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Exploring the potential of c-plane indium gallium nitride quantum dots for twin-photon emission

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Abstract

Non-classical light emission, such as entangled and single-photon emission, has attracted significant interest due its importance for future quantum technology applications. In this work, we study the potential of wurtzite (In,Ga)N/GaN quantum dots for novel non-classical light emission namely twin-photon emission. Our calculations, based on a fully atomistic many body framework, reveal that the combination of carrier localization due to random alloy fluctuations in the dot, spin-orbit coupling effects, underlying wurtzite crystal structure and built-in electric fields lead to an excitonic fine structure that is very different from more "conventional" zincblende (In,Ga)As dots, which have been used so far for twin photon emission. We show and discuss here that the four energetically lowest exciton states are all bright and emit linearly polarized light. Furthermore, three of these excitonic states are basically degenerate. All these results are independent of the alloy microstructure. Therefore, (In,Ga)N/GaN dots are very promising candidates to achieve efficient twin photon emission, potentially at high temperatures and over a wide emission wavelength range.

Keywords

Excitonic fine structure, quantum dots, twin-photon emission, InGaN

Introduction

Integrated non-classical light emitters such as single- and entangled-photon sources are a critical building block for realizing future quantum technologies.[1–4] These intriguing non-classical states are particularly attractive for long-distance distribution of quantum information due to their ease of manipulation and non-interacting nature of photons used in these schemes.[4,5] For this reason single- and entangled photon emitters have been in the center of research activities around the world in recent years. Especially semiconductor quantum dots (QDs) based on material systems such as (In,Ga)As/GaAs, GaN/AlN and (In,Ga)N/GaN have been of strong interest for such applications. For instance, thanks to their large band offsets, high excitonic binding energies and tunable emission wavelength, single photon emission from (In,Ga)N/GaN and (Al,Ga)N/AlN have been reported for temperatures of 200 K and above. This feature makes them highly attractive for novel and future non-classical light sources at ambient temperatures, in contrast to zincblende

(In,Ga)As/GaAs dots.[6–8]

Very recently, and in addition to entangled- and single-photon emission, further non-classical light emitters, so-called multi-photon sources, have been proposed in the literature.[9–11] These multi-photon states have been highlighted to have potential applications in quantum optical spectroscopy and quantum biology.[9–13] Such light sources are designed to release photons in the groups of N-photons, all having identical energy and optical polarization. Here, in particular twin-photon states ($N = 2$) attracted attention for studying nonlinear phenomena in quantum optics.[13] QDs are again ideal candidates for generating such twin photons by utilizing the biexciton (XX)-exciton (X) cascade.[9,11] For instance, Heindel *et al.*[9] demonstrated deterministic twin photon emission from (001)-oriented zincblende (In,Ga)As/GaAs QDs by manipulating the energies of excitonic states in a $XX - X$ cascade. The concept is schematically illustrated in Fig. 1 (a), showing the energy of the biexciton ground state (E_{XX}) and the four energetically lowest lying exciton states (E^i_X), in a (001)-oriented (In,Ga)As/GaAs QD. The light polarization characteristics of the emitted photons are either given by symbols next to the arrows (V-vertical; H-horizontal) or in the brackets (z-pol- polarized along growth direction; dark-not optically active). By tailoring the excitonic and biexcitonic energies of the dot, by e.g. varying dot size or shape, so that E_{XX}=$2E^2_X$, from the decay channel E_{XX}–E^2_X–0 of the $XX-X$ cascade (cf. Fig. 1) twin-photons can be generated.[9] In doing so, the authors were able to achieve superimposed emission of exciton and biexciton (H^2_X+H^2_{XX}) transition, therefore twin photons with H-polarization. A schematic representation of the polarization resolved photoluminescence (PL) spectra of such a QD is illustrated in Fig. 1 (b). A similar study was recently carried out by Moroni *et al.*,[11] extending this concept to (In,Ga)As QDs grown along the [111]-crystallographic direction.

So far, all studies on twin photon generation have utilized arsenide dots. However, as already highlighted above, nitride-based systems have attracted significant interest for non-

