Chapter 35
Nonlinear Optics

The polarization of a dielectric medium depends nonlinearly on the amplitude of the electromagnetic field. Nonlinear dielectric media are suitable for frequency conversion of radiation. Nonlinear media can be crystals, glasses, liquids or vapors. We discuss: frequency multiplication; difference frequency generation; parametric oscillation; four wave mixing; stimulated Raman scattering. In connection with four-wave mixing, we show how the frequencies of a frequency comb can be determined.

We will present only a very narrow view on the fascinating field of Nonlinear Optics. Our main aspect concerns the question: how can we convert coherent radiation of one frequency to coherent radiation of other frequencies?

35.1 Optics and Nonlinear Optics

In Maxwell's theory, the matter equations describe the electric properties of a dielectric medium are expressed by the relation between the dielectric polarization $\boldsymbol{P}$ of the medium and the electric field $\boldsymbol{E}$ in the medium,

$$\boldsymbol{P} = \boldsymbol{P}(\boldsymbol{E}). \tag{35.1}$$

In *Optics* (Linear Optics), the relation is

$$\boldsymbol{P} = \varepsilon_0 \chi^{(1)} \boldsymbol{E}, \tag{35.2}$$

where $\chi^{(1)}$ is the (complex) dielectric susceptibility. The susceptibility of an optically isotropic medium is a scalar. It is a tensor if a medium is anisotropic. The polarization has the same frequency as the electromagnetic field. The susceptibility $\chi^{(1)}(\omega)$ characterizes optical properties of a material. The study of $\chi^{(1)}(\omega)$ leads to an understanding of basic microscopic properties of matter.

K.F. Renk, *Basics of Laser Physics*, Graduate Texts in Physics,
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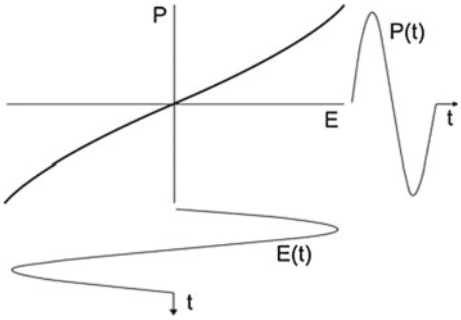

Fig. 35.1 Nonlinear polarization

The basis of *Nonlinear Optics* is the nonlinearity of the polarization at large amplitudes of the electric field (Fig. 35.1). We characterize the polarization by the relation

$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 + \ldots \tag{35.3}$$

We make use of two simplifications. We neglect the vector character of $\boldsymbol{E}$ and $\boldsymbol{P}$ as well as the tensor properties of $\chi^{(1)}$, $\chi^{(2)}$, $\chi^{(3)}$ etc. We assume that the field is spatially homogeneous in the direction of a light beam—we ignore that the field changes the phase during propagation. Thus, we neglect phase effects, which can be of great importance. Nevertheless, the simplified representation of the relation between polarization and field allows for developing an understanding of the principle of generation of radiation by means of the nonlinear polarization. Nonlinear polarization is applicable, for instance, to convert monochromatic radiation to radiation at other frequencies.

The electric field that causes a polarization can consist of fields of different frequencies. We can write, instead of (35.3),

$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E_1 E_2 + \varepsilon_0 \chi^{(3)} E_1 E_2 E_3 + \ldots, \tag{35.4}$$

where $E_1, E_2, \ldots$ are fields of different frequencies and where $E = E_1 + E_2 + \ldots$ is the sum of the fields.

35.2 Origin of Nonlinear Polarization

At which amplitude of an electromagnetic field do we expect a nonlinear polarization? We consider a hydrogen atom in a static electric field. We describe the H atom by Bohr's atomic model. In a distance of the Bohr radius ($a_0 = 0.053\,\text{nm}$) to the nucleus (proton), an electron experiences the field strength

$$|E_{\text{at}}| = \frac{e}{4\pi\varepsilon_0 a_0^2} \approx 10^{11}\ \text{V/m}. \tag{35.5}$$

A static external field polarizes an H atom. The center of the positive charge and the center of the negative charge do not coincide with each other. Therefore, the H atom represents an electric dipole. The dipole moment increases with the field strength. At large field strength, the dipole moment depends nonlinearly on the field strength. The nonlinearity is extremely large when the field strength is of the order of the internal field produced by the proton at the site of the electron.

