# Introduction to Quantum Optics I

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The preliminary programme for this lecture:

Motivation: experiments in Potsdam and elsewhere

I Interaction between light and atoms (light and matter)

- — relevant observables, statistics
- — the model of a two-level atom, a two-level medium
- — Bloch equations
- — quantum states of one qubit

II Photons – field quantization

- — elementary scheme with a mode expansion
- — states of the radiation field: Fock, coherent, thermal, squeezed; distribution functions in phase space
- — Jaynes-Cummings-Paul model (collapse and revival)
- — about spontaneous emission, quantum noise and vacuum energies

**Outlook** SS 2015: quantum optics II

- — master equations, photodetection
- — beamsplitter, homodyne detection
- — open systems, "system + bath" paradigm
- — quantum theory of the laser and the micromaser
- — correlations and fluctuations, spectral characterization
- — problems of current interest:
  two-photon interference, intensity correlations
  virtual vs real photons, strong coupling

These notes are a merger of previous years and contain more material than was actually delivered in WS 2017/18.

## Motivation

List of experiments, some of them performed at University of Potsdam. Try to answer the question: is this a quantum optics experiment? do we need quantum optics to understand it?

— A laser pulse is sent on a metallic surface and is (partially) absorbed there (see Problem 1.1(iv)).

— Small metallic particles are covered with a layer of quantum dots and deposited on a substrate. The absorption spectrum shows two peaks.

— When photons are absorbed in a semiconductor, they may create excitons that diffuse by hopping through the sample. The excitons can dissociate and generate charge carriers that create a current (solar cell).

— A femto-second short pulse is absorbed in a molecular beam and generates vibrational wavepackets.

— A polymer film is irradiated with an light pattern and develops a deformation that can be measured with a scanning microscope.

— A laser beam is exciting an optical cavity where one mirror is mobile: it is pushed a little due to radiation pressure. By adjusting the frequency of the laser, the motion of the mirror is cooled down.

— An intense laser is focused into a crystalline material and generates pairs of photons (also known as biphotons). The polarizations of the biphoton partners is uncertain, but anti-correlated with certainty: when one partner of the biphoton is polarized horizontally, the other has a vertical polarization and the other way around.

— When UV light is sent onto a metal, it generates free electrons whose energy increases linearly with the light frequency (photoelectric effect, photodetector). In theoretical chemistry and surface physics, this effect is modelled with a classical electromagnetic field inserted into the Schrödinger equation for the metallic electrons.

# Contents

# Chapter 1

# Matter-light interaction

In this chapter, we review some basic physics about polarizable matter and its interaction with light. The focus is on developing simple approximations that describe the coupling to near-resonant light fields. The model of a two-level atom will play an important role.

To begin the lecture, we describe in this chapter the field classically. The arguments can be made, however, with the quantized field as well. We shall give here and there a few formulas without going into the details (to be found in Chapter 2).

## 01 Nov 16: two-level medium

Two-level approximation: focus on energy levels with Bohr frequencies in the 'relevant spectrum' of light. The 'relevant spectrum' is defined by application, for example, by the laser source.

Quantum system with two states $|\mathrm{e}\rangle, |\mathrm{g}\rangle$: observables.

Probabilities $p_a$, projectors $\hat{\pi}_a = |a\rangle\langle a|$ $(a = \mathrm{e}, \mathrm{g})$. In condensed matter: occupation number densities $n_a(x)$. See Sec. 1.8 for details.

Dipole moment $\mathbf{d}$, medium polarisation $\mathbf{P}$, current density $\mathbf{j}$.

Matrix elements of dipole: selection rule $\langle a|\mathbf{d}|a\rangle = \mathbf{0}$. Proof: use definite parity of wave functions $\psi_a(\mathbf{r})$. Off-diagonal = transition dipole matrix element $\langle \mathrm{e}|\mathbf{d}|\mathrm{g}\rangle$.

Electric dipole interaction energy

$$V_{\mathrm{AL}} = -\mathbf{d} \cdot \mathbf{E}(\mathbf{r}_A, t) \tag{1.1}$$

also called 'multipolar coupling'. Long-wavelength approximation: electric field is spatially constant over the spatial extent of the electronic orbitals, evaluate at 'representative position of the atom', $\mathbf{E}(\mathbf{r}_A, t)$. See problem sets for numbers.

Current density 'inside' the electronic orbital

$$\mathbf{j}(x) = \frac{e}{m} \,\mathrm{Re}\,[\psi^*(x)(\mathbf{p} - e\mathbf{A})\psi(x)] \tag{1.2}$$

Conservation of probability / charge density.

Focus on 'total current' = spatial integral $\int \mathrm{d}^3r\, \mathbf{j}(\mathbf{r}, t)$, relevant for 'small atom' (= described as point dipole). Selection rule: diagonal matrix elements

$$\int \mathrm{d}^3r \psi_\mathrm{e}^*(\mathbf{r})\mathbf{p}\psi_\mathrm{e}(\mathbf{r}) = \mathbf{0} \tag{1.3}$$

by same parity argument.

Off-diagonal matrix elements and link to dipole operator

$$\langle \mathrm{e}|(\mathbf{p} - e\mathbf{A})|\mathrm{g}\rangle = \mathrm{i}m\omega_A \langle \mathrm{e}|\mathbf{r}|\mathrm{g}\rangle \tag{1.4}$$

with the two-level Bohr frequency $\hbar\omega_A = E_\mathrm{e} - E_\mathrm{g}$.

See Sec. 1.8: equations of motion for probabilities $p_a$ (rate equations) and their spatial densities. Processes: absorption, stimulated emission, spontaneous decay.

## 1.1 08 Nov 16: QM of two-level systems

Two-level Hamiltonian as matrix: Bohr frequency, atom+field Hamiltonian in dipole approximation (Sec.1.9). Rabi frequency (1.44):

$$\mathbf{d}_{eg} \cdot \mathbf{E}_L(\mathbf{x}_A, t) = -\frac{\hbar\Omega(t)}{2} \tag{1.5}$$

('Paris convention')

Equations of motion – for state (exercise): time-dependent Schrödinger equation.

$$\begin{aligned} \mathrm{i}\partial_t \tilde{c}_e &= -\frac{\omega_A - \omega}{2}\tilde{c}_e + \frac{\Omega(t)}{2}\,\mathrm{e}^{\mathrm{i}\omega t}\tilde{c}_g \\ \mathrm{i}\partial_t \tilde{c}_g &= +\frac{\omega_A - \omega}{2}\tilde{c}_g + \frac{\Omega(t)}{2}\,\mathrm{e}^{-\mathrm{i}\omega t}\tilde{c}_e \end{aligned} \tag{1.6}$$

in a ‘rotating frame’ at $\omega$. Typical choices are $\omega = \omega_A$ (‘interaction picture’) or $= \omega_L$ (‘frame rotating with the laser’).

– for observables: Heisenberg picture. Dipole operator

$$\hat{\mathbf{d}} = \mathbf{d}_{eg}^* \sigma + \mathbf{d}_{eg} \sigma^\dagger = \mathbf{d}_{eg} \sigma_1 \tag{1.7}$$

with transition dipole matrix element (often assumed real) and two-level lowering operator. In the Schrödinger picture, $\sigma = |\mathrm{g}\rangle\langle\mathrm{e}|$.

Python script with the numerical solution of the Schrödinger equation: see moodle.

Check that the following Hamiltonian generates the Schrödinger equation for $\tilde{c}_a$ [Eq.(1.6)] in the frame rotating at the laser frequency: $\omega = \omega_L$, detuning $\Delta = \omega_L - \omega_A$:

$$\tilde{H} = -\frac{\hbar\Delta}{2} (\pi_e - \pi_g) \underbrace{-\mathbf{d}_{eg} \cdot \mathbf{E}(x)}_{= +\hbar\Omega(t)/2} \left( \mathrm{e}^{+\mathrm{i}\omega_L t} \sigma^\dagger + \mathrm{e}^{-\mathrm{i}\omega_L t} \sigma \right) \tag{1.8}$$

## 1.2 15 Nov 16: resonance approximation, pure and mixed states

Rotating frame and resonance approximation (RWA = *rotating wave approximation*, Sec.1.9.3).

Typical questions:
– frequency and time scales (cw vs short pulses)
– envelope approximation (complex Rabi, RWA)
– numerical solutions with/out RWA, comparison
– Rabi oscillations in RWA (analytical: exercise)

Bloch equations: see Eq.(1.13).

## 1.3 22 Nov 16: dynamics in the Bloch sphere

Bloch vector = 3 real components in ‘abstract space’, represented as point (endpoint of vector) in sphere.

Animations with solution to Bloch equations: see Python script on Moodle.

Pure state = two-level wave function with amplitudes $c_g, c_e$. Gives Bloch vector on the sphere, $s^2 = s_1^2 + s_2^2 + s_3^2 = 1$. Geographic names: ‘North pole’ =

excited state ('spin up'), 'South pole' = ground state ('spin down'). On 'equator' = superpositions with equal weight/occupations $p_e = p_g = 1/2$, but definite relative phase.

Observation: to each point on the Bloch sphere corresponds a 'ray' of normalized two-level states that differ by a global phase factor. Since this phase is (usually) not observable, the Bloch representation is closer to the physical observables.

Observables: projection onto 'North-South axis' is the population difference $s_3 = p_e - p_g$. Projection onto equatorial plane gives real and imaginary parts of dipole expectation value: $\langle\sigma\rangle = (s_1 - \mathrm{i}s_2)/2$.

Dynamics:

(1) freely evolving two-level system, no laser = rotation (precession) of Bloch vector around North-South axis. Component $s_3$ is preserved. The rotation frequency is zero (in the rotating frame) if the laser is resonant with the atom. In the original frame, the precession frequency is the atomic frequency $\omega_A$ ('very fast').

(2) with a laser = rotation around a tilted axis. On resonance, the axis lies in the equatorial plane and rotates the Bloch vector from the ground state to the excited state and back. The rotation frequency is then the Rabi frequency. The projection onto the populations gives the Rabi oscillations.

Mixed states: Bloch vectors with length $s < 1$. Cannot be represented as state vectors, only as density matrix

$$\rho = \frac{1}{2}\begin{pmatrix} 1 + s_3 & s_1 - \mathrm{i}s_2 \\ s_1 + \mathrm{i}s_2 & 1 - s_3 \end{pmatrix} = \frac{1}{2}\left(\mathbb{1} + \mathbf{s}\cdot\boldsymbol{\sigma}\right) \tag{1.9}$$

Eigenvalues are $(1 \pm s)/2$, have the physical meaning of a probability. Are positive only when $s \le 1$.

(2) spontaneous decay of excited state = compression of Bloch sphere along the North-South axis and shift downwards so that the ground state is invariant.

(3) dephasing/decoherence of dipole = compression in the equatorial plane towards the North-South axis (from an 'orange' to a 'lemon' shape).

Generic dynamics: a combination of all these. Can lead to a spiralling-in towards a steady state in the lower half of the Bloch sphere. If the laser field is a continuous wave, the final state is typically close to the surface (close to a pure state).

Short laser pulse: a rotation around an axis defined by the complex Rabi frequency $\Omega(t)$ and the detuning $\Delta$. Recall full laser field projected onto atomic transition dipole

$$\mathbf{d}_{eg} \cdot \mathbf{E}_L(t) = -\frac{\hbar}{2} \left( \Omega(t)\, \mathrm{e}^{-\mathrm{i}\omega_L t} + \text{c.c.} \right) \tag{1.10}$$

For not-too-short laser pulses, $\Omega(t)$ is a slowly varying envelope on the scale set by the period $2\pi/\omega_L$.

## 1.4 29 Nov 16: complex Rabi frequency

From resonance approximation:

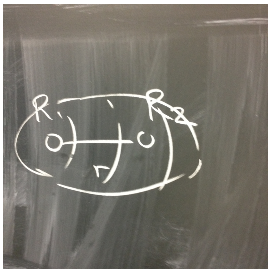
$$\Omega(t)\, \mathrm{e}^{\mathrm{i}\omega_L t} = -\frac{2\mathbf{d}}{\hbar} \cdot \mathbf{E}_L(t) \approx \Omega \tag{1.11}$$

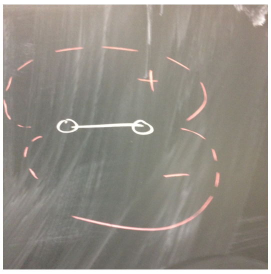
where $\Omega$ is proportional to the complex amplitude of the laser field. The neglected terms oscillate at $2\omega_L$ and provide small-amplitude fast oscillations around the solutions in the resonance approximation. Word from nuclear spin resonance: "rotating wave approximation" (RWA), is often used.

The complex Rabi frequency $\Omega$ has a phase that can even vary in time (slow compared to $\omega_L$) – this can be used to 'steer' the Bloch vector because its change is equivalent to a rotation of the 12-plane (equatorial plane) of the Bloch sphere.

## 1.5 06 Dec 16: short-pulse excitation of molecules

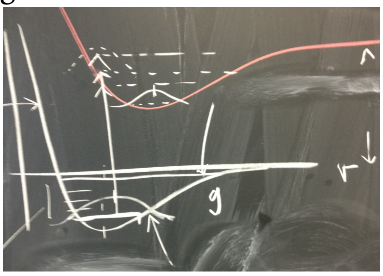
Electronic states in molecules have qualitative features that are similar to orbitals in atoms. Let us take as an example a diatomic molecule. The ground state is 'round' like an s-state (left), while an excited state is 'odd' and has a plane where the electronic wave function changes sign (right).





The right case is still ‘binding’ because there is still ‘enough’ electronic charge between the two nuclei. (This would be smaller with an orbital whose symmetry plane is in between the nuclei.)

The laser excitation of the molecule is often visualized in a two-potential diagram




where the lower/upper curves correspond to the ground/excited states. What is represented here are the eigenvalues of the Schrödinger equation for the electrons, at a fixed distance $r$ between the nuclei. In the so-called Born-Oppenheimer approximation, these eigenvalues can be used as potentials $V_g(r)$ and $V_e(r)$ for the relative motion of the molecule. At short distance, the Coulomb repulsion between the nuclei is dominant and the potentials are repulsive. At large distance, the electron can no longer ‘hop’ between the nuclei, and we get weak (van der Waals-like) interaction between an atom and an ion. In between is a minimum where one can read off the equilibrium length of the ‘chemical bond’ in the molecule (for the ground state). As usual in quantum mechanics, when we treat the distance $r$ as a dynamical variable, one gets bound states below the dissociation threshold, and continuum states above. The low-lying bound states have a characteristic frequency near the minima of the potential that sets the time scale for the vibrational motion. The period is in the hundred fs range, depending on the masses in the molecule.

With a short large pulse, one can excite from the ground state in $V_g$ a range of vibrational states in $V_e$, depending on the frequency spectrum of the pulse and the matching (overlap) between the vibrational wave functions. A typical rule is the ‘Franck-Condon principle’: with a high probability, one excites a vibrational state whose turning point in $V_e$ is close to the equilibrium position in $V_g$ (vertical arrows). But with a pulse, one typically excites several states and creates a wavepacket (superposition) that travels out in $V_e$ and comes back. With a second pulse, the process can be made to interfere, constructively or not, depending on the timing, the laser phase etc. One can also use a second laser with a different wavelength to excite the molecule to a third excited state – where it dissociates, for example. This pulse, also via the Franck-Condon principle, is sensitive to the excited-state amplitude in some spatial range and can probe in this way the arrival time, amplitude and shape of the wave packet. See lecture 'Quantum Dynamics and Wavepackets' with M. Gühr at U Potsdam (SS 16).

# 1.6 Spin language

The following sections 1.6–1.10 provide additional material: some has been covered in WS 16/17, some not.

## 1.6.1 Bloch vector

Bloch vector: expectation value of Pauli matrices

$$\mathrm{s}(t) = \langle \psi(t) | \boldsymbol{\sigma} | \psi(t) \rangle \tag{1.12}$$

Typical states, described by analogy to points on a sphere (spherical coordinates):

- ground state = south pole
- excited state = north pole
- superposition with equal weights $(|e\rangle + \mathrm{e}^{\mathrm{i}\varphi}|g\rangle)/\sqrt{2}$ = a point on the equator with longitude $\varphi$

Translation into macroscopic observables: occupations ($s_3$) and polarization ($s_1$, $s_2$).

## 1.6.2 Bloch dynamics

Dissipative dynamics for components of Bloch vector, contains the following processes:

Spontaneous decay rate $\gamma$ (time scale called $T_1$ in spin resonance).

Decoherence or dephasing rate $\Gamma$ (coherence time $T_2$).

Set of Bloch equations (assuming that $\Omega$ is real)

$$\begin{aligned}
\frac{\mathrm{d}s_1}{\mathrm{d}t} &= \Delta s_2 - \Gamma s_1 \\
\frac{\mathrm{d}s_2}{\mathrm{d}t} &= -\Delta s_1 - \Omega s_3 - \Gamma s_2 \\
\frac{\mathrm{d}s_3}{\mathrm{d}t} &= \Omega s_2 - \gamma (s_3 + 1)
\end{aligned} \tag{1.13}$$

in rotating frame at $\omega = \omega_L$, detuning $\Delta = \omega_L - \omega_A$, Rabi frequency $\Omega \sim \mathbf{E}_L$.

