# 1

# A Short Introduction to Laser Physics

To study the influence of light on the dynamics of an atom or a molecule experimentally, laser light sources are used most frequently. This is due to the fact that lasers have well-defined properties. The theory of the laser dates back to the 1950s and 1960s of the twentieth century and by now, 50 years later is textbook material. In this introductory chapter, we start by recapitulating some basic notions of laser theory, which will be needed to understand later chapters.

More recently, experimentalists have been focusing on pulsed mode operation of lasers with pulse lengths of the order of femtoseconds, allowing for time-resolved measurements. At the end of this chapter, we therefore put together some aspects of pulsed lasers that are important for their application to atomic and molecular systems.

## 1.1 The Einstein Coefficients

Laser activity may occur in the case of nonequilibrium, as we will see later. Before dealing with this situation, let us start by considering the case of equilibrium between the radiation field and an ensemble of atoms in the walls of a cavity. This will lead to the Einstein derivation of Planck's radiation law.

The atoms will be described in the framework of Bohr's model of the atom, allowing the electron to occupy only discrete energy levels. For the derivation of the radiation law, the consideration of just two of those levels is sufficient. They shall be indexed by 1 and 2 and shall be populated such that for the total number of atoms

$$N = N_1 + N_2 \tag{1.1}$$

holds. This means that $N_2$ of the atoms are in the excited state with energy $E_2$ and $N_1$ atoms are in the ground state with energy $E_1$. Transitions between the states shall be possible by emission or absorption of photons of the appropriate energy. The following processes can be distinguished:

- Absorption of light leading to a transition rate

$$\left.\frac{\mathrm{d}N_2}{\mathrm{d}t}\right|_{\mathrm{abs}} = \rho N_1 B_{12} \tag{1.2}$$

  from the ground to the excited state.
- Induced (or stimulated) emission of light leading to a transition rate

$$\left.\frac{\mathrm{d}N_1}{\mathrm{d}t}\right|_{\mathrm{emin}} = \rho N_2 B_{21} \tag{1.3}$$

  for the population change of the ground state.
- Spontaneous emission of light leading to a rate

$$\left.\frac{\mathrm{d}N_1}{\mathrm{d}t}\right|_{\mathrm{emsp}} = N_2 A \tag{1.4}$$

  which amounts to a further increase of the ground state population.

The first two processes are proportional to the energy density $\rho$ of the radiation field with the constants $B_{12}$, respectively, $B_{21}$. The process of spontaneous emission does not depend on the external field and is proportional to $A$. These coefficients are called Einstein's $A$- and $B$-coefficients.

In thermal equilibrium, the rate of transition from level 1 to 2 has to equal that from 2 to 1, leading to the stationarity condition

$$N_1 B_{12} \rho = N_2 B_{21} \rho + N_2 A. \tag{1.5}$$

This equation can be resolved for the energy density $\rho$ leading to

$$\rho = (N_1 B_{12}/(N_2 B_{21}) - 1)^{-1} A/B_{21}. \tag{1.6}$$

Furthermore, in thermal equilibrium, the ratio of populations is given by the Boltzmann factor according to

$$N_1/N_2 = \exp\left\{-\frac{E_1 - E_2}{kT}\right\} \tag{1.7}$$

with the temperature $T$ and the Boltzmann constant $k$.

As $T \to \infty$ also $\rho \to \infty$, and we can conclude that the $B$-coefficients have to be identical $B_{12} = B_{21} = B$. Using Bohr's postulate

$$E_2 - E_1 = h\nu, \tag{1.8}$$

where $\nu$ is the frequency of the light, we can conclude from (1.6) that

$$\rho = \left(\exp\left\{\frac{h\nu}{kT}\right\} - 1\right)^{-1} A/B. \tag{1.9}$$

holds. The ratio of Einstein coefficients $A/B$ can now be determined by comparing the formula above with the Rayleigh–Jeans law

$$\rho(\nu) = \left(8\pi/c^3\right)\nu^2 kT, \tag{1.10}$$

which is a very good approximation in the case of low frequencies (see Fig. 1.1). One then arrives at

$$A/B = \left(8\pi/c^3\right)h\nu^3 =: D(\nu)h\nu \tag{1.11}$$

for the ratio, where $D(\nu)\mathrm{d}\nu = 8\pi\nu^2/c^3\mathrm{d}\nu$ is the number of possible waves in the frequency interval from $\nu$ to $\nu+\mathrm{d}\nu$ in a cavity of unit volume [1]. Inserting this result into (1.9) yields Planck's radiation law

$$\rho\mathrm{d}\nu = (D(\nu)\mathrm{d}\nu)h\nu\left(\exp\left\{\frac{h\nu}{kT}\right\} - 1\right)^{-1}. \tag{1.12}$$

The last factor in this expression is the number of photons with which a certain wave is occupied. As a function of the wavelength, Fig. 1.1 shows a comparison of Planck's law with the two laws only valid in the limits of either long or short wavelength. These are the Rayleigh–Jeans and Wien's law, respectively.

