

Chapter 2

Theoretical Background

2.1 Passive Enhancement of Ultrashort Pulses

The setup of any femtosecond enhancement cavity (EC) experiment consists of three main components. Firstly, the pulses are generated by a mode-locked oscillator and optionally post-processed (e.g. amplified and/or spectrally broadened and temporally compressed etc.). The second component is the EC in which the laser pulses are overlapped coherently. If the EC is used for nonlinear conversion, it also contains the respective mechanism. And thirdly, one or more feedback loops are needed to ensure an interferometric overlap of the seeding laser pulses with the pulse(s) circulating in the enhancement cavity.

Ideally, a steady state is aimed for in which the enhanced electric field circulating in the passive cavity is a power scaled version of the seeding laser field. However, due to the fact that the spectrum emitted by a mode-locked laser consists of equidistant modes and the resonances of a passive cavity are in general not equidistant in the frequency domain, this task is highly challenging. To explain this challenge, we provide a theoretical background of the three main components. The different nature of the mechanisms underlying the generation of the pulses and their passive enhancement is addressed, and aspects particularly relevant for the experimental results presented in this thesis are emphasized.

2.1.1 Ultrashort Pulses from Mode-Locked Lasers

Mode-locking underlies the generation of the shortest light pulses directly from a laser oscillator. The advent of self-mode-locking techniques (see. e.g. [1]) merely 20 years ago and the subsequent development of phase stabilization in the last decade have had a significant impact on scientific research and technology. With this technique, light pulses comprising only a few oscillations of the electric field can be generated, confining the available laser energy to pulse durations on a femtosecond timescale.

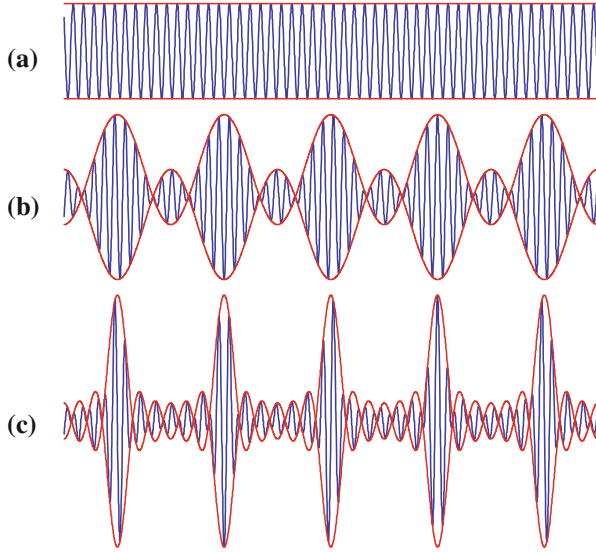


Fig. 2.1 The formation of ultrashort pulses: superposition of axial modes of an ideal Fabry-Perot resonator, oscillating with the same phase. The *red line* indicates the intensity profile for **a** a single mode, **b** for 3 and **c** for 7 modes

In addition, phase stabilization guarantees the pulse-to-pulse reproducibility of the electric field within the pulse intensity envelope. Pulses with these characteristics have marked the birth of ultrafast and attosecond science [1, 2], studying and controlling light-matter interactions at unprecedented intensity levels and atomic dynamics on previously inaccessible time scales. Phase-stabilized mode-locked lasers have also enabled high-precision optical frequency metrology. Their frequency spectrum is a regular structure of equidistant lines, called *frequency comb*, see e.g. [3–8]. The frequency comb can be parameterized by using only two radio frequencies (RF), which can be easily measured, e.g. with respect to the hyperfine transition of cesium at 9.193 GHz which provides the standard for one second. This constitutes a bridge transferring the measurement precision over the frequency gap between the RF spectrum and optical frequencies. A textbook-level description of the mode of operation of mode-locked lasers can be found e.g. in [9] or [10]. Here, we restrict the description to an overview of the basic mechanisms leading to the formation of a train of femtosecond pulses with a fixed phase relationship.

Mode locking refers to imposing a certain, fixed phase relation to the resonant axial (longitudinal) modes of a laser cavity. Figure 2.1 visualizes the intensity of (a) a single mode, i.e. continuous-wave operation, and the superposition of (b) 3 and (c) 7 modes of an ideal Fabry-Perot resonator, if these modes oscillate with identical phases. At a fixed time, or at a fixed space, the modes interfere constructively at equidistant maxima in space, or in time, respectively, which leads to the formation of light pulses. The more modes simultaneously resonant in the cavity, i.e. the broader the bandwidth

of contributing frequencies, the shorter the resulting pulses. However, in a real laser cavity, the circulating light pulses are subjected to dispersion, i.e. to a nonlinear dependence of the wavenumber $k(\omega)$ on the angular frequency ω . In general, for a pulse with a spectrum centered around the frequency ω_c , the wavenumber $k(\omega)$ can be expanded in a Taylor series:

$$k(\omega) = \frac{\omega_c}{v_{\text{ph}}} + \frac{\omega - \omega_c}{v_{\text{gr}}} + \frac{1}{2}GVD(\omega - \omega_c)^2 + \dots, \quad (2.1)$$

where v_{ph} , v_{gr} and GVD denote the phase velocity of propagation of the frequency component ω_c , the group velocity of the pulse and the group velocity dispersion, respectively (cf. e.g. Sect. 9.1 in [11]). The terms of orders 2 (i.e. GVD) and higher describe the dispersion accumulated by the pulse upon propagation and thus, the pulse lengthening in time. In the presence of non-zero net roundtrip dispersion, a pulse circulating in the laser cavity would broaden infinitely in time. In a mode-locked laser, however, the pulse reproduces after each round trip, which is equivalent to the fact that the effect of residual cavity dispersion on the pulse is compensated for upon each roundtrip. The intensity evolution of the pulse is given by the relative phase of the frequency components. Therefore, the nonlinear process of self phase modulation (SPM) can be used to perform this compensation and acts as a simultaneous switch for all active modes. The effect of SPM upon a roundtrip is determined by the interplay of the intracavity nonlinear mechanisms, such as the amplification and propagation in the active medium and (usually) an additional mode-locking mechanism. Dimensioning the phase effects in a mode-locked oscillator can be done by solving the nonlinear Schrödinger equation, see e.g. Sect. 10.3 in [11]. The interplay between the cavity dispersion, the mode-locking mechanism and the active material net gain bandwidth also determines the (achievable) output pulse duration of a mode-locked laser. A mathematical model of the mode-locking process can be found e.g. in [12, 13]. A decisive result is that the mode locking mechanism counterbalances the terms of order 2 and higher in the Taylor expansion in Eq. (2.1), implying:

$$k(\omega) = \frac{\omega_c}{v_{\text{ph}}} + \frac{\omega - \omega_c}{v_{\text{gr}}}. \quad (2.2)$$

This means in particular that the pulse can propagate with a different group velocity v_{gr} than the phase velocity v_{ph} of the wave with frequency ω_c . The following derivation will show that this fact is of crucial importance for the control of the electric field of the generated pulse train.

The steady-state resonance boundary condition of a linear Fabry-Perot oscillator with length L (or $2L$ for a ring cavity) is given by:

$$2k(\omega_N)L = 2\pi N, \quad (2.3)$$

where the integer N denotes the axial (longitudinal) mode number. The last two equations determine the frequencies of the modes of a mode-locked oscillator:

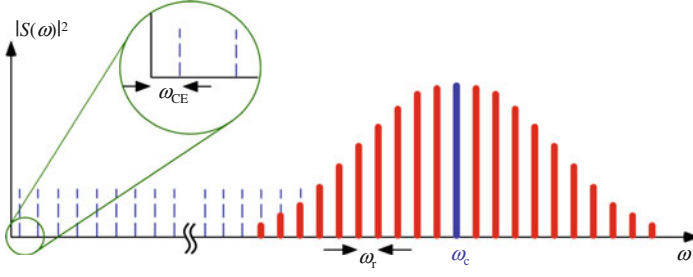


Fig. 2.2 Frequency-domain representation of the frequency comb emitted by a phase-stabilized mode-locked laser (as in [5]). The comb parameters are: the repetition frequency ω_r and the carrier-envelope frequency ω_{CE}

$$\omega_N = N \frac{2\pi}{2L} v_{gr} + \omega_c \left(1 - \frac{v_{gr}}{v_{ph}} \right). \quad (2.4)$$

Thus, the frequency difference between two adjacent modes amounts to:

$$\omega_{N+1} - \omega_N = \frac{2\pi}{2L} v_{gr} =: \omega_r \quad (2.5)$$

and corresponds to 2π times the repetition frequency of the pulses, which we will denote by ω_r .

