

Chapter 2

Theoretical Description of the Nonlinear Optical Pulse Propagation

An electric field $E(\vec{r}, t)$ interacts with the charged properties of matter. As electrons possess considerably less mass than the ionic cores, their dynamics excited by the driving field evolve on shorter timescales than the induced lattice oscillation.

The external field polarizes the material, inducing oscillating dipoles which themselves emit radiation. Therefore the propagation of the light inside the medium takes the form of a polariton, consisting of the electric field $E(\vec{r}, t)$ coupled to the polarization wave $P(\vec{r}, t)$. The propagation and interaction of $E(\vec{r}, t)$ and $P(\vec{r}, t)$ in the absence of free carriers and charges is described by the nonlinear wave equation [1]

$$-\nabla^2 E(\vec{r}, t) + \frac{1}{c^2} \frac{\partial^2 E(\vec{r}, t)}{\partial t^2} = -\frac{1}{c^2 \epsilon_0} \frac{\partial^2 P(\vec{r}, t)}{\partial t^2} \quad (2.1)$$

c is the speed of light in vacuum and ϵ_0 the vacuum permittivity. Equation (2.1) uses the MKS unit system. This convention will be employed throughout the entire thesis. If the amplitude of the external field becomes sufficiently large ($\geq 1 \frac{\text{V}}{\text{\AA}}$ in dielectrics) the dipoles cannot follow the driving field linearly anymore. This results in a nonlinear contribution $P^{NL}(\vec{r}, t)$ to the polarization response [2].

$$P(\vec{r}, t) = P^L(\vec{r}, t) + P^{NL}(\vec{r}, t) \quad (2.2)$$

$P^L(\vec{r}, t)$ represents the linear material response and accounts for the linear dispersion the different frequency components experience during propagation [1].

$$P^L(\vec{r}, t) = \epsilon_0 \chi^{(1)}(\vec{r}, t) E(\vec{r}, t) \quad (2.3)$$

$\chi^{(1)}(\vec{r}, t)$ is the lowest order susceptibility. In general $\chi^{(l)}(\vec{r}, t)$ are tensors of the order $l + 1$ and depend on the frequency of the applied field. For reasons of lucidity $\chi^{(l)}$ will be treated as scalars in the following and the material response is assumed to be instantaneous.

If the amplitude of the nonlinear contribution to the polarization is sufficiently small, it can be considered as a small perturbation of the system and approximated in a Taylor expansion [1].

$$P^{NL}(\vec{r}, t) = \epsilon_0 \chi^{(2)} E(\vec{r}, t)^2 + \epsilon_0 \chi^{(3)} E(\vec{r}, t)^3 + \dots \quad (2.4)$$

The susceptibility and the first order material permittivity $\epsilon^{(1)}$ are connected by $\epsilon^{(1)} = 1 + \chi^{(1)}$. Additionally $\epsilon^{(1)}$ is related to the linear refractive index $n_0^2 = \epsilon^{(1)}$. Under consideration of Eq. (2.1) and this dependency Eq. (2.2) can be written as [1]

$$-\nabla^2 E(\vec{r}, t) + \frac{\epsilon^{(1)} \partial^2 E(\vec{r}, t)}{c^2} = -\frac{1}{c^2 \epsilon_0} \frac{\partial^2 P^{NL}(\vec{r}, t)}{\partial t^2} \quad (2.5)$$

Fourier transformation of Eq. (2.5) into the frequency domain results in a differential equations for every frequency component $\tilde{E}_\omega(\vec{r})$. The individual equations are coupled by the nonlinear source term $\tilde{P}_\omega^{NL}(\vec{r})$ which involves all frequencies as it depends on the complete electric field.

$$\nabla^2 \tilde{E}_\omega(\vec{r}) + \frac{\epsilon^{(1)} \omega^2}{c^2} \tilde{E}_\omega(\vec{r}) = -\frac{\omega^2}{c^2 \epsilon_0} \tilde{P}_\omega^{NL}(\vec{r}) \quad (2.6)$$

with

$$E(\vec{r}, t) = \frac{1}{2\pi} \int \tilde{E}_\omega(\vec{r}) \exp(i\omega t) d\omega \quad (2.7)$$

$$P^{NL}(\vec{r}, t) = \frac{1}{2\pi} \int \tilde{P}_\omega^{NL}(\vec{r}) \exp(i\omega t) d\omega. \quad (2.8)$$

This thesis focuses on the examination of the nonlinear interaction of ultrashort light pulses with matter. Particularly for this case Brabec and Krausz [3] have further elaborated Eq. (2.6). The derived result is a generalized form of the nonlinear Schrödinger equation and describes a broad variety of nonlinear optical effects. To understand their physical origin it is very instructive to study the analysis of Brabec and Krausz in more detail. In the following the fundamental assumptions and the most important intermediate steps leading to the generalized nonlinear Schrödinger equation are presented.

