Corner and side localization of electrons in irregular hexagonal semiconductor shells

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Abstract

We discuss the low energy electronic states in hexagonal rings. These states correspond to the transverse modes in core–shell nanowires built of III–V semiconductors which typically have a hexagonal cross section. In the case of symmetric structures the 12 lowest states (including the spin) are localized in the corners, while the next following 12 states are localized mostly on the sides. Depending on the material parameters, in particular the effective mass, the ring diameter and width, the corner and side states may be separated by a considerable energy gap, ranging from few to tens of meV. In a realistic fabrication process geometric asymmetries are unavoidable, and therefore the particles are not symmetrically distributed between all corner and side areas. Possibly, even small deformations may shift the localization of the ground state to one of the sides. The transverse states or the transitions between them may be important in transport or optical experiments. Still, up to date, there are only very few experimental investigations of the localization-dependent properties of core–shell nanowires.

Keywords: core–shell nanowires, polygonal cross section, in-gap states, corner-localized states, side-localized states

(Some figures may appear in colour only in the online journal)

1. Introduction

Core–shell nanowires are radial heterojunctions consisting of a single-material nanowire (core) which is covered with one or more layers of different material (shells). Due to the crystallographic structure the cross section of such wires is usually hexagonal [1–7], but other shapes like triangles [8–14] or dodecagons [15] have already been obtained. These radial heterojunctions have been in the focus of extensive experimental [4–7, 16–21] and theoretical [22–24] studies in the recent years. This is mostly due to the possibility of controlling some of their physical properties, e.g. the band alignment. If the materials are properly adjusted, and also the geometric variables such as core diameter and shell thickness, then one may obtain type II band alignment at the heterojunction because of which the electrons are confined only in the shell volume and form conductive shells [1, 25]. Moreover, the core part may be etched out, and thus hollow systems or prismatic nanotubes of finite thickness may be obtained [2, 3].

Ever since the paper by Ferrari et al [23], it has been known that low-energy electrons confined in thin hexagonal tubes are accumulated along the edges, while the particles excited to higher energies occupy the facets. More generally, the shape of the cross section governs the energy structure of prismatic nanotubes [26–29]. If the cross section is a regular polygon with \(N\) corners, and the thickness of the wall is much smaller than the radius of the tube, the lowest \(2N\) energy states (including spin) are localized in the corners of the polygon. Moreover, the wave functions of the next group of \(2N\) states, on the energy scale, are localized on the sides of the polygon and are separated from the corner states by an energy gap which, depending on the geometry and material parameters, may be comparable or even larger than the room-temperature energy, especially for the triangular case [26, 27].

For regular polygons, both the corner and side states have an internal energy dispersion. Still, it decreases with the width, eventually reducing to a quasidegenerate group of corner states. On the contrary, the corner localization softens...
irregular conductance steps in ballistic transport within the gap singlet pairs of Coulomb coupled electrons with energies even for narrow shells structures can be well approximated with a cylindrical model shape of the wire and the results obtained from hexagonal most of the reported experimental studies do not resolve the predicted and they need the experimental detection. In reality, the absorption of photons from different spectral domains by a large energy gap implies very interesting physics, like the cross section. 

The presence of multiple corner and side states separated by a large energy gap implies very interesting physics, like the absorption of photons from different spectral domains [26, 27], multiple Majorana modes in interaction [30, 31], irregular conductance steps in ballistic transport [29], or spin-singlet pairs of Coulomb coupled electrons with energies within the gap [28, 32]. These mentioned phenomena are only predicted and they need the experimental detection. In reality, most of the reported experimental studies do not resolve the shape of the wire and the results obtained from a cylindrical model even for narrow shells [16, 33–35]. Only in rare situations the anisotropic electron distribution has been detected [4]. As a result, not much attention has been paid to the shape or to the internal structure of the wires and to the effects resulting from the inhomogeneous electron distribution within the cross section. Very recently, two experimental papers have reported complex photoluminescence spectra associated to irregular core–shell nanowires. One of them [36] describes polychromatic emission of InP–InAs–InP multi-shell nanowires with a diameter that increases along the nanowire axis. The other one [37] shows spectra that could be attributed to exciton recombinations on different facets situated on different regions along the length of an irregular GaAs shell embedded in a GaAs–AlGaAs multiple heterojunction.

Motivated by these recent experimental achievements, in this paper we compare the localization patterns of electrons on symmetric and irregular hexagonal rings. We consider several material parameters and show how the separation between corner and side states increases with decreasing the effective mass and how the corner and side localization and their separation evolve when the hexagonal symmetry is broken.

