Non-linear optics

The field of non-linear optics includes all of the processes whereby matter can alter an input frequency to yield one or more output frequencies. These are considered mixing processes that involve higher orders of the polarization. Absorption depends on a first-order in polarization, because susceptibility depends linearly on the electric field. However, there are higher order susceptibilities in matter, so that one can write the general expression for the polarization as,

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

Non-linear processes

- Second harmonic generation (SHG), or *frequency doubling*, generation of light with a doubled frequency (half the wavelength);
- 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);
- Difference frequency generation (DFG), generation of light with a frequency that is the difference between two other frequencies;
- 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);

Second order processes

Three wave mixing processes involving χ_2 are widely used in laser technology. There are six possible processes for this term. These are given by the formula.

$$E(t) = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} \\ + \textit{complex conjugate}$$

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

where (n_0, n_1, n_2, m_1, m_2)

(1,2,0,2,0) SHG, (1,0,2,0,2) SHG,

(2,2,0,0,0), (2,0,2,0,0) Optical rectification

(2,1,1,1,-1) DFG and (2,1,1,1,1) SFG

Second order processes

- The six combinations correspond to
- 1. Second harmonic generation of E_1
- 2. Second harmonic generation of E_2
- 3. Optically rectified signal of E_1
- 4. Optically rectified signal of E_2
- 5. The sum frequency
- 6. The difference frequency

Second harmonic generation

- A polarization is induced in the material at twice the input frequency.

$$P(2\omega) = \varepsilon_0 \chi_2 E(\omega)^2$$

- The SHG susceptibility is χ_2.

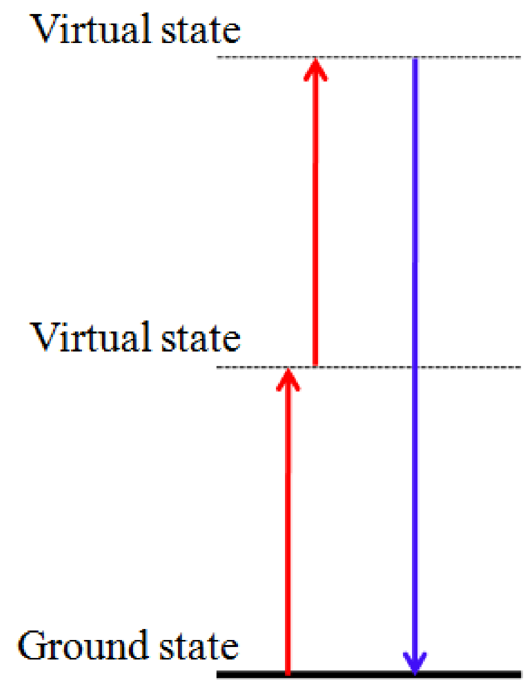
- SHG only occurs in a non-centrosymmetric crystal.

- It requires a birefringent crystal (i.e. a material that has two different indices of refraction).

Discovery of SHG

Second harmonic generation was first demonstrated by Franken, Hill, Peters, and Weinreich at the University of Michigan, Ann Arbor, in 1961.

An intense light source is required Since the SHG signal is so small. The invention of the laser was essential to demonstration of SHG. The first demonstration used the output of a Ruby laser (694 nm) in a quartz sample. The doubled signal at 347 nm was recorded on photographic paper. By mistake