Equation (2.6) can be simplified with the slowly-evolving-amplitude-approximation [1]. It implicates that the amplitude $A(\vec{r}, t)$ of the incident electric field $E(\vec{r}, t) = A(\vec{r}, t) \exp(-i(kz - \omega t)) + cc.$ does not vary significantly within one wavelength. For $E(\vec{r}, t)$ propagating in z direction the following ansätze can be employed to solve Eq. (2.6):

$$E(\vec{r}, t) = A(\vec{r}, t) \exp(-i(k_0 z - \omega_0 t)) + cc. \quad (2.9)$$

$$A(\vec{r}, t) = \frac{1}{2\pi} \int \tilde{A}_\omega(\vec{r}) \exp(i\omega t) d\omega \quad (2.10)$$

$$P^{NL}(\vec{r}, t) = p^{NL}(\vec{r}, t) \exp(-i(k_0 z - \omega_0 t)) + cc. \quad (2.11)$$

$$p^{NL}(\vec{r}, t) = \frac{1}{2\pi} \int \tilde{p}_\omega^{NL}(\vec{r}) \exp(i\omega t) d\omega \quad (2.12)$$

$\tilde{A}_\omega(\vec{r})$ and $\tilde{p}_\omega^{NL}(\vec{r})$ are the slowly varying amplitudes of the field and the nonlinear polarization wave in the frequency domain. ω_0 is the center frequency and $k_0 = \frac{2\pi}{\lambda_0}$ the wavevector at the fundamental wavelength λ_0 . Due to the slowly-evolving-amplitude-approximation $\frac{\partial^2 \tilde{A}_\omega(\vec{r})}{\partial z^2} \approx 0$ in Eq. (2.6) [4]. Higher order dispersion terms are considered with an expansion of the wavevector $k(\omega)$ into a Taylor series

$$k(\omega) = k_0 + k_1(\omega - \omega_0) + \underbrace{\frac{1}{2}k_2(\omega - \omega_0)^2 + \dots}_{\tilde{D}_\omega} \quad (2.13)$$

where $\tilde{D}_\omega = \sum \frac{1}{l!} k_l (\omega - \omega_0)^l$ for $l \geq 2$.

$$k_1 = \left[\frac{dk}{d\omega} \right]_{\omega=\omega_0} = \frac{1}{c} \left[n(\omega) + \omega \frac{dn(\omega)}{d\omega} \right]_{\omega=\omega_0} = \frac{1}{v_g(\omega_0)} \quad (2.14)$$

$$k_2 = \left[\frac{d^2 k}{d\omega^2} \right]_{\omega=\omega_0} = \frac{d}{d\omega} \left[\frac{1}{v_g(\omega_0)} \right]_{\omega=\omega_0} = \left[-\frac{1}{v_g^2(\omega_0)} \frac{dv_g}{d\omega} \right]_{\omega=\omega_0} \quad (2.15)$$

$n(\omega)$ is the refractive index of the medium. The linear term k_1 in the expansion stands for the inverse group velocity of the pulse v_g . k_2 and the higher order terms represent the group velocity dispersion.

Under consideration of the slowly-evolving-amplitude-approximation Eqs. (2.9), (2.11) and the dispersion treatment presented above, Eq. (2.6) is Fourier transformed back into time domain. Subsequently a retarded time frame moving with the light pulse is introduced

$$z = z' \quad \tau = t - k_1 z. \quad (2.16)$$

This leads to the following result [1, 4]

$$\begin{aligned} & \left[\Delta_\perp + 2ik_0 \frac{\partial}{\partial z'} \left(1 + \frac{ik_1}{k_0} \frac{\partial}{\partial \tau} \right) + 2k_0 D \left(1 + \frac{ik_1}{k_0} \frac{\partial}{\partial \tau} \right) \right] A(\vec{r}, \tau) \\ & = -\frac{\omega_0^2}{\epsilon_0 c^2} \left(1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right)^2 p^{NL}(\vec{r}, \tau) \end{aligned} \quad (2.17)$$

with the temporal representation of \tilde{D}_ω , $D = \sum \frac{1}{l!} k_l (i \frac{\partial}{\partial \tau})^l$.