In the case of nonequilibrium, an extension of the formalism just reviewed leads to the fundamentals of laser theory, as we will see in the following. The explicit calculation of the Einstein $B$-coefficient shall be postponed until Chap. 3.

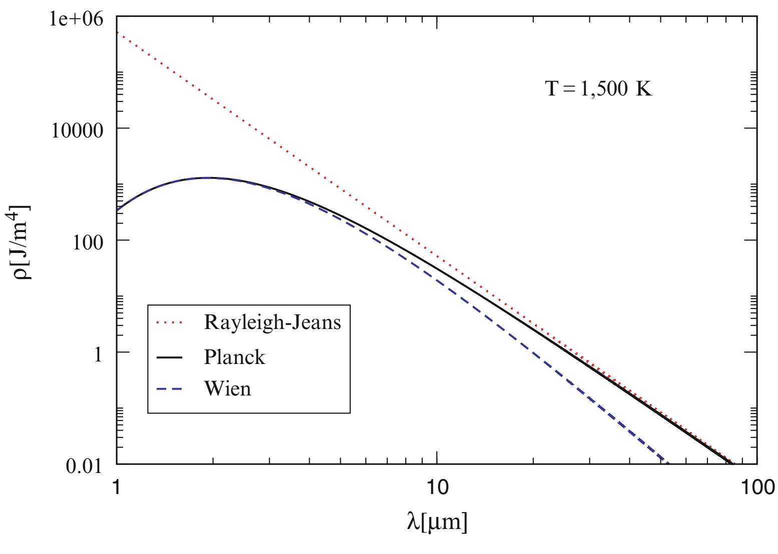


**Fig. 1.1.** Energy density (per wavelength interval) as a function of wavelength for different radiation laws at a temperature of $T = 1{,}500\,\mathrm{K}$

## 1.2 Fundamentals of the Laser

The derivation of laser activity can be done in a crude way by again considering the populations of two levels between which the laser transition occurs. The atoms are driven out of equilibrium by pumping and are interacting with a fixed frequency light field with photon number $n$ (considered to be a continuous variable in the following) in a resonator [2].

First, we consider the processes leading to a change in the populations. In addition to the ones introduced in Sect. 1.1, these are pump (or gain) and loss processes. We concentrate on laser activity and therefore spontaneous emission can be neglected for the time being. Secondly, the realization of the laser process, requiring more than a bare two-level system will be shortly discussed.

### 1.2.1 Elementary Laser Theory

In close analogy to the Einstein coefficients for the induced transition rates, coefficients can be defined that fulfill $W_{ij} = W_{ji} = W$ leading to an induced emission rate of $(N_2 - N_1)Wn$.[1] Including the gain and loss processes, depicted in Fig. 1.2, the rate equations

$$\frac{\mathrm{d}N_1(t)}{\mathrm{d}t} = \gamma_{12}N_2 - \mathit{\Gamma} N_1 + (N_2 - N_1)Wn, \tag{1.13}$$

$$\frac{\mathrm{d}N_2(t)}{\mathrm{d}t} = \mathit{\Gamma} N_1 - \gamma_{12}N_2 - (N_2 - N_1)Wn, \tag{1.14}$$

emerge. Subtracting the first from the second equation leads to a rate equation for the difference $D = N_2 - N_1$, which is also referred to as population inversion

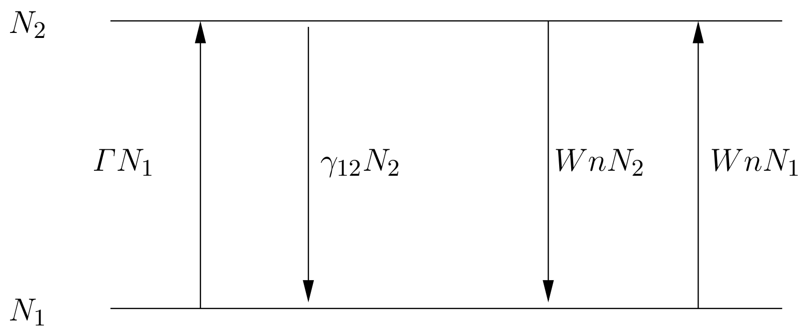


**Fig. 1.2.** Two-level system with elementary transitions (from the *left* to the *right*): Pump process, loss processes (e.g., by radiationless transitions), induced emission and absorption; spontaneous emission is not considered; adapted from [2]

