# 1
# Introduction

At the scale of nanometers, approaching the size of the fundamental constituents of materials, various subdisciplines of physics merge. To provide a closed description of many nanoscale phenomena, considerations from varied fields must be taken into account, such as quantum mechanics, optics, quantum optics, semiconductor physics, material science, atomic and molecular physics, and so on. Crucially, theory and experiments must go hand in hand in the discovery and elucidation of such effects. In this book, we present one example of such nanoscale science – the physics of spins in optically active quantum dots. The variety of areas of physics that must be brought to mind to understand these systems can be seen in the structure of this book, which contains chapters on materials growth and synthesis, solid state and quantum optics theory, and experimental methods.

This chapter will introduce the two key terms in the title: spin, the fundamental angular momentum of a particle, and nanometer-sized semiconductor structures, known as quantum dots. Specifically, we focus on quantum dots whose properties can be measured and controlled via their interaction with light, and how spins in these structures may be investigated and potentially used for novel applications such as quantum information processing. We expand the discussion on quantum dots in Chapter 2 by describing the two main fabrication techniques, semiconductor epitaxy and chemical synthesis. This is followed by some theoretical background on semiconductor physics and confined states in different types of quantum dots in Chapter 3. We then show in Chapter 4 that semiconductor diodes and optical cavities can be used to provide the knobs required to control the electrical, optical, and spin properties of optically active quantum dots for applications. To back up all the experimental findings and techniques, Chapter 5 provides the elementary theory of the coupling between confined states to electromagnetic radiation. The interactions between spins of carriers and a carrier and the nuclei in the dot's crystal lattice are then discussed in Chapter 6, before we switch back to experimental techniques. The rich toolkit to initialize, manipulate, and read out spins in quantum dots by optical means is opened and explained in Chapter 7. In the concluding Chapter 8 we will add another important part to this discussion, namely the coupling of quantum dots. In coupled quantum dots the interactions between charges and spins show a subtle interplay and provide us with the potential to use these optically active nanostructures for a scalable architecture for

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quantum information processing and also to observe fundamental phenomena in coupled quantum dots, analogous to the effects of coupled atoms in molecular physics.

## 1.1 Spin

Spin, the intrinsic angular momentum of a particle, was first described theoretically by George Uhlenbeck and Samuel Goudsmit in 1925, and formalized by Wolfgang Pauli in 1926. Experimentally, however, spin phenomena have been observed and put to practical use for much longer. The earliest known spin-based device is most likely the magnetic compass. Here, a freely rotating needle is constructed out of a material in which electron spins align with each other under their mutual exchange interaction. This leads to a macroscopic spin polarization in the needle (ferromagnetism), causing the needle to align with the Earth's magnetic field due to the Zeeman interaction of a spin with a magnetic field (see Eq. (1.5)). Written records from ancient China referring to these phenomena date back to the fourth century BC.

Over the next 2300 years or so, the knowledge of magnetism spread across the globe. Clever minds devised new uses for the phenomenon, and refined old ones, ranging from the electric motor, to the dynamo, to the posting of notes on a refrigerator. Despite its bountiful technological applications, at the beginning of the twentieth century, the physical origins of magnetism were still unclear.

As quantum mechanics was being developed in the 1920s, great strides were made in understanding atomic spectra by quantizing the orbital momentum of electrons around the atomic nucleus. However, results such as the Stern–Gerlach experiment, and unexplained splittings in atomic spectra (the "anomalous Zeeman effect", and hyperfine splitting) indicated that there were extra quantum degrees of freedom not being taken into account.

A natural candidate for this unknown quantity was the angular momentum of a particle. The idea was at first considered to be impossible. Given the known upper bound on the radius of the electron, the angular velocity of the electron would need to be impossibly high to provide the observed splittings. Nevertheless, Uhlenbeck and Goudsmit published the idea in 1925. Despite its apparent impossibility, the idea of "spin" nicely explained the observations. Originally a skeptic, Wolfgang Pauli warmed to the idea and ran with it, redefining spin not as an actual rotation of a particle, but as an angular momentum intrinsic to the particle, just as charge or mass are intrinsic properties. He then went on to develop a formalism for dealing with spin in (nonrelativistic) quantum mechanics (see, e.g., [1]).