In order to identify the different physical mechanisms which the individual terms in Eq. (2.17) represent it is instructive to restrict the perturbative expansion of the nonlinear source term to its lowest order contributions Eq. (2.4). In the course of this thesis only centrosymmetric materials are considered in which the even order contributions to the nonlinearity vanish. Hence $p^{NL}(\vec{r}, t)$ is approximated with

$$p^{NL}(\vec{r}, t) = 3\epsilon_0\chi^{(3)} |A(\vec{r}, t)|^2 A(\vec{r}, t). \quad (2.18)$$

To get an intuitive understanding of Eq. (2.17) the correction factors $\frac{ik_1}{k_0} \frac{\partial}{\partial \tau}$ and $\frac{i}{\omega_0} \frac{\partial}{\partial \tau}$ are neglected in a first step and group velocity dispersion is restricted up to second order k_2 Eq. (2.13). With these approximations Eq. (2.17) can be written as [1]

$$\frac{\partial A(\vec{r}, \tau)}{\partial z'} = \left[\frac{i}{2k_0} \Delta_{\perp} - \frac{ik_2}{2} \frac{\partial^2}{\partial \tau^2} + \frac{3i\omega_0}{2n_0 c} \chi^{(3)} |A(\vec{r}, \tau)|^2 \right] A(\vec{r}, \tau) \quad (2.19)$$

This representation of the nonlinear Schrödinger equation highlights that three different physical mechanisms influence the amplitude $A(\vec{r}, \tau)$ of the electric field on the left hand side of Eq. (2.19): the first term on the right side represents diffraction as it modifies the extension of the wave in x and y direction. The second contribution containing k_2 alters the temporal profile of the amplitude due to group velocity dispersion. The last term on the right hand side is proportional to $|A(\vec{r}, \tau)|^2$ and represents an intensity dependent nonlinear contribution known as the optical Kerr effect [1].

2.1 Self-Phase Modulation

By analogy with the relation between the linear refractive index n_0 , the first order permittivity $\epsilon^{(1)}$ and the susceptibility $\chi^{(1)}$

$$n_0^2 = \epsilon^{(1)} = 1 + \chi^{(1)} \quad (2.20)$$

the intensity dependence of the material's polarization response can be included in the description of the refractive index.

$$n(t)^2 = 1 + \chi^{(1)} + 3\chi^{(3)} |A(\vec{r}, t)|^2 + \dots \quad (2.21)$$

As only media with inversion symmetry are considered in the course of this thesis, even order contribution to the polarization are ignored.

Equation (2.21) can be written as

$$n(t) = n_0 + n_2 I(\vec{r}, t) \quad (2.22)$$

with the nonlinear Kerr coefficient $n_2 = \frac{3\chi^{(3)}}{n_0^2\epsilon_0 c}$ and the pulse intensity $I(\vec{r}, t) = \frac{1}{2}n_0\epsilon_0 c |A(\vec{r}, t)|^2$

This notation illustrates that the optical Kerr effect introduces an intensity dependent nonlinear phase contribution Φ^{NL}

$$\Phi^{NL}(t) = \omega_0 \frac{z}{c} n_2 I(t) \quad (2.23)$$

and modifies the instantaneous frequency ω of the electric field [2].

$$\delta\omega = -\frac{d\Phi^{NL}}{dt} \quad (2.24)$$

Figure 2.1 depicts the nonlinear phase $\Phi^{NL}(t)$ (dark line, Eq. (2.23)) and corresponding shift of the instantaneous frequency $\delta\omega$ (light line, Eq. (2.24)) a Gaussian pulse induces in a medium with $n_2 > 0$. As $\Phi^{NL}(t)$ is proportional to the intensity envelope, $\delta\omega$ is negative in the first half of the pulse and positive at its end. If the magnitude of the spectral modifications becomes comparable to the bandwidth of the incident field, the fundamental spectrum can be significantly broadened or narrowed depending on the initial chirp of the driving wave [2]. As the spectral width of the pulse $\Delta\omega$ is related to its duration τ_p by $\Delta\omega \propto \frac{2\pi}{\tau_p}$, self-phase modulation considerably alters the frequency structure of the fundamental if $\Phi_{max}^{NL} \geq 2\pi$.

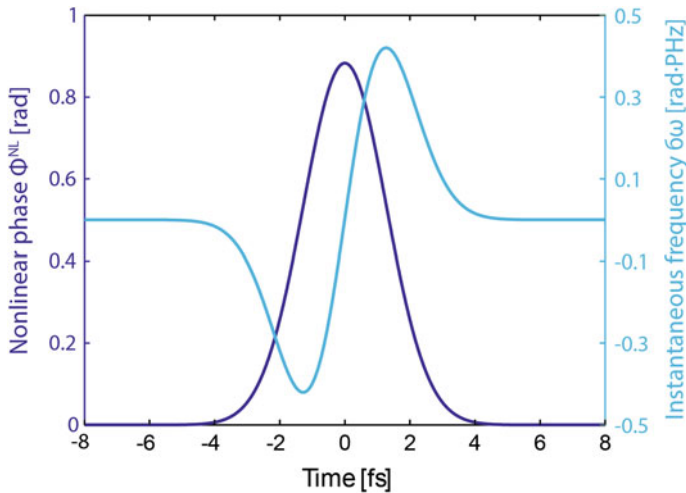


Fig. 2.1 In a medium with positive n_2 a Gaussian pulse generates the nonlinear phase $\Phi^{NL}(t)$ (dark line) proportional to its intensity envelope. $\Phi^{NL}(t)$ changes the instantaneous frequency of the carrier wave (light line)