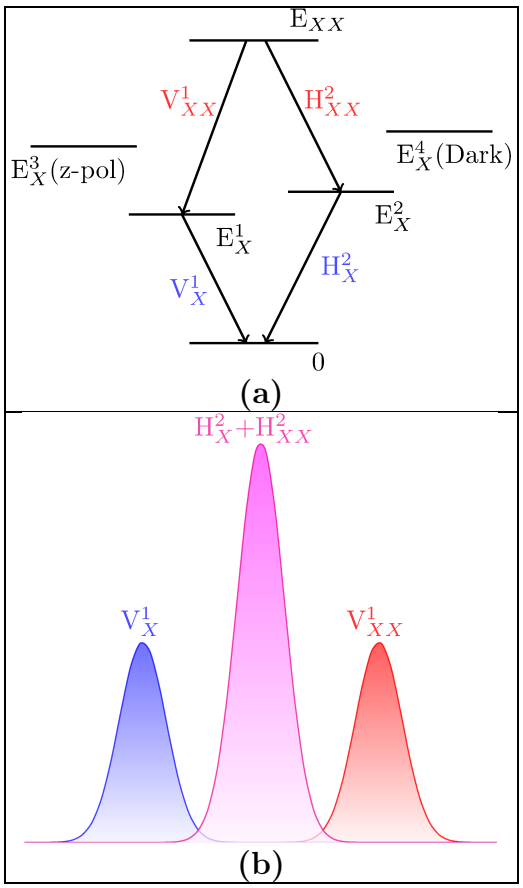


Figure 1: Schematic illustration of (a) biexciton-exciton cascade in a (001)-oriented (In,Ga)As/GaAs QD; (b) a corresponding luminescence spectra for the case that E_{XX}=$2E^2_X$.

classical light emission near or above room temperature. Therefore, given the growing interest in non-classical light emitters, it is key to understand the potential of III-N QDs for twin-photon emission. It is also important to note that the underlying wurtzite crystal structure of III-N materials leads to very different electronic and optical properties of nitride-based heterostructures when compared to zincblende (001)-oriented arsenide systems.[14,15] This starts with very strong electrostatic built-in fields and ranges over to strong carrier localization effects.[16] Both effects have been shown to impact the electronic and optical properties of (In,Ga)N-based heterostructures significantly and are essential for an accurate description of these systems.[16,17] Therefore, results obtained on (001)-oriented (In,Ga)As dots *cannot* be carried directly over to explain the excitonic fine structure of *c*-plane (In,Ga)N/GaN wurtzite

QDs. So far there exists no detailed theoretical analysis of the exciton fine structure of realistic (In,Ga)N/GaN dots accounting for carrier localization effects due to random alloy fluctuations and its consequences for multi-photon emission in these systems.

In this letter we address this question in the framework of atomistic electronic structure theory combined with many-body configuration interaction calculations for realistically sized *c*-plane (In,Ga)N/GaN QDs, given that these emitters are ideally suited to operate at much higher temperatures than for instance (In,Ga)As dots and at the same time allow for tuning of the emission wavelength in the visible spectral range. Our calculations reveal that the combination of carrier localization effects, spin-orbit coupling, underlying wurtzite crystal structure and strong electrostatic built-in fields results in a very different excitonic fine structure when compared conventional (001)-oriented (In,Ga)As/GaAs dots. For instance, we find that the four energetically lowest lying exciton states are all bright and one is left with three equivalent channels for twin-photon emission. All these findings are independent of the alloy microstructure and therefore highlight the potential insensitivity of these dots against structural variations. Overall, these peculiarities of the calculated excitonic fine structure indicate that (In,Ga)N/GaN QDs are good candidates for twin-photon emission. Furthermore, the intrinsic features of III-N nitride materials make them an intriguing choice for future non-classical light emitters operating near room temperature and potentially over a wide emission wavelength range.

Theoretical framework and QD geometry

To calculate the exciton fine structure of realistic *c*-plane (In,Ga)N/GaN QDs, in a first step their electronic structure has to be investigated. To resolve alloys fluctuations on a microscopic level, an atomistic approach is required. Here, we apply the atomistic nearest neighbor sp^3 tight-binding (TB) model introduced in detail in Refs. 18 and 19. Given the large lattice mismatch between InN and GaN, strain effects have to be taken into account. These calculations are performed in the frame of a valence force field (VFF)[20] model that accounts for electrostatic effects and has been been implemented in the software package LAMMPS.[18,21] Details of the VFF model are given in Ref. 18. (In,Ga)N/GaN heterostructures grown along the polar *c*-axis of the wurtzite crystal experience very strong electrostatic built-in fields.[15] These (local) polarization fields are included in the model via a local polarization theory, which is coupled to a point dipole model[19] to calculate the corresponding (local) built-in potential. The whole framework has been benchmarked against theory and experiment for bulk systems, and has shown to give a very good description of the electronic and optical properties of (In,Ga)N/GaN quantum well systems even for different growth planes.[22,23] Therefore this model presents an ideal starting point for atomistic electronic structure calculations of *c*-plane (In,Ga)N/GaN dots to account for alloy fluctuations on a microscopic level. For an accurate modeling of the exciton fine structure, spin-orbit coupling (SOC) has to be included in the calculations. Here, SOC effects are accounted for following Ref. 24, which is a widely used approach in the literature.

