

SOLID STATE AND MAGNETISM

Quantum dots: periodicity and confinement (spatial and magnetic)

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A quantum dot is a small crystal of nanometric size. It is built of a huge amount of electrons and nuclei. As in macroscopic crystals, atoms inside quantum dots are arranged according to an ordered pattern, displaying microscopic periodicity. However, nanocrystals and macroscopic crystals display quite different behaviour and properties. In a macroscopic crystal, despite its finite size, most of the properties are bulk-like. At the electronic scale, macroscopic crystal is so huge that electrons see the crystal border as if located at infinity. This allows to fully exploiting translational symmetry, drastically simplifying its quantum mechanical description. If we now introduce a heterojunction (or consider a nanocrystal) we still have local periodicity. However, the border is no longer felt at infinity. Instead, we may consider the border effect as a potential superimposed to the crystalline periodic potential. This extra potential is extremely smooth at the scale of a lattice constant. Physically, it behaves as a *confining potential* which renders a *discrete* character to the electron energy structure. The possibility to precisely control the size of a quantum dot enables to determine the wavelength of the absorption and emission, which in turn determines the colour of light the human eye perceives. Quantum dots can therefore be tuned during production to emit any desired colour light. The smaller the dot, the closer it is to the blue end of the spectrum, and the larger the dot, the closer to the red end. Quantum dots can even be tuned beyond visible light, into the infra-red or into the ultra-violet. In addition to this potential, hereafter referred to as *spatial confining potential*, externally applied fields or just the presence of interfaces may bring about additional sources of confinement, as the magnetic confinement. We can therefore understand why quantum dots are called *artificial atoms*, as the manufacturer can tune its properties in the fabrication process or/and by means of appropriate external sources of confinement. Along two lectures, we will provide a bird's eye view on periodicity and spatial and magnetic confinements on quantum dots. The aim of these talks, more than introducing the world of nanosystems is to revisit some basic knowledge viewed from and used for solid state and nanoscience.

1. PERIODICITY AND SPATIAL CONFINEMENT

1.1. Crystal structure: direct and reciprocal lattices

A crystal is a solid material whose constituent atoms, molecules or ions are arranged in an orderly repeating pattern. The pattern can be described as a motif (*basis*) which is repeated periodically at the points of an underlying lattice which looks the same from when viewed from any of its points (*Bravais lattice*). Because of symmetry restrictions, only a few types of Bravais lattices exist. The (smallest) region of the crystal that fills the entire space when repeated by translation is known as (primitive) unit cell. Of particular interest is the so-called *Wigner-Seitz cell*, a primitive unit cell which preserves the point symmetry of the crystal. The Wigner–Seitz cell around a lattice point is defined as the locus of points in space that are closer to that lattice point than to any of the other lattice points. To build it, lines are drawn to all nearby (closest) lattice points. At the midpoint of each line, another line (2D)/plane (3D) is drawn normal to each of the first set of lines.

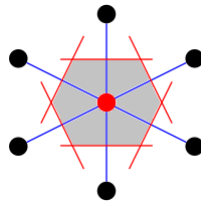


Figure 1.1: Construction of a Wigner–Seitz primitive cell.

The periodicity of a crystal, i.e., the existence of a lattice, is related to the translation symmetry and has profound consequences in the physical behaviour and the electronic structure of solids. For example, the observable electronic density $\rho(\mathbf{r})$ is periodic ($\rho(\mathbf{r}) = \rho(\mathbf{r}+\mathbf{t})$, with \mathbf{t} a translation vector). The square of the modulus of the wave function then is periodic, and therefore the wave function is periodic up to a phase. These phases are precisely the characters of the irreducible representations (irreps) of the translation group (since translations commute, the translation group is commutative, so all its irreps are one-dimensional). The linear momentum is the generator of translations ($\hat{T}_n = e^{i a n \hat{p}}$). Then, its eigenfunctions $\{e^{ikx}\}$ can be used as basis of the irreps, the calculation of their characters and the associated quantum number k as label for these irreps. However, while k can assume the value of all possible finite real numbers, the number of non-equivalent irreducible representations of the translation group is much smaller because $e^{ika} = 1$, where a is a point of the Bravais Lattice, that means $e^{ikx} = e^{ik(x+a)}$, i.e., k and $(k+a)$ are the same irrep as they have the same set of characters. We see then that for each point of the direct Bravais lattice (a)