$$\delta\omega_{max} \approx \frac{\Phi_{max}^{NL}}{\tau_p} \approx \frac{2\pi}{\tau_p} \rightarrow \Phi_{max}^{NL} \geq 2\pi \quad (2.25)$$

For a Gaussian pulse $\delta\omega(t)$ reaches its maximum and minimum amplitude at the inflection points of $\Phi^{NL}(t)$ Fig. 2.1. In between it can assume the same value at two different instants in time, resulting in an interference modulation of the nonlinearly modified spectrum [2].

2.2 Self-Focusing

The intensity dependence of the refractive index Eq. (2.22) does not only influence the spectral composition of the pulse but also its spatial structure. In the transverse profile of a Gaussian beam the intensity decreases with the distance from the optical axis. This results in a spatially inhomogeneous nonlinear modification of the optical material density across the beam area: for a medium with $n_2 > 0$ the refractive index becomes highest at the beam center and decreases towards its sides. Hence the nonlinear Kerr effect introduces a focusing lens Fig. 2.2 in the material. The location of the focus can be calculated with Fermat's principle [1], if the beam diameter is considerably larger than the wavelength of the electric field and diffraction can be neglected. Then the optical path of all rays at different transverse positions in the material must be the same.

Figure 2.2 illustrates how Fermat's principle determines the focusing angle θ in the medium. After entering the material the beams at different radial positions meet at the self-focusing length z_{sf} .

$$(n_0 + n_2 I) z_{sf} = \frac{n_0 z_{sf}}{\cos \theta} \quad (2.26)$$

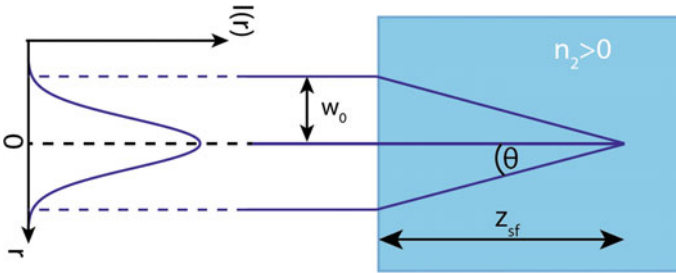


Fig. 2.2 The intensity dependent refractive index alters the optical path length for rays at different radial positions. In materials with $n_2 > 0$ the Gaussian intensity distribution leads to self-focusing of the beam after a propagation distance of z_{sf}

$$\cos \theta \approx 1 - \frac{1}{2} \theta^2 \rightarrow \theta = \sqrt{\frac{2n_2 I}{n_0 + n_2 I}} \quad (2.27)$$

As θ is related to the beam radius w_0 by $\theta = \frac{w_0}{z_{sf}}$, z_{sf} is given by

$$z_{sf} = w_0 \sqrt{\frac{n_0 + n_2 I}{2n_2 I}}. \quad (2.28)$$

2.3 Self-Steepening

To further investigate the impact of the Kerr effect on the nonlinear light-matter interaction, the factor $\frac{i}{\omega_0} \frac{\partial}{\partial \tau}$ on the left hand side of Eq. (2.17) is now taken into account. The terms $\frac{ik_1}{k_0} \frac{\partial}{\partial \tau}$ on the right side are further neglected. According to the slowly-evolving-amplitude-approximation $\left[1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau}\right]^2 \approx 1 + \frac{2i}{\omega_0} \frac{\partial}{\partial \tau}$.

