CONTRIBUTIONS to SCIENCE, **3** (4): 447–457 (2007)
Institut d'Estudis Catalans, Barcelona
DOI: 10.2436/20.7010.01.21 ISSN: 1575-6343 **www.cat-science.cat**

Nanoscopic semiconductor quantum rings

J.I. Climente[1, 2] and J. Planelles[2*]

1. National Research Council-Institute of Matter Physics (CNR-INFM S3), Modena, Itàlia
2. Departament de Química Física i Analítica, Universitat Jaume I, Castelló de la Plana

Resum

Els anells quàntics semiconductors són una classe particular de punts quàntics de forma toroïdal. De la mateixa manera que en punts quàntics, el nombre de portadors de càrrega confinats en aquestes estructures pot controlar-se un a un, i és factible un ampli control de les propietats electròniques i òptiques, motius que fan dels anells quàntics candidats prometedors per a la possible futura construcció d'aparells nanoelectrònics. L'estructura electrònica i la resposta magnètica dels anells quàntics és però molt diferent de la dels punts quàntics, a causa de la seva topologia doblement connectada. Al llarg de la darrera dècada s'ha fet un gran esforç per entendre les seves característiques fonamentals, sobretot en comparació amb les dels ja ben coneguts punts quàntics. En aquesta contribució, presentem una revisió del progrés realitzat cap a l'enteniment de les propietats dels anells autoordenats nanoscòpics a través dels nostres treballs, sempre dins el context de l'activitat experimental relacionada. Estudiem nombroses propietats d'aquestes estructures, des de la seva estructura energètica fins a l'espectroscòpia òptica, arribant a la formació de «molècules d'anells quàntics».

Paraules clau: anells quàntics, punts quàntics, efecte Aharonov-Bohm

Abstract

Semiconductor quantum rings are a particular type of toroidal shaped quantum dot-like structures. As in quantum dots, the number of carriers confined within these structures can be controlled one by one, and to a large extent their electronic and optical properties can be tailored, which makes them highly promising candidates for the eventual construction of nanoelectronic devices. However, the electronic structure and magnetic response of quantum rings is very different from that of quantum dots, owing to their doubly-connected topology. Over the last decade, much effort has been put into understanding the fundamental features of these structures, and comparing them to the well-known case of quantum dots. In this report, we present an overview of the progress in the understanding of self-assembled nanorings through our theoretical research related to experimental evidences. We have studied several properties of quantum rings, from their energy structure, to optical spectroscopy and the formation of "quantum ring molecules".

Keywords: quantum rings, quantum dots, Aharonov-Bohm effect

Quantum rings (QRs) are the simplest multiply connected nanostructures with three-dimensional confinement (see Figure 1). They have received much attention from researchers in the field of condensed matter over the last two decades [1-5], mostly due to their particular magnetic response. When a QR charged with an excess electron is pierced by a magnetic field perpendicular to the ring plane, the Aharonov-Bohm (AB) [6, 7] effect gives rise to persistent currents and oscillations of the electron ground state energy. The AB effect is a pure quantum effect, theoretically predicted in 1959 [6] and experimentally confirmed in mesoscopic QRs many years later [8, 9]. However, mesoscopic QRs are not the ideal playground for investigating the Aharonov-Bohm effect, as they are subject to heavy scattering coming from electron-electron, electron-impurity and electron-phonon interactions. All these phenomena tend to reduce the phase coherence of carriers confined in mesoscopic QRs. Thus, recent success in synthesizing QRs in the

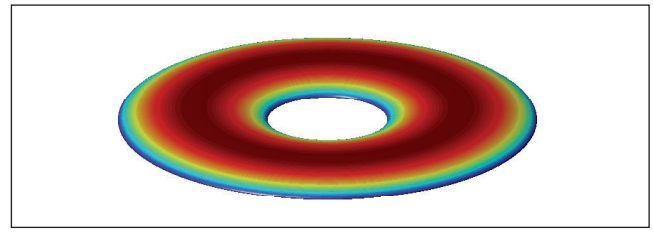

Figure 1. Schematic of a quantum ring.

* Author for correspondence: Josep Planelles. Departament de Química Física i Analítica. ESTCE. Campus Riu Sec. Universitat Jaume I. E-12071 Castelló de la Plana, EU. Tel. 34 964728090. Fax: 34 964728066. Email: josep.planelles@qfa.uji.es

nanoscopic regime (i.e. in the true quantum limit, with a small number of confined carriers and weak scattering) has been acknowledged as being a major achievement in the study of low-dimensional structures [1].

Nanoscopic QRs can be fabricated by means of self-assembly [10, 11, 12] or lithographic [13] techniques. While lithographic techniques provide better control of the QR dimensions, self-assembly offers the important advantage of forming samples of major optical quality, which renders them particularly attractive for practical applications. In this report, we present an overview of the progress in the understanding of nanoring properties through our theoretical research related to experimental evidences. We shall focus on the physics of self-assembled QRs, even though many of the features we will describe are valid for general nanoscopic rings. The report is organized as follows. Sections 1-5 deal with the study of single QRs. In Section 1 we deal with the energy structure of single, isolated QRs as revealed by optical spectroscopy. The following two sections deal with AB related effects. We will show that magnetization can be employed to determine the topology of buried nanostructures in a highly efficient manner, and we shall explore the possibility of electron-hole pairs confined in QRs having AB effects. In Section 4, we investigate the response of QR under tilted magnetic fields. Since AB effects are related to the magnetic flux penetrating the QR inner hole, most studies of the magnetic response of QRs carried out to date have focused on the axial field configuration. However, here we will show that in-plane and tilted magnetic fields yield complementary information about QR properties. In Section 5 we discuss a controversy found in the literature with regard to the exact shape of buried self-assembled QRs, and provide an answer based on the most recent experiments. Section 6 deals with the study of interacting QRs (also called quantum ring molecules). We show that the optical spectra of vertical, concentric and laterally coupled QRs undergo major changes as a function of the inter-ring distance. This affects the magnetic response of the system, in such a way that the optical and magnetic responses yield valuable information about the quantum ring molecule coupling regime. The paper ends with a brief section outlining our conclusions and future perspectives.

1. Optical spectroscopy

Self-assembled QRs were first grown by J. Garcia et al. following a similar procedure to that used in the synthesis of InGaAs self-assembled quantum dots (QDs) embedded in GaAs. [10] Atomic force microscopy (AFM) of these structures clearly revealed a ring-like morphology, with an inner radius of 10-15 nm, outer radius of about 60 nm, and a height of between 4 and 5 nm. [10, 14] However, self-assembled QRs need to be completely embedded in the semiconductor matrix to become operational. This meant that the observed QRs had to be buried with GaAs, and it was left unclear as to whether or not the ring morphology was preserved during the high-temperature capping. In order to find an answer, many spectroscopic experiments attempted to trace evidence of ring-like behavior in the buried samples. [15, 16, 17, 18, 19] These experiments revealed a different behavior from that of usual QDs, but theoretical confirmation that the observed response could actually be ascribed to QRs came afterwards, as we discuss below.

The key property to spectroscopically discriminate QDs and QRs is their response to magnetic fields. As shown in Figure 2A, the electron energy levels in a usual (lens-shaped) self-assembled QD display a characteristic evolution, where the levels forming the lowest Landau level do not cross each other. On the contrary, as shown in Fig. 2B, in self-assembled QRs the ground state displays quasi-periodic crossings vs. an increasing field, which is a manifestation of the AB effect. This striking difference inspired Lorke et al to measure the far-infrared magneto-absorption spectrum of self-assembled QRs charged with one and two electrons [15, 16]. They measured the position of some resonances versus the magnetic field, and showed that they could be ascribed to excitations of a quantum ring-like energy spectrum. However, the agreement between the experiment and their theoretical model was not conclusive: the model (which was two-dimensional, with parabolic in-plane confinement) contained a few fitting parameters, and yet it could neither explain all the experimental resonances, nor the relative intensities. Puente and Serra [17] improved the agreement by introducing anharmonic corrections to the parabolic confinement describing the inner edge of the ring, and as-