The paper is organized as follows. In the following section we present the model and the calculation method. Then, in section 3 we consider single (section 3.1) and many-body (section 3.2) energy levels and the localization of electrons confined in symmetric quantum rings. Next, in section 4 we study asymmetrically (section 4.1) and symmetrically (section 4.2) deformed structures. Finally, in section 5 we summarize the results.

2. Model

If the shells are so short that the separation between the two lowest longitudinal modes is larger than the energy dispersion of the interesting range of transversal states then the shells may be considered quantum rings. In our case it is enough if the separation exceeds the gap between corner and side states and their energy dispersion.

We model a polygonal quantum ring starting from a circular disk situated in the (x, y) plane. The plane is then discretized on a polar grid [38], on top of which we impose lateral boundary conditions, corresponding to infinite potential barriers, that enclose the hexagonal rings. The points that lie outside the defined boundaries are excluded, figure 1. The flexibility of this method allows us to model asymmetric samples without the necessity to adapt the background grid or redefining the Hamiltonian matrix elements.

The Hilbert space associated with the polar lattice is spanned by the vectors \( |q\rangle = |k, j, h\rangle = |k\rangle \), in which the radial \( (k) \) and the angular \( (j) \) coordinates are included. The single-particle eigenvalues \( E_a \) \( (a = 1, 2, 3, \ldots) \) and the eigenvectors \( \psi_a = \sum_q \psi(q, a) |q\rangle \) in the position representation are obtained through numerical diagonalization [26] of the Hamiltonian in the basis including the spin \( (\sigma = \pm 1) \), i.e. \( |k, j, \sigma\rangle \).
Effective mass $m_{\text{eff}}$, degeneracy pattern 2
that every four consecutive levels due to spin and orbital momentum, or due to spin only, such
ring is spin degenerate, and it is followed by alternating pairs of
The ground state of a single electron con
3.1. Single-particle energy levels and localization
3. Symmetric rings/shells
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The ground state of a single electron confined in a hexagonal
ring is spin degenerate, and it is followed by alternating pairs of four- and twofold degenerate levels. The degeneracy is either
due to spin and orbital momentum, or due to spin only, such
that every four consecutive levels (12 states) repeat the
degeneracy pattern 2–4–4–2. In figure 2(a) one can identify the
corner states as the lower group of states, separated from the
other states by an energy interval of $\Delta = 1.2$ meV. However, the corner peaks spread into the facets, where they overlap.
This effect is the strongest for the ground state, figure 2(b), and
with increasing the energy the maxima sharpen and the prob-
bility of finding an electron in the middle of a side decreases,
figures 2(c) and (d), or even vanishes for the highest level in the corner group, figure 2(e). The second group is built of
12 states associated with probability distributions which form
one maximum on each side. In the case of the lowest level, the
localization pattern consists of only 6 maxima and electrons
with this energy are depleted from corner areas, figure 2(f). For
the higher states another six peaks appear in the vicinity of the
vertices. Although they considerably increase with the energy,
they never reach the height of the side maxima, figures 2(g)–(i).
The number of maxima formed on each side increases by one
between consecutive groups of 12 states, and thus for the two
following groups on the energy scale there are 2 and 3 maxima
on every side, respectively (not shown).

The shape of the probability distributions is governed by the
aspect ratio between the side thickness and the external radius
($d/R_{\text{ext}}$). If it is large, i.e. for thick rings, the corner maxima
penetrate deeply into the sides and strongly overlap there,
figures 3(a) and (b). Such samples do not differ much from
circular ones, since in both cases the electrons may occupy the
whole circumference and easily rotate around the ring. If the
corners are rounded, or destroyed, as in a more realistic situation,
then the corner localization softens [26], and such samples
resemble the circular rings even more. While the corner maxima
sharpen with decreasing the aspect ratio, they still overlap in the
middle of the sides for a wide range of ring widths, figure 3,
except for very thin structures, figures 3(f) and (g).

