Chapter 2
Theoretical Foundations of Femtosecond Filamentation

In the following chapter, the theoretical modeling of femtosecond filamentation is discussed. For a detailed understanding of this phenomenon, the dynamical equation governing the evolution of the laser electric field have to be identified. As only femtosecond filaments in gases are considered here, these are provided by Maxwell’s equations [1] in an isotropic, homogeneous, non magnetizable dielectric. Thermal effects can be neglected here as they do not show up on a femtosecond timescale. In addition, the propagation equations admit further simplification as the radiation emitted by modern laser sources exhibits a highly directional character. In the following, the positive z-direction is chosen as the propagation direction of the beam. The electric field is decomposed into plane waves with wave vector \( \vec{k} \). Then, the notion of directional beam propagation along \( z \) implies \( k_z > 0 \) and \( k_\perp/|\vec{k}| \ll 1 \), where \( k_\perp \) is the modulus of the transverse wave vector, i.e., \( k_\perp = \sqrt{k_x^2 + k_y^2} \). With these assumptions, it can be shown that with good accuracy, the Maxwell equations can be factorized [2, 3], yielding a first order partial differential equation in \( z \), also known as Forward Maxwell’s Equation (FME) [4]. The latter governs the evolution of the directional laser field. Compared to Maxwell’s equations, the FME allows for a greatly simplified numerical treatment and speeds up the calculations. Moreover, the FME allows the description of ultra-short, ultra-broadband laser radiation emitted by modern, mode-locked femtosecond laser sources. The latter emit laser pulses with durations <10 fs. For laser radiation with a spectrum centered around 800 nm, this duration corresponds to less than three oscillations of the optical carrier wave. While the propagation of narrow-band optical pulses in a Kerr medium can be adequately described by a Nonlinear Schrödinger Equation (NLSE) [5], the slowly varying envelope approximation (SVEA) fails for these few-cycle pulses. Nevertheless, assuming moderate restrictions on the pulse and the propagation medium, a nonlinear envelope equation (NEE) [6] can be derived. The NEE is a generalization of the NLSE of Ref. [5] and turned out to be a successful model describing the dynamics of few-cycle femtosecond pulses, reproducing experimental results [7]. In fact, historically, the NEE may be considered an ancestor of the more general FME.
A complete description of intense laser radiation propagating in a dielectric medium further requires an appropriate modeling of the polarization $\vec{P}$ due to the response of bound electrons induced by the laser field. As in filamentation intensities of the order of $10^{13}$ W/cm$^2$ are involved [8], the polarization is expected to depend on the electric field in a nonlinear manner. Moreover, the intensity levels achieved in filamentation experiments are sufficiently high to ionize the medium, resulting in the generation of a dilute plasma. This gives rise to a non-zero electron density $\rho$ and an electron current density $\vec{J}$ coupling to the electric field. However, the laser wavelength typically used for the generation of femtosecond filaments is 800 nm (the characteristic wavelength of a Ti:sapphire amplifier), corresponding to a photon energy of $\approx 1.55$ eV. In contrast, the ionization potential of the gases relevant for filamentation experiments varies between 10 and 25 eV. This suggests that ionization does not proceed via direct (single-photon) photoionization. Rather, ionization proceeds in a highly nonlinear manner, via, e.g., multiphoton or tunneling ionization [9, 10] which leads to an equally nonlinear dependence of $\rho$ and $\vec{J}$ on the laser electric field.

Finally, the aim is to identify those mechanisms leading to the observed long-range propagation [11–13] of femtosecond filaments as well as other characteristic properties as will be detailed below. To this aim, the envelope equation is analyzed in certain limiting cases in order to isolate the dominant effects contributing to the specific phenomenon under consideration.

2.1 The Forward Maxwell Equations

Maxwell’s equations governing the evolution of an electromagnetic field in a dielectric material may be expressed as a coupled set of vector-wave equations for the electric field $\vec{E}$, the dielectric displacement $\vec{D}$ and the current density $\vec{J}$ according to [1, 8, 14]

$$\vec{\nabla}(\vec{\nabla} \cdot \vec{E}) - \vec{\nabla}^2 \vec{E} = -\mu_0 \left( \frac{\partial^2 \vec{D}}{\partial t^2} + \frac{\partial \vec{J}}{\partial t} \right)$$

(2.1)

$$\vec{\nabla} \cdot \vec{D} = \rho.$$  

(2.2)

Here, $\vec{D} = \varepsilon_0 \vec{E} + \vec{P}$ is the dielectric displacement which accounts for the bound-charge density due to the polarization $\vec{P}$ induced by the laser electric field. The polarization corresponds to an ensemble average of the atomic or molecular dipole moments induced by the laser field. Throughout the thesis, the paraxial approximation is used, assuming that the laser beam may be Fourier decomposed into plane waves with wave-vectors $\vec{k}$ satisfying

$$k_\perp \ll |\vec{k}|,$$

(2.3)

such that the angle between $\vec{k}$ and the optical axis is sufficiently small. As discussed in the introductory remarks to this chapter, this is a reasonable assumption as laser
beams exhibit a highly directional character and low beam divergences. Moreover, the polarization is decomposed according to

\[ \vec{P} = \vec{P}^{(1)} + \vec{P}_{\text{NL}}, \]  

(2.4)

where the first term \( \vec{P}^{(1)} \) varies linearly and the second term varies nonlinearly with the electric field. Thus, \( \vec{P}^{(1)} \) describes classical, linear optical phenomena, while the nonlinear response \( \vec{P}_{\text{NL}} \) leads to nonlinear optical effects and induces self-interactions of the optical field.

For an isotropic, homogeneous medium, \( \vec{P}^{(1)} \) is collinear to the electric field. In the following, it is often useful to treat Eq. (2.2) in the frequency-domain representation. The frequency-domain analogue \( \hat{G}(\omega) \) is related to the function \( G(t) \) via the Fourier-transform \( \mathcal{F} \), for which the following convention is adopted throughout the thesis,

\[ \hat{G}(\omega) = \mathcal{F}[G](\omega) \equiv \frac{1}{2\pi} \int G(t)e^{i\omega t} dt \]  

(2.5)

\[ G(t) = \mathcal{F}^{-1}[\hat{G}](t) \equiv \int \hat{G}(\omega)e^{-i\omega t} d\omega. \]  

(2.6)

Assuming local response, the frequency domain representation of the linear polarization may be written as [14]

\[ \hat{\vec{P}}^{(1)}(\vec{r}, \omega) = \epsilon_0 \chi^{(1)}(\omega) \hat{\vec{E}}(\vec{r}, \omega). \]  

(2.7)

The first order susceptibility \( \chi^{(1)} \) is related to the frequency dependent refractive index \( n(\omega) \) and absorption coefficient \( \alpha(\omega) \) via \( (n(\omega) + i\alpha(\omega)c/2\omega)^2 = \epsilon(\omega) \), where the dielectric permittivity is given by the relation \( \epsilon(\omega) = 1 + \chi^{(1)}(\omega) \). It has been shown in Refs. [3, 8] that the approximation \( \vec{\nabla} \cdot \vec{E} \approx 0 \) is justified if, in addition to the paraxiality criterion Eq. (2.3), the nonlinear polarization satisfies the inequality

\[ \frac{|P_{\text{NL},i}|}{\epsilon_0 n^2(\omega)} \ll |E_i|, \]  

(2.8)

where \( k(\omega) := |\vec{k}| = n(\omega)\omega/c \) describes the modulus of the wave vector and \( i = x, y, z \) labels the vector components. Thus, exploiting the condition Eq. (2.8), the frequency domain analogue of Eq. (2.2) reads

\[ \frac{\partial^2 \hat{\vec{E}}}{\partial z^2} + k^2(\omega) \hat{\vec{E}} + \nabla^2_{\perp} \hat{\vec{E}} = -\mu_0 \omega^2 \left( P_{\text{NL}} + i\frac{\hat{J}}{\omega} \right), \]  

(2.9)

1 Nonlocally responding media play a crucial role for the physics of negative refraction [15]. In these media, the susceptibility \( \chi^{(1)}(\omega, \vec{k}) \) depends both on the frequency \( \omega \) and the wave vector \( \vec{k} \). The non-local analogue to Eq. (2.7) therefore involves a convolution in the spatial domain.
where the imaginary part of the linear susceptibility has been neglected, i.e. \( k^2(\omega) = \omega^2 \varepsilon(\omega)/c^2 \), with a \textbf{real-valued} dielectric function \( \varepsilon(\omega) \). This is a suitable approximation for modeling femtosecond pulse propagation in gases at standard conditions, which exhibit negligible linear losses [8]. The latter approximation will be used throughout this work, unless otherwise stated. It is furthermore assumed that the nonlinear response is isotropic and homogeneous. In combination with the paraxiality assumption \( \mathbf{\nabla} \cdot \mathbf{E} \approx 0 \), this leads to a decoupling of the vectorial components \( \mathbf{E} = (E_x, E_y, E_z) \) in the propagation Eq. (2.9). Assuming linear polarization of the initial laser field, \( \mathbf{E} = (E_x, 0, 0) \), the polarization is then preserved during beam propagation in the paraxial regime, and throughout the thesis, it is justified to switch to a scalar description, setting

\[
\hat{E} = E\mathbf{e}_x, \quad \hat{P}_{NL} = P_{NL}\mathbf{e}_x, \quad \hat{J} = J\mathbf{e}_x
\]

with orthogonal unit vectors \( \mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z \). However, it should be noted that for large nonparaxiality, the latter assumptions cannot be maintained, leading to a nonlinear coupling of differently polarized states, as has recently been demonstrated in Ref. [16].

