Non-linear Optics I
(Electro-optics)

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Domain of Linear Optics

From electromagnetism courses we recall

$$\mathbf{D} = \varepsilon_0 \varepsilon_r \mathbf{E} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_0 \mathbf{E}(1+\chi) \tag{1}$$

Also at optical frequencies,

$$n = \sqrt{\varepsilon_r} = (1+\chi)^{1/2} \sim 1 + \frac{1}{2}\chi .... \tag{2}$$

$$P_i = \varepsilon_0 \sum \chi_{ij} E_j \tag{3}$$

The medium may not be isotropic and homogeneous; the polarisation P will not in general be collinear with E, and the susceptibility $\chi^{(n)}$ and the permittivity $\overline{\overline{\varepsilon}}$ are tensors (in this case of rank 2)

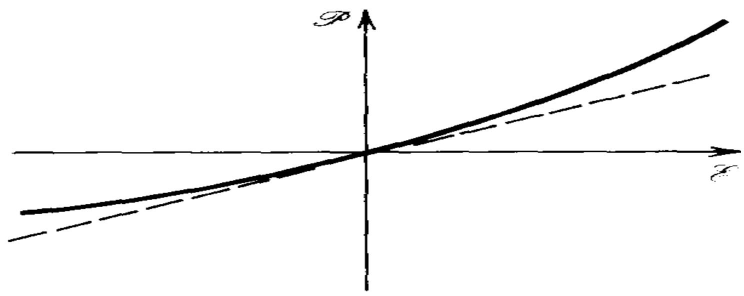

Figure 1: Linear versus nonlinear electric field effects

Domain of Non-linear Optics

$$P(\omega) = \varepsilon_0 \sum [\chi^{(1)}_{ij} E_j(\omega_1) + \chi^{(2)}_{ijk} E_j(\omega_1) E_k(\omega_2) + \chi^{(3)}_{ijk\ell} E_j(\omega_1) E_k(\omega_2) E_\ell(\omega_3) ......] \tag{4}$$

Typical values for the second order coefficient $d = \chi^{(2)}/2\varepsilon_0 = 10^{-24}$ to 10^{-21} AsV^{-2}. Typical values for the third order non-linear susceptibility $\chi^{(3)}$ is 10^{-29} to 10^{-34} (MKS units) for glasses, crystals, semiconductors and organics materials of interest. Only crystals with *NO* centre of symmetry[1] have a finite second order susceptibility; for other materials the first non-linear coefficient is $\chi^{(3)}$

[1] If a crystal possesses inversion symmetry the application of an electric field E along some direction causes a change $\Delta n = sE$ in the index. If the direction of the field is reversed the change becomes $\Delta n = s[-E]$, but inversion symmetry requires the two directions to be physically equivalent. This requires $s = -s$ which is possible only for $s = 0$. Thus, linear, Pockels cystals require NO centre of symmetry. Note also that these crystal are piezo-electric.

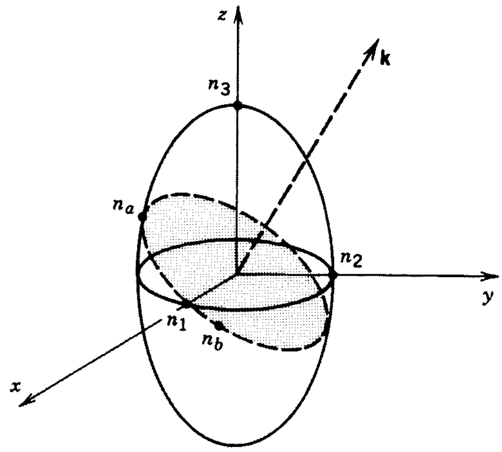

Figure 2: Index ellipsoid

Index Ellipsoid for a Uniaxial System

The optical properties of an anisotropic medium can be characterised by a geometric construction called the index ellipsoid where $\overline{\overline{\eta}}$ is the so-called impermeability tensor related to the refractive index as given above. The principal axes of the ellipse are the optical principal axes; the principal dimensions along these axes are the principal refractive indices: n_1, n_2, n_3. (Note also that the phase velocity of the wave is proportional to $1/n$). *Uniaxial* means $n_x = n_y \neq n_z$ (the optical axis). See appendix.

