Nonlinear optics

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Nonlinear optics is the branch of optics that describes the behaviour of light in *nonlinear media*, that is, media in which the polarization **P** responds nonlinearly to the electric field **E** of the light. This nonlinearity is typically only observed at very high light intensities such as provided by pulsed lasers.

Nonlinear optics gives rise to a host of optical phenomena:

Contents

Frequency mixing processes

- Second harmonic generation (SHG), or *frequency doubling*, generation of light with a doubled frequency (half the wavelength);
- Self-phase modulation (SPM), a $\chi^{(3)}$ effect.
- Sum frequency generation (SFG), generation of light with a frequency that is the sum of two other frequencies (SHG is a special case of this);
- Third harmonic generation (THG), generation of light with a tripled frequency (one-third the wavelength) (usually done in two steps: SHG followed by SFG of original and frequency-doubled waves);
- Difference frequency generation (DFG), generation of light with a frequency that is the difference between two other frequencies;
- Parametric amplification, amplification of a signal input in the presence of a higher-frequency pump wave, at the same time generating an *idler* wave (can be considered as DFG);
- Parametric oscillation, generation of a signal and idler wave using a parametric amplifier in a resonator (with no signal input);
- Parametric generation, like parametric oscillation but without a resonator, using a very high gain instead;
- Spontaneous parametric down conversion (SPDC), the amplification of the vacuum fluctuations in the low gain regime;
- Optical rectification, generation of quasi-static electric fields.

Other nonlinear processes

- Optical Kerr effect, intensity dependent refractive index;
  - Self focusing;
    - Kerr-lens modelocking (KLM).
  - Self-phase modulation (SPM);
    - Optical solitons.
  - Cross-phase modulation (XPM);
  - Four-wave mixing (FWM), can also arise from other nonlinearities.
- Raman scattering, interaction of photons with optical phonons;
  - Raman amplification,
  - Optical phase conjugation.
- Brillouin scattering, interaction of photons with acoustic phonons;
  - Optical phase conjugation.
- Two-photon absorption, simultaneous absorption of two photons, transferring the energy to a single electron;
- Multiple photoionisation, near-simultaneous removal of many bound electrons by one photon.

- Chaos in Optical Systems

Related processes

In these processes, the medium has a linear response to the light, but the properties of the medium are affected by other causes:

- Pockels effect, the refractive index is affected by a static electric field; used in electro-optic modulators;
- Acousto-optics, the refractive index is affected by acoustic waves (ultrasound); used in acousto-optic modulators.

Frequency-mixing processes

One of the most commonly-used frequency-mixing processes is **frequency doubling** or second-harmonic generation. With this technique, the 1064-nm output from Nd:YAG lasers or the 800-nm output from Ti:sapphire lasers can be converted to visible light, with wavelengths of 532 nm (green) or 400 nm (violet), respectively.

Practically, frequency-doubling is carried out by placing a special crystal in a laser beam under a well-chosen angle. Commonly-used crystals are BBO (β-barium borate), KDP (potassium dihydrogen phosphate), KTP (potassium titanyl phosphate), and lithium niobate. These crystals have the necessary properties of being strongly birefringent (necessary to obtain phase matching, see below), having a specific crystal symmetry and of course being transparent for and resistant against the high-intensity laser light. However, organic polymeric materials are set to take over from crystals as they are cheaper to make, have lower drive voltages and superior performance.

Theory

A number of nonlinear optical phenomena can be described as frequency-mixing processes. If the induced dipole moments of the material respond instantaneously to an applied electric field, the dielectric polarization (dipole moment per unit volume) $P(t)$ at time t in a medium can be written as a power series in the electrical field:

$$P(t) \propto \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \cdots.$$

Here, the coefficients $\chi^{(n)}$ are the n-th order susceptibilities of the medium. For any three-wave mixing process, the second-order term is crucial; it is only nonzero in media that have no inversion symmetry. If we write

$$E(t) = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + \text{c.c.},$$

where c.c. denotes the complex conjugate (E_1 and E_2 being the incident beams of interest), the second-order term in will read

$$P^{(2)}(t) \propto \sum \chi^{(2)} n_0 E_1^{n_1} E_2^{n_2} e^{i(m_1\omega_1 + m_2\omega_2)t} + \text{c.c.},$$

where the summation is over

$$(n_0, n_1, n_2, m_1, m_2) = (1,2,0,2,0),\ (1,0,2,0,2),\ (2,2,0,0,0),\ (2,0,2,0,0),\ (2,1,1,1,-$$

The six combinations (n_x,m_x) correspond, respectively, to the second harmonic of E_1, the second harmonic of E_2, the optically rectified signals of E_1 and E_2, the difference frequency, and the sum frequency. A medium that is thus pumped by the fields E_1 and E_2 will radiate a field E_3 with an angular frequency $\omega_3 = m_1\omega_1 + m_2\omega_2$.