Although the second order wave equation (2.9) provides a convenient simplification of the full model Eq. (2.2), both the paraxiality criterion and the condition Eq. (2.8) have not been fully exploited yet. In fact, as demonstrated in [2, 3, 17], the second order wave equation can be factorized to yield a first order differential equation in \( z \), a fact that greatly simplifies numerical beam propagation. A detailed derivation of this factorization procedure can be found in Refs. [2, 3]. Here, the method is outlined by means of the one-dimensional Helmholtz equation with an inhomogeneity \( h \),

\[
\frac{\partial^2 \hat{E}}{\partial z^2} + k^2 \hat{E} = \hat{h}
\]

where \( k = n(\omega)\omega/c \) and \( \hat{E}(z, \omega) \) denotes the frequency domain representation of the electric field \( E(z, t) \) in the time domain.

Fourier transform w.r.t \( z \), \( \hat{E}(z, \omega) \rightarrow \hat{E}_\beta(\beta, \omega) \), where \( \beta \) denotes the conjugate variable yields the equation

\[
\hat{E}_\beta = \frac{\hat{h}_\beta}{k^2 - \beta^2},
\]

where it was used that \( \partial / \partial z = -i\beta \) and the equation was formally solved for \( \hat{E}_\beta \). The rather formal manipulations leading to Eq. (2.12) can be substantiated by noting that the Fourier transform w.r.t. \( \beta \),

\[
G_\omega(z, z') = \int d\beta \frac{e^{-i\beta(z-z')}}{k^2(\omega) - \beta^2},
\]

(2.13)
corresponds to the Green’s function $G(z, z')$ of the one-dimensional Helmholtz equation. This allows the construction of a solution to the inhomogeneous Eq. (2.11) according to

$$\hat{E}(z, \omega) = \int dz' G_\omega(z, z') \hat{h}(z', \omega).$$  \hspace{1cm} (2.14)

However, note that appropriate boundary conditions [2] have to be supplied to solve the problem (2.11) using Eqs. (2.12) and (2.13).

Factorization of the Helmholtz equation is achieved by noting that Eq. (2.12) can be decomposed according to [3]

$$\hat{E}_\beta \equiv \hat{h}_\beta 2k - k^2 \hat{E}_\beta + \hat{E}_\beta, \hspace{1cm} (2.15)$$

where forward and backward propagating electric field components $\hat{E}_\beta^\pm$ were defined according to

$$\hat{E}_\beta^+ = -\hat{h}_\beta 2k \frac{1}{\beta + k}, \quad \hat{E}_\beta^- = \hat{h}_\beta 2k \frac{1}{\beta - k}.$$  \hspace{1cm} (2.16)

The Helmholtz equation in the $z$-domain is therefore equivalent to the set of first-order differential equations

$$(\partial_z + ik) \hat{E}^+ = \hat{h} 2k, \quad (\partial_z - ik) \hat{E}^- = \hat{h} 2k.$$  \hspace{1cm} (2.17)

The wave fields $E^\pm$ correspond to waveforms traveling into the positive and negative $z$ directions. In the linear regime, they evolve independently. The inhomogeneous three-dimensional Helmholtz Equation (2.9) allows a completely analogous factorization, with the subtle difference that the inhomogeneity $h$ may depend on the field $E$ to model pulse propagation in the nonlinear regime. In this case, the factorized Helmholtz equations for the forward- and backward propagating field components are nonlinearly coupled. However, it is shown in Ref. [3] that for an initial field $E = E^+ + E^-$ with a dominant forward-propagating field component $E^+$, the backward-propagating component $E^-$ stays small along $z$-propagation and can be neglected, as long as the paraxiality criterion $k_\perp/|\vec{k}| \ll 1$ and the condition (2.8) are fulfilled.

As shown in Sect. 2.3, these criteria are usually satisfied in filamentary propagation, which justifies the assumption $\hat{E} = \hat{E}^+$. The factorization procedure thus yields a first order partial differential equation for the forward-propagating field,

$$\frac{\partial \hat{E}}{\partial z} = \frac{i}{2k(\omega)} \nabla_\perp^2 \hat{E} + ik(\omega) \hat{E} + \frac{i \mu_0 \omega^2}{2k(\omega)} \left(P_{NL} + \frac{\hat{J}}{\omega}\right).$$  \hspace{1cm} (2.18)

This equation has originally been used in Ref. [4] as a starting point to analyze supercontinuum generation in photonic crystal fibers. While Eq. (2.18) describes
freely propagating pulses in a nonlinear medium, a rigorous derivation of an equation analogous to the FME, describing forward-propagating pulses in a guided geometry, has recently been given in Refs. [18, 19].

### 2.2 The Nonlinear Optical Response

This section is devoted to the nonlinear response of the material to the intense laser field. The basic assumption of perturbative nonlinear optics is that the nonlinear polarization $P_{NL}$ of an isotropic medium can be decomposed as

$$P_{NL} = P^{(3)} + P^{(5)} + P^{(7)} + \cdots .$$

(2.19)

As only isotropic, centrosymmetric media are examined in the following, all even-order contributions $P^{(2k)}$ vanish identically [20]. Demanding that the nonlinear response respects time-translational invariance of the dynamical equation leads to the following expression for the $n$-th order contribution in the time domain [22].

$$P^{(n)}(\vec{r}, t) = \epsilon_0 \int \int \cdots \int \int E(\vec{r}, t - \tau_1) E(\vec{r}, t - \tau_2) \cdots E_n(\vec{r}, t - \tau_n) \times R^{(n)}(\tau_1, \tau_2, ..., \tau_n)$$

(2.20)

In the frequency domain, this translates into

$$P^{(n)}(\vec{r}, \omega) = \epsilon_0 \int \cdots \int \chi^{(n)}(-\omega_\sigma; \omega_1, ..., \omega_n) E(\vec{r}, \omega_1) \cdots E(\vec{r}, \omega_n) \delta(\omega - \omega_\sigma) d\omega_1 \cdots d\omega_n$$

(2.21)

where $\omega_\sigma = \omega_1 + \omega_2 + \cdots + \omega_n$, and only homogeneous media are considered for which the response kernel $R^{(n)}$ and the susceptibilities $\chi^{(n)}$ are independent of position. The $n$-th-order contribution to the nonlinear polarization is frequently considered as resulting from an $n + 1$-photon process interacting with bound electronic states. From this point of view, the delta function in the integrand ensures conservation of photon energy, $\hbar \omega = \hbar \omega_1 + \cdots + \hbar \omega_n$.