Once this theoretical framework was in place, the experimental study of spin physics could now proceed hand-in-hand with theory, instead of the pure phenomenology of the previous millennia. Throughout the rest of the twentieth century numerous advances were made, such as a detailed understanding of magnetic materials, nuclear spin physics, and spin resonance phenomena. These discoveries

led to revolutionary technologies such as magnetic resonance imaging (MRI) and magnetic data storage (tapes, hard drives).

The reservoir of interesting spin phenomena is still far from dry. Recent advances in materials, electronics, and low temperature technologies have brought new untapped wells of spin physics within reach. One of the fruits of these new capabilities has been the development of quantum dots, which creates a straightforward way to isolate single or few spins for study or possible applications.

## 1.2 Spin-1/2 Basics

According to quantum mechanics, angular momentum as an observable can be described by two quantities: the angular momentum quantum number $l$ and the projection of angular momentum on the (say) $z$ axis, $m$. Throughout this book, we do not indicate operators with any special notation, assuming that the reader is familiar with the basics of quantum mechanics and that it is clear from the context which quantities are operators. The angular momentum quantum numbers are just the eigenvalues of the commuting operators $\mathbf{L}^2$ and $L_z$,

$$\mathbf{L}^2|\psi\rangle = l(l+1)\hbar^2|\psi\rangle \quad \text{and} \quad L_z|\psi\rangle = m\hbar|\psi\rangle \ , \tag{1.1}$$

where $\mathbf{L}^2 = L_x^2 + L_y^2 + L_z^2$, with $L_\alpha$ the angular momentum operator along the $\alpha$ direction, and $|\psi\rangle$ is a quantum mechanical state in Dirac notation. The quantum number $l$ can take on half-integer values, and for a given $l$, the projection $m$ can take on values $m = -l, -l+1, \ldots, l$.

In the case of a particle's spin, we consider an internal angular momentum with fixed quantum number $s$, and the projection of the spin can take on $2s+1$ values, from $-s$ to $s$. An electron has total spin $s = 1/2$ and projections $m_s = \pm 1/2$. Therefore, there are two eigenstates for $s = 1/2$, one with $m_s = +1/2$ denoted $|\uparrow\rangle$ and the other with $m_s = -1/2$ denoted $|\downarrow\rangle$. A general spin state of an electron is then given in a two-dimensional Hilbert space by a superposition of "spin up" and "spin down" states,

$$|\psi\rangle = \alpha|\uparrow\rangle + \beta|\downarrow\rangle \ , \tag{1.2}$$

where $\alpha$ and $\beta$ are complex numbers satisfying the normalization condition $|\alpha|^2 + |\beta|^2 = 1$.

In the "spin up" and "spin down" basis, it is convenient to represent the operators $S_\alpha$ in matrix form,

$$S_x = \frac{\hbar}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad S_y = \frac{\hbar}{2}\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad S_z = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

In this representation, the vector $(\alpha, \beta)^T$ represents the state given by Eq. (1.2), where $T$ indicates transposition.

The above matrices without the factor of $\hbar/2$ are known as Pauli matrices, and the vector $\mathbf{S} = (S_x, S_y, S_z)$ is the spin operator for the electron spin-1/2.

Note that there are four degrees of freedom in the two complex coefficients $\alpha$ and $\beta$ in Eq. (1.2). However, the normalization requirement removes one of these degrees of freedom, and another one is an overall phase that can be ignored as it cancels in matrix elements whenever an expectation value of an observable is calculated. Thus there are only two degrees of freedom that we care about, and Eq. (1.2) can be rewritten in the form

$$|\psi\rangle = \cos\frac{\theta}{2}|\uparrow\rangle + e^{i\phi}\sin\frac{\theta}{2}|\downarrow\rangle \ . \tag{1.3}$$