The second term on the right-hand side of Eq. (2.4) represents an offset frequency, which we will denote by ω_{CE} :

$$\omega_{CE} := \omega_c \left(1 - \frac{v_{gr}}{v_{ph}} \right). \quad (2.6)$$

The abbreviation CE stands for *carrier-envelope* due to the significance of ω_{CE} in the time domain, which we will address shortly. In conclusion, the spectrum emitted by a stable mode-locked laser is a *comb of equidistant modes* with a spacing ω_r and offset from a multiple of ω_r by ω_{CE} :

$$\omega_N = N\omega_r + \omega_{CE}. \quad (2.7)$$

Figure 2.2 shows such a spectrum. With a spectral amplitude envelope $S(\omega)$, the electric field of the pulses in the time domain can be written as a Fourier series:

$$E(t) = \sum_{N=-\infty}^{\infty} S(\omega_N) e^{-j(N\omega_r + \omega_{CE})t}. \quad (2.8)$$

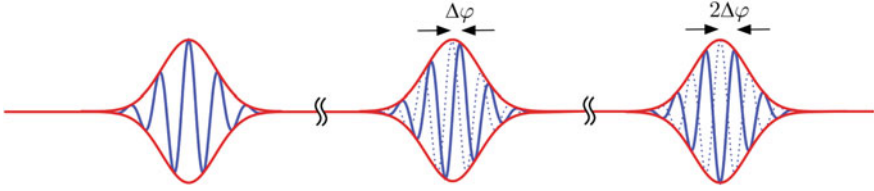


Fig. 2.3 Pulse-to-pulse evolution of the carrier-envelope offset for $\Delta\varphi = \pi/2$ (as in [5])

One cavity roundtrip period $T = 2\pi/\omega_r$ later, the electric field equals:

$$\begin{aligned}
 E(t + T) &= \sum_{N=-\infty}^{\infty} S(\omega_N) e^{-j(N\omega_r + \omega_{CE})(t+T)} \\
 &= E(t) e^{-j(2N\pi + 2\pi\omega_{CE}/\omega_r)} \\
 &= E(t) e^{-j\Delta\varphi}, \text{ with:}
 \end{aligned} \tag{2.9}$$

$$\Delta\varphi := 2\pi \frac{\omega_{CE}}{\omega_r}. \tag{2.10}$$

In other words, the electric field of each pulse produced by the laser is a copy of the electric field of the previous pulse, shifted by a phase $\Delta\varphi$. Due to the often used time-domain representation of the electric field as a continuously oscillating carrier wave times an envelope function, $\Delta\varphi$ is also called the “carrier-envelope offset phase slippage”, which also explains the name of ω_{CE} . Figure 2.3 visualizes the evolution of the carrier-envelope offset for the slippage phase $\Delta\varphi = \pi/2$. Stabilizing the pulse-to-pulse phase of a mode-locked laser is crucial for localizing the comb modes in the frequency domain as well as for time-domain applications which demand the reproducibility of the electric field rather than just that of the intensity profile. Carrier-envelope phase measurement and stabilization has been made possible in the last decade, see e.g. [3–8, 14–16].

The basic property enabling the enhancement of the pulses generated by a mode-locked oscillator in a passive cavity is the pulse-to-pulse reproducibility of the electric field, which is equivalent to the comb spectrum. Depending on the purpose of the enhancement, the pulses generated by the mode-locked oscillator can be further processed without affecting this basic property. Such processes include: *pulse picking*: reduction of the repetition rate; this generates new spectral components, in such a way that the comb spacing equals the reduced repetition rate; *sideband generation*: generation of additional frequency components through fast modulation, particularly useful for the Pound-Drever-Hall locking scheme (see Sect. 2.1.3); *pulse chirping*: manipulation of the spectral phase, in particular, allows for a variation of the pulse peak power while keeping the spectral components and the energy constant; *linear*

power amplification: scaling up the pulse energy while keeping the other parameters constant; *comb-preserving harmonic generation*: creates a frequency comb with the original comb spacing at a multiple of the central frequency.

2.1.2 Passive Enhancement in an External Cavity

Similar to a mode-locked laser, the steady-state condition of an enhancement cavity requires a pulse which is reproduced after each roundtrip. However, in contrast to a mode-locked oscillator, the latter does not incorporate an intracavity mechanism for dispersion compensation. Rather, the optical resonances of cavity, which are continuously excited by the seeding frequency comb modes, are not equidistant in the frequency domain due to roundtrip dispersion. This distinction from the seeding frequency comb affects the interference of the seeding with the circulating field, representing an enhancement limitation. Moreover, the fact that the circulating field has a different spectral amplitude and phase distribution than the seeding field requires increased attention when controlling the interference of the two. In the following, we first derive an analytical model for the steady state of a passive cavity excited by a frequency comb. Then, we use this model to discuss the trade-off between enhancement finesse and bandwidth.

The Steady State

Figure 2.4 shows the complex electric fields at the input coupler (IC) of an enhancement cavity in the steady state. Usually, enhancement cavities for nonlinear conversion are implemented as ring cavities. This is to avoid a double pass in opposite directions through the nonlinear material and to enable a straightforward spatial separation of the field seeding the cavity and the field reflected by the IC.¹ The following derivation, however, holds for both linear and ring cavities. Moreover, we assume perfect transverse mode matching at the IC, i.e. that the seeding laser beam is matched to the excited cavity transverse mode. For each frequency component ω , the single-roundtrip power attenuation and accumulated phase are denoted by $A(\omega)$ and $\theta(\omega)$, respectively. Thus, one cavity roundtrip of the circulating electric field component $\tilde{E}_c(\omega)$ can be completely described by its multiplication by $\sqrt{A(\omega)} \exp[j\theta(\omega)]$. Let $R(\omega)$ denote the IC power reflectivity, so that a reflection on the cavity side at the IC implies the multiplication of the impinging electric field $\tilde{E}_c(\omega)$ by $\sqrt{R(\omega)}$. Let $T(\omega)$ denote the IC power transmission. Resonant enhancement requires the constructive interference of the intracavity field reflected by the IC, i.e., $\sqrt{R(\omega)}\tilde{E}_c(\omega)\sqrt{A(\omega)}\exp[j\theta(\omega)]$ with the portion of the input field transmitted through the IC, i.e., $\sqrt{T(\omega)}\tilde{E}_i(\omega)$. In the steady state the sum of these two interfering fields equals $\tilde{E}_c(\omega)$:

¹ This comes in handy especially when using seeding systems sensitive to optical feedback.

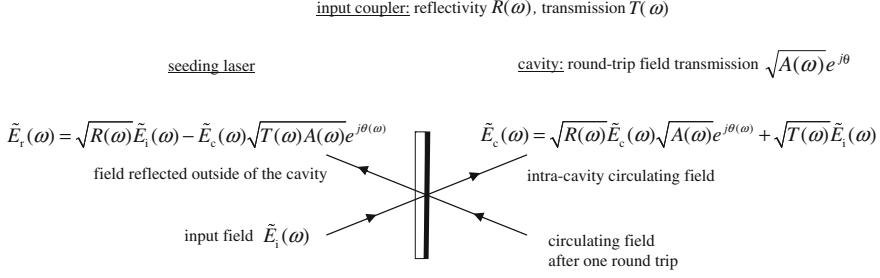


Fig. 2.4 Electric fields at the input coupler of an enhancement cavity in the steady state

$$\tilde{E}_c(\omega) = \sqrt{R(\omega)}\tilde{E}_c(\omega)\sqrt{A(\omega)}\exp[j\theta(\omega)] + \sqrt{T(\omega)}\tilde{E}_i(\omega) \quad (2.11)$$

$$\Leftrightarrow \tilde{H}(\omega) := \frac{\tilde{E}_c(\omega)}{\tilde{E}_i(\omega)} = \frac{\sqrt{T(\omega)}}{1 - \sqrt{R(\omega)A(\omega)}\exp[j\theta(\omega)]}. \quad (2.12)$$

For linear, i.e. intensity-independent behavior of the cavity, the Airy function $\tilde{H}(\omega)$ defined in Eq. (2.12) represents the transfer function of the cavity. However, the ratio $\tilde{H}(\omega)$ evaluated over the spectrum of the input field also bears significance if intracavity nonlinear processes are involved. In particular, $\tilde{H}(\omega)$ can be measured with high accuracy (see Sect. 4.2.2) providing information on the nonlinear process itself.

In a similar fashion, the superposition $\tilde{E}_r(\omega)$ of the input field reflected by the IC outside of the cavity and the intracavity field transmitted through the IC can be calculated:

$$\tilde{E}_r(\omega) = \sqrt{R(\omega)}\tilde{E}_i(\omega) - \tilde{E}_c(\omega)\sqrt{T(\omega)A(\omega)}\exp[j\theta(\omega)]. \quad (2.13)$$

Note that the minus sign between the two terms stems from a phase shift of opposed sign compared to the one between the two terms on the right-hand side of Eq. (2.11). This follows from the Fresnel equations and can also be explained by energy conservation. By plugging Eq. (2.12) in the above equation, we obtain:

$$\frac{\tilde{E}_r(\omega)}{\tilde{E}_i(\omega)} = \frac{\sqrt{R(\omega)} - [R(\omega) + T(\omega)]\sqrt{A(\omega)}\exp[j\theta(\omega)]}{1 - \sqrt{R(\omega)A(\omega)}\exp[j\theta(\omega)]}. \quad (2.14)$$

The entire seeding energy is coupled to the cavity if the numerator of the right-hand side of Eq. (2.14) equals 0. This *impedance matching* condition is fulfilled for a frequency ω , if on the one hand the roundtrip phase at this frequency is a multiple of 2π and on the other hand the reflectivity and transmission of the IC are matched to the roundtrip amplitude losses. For a lossless IC, i.e. if $R(\omega) + T(\omega) = 1$ holds, the second impedance matching condition becomes $R(\omega) = A(\omega)$.