The frequency dependence of $\chi^{(3)}$ is taken into account with the lowest order terms of a Taylor expansion

$$\chi^{(3)}(\omega) \approx \chi^{(3)}(\omega_0) + (\omega - \omega_0) \frac{d\chi^{(3)}}{d\omega} [5].$$

Back in the laboratory reference frame Eq. (2.17) can be written as

$$\begin{aligned} \frac{\partial A(\vec{r}, t)}{\partial z} - k_1 \frac{\partial A(\vec{r}, t)}{\partial t} &= \frac{i}{2k_0} \Delta_{\perp} A(\vec{r}, t) - \frac{ik_2}{2} \frac{\partial^2 A(\vec{r}, t)}{\partial t^2} + \frac{3i\omega_0}{2n_0 c} \chi^{(3)}(\omega_0) |A(\vec{r}, t)|^2 A(\vec{r}, t) \\ &+ \frac{3i\omega_0}{2n_0 c} \chi^{(3)}(\omega_0) \left[2 + \frac{\omega_0}{\chi^{(3)}(\omega_0)} \frac{d\chi^{(3)}}{d\omega} \right] \frac{i}{\omega_0} \frac{\partial}{\partial t} |A(\vec{r}, t)|^2 A(\vec{r}, t). \end{aligned} \quad (2.29)$$

The first three terms on the right hand side have been previously identified as the contribution of diffraction, group velocity dispersion and self-phase modulation, respectively. By computing $\frac{\partial}{\partial t} (|A(\vec{r}, t)|^2 A(\vec{r}, t)) = 2|A(\vec{r}, t)|^2 \frac{\partial A(\vec{r}, t)}{\partial t} + A(\vec{r}, t)^2 \frac{\partial A^*(\vec{r}, t)}{\partial t}$ Eq. (2.29) becomes

$$\begin{aligned} \frac{\partial A(\vec{r}, t)}{\partial z} - \left[k_1 + \frac{4\gamma c}{\omega_0} |A(\vec{r}, t)|^2 \right] \frac{\partial A(\vec{r}, t)}{\partial t} &= \frac{i}{2k_0} \Delta_{\perp} A(\vec{r}, t) - \frac{ik_2}{2} \frac{\partial^2 A(\vec{r}, t)}{\partial t^2} \\ &+ \frac{3i\omega_0}{2n_0 c} \chi^{(3)}(\omega_0) |A(\vec{r}, t)|^2 A(\vec{r}, t) \\ &- \frac{2\gamma}{\omega_0} A(\vec{r}, t)^2 \frac{\partial A(\vec{r}, t)}{\partial t} \end{aligned} \quad (2.30)$$

with $\gamma = \frac{3\omega_0}{2n_0c} \chi^{(3)}(\omega_0) \left[1 + \frac{\omega_0}{2\chi^{(3)}(\omega_0)} \frac{d\chi^{(3)}}{d\omega} \right]$. The second term on the left hand side shows that an intensity dependent contribution to the inverse group velocity emerges in the nonlinear light-matter interaction. It can be described with the nonlinear group index $n_2^{(g)}$ which is related to the effective group velocity index by $n_{eff}^{(g)} = n_0^{(g)} + n_2^{(g)} I$.

$$n_2^{(g)} = \frac{12}{n_0^2 c \epsilon_0} \chi^{(3)}(\omega_0) \left[1 + \frac{\omega_0}{2\chi^{(3)}(\omega_0)} \frac{d\chi^{(3)}}{d\omega} \right] \quad (2.31)$$

For $n_2^{(g)} > 0$, v_g decreases proportional to the intensity envelope of the driving field. Hence the central part of the pulse is decelerated to a higher extent than its flanks. This delays the first moment of the pulse and increases the slope at the trailing flank. The pulse front is flattened. The described phenomena is known as self-steepening and illustrated in Fig. (2.3b) [1]. For few-cycle pulses (in dispersionless media) self-steepening can increase the slope at the pulse end to infinity and form an optical shock wave Fig. (2.3c). When the central part of the pulse is even further delayed and overtaken by the components at the tail, the pulse breaks similar to the behavior known from water waves [6].

The analysis of the nonlinear interaction presented so far illustrates that various physical mechanisms influence the propagation of an intense light pulse through matter Eq. (2.30). The characteristic length scales on which the different effects significantly modify the pulse properties can be used to classify the relevance of the contribution. If the characteristic length of the nonlinear effect is small, its impact in the interaction is high.

The dispersion length $L_{dispersion}$ constitutes a measure for the amplitude of group velocity dispersion [1]

$$L_{dispersion} = \frac{\tau_p^2}{4 \ln(2) |k_2|}. \quad (2.32)$$

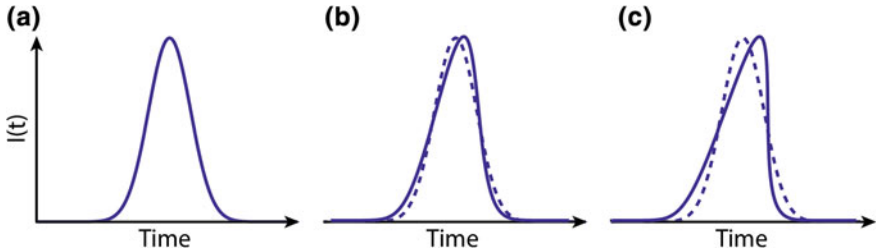


Fig. 2.3 Panel **a** the intensity envelope of a Gaussian pulse. Panel **b** the intensity envelope of the Gaussian pulse is altered by self-steepening in a material with $n_2^{(g)} > 0$. Panel **c** for ultrashort pulses severe self-steepening can induce the formation of optical shock waves

