Nonlinear polarization of the medium

At the time of the pre-laser optics it was reasonably believed that the medium characteristics are independent of the intensity of light passing through this medium. Pre-laser light sources produced light field intensities not exceeding 10^5 V*/m*. Whereas, the intra-atomic fields are characterized by intensities of the order of 10^8 to 10^{12} *V/m* makes it evident why light waves of such sources could not produce a somewhat detectable effect on the atomic fields and, consequently, on the medium behaviour. Therefore, the medium response, in the form of polarization *P,* to the external perturbation, in the form of a light wave electric field *E,* turned out to be linear

$$P = \chi E$$

Where, χ is the dielectric susceptibility of the medium. This relationship coined the term "linear optics" used in relation to the pre-laser (incoherent) optics. On the other hand, lasers yield light fields with intensities as high as 10^{10} to 10^{11} *V/m.* This electric field strength are comparable with those within the atom. Now the dielectric susceptibility of the medium becomes a function of the light field intensity as

$$\chi(E) = \chi_o + \chi_1 E + \chi_2 E^2 + \chi_3 E^3 + \chi_4 E^4 + \ldots..$$

Where χ_o, χ_1, χ_2, χ_3, χ_4….. etc. are the medium parameters which define its polarizability. The polarization P becomes

$$P=\chi E = \chi_0 E + \chi_1 E^2 + \chi_2 E^3 + \chi_3 E^4 + \ldots..$$

The polarization P becomes nonlinear in the light field intensity.

The nonlinear term $P_{NL} = \chi_1 E^2$ describes the nonlinear polarization of the medium, χ_1 being the nonlinear susceptibility.

Interaction of light waves in nonlinear media

Let a plane monochromatic light wave of frequency ν travel at velocity v in the z direction of a nonlinear medium (exhibiting a nonlinear polarization for this wave). The electric field strength of this wave may be defined as

$$E(z,t) = E_0 cos\left[2\pi\nu\left(t - \frac{z}{v}\right)\right]$$

The nonlinear polarization becomes

$$P_{NL}(z,t) = \frac{1}{2}\chi_1 E_0^2 + \frac{1}{2}\chi_1 E_0^2 cos\left[4\pi\nu\left(t - \frac{z}{v}\right)\right]$$

The second term of this equation indicates that there is a wave of polarization propagating in the medium in the same direction and at the same speed, but this wave oscillates at the twice higher frequency 2ν, rather than ν. This wave of polarization may be thought of as the kind of "radiation aerial" traversing the medium at velocity *v*. Under certain conditions this "aerial" may cause emission of a new light wave at the frequency of the wave of polarization. This is called second harmonic generation.

Consider now the case with two waves, one at frequency ν_1 and the other at frequency ν_2 being launched into the non-linear medium.

$$E_1 = E_{01}\cos\left\{2\pi\nu_1\left(t - z/v_1\right)\right\}$$

$$E_2 = E_{02}\cos\left\{2\pi\nu_2\left(t - z/v_2\right)\right\}$$

$$E(z,t) = E_{01}\cos\left\{2\pi\nu_1\left(t - z/v_1\right)\right\} + E_{02}\cos\left\{2\pi\nu_2\left(t - z/v_2\right)\right\}$$

Nonlinear polarization P_{NL}

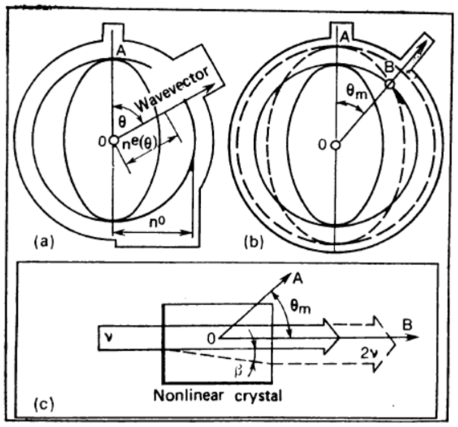
$$P_{NL}(z,t) = \varepsilon_0\chi_1\left[E_{01}\cos\left\{2\pi\nu_1\left(t - z/v_1\right)\right\} + E_{02}\cos\left\{2\pi\nu_2\left(t - z/v_2\right)\right\}\right]^2$$