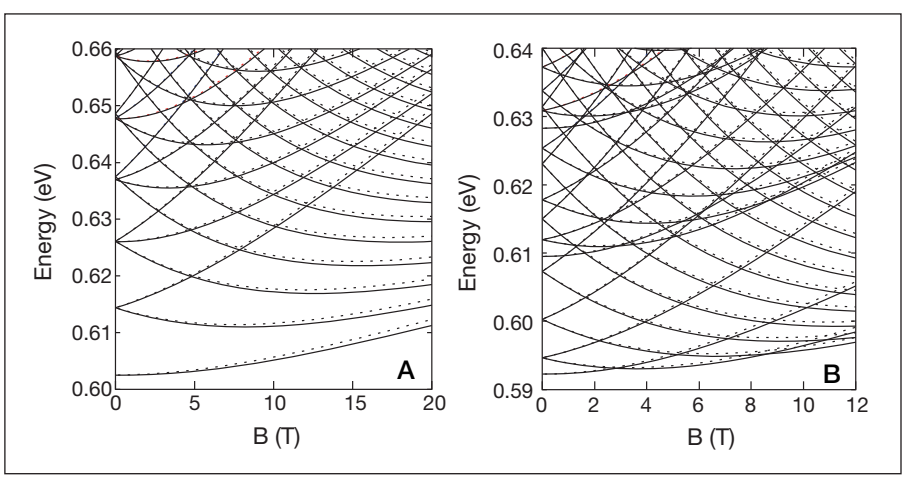

Figure 2. Single electron energy levels vs. magnetic field in: (A) InAs/GaAs QD with height 4.5 nm and radius 60 nm; (B) QR with identical composition and external dimensions, plus an inner cavity radius of 5 nm. Dotted and solid lines represent the up and down spin Zeeman sublevels. While the QD ground state does not undergo level crossings with the field, the QR displays many crossings.

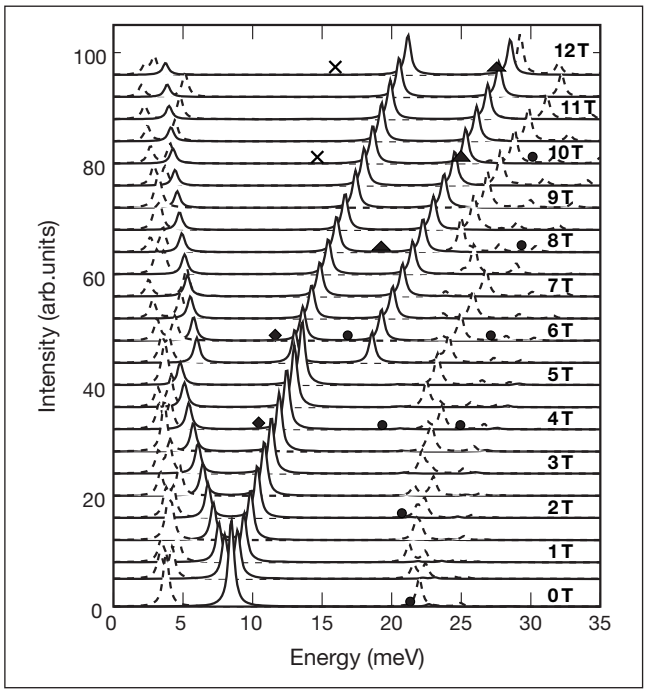

Figure 3. Far-infrared magneto-absorption spectrum of two electrons in two types of QRs, represented by solid and dashed lines. The spectra are calculated for T=0 K. The symbols indicate the energy of the experimental resonances. Reprinted with permission from [20].

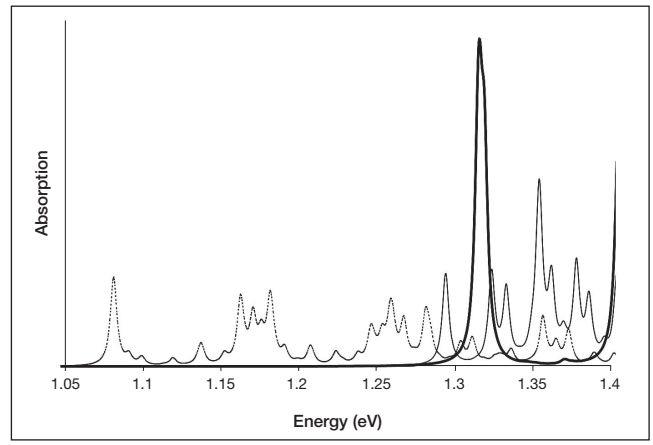

Figure 4. Near-infrared absorption spectrum of one electron at zero magnetic field. The dashed line corresponds to a usual self-assembled QD, the thick solid line to a usual self-assembled QR, and the thin solid line to a QD with similar dimensions to the ring. Note the characteristically strong absorption peak of QRs as compared to QDs. Reprinted with permission from [21].

suming that not one, but two different kinds of QR were present in the sample. The latter assumption was actually supported by related magneto-photoluminescence experiments [19]. However, a few resonances observed in the experiment could still not be interpreted. Definitive evidence that the complete set of resonances could be assigned to magneto-excitations of QRs was given by our group (Ref. [20]) using a three-dimensional, fitting-free model, with a realistic description of the confinement potential, electron-electron interactions and strain fields. This is shown in Fig. 3 for the case of two-electron QRs. Solid and dashed lines represent the absorption spectra from two different kinds of QRs, which add up to give similar resonance positions and intensities as compared to the experiment (symbols in the figure). It was also shown that similar agreement was not possible if the sample was composed of QDs rather than QRs [20].

Further experiments investigating the energy structure of QRs were carried out by Petterson et al. [18]. They measured the near-infrared absorption of self-assembled QRs (i.e., absorption due to electron-hole recombination) at zero magnetic field, and compared this with that of the self-assembled QDs from which the rings had originated. The spectrum of the two kinds of samples exhibited clear differences. Namely, the QR signals showed up at larger energies than those of QDs, and they were of larger intensities. The authors of the experiment argued that these differences might be due to the stronger vertical confinement of QRs observed with AFM [14, 15], which would lead to higher energy gaps, as well as to the weaker lateral confinement, which would lead to stronger Coulomb attraction between the electron-hole pair, and hence to increased oscillator strength. However, conclusive evidence was not given for this either, since the same features could be expected if the QRs had evolved into ordinary QD with strong vertical and weak lateral confinement during the capping process. Numerical calculations simulating Petterson's experiment were presented by our group in Ref. [21], where it was shown that good agreement with the experiment could be obtained by taking into account the different spatial confinements of QDs and QRs (see Fig. 4). Notably, it was also shown that a QD of the same dimensions as the QR could not yield fundamental transitions having the strength observed in the experiment (thin solid lines in Fig. 4), since this feature arises from the particular energy structure of the QRs conduction band, with closely-packed azimuthal energy levels. This further supported the belief that QRs preserve their topology after capping.

Magneto-photoluminescence measurements of self-assembled QRs were reported by Haft and co-workers [19], and again the abovementioned theoretical model gave good agreement under the assumption of toroidal geometry [21].

2. Magnetization

An intrinsic problem of optical magneto-absorption experiments with self-assembled QRs is the low resolution with current technology. [15, 16] This makes a complementary, efficient technique to determine the (simply or doubly-connected) topology of buried nanostructures highly desirable.

Magnetization measurements have proved useful in the study of the electronic structure of QDs, where they outperform other techniques such as far-infrared absorption or transport experiments. [22, 23, 24] As a matter of fact, the so-called Kohn theorem states that electron-electron interaction effects in QDs with parabolic-like confinement cannot be observed through dipolar transitions. [25] In comparison, magnetization studies have successfully revealed some of these effects, such as the spin-singlet-spin-triplet transition of the two-electron QD ground state. [26] In this context, the results obtained by our

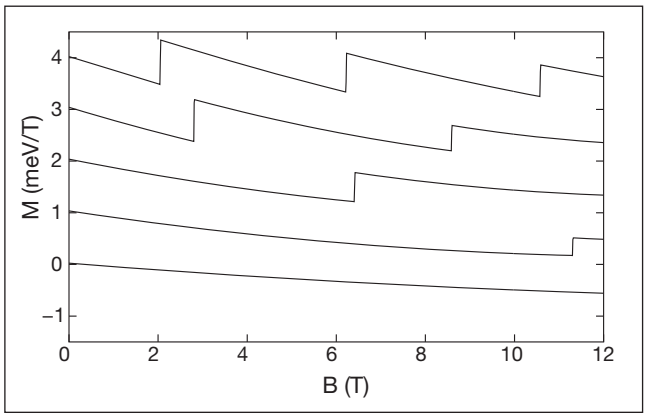

Figure 5. Magnetization of a single-electron in self-assembled quantum rings with increasing inner radius R. From bottom to top, R=0 (QD limit), 1, 2, 5 and 10 nm. Clearly, the introduction of a very small inner cavity brings about magnetization steps at relatively weak fields. Reprinted with permission from [27].

