plane polarized because the hole \( P \) states do not show any preferential in-plane orientation. In addition, we also find weaker transitions in the interval 40-60 meV, which arise from the mixing between the \( D \)-shell and \( P \)-shell hole states. For dot A, the lowest valence intraband transition shows a higher intensity when light is polarized along the [110] direction, as in the experiment of Preisler et al.22

Calculations based on simplified models are not capable of explicitly introducing alloy fluctuations. Instead, these fluctuations are naturally included within our atomistic approach. Figure 4 illustrates the effect of random alloy fluctuations on the polarization properties of the conduction intraband transitions in In_{0.5}Ga_{0.5}As/GaAs dots with the same size as dot A but with different random alloy fluctuations: (i) Under [110] polarization, dot B presents transition 1S-1P1 nearly fully polarized and transition 1S-1P2 nearly forbidden; conversely, for \( \hat{e} \parallel [100] \) transition 1S-1P2 is nearly fully polarized and 1S-1P1 nearly forbidden. These polarization properties are switched when compared to dot A. In addition, that the lowest conduction intraband transition in dot \( B \) is mainly polarized along [110] in both is in agreement with the experiment of Zibik et al.23 (ii) In contrast, dot C presents both transitions allowed for polarizations [110] and [100], with very small in-plane polarization anisotropies. We find that transition 1S-1P1 is polarized along [100], with transition 1S-1P2 forbidden, and that for \( \hat{e} \parallel [010] \) transition 1S-1P2 is allowed while 1S-1P1 is forbidden.

### B. Out-of-plane polarization— intra-valence transitions

The inset of Fig. 3(a) shows the valence intraband transition for \( \hat{e} \parallel [001] \). We find a strong feature originated from the 1Sh-1Sth transition, which involves a weakly confined, highly excited hole state with predominant light-hole character. This transition is nearly three times as intense as the in-plane valence transitions. This transition is consistent with the selection rule \( \Delta L_z = 0 \)
The configuration-interaction singlet (CI s) and triplet (CI t) are indicated. The latter being forbidden.

The energy of a many-particle state can be expressed as superposition of a Hartree-Fock term \( E^{(HF)} \) and correlation component \( \Delta \); namely,

\[
E^{(CI)}(N) = E^{(HF)}(N) + \Delta(N).
\]

For the conduction-to-conduction intraband transition energies and shifts, we compare results for the conduction intraband transition energies and shifts obtained within the Hartree-Fock approximation and CI calculations.

### A. Intra-conduction transitions vs \( N_e \)

We present Hartree-Fock (HF) expressions for the intraband transition energies when the dot is charged with \( N_e = 1, 2, 3 \) and then compare to configuration-interaction calculations. To illustrate our findings, we consider dot \( B \), which has \( 1P_1 \) and \( 1P_2 \) states that are nearly fully polarized along \([110]\) and \([1\bar{1}0]\), respectively, and focus on light polarization along \([110]\).

- \( N_e = 1 \): The ground state is \( |\Psi_0\rangle = |e_0^1\rangle \) and under IR light the final state is \( |e_1^1\rangle \). (Both states are twofold degenerate.) For this occupation, there are no many-body effects and the energy of the initial and final state are simply given by

\[
E_0(1) = \xi^{(c)}_{1S}; \quad E_f(1) = \xi^{(c)}_{1P_1}.
\]

The intraband transition energy is

\[
\hbar \omega_{f}(1) = E_f - E_i = \xi^{(c)}_{1P_1} - \xi^{(c)}_{1S}.
\]

- \( N_e = 2 \): The closed-shell (nondegenerate) state \( |\Psi_0\rangle = |e_0^2\rangle \) is the ground state, and there are four possible final states originating from \( |e_0^2\rangle \). At the single-particle level the four final states are degenerate, but within the HF approximation these states split in a singlet and triplet, and, as expected, the singlet is an allowed final state. Thus, we have the following energies for the initial and final state:

\[
E_0(2) = 2\xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + \delta_0(2) \quad \text{and} \quad E_f(2) = \xi^{(c)}_{1P_1} + \xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + K^{(ee)}_{SP_1} + \delta_f(2).
\]

