Chapter 1

Quantum-Mechanical Fundamentals of Lasers

In this chapter we will investigate the basic quantum-mechanical effects and relations that allow the realization of a laser and determine the properties of laser operation. These are the fundamental processes of absorption, spontaneous emission and stimulated emission of light and their quantum-mechanical description.

1.1 Einstein Relations and Planck's Law

It was is the early years of quantum physics, when Planck found a theoretical description of the spectral distribution of the blackbody radiation. This radiation, which is emitted, e.g., from a small hole in the walls of a hohlraum (the blackbody) kept at a temperature T as shown in Fig. 1.1, shows a characteristic spectrum. Its spectral distribution and the peak of the emission intensity are only a function of the blackbody temperature. In Planck's derivation of this spectrum he assumed that electromagnetic radiation cannot be emitted or absorbed continuously, but only in fixed amounts of energy, the quanta, with a corresponding energy of

$$E = h\nu = \frac{hc}{\lambda}. \tag{1.1}$$

Today we know that these quanta are the photons of the electromagnetic field that can be described by their frequency ν or their wavelength λ.

Einstein also tried to find a derivation of this spectral distribution, starting from the fundamental interactions of absorption and emission between a quantum-mechanical system (atom, ion, molecule, electronic states in condensed matter for

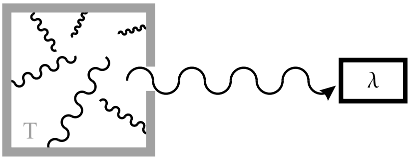

Fig. 1.1 Measurement of the spectral distribution of the blackbody radiation emitted by a hohlraum at a temperature T

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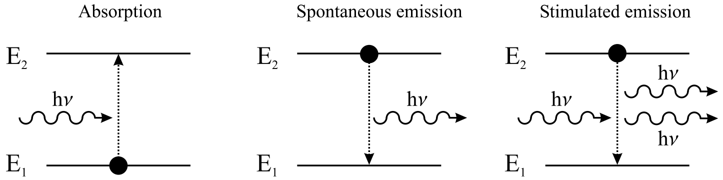


Fig. 1.2 Interactions between a two-level system and a photon according to Einstein

example) and a photon. According to Einstein, this can be described by three basic processes as shown in Fig. 1.2 for a simple two-level system. These processes are:

- The **absorption** of a photon of energy $h\nu = E_2 - E_1$, causing a transition from level $|1\rangle$ to level $|2\rangle$.
- The **spontaneous emission**, in which the system emits a photon of energy $h\nu$ by returning from level $|2\rangle$ to level $|1\rangle$. This is called spontaneous emission as the moment of emission (i.e. the phase ϕ of the radiation), the polarization $\vec{\epsilon}$ and the propagation direction, i.e. the direction of the wave vector $\vec{k}$, is random. Thus the spontaneous emission causes incoherent radiation and is responsible for the fluorescence of excited media.
- The **stimulated emission**, in which an incoming photon induces a resonant transition from the excited level $|2\rangle$ to level $|1\rangle$, emitting a second photon of energy $h\nu$. As photons are Bosons, i.e. they are allowed to be in the same quantum-mechanical state, and as stimulated emission is a resonant process, both photons are identical in all their properties. This effect, therefore, allows the amplification of light, the fundamental process of any laser.

The fundamental process that allows us to realize a laser is the stimulated emission process occurring in excited quantum-mechanical systems, giving rise to the possibility of photon amplification. It was the existence of the stimulated emission process that Einstein postulated in 1917 in order to derive the well-known **Planck's law** of the spectral energy density of electromagnetic radiation per volume $u(\nu, T)$ in the spectral range ν to $\nu + d\nu$ (or in wavelengths $u(\lambda, T)$ in the spectral range λ to $\lambda + d\lambda$),

$$u(\nu, T)d\nu = \frac{8\pi h\nu^3}{c^3}\frac{1}{e^{\frac{h\nu}{k_B T}} - 1}d\nu \tag{1.2}$$

$$u(\lambda, T)d\lambda = \frac{8\pi hc}{\lambda^5}\frac{1}{e^{\frac{hc}{\lambda k_B T}} - 1}d\lambda, \tag{1.3}$$

which is shown in Fig. 1.3. Therein, also the classical Rayleigh-Jeans law is shown, which we will need later to find some relations in Einstein's derivation.

In this derivation [1] Einstein assumed that an ensemble of $N = N_1 + N_2$ non-degenerate two-level systems with an energy difference $\Delta E = h\nu = E_2 - E_1$ is in thermal equilibrium with its environment kept at a temperature T. The absorption

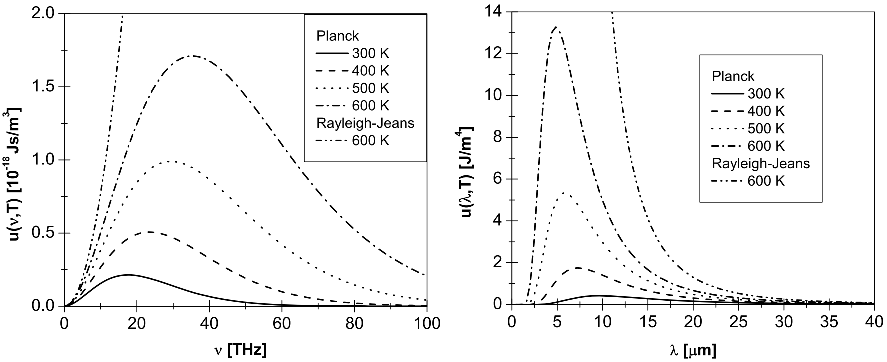


Fig. 1.3 Plot of the spectral energy density of electromagnetic radiation per volume as a function of frequency or wavelength for different temperatures

of the radiation then causes a transition rate from level $|1\rangle$ to level $|2\rangle$,

$$\left(\frac{dN_2}{dt}\right)_{abs} = -\left(\frac{dN_1}{dt}\right)_{abs} = B_{12}u(\nu, T)N_1, \tag{1.4}$$

that is proportional to the (unknown) radiation energy density $u(\nu, T)$ and the number of absorbers N_1 with a proportionality constant B_{12}. The stimulated emission of the radiation causes a transition from level $|2\rangle$ to level $|1\rangle$ with the rate

$$\left(\frac{dN_2}{dt}\right)_{stim} = -\left(\frac{dN_1}{dt}\right)_{stim} = -B_{21}u(\nu, T)N_2, \tag{1.5}$$

which is also proportional to the radiation density $u(\nu, T)$ and the number of emitters N_2 with a proportionality constant B_{21}. The spontaneous emission is only proportional to the number of the possible emitters N_2 and causes a rate

$$\left(\frac{dN_2}{dt}\right)_{spont} = -\left(\frac{dN_1}{dt}\right)_{spont} = -A_{21}N_2. \tag{1.6}$$

The proportionality constants B_{12}, B_{21} and A_{21} are called **Einstein coefficients**.

