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Introduction

The interaction between matter and radiation has fascinated physicists for a long time. On the material side, the most detailed investigations of these processes concentrate on atoms, the basic constituents of matter. The radiation that is involved in these processes is primarily light, i.e., radiation whose wavelength is in the range of a few tenths of a micron to a few microns. Under today's laboratory conditions, this radiation is generally produced by a laser. This introduction outlines our picture of these constituents and presents some of the concepts and models that we will use throughout this book.

1.1 Atoms

1.1.1 Historical

Early models: atoms as building blocks

The term "atom" was coined by the Greek philosopher Democritus of Abdera (460–370 B.C.), who tried to reconcile change with eternal existence. His solution to this dilemma was that matter was not indefinitely divisible, but consisted of structureless building blocks that he called atoms. According to Democritus and other proponents of this idea, the diverse aspects of matter, as we know it, are a result of different arrangements of the same building blocks in empty space (Melsen 1957; Simonyi 1990). The most important opponent of this theory was Aristotle (384–322 B.C.), and his great influence is probably the main reason that the atomic hypothesis was not widely accepted, but lay dormant for two thousand years. It reappeared only in the eighteenth century, when the emerging experimental science found convincing evidence that matter does indeed consist of elementary building blocks. Chemists discovered that elements react in constant proportions with each other, and that these proportions are related by fractions of small integer numbers. Aristotle's teachings could not explain these experimental findings, but the atomic hypothesis

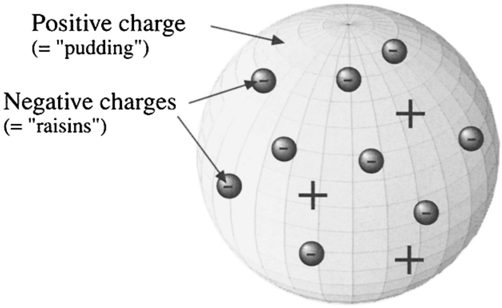

Figure 1.1. Sir Joseph Thomson's model of the atom as a "pudding with raisins."

gave a convincing explanation. During the nineteenth century, most types of atoms were discovered and classified in the periodic system of the elements. The atomic hypothesis also gained recognition as it could explain diverse findings, like the variation of the pressure of gases with temperature or Brownian motion. Although nobody had yet seen an atom, the atomic hypothesis was already quite well established by the end of the nineteenth century.

Also during the nineteenth century, pieces of evidence emerged that indicated that atoms were not the unchanging and structureless ultimate constituents of matter that Democritus had postulated. The discovery of radioactivity showed that they were not eternal, but subject to decay and change. In lightning, cathode rays, and in electrolysis, particles showed up that apparently were smaller constituents of the atom. It soon turned out that these negatively charged particles, which were called electrons, carried only a small part of the atomic mass. Thus, most of the mass had to be concentrated in the remaining part. To make the atom electrically neutral, this remaining part had to carry a positive electric charge. A model for the atomic structure that was quite popular at this time was Sir Joseph Thomson's (1856–1940) "pudding with raisins" that considered the electron as the "raisins" in the much larger, positively charged "pudding" (see Figure 1.1).

Internal structure

It therefore came as a big surprise when the scattering experiments which Ernest Rutherford (1871–1937) performed in the years 1911–1913, showed that the positive charge was concentrated in a region many orders of magnitude smaller than the volume of the whole atom. Although a model of electrons orbiting the positively charged nucleus could explain the apparent size of the atoms, it was in direct contradiction to the newly established field of

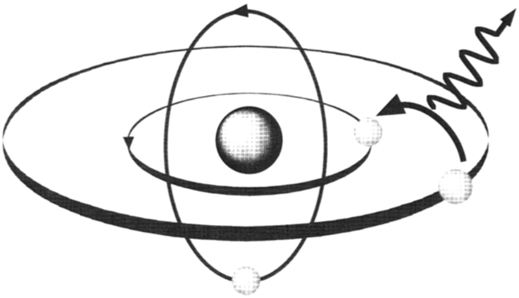

Figure 1.2. Bohr's model of the atom.

electrodynamics: Accelerated charges were known to radiate. Electrons orbiting a nucleus should therefore radiate and lose energy. This would cause them to fall into the nucleus on a short timescale. This prediction of classical electrodynamics was evidently in contradiction to the experimental fact of the stability of atoms and constituted one of the famous inconsistencies that were later on resolved by the quantum theory. Another important hint came from spectroscopy, where Joseph von Fraunhofer (1787–1826) had discovered dark lines in the solar spectrum and Michael Faraday (1791–1867) had shown that magnetic fields could influence the optical properties of various substances.