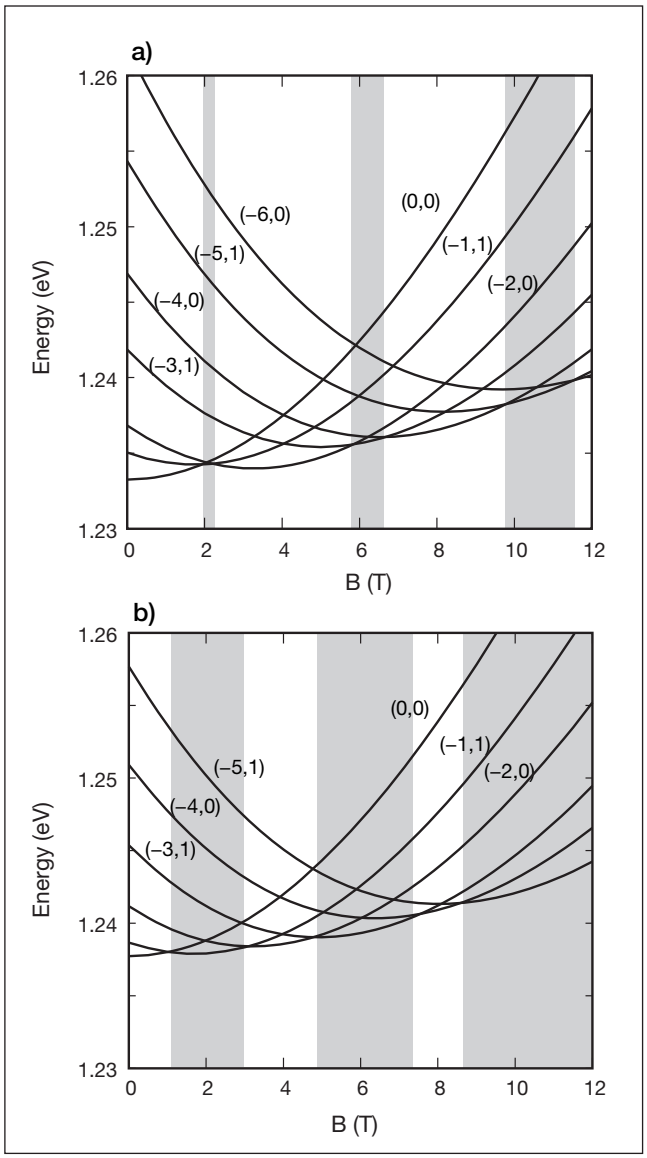

Figure 6. Energy levels vs. magnetic field of two non-interacting (a), and interacting (b) electrons in a self-assembled QR. Only levels that become the ground state are shown for clarity. Grey-shaded regions indicate windows where the ground state is a triplet. The total angular momentum and total spin quantum numbers (M, S) are given for each level. Reprinted with permission from [30].

group in Ref. [27] showed that magnetization is also an excellent tool for probing the energy structure of QRs.

The underlying idea is that the single-electron ground state of QDs does not exhibit any crossings as we increase magnetic field, while that of QRs does, due to the AB effect (recall Fig. 2). These crossings should induce peaks in the magnetization, in such a way that a saw-teeth magnetization spectrum can be expected for the ring [28], as opposed to the smooth response of the QD. Fig. 5 shows the magnetization calculated for a lens-shaped nanostructure with a constant outer radius and vertical confinement, and an inner cavity of increasing radius R (i.e., for an inner radius R=0, the structure corresponds to a QD, for R>0 to QRs with an ever-larger cavity). While for the QD the magnetization changes smoothly, a very small inner cavity (R=1 nm) already induces a clear step at magnetic fields attainable in the laboratory, and with cavities of the size seen in AFM experiments of self-assembled QRs [15] (R=10 nm), several steps are clearly visible. Hence, magnetization appears to be a highly sensitive magnitude for unambiguously identifying the topology of zero-dimensional nanostructures.

So far the energy spectrum of QRs has been described within a single-particle picture. However, QRs (like QDs) are interesting systems for investigating electron-electron interaction on a fundamental level. As a particular result of the toroidal confinement, it has been theoretically predicted that Coulomb interaction in QRs lifts degeneracies of the energy spectrum in such a way that the AB oscillation periods calculated at an independent-particle level are divided by *N*, where *N* is the number of electrons confined in the ring. For example, the single-particle periods are halved for *N=2* electrons, they narrow down to one third for *N=3*, etc. This is the so-called fractional Aharonov-Bohm effect. [29] In fact, the fractional AB effect was originally proposed for a two-dimensional (flat) QR, neglecting the spin Zeeman splitting. Our research (Ref. [30]) has shown that the situation in real self-assembled rings is far more complex than this ideal picture. If one computes the energy levels of a three dimensional, two-electron QR, accounting for strain fields and spin Zeeman splitting, the spectrum obtained resembles that of Fig. 6 for the case where Coulomb interaction is neglected (top panel) or included (bottom panel).

When Coulomb interaction is neglected, the ground state is mostly a singlet, with narrow magnetic field windows where it becomes a triplet due to spin Zeeman splitting (shaded areas). When Coulomb interaction is switched on, the triplet windows become significantly wider, as high-spin states are energetically favored by exchange interaction. Indeed, for large magnetic fields the triplet windows are even wider than the singlet ones, which constitutes a visible deviation from the ideal system. [29]. In Ref. [27] we proposed magnetization experiments as an optimal tool for estimating the accuracy of the fractional AB effect in self-assembled QRs, e.g. by comparing the *B* values at which the magnetization steps of one and two electrons appear.

We conclude this section by pointing out that magnetization experiments to measure the AB effect in self-assembled QRs have been very recently carried out by Kleemans et al. [31]

3. Optical Aharonov-Bohm effect

While charged carriers confined in QRs exhibit experimentally observable energy oscillations with the magnetic field (AB oscillations), neutral carriers such as excitons do not seem to do so. [13] The reason for this is the absence of a net charge (and hence of the vector potential term in the Hamiltonian). [32, 33]

Recently, it has been claimed that the AB effect in QRs may even be detected through the magneto-photoluminescence (PL) spectrum for neutral excitons. [34, 35, 36] The reason for this is that excitons, despite their overall neutral charge, are polarized entities. When a magnetic field perpendicular to the ring plane is applied, periodic oscillations of both electron and hole levels take place (see e.g. Fig. 2B for electrons). Since electrons and holes have different effective mass and average charge density radius (they are polarized), they will oscillate at different rates. Consequently, for certain magnetic field windows it may be that the exciton angular momentum (i.e., the sum of the electron and hole angular momenta) is different from zero. If this occurs, the exciton does not fulfil the selection rule for electron-hole recombination and the PL intensity should be strongly suppressed. [34, 35, 36] This is the so-called "optical" Aharonov-Bohm effect. Actually, real QRs are not exactly circular [10, 11, 12, 37], so the angular momentum is at best an approximate quantum number and PL intensity may not really drop abruptly. Yet, visible fluctuations are to be expected. This is in contrast to existing experiments, which up to 9 T showed no trace of the optical AB effect in self-assembled QRs. [19]

In order to understand this experimental result, our group investigated the energy levels of electron-hole pairs in a typical InGaAs self-assembled QR in Ref. [21]. It is worth mentioning that the models which had been used to introduce the optical AB effect were not suitable for realistic modeling, as they lacked some features that are important for determining the oscillation periods of electrons and holes, such as the QR thickness [32, 33] and the heavy-hole light-hole mixing of the valence band. Using a theoretical model which accounts for these features, we obtained (Ref. [21]) the energy spectrum plotted in Fig. 7. In the figure, the symmetry of the states is given by the label (*m, Fz*), where *m* is the electron azimuthal angular momentum and *Fz* the *z*-projection of the hole total angular momentum. The electron-hole recombination selection rule is fulfilled when *m=Fz-Jz*, where *Jz=3/2, 1/2, –1/2, –3/2* are the possible z-projections of the hole Bloch angular momentum. This condition is met for any magnetic field $B<9$ T, which is in agreement with the fact that no PL intensity suppression was observed in Ref. [19] experiments. However, for larger fields Fig. 6 reveals a magnetic field window where the selection rule is violated, which should lead to the manifestation of the AB effect (shaded area). Certainly, the window is rather narrow, and the prediction can only be considered qualitative, because spin Zeeman splitting and electron-hole Coulomb interaction were neglected. However, the same theoretical model gave good agreement with other experiments involving self-assembled QRs, and one can conclude from Fig. 7 that the optical AB effect *may* be found for magnetic fields $B>9$ T.

