This chapter provides a brief introduction into the basic nonlinear-optical phenomena and discusses some of the most significant recent advances and breakthroughs in nonlinear optics, as well as novel applications of nonlinear-optical processes and devices.

Nonlinear optics is the area of optics that studies the interaction of light with matter in the regime where the response of the material system to the applied electromagnetic field is nonlinear in the amplitude of this field. At low light intensities, typical of non-laser sources, the properties of materials remain independent of the intensity of illumination. The superposition principle holds true in this regime, and light waves can pass through materials or be reflected from boundaries and interfaces without interacting with each other. Laser sources, on the other hand, can provide sufficiently high light intensities to modify the optical properties of materials. Light waves can then interact with each other, exchanging momentum and energy, and the superposition principle is no longer valid. This interaction of light waves can result in the generation of optical fields at new frequencies, including optical harmonics of incident radiation or sum- or difference-frequency signals.

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Although the observation of most nonlinear-optical phenomena requires laser radiation, some classes of nonlinear-optical effects were known long before the invention of the laser. The most prominent examples of such phenomena include Pockels and Kerr electrooptic effects [4.1], as well as light-induced resonant absorption saturation, described by Vavilov [4.2, 3]. It was, however, only with the advent of lasers that systematic studies of optical nonlinearities and the observation of a vast catalog of spectacular nonlinear-optical phenomena became possible.

In the first nonlinear-optical experiment of the laser era, performed by Franken et al. in 1961 [4.4], a ruby-laser radiation with a wavelength of 694.2 nm was used to generate the second harmonic in a quartz crystal at the wavelength of 347.1 nm. This seminal work was followed by the discovery of a rich diversity of nonlinear-optical effects, including sum-frequency generation, stimulated Raman scattering, self-focusing, optical rectification, four-wave mixing, and many others. While in the pioneering work by Franken the efficiency of second-harmonic generation (SHG) was on the order of $10^{-8}$, optical frequency doublers created by early 1963 provided 20%–30% efficiency of frequency conversion [4.5, 6]. The early phases of the development and the basic principles of nonlinear optics have been reviewed in the most illuminating way in the classic books by Bloembergen [4.7] and Akhmanov and Khokhlov [4.8], published in the mid 1960s.

Over the following four decades, the field of nonlinear optics has witnessed an enormous growth, leading to the observation of new physical phenomena and giving rise to novel concepts and applications. A systematic introduction into these effects along with a comprehensive overview of nonlinear-optical concepts and devices can be found in excellent textbooks by Shen [4.9], Boyd [4.1], Butcher and Cotter [4.10], Reintjes [4.11] and others. One of the most recent up-to-date reviews of the field of nonlinear optics with an in-depth discussion of the fundamental physics underlying nonlinear-optical interactions was provided by Flytzanis [4.12]. This chapter provides a brief introduction into the main nonlinear-optical phenomena and discusses some of the most significant recent advances in nonlinear optics, as well as novel applications of nonlinear-optical processes and devices.
4.1 Nonlinear Polarization and Nonlinear Susceptibilities

Nonlinear-optical effects belong to a broader class of electromagnetic phenomena described within the general framework of macroscopic Maxwell equations. The Maxwell equations not only serve to identify and classify nonlinear phenomena in terms of the relevant nonlinear-optical susceptibilities or, more generally, nonlinear terms in the induced polarization, but also govern the nonlinear-optical propagation effects. We assume the absence of extraneous charges and currents and write the set of Maxwell equations for the electric, \( \mathbf{E}(r, t) \), and magnetic, \( \mathbf{H}(r, t) \), fields in the form

\[
\nabla \times \mathbf{E} = \frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \tag{4.1}
\]
\[
\nabla \times \mathbf{B} = \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, \tag{4.2}
\]
\[
\nabla \cdot \mathbf{D} = 0, \tag{4.3}
\]
\[
\nabla \cdot \mathbf{B} = 0. \tag{4.4}
\]

Here, \( \mathbf{B} = \mathbf{H} + 4\pi \mathbf{M} \), where \( \mathbf{M} \) is the magnetic dipole polarization, \( c \) is the speed of light, and

\[
\mathbf{D} = \mathbf{E} + 4\pi \int_{-\infty}^{t} \mathbf{J}(\xi) \, d\xi. \tag{4.5}
\]

where \( \mathbf{J} \) is the induced current density. Generally, the equation of motion for charges driven by the electromagnetic field has to be solved to define the relation between the induced current \( \mathbf{J} \) and the electric and magnetic fields. For quantum systems, this task can be fulfilled by solving the Schrödinger equation. In Sect. 4.5 of this chapter, we provide an example of such a self-consistent analysis of nonlinear-optical phenomena in a model two-level system. Very often a phenomenological approach based on the introduction of field-independent or local-field-corrected nonlinear-optical susceptibilities can provide an adequate description of nonlinear-optical processes.

Formally, the current density \( \mathbf{J} \) can be represented as a series expansion in multipoles:

\[
\mathbf{J} = \frac{\partial}{\partial t} (\mathbf{P} - \nabla \cdot \mathbf{Q}) + c \left( \nabla \times \mathbf{M} \right), \tag{4.6}
\]

where \( \mathbf{P} \) and \( \mathbf{Q} \) are the electric dipole and electric quadrupole polarizations, respectively. In the electric dipole approximation, we keep only the first term on the right-hand side of (4.6). In view of (4.5), this gives the following relation between the \( \mathbf{D}, \mathbf{E} \), and \( \mathbf{P} \) vectors:

\[
\mathbf{D} = \mathbf{E} + 4\pi \mathbf{P}. \tag{4.7}
\]

We now represent the polarization \( \mathbf{P} \) as a sum

\[
\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{nl}, \tag{4.8}
\]

where \( \mathbf{P}_L \) is the part of the electric dipole polarization linear in the field amplitude and \( \mathbf{P}_{nl} \) is the nonlinear part of this polarization.