---

2 As in the case of the linear polarization, spatial dispersion modeled by a wave-vector dependent nonlinear susceptibility $\chi^{(n)}(\omega_1, \cdots, \omega_n, \vec{k}_1, \cdots, \vec{k}_n)$ was disregarded. Spatially dispersive nonlinearities involve a nonlocal optical response and can arise from thermal effects or may occur in dipolar Bose-Einstein condensates [21].
2.2 The Nonlinear Optical Response

2.2.1 Third-Order Response to a Monochromatic Wave

In the following, the impact of the first non-vanishing order \( P^{(3)} \) on a monochromatic plane wave of frequency \( \omega_0 \) and amplitude \( E_0 \) propagating into the positive \( z \)-direction with wave-vector \( k_0 \equiv k(\omega_0) = n(\omega_0)\omega_0/c \),

\[
E(\vec{r}, t) = E_0 \cos(\omega_0 t + kz + \varphi)
\]  

(2.22)

will be discussed. With Euler’s formula for the cosine, this may be decomposed according to

\[
E(\vec{r}, t) = \frac{1}{2} \left( A e^{i\omega_0 t + ik_0 z} + A^* e^{-i\omega_0 t - ik_0 z} \right),
\]  

(2.23)

where

\[
A = E_0 e^{i\varphi}.
\]  

(2.24)

With the help of Eq. (2.23), the frequency-domain representation Eq. (2.21) of the third-order nonlinear polarization induced by a monochromatic plane wave may be written as [20]

\[
P^{(3)}(\omega) = \frac{3}{8} \epsilon_0 \chi^{(3)}(-\omega_0; \omega_0, \omega_0, -\omega_0) |A|^2 A \delta(\omega - \omega_0) e^{ikz}
\]

\[
+ \frac{3}{8} \epsilon_0 \chi^{(3)}(\omega_0; -\omega_0, -\omega_0, \omega_0) |A|^2 A^* \delta(\omega + \omega_0) e^{-ikz}
\]

\[
+ \frac{1}{8} \epsilon_0 \chi^{(3)}(-3\omega_0; \omega_0, \omega_0, \omega_0) A^3 \delta(\omega - 3\omega_0) e^{i3kz}
\]

\[
+ \frac{1}{8} \epsilon_0 \chi^{(3)}(3\omega_0; -\omega_0, -\omega_0, -\omega_0) A^3 \delta(\omega + 3\omega_0) e^{-i3kz}.
\]  

(2.25)

It follows that the polarization \( P^{(3)} \) oscillates at frequencies \( \pm 3\omega_0 \) and \( \pm \omega_0 \). While the latter give rise to a nonlinear refractive index change as will be detailed below, the former correspond to the generation of a third-harmonic wave copropagating with the fundamental wave, a phenomenon known as third-harmonic generation (THG). However, the expression (2.25) shows that there exists a mismatch between the wave-vector \( 3k(\omega_0) \) of the polarization and the wave-vector \( k(3\omega_0) \) of the radiated harmonic wave, \( \Delta k = k(3\omega_0) - 3k(\omega_0) \), whenever the medium exhibits nontrivial dispersion \( n(3\omega_0) \neq n(\omega_0) \)[20]. In general, this will lead to destructive interference of the third harmonic waves generated at different positions unless suitable phase-matching techniques [20] are applied which ensure vanishing of the wave-vector mismatch \( \Delta k \). Harmonic generation is therefore disregarded in the following, focusing the attention to self-induced refractive index changes.
2.2.2 Third-Order Response to an Optical Pulse

While Eq. (2.25) was derived for a monochromatic plane wave, filamentation is only observed for sufficiently high peak powers of the laser pulse of the order of 100 GW, which is impossible to achieve with monochromatic light. Instead, only pulsed laser sources generating ultrashort pulses with durations of the order of some ten femtoseconds are capable of providing the required peak optical powers. Equation (2.25) therefore has to be generalized for ultrashort optical pulses. The subsequent discussion is greatly simplified by introducing so-called complex-valued analytic signals. With the decomposition (2.23), the real-valued monochromatic wave is seen to consist of positive and negative frequency components. This can be generalized for arbitrary time-dependence of the electric field, using that the Fourier transform of any real-valued function \( F(t) \) satisfies \( \hat{F}(-\omega) = \hat{F}^*(\omega) \). This reveals that the information contained in the negative frequency components of \( F \) can be considered redundant, and instead of the real-valued electric field \( E \), the so called analytic signal \( E_A \) is considered in the following. This is composed of the positive frequency components of \( E \) according to

\[
E_A(\vec{r}, t) = 2 \int_{0}^{\infty} d\omega \hat{E}(\vec{r}, \omega) e^{-i\omega t}. \tag{2.26}
\]

From this, the electric field may easily be reconstructed according to

\[
E(\vec{r}, t) = \frac{1}{2} (E_A(\vec{r}, t) + E_A^*(\vec{r}, t)). \tag{2.27}
\]

It is moreover useful to factorize the analytic signal \( E_A \) into an envelope \( A \) and an exponential oscillating at the carrier-frequency \( \omega_0 \) of the laser field,

\[
E_A(\vec{r}, t) = A(\vec{r}, t) e^{-i\omega_0 t}, \tag{2.28}
\]

where the carrier frequency \( \omega_0 \) denotes the mean frequency \[23]\]

\[
\omega_0 = \frac{\int_{-\infty}^{\infty} d\omega |\hat{E}|^2 \omega}{\int_{-\infty}^{\infty} d\omega |\hat{E}|^2}. \tag{2.29}
\]

In the frequency domain, the definition (2.28) corresponds to the identity \( \hat{A}(\vec{r}, \omega) = \hat{E}_A(\vec{r}, \omega + \omega_0) \), which shows that \( A \) has zero mean frequency, corresponding to the removal of the fast carrier oscillations at \( \omega_0 \), leaving only a pulse envelope. In what follows, the generalization of Eq. (2.25) for short laser pulses shall be discussed. However, experimental or theoretical data describing the dispersion of \( \chi^{(3)} \) over a large frequency range often vary by orders of magnitude \[24\]. More reliable data is
available from measurements or calculations of $\chi^{(3)}$ at a single frequency. Consequently, it is henceforth assumed that the spectral bandwidth of the pulse is small with respect to the frequency scale on which $\chi^{(3)}$ shows notable variation. Then, it is possible to show [8] that the third-order polarization induced by the electromagnetic pulse is given by

$$P^{(3)}(\vec{r}, t) = \frac{3}{8} \epsilon_0 c \chi^{(3)}(-\omega_0; \omega_0, \omega_0, -\omega_0)|A(\vec{r}, t)|^2 A(\vec{r}, t)e^{-i\omega_0 t} + c.c. + \frac{1}{8} \epsilon_0 c \chi^{(3)}(-3\omega_0; \omega_0, \omega_0, \omega_0)A^3(\vec{r}, t)e^{-i3\omega_0 t} + c.c. \quad (2.30)$$

Neglecting again the THG term oscillating at $3\omega_0$, the third order polarization gives rise to an intensity dependent change of the refractive index. This is due to the fact that sufficiently strong electromagnetic fields can distort the electronic distribution within in the medium, which gives rise to a modified refractive index. This effect is also referred to as the all-optical Kerr effect [25] and should not be confused with the electro-optic (DC) Kerr effect [26], where a static electric field induces birefringence in the material. In order to further evaluate the third order contribution to the intensity dependent refractive index (IDRI), it is useful to introduce the optical intensity $I$. As the energy density of an electric field is proportional to the square of the electric field strength, it follows that the optical intensity is given by [23]

$$I(\vec{r}, t) = \epsilon_0 c n_0 \frac{1}{T} \int_{t-T/2}^{t+T/2} E^2(\vec{r}, t')dt' \quad (2.31)$$

where $n_0 \equiv n(\omega_0)$ denotes the refractive index at the center frequency, and the average over one optical cycle of duration $T = 2\pi/\omega_0$ was taken. Demanding that the envelope $A$ defined in Eq. (2.28) varies slowly compared to the carrier oscillation at $\omega_0$, it follows that the above relation for the cycle-averaged intensity can be evaluated to give

$$I = \frac{1}{2} n_0 \epsilon_0 c |A|^2 \quad (2.32)$$

Including only the third-order nonlinear polarization, it can be deduced from Eq. (2.30) that the IDRI due to the all-optical Kerr effect is given by

$$n(I) = n_0 + n_2 I \quad (2.33)$$

where $n_2$ denotes the second order nonlinear refractive index which is given by

$$n_2 = \frac{3}{4n_0^2 \epsilon_0 c} \chi^{(3)} \quad (2.34)$$
Note that for the latter derivation, both linear and nonlinear absorption were disregarded, which allows to impose $\text{Im} \chi^{(1)} = \text{Im} \chi^{(3)} = 0$. In fact, this approximation is frequently justified in the context of femtosecond filamentation [8, 12]. A more detailed discussion of higher-order nonlinear refraction and absorption coefficients and their relation to the nonlinear susceptibilities $\chi^{(n)}$ is provided in Sect. 2.3. Indeed, it is one of the main conclusions both, of recent experimental results [27–29], and of the theoretical investigations in Chap. 4, that higher-order nonlinearities $\chi^{(n)}$ for $n > 3$ actually play a greater role than previously supposed.

