

Non linear optics and second harmonic generation

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Electric polarization:

Polarization is the vector field that expresses the density of permanent or induced electric dipole moments in a dielectric material. When a dielectric is placed in an external electric field, its molecules gain electric dipole moment and the dielectric is said to be polarized. The electric dipole moment induced per unit volume of the dielectric material is called the electric polarization of the dielectric.

Electric susceptibility

In electricity, the electric susceptibility is a dimensionless proportionality constant that indicates the degree of polarization of a dielectric material in response to an applied electric field.

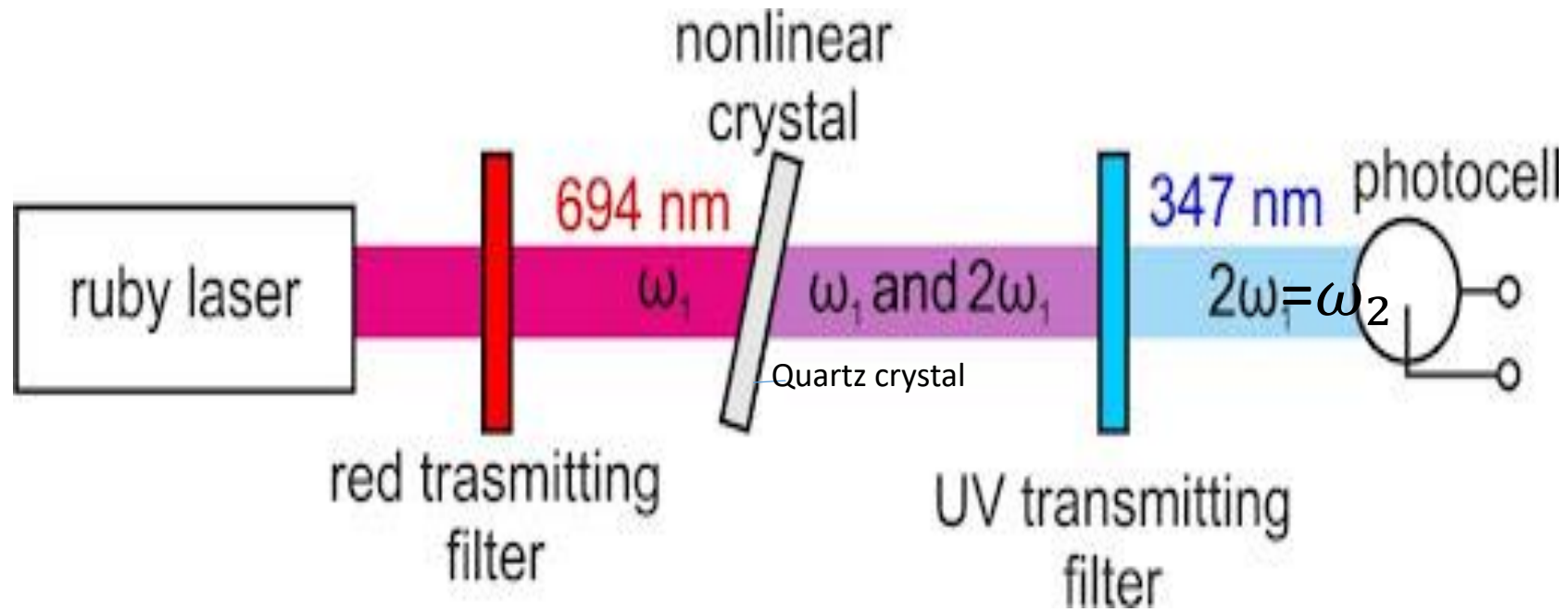
Permittivity is the ability to resist external electric field. This means a substance with high permittivity requires high external electric field to polarize. On the other hand, susceptibility is defined as the ability to polarize. So a substance with higher susceptibility should polarize easily. This means both the quantities are inversely proportional to each other but mathematically they are linearly dependant.

Nonlinear optics

Nonlinear optics is the branch of optics that describes the behaviour of light in nonlinear media. It happens with very intense light. In a non linear medium polarization density P responds non-linearly to the electric field E of the light.