The linear polarization governs linear-optical phenomena, i.e., it corresponds to the regime where the optical properties of a medium are independent of the field intensity. The relation between \( \mathbf{P}_L \) and the electric field \( \mathbf{E} \) is given by the standard formula of linear optics:

\[
\mathbf{P}_L = \int \chi^{(1)}(t - t') \mathbf{E}(t') \, dt', \tag{4.9}
\]

where \( \chi^{(1)}(t) \) is the time-domain linear susceptibility tensor. Representing the field \( \mathbf{E} \) and polarization \( \mathbf{P}_L \) in the form of elementary monochromatic plane waves,

\[
\mathbf{E} = \mathbf{E}(\omega) \exp(i \mathbf{k} \mathbf{r} - \omega t) + c.c., \tag{4.10}
\]

and \( \mathbf{P}_L = \mathbf{P}_L(\omega) \exp(i \mathbf{k} \mathbf{r} - \omega t) + c.c., \tag{4.11} \) we take the Fourier transform of (4.9) to find

\[
\mathbf{P}_L(\omega) = \chi^{(1)}(\omega) \mathbf{E}(\omega), \tag{4.12}
\]

where

\[
\chi^{(1)}(\omega) = \int \chi^{(1)}(t) \exp(i \omega t) \, dt. \tag{4.13}
\]

In the regime of weak fields, the nonlinear part of the polarization \( \mathbf{P}_{nl} \) can be represented as a power-series expansion in the field \( \mathbf{E} \):

\[
\mathbf{P}_{nl} = \iint \chi^{(2)}(t - t_1, t - t_2) : \mathbf{E}(t_1) \mathbf{E}(t_2) \, dt_1 \, dt_2 + \iint \iint \chi^{(3)}(t - t_1, t - t_2, t - t_3) \mathbf{E}(t_1) \mathbf{E}(t_2) \mathbf{E}(t_3) \, dt_1 \, dt_2 \, dt_3 + \cdots, \tag{4.14}
\]

where \( \chi^{(2)} \) and \( \chi^{(3)} \) are the second- and third-order nonlinear susceptibilities. Representing the electric field in the form of a sum of plane monochromatic waves,

\[
\mathbf{E} = \sum_i \mathbf{E}_i(\omega_i) \exp(i \mathbf{k}_i \mathbf{r} - \omega_i t) + c.c., \tag{4.15}
\]

we take the Fourier transform of (4.14) to arrive at

\[
\mathbf{P}_{nl}(\omega) = \mathbf{P}_{2}(\omega) + \mathbf{P}_{3}(\omega) + \cdots, \tag{4.16}
\]
where

\[
\mathbf{P}^{(2)}(\omega) = \chi^{(2)}(\omega; \omega_1, \omega_2) : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2),
\]

and

\[
\mathbf{P}^{(3)}(\omega) = \chi^{(3)}(\omega; \omega_1, \omega_2, \omega_3) : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \mathbf{E}(\omega_3),
\]

\[
\chi^{(2)}(\omega; \omega_1, \omega_2) = \chi^{(2)}(\omega = \omega_1 + \omega_2) = \int \int \chi^{(2)}(t_1, t_2) \exp[i(\omega t_1 + \omega t_2)] \, dt_1 \, dt_2
\]

is the second-order nonlinear-optical susceptibility and

\[
\chi^{(3)}(\omega; \omega_1, \omega_2, \omega_3) = \chi^{(3)}(\omega = \omega_1 + \omega_2 + \omega_3)
\]

\[
= \int \int \int \chi^{(3)}(t_1, t_2, t_3) \exp[i(\omega t_1 + \omega t_2 + \omega t_3)] \, dt_1 \, dt_2 \, dt_3
\]

is the third-order nonlinear-optical susceptibility.

The second-order nonlinear polarization defined by (4.17) gives rise to three-wave mixing processes, optical rectification and linear electrooptic effect. In particular, setting \(\omega_1 = \omega_2 = \omega_0\) in (4.17) and (4.19), we arrive at \(\omega = 2\omega_0\), which corresponds to second-harmonic generation, controlled by the nonlinear susceptibility \(\chi^{(2)}(2\omega_0; \omega_0, \omega_0)\). In a more general case of three-wave mixing process with \(\omega_0 = \omega_1 \neq \omega_2\), the second-order polarization defined by (4.17) can describe sum-frequency generation (SFG) \(\omega_{sfg} = \omega_1 + \omega_2\) Fig. 4.1 or difference-frequency generation (DFG) \(\omega_{dfg} = \omega_1 - \omega_2\), governed by the nonlinear susceptibilities \(\chi^{(2)}_{SFG} = \chi^{(2)}(\omega_{sfg}; \omega_1, \omega_2)\) and \(\chi^{(2)}_{DFG} = \chi^{(2)}(\omega_{dfg}; \omega_1, -\omega_2)\), respectively.

The third-order nonlinear polarization defined by (4.18) is responsible for four-wave mixing (FWM), stimulated Raman scattering, two-photon absorption, and Kerr-effect-related phenomena, including self-phase modulation (SPM) and self-focusing. For the particular case of third-harmonic generation, we set \(\omega_0 = \omega_2 = \omega_0\) in (4.18) and (4.20) to obtain \(\omega = 3\omega_0\). This type of nonlinear-optical interaction, in accordance with (4.18) and (4.20), is controlled by the cubic susceptibility \(\chi^{(3)}(3\omega_0; \omega_0, \omega_0, \omega_0)\). A more general, frequency-nondegenerate case can correspond to a general type of an FWM process. These and other basic nonlinear-optical processes will be considered in greater details in the following sections.

### 4.2 Wave Aspects of Nonlinear Optics

In the electric dipole approximation, the Maxwell equations (4.1-4.4) yield the following equation governing the propagation of light waves in a weakly nonlinear medium:

\[
\nabla \times (\nabla \times \mathbf{E}) - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2}.
\]

The nonlinear polarization, appearing on the right-hand side of (4.21), plays the role of a driving source, inducing an electromagnetic wave with the same frequency \(\omega\) as the nonlinear polarization wave \(\mathbf{P}_{nl}(r, t)\). Dynamics of a nonlinear wave process can be then thought as a result of the interference of induced and driving (pump) waves, controlled by the dispersion of the medium.