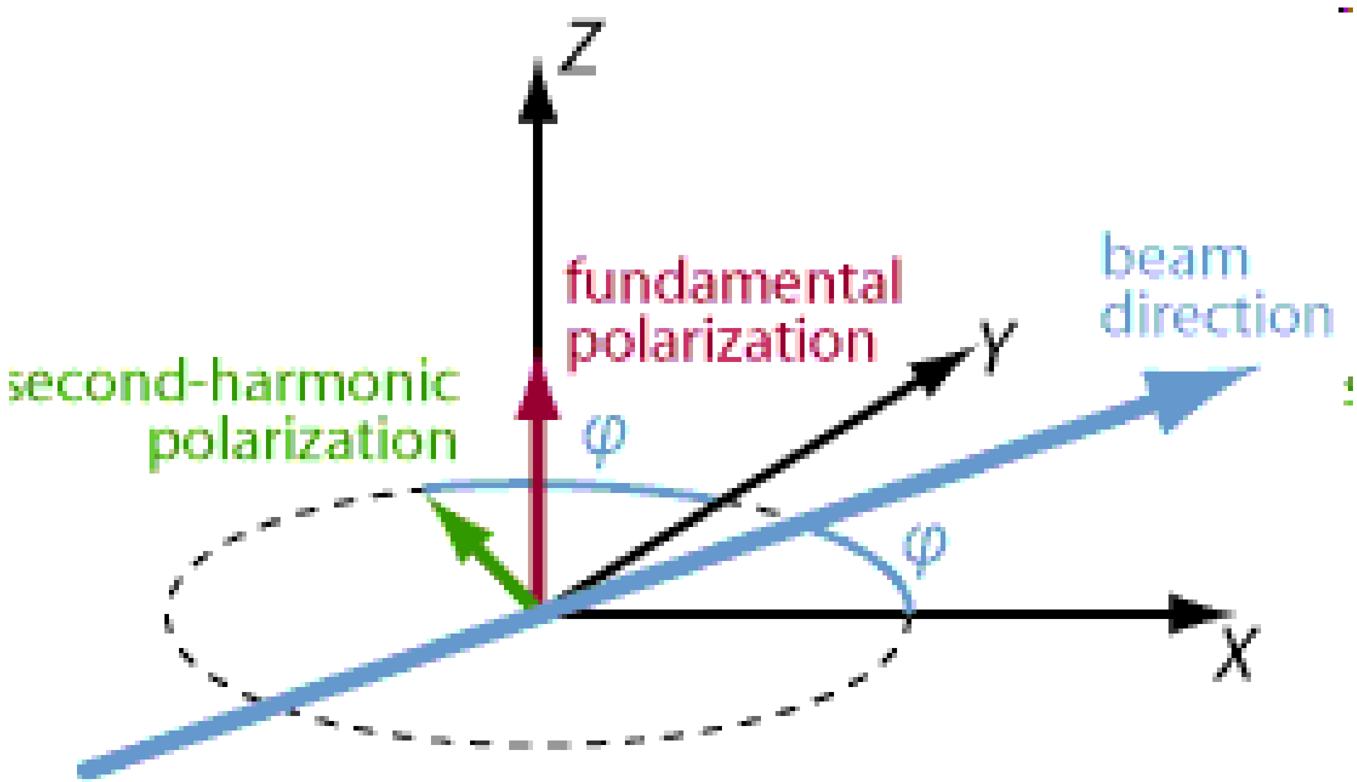


The editor of *Physical Review Letters* removed the signal on the photographic paper believing it was a speck of dirt.

Phase matching

Critical phase matching (also called angle phase matching) is a technique for obtaining phase matching of some nonlinear process in a birefringent crystal. The interacting beams are aligned at some angle(s) to the axes of the index ellipsoid. In almost all cases, there are one or two waves polarized along one axis of the index ellipsoid (*ordinary beam*), while another one or two waves are polarized at some variable angle with the plane spanned by the other two axes (*extraordinary beams*). Adjustment of the propagation angle affects the refractive index of the extraordinary beam (called *extraordinary refractive index*), whereas the ordinary index stays constant. For some angular position, phase matching may be achieved.

Beams in a birefringent crystal



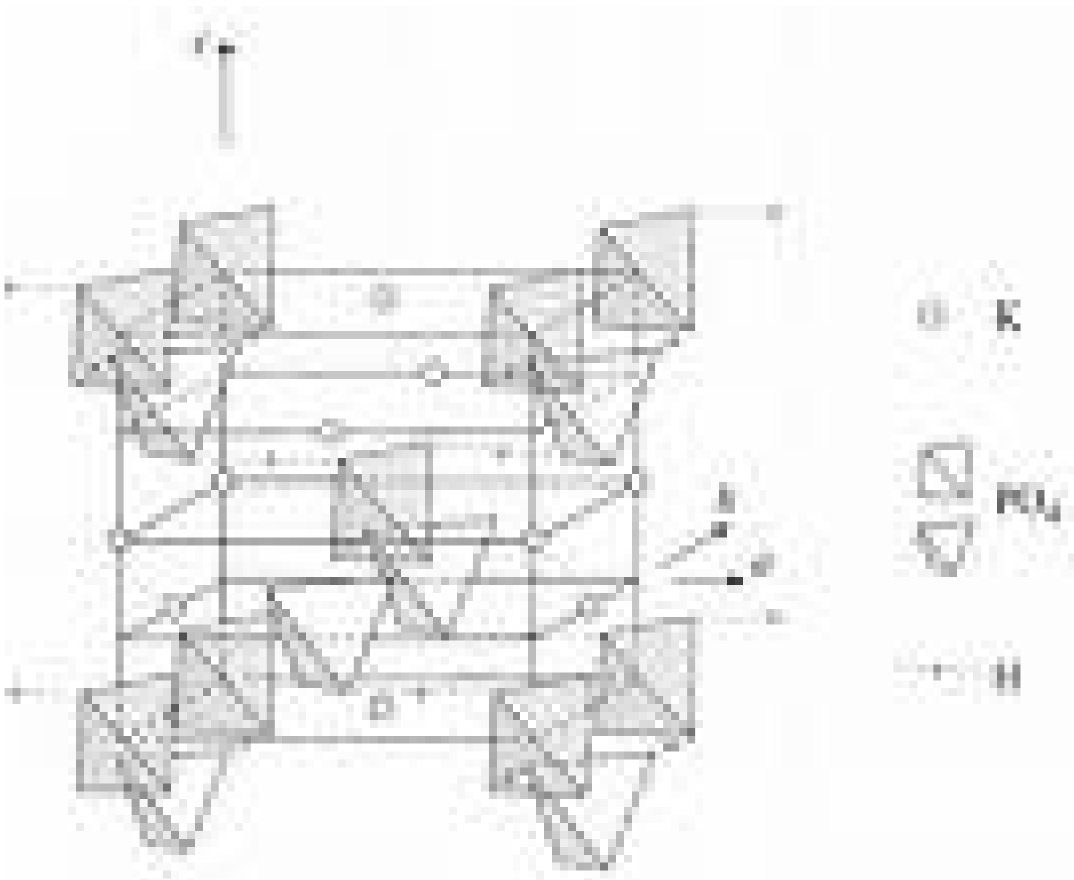
Example of phase matching in a birefringent crystal.