$$\frac{x^2+y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1 \tag{5}$$

Thus for an arbitrary angle θ to the $z-$axis as shown,

$$n(\theta) = \left\{ \frac{\cos^2\theta}{n_0^2} + \frac{\sin^2\theta}{n_e^2} \right\}^{-1/2} \tag{6}$$

Linear Electro-optic Effect (Pockels).

When a steady electric field E with components (E_1, E_2, E_3) is applied to the crystal the elements of the tensor $\overline{\overline{\eta}}$ are altered so that each of the 9 elements becomes a function of E and the ellipsoid changes shape. Thus, in equation 4 we let $E_k(\omega_2) = E^0$ - a d.c. electric field so that,

$$P(\omega) = \varepsilon_0[\chi_{ij}^{(1)} + \chi_{ijk}^{(2)}E^0]E(\omega) \tag{7}$$

Since χ is related to ϵ, the equation can be re-written in terms of the refractive index where each of the elements $\eta_{ij}(E)$, is a function of the appropriate field components, i.e.

$$\eta_{ij}(E) = \varepsilon_0/\varepsilon = 1/n^2 = \eta_{ij} + \textstyle\sum_k r_{ijk}E_k + \sum_{k,l} s_{ijkl}E_kE_l........$$

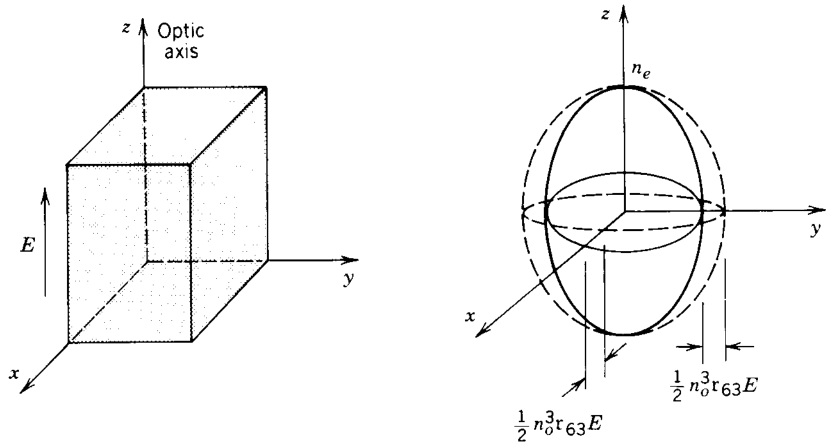

Figure 3: Index ellipsoid for a $\bar{4}$2m crystal

Terms linear in the applied field represent the Pockels effect; those quadratic in E represent the Kerr effect. Alternatively, we could write, by Taylor expansion,[2] the refractive index in the presence of an electric field as follows,

$$n(E) = n_0 + a_1 E + \tfrac{1}{2} a_2 E^2......$$

This introduces the connection between the linear electro-optic coefficients and the polarisation of the medium, see equation (9).