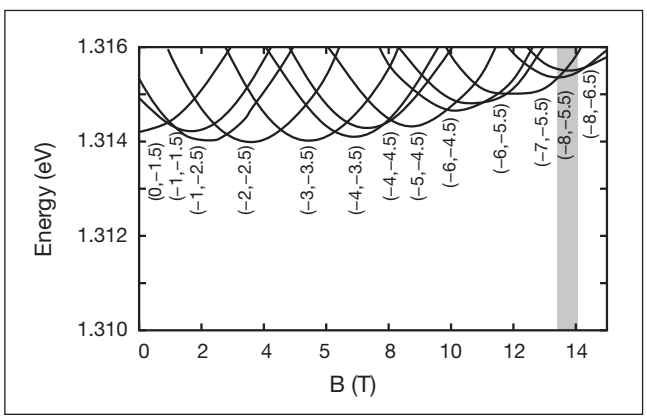

Figure 7. Energy levels of the lowest electron-hole pairs vs. magnetic field in a self-assembled QR. Only levels that become the ground state are shown for clarity. The states are labeled by (*m, F_z*). The magnetic field window for which the ground state is dark is grey shaded. Reprinted with permission from [21].

4. Tilted magnetic fields

The magnetic response of QRs is usually studied for fields perpendicular to the ring plane, because this configuration preserves the axial symmetry of the system and gives the largest magnetic flux trapping, which is desirable for observing AB effects. However, some recent experiments have started using in-plane magnetic fields as a useful way of elucidating the ground state spin of QRs. [38] In fact, the method used in the experiment was based on the theory of QDs subject to in-plane magnetic fields, which is well-known. [39, 40] Whether the response of QRs to in-plane fields was similar to that of QDs, and what the effect of tilting the field in QRs is, were questions only later addressed by our group in Ref. [41]. The main result of that work is summarized in Fig. 8, where a comparison of the single-electron energy spectrum of a nanoscopic GaAs/AlGaAs QR (upper panels) and that of a QD of the same external dimensions, but with no inner cavity (lower panels), is carried out. In Fig. 8, θ represents the angle of the applied magnetic field with respect to the vertical axis (e.g., θ=0º(90º) is perpendicular (parallel) to the ring plane). Clearly, QRs and QDs display very different magnetic responses. For perpendicular *B*, the different behavior is due to the AB effect in QRs, as discussed above for Fig. 2. For in-plane *B* (applied, say, along the *x*-axis), despite the absence of any magnetic flux trap and hence of AB effects, QRs and QDs continue to exhibit different behaviors. In particular, it can be seen that at large fields the QR energy levels arrange into pairs. This fundamental difference is due to the inner hole of the QR which, together with the magnetic field-induced wave function compression in the *y*-direction, leads to the formation of quasi-degenerate double quantum well solutions along *x*. For $\theta = 45º$, an intermediate behavior between the in-plane and perpendicular field cases is found: the spectrum roughly resembles that of $\theta = 0º$, but the AB periods are now larger (owing to the reduced magnetic field trapping) and the formation of double quantum well solutions start to become visible for the lowest-lying energy levels at high fields.

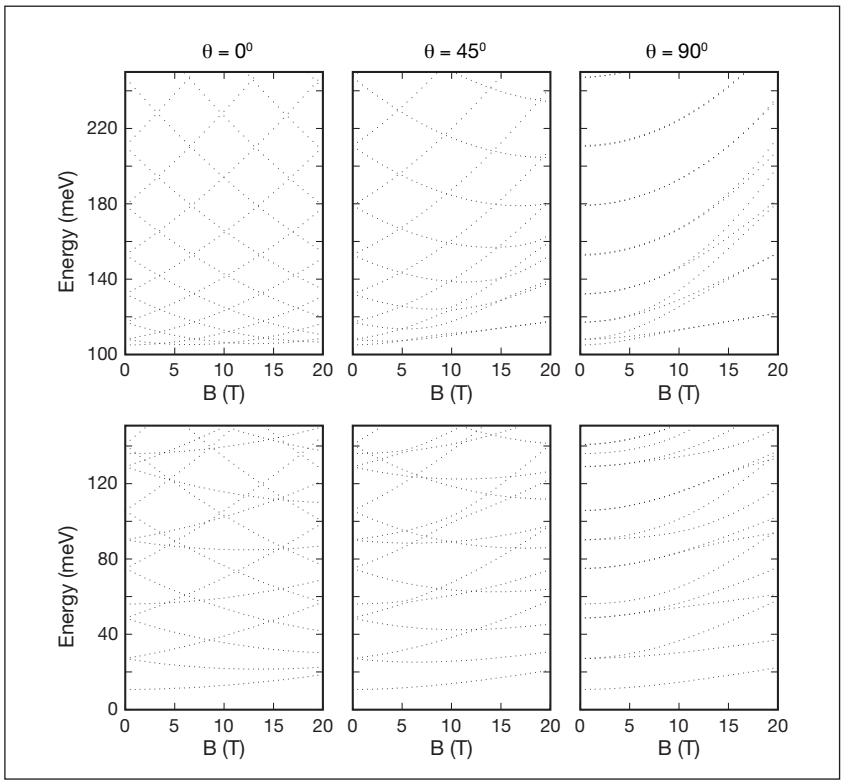

Figure 8. Energy levels in a QR (upper panels) and a QD (lower panels) vs. magnetic fields tilted by an angle θ with respect to the axial direction of the structure. The magnetic response of the QR and the QD is clearly visible not only for axial but also for in-plane fields. Reprinted with permission from [41].

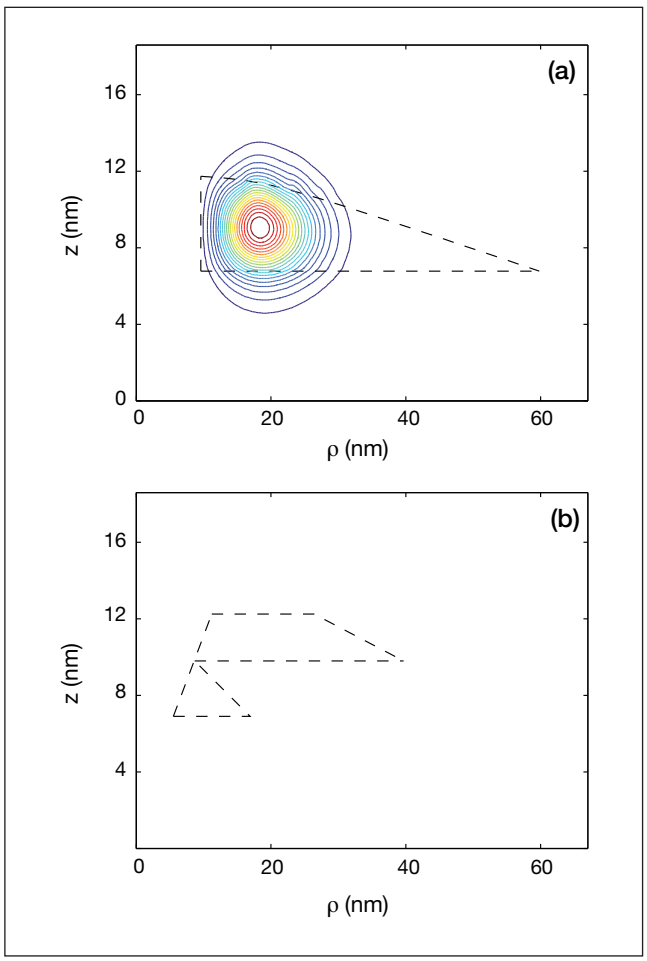

Figure 9. Confinement potential cross-section in the (ρ, z) plane of our QR (a), and of that used in Ref. [58] (b). In the upper panel, the contour represents the ground state charge density, which mostly localizes near the inner wall of the ring, leaving the external part of the ring almost empty due to the strong vertical confinement potential.