The two parameters $\theta$ and $\phi$ can be thought of as the polar and azimuthal angles defining a point on a sphere. This is known as the Bloch sphere, shown in Figure 1.1, and turns out to be a very useful way of picturing a spin 1/2 or any other two-level quantum system. The usefulness of this picture can be seen by looking at the expectation values of the spin in the $x$, $y$, and $z$ directions. Using the matrix forms of the $S_\alpha$ operators given above, it is easy to show that the corresponding expectation values are

$$\langle S_x\rangle = \frac{\hbar}{2}\cos\phi\sin\theta \quad \langle S_y\rangle = \frac{\hbar}{2}\sin\phi\sin\theta \quad \langle S_z\rangle = \frac{\hbar}{2}\cos\theta \ . \tag{1.4}$$

These expectation values are equivalent to the $x$, $y$, and $z$ components of the Bloch vector, as shown in Figure 1.1. Therefore, it is correct in some sense to think of the spin as actually "pointing" along the vector on the Bloch sphere. This one-to-one correspondence between the quantum state in a two-dimensional Hilbert space and the intuitive picture of a classical angular momentum vector in Euclidean space is apparently just a coincidence. Note, however, that the corresponding groups acting on the spin in these spaces, SU(2) and SO(3), are not isomorphic [1], giving room for purely quantum phase effects not captured in the Bloch

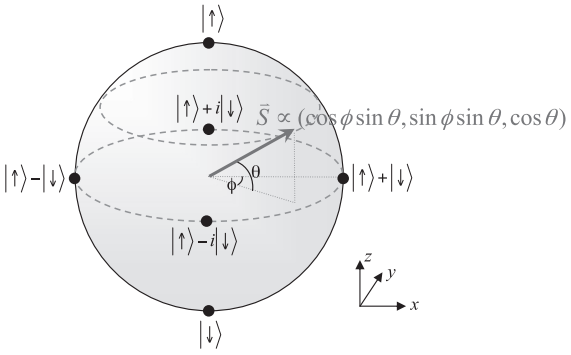



**Fig. 1.1** The Bloch sphere. The vectors pointing to the north and south poles of the Bloch sphere represent the "up" and "down" eigenstates, with the rest of the sphere representing superpositions of "up" and "down".

sphere picture. For spins larger than 1/2 it is harder to think of a corresponding visualization in real space. But since here we are typically interested in electron spins, the Bloch sphere provides a useful and intuitive way of thinking about quantum spin states.

A spin in a magnetic field **B** has a contribution to its energy from the Zeeman Hamiltonian,

$$H_Z = \frac{g\mu_B}{\hbar}\mathbf{B}\cdot\mathbf{S}\,, \tag{1.5}$$

where $\mu_B = 9.274\times10^{24}$ J/T is the Bohr magneton. For an electron in vacuum, the electron gyromagnetic factor or *g*-factor is approximately $g = 2$. However, this is not a universal property. The spin–orbit interaction modifies this quantity, in some crystals even up to an extent that its sign is reversed. For example, electrons in the conduction band of GaAs have $g = -0.44$.

The result of the Zeeman effect on an electron spin is clearly seen by choosing the $z$ axis to be along the magnetic field, leading to

$$H_Z = \frac{1}{2}g\mu_B B_z \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \tag{1.6}$$

The spin eigenstates $|\uparrow\rangle$ and $|\downarrow\rangle$ are split by the Zeeman splitting $\Delta E = g\mu_B B_z$. If the spin is not in an eigenstate, then it evolves in time, depending on the Zeeman splitting. For a spin in the state given by Eq. (1.3) at $t = 0$, the state evolves according to (again, ignoring the overall phase)

$$|\psi(t)\rangle = \cos\frac{\theta}{2}|\uparrow\rangle + e^{i(\omega_L t+\phi)}\sin\frac{\theta}{2}|\downarrow\rangle\,, \tag{1.7}$$

where $\hbar\omega_L = g\mu_B B_z$ is the Zeeman splitting. The angular frequency $\omega_L$ is known as the Larmor frequency. In the Bloch sphere picture, this corresponds to the spin vector precessing about the $z$ axis at the Larmor frequency,

$$\mathbf{S}(t) = (\cos(\omega_L t+\phi)\sin\theta\,, \sin(\omega_L t+\phi)\sin\theta\,, \cos\theta)\,. \tag{1.8}$$

This phenomenon is referred to as Larmor precession.