$$= \frac{1}{2}\varepsilon_0\chi_1 E_{01}^2 + \frac{1}{2}\varepsilon_0\chi_1 E_{02}^2 + \frac{1}{2}\varepsilon_0\chi_1 E_{01}^2\cos\left\{2(2\pi)\nu_1\left(t - z/v_1\right)\right\}$$
$$+ \frac{1}{2}\varepsilon_0\chi_1 E_{02}^2\cos\left\{2(2\pi)\nu_2\left(t - z/v_2\right)\right\}$$
$$+ \varepsilon_0\chi_1 E_{01}E_{02}\cos\left\{2\pi(\nu_1 + \nu_2)t - 2\pi\left(\frac{\nu_1}{v_1} + \frac{\nu_2}{v_2}\right)z\right\}$$
$$+ \varepsilon_0\chi_1 E_{01}E_{02}\cos\left\{2\pi(\nu_1 - \nu_2)t - 2\pi\left(\frac{\nu_1}{v_1} - \frac{\nu_2}{v_2}\right)z\right\}$$

The interaction of light waves at frequencies ν_1 and ν_2 launched in a material with the nonlinear polarization may cause generation of new light waves at the sum (ν_1+ν_2), difference (ν_1-ν_2) and double $2\nu_1$ and $2\nu_2$ frequencies.

Second harmonic generation in nonlinear crystals

Under certain conditions the wave of nonlinear polarization may give birth to the second optical harmonic, a generated light wave at frequency 2ν. The polarization wave propagates in the medium at the velocity $v_I = c/n(\nu)$, while the second harmonic propagates at the velocity $v_2 = c/n(2\nu)$. $n(\nu)$ and $n(2\nu)$ are the refractive indices of the medium for frequency ν and 2ν. For the transfer of energy from the polarization wave to the new light wave to be efficient, the waves must be matched in velocity. This leads to the condition n (v) = n (2v), referred to as the *phase-matching condition.* The refractive index depends on the direction in the anisotropic crystal. A light wave launched in an anisotropic crystal splits into two waves travelling at different velocities. In a large group of anisotropic crystals, called uniaxial crystals, one of these light waves is called the *ordinary wave;* its refractive index is independent of the direction of propagation. The other light wave is called the *extraordinary wave* and the corresponding refractive index depends on the direction of propagation. The different refractive index behaviour in an anisotropic crystal is usually described in terms of the so-called *index ellipsoid.* Figure a shows a section through the surfaces of refractive indices for the ordinary (sphere) and extraordinary (ellipsoid) waves. The ellipsoid axis *OA* is the optic axis of the uniaxial crystal. As can be seen, the refractive indices are the same along the optic axis, therefore a light wave travelling in this direction does not split into the ordinary and extraordinary waves. In case the wave-vector forms an angle θ with the crystal optic axis *OA,* the "splitting" does take place, as away from the axis the ordinary refractive index is n_O and the extraordinary refractive index is



$n^e(\theta)$.The extraordinary wave is polarized in the plane passing through the wave vector and the optic axis (exactly this plane is shown in Figure a), whereas the ordinary wave is polarized normally to this plane. The section through the surfaces of refractive index, shown in figure a, corresponds to a certain frequency of the incident light. Suppose now that this frequency doubles. The refractive index normally increases with frequency. Therefore, the dimensions of the refractive-index sphere and ellipsoid increase accordingly. Figure b shows for comparison the sections through these surfaces plotted for a frequency ν (solid lines) and the doubled frequency 2ν (dashed lines). The dashed ellipse is seen to intersect with the solid circle; one of the points of intersection is point *B*. This means that for light waves propagating in the *OB* direction (i.e, close to the cone where *OB* is a generating element) the phase-matching condition is satisfied n_o (ν) = n_e (2ν) The cone angle θ_m is obviously the phase-matching angle. For all directions lying on this cone the ordinary refractive index at frequency ν equals the extraordinary refractive index at frequency 2ν.