2.2.3 Plasma Response

Besides the all-optical Kerr effect, an important contribution to the nonlinear refractive index is given by free carriers. In fact, the intensities achieved within femtosecond filaments are sufficiently high to trigger photoionization processes. The femtosecond laser pulse thus propagates in a self-generated plasma. The current density $J$ taking into account the generation of free carrier by photoionization can be decomposed according to

$$J = J_{\text{FC}} + J_{\text{PI}},$$

where $J_{\text{FC}}$ is the current density of free carriers subject to the electric field $E$, while $J_{\text{PI}}$ accounts for losses due to photoionization. Both quantities couple to the FME Eq. (2.18). The dynamics of the free carriers is treated in terms of the Drude model [30, 31] according to

$$\frac{\partial J_{\text{FC}}}{\partial t} + \frac{J_{\text{FC}}}{\tau_c} = \frac{q_e^2 \rho}{m_e} E,$$

Here, $q_e$ and $m_e$ denote electron charge and mass, respectively, $\rho$ denotes the number of free carriers per unit volume and $\tau_c$ represents the mean time between collision of free carriers. In the frequency domain, Eq. (2.36) can be formally solved for the Fourier transform $\hat{J}_{\text{FC}}$, and it is found that the current of free carriers (2.18) is given by [8]

$$-\frac{\mu_0 \omega}{2k(\omega)} \hat{J}_{\text{FC}} = \frac{1}{2k(\omega)} \left( -\frac{\omega n_0 \sigma(\omega)}{c} - i \frac{\omega^2}{c^2 \rho_c (1 + \nu_e^2/\omega^2)} \right) \hat{\rho} E$$

where $n_0 = n(\omega_0)$ is the refractive index at the carrier frequency, $\nu_e = 1/\tau_c$ and $\rho_c = \omega_0^2 m_e e_0/q_e^2$ is the critical density of free carriers for which the plasma becomes opaque for a laser beam of carrier frequency $\omega_0$. The cross-section for collision of free carriers is given by

$$\sigma(\omega) = \frac{q_e^2}{m_e e_0 n_0 c \nu_e (1 + \omega^2/\nu_e^2)}.$$
In Eq. (2.37), the loss term involving the cross-section $\sigma(\omega)$ accounts for collisional ionization by free carriers accelerated in the laser field. As this process consumes electromagnetic energy, it is frequently referred to as inverse Bremsstrahlung. In contrast, the term involving the purely imaginary prefactor of $\hat{\rho}E$ corresponds to the change of the refractive index due to the plasma and will be discussed below.

In addition, direct photoionization of neutral atoms takes energy from the laser field. This requires the introduction of the loss current [8],

$$J_{\text{PI}} = \frac{k_0}{\omega_0 \mu_0} \frac{U_i w(I)}{I} (\rho_0 - \rho) E. \quad (2.39)$$

This quantity depends on the ionization potential $U_i$ of the gas species and on the neutral density $\rho_0$. Furthermore, it depends on the ionization rate $w(I)$. A theoretical derivation of the ionization rate of atoms or molecules subject to intense laser fields has been performed by several independent researchers [10, 32–36]. Throughout this work, the results of Perelomov, Popov and Terent’ev (PPT) [32, 35] are applied. A deeper discussion of the PPT model is presented in Chap. 4 of this thesis. As the ionization depends highly nonlinear on the intensity, it is justified to assume that only frequency components of the pulse close to the carrier frequency $\omega_0$ contribute to ionization processes. Therefore, for the collisional cross section the replacement $\sigma(\omega) \rightarrow \sigma(\omega_0)$ is performed throughout. It then follows that the density $\rho$ of the self-generated plasma satisfies the rate equation

$$\frac{\partial \rho}{\partial t} = w(I)(\rho_0 - \rho) + \frac{\sigma(\omega_0)}{U_i} \rho I. \quad (2.40)$$

While typical timescales relevant in filamentation are of the order of $10^{-13} - 10^{-14}$ s, recombination of ions and electrons takes place on a nanosecond timescale. This justifies to neglect recombination effects in Eq. (2.40). The first term on the r.h.s. of Eq. (2.40) accounts for photoionization, while the second term models the contribution of collisional ionization to the electron density.

### 2.3 Contributions to the Nonlinear Refractive Index

#### 2.3.1 Plasma Contributions

For the case of a monochromatic plane wave of frequency $\omega = \omega_0$, leading to $k(\omega) = k(\omega_0) = k_0$ and $n(\omega) = n(\omega_0) = n_0$, the FME Eq. (2.18) reduces to

$$\frac{\partial \hat{E}}{\partial z} = -i \frac{\omega_0}{c} (n_0 + \Delta n_p) \hat{E}, \quad (2.41)$$
where additionally, losses due to collisional ionization, i.e., $\nu_e \rightarrow 0$, and the nonlinear polarization were neglected. This shows that for $\nu_e = 0$, the contribution of the free carriers to the refractive index is given by $\Delta n_p = -\rho/2n_0^2\rho_c$. In contrast, using the Drude model of a collisionless plasma and the wave Eq. (2.9), it turns out that the presence of plasma in a medium with neutral refractive index $n_0$ lowers the refractive index according to [37]

$$n = \sqrt{n_0^2 - \frac{\omega_p^2}{\omega^2}}, \quad (2.42)$$

where $\omega_p = \sqrt{\rho q_e^2/m_e\epsilon_0} = \omega_0\sqrt{\rho/\rho_c}$ is the plasma frequency. The obvious discrepancy arises from the approximations introduced with the FME: the term accounting for the linear polarization $\sim k^2(\omega)\hat{E}$ exhibits a quadratic dependence on the wave vector $k$, while the current density $J_{FC}$ enters linearly. In contrast, due to the factorization procedure, the linear polarization gives rise to a term $\sim k(\omega)\hat{E}$ on the r.h.s. of Eq. (2.9), while the term containing the current is not affected by the factorization and enters linearly. However, for $\rho \ll \rho_c$, Eq. (2.42) may be approximated according to

$$n = n_0 - \frac{\rho}{2n_0^2\rho_c}, \quad (2.43)$$

which corresponds to the plasma induced refractive index change derived from the FME. Thus, the inequality $\rho \ll \rho_c$ provides an additional criterion for the validity of the FME. As an example of practical relevance, filamentary propagation of a pulsed femtosecond laser beam emitted by a Ti:sapphire amplifier (with a center wavelength of 800 nm) is considered. Assuming that the pulse is propagating in a gaseous medium of atmospheric pressure, the ratio of ionized particles is of the order of $10^{-3}$ [38], i.e., $\rho \approx 3 \times 10^{16} \text{cm}^{-3}$, while the critical plasma density for the given wavelength is $\rho_c \approx 2 \times 10^{21} \text{cm}^{-3}$. Under these assumptions the plasma-induced refractive index change in femtosecond filaments is therefore of the order of $\rho/\rho_c \approx 10^{-5}$, which justifies the approximations introduced with the FME Eq. (2.18).