A strong high frequency field with a sinusoidal time dependence applied to a nonlinear medium leads to a time dependent polarization

$$P(t) = \varepsilon_0 \chi^{(1)} E(t) + \varepsilon_0 \chi^{(2)} E^2(t) + \ldots . \tag{35.6}$$

We describe the nonlinear polarization in the classical model of an atom (Sect. 4.8). Under the action of a strong electric field, an electron oscillates unharmonically—leading to a nonlinear dipole moment. Accordingly, the polarization of a medium depends nonlinearly on the amplitude of the electric field. The time-dependent polarization, which depends nonlinearly on the electric field, contains frequency components not only at the driving frequency but also at other frequencies. Therefore, the nonlinear polarization is the source of electromagnetic radiation at frequencies that differ from the driving frequency.

Atoms, ions, or molecules in gases, liquids and solids show nonlinear polarization. The strength of the nonlinearity strongly depends on the specific material. Especially large nonlinear susceptibilities are known for a variety of crystals (e.g., KDP, $LiNbO_3$).

We will now discuss applications that are based on the nonlinear polarization.

35.3 Optical Frequency Doubler

A frequency doubler (Fig. 35.2a) converts radiation of frequency ν to radiation at the doubled frequency (2ν). A filter blocks the radiation of frequency ν that is not converted. The frequency doubling makes use of the quadratic term of the polarization,

$$P = \varepsilon_0 \chi^{(1)} E(\omega) + \varepsilon_0 \chi^{(2)} E^2(\omega). \tag{35.7}$$

An electric field

$$E = A \cos \omega t \tag{35.8}$$

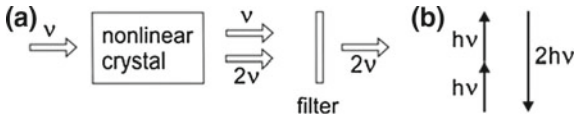

Fig. 35.2 Optical frequency doubler. **a** Principle. **b** Elementary process

causes a polarization

$$P = \varepsilon_0 \chi^{(1)} A \cos \omega t + \varepsilon_0 \chi^{(2)} A^2 \cos^2 \omega t. \tag{35.9}$$

It follows, with $\cos^2 \omega t = \frac{1}{2} + \frac{1}{2} \cos(2\omega t)$, that

$$P = \varepsilon_0 \chi^{(1)} A \cos \omega t + \frac{1}{2} \varepsilon_0 \chi^{(2)} A^2 + \frac{1}{2} \varepsilon_0 \chi^{(2)} A^2 \cos 2\omega t. \tag{35.10}$$

We obtain a polarization at the frequency 2ω that is the source of radiation at the frequency 2ω. The additional term corresponds to a static polarization (optical rectification). In an elementary process of frequency doubling (Fig. 30.2b), two photons of the quantum energy $h\nu$ are annihilated and a photon of energy $2h\nu$ is created.

Examples

- A frequency doubler converts infrared radiation to green radiation. As nonlinear crystals, $LiNbO_3$ or KDP are suitable: the conversion efficiency can reach 40%.
- In a titanium–sapphire laser, a frequency doubler located within the laser resonator produces frequency-doubled radiation in the violet and green (according to the tunability of the laser).
- Frequency doubling of green or blue radiation leads to UV radiation.

35.4 Difference Frequency Generator

In a difference frequency generator (Fig. 35.3a), two sinusoidal fields of different frequencies (ω_1 and ω_2; with $\omega_1 > \omega_2$) produce a nonlinear polarization in a nonlinear crystal. The nonlinear polarization is the source of an electromagnetic field at the difference frequency (beat frequency)

$$\omega_3 = \omega_1 - \omega_2. \tag{35.11}$$

An electric field of frequency ω_1

$$E_1 = A_1 \cos \omega_1 t \tag{35.12}$$

and another field

$$E_2 = A_2 \cos \omega_2 t \tag{35.13}$$

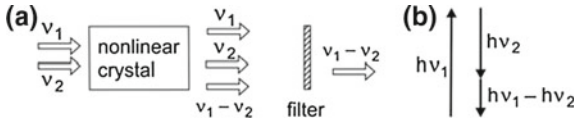

Fig. 35.3 Difference frequency generator. **a** Principle. **b** Elementary process

superimposed to each other lead to the field

$$E = E_1 + E_2. \tag{35.14}$$

This produces the polarization

$$\begin{aligned} P = {} & \varepsilon_0 \chi^{(1)} (A_1 \cos \omega_1 t + A_2 \cos \omega_2 t) \\ & + \varepsilon_0 \chi^{(2)} (A_1 \cos \omega_1 t + A_2 \cos \omega_2 t)^2. \end{aligned} \tag{35.15}$$