Assuming that the fields have the form of quasi-monochromatic plane waves propagating along the \(z\)-axis, we represent the field \(\mathbf{E}\) in (4.21) by

\[
\mathbf{E}(r, t) = \text{Re} \left[ e^{iA(z, t)} \exp(ikz - \omega t) \right]
\]

and write the nonlinear polarization as

\[
\mathbf{P}_{nl}(r, t) = \text{Re} \left[ e^{iA} \mathbf{P}_{nl}(z, t) \exp(ik\Delta z - \omega t) \right],
\]

where \(k\) and \(A(z, t)\) are the wave vector and the envelope of the electric field, \(k_p\) and \(P_{nl}(z, t)\) are the wave vector and the envelope of the polarization wave.

If the envelope \(A(z, t)\) is a slowly varying function over the wavelength, \(|\partial^2 A/\partial z^2| \ll |k| \partial A/\partial z\), and \(\partial^3 \mathbf{P}_{nl}/\partial t^3 \approx -\omega^2 \mathbf{P}_{nl}\), (4.21) is reduced to [4.9]

\[
\frac{\partial A}{\partial t} + \frac{1}{u} \frac{\partial A}{\partial u} = \frac{2\pi \omega^2}{kc^2} \mathbf{P}_{nl} \exp(i\Delta k z),
\]
where \( u = (\partial k / \partial \omega)^{-1} \) is the group velocity and \( \Delta k = k_p - k \) is the wave-vector mismatch.

In the following sections, this generic equation of slowly varying envelope approximation (SVEA) will be employed to analyze the wave aspects of the basic second- and third-order nonlinear-optical phenomena.

### 4.3 Second-Order Nonlinear Processes

#### 4.3.1 Second-Harmonic Generation

In second-harmonic generation, a pump wave with a frequency of \( \omega \) generates a signal at the frequency \( 2\omega \) as it propagates through a medium with a quadratic nonlinearity (Fig. 4.1). Since all even-order nonlinear susceptibilities \( \chi^{(n)} \) vanish in centrosymmetric media, SHG can occur only in media with no inversion symmetry.

Assuming that diffraction and second-order dispersion effects are negligible, we use (4.24) for a quadratically nonlinear medium with a nonlinear SHG susceptibility \( \chi_{\text{SHG}}^{(2)} = \chi^{(2)}(2\omega, \omega, \omega) \) to write a pair of coupled equations for the slowly varying envelopes of the pump and second-harmonic fields \( A_1 = A_1(z, t) \) and \( A_2 = A_2(z, t) \):

\[
\begin{align*}
\frac{\partial A_1}{\partial z} + \frac{1}{u_1} \frac{\partial A_1}{\partial t} &= i\gamma_1 A_1^* A_2 \exp(i\Delta k z), \\
\frac{\partial A_2}{\partial z} + \frac{1}{u_2} \frac{\partial A_2}{\partial t} &= i\gamma_2 A_1^2 \exp(-i\Delta k z),
\end{align*}
\]

where

\[
\gamma_1 = \frac{2\pi\omega_0^2}{k_1 c^2} \chi^{(2)}(\omega, 2\omega, -\omega)
\]

and

\[
\gamma_2 = \frac{4\pi\omega_0^2}{k_2 c^2} \chi_{\text{SHG}}^{(2)}
\]

are the nonlinear coefficients, \( u_1 \) and \( u_2 \) are the group velocities of the pump and second-harmonic pulses, respectively, and \( \Delta k = 2k_1 - k_2 \) is the wave-vector mismatch for the SHG process.

If the difference between the group velocities of the pump and second-harmonic pulses can be neglected for a nonlinear medium with a given length and if the intensity of the pump field in the process of SHG remains much higher than the intensity of the second-harmonic field, we set \( u_1 = u_2 = u \) and \( |A_1|^2 = |A_{10}|^2 = \text{const.} \) in (4.25) and (4.26) to derive in the retarded frame of reference with \( z' = z \) and \( \eta = t - z/u \)

\[
A_2(L) = i\gamma_2 A_{10}^2 \frac{\sin\left(\frac{\Delta k L}{2}\right)}{\Delta k L} L \exp\left(i\frac{\Delta k L}{2}\right), \quad (4.29)
\]

where \( L \) is the length of the nonlinear medium.

The intensity of the second-harmonic field is then given by

\[
I_2(L) \propto \gamma_2^2 I_{10}^2 \left(\frac{\sin\left(\frac{\Delta k L}{2}\right)}{\Delta k L}\right)^2 L^2, \quad (4.30)
\]

where \( I_{10} \) is the intensity of the pump field.

Second-harmonic intensity \( I_2 \), as can be seen from (4.30) oscillates as a function of \( L \) Fig. 4.2 with a period \( L_c = \pi / |\Delta k| = \lambda_1 (4|n_1 - n_2|)^{-1} \), where \( \lambda_1 \) is the pump wavelength and \( n_1 \) and \( n_2 \) are the values of the refractive index at the frequencies of the pump field and its second harmonic, respectively. The parameter \( L_c \), defining the length of the nonlinear medium providing the maximum SHG efficiency, is referred to as the coherence length.

![Fig. 4.2 Second-harmonic intensity as a function of the length \( L \) of the nonlinear medium normalized to the coherence length \( L_c \); (dashed line) \( L_{c1} \) and (solid line) \( L_{c2} = 2L_{c1} \)](image)
Although the solution (4.29) describes the simplest regime of SHG, it is very instructive as it visualizes the significance of reducing the wave-vector mismatch $\Delta k$ for efficient SHG. Since the wave vectors $k_1$ and $k_2$ are associated with the momenta of the pump and second-harmonic fields, $p_1 = \hbar k_1$ and $p_2 = \hbar k_2$, with $\hbar$ being the Planck constant, the condition $\Delta k = 0$, known as the phase-matching condition in nonlinear optics, in fact, represents momentum conservation for the SHG process, where two photons of the pump field are required to generate a single photon of the second harmonic.