Examples of inorganic nonlinear crystals

Material	Symbol	Formula
Potassium dihydrogen phosphate	KDP	KH_2PO_4
Ammonium dihydrogen phosphate	ADP	$NH_4H_2PO_4$
Cesium dihydrogen arsenate	CDA	CsH_2AsO_4
Cesium dideuterium arsenate	D-CDA	CsD_2AsO_4
Lithium niobate	—	$LiNbO_3$
Lithium iodate	—	$LiIO_3$
Barium-sodium niobate	—	$Ba_2NaNb_5O_{15}$

Self-focusing of light in a nonlinear medium

For an isotropic medium or crystal having a centre of symmetry

$$P(-E) = -P(E)$$

This means that all the terms containing an even number of multipliers E must vanish

$$P = \varepsilon_0 \chi E + \varepsilon_0 \chi_1 E^2 + \varepsilon_0 \chi_2 E^3 + \ldots\ldots\ldots\ldots\ldots\ldots$$

Hence the first nonlinear correction is given by the cubic term and the polarization assumes the form

$$\begin{aligned} P &= \varepsilon_0 \chi E + \varepsilon_0 \chi_3 E^3 \\ &= \varepsilon_0 \chi E + aE^3 \end{aligned}$$

This gives

$$\begin{aligned} D &= \varepsilon E \\ &= \varepsilon_0 E + P \\ &= \varepsilon_0 E + \varepsilon_0 \chi E + aE^3 \\ &= \left[\varepsilon_0 (1+\chi) + aE^2\right] E \end{aligned}$$

$$\varepsilon = \varepsilon_0 (1+\chi) + aE^2$$

The refractive index, $n = \sqrt{\dfrac{\varepsilon}{\varepsilon_0}}$

$$= \sqrt{(1+\chi) + aE^2 / \varepsilon_0}$$

$$= \sqrt{(1+\chi)} \left(1 + \frac{aE^2}{2\varepsilon_0 (1+\chi)}\right)$$

$$\Rightarrow \qquad n = n_0 + n_2 E_0^2$$

Hence, the square of the electric field strength has been averaged over a period of oscillation and hence E_0 is the amplitude of the electric field strength of the wave.

Self-focusing length

The self-focusing length is the distance l over which the beam is focused at the axis or collapses to the axis in a nonlinear medium

Let,

'a' is the radius of a laser beam

'E_0' is the amplitude of the electric field strength at the axis

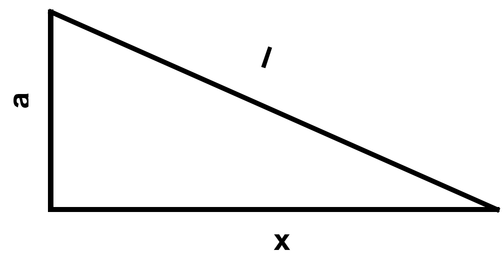
The amplitude at a distance 'a' from the axis is assumed to be equal to zero

A phase difference, over the path difference *l* is

$$= \frac{2\pi}{\lambda} l\,\Delta n \qquad \Delta n = n_2 E_2^2$$

$$= \frac{2\pi\nu}{c} l\,\Delta n$$

$$= \frac{\omega\,\Delta n}{c} l$$



Appears between the axis ray and the extreme ray is b = l - x

$$b = l - \sqrt{l^2 - a^2}$$

$$= l - l\left(1 - \frac{a^2}{l^2}\right)^{1/2}$$

$$= l - l\left(1 - \frac{a^2}{2l^2}\right) \quad \because \quad \frac{a}{l} \ll 1$$

$$= l - l + \frac{a^2}{2l}$$

$$= \frac{a^2}{2l}$$

Phase difference for b is $= \frac{2\pi}{\lambda}(n_0 b) = \frac{2\pi\nu}{c} n_0 b$

$$= \frac{\omega n_0 b}{c} = \frac{\omega n_0 a^2}{2lc} \qquad \because b = \frac{a^2}{2l}$$

The phase difference for two paths must be equal

$$\therefore \frac{\omega\,\Delta n\, l}{c} = \frac{\omega n_0 a^2}{2lc}$$

$$\Rightarrow \qquad \Delta n\, l = \frac{n_0 a^2}{2l}$$

$$\Rightarrow \qquad n_2 E_0^2\, l = \frac{n_0 a^2}{2l}$$

$$\Rightarrow \qquad l^2 = \frac{n_0 a^2}{2 n_2 E_0^2}$$

$$\Rightarrow \qquad l = \frac{a}{E_0}\sqrt{\frac{n_0}{2 n_2}}$$