From Eq. (1.6) it can be deduced that in absence of other processes the population of level $|2\rangle$ decays exponentially with a time constant $\tau_{21} = A_{21}^{-1}$, called the natural lifetime of the level $|2\rangle$. Therefore, the evolution of the externally measurable fluorescence intensity $I(t) \propto \frac{dN_2}{dt}$ is given by

$$I(t) = I(0)e^{-\frac{t}{\tau_{21}}}. \tag{1.7}$$

This exponential decay is shown in Fig. 1.4.

In thermal equilibrium the populations of the levels $|1\rangle$ and $|2\rangle$ are constant, i.e.

$$\frac{dN_2}{dt} = \left(\frac{dN_2}{dt}\right)_{abs} + \left(\frac{dN_2}{dt}\right)_{stim} + \left(\frac{dN_2}{dt}\right)_{spont} = 0, \tag{1.8}$$

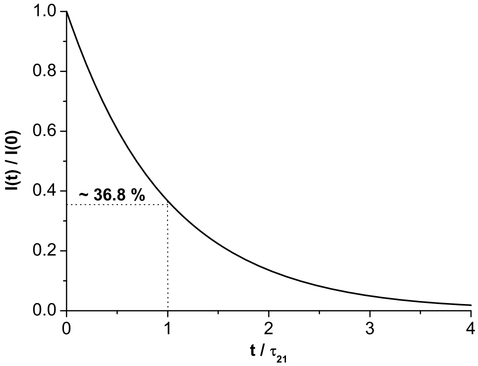


Fig. 1.4 Decay of the fluorescence intensity of an excited sample

$$\frac{dN_1}{dt} = \left(\frac{dN_1}{dt}\right)_{abs} + \left(\frac{dN_1}{dt}\right)_{stim} + \left(\frac{dN_1}{dt}\right)_{spont} = 0, \tag{1.9}$$

and their ratio can be described by a Boltzmann distribution resulting in

$$\frac{N_2}{N_1} = \frac{B_{12}u(\nu, T)}{A_{21} + B_{21}u(\nu, T)} \overset{!}{=} e^{-\frac{E_2 - E_1}{k_B T}}. \tag{1.10}$$

Therefore, the spectral energy density $u(\nu, T)$ has to have the form

$$u(\nu, T) = \frac{A_{21}}{B_{12}e^{\frac{h\nu}{k_B T}} - B_{21}}. \tag{1.11}$$

In order to find the relations between the Einstein coefficients, two limits are investigated: In the high temperature limit $T \to \infty$ the spectral energy density diverges, forcing

$$B_{21} = B_{12}. \tag{1.12}$$

This result is very important as it shows that absorption and stimulated emission are completely equivalent processes.

For the low photon energy limit $h\nu \ll k_B T$, $u(\nu, T)$ needs to be consistent with the classical **Rayleigh-Jeans law**

$$u_{RJ}(\nu, T) = \frac{8\pi\nu^2}{c^3} k_B T, \tag{1.13}$$

which itself was proven experimentally and which can be deduced in the scope of classical Maxwellian electrodynamics, as it does not contain h. This comparison results in

$$A_{21} = \frac{8\pi h\nu^3}{c^3} B_{12}, \tag{1.14}$$

stating that absorption and spontaneous emission are proportional to each other (**Kirchhoff's law**). Including both limits into Eq. (1.11) then gives Planck's law as in Eq. (1.2).

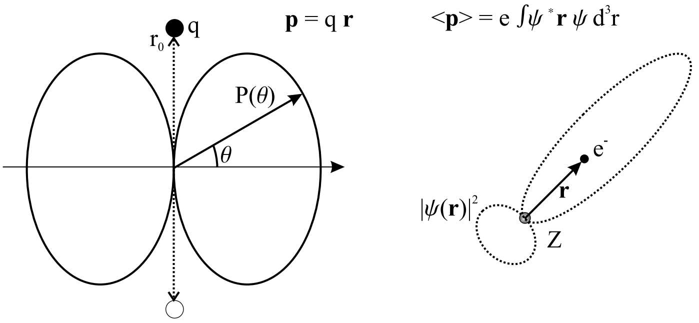


Fig. 1.5 Spatial radiation power of a classical dipole and dipole moment of an electron in quantum mechanics

1.2 Transition Probabilities and Matrix Elements

In this section we will derive the relations between the Einstein coefficients and the quantum-mechanical properties of a dipole transition [2].

1.2.1 Dipole Radiation and Spontaneous Emission

In classical electrodynamics, a dipole consisting of a charge q oscillating at a frequency $\omega = 2\pi\nu$ with a spatial amplitude $r_0 = |\vec{r}_0|$ possesses an electric dipole moment

$$\vec{p}(t) = q\vec{r}(t) = q\vec{r}_0 \sin\omega t. \tag{1.15}$$

As the oscillating dipole is an accelerated charge, it will give rise to a dipole radiation, see Fig. 1.5. The total radiated average power $\overline{P}$ can be derived in the scope of classical electrodynamics and results in **Larmor's formula**,

$$\overline{P} = \frac{2}{3}\frac{\overline{\vec{p}^2}\omega^4}{4\pi\epsilon_0 c^3}. \tag{1.16}$$

Therein,

$$\overline{f} = \frac{1}{T}\int_0^T f\,dt \tag{1.17}$$

is the time-average over one period $T = \frac{2\pi}{\omega}$, leading to

$$\overline{\vec{p}^2} = \frac{1}{2}q^2|\vec{r}_0|^2. \tag{1.18}$$

In quantum mechanics, the average dipole moment of an electron with its elementary charge e, described by the wave function ψ, is given by

$$\langle\vec{p}\rangle = \langle\psi|e\vec{r}|\psi\rangle = \int \psi^* e\vec{r}\psi\,dV. \tag{1.19}$$

Accordingly, we define for a transition between two levels $|1\rangle$ and $|2\rangle$ the transition dipole moment

$$\vec{M}_{21} = \langle \psi_2 | e\vec{r} | \psi_1 \rangle = \int \psi_2^* e\vec{r} \psi_1 dV \tag{1.20}$$

and its absolute value

$$M_{21} = \left| \langle \psi_2 | e\vec{r} | \psi_1 \rangle \right| = \left| \int \psi_2^* e\vec{r} \psi_1 dV \right|. \tag{1.21}$$

In this transition to quantum mechanics we also have to exchange the classical average of $\overline{\vec{p}^2}$ by the quantum-mechanical expression [3]

$$\overline{\vec{p}^2} \rightarrow \frac{1}{2}(M_{21} + M_{12})^2 = 2M_{21}^2. \tag{1.22}$$

Inserting this in Eq. (1.16) results in an emitted power given by

$$\langle P_{21} \rangle = \frac{4}{3} \frac{\omega^4}{4\pi\epsilon_0 c^3} M_{21}^2, \tag{1.23}$$

with $\omega = (E_2 - E_1)/\hbar$. According to Eq. (1.6), the total average fluorescence power P_f emitted by N_2 excited levels corresponds to

$$P_f = h\nu A_{21} N_2 \overset{!}{=} \langle P_{21} \rangle N_2. \tag{1.24}$$

This comparison now allows us to deduce the explicit form of the Einstein coefficient A_{21} as

$$A_{21} = \frac{2}{3} \frac{e^2\omega^3}{h\epsilon_0 c^3} |\langle \psi_2 | \vec{r} | \psi_1 \rangle|^2 = \frac{2}{3} \frac{e^2\omega^3}{h\epsilon_0 c^3} \left| \int \psi_2^* \vec{r} \psi_1 dV \right|^2. \tag{1.25}$$

For an atom or molecule with many different levels for which the wave functions are known, the spontaneous emission rates A_{ji} may now be calculated for all possible transitions between the levels j and i, resulting in a matrix $A_{[j,i]}$. Therefore the M_{ji} in Eq. (1.21) are also called matrix elements.