Two further notions, historically stemming from optical interferometry, are important in the context of passive cavities. The *free spectral range* FSR of a resonator (or interferometer) denotes the resonance spacing in the frequency domain. For a dispersive cavity, the FSR is ω -dependent and $FSR(\omega) = \pi c_0 / [2n(\omega)L]$ holds, where c_0 , $n(\omega)$ and $2L$ denote the speed of light in vacuum, the frequency-dependent refractive index for the intracavity propagation and the cavity geometrical length, respectively. The cavity *fineness* $\mathcal{F}$ is a measure of the resolving power of the resonator used as a transmission filter (in the sense of an etalon or interferometer). It is defined as the ratio of the FSR to the FWHM bandwidth of a cavity resonance $\Delta\omega_{\text{cav}}$, see e.g. Sect. 11.5 in [11]:

$$\mathcal{F}(\omega) := \frac{FSR(\omega)}{\Delta\omega_{\text{cav}}}. \quad (2.15)$$

With the field propagation coefficient $r_{\text{tot}}(\omega) := \sqrt{R(\omega)A(\omega)}$ summing up the total losses of the frequency ω upon a roundtrip propagation in the passive resonator, the following equation holds for $\Delta\omega_{\text{cav}}$ (cf. Eq. (52) in [11], Sect. 11.5):

$$\Delta\omega_{\text{cav}} = \frac{4c}{2L} \arcsin \left(\frac{1 - r_{\text{tot}}(\omega)}{2\sqrt{r_{\text{tot}}(\omega)}} \right) \quad (2.16)$$

$$\approx \left(\frac{1 - r_{\text{tot}}(\omega)}{\pi\sqrt{r_{\text{tot}}(\omega)}} \right) \cdot FSR(\omega). \quad (2.17)$$

It follows for the finesse:

$$\mathcal{F}(\omega) \approx \frac{\pi\sqrt{r_{\text{tot}}(\omega)}}{1 - r_{\text{tot}}(\omega)} \quad (2.18)$$

$$\approx \frac{2\pi}{1 - r_{\text{tot}}^2(\omega)} = \frac{2\pi}{1 - R(\omega)A(\omega)}. \quad (2.19)$$

Thus, the finesse is given by the resonator losses and is independent of the resonator length.

Summary of Useful Formulas

In the following, we sum up the main equations for two important special cases:

- *Assumptions.* No dispersion, i.e. ω -dependence is discarded, lossless IC, i.e. $R + T = 1$ holds, and the cavity is on resonance. Then Eqs. (2.12) and (2.14) imply for the power enhancement P and the reflected portion of the input light $Refl$:

$$P := |\tilde{H}(\omega)|^2 = \left| \frac{\tilde{E}_c(\omega)}{\tilde{E}_i(\omega)} \right|^2 = \frac{T}{(1 - \sqrt{RA})^2}, \quad (2.20)$$

$$Refl := \left| \frac{\tilde{E}_r(\omega)}{\tilde{E}_i(\omega)} \right|^2 = \left(\frac{\sqrt{R} - \sqrt{A}}{1 - \sqrt{RA}} \right)^2. \quad (2.21)$$

- *Assumptions.* Like above but also impedance matching is given, i.e. $R = A$ holds:

$$P = \frac{1}{T}, \quad (2.22)$$

$$Refl = 0, \quad (2.23)$$

$$\mathcal{F} = \pi P. \quad (2.24)$$

Enhancement Trade-off: Finesse Versus Bandwidth

The roundtrip phase $\theta(\omega)$ describes the phase changes of a wave of frequency ω upon propagating in the cavity (usually in a transverse eigen-mode of the cavity) and interacting with its optics. Let us assume that a certain frequency ω_c is resonant in the cavity, i.e. the roundtrip phase accumulated by a wave with this frequency is a multiple of 2π . Then, $\theta(\omega)$ can be expanded in a Taylor series about ω_c :

$$\theta(\omega) = \theta_0 + GD(\omega - \omega_c) + GDD(\omega - \omega_c)^2 + \dots, \quad (2.25)$$

where θ_0 , GD and GDD denote a constant phase, the group delay and the group delay dispersion,² respectively. The constant phase term θ_0 has only a physical significance if a phase difference is considered, e.g. between two transverse modes of different orders (cf. Sect. 2.1.4). The group delay is determined by the repetition frequency of the pulse circulating in the cavity according to $GD = 2\pi/\omega_r$ and can usually be easily varied by tuning the geometrical cavity length.

Figure 2.5 shows the magnitude and phase of $\tilde{H}(\omega)$ for an impedance-matched (dispersion-free) cavity with a power enhancement of 100, which is resonant at ω_c , over a frequency range extending 5% of ω_r to the left and to the right of ω_c . If a continuous wave with a slightly different frequency than ω_c is exciting this cavity resonance (see e.g. green line in Fig. 2.5), the wave oscillating in the resonator will not only experience a smaller power enhancement than that of a wave with ω_c , but also a constant phase shift with respect to the exciting wave.

Equation (2.12) implies that roundtrip dispersion, i.e. GDD and higher-order terms in Eq. (2.25) and also frequency-dependencies of $\sqrt{A(\omega)}$ affect both the amplitude and the phase of $\tilde{H}(\omega)$. In particular, this means that the resonances of a real cavity are not equidistant in contrast to the seeding frequency comb. Thus, some of the

² Note that the GDD and the GVD are linked by the relationship $\theta(\omega) = k(\omega)d$ which implies $GDD(\omega) = GVD(\omega)d$, where d is a distance. While the GVD measures the dispersion upon propagation through a material in general, the GDD is used to characterize the dispersion of optical elements with a fixed geometry.

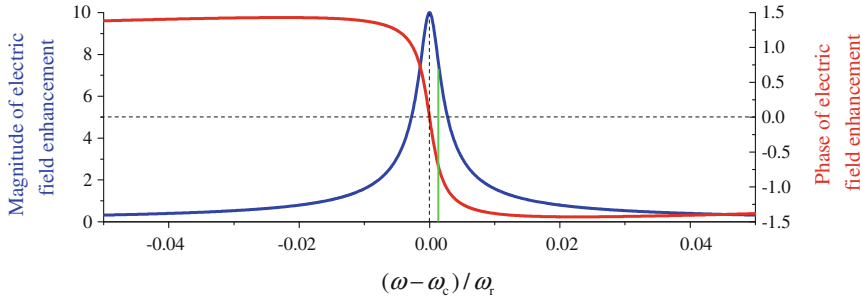


Fig. 2.5 Resonance of an impedance-matched cavity with a power enhancement of 100, centered at ω_c . *Green line* continuous wave with a slightly different frequency than ω_c , would experience a lower enhancement and a phase shift in the cavity

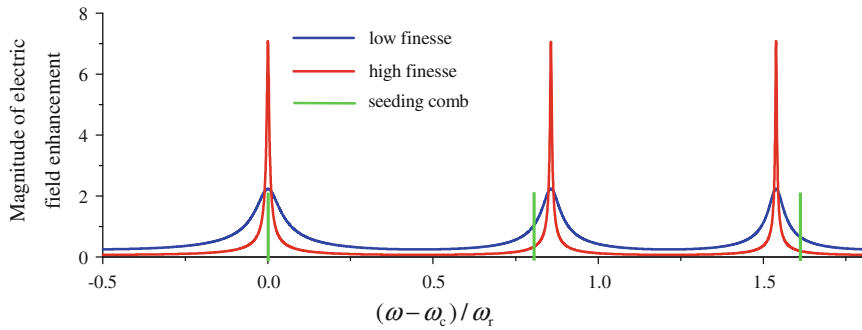


Fig. 2.6 Qualitative illustration of the enhancement finesse versus bandwidth trade-off. *Green* seeding comb frequencies, *red* high-finesse cavity with dispersion, *blue* low-finesse cavity with same dispersion. Spectral filtering and phase distortions are less pronounced for the low-finesse cavity. However, the power enhancement at the central mode is also smaller than in the high-finesse case

comb modes will necessarily be offset from the centers (magnitude peaks) of the corresponding cavity resonances, having a twofold effect on the circulating field. Firstly, the spectrum will not be evenly enhanced, meaning a spectral filtering of the seeding frequencies. Secondly, the phase of the circulating field will differ from the one of the input field. The dispersion imposed on the circulating field amounts to approximately the power enhancement factor times the roundtrip dispersion of the cavity (a derivation hereof can be found in Sect. 2.4 of [17]). A higher cavity finesse increases both effects. Thus, when dimensioning an enhancement cavity, a trade-off between finesse and bandwidth is necessary. Figure 2.6 illustrates this trade-off. The same frequency comb (green lines) is enhanced in a cavity with high finesse and in a cavity with low finesse. The high-finesse cavity enhances the central mode to a high degree but filters out the other modes strongly and also strongly affects the phase of the circulating field. The spectral filtering and the applied dispersion are much smaller in the case of the low-finesse cavity. However, the power enhancement in the

latter is also smaller. This trade-off is clearly noticeable in the enhancement cavity systems demonstrated so far, cf. Appendix 7.1.