### 2.3.2 Contributions Due to the All-Optical Kerr Effect

In linear optics, the refractive index $n_0$ and absorption coefficient $\alpha_0$ are related to the complex dielectric permittivity $\epsilon$ according to

$$(n_0 + i\alpha_0 c/2\omega)^2 = \epsilon. \quad (2.44)$$

Using

$$\hat{D} \equiv \epsilon_0\epsilon\hat{E} = \epsilon_0\hat{E} + \hat{E}^{(1)} \quad (2.45)$$
satisfied by the dielectric displacement and Eq. (2.7) for the linear polarization $\hat{P}^{(1)}$ it follows that $\epsilon(\omega) = 1 + \chi^{(1)}(\omega)$. This consideration can be generalized to the case of nonlinear optics if it is assumed that the spectral bandwidth of the optical pulse $E$ is small compared to the frequency scale on which the nonlinear susceptibilities $\chi^{(n)}$ show notable dispersion. In analogy to the reasoning that led to Eq. (2.30) for the third-order susceptibility $\chi^{(3)}$, an envelope description (Eq. 2.28) is introduced to identify the self-refraction terms contributing to the nonlinear polarization $P^{(n)}$. This yields an intensity dependent dielectric permittivity [39]

$$\epsilon(I) = 1 + \chi^{(1)}(\omega_0) + \sum_{k \geq 1} C^{(k)}(\omega_0) I^k \chi^{(2k+1)}|A|^2,$$

(2.46)

where the intensity $I$ is related to the envelope $A$ according to Eq. 2.32. The factor $C^{(k)}$ follows from combinatorial considerations [40] and is given by

$$C^{(k)} = \frac{(2k + 1)!}{22^k k!(k + 1)!},$$

(2.47)

and $\chi^{(n)}(\omega_0)$ denotes the $n$th-order nonlinear susceptibility associated to self-refraction, e.g., for the third order polarization, $\chi^{(3)}(\omega_0) = \chi^{(3)}(-\omega_0, \omega_0, \omega_0, -\omega_0)$, while $\chi^{(1)}(\omega_0)$ denotes the linear susceptibility at frequency $\omega_0$. From Eq. (2.46), a nonlinear refractive index $n(I)$ and a nonlinear absorption coefficient $\alpha(I)$ can be defined by generalizing Eq. (2.44) according to

$$(n(I) + i \alpha(I)c/2\omega)^2 = \epsilon(I).$$

(2.48)

Compact approximate expressions for $n(I)$ and $\alpha(I)$ can be derived if it is assumed that the nonlinear refraction and absorption changes $\Delta n(I) = n(I) - n_0$ and $\Delta \alpha(I) = \alpha(I) - \alpha_0$ are sufficiently small such that only first order contributions of these quantities have to be considered. In addition, it is assumed that the linear absorption coefficient $\alpha_0$ satisfies $\alpha c/\omega \ll n_0$ [22], which leads to the following expressions for the nonlinear refractive index and absorption coefficient,

$$n(I) = n_0 + \sum_{k \geq 1} n_{2k} I^k$$

$$\alpha(I) = \alpha_0 + \sum_{K \geq 2} \beta_K I^{K-1}.$$

(2.49)

The coefficients $n_{2k}$ and $\beta_K$ are related to the real and imaginary part of the nonlinear susceptibilities $\chi^{(2k+1)}$ pursuant to

$$n_{2k} = \frac{2^{k-1} C^{(k)}}{n_0(n_0c)^k} \text{Re}\chi^{2k+1},$$

(2.50)
\[ \beta_K = \frac{\omega_0}{c} \frac{2^{K-1} C^{(K-1)}}{n_0(n_0 \epsilon_0 c)^{K-1}} \Im \chi^{2K-1}. \] (2.51)

It is interesting to note that the approximations involved in defining a nonlinear refractive index are closely related to the approximation (2.8) made during the derivation of the FME. In fact, in terms of refractive index changes, the condition on \( P_{NL} \) translates itself into \( \Delta n(I) \ll n_0 \).

In Chap. 4 it is shown that the nonlinearly induced refractive index changes \( \Delta n \) are small for the noble gases helium, neon, argon, krypton and xenon, which are the most commonly used media in experimental femtosecond filamentation. For these gases, it can be shown that \( \Delta n \) varies between \( \sim 10^{-5} \) and \( \sim 10^{-7} \) for intensities up to 40 TW/cm\(^2\) (xenon) and 300 TW/cm\(^2\) (helium), respectively. As the error introduced by approximation (2.49) is of the order \( \Delta n^2 \), the use of the FME is clearly justified.

### 2.4 An Envelope Equation for Few-Cycle Optical Pulses

A further simplification of the FME may be obtained by imposing certain restrictions on the envelope \( A \). Besides assuming that the envelope varies slowly in time, it has to be imposed that the envelope varies slowly in the spatial coordinate \( z \). Thus, for the following, besides subtracting the carrier oscillations at \( \omega_0 \) in time, a subtraction of the spatial oscillations along the propagation direction \( z \) is necessary. These oscillations are governed by the \( z \)-component \( k_z \) of the wave-vector. However, assuming paraxial propagation, it is found that \( k_\perp / k \ll 1 \) which is equivalent to \( k_z \approx k_0 \). The electric field is then rewritten in terms of amplitudes that are slowly varying both in time and space pursuant to

\[ E(\vec{r}, t) = \sqrt{c_1} \left( \mathcal{E}(\vec{r}, t)e^{ik_0 z - i\omega_0 t} + \mathcal{E}^*(\vec{r}, t)e^{-ik_0 z + i\omega_0 t} \right). \] (2.52)

The normalization factor \( c_1 = \mu_0 / (n_0^2 \epsilon_0) \) is chosen such that \( I = |\mathcal{E}|^2 \). The envelopes \( \mathcal{E} \) and \( \mathcal{A} \) are related by \( \mathcal{E} = \mathcal{A} \exp(-ik_0 z) / 2 \sqrt{c_1} \). The requirements leading to an envelope \( \mathcal{E} \) varying slowly both in \( t \) and \( z \) then read

\[ \left| \frac{\partial}{\partial z} \mathcal{E} \right| \ll k_0 |\mathcal{E}|, \] (2.53)

and

\[ \left| \frac{\partial}{\partial t} \mathcal{E} \right| \ll \omega_0 |\mathcal{E}|. \] (2.54)

These restrictions provide the slowly varying envelope approximation (SVEA) [41]. With these conditions, a simple first order PDE in \( z \) for the envelope \( \mathcal{E} \) was obtained [5], which, neglecting plasma response and nonlinearities higher than third
order, corresponds to the Nonlinear Schrödinger Equation, see Appendix A for a brief mathematical introduction. This equation has successfully been applied to explain various phenomena during the early days of nonlinear optics. However, the latter equation fails to correctly describe ultra-broadband pulses as they arise for example in femtosecond filaments. This is due to the fact that for ultra-broadband pulses, the slowly varying envelope ceases to be a meaningful concept, especially for pulses consisting only of a few-cycles of the optical carrier field. However, a generalized envelope equation capable of describing the propagation of few-cycle pulses can be derived from the FME, which yields [6]

\[
\frac{\partial_z \mathcal{E}}{2k_0} = i \frac{T^{-1}}{\Delta_2} \mathcal{E} + i \mathcal{D} \mathcal{E} + i \frac{\omega_0}{c} n_2 T |\mathcal{E}|^2 \mathcal{E} - i \frac{k_0}{2\rho_c} T^{-1} \frac{\partial (\rho n_t - \rho)}{\partial t} \mathcal{E},
\]

\[
- \frac{\sigma}{2} \rho \mathcal{E} - \frac{U_i w(I)(\rho n_t - \rho)}{2I} \mathcal{E},
\]

\[
\frac{\partial_t \rho}{\partial_t} = w(I)(\rho n_t - \rho) + \frac{\sigma}{U_i} \rho I.
\]

In the above equation also referred to as the nonlinear envelope equation (NEE), only the third-order nonlinearity \( \chi^{(3)} \) was taken into account. Furthermore, a transformation of variables \( t \rightarrow t - z/v_g(\omega_0) \) to a frame comoving with the group velocity \( v_g(\omega) = (dk(\omega)/d\omega)^{-1} \) of the laser pulse was performed, and it was used that \( v_g \approx c \) for gaseous media at standard conditions. The operator \( T \) ensures validity of the model in the few-cycle domain and is given by

\[
T = 1 + \frac{i}{\omega_0} \partial_t.
\]

while the operator \( \mathcal{D} \) is given, in the frequency domain, by

\[
\mathcal{D}(\omega) = k(\omega) - k_0 - (\omega - \omega_0) \frac{\partial k}{\partial \omega} \bigg|_{\omega = \omega_0}
\]

A Taylor expansion of this expression followed by a Fourier transform, yields the following expression for the operator \( \mathcal{D} \) in the time domain,

\[
\mathcal{D} = \frac{1}{2!} \beta_2 \frac{\partial^2}{\partial t^2} + \frac{1}{3!} \beta_3 \frac{\partial^3}{\partial t^3} + \cdots
\]

where \( \beta_n = d^n k/d\omega^n \), evaluated at \( \omega = \omega_0 \), and it was used that the Fourier transform \( \mathcal{F} \) satisfies

\[
\mathcal{F}^{-1}(\omega G(\omega)) = i \frac{d}{dt} G(t)
\]