there is a point k in the so-called *reciprocal* space (units 1/length). The full set of k points form a Bravais lattice (K) whose Wigner-Seitz cell is known as the *first Brillouin zone*. The reciprocal lattice is extremely useful in the study of crystals.

1.2. Translations in 3D: Bloch functions

The extension to three dimensions implies replacing x by \mathbf{r} and k by \mathbf{k} . Now the label of the three-dimensional translation group is not a number k but a vector \mathbf{k} . The most general basis of the irreps are the so-called Bloch functions: $\Psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u(\mathbf{r})$; $u(\mathbf{r} + \mathbf{a}) = u(\mathbf{r})$. We also can define in 3D the reciprocal lattice and identify the labels of all non-equivalent irreps within the 3D first Brillouin zone.

1.3. Solving the Schrödinger equation

In order to solve the Schrödinger equation of a crystal we use translational symmetry and the characters of the irreducible representation to set the required boundary conditions: since $\hat{T}_{\mathbf{a}} \Psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{a}} \Psi(\mathbf{r})$ then, $\Psi_{\mathbf{k}}(-\mathbf{a}/2) = e^{i\phi} \Psi_{\mathbf{k}}(\mathbf{a}/2)$, $\phi \in [-\pi, \pi]$. We solve the Schrödinger equation for each k value and find a set of low-lying eigenvalues. The plot vs. k of the n -th eigenvalue, $E_n(k)$, represents an energy band.

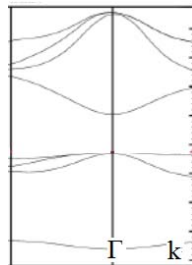


Figure 1.2: energy bands

1.4 Energy band structures

As a starting point to study the energetics of crystal solids, we consider an electron travelling along the crystal. The *quasi-free electron* feels the Coulomb potential exerted by the lattice ions as a perturbation. This kind of carriers is responsible for most of the solid's collective properties.

Let us first assume that the kinetic energy of the electron greatly exceeds the potential, $\epsilon \gg V$ (empty lattice). The Hamiltonian describing the system and its solutions are given by:

$$\frac{p^2}{2m} \Psi(\vec{r}) = \epsilon_k \Psi(\vec{r}), \quad \Psi(\vec{r}) = N e^{i\vec{k}\cdot\vec{r}}, \quad \epsilon_k = \frac{\hbar^2 k^2}{2m}.$$

With $p = \hbar k$ the linear momentum and N the normalization constant. Such a wave function is

known as *plane wave*, and its energy structure vs k is shown in figure 1.3a. At zero temperature, the electrons in a solid will occupy the lowest orbitals allowed by the Pauli exclusion principle, from $k=0$ till $k=k_F$. We then define the *Fermi energy* as $\epsilon_F = \hbar^2 k_F^2 / 2m$.

Let us next include the lattice as a weak perturbation. It can be shown that electrons will not feel the lattice unless $k \sim \pi/a$, where a is the lattice constant. If, $k \sim \pi/a$, the plane wave experiences Bragg reflection. This leads to forbidden values of k , where energy gaps appear (figure 1.3b). Depending on the magnitude of the gap and the position of the Fermi energy, we classify solids as insulators, semiconductors and conductors (metals) –figure 1.3c-.