Several strategies have been developed to solve the phase-matching problem for SHG. The most practically significant solutions include the use of birefringent nonlinear crystals [4.13, 14], quasi-phase-matching in periodically poled nonlinear materials [4.15, 16] and waveguide regimes of nonlinear interactions with the phase mismatch related to the material dispersion compensated for by waveguide dispersion [4.7]. Harmonic generation in the gas phase, as demonstrated by Miles and Harris [4.17], can be phase-matched through an optimization of the content of the gas mixture. Figure 4.3 illustrates phase matching in a birefringent crystal. The circle represents the cross section of the refractive-index sphere $n_0(\omega)$ for an ordinary wave at the pump frequency $\omega$. The ellipse is the cross section of the refractive-index ellipsoid $n_{\pm}(2\omega)$ for an extraordinary wave at the frequency of the second harmonic $2\omega$. Phase matching is achieved in the direction where $n_0(\omega) = n_{\pm}(2\omega)$, corresponding to an angle $\theta_{pm}$ with respect to the optical axis $c$ of the crystal in Fig. 4.3.

When the phase-matching condition $\Delta k = 0$ is satisfied, (4.29) and (4.30) predict a quadratic growth of the second-harmonic intensity as a function of the length $L$ of the nonlinear medium. This scaling law holds true, however, only as long as the second-harmonic intensity remains much less than the pump intensity. As $|A_2|$ becomes comparable with $|A_1|$, depletion of the pump field has to be taken into consideration. To this end, we introduce the real amplitudes $\rho_j$ and phases $\varphi_j$ of the pump and second-harmonic fields, $A_j = \rho_j \exp(\varphi_j)$, with $j = 1, 2$. Then, assuming that $u_1 = u_2 = u$ and $\gamma_1 = \gamma_2 = \gamma$, we derive from (4.25) and (4.26)

$$\rho_1(\eta, z) = \rho_{10}(\eta) \sech[\gamma \rho_{10}(\eta) z], \quad (4.31)$$

$$\rho_2(\eta, z) = \rho_{10}(\eta) \tanh[\gamma \rho_{10}(\eta) z]. \quad (4.32)$$

The solutions (4.31) and (4.32) show that the entire energy of the pump field in the phase-matching regime can be transferred to the second harmonic. As the pump field becomes depleted Fig. 4.4, the growth of the second-harmonic field saturates.

Effects related to the group-velocity mismatch become significant when the length of the nonlinear medium $L$ exceeds the length $L_g = \tau_1 / [u_2^{-1} - u_1^{-1}]$, where $\tau_1$ is the pulse width of the pump field. The length $L_g$ characterizes the walk-off between the pump and second-harmonic pulses caused by the group-velocity mismatch. In this nonstationary regime of SHG, the amplitude of the second harmonic in the constant-pump-
field approximation is given by
\[ A_2(z, t) = i\gamma_3 \int_0^Z A_{10}^* \left[ t - z/u_2 + i \left( \frac{\pi}{163} \right) \right] \times \exp \left(-i\Delta k\xi\right) d\xi . \]  
(4.33)

Group-velocity mismatch may lead to a considerable increase in the pulse width of the second harmonic \( \tau_2 \). For \( L \gg L_0 \), the second harmonic pulse width, \( \tau_2 \approx |u_2^{-1} - u^{-1}|L \), scales linearly with the length of the nonlinear medium and is independent of the pump pulse width.

### 4.3.2 Sum- and Difference-Frequency Generation and Parametric Amplification

In sum-frequency generation Fig. 4.1, two laser fields with frequencies \( \omega_1 \) and \( \omega_2 \) generate a nonlinear signal at the frequency \( \omega_3 = \omega_1 + \omega_2 \) in a quadratically nonlinear medium with a nonlinear susceptibility \( \chi_{\text{SFG}}^{(2)} = \chi^{(2)}(\omega_3; \omega_1, \omega_2) \). In the first order of dispersion theory, the coupled equations for slowly varying envelopes of the laser fields \( A_1 = A_1(z, t) \) and \( A_2 = A_2(z, t) \) and the nonlinear signal \( A_3 = A_3(z, t) \) are written as
\[
\begin{align*}
\frac{\partial A_1}{\partial z} + \frac{1}{u_1} \frac{\partial A_1}{\partial t} &= i\gamma_1 A_3 A_1^* \exp(\text{i}\Delta kz) , \\
\frac{\partial A_2}{\partial z} + \frac{1}{u_2} \frac{\partial A_2}{\partial t} &= i\gamma_2 A_3 A_2^* \exp(\text{i}\Delta kz) , \\
\frac{\partial A_3}{\partial z} + \frac{1}{u_3} \frac{\partial A_3}{\partial t} &= i\gamma_3 A_1 A_2^* \exp(-\text{i}\Delta kz) ,
\end{align*}
\]
(4.34)
(4.35)
(4.36)

where
\[
\begin{align*}
\gamma_1 &= \frac{2\pi\omega_2}{k_1 c^2} \chi^{(2)}(\omega_1; \omega_3, -\omega_2) , \\
\gamma_2 &= \frac{2\pi\omega_3}{k_2 c^2} \chi^{(2)}(\omega_2; \omega_3, -\omega_1) , \\
\gamma_3 &= \frac{2\pi\omega_1}{k_3 c^2} \chi_{\text{SFG}}^{(2)} ,
\end{align*}
\]
(4.37)
(4.38)
(4.39)

are the nonlinear coefficients, \( u_1, u_2, \) and \( u_3 \) and \( k_1, k_2, \) and \( k_3 \) are the group velocities and the wave vectors of the fields with frequencies \( \omega_1, \omega_2, \) and \( \omega_3, \) respectively, and \( \Delta k = k_1 + k_2 - k_3 \) is the wave-vector mismatch for the SFG process.