The impact of self-phase modulation can be estimated with the nonlinear length L_{NL} [2]

$$L_{NL} = \frac{c}{\omega n_2 I}. \quad (2.33)$$

For self-steepening the self-steepening distance L_{SS} is defined

$$L_{SS} = \frac{c\tau_p}{2\sqrt{\ln(2)}n_2^{(g)}I}. \quad (2.34)$$

2.4 Frequency Tripling

For reasons of lucidity only components of the nonlinear polarization at the fundamental frequency ω of the driving field have been considered so far Eq. (2.18). However in the nonlinear light-matter interaction also higher order harmonics $n \times \omega$ ($n \in \mathbb{N}$) are generated Eq. (2.4). In the $\chi^{(3)}$ process, 3 photons of the fundamental can be transformed into coherent radiation oscillating at a frequency of 3ω Fig. 2.4 [1].

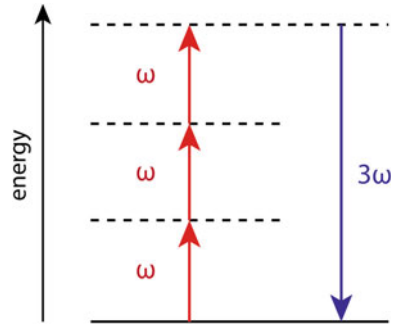
$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E(t)^3 = \epsilon_0 \chi^{(3)} A(t)^3 \exp(-i(3\omega t)) \quad (2.35)$$

To evaluate the efficiency of the nonlinear process the magnitude of $|\overleftrightarrow{\chi}^{(i)}|$ Eq. (2.35) is estimated. The interatomic atomic force E_{at} can be approximated with

$$E_{at} = \frac{e}{4\pi\epsilon_0 a_0^2} = 5.14 \times 10^{11} \frac{V}{m} \quad (2.36)$$

where $e = 1.6 \times 10^{-19} C$ is the electron charge, $\epsilon_0 = 8.85 \times 10^{-12} \frac{F}{m}$ the electric permittivity of free space and $a_0 = 5.29 \times 10^{-11} m$ the Bohr radius. If the amplitude of the external driving field is of the same order of magnitude as E_{at} , the amplitude of the linear material response $P^{(L)}(t)$ Eq. (2.3) is of similar size as for $P^{(NL)}(t)$ Eq. (2.4). As $\chi^{(1)} \approx 1$

Fig. 2.4 Schematic principle of frequency tripling [1]: 3 photons of the fundamental wave with frequency ω are absorbed by a two level system. In return a high energy photon with a frequency of 3ω is emitted



$$\epsilon_0 \chi^{(1)} E_{at} \approx \epsilon_0 \chi^{(i)} E_{at}^i \rightarrow \chi^{(i)} = \frac{1}{E_{at}^{i-1}}. \quad (2.37)$$

With increasing order of the nonlinearity $\chi^{(i)}$ decreases [7]. For second order processes $\chi^{(2)} \approx 19.5 \times 10^{-13} \frac{\text{m}}{\text{V}}$. For third order nonlinearities the efficiency drops considerably as $\chi^{(3)} \approx 38.0 \times 10^{-24} \frac{\text{m}^2}{\text{V}^2}$. The conversion rate for frequency tripling of VIS/NIR photons into the UV range in the gaseous phase is less than $<1\%$.

2.5 Intrapulse Raman-Scattering

As electrons possess less mass than the ionic cores most electronic excitations in solids oscillate at higher frequencies than the vibrations of the lattice. This is the reason why the interaction of ultrashort pulses with matter can be assumed to be dominated by the electronic contribution. However the extent to which Raman modes contribute to the induced polarization is still unclear [2].