Linear Electro-optic Tensor

The change to the index ellipsoid when an electric field is applied can be written as follows,

$$\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} + \frac{2yz}{n_4^2} + \frac{2xz}{n_5^2} + \frac{2xy}{n_6^2} = 1 \tag{8}$$

Clearly if the indices $1, 2, 3,$...are chosen to be coincident with the principal dielectric axes equation 8 must reduce to equation 5 in the absence of the electric field, *i.e.* that $1/n_{4,5,6} = 0$

This introduces the linear electro-optic tensor r^{LEO}

$$\Delta\left(\frac{1}{n^2}\right) = \sum r_{ijk} E_k^0 \tag{9}$$

This is a $3\times 3\ \times 3$ matrix, *i.e.,* it has 27 elements. Of these, physical symmetry reduces the number to 18 independent elements, written as a 3×6 matrix. [$r_{ijk} = \partial\eta_{ij}/\partial E_k$ where $\eta = \varepsilon_0\varepsilon^{-1}$ and the index ellipsoid is given by $\sum \eta_{ij} x_i x_j = 1$ where $i, j = 1, 2, 3$ with principal indices of refraction n_1, n_2, n_3 (see footnote 2) and η is symmetric with respect to interchange of indices i, j. Thus, it follows r (and d) are also invariant under i, j interchange. It is therefore conventional to reduce the i, j index to one symbol I with the correspondence as given in the "look up" table 1]

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[2] By Taylor expanding the refractive index about $E = 0$ we can write
$n(E) = n_0 + a_1 E + \frac{1}{2} a_2 E^2$...
where the coefficients are derivatives of the refractive index with E in the normal way. Defining $r = -2a_1/n^3$ and $s = -a_2/n^3$ we have for $\eta = \varepsilon_0/\varepsilon = 1/n^2$ the following field dependent change $\Delta\eta = (d\eta/dn)\Delta n = (-2/n^3)(-\frac{1}{2}rn^3E - \frac{1}{2}sn^3E^2..)$.

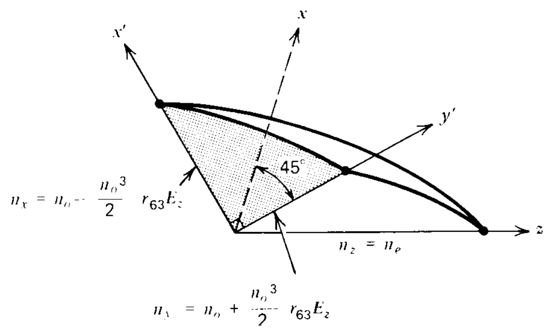

Figure 4: Rotation of axes by 45^0 about the optical axis.

$j \downarrow$ i$\longrightarrow$	1	2	3
1	1	6	5
2	6	2	4
3	5	4	3

Table 1 Look up table for $i, j \longrightarrow I$

Any particular Pockels crystal will further reduce the number of non-zero elements as follows. As an example, consider the uniaxial crystal ADP (ammonium dihydrogen phosphate) which has tetragonal ($\bar{4}$2m) symmetry. The index ellipsoid (see figure 3) is represented by

$$\begin{bmatrix} \Delta\left(\frac{1}{n^2}\right)_1 \\ \Delta\left(\frac{1}{n^2}\right)_2 \\ \Delta\left(\frac{1}{n^2}\right)_3 \\ \Delta\left(\frac{1}{n^2}\right)_4 \\ \Delta\left(\frac{1}{n^2}\right)_5 \\ \Delta\left(\frac{1}{n^2}\right)_6 \end{bmatrix} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{52} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \begin{bmatrix} E_1^0 \\ E_2^0 \\ E_3^0 \end{bmatrix} \tag{10}$$

The crystal is now *biaxial.*

Or in terms of the polarisation of the medium[3], which we shall use for optical fields in harmonic generation,

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix} \begin{bmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix} \tag{11}$$

If we take as the direction of the applied d.c. field $E^0 = E_z^0 = E_3^0$ then the new index ellipsoid will given by

$$\frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} + 2r_{63}xyE_z^0 = 1 \tag{12}$$

[3] The coefficients d and r are related as follows: $d = \frac{\varepsilon_0\chi^{(2)}}{2}$and $r \sim -\frac{4d}{\varepsilon_0 n^4}$ Be careful about factors of 2 arising from the use of a complex field. For the Pockels case let the d.c. and optical fields be represented as $E(t) = E^0 + \mathrm{Re}\{E(\omega)\exp(-i\omega t)\}$.For the case of H.G. let the coupled optical fields be represented as $E(t) = \mathrm{Re}\{E(\omega_1)\exp(-i\omega_1 t) + E(\omega_2)\exp(-i\omega_2 t)\}$.