Calculating the exciton fine structure is inherently a many-body problem. To do so we employ the configuration interaction (CI) scheme and include both direct and electron-hole exchange interactions. The corresponding Coulomb matrix elements are calculated directly from the TB wave functions as detailed for instance in Refs. 25 and 26 and by other groups.[27–29] In the following we include six electron and ten hole states in the CI expansion, reflecting the smaller energetic separation of hole states when compared to electrons. To evaluate optical spectra, Fermi's golden rule is applied and the required dipole matrix elements are calculated from the TB wave functions as described in Ref. 26.

Additionally, the theoretical framework requires QD geometry and In atom distribution as input. Here, we consider a truncated-cone

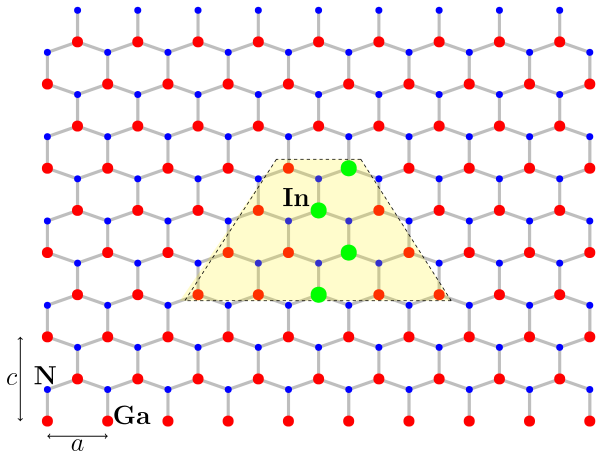


Figure 2: Schematic illustration of a truncated-cone shaped c-plane (In,Ga)N QD, including alloy fluctuations, buried in a GaN matrix.

shaped (In,Ga)N QD embedded inside a GaN matrix, which is in good agreement with recent transmission electron microscopy (TEM) studies on c-plane (In,Ga)N QDs.[30] The experimental study in Ref. 30 revealed also average diameters of the investigated c-plane QDs of ≈10-20 nm and dot heights varying between 3-5 nm. Following this recent experimental data, a base diameter of 13 nm and a height of 3 nm has been assumed in the modeling. Previous theoretical studies on c-plane (In,Ga)N QDs have assumed similar geometries.[31,32] The QD geometry is schematically shown in Fig. 2.

As mentioned above, information about the In content and the distribution of In atoms inside the dot is required. Usually, dots with In contents between 20 % and 25% are reported in the literature,[33,34] with only few reports targeting (In,Ga)N dots with very high In contents of 40% to achieve red emission.[35] Following more conventional experimental data, we work in this letter with 20% In. Regarding the In atom distribution, based on atom probe tomography data on (In,Ga)N alloys and quantum wells with In contents near or below 20%,[36,37] a random alloy assumption has been made here. This means in a selected area (shape of a truncated cone with above given dimensions, as indicated in Fig. 2) of our supercell, Ga atoms have been randomly replaced by In atoms. To study the impact of the alloy microstructure on the results, the calculations have been repeated 5 times, meaning 5 different microscopic configurations have been generated.

All calculations have been performed on supercells with periodic boundary conditions and a dimension of ≈29.33 nm×25.40 nm×18.66 nm (1,218,816 atoms). Bearing in mind that the symmetry of the combined system of underlying wurtzite lattice and assumed QD geometry (truncated cone) ideally is C_{3v} when neglecting alloy fluctuations by using for instance a virtual crystal approximation (VCA), the supercell has to be sufficiently large to avoid artifacts arising from the cubic supercell boundaries. To this end, a VCA calculation has been performed to ensure that the simulation box is large enough so that strain and built-in fields do not lift degeneracies in the single-particle states and/or the excitonic fine structure. These results will be discussed in detail elsewhere, since our focus here is on a realistic, beyond VCA, description of the excitonic properties and its consequences for twin photon emission from (In,Ga)N/GaN dots.

Results and discussion

As already pointed out in the introduction, to study twin photon emission from c-plane (In,Ga)N dots, it is important to analyze the excitonic fine structure and the light polarization characteristics of the emitted photons. However, before investigating these questions we turn our attention to the electronic structure of c-plane (In,Ga)N/GaN dots and discuss the impact of the alloy fluctuations on the results.