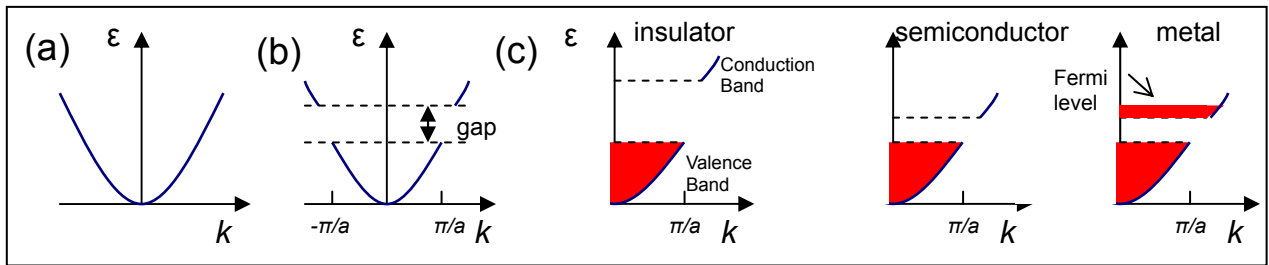


Figure 1.3.

1.5 The $k \cdot p$ model. Effective mass.

A computationally affordable yet reliable description of many semiconductor crystal is obtained using the $k \cdot p$ method. The idea is to project the crystal Hamiltonian, $H = p^2/2m + V_{cr}(\mathbf{r})$, onto a general Bloch function $\psi(\mathbf{r}) = N e^{i\mathbf{k}\mathbf{r}} u_{\mathbf{k}}(\mathbf{r})$. After operating and left-multiplying by $e^{-i\mathbf{k}\mathbf{r}}$, one gets the $k \cdot p$ Hamiltonian:

$$\left(\frac{p^2}{2m} + V_{cr}(\vec{r}) + \frac{\hbar^2 k^2}{2m} + \hbar \mathbf{k} \cdot \mathbf{p} \right) u_{\mathbf{k}} = \epsilon_{\mathbf{k}} u_{\mathbf{k}}.$$

If we have some information about $u_{\mathbf{k}}$ at a given point $k=0$ (Γ point, at the centre of the Brillouin zone), we can solve the Hamiltonian perturbationally for k near $k=0$. To this end, we expand the Hamiltonian in terms of a basis of functions $u_{\mathbf{k}}(\mathbf{r}) = \sum_n c_{n\mathbf{k}} u_{n0}(\mathbf{r})$. A general matrix element is of the form:

$$\langle u_{n0} | \hat{H}_{kp} | u_{l0} \rangle = \left(\epsilon_{n0} + \frac{\hbar^2 k^2}{2m} \right) \delta_{nl} + \frac{1}{m} \vec{k} \cdot \vec{P}_{nl},$$

Where ϵ_{n0} is the *energy gap* and $\mathbf{P}_{nl} = \langle u_{n0} | \mathbf{p} | u_{l0} \rangle$ is the *Kane parameter*. Both magnitudes can be inferred from spectroscopy experiments. Depending on the number n of functions in our basis, we speak about n -band $k \cdot p$ models. Typical models employed to describe zinc-blende semiconductors are the 1-band model for conduction electrons, 4-band model for valence holes and 8-band model to include coupling between conduction and valence bands.

The 1-band model (all models indeed) is usually solved up to second-order perturbation

theory, leading to energies:

$$\varepsilon_{nk} = \varepsilon_{n0} + \sum_{\alpha=x,y,z} \frac{k_{\alpha}^2}{2} \left(\frac{1}{m} + \frac{2}{m} \sum_l \frac{P_{nl}^{(\alpha)2}}{(\varepsilon_{n0} - \varepsilon_{nl})} \right) = \varepsilon_{n0} + \sum_{\alpha=x,y,z} \frac{k_{\alpha}^2}{2m_{\alpha}^*}.$$

Note that the energy is simply that of a free electron but with an *effective mass* m^* , which depends on the coupling with other bands. The effective mass can have small and even negative values, which leads to lots of interesting physics in semiconductors.