For S.H.G. in particular let $\omega_1 = \omega_2$

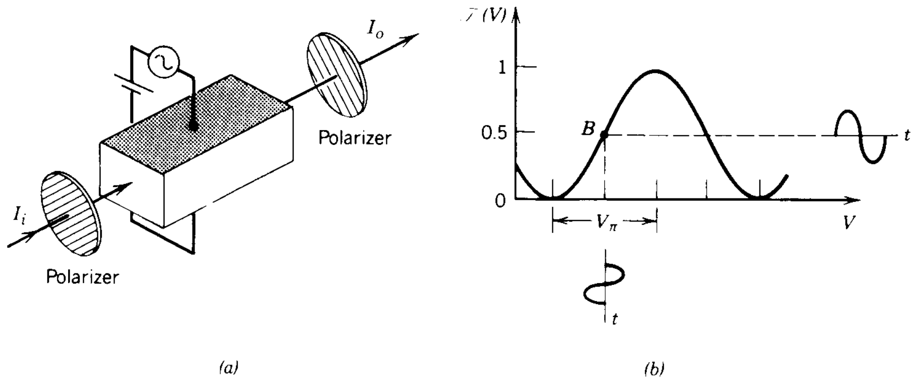

Figure 5: Electro-optic modulator used as an intensity modulator.

A rotation of the axes by $+45^0$ to (x', y', z') then transforms the ellipsoid equation as follows

$$\left(\frac{1}{n_0^2} + r_{63}E_z^0\right) x'^2 + \left(\frac{1}{n_0^2} - r_{63}E_z^0\right) y'^2 + \frac{z'^2}{n_e^2} = 1 \tag{13}$$

Thus, given that $r_{63}E_z^0 << n_o^{-2}$ and that $n_{x'}^{-2} = n_o^{-2} + r_{63}E_z^0$ which gives $n_{x'} = n_0(1 + n_0^2 r_{63} E_z^0)^{-1/2}$ and similarly for $n_{y'}$ we have finally

$$\Delta n = |n_{x'} - n_{y'}| = n_0^3 r_{63} E_z^0 \tag{14}$$

To act as a half-wave plate the phase induced by the field must be π radians, so

$$\phi = \frac{2\pi}{\lambda}\Delta n d = \pi \tag{15}$$

and the half-wave voltage is

$$V_\pi = \frac{\lambda}{2n_0^3 r_{63}} \tag{16}$$

Appendix

Wave-vector surface

Linear but tensorial χ

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \tag{17}$$

The wave equation including polarisation but not conduction currents J is of the form

$$\boldsymbol{\nabla}\times(\boldsymbol{\nabla}\times\mathbf{E}) + \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = -\frac{1}{c^2}\boldsymbol{\chi}\frac{\partial^2\mathbf{E}}{\partial t^2} \tag{18}$$

The transparent, insulating crystal can thus sustain a plane monochromatic wave ($E_0 \exp i\{\mathbf{k}.\mathbf{r} - \omega t\}$) provided the propagation vector satisfies the equation

$$\mathbf{k}\times(\mathbf{k}\times\mathbf{E}) + \frac{\omega^2}{c^2}\mathbf{E} = -\frac{\omega^2}{c^2}\boldsymbol{\chi}\mathbf{E} \tag{19}$$