Note: in this description, $\chi^{(2)}$ is a scalar. In reality, $\chi^{(2)}$ is a tensor whose components depend on the combination of frequencies.

Parametric generation and amplification is a variation of difference frequency generation, where the lower-frequency one of the two generating fields is much weaker (parametric amplification) or completely absent (parametric generation). In the latter case, the fundamental quantum-mechanical uncertainty in the electric field initiates the process.

Phase matching

The above ignores the position dependence of the electrical fields. In a typical situation, the electrical fields are traveling waves described by

$$E_j(\mathbf{x}, t) = e^{i(\omega_j t - \mathbf{k}_j \cdot \mathbf{x})},$$

at position $\mathbf{x}$, with the wave vector $\mathbf{k}_j = n(\omega_j)\omega_j/c$, where c is the velocity of light and $n(\omega_j)$ the index of refraction of the medium at angular frequency ω_j. Thus, the second-order polarization angular frequency ω_3 is

$$P^{(2)}(\mathbf{x}, t) \propto E_1^{n_1} E_2^{n_2} e^{i(\omega_3 t - (m_1 \mathbf{k}_1 + m_2 \mathbf{k}_2) \cdot \mathbf{x})}.$$

At each position $\mathbf{x}$, the oscillating second-order polarization radiates at angular frequency ω_3 and a corresponding wave vector $\mathbf{k}_3 = n(\omega_3)\omega_3/c$. Constructive interference, and therefore a high intensity ω_3 field, will occur only if

$$\mathbf{k}_3 = m_1 \mathbf{k}_1 + m_2 \mathbf{k}_2.$$

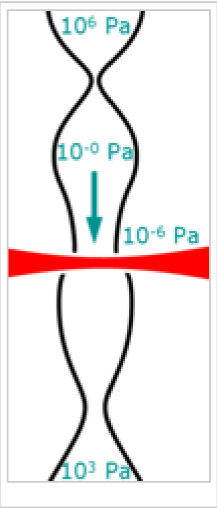
The above equation is known as the *phase matching condition*. Typically, three-wave mixing is done in a birefringent crystalline material (I.e., the refractive index depends on the polarization and direction of the light that passes through.), where the polarizations of the fields and the orientation of the crystal are chosen such that the phase-matching condition is fulfilled. This phase matching technique is called angle tuning.

One undesirable effect of angle tuning is that the optical frequencies involved are not collinear with each other. This is due to the fact that the extraordinary wave propagating through a birefringent crystal possesses a Poynting vector that is not parallel with the propagation vector. This would lead to beam walkoff which limits the nonlinear optical conversion efficiency. Two other methods of phase matching avoids beam walkoff by forcing all frequencies to propagate at a 90 degree angle with respect to the optical axis of the crystal. These methods are called temperature tuning and quasi-phase-matching.

Temperature tuning is where the pump (laser) frequency polarization is orthogonal to the signal and idler frequency polarization. The birefringence in some crystals, in particular Lithium Niobate is highly temperature dependent. The crystal is controlled at a certain temperature to achieve phase matching conditions.

The other method quasi-phase matching. In this method the frequencies involved are not constantly locked in phase with each other, instead the crystal axis is flipped at a regular interval Λ, typically 15 micrometres in length. Hence, these crystals are called periodically-poled. This results in the polarization response of the crystal to be shifted back in phase with the pump beam by reversing the nonlinear susceptibility. This allows net positive energy flow from the pump into the signal and idler frequencies. In this case, the crystal itself provides the additional wavevector $k=2\pi/\lambda$(and hence momentum) to satisfy the phase matching condition. Quasi-phase matching can be expanded to chirped gratings to get more bandwidth and to shape a SHG pulse like it is done in a dazzler. SHG of a pump and Self-phase modulation (emulated by second order processes) of the signal and an optical parametric amplifier can be integrated monolithically.