Electronic structure: Impact of alloy fluctuations

Figure 3 depicts the electron $|\phi^e_{\rm GS}|^2$ (red) and hole (green) ground state $|\phi^h_{\rm GS}|^2$ probability densities for two arbitrarily chosen alloy configurations, namely Config. 3 and Config. 4. The probability densities are plotted at 10% and 50% of the respective maximum values. The gray shaded area indicates the QD region and the dashed lines the dot barrier interfaces. The data are shown for a top view along the wurtzite

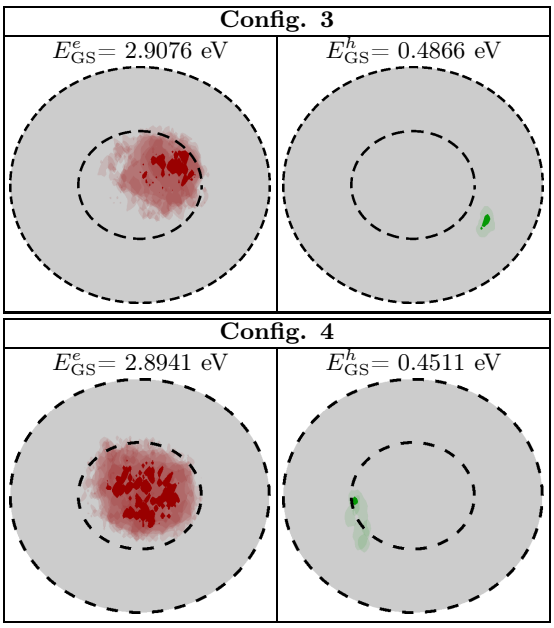


Figure 3: Isosurface plots of single-particle electron (first column) and hole (second column) ground state charge densities $|\phi^e_{GS}|^2$ and $|\phi^h_{GS}|^2$, respectively, for a $In_{0.20}Ga_{0.80}N$ QD. The results are shown for the alloy configurations Config. 3 (upper row) and Config. 4 (lower row). Here, the data are shown in a top view (along $c-$axis) for 10% (light) and 50% (dark) of the respective maximum charge densities. The corresponding energies are denoted by E^e_{GS} and E^h_{GS} for electrons and holes, respectively.

c-axis. Several interesting features are visible from these plots. First, we find that the ground state charge densities (especially hole) are deformed due to the random distribution of In and Ga atoms in the dot. This is in strong contrast to previous theoretical results studying c-plane (In,Ga)N/GaN QDs in the frame of VCA type calculations and/or continuum-based $\mathbf{k}\cdot\mathbf{p}$ theory.[38] Second, hole localization effects are more pronounced when compared to the electrons. We attribute this effect to the higher effective mass of holes when compared to electrons.[18] Finally, the hole wave functions do not necessarily localize directly under the electron wave functions. Again this finding is very different to conclusions that are usually drawn from continuum or VCA type calculations.[38] The other configuration (Config. 4) illustrated in Fig. 3 reveals similar features. We stress here that all these effects are not a peculiarity of the chosen microscopic configurations and all other configurations show a similar behavior. Furthermore, in terms of wave function localization effects due to random alloy fluctuations, our findings are consistent with observations made by several independent groups on (In,Ga)N/GaN quantum wells.[16,17,39,40] Overall, our analysis and results highlight that the alloy microstructure significantly affects the electronic structure of c-plane (In,Ga)N/GaN QDs and introduces effects that are very different from a continuum-based approximation.

It is also important to note that these carrier localization effects are much more pronounced in the here studied c-plane (In,Ga)N/GaN QDs when compared to (001)-oriented (In,Ga)As/GaAs dots.[41] But, in (In,Ga)As dots alloy fluctuation can already significantly affect the excitonic fine structure.[42] Therefore, given the here observed strong localization effects in (In,Ga)N QDs, the question remains how does this affect the excitonic fine structure and what are the implications for twin-photon emission from such dots? Before presenting the numerical results, we first apply group theory to understand how the symmetry/reduced symmetry will affect the excitonic fine structure.

Excitonic fine structure of c-plane (In,Ga)N QDs: Insights from group theory

Starting with an ideal c-plane (In,Ga)N/GaN QD in the absence of random alloy fluctuations, e.g. VCA, several studies have already concluded that the symmetry of the combined system of QD geometry (e.g. truncated cone, lens) and wurtzite lattice is C_{3v}.[25,43] In the presence of SOC, the single-particle states can be represented by the irreducible representations (IRR) of the C_{3v} double group ($\bar{C}_{3v}$). One can show that the hole ground state ϕ^h_{GS} transforms according to a two-dimensional $E_{3/2}$ representation.[44] On the other hand, the electron ground state ϕ^e_{GS} transforms according to the two-dimensional IRR $E_{1/2}$. Following Refs. 44 and 45, the exciton ground states transform ac-