Niels Bohr's (1885–1962) model of the atom (see Figure 1.2) tackled these problems by postulating stationary states in which the electron did not evolve in time. Energy exchange through absorption or emission of light would be associated with discrete jumps of the electron between different stationary states. A few years later, justification for these assumptions was provided by the quantum mechanics of Erwin Schrödinger (1887–1961) and Werner Karl Heisenberg (1901–1976).

1.1.2 Quantum mechanics

Energy levels

The quantum mechanical picture, still relevant today, tells us that the electrons do not orbit the nucleus in planetlike trajectories, but in spatially extended "orbitals." Depending on the energy of the system, the atom can be in one of an infinite number of stationary states, which are represented mathematically as the eigenstates of the Hamiltonian of the system (Weissbluth 1978).

Figure 1.3 shows the usual representation of the lowest of these orbitals for the hydrogen atom. The lowest state is spherically symmetric, whereas higher lying states have lower symmetry. This description implies that the atom has

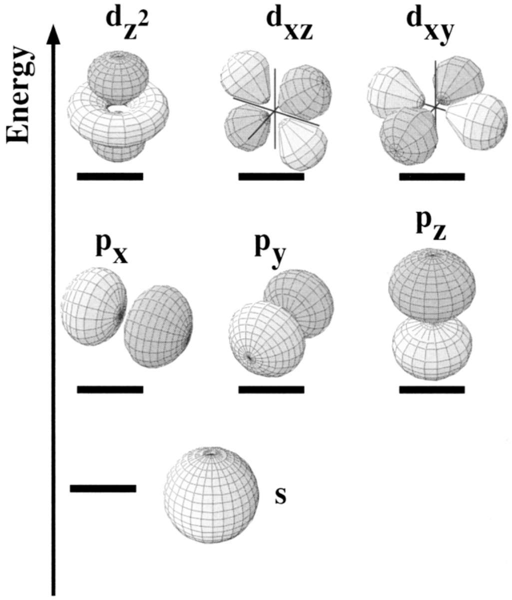

Figure 1.3. Hydrogenlike orbitals.

only a single electron, but even for atoms with a large number of electrons, it has turned out to be a useful model.

Since the main subject of this book is not the structure of the atom, but its interaction with radiation, we do not describe the atom in detail. In particular, we will not have to consider the complete set of energy levels. The most important ones are the energetically highest of the occupied orbitals and the lowest unoccupied orbital, since absorption and emission of light are often accompanied by transitions between these two levels.

Sublevels

As Figure 1.3 indicates, the energy level structure of atoms and molecules consists in most cases of multiplets of degenerate or near-degenerate states. We refer to these states as substates or sublevels. Although their energy differs very little, other physical properties, in particular the angular momentum, exhibit significant variations between them. If a measurement is performed on such a system without distinguishing the various sublevels, the result is a weighted average of the contributions from all states. This is the typical situation in many experiments that probe material properties with electromagnetic fields. Since the interaction between the material and the probing radiation is strongly enhanced when the photon energy is close to an allowed electronic transition, distinguishing the different states is in most cases achieved through their energy. This method cannot easily distinguish between states that are energetically degenerate, however.

Historically, the energy level structure of atomic systems has been investigated primarily with optical spectroscopy. From this point of view, the energy differences are usually classified into electronic transitions, fine structure, hyperfine structure, and Zeeman multiplets. Whereas the fine structure and in many cases the hyperfine structure of these systems can be resolved by optical spectroscopy, the Zeeman level splitting is considerably smaller than the natural linewidth of the optical transitions unless the applied magnetic field is significantly stronger than that of the earth.