5. The shape of self-assembled quantum rings

The difficulty of imaging buried QRs has turned the determination of the geometry of self-assembled QRs into a matter of controversy. As a matter of fact, a recent theoretical work [58], which reports a quantitative reproduction of the capacitance spectroscopy experiments in Ref. [15], has claimed that the ideal QR picture shown in Fig. 1 is oversimplified, and one has to consider the more complex geometry resulting from surface tension considerations proposed by Blossey and Lorke. [59] In our works, however, we have obtained good agreement with many experiments by modeling the QRs as volcano structures, as shown by the dashed lines of Fig. 9A, with a vertical inner wall and a spherical casket describing the outer radius. This is different from the geometry used in Refs. [58], Fig. 9B. In both cases, the height and radius of the QRs have been taken in accordance with AFM measurements in Ref. [14], but in Ref. [58] the total height (4-5 nm) was distributed between the top (wider) part of the ring and the bottom (narrower) one.

The question of which geometry is more realistic can now be addressed thanks to combined AFM-tunneling electron microscopy experiments, which have revealed the actual shape of capped self-assembled QRs. [37, 43] It turns out that a double QR structure does indeed exist, but the bottom ring is richer in In than the top one so that, in principle, most of the charge density localizes within it, leaving the top ring somewhat unoccupied. This makes the ideal single QR picture a good approximation. Moreover, the average height of the bottom ring is about 3.6 nm, which is close to the height considered in our papers [20, 21, 27, 30, 46]. Finally, the radius of the bottom ring turns out to be significantly smaller than that observed in AFM (which corresponds to the upper ring). However, we be-

lieve our model remains valid because the low-lying energy levels localize mostly on the inner side of the QR (see charge density contour in Fig. 9A), where vertical confinement is weaker. We then conclude that the geometry employed in our works is in reasonable agreement with the expectations derived from the most recent experiments.

6. Quantum ring molecules

QDs and QRs are often called 'artificial atoms', because carriers confined within them give rise to a discrete energy spectrum and shell structure in a similar way to atoms. [1] Drawing a similar parallel, when two QDs or QRs are placed close together, the charge density may tunnel between the two structures, such that we can describe 'diatomic artificial molecules' and molecular-like states. QD molecules have been studied extensively in recent years, motivated not only by fundamental physics but also by promising proposals for using coupled QDs as universal, scalable quantum gates for quantum computation. [42] The difficulty of synthesizing high-quality self-assembled QRs has delayed the appearance of the first studies of QR molecules. Only very recently have experiments demonstrated the controlled fabrication of QR molecules. In particular, two kinds of molecules have been synthesized, namely vertically [37, 43] and concentrically [12] coupled QRs.

Vertically coupled quantum rings (VCQRs) are formed by stacking layers of self-assembled QRs on top of each other. The strain fields favor the nucleation of upper rings right on top of bottom rings, so that the circular symmetry of the QR system is preserved. [43] As a primary approach to investigating the electronic structure of VCQRs, Malet et al. calculated the addition energy spectrum of two vertically coupled rings, [44] using a model based on the local density spin approximation that had given excellent agreement with experiments in the study of coupled QDs. [45] The addition energy is defined as:

$\varnothing_2$ = (E[N+1] – E[N]) – (E[N] – E[N-1]) , where E[N] is the total energy of the N-electron ground state. It represents the amount of energy one has to provide in order to add one electron to the QR, as compared to the energy required to add the last electron (chemical potential differences). The addition energy spectrum of symmetric QDs or QRs displays a regular pattern which reveals the shell structure of the low-dimensional system, with local maxima at filled and half-filled shells. [45] Figure 10 depicts the addition energy spectrum of two VCQRs as a function of the inter-ring vertical distance *d*. The addition spectrum of a single ring is also shown (bottom panel) for comparison. It can be seen that the single QR spectrum exhibits strong peaks at N=2, 6, 10, and moderate peaks at N=4, 8. To understand this result, we have to consider that the lateral and vertical motion of the electron in a usual QR can be decoupled, because the lateral confinement is much weaker than the vertical one. As a result, the bottom part of the energy spectrum can be described by the lateral eigenstates of the QR, considering that the electrons are "frozen" in the lowest vertical eigenstate. The lateral eigenstates are of circular symmetry and, according to the Aufbau logic and Hund rules, this corresponds to filled shell configurations at N = 2 + 2*k, and to half-filled shell configurations at N = 1 + 2*k, where k = 0, 1, 2... This is indeed what one observes for the bottom part of the single QR addition spectrum. For a larger number of electrons N, though, Coulomb exchange and correlation energies become comparable to confinement energies and Aufbau/Hund rules are often violated.

In VCQRs, the inter-ring charge density coupling essentially takes place in the vertical (*z*-)direction. For the description of the vertical motion eigenstates, one can no longer consider the lowest eigenstate only, and at least the two lowest vertical eigenstates of this 'artificial molecule' must be taken into account. We label these states according to their parity with respect to the z-inversion as symmetric (|S>) and antisymmetric (|AS>).

Strong, intermediate and weak coupling regimes can be distinguished in VCQRs, depending on the inter-ring separation *d*. An example of strong coupling regime is the *d*=2 nm panel in Fig. 10. In such a case, the addition spectrum is very similar to the single QR one, because the tunneling energy between the ring is very large and |AS> solutions are highly excited. On the other hand, in the weak coupling regime (*d*=6 nm), when the tunneling energy is smaller than the lateral confinement energy, |S> and |AS> states are quasi-degenerate and thus the low-ly-

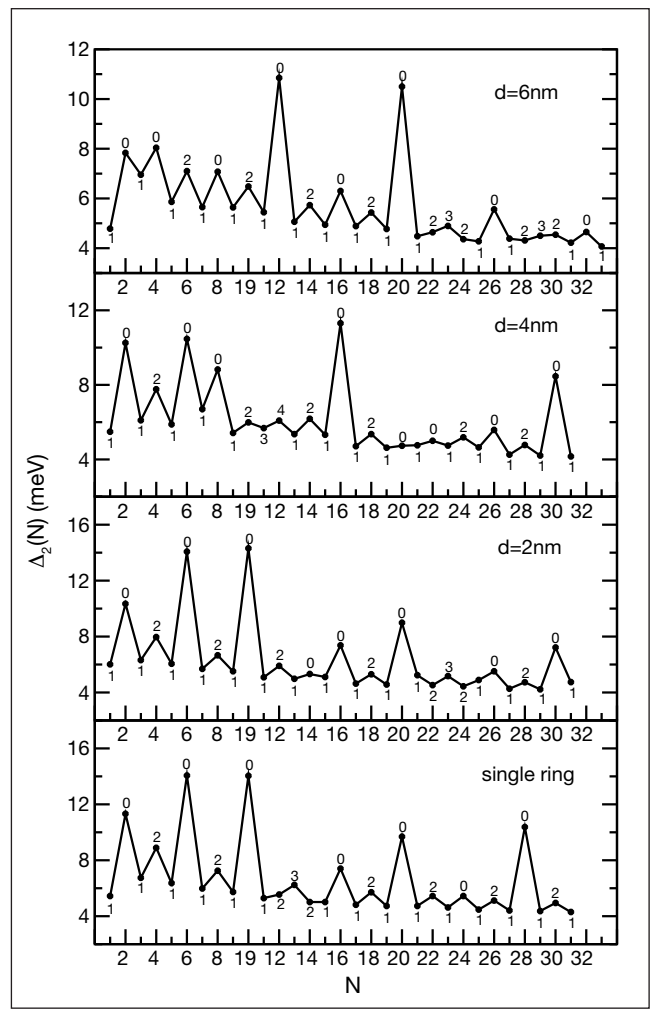

Figure 10. Addition energy spectrum of two VCQRs with inter-ring distances d = 2, 4 and 6 nm. Also shown is the reference spectrum for a single QR. The value of $2S_z$ is indicated next to each peak. Reprinted with permission from [44].

ing part of the spectrum is no longer well-described considering the |S> state only. Indeed, the system is governed by Coulomb interactions and no clear shell structure is observed.