As long as the intensity of the sum-frequency field remains much less than the intensities of the laser fields, the amplitudes of the laser fields can be assumed to be given functions of \( t, A_1(z, t) = A_{10}(t) \) and \( A_2(z, t) = A_{20}(t) \), and the solution of (4.36) yields
\[
\begin{align*}
A_3(z, t) &= i\gamma_3 \int_0^Z A_{10}^* \left[ t - z/u_3 - i \left( \frac{\pi}{163} \right) \right] \times A_{20} \left[ t - z/u_3 + i \left( \frac{\pi}{163} \right) \right] \times \exp(-\text{i}\Delta k\xi) d\xi .
\end{align*}
\]
(4.40)

The efficiency of frequency conversion, as can be seen from (4.40) is controlled by the group delays \( \Delta_{21} \approx |u_2^{-1} - u^{-1}|L, \Delta_{31} \approx |u_3^{-1} - u^{-1}|L, \) and \( \Delta_{32} \approx |u_3^{-1} - u_2^{-1}|L \) between the pulses involved in the SFG process. In particular, the laser fields cease to interact with each other when the group delay \( \Delta_{21} \) starts to exceed the pulse width of the faster laser pulse.

In difference-frequency generation (DFG), two input fields with frequencies \( \omega_1 \) and \( \omega_2 \) generate a nonlinear signal at the frequency \( \omega_3 = \omega_1 - \omega_2 \). This process is of considerable practical significance as it can give rise to intense coherent radiation in the infrared range. In the limiting case of \( \omega_1 \approx \omega_2 \), this type of nonlinear-optical interaction corresponds to optical rectification, which has been intensely used over the past two decades for the generation of terahertz radiation.

If the field at the frequency \( \omega_1 \) is strong and remains undepleted in the process of nonlinear-optical interaction, \( A_1(z, t) = A_{10}(t) \), the set of coupled equations governing the amplitudes of the remaining two fields in the stationary regime is written as
\[
\begin{align*}
\frac{\partial A_2}{\partial z} + \frac{1}{u_2} \frac{\partial A_2}{\partial t} &= i\gamma_2 A_1 A_2^* \exp(\text{i}\Delta k z) , \\
\frac{\partial A_3}{\partial z} + \frac{1}{u_3} \frac{\partial A_3}{\partial t} &= i\gamma_3 A_1 A_2^* \exp(-\text{i}\Delta k z) ,
\end{align*}
\]
(4.41)
(4.42)

where,
\[
\begin{align*}
\gamma_2 &= \frac{2\pi\omega_3}{k_2 c^2} \chi^{(2)}(\omega_2; \omega_1, -\omega_3) , \\
\gamma_3 &= \frac{2\pi\omega_1}{k_3 c^2} \chi_{\text{SFG}}^{(2)} .
\end{align*}
\]
(4.43)
(4.44)

are the nonlinear coefficient and \( \Delta k = k_1 - k_2 - k_3 \) is the wave-vector mismatch for the DFG process.

With no signal at \( \omega_3 \) applied at the input of the nonlinear medium, \( A_3(0, t) = 0 \), the solution to (4.41) and (4.42) in the stationary regime is given by \[4.12\]
\[
A_2(z) = A_{20}(0) \left[ \cosh(\kappa z) + \frac{\Delta k}{2\kappa} \sinh(\kappa z) \right] ,
\]
(4.45)
\[
A_3(z) = i A_{20}(0) \sinh(\kappa z) .
\]
(4.46)
where
\[ \kappa^2 = 4\gamma_2 A_2^4 \left| A_1 \right|^2 - (\Delta k)^2. \]

Away from the phase-matching condition, the amplification of a weak signal is achieved only when the intensity of the pump field exceeds a threshold,
\[ I_1 > I_{th} = \frac{n_1 n_2 n_3 c^3 (\Delta k)^2}{32\pi^3 |\chi^{(2)}_{DFG}|^2 \alpha_2 \omega_3}, \]

where we took
\[ \chi^{(2)}(\omega_2; \omega_1, -\omega_3) \approx \chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \chi^{(2)}_{DFG}. \]

Above this threshold, the growth in the intensity \( I_2 \) of a weak input signal is governed by
\[ I_2 (z) = I_2 (0) \left( \frac{2\gamma_2 A_2^4 \left| A_1 \right|^2}{\kappa^2} \sin^2 (kz) + 1 \right). \]

This type of three-wave mixing is often referred to as optical parametric amplification. A weak input field, referred to as the signal field (the field with the amplitude \( A_2 \) in our case), becomes amplified in this type of process through a nonlinear interaction with a powerful pump field (the undepleted field with the amplitude \( A_1 \) in the case considered here). In such a scheme of optical parametric amplification, the third field (the field with the amplitude \( A_3 \)) is called the idler field.

We now consider the regime of optical parametric amplification \( \omega_1 = \omega_2 + \omega_3 \) where the pump, signal and idler pulses are matched in their wave vectors and group velocities. Introducing the real amplitudes \( \rho_j \) and phases \( \varphi_j \) of the pump, signal, and idler fields, \( A_j = \rho_j \exp (i\varphi_j) \), where \( j = 1, 2, 3 \), assuming that \( \gamma_2 = \gamma_3 = \gamma \) in (4.35) and (4.36), \( A_1 (z, t) = A_{10}(t) \) and \( A_3(0, t) = 0 \), we write the solution for the amplitude of the signal field as [4.18]
\[ A_2 (\eta, z) = A_{20} (\eta) \cosh \left[ \gamma \varphi_{10} (\eta) z \right]. \]

The idler field then builds up in accordance with
\[ A_3 (\eta, z) = A_{30}^* (\eta) \exp [i\varphi_{10} (\eta)] \sinh \left[ \gamma \varphi_{10} (\eta) z \right]. \]

As can be seen from (4.50), optical parametric amplification preserves the phase of the signal pulse. This property of optical parametric amplification lies at the heart of the principle of optical parametric chirped-pulse amplification [4.19], allowing ultrashort laser pulses to be amplified to relativistic intensities. It also suggests a method of efficient frequency conversion of few-cycle field waveforms without changing the phase offset between their carrier frequency and temporal envelope, making few-cycle laser pulses a powerful tool for the investigation of ultrafast electronic dynamics in atomic and molecular systems.