The derivation above, as a result of Eqs. (1.16), (1.25), is valid only in the dipole approximation, i.e. as long as the wavelength of the emitted radiation is longer than the spatial dimension of the dipole $\lambda \gg r_0$. This is true for $\lambda > 1$ nm, and therefore, for all visible and infrared lasers.

From the ω^3 dependence in Eq. (1.25) it can also be concluded that the spontaneous emission increases dramatically for short wavelengths, resulting in very short lifetimes of the corresponding upper level. As will be shown in Chap. 2, this affords very high pump and laser intensities to saturate the optical transition, making the realization of deep-UV and X-ray lasers based on electronic transitions very difficult.

1.2.2 Stimulated Emission and Absorption

In contrast to the description of the spontaneous emission presented before, the stimulated emission or absorption of a photon by our two-level system is a quantum-mechanical process that involves the interaction between the system and the electromagnetic field. Therefore, it is necessary to make a short excursion into time-dependent quantum-mechanics and perturbation theory [4]:

Let $\mathbb{H}_0$ be the Hamiltonian for the unperturbed system, i.e. the system without the electromagnetic field, which is therefore described by the time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\big|\psi^0(t)\big\rangle = \mathbb{H}_0\big|\psi^0(t)\big\rangle \tag{1.26}$$

for $t < t_0$ with $|\psi^0(t)\rangle$ being the state of the system before the perturbation occurs. After the application of the time-dependent perturbation $\mathbb{V}(t)$, which is assumed to be small compared with $\mathbb{H}_0$, the system will occupy the state $|\psi(t)\rangle$ and will evolve according to

$$i\hbar\frac{\partial}{\partial t}\big|\psi(t)\big\rangle = \big(\mathbb{H}_0 + \mathbb{V}(t)\big)\big|\psi(t)\big\rangle. \tag{1.27}$$

The time dependent perturbation theory allows to calculate the transition rates between different states. The exact derivation of the following formulas can be found, e.g., in [4]. Here we only quote the results that we need to investigate the stimulated emission and absorption processes. As the electromagnetic field of the incident photon can be treated as a periodic perturbation, we use the corresponding results of perturbation theory for a periodic perturbation oscillating at a frequency $\omega = 2\pi\nu$ of the form

$$\mathbb{V}(t) = \mathbb{F}e^{-i\omega t} + \mathbb{F}^\dagger e^{i\omega t}, \tag{1.28}$$

in which $\mathbb{F}$ is an operator defining the nature of the perturbation. Then the transition rate, i.e. the transition probability per unit time, for the transition from state j to state i can be calculated by **Fermi's golden rule**, resulting in

$$R_{ji} = \frac{2\pi}{\hbar}\big(\delta(E_j - E_i - \hbar\omega)\big|\langle\psi_j|\mathbb{F}|\psi_i\rangle\big|^2 + \delta(E_j - E_i + \hbar\omega)\big|\langle\psi_j|\mathbb{F}^\dagger|\psi_i\rangle\big|^2\big). \tag{1.29}$$

The two δ-functions describe the conservation of energy (or the resonance of the process) and the matrix elements of the perturbation $\mathbb{F}$ account for the strength of the transition. Owing to $\omega > 0$ it follows that for $E_j > E_i$ the first term describes the stimulated emission and for $E_j < E_i$ the absorption process is described by the second term.

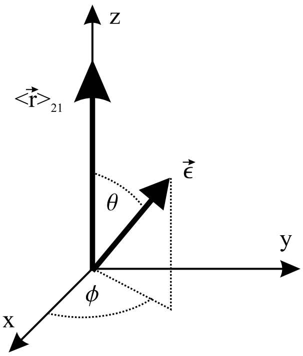
For the stimulated emission from level $|2\rangle$ to level $|1\rangle$ or the absorption from level $|1\rangle$ to level $|2\rangle$ in our two-level system the perturbation is given by the electric field of the incoming photon

$$\vec{E}(t) = \vec{E}_0 e^{i\vec{k}\vec{r}} e^{-i\omega t}, \tag{1.30}$$

causing a transition rate [5]

$$R_{21} = \frac{\pi e^2}{2\hbar^2}\big|\langle\psi_2|\vec{E}_0\vec{r}e^{i\vec{k}\vec{r}}|\psi_1\rangle\big|^2\delta(\omega_0 - \omega) = \frac{\pi e^2}{2\hbar^2}\left|\int\psi_2^*\vec{E}_0\vec{r}e^{i\vec{k}\vec{r}}\psi_1 dV\right|^2\delta(\omega_0 - \omega). \tag{1.31}$$

Fig. 1.6 Local coordinate frame for the calculation of the average over all polarizations



Therein $\vec{k}$ is the wave vector of the electromagnetic wave with $|\vec{k}| = \frac{2\pi}{\lambda}$ and $\omega_0 = \frac{E_2 - E_1}{\hbar}$ is the resonance frequency.

In the same dipole approximation as used for the spontaneous emission, $\lambda \gg r_0$, i.e. $\vec{k}\vec{r} \ll 1$, this rate can be approximated by taking $|e^{i\vec{k}\vec{r}}| \approx 1$ to give

$$R_{21} = \frac{\pi e^2}{2\hbar^2} E_0^2 \left|\vec{\epsilon}\langle\psi_2|\vec{r}|\psi_1\rangle\right|^2 \delta(\omega_0 - \omega) = \frac{\pi e^2}{2\hbar^2} E_0^2 \left|\vec{\epsilon}\int \psi_2^* \vec{r}\psi_1 dV\right|^2 \delta(\omega_0 - \omega), \tag{1.32}$$

with $\vec{\epsilon}$ describing the polarisation of the wave. It shows that the electric field needs to be applied in the same direction as the dipole orientation, in order to produce the maximum transition rate.