2.1.3 Interferometric Stabilization

The strict condition of interferometric overlap of the input field with the intracavity circulating field at all times, usually implies the need for active locking of the seeding comb to the cavity resonances or viceversa. The example in Appendix 7.2 shows that even small length deviations on a picometer scale, common to a normal lab environment, can alter the enhancement significantly. In this section we first address the control variables in the context of comb-cavity locking and review the most common locking schemes. Then, we derive quantitative conditions under which the lock of the seeding comb and the enhancement cavity can be realized with a single feedback loop. This is particularly relevant for the design of an experimental setup.

In our discussion we assume (i) that all fluctuations are slow enough and (ii) that the cavity response $\tilde{H}(\omega)$ is linear, so that the steady states described in the previous two sections can be assumed. These assumptions usually apply for most mechanical and electronic distortions and they allow the discussion to be carried out in the frequency domain by using the comb and the cavity models given by Eqs. (2.7) and (2.12), respectively. We stress that processes on shorter time scales, such as nonlinear intracavity interactions, might introduce additional fluctuations, which are not accounted for by this discussion. Sect. 3.2.2) addresses this in more detail.

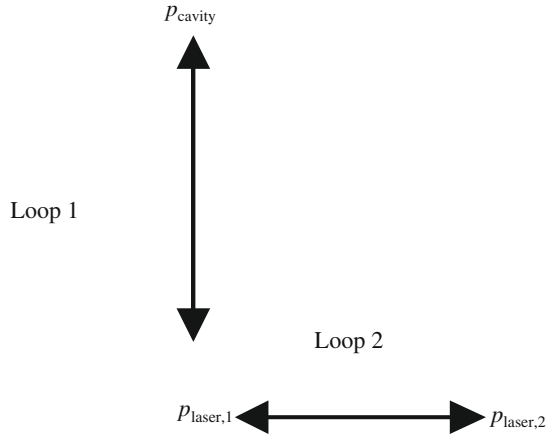
The Optimum Overlap

The frequency comb described by Eq. (2.7) can be parameterized by using two independent parameters. The two parameters need not necessarily be ω_r and ω_{CE} and for the sake of generality we will call them $p_{\text{laser},1}$ and $p_{\text{laser},2}$. In contrast, under the assumptions made above, the comb-like³ structure of the enhancement cavity response in the frequency domain, given by Eq. (2.12), is completely described by the propagation length along the cavity optical path for a given set of (dispersive) cavity optics. In other words, the cavity transfer function can be parameterized by a single parameter (once the cavity optics are given), which we will refer to as p_{cavity} .

In analogy to the trade-off between finesse and enhancement bandwidth discussed in the previous section, the spectral filtering property of the cavity owed to dispersion implies that different combinations of $p_{\text{laser},1}$, $p_{\text{laser},2}$ and p_{cavity} might be optimal for different enhancement purposes. For instance, obtaining the shortest possible circulating pulse or obtaining the highest enhancement in a certain spectral region might call for a different overlap between the frequency comb modes and the cavity

³ By the formulation “comb-like” we wish to emphasize that the cavity resonances are not equidistant.

Fig. 2.7 Feedback-loop connection of the parameters $p_{\text{laser},1}$, $p_{\text{laser},2}$ and p_{cavity} , enabling optimum overlap of the seeding comb with the cavity resonances. Active locking is indicated by the arrows



resonances in the same setup (cf. e.g. [18]). In general, there exists an *optimum overlap* of the seeding comb modes with the cavity resonances, which can be described by a well defined combination of $p_{\text{laser},1}$, $p_{\text{laser},2}$ and p_{cavity} .

The train of thought followed so far has two immediate consequences. On the one hand, the single cavity parameter does not in general suffice to reach optimum overlap, since the seeding comb requires two parameters for a full description. Thus, at least one degree of freedom of the overlap needs to be controlled by means of the seeding comb. On the other hand, if one of the comb parameters, say $p_{\text{laser},1}$, and the cavity parameter p_{cavity} are locked, the other comb parameter, i.e. $p_{\text{laser},2}$, is unambiguously determined by $p_{\text{laser},1}$ according to a constraint set by the optimum overlap (e.g. shortest intracavity pulse or highest intracavity power). In particular, this means that $p_{\text{laser},2}$ can be locked to $p_{\text{laser},1}$, without a direct feedback from the cavity, which is illustrated in Fig. 2.7.

Choice of the Control Parameters: Practical Considerations

The main fluctuations causing deviations from the optimum overlap and occurring in every real experimental setup are of mechanical and electrical nature. In general, mechanical vibrations affect all parameters, while electrical fluctuations affect mostly the laser (see e.g. [19, 20]), since the cavity is passive. Due to the effect of mechanical vibrations on the lengths of both the oscillator cavity and the enhancement cavity, it is common to stabilize the length of one of these cavities to match the length of the other one. If an enhanced frequency comb is desired for which the absolute frequencies are highly precise (e.g. for spectroscopy), then it is customary to lock the enhancement cavity to the seeding laser (which is locked to an external reference). However, if the precise knowledge of the absolute value of the enhanced modes is not mandatory, as is the case with most time-domain applications, the passive cavity

can be used as a reference and one of the oscillator parameters can be locked to the cavity parameter. In practice this implementation is usually less demanding due to the lower power in the oscillator cavity (see also Sect. 4.1). In conclusion, we can thus assume without loss of generality that the cavity resonances are fixed, i.e. p_{cavity} is constant, and the two comb parameters are used to compensate deviations from the optimum overlap, which can be divided into two independent locking tasks (cf. Fig. 2.7):

- Loop 1: $p_{\text{laser},1}$ controls the frequency of a comb mode (e.g. ω_c), so that this mode is locked to (usually the peak of) a cavity resonance, which is determined by p_{cavity} .
- Loop 2: $p_{\text{laser},2}$ is set by using only feedback from $p_{\text{laser},1}$, with the boundary condition that optimum overlap is reached.

The complex dynamics of mode locking (see e.g. [13, 19]) makes it very difficult to vary the two comb parameters ω_r and ω_{CE} completely independently. In general, the available control mechanisms (like piezoelectric actuators, acousto- and electro-optical modulators, dispersion management, optical pump power variation, etc.) act on both ω_r and ω_{CE} . Therefore, in practice, the parameters $p_{\text{laser},1}$ and $p_{\text{laser},2}$ are two linear combinations of ω_r and ω_{CE} , which need to satisfy two necessary conditions: (i) they need to be linearly independent and (ii) they need to be tunable over a region large enough around the optimum overlap so that all occurring fluctuations can be compensated for. Under these two conditions, the comb needs to be brought in the vicinity of the optimum overlap in the parameter space defined by $p_{\text{laser},1}$ and $p_{\text{laser},2}$ by coarse adjustment and, subsequently, active stabilization will ensure an optimum overlap. In the following paragraph, we review the most common schemes for the two locking tasks, i.e. $p_{\text{laser},1}$ to p_{cavity} and $p_{\text{laser},2}$ to $p_{\text{laser},1}$.

Common Locking Schemes

An active feedback loop consists of three main components: an error signal, indicating the deviation of the actuating variable from the desired optimum, locking electronics setting the control mechanism according to this error signal, and the control mechanism itself. For the lock of a comb line to a Fabry-Perot cavity with an Airy-function response as given by Eq. (2.12) and plotted in Fig. 2.5, the control mechanism can be a piezo-actuated mirror, the optical pump power of the oscillator, an acousto- or electro-optical modulator etc.. The locking electronics are usually a standard, commercially available phase-locked loop. Rather than providing a deep insight into the mode of operation of these standard electronic components, we address in the following, the generation of the error signal, which represents the interface of the optical setup, and the task of interferometric stabilization, which is an electronic one.

The necessary property every error signal needs to fulfill, is bipolarity, i.e. the signal should indicate unambiguously the direction (and magnitude) of the drift which needs to be compensated for. For example, the intensity of the optical signal transmitted through a cavity mirror, as a function of the mismatch between the driving

frequency and the cavity resonance, is not suited as an error signal for locking the peak of the resonance, since the Airy function is an even function (see Fig. 2.5). However, this signal is suited to lock the driving frequency to a sub-maximum level on one side of the Airy fringe, e.g. the position indicated by the green comb line in Fig. 2.5. This scheme is usually referred to as *side-of-fringe locking* and was first employed to stabilize (CW) lasers to a reference cavity [21] rather than to enhance a field with the purpose of increasing the available power.