If perfectly isolated from the environment, a spin in a static magnetic field would obey the dynamics described above forever. In reality, there are a number of effects that damp the evolution in time of an electron spin in a semiconductor. These effects can be divided into two categories: those that randomize the relative phase $\phi$, and those that affect $\theta$ in Eq. (1.7). The randomization of $\theta$ is referred to as longitudinal spin relaxation, and is characterized by a time $T_1$. The loss of the relative phase information $\phi$ is referred to as transverse spin decoherence, occurring in time $T_2$. To illustrate these two time scales, we can say that a spin prepared in the excited state will relax into equilibrium on the time scale $T_1$. A spin that precesses in the plane of the equator of the Bloch sphere, as for $\theta = \pi/2$ in Eq. (1.7), will be distributed randomly on the equator after the characteristic time $T_2$.

This type of damping of the Larmor precession is taken into account in the Bloch equations,

$$\dot{\mathbf{S}}(t) = \mathbf{S} \times \mathbf{h} - R(\mathbf{S} - \mathbf{S}_\infty) \,. \tag{1.9}$$

Here, the first term on the right-hand side describes the precession of the spin components due to a magnetic field $\mathbf{B}$ along $z$, contained in $\mathbf{h} = (0, 0, \omega_\mathrm{L})$, where $\omega_\mathrm{L}$ is again the Larmor frequency. The second term describes relaxation and decoherence of $\mathbf{S}$ towards the equilibrium spin polarization $\mathbf{S}_\infty = (0, 0, \tilde{S})$, which occurs due to

$$R = \begin{pmatrix} 1/T_2 & 0 & 0 \\ 0 & 1/T_2 & 0 \\ 0 & 0 & 1/T_1 \end{pmatrix}. \tag{1.10}$$

In this description it is intuitively clear that the decoherence time $T_2$ is called the transverse spin lifetime, as it acts on the transverse spin components, $S_x$ and $S_y$. The relaxation time $T_1$, in turn, affects the $z$ component and is therefore called the longitudinal spin lifetime. The single-spin Bloch equation can be written in the more compact form

$$\dot{\mathbf{S}}(t) = -\Omega\,(\mathbf{S} - \mathbf{S}_\infty) \,, \tag{1.11}$$

since $\mathbf{h} \times \mathbf{S}_\infty = 0$. The solution of Eq. (1.9) is now just given by

$$\mathbf{S}(t) = \mathrm{e}^{-\Omega t}\mathbf{S}(0) + (1 - \mathrm{e}^{-\Omega t})\mathbf{S}_\infty \,, \tag{1.12}$$

with the components

$$S_x(t) = S_x(0)\mathrm{e}^{-t/T_2}\cos(\omega_\mathrm{L} t) + S_y(0)\mathrm{e}^{-t/T_2}\sin(\omega_\mathrm{L} t) \,, \tag{1.13}$$

$$S_y(t) = -S_x(0)\mathrm{e}^{-t/T_2}\sin(\omega_\mathrm{L} t) + S_y(0)\mathrm{e}^{-t/T_2}\cos(\omega_\mathrm{L} t) \,, \tag{1.14}$$

$$S_z(t) = S_z(0)\mathrm{e}^{-t/T_1} + \tilde{S}\left(1 - \mathrm{e}^{-t/T_1}\right). \tag{1.15}$$

In the above solution of the Bloch equations we can nicely see the decay of the precession amplitude with the characteristic time $T_2$ and the relaxation into an equilibrium spin polarization $\tilde{S}$ along $z$ with the characteristic time $T_1$.

As a caveat we would like to mention that not all baths that damp the evolution of a spin lead to nice exponential decays, as implicitly assumed by the above characteristic decay times $T_1$ and $T_2$. For example, the bath of nuclear spins in a quantum dot can lead to a power-law decay of electron spin coherence. It has also been observed that if, for example, the bath consists of only relatively few electron spins, the coherence of a central spin (in one particular case the spin of the so-called nitrogen vacancy center in diamond [2]) can also decay according to a power law rather than an exponential law.