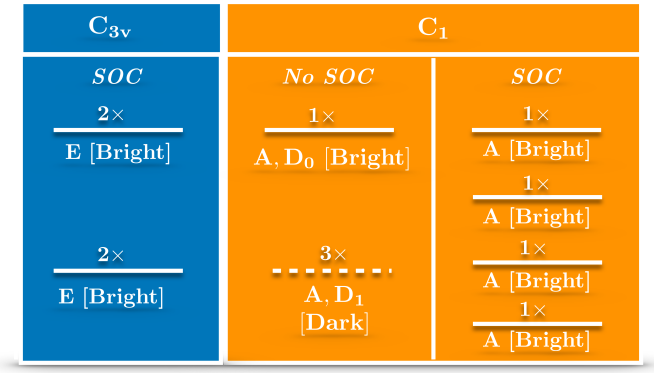


Figure 4: Exciton fine structure for systems with different point group symmetries and when accounting for electron-hole exchange interactions. Notations are explained in the main text.

cording to $E_{1/2} \otimes E_{3/2} = E \oplus E$. Since E is a two-dimensional IRR of $\bar{C}_{3v}$, the four energetically lowest exciton states form two doublets of bright states, which are polarized in the x-y-plane/in the c-plane. This has been discussed for instance in detail in Ref. 44 and a schematic illustration is given in Fig. 4.

However, this analysis is only true for highly symmetric dots. Our calculations on the single-particle states in the previous section show already that this is not the case and the C_{3v} symmetry is lost due to random alloy fluctuations and the connected carrier localization effects. Therefore, the system is described by the point group C_{1v}.[45] Since the SOC is small in InN ($\Delta_{\mathrm{SOC}}^{\mathrm{InN}} = 5$ meV) and GaN ($\Delta_{\mathrm{SOC}}^{\mathrm{GaN}} = 17$ meV),[46] at least when compared to InAs ($\Delta_{\mathrm{SOC}}^{\mathrm{InAs}} = 390$ meV) or GaAs ($\Delta_{\mathrm{SOC}}^{\mathrm{GaAs}} = 341$ meV),[47] we start our group theoretical analysis by *neglecting* this contribution. The C_{1v} single group contains only one element, namely A, which is a one-dimensional IRR. Thus, the exciton ground state is of $A \otimes A = A$ symmetry. When including spin in the description but neglecting SOC, so that the spin is still a good quantum number, one expects, due to the electron-hole exchange interaction, a singlet (D_0 symmetric) and triplet (D_1 symmetric) splitting in the four energetically lowest exciton states.[45,48] The situation is schematically illustrated in Fig. 4. In this case three dark (triplet) and one bright state (singlet) are expected.

Turning to the realistic case of a C_{1v} symmetric system with SOC, thus dealing with the double group of C_{1v}, based on the IRR of this group, the exciton states transform according to $A_{1/2} \otimes A_{1/2} = A$ symmetry. Again, since the double group $\bar{C}_{1v}$ has only one element, $A_{1/2}$, and A is a one dimensional IRR, the four energetically lowest excitonic states should all be non-degenerate and bright.[45] This is again schematically displayed in Fig. 4. Equipped with this information, we now turn to our numerical data and discuss the implications of the resulting excitonic fine structure for twin-photon emission.

Excitonic fine structure and twin-photon generation

To analyze twin-photon emission from c-plane (In,Ga)N/GaN dots we focus in the following on (i) the excitonic fine structure and (ii) the light-polarization characteristics of the involved excitonic states. In a first step, we look at the excitonic fine structure. Figure 5 (a) depicts the energies of the four energetically lowest exciton states as a function of the microscopic configuration number (inset shows the exciton binding energy E_b^X, which will be discussed in more detail below). For the four energetically lowest exciton states, the zero of energy is always chosen to be the energetically lowest lying state of each configuration. In doing so, the calculated fine structure splitting can be studied and compared more easily. Before looking at the fine details, Fig. 5 (a) clearly demonstrates that the exciton fine structure, *independent* of the microscopic configuration, is always the same: we find a single non-degenerate state lying energetically above three states which are approximately degenerate in energy.

At first glance this result is in contrast to our group theoretical investigation of the C_{1v} double group, where four non-degenerate states are expected. Instead, even though the calculations include SOC effects, the obtained excitonic fine structure gives the impression to reflect the results expected for the C_{1v} single group, thus where the spin is a good quantum number. Using our group theoretical analysis, one might be tempted to classify the excitonic