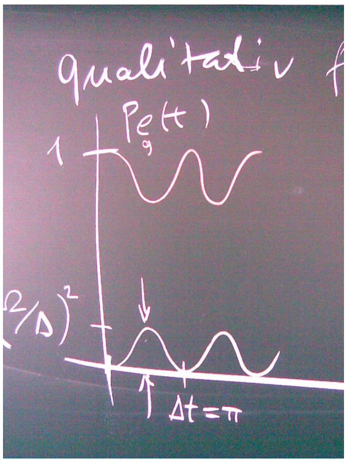
Geometric moves:

– rotation (from Hamiltonian, axis defined by detuning $\Delta$ and Rabi frequency $\Omega$)

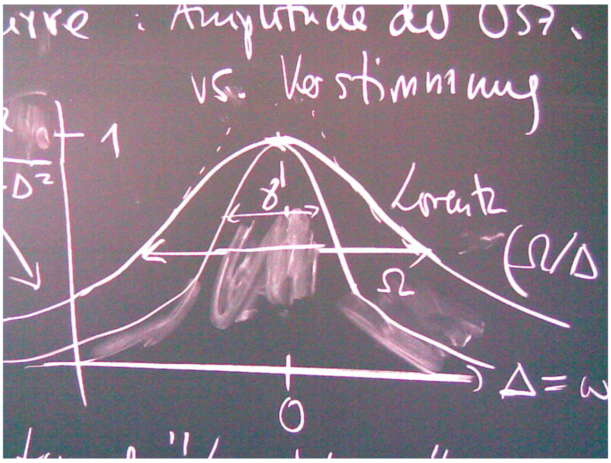
– contraction (from decoherence rate $\Gamma$ and decay rate $\gamma$), squeezes sphere into 'lemon' with axis in the 'north-south' direction

– displacement towards the south pole (ground state), from the decay rate $\gamma$

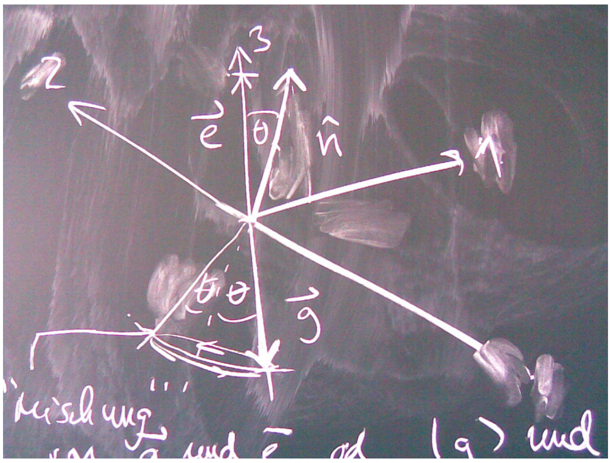
Solution to time-dependent Schrödinger equation: Rabi oscillations, Sec.1.9.4.



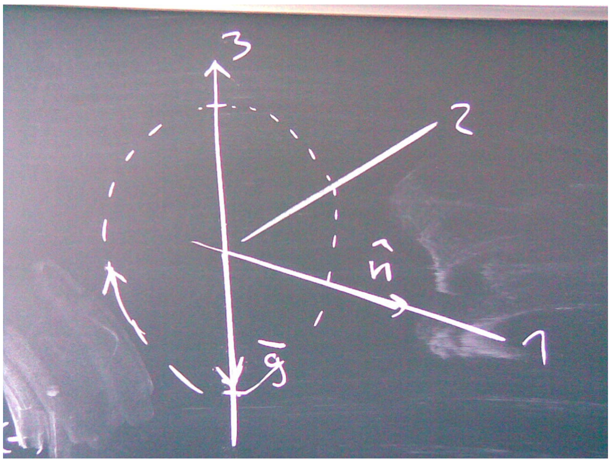

Rabi oscillations with nonzero detuning: populations of the two levels, $p_e(t)$ and $p_g(t)$, vs. time.




'Absorption spectrum' = amplitude of Rabi oscillations in $p_e(t)$ vs. laser frequency. Lorentz = name for the line shape = dependence on detuning $\Delta = \omega_L - \omega_A$. $\gamma$ = 'natural linewidth' = minimal width of the spectrum when the Rabi frequency $\Omega$ becomes small.



Sketch for rotation axis $\hat{\mathbf{n}}$ of the spin vector representing the state of the atom. Initial states $\vec{\mathrm{g}}$ = ground state ('spin down'), $\vec{\mathrm{e}}$ = excited state ('spin up'). The angle $\theta$, proportional to the Rabi frequency, gives the opening angle of a cone on which the spin vector rotates.




Rotation axis for a laser field on resonance: the spin vector rotates 'full circle' from spin down to spin up and back. Geometric representation of Rabi oscillations (on resonance, $\Delta = 0$).

General Hamiltonian is decomposed into unit matrix and Pauli matrices

$$\hat{H} = H_0 \mathbb{1} + \sum_j H_j \sigma_j \tag{1.14}$$

This gives the 'coherent dynamics'

$$\frac{\partial \mathbf{s}}{\partial t} = \frac{2}{\hbar} \mathbf{H} \times \mathbf{s} \tag{1.15}$$

= rotation of the Bloch vector. By analogy to the precession of a spin in a magnetic field, one calls the components $H_j$ in Eq.(1.14) the 'effective magnetic field'.

Rotations preserve the length of a vector: if one starts with a Bloch vector on the surface of the sphere, it will remain there if the time evolution is coherent (purely Hamiltonian). The length of the Bloch vector is not preserved by dissipative processes: they push the state into the interior of the Bloch sphere where 'incoherent states' occur (see Sec.1.10).

### 1.6.3 Limit of rate equations

The Bloch equations are a 'more refined' description compared to the rate equations (1.27). By making a suitable approximation, we can recover them, this is the technique of

- adiabatic elimination of coherences

We solve the equations for $s_1$ and $s_2$ with the assumption that the coherence time $1/\Gamma$ is the 'shortest time scale'. These two components of the Bloch vector (also known as 'coherences') then relax rapidly to their stationary values. Setting $\mathrm{d}s_{1,2}/\mathrm{d}t = 0$, we find for example

$$s_2 \approx -\frac{\Gamma\Omega}{\Gamma^2 + \Delta^2} s_3 \tag{1.16}$$

This is now inserted into the differential equation for $s_3$, assuming that the population difference $s_3$ evolves 'slowly enough' so that the coherence $s_2$ can 'follow adiabatically'. This gives the equation

$$\frac{\mathrm{d}s_3}{\mathrm{d}t} = -\gamma(s_3 + 1) - \Gamma \frac{\Omega^2}{\Gamma^2 + \Delta^2} s_3 \tag{1.17}$$

Adding an intelligent zero, $(\mathrm{d}/\mathrm{d}t)(p_e + p_g) = 0$, we get the rate equation for the excited state probability

$$\frac{\mathrm{d}p_e}{\mathrm{d}t} = -\gamma p_e - \Gamma \frac{\Omega^2}{\Gamma^2 + \Delta^2}(p_e - p_g) \tag{1.18}$$

The first term gives the spontaneous decay, the second one gives stimulated decay (negative) and absorption (positive). By comparing to the rate equations (1.27), we get the following information

- stimulated decay and absorption both scale with the light intensity, with the same proportionality factor

- the absorption cross section $\sigma$ can be found by identifying the absorption rate

$$\sigma I = \frac{\Gamma}{2} \frac{\Omega^2}{\Gamma^2 + \Delta^2} \tag{1.19}$$

Note the Lorentzian line shape of the absorption vs. laser frequency. For the photon flux $I$ (number of photons per time and area), we have from electrodynamics

$$I = \frac{c\varepsilon_0 |\mathbf{E}_L|^2}{\hbar\omega_L} \tag{1.20}$$

while the squared Rabi frequency is $\Omega^2 = 4d_{eg}^2 |\mathbf{E}_L|^2/\hbar^2$ (assuming a transition dipole parallel to the field). We get

$$\sigma = \frac{\Gamma^2}{\Gamma^2 + \Delta^2} \frac{2d_{eg}^2\omega_L}{\hbar c\varepsilon_0\Gamma} = \frac{\Gamma^2}{\Gamma^2 + \Delta^2} \frac{\omega_L}{\omega_A} 6\pi \left(\frac{c}{\omega_L}\right)^2 \tag{1.21}$$

where we have used the expression for the purely radiative decoherence rate

$$\Gamma = \frac{d_{eg}^2 \omega_A^3}{3\pi\varepsilon_0 \hbar c^3} \tag{1.22}$$

that Dirac (1927) has first found when he quantized the electromagnetic field. On resonance, Eq.(1.21) reproduces an absorption cross section given by the squared wavelength, much larger that the atomic size itself:

$$\text{resonant absorption cross section :} \qquad \sigma_{\text{abs}} = \frac{3}{2\pi}\lambda_A^2 \tag{1.23}$$

## 1.7 Selection of physical phenomena in light-matter interaction

The description of atom+light interaction that we have developed so far, complemented by techniques to quantize the field (see following chapter) is the basis for most of the phenomena that have been studied in the quantum optics of two-level system. One can discuss the following topics (we give a selection in this lecture):

- Rabi oscillations in a classical monochromatic field;
- spontaneous decay of an excited atom into the continuum of vacuum field modes (initially in the ground state);
- interaction of light with a medium of two-level atoms. One has to re-interpret the density matrix as giving the state of a macroscopic number of atoms. The occupations $p_e$, $p_g$, for example, then are proportional to the number of atoms (or molecules) in the excited and ground state. The atomic dipole becomes, after multiplication with the atom density, the polarization field (electric dipole moment per volume). Coupled to the Maxwell equations where this polarization field enters as a source term, one then has a simple "semiclassical" description for a laser, for a solar cell, for a semi-conductor. The Bloch equations in this case may contain more complicated terms.
- collapse and revival of Rabi oscillations when the atom couples to a single quantized field mode. The collapse and the revival occurs because the

Rabi frequency depends on the photon number, and the oscillations for the different Fock state components of a field state (a coherent state, for example) get out of phase;

- resonance fluorescence is the radiation emitted by an atom driven by a near-resonant laser field. This combines Rabi oscillations in a classical field with the emission of photons into the empty mode continuum. Of particular interest is the spectrum of this emission: it contains, for sufficiently strong driving, two sidebands, split by the Rabi frequency from the central line (centered at the laser frequency). The central line contains a monochromatic component ('elastic scattering', related to the laser-induced dipole moment as in classical electrodynamics) and a broadened component of Lorentzian shape, related to spontaneous emission. This spectrum is a cornerstone of quantum optics and one of the few examples of a non-perturbative calculation in quantum electrodynamics.

There are also physical effects which *cannot* be captured by our two-level description. They are related to the failure of the resonance approximation:

- the Lamb shift, a displacement of certain energy levels in atoms, arises from a very wide frequency continuum of vacuum field modes. One does get a wrong result if in the atom+field interaction, non-resonant processes are neglected.
- for short light pulses (roughly in the 1 fs range), one needs more than two energy levels, and even continuum states may be necessary for an accurate description. These pulses can generate 'high harmonics' as an electron is pulled away from the atomic core and pushed back under the influence of so-called 'few-cycle pulses' which can be very intense (electric fields comparable to those in the atom itself, Rabi frequencies comparable to atomic Bohr frequencies).

The following sections give more details on the atom+field coupling and are recommended for further reading. All details have not been covered in depth in WS 16/17.

## 1.8 Relevant observables

matter responds with
– absorption of light
– electric dipole moments, i.e. a polarization field $\mathbf{P}(\omega, \mathbf{r})$

... two different processes. A minimal description that contains the two is a *two-level model*.

### 1.8.1 Reminder on energy levels, occupations

Energy level scheme to model absorption: from quantum mechanics. Suppose that constituent atoms (molecules, ... ) are in either state $|\mathrm{g}\rangle$ or $|\mathrm{e}\rangle$. In a semiconductor, electronic bands (valence band, separated by an energy gap from the conduction band).

In a single atom, quantum mechanics provides probabilities $p_\mathrm{g}$ and $p_\mathrm{e}$: if $|\psi\rangle$ is the state of the system, then

$$p_a = |\langle a|\psi\rangle|^2, \qquad a = \mathrm{g}, \mathrm{e} \tag{1.24}$$

In a macroscopic piece of matter where $n(\mathbf{r})$ is the number density of atoms, we have level densities

$$n_a(\mathbf{r}, t) = n(\mathbf{r}) p_a(t; \mathbf{r}) \tag{1.25}$$

where the probability $p_a$ depends on $\mathbf{r}$ because it involves the local light fields, as we shall see in a moment.[1]

The total probability is conserved

$$p_\mathrm{g} + p_\mathrm{e} = 1 \tag{1.26}$$

and similarly the sum over the densities $n_a(\mathbf{r}, t)$. If this would not hold true, then a Hilbert space with only two states is 'too small'.

### 1.8.2 'Incoherent' dynamics: rate equations

The word 'incoherent' means here: a description with only populations $p_a$ is sufficient. Is often the case in condensed matter. Also for atoms in the gas phase

[1] Distinction between 'local' and 'macroscopic' field. Not a well-defined question, actually.

as long as the time scales are 'long enough'. Rate equations for absorption of light

$$\begin{aligned}\frac{\mathrm{d}p_{\mathrm{e}}}{\mathrm{d}t} &= \sigma I(\mathbf{r})p_{\mathrm{g}} - \gamma p_{\mathrm{e}} \\ \frac{\mathrm{d}p_{\mathrm{g}}}{\mathrm{d}t} &= -\sigma I(\mathbf{r})p_{\mathrm{g}} + \gamma p_{\mathrm{e}}\end{aligned} \tag{1.27}$$

where $I(\mathbf{r})$ is the 'photon intensity' (flux of photons per second, related to standard intensity by factor $\hbar\omega_L$) and $\sigma$ the absorption cross section. The time $1/\gamma$ is the lifetime of the excited state.

*Simple exercise*: stationary solution. $p_{\mathrm{e}}/p_{\mathrm{g}} = \sigma I/\gamma$. Typical numbers: $1/\gamma \sim 10\,\mathrm{ns}$, $\sigma \sim 10^{-16}\,\mathrm{cm}^2$, $I\,hf = 1\,\mathrm{mW/cm}^2$ give $p_{\mathrm{e}}/p_{\mathrm{g}} \sim 10^{-9}$. Can be much larger on resonance and with higher laser power.

*Exercise*: think about link between absorption cross section, absorption and complex refractive index of a medium. Check typical numbers.

### 1.8.3 Rate equations in a medium

Basic quantities are now spatial number densities $n_a(\mathbf{x}, t)$ ($a = e, g$) for atoms/molecules in the 'internal states' $e$ or $g$. A typical model looks like

$$\partial_t n_e = -\gamma n_e + \sigma I(n_g - n_e) + \nabla \cdot D\nabla n_e \tag{1.28}$$

$$\partial_t n_g = +\gamma n_e + \sigma I(n_e - n_g) + \nabla \cdot D\nabla n_g \tag{1.29}$$

The notation is as in Eqs.(1.27). In the first equation, we have two additional terms:

- *stimulated emission* described by the term with $-\sigma I n_e$: in the presence of photons, the quantum jump to the ground state happens more frequently. This is the key process for the working of a laser because it increases the photon number.
- *spatial diffusion* described by the second derivatives proportional to $D\nabla^2$: this is a typical 'macroscopic' consequence of collisions between particles (in a gas) or with the 'background medium' (in a solid medium). Diffusion involves the Fick-Fourier law for the current density

$$\mathbf{j}_a = -D\nabla n_a \tag{1.30}$$

In words: if there is a concentration gradient, diffusion tries to flatten it out by moving particles in the direction of less concentration. The diffusion coefficient $D$ (units: $\mathrm{m}^2/\mathrm{s}$) gives the mean square displacement per unit time, as in the simple model of a random walker (Brownian motion).

The total number of molecules (spatial integral of $n_e + n_g$) is conserved, up to boundary terms. At the boundary of the system, one may impose, for example, that particles in a given states are 'pumped' into it.

### Remarks

• With these rate equations, one may describe for example the dynamics of charge carriers (electrons and holes) generated in a solar cell. A few additional terms are needed to describe their generation by absorption of light, the electric field that drives them to the boundaries of the cell, and the recombination of electrons and holes.[2]

• A simplified laser model also works with rate equations. One needs, of course, an additional equation of motion for the photon intensity

$$\partial_t I = \ldots \text{(Maxwell)} \tag{1.31}$$

that will be based on the Maxwell equations to describe photon propagation in space.

• Such a set of rate equations for a laser gives a *nonlinear* system because of the processes of absorption and stimulated emission that involves the products $In_a$. In the field of 'nonlinear dynamics', a simplified version has become known as the Lorenz model; it is a well-known example of chaotic behaviour.

### Reminder on polarization

Meaning of polarization used here[3]: spatial density of electric dipole moments (a vector field). This is a 'micro-macro relation' typical for electrodynamics

$$\mathbf{P}(\omega, \mathbf{r}) = n_{\mathrm{at}}(\mathbf{r})\mathbf{d}(\omega) \tag{1.32}$$

[2]A recent example is the paper by Gélinas & al. (2014) where a spatial separation of electrons and holes was observed that was even faster than expected from the diffusion model above.

[3]Other meaning for polarization: orientation of the electric field vector in an electromagnetic wave. Linear, circular, elliptic polarization. Unpolarized, partially polarized light.

where $n_{\rm at}(\mathbf{r})$ is the spatial density of constituent 'atoms' (or molecules) and $\mathbf{d}(\omega)$ the dipole moment per atom. (Electrons are displaced relative to positive atomic cores, overall charge remains zero.)

Link to refractive index $n(\omega, \mathbf{r})$: the polarization field is the 'linear response' to the electric field. The linear coefficient involves the index:

$$\mathbf{P}(\omega, \mathbf{r}) = \varepsilon_0(n^2 - 1)\mathbf{E}(\omega, \mathbf{r}) \tag{1.33}$$

At the microscopic level: the linear response coefficient is called the polarizability $\alpha(\omega)$ = response of dipole moment to an electric field

$$\mathbf{d}(\omega) = \alpha(\omega)\mathbf{E}(\omega, \mathbf{r}) \tag{1.34}$$

where $\mathbf{r}$ is now the position of the atom in the field. In suitable units: $\alpha(\omega)/\varepsilon_0$ has the units of volume, typically comparable to the volume occupied by electrons in an atom.

## 1.9 Quantum mechanics of two-level system

We come back to a single two-level atom and start with a quantum mechanics description. The general state is a superposition

$$|\psi(t)\rangle = \sum_a c_a(t)|a\rangle = c_e(t)|e\rangle + c_g(t)|g\rangle \tag{1.35}$$

which contains more information that just the probabilities $p_a = |c_a|^2$. We shall see that the phases of the complex numbers $c_e$, $c_g$ are related to the polarization or the dipole moment of the atom.