A birefringent crystal has two different indices of refraction along different directions (polarizations).

Types of SHG

- Second harmonic generation occurs in two types, denoted I and II.
- In **Type I SHG** two photons having ordinary polarization with respect to the crystal will combine to form one photon with double the frequency and extraordinary polarization.
- In **Type II SHG**, two photons having orthogonal polarization will combine to form one photon with double the frequency and extraordinary polarization. For a given crystal orientation, only one of these type of SHG occurs.

Structure of potassium dihydrogen phosphate (KDP)



International Union of Crystallography

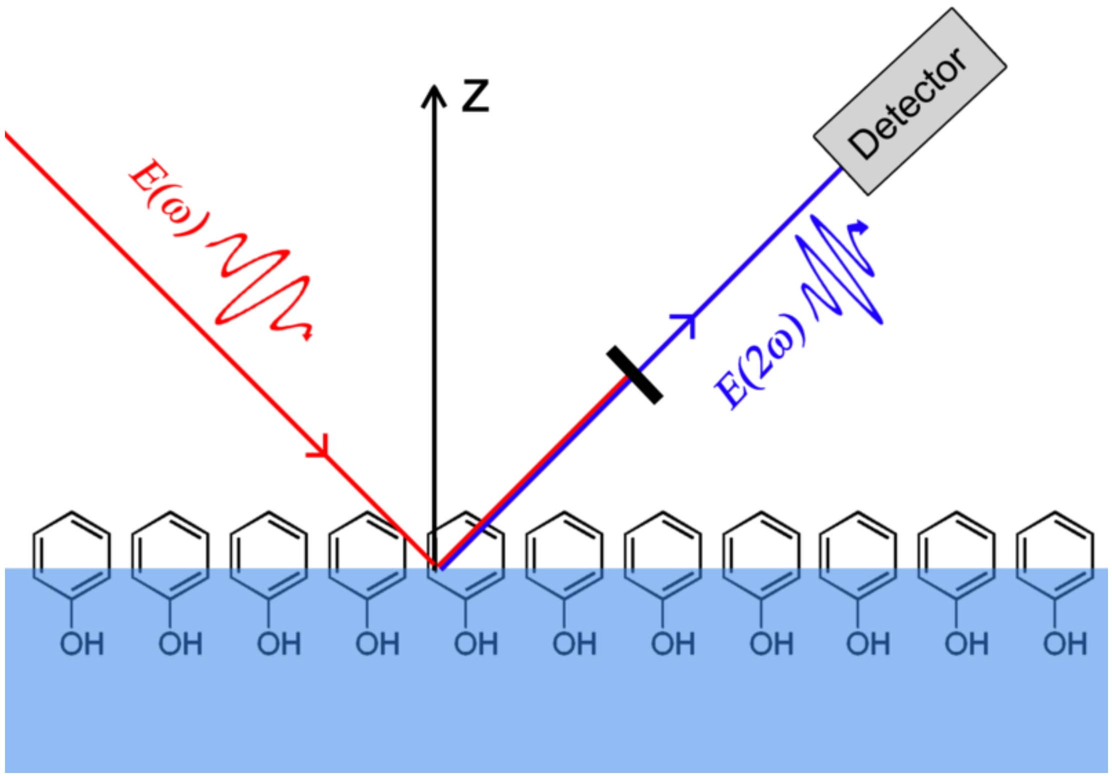
Grow your own KDP doubling crystal

This student is removing the first KDP crystal grown at Monte Vista High School.