The operator \( \mathcal{D} \) describes the dispersion of the temporal pulse profile of the pulse due to the fact that different frequency components of the pulse propagate with
different velocities. For a narrow-band pulse, it often suffices to employ the power series representation Eq. (2.59) up to some finite order. However, for ultra-broadband pulses, it is more appropriate to evaluate $D(\omega)$ by using Sellmeier type equations for $n(\omega)$ [41, 42]. Alternatively, in the context of fiber optics, a recent approach involves approximation of $D(\omega)$ using rational functions [43]. The frequency domain representation $\hat{T}$ of $T$ reads

\[ \hat{T} = 1 + \frac{\omega}{\omega_0} \]  

(2.61)

This operator emerges by setting $k(\omega) \approx n_0 \omega / c$ on the r.h.s. of the FME (2.18), where it is assumed that the carrier wavelength is far from a medium resonance such that $n(\omega)$ exhibits weak dispersion. Subsequently, in the $\omega$ dependent prefactors of $\hat{E}$, $\hat{P}_{NL}$ and $\hat{J}$, the identity $\omega = \omega_0 (1 + (\omega - \omega_0) / \omega_0)$ is employed. The operator $T$ then emerges by noting that $\omega - \omega_0 \to i \partial_t$, where $\partial_t$ is restricted to act on the pulse envelope $E$. With these approximations, the envelope Eq. (2.55) is obtained after performing the Fourier transform into the time domain. It suitably models the propagation of few-cycle pulses given that the electric field $E$ satisfies

\[ \left| \frac{\partial E}{\partial z} \right| \ll k_0 |E|, \]  

(2.62)

This holds when the electric field evolves slowly during $z$-propagation. Therefore, the approximations leading to the NEE are referred to as the Slowly Evolving Wave Approximation (SEWA). Note that the SVEA corresponds to setting $T = 1$ in the NEE (2.55). In contrast to the SVEA, the SEWA does not imply a limitation of the pulse duration and can be used to model the propagation of few-cycle pulses in media subject to the additional limitation

\[ \left| k_0 - \frac{\omega_0}{v_g(\omega_0)} \right| / k_0 \ll 1, \]  

(2.63)

demanding that the group velocity differs from the phase velocity only marginally.

Historically, the generalized envelope equation was derived by Brabec and Krausz prior to the FME. However, as shown above, the envelope Eq. (2.55) can be derived from the more general FME in a rather straightforward manner. When wave-mixing phenomena like THG or sum frequency generation in filaments are investigated, the FME should clearly be preferred against the envelope description [44]. Nevertheless, only self-refraction effects are considered throughout this thesis, as the radiation due to THG is typically poorly phase-matched in femtosecond filaments and can be neglected. Further on, like the FME, the NEE provides a valid model for propagation of few-cycle pulses as shown in the above-cited references.

Therefore, the Eqs. (2.55) and (2.66) suitably describe femtosecond filamentation for the medium and input pulse parameters considered here.
2.4 An Envelope Equation for Few-Cycle Optical Pulses

2.4.1 Reduction to the Cylindrically Symmetric Case

It turns out that the complexity of solving Eq. (2.55) numerically can be strongly reduced by imposing an additional symmetry constraint, i.e. cylindrical symmetry along the propagation direction \( z \). As discussed in Sect. 2.5, the evolution Eq. (2.55) is subject to an azimuthal modulation instability which amplifies small amplitude, radially asymmetric perturbations on a cylindrically symmetric beam, leading to a loss of cylindrical symmetry and eventually a multiple spatial break-up of the beam also known as multifilamentation. However, it follows from the experimental observations of Ref. [45] that the radial symmetry of the input beam is preserved during filamentary propagation for input powers not exceeding roughly \( 5 - 6 P_{\text{cr}} \) [45], where \( P_{\text{cr}} \) is the critical power for self-focusing, as discussed in detail in Sect. 2.5. In the latter experimental work, 45 fs, 5 mJ-pulses emitted by a regenerative Ti:sapphire amplifier were focused into a 1.5 m long cylindrical gas cell filled with argon. For the chosen input beam parameters, the above constraint on the input power translates itself into a constraint for the pressure within the argon gas cell to values below 60 kPa, thereby limiting the nonlinearity \( n_2 \). Alternatively, the energy of the input pulse can be diminished using an adjustable diaphragm. Indeed, a carefully adjusted diaphragm has frequently been proven to be a suitable measure to avoid multiple filamentation [46]. Under these assumptions, it suffices to consider cylindrically symmetric solutions \( E(r, z, t) \) of Eq. (2.55). In this case the Laplacian in Eq. (2.55) can be reduced to its radial component

\[
\Delta_\perp = \frac{1}{r} \partial_r r \partial_r,
\]

where the radial coordinate is given by \( r = \sqrt{x^2 + y^2} \).

2.5 Properties of Filamentary Propagation

In the following, various limiting cases of the envelope Eq. (2.55) are considered, and a discussion of the phenomena relevant in the respective regimes is provided.

2.5.1 Dispersion

Dispersion is usually referred to as the frequency dependence of certain material properties governing the response to an external optical field, such as the refractive index \( n = n(\omega) \) or the nonlinear susceptibilities [14]. In linear optics, an external optical field induces a frequency dependent polarization, which may reshape the irradiated optical pulse during propagation, as different frequency components of the pulse propagate with unequal phase velocities in the medium. Neglecting the
nonlinear response terms and assuming that the pulse is sufficiently long such that the SVEA can be applied, it is a reasonable approximation to set $T = 1$ in Eq. (2.55). In addition, only plane waves propagating into the positive $z$-direction are considered. Then, it follows that the NEE reduces to

$$\partial_z \mathcal{E} = i D \mathcal{E}. \quad (2.65)$$

The dispersion operator accounts for group-velocity dispersion (GVD) governed by $\beta_2$, third-order dispersion (TOD) governed by $\beta_3$ and higher-order dispersion terms [41]. Here, the effect of GVD on an initially Gaussian-shaped temporal profile,

$$\mathcal{E}(0, t) = N(0)e^{-t^2/t_p(0)^2} \quad (2.66)$$

will be discussed. Neglecting all higher order terms but GVD, Eq. (2.65) can be integrated to yield [41]

$$\mathcal{E}(z, t) = N(z)e^{-t^2/t_p(z)^2} - i C(z)t^2/t_p(z)^2 \quad (2.67)$$

with

$$N(z) = \frac{N(0)}{\sqrt{1 - i z/L_D}}, \quad t_p(z) = t_p(0)\sqrt{1 + (z/L_D)^2}, \quad C(z) = z/L_D, \quad (2.68)$$

where $L_D = t_p^2(0)/2|\beta_2|$. The expressions for the normalization constant $N(z)$ and the pulse duration $t_p(z)$ show that the amplitude of the pulse decreases, while the duration $t_p(z)$ increases along $z$. The chirp factor $C(z)$ shows that GVD introduces a linear frequency chirp on the pulse. A discussion of chirped Gaussian pulses can be found in the introduction to the subsequent chapter dealing with pulse compression techniques. A characteristic length-scale on which these processes take place is provided by $L_D$.

While group velocity dispersion $\beta_2$ introduces a symmetric temporal broadening of the pulse envelope, the odd-order terms $\beta_{2k+1}$ terms introduce an asymmetric temporal stretching. Especially, the impact of TOD on Gaussian pulses can be described analytically in terms of Airy functions [41]. In filamentation, one typically focuses pulsed femtosecond laser beam into noble gases or air at atmospheric pressure. However, these exhibit a relatively weak dispersion [42]. For example, argon at standard conditions has $\beta_2 = 0.2$ fs$^2$/cm, such that the characteristic length scale $L_D = 62.5$ m for a 50 fs initial pulse. In fact, one may estimate that GVD only becomes relevant for pulses with initial duration $<10$ fs, for which $L_D$ approaches the typical propagation distances of the order of $\sim 1$ m used in experimental investigations of filamentary propagation.
2.5.2 Self-Phase Modulation

Self-phase modulation arises from the intensity dependence of the refractive index, \( n = n_0 + n_2 I \). This nonlinear effect can result in a substantial spectral broadening of an optical pulse, leading to the formation of a white-light supercontinuum. In condensed media, this was first observed in Ref. [47].