[1] Note that in the previous section the rate was proportional to $\rho$ and here it is proportional to the dimensionless variable $n$; we therefore have to use a different symbol for the coefficients.

$$\frac{\mathrm{d}D}{\mathrm{d}t} = -2WnD - \frac{1}{T_1}(D - D_0). \tag{1.15}$$

Here the definitions of the unsaturated inversion $D_0 = N(\mathit{\Gamma} - \gamma_{12})/(\mathit{\Gamma} + \gamma_{12})$, which will become clear below, and the relaxation time $T_1 = (\mathit{\Gamma} + \gamma_{12})^{-1}$ have been introduced. Including loss effects of the optical cavity via a parameter $t_{\mathrm{cav}}$, the rate equation for the photon number

$$\frac{\mathrm{d}n}{\mathrm{d}t} = WnD - \frac{n}{t_{\mathrm{cav}}}, \tag{1.16}$$

follows, where the first term is due to the increase of radiation by stimulated processes and the effect of spontaneous emission has been neglected. Equation (1.15) for the inversion together with (1.16) for the photon number are a simplified version of the full quantum mechanical laser equations, allowing one to understand some basic laser properties [2, 3].

For an amplification of the light field to occur by starting from a low initial photon number $n_0$ with unsaturated inversion $D_0$, the right-hand side of (1.16) has to be larger than zero. For reasons of simplicity, let us here just consider the steady state defined by

$$\frac{\mathrm{d}n}{\mathrm{d}t} = 0 \qquad \frac{\mathrm{d}D}{\mathrm{d}t} = 0, \tag{1.17}$$

however. For the inversion we get

$$D = D_0/(1 + 2T_1Wn), \tag{1.18}$$

i.e., a reduction for a finite photon number as compared to the unsaturated value $D_0$. The photon number in the steady state follows from

$$n\left(\frac{WD_0}{1 + 2T_1Wn} - \frac{1}{t_{\mathrm{cav}}}\right) = 0, \tag{1.19}$$

leading to two different solutions:

(1) $n_0 = 0$
(2) $n_0 = (D_0 - D_{\mathrm{thr}})\frac{t_{\mathrm{cav}}}{2T_1}$

In order for the nontrivial solution to be larger than zero, the inversion has to be larger than a threshold value $D_{\mathrm{thr}} = 1/(Wt_{\mathrm{cav}})$. As a function of $D_0$, the transition from the trivial solution to the one with a finite number of photons is depicted in Fig. 1.3.

In principle, laser theory has to be formulated quantum theoretically. This is done e.g., in [2]. There the transition from a standard light source to a laser above threshold is explained in a consistent framework. For large photon numbers one finds the phenomenon of anti-bunching, i.e., the photons leave the cavity equidistantly. The corresponding laser light has a constant amplitude. Therefore in the applications part of this book, we will assume that the field can be described classically by using a sinusoidal oscillation.