It must be pointed out that perturbation theory becomes quite more complex for many-band models and that, actually, nobody calculate the huge amount of integrals involved in the perturbation expansions, but group them and fit to the experiment. A good (simpler and deeper) alternative to perturbation expansions for reaching the Hamiltonian is to determine it by pure symmetry considerations: the theory of Invariants comes to play. The fundamental underlying idea is that a product $A \cdot B$ is invariant (A_1 symmetry) if A, B are of the same symmetry. Since the Hamiltonian must be quadratic in k (as it comes from a second order perturbation), then $H = \sum M_{ij} k_i k_j$. Therefore, we must write down symmetry-adapted $k_i k_j$

products (as are given in the character table of the involved point group, T_d for zinc-blende semiconductors) and find out appropriate partner matrices (of the same symmetry). To this end one can employ symmetry-adapted products of matrix representations of the angular

momentum. The resulting Hamiltonian can be written $H = \sum_i^{\dim(\Gamma)} \sum_{\Gamma} a_{\Gamma} N_i^{\Gamma} k_i^{\Gamma}$, where N_i^{Γ} is the symmetry-adapted matrix, k_i^{Γ} the symmetry-adapted $k_i k_j$ product and a_{Γ} a *fitting* parameter that cannot be determined by symmetry considerations.

1.6 Heterostructures. Quantum wells, wires and dots.

So far we have considered homogeneous crystals. However, current semiconductor research is largely focused on semiconductors made of different materials. How do we model a system with two semiconductor crystals A and B (figure 1.4a)? If both materials have the same crystal structure and similar lattice constant, one usually employs the *envelope function approximation*. We replace the usual Bloch function by: $\psi(\mathbf{r}) = N e^{ik_{\perp} \cdot \mathbf{r}_{\perp}} \chi(z) u_k(\mathbf{r})$, where $\chi(z)$ is an unknown function in the direction where the translational symmetry has been broken. Noteworthy, $f(\mathbf{r}) = e^{ik_{\perp} \cdot \mathbf{r}_{\perp}} \chi(z)$, varies slowly as compared to the unit cell function $u_k(\mathbf{r})$. Thus, it is often referred to as the envelope function, which is modulated inside each unit cell by u_k . Next, one proceeds as in the homogeneous case, with the additional consideration that the

integral $\int_{\Omega} f(\mathbf{r})u(\mathbf{r}) d\mathbf{r}^3 \approx 1/\Omega (\int_{\Omega} u(\mathbf{r}) d\mathbf{r}^3) (\int_{\Omega} f(\mathbf{r}) d\mathbf{r}^3)$, with Ω standing for the crystal volume. For a one-band model, eventually this leads to the eigenvalue equation:

$$\left(\frac{p_z^2}{2m} + V(z) + \frac{\hbar^2 k_{\perp}^2}{2m} + \varepsilon_0^A \right) \chi(z) = \varepsilon \chi(z),$$

where $V(z)$ is the spatial confinement potential defined by the alignment between the band gaps of materials A (ε^A) and B (ε^B). As shown in figure 1.4b, for a B/A/B heterostructure the potential can be that of a *quantum well*, and the eigenstates $\chi(z)$ the solutions particle in the box. Thus, semiconductor heterostructures provide the experimental realization of fundamental systems which had hitherto been but theoretical idealizations. It is also possible to fabricate heterostructures where translational symmetry is broken in two and three directions of the space, which gives rise to *quantum wires* and *quantum dots*, respectively. Of particular interest are quantum dots, where a strong spatial confinement in the three directions leads to discrete energy levels, very much as in atoms. Because of this quantum dots are often referred to as artificial atoms or macroatoms. Unlike natural atoms, however, quantum dots can be built with different sizes, shapes and composition, so that the energy spectrum is designed at will. This has opened great prospects for fundamental research and technological developments over the last two decades.