This results in a \( 1S-1P \) transition energy in the presence of an extra electron that can be written as

\[
\hbar \omega_{f}(2) = \xi^{(c)}_{1P_1} - \xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + K^{(ee)}_{SP_1} - J^{(ee)}_{SP_1} + \delta_f(2) - \delta_0(2)\]

Here, \( J^{(ee)}_{SS} \) and \( J^{(ee)}_{SP_1} \) are, respectively, the direct Coulomb interaction between two electrons in \( |S\rangle \), and one electron in \( |S\rangle \) and another in \( |P_1\rangle \). For dot \( B \) our atomistic calculation predicts \( J^{(ee)}_{SS} = 20.8 \) meV and \( J^{(ee)}_{SP_1} = 16.6 \) meV, and \( K^{(ee)}_{SP_1} = 4.3 \) meV.

Our CI calculations for \( N_e = 2 \) are shown in Fig. 4(a). We find that the four states arising from configuration \( |e_0^2\rangle \) indeed split in a singlet and triplet and, as expected, only the intraband transition from the ground-state to the singlet is allowed—strong feature at \( \approx 45 \) meV. The Hartree-Fock predicted transition energy [first five terms

\[
E_0(2) = 2\xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + \delta_0(2) \quad \text{and} \quad E_f(2) = \xi^{(c)}_{1P_1} + \xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + K^{(ee)}_{SP_1} + \delta_f(2).
\]

The intraband transition energy is

\[
\hbar \omega_{f}(2) = \xi^{(c)}_{1P_1} - \xi^{(c)}_{1S} + J^{(ee)}_{SP_1} + K^{(ee)}_{SP_1} - J^{(ee)}_{SP_1} + \delta_f(2) - \delta_0(2)\]

The intraband optical absorption vs dot occupation

Absorption (arb. units)

\[10^1 \quad 10^2 \quad 10^3 \quad 10^4 \quad 10^5 \quad 10^6 \]

Intraband optical absorption vs dot occupation

Absorption (arb. units)

\[10^1 \quad 10^2 \quad 10^3 \quad 10^4 \quad 10^5 \quad 10^6 \]

FIG. 5: (Color online) For in-plane polarization \( \hat{e} \parallel [110] \), intraband conduction-to-conduction (a) and valence-to-valence (b) transitions calculated at the CI level versus number of electrons \( N_e \) and holes \( N_h \) in the dot. Black, short bars in panel (a) for \( N_e = 2 \) and 3 show the 1S-1P transition energies predicted within the Hartree-Fock approximation—Eqs. (13) and (17), respectively. For \( N_e = 2 \), the HF singlet (HF s) and the configuration-interaction singlet (CI s) and triplet (CI t) are indicated. The latter being forbidden.

for this light polarization.
in Eq. (12) for dot B appears as a short black bar in Fig. 5(a) and is 43.7 meV, while CI predicts 44.9 meV, so the correlation correction [terms in brackets in Eq. (12)] is 1.2 meV.

The spectroscopic shift between the transitions in $N_e = 1$ and 2 is thus given by

$$\Delta(2) = J^{(ee)}_{S} - J^{(ee)}_{SS} + K^{(ee)}_{SP} + [\delta f(2) - \delta_0(2)]$$

(13)

The Hartree-Fock component of this shift is 0.1 meV, whereas our CI calculation reveals a blue-shift of $\Delta(2) = 1.3$ meV; i.e., 1.2 meV from correlation effects [terms in bracket in Eq. (13)]. This shows correlation effects determine the spectroscopic shift.

$N_e = 3$: The ground state of the initial state is $|e_0^2e_1^1\rangle$ (twofold degenerate) and in addition to the final states originating from the P shell: $|e_0^3e_2^1\rangle$ and $|e_0^2e_3^1\rangle$, one now has six states arising from transitions to the D shell: $|e_0^3e_2^1\rangle$, $|e_0^2e_3^1\rangle$, and $|e_0^2e_4^1\rangle$. The energy of the ground state and the P-shell derived final are the following:

$$E_0(3) = 2\varepsilon_1^{(c)} + \varepsilon_2^{(c)} + \varepsilon_3^{(c)} + 2J^{(ee)}_{SP} - K^{(ee)}_{SP} + \delta(3)$$

$$E_{PP}(3) = 2\varepsilon_1^{(c)} + \varepsilon_2^{(c)} + 2J^{(ee)}_{SP} + J^{(ee)}_{PP} + \delta f(3).$$