We will now simplify this relation for the case of a thermal radiation that is isotropically distributed in space. Therefore, Eq. (1.32) is averaged over all possible polarisation orientations $\vec{\epsilon}$, noting that $\langle\vec{r}\rangle_{21} = \int \psi_2^* \vec{r}\psi_1 dV$ is a constant vector after the integration has been performed. By defining a local frame with its z-axis aligned with $\langle\vec{r}\rangle_{21}$ as in Fig. 1.6, the average over all orientations of

$$\vec{\epsilon} = \begin{pmatrix} \sin\theta\cos\phi \\ \sin\theta\sin\phi \\ \cos\theta \end{pmatrix} \tag{1.33}$$

is calculated in polar coordinates to give

$$\left\langle|\vec{\epsilon}\langle\vec{r}\rangle_{21}|^2\right\rangle = \frac{1}{4\pi}\int_0^{\pi}\int_0^{2\pi} \cos^2\theta \left|\langle\vec{r}\rangle_{21}\right|^2 \sin\theta d\theta d\phi = \frac{1}{3}\left|\langle\vec{r}\rangle_{21}\right|^2. \tag{1.34}$$

This results in the averaged transition rate

$$\langle R_{21}\rangle = \frac{\pi e^2}{6\hbar^2} E_0^2 \left|\langle\psi_2|\vec{r}|\psi_1\rangle\right|^2 \delta(\omega_0 - \omega) = \frac{\pi e^2}{6\hbar^2} E_0^2 \left|\int \psi_2^* \vec{r}\psi_1 dV\right|^2 \delta(\omega_0 - \omega), \tag{1.35}$$

which can be further simplified by introducing the spectral energy density of the electric field at resonance,

$$u(\nu) = \frac{1}{2}\epsilon_0 E_0^2 \delta(\nu_0 - \nu) = \pi\epsilon_0 E_0^2 \delta(\omega_0 - \omega), \tag{1.36}$$

using $\delta(ax) = \frac{1}{a}\delta(x)$. This yields

$$\langle R_{21}\rangle = \frac{2}{3}\frac{\pi^2 e^2}{3\epsilon_0 h^2}\left|\langle\psi_2|\vec{r}|\psi_1\rangle\right|^2 u(\nu) = \frac{2}{3}\frac{\pi^2 e^2}{3\epsilon_0 h^2}\left|\int \psi_2^* \vec{r}\psi_1 dV\right|^2 u(\nu). \tag{1.37}$$

The direct comparison with Eq. (1.5) results in the expression for the Einstein coefficient of stimulated emission,

$$B_{21} = \frac{2}{3}\frac{\pi^2 e^2}{\epsilon_0 h^2}\left|\langle\psi_2|\vec{r}|\psi_1\rangle\right|^2 = \frac{2}{3}\frac{\pi^2 e^2}{\epsilon_0 h^2}\left|\int \psi_2^* \vec{r}\psi_1 dV\right|^2. \tag{1.38}$$

It is, in contrast to A_{21}, independent of the transition frequency or wavelength, and does only depend on the quantum-mechanical properties of the transition enclosed in the matrix elements. By comparing this result with the Einstein coefficient of spontaneous emission A_{21} in Eq. (1.25), we again find the relation shown in Eqs. (1.12), (1.14).

1.3 Mode Structure of Space and the Origin of Spontaneous Emission

Spontaneous emission can be seen as a statistical process, i.e. each atom, ion or molecule decays independently by emitting a photon at a certain time in a single process whilst the observation of the fluorescence of an ensemble of many atoms, ions or molecules shows the well-known exponential decay law of Eq. (1.7). However, this statistical view cannot explain why a single atom "decides" to emit the photon at a certain time. In order to answer this question, we need to have a look into the mode structure of space, i.e. the structure of the allowed eigenmodes of electromagnetic radiation in vacuum, and the nature of the photons occupying these states.

1.3.1 Mode Density of the Vacuum and Optical Media

In order to determine the mode density of the vacuum and of optical transparent media with a refractive index $n > 1$, we first calculate the number of eigenmodes of a cubic hohlraum resonator of length a and volume a^3 up to the frequency ω. As we assume infinitely conductive walls the tangential components of the electric field must vanish on these walls. Therefore, the set of eigenmodes can be represented by

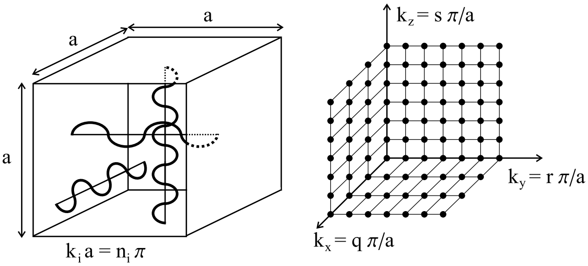


Fig. 1.7 Standing waves in a conductive hohlraum and representation of the eigenmodes in reciprocal space

the standing waves inside the hohlraum as shown in Fig. 1.7 with the wave vectors given by

$$\vec{k} = \frac{\pi}{a}\begin{pmatrix} q \\ r \\ s \end{pmatrix} \quad \text{with } q, r, s \in \mathbb{Z}. \tag{1.39}$$

The corresponding electric field can be written as

$$\vec{E} = \vec{E}_0 \cos \omega t, \tag{1.40}$$

with the spatial components

$$\vec{E}_0 = \begin{pmatrix} E_{0x} & \cos \frac{\pi q}{a} x & \sin \frac{\pi r}{a} y & \sin \frac{\pi s}{a} z \\ E_{0y} & \sin \frac{\pi q}{a} x & \cos \frac{\pi r}{a} y & \sin \frac{\pi s}{a} z \\ E_{0z} & \sin \frac{\pi q}{a} x & \sin \frac{\pi r}{a} y & \cos \frac{\pi s}{a} z \end{pmatrix}, \tag{1.41}$$

the wave vector

$$|\vec{k}_{qrs}| = \frac{\pi}{a}\sqrt{q^2 + r^2 + s^2} \tag{1.42}$$

and the possible resonance frequencies

$$\omega_{qrs} = \frac{\pi c}{a}\sqrt{q^2 + r^2 + s^2}, \tag{1.43}$$

that result from the dispersion relation

$$\omega = c|\vec{k}| \tag{1.44}$$

of electromagnetic waves in vacuum.

In reciprocal space or k-space, where all eigenmodes are represented by a three-dimensional point lattice with a lattice constant $\frac{\pi}{a}$, Eq. (1.42) describes a sphere with radius $|\vec{k}| = \frac{\omega}{c}$. For high frequencies, i.e. large mode numbers $q^2 + r^2 + s^2 \gg 1$, the discrete lattice can be approximated by a homogeneous k-space density $\rho_k = (\frac{a}{2\pi})^3$, which takes into account that, e.g., $-q$ and q describe the same mode. This allows an easy calculation of the volume density of the number of modes in the hohlraum up to the frequency ν:

$$M(\nu) = 2a^{-3}\int \rho_k d^3k = 8\pi \int \frac{k^2 dk}{(2\pi)^3} = \frac{8\pi n^3}{c^3}\int \nu^2 d\nu = \frac{8\pi n^3 \nu^3}{3c^3}. \tag{1.45}$$

Therein the factor of 2 represents the two independent polarisations of the electric field and n the refractive index in the case of a hohlraum filled with an optical medium. This result is independent from the external dimension or orientation of the hohlraum. We can thus let $a \to \infty$ and find the spectral mode density of space as

$$\tilde{M}(\nu) = \frac{\partial M}{\partial \nu} = \frac{8\pi n^3 \nu^2}{c^3}. \tag{1.46}$$

One can recognize this spectral mode density in Planck's law, Eq. (1.2) and in the Rayleigh-Jeans law, Eq. (1.13), which simply states that each of these modes is excited with an energy of $k_B T$ in thermal equilibrium.

