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

Gustavo A. Narvaez* and Alex Zunger
Na a Re e ab e E e Lab a , G de , C ad 80401, USA
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V_{SO} is a nonlocal pseudopotential that accounts for the spin-orbit interaction.[23] 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 ψ to solve Eq. (1) is a linear combination of f $\backslash$ e Bloch bands of the underlying solids.[24] Thus, this method incorporates multiband and multivalley coupling, band nonparabolicity, and spin-orbit effects, as well as the effects of the underlying strain in the dot and barrier.

To solve for the many-particle states $\{\Psi_\nu(\mathcal{N}), E_\nu(\mathcal{N})\}$ of the dot with $\mathcal{N}$ carriers, where $\mathcal{N}=N_e$ electrons or N holes, we use a configuration-interaction (CI) approach with screened direct (J) electron-electron and hole-hole Coulomb interaction and exchange (K).[25] This method has been recently applied to the calculation of electronic and optical properties of (In,Ga)As/GaAs dots such as electron and hole charging,[26] radiative lifetimes of neutral and charged excitons,[28] relaxation times of electrons due to electron-hole Auger scattering,[29] and fine-structure splittings of neutral and charged excitons.[30]

At low temperatures such that only the ground state $\Psi_0(\mathcal{N})$ of the $\mathcal{N}$-carrier dot is significantly occupied, the optical absorption spectrum for light polarized along $\mathbf{e}$ is given by

$$I(\hbar\omega;\mathbf{e}) = \sum_{\nu'} |\langle\Psi_{\nu'}|\mathbf{e}\ \ \mathbf{p}|\Psi_0\rangle|^2 \delta(E_{\nu'}\ \ E_0\ \ \hbar\omega). \quad (2)$$

In the results we present subsequently, we have phenomenologically broadened the spectra with a Gaussian of width σ =0.25 meV. Such a broadening has also been used in other simulations of optical absorption ier a rattm273 Tmr78 TDITjthe

ute to this P-P splitting. In addition, each of these P states present 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 (nonalloyed) dot $1P_1$ is oriented along $[\bar{1}10]$ 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 $[\bar{1}10]$ 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 ϕ 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 nondegenerate $1D_1$, $1D_2$, 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 nonalloyed and alloyed dots confine a large ($M > 20$) number of single-particle levels. Due to the multiband 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].

IV. INTRACONDUCTION AND INTRAVALENCE TRANSITIONS IN DOTS WITH A SINGLE CARRIER

Assuming that only the $1S$ state is occupied by doping, for in-plane polarized light we expect intraconduction 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 (nonalloyed) [illegible]TTA-390d7no5GaAs-286.3dot,s-28645w

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 $In_{0.6}Ga_{0.4}As/GaAs$ dots with the same size as dot A but different random alloy fluctuations: (i) Under $[\bar{1}10]$ polarization, dot B presents transition $1S$-$1P_1$ nearly fully polarized and transition $1S$-$1P_2$ nearly forbidden; conversely, for $\mathbf{e}\|[110]$ transition $1S$-$1P_2$ is nearly fully polarized and $1S$-$1P_1$ nearly forbidden. These polarization properties are , c ed when compared to dot A. In addition, that the lowest conduction intraband transition in d B is mainly polarized along $[\bar{1}10]$ in both is in agreement with the experiment of Zibik e a .[16] (ii) In contrast, dot C presents both transitions allowed for polarizations $[110]$ and $[\bar{1}10]$, with very small in-plane polarization anisotropies. We find that transition $1S$-$1P_1$ is polarized along $[100]$, with transition $1S$-$1P_2$ forbidden, and that for $\mathbf{e}\|[010]$ transition $1S$-$1P_2$ is allowed while $1S$-$1P_1$ is forbidden.

B. Out-of-plane polarization—intravalence transitions

The inset of Fig. 3(a) shows the valence intraband transition for $\mathbf{e}\|[001]$. We find a strong feature originated from the $1S$ -$1S$ 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 $\Delta L_z = 0$ for this light polarization.

V. INTRACONDUCTION AND INTRAVALENCE TRANSITIONS IN DOTS WITH A FEW CARRIERS

We now study the conduction and valence intraband transitions for $N=2$, and 3 carriers occupying the dot.

The energy of conduction and valence band transitions in the presence of N_e electron or N holes is dictated by differences in total energies. [See Eq. (2).

$N_e=2$: 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 $|e_0^1 e_1^1\rangle$. At the single-particle level the four final states are degenerate, but within the HF approximation these states split in a triplet ($\uparrow\uparrow$) and a singlet ($\uparrow\downarrow$): $|e_0^1 e_1^1\rangle_{\uparrow\uparrow}$ and $|e_0^1 e_1^1\rangle_{\uparrow\downarrow}$.

$$\hbar\omega_3(3) = \mathcal{E}^{(e)}_{2S} \quad \mathcal{E}^{(e)}_{1P_1} + 2J^{(ee)}_{SP_1} \quad K^{(ee)}_{SP_1} \quad 2J^{(ee)}_{S2S} \quad K^{(ee)}_{S2S} + \delta_3(3). \tag{20}$$

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 ~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 e_l. So we discuss directly the results of our CI calculations. Figure 5(b) shows the valence intraband transitions for $N=1$, 2, and 3 for light polarized along $\mathbf{e}\|[\bar{1}10]$. In general, compared to the conduction case, the valence intraband spectra are more sensitive to the number of holes in the dot.

$N=2$: The $1S$-$1P_1$ transition (lowest feature in $N=1$) appears redshifted 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 ~1 meV. Similarly, transition $1S$-$1P_2$ splits in two transitions: One transition at ~14 meV, with an ensuing redshift of 6 meV, and another at ~20 meV that appears slightly blueshifted (~1 meV) with respect to the transition at $N=1$.

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 $\begin{smallmatrix}1 & 1\\ 0 & 2\end{smallmatrix}$ 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.

$N=3$: While the ground state is well described by $|\begin{smallmatrix}2 & 1\\ 0 & 1\end{smallmatrix}\rangle$, 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 ~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 [illegible] 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 ~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 $\begin{smallmatrix}2 & 1\\ 0 & 1\end{smallmatrix}$ - $\begin{smallmatrix}1 & 2\\ 0 & 2\end{smallmatrix}$ transition. In turn, the higher-energy peak is a mixture of $1S$-$1P_2$ and $1P_1$-D_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 [illegible] 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 [illegible] spectra of (In,Ga)As/GaAs dots.[33]

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 $In_{0.6}Ga_{0.4}As$/GaAs dot with diameter $b=252$ Å and height $=35$ Å. And as a benchmark, for dots charged with a single carrier, we provided results for a pure nonalloyed InAs/GaAs dot with the same size. We have made specific predictions that could be probed in [illegible] infrared spectroscopy of -doped and -doped dot:

(i) In pure, nonalloyed 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 $In_{0.6}Ga_{0.4}As$/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 multiband characteristic of holes states result in important differences between the [illegible] valence and conduction intraband spectra. Spectroscopic shifts can only be determined unambiguously for conduction transitions.

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*Present address: Eclipse Energy Systems, Inc., St. Petersburg, Florida 33710; Electronic email: gnarvaez@eclipsethinfilms.com
Electronic address: alex_zunger@nrel.gov

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