In the nonstationary regime of optical parametric amplification, when the pump, signal, and idler fields propagate with different group velocities, useful and important qualitative insights into the phase relations between the pump, signal, and idler pulses can be gained from energy and momentum conservation, \( \omega_1 = \omega_2 + \omega_3 \) and \( k_1 = k_2 + k_3 \). These equalities dictate the following relations between the frequency deviations \( \delta \omega_j \) in the pump, signal, and idler fields \( (j = 1, 2, 3) \):
\[ \delta \omega_1 = \delta \omega_2 + \delta \omega_3 \]
and
\[ \delta \omega_1 / \omega_1 = \delta \omega_2 / \omega_2 + \delta \omega_3 / \omega_3. \]

In view of (4.52) and (4.53), we find
\[ \delta \omega_2 = q_2 \delta \omega_1 \]
and
\[ \delta \omega_3 = q_3 \delta \omega_1, \]
where \( q_2 = (u_1^{m-1} - u_3^{m-1}) / (u_2^{m-1} - u_3^{m-1}) \), \( q_3 = 1 - q_2 \).

In the case of a linearly chirped pump, \( \varphi_{1}(t) = \alpha_1 t^2 / 2 \), the phases of the signal and idler pulses are given by \( \varphi_{m}(t) = \alpha_m L^2 / 2 \), where \( \alpha_m = \alpha_m \omega_1 / m = 2, 3 \). With \( q_m > 1 \), the chirp of the signal and idler pulses can thus considerably exceed the chirp of the pump field.

### 4.4 Third-Order Nonlinear Processes

Optical nonlinearity of the third order is a universal property, found in any material regardless of its spatial symmetry. This nonlinearity is the lowest-order nonvanishing nonlinearity for a broad class of centrosymmetric materials, where all the even-order nonlinear susceptibilities are identically equal to zero for symmetry reasons. Third-order nonlinear processes include a vast variety of four-wave mixing processes, which are extensively used for frequency conversion of laser radiation and as powerful methods of nonlinear spectroscopy. Frequency-degenerate, Kerr-effect-type phenomena constitute another important class of third-
order nonlinear processes. Such effects lie at the heart of optical compressors, mode-locked femtosecond lasers, and numerous photonic devices, where one laser pulse is used to switch, modulate, or gate another laser pulse. In this section, we provide a brief overview of the main third-order nonlinear-optical phenomena and discuss some of their practical applications.

### 4.4.1 Self-Phase Modulation

The third-order nonlinearity gives rise to an intensity-dependent additive to the refractive index:

\[
n = n_0 + n_2 I(t) \tag{4.56}
\]

where \(n_0\) is the refractive index of the medium in the absence of light field, \(n_2 = (2\pi/n_0)^2 \chi^{(3)}(\omega; \omega, -\omega)\) is the nonlinear refractive index, \(\chi^{(3)}(\omega; \omega, -\omega)\) is the third-order nonlinear-optical susceptibility, referred to as the Kerr-type nonlinear susceptibility, and \(I(t)\) is the intensity of laser radiation. Then, the nonlinear (intensity-dependent) phase shift of a pulse at a distance \(L\) is given by

\[
\Phi(t) = \frac{\omega}{c} n_2 L I(t) \tag{4.57}
\]

Due to the time dependence of the radiation intensity within the light pulse, the nonlinear phase shift is also time-dependent, giving rise to a generally time-dependent frequency deviation:

\[
\Delta \omega(t) = \frac{\omega}{c} n_2 L \frac{\partial I}{\partial t} \tag{4.58}
\]

The resulting spectral broadening of the pulse can be estimated in the following way:

\[
\Delta \omega = \frac{\omega}{c} n_2 L I_0 \tau \tag{4.59}
\]

where \(I_0\) is the peak intensity of the light pulse and \(\tau\) is the pulse duration.

The first-order dispersion-theory equation for the slowly varying envelope \(A(t, z)\) of a laser pulse propagating in a medium with a Kerr-type nonlinearity is written as [4,9]

\[
\frac{\partial A}{\partial z} + \frac{1}{a} \frac{\partial A}{\partial \tau} = i \gamma |A|^2 A \tag{4.60}
\]

where \(a\) is the group velocity of the laser pulse and

\[
\gamma = \frac{3\pi n_0^2}{2} \chi^{(3)}(\omega; \omega, -\omega, \omega) \tag{4.61}
\]

is the nonlinear coefficient.

In the retarded frame of reference, \(z' = z\) and \(\eta = t - z/\tau\), the solution to (4.60) is written as

\[
A(\eta, z) = A_0(\eta) \exp \left[ i \gamma |A_0(\eta)|^2 z \right] \tag{4.62}
\]

where \(A_0(\eta)\) is the initial field envelope.

Since the group-velocity dispersion was not included in (4.60), the shape of the pulse envelope remains unchanged as the pulse propagates through the nonlinear medium. The intensity-dependent change in the refractive index gives rise to a nonlinear phase shift

\[
\varphi_{nl}(\eta, z) = \gamma_{\text{SPM}} I_0(\eta) z \tag{4.63}
\]

where \(\gamma_{\text{SPM}} = 2\pi n_2/\lambda\) and \(I_0(\eta)\) is the initial intensity envelope.