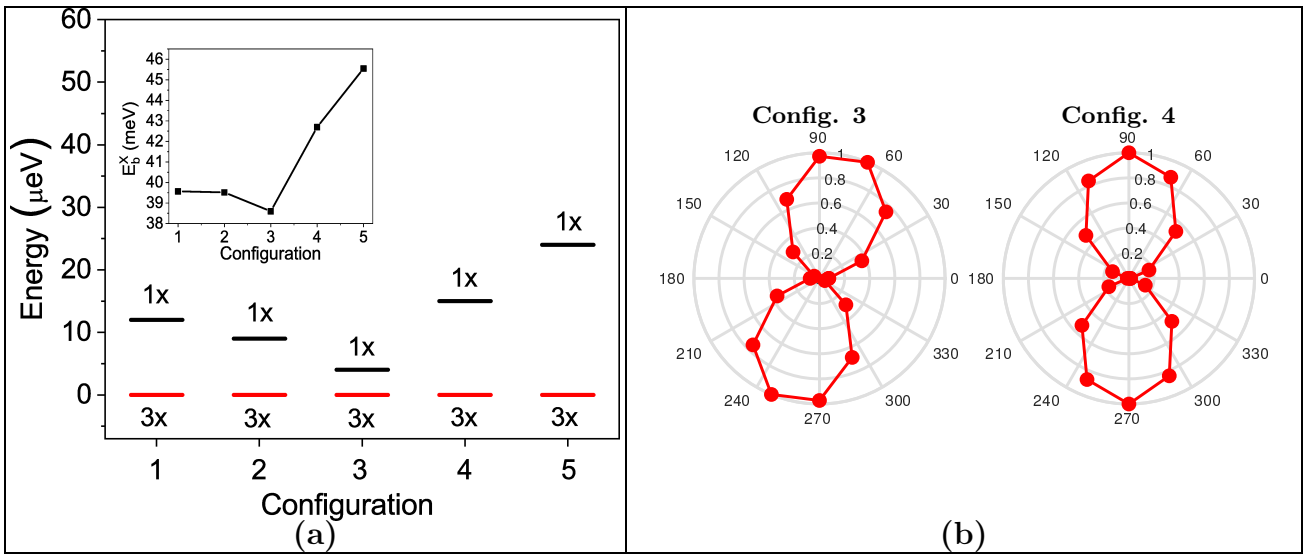


Figure 5: (a) Excitonic fine structure for the five different microscopic configurations. The degree of degeneracy is indicated by the numbers. Inset: Excitonic binding energy E_b^X as a function of the microscopic configuration. (b) polar plot of the optical intensity of the exciton ground state emission as a function of the in-plane light polarization vector for Configs. 3 and 4.

states as singlet (non-degenerate) and triplet (three-fold degenerate) states. However, given that the energetically lower lying states are not exactly degenerate, this indicates that the SOC introduces singlet-triplet mixing effects as discussed in other systems.[49] We come back to this below. To further validate and analyze our fine structure results, we have also performed a VCA calculation for our *c*-plane truncated-cone shaped $In_{0.2}Ga_{0.8}N$/GaN dot, leaving us with a C_{3v} symmetric system.[25,43] Our calculations of the exciton fine structure reveal two doublets of degenerate states (not shown), as expected and predicted from our group theoretical analysis of the $\bar{C}_{3v}$ group. Overall this highlights two important factors (i) that including carrier localization effects significantly affects the excitonic fine structure of *c*-plane (In,Ga)N/GaN QDs and (ii) that even though the SOC is weak in InN and GaN systems, the $\bar{C}_{3v}$ double group has to be used for understanding the results; otherwise, for instance in the VCA case, the splitting into two doublets would not occur.

We note also that our calculated excitonic fine structure splittings are much smaller than typical fine structure splittings (bright-bright, dark-bright, dark-dark) reported for InAs/GaAs QDs. In (In,Ga)As/GaAs dots values of the order of 60 μeV (bright-bright) and 500 μeV (dark-bright) are found in the literature.[50] In our case the maximum fine structure splitting is around 25 μeV, with more configurations showing values close to 10 to 15 μeV. We attribute the much smaller excitonic fine structure splittings in *c*-plane (In,Ga)N/GaN QDs to the presence of the strong electrostatic built-in field along the growth direction in these systems. This field spatially separates electron and hole wave functions along the *c*-axis and reduces the wave function overlap.[26,38] As a consequence, the electron-hole exchange matrix elements, which mainly give rise to fine structure splittings, are reduced. This small exchange contribution, when compared to a InAs/GaAs dot for instance, will also affect the singlet-triplet state mixing, given that this contribution affects the energetic separation of these states.