### 1.9.1 Atom Hamiltonian

An operator, not consistently written with a hat

$$\hat{H} = H_A + H_{AL} \tag{1.36}$$

'Free atom Hamiltonian' for a system with just two energy levels (states $|e\rangle$ and $|g\rangle$)

$$H_a = E_e|e\rangle\langle e| + E_g|g\rangle\langle g| \tag{1.37}$$

Atom+light interaction Hamiltonian: copied by correspondence principle from electrodynamics = energy of electric dipole in a field

$$H_{AL} = -\hat{\mathbf{d}} \cdot \mathbf{E}_L(\mathbf{x}_A, t) \tag{1.38}$$

The 'dipole approximation' is made here: the electric field is evaluated at the position $\mathbf{x}_A$ of the 'atom as a whole', neglecting its variation across the size of the electron orbitals. In this chapter, we shall treat the electric field as a classical (time-dependent) field and not as an operator. This is called the 'semiclassical model'.

The dipole can be written as the following operator

$$\hat{\mathbf{d}} = \mathbf{d}_{eg}\Big(|e\rangle\langle g| + |g\rangle\langle e|\Big) \tag{1.39}$$

Its matrix elements between the two states can be arranged in a $2 \times 2$ matrix

$$\begin{pmatrix} \langle e|\hat{\mathbf{d}}|e\rangle & \langle e|\hat{\mathbf{d}}|g\rangle \\ \langle g|\hat{\mathbf{d}}|e\rangle & \langle g|\hat{\mathbf{d}}|g\rangle \end{pmatrix} = \begin{pmatrix} \mathbf{0} & \mathbf{d}_{eg} \\ \mathbf{d}_{eg} & \mathbf{0} \end{pmatrix} \tag{1.40}$$

Since this must be a hermitean matrix, we have assumed here that the dipole matrix elements are real: $\mathbf{d}_{ge} = \mathbf{d}_{eg}^* = \mathbf{d}_{eg}$. This can be done very often, for example when time-reversal holds. For other choices of the states $e$, $g$, however, $\mathbf{d}_{eg}$ remains complex and one has to adapt Eq.(1.39) accordingly.

We have assumed here that the states $|e\rangle$ and $|g\rangle$ have no 'average dipole moment' which is the quantum mechanical word for the diagonal matrix elements being zero. This is the result of a so-called 'selection rule' that applies as long as the two states have a well-defined parity (symmetry under spatial inversion of the electron coordinates). Conversely, the transition dipole moment $\mathbf{d}_{eg}$ is nonzero only when $|e\rangle$ and $|g\rangle$ have opposite parity, for example between an s-ground state (angular momentum $l = 0$) and a p-excited state ($l = 1$). More details on this can be found in Sec.1.11.2.

Eq.(1.40) is a vector-valued matrix, this happens quite often in quantum field theory. Recall vector $\boldsymbol{\sigma}$ of Pauli matrices. These matrices can also be used to re-write the atom Hamiltonian. The matrix representation is based on the identification of the basis vector $|e\rangle \leftrightarrow (1, 0)^T$ (my personal convention) and takes the form

$$H = \frac{E_e + E_g}{2}\mathbb{1} + \frac{E_e - E_g}{2}\sigma_3 - \mathbf{d}_{eg} \cdot \mathbf{E}_L(t)\sigma_1$$

where the term with the unit matrix $\mathbb{1}$ is often suppressed by a suitable choice of the zero of energy.

Finally, if the atom is illuminated by laser light, we may assume that the electric field is monochromatic with an amplitude and a frequency $\omega_L$. One often-used convention is to write

$$\mathbf{E}_L(\mathbf{x}, t) = \mathbf{E}_L(\mathbf{x})\, \mathrm{e}^{-\mathrm{i}\omega_L t} + \text{c.c.} \tag{1.41}$$

where 'c.c.' means 'complex conjugate'. Other conventions use the real part of the first term, but then the amplitude differs by a numerical factor. One has to check carefully which convention is used. An alternative convention is also using the 'other' sign in the exponential, $\mathrm{e}^{+\mathrm{i}\omega_L t}$, often used in optics and electrical engineering. (Sometimes, it may help to write $\mathrm{j} = -\mathrm{i}$ and avoid the confusion.)

### 1.9.2 Schrödinger equation

We want to work out the dynamics of the two-level system. The time-dependent amplitudes $c_a(t)$ in Eq.(1.35) can be found by taking the time derivative and using the Schrödinger equation:

$$\mathrm{i}\hbar c_a(t) = \langle a|H|\psi(t)\rangle \tag{1.42}$$

To work this out, we introduce the notation

$$\begin{aligned} E_e + E_g &= 0 \\ E_e - E_g &= \hbar\omega_A \qquad (1.43) \\ -\mathbf{d}_{eg} \cdot \mathbf{E}_L(\mathbf{x}_A, t) &= \frac{\hbar\Omega(t)}{2} \qquad (1.44) \end{aligned}$$

Eq.(1.85) is the formula of Bohr for the spectral line associated with the two energy levels. Eq.(1.44) defines the 'Rabi frequency' (in the Paris convention), which is proportional to the electric field of the laser. In the exercises, you have checked that typically, the Rabi frequency is much smaller than the Bohr frequency

$$|\Omega| \ll \omega_A \tag{1.45}$$

unless one works with 'strong laser fields'.

With this notation, the Schrödinger equation becomes

$$\begin{aligned} \mathrm{i}\partial_t c_e &= \frac{\omega_A}{2} c_e + \frac{\Omega(t)}{2} c_g \\ \mathrm{i}\partial_t c_g &= \frac{\omega_A}{2} c_g + \frac{\Omega(t)}{2} c_e \qquad (1.46) \end{aligned}$$

This is a coupled set of linear, ordinary differential equations with time-dependent coefficients. This time dependence makes the solution more complicated, and one has to apply an approximation to pursue.

In mathematical physics, a more systematic technique is based on a separation of multiple time scales. The 'fast time scale' is given by the Bohr frequency $\omega_A$, and the 'slow time scale' by the Rabi frequency $\Omega$, see Ineq.1.45. One can then derive small corrections to the resonance approximation we find below.

### 1.9.3 Resonance approximation

The equations can be simplified by adopting a transformation of the probability amplitudes $c_a$ with the following *Ansatz*:

$$
\begin{aligned}
c_e(t) &= \tilde{c}_e(t)\,\mathrm{e}^{-\mathrm{i}\omega t/2} \\
c_g(t) &= \tilde{c}_g(t)\,\mathrm{e}^{\mathrm{i}\omega t/2}
\end{aligned}
\tag{1.47}
$$

We shall assume that the prefactors $\tilde{c}_a(t)$ are 'slowly varying envelopes', while the exponentials give 'fast carrier waves'. (Think of radio signals that are modulated with a slow acoustic signal.) The frequency $\omega$ is for the moment a free parameter. In quantum optics, one says that the slow amplitudes $\tilde{c}_a(t)$ live 'in frame rotating at the frequency $\omega$'. (This picture has to do with spin resonance and the Bloch sphere, see later.)

Working out the time-derivative of Eqs.(1.47), one finds (no approximation yet) the equations of motion

$$
\begin{aligned}
\mathrm{i}\partial_t \tilde{c}_e &= \frac{\omega_A - \omega}{2}\tilde{c}_e + \frac{\Omega(t)\,\mathrm{e}^{\mathrm{i}\omega t}}{2}\tilde{c}_g \\
\mathrm{i}\partial_t \tilde{c}_g &= \frac{\omega - \omega_A}{2}\tilde{c}_g + \frac{\Omega(t)\,\mathrm{e}^{-\mathrm{i}\omega t}}{2}\tilde{c}_e
\end{aligned}
\tag{1.48}
$$

The term with $\omega$ appears because the transformation (1.47) into the rotating frame is time-dependent. One can now distinguish two choices

- *interaction picture*: $\omega = \omega_A$. The first term, from the free atom Hamiltonian $H_A$ drops out, and the dynamics depends only on the atom+field coupling. This is the starting point for (time-dependent) perturbation theory.

- *rotating frame* at the laser frequency: $\omega = \omega_L$. In combination with the resonance approximation, we can then find a Schrödinger equation with constant coefficients which is easier to solve. This is the starting point for many applications in quantum optics.

We adopt the second choice and fix the free parameter $\omega$ to the laser frequency. This motivates the notation for the

$$\text{detuning:} \qquad \Delta = \omega_L - \omega_A \tag{1.49}$$

(again: Paris convention, other signs exist). The term with the Rabi frequency becomes, using the complex notation of Eq.(1.41):

$$\Omega(t)\,\mathrm{e}^{\mathrm{i}\omega t} = \left(\Omega\,\mathrm{e}^{-\mathrm{i}\omega_t} + \text{c.c.}\right)\mathrm{e}^{\mathrm{i}\omega_L t} = \Omega + \Omega^* \mathrm{e}^{2\mathrm{i}\omega_L t} \tag{1.50}$$

where the first term is constant and the second 'varies rapidly'. We now apply the

- *resonance approximation* (or 'rotating wave approximation' RWA): we assume that the atomic dynamics is slow on the time scale $2\pi/\omega_L$ of the laser period and time-average the Schrödinger equation (1.48) over one period. The second term in Eq.(1.50) averages to zero and we get

$$\begin{aligned} \mathrm{i}\partial_t \tilde{c}_e &= -\frac{\Delta}{2}\tilde{c}_e + \frac{\Omega}{2}\tilde{c}_g \\ \mathrm{i}\partial_t \tilde{c}_g &= \frac{\Delta}{2}\tilde{c}_g + \frac{\Omega^*}{2}\tilde{c}_e \end{aligned} \tag{1.51}$$

This is nicer equation because the coefficients are constant in time.

The argument leading to the RWA, based on time-averaging, is 'heuristic' and has to be applied with care. If we did the averaging on Eq.(1.46), for example, the atom+field coupling would disappear.

There are other arguments that motivate the resonance approximation. For example, when the field is also described quantum mechanically, it becomes an operator that generates and destroys photons. A process that one neglects in the RWA corresponds to a 'non-resonant excitation': the atom jumps to the upper state $|e\rangle$ and generates a photon. This process involves an 'energy mismatch' $\mathcal{O}(\hbar\omega_A + \hbar\omega_L)$ and is therefore called 'non-resonant'. It is not forbidden in quantum mechanics, but it happens with a small amplitude. In the RWA, this small amplitude is neglected.

There are observable effects where non-resonant (also known as 'virtual') processes play a role and where the RWA must not be applied. An example are energy shifts in quantum-electrodynamics like the Lamb shift or the Casimir-Polder shift.

### 1.9.4 Rabi oscillations

The simplest case of atom-laser dynamics is a laser 'on resonance', i.e., $\omega_L = \omega_A$. The Schrödinger equation (1.51) yields (we drop the tildes)

$$\mathrm{i}\dot{c}_\mathrm{e} = \frac{\Omega}{2} c_\mathrm{g} \tag{1.52}$$

$$\mathrm{i}\dot{c}_\mathrm{g} = \frac{\Omega}{2} c_\mathrm{e}. \tag{1.53}$$

where the Rabi frequency is chosen real for simplicity. With the initial conditions $c_\mathrm{g}(0) = 1,\ c_\mathrm{e}(0) = 0$, the solution is

$$\dot{c}_\mathrm{e} = -\mathrm{i}\sin(\Omega t/2) \tag{1.54}$$

$$c_\mathrm{g} = \cos(\Omega t/2). \tag{1.55}$$

The excited state probability thus oscillates between $0$ and $1$ at a frequency $\Omega/2$. This phenomenon is called 'Rabi flopping'. It differs from what one would guess from ordinary time-dependent perturbation theory where one typically gets linearly increasing probabilities. That framework, however, applies only if the final state of the transition lies in a continuum which is not the case here. Rabi flopping also generalizes the perturbative result (1.110) which would give a quadratic increase $|c_\mathrm{e}|^2 \propto t^2$ that cannot continue for long times. But instead of saturating, the atomic population returns to the ground state.

Every experimentalist is very happy when s/he observes Rabi oscillations. It means that any dissipative processes have been controlled so that they happen at a slower rate. In a realistic setting, one gets a damping of the oscillation amplitude towards equilibrium populations.

**Non-resonant case.** Solved with the exponential of a linear combination of Pauli matrices: write

$$H = -\frac{\hbar}{2} R\left(\sigma_3 \cos\theta + \sigma_1 \sin\theta\right) = -\frac{\hbar}{2} R\sigma_R \tag{1.56}$$

with $\tan\theta = -\Omega/\Delta$ and $R^2 = \Delta^2 + \Omega^2$. The matrix $\sigma_R$ has the property $\sigma_R^2 = \mathbb{1}$ and $\sigma_R^3 = \sigma_R$. This permits to re-sum even and odd terms in the series expansion of the exponential into cosine and sine functions. The unitary time evolution operator becomes (in the rotating frame only)

$$U(t) = \exp(-\mathrm{i}Ht/\hbar) = \mathbb{1}\cos(Rt/2) + \mathrm{i}\sigma_R \sin(Rt/2) \tag{1.57}$$

If this is applied to the ground state, $(0, 1)^\mathsf{T}$ as a two-component vector, we can read off the state at time $t$.

Details: exercise. Excited state population oscillates at frequency $R$ ('generalized Rabi frequency'), but with an amplitude $< 1$. The dependence of this amplitude on the laser frequency (detuning $\Delta$) can be called an 'absorption line shape', it decays as $1/\Delta^2$ for large detuning. The width is of the order of $\Omega$: this is called 'power broadening'.

Picture becomes wrong as the Rabi frequency gets weaker because the spontaneous emission rate $\gamma$, another relevant frequency scale, sets in: the absorption line shape cannot become narrower than $\gamma$.

## 1.10 Mixed states and dissipation

The concept of the density operator generalizes the state vector familiar from quantum mechanics. The main reason is that we also want to handle states (or ensembles) which have a nonzero entropy (like thermal states have) or which arise from processes that do not conserve entropy, like spontaneous decay.

See the *exercises* for a discussion of entropy.

In the field of quantum information, one adopts sometimes a quite mathematical language. We shall follow this route and define a density operator by the following properties

A density operator $\rho$ is a hermitean operator on the Hilbert space $\mathcal{H}$ of the system. (A $N \times N$-matrix if $\dim \mathcal{H} = N$ is finite.)

$\rho$ is normalized to unit trace, $\operatorname{tr} \rho = 1$.

$\rho$ is positive: for all $|\psi\rangle \in \mathcal{H}$, we have

$$0 \le \langle\psi|\rho|\psi\rangle \le \langle\psi|\psi\rangle \tag{1.58}$$

The expectation value of a system observable $\hat{A}$ is given by the 'trace rule'

$$\langle\hat{A}\rangle = \operatorname{tr}(\hat{A}\rho) = \operatorname{tr}(\rho\hat{A}) \tag{1.59}$$

where we have used a cyclic permutation under the trace.

These conditions imply that the eigenvalues of $\rho$ are real numbers in the interval $0 \ldots 1$ whose sum is unity. They can therefore be interpreted as probabilities.

Indeed the diagonal matrix element $\langle\psi|\rho|\psi\rangle$ in Eq.(1.58) is physically interpreted as the probability to find the system in the state $|\psi\rangle$ when it has been prepared in the density operator $\rho$. (This does not tell anything how this measurement is implemented or what observable one has to measure.)

### 1.10.1 Projectors = pure states

One class of states is familiar from ordinary quantum mechanics:

If $|\psi\rangle$ is a normalized state vector in the Hilbert space, then the projector

$$\mathbb{P}_{|\psi\rangle} = |\psi\rangle\langle\psi| \tag{1.60}$$

is a density operator. It is *the* density operator for a system prepared in the state $|\psi\rangle$.

It is simple the check that $\mathbb{P}_{|\psi\rangle}$ satisfies all conditions for a density operator. As an example, let us work out its trace by using a set of basis vectors $|n\rangle$

$$\mathrm{tr}(|\psi\rangle\langle\psi|) = \sum_n \langle n|\psi\rangle\langle\psi|n\rangle = \sum_n |\langle n|\psi\rangle|^2 = 1 \tag{1.61}$$

In the last sum, we recognize the sum over the squares of the coefficients $c_n = \langle n|\psi\rangle$ that represent the vector $|\psi\rangle$ in the chosen basis. This is just the norm of the state. Alternatively, we could have used the completeness relation $\sum_n |n\rangle\langle n| = \mathbb{1}$.

By a similar calculation, one also checks that the trace rule is equivalent to the standard expectation value of observables:

$$\mathrm{tr}(\hat{A}\rho) = \sum_n \langle n|\hat{A}|\psi\rangle\langle\psi|n\rangle = \sum_n \langle\psi|n\rangle\langle n|\hat{A}|\psi\rangle = \langle\psi|\hat{A}|\psi\rangle \tag{1.62}$$

Density operators that are built from projectors are called *pure states*. A formal definition that requires only the knowledge of the density operator $\rho$ is based on a well-known property of projectors:

A density operator $\rho$ is called a pure state when $\rho^2 = \rho$.

It is easy to see that for a pure state, the eigenvalues $p_n$ must satisfy the property $\sum_n p_n^2 = 1$. Since we have $0 \le p_n \le 1$, this can only be satisfied if exactly one term in the sum is nonzero, say $p_1 = 1$, while all others are zero. But this means, using the eigenvector $|\psi_1\rangle$, that the 'spectral representation' of the density operator reduces to a single term:

$$\rho = \sum_n p_n |\psi_n\rangle\langle\psi_n| = |\psi_1\rangle\langle\psi_1| \tag{1.63}$$

in other words: $\rho$ is a projector.

### 1.10.2 Purity and entropy

The definition of a pure state above can be exploited to define a quantitative measure of how far a state is from the set of pure states:

The 'mixedness' $\mathrm{Mx}(\rho)$ of a density operator is given by

$$\mathrm{Mx}(\rho) = \frac{\mathrm{tr}(\rho - \rho^2)}{\mathrm{tr}\,\rho} = 1 - \mathrm{tr}\,\rho^2 \tag{1.64}$$

where the last equation holds for trace-normalized density operators. Conversely, the *purity* is $\mathrm{Pu}(\rho) = 1 - \mathrm{Mx}(\rho)$.

For a pure state, we have $\mathrm{Mx}(\rho) = 0$ and $\mathrm{Pu}(\rho) = 1$ States with $\mathrm{Mx}(\rho) > 0$ ($\mathrm{Pu}(\rho) < 1$) are called non-pure or *mixed*.