Schemes able to lock a driving frequency to the peak of a cavity resonance require a bipolar error signal that equals 0 at the peak of the resonance and changes its sign when the mismatch passes through this zero. Such signals are usually obtained from observing the interference of a portion of light in phase with the driving frequency (“reference”) and a portion of intracavity light that contains the phase information of the cavity (“sample”).

The *Hänsch-Couillaud* or *polarization locking* scheme [22] works for polarization discriminating cavities. The linearly polarized light seeding the cavity needs to contain nonzero components in two orthogonal polarization directions, e.g. parallel and perpendicular to the intracavity polarization discrimination direction. For each polarization component, the overall field reflected by the cavity is given by Eq. (2.14) and represents the superposition of a portion of the seeding light which serves as the “reference” and a portion of transmitted light, which carries the phase information of the cavity and serves as the “sample” part of the interferometer. The “sample” portion is linearly polarized, but, due to the polarization discrimination, its decomposition along the two polarization directions has a different ratio of coefficients than the reference part of the beam. When the cavity is on resonance, the phase difference between the two linearly polarized parts is 0 so that their superposition is linearly polarized. However, when the cavity is off-resonance, the sample part has a frequency-dependent phase shift with respect to the reference part, manifesting itself as an elliptical polarization of the reflected beam. The direction and the magnitude of the ellipticity can readily be detected with an analyzer consisting of a quarter-wave plate, a polarizing beam splitter and a difference photodiode, providing the error signal. The cavity in the proof-of-principle paper [22] contains a Brewster plate which provides strong polarization discrimination. However, recently we have shown that the nonorthogonal incidence of the beam on the cavity mirrors provides a polarization discrimination large enough for a stably working Hänsch-Couillaud lock [17, 23]. The results presented in this thesis have been obtained using the Hänsch-Couillaud locking scheme.

Several locking schemes, which we summarize under the name of *transverse mode mismatch locking*, use the fact that an optically stable cavity has a set of well-defined transverse eigen-modes. If this cavity is excited externally and a mismatch between the exciting beam and the excited transverse mode is introduced, a spatial interference of the part of the input beam which is not matched to the cavity mode (reference) with the beam resonant in the cavity (sample) can be produced. A photodetector placed at a proper position generates a bipolar error signal. The mismatch can be e.g. given by a non-mode-matched input beam [24, 25] or by a tilted input beam [26]. While these methods are easy to implement and low-cost, they are inherently prone

to misalignment and to changes of the input beam and the excited cavity transverse mode.

One of the most robust and widely used stabilization schemes today is the *Pound-Drever-Hall locking* scheme, see e.g. [27–29] and references therein. The method was initially developed by Pound [30] for microwave frequency stabilization of and later adapted for optical oscillators [27]. By modulating the input light with an RF source (local oscillator), sidebands of the frequency to be locked are produced. If the distance of the sidebands from the resonant frequency in the frequency domain is large enough so that these sidebands are not resonant, the sidebands can be used as the reference. The signal reflected by the input coupler of the resonant cavity carries the desired phase information and can be used as the sample signal in a phase-sensitive heterodyne detection scheme which demodulates the reflection signal against the RF source [27].

The second stabilization task consists in locking $p_{\text{laser},2}$ to $p_{\text{laser},1}$. As discussed in the beginning of this subsection, this stabilization concerns the laser only. While the repetition frequency of the laser is easy to access as an RF signal from a photodiode, the detection of the CE phase slippage $\Delta\varphi$ is somewhat more intricate. The most common and straight-forward scheme for the detection of ω_{CE} is based on an f -to- $2f$ interferometer, see e.g. [5] and references therein. Here, an octave-spanning frequency comb is employed to generate a beat signal between the mode with the number N and the one with the number $2N$. Such a broad spectrum can be generated e.g. by self-phase modulation in a nonlinear crystal. The two frequencies $\omega_N = N\omega_r + \omega_{\text{CE}}$ and $\omega_{2N} = 2N\omega_r + \omega_{\text{CE}}$ generate a beat note with an RF frequency corresponding to their frequency difference, i.e. $\omega_{2N} - \omega_N = \omega_{\text{CE}}$. Controlling ω_{CE} can be done in manifold ways. A slow control can be obtained by varying the length of the oscillator beam path through a dispersive material, which influences the ratio of the group to the phase velocity, see e.g. [31]. Recently, a related method was demonstrated in which a composite plate is used to vary this ratio without affecting the repetition rate [32]. Faster controls can be achieved by modulating the power of the pumping beam, see e.g. [19, 20, 33] or by shifting the pumping beam with respect to the cavity mode [34].

Locking a Single Degree of Freedom

The necessity to actively control a second degree of freedom (i.e. the lock of $p_{\text{laser},2}$ to $p_{\text{laser},1}$) mainly depends on the jitter, on the optical bandwidth, on the cavity finesse and on the required stability of the enhanced frequency comb. For many practical applications, fluctuations of two orthogonal degrees of freedom, e.g. of ω_r and ω_{CE} around the state of optimum overlap have a very similar effect on the enhancement, making a second active loop unnecessary. This will be shown quantitatively in the following calculation, in close analogy to Sect. 2.1.2 of our paper [35].

For our derivation, we assume that the central optical frequency ω_c is being locked to the peak of a corresponding cavity resonance by actively controlling solely the repetition frequency ω_r of the comb, and we calculate the effect of a variation $\Delta\omega_{\text{CE}}$

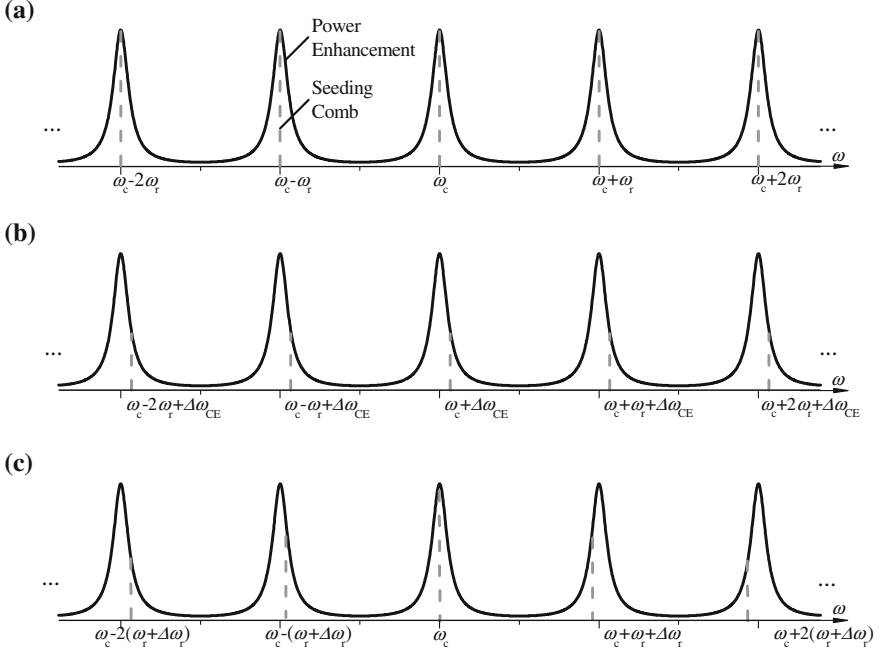


Fig. 2.8 Illustration of the effect of a shift $\Delta\omega_{\text{CE}}$ of the carrier-envelope offset frequency ω_{CE} on the enhanced frequency comb, if the laser-cavity lock is implemented by controlling the repetition frequency ω_r so that the central optical frequency ω_c is locked to the peak of a cavity resonance. **a** Optimum overlap. **b** Shift of ω_{CE} by $\Delta\omega_{\text{CE}}$. **c** Compensation of $\Delta\omega_{\text{CE}}$ through $\Delta\omega_r$ at ω_c

of the other comb parameter ω_{CE} on the enhancement. In the context of the laser-cavity lock the following description of $\theta(\omega)$ is convenient:

$$\theta(\omega) = \tau\omega + \psi(\omega) = 2\pi \frac{\omega}{\omega_r} + \psi(\omega), \quad (2.26)$$

where $\tau = 2\pi/\omega_r$ is the round trip group delay and $\psi(\omega)$ is a term describing the residual intra-cavity dispersion. For simplicity, here we assume that $\psi(\omega) = 0$ and that an initial ω_{CE} is given, which leads to an optimum overlap of the seeding comb modes with the cavity resonances, see Fig. 2.8a. Let N_c denote the mode number of ω_c , so that

$$\omega_c = N_c\omega_r + \omega_{\text{CE}} \quad (2.27)$$

holds. A variation of the comb parameter ω_r , meaning a change of the comb mode spacing, can be entirely compensated for by the active control. In contrast, a variation $\Delta\omega_{\text{CE}}$ of the carrier-envelope offset frequency shifts the entire frequency comb, as depicted in Fig. 2.8b. The active control compensates the drift of ω_c by introducing a variation $\Delta\omega_r$ to ω_r :

$$\omega_c = N_c(\omega_r - \Delta\omega_r) + \omega_{CE} + \Delta\omega_{CE}. \quad (2.28)$$

The last two equations imply:

$$\Delta\omega_r = \Delta\omega_{CE}/N_c. \quad (2.29)$$

While the mode with the number N_c retains its initial frequency, the mode with the number $N_c + m$ will experience a frequency shift equal to $m\Delta\omega_r$. This situation is depicted in Fig. 2.8c.