The regime of electronic coupling in VCQRs can be accurately determined by means of far-infrared spectroscopy. If one measured the far-infrared absorption of electrons in VCQRs, two sets of resonances would be obtained. One is connected with transitions involving the selection rule $|\varnothing m| = 1$, while the other involves transitions with selection rule $\varnothing m = 0$. It turns out that in QRs, owing to their particular electronic structure, $|\varnothing m|=1$ transitions take place between electron levels associated with different lateral eigenstates, while $\varnothing m = 0$ transitions take place between levels associated with different vertical eigenstates (e.g. |S> → |AS>). The two kinds of transitions behave differently as a function of axial magnetic fields, inter-ring distance and light-polarization. [46] Therefore, one could conceive an experiment where the far-infrared absorption of self-assembled VCQRs is measured, the nature of each resonance is identified and then, by comparing the excitation energy of the transitions, the energy scale of lateral confinement and tunneling energy is accurately determined. This is shown in Fig. 11, which simulates the single-electron absorption resonances of two InGaAs/GaAs VCQRs for several inter-ring distances *d*. The dashed line is a guide for the eye to follow the |S> → |AS> transition. As *d* increases, we move from a strong coupling regime, where the high energy of the |S> → |AS> transition indicates that |AS> is highly excited, to a weak coupling regime where such a transition is comparable to those between lateral eigenstates (independent of *d*). This means that lateral confinement and tunneling energies are already of the same order.

Concentrically coupled double quantum rings (CCQRs) were first synthesized by means of droplet epitaxy in 2005 [12], and many experimental and theoretical works soon followed that studied the electronic and excitonic properties of these complexes. [47, 48, 49, 50, 51, 52] The interest of these QR structures is that they constitute a unique class of artificial molecule, where lateral coupling is achieved preserving circular symmetry.

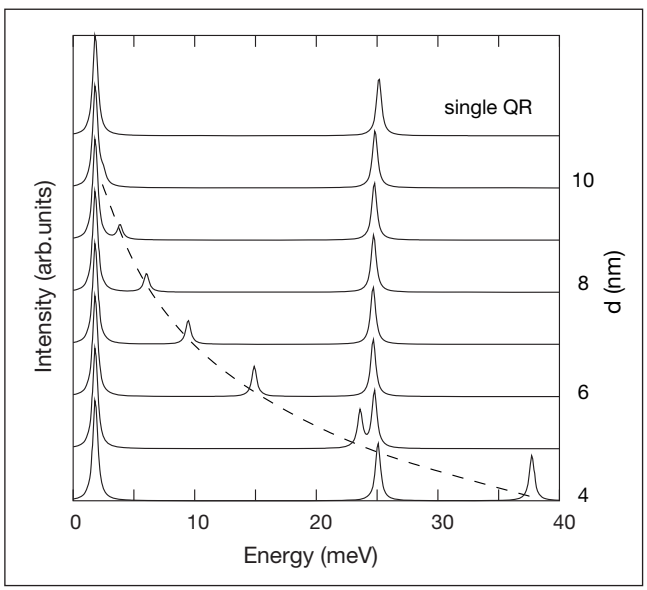

Figure 11. Far-infrared absorption of a single-electron in two VCQRs as a function of inter-ring distance. The dashed line is a guideline for the eye showing the evolution of the |S>-|AS> transition resonance. Note that this resonance is the only one which significantly depends on the inter-ring distance. Reprinted with permission from [46].

The single-electron magnetic response and localization in CCQRs was first studied theoretically by Fuster et al. [48], and soon after by Szafran and Peeters [49], obtaining some contradictory conclusions. Fuster's calculations described the energy magneto-spectrum as two sets of energy levels with well-distinguished AB oscillation periods. This seemed to indicate that the electron states localize either in the inner or in the outer ring. In comparison, Szafran predicted intermediate localization and therefore intermediate AB oscillations periods. In Ref. [50], our group studied the electron and hole energy spectrum in CCQRs of similar dimensions to those of Ref. [12] experiments, using a three-dimensional, effective mass Hamiltonian. Our results were in agreement with those of Fuster, in the sense that a bimodal distribution of energy levels was found. This is illustrated in Fig. 12, where the lowest lying states with radial quantum number $n = 0, 1, 2$ are plotted. One can see that the $n = 0$ and $n = 2$ levels have the same AB oscillation period, which is much shorter than that of $n = 1$. This suggests that $n = 0$ and $n = 2$ states are localized in the outer ring, while $n = 1$ states are localized in the inner ring, thus trapping less magnetic flux and showing slower AB oscillations. These results seem to agree with experimental findings, which are consistent with electrons localizing either in the inner or in the outer ring, depending on

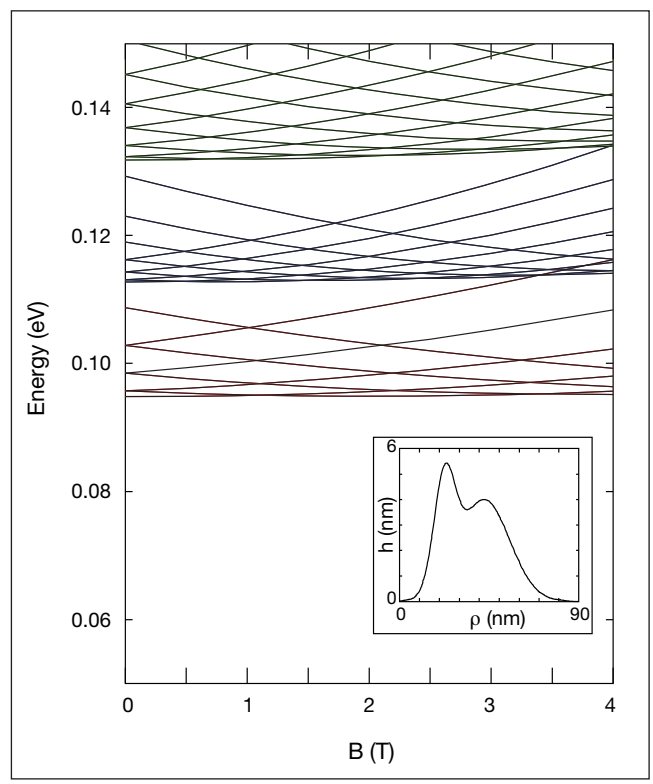

Figure 12. Energy levels vs. magnetic field of one electron in a CCQR with inner ring radius 22.5 nm and outer ring radius 42.5 nm. For the sake of clearness, only the low energy levels with $n = 0, 1, 2$ (n is the number of radial nodes) are displayed. The inset illustrates the cross-section profile. The oscillation period of the $n = 0$ and $n = 2$ levels is similar, and different from the $n = 1$ one. This is a consequence of the electron state localization in the inner or outer ring. Reprinted with permission from [50].

number of radial nodes. [12, 47] Differences with respect to Szafran's predictions were traced back to the confinement potential assumptions: their model represented a two-dimensional (flat) QR system, accounting for the ring-ring coupling by means of a parabolic potential similar to that used in the study of laterally coupled QDs. However, our results proved (see Ref. [50]) that a realistic modeling of the strong vertical confinement was necessary for a realistic description of CCQRs, since small changes could have a strong influence on the localization of the particles. This belief was confirmed later by few-electron calculations [51] using both a parabolic-like coupling potential as the one described in Ref. [49], and a realistic three-dimensional one as in Ref. [50], which enabled direct comparison with the many-body results of Szafran and Peeters.

In Ref. [51] we also showed a novel, interesting feature of CCQRs, namely the formation of a strongly correlated system for inter-ring distances where the electron orbitals localized in the inner and outer rings are quasi-degenerate. In this regime, Coulomb interactions become critical in determining the electron localization within the double ring structure, and one may even obtain spin-dipolar structures. This is illustrated in Fig. 13, which represents the radial profile for spin up (dashed line) and spin down (solid line) charge density.

We finally mention a different type of QR molecules which may be found in the literature, namely laterally coupled quantum rings (LCQRs). These structures are often formed during the self-assembly of QRs, as shown by AFM imaging. [53] The main problem is that the inter-ring distance in these systems is very difficult to control, the spatial distribution of self-assembled nanostructures being fairly random. [54] However, LCQRs exist in current experimental samples and may be selectively addressed by local spectroscopy, so one could also envisage future experiments in which the inter-ring distance is controlled by means of on-template growth. [55] Thus, our group has recently investigated in Ref. [56] the electronic structure of LCQRs, and showed that inter-ring distances induce strong, qualitative changes in the energy spectrum. Moreover, it was shown that the magnetic response can provide valuable information about the coupling regime as well as about the spatial orientation of the QR molecule.