The cartesian components of this equation are thus

$$\left(-k_y^2 - k_z^2 + \frac{\omega^2}{c^2}\right)E_x + k_xk_yE_y + k_xk_zE_z = -\frac{\omega^2}{c^2}\chi_{11}E_x \tag{20}$$

and similarly for $y-$ and $z-$ components.
To interpret this result let the wave propagate along one of the principal axes of the crystal, say z. In this case $k_z = k$ and $k_x = k_y = 0$ and the components become

$$\left(-k^2 + \frac{\omega^2}{c^2}\right)E_x = -\frac{\omega^2}{c^2}\chi_{11}E_x \tag{21}$$

$$\left(-k^2 + \frac{\omega^2}{c^2}\right)E_y = -\frac{\omega^2}{c^2}\chi_{22}E_y \tag{22}$$

$$\frac{\omega^2}{c^2}E_z = -\frac{\omega^2}{c^2}\chi_{33}E_z \tag{23}$$

The last equation suggests $E_z = 0$ because neither χ or ω is zero. The wave is transverse. On the other hand, the first two equations show

$$k = \frac{\omega}{c}\sqrt{1+\chi_{11}} = \frac{\omega}{c}\sqrt{K_{11}} = \frac{\omega}{c}n_1 \tag{24}$$

$$k = \frac{\omega}{c}\sqrt{1+\chi_{22}} = \frac{\omega}{c}\sqrt{K_{22}} = \frac{\omega}{c}n_2 \tag{25}$$

where n_1 , n_2 and n_3 are the principal indices of refraction.

Now the equations for the components [20] lead to the following condition for non-trivial solution for the field components not to vanish, *i.e.*

$$\begin{vmatrix} (\frac{n_1\omega}{c})^2 - k_y^2 - k_z^2 & k_xk_y & k_xk_z \\ k_yk_x & (\frac{n_2\omega}{c})^2 - k_x^2 - k_z^2 & k_yk_z \\ k_zk_x & k_zk_y & (\frac{n_3\omega}{c})^2 - k_x^2 - k_y^2 \end{vmatrix} = 0 \tag{26}$$

This equation gives the wave-vector surface for propagation in the crystal. Thus, for example in the $k_z = 0$ plane the determinant gives a product of two factors either or both of which must reduce to zero. This condition gives