In Fig. 2.9, the effect of a variation $\Delta\omega_{CE}$, corrected by a variation of ω_r as described above, on the enhanced pulse is plotted for several enhancement factors and several optical bandwidths. As a result of this correction the circulating power decreases and the intra-cavity spectrum is narrowed relative to the case of optimum overlap of the seeding comb with the cavity resonances. The relative changes are plotted as functions of $\Delta\omega_{CE}$ and of $\Delta\omega_{CE}/(2\pi)\Delta\omega_{cav}$, where $\Delta\omega_{cav}$ is the full-width-half-maximum of the cavity resonance. The optical bandwidths used in the calculated examples represent typical values used in femtosecond enhancement cavity experiments. The majority of the experiments presented in this thesis are performed with parameters close to the 7-nm case shown in Fig. 2.9c. Throughout these experiments, the seeding comb could be locked to the cavity by using a single feedback loop, cf. Sect. 4.1. However, a high-finesse enhancement of pulses with broader bandwidths might require a second feedback loop, cf. Sect. 5.1.2.

2.1.4 Transverse Modes, Cavity Scan and Quasi-Imaging

The transverse modes of a resonator (and also of free space or a waveguide) are self-consistent transverse electromagnetic field distributions. A textbook treatment of optical resonator transverse modes is given in Sects. 1.6 and 14.3 of Siegman's book *Lasers* [11]. The most relevant theoretical aspects for the work presented in this thesis are presented in Sect. 2 of the paper [36], see also Chap. 6. In this section we emphasize two further aspects.

Cavity Scan

The scan of the oscillator length generating the seeding pulses around the value of the enhancement cavity length is explained in Sect. 2.3 of the paper [36] (see also Chap. 6) and in Figs. 3 and 5b therein, in the context of adjusting the relative phase between higher-order transverse modes. This is particularly useful to enable the simultaneous resonance of a subset of cavity eigen-modes, which can then be coupled by means of obstacles in the beam path to tailor new low-loss transverse modes. This technique, which we call *quasi-imaging*, is addressed in the next paragraph. Besides enabling quasi-imaging adjustment the scan pattern is also a powerful diagnostic

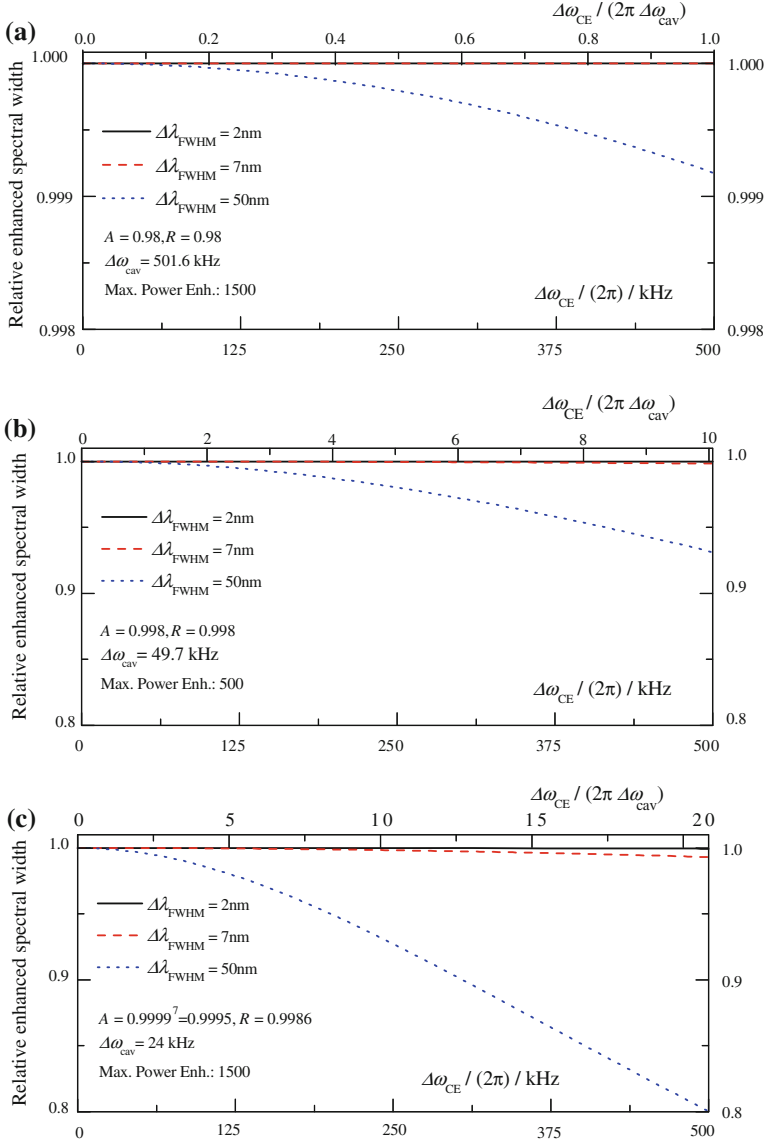


Fig. 2.9 Calculated reduction of the enhanced spectral width (FWHM) after a carrier-envelope offset frequency drift $\Delta\omega_{\text{CE}}$ has been compensated for by a shift of the repetition frequency ω_r , locked at the central optical frequency ω_c . The reduction is related to the case of optimum overlap between the frequency comb modes and the cavity resonances. A Gaussian input spectrum is assumed. In a good approximation, the total power enhancement is proportional to the spectral narrowing and the pulse lengthening is inversely proportional to the spectral narrowing. **a** and **b** impedance matched cases with power enhancements at ω_c of 50 and 500, respectively. **c** power enhancement of 1,500 at ω_c , not impedance matched, corresponding to our experimental setup

tool, as discussed in the following.⁴ Section 4.2.2 includes examples for all points listed here.

- *Optimum overlap of the axial comb modes of the seeding pulses with cavity resonances.* The scan pattern can be used to adjust the two laser parameters and the cavity parameter manually and coarsely so as to be in the vicinity of the optimum overlap so that subsequent active stabilization ensures a steady state in the optimum overlap, as discussed in the previous section. For sufficiently small cavity dispersion, this is achieved when the main resonance in the scan pattern reaches a maximum and the peaks of the two resonances, one free spectral range to the left and to the right of the main resonance, have equal heights.
- *Transverse mode matching.* The scan pattern also provides information on the amount of light coupled to transverse modes other than the desired one. For instance, in Sect. 3 of [36] the peaks corresponding to higher-order transverse modes are used to calculate the overlap of the input beam with the excited modes. If the target alignment is a perfect transverse mode matching then these peaks must vanish.
- *Error signal adjustment.* As discussed in the previous section, the bipolar error signal for locking should indicate unambiguously the direction and magnitude of the drift which needs to be compensated. Important parameters of the error signal, like amplitude and balancing can be adjusted while scanning over a cavity resonance.
- *Finesse and nonlinearities.* For a given seeding frequency comb, the ratio of the main peak to the ones located a free spectral range to the left and to the right provides a measure for the spectral filtering of the cavity (as discussed in the previous section), which increases with a higher finesse and with dispersion, both of which might also be intensity-dependent. This diagnostic is particularly useful in power scaling experiments, where the (average or peak) power of the seeding comb is increased while all other parameters are kept constant.
- *Stability range detuning and roundtrip phase difference between the sagittal and tangential directions.* As explained in [36], the scan pattern contains information about the detuning from the center of the stability range (see also next paragraph). Moreover, a difference of the Gouy parameters for the x and the y directions in the cavity leads to different on-axis phases⁵ of the higher-order modes with the same sum of x and y indices. This results in a multitude of peaks in the scan pattern corresponding to a group of resonances with identical index sum. Conversely, the roundtrip phase difference between the x and the y directions can be determined from this scan pattern (if the finesse is large enough so that these peaks are well resolved). For example, Fig. 5b in Sect. 3 of [36] shows a case in which the Gouy

⁴ The frequency-domain scan of a Fabry-Perot resonator is not only a helpful diagnostic for the enhancement in this resonator, but a measurement tool in its own right: a cavity with variable length can be used as a passive tunable filter or as a scanning interferometer, see e.g. Sect. 11.5 in [11], scanning a CW laser through a cavity resonance can be employed for ringdown measurements [37] etc.

⁵ The accumulation of different on-axis phases manifests itself in different *optical cavity lengths*.

parameters are $\psi_x = 0.78 \cdot 2\pi$ and $\psi_y = 0.76 \cdot 2\pi$. In a stable resonator, in which the beam lies in a single plane, this difference is caused by the difference of the effective radius of curvature of the (de)focusing mirrors thus, indicating the angles of incidence on these mirrors. In an out-of-plane stable resonator this diagnostic can e.g. be used to adjust the angles of incidence on the curved mirrors so that the Gouy parameters in both directions are equal. In this case all peaks corresponding to modes with the same sum of x and y indices coincide. In particular, this could be used to adjust quasi-imaging in both directions, see Sect. 5.2.