An alternative deduction of Planck's law, which is the one Planck used, starts from this spectral mode density $\tilde{M}(\nu)$, which is multiplied by the energy per photon $h\nu$ and by the number of thermally excited photons per mode

$$n(\nu, T) = \frac{1}{e^{\frac{h\nu}{k_B T}} - 1}, \tag{1.47}$$

to yield the spectral energy density in thermal equilibrium. $n(\nu, T)$ is given by the **Bose-Einstein distribution** as photons, spin 1 particles, are bosons.

1.3.2 Vacuum Fluctuations and Spontaneous Emission

We now know the spectral mode density of space and the fact that photons are bosons, which means especially that the number of photons in one quantum-mechanical state, i.e. in one mode, is not restricted. But what kind of state is a mode, i.e. what energy potential creates this state? In order to answer this question we have to look at the quantum structure of the electromagnetic field.

In classical electrodynamics [1], as we did for the determination of the spectral mode density, we can see a mode as a monochromatic wave, e.g. propagating along the x-axis and polarized along the z-axis with an electric field given by

$$\vec{E}(t) = \begin{pmatrix} 0 \\ 0 \\ p(t)\sin kx \end{pmatrix}, \tag{1.48}$$

with a temporal evolution described by $p(t)$. The corresponding magnetic field is given by the Maxwell-Equation $\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$, resulting in

$$\vec{B}(t) = \begin{pmatrix} 0 \\ \frac{1}{c} q(t)\cos kx \\ 0 \end{pmatrix}, \tag{1.49}$$

with $q(t)$ being given by

$$\frac{dq(t)}{dt} = \omega p(t) \tag{1.50}$$

using Eq. (1.44). By inserting both fields into the Maxwell-Equation $\vec{\nabla} \times \vec{B} = \epsilon_0 \mu_0 \frac{\partial \vec{E}}{\partial t}$, we get

$$\frac{dp(t)}{dt} = -\omega q(t), \tag{1.51}$$

which we can combine with Eq. (1.50) to result in

$$\frac{d^2 q(t)}{dt^2} + \omega^2 q(t) = 0. \tag{1.52}$$

This is the equation of motion of a harmonic oscillator, that, in terms of classical mechanics, can be described by a Hamilton function, i.e. a total energy, of

$$H = \frac{1}{2}\omega\big(p^2 + q^2\big). \tag{1.53}$$

The quantization of the electromagnetic field can now be done by formally identifying this result with the quantum-mechanical harmonic oscillator of mass m described by the Hamiltonian

$$\mathbb{H}_{HO} = \frac{1}{2m}\mathbb{P}^2 + \frac{1}{2}m\omega^2\mathbb{Q}^2 = \hbar\omega\left(\mathbb{N} + \frac{1}{2}\right), \tag{1.54}$$

with $\mathbb{P}$ and $\mathbb{Q}$ being the momentum and position operators, respectively. For this formal identification the mass m is a free, non-used parameter that we can simply set to $m = \omega^{-1}$ and compare the result with the classical Eq. (1.53). The different operators in this Hamiltonian are given by

$$\mathbb{N} = \mathbb{A}^\dagger \mathbb{A} \tag{1.55}$$

$$\mathbb{A} = \frac{1}{\sqrt{2\hbar}}\left(\sqrt{m\omega}\mathbb{Q} + \frac{i}{\sqrt{m\omega}}\mathbb{P}\right) \tag{1.56}$$

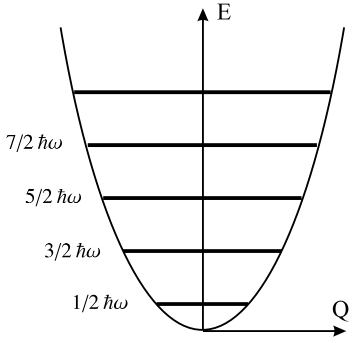
$$\mathbb{A}^\dagger = \frac{1}{\sqrt{2\hbar}}\left(\sqrt{m\omega}\mathbb{Q} - \frac{i}{\sqrt{m\omega}}\mathbb{P}\right), \tag{1.57}$$

with $\mathbb{N}$ being the **occupation number operator** and $\mathbb{A}$ and $\mathbb{A}^\dagger$ the **annihilation** and **creation operators**, respectively, as they decrease or increase the quantum number n of the eigenstate $|n\rangle$ by one, i.e. $\mathbb{A}|n\rangle = \sqrt{n}|n-1\rangle$ and $\mathbb{A}^\dagger|n\rangle = \sqrt{n+1}|n+1\rangle$. For the quantum-mechanical oscillator, we know that the possible energy levels are evenly spaced with a separation of $\hbar\omega$ as shown in Fig. 1.8 and that the energy eigenvalues are given by

$$\mathbb{H}_{HO}|n\rangle = E_n|n\rangle \quad \text{with } E_n = \hbar\omega\left(n + \frac{1}{2}\right). \tag{1.58}$$

As a result, we can state that a photon mode consisting of n photons can be seen as a harmonic quantum oscillator occupying the state with quantum number n. The linear relationship between the number of photons n and the total energy E_n of the mode is as expected, since adding a photon just should increase the energy of that mode by $\hbar\omega$. However, as it is well known from the quantum oscillator,

Fig. 1.8 Potential and corresponding eigenstates of the quantum-mechanical harmonic oscillator



the lowest state $|0\rangle$ has a finite energy of $\frac{1}{2}\hbar\omega$, and since usually the quantum-mechanical system is found in its ground state at absolute zero, i.e. at $T = 0$ K, this energy is called **zero-point energy**. We thus found that in quantum electrodynamics the vacuum is not just an empty space. Electrodynamically, it can be described by an infinite set of modes that are all in their ground state, i.e. unoccupied. However, from **Heisenberg's uncertainty relation**

$$\Delta E \Delta t \geq \frac{\hbar}{2}, \tag{1.59}$$

we can deduce that it is quantum dynamically allowed that a photon of energy $\hbar\omega$ may be spontaneously created "out of nothing' for a short period of time $\Delta t \leq \frac{1}{2\omega}$, so that the necessary energy $\hbar\omega$ is within the theoretically allowed uncertainty

$$\Delta E \geq \frac{\hbar}{2\Delta t} \geq \hbar\omega. \tag{1.60}$$

Without going into detail here, we just state the result that these "virtual" photons do exist and are called **vacuum fluctuations**. In a classical picture this explanation is not possible as the time $\Delta t \leq \frac{1}{2\omega}$ does not allow a full oscillation to occur.