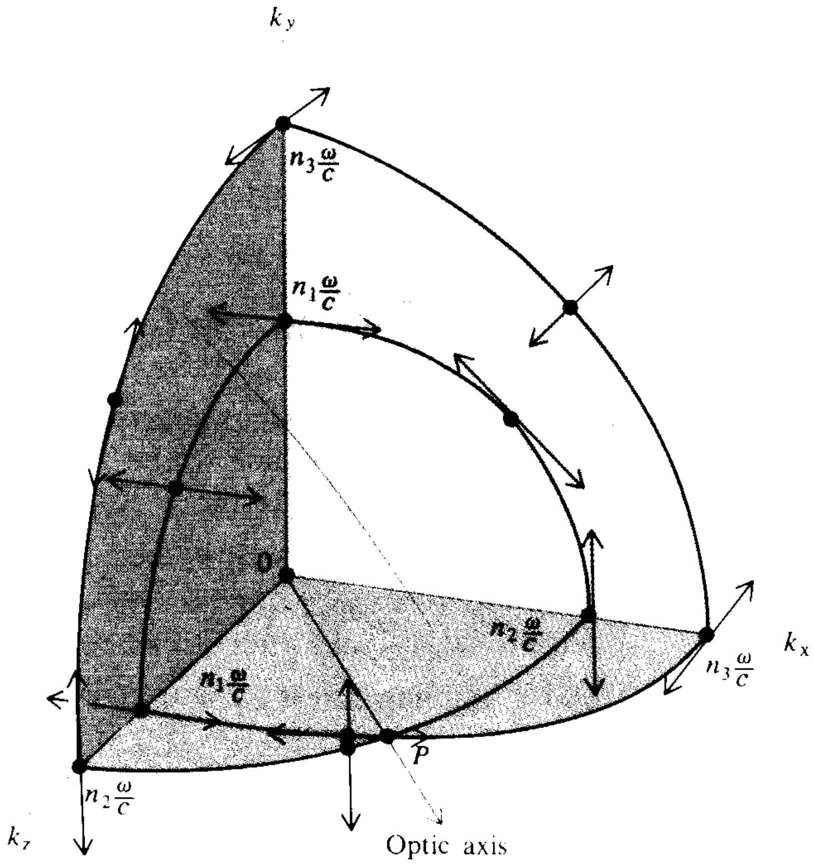

Figure 6: Wave-vector surface for an anisotropic crystal

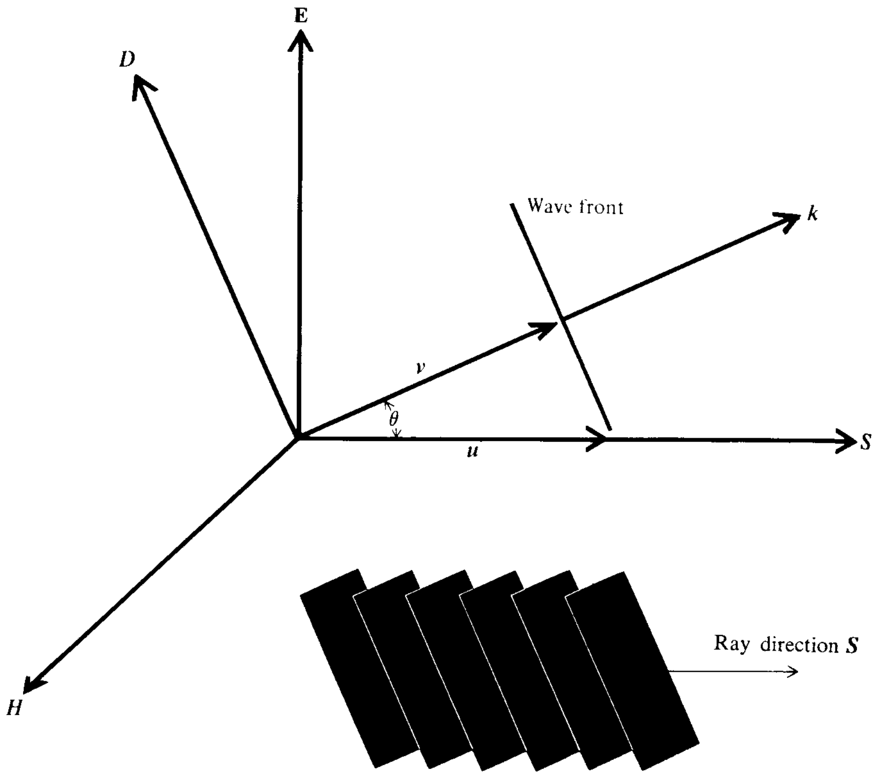

Figure 7: Walk off. Poynting's vector and k are no longer collinear

$$k_x^2 + k_y^2 = \left(\frac{n_3\omega}{c}\right)^2 \tag{27}$$

$$\frac{k_x^2}{(n_2\omega/c)^2} + \frac{k_y^2}{(n_1\omega/c)^2} = 1 \tag{28}$$

which can be seen as the equation of a circle and an ellipse respectively. For a *uniaxial* crystal $n_1 = n_2 \neq n_3$ while for *biaxial* crystal all principal indices are different.

Recognising that $\mathbf{k} = \mathbf{v}(\omega/v^2)$ we can derive the corresponding determinant equation and construct the phase-velocity surface. Finally we can consider the ray-velocity defined by considering the propagation of a narrow beam of light in the crystal. The surfaces of constant phase with velocity u given by

$$u = \frac{v}{\cos\theta} \tag{29}$$

where θ represents the angle between Poynting vector $\mathbf{S}$ (which gives energy flow) and the $\mathbf{k}-$vector. When we come to discussing harmonic frequency generation in crystals this effect will be referred to as *walk off*.

References

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