The polarization contains the term

$$P_{\omega_1 - \omega_2} = \frac{1}{2} \varepsilon_0 \chi^{(2)} A_1 A_2 \cos(\omega_1 - \omega_2) t \tag{35.16}$$

that is the source of the field at the difference frequency. The polarization and, accordingly, the field at the difference frequency $\omega_1 - \omega_2$ are proportional to the product of the amplitudes A_1 and A_2. In an elementary process of difference frequency generation, a photon (energy $h\nu_1$) is annihilated, a photon at the energy $h\nu_2$ and another photon at the energy $h\nu_3 = h\nu_1 - h\nu_2$ are created (Fig. 35.3b).

The frequency difference generation obeys the *Manley-Rowe rule*. A photon of the quantum energy $h\nu_1$ can only produce one photon of energy $h\nu_3$. This corresponds to the energy conservation law of the elementary process. Thus, the efficiency of conversion of radiation at frequency ν_1 to radiation of frequency ν_3 is

$$\eta_{\text{diff}} = \nu_3 / \nu_1. \tag{35.17}$$

If the frequency ν_3 is much smaller than ν_1 (and ν_2), only a small portion of the power of radiation at the frequency ν_1 is converted to power of radiation at the difference frequency.

Application. The superposition of two visible or near infrared laser fields of different frequencies can lead to generation of far infrared radiation.

35.5 Optical Parametric Oscillator

An optical parametric oscillator (OPO) converts radiation of a pump frequency (ν_p) to tunable radiation at two other frequencies (ν_1 and ν_2). Radiation of one of the frequencies ν_1 or ν_2 (or of both frequencies) is stored in a resonator in order to produce a feedback to the nonlinear crystal (Fig. 35.4a). The OPO shows threshold behavior. Above a threshold amplitude of the pump field, optical parametric oscillation sets in. The oscillation frequency ν_1 depends on the resonator. Changing the eigenfrequency of the resonator leads to a variation of ν_2 and ν_3.

(Together with the change of the resonance frequency, phase matching of the fields of different frequencies is a necessary condition for operation of a parametric

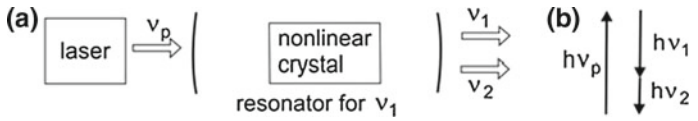

Fig. 35.4 Optical parametric oscillator; OPO. **a** Principle. **b** Elementary process

oscillator. Phase matching can be achieved by the choice of an appropriate orientation of the nonlinear crystal, e.g., of a $LiNbO_3$ crystal; changing the frequency of the signal wave then requires a rotation of the crystal. Another possibility is the change of the crystal temperature, suitable for KDP.)

In the photon picture (Fig. 35.4b), the elementary process in the OPO crystal corresponds to the decay of a photon into two photons of smaller quantum energy. The energy conservation law holds,

$$h\nu_\mathrm{p} = h\nu_1 + h\nu_2. \tag{35.18}$$

An optical parametric oscillator, pumped with radiation near 1 μm, is suitable for generation of tunable infrared radiation with frequencies in the range from 1 THz to 15 THz; the threshold pump power of a parametric oscillator is of the order of 1 MW/cm^2 (for $LiNbO_3$ as the nonlinear crystal).

The notations—ν_p = pump frequency, $\nu_1 = \nu_\mathrm{s}$ = signal frequency and $\nu_2 = \nu_\mathrm{i}$ = idler frequency—have originally been introduced in the fields of high frequency technique and of microwave technique. "Parametric" means that the pump field modulates a parameter and that the parameter gives rise to a frequency converting process. In the OPO, the parameter is the refractive index of the nonlinear medium.

35.6 Third-Order Polarization

As an example of third-order polarization $P^{(3)} = \varepsilon_0 \chi^{(3)} E^3$, we consider the effect of a harmonic field $E = A\cos\omega t$ and find

$$P^{(3)} = \frac{1}{4}\varepsilon_0 \chi^{(3)} A^3 \cos 3\omega t + \frac{3}{4}\varepsilon_0 \chi^{(3)} A^3 \cos\omega t. \tag{35.19}$$

The third-order polarization due to a monochromatic field causes two different effects, frequency tripling and a change of the refractive at the frequency ω.