The deviation of the instantaneous frequency of the pulse is given by

\[
\delta \omega(\eta, z) = -\frac{\partial \varphi_{nl}(\eta, z)}{\partial \eta} = -\gamma_{\text{SPM}} \frac{\partial I_0(\eta)}{\partial \eta} z \tag{4.64}
\]

A quadratic approximation of the pulse envelope,

\[
I_0(\eta) \approx I_0(0) \left( 1 - \frac{\eta^2}{\gamma_{\text{SPM}}^2} \right) \tag{4.65}
\]

where \(\tau_0\) is the pulse width, which is valid around the maximum of the laser pulse, gives a linear chirp of the

![Fig. 4.5](image_url) Self-phase-modulation-induced spectral broadening of a laser pulse with a central wavelength of 1.03 μm (an input spectrum is shown by curve 1) in a fused-silica optical fiber with \(n_2 = 3.2 \times 10^{-16}\) cm³/W: \(\gamma_{\text{SPM}} I_0(0)\tau = 1.25\) (curve 2), 2.50 (curve 3), and 6.25 (curve 4).
pulse
\[ \delta \omega (\eta, z) \approx -2 \gamma_{\text{SPM}} \frac{l_0(0)}{\tau_0} \eta z. \]  

The spectrum of a self-phase-modulated pulse is given by
\[ S(\omega) = \left| \int_0^\infty I(\eta) \exp[i \omega \eta + i \varphi_{\text{NL}}(\eta)] \, d\eta \right|^2. \]

Figure 4.5 illustrates SPM-induced spectral broadening of a short laser pulse with a central wavelength of 1.03 μm, typical of ytterbium fiber lasers, in a fused-silica optical fiber with nonlinear refractive index \( n_2 = 3.2 \times 10^{-16} \text{ cm}^2/\text{W} \).

Thus, self-phase modulation results in spectral broadening of a light pulse propagating through a hollow fiber. This effect allows compression of light pulses through the compensation of the phase shift acquired by the pulse in a hollow fiber. Compensation of a linear chirp, corresponding to a linear time dependence of the instantaneous frequency, is straightforward from the technical point of view. Such a chirp arises around the maximum of a light pulse, where the temporal pulse envelope can be approximated with a quadratic function of time [see (4.65, 4.66)].

It is physically instructive to consider the compression of chirped light pulses in the time domain. Since the frequency of a chirped pulse changes from its leading edge to its trailing edge, dispersion of our compressor should be designed in such a way as to slower down the leading edge of the pulse with respect to the trailing edge of the pulse. In other words, the group velocities for the frequencies propagating with the leading edge of the pulse should be lower than the group velocities for the frequencies propagating with the trailing edge of the pulse. This can be achieved by designing a dispersive element with the required sign of dispersion and appropriate dispersion relation. Systems of diffraction gratings and, recently, multilayer chirped mirrors [4.20] are now widely used for the purposes of pulse compression. In certain regimes of pulse propagation, self-phase modulation and pulse compression may take place in the same medium.

### 4.4.2 Temporal Solitons

The nonlinear phase shift acquired by a laser pulse propagating through a medium with a Kerr nonlinearity can be balanced by group-velocity dispersion, giving rise to pulses propagating through the nonlinear dispersive medium with an invariant or periodically varying shape: optical solitons.

Optical solitons is a special class of solutions to the nonlinear Schrödinger equation (NLSE)
\[ i \frac{\partial q}{\partial \xi} + \frac{1}{2} \frac{\partial^2 q}{\partial \tau^2} + |q|^2 q = 0. \]

The NLSE can describe the evolution of optical wave packets including the dispersion \( \beta(\omega) \) of optical waves in a bulk material or in a waveguide structure through the power series expansion
\[ \beta (\omega) \approx \beta (\omega_0) + \frac{1}{u} (\omega - \omega_0) \]
\[ + \frac{1}{2} \beta_2 (\omega - \omega_0)^2 + \ldots, \]

where \( \omega_0 \) is the central frequency of the wave packet, \( u = (\partial \beta / \partial \omega)|_{\omega=\omega_0} \) is the group velocity, and \( \beta_2 = \partial^2 \beta / \partial \omega^2 |_{\omega=\omega_0} \). Thus, with the NLSE (4.68) projected on laser pulses propagating in a nonlinear medium, \( q \) is understood as the normalized pulse envelope, \( q = A/(P_0)^{1/2} \), with \( \xi \) being the normalized propagation coordinate, \( \xi = z/L_4 \), \( L_4 = \tau_0^2/|\beta_2| \) being the dispersion length, \( P_0 \) and \( \tau_0 \) defined as the pulse width and the pulse peak power, respectively, and \( \tau = (t - z/u)/\tau_0 \).

The canonical form of the fundamental soliton solution to (4.68) is [20]
\[ q (\xi, \tau) = \text{sech} (\tau) \exp \left( i \frac{\xi}{2} \right). \]

The radiation peak power required to support such a soliton is given by
\[ P_0 = |\beta_2|/(\gamma \tau_0^2). \]

Solitons retain their stable shape as long as their spectrum lies away from the spectrum of dispersive waves that can propagate in the medium. High-order dispersion perturbs solitons, inducing Cherenkov-type wave-matching resonances between solitons and dispersive waves [4.21, 22]. Under these conditions solitons tend to lose a part of their energy in the form of blueshifted dispersive-wave emission. For low pump-field powers, the generic wave-matching condition for such soliton–dispersive wave resonances is written [4.22]
\[ \Omega = 1/2e, \] where \( \Omega \) is the frequency difference between the soliton and the resonant dispersive wave and \( e \) is the parameter controlling the smallness of perturbation of the nonlinear Schrödinger equation, which can be represented as \( e = |\beta_3/6\beta_2| \) for photonic-crystal fibers.
(PCFs) with second-order dispersion \( \beta_2 = \frac{\partial^2 \beta}{\partial \omega^2} \) and third-order dispersion \( \beta_3 = \frac{\partial^3 \beta}{\partial \omega^3} \). This dispersive-wave emission of solitons is an important part of supercontinuum generation in nonlinear optical fibers, including photonic-crystal fibers.

4.4.3 Cross-Phase Modulation

Cross-phase modulation (XPM) is a result of nonlinear-optical interaction of at least two physically distinguishable light pulses (i.e., pulses with different frequencies, polarizations, mode structures, etc.) related to the phase modulation of one of the pulses (a probe pulse) due to the change in the refractive index of the medium induced by another pulse (a pump pulse).