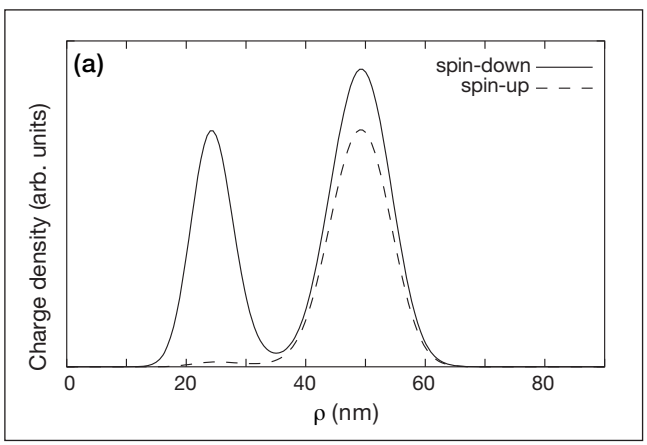

Figure 13. Radial profile of the charge density in a 12-electron CCQR. Note that the inner ring contains only spin-down charge, while the outer ring contains both spin-up and spin-down charge, in such a way that the double ring structure is spin-polarized. Reprinted with permission from [51].

Conclusions and future perspectives

Nanoscopic QRs have undergone spectacular development since they were first synthesized in 1997. Paralleling QD research, a large number of experimental and theoretical studies have dealt with these structures. Much of the interest in QRs stems from their toroidal, doubly-connected topology, which provides them with special quantum properties, most notably those related to the Aharonov-Bohm effect. The fundamental properties of single QRs are now reasonably well understood, as shown in this review of our contributions in relation to theoretical and experimental activity. Thus, a complete assignation of the experimentally reported magneto-optical resonances of self-assembled QRs has been achieved, combined AFM-tunneling electron microscopy experiments have neatly shown the shape of these QRs, magnetization techniques have revealed AB related effects and, more recently, controlled fabrication of QR molecules (both, vertically and concentrically coupled) has been carried out, followed and preceded by theoretical studies on their electronic and excitonic properties. For this reason, current theoretical research partly aims at understanding the deviations from the ideal case of circular, homogeneous QRs. [57] From the experimental point of view we can say that phase coherence in nanoscopic QRs has already been clearly demonstrated, and much of the current activity is being aimed at improving the size and shape control of self-assembled QRs, as well as the extension of single QRs to QR molecules.

Acknowledgments

Continuous support from MEC-DGI projects CTQ2004-02315/BQU, and UJI-Bancaixa project P1-1B2006-03 is gratefully acknowledged. One of our team (J.I.C.) was supported by the EU as part of the Marie Curie IEF project NANO-CORR MEIF-CT-2006-023797.

References

[1] T. Chakraborty (1999), Quantum Dots; Elsevier Science BV, Amsterdam.

[2] M. Büttiker, Y. Imry, and R. Landauer (1983), Josephson behavior in small normal one-dimensional rings, Phys. Lett. 96A, 365-367.

[3] A.G. Aronov, and Yu. V. Sharvin (1987), Magnetic flux effects in disordered conductors, Rev. Mod. Phys. 59, 755-779.

[4] L. Wendler, and V.M. Fomin (1995), Persistent currents in finite-width mesoscopic rings: the role of the interchannel coupling and of the electron-electron interaction, Phys. Status Solidi (B) 191, 409-447.

[5] S. Viefers, P. Koskinen, P. Singha Deo, M. Maninen

(2004), Quantum rings for beginners: energy spectra and persistent currents, Physica E 21, 1-35.

[6] Y. Aharonov, and D. Bohm (1959), Significance of electromagnetic potentials in the quantum theory, Phys. Rev. 115, 485-491.

[7] J. Planelles, J.I. Climente, and J.L. Movilla (2006), Aharonov-Bohm effect for pedestrian, in: Symmetry, Spectroscopy and SCHUR, Proceedings of the Prof. Brian Wybourne Commemorative Meeting, p.1-8.

[8] S. Olariu, and I.I. Popescu (1985), The quantum effects of electromagnetic fluxes, Rev. Mod. Phys. 57, 339-436.

[9] V. Chandrasekhar, R.A. Webb, M.J. Brady, M.B. Ketchen, W.J. Galagher, and A. Kleinsasser (1991), Magnetic response of a single, isolated gold loop, Phys. Rev. Lett. 67, 3578-3581.

[10] J.M. Garcia, G. Medeiros-Ribeiro, K. Schmidt, T. Ngo, J.L. Feng, A. Lorke, J. Kotthaus, and P.M. Petroff (1997), Intermixing and shape changes during the formation of InAs self-assembled quantum dots, Appl. Phys. Lett. 71, 2014-2016.

[11] B.C. Lee, O. Voskoboynikov, and C.P. Lee (2004), III-V Semiconductor nano-rings, Physica E 24, 87-91.

[12] T. Mano, T. Kuroda, S. Sanguinetti, T. Ochiai, T. Tateno, J. Kim, T. Noda, M. Kawabe, K. Sakoda, G. Kido, and N. Koguchi (2005), Self-assembly of concentric quantum double rings, NanoLetters 5, 425-428; J.H. Lee, Zh. Mwang, Z.Y. Abuwaar, N.W. Strom, G.J. Salamo (2006), Evolution between self-assembled single and double ring-like nanostructures, Nanotechnology 17, 3973-3976.

[13] M. Bayer, M. Korkusinski, P. Hawrylak, T. Gutbrod, M. Michel, and A. Forchel (2003), Optical detection of the Aharonov-Bohm effect on a charged particle in a nanoscale quantum ring, Phys. Rev. Lett. 90, 186801.

[14] J.A. Barker, R.J. Warburton, and E.P. O'Reilly (2004), Electron and hole wave functions in self-assembled quantum rings, Phys. Rev. B 69, 035327.

[15] A. Lorke, R.J. Luyken, A.O. Govorov, J.P. Kotthaus, J.M. Garcia, and P.M. Petroff (2000), Spectroscopy of nanoscopic semiconductor rings, Phys. Rev. Lett. 84, 2223-2226.

[16] A. Emperador, M. Pi, M. Barranco, and A. Lorke (2000), Far-infrared spectroscopy of nanoscopic InAs rings, Phys. Rev. B 62, 4573-4577.

[17] R.J. Warburton, C. Schaflein, D. Haft, F. Bickel, A. Lorke, K. Karrai, J.M. Garcia, W. Schoenfeld, and P.M. Petroff (2000), Optical emission from a charge-tunable quantum ring, Nature 405, 926-929.

[18] H. Petterson, R.J. Warburton, A. Lorke, K. Karrai, J.P. Kotthaus, J.M. Garcia, and P.M. Petroff (2000), Excitons in self-assembled quantum ring-like structures, Physica E 6, 510-513.

[19] D. Haft, C. Schulhauser, A.O. Govorov, R.J. Warburton, K. Karrai, J.M. Garcia, W. Schoenfeld, and P.M. Petroff (2002), Magneto-optical properties of ring-shaped self-assembled InGaAs quantum dots, Physica E 13, 165-169.

[20] J. Planelles, and J.I. Climente (2005), Far-infrared absorption of self-assembled semiconductor rings, Collect. Czech. Commun. 70, 605-620.

[21] J.I. Climente, J. Planelles, and W. Jaskolski (2003), Magneto-optical transitions in nanoscopic rings, Phys. Rev. B 68, 075307; J. Planelles, W. Jaskolski, and J.I. Aliaga (2001), Energy structure of quantum rings in a magnetic field, Phys. Rev. B 65, 033306.

[22] V. Gudmundsson, S.I. Erlingsson, and A. Manolescu (2000), Magnetization in short-period mesoscopic electron systems, Phys. Rev. B 61, 4835-4843.

[23] T.H. Oosterkamp, S.F. Godijn, M.J. Uilenreef, Y.V. Nazarov, N.C. van der Vaart, and L.P. Kouwenhoven (1998), Changes in the magnetization of a double quantum dot, Phys. Rev. Lett. 80, 4951-4954.

[24] M.P. Schwarz, D. Grundler, M. Wilde, Ch. Heyn, and D. Heitman (2002), Magnetization of semiconductor quantum dots, J. Appl. Phys. 91, 6875.-6877

[25] L. Jacak, P. Hawrylak, and A. Wojs (1998), Quantum Dots, Springer-Verlag, Berlin.

[26] P.A. Maksym, and T. Chakraborty (1992), Effect of electron-electron interactions on the magnetization of quantum dots, Phys. Rev. B 45, 1947-1950.

[27] J.I. Climente, J. Planelles, and J.L. Movilla (2004), Magnetization of nanoscopic quantum rings and dots, Phys. Rev. B 70, 081301R.