In most experimental situations, one does not deal with individual atoms, but with a large number, typically $\geq 10^9$. The observable properties depend then on the population of the various levels, i.e., on the number of atoms occupying a particular eigenstate of the Hamiltonian. According to equilibrium statistical mechanics, these populations depend only on the energy of the states and the temperature of the ensemble. For degenerate states, they are identical, and a measurement performed on such a system averages over all degenerate sublevels with equal weights.

As long as the system is in internal equilibrium, the influence of the sublevel structure on the macroscopically observable properties is small and does not depend on the details of the experimental situation. If the interaction with external fields drives the system far from internal equilibrium, however, the distribution of the sublevel populations may become nonthermal, as in the right-hand side of Figure 1.4. The population of each sublevel, and therefore the weighting coefficients in the averaging process, thus depend on the details of the experimental parameters and usually also on the history of the system. In addition, the external fields may put the atoms not into a single eigenstate of the Hamiltonian, but into superposition states of two or more eigenstates. These superposition states can have physical properties qualitatively different from either of the constituent states, e.g., a nonvanishing electric dipole moment. Without a detailed knowledge of the internal state of the microscopic system, it is difficult to make predictions about its macroscopic properties.

This situation represents the main theme of this book, which concentrates on methods for obtaining detailed and precise information about the stationary properties and on the dynamics of the internal degrees of freedom of atomic multilevel systems. In a somewhat different context, the investigation of the sublevel structure is also the subject of magnetic resonance spectroscopy,

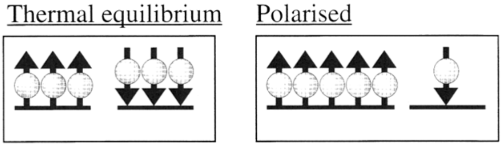

Figure 1.4. Change of overall properties by polarisation of sublevels.

where radio frequency fields are used to induce and observe transitions between different substates. This field, which was the first to introduce coherent methods to spectroscopy, has had a large impact on laser spectroscopy and we will use many of its results in the discussion of optical experiments.

1.2 Light

1.2.1 The quantum theory of light

Introduction

The interaction of light with matter, in particular blackbody radiation and the photoelectric effect, were among the major experimental discoveries that initiated the development of quantum mechanics. The new theory quickly allowed a better description of the material world, in particular atoms. Almost in parallel, P. A. M. Dirac (1902–1984), Heisenberg and Wolfgang Pauli (1900–1958) formulated in 1928 a quantum mechanical theory of light, which was later extended by Shinichiro Tomonaga (1906–1979), Julian Schwinger (1918–1994) and Richard Feynman (1918–1988) (Dyson 1949) and others. This theory, known as quantum electrodynamics, or QED, is today among those physical theories that have been most thoroughly tested experimentally. In all respects, these tests have confirmed the predictions of the theory and today it has an extraordinarily good status – both experimentally and theoretically.

Nevertheless, most theoretical descriptions of light and its interaction with matter describe the radiation field classically, using Maxwell's equations (James Clerk Maxwell, 1831–1879), as we will for most of this book. In a few cases, however, particularly when the conservation laws for energy, momentum and angular momentum are involved, the quantum nature of the light is important. For this reason, we include here a brief, qualitative outline of the main features of the quantum mechanical description of light.

Modes

Even the quantum mechanical theory uses Maxwell's equations to describe the propagation of the light. Quantum mechanical aspects are important only

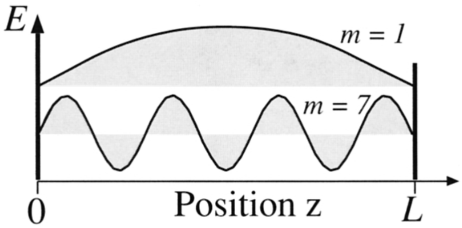

Figure 1.5. One-dimensional resonator. The two curves represent the field between two conducting surfaces for two different modes.

for the interaction of light with matter and when statistical features become important. The usual formulation of quantum electrodynamics expands the radiation field in the eigenmodes of optical resonators with fixed frequency, polarisation, and field distribution. Although the selection of modes is in principle arbitrary, the most popular expansion uses the eigenmodes of rectangular boxes with perfectly conducting walls.

