3. Optical Nonlinearities with Ultrashort Pulses

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With 27 Figures

Picosecond and femtosecond pulses provide a unique tool for the study of ultrafast processes accompanying light-matter interaction. Novel measuring techniques, on the one hand, expanded the experimental capabilities in this field while, on the other hand, a great demand for suitable pulses in various spectral regions was created that cannot be directly fulfilled by available laser systems. A basic solution of this problem is offered by quantum optics which can convert pulse frequencies with the help of nonlinear optical effects. A second motivation for the study of short pulse nonlinearities is the high power level necessarily connected with shorter and shorter pulses of finite energy content. Consequently, nonlinear effects cannot be ignored in spectroscopic applications for an adequate understanding of the coupling between the short pulse and the dynamical processes under investigation. Without exaggeration one can say that nonlinear optics is an inherent and fundamental part of the generation, analysis and application of ultrashort laser pulses.

It is beyond the scope of the present chapter to review the abundant literature on nonlinear optics. The following discussion will be restricted to investigations with pulses shorter than 1 ns. Special emphasis will be placed on applications e.g. conversion of the pulses frequencies. Related work with longer time scales will only be briefly mentioned. For background information the reader is referred to text books on quantum optics, to the second volume of the Laser Handbook (Arecchi and Schulz-Dubois 1972), to the monograph on parametric processes by Reintjes (1984) and to other volumes of the series Quantum Electronics – Principles and Applications. Applications of ultrafast nonlinear optics to mode-locking techniques and to spectroscopic investigations are outlined in other chapters of this volume.

3.1 Nonlinear Polarization

The concept of nonlinear optics is considerably older than the laser. Interactions involving two or more quanta, e.g. two-photon absorption and stimulated Raman scattering were described theoretically as early as 1931 by Goeppert-Mayer. Other nonlinear optical interactions such as the saturation of an electronic transition of atoms were also observed before the invention of the laser.
At the high light intensities of \( \lesssim 10^{12} \text{ W/cm}^2 \) which, with ultrashort pulses, can be applied to condensed matter without irreversible damage, nonlinear phenomena turn out to be amazingly efficient.

The intense photon flux favours the use of the rather simple classical theory of electromagnetism. The response of the medium to the incident optical radiation is formulated in terms of the induced macroscopic polarisation. Familiar processes like (linear) absorption, refraction and scattering arise from polarizations proportional to the first power of the optical field. To include nonlinear phenomena higher order terms in the Taylor expansion of the polarization have to be considered. In the electric dipole approximation one may write

\[
P(E) = \chi^{(1)}E + \chi^{(2)}:EE + \chi^{(3)}:EEE + \cdots.
\]

The constant term has been omitted in (3.1) since it cannot contribute to nonlinear processes. \( \chi^{(2)} \) and \( \chi^{(3)} \) denote the second- and third-order susceptibilities describing three- and four-wave interactions, respectively. Similar to the well-known linear susceptibility \( \chi^{(1)} \), the higher order coefficients \( \chi^{(n)} \) display a distinct frequency dependence, corresponding, in general, to time-dependent nonlinear susceptibilities in the time domain. Simple use of (3.1) for very short pulses is therefore possible only for special cases with negligible frequency dependency, i.e. far-off resonances.

For an understanding of the nonlinear interaction, the macroscopic susceptibilities have to be related to a microscopic picture of the physical mechanisms. Quantum mechanical derivations were discussed by Armstrong et al. (1962) and more recently by Flytzanis (1975). For short pulse nonlinearities, e.g. close to material resonance it is often convenient to avoid the \( \chi \)-formalism and to treat the transient response of the medium directly by microscopic equations of motion. Typical time constants characterizing the dynamics are \( 10^{-13} - 10^{-10} \text{ s} \) in condensed matter (and somewhat longer in gases) and are directly measurable with femto- and picosecond pulses.

Equation (3.1) allows a classification of nonlinear processes into three- or four-wave interactions etc. This ordering will be used in Sects. 3.2, 3. In Sect. 3.4 particular physical systems that have attracted great interest in current research work and which promise to be important in the future will be described.

### 3.2 Three-Wave Interactions

#### 3.2.1 Second Harmonic Generation

After its first observation by Franken et al. (1961), second harmonic (SH) generation quickly became an important process for the extension of the wavelength range accessible with available laser sources because of its simple, reliable application and good conversion efficiency. The process is of second order in the