The energy spacings between adjacent energy levels, and
in particular the gap separating the corner from side states
($\Delta$), strongly depend on the aspect ratio ($d/R_{\text{ext}}$) and the
diameter ($2R_{\text{ext}}$). For the 10 nm thick InP ring with the
external diameter of 120 nm $\Delta$ is equal to 1.2 meV,
figure 2(a). Decreasing the side thickness by 4 nm, while the
diameter is kept constant, results in an increase of over four
times of this gap, figure 4(a). Even though $\Delta$ becomes
thereby the dominant energy gap, which exceeds the disper-
sion of the corner states, it is still much smaller than for a
triangular ring of the same thickness and external radius [26].
The energy spectrum scales with the effective mass ($m_{\text{eff}}$), i.e.
the smaller the effective mass of the material is the larger the
separation of the corner states and their energy dispersion are.
In figure 4 we compare the energy spectra for four rings made of
different semiconducting materials. A substantial fraction of
experiments was performed on core–shell nanowires with the
shell made of InP or GaAs. Both of these materials are
characterized by relatively large values of the effective mass,
table 1, and thus even though for the 6 nm thick ring the gap
between corner and side states is considerably larger than
other energy intervals in the system, it is still below the
resolution of most experiments, figures 4(a) and (b). Still, $\Delta$
increases to 17 meV for InAs, figure 4(c), and for the InSb
ring of the same shape it becomes 28 meV, figure 4(d).

| Table 1. Material parameters used in the numerical simulations. |
|-----------------|---------|---------|---------|---------|
|                  | InP     | GaAs    | InAs    | InSb    |
| Effective mass   | $m_{\text{eff}}$ | 0.08 $m_e$ | 0.067 $m_e$ | 0.023 $m_e$ | 0.014 $m_e$ |
| Relative permittivity | $\epsilon$ | 12.5 | 12.9 | 15.0 | 16.8 |

Note. $m_e$ is the electron mass.

In the absence of an external electromagnetic field the system evolution is governed by the Schrödinger equation

$$\frac{p^2}{2m_{\text{eff}}} \psi_n = E_n \psi_n,$$  (1)

The corresponding Hamiltonian matrix elements are

$$\langle \ell j | \frac{p^2}{2m_{\text{eff}}} | \ell' j' \rangle = T \delta_{\ell, \ell'} \delta_{j, j'} + V \delta_{\ell, \ell'} (\delta_{j, j'} - \delta_{j, j'+1}) + h.c.,$$

where $T = \hbar^2 / (2m_{\text{eff}} R_{\text{ext}}^2)$ is a reference energy with $m_{\text{eff}}$ being the effective mass of the electrons in the ring material (table 1), and $R_{\text{ext}}$ the external radius of the polygonal shell. The factors $T = (R_{\text{ext}}/\delta)^2$ and $V = (R_{\text{ext}}/\delta\phi)^2$. Finally, $\delta r$ and $\delta \phi$ are the distance between neighboring sites with the same angle and the angle difference between adjacent sites with the same radius, respectively.

The next step in the calculations is to use a subset of the single-particle eigenvectors $\psi_n$ as a basis for the many-body problem. The electron–electron interaction within the poly-
gonal ring is considered by taking the Schrödinger equation in the second quantization formalism

$$\hat{H} = \sum_a E_a c_a^\dagger c_a + \frac{1}{2} \sum_{a,b,c,d} V_{abcd} c_a^\dagger e_b^\dagger c_d e_c,$$  (2)

where $c^\dagger$ and $c$ are the well known creation and annihilation operators. The elements of the Coulomb potential are calculated as

$$V_{abcd} = \langle \psi_e \psi_h | \frac{e^2}{4\pi\epsilon \epsilon_0 |\mathbf{r} - \mathbf{r}'|} | \psi_e \psi_h \rangle,$$  (3)

where $\mathbf{r} = (x, y)$ is the position of electrons in the plane and $\epsilon$ represents the relative dielectric permittivity of the shell material, table 1. The Hamiltonian (2) is then diagonalized numerically using a subspace of up to 24 corner and side states with the lowest energy (configuration-interaction method) [32].

3. Symmetric rings/shells
3.1. Single-particle energy levels and localization

The ground state of a single electron confined in a hexagonal
ring is spin degenerate, and it is followed by alternating pairs of four- and twofold degenerate levels. The degeneracy is either
due to spin and orbital momentum, or due to spin only, such
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Figure 2. (a) Energy levels of a single electron confined in a symmetric InP ring for which the side thickness and external radius are equal to 10 and 60 nm, respectively. \( \Delta \) stands for the gap between corner and side states. (b)–(i) The corresponding probability distributions in increasing energy order.

Figure 3. Probability distributions corresponding to the ground state of a single electron confined in symmetric rings restricted by the external radius of 60 nm versus side thickness \( d \).

Figure 4. Energy levels of a single electron confined in symmetric quantum rings of different materials. In all the cases the side thickness and external radius are equal to 6 and 60 nm, respectively. \( \Delta \) stands for the gap between corner and side states.