Figure 1.5 illustrates this expansion for the one-dimensional case: The mirrors at positions 0, L impose the boundary conditions that the field vanish at those positions. The eigenmodes have field distributions that vary with $\sin(m\pi z/L)$, where m is the mode number, z the spatial variable, and L the separation of the two mirrors. The spatiotemporal variation of the field is, in complex notation,

$$E_m = \sin\left(m\,\pi\frac{z}{L}\right)e^{i\omega_m t} \qquad (m = 1, 2, \ldots) \tag{1.1}$$

where the angular frequency is

$$\omega_m = m\frac{\pi\,c}{L} \tag{1.2}$$

The dynamics of each mode of the field is governed by the Hamiltonian

$$\mathcal{H}_m = \frac{1}{2}(\omega_m^2 q^2 + p^2) \tag{1.3}$$

which is equivalent to that of a harmonic oscillator with unit mass. Here and through most of the book, we use frequency units for the energy, which correspond to units in which $\hbar = 1$. This not only allows a more compact notation, but also emphasises that energy differences will always be measured in

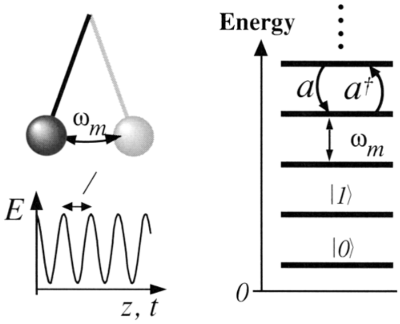

Figure 1.6. Energy levels of the harmonic oscillator, which may represent a pendulum or a single mode of the radiation field.

the form of frequencies. In the harmonic oscillator case, the variables p and q correspond to momentum and position of the harmonic oscillator and in the electromagnetic case to the electric and magnetic components of the field. In the mechanical harmonic oscillator, the energy oscillates between kinetic and potential energy. In the case of the radiation field, it oscillates between the electric and the magnetic fields.

The eigenvalues of the harmonic oscillator

$$\mathcal{E}_{m,n} = \left(n + \frac{1}{2}\right) \omega_m \qquad (n = 0, 1, 2, \ldots) \tag{1.4}$$

increase in units of the oscillator frequency ω_m. The corresponding eigenstates are commonly written as $|n\rangle$, as indicated in Figure 1.6.

In contrast to the classical oscillator, the energy of the lowest level is not zero but $\omega_m/2$. This zero-point energy is an important distinction between the quantum mechanical and the classical systems. It cannot be extracted from the system but nevertheless has observable consequences. It is responsible, e.g., for spontaneous emission (Weisskopf and Wigner 1930), the Lamb shift (Lamb and Retherford 1947), the electron g factor, and the shot noise in the detection of light (Walls 1979).

Photons

The operators p and q are closely related to the electric and magnetic fields in the mode

$$E(z,t) = 2\,q\,\sqrt{\frac{\hbar\omega_m}{\epsilon_0 V}}\,\sin\!\left(m\,\pi\,\frac{z}{L}\right) \quad H(z,t) = 2\,p\,\sqrt{\frac{\hbar - \omega_m}{\mu_0 V}}\,\cos\!\left(m\,\pi\,\frac{z}{L}\right) \tag{1.5}$$

where V is the resonator volume. In the quantum mechanical analysis, one often uses linear combinations of the operators p and q

$$a = \frac{1}{\sqrt{2\hbar\omega_m}}(\omega_m q + \mathrm{i}p) \qquad a^t = \frac{1}{\sqrt{2\hbar\omega_m}}(\omega_m q - \mathrm{i}p) \tag{1.6}$$

which act on the eigenstates of the Hamiltonian as

$$a\,|n\rangle = \sqrt{n}\,|n-1\rangle \qquad a^\dagger\,|n\rangle = \sqrt{n+1}\,|n+1\rangle \tag{1.7}$$