The cross-action of a pump pulse with a frequency \( \omega_1 \) on a probe pulse with a frequency \( \omega_2 \) gives rise to a phase shift of the probe pulse, which can be written as [4.23],

\[
\Phi_{XPM}(\eta, z) = \frac{3\pi\omega_2^2}{c^2k_2} \chi^{(3)}(\omega_1, \omega_2, -\omega_1, -\omega_2) \times \int_0^z \left[ A_\eta \left( \eta - \frac{\xi}{\sigma}, 0 \right) \right]^2 d\xi,
\]

where \( \chi^{(3)}(\omega_1, \omega_2, -\omega_1, -\omega_2) \) is the third-order nonlinear-optical susceptibility of the medium; \( 1/\sigma = 11/u_1 - 1/u_2; u_1 \) and \( u_2 \) are the group velocities of the pump and probe pulses, respectively; and \( k_2 \) is the wave number of the pump pulse. Taking the time derivative of the nonlinear phase shift, we arrive at the following expression for the frequency deviation of the probe pulse

\[
\delta \omega_{XPM}(\eta, z) = -\frac{3\pi\omega_2^2}{c^2k_2} \chi^{(3)}(\omega_1, \omega_2, -\omega_1, -\omega_2) \times \sigma \left[ \left| A_\eta(\eta, 0) \right|^2 - \left| A_\eta \left( \eta - \frac{\xi}{\sigma}, 0 \right) \right|^2 \right].
\]

Similarly to self-phase modulation, cross-phase modulation can be employed for pulse compression. The dependence of the chirp of the probe pulse on the pump pulse intensity can be used to control the parameters of ultrashort pulses [4.24]. Cross-phase modulation also opens the ways to study the dynamics of ultrafast nonlinear processes, including multiphoton ionization, and to characterize ultrashort light pulses through phase measurements on a short probe pulse [4.25].

4.4.4 Self-Focusing

Self-focusing is a spatial counterpart of self-phase modulation. While SPM originates from the time-dependent change in the refractive index induced by a laser pulse with an intensity envelope \( I(t) \) varying in time, self-focusing is related to a nonlinear lens induced by a laser beam with a spatially nonuniform intensity distribution \( I(r) \). Given a transverse intensity profile \( I(r) \), the nonlinear additive to the refractive index is written as

\[
n(\mathbf{r}) = n_0 + n_2 I(\mathbf{r}) \cdot \frac{\partial}{\partial \omega} \ln I(\mathbf{r}).
\]

If the field intensity peaks at the center of the beam at \( r = 0 \), the nonlinear change in the refractive index also reaches its maximum at \( r = 0 \), yielding a focusing or defocusing lens, depending on the sign of \( n_2 \).

The stationary regime of self-focusing is governed by [4.9]

\[
2i k \frac{\partial A}{\partial z} + \Delta_{\perp} A = -2k^2 \frac{\Delta n}{n_0} A,
\]

where \( \Delta n = n_2 I = n_2 |E|^2 \), \( \Delta_{\perp} \) is the transverse part of the Laplacian.

We consider a Gaussian beam and assume that this beam retains its profile as it propagates through the nonlinear medium,

\[
A(\mathbf{r}, z) = A_0 e^{-r^2/2a_0^2} \exp \left[ -\frac{r^2}{2a_0^2 f^2(z)} + i\psi(z) \right],
\]

where \( a_0 \) is the initial beam size, \( f(z) \) characterizes the evolution of the beam size along the propagation coordinate \( z \) [\( f(0) = 1 \)], and the function \( \psi(z) \) describes the spatial phase modulation of the field.

In the paraxial approximation, \( r \ll a_0 f(z) \), (4.75) and (4.76) give [4.18]

\[
\frac{\partial^2 f}{\partial z^2} = \frac{L_{diff}^{-2} - L_{nl}^{-2}}{f^2(z)},
\]

where \( L_{diff} = 2\pi a_0^2/\lambda \) and \( L_{nl} = a_0 |2n_0/(n_2 |E|^2)|^{1/2} \) are the characteristic diffraction and nonlinear lengths, respectively.

Solving (4.77), we arrive at

\[
f^2(z) = 1 + \left( \frac{z}{L_{diff}} \right)^2 \left( 1 - \frac{P_0}{P_{crit}} \right),
\]

where \( P_0 \) is the total power of the laser beam and

\[
P_{crit} = \frac{c\lambda^2}{16\pi^2 n_2}
\]

is the critical power of self-focusing. The focal length of the nonlinear lens is given by

\[
L_{diff} = \left( \frac{P_0}{P_{crit}} - 1 \right)^{1/2}.
\]
With this tool it should be possible to precisely control the motion of energetic electron wave packets around atoms on attosecond timescales just as the motion of nuclear wave packets in molecules can be controlled within a few femtoseconds. The single sub-femtosecond electron bunches and (XUV/X-ray) photon bursts that arise from the recently gained ability to control electron wave packets will enable the scientific community to excite and probe atomic dynamics on atomics time scales.

### 4.12.5 Some Recent Developments

Attosecond science is rapidly evolving [4.344] and the last three years have seen important progress both in the performances of femtosecond and attosecond light pulses based on high-order harmonic generation in gases and in their applications in different scientific areas.

Harmonic sources now reach pulse energies in the microjoule range [4.345] and their spectra extend to energies of several keV (though with a lower throughput) [4.346]. Applications are flourishing, going from the determination of vibration frequencies in molecules [4.347] to microscopy [4.348, 349] and even recently to seeding of X-ray laser plasmas [4.350] and possibly, in the future, free-electron lasers.

Attosecond pulses have been studied in more details with different techniques [4.351, 352] and the method to isolate a single pulse refined [4.353, 354]. Their time-frequency characteristics have been mapped out [4.355–357] and ways to control both the individual pulses [4.358] and the train structure [4.359] have been developed. The shortest isolated pulse produced to date is 130 as, using the polarization gating technique for the temporal confinement [4.360, 361]. Applications now include characterization of electromagnetic fields [4.362], tomography of molecular orbitals [4.363], molecular dynamics studies of simple molecules [4.364, 365], dynamical studies and interferometric measurements of electron wavepackets [4.366, 367], and time-resolved inner-shell spectroscopy in atoms and solids [4.368].

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