## 1.3 Quantum Dots

For many practical applications of spins – for example to realize a quantum bit – solid state implementations are an attractive option. When electrons or holes – the mobile carriers of spin in semiconductors – are confined within a tiny structure with one or more dimensions smaller than the extent of the bulk wavefunctions, the electronic properties are drastically modified. When this confinement is along all three spatial directions, a "quantum dot" forms in which, like in a particle-in-a-box, as shown in Figure 1.2a, the energy levels of carriers are quantized. The band edges along one of the three spatial directions with resulting discrete energy levels for electrons and holes are depicted in Figure 1.2b. These energy levels can, following Pauli's exclusion principle, hold two electrons or holes of opposite spin direction. These "orbital states" can be filled sequentially starting from the lowest levels, the ground state of the quantum dot for each carrier species. Atomic physics holds an equivalent of this shell filling known as "Hund's rule". Due to these remarkable analogies to real atoms quantum dots are often referred to as their "artificial" counterparts [3]. Instead of continuous bands of conduction and valence band states, the energy eigenstates are now spatially localized within the dot, and separated by an energy that increases with increasing confinement.

When the temperature ($T$) is low enough such that $k_B T$ is smaller than the quantum dot energy level spacing ($k_B$ is Boltzmann's constant), the quantized nature of the energy levels becomes apparent. For temperatures around 4 K, this requires a QD size of the order of 100 nm, consisting of $\sim 10^5$ atoms. In this regime, quantum dots will exhibit an atom-like spectrum of absorption and emission lines. Figure 1.3 shows a comparison of the emission lines of helium atoms and a semiconductor quantum dot. This correspondence between atoms and quantum dots provides a useful analogy, and the physics of quantum dots can be understood by borrowing ideas and concepts from atomic physics. For example, the optical pumping and control of quantum dot states has been demonstrated using the same tech-

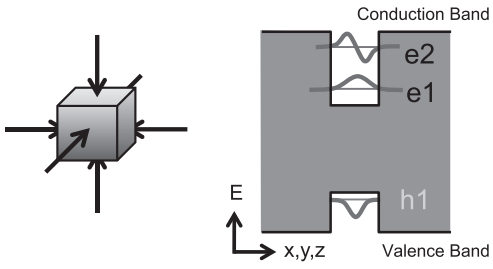
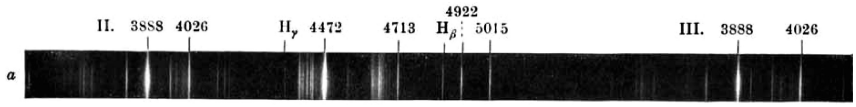
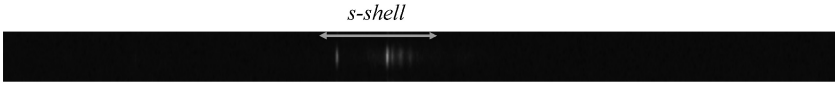



**Fig. 1.2** (a) Schematic of a "quantum dot" in which carriers are confined in all three spatial direction in an area smaller than the de Broglie wavelength of the particle. (b) The confining potential, energy levels, and wavefunctions in a simple particle-in-a-box picture are illustrated for one spatial direction.




(a) Helium–"real atom"




(b) Quantum dot–"artifical atom"

**Fig. 1.3** Comparison of a part of the Helium atom spectrum (a) recorded on a photographic plate and a quantum dot spectrum (b) recorded using a CCD detector array. Figure reprinted with permission from [4]. Copyright by Wiley-VCH Verlag GmbH & Co. KGaA.

niques that were developed for controlling atomic states. On the other hand, the study of quantum dots offers much more flexibility than atomic systems. Many properties of quantum dots are tunable, including their size, shape, and material, which gives us a large degree of control. Even "artificial molecules" in which the interaction between two quantum dots can be switched "on" and "off" can be realized.

### 1.3.1 Spin-Based Quantum Information Processing with Artificial Atoms

In a classical computer, information is stored and processed in bits, each of which can take on one of two logic values. Once the values "0" and "1" have been assigned to the two eigenstates of a two-level system, such as a spin 1/2, the quantum mechanical spin dynamics can be viewed as the processing of information. A peculiarity of this information is that the binary values of a single bit can be brought into a coherent superposition. A bit with this property is called a quantum bit or qubit. According to the postulates of quantum mechanics, when a qubit is measured, it is always projected into one of its eigenstates, providing, for example a classical binary output after the end of a computation. In general, exploiting such uniquely quantum effects in spins or other two-level systems via unitary operations goes by the name of quantum information processing. This field can be divided into two categories: quantum computing, and quantum communication. These topics would already fill more than the space provided in this book. We only mention a few ideas here and refer the reader to the literature for more details.