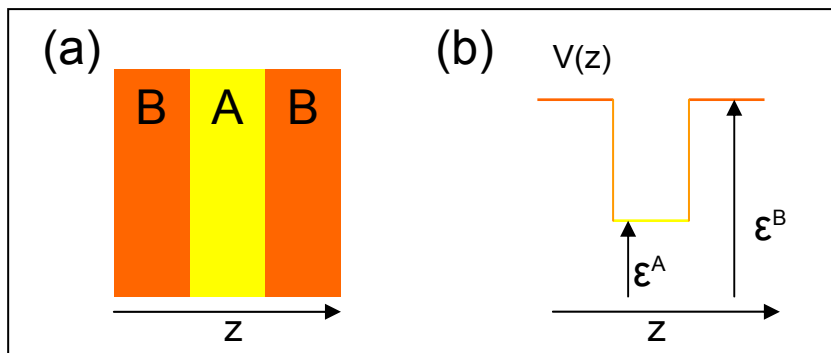


Figure 1.4.

2. MAGNETIC CONFINEMENT

2.1. Overview: Lagrange equations with velocity-dependent potentials

The concept of a Lagrangian L is introduced in order to reformulate classical Newton mechanics. In the case of conservative systems, the Lagrangian is defined as the velocity-dependent kinetic energy T of the system minus its coordinate-dependent potential energy V , $L=T-V$, and the Lagrange equation,

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{x}}\right) - \left(\frac{\partial L}{\partial x}\right) = 0,$$

can be seen as a rewriting of the Newton law $\frac{d}{dt}(p) - F = 0$, as can be easily checked. When the potential does not depend on the velocity, kinetic momentum (velocity-derivative of the kinetic energy) and canonic momentum (velocity-derivative of the Lagrangian) coincide. In this case, the Hamiltonian (Lagrange transformation of the Lagrangian function) is just the total energy $H=T+V$.

When the potentials also depend on velocity, $U(x, v)$, kinetic π and canonic p momenta are different and the Hamiltonian coincides with the kinetic energy. It can be checked in the case of a particle in a magnetic field, described by the potential $U = -e(v \cdot A)$, with A being the potential vector, related to the magnetic field by the equation $B = \nabla \wedge A$. One can easily find that $p = \pi + eA$, and that the Hamiltonian can be written as $H = \frac{1}{2m}(p - eA)^2$.

2.2. Coulomb gauge and quantification

We can realize that, as far as $\chi(x)$ is a scalar potential, two vector functions $A(x)$ and $A(x) + \nabla\chi(x)$ yield the same magnetic field (since $\nabla \wedge \nabla\chi(x) = 0$). This means that we have some freedom to select the potential vector. Having in mind the transition to quantum mechanics, where momentum ($-i\hbar\nabla$) and coordinate do not commute, it is customary to select $\chi(x)$ such that it yields a null divergence of A ($\nabla \cdot A = 0$). This is called the Coulomb gauge. Assuming this gauge, the quantification ($p \rightarrow -i\hbar\nabla, x \rightarrow x$) of the Hamilton function yields

$$H = \frac{\hat{p}^2}{2m} - \frac{e}{m} A \cdot \hat{p} + \frac{e^2}{2m} A^2$$

In the case of a uniform axial magnetic field, $B = B_0 \vec{k}$, we can select a potential

vector $A = [-yB_0/2, xB_0/2, 0]$ and this equation turns into:

$$H = -\frac{\hbar^2}{2m}\nabla^2 + \frac{e^2 B_0^2}{8m}\rho^2 - \frac{e B_0}{2m}\hat{L}_z, \text{ with } \rho = \sqrt{x^2 + y^2}.$$

2.3. Magnetic Confinement: Landau levels

By introducing the notation $\omega = -e B_0 / 2m$, the above equation can be rewritten as:

$$H = -\frac{\hbar^2}{2m}\nabla^2 + \frac{1}{2}m\omega^2\rho^2 + \omega\hat{L}_z = \frac{\hat{p}_z^2}{2m} + H_{HO}^{2D} + \omega\hat{L}_z$$

Since the two-dimensional harmonic oscillator Hamiltonian, \hat{H}_{2D}^{HO} , commutes with the z-component of the angular momentum, \hat{L}_z , in addition to the kinetic energy along z, the energy of the system is just $E(m, M) = (2n + |M| + M + 1)\omega$, where ω is proportional to the magnetic field. These are the so-called Landau levels of a free electron in a uniform magnetic field. As we see, the energy levels grow linearly without crossings as the magnetic field increases.