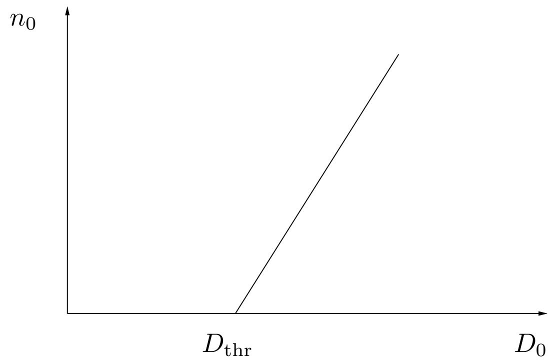


**Fig. 1.3.** Steady state photon number versus unsaturated inversion

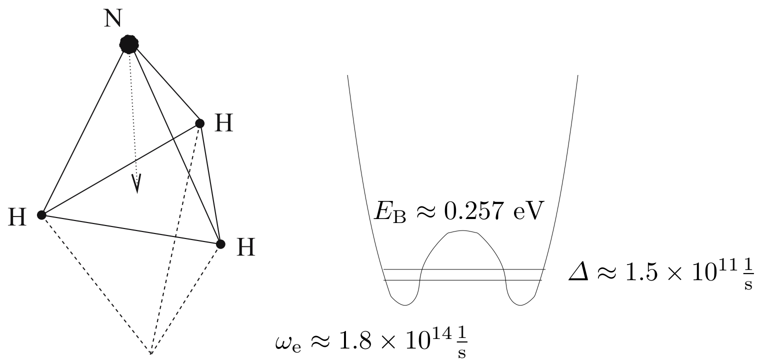


**Fig. 1.4.** Umbrella mode of $NH_3$ indicated by the *arrow* and schematic double well (frequency of oscillation around the minima $\omega_e$, tunneling frequency $\Delta$, and barrier height $E_B$ are indicated) with the two levels (their separation is vastly exaggerated for reasons of better visibility) used for the maser process

### 1.2.2 Realization of the Laser Principle

As we have just seen, nonequilibrium, characterized by population inversion, is crucial for operating a laser. Since the invention of the first maser[2] it has been shown that inversion can be achieved in many different ways. A small collection of possibilities (including also the microwave case) will now be discussed.

#### The Ammonia Maser

In the $NH_3$-maser [4], the umbrella mode (see Fig. 1.4) leads to a double well potential and thus quantum mechanically tunneling is possible. A corresponding doublet of levels in the double well exists, which is used for the

[2] Maser stands for "Microwave amplification by stimulated emission of radiation".

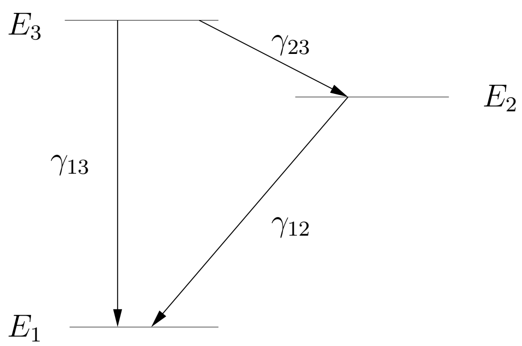


**Fig. 1.5.** Three-level system of the ruby laser with the metastable level $E_2$

maser process. Inversion is created by separating the molecules in the upper level from the ones in the lower level by using the quadratic Stark effect in an inhomogeneous electric field.

This principle cannot be applied in the optical (i.e. laser) case, however, since typically $h\nu \gg kT$ at optical frequencies and therefore $N_2 \ll N_1$. Increasing the number of atoms in the upper level via pumping is therefore necessary.

### The Ruby Laser

To achieve inversion in a laser, more than two levels are needed. Solid-state lasers like the three-level ruby laser [5] are pumped optically. Lasing is then done out of the metastable level $E_2$, shown in Fig. 1.5. By considering just the pumping and the loss terms in the rate equations for the three-level system one can show that

$$\Gamma > \gamma_{12}\left(1+\frac{\gamma_{13}}{\gamma_{23}}\right) \tag{1.20}$$

has to hold for $N_2 > N_1$, which can be fulfilled with moderate pumping under the conditions $\gamma_{12} \ll \gamma_{13}$ and $\gamma_{23} \gg \gamma_{13}$ [3].

**Exercise 1.1** *Consider an extension of the rate equations to the three-level case and neglect the induced terms. Under which condition for the pumping rate $\Gamma$ can population inversion between the second and first level be achieved?*

### Other Types of Lasers

Other types of lasers are gas lasers, in which the laser active medium is pumped by collisions with electrons or atoms and the transitions can be either electronic (He–Ne laser) or ro-vibronic ones ($CO_2$ laser).

In addition, there are semiconductor-based lasers, dye lasers, excimer lasers, to name but a few. Their working principles are described in some detail in [2, 6]. Another special laser type is the free electron laser (FEL), where a high speed electron beam is accelerated in a spatially modulated magnetic field and thereby emits coherent light. Recently, the principle of the FEL has been realized in two new large scale experiments. An infrared FEL has e.g., been built in Dresden (Rossendorf) and the FEL FLASH (formerly VUV-FEL) at DESY in Hamburg generates radiation in the soft X-ray regime.

A common principle in the experimental setup of all lasers is the fact that spontaneous emission (being a form of isotropic noise) should be suppressed. This is a difficult task, especially for high frequencies, however, due to the fact that $A \sim B\nu^3$, see (1.11), holds for the Einstein coefficients. Details of the experimental setup as e.g., the quality factor of the cavity have to be considered to understand how temporal fluctuation tend to get washed out, see e.g., [7].