[28] T. Chakraborty, and P. Pietiläinen (1994), Electron-electron interaction and the persistent current in a quantum ring, Phys. Rev. B 50, 8460-8468.

[29] K. Niemela, P. Pietiläinen, P. Hyvönen, and T. Chakraborty (1996), Fractional oscillations of electronic states in a quantum ring, Europhys. Lett. 36, 533-538.

[30] J.I. Climente, J. Planelles, and F. Rajadell (2005), Energy structure and far-infrared spectroscopy of two electrons in a self-assembled quantum ring, J. Phys.: Condens. Matter 17, 1573-1582.

[31] N.A.J.M. Kleemans, I.M.A. Bominaar-Silkens, V.M. Fomin, V.N. Gladilin, D. Granados, A.G. Taboada, J.M. Garcia, P. Offermans, U. Zeitler, P.C.M. Christianen, J.C. Maan, J.T. Devreese, and P.M. Koenraad (2007), Phys. Rev. Lett. 99,146808.

[32] J. Song, and S.E. Ulloa (2001), Magnetic field effects on quantum ring excitons, Phys. Rev. B 63, 125302.

[33] H. Hu, J.L. Zhu, D.J. Li, and J.J. Xiong (2001), Aharonov-Bohm effect of excitons in nanorings, Physical Review B 63, 195307.

[34] A.O. Govorov, A.V. Kalameitsev, R.J. Warburton, K. Karrai, and S.E. Ulloa (2002), Excitons in quantum ring structures in a magnetic field: optical properties and persistent currents, Physica E 13, 297-300.

[35] S.E. Ulloa, A.O. Govorov, A.V. Kalameitsev, R.J. Warburton, and K. Karrai (2002), Magnetoexcitons in quantum ring structures: a novel magnetic interference effect, Physica E 12, 790-793.

[36] A.O. Govorov, S.E. Ulloa, K. Karrai, and R.J. Warburton (2002), Polarized excitons in nanorings and the optical Aharonov-Bohm effect, Phys. Rev. B 66, 081309R.

[37] P. Offermans, P.M. Koenraad, J.H. Wolter, D. Granados,

J.M. Garcia, V.M. Fomin, V.N. Gladilin, and J.T. Devreese (2005), Atomic-scale structure of self-assembled In(Ga)As quantum rings in GaAs, Appl. Phys. Lett. 87, 131902.

[38] T. Ihn, A. Fuhrer, K. Ensslin, W. Wegscheider, and M. Bichler (2005), Spin effects in a quantum ring, Physica E 26, 225-230.

[39] B. Meurer, D. Heitmann, and K. Ploog (1994), Quantum dots in tilted magnetic fields, Surface Sci. 305, 610-614.

[40] T. Pyragiene, and A. Matulis (2000), Parabolic dot in tilted magnetic field, Physica E 8, 230-238.

[41] J. Planelles, J.I. Climente, and F. Rajadell (2006), Quantum rings in tilted magnetic fields, Physica E 33, 370-375.

[42] D. Loss, and D.P. DiVicenzo (1998), Quantum computation with quantum dots, Phys. Rev. A 57, 120-126; G. Burkard, D. Loss, and D.P. DiVicenzo (1999), Coupled quantum dots as quantum gates, Phys. Rev. B 59, 2070-2078.

[43] D. Granados, J.M. Garcia, T. Ben, and S.I. Molina (2005), Vertical order in stacked layers of self-assembled In(Ga)As quantum rings on GaAs (001), Appl. Phys. Lett. 86, 071918.

[44] F. Malet, M. Barranco, E. Lipparini, R. Mayol, M. Pi, J.I. Climente, and J. Planelles (2006), Vertically coupled double quantum rings at zero magnetic field, Phys. Rev. B 73, 245324.

[45] M. Pi, A. Emperador, M. Barranco, F. Garcias, K. Muraki, S. Tarucha, and D.G. Austing (2001), Dissociation of vertical semiconductor diatomic artificial molecules, Phys. Rev. Lett. 87, 066801.

[46] J.I. Climente, and J. Planelles (2005), Far-infrared absorption of vertically coupled self-assembled quantum rings, Phys. Rev. B 72, 155322.

[47] T. Kuroda, T. Mano, T. Ochiai, S. Sanguinetti, K. Sakoda, G. Kido, and N. Koguchi (2005), Optical transitions in quantum ring complexes, Phys. Rev. B 72, 205301.

[48] G. Fuster, M. Pacheco, and Z. Barticevic (2004), Electronic properties of coupled quantum rings in the presence of a magnetic field, Braz. J. Phys. 34, 666-668.

[49] B. Szafran, and F.M. Peeters (2005), Few-electron eigenstates of concentric double quantum rings, Phys. Rev. B 72, 155316.

[50] J. Planelles, and J.I. Climente (2005), Semiconductor concentric double rings in a magnetic field, Eur. Phys. J. B 48, 65-70.

[51] J.I. Climente, J. Planelles, M. Barranco, F. Malet, and M. Pi (2005), Electronic structure of few-electron concentric double quantum rings, Phys. Rev. B 73, 235327.

[52] F. Malet, M. Pi, M. Barranco, E. Lipparini, and Ll. Serra (2006), Optical response of two-dimensional few-electron concentric double quantum rings: A local-spin-density-functional theory study, Phys. Rev. B 74, 193309.

[53] B.C. Lee, and C.P. Lee (2004), Formation of semiconductor quantum rings using GaAs/AlAs partially capped layers, Nanotechnology 15, 848-850; A. Schramm, T. Kipp, F. Wilde, J. Schaefer, Ch. Hyen, and W. Hansen (2006), Shape transformation of self-assembled InAs quantum dots during overgrowth with AlAs, J. Cryst. Growth 289, 81-88.

[54] M. Meixner (2002), Simulation of self-organized growth kinetics of quantum dots, Ph. Dissertation, Technischen Universität Berlin, Berlin.

[55] D. Chithrani, R.L. Williams, J. Lefebvre, P.J. Poole, and G.C. Aers (2003), Optical spectroscopy of single, site-selected, InAs/InP self-assembled quantum dots, Appl. Phys. Lett. 84, 978-980.

[56] J. Planelles, F. Rajadell, J.I. Climente, M. Royo, and J.L. Movilla (2007), Electronic states of laterally coupled quantum rings, Journal of Physics: Conference Seriers 61, 936-941. See also cond-mat/0606438.

[57] J. Planelles, F. Rajadell, and J.I. Climente (2007), Quantum rings with structural distortions under axial or in-plane magnetic fields, Nanotechnology, 18, 375402.1-11.

[58] I. Filikhin, V.M. Suslov, and B. Vlahovic (2006), Modeling of InAs/GaAs quantum ring capacitance spectroscopy in the nonparabolic approximation, Phys. Rev. B 73, 205332.

[59] R. Blossey, and A. Lorke (2002), Wetting droplet instability and quantum ring formation, Phys. Rev. E 65, 021603.

About the authors

J.I. Climente received his Ph.D. from Universitat Jaume I *in 2005, in recognition of his work in the study of the energy structure of semiconductor QDs within the k·p theory. He then became a Marie Curie post-doctoral fellow in the Italian National Research Council-Institute of Matter Physics-S3 in Modena, Italy, where he is investigating the effects of electronic correlations on different aspects of quantum dot physics. He collaborates with the Quantum Chemistry group at the* Universitat Jaume I*, the Many-Body Interacting Systems Group at the* Universitat de Barcelona *and the Quantum Theory group at the Institute of Microstructures (Ottawa, Canada).*

J. Planelles is currently a full professor at the Universitat Jaume I *(Castelló, Spain) and the head of the Quantum Chemistry research group. He received his Ph.D degree in chemistry with a special award in 1984 from the* Universitat de València*, where he later became a professor, for his theoretical studies on Petrochemistry. As a visiting professor, he has worked at different research centers: CSIC (Madrid, Spain), Univ. M. Kopernika (Torun, Poland), Univ. Waterloo (Waterloo, Ontario, Canada), his research work covering several fields, mainly: catalysis and petrochemistry, the mathematical properties of density operators in relation to statistical spectroscopy of Hamiltonian operators and spin-adapted reduced Hamiltonians, new developments in coupled cluster theory, and studies of confined model systems. In recent years his scientific interests have focused on the study of the physical properties of semiconductor nanocrystals. His output has included more than one hundred publications, most of which have been scientific papers in internationally refereed journals but he has also published lecture notebooks for students.*