2.4. Spatial and magnetic confinement in quantum dots

When an additional, spatial confining potential co-exists with the magnetic confinement, the above Landau levels mix and we find out a competition between linear and the quadratic terms of the magnetic field. For example, the case of a spherical quantum dot in a homogeneous magnetic field is described by the following eigenvalue equation

$$\left(-\frac{\hbar^2}{2m_e}\nabla^2 + \frac{e^2 B^2 \rho^2}{8m_e} + \frac{e B M}{2m_e} + V_e(\rho, z) \right) \Phi_{n,M} = E_{n,M} \Phi_{n,M}$$

We find a very different behaviour for short/large radius, owing to the ρ^2 factor appearing in the second term of the above equation. As a result, for small radius the response to the magnetic field is atomic-like (just an energy splitting due to $W = -\mu B$, see figure 3.1a) while for larger systems one may envisage the formation of Landau levels for extremely large magnetic fields, where the magnetic confinement overpasses the spatial one –figure 3.1b-.

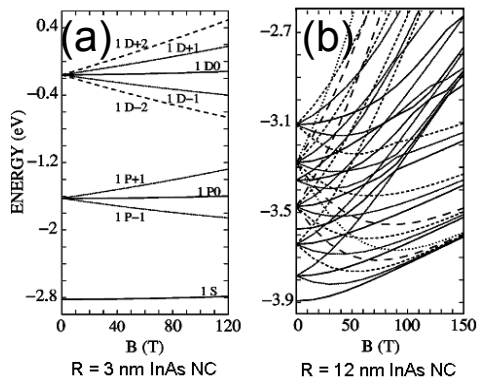


Figure 3.1

2.5. Multiple-connected quantum systems: Aharonov-Bohm effect

The response of a system with a multiply connected topology, as for example a quantum ring, is even more complex, showing level crossings as the magnetic field increases. This is related to the so-called Aharonov-Bohm effect. In figure 3.2 (from J. Planelles, W. Jaskólski, and I. Aliaga, Phys. Rev. B 65 (2001) 033306) we show the two low-lying bands of a prototypical InAs quantum ring.

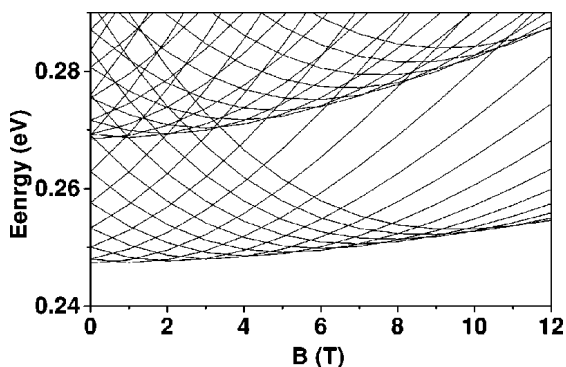


Figure 3.2

The most intriguing aspect of the Aharonov-Bohm effect is that these crossings can be predicted even in the case that the magnetic field pierces the inner hole of the ring only, i.e. no magnetic field acts on the ring section, the region where the electron is located. The mathematical reason comes from the fact that it is the potential vector and not the magnetic field itself that comes into the Hamiltonian, so that an electron can be influenced by the potentials even if no fields act upon it. The physical counterpart is a set of experimental observations that can only be understood as coming from this purely quantum effect without a classical correspondence.

2.6. Lattices and magneto-translations: Hofstadter butterfly

When one proceeds to calculate the energy structure of a two-dimensional periodic system

pierced by an axial magnetic field, a fractal-like spectrum is obtained, which is referred to as Hofstadter butterfly (in honour to Douglas Hofstadter, which described it for the first time in Phys. Rev. B 14 (1976) 2239), see figure 3.3a.