The first term is the source of an electric field at the frequency 3ν (Fig. 35.5a). Radiation of frequency ν is converted to radiation of frequency 3ν. In an elementary process, three photons of energy $h\nu$ are annihilated and a photon of energy $3h\nu$ is created (Fig. 35.5b).

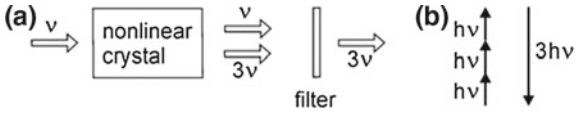

Fig. 35.5 Frequency tripler. **a** Principle. **b** Elementary process

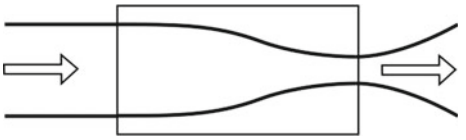

Fig. 35.6 Self-focusing

The polarization at the frequency ω can be written in the form:

$$P = \varepsilon_0 \chi^1 A \cos \omega t + \frac{3}{4} \varepsilon \chi^{(3)} A^3 \cos \omega t = \varepsilon_0 \left(\chi^{(1)} + \frac{3}{4} \chi^{(3)} A^2 \right) A \cos \omega t$$
$$= \varepsilon_0 \chi^{(1)} \left(1 + \frac{3\chi^{(3)}}{4\chi^{(1)}} A^2 \right) A \cos \omega t. \tag{35.20}$$

It follows, with $n = \sqrt{1 + \chi^{(1)}}$, that $n = n_0 + n_2 I$, where n_0 is the refractive index and

$$n_2 = \frac{6\chi^{(3)}}{c\chi^{(1)}\varepsilon_0} \tag{35.21}$$

is a factor that accounts for the change of the refractive index due to third-order nonlinearity (Problems 35.2 and 35.3). The intensity-dependent refractive index gives rise to self-focusing (Fig. 35.6; the Kerr lens mode locking (Sect. 13.2) makes use of self-focusing).

35.7 Four-Wave Mixing and Optical Frequency Analyzer

In a four-wave mixing experiment (Fig. 35.7a) two fields (frequency ν_1 and ν_2) produce fields at two other frequencies (ν_3 and ν_4). The polarization

$$P = \varepsilon_0 \chi^{(3)} E_1 E_2 E_3, \tag{35.22}$$

with $E_1 = A_1 \cos \omega_1 t$, $E_2 = A_2 \cos \omega_2 t$ and $E_3 = A_3 \cos \omega_3 t$, is responsible for the mixing process. The polarization P is the source of a field $E_4 = A_4 \cos \omega_4 t$. The nonlinear medium can be a crystal or a glass. The elementary process of the four-wave mixing (Fig. 35.7b) corresponds to the conversion of two photons (energy $h\nu_1$ and $h\nu_2$) to two other photons (energy $h\nu_3$ and $h\nu_4$). Four-wave mixing is a stimulated process. It occurs at strengths above appropriate threshold fields of E_1 and E_2.

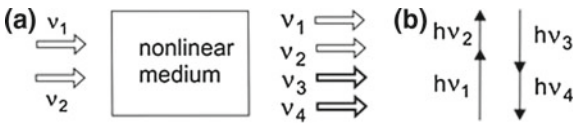

Fig. 35.7 Four-wave mixing. **a** Principle. **b** Elementary process

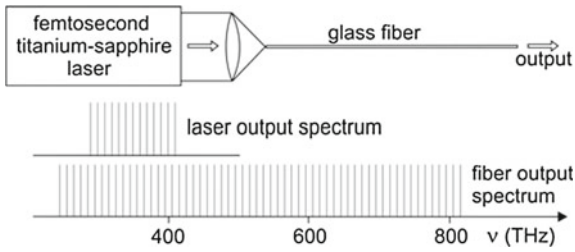

Fig. 35.8 Four-wave mixing of a frequency comb

Four-wave mixing has many applications (*see* books on nonlinear optics). Here, we discuss as an example the role of four-wave mixing for the optical frequency analyzer.

Example four-wave mixing of radiation consisting of an optical frequency comb.