## 1.3 Pulsed Lasers

Experimentally, lasers have led to a revolution in the way spectroscopy is performed. This is due to the fact that lasers are light sources with well-defined properties. They can be operated continuously in a single mode modus with a fixed or a tunable frequency or in a multi-mode modus [6]. However, more important for the remainder of this book is the possibility to run lasers in a pulsed mode. There the laser only oscillates for a short time span (e.g., some femtoseconds) with the central frequency of the atomic transition that is used.

### 1.3.1 Frequency Comb

Experimentally, ultrashort laser pulses can be created by using the principle of mode locking, explained in detail e.g., in [6]. We will shortly discuss the superposition of a central mode with side bands, underlying that principle below. The net result is shown in Fig. 1.6, where a train of femtosecond pulses coupled out of a cavity is depicted. Among other possible applications to be

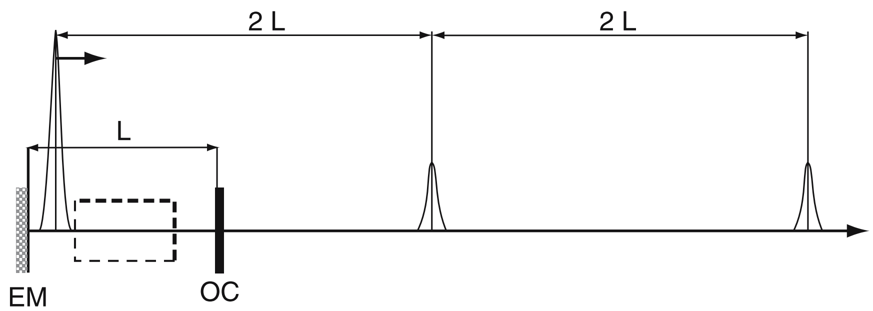


**Fig. 1.6.** Laser with end mirror (EM), output coupler (OC) and a pulse, propagating between EM and OC and being partially transmitted, from [9]

discussed in detail in later chapters, a pulse train can be used to measure frequencies very precisely [8].

How are the side bands obtained experimentally, and why does their superposition together with the central frequency $\nu$ lead to a train of pulses? The first question can be answered by considering the periodic modulation of the inversion with the frequency

$$\delta\nu = c/(2L) = 1/T_{\mathrm{RT}}, \tag{1.21}$$

corresponding to the round trip time $T_{\mathrm{RT}}$ of the light in the resonator. With the modulator placed at some position inside the cavity, the possible resonator modes with the angular frequencies

$$\omega \pm 2\pi n\delta\nu \tag{1.22}$$

with $\omega = 2\pi\nu$ and $n = 0, 1, 2, 3, \ldots$ are amplified [6]. Peaks at these equidistantly spaced frequencies are called the frequency comb.

To answer the second question, the amplitude of the electric field at a fixed point in space,

$$\mathcal{E}(t) = \sum_{n=-p}^{p} \mathcal{E}_n \cos[(\omega + 2\pi n\delta\nu)t + \varphi_n], \tag{1.23}$$

has to be considered, where $\varphi_n = n\alpha$ are the locked phases. A total of $2p+1$ modes shall have a gain above the threshold value. In the case of $\mathcal{E}_n = \mathcal{E}$ and for $\alpha = 0$ this leads to an intensity of

$$I(t) \sim \mathcal{E}^2 \left| \frac{\sin[(2p+1)\pi\delta\nu t]}{\sin(\pi\delta\nu t)} \right|^2 \cos^2(\omega t). \tag{1.24}$$

**Exercise 1.2** *Derive a closed expression for the electric field and the corresponding intensity in the case of the mode locked laser by using the geometric series.*

The intensity of (1.24) contains a term describing a fast oscillation with the central frequency and an envelope function leading to peaks separated by the round trip time $T_{\mathrm{RT}} = 1/\delta\nu$. Furthermore, the pulse length[3] is $T_{\mathrm{p}} \approx 1/\Delta\nu$, with the inverse width parameter $\Delta\nu = (2p+1)\delta\nu$, increasing linearly with the number of participating modes. The intensity as a function of time for three different total numbers of contributing modes is displayed in Fig. 1.7. The peak intensity increases proportional to $(2p+1)^2$, whereas the pulse length decreases with $1/(2p+1)$.

