Part E

Materials for Non-resonant Nonlinear Interactions

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NLO materials are materials in which the intensity of light input, including its frequency, is not directly proportional to the intensity of light output.

Because of this NL behavior, an intense light beam propagating through a NLO material will produce new effects that can't be seen with weak light beams, e.g., an intense light beam propagating through a NL material can generate harmonics of the original light frequency. This means that the red beam from a ruby laser can create UV beam as it passes through the NLO material.

Inorganic Crystals

Crystals are used for all kinds of frequency transformation technologies such as harmonic generation, frequency mixing and electro-optical effects. They may be classified into two groups:

- *grown from solution*: These crystals are hygroscopic and in contrast to the group below thermal shock sensitive, mostly fragile and comparatively soft. But they are available in large sizes of good optical quality and are mostly cheaper.
- *grown from melt*: These crystals are nonhygroscopic and thus much more useful than those above.

Known materials are, e.g. KDP, KD*P, ADP, AD*P and $LiNbO_3$. More recently developed crystals are KTP, CDA, CD*A, RDA, RDP, BBO, BIBO, LBO and BANANA.

Organic Materials

Organic materials can show very high nonlinear coefficients, high damage threshold and good transparency at short wavelengths .

Because of the large variety of these compounds an inestimable number of possibilities exist in principle. Molecules with a large conjugated π-electron system from a large number of multiple bonds will show a large inducable dipole moment from these delocalized electrons. This can even be enhanced by donor (N-atoms) and acceptor (O-atoms) groups.

Some of these organic molecules can be crystallized with sufficient optical quality. Known examples of crystals without an inversion center are urea [M33], DAN, MNA, MAP, COANP, PAN and MBANP.

The inversion symmetry can be broken by applying these materials at surfaces. SHG with high efficiencies was demonstrated this way. Liquid crystals are especially applied in such setups.

Amorphous organic matter will find new applications in photonics as in optical fibers, in optical switches and storage or in optical phase conjugation. Many polymer materials have been proposed and are still used.

Liquids

Organic liquids or solutions are applied in nonresonant photonic applications for "white light" generation , optical phase conjugation, and Raman shifting of the incident light. These are different from the nonlinear absorbers and laser materials for dye lasers in resonant applications, and in this case the transparent matter operates again by its induced dipole moments, based on the electron distribution in different electronic or vibrational states of the organic molecules in a similar way as that just described above.

Useful materials are, e.g. CS_2, CCl_4, $TiCl_4$, Freon, hexene, benzene, alcohol and almost all other solvents as described in Sect. 4.5.9 (p. 224). For nonlinear applications with high light powers the chemically specified purity is sometimes not sufficient. Small particles which are usually not specified can disturb the nonlinear interaction and promote optical break down by the resulting inhomogeneous high local field. Therefore lavish cleaning with filters or "pump and freeze" procedures may be necessary before use [4.408].

Liquid Crystals

The geometrical orientation and order of molecular systems in liquid crystals [M31, 4.750–4.758] can be applied in photonics for changing the polarization of a transmitting light beam. This is achieved in liquid crystal displays (LCD) of all kinds and projectors based on electro-optically switching the orientation of the molecules by an external electric field. Other electro-optic devices such as phase modulators were build based on liquid crystals.

However, the orientation of the molecules in the liquid crystal can be changed via polarized pump light, too, and thus opto-optical switching becomes possible. Furthermore, liquid crystals can be used for frequency conversion and four-wave mixing techniques

Gases

Noble gases and gases of organic molecules are used in a way similar to liquids for optical phase conjugation and Raman shifting. The mechanisms are the same as described for liquids and references are given there. Gases show the advantage of easy "self-repairing" if damage threshold is exceeded and, e.g. optical breakdown occurred. Thus high-power applications are possible with nonlinear processes in gases.

Typical applied nonlinear gases are SF_6, N_2, Xenon, CH_4, C_2F_6, CO and CO_2 used at pressures of 10–100 bar. The damage threshold in gases is mostly determined by impurities. It can be improved by at least one order of magnitude by cleaning, e.g. with high electric fields