Their effect on the eigenstate |n⟩ is thus a decrease or increase of the energy by an amount ω_m. These excitations of the field mode may be taken as the constituents of the radiation field and are known as photons. The operators $a^\dagger$ and a create and destroy photons, usually during the interaction with atoms and are referred to as creation and annihilation (or raising and lowering) operators. The relevant properties of the photons include, apart from their energy $\mathscr{E}_\phi = \hbar\omega$, a linear momentum $p_\phi = \hbar k = \mathscr{E}_\phi/c$ and an angular momentum $S_\phi = \hbar$.

Each eigenstate of the Hamiltonian corresponds to a definite number of photons in the field and is therefore known as a number state. The operator

$$a^\dagger\, a\,|n\rangle = n\,|n\rangle \tag{1.8}$$

counts the number of photons in each state; it is known as number operator and may be used to rewrite the Hamiltonian as $\mathscr{H}_m = \omega_m(a^\dagger a + 1/2)$.

Field states

Although the number states $|n\rangle$ are useful to describe some properties of the isolated field, they have an important drawback when it comes to describing the interaction with matter. As an evaluation of the field operator $q = a + a^\dagger$ shows, the expectation value of the field vanishes for number states. This is a consequence of the Heisenberg uncertainty relation: Since the eigenstates of the Hamiltonian have a definite amplitude, their phase, which is the conjugate variable of the amplitude, must be completely uncertain. This contrasts with the classical description of a field mode: For a complete description, we need to specify two parameters, e.g., amplitude and phase, or cosine and sine components of the field. The state of the field is thus a point in a two-dimensional coordinate system, as shown in Figure 1.7.

Quantum mechanically, such a state would violate the uncertainty relation, since the two variables correspond to operators that do not commute with each other. For the number state, the uncertainty relation is satisfied by the complete phase uncertainty. This behaviour makes such a field state unsuitable for

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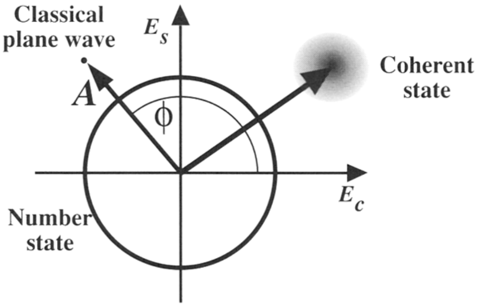

Figure 1.7. Probability distribution for the field of a number state. The two coordinate axes are the cosine and sine components; the polar coordinates A and ϕ represent amplitude and phase of the field.

describing phase-sensitive experiments, such as interference phenomena, and differs markedly from what we expect for a laser field.

The output of a laser is best approximated by a coherent state (Glauber 1963b; Glauber 1963a), which is the closest quantum mechanical approximation to a classical field. It can be expanded in the basis of the number states as

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_n \frac{\alpha^n}{\sqrt{n!}} |n\rangle \tag{1.9}$$

The complex parameter α characterises the state completely; it represents the mean excitation of the field. Coherent states are states with minimum uncertainty, i.e., the uncertainty is the minimum permitted by the Heisenberg relation. In addition, the uncertainty is distributed equally between conjugate variables like amplitude and phase.

As Figure 1.8 shows, the coherent state has a nonvanishing mean field, in contrast to the number state, where the probabilities at positive and negative field are equal, thus cancelling each other. The width of the field distribution is independent of the mean excitation. The relative uncertainty decreases correspondingly with the inverse square root of the mean photon number $1/\sqrt{\bar{n}}$. For large excitations, it is often sufficient to use a classical description that approximates the field with its mean. For most of this book, this approximation will be sufficient and we describe the field classically.

1.2.2 The classical description

Formalism

The corresponding classical description (Born and Wolf 1986) uses the Maxwell equations to describe the electromagnetic field. Here, we introduce