In our earlier treatment of the optical spectrum analyzer, we have seen that the pulses emitted by a femtosecond laser are not exactly multiples of the round trip frequency f_r of the radiation pulses in the laser, but that the frequencies are shifted by a frequency offset f_o. The frequency of the nth line of the optical frequency comb is equal to

$$f_n = nf_r + f_o. \tag{35.23}$$

We now discuss how the frequencies f_r and f_o can be determined (Fig. 35.8). A frequency comb generated by a titanium–sapphire laser is strongly focused to a glass fiber (inner diameter $\sim$1 μm). In a four-wave mixing process, the frequency comb broadens and all lines show the same frequency shift f_o. This follows from an analysis of the polarization. The term $\cos\omega_1 t \cos\omega_2 t \cos\omega_3 t$ of the polarization contains the angular frequency $\omega_1 + \omega_2 - \omega_3$ and is the source of a field of frequency

$$f_4 = f_1 + f_2 - f_3 = n_4 f_r - f_o, \tag{35.24}$$

where $n_4 = n_1 + n_2 - n_3$ is an integer. By the mixing of radiation of different frequencies f_1, f_2, and f_3, the frequency comb becomes very broad. The optical frequency analyzer involves the following frequencies.

- f_r = repetition rate of the femtosecond pulses = number of pulses per second, measured by counting the pulses.
- f_o = offset frequency; measured by mixing of frequency-doubled radiation.
- $2f_n = 2nf_r + 2f_o$ = frequencies of frequency-doubled radiation. The mixing of the frequency-doubled radiation with radiation generated by four-wave mixing provides the difference frequencies

$$2f_n - f_m = (2n - m)f_r - f_o, \tag{35.25}$$

where n and m are integers. For $2n = m$, the difference frequency is f_o. By measuring different combinations of $(2n - m)$, also f_r can be determined by a mixing experiment: a photodiode serves as nonlinear device, which produces microwaves at the difference frequencies.

In an optical frequency analyzer, the frequency f_o is kept constant. For this purpose, the laser resonator of the titanium–sapphire laser is stabilized: the distance between the resonator mirrors is piezoelectrically controlled. After starting a femtosecond titanium–sapphire laser and reaching stable operation, the offset frequency f_o is kept constant

35.8 Stimulated Raman Scattering

An efficient way of converting monochromatic radiation into coherent radiation of another frequency is the *stimulated Raman scattering*.

In a Raman scattering process (Fig. 35.9), the energy $\hbar\omega_1$ of a photon is converted to energy $\hbar\omega_2$ of another photon and internal excitation energy E_{int} of a medium. The Raman scattered light is incoherent. Above a threshold pump field, stimulated Raman scattering results in the generation of a coherent field. Internal excitations can be phonons in crystals, phonons in glasses, vibrational-rotational excitations, or rotational excitations in molecular gases.

Example Stimulated Raman scattering of radiation of a CO_2 laser at molecules (e.g., CH_3F molecules in a gas) can lead to coherent far infrared radiation; the internal excitation is a vibrational-rotational state in a molecular gas (Sect. 14.9).

References [12, 297–307].

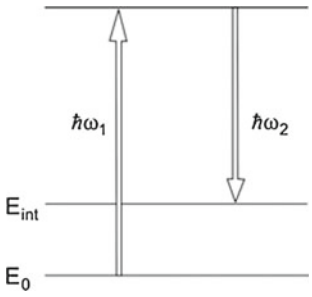

Fig. 35.9 Raman scattering

Problems

35.1 Two monochromatic optical waves (wavelengths near 600 nm) are focused on a photodetector. The photodetector generates a microwave signal at beat frequencies. The smallest beat frequency is 200 MHz. Calculate the wavelength difference of the two optical waves.

35.2 Nonlinear polarization.

(a) Show that a strong electric field applied to a hydrogen atom gives rise to nonlinear polarization of any order. [*Hint*: make use of the Taylor expansion of $(1 + x)^{-2}$].
(b) Estimate the values of $\chi^{(2)}$, $\chi^{(3)}$ and $\chi^{(4)}$; $\chi^{(1)}$ is of the order of unity. [*Hint*: the lowest-order correction term $P^{(2)}$ would be comparable to $P^{(1)}$ if the amplitude A of the field is of the order of the strength of field acting on an electron in an H atom.]

35.3 Show that (35.21) follows from (35.20). Estimate the value of n_2. [*Hint*: use the estimate of $\chi^{(3)}$ in the preceding problem.]