Before touching on these two developments of quantum information processing, let us briefly consider a few particularities of qubits, namely, coherence and entanglement. We have seen above that coherence is basically the stability of the relative phase $\phi$ in Eq. (1.7) between the two eigenstates. Preservation of coherence is obviously a necessary condition for an undisturbed quantum computation. The

decoherence time therefore imposes a limit on the minimal speed required for successful qubit operations. We already mentioned earlier that certain quantum effects are not captured in a semiclassical framework. A particularly important quantum property without classical counterpart is the entanglement of quantum states. Two spins are entangled if their total wavefunction cannot be written as a direct product of two single-spin states, such as $|\psi_1\rangle_1 \otimes |\psi_2\rangle_2$. Probably the most famous representatives of entangled states are the spin singlet,

$$|S\rangle = \frac{1}{\sqrt{2}} \left(|\downarrow\rangle_1|\uparrow\rangle_2 - |\uparrow\rangle_1|\downarrow\rangle_2\right) \tag{1.16}$$

and the spin triplet with zero spin projection along the quantization axis $z$,

$$|T_0\rangle = \frac{1}{\sqrt{2}} \left(|\downarrow\rangle_1|\uparrow\rangle_2 + |\uparrow\rangle_1|\downarrow\rangle_2\right). \tag{1.17}$$

Clearly, the remaining two triplet states, $|T_+\rangle = |\uparrow\rangle_1|\uparrow\rangle_2$ and $|T_-\rangle = |\downarrow\rangle_1|\downarrow\rangle_2$ factorize and are not entangled.

Quantum computing exploits the additional computational possibilities due to quantum mechanical complexity and parallelism in certain algorithms [5]. A famous example is the quantum algorithm by Peter Shor for the prime factorization of integers, which provides an enormous speed-up potential when factorizing large numbers. The crucial difference here lies in different scaling, as the time needed to factorize an integer on a classical computer grows exponentially with the number of digits $\log N$, while with Shor's algorithm it only scales polynomially [6]. The second famous algorithm is Lov Grover's quantum algorithm for search in an unstructured database [7].

David DiVincenzo has formulated criteria that need to be satisfied for quantum computation [8]. First, a suitable realization of a qubit must be found, in which information can be written, manipulated, and read out. Then, a register of qubits needs to be initialized at the beginning of a computation. The qubits must be sufficiently isolated from the environment to provide long enough decoherence times. In order to process quantum information, gate operations must be implemented. This requires high-precision control of single-qubit rotations and of switchable two-qubit interactions. It has been shown that single- and two-qubit operations are sufficient to implement any computation, that is, they form a universal set of gates [9, 10]. Finally the qubit register must be read out. The scalability is an additional criterion that needs to be met in practical implementations.

An electron spin in a quantum dot is a popular candidate for a qubit, since it is a natural two-state system. Electron spins in semiconductors have received much attention for quantum information applications because (i) semiconductor processing technology should make the scaling to large systems easier, (ii) electron spins in semiconductors have been found to have long coherence times relative to the expected times for gate operations, and (iii) spins and charge excitations can be initialized, addressed, manipulated, and read *both* by electrical and optical means. In recent years, a number of schemes have been demonstrated for achieving the

requirements of state initialization, readout, and control for spin qubits; this is further discussed in later chapters. Nevertheless, there is still a long way to go before these elements can be put together in a functioning quantum computer. For a review on the recent status of spin-based quantum computing, see, for example, Cerletti *et al*. [11].