The effect of the pulse generation can also be understood in the photon picture. Those photons passing through the modulator at times where its

[3] Defined as the full width at half maximum of the intensity curve.

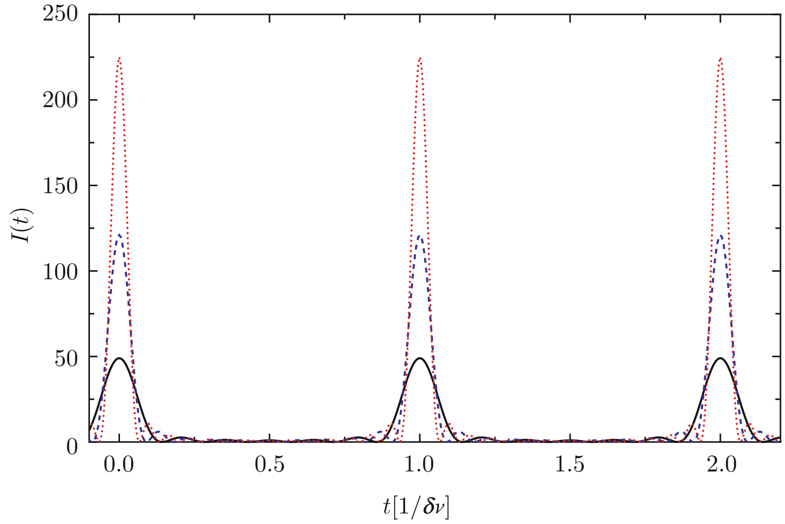


**Fig. 1.7.** Envelope of the intensity (arbitrary units) of a pulse train as a function of time for the superposition of 7 (*solid line*) 11 (*dashed line*) and 15 modes (*dotted line*)

transmission has a maximum will experience a minimum loss and the corresponding light will be maximally amplified. Enormously high intensities on the order of $10^{16}\,\mathrm{W\,cm^{-2}}$ can be generated using the principle of passive mode locking [6]. They prevail only for short times on the order of several femtoseconds, however. Pulses with 6 fs length are nowadays generated with Ti:Sapphire lasers with Kerr lens mode locking and operate at a center wavelength of 800 nm [10]. Only about two oscillations of the field are contained in such a short pulse at those wavelengths. The light is therefore extremely polychromatic. Many further details regarding experimental realization can be found in Chap. 3 of [9].

### 1.3.2 Carrier Envelope Phase

Let us look at the electric field of the last section in a bit more detail. It consists of an oscillation with the central frequency $\omega$ under an envelope and is plotted for a certain choice of parameters in Fig. 1.8.

The parameters in Fig. 1.8 have been chosen such that the peak separation does coincide with a half integer multiple of the period of the fundamental oscillation. This results in the fact that the phase of the fundamental oscillation is different by $\pi$, whenever the envelope has reached its next maximum. In general, this phase difference is the so-called carrier envelope phase (CEP) $\Delta\varphi$, and later-on we will frequently adopt the form

$$\mathcal{E}(t) = \mathcal{E}_0 f(t) \cos(\omega(t)t + \Delta\varphi) \tag{1.25}$$

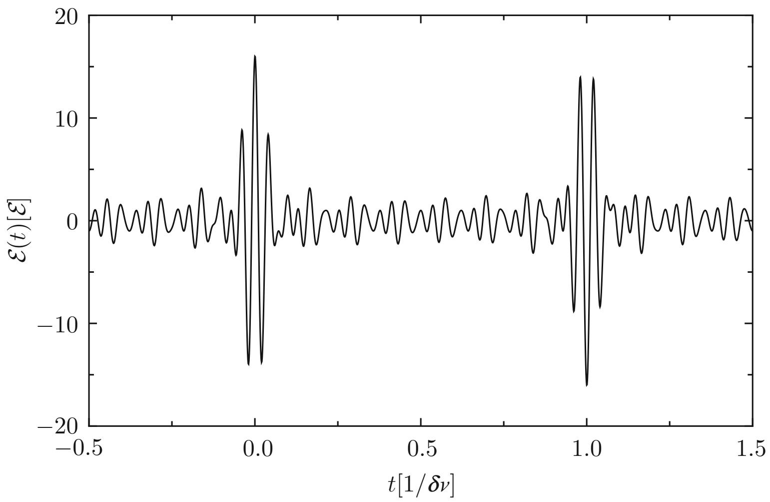


**Fig. 1.8.** Laser field consisting of the superposition of 17 modes with central frequency $\nu = 4$, $L = 3.0625$, and $\alpha = 0$ (all quantities in arbitrary units) as a function of time

