Part D

Higher-Order Nonlinear Effects

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$$P_{tot} = \epsilon_O \chi^{(1)} E + \epsilon_O \chi^{(2)} E^2 + \epsilon_O \chi^{(3)} E^3 + \epsilon_O \chi^{(4)} E^4 + \epsilon_O \chi^{(5)} E^5 + \ldots\ldots$$

Nonlinear effects of higher order than 2 or 3 are present in many high-power laser experiments but are not dominant and are thus difficult to detect. Explicitly reported are the generation of higher-order frequency harmonics mainly in noble gases [4.693–4.743]. For technical applications the stepwise frequency transformation based on second and third harmonic effects is usually more efficient. This is a consequence of the much larger nonlinear coefficients $\chi^{(2)}$ and $\chi^{(3)}$ compared to, e.g. $\chi^{(4)}$, $\chi^{(5)}$ and so on for known materials. Further these materials have to be transparent over a wide spectral range because absorption especially of the short wavelength harmonics will decrease the efficiency. On the other hand the resonance effect working with wavelengths close to matter absorption can increase the nonlinear effect drastically [e.g. 4.693]. Therefore a suitable compromise can enhance the harmonic output.

The usually applied atom vapors for generation of higher frequencies do not automatically give phase matching. Thus by tuned mixing of different atoms with different refractive indices at the wavelength of the fundamental and the high harmonics, phase matching can be achieved in isotropic materials

As an example the generation of the fifth harmonic in Ne vapor is described, which in combination with the generation of twice the second harmonic, finally results in the generation of the 20th harmonics of the original Nd:YAG laser light . The process is depicted in Fig. 4.56.

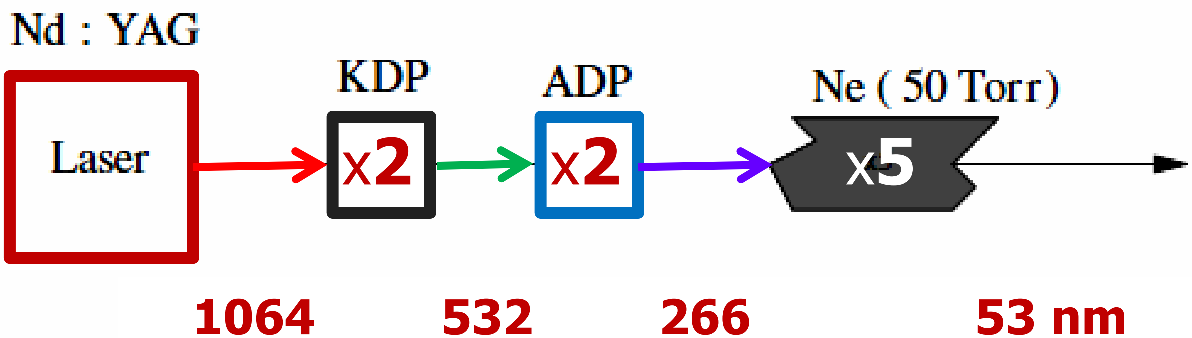

Fig. 4.56. Generation of the fifth harmonic in Ne vapor by pumping with the fourth harmonic of a Nd:YAG laser resulting in the 20th harmonic of the laser radiation

The efficiency in this experiment was less than 10^{-6} although it was performed close to the resonance of Ne atomic absorption and thus the nonlinear coefficient is distinctly enlarged.

Another example is the reported seventh harmonic generation of the radiation of a Krypton fluoride excimer laser with a wavelength of 248 nm in He vapor The resulting seventh harmonic shows a wavelength of 35.4 nm.

The generation of even higher harmonics for generating coherent light at wavelengths below 20 nm was obtained using very high powers in the range of TW and more with short pulses in the ps or fs range. The fourth-order nonlinear processes are discussed in [4.701–4.704], the generation of the fifth-harmonic was obtained besides the seventh-harmonic . Much higher harmonics are observed . For example, the 221th-harmonic as discrete harmonic peak of coherent light with a wavelength of 3.6 nm was observed in He using a high-power Ti:sapphire laser pulse with a width of 26 fs, an energy of 20 mJ, a wavelength of 800 nm and a focal spot diameter of 100 μm resulting in an intensity of 6×10^{15} W cm^{-2}. Ne or He gas was used at 8 Torr. Coherent emission was observed up to the 297th harmonic of the laser light corresponding to a wavelength of 2.7 nm.