An alternative measure of purity is provided by the entropy:

The *entropy* of a density operator is given formally and in terms of its eigenvalues $p_n$ by

$$S(\rho) = -\mathrm{tr}\,\rho \log \rho = -\sum_n p_n \log p_n \tag{1.65}$$

A pure state has entropy $S(\rho) = 0$ if we remember the limiting value $\lim_{x\to 0} x \log x = 0$. (Apply the De L'Hôpital rule to the ratio $x/(1/\log x)$ or to $\log x/(1/x)$.)

If we make the formal expansion $\log \rho \approx \log \mathbb{1} + \rho - \mathbb{1} + \ldots$, define $\log \mathbb{1} = 0$, and stop after the linear term, we recover the purity defined above. The purity has the advantage that the logarithm of the matrix is never needed (and no diagonalization neither). In quantum information, one uses other types of entropy measures that share similar monotony properties: they are zero for pure states and increase as the state becomes mixed.

### Mixed states and convexity

As an example of a mixed state, let us consider two pure states $|\psi\rangle$, $|\chi\rangle$ and form the following linear combination of projectors

$$\rho = p|\psi\rangle\langle\psi| + q|\chi\rangle\langle\chi|, \qquad p, q > 0, \quad p + q = 1 \tag{1.66}$$

This is called a *statistical mixture* of the states $|\psi\rangle$, $|\chi\rangle$.

Let us interpret this as a state preparation procedure with incomplete knowledge: a machine prepares a state $|\psi\rangle$ with probability $p$ and otherwise the state $|\chi\rangle$. The only possible prediction of the average value of any observable $\hat{A}$ that we can then make is the following

$$\langle\hat{A}\rangle = p\langle\psi|\hat{A}|\psi\rangle + q\langle\chi|\hat{A}|\chi\rangle = \mathrm{tr}(\hat{A}\rho) \tag{1.67}$$

which is nothing but the definition of the average for a density operator. This equation combines in an elegant way 'quantum' and 'classical' probabilities: indeed, the expectation values $\langle\psi|\hat{A}|\psi\rangle$ are typical for quantum mechanics, with the distribution of possible values for $\hat{A}$ given by the wave function of $|\psi\rangle$ (the expansion of the state over a basis of eigenstates of $\hat{A}$). On the other hand, the weighting factors $p$, $q$ in Eq.(1.67) are simply what one would do in classical statistics, when events occur with some probabilities and an average outcome is asked for.

In a thermal state (statistical physics), a typical mixture occurs in thermal equilibrium: the states $|\psi\rangle$ and $|\chi\rangle$ are energy eigenstates (with energies $E_\mathrm{e}$ and $E_\mathrm{g}$, say). The corresponding probabilities are proportional to the Boltzmann factor $p \sim \mathrm{e}^{-E_\mathrm{e}/kT}$ where $T$ is the temperature of the system (and $k$ the Boltzmann constant). The average value $\langle\hat{A}\rangle$ in thermal equilibrium is then the combination of quantum expectation values in the energy eigenstates, averaged over the classical probabilities of finding these states in the thermal ensemble. For a system with $n$ energy levels (subscript $T$ for thermal equilibrium):

$$\langle\hat{A}\rangle_T = \sum_n \frac{\mathrm{e}^{-E_n/kT}}{Z}\langle n|\hat{A}|n\rangle \tag{1.68}$$

Here, $Z = \sum_n \mathrm{e}^{-E_n/kT}$ is the partition function (*Zustandssumme*) that provides the normalization for the Boltzmann probabilities.

From a geometrical viewpoint, the 'mixing rule' (1.66) implies that for any two points in the set of density operators, also the straight line that joins them is included in the set. This straight line is just the linear combination with real coefficients between $0$ and $1$. In mathematics, this is called a *convex* linear combination. A set is called convex if it contains for any two points, also the line between the two.

We can of course generalize Eq.(1.66) to the mixing of any two density operators $\rho_1$, $\rho_2$

$$\rho = p\rho_1 + q\rho_2, \qquad p, q > 0, \quad p + q = 1 \tag{1.69}$$

It is easy to check that $\rho$ defined in this way is again a density operator. The physical interpretation can again be formulated in terms of an incomplete knowledge about state preparation.

In mathematics, convex sets are conveniently characterized when their 'extreme points' are known. Intuitively speaking, extreme points correspond to 'corners' of the convex set from which lines into its interior can be drawn. In quantum information, extreme points are closely related to pure states. We shall encounter below a very simple convex set: a sphere (the *Bloch sphere*). Its extreme points are all those on the surface of the sphere, while the points in the interior correspond to mixed states.

One even more formal way of introducing a density operator or a quantum state: in the axiomatic language of quantum information, a 'state' $\rho$ is a mapping from a set of observables to their expectation values

$$\rho : A \mapsto \rho(A) = \langle A \rangle_\rho \tag{1.70}$$

Linear map with $\rho(\mathbb{1}) = 1$ (real or complex coefficients depending on choice of observable algebra) and $\rho(A)$ real for a hermitean $A$.

Consider the familiar linear combinations in Hilbert space, $|\phi\rangle = \alpha|\mathrm{e}\rangle + \beta|\mathrm{g}\rangle$, with basis vectors $|\mathrm{e}\rangle$ and $|\mathrm{g}\rangle$, say. These states play a special role and are called *pure states*. They also correspond to special observables: *projectors*

$$\mathbb{P}_\phi = |\phi\rangle\langle\phi| \tag{1.71}$$

This is also a hermitean operator with eigenvalues $0$ or $1$. A physical state has the property that it is *positive*:

$$\rho(\mathbb{P}_\phi) \geq 0 \quad \text{for all } |\phi\rangle \tag{1.72}$$

Physical interpretation: this is the probability of finding the system in the pure state $|\phi\rangle$, which clearly must be a positive number.

Definition of *density matrix* (or *density operator*): any linear map on the vector space of observables can be represented by a suitable linear form

$$\rho(A) = \mathrm{tr}(\bar{\rho}A) \tag{1.73}$$

where $\bar{\rho}$ is a hermitean operator and $\mathrm{tr}(\hat{A}\hat{B})$ is a natural scalar product on the space of (hermitean) observables. This rule corresponds to the usual calculation of expectation values for mixed states in quantum statistics. In a finite-dimensional system, it corresponds to the duality between linear forms and vectors: each linear form can be represented as a scalar product with a suitable vector. This becomes the Riesz representation theorem in an infinite-dimensional Hilbert space.

### 1.10.3 Two-level systems: Bloch sphere

**Density matrix**

Let us analyze the set of density operators for a two-level system using a simple parametrization. The operator $\rho$ has a matrix representation in the basis $|\mathrm{e}\rangle$, $|\mathrm{g}\rangle$

$$\rho = \begin{pmatrix} p & \rho_{\mathrm{eg}} \\ \rho^*_{\mathrm{eg}} & q \end{pmatrix}, \qquad p + q = 1 \tag{1.74}$$

We have $p, q > 0$ because these elements correspond to diagonal matrix elements $p = \langle \mathrm{e}|\rho|\mathrm{e}\rangle = (1, 0)\rho \binom{1}{0} \geq 0$. Their sum is unity because of the trace-normalization. A single complex number $\rho_{\mathrm{eg}}$ gives both off-diagonal elements:

The off-diagonal elements of a density matrix are called *coherences*. They quantify to what extent (in this basis), the quantum state is rather a 'coherent superposition' as opposed to a 'classical mixture'. In quantum optics, 'states with large coherence' are often very useful, but also fragile.

The coherences cannot be very large in order not to spoil the positivity of $\rho$. This can be worked out from $\langle\phi|\rho|\phi\rangle$ with an 'optimized' superposition state $|\phi\rangle$. We follow here a faster route and compute simply the mixedness of the density operator. It must be positive or zero:

$$\mathrm{Mx}(\rho) = 1 - \mathrm{tr}\,\rho^2 = 1 - (p^2 + |\rho_{\mathrm{eg}}|^2 + |\rho_{\mathrm{eg}}|^2 + q^2) = 1 - p^2 - q^2 - 2|\rho_{\mathrm{eg}}|^2 \geq 0 \tag{1.75}$$

We thus have an upper bound for the modulus $|\rho_{\mathrm{eg}}|$. For equal probabilities $p = q = \frac{1}{2}$ (which looks like a 'mixed state'), we get the inequality $|\rho_{\mathrm{eg}}| \leq 1/2$. At the upper limit $|\rho_{\mathrm{eg}}| = 1/2$, we even get a pure state! On the other side, if $p = 1$, then necessarily $\rho_{\mathrm{eg}} = 0$: in the vicinity of the basis states, there is little space for 'coherence'. (We shall see in the next section how significant coherences can be achieved nevertheless.)

To summarize, we have found three independent real parameters ($p$, $\mathrm{Re}\,\rho_{\mathrm{eg}}$, and $\mathrm{Im}\,\rho_{\mathrm{eg}}$) whose range is some compact domain. What can be said about the geometry of this domain?

### Bloch vector

Any hermitean $2 \times 2$ matrix with trace $1$ can be written as a linear combination of hermitean matrices that are well known from quantum mechanics as the Pauli matrices (check the signs in $\sigma_2$)

$$\rho = \frac{1}{2}\left(\mathbb{1} + s_3\sigma_3 + s_1\sigma_1 + s_2\sigma_2\right) = \frac{1}{2}\begin{pmatrix} 1+s_3 & s_1+\mathrm{i}s_2 \\ s_1-\mathrm{i}s_2 & 1-s_3 \end{pmatrix} \tag{1.76}$$

The three numbers $s_i$ $(i = 1, 2, 3)$ are called the components of the *Bloch vector*. If we remember our notation $p$, $q$ for the diagonal elements, we have $s_3 = p - q$. Similarly, $s_1$ and $s_2$ are proportional to the real and imaginary parts of $\rho_{\mathrm{eg}}$. The inequality (1.75) for the real parameters takes a very simple form for the Bloch vector

$$s_3^2 + s_1^2 + s_2^2 \leq 1 \tag{1.77}$$

This means that the vector $\mathrm{s} = (s_1, s_2, s_3)$ lies inside a sphere of radius $1$ in three-dimensional space.

*Exercises*: the components of the Bloch vector are the expectation values of the Pauli matrices:

$$s_i = \langle \sigma_i \rangle = \mathrm{tr}(\sigma_i \rho) \tag{1.78}$$

The mixedness is determined by the length of the Bloch vector only

$$\mathrm{Mx}(\rho) = 1 - \mathrm{tr}(\bar{\rho}^2) = \ldots = \frac{1}{2}(1 - \mathbf{s}^2) \tag{1.79}$$

• picture!

### Pure states

The language of a spin 1/2 is often used to visualize the dynamics of a two-level atom. Let us compute the components of the average spin vector $(s_1, s_2, s_3)^T \equiv \langle \boldsymbol{\sigma} \rangle$ (the 'Bloch vector') in the pure state $|\psi(t)\rangle = (c_{\mathrm{e}}(t), c_{\mathrm{g}}(t))^T$:

$$s_3 = |c_e|^2 - |c_g|^2 \tag{1.80}$$

$$s_1 - \mathrm{i}s_2 = 2c_g^* c_e \tag{1.81}$$

Observe that $\mathrm{s}^2 = s_1^2 + s_2^2 + s_3^3 = 1$ for a pure state. The component $s_3$ is related to the occupation probabilities $|c_{\rm e}(t)|^2$, $|c_{\rm g}(t)|^2$ (the 'populations'): it gives the *inversion*, i.e., the difference of the ground and excited state populations. In the ground state, one has $\langle \sigma_3 \rangle = -1$. The other two components (1.81) are only nonzero when the atom is in a superposition of the ground and excited states.

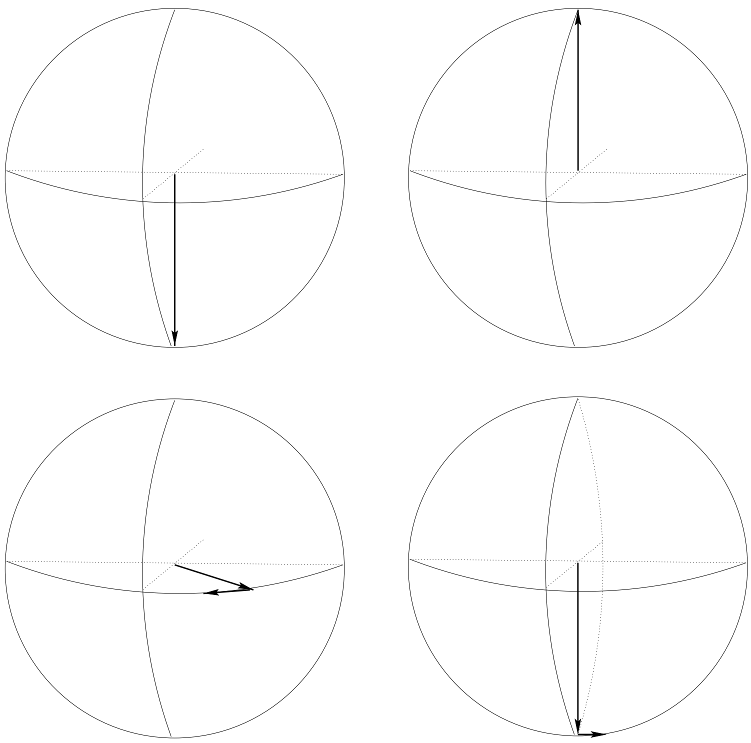


Figure 1.1: Upper left: Bloch vector for an atom in the ground state. Upper right: excited state. Lower left: superposition of ground and excited states with equal weight. The arrow along the 'equator' indicates the direction of free rotation of the Bloch vector without a laser field. Lower right: an initial ground state starts to undergo resonant Rabi oscillations, as indicated by the arrow tangent to the 'south pole'. The $x_1$-axis points to the right, the $x_3$-axis upwards.

In the exercises, you show that the Bloch vector is in general constrained by $\mathrm{s}^2 \le 1$, due to the positivity condition (1.150) of the density matrix. It can thus be represented by a point in a sphere, the 'Bloch sphere' (see Fig.1.1). On the sphere surface are located the pure states: the 'south pole' corresponds to an atom in the ground state, while an excited atom is at the 'north pole'. In between these two, the atom is in a superposition of ground and excited states. In particular, around the equator, both ground and excited state are occupied with probability $1/2$ (the inversion is zero). The longitude along the equator is fixed by the phase difference between ground and excited state.

If no external laser field is applied, the Bloch vector rotates at the frequency $\omega_A$ clockwise around the vertical axis. In particular, the occupation probabilites do not change with time: the inversion, being the projection onto the vertical axis, is constant. With the laser on, we shall see that the rotation axis gets tilted so that an atom initially in the ground state develops an excited component.

### 1.10.4 Bloch equations

Two-level system with atom+field Hamiltonian in resonance approximation, plus spontaneous emission and dephasing. The equations of motion for the Bloch vector are called the (optical) Bloch equations

$$\begin{aligned} \frac{\mathrm{d}s_1}{\mathrm{d}t} &= \Delta s_2 - \Gamma s_1 \\ \frac{\mathrm{d}s_2}{\mathrm{d}t} &= -\Delta s_1 - \Omega s_3 - \Gamma s_2 \\ \frac{\mathrm{d}s_3}{\mathrm{d}t} &= \Omega s_2 - \gamma(s_3 + 1) \end{aligned} \tag{1.82}$$

in rotating frame at $\omega = \omega_L$, detuning $\Delta = \omega_L - \omega_A$, Rabi frequency $\Omega \sim \mathbf{E}_L$.

Pictures from numerical solution: Bloch vector is spiralling around an axis given by the Hamiltonian (1.56): tilted by the angle $\theta$ with respect to the 'north-south-axis'. The spiralling is due to decoherence and spontaneous emission: decoherence (rate $\Gamma$) 'contracts' the Bloch sphere towards to the north-south-axis, while the spontaneous decay rate $\gamma$ contracts the sphere vertically and pushes it 'down' towards the ground state.

| | | |
|---|---|---|
| hydrogen | | H |
| lithium | | Li |
| sodium | Natrium | Na |
| potassium | Kalium | K |
| rubidium | | Rb |
| cesium | | Cs |
| francium | | Fr |

| $n$ | $l = 0$ | $l = 1$ | $l = 2$ | $\ldots$ |
|---|---|---|---|---|
| $\ldots$ | $\ldots$ | $\ldots$ | $\ldots$ | $\ldots$ |
| 3 | 3s | 3p | 3d | |
| 2 | 2s | 2p | | |
| 1 | 1s | | | |

Table 1.1: Left: the series of alkaline atoms. Right: Spectroscopic notation for energy levels of hydrogen-like atoms.

## 1.11 More atomic physics

Detailed treatment not covered in WS 16/17.

An atom can be modelled as a collection of charged point particles. The simplest Hamiltonian one can write is therefore

$$H_A = \sum_\alpha \frac{\mathbf{p}_\alpha^2}{2M_\alpha} + \frac{1}{2} \sum_{\alpha \neq \beta} \frac{e_\alpha e_\beta}{4\pi\varepsilon_0 |\mathbf{x}_\alpha - \mathbf{x}_\beta|} \tag{1.83}$$

where $\alpha$ labels the particles, $M_\alpha, e_\alpha$ are their masses and charges. We try to use in this lecture SI units. (In cgs units, drop the $4\pi\varepsilon_0$.) The interaction term corresponds to the electrostatic (or Coulomb) field created by the charges.

More advanced atomic models adopt a relativistic viewpoint, take into account the electron spin, the magnetic field created by the motion of the particles, the corresponding spin-orbit interaction, the spin-spin coupling, the hyperfine interaction etc. Theoretical atomic physics computes all these corrections to the energy levels and matrix elements 'from first principles' (see the textbook by Haken & Wolf (2000), for example). Typically, no simple analytical results can be found for atoms with more than two or three electrons, say.