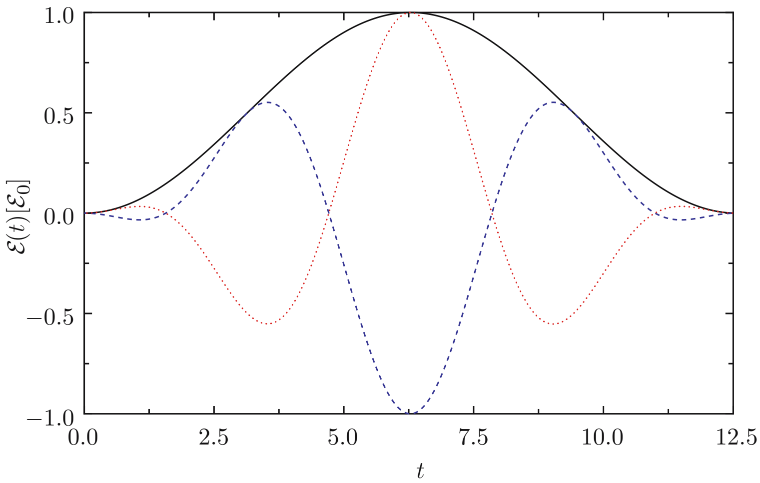


**Fig. 1.9.** Schematic laser field oscillation under a *single* pulse envelope (in units of $\mathcal{E}_0$) but with two different values of the carrier envelope phase as a function of time in arbitrary units, analogous to the two different pulses depicted in Fig. 1.8

of the laser field with an amplitude $\mathcal{E}_0$ and an envelope function $f(t)$, which is chosen from a large variety of suitable analytic functions. In addition, the frequency $\omega$ might be time-dependent and the carrier envelope phase can be varied, leading to tremendous effects as we will see later. In Fig. 1.9, a *single* pulse with oscillations corresponding to two different values of $\Delta\varphi$ is shown.

### 1.3.3 Husimi Representation of Laser Pulses

As mentioned in Sect. 1.3.2, we will model a laser pulse by an oscillation times a freely chosen pulse envelope. This is reasonable due to the fact that arbitrarily formed laser pulses can be generated experimentally by so-called pulse shapers [9]. We will make use of this fact in Chap. 5 in connection with the control of chemical reactions. In the following, the representation of the frequency content of general laser fields will be discussed.

A very intuitive way to characterize a laser pulse is given by a "windowed" Fourier transformation (or Husimi transformation)

$$F(\tau,\Omega) = \left| \int_{-\infty}^{\infty} \mathrm{d}t g(t-\tau)\mathcal{E}(t)\mathrm{e}^{-\mathrm{i}\Omega(t-\tau)} \right|^2 \tag{1.26}$$

with the window function

$$g(t) = \exp[-t^2/(2\sigma^2)]/\sqrt{2\pi\sigma^2}. \tag{1.27}$$

The function $F(\tau,\Omega)$ depending on a time-like variable, as well as on a frequency is also referred to as a spectrogram. It tells us at which time $\tau$ a certain frequency $\Omega$ is present in the original signal $\mathcal{E}(t)$. The term frequency resolved optical gating (FROG) is used for a measurement technique of a pulse which is designed by using (1.26) [9, 11]. In the field of molecular spectra, the term vibrogram [12] is used for a quantity which is constructed in a similar way from a time-signal called auto-correlation function, to be defined in the next chapter.

The case of two pulses which are temporally delayed with respect to each other will occur frequently later-on. For such a so-called pump–dump pulse, with slightly different central frequency of the pump versus the dump pulse, a spectrogram is shown in Fig. 1.10. The frequency change and also the temporal delay is clearly visible in the spectrogram. Also the case of a single pulse with a so-called "up chirp" (central frequency increasing as a function of time) or a "down chirp" (central frequency decreasing) are very obvious in a corresponding Husimi plot. To verify this for a simple Gaussian pulse envelope, a Gaussian integral has to be performed. This is by far not the last one that appears in this book and for convenience some Gaussian integrals are collected in Appendix 1.A.