(15)

Here, $J^{(ee)}_{PP}$ is the direct Coulomb interaction of two electrons occupying $P_1$. For dot B, $J^{(ee)}_{PP} = 16.0$ meV. The resulting 1S-1P$_1$ intraband transition energy is

$$\hbar\omega_{0f}(3) = \varepsilon_1^{(c)} - \varepsilon_1^{(c)} + J^{(ee)}_{PP} - J^{(ee)}_{SP} + \delta f(3) - \delta_0(3).$$

(16)

For dot B, the HF component amounts to 43.1 meV [black bar in Fig. 5(a)] and the term due to correlations (in brackets) amounts to 2.9 meV.

The spectroscopic shift between the 1S-1P$_1$ transitions in $N_e = 2$ and 3 is

$$\Delta(3) = J^{(ee)}_{PP} - J^{(ee)}_{SP} + [\delta f(3) - \delta_0(3) + \delta_0(2)].$$

(17)

The HF part is a red-shift of $\Delta(3) = -0.6$ meV. The value predicted by EMA is nearly twice as big: namely, $[\Delta(3)]_{EMA} = -1.3$ meV. Yet, our CI calculation shows [Fig. 5(a)] for $N_e = 3$ that the transition is actually blue-shifted by 1.1 meV with respect to that in $N_e = 2$.

The correlations contribution [in brackets in Eq. (17)] being 1.7 meV. Note also that the intensity of transition 1S-1P$_1$ is bleached (reduced) by nearly 50% as a consequence of Pauli blocking—one electron occupies $|1P_1\rangle$.

Regarding the P-D transitions, within the HF approximation we expect them to occur at the following energies:

$$\hbar\omega_{1}(3) = \varepsilon_1^{(c)} - \varepsilon_1^{(c)} + 2J^{(ee)}_{SP} - K^{(ee)}_{SP} + \delta(3)$$

(18)

$$\hbar\omega_{2}(3) = \varepsilon_2^{(c)} - \varepsilon_1^{(c)} + 2J^{(ee)}_{SP} - K^{(ee)}_{SP} + \delta(3)$$

(19)

$$\hbar\omega_{3}(3) = \varepsilon_2^{(c)} - \varepsilon_2^{(c)} + 2J^{(ee)}_{SP} - K^{(ee)}_{SP} + \delta(3)$$

(20)

We find in our CI calculations [Fig. 5(a)] that the strong feature around 49 meV corresponds to the (nearly overlapping) 1P-1D$_1$ and 1P-1D$_2$ transitions. The weak transition at $\sim 52$ meV arises from 1P$_1$-2S and because $|2S\rangle$ in the alloyed dot is primarily oriented (polarized) along [110] the transition is weak.

B. Valence transitions vs $N_h$

Earlier calculations assumed simple models with the incorrect symmetry and neglected the multiband nature of the hole single-particle states, which leads to an incorrect treatment of the hole-hole interaction. Within our atomistic approach, spin-orbit coupling and the multiband nature of the hole single-particle states prevent us from writing meaningful HF expressions in the case of holes. So we discuss directly the results of our CI calculations. Figure 5(b) shows the valence intraband transitions for $N_h = 1, 2,$ and 3 for light polarized along $\hat{e} \parallel [110]$. In general, compared to the conduction case, the valence intraband spectra are more sensitive to the number of holes in the dot.

$N_h = 2$: The 1S-1P$_1$ transition (lowest feature in $N_h = 1$) appears red-shifted by nearly 6 meV and split—two peaks between 8-11 meV. Due to the hole-hole exchange interaction this transition is split in a pair of low-energy, nearly doubly degenerate states and two higher-lying states mutually split by $\sim 1$ meV. Similarly, transition 1S-1P$_2$ splits in two transitions: One transition at $\sim 14$ meV, with an ensuing red-shift of 6 meV, and another at $\sim 20$ meV that appears slightly blue-shifted.
(\sim 1 \text{ meV}) \text{ with respect to the transition at } N_h = 1.