For our purposes, we are not interested in so much detail. Instead, we use a simplified description of the atom states that captures their essential properties. Good examples are atoms with a single electron in the outer shell (the alkaline series), as listed in table 1.1. Their energy levels are to a good approximation given by a modified Balmer formula

$$E_{nl} = -\frac{e^2}{8\pi\varepsilon_0 a_0 (n + \delta_l)^2} = -\frac{\text{Ryd}}{(n + \delta_l)^2}, \qquad \begin{array}{rcl} n &=& 1, 2 \ldots \\ l &=& 0, \ldots n - 1 \end{array} \tag{1.84}$$

The Bohr radius $a_0 = 4\pi\varepsilon_0\hbar^2/me^2 \approx 0.5\,\text{Å}$ gives the typical size of the electron cloud, and the 'quantum defect' $\delta_l$ lifts the degeneracy of the hydrogen levels.The energy scale is given by the Rydberg constant $\text{Ryd} \approx 13.6\,\text{eV}$.

The charge $Z|e|$ of the nucleus enters the Balmer formula via the quantum defects $\delta_l$. In fact, the outer electron 'sees' the nucleus screened by the core electrons. This gives a Coulomb potential as for the hydrogen atom, with some modifications due to the core electrons. These are responsible for the lifted degeneracy between the $l$ states.

The frequency of electromagnetic radiation emitted by atoms in a process $|i\rangle \to |f\rangle$ is given according to Bohr by

$$\omega_{if} = \frac{E_i - E_f}{\hbar} \tag{1.85}$$

For two typical energy levels $E_{i,f}$, $\hbar\omega_{if}$ is also of the order of $1\,\text{Ryd}$. If we compute the wavelength of the corresponding electromagnetic radiation, we find

$$\lambda_{if} \sim \frac{2\pi\hbar c}{\text{Ryd}} = \frac{4\pi}{\alpha_{\text{fs}}} a_0 \gg a_0 \tag{1.86}$$

$$\text{fine structure constant } \alpha_{\text{fs}} = \frac{e^2}{4\pi\varepsilon_0\hbar c} \approx \frac{1}{137}, \tag{1.87}$$

which is much longer than the typical size of the atom (the Bohr radius gives the extension of the electronic orbitals). For typical light fields, the atom thus appears like a pointlike object. This property justifies the 'long wavelength approximation' that simplifies the Hamiltonian for the atom–light interaction.

Another way of looking at the result (1.86) is to interpret the inverse fine structure constant, $\alpha_{\text{fs}}^{-1} = c(4\pi\varepsilon_0\hbar/e^2)$ as the ratio between the speed of light $c$ and the typical velocity for an electron in the Hydrogen atom (the natural velocity scale in the so-called 'atomic units'). We see that this velocity is only a few percent of $c$, hence we expect that the non-relativistic description we have used so far is a good approximation.

### 1.11.1 Atom-light interaction

#### Minimal coupling

According to the rules of electrodynamics, the interaction between a collection of charges with a given electromagnetic field is described by the 'minimal coupling' Hamiltonian. This corresponds to the replacement $\mathbf{p}_\alpha \mapsto \mathbf{p}_\alpha - e_\alpha \mathbf{A}(\mathbf{x}_\alpha, t)$ where $\mathbf{A}(\mathbf{r}, t)$ is the vector potential. In this chapter, this is a given time-dependent function. It will become an operator when the field is quantized. In addition, there is the potential energy due to an 'external' scalar potential $\phi_{\rm ext}(\mathbf{x}, t)$, so that we get

$$H_{AF} = \sum_\alpha \frac{(\mathbf{p}_\alpha - e_\alpha \mathbf{A}(\mathbf{x}_\alpha, t))^2}{2M_\alpha} + \frac{1}{2}\sum_{\alpha \neq \beta} \frac{e_\alpha e_\beta}{4\pi\varepsilon_0 |\mathbf{x}_\alpha - \mathbf{x}_\beta|} + \sum_\alpha e_\alpha \phi_{\rm ext}(\mathbf{x}_\alpha, t). \quad (1.88)$$

The minimal coupling prescription is related to the freedom of choosing the phase reference of the wave function, as is seen in more detail in the exercises.

**Remark.** This freedom is also called 'local $U(1)$ gauge invariance' because phase factors form the unitary group $U(1)$. Local changes in the phase of the wave function generate terms in the Schrödinger equation that can be combined with gauge transformations for the electromagnetic potentials. This connection to the electromagnetic gauge transformations is of great importance for quantum field theory. It allows to construct the coupling to the electromagnetic field from the symmetry properties of the quantum fields. For example, there are theories where electrons and neutrinos are combined into a two-component field, and the interactions are invariant under $SU(2)$ transformations that mix these two, plus $U(1)$ transformations of the phase common to the two components. The group $SU(2) \times U(1)$ is four-dimensional and has four 'generators'. Each of them corresponds to a vector potential that interacts with the two-component field. In addition to the standard electromagnetic potential (the 'photon'), there are interactions associated to the 'massive vector bosons', called $W^\pm$ and $Z^0$. They convey the 'weak interaction' that is responsible for $\beta$ decay. More details in any book on quantum field theory. I sometimes use the one by Itzykson & Zuber (2006).

When the minimal coupling Hamiltonian (1.88) is expanded to lowest order in the charges $e_\alpha$, we obtain the so called '$p \cdot A$' interaction

$$H_{\rm int} = -\sum_\alpha \frac{e_\alpha}{2M_\alpha} \{\mathbf{p}_\alpha \cdot \mathbf{A}(\mathbf{x}_\alpha, t) + \mathbf{A}(\mathbf{x}_\alpha, t) \cdot \mathbf{p}_\alpha\} + \sum_\alpha e_\alpha \phi_{\rm ext}(\mathbf{x}_\alpha, t). \quad (1.89)$$

In the Coulomb gauge where $\nabla \cdot \mathbf{A} = 0$, the ordering of the operators is irrelevant. This interaction is linear in the vector potential, but there is also a second-order (or 'diamagnetic') term

$$H_{\rm dia} = \sum_\alpha \frac{e_\alpha^2 \mathbf{A}^2(\mathbf{x}_\alpha, t)}{2M_\alpha} \quad (1.90)$$

When calculations are pushed to second order in the $p \cdot A$-coupling, the diamagnetic interaction must be included as well, for consistency. This makes the 'book-keeping' in perturbation theory complicated.

**Gauge transformation.** There are essentially two schools that treat the scalar potential in very different ways.

(1) Either one is interested in electromagnetic fields on short scales compared to the wavelength (called "non-retarded limit"). Then one can ignore the vector potential and use only the scalar potential $\phi_{\text{ext}}$ to describe the matter-field interaction. This applies, for example, to the interaction of atoms or matter with electrons (scattering experiments).

(2) Or the wavelength is an important scale. Then one can even choose a gauge where $\phi_{\text{ext}} = 0$, and only the vector potential is nonzero.

Sometimes a mixture of the two schemes is needed, for example when light creates charge carriers, like in semiconductors or in the photoelectric effect.

### Electric dipole coupling

A simpler approach is possible, however. We first make the approximation that the field varies slowly on the scale of the displacements $\mathbf{x}_\alpha$ of the charges in the atom. Then we can replace, to lowest order, $\mathbf{A}(\mathbf{x}_\alpha, t) \approx \mathbf{A}(\mathbf{R}, t)$ where $\mathbf{R}$ is the atomic center of charge. This is called the 'long-wavelength approximation' that is well justified for fields near-resonant with typical atomic transitions. Within this approximation, we can find a simpler interaction Hamiltonian that is linear in the electromagnetic field. It is called the '$d \cdot E$' coupling and is strictly linear in the electric field:

$$\begin{aligned} H_{\text{int}} &= -\mathbf{d} \cdot \mathbf{E}(\mathbf{R}, t), \\ \mathbf{d} &= \sum_\alpha e_\alpha (\mathbf{x}_\alpha - \mathbf{R}), \end{aligned} \tag{1.91}$$

where $\mathbf{d}$ is the electric dipole moment of the atom relative to $\mathbf{R}$. This version of the $d \cdot E$ interaction can be derived from the minimal coupling Hamiltonian with a gauge transformation (see the exercises) from the minimal coupling Hamiltonian in the long-wavelength approximation, without invoking an additional approximation.

**Gauge transformation.** Change from the vector potential $\mathbf{A}(\mathbf{x}, t)$ to

$$\mathbf{A}'(\mathbf{x}, t) = \mathbf{A}(\mathbf{x}, t) - \nabla\chi(\mathbf{x}, t), \qquad \chi(\mathbf{x}, t) = (\mathbf{x} - \mathbf{R}) \cdot \mathbf{A}(\mathbf{R}, t) \tag{1.92}$$

where $\chi(\mathbf{x}, t)$ is called the 'gauge function'. It ensures that $\mathbf{A}'(\mathbf{R}, t) = 0$ at all times. Since the gauge function is time-dependent, the scalar potential also changes:

$$\phi'(\mathbf{x}, t) = \phi(\mathbf{x}, t) + \partial_t \chi(\mathbf{x}, t) = \phi(\mathbf{x}, t) + (\mathbf{x} - \mathbf{R}) \cdot \partial_t \mathbf{A}(\mathbf{R}, t) \tag{1.93}$$

If we simply insert these 'new potentials' into the minimal coupling Hamiltonian, we get

$$H'_{\text{dia}} = 0, \qquad H'_{\text{int}} = +\sum_{\alpha} e_\alpha \left\{ \phi_{\text{ext}}(\mathbf{x}_\alpha, t) + (\mathbf{x}_\alpha - \mathbf{R}) \cdot \partial_t \mathbf{A}(\mathbf{R}, t) \right\} \tag{1.94}$$

Note that the diamagnetic term cancels without further approximation. We expand the scalar potential for small $\mathbf{x}_\alpha - \mathbf{R}$ and get

$$H'_{\text{int}} = +\phi_{\text{ext}}(\mathbf{R}, t) \sum_{\alpha} e_\alpha + \sum_{\alpha} e_\alpha (\mathbf{x}_\alpha - \mathbf{R}) \cdot \left\{ \nabla \phi_{\text{ext}}(\mathbf{R}, t) + \cdot \partial_t \mathbf{A}(\mathbf{R}, t) \right\} \tag{1.95}$$

The first term cancels if the system of charges is globally neutral. In the second term, we recognize the expression of the electric field $\mathbf{E}(\mathbf{R}, t)$ in terms of the potentials. Hence we find the electric dipole Hamiltonian (1.91).

The advantages of the electric dipole coupling are: the atom couples directly to the field; there is no quadratic interaction term. One must not forget that between the two interactions, the wave function (the atomic state) differs by a unitary transformation. Otherwise, some matrix elements or transition rates may come out differently. This issue is discussed in great detail in the book 'Molecular Quantum Electrodynamics' by Craig & Thirunamachandran (1984) and in Chap. IV of 'Photons and Atoms – Introduction to Quantum Electrodynamics' by Cohen-Tannoudji & al. (1987).

### 1.11.2 Selection rules

Since the electric dipole moment determines the interaction with the light field, a few remarks on its matrix elements are in order. We take as a starting point the basis of the stationary states of an atom, described by the Hamiltonian (1.83). These states are typically described by quantum numbers like parity, angular momentum etc. The 'selection rules' specify for which states we know by symmetry that the matrix elements of the electric dipole moment vanish. In that case, the corresponding states are not connected by an 'electric dipole transition', or the transition is 'dipole-forbidden'.

**Parity.** We say that a state $|a\rangle$ has a defined parity $P_a = \pm 1$ when the electronic wave function $\psi(\{\mathbf{x}_\alpha\})$ "transforms like" $P_a\,\psi(\{\mathbf{x}_\alpha\})$ when all coordinates are transformed as $\mathbf{x}_\alpha \mapsto -\mathbf{x}_\alpha$. This means that

$$(\hat{P}\psi)(\{-\mathbf{x}_\alpha\}) = \psi(\{-\mathbf{x}_\alpha\}) = P_a\,\psi(\{\mathbf{x}_\alpha\}) \tag{1.96}$$

where $(\hat{P}\psi)$ denotes the action of the parity operator on the wave function.[4] If now $|a\rangle$ has a well-defined parity, then it is easy to show that $\langle a|\mathbf{d}|a\rangle = 0$ (see lecture). In addition, the matrix element $\langle a|\mathbf{d}|b\rangle$ is only nonzero when $|a\rangle$ and $|b\rangle$ have different parity. This is an example of a "selection rule". It provides a simple argument to exclude certain transitions from happening under the electric-dipole coupling. We shall see below that an off-diagonal matrix element like $\langle a|\mathbf{d}|b\rangle$ is essential when one wants to induce a "quantum jump" from one level to another with light.

**Energy.** Selection rules often arise when the system has certains symmetries. See a few examples below. The simplest symmetry is "translation in time", i.e., the system Hamiltonian does not depend on time. We then know from classical mechanics that energy is a conserved quantity. This is also true in quantum mechanics and quantum optics. The corresponding selection rule for atom-light interaction is the Bohr-Sommerfeld rule for the photon (angular) frequency $\omega$ that can induce a transition between two levels $|a\rangle$ and $|b\rangle$:

$$\hbar\omega = |E_b - E_a|. \tag{1.97}$$

This formula is in fact one of the birth certificates of quantum theory – remember that quantum mechanics was developed to explain the discrete frequencies observed in the radiation spectra of atoms.

**Angular momentum.** If there is no electron spin, this is given by $l$, and by $j = l \pm \frac{1}{2}$ for hydrogen-like atoms where one spin of a non-paired electron is present. The vector operator $\mathbf{d}$ transforms under rotation like a spin 1 (there are three different basis vectors). One can introduce a basis $\mathbf{e}_q$ ($q = -1, 0, 1$) that are eigenvectors of $L_3$ as well and write $\mathbf{d} = \sum_q d_q \mathbf{e}_q$. The product $\mathbf{e}_q|l, m\rangle$ then is an eigenstate of $L_3$ with eigenvalue $q + m$. Therefore, the matrix element with $|l', m'\rangle$ is only nonzero when $m' = q + m$. We find the selection rule

$$|m - m'| \leq 1.$$

[4]In the simple one-dimensional problems you remember from Quantum Mechanics I, wave functions that are 'even' or 'odd' have a well-defined parity. But there are also wave function that are neither even nor odd. One says that these do not have a well-defined parity.

In addition, the product states $\mathbf{e}_q|l,m\rangle$ can be expanded onto eigenstates of $\mathbf{L}^2$. The rules for the 'addition of angular momentum' imply that only angular momenta $l' = l-1, l, l+1$ occur in this expansion. This gives the selection rule

$$|l - l'| \le 1.$$

**Total momentum.** An atom that is in a plane wave state regarding its centre-of-mass motion, with momentum $\mathbf{P}$ receives an additional momentum $\hbar\mathbf{k}$ when a photon from a plane electromagnetic wave with wave vector $\mathbf{k}$ is absorbed. The corresponding 'recoil velocity' $\hbar\mathbf{k}/M$ is of the order of a few mm/s to a few cm/s for typical atoms. The atomic recoil plays an important role for atom deceleration and cooling with laser light.

### 1.11.3 Two-level atoms

For the rest of this lecture, it will be sufficient to write the atomic Hamiltonian in the form

$$H_A = \sum_n E_n |n\rangle\langle n|, \tag{1.98}$$

where the states $|n\rangle$ are the stationary states corresponding to the energy eigenvalue $E_n$. But even this form is too complicated: it contains too many terms when dealing with near-resonant laser light. This is the setting we shall focus on here. One can then retain only a few states to describe the atom.

#### Two-level observables

**Atomic Hamiltonian.** The standard notation for the two states is $|\mathrm{g}\rangle$ for the ground state and $|\mathrm{e}\rangle$ for the excited state. The Bohr frequency is often written $\omega_A = \omega_\mathrm{e} - \omega_\mathrm{g} > 0$. The atomic energy levels are often referenced to a zero energy lying between both states, this gives:

$$H_A = \frac{\hbar\omega_A}{2}|\mathrm{e}\rangle\langle\mathrm{e}| - \frac{\hbar\omega_A}{2}|\mathrm{g}\rangle\langle\mathrm{g}| \tag{1.99}$$

It is also useful to identify the two-dimensional Hilbert space of the two-level atom with the $\mathbb{C}^2$, using the basis vectors $(1,0)^T \leftrightarrow |\mathrm{e}\rangle$ and $(0,1)^T \leftrightarrow |\mathrm{g}\rangle$. The Hamiltonian then becomes the diagonal matrix

$$H_A = \frac{\hbar\omega_A}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = \frac{\hbar\omega_A}{2}\sigma_3 \tag{1.100}$$

where $\sigma_3$ is the third Pauli matrix. Indeed, it is obvious that a two-dimensional Hilbert space can be identified with the Hilbert space of a spin $1/2$.

Observables energy, inversion, dipole.

$$\mathbf{d} = \mathbf{d}_{ge}\sigma + \mathbf{d}_{ge}^{*}\sigma^{\dagger} = \mathbf{d}_{ge}|\mathrm{g}\rangle\langle\mathrm{e}| + \mathbf{d}_{ge}^{*}|\mathrm{e}\rangle\langle\mathrm{g}| \qquad (1.101)$$

where the vector of matrix elements of the dipole operator is $\mathbf{d}_{\mathrm{ge}} = \langle\mathrm{g}|\mathbf{d}|\mathrm{e}\rangle$. Only off-diagonal matrix elements because of the parity selection rule.

### 1.11.4 Resonance approximation

#### Interaction Hamiltonian

The interaction with a monochromatic laser field can be described by the Hamiltonian

$$\begin{aligned} H &= H_A - \mathbf{d}\cdot\mathbf{E}(t) \qquad (1.102) \\ \mathbf{E}(t) &= \mathbf{E}\,\mathrm{e}^{-\mathrm{i}\omega_L t} + \mathrm{c.c.} \\ \mathbf{d} &= \mathbf{d}_{ge}|\mathrm{g}\rangle\langle\mathrm{e}| + \mathrm{h.c.} \end{aligned}$$

where the complex vector $\mathbf{E}$ gives the amplitude of the electric field. It is evaluated at the position of the atom, we drop this dependence here. The laser (angular) frequency is $\omega_L$. The term $\mathbf{E}\,\mathrm{e}^{-\mathrm{i}\omega_L t}$ is called the 'positive frequency part' of the field: its time evolution is the same as for a solution of the time-dependent Schrödinger equation (with positive energy $\hbar\omega_L$).