While the excitonic fine structure splitting depends mainly on the electron-hole exchange matrix elements, the exciton binding energy, defined as the difference between the single-particle and many-body energy gap, is mainly determined by the *direct* Coulomb interaction. Previous studies have already revealed (very) high exciton binding energies (>50 meV) in *c*-

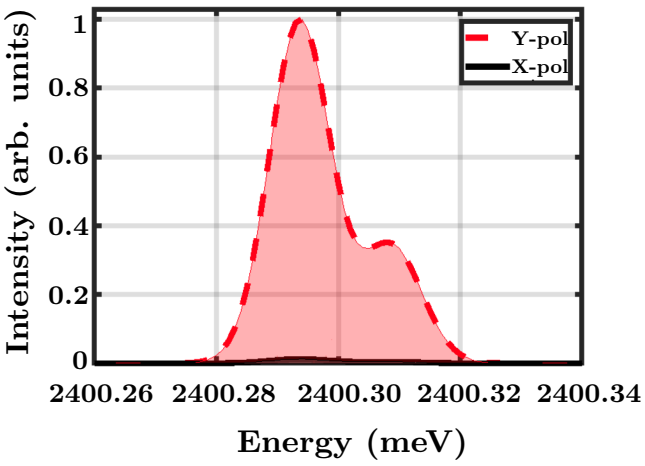


Figure 6: Excitonic ground state emission spectrum of Config. 4 for two orthogonal in-plane light polarizations (X-pol (red-dashed); Y-pol (black solid)). The emitted light is mainly polarized along the y-axis (red-dashed line); this high degree of optical polarization reflects the behavior in Fig. 5 (b) for Config. 4. The two peaks in the spectrum correspond to threefold (lower energy) and non-degenerate excitonic states.

plane (In,Ga)N/GaN QDs.[38] These high values originate from higher effective masses and lower dielectric constants in III-N materials when compared to zincblende (In,Ga)As systems. The inset of Fig. 5 (a) depicts the calculated exciton binding energy E_b^X for the five different microscopic configurations. Two interesting aspects can be deduced from this figure. First, the magnitude of E_b^X follows the trend of the fine structure splitting (lower fine structure splitting lower E_b^X value), indicating that the microscopic configuration affects both exchange and direct Coulomb interaction in a similar way. Second, the calculated exciton binding energies are in the range of 38 - 46 meV, which exceed the thermal energy at room temperature ($\approx$ 26 meV). This is thus indicative of stable excitons at room temperature, a significant advantage over (In,Ga)As dots for twin-photon emission.

Having discussed the excitonic fine structure (splitting), we now turn to the second important factor for twin-photon emission, the light polarization characteristics of the photons emitted from transitions involving the different excitonic states. Using Fermi's Golden rule, we have calculated the excitonic emission spectra of our dots for different microscopic alloy configurations. Figure 5 (b) depicts the polar plot of the optical intensity of the exciton ground state emission as a function of the in-plane light polarization vector for Configs. 3 and 4. Figure 5 (b) clearly reveals that the excitonic ground state emission in both cases is strongly linearly polarized. We find that this is the case for all configurations studied. Overall, this result is in excellent agreement with experimental data, which reports strongly linearly polarized excitonic emission spectra from single c-plane (In,Ga)N/GaN QDs with 20% In content.[51]

Furthermore, we find that the four energetically lowest states are all optically active and show the same polarization properties as the lowest state. As an example, Fig. 6 shows the excitonic ground state emission spectrum of Config. 4. This indicates that the combination of the small electron-hole exchange interaction in combination with SOC effects lead to singlet-triplet mixing effects; otherwise for pure singlet and triplet states, bright (singlet) and dark (triplet) states should be expected.[52,53] Overall, we stress here that the observed excitonic fine structure is very different from a C_{2v} symmetric (In,Ga)As/GaAs QD, where usually two bright and two dark states are predicted by theory and observed in experiment.[54,55]

Taking all this together it allows us now to illustrate the $XX-X$ cascade in c-plane (In,Ga)N/GaN dots, which is presented in Fig. 7 (a). We highlight here that all of the decay channels of the $XX-X$ cascade are bright and emit linearly polarized light. Such a system is therefore highly attractive for twin photon emission for the following reasons. First, all emitted photons from the $XX-X$ cascade should be linearly polarized. As discussed in the introduction, a prerequisite for twin-photon emission is that the photons have identical optical polarization. Second, and similar to C_{2v} symmetric (In,Ga)As/GaAs dots, cf. Fig. 1, when tuning the biexciton and excitonic energies of the dot, by varying for instance dot size and shape, one is left with two potential "energy" channels for twin-photon emission, namely the cascades (E_{XX}=$2E_X^1$ $\approx$$2E_X^2$ $\approx$$2E_X^3$)