A conventional light source of low intensity are unable to change the optical parameter of the medium. But for a moderately powerful laser of electric field strength 3×10^8 v/m which is within the range of atomic field, the relation between the electric polarisation vector \mathbf{P} and field strength E ceases to be linear and some non linear effect happen which leads to non linear optics.

Experimental arrangement for non linear optics



A ruby laser beam of wavelength 6943 \AA with average power 10 kw is focused on a quartz crystal slab. The transmitted light is then allowed to pass through a filter which cuts the red light and allows to pass through it the UV light. This emerging light is then allowed to pass through a photo cell. The transmitted light from the cell is found to be of wavelength 3471 \AA .

Theory:

A dielectric molecule when placed in an electric field, each molecule behaves as a dipole with dipole moment p_i . So the dipole moment vector per unit volume is given by

$$P_i = \sum p_i \dots \dots \dots (1)$$

We know that \mathbf{P} depends on the properties of medium and on the electric field strength

$$\therefore \mathbf{P} = \epsilon_0 \chi \mathbf{E} \dots\dots\dots(2)$$

Here χ electric polarizability or electric susceptibility of the medium.

The value of χ is constant and its magnitude is a function of frequency and it is independent of electric field E .

For sufficiently intense light field the relation (2) generalised as

$$\mathbf{P} = \epsilon_0 [\chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{E}^2 + \chi^{(3)} \mathbf{E}^3 + \dots\dots\dots] \dots\dots(3)$$

Here $\chi^{(1)} = \chi$ and $\chi^{(2)}, \chi^{(3)}$ ----- are known as non linear susceptibilities.

In case of ordinary light only the first term retain but for intense light of high intensity electric field the higher order terms become significant.

Let us now consider that electric field equation of incident radiation is

$$E = E_0 \cos \omega t \text{ -----(4)}$$

Substituting this value in equation (3) and applying the trigonometric relation we get

$$P = \frac{1}{2} \epsilon_0 \chi^{(2)} E_0^2 + \epsilon_0 \left(\chi^{(1)} + \frac{3}{4} \chi^{(3)} E_0^2 \right) E_0 \cos \omega t + \frac{1}{2} \epsilon_0 \chi^{(2)} E_0^2 \cos 2\omega t + \frac{1}{4} \epsilon_0 \chi^{(3)} E_0^3 \cos 3\omega t + \text{-----} \text{ (5)}$$

Here 1st term represents the constant dc field across the medium.

The 2nd term is due to external polarisation and is called 1st or fundamental harmonics of polarisation, the third term is called as 2nd harmonic of polarisation etc.

Second-harmonic generation (SHG):

It is a nonlinear optical process in which two photons with the same frequency interact with a nonlinear material, are combined, and generate a new photon with twice the energy of the initial photons. So the frequency of the new photon is twice the original one. SHG was firstly demonstrated by Franken and his colleagues in 1961 (Franken et al., 1961).

A polarization wave oscillating at frequency 2ω , radiates wave of same characteristic and monochromaticity as the incident wave .

The non linear polarizability $\chi^{(2)}$ depends on the direction of propagation, polarization of the electric field and orientation of the optic axis of the crystal. Since vector P and E are not parallel χ must be treated as tensor. Then it can be represented by

$$P_i^{(2)} = \epsilon_0 \sum_{ijk} \chi_{ijk}^{(2)} E_j E_k$$

i, j, k represent the x, y, z co-ordinates.

The 2nd harmonic generation occurs only in certain types of crystal.

In isotropic medium 2nd harmonic generation does not occur.

Crystals that lack of inversion symmetry shows 2nd harmonic generation.

As for example, if we change the direction of x, y, z axes leaving electric field and dipole moment unchanged then we get

$$\begin{aligned} -P_i^{(2)} &= \epsilon_0 \sum_{j,k} \chi_{ijk}^{(2)} (-E_j)(-E_k) \\ &= + P_i^{(2)} \\ \therefore 2P_i^{(2)} &= 0 \quad \text{so} \quad P_i^{(2)} = 0 \end{aligned}$$

$\chi_{ijk}^{(2)} = 0$ which shows that 2nd harmonic generation can not occur in an isotropic medium.