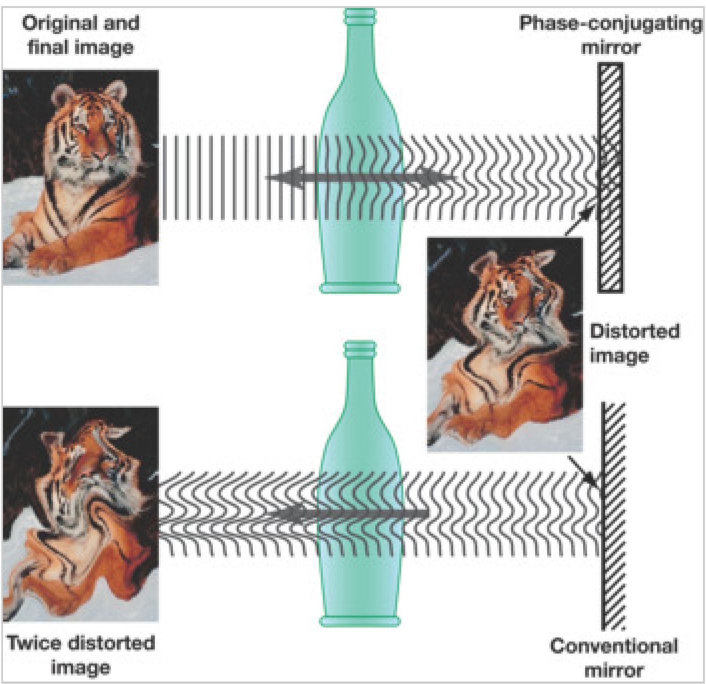
Higher-order frequency mixing



The above holds for $\chi^{(2)}$ processes. It can be extended for processes where $\chi^{(3)}$ is nonzero, something that is generally true in any medium without any symmetry restrictions. Third-harmonic generation is a $\chi^{(3)}$ process, although in laser applications, it is usually implemented as a two-stage process: first the fundamental laser frequency is doubled and then the doubled and the fundamental frequencies are added in a sum-frequency process. The Kerr effect can be described as a $\chi^{(3)}$ as well.

At high intensities the Taylor series, which led the domination of the lower orders, does not converge anymore and instead a time based model is used. When a noble gas atom is hit by an intense laser pulse, which has an electric field strength comparable to the Coulomb field of the atom, the outermost electron may be ionized from the atom. Once freed, the electron can be accelerated by the electric field of the light, first moving away from the ion, then back toward it as the field changes direction. The electron may then recombine with the ion, releasing its energy in the form of a photon. The light is emitted at every peak of the laser light field which is intense enough, producing a series of attosecond light flashes. The photon energies generated by this process can extend past the 800th harmonic order up to 1300 eV. This is called *high-order harmonic generation*. The laser must be linearly polarized, so that the electron returns to the vicinity of the parent ion. High-order harmonic generation has been observed in noble gas jets, cells, and gas-filled capillary waveguides.

Optical phase conjugation



Comparison of a phase conjugate mirror with a conventional mirror. With the phase conjugate mirror the image is not deformed when passing through an aberrating element twice.

It is possible, using nonlinear optical processes, to exactly reverse the propagation direction and phase variation of a beam of light. The reversed beam is called a *conjugate* beam, and thus the technique is known as **optical phase conjugation** (also called *time reversal*, *wavefront reversal* and *retroreflection*).

One can interpret this nonlinear optical interaction as being analogous to a real-time holographic process. In this case, the interacting beams simultaneously interact in a nonlinear optical material to form a dynamic hologram (two of the three input beams), or real-time diffraction pattern, in the material. The third incident beam diffracts off this dynamic hologram, and, in the process, reads out the phase-conjugate wave. In effect, all three incident beams interact (essentially) simultaneously to form several real-time holograms, resulting in a set of diffracted output waves that phase up as the "time-reversed" beam. In the language of nonlinear optics, the interacting beams result in a nonlinear polarization within the material, which coherently radiates to form the phase-conjugate wave.

The most common way of producing optical phase conjugation is to use a four-wave mixing technique, though it is also possible to use processes such as stimulated Brillouin scattering. A device producing the phase conjugation effect is known as a phase conjugate mirror (PCM).