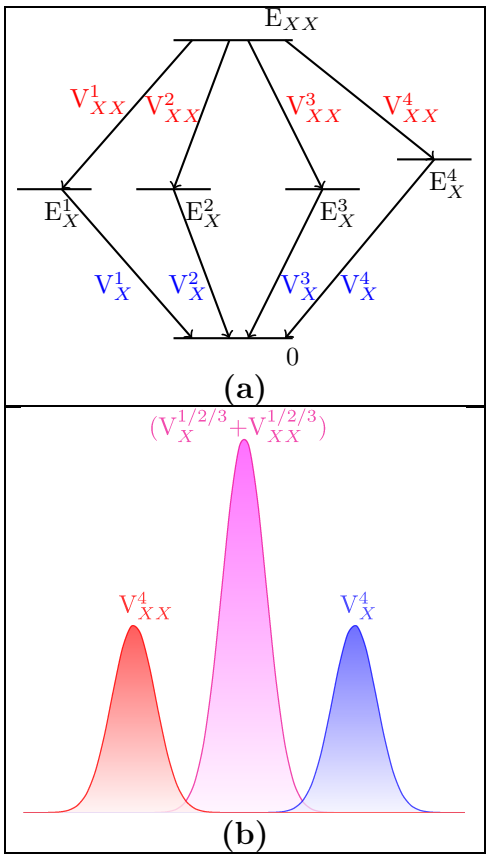


Figure 7: (a) Schematic representation of biexciton-exciton cascade in a *c*-plane (In,Ga)N/GaN QD. (b) Illustration of photoluminescence spectra of such a dot with E_{XX}=$2E^1_X$ ≈$2E^2_X$ ≈$2E^3_X$.

and (E_{XX}=$2E^4_X$), cf. Fig. 7 (a). This reveals already the benefit of a *c*-plane (In,Ga)N/GaN QD system for twin photon emission. Given that the energy separation between E^1_X, E^2_X, and E^3_X is very small and should be difficult to resolve experimentally, these three decay channels are equivalent in energy and polarization properties. Therefore, twin photon emission is possible through these channels with the same probability. A schematic illustration of a resulting PL spectrum, indicating also light polarization characteristics, is given in Fig. 7 (b). Thus, if E_{XX}=$2E^1_X$ ≈$2E^2_X$ ≈$2E^3_X$, one expects with a probability of 75% that the $XX-X$ cascade emits a twin-photon pair. In a C_{2v} symmetric dot, bearing in mind the larger bright-bright fine structure splitting and the presence of dark excitonic states, only a probability of 25% could be reached, since only one decay path is able to emit twin-photon pairs.

To further study twin-photon emission from *c*-plane (In,Ga)N/GaN dots, detailed investigations on tailoring their excitonic and biexcitonic energies are required. In general, this can be achieved by changing the dot geometry, aspect ratio or In content as previous investigations indicated already.[56] Furthermore, the optical properties of the $XX-X$ cascade depend also on the symmetry of the biexcitonic complex.[57] While in previous studies of (In,Ga)N/GaN dots, "conventional" biexcitonic ground states with a net spin of zero ($s=0$) have been observed[51,56] and successfully used to explain experimental results,[51,58] recently in *GaN/AlN* QDs so-called "hybrid" biexcitons, which exhibit a net spin of $s=\pm 3$, have been found and discussed.[57] Thus, when tailoring the excitonic and biexcitonic properties of (In,Ga)N/GaN dots for twin-photon emission, the biexcitonic properties could reveal further new and interesting physics. Finally, by engineering the geometrical aspects of *c*-plane (In,Ga)N/GaN dots, exciton phonon interactions as well as the radiative lifetimes can be modified and investigated.[59,60] Due to the presence of the electrostatic built-in field in these systems and the resulting large exciton-dipole moment, exciton-phonon interactions are pronounced and can lead for instance to linewidth broadening.[59] Also the strong built-in fields lead to radiative lifetimes of the order of nanoseconds.[60] To reduce the exciton dipole moment and the radiative lifetimes, nonpolar (In,Ga)N/GaN dots could be an attractive alternative.[3,60] However, further studies on the exciton fine-structure of these systems are required to investigate their potential for non-classical light emission in general.

Conclusion

In this work we have presented a fully atomistic many body analysis of the potential of *c*-plane (In,Ga)N/GaN QDs for novel non-classical light emission via twin-photon emission. Our atomistic calculations show that wurtzite crystal structure, random alloy fluctuations, small electron-hole exchange interaction

and spin-obit coupling effects lead to an excitonic fine structure that is very different from QDs based on more conventional III-V materials such as (In,Ga)As/GaAs dots. More specifically, we find that the three energetically lowest excitonic states are basically degenerate in energy and bright. Using these three states in the $XX - X$ exciton cascade, in comparison with a zincblende (In,Ga)As/GaAs QD a significantly higher probability for twin-photon emission can be achieved in the here studied *c*-plane (In,Ga)N/GaN dot systems. Furthermore, our calculations reveal large exciton binding energies (>35 meV), which exceed the thermal energy at room temperature. In combination with the large band offsets, the results obtained here for the energetically lowest exciton states indicate that this system should be highly attractive for novel twin photon emission near room temperature and a wide emission wavelength range when changing the In content in the dot.

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