In order to investigate its impact on the evolution of a laser pulse, it is assumed that the optical intensity is sufficiently low not to trigger photo-ionization. Further on, it is assumed that the SVEA can be applied, leading to the condition that the pulse has to be much longer than the optical cycle, such that setting \( T = 1 \) in Eq. (2.55) is justified. If one additionally assumes that the dispersion length \( L_D \) is large, it is possible to neglect dispersion, setting \( D = 0 \). Using a polar decomposition of the complex envelope \( E(z, t) \) of a plane wave propagating in the \( z \)-direction according to \( E(z, t) = |E(z, t)| e^{-i\phi(z, t)} \), it can be inferred from the dynamical Eq. (2.55) that the temporal phase \( \phi(z, t) \) of the pulse acquires a self-induced temporal phase-shift according to [12, 20]

\[
\phi(z + \Delta z, t) = \phi(z, t) - \frac{\omega_0}{c} n_2 |E(z, t)|^2 \Delta z. \tag{2.69}
\]

From this, the SPM induced change of the instantaneous frequency, calculated as the time derivative of the instantaneous phase \( \phi(t) \), is given by

\[
\Delta \omega(t) = -\frac{\omega_0}{c} n_2 \frac{\partial}{\partial t} |E(z, t)|^2 \Delta z. \tag{2.70}
\]

Assuming a Gaussian temporal shape of the pulse \( E \sim \exp(-t^2/t_p^2) \), it follows that the SPM induced change of the instantaneous frequency satisfies

\[
\Delta \omega(t) \sim t e^{-t^2/t_p^2}. \tag{2.71}
\]

This reveals that action of SPM on the leading edge \((t < 0)\) of the pulse produces a spectral redshift, while a blueshift is produced in the trailing edge \((t > 0)\). In summary, SPM generates new spectral content, leading to a broadening of the frequency spectrum of the pulse [20, 41]. Under the above approximations, SPM only affects the temporal phase, leaving the temporal profile \( |E(z, t)| \) unchanged. This ceases to be true when dispersion can no longer be neglected. Assuming, for simplicity, that the pulse is subject to GVD only, it can be shown that the combined action of normal GVD \((\beta_2 > 0)\) and SPM leads to the phenomenon of optical wavebreaking [41]. This becomes noticeable as a steepening both of the leading and trailing edges of the pulse, which in turn yields a strong impact of GVD on the steepened pulse edges. The latter causes rapid oscillations of the pulse envelope in leading and trailing edge. The formation of pronounced spectral sidelobes are the frequency domain analog of optical wavebreaking.
If the initial pulse is symmetric in time, the aforementioned interplay of GVD and SPM preserves this symmetry as the pulse propagates along $z$. However, the situation dramatically changes when the pulse duration approaches the order of the optical cycle. In this case, the operator $T$ in Eq. (2.55) becomes essential for a physically reasonable description, and the dynamical equation describing SPM in the few-cycle regime reads

$$\partial_z \mathcal{E} = -i \frac{\beta_2}{2} \frac{\partial^2}{\partial t^2} \mathcal{E} + i \frac{\omega_0}{c} n_2 T |\mathcal{E}|^2 \mathcal{E}. \quad (2.72)$$

Here, the operator $T$ can be regarded to account for an intensity dependence of the group velocity. For positive $n_2$, it takes into account that more intense parts of the pulse propagate slower and are delayed with respect to the less intense parts. This behavior causes a steepening of the trailing edge of the pulse, while the leading edge is unaffected by steepening effects. This characteristically asymmetric effect is known as self-steepening [48]. Depart of the characteristically asymmetric temporal pulse profile, SPM in the few-cycle domain also leads to a strong asymmetry in the spectrum. In fact, the generation of new frequency components by SPM is most pronounced when the envelope exhibits a strong temporal gradient. Therefore, the generation of blue spectral content in the steepened trailing edge of the pulse is strongly enhanced compared to SPM based generation of red frequencies in the slowly rising leading edge of the pulse. It follows that few-cycle pulses subject to SPM typically exhibit a strongly asymmetric spectrum, with a pronounced blueshifted spectral tail. In addition, as the blue spectral components are dominantly generated in the vicinity of the self-steepened trailing edge of the pulse, they are strongly localized in the temporal domain, leading to a nearly flat spectral phase in the blueshifted spectral range.

### 2.5.3 Self-Focusing

Besides the modulation of the temporal phase leading to spectral broadening and optical wavebreaking, the IDRI can cause a modulation of the spatial phase of the pulse. Assuming a monochromatic cw-beam propagating in a medium with vanishing plasma response, one finds that in this regime, setting $\mathcal{E} = \mathcal{E}(x, y, z)$, $T = 1$ and $D = W = \rho = 0$ leads to the simplified Eq. [5]

$$\partial_z \mathcal{E} = i \frac{k_0}{2} \Delta_L \mathcal{E} + i \frac{\omega_0}{c} n_2 |\mathcal{E}|^2 \mathcal{E} \quad (2.73)$$

This is the Nonlinear Schrödinger equation in two transverse spatial dimensions ($x$, $y$) and one dimension corresponding to the propagation direction $z$. It corresponds to the paraxial approximation of the Helmholtz equation, augmented by a contribution due to the IDRI $n = n_0 + n_2 I$. The nonlinear part of the refractive index gives rise to a self-induced modulation of the spatial phase,
2.5 Properties of Filamentary Propagation

\[ \phi(x, y, z) \rightarrow \phi(x, y, z) + \frac{\omega_0}{c} n_2 I(x, y, z) \Delta z. \] (2.74)

For a Gaussian beam and positive \( n_2 \), the self-induced spatial phase exhibits negative curvature and mimics the action of a focusing lens. This may lead to persistent self-focusing of the beam until the intensity blows up, as was first observed in Ref. [49]. The nonlinear Schrödinger equation can be analyzed by several approximate approaches, considering for instance the propagation of rays in a self-induced refractive index profile, or, alternatively, the method of moments [50]. All these approaches predict that a Gaussian beam in a medium with positive \( n_2 \) will tend to self-focus until the amplitude blows up at a finite distance \( z_{cr} \), and the solution diverges, given that its optical power \( \sim \int |\mathcal{E}(x, y, z)|^2 dx\,dy \) exceeds a certain critical value \( P_{cr} \). The numerical value of this critical power differs slightly between the various approximative approaches. An analytical treatment of Eq. (2.74) reveals that the critical power for self-focusing is given by [50–52]

\[ P_{cr} = \frac{11.69 \lambda^2}{8\pi^2 n_0 n_2}. \] (2.75)

Here, \( P_{cr} \) is the optical power \( P \) of specific transverse profile, the so-called Townes mode [52], which provides a family of stationary solutions to Eq. (2.73). A more detailed discussion of the mathematical background of self-focusing described by the Nonlinear Schrödinger equation can be found in Appendix A. It can be shown that an arbitrarily shaped beam will not collapse if its optical power \( P \) satisfies \( P < P_{cr} \) [53]. Therefore, \( P > P_{cr} \) provides a necessary, but not sufficient condition for collapse to occur. For Gaussian beams, collapse does not occur unless \( P > P_{cr}^G \approx 1.02 P_{cr} \) [50].

Note that a frequently used approximation for the critical power is given by \( P_{cr} = \lambda_0^2/\left(2\pi n_0 n_2\right) \) which is used throughout the thesis. This approximate value can, as will be shown in Sect. (3.1), be derived using a variational approach (see Appendix A). This variational approach is closely related to the aforementioned method of moments. Also, it has to be emphasized that the Townes profile provides a stationary, yet unstable solution to (2.75)[53–55]. This is due to the fact that any arbitrary small perturbation will either cause collapse or decay of the Townes mode. In this aspect, the Townes solution differs from the familiar fiber solitons [41] in the anomalous dispersion regime \( \beta_2 < 0 \), which are governed by a (1+1)-dimensional NLSE analogous to (2.74) and are unconditionally stable. A semiempirical formula for the finite propagation distance \( z_{cr} \) at which a Gaussian beam with beam waist \( w_0 \) at \( z = 0 \) collapses was provided by Marburger [56], pursuant to

\[ z_{cr} = \frac{0.376 z_0}{\sqrt{\left(\frac{P_{in}}{P_{cr}} - 0.852\right)^2 - 0.0219 + \frac{z_0}{f}}}, \] (2.76)

where \( z_0 = \pi n_0 w_0^2/\lambda_0 \) is the Rayleigh range of a collimated Gaussian beam, \( f = R/2 \) the focal point, and \( R \) is the radius of curvature of the beam wavefront.
A detailed analysis of this formula shows that $z_{cr}$ may be smaller than $f$ such that the beam collapses at a position prior to the position of the linear focus. Thus, in order to distinguish it from the geometrical focus of linear theory, $z_{cr}$ is often termed nonlinear focus. Of course, beam collapse does not occur in physical systems, as counteracting effects come into play as the beam intensity blows up. For example, it has been shown that nonparaxiality and vectorial effects can stop the collapse of the beam \([16, 57]\). However, these effects become relevant at intensities far beyond the photoionization threshold. While photoionization is currently supposed to be the prevalent mechanism to stop beam collapse, as discussed below, the discussion of Chap. 4 reveals the possible role played by higher-order Kerr nonlinearities. Besides, also GVD turns out to be able to saturate the self-focusing collapse, this will be discussed below, and, in more detail, in Sect. 3.3.