Figure 5. Energy levels of a pair of electrons confined in InP (a) and InSb (b) quantum rings of 6 nm thickness. The blue diamonds correspond to noninteracting electrons, while the red circles represent Coulomb interacting particles. In the inset to figure (b) we show the degeneracy of the in-gap states.
3.2. Many-body states

The Coulomb repulsion between electrons adds to the energy of the quantum states. A question is how this energy compares to the energy $\Delta$. If $\Delta$ is smaller than the characteristic Coulomb energy, many-body corner and side states will mix up. On the contrary, if $\Delta$ is the largest energy, then pure corner many body states may have energies within this gap [32]. In the examples shown in figure 5, 66 eigenstates of two non-interacting electrons are separated from the higher states by approximately $\Delta$ (blue diamonds). The Coulomb interaction shifts all states to higher energies and rearranges them according to the particle distribution within the ring. In particular, the lowest states correspond to particles localized around the opposite or alternating corners, respectively. The 24 states representing electrons occupying neighboring corners form a quasidegenerate level split from the lower corner states by a small gap, resulting from the decreased spatial separation of the particles. The number of separated corner states of the interacting system is reduced by 6 with respect to the noninteracting case. The remaining states correspond to pairs of electrons in a spin singlet configuration accumulated in the same corner area for which the contribution due to the electrostatic repulsion exceeds $\Delta$, and thus these states mix with the states above the gap (red circles in figure 5(a)). The splitting between single-particle corner and side states increases for materials with smaller effective mass. Moreover, for the set of studied materials the relative permittivity increases (table 1), and thus the contributions due to Coulomb interaction decrease. Consequently, for the InSb ring the gap $\Delta$ is larger than the Coulomb-induced shift of the pairs of close by electrons. As a result, the corresponding six states stay below the states associated with mixed corner-side probability distributions, i.e. in the gap, figure 5(b). Such in-gap states were previously obtained for triangular rings where the gap $\Delta$ is much larger than in the hexagonal case [32]. In principle, states of this type should also appear for ultra thin InP and GaAs hexagonal shells, but such rings are beyond our computational limitations.

4. Deformed rings

4.1. Side thickness and corner deformations

In spite of high-precision manufacturing technologies it is still impossible to obtain perfectly symmetric wires, and it is even more difficult to cover the wires with shells of constant thickness. Core-shell wires are grown in sets of close-by vertical cores which are later covered with layers of a different material. Due to the screening of neighboring wires, the shell thickness varies along the cross section circumference [36, 37]. In general, the corner localization is very sensitive to the ring symmetry and to the size of the corner area [27]. Ballester et al [39] analyzed a hexagonal ring with one thicker side and showed that the lowest single-particle states are localized on that side. Starting with a symmetric hexagon and increasing the thickness of one side, the probability distribution corresponding to the ground state forms initially two maxima at the ends of the thinnest side, which further merge to form one maximum along the whole facet. The number of states localized on the widest side depends on the ratio between the thickness of this side and the width of the others. The following group on the energy scale consists of four corner states, associated with probability distributions forming four peaks localized around the corner areas of the same size [39].

We consider the opposite case, i.e. the situation when one of the sides is thinner than the other ones. Here the electrons are depleted from the thinner side and the structure acts as a system with five facets. The low-energy states are distributed between the four larger corner areas, while the electrons excited to the higher levels are delocalized over the five sides. Obviously, the probability distributions corresponding to both types of states do not reproduce the symmetry of the sample, but only the mirror symmetry with respect to the thinnest side, figures 6(b)–(d). As for square rings [26], there are eight corner-localized states, spread within a narrow energy range and separated from the ten higher side-localized states, i.e. the corner states are still protected by a considerable energy gap, but slightly smaller than in the case of a square ring of the same diameter and width, figure 6(a).

During the fabrication process, the coverage of the core with the shell material may be nonuniform, such that some regions of the shell may be very thin or even absent [13, 40]. We illustrate such examples in figure 7. In such cases, the lowest energy levels are also associated with corner-localized probability distributions. The corner localization is not surprising since such structures resemble multibent wires and it had already been shown that in the bent parts of such structures effective quantum wells are formed [41]. Interestingly, the separation of the corner states from the side ones initially,

![Figure 6. Single-particle energies for a ring with one thinner side (side $d'$ in figure (b)) (a), and the examples of the corresponding probability distributions (b)–(d).](image-url)
i.e. when one of the corners is excluded, drops down and later increases and fluctuates with the subsequent reduction of the number of corners, figures 8(a)–(c). More importantly, if the shell is made of InSb material, then the in-gap states are obtained down to the two-corner system, figures 8(d)–(f).