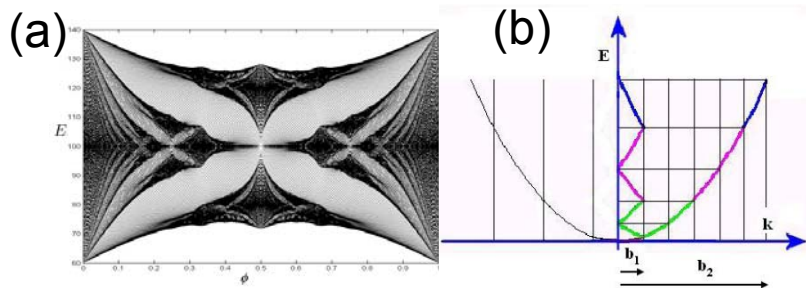


Figure 3.3

We may understand this fractal structure using the concept of magnetotranslation (E. Brown, Phys. Rev. 133 (1964) A1038). In the absence of magnetic field, the Bloch functions are simultaneously eigenfunctions of the Hamiltonian and the translation operator. In the presence of magnetic field, magnetotranslations commute with the Hamiltonian but the size of the magnetic unit cell (cell pierced by a flux unit) depends on the magnetic field. In order to get Bloch-like functions, spatial and magnetic unit cells must have a commensurate relation. So we meet a two-fold periodicity, spatial and magnetic. As we increase the magnetic field from zero, the magnetic unit cell in the direct space becomes extremely large (and then very small in the reciprocal space) thus producing the splitting the zero field band into a huge set of minibands (whose number and distribution depends on the magnetic field). As the magnetic flux on the spatial unit cell approaches unity, magnetic and spatial unit cells coincide and the system recover a unique band, as at $B=0$.

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3. HANDS ON SESSION

The aim of this session is to introduce the students to the task of identifying a system, translate it into a differential equation, numerically integrate it and plot the results. To this end we propose a set of elementary 1D/2D examples of increasing difficulty. The first and most simple case of determining the bottom of the bands in a quantum well (or equivalently, the energy spectrum of a 1D QD) is fully developed and used to introduce some key matlab commands that will be used in the later examples. Also some basic on differences finites to numerically integrate differential equations, choice of appropriate boundary conditions, building up appropriate matrix representation and further diagonalization are outlined. Then the rest of examples are sketched with some hints.

Exercise 1. Write a code to calculate the electron energy spectrum of a 1D GaAs/AlGaAs QD as a function of the size. Consider GaAs effective mass, $m^*=0.05$, all over the structure. Justify the assumed simplification. The GaAs/AlGaAs band off set amounts 0.25 eV. Assume a well width $L=25\text{nm}$. Repeat the calculations for L ranging in $[5, 50]$ nm and plot a few low-lying energies vs. L . Discuss the obtained results.

Exercise 2. Write a code to calculate the electron energy spectrum of two coupled GaAs/AlGaAs QDs as a function of their separation S . Plot the two lowest states for $S=1$ nm and $S=10$ nm and discuss the obtained results.

Exercise 3. Write a code to calculate the electron energy spectrum of $N=20$ coupled GaAs/AlGaAs QDs as a function of their separation S . Plot the charge density of the $n=1, 2$ and $n=21, 22$ states for $S=1$ nm and $L=5$ nm.

Exercise 4. Write a code to calculate the energies of an electron in a 2D GaAs/AlGaAs cylindrical quantum ring with inner radius R_{in} and outer radius R_{out} , subject to an axial magnetic field B . The system (ring) is made of GaAs. The surrounding matrix and inner hole of the ring is made of AlGaAs. Consider a finite width L_b for the surrounding AlGaAs matrix, $L_b=10$ nm, (i.e., assume that beyond of this distance the wave function is zero). Calculate the energies as a function of B in the range 0-20 T, both, for a structure with $(R_{\text{in}}, R_{\text{out}})=(0, 30)$ nm –i.e. a quantum disk- and for $(3, 30)$ nm –a quantum ring-. Discuss the role of the linear and quadratic magnetic terms in each case.