Quantum communication involves the transmission of quantum information from one place to another. This has applications, for example, in secure communication (cryptography), in teleportation of quantum states, and in superdense coding [12]. Cryptography involves sharing a secret key between two parties, such that the communication can be executed via a traditional communication channel, protected by a safe encryption. Quantum mechanics helps in distributing a secret key with the very issue we have when measuring a state: we project it into an eigenstate. For example, if polarization encoded photons are measured on the way by a third party in a different basis than the encryption scheme is using (which is the most probable case), the photon polarizations received in the end will just be randomized, which is detectable. Other schemes that have recently been implemented with quantum dot single-photon sources [13] involve the encoding of a key in a stream of single photons. Here, a third party would obviously quench the flow of information with a beam splitter attack, when detecting photons in a photon counter at the third party's site, which is also easily detectable in a communication scheme using control sequences. Quantum teleportation, in turn, consists of transmitting the quantum state of an object, for example a photon or an atom, faithfully onto a second object by performing a clever set of local measurements and by sharing a pair of entangled particles, for example two photons. Quantum teleportation has also been implemented using a single photon source provided by a quantum dot [14]. At this point we do not delve more deeply into these particular realizations.

Quantum communication necessitates a "flying qubit" – a carrier of quantum information that can be moved from place to place. Though spins and other qubit candidates such as single atoms can be moved over micron-scale distances within their coherence time, the only practical qubits for long-distance quantum communication are photon-based. Photons make ideal carriers of quantum information because they travel fast, and they have very long coherence times. The flip side to the long coherence time is that photons interact very weakly with each other – and a strong controllable interaction is a desired feature for quantum information processing. This leads us to consider a hybrid system with stationary qubits used for quantum information processing at either end, and flying qubits communicating faithfully between the two. This requires a way of converting stationary qubits to flying qubits. Fortunately, spins in optically active quantum dots couple to photons in a variety of ways, making this an intriguing platform for a potential hybrid quantum computing/communication system. In this book we want to introduce not only various ways how quantum information can be transferred between the photonic and spin domains but also how the electromagnetic wave nature of light can be used to coherently initialize, manipulate, and read-out the spin of an electron confined in a quantum dot.

### 1.3.2 Optically Active Quantum Dots

For a quantum dot to be considered "optically active" the interaction between light and carriers in the quantum dot must be sufficiently strong so as to make it useful for scientific investigations or technological applications. The definition of an "optically active" quantum dot is not entirely strict since virtually all quantum dots have some measurable interaction with electromagnetic radiation in the optical domain, that is, light. Nonetheless, here we will concern ourselves with those types of quantum dots that are primarily investigated by optical means. The confinement potential of these dots is typically such that they can hold *both* electrons and holes giving rise to a particularly strong interaction with light.

The primary way that light interacts with an optically active quantum dot is through transitions between the valence band and conduction band states in the quantum dot or in the surrounding semiconductor material. When light with a sufficiently small wavelength is incident on a quantum dot, transitions can be driven that serve to excite electrons from the valence band to the conduction band. Likewise, the inverse process occurs when an electron relaxes from a conduction band state to an unoccupied valence band state – light is emitted. This gives two measurable quantities for investigating the properties of a quantum dot: optical absorption and luminescence.

Further optical properties can also be observed and exploited in optically active quantum dots. Off-resonant interactions, such as the Faraday effect or Raman transitions are particularly useful for spin readout and manipulation, respectively.

### 1.3.3 "Natural" Quantum Dots

A very simple quantum dot system, which moreover exhibits an extremely high optical quality, are so-called "natural" quantum dots. The name originates from the fact that these dots form naturally in thin quantum wells. Such quantum wells are fabricated by deposition of two-dimensional films of semiconductor materials with different bandgaps. For example, if a thin layer (with a width $d \sim 7\,\mathrm{nm}$) of gallium arsenide (GaAs) is sandwiched between barriers made of aluminum gallium arsenide (AlGaAs) a potential well for both electrons and holes is formed perpendicular to the layers as shown in Figure 1.2b. The energy of the lowest level with respect to the band edges of the quantum well material (GaAs) depends on the width $d$ of the well, that is, the GaAs layer. In the simplest approximation of a square well potential with infinite barriers we would expect a $\propto d^{-2}$ dependence of the ground state energy and, therefore, a wider well has a deeper lying ground state. As a matter of fact, semiconductor quantum wells are not always perfectly flat but exhibit monolayer-high steps. This situation is sketched schematically in Figure 1.4a. If the areas in which the well is thicker have the appropriate size (typically $\sim 100\,\mathrm{nm}$) monolayer fluctuations form quantum dots, which are therefore also sometimes referred to as "interface fluctuation quantum dots" (IFQD). These