**Exercise 1.3** *For the case of a linearly chirped frequency*

$$\omega(t) = \omega_0 \pm \lambda t/2$$

*calculate and schematically depict the Husimi transform of the pulsed field*

$$\mathcal{E}(t) = \mathcal{E}_0 \exp\left[-\frac{(t-t_0)^2}{2\sigma^2} + \mathrm{i}\omega_0(t-t_0) \pm \mathrm{i}\frac{\lambda}{2}(t-t_0)^2\right].$$

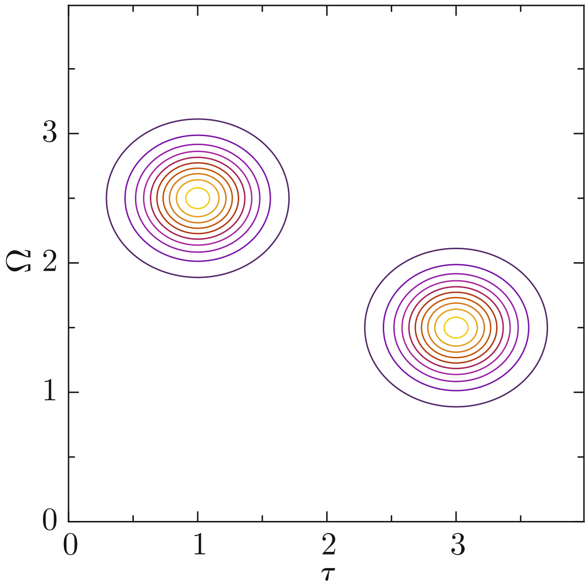


**Fig. 1.10.** Husimi transform of a pump–dump pulse as a function of $\tau$ and $\Omega$ in arbitrary units

## 1.A Some Gaussian Integrals

Throughout this book, Gaussian integrals will be encountered. For complex-valued parameters $a$ and $b$ with $\mathrm{Re}\, a \geq 0$, the following formulae hold:

$$\int_{-\infty}^{\infty} \mathrm{d}x \exp\{-ax^2\} = \sqrt{\frac{\pi}{a}}, \tag{1.28}$$

$$\int_{-\infty}^{\infty} \mathrm{d}x\, x \exp\{-ax^2\} = 0, \tag{1.29}$$

$$\int_{-\infty}^{\infty} \mathrm{d}x\, x^2 \exp\{-ax^2\} = \left(\frac{1}{2a}\right)\sqrt{\frac{\pi}{a}}, \tag{1.30}$$

$$\int_{-\infty}^{\infty} \mathrm{d}x \exp\{-ax^2 + bx\} = \sqrt{\frac{\pi}{a}} \exp\left\{\frac{b^2}{4a}\right\}, \tag{1.31}$$

$$\int_{-\infty}^{\infty} \mathrm{d}x\, x \exp\{-ax^2 + bx\} = \left(\frac{b}{2a}\right)\sqrt{\frac{\pi}{a}} \exp\left\{\frac{b^2}{4a}\right\}, \tag{1.32}$$

$$\int_{-\infty}^{\infty} \mathrm{d}x\, x^2 \exp\{-ax^2 + bx\} = \left(\frac{1}{2a}\right)\left(1 + \frac{b^2}{2a}\right)\sqrt{\frac{\pi}{a}} \exp\left\{\frac{b^2}{4a}\right\}. \tag{1.33}$$

A generalization of one of the formulae given above to the case of a $d$-dimensional integral that is helpful is

$$\int \mathrm{d}^d x \, \exp\{-\boldsymbol{x} \cdot \mathbf{A}\boldsymbol{x} + \boldsymbol{b} \cdot \boldsymbol{x}\} = \sqrt{\frac{\pi^d}{\det \mathbf{A}}} \exp\left\{\frac{1}{4}\boldsymbol{b} \cdot \mathbf{A}^{-1}\boldsymbol{b}\right\}. \tag{1.34}$$

As in the 1d-case, it can be proven by using a "completion of the square" argument. Furthermore, the convention that non-indication of the boundaries implies integration over the whole range of the independent variables has been used.

## Notes and Further Reading

The theory of the laser is treated on the level of the rate equations as well as in its full quantum version in the book by Haken [2] (the first book of the series [1,2] contains the derivation of Plancks's law, that we have followed) and by Shimoda [3]. In these books one can also find a more detailed discussion of the rate equations beyond the steady-state solution, especially concerning the build up of the oscillation.

A lot of information on the experimental aspects of lasers and about mode locking are contained in the book by Demtröder [6]. The handbook article by Wollenhaupt et al. deals with the properties, the creation via mode locking, and the measurement of femtosecond laser pulses [9]. It also contains a long list of additional references. The characterization of short pulses by using FROGs is the topic of [9, 11].

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