We re-write Eq.(1.102) in terms of two separately hermitean operators

$$\begin{aligned} -\mathbf{d}\cdot\mathbf{E}(t) &= \frac{\hbar}{2}\left(\Omega\,\mathrm{e}^{-\mathrm{i}\omega_L t}|\mathrm{e}\rangle\langle\mathrm{g}| + \mathrm{h.c.}\right) \\ &\quad + \frac{\hbar}{2}\left(\Omega'\,\mathrm{e}^{\mathrm{i}\omega_L t}|\mathrm{e}\rangle\langle\mathrm{g}| + \mathrm{h.c.}\right) \qquad (1.103) \\ \frac{\hbar\Omega}{2} &= -\mathbf{d}_{ge}^{*}\cdot\mathbf{E}, \qquad (\text{ Rabi frequency }) \qquad (1.104) \end{aligned}$$

where $\Omega$ is a complex-valued frequency, and $\Omega'$ has a similar expression as Eq.(1.104). (We follow the Paris convention and notation for $\Omega$.)

If the field is quantized, Eq.(1.102) applies in similar form in the interaction picture and involves photon annihilation operators $a_L$ in place of the complex amplitude $\mathbf{E}$ (and creation operators $a_L^{\dagger}$ in place of $\mathbf{E}^{*}$. The fully quantized interaction Hamiltonian is derived from the quantized field operator and takes the form:

$$\begin{aligned} &-\mathbf{d}(t)\cdot\mathbf{E}(t) \mapsto \\ &-\sum_k \mathcal{E}_k\left(a_k(t)\mathbf{f}_k\cdot\mathbf{d}(t) + a_k^{\dagger}(t)\mathbf{f}_k^{*}\cdot\mathbf{d}(t)\right) \qquad (1.105) \end{aligned}$$

where $\mathcal{E}_k = \sqrt{\hbar\omega_k/(2\varepsilon_0 V)}$ is the electric field amplitude at the one-photon level and $\mathbf{f}_k = \mathbf{f}_k(\mathbf{R})$ is the normalized mode function, evaluated at the position $\mathbf{R}$ of the atom. We have written Eq.(1.105) in the "interaction picture" where all operators carry their "free" time dependence. For the photon annihilation operator $a_k(t) = a_k\,\mathrm{e}^{-\mathrm{i}\omega_k t}$, which is the operator in the Heisenberg picture under the free field Hamiltonian. The time dependence of the (freely evolving) dipole operator is given by $\mathbf{d}(t) = \exp(\mathrm{i}H_A t)\mathbf{d}\exp(-\mathrm{i}H_A t)$.

In order to examine what happens to an atom illuminated by a laser field, we make the *Ansatz*

$$|\psi(t)\rangle = \tilde{c}_e(t)\,\mathrm{e}^{-\mathrm{i}\omega t/2}|\mathrm{e}\rangle + \tilde{c}_g(t)\,\mathrm{e}^{+\mathrm{i}\omega t/2}|\mathrm{g}\rangle \tag{1.106}$$

where the frequency $\omega$ is chosen later. The amplitudes describe the two-level system in a *picture* that differs from the usual choice $c_e(t)$, $c_g(t)$. The Schrödinger equation for $\tilde{c}_e$ contains a correction term because of the time-dependent exponential. One gets

$$\begin{aligned}\mathrm{i}\hbar\partial_t\tilde{c}_e = \left(E_e - \frac{\hbar\omega}{2}\right)\tilde{c}_e &+ \frac{\hbar}{2}\Omega\,\mathrm{e}^{-\mathrm{i}(\omega_L-\omega)t}\,\tilde{c}_g \\ &+ \frac{\hbar}{2}\Omega'\,\mathrm{e}^{\mathrm{i}(\omega_L+\omega)t}\,\tilde{c}_g\end{aligned} \tag{1.107}$$

Now, we can take the choice $E_e = \frac{1}{2}\hbar\omega_A$ for the excited state energy. There are now two "natural choices" for $\omega$:

(1) **interaction picture**: $\omega = \omega_A$, and the first term disappears. The time dependence of $\tilde{c}_e$ is then only due to the atom-laser interaction. This picture is suitable for perturbation theory.

(2) **rotating frame**: $\omega = \omega_L$, and the second term becomes time-independent. This picture is suitable for concrete calculations in quantum optics, once we have convinced outselves what is the meaning of the third term.

### Time-dependent perturbation theory

We now make the choice (1) of the interaction picture and solve the equation for $\tilde{c}_e$ with the help of time-dependent perturbation theory. This proceeds by identifying the interaction Hamiltonian as a "small term" and by counting ascending powers.

At the order zero, $H_A$ is the only Hamiltonian. Keeping in mind the choice $\omega = \omega_A$, Eq.(1.107) reduces to

$$\partial_t \tilde{c}_e^{(0)} = 0, \qquad \partial_t \tilde{c}_g^{(0)} = 0 \tag{1.108}$$

where the equation for $\tilde{c}_g$ is similar to Eq.(1.107). As expected from the analogy to the time-dependent Schrödinger equation (for the “free” atom), the amplitudes are constants at order zero. The natural initial condition “atom is in state $|\mathrm{g}\rangle$ translates into

$$\tilde{c}_e^{(0)}(t) = 0, \qquad \tilde{c}_g^{(0)}(t) = 1. \tag{1.109}$$

To the first order, we get from Eq.(1.107)

$$\mathrm{i}\hbar\partial_t \tilde{c}_e^{(1)} = \frac{\hbar}{2}\Omega\, \mathrm{e}^{-\mathrm{i}(\omega_L - \omega_A)t}\, \tilde{c}_g^{(0)} + \frac{\hbar}{2}\Omega'\, \mathrm{e}^{\mathrm{i}(\omega_L + \omega_A)t}\, \tilde{c}_g^{(0)} \tag{1.110}$$

Now, since we know $c_g^{(0)}(t)$ as a (constant) function of time, this can be integrated immediately to give

$$c_e(t) = c_e(0) + \frac{\Omega}{2}\frac{\mathrm{e}^{-\mathrm{i}(\omega_L - \omega_A)t} - 1}{\omega_L - \omega_A} - \frac{\Omega'}{2}\frac{\mathrm{e}^{\mathrm{i}(\omega_A + \omega_L)t} - 1}{\omega_A + \omega_L} \tag{1.111}$$

The two terms in this result have distinct physical interpretations, related to the denominators.

**Absorption.** The first denominator leads to a ‘large’ result when $\omega_A = \omega_L$. One says that the atom went from the state $|g\rangle$ to the higher-lying state $|e\rangle$ by absorbing one ‘energy quantum’ (‘photon’). (Recall that the amplitude $c_e$ for the state $|e\rangle$ is increased in Eq.(1.111).) This process is governed by the ‘positive frequency’ component $\Omega\, \mathrm{e}^{-\mathrm{i}\omega_L t}$ of the interaction Hamiltonian (corresponding to the positive frequency component of the electromagnetic field). In the quantized description of the light field, this component corresponds to an ‘annihilation operator’ that removes one photon from the field. If we fix the states $|g\rangle$ and $|e\rangle$ such that the condition for absorption is satisfied, then the second term in Eq.(1.111) has a ‘large’ denominator, $\omega_A + \omega_L \approx 2\omega_A$. This term is therefore much smaller than the first one, by a factor of the order $\mathcal{O}(10^{-6})$ for laser fields of reasonable intensity (see exercise). This suggests that we can neglect this term. This approximation is called the ‘resonance approximation’ (or the ‘rotating wave approximation’, an admittedly strange name). If we keep the non-resonant term, we deal in the quantum theory with a ‘virtual’ process where the atom passes into a state with a higher energy and at the same time, a photon is created.

**Emission.** If we had started with the atom in the excited state $|\mathrm{e}\rangle$, one would get a resonant contribution again for $\omega_L = \omega_A$, with a large amplitude being created in $|\mathrm{g}\rangle$. This corresponds to a transition with the energy balance $E_e = E_g + \hbar\omega_L$: the atom makes a transition to a lower-lying state, and in the quantized field description, a 'photon' is created (by the creation operator $a_k^\dagger$ in the expansion of the field operator). The non-resonant term in this setting would correspond to the atom decaying to the ground state and absorbing a photon, clearly a virtual process.

To summarize, in the resonance approximation, we only retain those parts of the interaction Hamiltonian where the excitation of the atom (the operator $\sigma^\dagger = |\mathrm{e}\rangle\langle\mathrm{g}|$ is accompanied by a positive frequency laser field $\mathbf{E}\,\mathrm{e}^{-\mathrm{i}\omega_L t}$. This approximation is consistent with the two-level approximation where right from start, we only considered atomic levels whose Bohr frequencies are near-resonant with the laser.

This approximation is possible for a 'detuning' $\Delta = \omega_L - \omega_A$ small compared to the typical differences between atomic transition frequencies. This condition is easily achieved since transition frequencies (spectral lines) differ easily by energies of the order of $1\,\mathrm{eV}$, and this is a 'huge' detuning to drive an atomic transition.

**Remark.** The description of absorption and emission, as we encounter it here, does not explicitly require the quantization of the light field. These processes also occur in a 'classical' time-dependent potential because energy is not conserved there, as is well known in classical mechanics. One can push the analogy even further: a weak monochromatic excitation of a mechanical system reveals the system's 'resonance frequencies'. For an atom, these are apparently given by the Bohr frequencies. The only difference to a mechanical system is that we are inclined to use different names for the excitations with positive and negative frequencies, since in the atomic energy spectrum, there is a definite difference between 'going up' and 'going down' (there exists a ground state).

### Atomic polarizability

The perturbative calculation can be used to determine the dipole moment that the laser field induces in the atom. This dipole is, within the lowest order in the perturbation, linear in the amplitude $\mathbf{E}$ of the laser, or equivalently, in the Rabi frequency $\Omega$. As is discussed in more detail in the exercises (Problem 3.2), the polarizability is defined by equating the average (induced) dipole moment with a function linear in the laser amplitude,

$$\langle\psi(t)|\,(\mathbf{d}_{\rm ge}\sigma + {\rm h.c.})\,|\psi(t)\rangle = \alpha(\omega_L)\mathbf{E}\,{\rm e}^{-{\rm i}\omega_L t} + {\rm h.c.} \tag{1.112}$$

Note that the asymptotic regime $t \to \infty$ is taken here where the atomic dipole oscillates at the frequency of the external field. Now, for the initial condition that the atom starts in its ground state, the polarizability is

$$\alpha_{\rm g}(\omega) = \frac{(2\omega_{\rm eg}/\hbar)\mathbf{d}_{\rm ge}\otimes\mathbf{d}^*_{\rm ge}}{\omega^2_{\rm eg} - \omega^2} \tag{1.113}$$

where two peaks at $\omega = \pm\omega_{\rm eg}$ appear. These peaks are not damped – which is an artefact because we ignored any damping processes to far. In practice, processes called spontaneous emission, thermal absorption, and dephasing remove the divergence of the polarizability $\omega = \pm\omega_{\rm eg}$ and lead to a Lorentzian profile with a nonzero linewidth. To calculate this, we need the quantum theory of the light field.

**RWA Hamiltonian in the rotating frame**

We come back to the (near-)resonant interaction: it can be described by the (effective) Hamiltonian

$$H_{AL} = -\mathbf{d}^*_{ge}\mathbf{E}\,{\rm e}^{-{\rm i}\omega_L t}\sigma^\dagger - \mathbf{d}_{ge}\mathbf{E}^*\,{\rm e}^{{\rm i}\omega_L t}\sigma \tag{1.114}$$

This is called the "rotating wave approximation", a physically more transparent name would be "resonance approximation". The change into the picture (2) (rotating frame) mentioned on p.45 above ($\omega = \omega_L$) corresponds to the unitary transformation

$$|\psi(t)\rangle = {\rm e}^{-{\rm i}\frac{\omega_L t}{2}\sigma_3}|\tilde{\psi}(t)\rangle \tag{1.115}$$

This gives for the state $|\tilde{\psi}(t)\rangle$ the following Hamiltonian

$$H = -\frac{\hbar\Delta}{2}\sigma_3 + \frac{\hbar}{2}\left(\Omega^*\sigma + \Omega\sigma\right) \qquad \Delta = \omega_L - \omega_A \tag{1.116}$$

where the time-dependence of Eq.(1.114) has disappeared (exactly) and where only the *detuning* $\Delta$ instead of the laser frequency appears (Paris convention for the sign of $\Delta$).

With a quantized field, add the Hamiltonian $H_F$ for the field and make the replacement

$$\frac{\hbar}{2}\left(\Omega^* \sigma + \sigma^\dagger \Omega\right) \mapsto -\sum_k \mathcal{E}_k \left\{ a_k^\dagger \mathbf{f}_k^* \cdot \mathbf{d}_{\mathrm{eg}}^* \sigma + \sigma^\dagger a_k \mathbf{f}_k \cdot \mathbf{d}_{\mathrm{eg}} \right\} \tag{1.117}$$

One could also read this as an operator-valued Rabi frequency per mode, $\hat{\Omega}_k$.

**Spin 1/2 analogy.** We now come back to the spin 1/2 analogy. The Hamiltonian (1.114) with the atomic energies and the atom–laser interaction has the same form as the Hamiltonian for a spin 1/2 in a time-dependent magnetic field,

$$H_{\mathrm{spin}} = \boldsymbol{\sigma} \cdot \mathbf{B}(t) \tag{1.118}$$

where $\boldsymbol{\sigma}$ is the vector of Pauli matrices and the 'magnetic field' $\mathbf{B}(t)$ actually has the dimensions of an energy (we took a unity magnetic moment). The magnetic field rotates at the laser frequency around the $x_3$-axis:

$$\mathbf{B}(t) = \frac{\hbar}{2} \begin{pmatrix} \Omega \cos \omega_L t \\ \Omega \sin \omega_L t \\ \omega_A \end{pmatrix} \tag{1.119}$$

It is useful to change the coordinate frame such that it co-rotates with this field (this is the 'rotating frame'). In this frame, the 'effective magnetic field' is static[5],

$$\mathbf{B}_{\mathrm{eff}} = \frac{\hbar}{2} \begin{pmatrix} \Omega \\ 0 \\ \omega_A \end{pmatrix}. \tag{1.120}$$

The transformation into the rotating frame also changes the wave function of our two-state particle by a unitary transformation (this is the way a two-component spinor transforms under a rotation of the coordinate axes)

$$U(t) = \exp\{-\mathrm{i}\omega_L t \sigma_3/2\} = \begin{pmatrix} \mathrm{e}^{-\mathrm{i}\omega_L t/2} & 0 \\ 0 & \mathrm{e}^{\mathrm{i}\omega_L t/2} \end{pmatrix} \tag{1.121}$$

We observe that this is the transformation we used in Eq.(1.106) to go into the interaction picture (on resonance where $\omega_L = \omega_A$). This unitary transformation being time-dependent, we get also a modification of the Hamiltonian proportional to $-\mathrm{i}\hbar U^\dagger \partial_t U = -\hbar\omega_L \sigma_3$. All told, we find the Hamiltonian *in the rotating frame*

$$\boxed{H = -\frac{\hbar\Delta}{2}\sigma_3 + \frac{\hbar\Omega}{2}\sigma_1} \tag{1.122}$$

where the *detuning* is given by the difference between the laser frequency and the atomic transition frequency

$$\Delta = \omega_L - \omega_A. \tag{1.123}$$

[5]If we had kept the nonresonant terms in the Hamiltonian, the magnetic field would also show a time-dependent component rotating at the frequency $2\omega_L$.

Note that the laser and atomic frequencies have disappeared from the Hamiltonian and only their difference (the detuning) occurs. As a consequence, the relevant time scales (given by $1/\Delta$ and $1/\Omega$) are typically much longer than the optical period $2\pi/\omega_L$. On these long time scales, nonresonant processes remain 'virtual' and cannnot be directly observed. This is consistent with the neglect of nonresonant levels (two-state approximation) and of the nonresonant two-state coupling (rotating wave approximation).

### 1.11.5 Rabi oscillations

The most simple case of atom-laser dynamics is a laser 'on resonance', i.e., $\omega_L = \omega_A$. The Schrödinger equation for the Hamiltonian (1.116) yields (we drop the tildes)

$$\mathrm{i}\hbar\dot{c}_\mathrm{e} = \frac{\hbar\Omega}{2}c_\mathrm{g} \tag{1.124}$$

$$\mathrm{i}\hbar\dot{c}_\mathrm{g} = \frac{\hbar\Omega}{2}c_\mathrm{e}. \tag{1.125}$$

where the Rabi frequency is chosen real for simplicity. With the initial conditions $c_\mathrm{g}(0) = 1,\ c_\mathrm{e}(0) = 0$, the solution is

$$\dot{c}_\mathrm{e} = -\mathrm{i}\sin(\Omega t/2) \tag{1.126}$$

$$c_\mathrm{g} = \cos(\Omega t/2). \tag{1.127}$$

The excited state probability thus oscillates between $0$ and $1$ at a frequency $\Omega/2$. This phenomenon is called 'Rabi flopping'. It differs from what one would guess from ordinary time-dependent perturbation theory where one typically gets linearly increasing probabilities. That framework, however, applies only if the final state of the transition lies in a continuum which is not the case here. Rabi flopping also generalizes the perturbative result (1.110) which would give a quadratic increase $|c_\mathrm{e}|^2 \propto t^2$ that cannot continue for long times. But instead of saturating, the atomic population returns to the ground state.

Every experimentalist is very happy when s/he observes Rabi oscillations. It means that any dissipative processes have been controlled so that they happen at a slower rate. In a realistic setting, one gets a damping of the oscillation amplitude towards equilibrium populations.