### 2.5.4 Modulation Instabilities

The Townes mode is a radially symmetric solution to \((2.74)\). However, besides the previously discussed self-focusing instability, solutions of \((2.74)\) suffer from the so-called azimuthal modulation instability, which is able to break the radial symmetry of a given solution. To be more precise, an infinitesimally small, radially asymmetric perturbation to the radially symmetric initial field will, under certain conditions, be exponentially amplified, leading to a spatial break-up and loss of radial symmetry of the initial solution. Theoretically, this was first observed by Bespalov and Talanov \([58]\) by means of a plane wave propagating in the $z$ direction, being perturbed by a small amplitude wave with nonvanishing transverse wave vector $\vec{k}_\perp = (k_x, k_y)$.

In the context of femtosecond filamentation, this phenomenon is known as multifilamentation and is expected to be observed when the input power of the pulsed laser beam strongly exceeds the critical power $P_{cr}$ for self-focusing. Nevertheless, for beam powers up to five critical powers, it has been demonstrated that the onset of multifilamentation can be circumvented by means of suitably aperturing the input beam \([45]\).

In addition, modulation instability is the cause of another phenomenon occurring in the context of femtosecond filamentation, namely the generation of hyperbolic shock-waves, X-waves and conical emission \([59]\). The latter instability occurs due to the interplay of self-focusing and normal GVD. In fact, it can be shown that GVD with $\beta_2 > 0$ is able to counteract the optical collapse induced by the Kerr nonlinearity \([60]\). Considering the evolution of long input pulses such that the SVEA ($T = 1$) can applied, and further neglecting plasma response and higher order dispersion $\beta_k$ for $k > 2$, the envelope Eq. \((2.55)\) reduces to

$$
\partial_z \mathcal{E} = \frac{i}{2k_0} \Delta_{\perp} \mathcal{E} - i \frac{\beta_2}{2} \frac{\partial^2}{\partial t^2} \mathcal{E} + i \frac{\omega_0}{c} n_2 |\mathcal{E}|^2 \mathcal{E}.
$$

\((2.77)\)
For $\beta_2 > 0$, this is a Nonlinear Schrödinger Equation in $(2+1)$-dimensions with a hyperbolic wave operator $\alpha \Delta_\perp - \gamma \frac{\partial^2}{\partial t^2}$ with $\alpha, \gamma > 0$. The latter equation admits identical stationary solution as Eq. (2.74), namely a monochromatic beam with a transverse beam profile given by the Townes mode. However, the detailed analysis of [61] reveals that a small perturbation to the stationary solution may acquire exponential gain, leading to the formation of so-called X-waves. The latter have recently been related to the phenomenon of conical emission frequently observed in filamentation [59].

### 2.5.5 Space-Time Focusing

Space-time focusing is not a phenomenon restricted to filamentary propagation. Rather, it describes the impact of linear diffraction on ultrashort, ultra-broadband optical pulses. Diffraction, i.e. the spreading of a laser beam in the transverse plane, is governed by the dispersion relation

$$n^2(\omega) \frac{\omega^2}{c^2} = k_x^2 + k_y^2 + k_z^2.$$  \hspace{1cm} (2.78)

In the case of paraxial propagation into the positive $z$-direction, the latter relation may be approximated using $\sqrt{1+x} \approx 1 + x/2$, which yields

$$k_z = k(\omega) \left(1 - \frac{k_\perp^2}{2k^2(\omega)} \right).$$  \hspace{1cm} (2.79)

However, it was noticed in Ref. [62] that the SVEA, which corresponds to the replacement $k(\omega) \rightarrow k_0$ in the latter equation, fails to describe spatiotemporal coupling effects which occur for ultrashort, diffracting laser pulses. Indeed, it can be deduced from the dispersion relation Eq. (2.78) that blue spectral components of a pulse diffract slower than their red spectral counterparts, which eventually gives rise to a narrowing of the on-axis spectrum of the beam. This corresponds to a pulse broadening in the temporal domain. Therefore, it became necessary to include a correction term to the SVEA model to account for the spatiotemporal coupling effect governing the diffraction of ultrashort pulses. In the NEE model (2.55), this correction is provided by the augmented diffraction term $\sim T^{-1} \Delta_\perp$. In contrast to the NEE model, the factorization procedure leading to the FME naturally respects the dispersion relation (2.79).
2.5.6 Intensity Clamping and the Dynamic Spatial Replenishment Model

The intensity clamping model [63] of femtosecond filamentation assumes that the onset of photoionization and the subsequent defocusing effect due to free carriers is the dominant mechanism to arrest the beam collapse triggered by Kerr self-focusing. Using the expression for the IDRI $\Delta n = n_2 I$ and the corresponding expression for the plasma-induced refractive index change according to Eq. (2.43), the nonlinear change of the refractive index is given by

$$\Delta n = n_2 I - \frac{\rho}{n_0^2 \rho_c}$$

(2.80)

For a self-focusing medium $n_2 > 0$, the plasma contribution to the refractive index has a sign opposite to that of the Kerr nonlinearity. Therefore, the plasma-induced refractive index profile acts like a defocusing lens, as the density of free carriers generally increases towards the optical axis. The intensity clamping model provides an estimate for the maximal intensity achieved within femtosecond filaments. At the clamping intensity, defined as the solution of

$$\Delta n = n_2 I - \frac{\rho}{n_0^2 \rho_c} = 0,$$

(2.81)

the plasma induced refractive index change is sufficient to generate a defocusing effect that balances Kerr induced self-focusing. Using (2.56) and neglecting plasma losses, the rate equation for $\rho$ can be integrated to yield

$$\rho = \rho_0 \left(1 - \exp\left[-\int_{-\infty}^{t} w[I(t')]dt'\right]\right).$$

(2.82)

However, in order to obtain an estimate of the clamping intensity, it suffices to consider the peak plasma density and peak pulse intensities, $\rho_{\text{max}}$ and $I_{\text{max}}$. Using $\exp(-x) \approx 1 - x$ for not too high plasma densities, one finds that $\rho_{\text{max}} = t_p \rho_0 w(I_{\text{max}})$, leading to the following equation for the clamping intensity [8]

$$I_{\text{max}} = \frac{t_p \rho_0 w(I_{\text{max}})}{2 \rho_c n_0 n_2}.$$

(2.83)

It has been shown that this expression generally provides a good estimate for the intensity in the nonlinear focus [13]. However, intensity clamping does not imply that long-range filamentary propagation is a stationary process, with constant intensity $I_{\text{max}}$ along the entire longitudinal extension of the filament. Rather, filamentation is a highly dynamical process, and a detailed examination of a femtosecond filament reveals that it consists of recurrent nonlinear foci along its longitudinal axis. Besides,
it has been demonstrated in Ref. [64] that the different temporal sections of the pulse are subject to repeated focusing and refocusing cycles such that the pulse fluence, i.e. the time integrated intensity given in units of J/m², appears stationary and, according to [64]: “[...]produce[s] the illusion of long-distance propagation of one self-guided pulse”. This model was termed Dynamic spatial replenishment by the authors of [64].

2.5.7 Pulse Self-Compression

Pulse self-compression probably is the most intriguing phenomenon observed in femtosecond filamentation. Unless traditional schemes for laser pulse compression, it enables the generation of ultrashort pulses consisting only of a few cycles of the optical carrier field without the need for any dispersive compression techniques. The following Chap. 3 will discuss filamentary self-compression in detail, revealing both theoretical and experimental aspects of the topic.

References

1. J.C. Maxwell, On physical lines of force. Philos Mag. 21, 161(1861)
34. L.V. Keldysh, Ionization in the field of a strong electromagnetic wave. Sov. Phys. JETP 20, 1307 (1965)
References

Nonlinear Optics in the Filamentation Regime
Brée, C.
2012, XVI, 128 p., Hardcover
ISBN: 978-3-642-30929-8