Mode Degeneracy and Quasi-Imaging

A phenomenon which can be very useful if controlled properly, and harmful if unwanted, is transverse mode degeneracy. Several transverse modes of different orders can be simultaneously resonant. Intracavity phase front distortions can then lead to a coupling of these modes [38], influencing the excited mode in the cavity.

On the one hand, if fundamental-mode operation is desired this phenomenon leads to a degradation of the beam quality, as discussed in [38]. Such phase front distortions can for instance be induced by thermal or nonlinear effects in the resonator optics. We have observed this in experiments involving an intracavity Brewster plate for XUV output coupling, cf. Sect. 4.4.1. It has also been reported that the gas target can lead to such phase front distortions [39]. The consequence of this unwanted effect is an increase of the roundtrip losses for the distorted mode, possibly a decrease of the overlap with the exciting mode and thus, overall a decrease of the enhancement and of the HHG efficiency. The scan pattern can be used to adjust the relative phases of the transverse modes to avoid resonant coupling among them.⁶

On the other hand, the phenomenon of transverse mode degeneracy, together with the proper choice of obstacles in the beam path and of the seeding transverse mode, enables the controlled combination of eigen-modes of a cavity resulting in a tailored mode with desired properties. An example thereof is the quasi-imaging (QI) concept presented in the paper [36], see also Chap. 6 (and also Sect. 5.2). The initial motivation of the development of QI was to obtain a field distribution which simultaneously exhibits an on-axis maximum for HHG in a gas target close to a cavity focus and, after a certain propagation distance, an on-axis low-intensity region for coupling out the intracavity generated XUV radiation through an opening in a cavity mirror in this region. However, this application can be regarded as a special case of the more general concept of exciting a combination of degenerate transverse modes in a cavity, among which the coupling can be tuned with high precision by means of the position in the stability range. This enables the confinement of different processes to spatially separated regions within the cavity and, at the same time, a precise control of the energy coupling between these regions and therefore between the respective

⁶ However, the average power during the scan is different from the steady-state regime. Therefore, distortions due to thermal effects might not be visible in the scan pattern.

processes. This concept might prove to be useful for tasks other than providing a direct on-axis access to a high-finesse cavity.⁷

2.2 High-Harmonic Generation in a Gas

For longer than the past two decades the process of high-harmonic generation (HHG) has been extensively studied, both experimentally and theoretically. Owing to its immediate relation to the generation of XUV radiation, such studies have been playing a central role at our institute. In particular, in the context of enhancement cavities, theoretical descriptions of HHG have recently found their way into several PhD theses, e.g. [40–43]. Rather than reproducing in detail the different models describing HHG, this section briefly outlines the semiclassical HHG model known as the *three step model*, and subsequently addresses phase matching considerations, aimed at providing an intuitive introduction to HHG and at enabling the interpretation of the experimental results presented here as well as of the challenges faced towards further power scaling. At the end of the section, we sum up scaling laws for HHG.

2.2.1 The Three-Step Model and Phase-Matching

The Three-Step Model of Single-Atom HHG

The specific structure of high-harmonic spectra generated by multi-cycle laser pulses consists of odd harmonics, whose envelope exhibits: (i) an exponential fall-off (perturbative region) for the lower-order harmonics, (ii) a plateau region of nearly equal power per harmonic, succeeded by (iii) a cutoff in the short-wavelength part of the spectrum. Experimentally, this structure has first been observed in the late eighties [44, 45]. The three step model, developed by Corkum et al., see [46] and references therein, provides a semiclassical, intuitive approach to HHG, explaining many of its properties. In 1994, Lewenstein et al. justified this model with quantum mechanical considerations [47]. In the following, we outline the three step model, explaining the response of a single atom to a high-intensity field. The model is illustrated in Fig. 2.10.

In the first step, the highly intense driving electric field bends the Coulomb potential barrier of the atom so that an electron tunnels from the atomic ground state. A necessary condition for this model to hold is that the so-called Keldysh parameter

⁷ For instance, it might enable the implementation of the seeding oscillator resonator (with its active medium and mode-locking mechanism) and the passive enhancement resonator in the same cavity (here, *cavity* designates a hollow space rather than a resonator). This could simplify the experimental setup considerably and in particular, spare the necessity of active synchronization.

Fig. 2.10 Three step model: 1 tunnel ionization, 2 acceleration through the laser field, 3 recombination to the ground state and emission of an XUV photon. This image is reproduced from [48] by courtesy of Matthias Kling

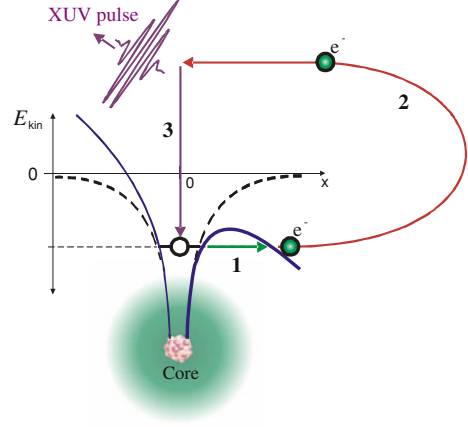


Table 2.1 Ionization potentials (from Sect. 12.2.1 in [49]).

Gas	Xe	Kr	Ar	Ne	He
U_i/ev	12.13	13.99	15.76	21.6	24.6

$$\gamma = \sqrt{\frac{U_i}{2U_p}} \quad (2.30)$$

is smaller than one [40]. Here, U_i is the ionization potential of the atom, see Table 2.1, and U_p is the ponderomotive energy, i.e. the average kinetic energy of an electron in a harmonic field, given by:

$$U_p = \frac{e^2 E_0^2}{4\omega^2 m_e} \quad (2.31)$$

$$= \frac{e^2}{4\omega^2 m_e} \cdot \frac{2I}{c\epsilon_0}, \quad (2.32)$$

where e , m_e , ω , ϵ_0 , E_0 and I are the electron charge and mass, the optical frequency, the vacuum permittivity, the electric field amplitude and the laser beam intensity, respectively. Plugging in the natural constants and using the common measurement units W/cm^2 and μm for I and λ , respectively, yields:

$$U_p \approx 9.33 \cdot 10^{-14} I \lambda^2. \quad (2.33)$$

For $\gamma < 1$ the tunneling times of the electron through the bent Coulomb barrier are short compared to the optical cycle, and the tunnel ionization model implies the

formation of a sequence of wave packets, one close to each electric field maximum of the driving multi-cycle laser pulse.

The second step concerns the acceleration of the electron in the laser electric field. It uses classical mechanics to describe well-defined trajectories for the electrons. The trajectories along which the electron wave packets return to the parent ion lead to recombination, which constitutes the third step. Upon recombination, a photon is emitted with an energy equal to the sum of the kinetic energy acquired during acceleration and the ionization potential, i.e. $\hbar\omega = E_{\text{kin}} + U_i$. The trajectory and thus, the energy of the emitted photon, depends on the time, i.e. the phase of the driving field, at which the ionization takes place. The model implies that the highest kinetic energy is reached if the electron tunnels at $\omega t \approx 0.3 + k\pi$ rad for any integer k . In this case, the absolute maximum of the kinetic energy amounts to $3.17U_p$, which defines the cutoff frequency via:

$$\hbar\omega_{\text{cutoff}} = 3.17U_p + U_i. \quad (2.34)$$

The typical odd-harmonic spectrum originates from the interference of the spectra of the radiation emitted by the different optical cycles of the driving pulse. The cycle-to-cycle repetition of the wave packets passing the parent ions implies that any light that is emitted will be at an odd harmonic of the laser frequency. In the case of single-cycle driving pulses, this interference vanishes and the harmonic spectrum exhibits a rather continuous shape.

Another important result is that only close-to-linear polarization leads the tunneled electron back to the atom and thus to recombination. This is also the key to all isolated as-pulse generation schemes involving polarization gating, see Sect. 5.3.

Multi-Atom HHG: Phase Matching

The response of a macroscopic target, such as a gas jet or cell, to a driving field depends on the spatial phase relation between the radiation contributions of each atom involved in the HHG process, i.e. on phase matching. On the one hand, the coherent addition of these contributions is essential for efficient XUV generation. On the other hand, multiple factors, like the intensity-dependent phases of the excited dipoles, the macroscopic properties of the emitting medium as well as the driving beam space and time properties influence phase matching, making it a complex problem. A thorough treatment of HHG phase matching exceeds the scope of this thesis. Here, the discussion is restricted to an overview of the main factors determining phase matching.

Firstly, the atomic phase of the participating dipoles is intensity-dependent and thus, a function of space and time. Section 11.2 of [49] gives an introduction hereof. An important result is that two geometries are found for which good phase matching is achieved. One of them corresponds to collinear phase matching and results in a Gaussian-like emitted harmonic beam. The other one corresponds to noncollinear phase matching and yields an annular beam.