**Rabi pulses.** Rabi oscillations with a fixed interaction time are often used to implement coherent operations on an atom or spin. The corresponding evolution operator is given by (we focus on the resonant case)

$$U_\theta = \exp\{-\mathrm{i}\theta\sigma_1/2\} = \cos(\theta/2) - \mathrm{i}\sigma_1 \sin(\theta/2) \tag{1.128}$$

with $\theta = \Omega t$. After one cycle of Rabi oscillations, $\Omega t = 2\pi$ (a '$2\pi$-pulse'), the atom returns to its ground state — but its wave function has changed sign. This sign change is well-known from spin 1/2 particles: the corresponding unitary transformation reads

$$U_{2\pi} = \cos(\pi) - \mathrm{i}\sigma_1 \sin(\pi) = -1 \tag{1.129}$$

A more interesting manipulation is a '$\pi$-pulse', $\Omega t = \pi$, that flips the ground and excited state:

$$U_\pi = \cos(\pi/2) - \mathrm{i}\sigma_1 \sin(\pi/2) = -\mathrm{i}\sigma_1 \tag{1.130}$$

Finally, a '$\pi/2$-pulse' takes the atom into a superposition of ground and excited states with equal weight (a Bloch vector on the equator of the Bloch sphere)

$$\begin{aligned} U_{\pi/2} &= \cos(\pi/4) - \mathrm{i}\sigma_1 \sin(\pi/4) = \frac{1 - \mathrm{i}\sigma_1}{\sqrt{2}} \\ U_{\pi/2}|g\rangle &= \frac{1}{\sqrt{2}}|g\rangle - \frac{\mathrm{i}}{\sqrt{2}}|e\rangle \end{aligned}$$

If the laser is shut off after such a pulse, the Bloch vector will continue to rotate along the equator at the frequency $\Delta$.

## 1.12 Dissipation and open system dynamics

We now describe how the dynamics of the atomic Bloch vector is modified when so-called dissipative processes are taken into account. These processes occur because the two-level system is not closed: it is in contact with the electromagnetic field that carries away energy and information (entropy). In addition, it is subject to vacuum fluctuations (see Chapter **??**). The challenge of including dissipation into quantum optics is that the equations of motion must be compatible some basic principles of quantum mechanics: states cannot evolve in an arbitrary way because probabilities remain positive, for example.

### 1.12.1 Spontaneous emission

As a consequence of the coupling to the quantized electromagnetic field, the excited state of the two-level atom decays by emitting a photon into an 'empty' mode of the electromagnetic field. This phenomenon can conveniently be described by the equations of 'radioactive decay' (a pair of 'rate equations')

$$\frac{\mathrm{d}p_{\mathrm{e}}}{\mathrm{d}t} = -\gamma p_{\mathrm{e}}, \qquad \frac{\mathrm{d}p_{\mathrm{g}}}{\mathrm{d}t} = +\gamma p_{\mathrm{e}} \tag{1.131}$$

The rate $\gamma$ gives the probability per unit time of emitting a photon and putting the atomic population from the excited state down to the ground state. The total population is conserved, as it should be for a process where the atom just changes its internal state. (In radioactive decay, 'e' would by a plutonium and 'g' an uranium atom, and the 'photon' an $\alpha$-particle.) In terms of the third component of the Bloch vector (the inversion), we have the following equation

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_3\rangle\right|_{\mathrm{decay}} = -\gamma(\langle\sigma_3\rangle + 1) \tag{1.132}$$

We also need a prescription how to take into account such a process in the dynamics of off-diagonal elements of the density matrix like $\rho_{\mathrm{eg}}$. These equations cannot be chosen arbitrarily because we require that the density operator $\rho$ remains positive under time evolution. We discuss this in more detail later in the lecture. The result is that also the dipole components of the Bloch vector decay exponentially

$$\left.\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma\rangle\right|_{\mathrm{decay}} = -\Gamma\langle\sigma\rangle \tag{1.133}$$

The rate must satisfy the inequality $\Gamma \geq \gamma/2$, otherwise one can find initial conditions that evolve into states outside the Bloch sphere (i.e., a density matrix with negative probabilities). The process (1.133) is sometimes called "dephasing" or "decoherence" because it happens when the relative phase of a superposition state, $\alpha|\mathrm{g}\rangle + \beta\,\mathrm{e}^{\mathrm{i}\theta}|\mathrm{e}\rangle$ is "diffusing" in time (with a variance that increases linearly with $t$ like in Brownian motion). The off-diagonal elements of the density matrix are sometimes called "coherences", they determine to what extent one has a genuine quantum superposition, distinct from a "classical" (or thermodynamical) mixture.

To compute the spontaneous decay rate $\gamma$, we need Fermi's Golden Rule, a standard result from time-dependent perturbation theory. We derive this after we have learned about the quantization of the electromagnetic field, but the result is

$$\gamma = \frac{|\mathbf{d}_{\rm ge}|^2 \omega_{\rm eg}^3}{3\pi\varepsilon_0 \hbar c^3} \tag{1.134}$$

with a typical value $1/\gamma \sim 10\,\mathrm{ns}$ for transitions in the visible range and dipole moments of the order of the Bohr magneton. We just note the scaling with the fine structure constant

$$\frac{\gamma}{\omega_{\rm eg}} \sim \frac{e^2}{3\pi\varepsilon_0 \hbar c} \frac{a_0^2}{\lambda_{\rm eg}^2} \sim \alpha_{\rm fs}^3 \tag{1.135}$$

On the scale of the Bohr frequencies in the atom, the decay is thus very slow.

### 1.12.2 Bloch equations

One finds by applying the model for spontaneous decay that the average spin vector evolves according to the following set of equations, now including dissipation,

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma\rangle = -\left(\mathrm{i}\omega_A + \Gamma\right)\langle\sigma\rangle + \mathrm{i}(\Omega/2)\,\mathrm{e}^{-\mathrm{i}\omega_L t}\,\langle\sigma_3\rangle \tag{1.136}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_3\rangle = -\gamma(\langle\sigma_3\rangle + 1) + \mathrm{i}\left(\Omega^*\,\mathrm{e}^{\mathrm{i}\omega_L t}\langle\sigma\rangle - \Omega\,\mathrm{e}^{-\mathrm{i}\omega_L t}\langle\sigma^\dagger\rangle\right) \tag{1.137}$$

This is written within the rotating wave approximation (the Hamiltonian (1.114)) but not yet in the frame rotating at the laser frequency $\omega_L$ (choice (2) on p.45). Using the transformation (1.115), one finds $\sigma(t) = \tilde\sigma(t)\,\mathrm{e}^{-\mathrm{i}\omega_L t}$ with

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\tilde\sigma\rangle = (\mathrm{i}\Delta - \Gamma)\,\langle\tilde\sigma\rangle + \mathrm{i}(\Omega/2)\,\langle\sigma_3\rangle \tag{1.138}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_3\rangle = -\gamma(\langle\sigma_3\rangle + 1) + \mathrm{i}\left(\Omega^*\langle\tilde\sigma\rangle - \Omega\langle\sigma^\dagger\rangle\right) \tag{1.139}$$

Here, the frequencies enter via the detuning $\Delta = \omega_L - \omega_A$ (Paris convention, some authors use the other sign).

### 1.12.3 Rate equation limit

Assume that dephasing rate $\Gamma$ is "faster" than all other time scales. "Adiabatic elimination of coherences" leads to

$$\langle\tilde\sigma\rangle_{\rm ad} \approx \frac{\mathrm{i}(\Omega/2)\,\langle\sigma_3\rangle}{\Gamma - \mathrm{i}\Delta} \tag{1.140}$$

found by solving Eq.(1.138) in the steady state. *Idea*: find stationary state after an initial transient. Time scale for transient is $1/\Gamma$, hence short by assumption. Assume that $\langle\sigma_3\rangle$ evolves slowly on this scale ("adiabatic following" of $\langle\tilde{\sigma}\rangle$).

Insert into Eq.(1.139) for the inversion gives the *rate equation*

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle\sigma_3\rangle &= -\gamma(\langle\sigma_3\rangle + 1) - 2\,\mathrm{Im}\,(\Omega^*\langle\tilde{\sigma}\rangle_{\mathrm{ad}}) \\ &= -\gamma(\langle\sigma_3\rangle + 1) - \underbrace{\frac{\Gamma|\Omega|^2}{\Gamma^2+\Delta^2}}_{\text{absorption}}\langle\sigma_3\rangle \end{aligned} \qquad (1.141)$$

The last term is the rate by which the two-level system absorbs energy from the laser (and gets excited). Indeed, for initial conditions in the ground state $\langle\sigma_3\rangle = -1$, one gets a positive derivative from Eq.(1.141).

**Exercise.** Calculate the time-averaged power absorbed by the two-level atom using the formula from mechanics, $P_{\mathrm{abs}} = \langle\dot{\mathbf{d}}(t)\cdot\mathbf{E}(t)\rangle$ by taking the time average and the quantum expectation value in the stationary state. Compare $P_{\mathrm{abs}}/\hbar\omega_L$ to the absorption rate in Eq.(1.141).

Rate equations are often used in condensed matter when fluorescent systems like molecules or quantum dots are embedded in a solid environment. In that case, the contact with the surrounding atoms and molecules leads to a large value for $\Gamma$. In this limit, the induced dipole moment is quite small (see Eq.(1.140)), and the relevant dynamics is well approximated by considering only probabilities (occupation numbers, the inversion).

### 1.12.4 Collapse and revival

In this paragraph, we are using concepts from the quantized description of the field. Details on that in the Chapter in field quantization.

If the light field is described as a single quantized mode, an additional feature occurs in the Rabi oscillations. The key point is that the coupling Hamiltonian, $g(a^\dagger\sigma + \sigma^\dagger a)$, now couples the states $|\mathrm{g}, n\rangle$ and $|\mathrm{e}, n-1\rangle$ where $n$ is the photon number. These states are split (on resonance) in energy by the 'Rabi splitting' $g\sqrt{n}$. Recall that this splitting was $|\Omega|$ for a classical laser field, proportional to the field amplitude. This is mimicked by the scaling with $\sqrt{n}$ since the photon number $n$ is proportional to the field intensity.

In each sub-space spanned by $|\mathrm{g}, n\rangle$ and $|\mathrm{e}, n-1\rangle$, the system thus performs Rabi oscillations with a slightly different frequency. If one starts with a coherent state $|\alpha\rangle$ for the field mode, the Rabi oscillations will thus evolve at a mean frequency $\approx g|\alpha|$, but at large times, the oscillations will 'get out of phase'. This leads to a 'collapse' of the Rabi oscillation amplitude, as illustrated in Figure 1.2. It happens on the time scale $1/g$ which is a factor $|\alpha|$ times longer than the period

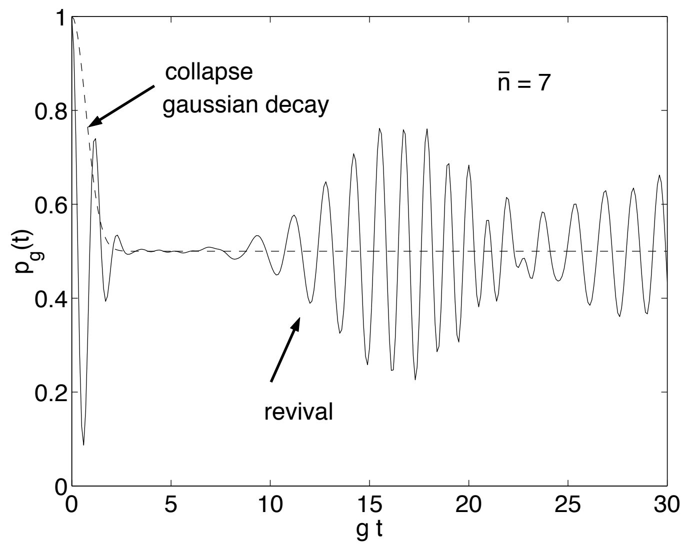



Figure 1.2: Ground state occupation $p_{\mathrm{g}}(t)$ for a two-level atom coupled to a single mode, initially in the coherent state $|\alpha\rangle$ with $|\alpha|^2 = 7$. Time is in units of the 'single-photon Rabi frequency' $g$.

of the initial Rabi oscillations. At still larger times, of order $|\alpha|/g$, the amplitude of the oscillations 'revives' again. This is due to the fact that the Rabi frequencies form a discrete, incommensurable set (the frequencies are proportional to the irrational numbers $\sqrt{n}$, on resonance). A more detailed analysis is presented in a later section.

## 1.13 More notes on quantum dissipation

Material not covered in WS 16/17.

### 1.13.1 State of a two-level system

This section contains details on a somewhat more axiomatic approach than what we did in the lecture. You may jump directly to Eqs.(1.151, 1.152) that have been discussed in the Problem sessions.

*The expectation values $\langle\sigma_3\rangle$ and $\langle\sigma\rangle$ completely specify the state of the two-level system.*

Why is this so? A general observable is a hermitean $2 \times 2$ matrix. All these matrices are linear combinations of Pauli matrices

$$\begin{aligned} A &= \begin{pmatrix} a_{ee} & a_{eg} \\ a_{ge} & a_{gg} \end{pmatrix} = \frac{a_{ee} + a_{gg}}{2}\mathbb{1} + \frac{a_{ee} - a_{gg}}{2}\sigma_3 + a_{ge}\sigma + a_{eg}\sigma^\dagger \\ &= \frac{a_{ee} + a_{gg}}{2}\mathbb{1} + \sum_j a_j\sigma_j, \qquad (1.142) \\ \sigma_1 &= \sigma + \sigma^\dagger \qquad (1.143) \\ \sigma_2 &= \mathrm{i}(\sigma - \sigma^\dagger) \qquad (1.144) \end{aligned}$$

with real coefficients $a_j$.

The above statement is true even for a more general definition of a *state* than you may be used to. In the axiomatic language of quantum information, a state is a mapping from a set of observables to their expectation values

$$\rho : A \mapsto \rho(A) = \langle A\rangle_\rho \qquad (1.145)$$

Linear map with $\rho(\mathbb{1}) = 1$ (real or complex coefficients depending on choice of observable algebra) and $\rho(A)$ real for a hermitean $A$.

Now, the action of this map is determined by evaluation on basis vectors = Pauli matrices for a two-level system:

$$\rho(A) = \frac{a_{ee} + a_{gg}}{2}\rho(\mathbb{1}) + \sum_j a_j\rho(\sigma_j) = \frac{a_{ee} + a_{gg}}{2}\rho(\mathbb{1}) + \sum_j a_j s_j \qquad (1.146)$$

with components of *Bloch vector* $\mathrm{s} = (s_1, s_2, s_3)$.

This definition is more general than complex linear combinations of $|\mathrm{e}\rangle$ and $|\mathrm{g}\rangle$. These states play a special role and are called *pure states*. They also correspond to special observables: *projectors*

$$\mathbb{P}_\phi = |\phi\rangle\langle\phi| \qquad (1.147)$$

This is also a hermitean operator with eigenvalues $0$ or $1$. A physical state has the property

$$\rho(\mathbb{P}_\phi) \geq 0 \quad \text{for all } |\phi\rangle \tag{1.148}$$

Physical interpretation: this is the probability of finding the system in the pure state $|\phi\rangle$, which clearly must be a positive number.

Definition of *density matrix* (or *density operator*): any linear map on the vector space of observables can be represented by a suitable linear form

$$\rho(A) = \mathrm{tr}(\bar{\rho}A) \tag{1.149}$$

where $\bar{\rho}$ is a hermitean operator. This rule corresponds to the usual calculation of expectation values for mixed states in quantum statistics. In a finite-dimensional system, it corresponds to the duality between linear forms and vectors: each linear form can be represented as a scalar product with a suitable vector. This becomes the Riesz representation theorem in an infinite-dimensional Hilbert space.

Using this for projector observables, we find from Eq.(1.148):

$$0 \leq \rho(\mathbb{P}_\phi) = \mathrm{tr}(\bar{\rho}|\phi\rangle\langle\phi|) = \langle\phi|\bar{\rho}|\phi\rangle \tag{1.150}$$

Hence the diagonal elements of the density matrix are positive, in any basis. This connects again to the interpretation of the probability of finding the system in the state $|\phi\rangle$.

Density operator as observable itself. Expectation value is called purity

$$\mathrm{Pu}(\rho) = \langle\bar{\rho}\rangle_\rho = \mathrm{tr}(\bar{\rho}^2) = \ldots = \frac{1}{2}(1 + \mathbf{s}^2) \tag{1.151}$$

Calculation uses representation in terms of Bloch vector and Pauli matrices

$$\bar{\rho} = \frac{1}{2}\left(\mathbb{1} + \sum_j s_j \sigma_j\right) = \frac{\mathbb{1} + \mathbf{s}\cdot\boldsymbol{\sigma}}{2} \tag{1.152}$$

### How to generate mixed states

If a quantum system is closed and can be prepared in a pure state, then the time evolution is simply Hamiltonian, $|\psi(t)\rangle = U(t)|\psi(0)\rangle$, and we don't have to talk about quantum dissipation. This is not so in many settings, however.

There are a few examples how mixed (or non-pure) states arise.

**(i) Initial mixed state.** If the initial state is prepared within some probabilistic scheme, we have to work with an initial density matrix $\rho(0) \neq |\psi(0)\rangle\langle\psi(0)|$. This translates our incomplete knowledge about the initial conditions. Recall that density matrices can be "mixed" by forming so-called convex linear combinations

$$\rho = p\rho_1 + q\rho_2, \qquad p + q = 1, \quad p, q \geq 0, \quad \operatorname{tr}\rho_{1,2} = 1 \tag{1.153}$$

where the two density matrices $\rho_{1,2}$ are both normalized and $p$, $q$ can be interpreted as probabilities for preparing the two.

The time evolution is still simple if the system is closed (Hamiltonian evolution):

$$\rho(t) = U(t)\rho(0)U^\dagger(t) \tag{1.154}$$

or in differential form (the von Neumann equation)

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho = \frac{1}{\mathrm{i}\hbar}[H, \rho] \tag{1.155}$$

A typical example is an initial state prepared with a given temperature, $\rho(0) \propto \exp(-H_I/T)$. Interesting dynamics then happens only if $H_I \neq H$.

**Exercise.** We actually don't need to solve the von Neumann equation (1.155): by expanding $\rho(0)$ in terms of its eigenvectors, we can just evolve these eigenvectors under Schrödinger's equation and mix the final states. By linearity, the result is the same.