An experimental proof of the vacuum fluctuations, e.g., the **Casimir force**, an additional attractive force component between two uncharged parallel metallic plates placed close to each other. It can be explained by the fact that between the plates only those vacuum fluctuations occur that are consistent with the allowed standing modes, see illustration in Fig. 1.9, whilst in the outer space all vacuum fluctuations occur. Thus an external pressure is created that pushes the two plates together.

Now that we know that the physical vacuum is no "quiet" space, but that vacuum fluctuations occur, we can understand the spontaneous emission in a much more fundamental way. By using the quantum nature of the electromagnetic field, being present in the form of modes that are occupied by a number of photons, we can separate this electromagnetic field into a real part consisting of real photons, and a virtual part, consisting of the virtual photons of the vacuum fluctuations. Therefore, we can describe the spontaneous emission process as a stimulated emission process, triggered by a virtual photon from the vacuum fluctuations. As in standard stimulated emission triggered from a real photon, the emitted photon here is an exact

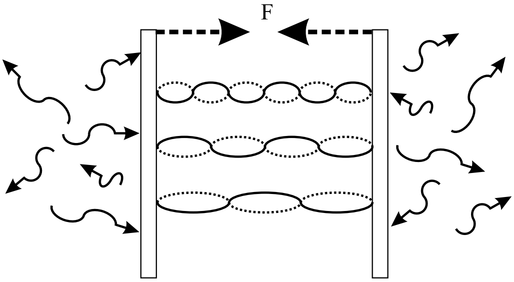


Fig. 1.9 Casimir force between two metallic plates

copy of the input virtual photon. However, no real photon amplification occurs as the virtual photon has to disappear after the process to obey Eq. (1.59). The statistical behaviour of the vacuum fluctuations, i.e. the random nature of the time of creation of the virtual photon as well as the mode in which it occurs, is therefore, transferred on to the whole emission process, explaining the statistical nature of the spontaneous emission. As a result of this more fundamental view, we find that any state of a quantum-mechanical system that couples to the electromagnetic field, and which is not the ground state of the system, will show spontaneous emission towards the energetically lower lying states.

1.4 Cross Sections and Broadening of Spectral Lines

We will introduce in this section the spectroscopic properties that describe a laser medium as well as the line broadening mechanisms that influence the spectral behaviour and the efficiency of a laser. This allows us to quantify the different properties of the optical transitions in a laser medium and results in a general mathematical description of lasers that will be presented in Chap. 2.

1.4.1 Cross Sections of Absorption and Emission

When an electromagnetic wave propagates in an absorbing medium along the z-axis, its intensity $I(z)$ will be attenuated during propagation. In this process each frequency or wavelength component of the radiation may suffer from a different absorption strength. Therefore, we introduce the spectral intensity $\tilde{I}(z,\lambda)$, which is defined by

$$I(z) = \int \tilde{I}(z,\lambda)d\lambda. \tag{1.61}$$

By passing an infinitesimal propagation distance dz each wavelength component is attenuated proportional to the incident spectral intensity according to

$$\frac{d\tilde{I}(z,\lambda)}{dz} = -\alpha(\lambda)\tilde{I}(z,\lambda), \tag{1.62}$$

as shown in Fig. 1.10.

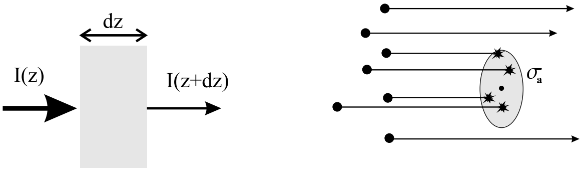


Fig. 1.10 Absorption of light in a medium and geometrical interpretation of the cross section in an absorption process of particles

Integrating this equation under the assumption of a spatially constant absorption coefficient $\alpha(\lambda)$ leads to the **Lambert-Beer law** of absorption

$$\tilde{I}(z,\lambda) = \tilde{I}(0,\lambda)e^{-\alpha(\lambda)z}. \tag{1.63}$$

In the important case in which the absorption is caused by an optical transition from a lower state $|1\rangle$ to an upper state $|2\rangle$, as described by Eq. (1.4), the absorption coefficient is proportional to the number density N_1 of atoms, ions, molecules or other laser species in state $|1\rangle$ and can be written as

$$\alpha(\lambda) = \sigma_a(\lambda)N_1. \tag{1.64}$$

The proportionality constant $\sigma_a(\lambda)$ is the **absorption cross section**. It has a dimension of an area and can be interpreted as an effective "cross-sectional area" attached to, e.g., an atom that absorbes the incident photons as shown in Fig. 1.10. However, depending on the strength of the transition it can vary for different transitions in one atom and one should not confuse it with the geometrical size of the atom itself.

In the same way also the stimulated emission can be described as an amplification of the incident light according to

$$\frac{d\tilde{I}(z,\lambda)}{dz} = \gamma(\lambda)\tilde{I}(z,\lambda), \tag{1.65}$$

resulting in

$$\tilde{I}(z,\lambda) = \tilde{I}(0,\lambda)e^{\gamma(\lambda)z}. \tag{1.66}$$

In analogy to the absorption coefficient, the emission coefficient is proportional to the number density N_2 of atoms in the upper state

$$\gamma(\lambda) = \sigma_e(\lambda)N_2, \tag{1.67}$$

for which the proportionality constant $\sigma_e(\lambda)$ is the **emission cross section**. Taking both processes together leads to the total evolution of the spectral intensity given by

$$\tilde{I}(z,\lambda) = \tilde{I}(0,\lambda)e^{(\sigma_e(\lambda)N_2-\sigma_a(\lambda)N_1)z} \tag{1.68}$$

In the special case of the two-level system in Fig. 1.2, where N_i denote the population densities of the two levels $|i\rangle$, it follows from Eq. (1.12) that the emission and absorption cross sections related to the intrinsic transition are equal, i.e. $\sigma_e(\lambda) = \sigma_a(\lambda)$. They are, therefore, called intrinsic cross sections. However, as we will see in Chap. 2, more complex level schemes exist, especially for ionic levels in solids. Then the levels are split up by the Stark effect and create manifolds, for which it is easier to refer to N_i as the total manifold population and to include e.g. the thermal population distributions within one manifold into the externally measured spectroscopic cross sections, for which $\sigma_e(\lambda)$ and $\sigma_a(\lambda)$ are usually different. This will be explained in more detail in Chap. 2.