The principle of spontaneous inelastic Raman scattering is displayed in Fig. 2.5. An external pump field at frequency ω excites electrons from the ground state into a virtual level associated with an excited state. The electron decays into a vibrational level by emitting a photon at the Stokes-frequency $\omega_{\text{Stokes}} < \omega$. As the final vibration state is located energetically above the ground state energy is transferred from the external field to the material Fig. (2.5a). In the anti-Stokes process the electron is excited from the vibrational state into the virtual level by the external field and decays into the ground state. Thereby it emits an Anti-Stokes photon $\omega_{\text{Anti-Stokes}} > \omega$. In this case the pump field gains energy from the medium Fig. (2.5b). However as the electron decays into the populated ground state, the transition probability for Anti-Stokes scattering is by orders of magnitude smaller than for the Stoke process. The

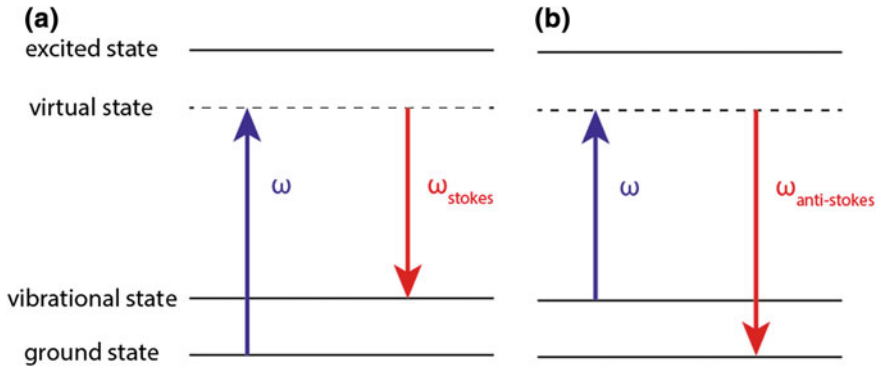


Fig. 2.5 The principle of spontaneous Raman scattering for the Stokes (Panel a) and Anti-Stokes (Panel b) process

cross section of both Raman processes can be considerably increased by exposing the energy system to radiation with ω_{Stokes} or $\omega_{Anti-Stokes}$ additionally to the pump field ω [2].

An ultrashort few-cycle optical pulse covers a bandwidth of >1 octave. Therefore it can deliver ω and ω_{Stokes} for stimulated Stokes scattering. In the Raman process energy is transferred from the high frequency part of the pulse spectrum to the long wavelength range. If the lifetime of the virtual level lies in the femtosecond range, intensity is redistributed from the center of the pulse to its red shifted tail. This phenomena is known as intrapulse Raman-scattering [2].

Absorption can be included in Eq. (2.5) under consideration of the general relation between the permittivity ϵ and n with the absorption coefficient α

$$\epsilon = \left(n + \frac{i\alpha c}{2\omega} \right)^2. \quad (2.38)$$

The time delayed Raman decay and its impact on the pulse structure can be considered by the temporal and dispersive profile of $\chi^{(3)}$ [2].

2.6 Plasma Dynamics

2.6.1 The Contribution of Free Electrons to the Refractive Index

As presented in the previous sections many nonlinear optical phenomena resulting from the oscillatory motion of electrons around their equilibrium position induced by a strong external field are well described with the perturbative approach Eq. (2.4). If the amplitude of the incident field becomes comparable to the interatomic forces E_{at} Eq. (2.36) this model breaks down as the electronic structure of the medium is altered considerably when the carriers gain sufficient energy to escape the attractive potential of their parent ion. To understand the dynamics in a plasma it is instructive to study the classical motion of a free electron with mass m_e and charge e under the influence of an external electric field $E(t) = E(t)\hat{x}$. It is well described by the harmonic oscillator model [1].

$$m_e \ddot{x} = -eE(t) \quad (2.39)$$

$$E(t) = E_0 \exp(-i\omega t) + cc. \quad (2.40)$$

$x(t)$ is the position of the electron in direction of the oscillating wave $E(t)$. This differential equation is fulfilled if the electron follows $E(t)$ instantaneously on the trajectory $x(t) = x_0 \exp(-i\omega t) + cc.$ with a maximum amplitude of $x_0 = \frac{eE_0}{m_e\omega^2}$.

On the one hand the polarization is the average over all induced dipole momenta $d(x) = -ex(t)$. For one electron $P(t) = -ex(t)$. On the other hand according to Eq. (2.3) P is given by $P(t) = \epsilon_0 \alpha(\omega) E(t)$ with the molecular polarizability $\alpha(\omega)$. ($\alpha(\omega)$ is the polarizability of a single dipole, for N charges $\chi^{(1)} = N\alpha(\omega)$) [1]. By exploiting this identity $\alpha(\omega)$ can be written as

$$\alpha(\omega) = -\frac{e^2}{m_e \epsilon_0 \omega^2} \quad (2.41)$$

The permittivity ϵ of an ensemble of free electrons with the carrier density n_e is connected to the polarizability by

$$\epsilon(\omega) = 1 + n_e \alpha(\omega) = 1 - \frac{n_e e^2}{m_e \epsilon_0 \omega^2} = 1 - \frac{\omega_p^2}{\omega^2} \quad (2.42)$$

with the plasma frequency $\omega_p^2 = \frac{n_e e^2}{m_e \epsilon_0}$.