**(ii) Reduced density matrix.** The second example is that of a "system" $S$ coupled to another one, let's call it "bath" or "environment" $B$. In this setting, we restrict ourselves (*by construction*) to observables that do not give any information about the state of the environment. These observables can be written in the form $\hat{A} \otimes \mathbb{1}_B$ where $\mathbb{1}_B$ is the unit operator in the environment's Hilbert space. The key observation is that the expectation values for all system observables of this type can be calculated with the help of a density operator $\rho$ for the system,

$$\langle \hat{A} \otimes \mathbb{1}_B \rangle_{S+B} = \operatorname{tr}(\hat{A}\rho_S) \tag{1.156}$$

Note that there are many authors who do not make the distinction between $\hat{A} \otimes \mathbb{1}_B$ and $\hat{A}$. The object $\rho_S$ is called a *reduced density operator* (or matrix). It is sometimes written

$$\rho_S = \operatorname{tr}_B \rho_{S+B} = \operatorname{tr}_B |\psi_{S+B}\rangle\langle\psi_{S+B}| \tag{1.157}$$

where the last writing assumes that system+environment are in a pure state $|\psi_{S+B}\rangle$. This procedure is called "taking the partial trace" over the environment (symbolic: $\operatorname{tr}_B$), *tracing out the environment*, or "projecting into the system Hilbert space". More precisely, the partial trace and the reduced density operator can be written in terms of the matrix elements ($|a\rangle$, $|b\rangle$ are arbitrary system states)

$$\langle a|\rho_S|b\rangle = \sum_n \langle a, n|\rho_{S+B}|b, n\rangle \tag{1.158}$$

where the $\{|n\rangle\}$ form a complete basis for the environment. You will encounter sometimes the writing

$$\mathrm{tr}_B \rho_{S+B} = \sum_n \langle n|\rho_{S+B}|n\rangle \qquad \text{(symbolic)} \tag{1.159}$$

where the object on the rhs has to be understood as having still the character of an operator in the Hilbert space of the system.

The time evolution of a system coupled to an environment produces mixed states in a dynamical way:

$$\rho_S(t) = \mathrm{tr}_B \left[ U_{S+B}(t)\rho_{S+B}(0)U^\dagger_{S+B}(t) \right] \tag{1.160}$$

even if $\rho_{S+B}(0)$ starts off in a pure state. This is called the "Nakajima-Zwanziger" projection. This construction is, of course, only relevant if (i) the initial state is not factorized (it is entangled) or (ii) there is some interaction between $S$ and $B$. Otherwise $U_{S+B}(t)$ factorizes, and the partial trace simply reduces to

$$\begin{aligned}
&\mathrm{tr}_B (U_S \otimes U_B)(\rho_S \otimes \rho_B)(U_S \otimes U_B)^\dagger \\
&= \mathrm{tr}_B (U_S \rho_S U_S^\dagger) \otimes (U_B \rho_B U_B^\dagger) \\
&= U_S \rho_S U_S^\dagger \, \mathrm{tr}_B (U_B \rho_B U_B^\dagger) \\
&= U_S \rho_S U_S^\dagger
\end{aligned} \tag{1.161}$$

The Nakajima-Zwanziger projection (1.160) shares many physically interesting features and is at the basis of many generalizations of the Schrödinger equation to "open quantum systems". The system+environment setting thus provides a conceptual framework to introduce dissipation into quantum mechanics. We shall use it in the later parts of the quantum optics course.

**(iii) Measure and forget.** This procedure of mixing states is related to the system+environment setting, but it arises from the basic postulates and can be formulated without introducing explicitly an environment. We recall the standard rule (von Neumann and Lüders) of what happens to a quantum state when an observable $\hat{A}$ has been measured (with eigenvalue $a$):

$$|\psi\rangle \mapsto |a\rangle \tag{1.162}$$

The system has "collapsed" to an eigenstate $|a\rangle$ of the observable. This is still a pure state and corresponds to a "perfect" or projective measurement.

Now introduce probabilities and forgetting. The probability that we get the eigenvalue $a$ is, of course, given by $p(a) = |\langle a|\psi\rangle|^2 = \mathrm{tr}\,(|a\rangle\langle a|\rho)$ where $\rho = |\psi\rangle\langle\psi|$ for an initially pure state. Hence if we start off with a non-pure state, the von-Neumann-Lüders rule reads

$$\rho \mapsto |a\rangle\langle a| \tag{1.163}$$

In this way, we can even "purify" a mixed state! After all, the states in quantum mechanics just reflect the knowledge we have about the system.

The perfect measurement is often quite difficult to perform, however, and many states can be found that are still compatible with the measured eigenvalue $a$. In other words, our measurement cannot distinguish precisely among the different eigenstates $|a\rangle$. This is the typical scenario if the eigenvalues are continuously distributed.

Now let us imagine that we only know that we have performed the measurement "Is the system in state $|a\rangle$?", but have forgotten the result. We know that with probability $p(a)$, the state has collapsed (projective measurement). But with probability $1 - p(a)$, something else has happened. Let us assume that the state remained unchanged. By forgetting the result of the measurement, we are forced to assign to the system a mixed state:

$$\rho \mapsto (1 - p(a))\rho + p(a)|a\rangle\langle a| \qquad \text{(simplest approximation)} \tag{1.164}$$

This scenario is called an "imperfect" or weak measurement. If the probability $p(a)$ is small, the state change is also small. This is the scenario we shall use to motivate the dissipative evolution of a two-level system. An alternative notation for the probabilistic mixture of the two states can be given

$$\rho \mapsto \begin{cases} \rho & \text{with prob } 1 - p(a) \\ |a\rangle\langle a| & \text{with prob } p(a) \end{cases} \tag{1.165}$$

**Remark.** We can re-phrase this procedure within a system+environment setting. Suppose that we couple the system to an environment that can "measure" whether the system is in state $|a\rangle$. After some evolution time, we get an entangled state ($|\psi\rangle$ is the initial system density state, assumed pure and $|0\rangle$ the initial environment state)

$$|\psi, 0\rangle \mapsto \langle a|\psi\rangle|a, 1_a\rangle + U_{S+B}|\psi_\perp, 0\rangle$$

where $|1_a\rangle$ is the ("conditional") environment state and $|\psi_\perp\rangle$ is the (non-normalized) system state orthogonal to $|a\rangle$. We construct the reduced density operator and get a mapping (between system operators)

$$|\psi\rangle\langle\psi| \mapsto \mathrm{tr}_B(\langle a|\psi\rangle|a, 1_a\rangle + U_{S+B}|\psi_\perp, 0\rangle)(\langle a|\psi\rangle|a, 1_a\rangle + U_{S+B}|\psi_\perp, 0\rangle)^\dagger$$

Now comes the key assumption: the coupling to the environment has been sufficiently strong so that one can distinguish the environment states $|1_a\rangle$, $|0\rangle$, and the environment states contained in $U_{S+B}|\psi_\perp, 0\rangle$. The best we can do is that these states are orthogonal

$$\langle 1_a|0\rangle \approx 0, \qquad \mathrm{tr}_B\, U_{S+B}|\psi_\perp, 0\rangle\langle a, 1_a| \approx 0 \tag{1.166}$$

This removes the mixed (crossed) terms in the partial trace, and we get a mixture (with $p(a) = |\langle a|\psi\rangle|^2$ as in QM I)

$$|\psi\rangle\langle\psi| \mapsto |a\rangle\langle a|p(a) + \mathrm{tr}_B\, U_{S+B}|\psi_\perp, 0\rangle\langle\psi_\perp, 0|U^\dagger_{S+B}$$

where the first term contains the projection onto the eigenstate. The simplest assumption for the second term is that the environment does not evolve at all, provided the system is in the orthogonal state $|\psi_\perp\rangle$. Then $U_{S+B}|\psi_\perp, 0\rangle \approx |\psi_\perp, 0\rangle$, and the partial trace gives

$$|\psi\rangle\langle\psi| \mapsto |a\rangle\langle a|p(a) + \mathbb{P}_\perp|\psi\rangle\langle\psi|\mathbb{P}_\perp$$

where $\mathbb{P}_\perp$ projects into the subspace orthogonal to $|a\rangle$. The last term has a trace $1 - p(a)$, as in Eq.(1.164), but differs slightly because of the projection. We come back to this when discussing spontaneous emission.

See the introductory article "Decoherence and the transition from quantum to classical" by Zurek (1991) for more details on this discussion. The main message is that the coupling to an environment can provide the same physics as measuring a quantum system.

### 1.13.2 Quantum dissipation in a two-level system

Evolution over time step $\Delta t$. "Sufficiently small" in some sense to put together Hamiltonian evolution and measurement ("monitoring") by an environment.

Pure Hamiltonian (for simplicity, time-independent, applies in rotating frame) ($\hbar = 1$)

$$\rho(t + \Delta t) \approx (\mathbb{1} - \mathrm{i}H\Delta t)\rho(t)(\mathbb{1} + \mathrm{i}H\Delta t) = \rho(t) - \mathrm{i}\,[H\Delta t, \rho(t)] + \mathcal{O}(\Delta t^2) \tag{1.167}$$

Now observing and forgetting about the results. We consider two scenarios.

**Dephasing: measuring energy states.** We assume that with a probability $\Delta p$, we have been able to determine in which energy eigenstate the two-level system is. This can be achieved, for example, by performing measurements on the environment. The rule for "measure and forget" then gives (we have three outcomes)

$$\rho(t + \Delta t) = \begin{cases} \rho(t) & \text{with prob } 1 - \Delta p \\ |\mathrm{g}\rangle\langle\mathrm{g}| & \text{with prob } \Delta p\, \rho_{\mathrm{gg}}(t) \\ |\mathrm{e}\rangle\langle\mathrm{e}| & \text{with prob } \Delta p\, \rho_{\mathrm{ee}}(t) \end{cases} \tag{1.168}$$

This gives the mixed state, as a simple calculation shows

$$
\begin{aligned}
\rho(t+\Delta t) &= (1-\Delta p)\rho(t) + \Delta p \sum_{a\,=\,\mathrm{g,e}} \langle a|\rho(t)|a\rangle\ |a\rangle\langle a| \\
&= (1-\tfrac{1}{2}\Delta p)\rho(t) + \tfrac{1}{2}\Delta p\, \sigma_3 \rho(t) \sigma_3
\end{aligned}
\qquad (1.169)
$$

Concatenate the two elementary processes (1.167, 1.169) and construct an approximate time derivative

$$
\frac{\Delta \rho}{\Delta t} \approx -\mathrm{i}\,[H, \rho(t)] + \frac{\Delta p}{2\Delta t}\left\{\sigma_3 \rho(t) \sigma_3 - \rho(t)\right\} \qquad (1.170)
$$

This is the dynamical equation for a system subject to dephasing. The equation is in the so-called Lindblad form (see Eq.(1.178)), a general form for the time evolution of an open system that we shall derive later in the lecture. The rate $\Delta p / 2\Delta t$ is called the "dephasing rate".

**Exercise.** Switch to the Heisenberg picture and calculate from Eq.(1.170) the rate of change $\langle \Delta \boldsymbol{\sigma} / \Delta t \rangle$ of the Bloch vector. Show that the non-Hamiltonian terms give

$$
\left.\frac{\langle \Delta \sigma \rangle}{\Delta t}\right|_{\mathrm{non-H}} \approx -\frac{\Delta p}{2\Delta t} \langle \sigma \rangle \qquad (1.171)
$$

while $\langle \Delta \sigma_3 / \Delta t \rangle = 0$. The monitoring of the energy levels thus does not change the inversion which is not surprising, since we made the assumption that the measured eigenstate is not changed. The dipole, that captures the relative phase of superposition states in the energy basis, however, decays with a rate $\Delta p / 2\Delta t$. We can thus interpret the dipole relaxation rate as the rate at which the environment acquires information about the system's energy. Note also that the decay of the dipole is the price to pay for the measurement in the energy basis – the quantum-mechanical rule that "any measurement perturbs the system" still holds.

**Spontaneous emission: quantum jumps.** The second scenario is based on the observation of the photons that a two-level atom can emit. We assume that over the evolution time $\Delta t$, the probability to detect an emitted photon is $\Delta p\, \rho_{\mathrm{ee}}(t)$. We have clearly $\Delta p = \gamma \Delta t$ according to the law of radioactive decay. In addition, once this photon has been detected, we know that the atom must be in the ground state $|\mathrm{g}\rangle$. This feature is different from the previous scenario where the measurement perturbed the system in a weaker way.

Now imagine that we throw away the information that a photon has been emitted. The state then mixes into

$$
\rho(t+\Delta t) = \begin{cases} \rho' & \text{with prob } 1-\Delta p \\ |\mathrm{g}\rangle\langle \mathrm{g}| & \text{with prob } \Delta p\, \rho_{\mathrm{ee}}(t) \end{cases} \qquad (1.172)
$$

where the state $\rho'$ is normalized and corresponds to the event "no photon detected". This can be translated into

$$\rho(t+\Delta t) = \rho'' + \Delta p\, \rho_{\rm ee}(t)\; |{\rm g}\rangle\langle{\rm g}| = \rho'' + \Delta p\, \sigma\rho(t)\sigma^\dagger \tag{1.173}$$

where $\rho''$ is a non-normalized state with trace $1 - \Delta p\, \rho_{\rm ee}(t)$. The second term appearing here is called a "quantum jump": the photon emission happens when the atom jumps from the excited to the ground state $|{\rm e}\rangle \to |{\rm g}\rangle$. The ladder (or annihilation) operator $\sigma$ plays here a very intuitive role. If $\rho(t) = |\psi(t)\rangle\langle\psi(t)|$ is a pure state, the system jumps to the state $\sigma|\psi(t)$ at the photon emission.

The first term $\rho''$ in Eq.(1.173) now takes care of the conservation of probabilities. In our first guess (1.164) we simply took $\rho'' = (1 - \Delta p\, \rho_{\rm ee})\rho$. This recipe must be refined here, for two reasons.

One reason is more formal: we want that $\rho(t + \Delta t)$ to be expressed as a linear map of the state $\rho(t)$, but $\Delta p\, \rho_{\rm ee}\, \rho$ is quadratic in $\rho$. This reason is deeply rooted in the linearity of quantum mechanics. The Nakajima-Zwanziger scheme (1.160) which provides a very general framework for quantum dissipation, is also a linear map between density matrices.

The second reason is that the event "no photon has been detected" actually *changes our knowledge* about the system. Qualitatively speaking, it increases our confidence that the system might be in the ground state. We are having less the tendency to think it is in the excited state. After all, if the system is in the excited state, we would expect at some point a photon to appear! In the opposite limit, if over a very long time we do not detect any photons, the system must be (with a very high probability) in the ground state, and we have gained this knowledge from the sequence of "no-photon" events.

We are thus led to the following refined approximation

$$\begin{aligned}\rho'' &\approx \left(\mathbb{1} - \tfrac{1}{2}\Delta p\, |{\rm e}\rangle\langle{\rm e}|\right) \rho(t) \left(\mathbb{1} - \tfrac{1}{2}\Delta p\, |{\rm e}\rangle\langle{\rm e}|\right) \\ &\approx \rho(t) - \tfrac{1}{2}\Delta p\, \left\{\sigma^\dagger\sigma\, \rho(t) + \rho(t)\, \sigma^\dagger\sigma\right\}\end{aligned} \tag{1.174}$$

Putting Eqs.(1.173, 1.174), together, we find the so-called "master equation" for a two-level system with spontaneous decay

$$\frac{\Delta\rho}{\Delta t} \approx -{\rm i}\,[H, \rho(t)] + \frac{\Delta p}{\Delta t}\left\{\sigma\rho(t)\sigma^\dagger - \tfrac{1}{2}\sigma^\dagger\sigma\rho(t) - \tfrac{1}{2}\rho(t)\sigma^\dagger\sigma\right\} \tag{1.175}$$

More details on this derivation can be found in the paper by Dalibard & al. (1992) on a "Wave-Function Approach to Dissipative Processes in Quantum Optics".

**Exercise.** By working out matrix elements of this equation, identify $\gamma = \Delta p/\Delta t$ with the spontaneous decay rate. The important message of this equation is that spontaneous emission changes *both* the inversion and the dipole:

$$\left.\frac{\langle\Delta\sigma\rangle}{\Delta t}\right|_{\rm non-H} \approx -\tfrac{1}{2}\gamma\langle\sigma\rangle \tag{1.176}$$

$$\left.\frac{\langle\Delta\sigma_3\rangle}{\Delta t}\right|_{\rm non-H} \approx -\gamma(\langle\sigma_3\rangle + 1) \tag{1.177}$$

### 1.13.3 Lindblad master equation

Without going into the details of the derivation, we just state here that the generalized von-Neumann equations (1.170, 1.175) are special cases of a general theorem about the time evolution of a quantum system, the

**Lindblad theorem.** If a time evolution $T_t : \rho(0) \mapsto \rho(t)$ satisfies the following conditions:

- the map $T_t$ is linear and maps density matrices onto density matrices;
- the map $T_t$ is completely positive[6];
- expectation values evolve continuous in time;
- repeating the map corresponds to adding time lapses, $T_t T_{t'} = T_{t+t'}$,

then there exists a hermitean operator $H$ and a countable set of system operators $L_k$ ($k = 1 \ldots K$) such that the state $\rho(t) = T_t\rho(0)$ solves the differential equation

$$\frac{{\rm d}\rho}{{\rm d}t} = -{\rm i}\,[H,\rho] + \sum_{k=1}^{K}\left\{L_k\rho L_k^\dagger - \tfrac{1}{2}L_k^\dagger L_k\rho - \tfrac{1}{2}\rho L_k^\dagger L_k\right\} \tag{1.178}$$

The operators $L_k$ are called Lindblad or jump operators.

For spontaneous emission and dephasing, only a single Lindblad operator appears, as shown in this Table:

| | spont. decay | dephasing |
|---|---|---|
| Lindblad operator | $\sqrt{\gamma}\,\sigma$ | $\sqrt{\kappa/2}\,\sigma_3$ |

where $\kappa$ is the dephasing rate. Keeping both Lindblad operators in the time evolution, gives the Bloch equations (1.138, 1.139) with a dephasing rate $\Gamma = \kappa + \gamma/2$. The Lindblad theorem is proven later on in the quantum optics lecture. A simple proof can be found in Nielsen & Chuang (2011) and Henkel (2007).

[6] Qualitatively speaking: density matrices are mapped onto density matrices even if the system is augmented by some environment and the map $T_t$ augmented by "nothing happens with the environment".

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