For the four-wave mixing technique, we can describe four beams (j = 1,2,3,4) with electric fields:

$$\Xi_j(\mathbf{x},t) = \frac{1}{2}E_j(\mathbf{x})e^{i(\omega_j t - \mathbf{k}\cdot\mathbf{x})} + \mathrm{c.c.}$$

where E_j are the electric field amplitudes. Ξ_1 and Ξ_2 are known as the two pump waves, with Ξ_3 being the signal wave, and Ξ_4 being the generated conjugate wave.

If the pump waves and the signal wave are superimposed in a medium with a non-zero $\chi^{(3)}$, this produces a nonlinear polarization field:

$$P_{\mathrm{NL}} = \varepsilon_0\chi^{(3)}(\Xi_1 + \Xi_2 + \Xi_3)^3$$

resulting in generation of waves with frequencies given by $\omega = \pm\omega_1 \pm\omega_2 \pm\omega_3$ in addition to third harmonic generation waves with $\omega = 3\omega_1, 3\omega_2, 3\omega_3$.

As above, the phase-matching condition determines which of these waves is the dominant. By choosing conditions such that $\omega = \omega_1 + \omega_2 - \omega_3$ and $\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$, this gives a polarization field:

$$P_\omega = \frac{1}{2}\chi^{(3)}\epsilon_0 E_1 E_2 E_3^* e^{i(\omega t - \mathbf{k}\cdot\mathbf{x})} + \mathrm{c.c.}.$$

This is the generating field for the phase conjugate beam, Ξ_4. Its direction is given by $\mathbf{k}_4 = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$, and so if the two pump beams are counterpropagating ($\mathbf{k}_1 = -\mathbf{k}_2$), then the conjugate and signal beams propagate in opposite directions ($\mathbf{k}_4 = -\mathbf{k}_3$). This results in the retroreflecting property of the effect.

Further, it can be shown for a medium with refractive index n and a beam interaction length l, the electric field amplitude of the conjugate beam is approximated by

$$E_4 = \frac{i\omega l}{2nc}\chi^{(3)}E_1E_2E_3^*$$

(where *c* is the speed of light). If the pump beams E_1 and E_2 are plane (counterpropagating) waves, then:

$$E_4(\mathbf{x}) \propto E_3^*(\mathbf{x});$$

that is, the generated beam amplitude is the complex conjugate of the signal beam amplitude. Since the imaginary part of the amplitude contains the phase of the beam, this results in the reversal of phase property of the effect.

Note that the constant of proportionality between the signal and conjugate beams can be greater than 1. This is effectively a mirror with a reflection coefficient greater than 100%, producing an amplified reflection. The power for this comes from the two pump beams, which are depleted by the process.

The frequency of the conjugate wave can be different from that of the signal wave. If the pump waves are of frequency $\omega_1 = \omega_2 = \omega$, and the signal wave higher in frequency such that $\omega_3 = \omega + \Delta\omega$, then the conjugate wave is of frequency $\omega_4 = \omega - \Delta\omega$. This is known as *frequency flipping*.

Common SHG materials

- 806 nm light : lithium iodate ($LiIO_3$)
- 860 nm light : potassium niobate ($KNbO_3$)
- 980 nm light : $KNbO_3$
- 1064 nm light : monopotassium phosphate (KH_2PO_4, KDP), lithium triborate (LBO) and β-barium borate (BBO).
- 1319 nm light : $KNbO_3$, BBO, KDP, $LiIO_3$, lithium niobate ($LiNbO_3$), and potassium titanyl phosphate (KTP)

References

1. *Scientific American*, December 1985, "Phase Conjugation," by Vladimir Shkunov and Boris Zel'dovich.
2. *Scientific American*, January 1986, "Applications of Optical Phase Conjugation," by David M. Pepper.
3. *Scientific American*, October 1990, "The Photorefractive Effect," by David M. Pepper, Jack Feinberg, and Nicolai V. Kukhtarev.

See also

- Born-Infeld action
- Filament propagation
- Category:Nonlinear optical materials

External links

- Encyclopedia of laser physics and technology (http://www.rp-photonics.com/encyclopedia.html), with content on nonlinear optics, by Rüdiger Paschotta
- Nonlinear Optics, Quantum Optics: Concepts in Modern Optics (http://www.oldcitypublishing.com/NLOQO/NLOQO.html), journal of basic science and practical applications, edited by Takayoshi Kobayashi

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