In the absence of absorption the refractive index n is connected to $\epsilon = n^2$. For small changes $\Delta n \ll 1$ and materials with $n \approx 1$

$$n^2 = n^2 + \Delta n^2 + 2n\Delta n \approx 1 + 2n\Delta n \quad (2.43)$$

$$1 + 2n\Delta n = 1 - \frac{n_e e^2}{m_e \epsilon_0 \omega^2} \rightarrow \Delta n = -\frac{n_e e^2}{2nm_e \epsilon_0 \omega^2} < 0 \quad (2.44)$$

the free electron contribution decreases the refractive index of the material [1].

2.6.2 The Nonresonant Contribution of Bound Electrons to the Refractive Index

The model of the harmonic oscillator can also be employed to derive the modification of the refractive index by bound electrons. In this case a term representing the restoring force of the atomic potential is introduced in Eq. (2.39). ω_0 is the resonance frequency of the system.

$$m_e \ddot{x} + m_e \omega_0^2 x = -eE(t) \quad (2.45)$$

$$E(t) = E_0 \exp(-i\omega t) + cc \quad (2.46)$$

Equation (2.45) is solved by $x(t) = \frac{eE(t)}{m_e(\omega^2 - \omega_0^2)}$. Analogue to the derivation presented in the previous section, far off resonance $\omega \ll \omega_0$ the polarizability of the system is found to be

$$\alpha(\omega) = \frac{e^2}{m_e \epsilon_0 \omega_0^2}. \quad (2.47)$$

According to Eqs. (2.41) and (2.42) the bound electrons alter the refractive index by

$$\Delta n = \frac{n_e e^2}{2nm_e \epsilon_0 \omega_0^2} > 0. \quad (2.48)$$

It is apparent that the effect of bound and free carriers on the optical material density is of opposite sign. The plasma contribution Eq. (2.44) decreases the refractive index, whereas the response of bound carriers Eq. (2.48) increases its value. The absolute value of the free electron contribution exceeds the magnitude of the bound carrier effect. This difference is intuitively comprehensible as the excursion of the induced free carrier motion is bigger than the path length covered by the oscillating bound charges [1].

2.6.3 The Polarization in a Plasma

If collective particle motion, collision processes and relativistic effects are neglected the nonlinear polarization in an underdense plasma ($\omega_p < \omega$) is the sum over all the induced dipole momenta. However the contribution of the ionic cores can be neglected as they are by orders of magnitude heavier and therefore slower than the freed electrons [8]. This means that the nonlinear polarization can be written as

$$P_{NL}(t) = n_e(t)e \langle x(t) \rangle \quad (2.49)$$

where $\langle x(t) \rangle$ is the expectation value of the electronic wavefunction. The temporal derivative of $P_{NL}(t)$ is given by

$$\frac{dP_{NL}(t)}{dt} = \frac{dn_e(t)}{dt} e x_0(t) + n_e(t) e \frac{d \langle x(t) \rangle}{dt}. \quad (2.50)$$

$x_0(t) = \frac{I_p}{eE(t)}$ is the distance from the ionic core at which the electron appears after tunneling through the atomic potential suppressed by the external electric field [8]. The two terms on the right hand side of Eq. (2.50) represent the different physical processes contributing to the nonlinear polarization response. The first term describes the ionization rate at which new electrons appear outside the Coulomb barrier at position x_0 . Ionization happens with a maximum probability at the peaks of the driving field in each subcycle. The second term accounts for the acceleration of the freed carriers in the field. They acquire their maximum velocity at the zero crossings of the oscillating wave [8].

References

1. Boyd R (2008) Nonlinear optics, 3rd edn. Academic Press, Burlington
2. Agrawal G (2006) Nonlinear fiber optics, 4th edn. Academic, San Diego
3. Brabec T, Krausz F (1997) Nonlinear optical pulse propagation in the single-cycle regime. *Phys Rev Lett* 78:3282–3285
4. Brabec T, Krausz F (2000) Intense few-cycle laser fields: Frontiers of nonlinear optics. *Rev Mod Phys* 72:545–591
5. Diels J-C, Wolfgang R (2006) Ultrashort laser pulse phenomena. Academic, San Diego
6. Gaeta L, Alexander L (2000) Catastrophic collapse of ultrashort pulses. *Phys Rev Lett* 84:3582–3585
7. Reider GA (2005) Photonik, eine Einführung in die Grundlagen., zweite, überarbeitete und erweiterte Auflage Springer, Wien
8. Geissler M et al (1999) Light propagation in field-ionizing media: Extreme nonlinear optics. *Phys Rev Lett* 83:2930–2933

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