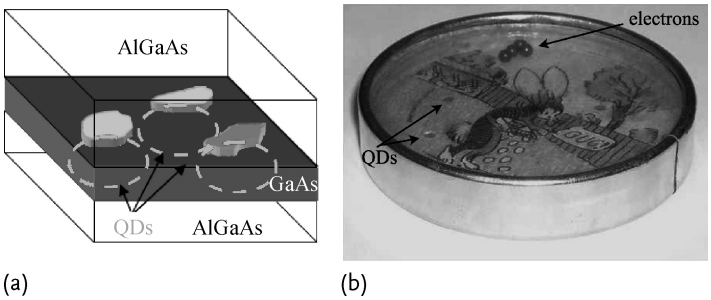



**Fig. 1.4** (a) Illustration of interface fluctuation quantum dots: Quantum dots form at localized monolayer fluctuations of well thickness. (b) Toy with little balls and dimples.

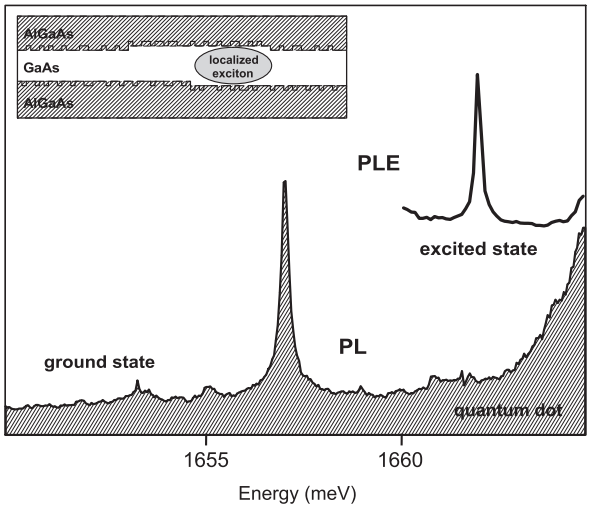



**Fig. 1.5** Photoluminescence and PL excitation spectra of a "natural" quantum dot. The quantum dot geometry is shown schematically as an inset. Figure courtesy of W. Heller.

interface fluctuation quantum dots act like the little dimples in the toy shown in Figure 1.4b where little balls (representing electrons or holes) can be caught[1)].

In the case of a GaAs-AlGaAs quantum dot the confined electrons and holes can recombine by emitting light, which is called photoluminescence (PL) if the electrons and holes were previously generated by light, for example by a laser. A typical example of a PL spectrum and a schematic of the quantum dot geometry is shown in Figure 1.5. This quantum dot shows a sharp atom-like PL line at and energy of 1657 meV. The observed linewidth is determined entirely by the resolution of

1) Since this toy does not know about spins, each dimple can only hold one ball. For deeper dimples higher occupancy states can be achieved.

the experimental apparatus, which underlines the high optical quality of the dot. In this figure the excited state absorption of the same quantum dot is also shown. It is measured in a PL excitation (PLE) experiment in which the intensity of the ground state PL signal is recorded as a function of the excitation laser energy. In such an experiment a high signal is only observed at laser energies at which the QD absorbs, which in this example occurs at an energy 5 meV higher than the ground state. Furthermore, on the right, at even higher energy, the onset of emission from the quantum well in which the dot is formed (peak at 1670 meV) can be seen.

The preceding discussions of quantum dots and of electron spin dynamics provide an introduction to these topics that will be delved into more deeply throughout this book. Chapters 3, 5, and 6 will treat the physics of spins in quantum dots in much more detail. Moreover, Chapters 7 and 8 describe a number of experimental observations of these phenomena. But first we will continue in Chapter 2 with different physical realizations of QDs. We will start by explaining the basics of semiconductor heteroepitaxy, which is the underlying method for the fabrication of embedded QD structures like "natural" dots, which we introduced in the previous section. We will also discuss the other prominent example of epitaxial quantum dots, the so-called self-assembled QDs after which we continue with a different technique to fabricate nanometer-size colloidal quantum dots.