To connect this spectroscopic description to the Einstein coefficients, the emitted spectral power density per volume resulting from stimulated emission in the medium of volume $dV = dAdz$ is investigated. Therefore, we use the description of Eq. (1.5) and assume that each emission process emits the energy of one photon $h\nu$ into the propagating mode. The photon energies itself are taken to be distributed around $h\nu_0 = E_2 - E_1$, given by a normalized distribution $\rho_f(\nu)$ describing this fluorescence, with

$$\int \rho_f(\nu)d\nu = 1. \tag{1.69}$$

$\rho_f(\nu)$ thus determines which frequencies are amplified by the stimulated emission. Then the emitted spectral power density per volume is given by

$$\frac{\partial \tilde{P}}{\partial V} = -h\nu\rho_f(\nu)\frac{\partial N_2}{\partial t} = h\nu\rho_f(\nu)B_{21}u(\nu)N_2. \tag{1.70}$$

The spectroscopic view on the other hand results in

$$\frac{\partial \tilde{P}}{\partial V} = \frac{\partial \tilde{I}}{\partial z} = \gamma(\nu)\tilde{I}(\nu) = N_2\sigma_e(\nu)\frac{c}{n}u(\nu) \tag{1.71}$$

Therein, it was assumed that $\tilde{I}(\nu)$ describes a collimated homogeneous beam, which is related to the energy density by $\tilde{I}(\nu) = \frac{c}{n}u(\nu)$ and describes that the energy "flows" with the velocity of light $\frac{c}{n}$ in a medium with a refractive index n.

Comparing Eq. (1.70) and Eq. (1.71) results in the relation

$$\sigma_e(\nu) = \frac{h\nu n}{c}B_{21}\rho_f(\nu). \tag{1.72}$$

By using the relation for the Einstein coefficients Eq. (1.14) and Eq. (1.12), which in a medium with a refractive index n changes owing to the changed mode density of Eq. (1.46) to

$$A_{21} = \frac{8\pi h\nu^3 n^3}{c^3}B_{21}, \tag{1.73}$$

the important relation

$$\sigma_e(\nu) = \frac{c^2}{8\pi n^2\nu^2\tau_{21,sp}}\rho_f(\nu) \tag{1.74}$$

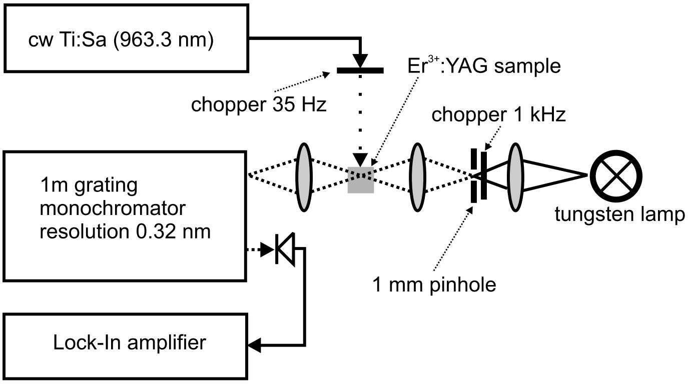


Fig. 1.11 Measurement setup for the determination of absorption and emission cross sections, e.g. of an Er^{3+}:YAG sample

results. Therein, it has been explicitly written that the A_{21} coefficient is related to the spontaneous decay with a decay time $\tau_{21,sp}$ as

$$A_{21} = \frac{1}{\tau_{21,sp}}. \tag{1.75}$$

Thus, the spectral distribution $\rho_f(\nu)$ of the light emitted from the volume dV is closely linked to the spectroscopic emission cross section $\sigma_e(\nu)$. Finally, by exploiting the normalization of $\rho_f(\nu)$ a relation between the upper state lifetime and the integral emission cross section can be deduced,

$$\frac{1}{\tau_{21,sp}} = \frac{8\pi n^2}{c^2} \int \sigma_e(\nu)\nu^2 d\nu = 8\pi n^2 c \int \frac{\sigma_e(\lambda)}{\lambda^4} d\lambda. \tag{1.76}$$

In the last step we used $|d\nu| = \frac{c}{\lambda^2}|d\lambda|$.

Equation (1.76) is called **Füchtbauer-Ladenburg relation**, for which it has to be noted that it is also valid for spectroscopic cross sections and that λ always refers to vacuum wavelengths. It allows the calculation of the **spontaneous emission lifetime** $\tau_{21,sp}$, also called **radiative lifetime**, from measured spectra, or, in the reverse sense, the calibration of measured spectral intensities $\tilde{I}(\lambda)$ to deduce the absolute values of the emission cross section $\sigma_e(\lambda)$.

For this application, the spectral fluorescence is recorded from an excited sample and the emission cross section is then calculated by

$$\sigma_e(\lambda) = \frac{\lambda^4 \tilde{I}(\lambda)}{8\pi n^2 c \tau_{21,sp} \int \tilde{I}(\lambda) d\lambda}. \tag{1.77}$$

A scheme of this measurement setup is shown in Fig. 1.11. An Er^{3+}:YAG sample is excited by the emission from a Ti:sapphire laser and its fluorescence is recorded by a $1-\mathrm{m}$ spectrometer. As the number of fluorescence photons that are captured by the spectrometer is often low, the excitation beam is modulated at a frequency f_{mod}, so that only the detected signal with this frequency component is recorded using a lock-in technique. This enables an increase in signal-to-noise ratio, especially when low cross sections are to be determined. In order to measure the absorption cross section, a broad-spectrum tungsten lamp is used as the source and the spectrometer records the spectrum of the intensity transmitted by the sample. After correcting the data for the spectral emission characteristics of the lamp (cf. Planck's law), the absorption cross section can be calculated by the relative intensity change using

$$\sigma_a(\lambda) = -\frac{1}{LN_1} \ln \frac{I_t(\lambda)}{I_{0,t}(\lambda)}. \tag{1.78}$$

Therein, N_1 equals the Er^{3+} ion density as the excitation power from the lamp is chosen to be low enough in order not to bleach the ground state, L is the length of the sample, $I_t(\lambda)$ the transmitted intensity signal and $I_{0,t}(\lambda)$ the reference recorded with an undoped sample in place.

1.4.2 Natural Line Width and Broadening of Spectral Lines

As a result of the Heisenberg uncertainty principle, Eq. (1.59), a transition between two quantum-mechanical levels cannot be infinitely sharp when the corresponding upper level has a finite lifetime τ, i.e. the corresponding cross sections $\sigma_e(\nu)$ and $\sigma_a(\nu)$ as well as the fluorescence distribution $\rho_f(\nu)$ discussed previously are no δ-functions. As shown in Sect. 1.3.2, each level above the ground state will at least have its natural lifetime that is determined by the spontaneous emission, and thus, by the vacuum fluctuations. Therefore, any optical transition will show a minimum line width, called the **natural line width** of the transition and the spectral line can be represented by its line form function $g(\nu)$, which is identical with the fluorescence distribution $\rho_f(\nu)$.

The fact that a laser medium usually consists of many identical absorption and emission systems, i.e. the atoms, ions or molecules, divides the interaction between them and the electromagnetic field into two cases that define the two different line broadening mechanisms:

- **Homogeneous line broadening**: In this case all systems show the same transition frequency ν_0, line width $\Delta\nu$ and form function $g(\nu)$. Therefore, they all contribute to the emission or absorption of a photon of energy $h\nu$ in the same way, i.e. with the same probability. They can be described by processes that reduce the upper level lifetime in a homogeneous way for all the systems, thus causing an equal broadening of all systems around the same resonance frequency ν_0, e.g. spontaneous emission (**natural line width**), lattice vibrations (phonons) of the crystal matrix in solid-state lasers causing **multi-phonon relaxation**, atomic collisions in gas lasers causing collisional relaxation (**pressure broadening**).