An 800 pound KDP doubling crystal

National Ignition Facility and Photon Science
Lawrence Livermore National Laboratory

SHG on surfaces



SHG and SFG on surfaces

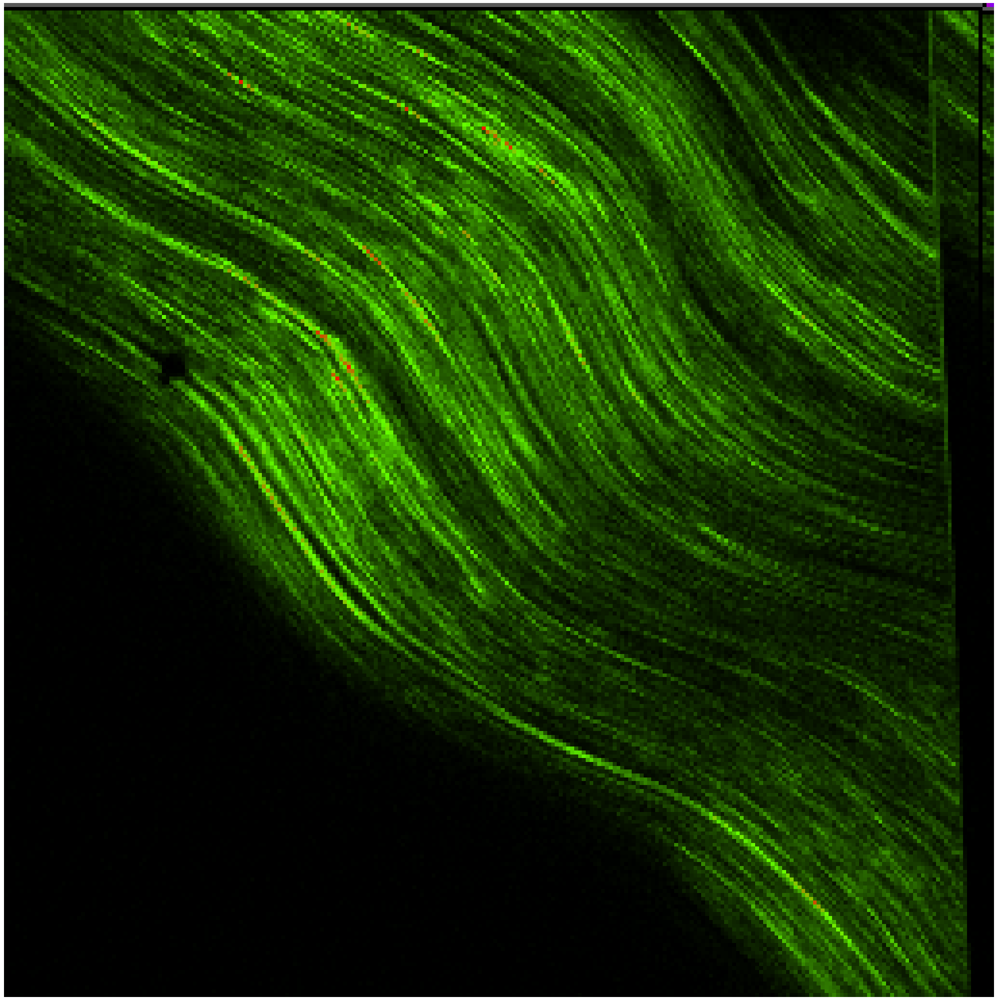
Although it was believed that SHG would require a non-centrosymmetric crystal, there were observations of the effect on centrosymmetric crystals. However, it was later understood that this effect arose because of the surface sensitivity of SHG. SHG has been used on metal and Insulator surfaces and even at the air water interface.

Perhaps even more interesting that SHG is the use of sum frequency generation on surfaces to detect vibrational spectra of surface attached molecules, e.g. self-assembled monolayers, Langmuir-Blodgett bilayers etc.

SHG microscopy

In biological and medical science, the effect of second harmonic generation is used for high-resolution optical microscopy. Due to the phase-matching condition only non-centrosymmetric structures are capable of producing SHG light. One such structure is collagen, which is found in most load-bearing tissues. Using a short-pulse laser such as a femtosecond laser and a set of appropriate filters the excitation light can be easily separated from the emitted, frequency-doubled SHG signal. This allows for very high axial and lateral resolution comparable to that of confocal microscopy without having to use pinholes.

Image of collagen



Texas A&M Imaging Center