Secondly, for a Gaussian beam with the confocal parameter

$$b = \frac{2\pi w_0^2}{\lambda}, \quad (2.35)$$

where w_0 is the $1/e^2$ -intensity decay focus radius, the geometrical phase shift with respect to a plane wave (Gouy phase shift), depends on both the axial and radial coordinates, z and r , respectively. For a Gaussian beam with a focus at $z = 0$ and a wavefront curvature $R(z)$, the geometrical phase is given by the expression:

$$\phi_{\text{geom}}(r, z) = -\arctan\left(\frac{2z}{b}\right) + \frac{\pi r^2}{\lambda R(z)}. \quad (2.36)$$

The intensity-dependence of the atomic dipole moment together with the geometrical phase lead to a substantial dependence of the coherence properties of the generated harmonics on the position of the nonlinear medium with respect to the focus and on its geometry (see e.g. [50] for details). Assuming a homogenous transverse distribution of the nonlinear medium, the geometry of the interaction region is mainly described by its length l_{med} . If $l_{\text{med}} < b$ holds, the system is referred to as exhibiting *weak (or loose) focusing*. For $l_{\text{med}} > b$ the focusing is said to be *tight*. As a rule, optimum phase matching is reached employing loose focusing conditions [51–53]. The high quality of the transverse mode of an enhancement cavity favors phase matching.

Thirdly, the atomic density plays an essential role in phase matching since dispersion introduces a phase mismatch $\Delta k = k_q - qk_1$ between the phase k_q of the harmonic field of order q and the phase k_1 of the fundamental (polarization) field. In a dispersive medium, the phase lag between the nonlinear polarization introduced by the fundamental laser, oscillating with the frequency ω , at the frequency $q\omega$ and the free Gaussian beam propagating in the medium is [54]:

$$\Delta\phi(z) = (1 - q) \arctan\left(\frac{2z}{b}\right) + \Delta kz. \quad (2.37)$$

The values of Δk for a fundamental wavelength close to the one employed in our experiment are given in Table 1 in [55] for q between 3 and 33. Another important contribution to the phase mismatch is caused by the free electrons of the ionized gas, see also Table 1 in [55]. The influence of several gas target parameters on the harmonic yield is discussed in Sect. 12.2.2 in [49]. In general, an optimum interplay of the mechanisms contributing to phase matching can be found empirically, by adjusting the geometry, the position and the pressure of the gas nozzle.

2.2.2 Scaling Laws

For the design (dimensioning) of any HHG device, the knowledge of the scaling laws of this highly nonlinear process is imperative. However, as opposed to the cases of many other nonlinear processes, in the case of HHG in a gas, a simple and general scaling model, consisting of closed expressions of all relevant experimental parameters, which predict the spectrally resolved intensity of the generated high harmonics, still lacks.⁸ This is partly due to the complexity of the process and the multitude of involved parameters, but also due to the absence of HHG experiments enabling the validation of theoretical models over large parameter variation ranges. In particular, the strong intensity-dependence of the process makes the comparison between different experiments difficult. In this section we sum up the most relevant HHG scaling laws, which have been confirmed by all experiments so far.

For a better overview we first list the formulas linking the pulse energy E_{pulse} , the average power P_{av} , the repetition frequency (rate) f_{rep} , the pulse peak power P_{peak} , the pulse duration τ_{pulse} and the peak intensity I of a pulse with a Gaussian distribution in time and space:

$$E_{\text{pulse}} = \frac{P_{\text{av}}}{f_{\text{rep}}}, \quad (2.38)$$

$$P_{\text{peak}} = 0.94 \frac{E_{\text{pulse}}}{\tau_{\text{pulse}}}, \quad (2.39)$$

$$I = 2 \frac{P_{\text{peak}}}{A_{\text{focus}}} = 2 \frac{P_{\text{av}}}{f_{\text{rep}} \tau_{\text{pulse}} \pi w_0^2}. \quad (2.40)$$

Equations (2.31) and (2.34) imply for the cutoff frequency:

$$\hbar\omega_{\text{cutoff}} = 2.96 \cdot 10^{-13} I \lambda^2 + U_i. \quad (2.41)$$

Thus, higher harmonics are generated using a longer driving wavelength. In particular, among the most common mode-locked lasers, this constitutes an advantage of Yb-based systems (centered around 1,030 nm) over Ti:Sa systems (centered around 800 nm). Scaling the cutoff by means of the intensity can be done by (see Eq. (2.40)): (i) increasing the pulse peak power, meaning increasing P_{av} and/or decreasing τ_{pulse} and/or decreasing f_{rep} . The challenges related to these scaling measures from the point of view of enhancement cavities are discussed in Sect. 3.2.1. (ii) by tightening the focusing. However, this usually alters phase matching (see previous section). Moreover, increasing the intensity is limited by ionization. The probability for the latter increases with the intensity, leading to an eventual breakdown of HHG. For example, for Xe the optimum intensity for HHG lies in the range $\sim 10^{13} - 10^{14} \text{ W/cm}^2$.

⁸ First closed-form relations for the conversion efficiency under the assumption of a square-shaped pulse are provided in [56]. However, the focusing conditions and intensity-dependent phase matching are left out of this model.

Furthermore, the cutoff can be pushed towards higher frequencies by choosing lighter atoms with a larger ionization potential (see Table 2.1) since ω_{cutoff} depends on U_i via Eq. (2.34).

Concerning the conversion efficiency, a compendium of several HHG experiments is provided in Sect. 3.1.2 of [57]. A theoretical study [58] for driving lasers from the near-visible (800 nm) to the mid-infrared (2 μm), has recently predicted a dramatic conversion efficiency (CE) improvement for HHG driven by shorter wavelengths:

$$CE \propto \lambda^{-6} \dots \lambda^{-5}. \quad (2.42)$$

This scaling law has been experimentally verified in the range 800–1850 nm for Xe ($CE \propto \lambda^{-6.3 \pm 1.1}$) and Kr ($CE \propto \lambda^{-6.5 \pm 1.1}$) [59].

In weak focusing operation (i.e. for $l_{\text{med}} < b$), at constant driving intensity, the intensity of plateau harmonics I_{plateau} is observed to scale according to the following law (see Sect. 12.2.1 in [49, 60]):

$$I_{\text{plateau}} \propto b^3. \quad (2.43)$$

The transverse mode characteristics of the generated harmonics depend strongly on the fundamental beam shape, the intensity and the phase matching conditions. In practice, spatial coherence measurements can be performed (cf. e.g. Sect. 12.1.2 in [49]). For HHG with a focused Gaussian beam the following approximation of the divergence of the harmonics holds (cf. Sect. 4.2.1 in [40]):

$$\frac{\theta_{\text{perturb}}^q}{\theta_f} \propto \frac{1}{\sqrt{q}}, \quad (2.44)$$

$$\frac{\theta_{\text{plateau}}^q}{\theta_f} \propto \frac{1}{q}, \quad (2.45)$$

where q , θ_f , $\theta_{\text{perturb}}^q$ and $\theta_{\text{plateau}}^q$ denote the harmonic order, the divergence of the fundamental beam with $\theta_f = \lambda/(\pi w_0)$, the divergence of the q th harmonic in the perturbative regime (exponential decay, low harmonics) and the divergence of the q th plateau harmonic, respectively.

Like in the case of the $\lambda^{-(6 \dots 5)}$ and the b^3 -laws in Eqs. (2.42) and (2.43), respectively, formulating a scaling law for the harmonic yield with respect to the pulse duration τ_{pulse} requires a constant intensity. However, unlike in the aforementioned two cases, where the variables λ and b have a well-defined influence on the HHG process, the variation of τ_{pulse} and the subsequent adjustment of the intensity to a constant level, is not a well-defined operation. For instance, τ_{pulse} can be varied by modifying the amplitude spectrum of the pulse and/or the spectral phase. The latter influences the distances between the zeros of the electric field rather than their number, which determines the number of HHG events. Moreover, the *intensity envelope* of the pulse, which can exhibit a complex shape, influences the ionization of the gas

during the interaction with the pulse, implying saturation and dispersion processes. Thus, the terms *pulse duration* and *intensity* are too coarse to fully describe the multitude of possibilities related to these quantities. Despite this shortcoming several special cases can be found in the literature. In [49, 60] it is stated that the harmonic yield is proportional to τ_{pulse} . This however, follows from the assumption that the number of XUV photons generated at a given intensity is proportional to the pulse duration, which only holds in the absence of ionization.⁹ In [61] and [62] the pulse duration is reduced from 750 to 120 fs and from 200 to 25 fs, respectively, by removing an imposed chirp. In both cases the intensity is kept constant by decreasing the pulse energy along with its temporal compression. While the first publication reports no dependence of the harmonic distribution on the pulse duration, the second one states that shorter pulses lead to an increased conversion efficiency. Moreover, fewer oscillations per pulse, which in the transfer-limited case means shorter pulses, imply broader spectra of the individual harmonics.

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⁹ For shorter pulses, the saturation intensity is higher, allowing to drive HHG at higher intensities. In a certain sense, this contradicts the τ_{pulse} -proportionality of the harmonics yield.

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