### 4.2. Angular deformations

Next, we consider hexagonal rings with angular deformations. This class includes star shaped polygons which have been recently obtained [42]. We assume that three alternating external radii \((R_1)\) do not change, while the other three are reduced \((R_2)\), figure 9. As a result, the system is now threefold rotationally symmetric, i.e. it is built of three identical elements. The two different angles split the corner domain into two groups of corner states [43], separated by a gap \((\delta)\), which is comparable to the separation of the corner states from side states \((\Delta)\) for small differences between the two radii, figure 9(a). The probability distributions corresponding to the lowest states are localized around the sharpest corners, figure 9(b), while those associated with the second group of corner states form maxima centered around the bent parts, figure 9(c). These, relatively small, differences of the corner areas do not affect the side localization, which remains symmetric, figure 9(d). Decreasing the ratio \(R_2/R_1\) results in the sharpening of three corner areas and softening of the alternating ones. This considerably increases the separation between the corner states \((\delta)\) such that it becomes the dominant gap of the spectrum, while the separation of the corner from side states slightly decreases, figure 9(e). The lowest states are strongly localized in the vicinity of the sharpest corners, figure 9(f), while the probability distributions associated with the second group of corner states form elongated maxima centered around the bent parts, figure 9(g). In this case the side-localized maxima are shifted towards the softer corners, figure 9(h). The decrease of the ratio \(R_2/R_1\) results in obtaining triangular quantum rings which were analyzed elsewhere [26–28, 32], and star-like structures. For the latter ones the gap separating the corner from side states \((\Delta)\) slightly increases, but the lower gap \((\delta)\) exceeds it by nearly one order of magnitude, figure 9(i). The ratio of external radii \((R_2/R_1)\) which is lower than that of a triangle \((0.5)\) induces the formation of six quasidegenerate corner-states. These states are well-separated from the higher, formally of corner-type, states but spatially elongated similarly to the side states of triangular rings. Further decrease of the ratio \(R_2/R_1\) leads to the reduction of the wider corner areas, and thus sharpening of the corresponding localization peaks, figure 9(o). This results in the increase of the separation of this states from the
side-localized states, figure 9(m), but does not affect much the localization of the ground state and the side states, figures 9(n) and (p), respectively. Similarly to triangular rings \[32\] the star-like shells allow to obtain three in-gap states in the largest gap and on top of that, they enable the formation of well-separated levels in the second gap of the many-body spectrum of a pair of Coulomb coupled carriers, figure 10.

5. Conclusions

We studied the energy levels and localization of single and two Coulomb interacting electrons confined in symmetric and nonsymmetric hexagonal quantum rings made of commonly used semiconductor materials: InP, GaAs, InAs, and InSb. The results that we showed are relevant not only for quantum rings, but also for the transverse modes in long nanowires of core–shell type.

The wave functions of the low energy states are distributed either in the corner areas or on the sides of the hexagonal shell. The separation of the corner and side states, both in terms of localization and in energy, depends on the

Figure 9. Single-particle energies for InSb rings with different ratios of the neighboring external radii \(R_2/R_1\) (first column), and the probability distributions corresponding to the ground state (second column), the second corner level (third column), and the lowest side level (fourth column).

Figure 10. Energy levels for a pair of electrons confined in an InSb ring for which \(R_2/R_1 = 0.2\).
geometry parameters and on the material, via the effective mass. In particular, with InSb for which the effective mass is the smallest, the single-particle corner states may be separated from the side-localized states by a gap of 28 meV, i.e. comparable to the room-temperature energy, for a ring of 60 nm radius and 6 nm thickness. In this case, the many-body energy spectrum resembles the one of triangular rings, i.e. the lowest states are spread within a small energy range and are followed by in-gap states.

The separation of the in-gap states from the lower corner states, and also from the higher corner-side states, may considerably exceed the energy dispersion of the lowest group of corner states. External electric and magnetic fields may easily change the arrangement of the latter states, and thus the spin configuration of the ground state, without altering much the in-gap states. Since all in-gap states are of the spin-singlet type, in the absence of spin-orbit interaction they may be optically excited only from a state of the same spin component. This allows to block the excitation of these states in the presence of an external field which changes the ground state, i.e. as for the triangular rings, such energy structure allows for the contactless field which changes the ground state, i.e. as for the

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