Note that contrary to the case of electrons, and due to the presence of spin-orbit interaction, the four states arising from the two-hole configuration \( h_0^1 h_2^1 \) do not split in a triplet and one singlet. Instead, these four states split in two doublets that are allowed under IR light excitation. More importantly, in the commonly used EMA with two-dimensional harmonic confinement and without spin-orbit coupling one would not find these double-peak structure of allowed transitions, but instead one would find a spectra that resembles that of the \( N_e = 2 \) electron case.

\( N_h = 3 \): While the ground state is well described by \( \{h_0^2 h_1^1\} \), the effect of configuration mixing in the final states (upon absorption) due to hole-hole interaction is remarkably pronounced and leads to a complex spectrum. As a result, it is not possible to determine unambiguously the spectroscopic shifts \( \Delta(3) \). Prominent features are the following.

(i) The lowest-energy peak at nearly 9 meV corresponds to transition \( 1S-1P_1 \). Also, the peak at \sim 15 meV is mainly \( 1S-1P_1 \), but mixed with \( 1P_1-2S \) and \( 1P_1-2S \). This mixing leads to the high intensity of this transition. Remarkably, we find that in contrast to the \( N_e = 3 \) case, the \( 1S-1P_1 \) transition is not bleached by having a hole occupying the \( 1P_1 \) state.

(ii) The peak at 10 meV correspond to transition \( 1S-1P_2 \), while the weaker feature at \sim 11 meV is due to a transition with \( 1P_1-1D_1 \) predominant character.

(iii) Above 15 meV the features in the spectrum correspond to transitions to heavily mixed final configurations: (a) The lower-energy peak in the double-peak structure around 20 meV corresponds to a mixture of the allowed \( 1S-1P_1 \) and \( 1S-1P_2 \) transitions in addition to a sizeable component (16\%) of the forbidden \( h_0^2 h_1^1 - h_0^1 h_2^1 \) transition. In turn, the higher-energy peak is a mixture of \( 1S-1P_2 \) and \( 1P_1-1D_1 \) transitions. (b) The peak at 26 meV arises from two nearly overlapping transitions. These transitions are a mixture of allowed \( P-D \) and \( P-F \) transitions, as well as forbidden transitions.

(iv) Although significantly weaker than the other features in the spectrum, the peak at 24 meV corresponds to a forbidden transition made allowed by configuration mixing with allowed transitions. We also have found this type of transitions in the interband spectra of (In,Ga)As/GaAs dots.\textsuperscript{23}

VI. SUMMARY

By combining an atomistic, pseudopotential-based approach with the configuration method, we have calculated the conduction and intraband transitions in (In,Ga)As/GaAs quantum dots with up to three carriers. We illustrated our calculations with a prototypical lens-shaped \( \text{In}_{0.6}\text{Ga}_{0.4}\text{As/GaAs} \) dot with diameter \( b = 252 \text{ Å} \) and height \( h = 35 \text{ Å} \). And as a benchmark, for dots charged with a single carrier, we provided results for a pure non-alloyed InAs/GaAs dot with the same size. We have made specific predictions that could be probed in single-dot infrared spectroscopy of n-doped and p-doped dot:

(i) In pure, non-alloyed InAs/GaAs dots, the \( 1S-1P \) conduction intraband transitions are fully in-plane polarized, while valence transitions are weakly polarized because the hole \( P \) states do not show any in-plane preferential orientation.

(ii) In alloyed \( \text{In}_{0.6}\text{Ga}_{0.4}\text{As/GaAs} \) dots the in-plane polarization of \( 1S-1P \) conduction intraband transitions strongly depend on alloy fluctuations, which change the in-plane orientation of the nearly generate P-shell states. The polarization of valence intraband transitions is insensitive to changes in alloy fluctuations.

(iii) Upon changing the number of carriers in the dot, the intraband transitions display spectroscopic shifts of about 1-2 meV. These shifts are not well described within Hartree-Fock, instead their magnitude is determined by correlation effects.

(iv) Spin-orbit coupling and the multi-band characteristic of holes states result in important differences between the many-particle valence and conduction intraband spectra. Spectroscopic shifts can only be determined unambiguously for conduction transitions.

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In reality one expects the dots to be alloyed due to interdiffusion, making pure InAs/GaAs dots an idealization. However, Schmidt et al. [Phys. Rev. B 54, 11346 (1996)] and others have presented data that show exciton recombination at about 1 eV, which has been attributed to pure non-alloyed InAs/GaAs dots.