- **Inhomogeneous line broadening**: In this case, the transition frequency of the different systems varies, resulting in different interaction probabilities between a photon of energy $h\nu$ and the different systems. This inhomogeneous distribution of the resonance frequencies over the different systems may be temporally constant as e.g. in ion-doped amorphous solids as fibers, in which the **Stark effect** shifts the energy levels by a fixed amount for a given ion but varies from site to site in the glass matrix. It can also be time dependent for a given system as e.g. the Doppler-shift in gas lasers, which depends on the local velocity of an atom or molecule (**Doppler broadening**). Thus, the atom itself changes its resonance frequency over time resulting from its collisions and the corresponding changes in velocity and direction. However, for the ensemble, a constant effective average results from the Maxwell distribution.

Homogeneous Broadening

To find the line width function of a homogeneously broadened line $g(\nu)$ we will investigate a classical example [6]. The spontaneous emission can be explained by a suddenly emitted electric field oscillating at the resonance $\hbar\omega_0 = h\nu_0 = E_2 - E_1$ with an exponentially decaying amplitude with a time constant of 2τ (the time constant τ corresponds to the intensity $I \propto E^2$),

$$E(t) = \begin{cases} 0 & \text{for } t \le 0 \\ E_0 e^{-\frac{t}{2\tau}} \cos\omega_0 t & \text{for } t > 0 \end{cases} = \begin{cases} 0 & \text{for } t \le 0 \\ \frac{E_0}{2} e^{-\frac{t}{2\tau}} \left(e^{i\omega_0 t} + e^{-i\omega_0 t}\right) & \text{for } t > 0 \end{cases}. \tag{1.79}$$

The spectral components of this electric field are given by its Fourier transformation

$$E(\nu) = \int_{-\infty}^{\infty} E(t) e^{-i2\pi\nu t} dt = i\frac{E_0}{4\pi}\left(\frac{1}{\nu_0 - \nu + \frac{i}{4\pi\tau}} - \frac{1}{\nu_0 + \nu - \frac{i}{4\pi\tau}}\right), \tag{1.80}$$

resulting in the spectral intensity given by a **Lorentzian function**

$$\tilde{I}(\nu) = I g(\nu) = \sqrt{\frac{\epsilon_0}{\mu_0}} \left|E(\nu)\right|^2 = I\frac{2}{\pi}\frac{\Delta\nu}{4(\nu - \nu_0)^2 + (\Delta\nu)^2}. \tag{1.81}$$

Therein, $\Delta\nu = \frac{1}{2\pi\tau}$ is the natural line width, given as the full width at half maximum (FWHM) of the line.

Inhomogeneous Broadening

The most important broadening process in gas lasers is Doppler broadening, which will be described here as an example for inhomogeneous broadening. In a gas the different atoms or ions will show a kinetic velocity distribution with respect to one propagation direction, e.g. the z-axis, that is given by the normalized Maxwell distribution [6]

$$P(v_z) = \sqrt{\frac{m}{2\pi k_B T}} e^{-\frac{m v_z^2}{2 k_B T}}. \tag{1.82}$$

Therein, m is the mass of the atom, v_z its velocity component along the z-axis and T the kinetic gas temperature. While the atoms itself still have the same resonance frequency ν_0 in their local rest frame, an external observer looking along the z-axis, however, will see the Doppler shifted frequency

$$\nu = \nu_0 \left(1 + \frac{v_z}{c}\right), \tag{1.83}$$

which is taken here in the non-relativistic limit. As the velocity distribution directly determines the probability of that velocity component in the gas, the line form function can be directly deduced as

$$g(\nu) = \frac{2\sqrt{\pi \ln 2}}{\pi \Delta\nu_D} e^{-(2\frac{\nu-\nu_0}{\Delta\nu_D})^2 \ln 2}, \tag{1.84}$$

with a Doppler line width of

$$\Delta\nu_D = \nu_0 \sqrt{\frac{8 k_B T \ln 2}{m c^2}}. \tag{1.85}$$

In contrast to the homogeneous broadening, the form function of the inhomogenous Doppler broadening is a **Gaussian function**.

Simultaneous Broadening Processes

In the case of different homogeneous broadening processes acting simultaneously, e.g. spontaneous emission and multi-phonon relaxation in a solid-state laser, each process contributes to the total decay of the upper level and can be described by its own lifetime or decay constant. In this example, they are the spontaneous lifetime τ_{sp} and the non-radiative relaxation lifetime τ_r, that both contribute to the decay of the upper level as

$$\frac{dN_2}{dt} = -\frac{N_2}{\tau_{sp}} - \frac{N_2}{\tau_r} = -\frac{N_2}{\tau}. \tag{1.86}$$

Therefore, for different homogeneous broadening processes the different lifetimes add inversely to the total lifetime τ like the parallel connection of resistors,

$$\frac{1}{\tau} = \sum_i \frac{1}{\tau_i}, \tag{1.87}$$

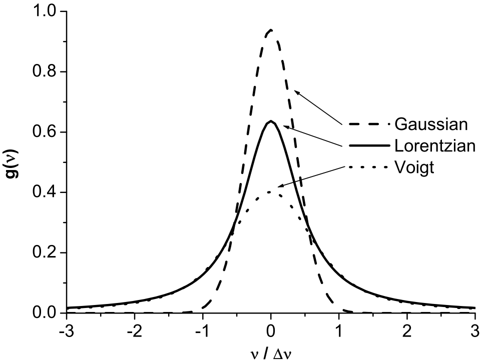


Fig. 1.12 Fundamental Lorentzian and Gaussian line shape functions and their convolution, the Voigt profile

and the corresponding line widths add directly like the parallel connection of capacitors,

$$\Delta\nu = \sum_i \Delta\nu_i. \tag{1.88}$$

The line shape function $g(\nu)$ of the combined line shape is again a Lorentzian function.

For the case of two inhomogeneous processes with line form functions $g_1(\nu)$ and $g_2(\nu)$, or a mixing between a homogeneous and an inhomogeneous process, the total line form function is in general given by the convolution of the line form functions of the different processes [6],

$$g(\nu) = \int_{-\infty}^{\infty} g_1(\nu')g_2(\nu - \nu')d\nu'. \tag{1.89}$$

Two Gaussian line shapes thus result in a new Gaussian line shape with the line width

$$\Delta\nu = \sqrt{\Delta\nu_1^2 + \Delta\nu_2^2}, \tag{1.90}$$

while for a mixing between a Lorentzian and a Gaussian line form function the convolution cannot be solved analytically and results in the **Voigt profile**. All three line shapes are shown for comparison in Fig. 1.12.

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