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)

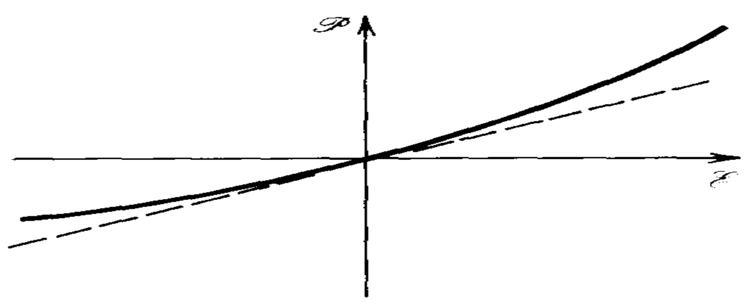

Figure 1: Linear versus nonlinear electric field effects

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 crystals are piezo-electric.

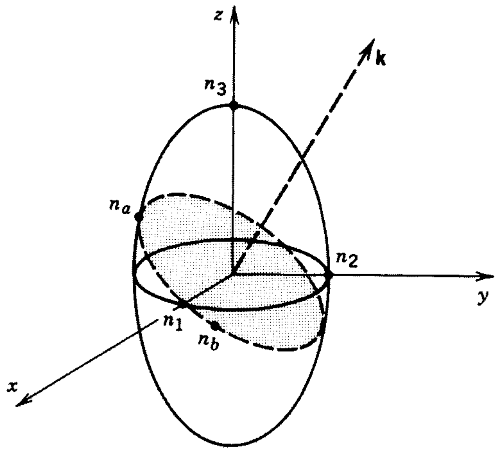

Figure 2: Index ellipsoid

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). In the appendix we give an alternative way of looking at the variation of the refractive index with direction in term of the k-vector surface; here the general idea is to compare plane waves of equal energy density travelling in different directions in a crystal. In this description we recognise that for the energy density to remain constant the lengths of the vectors **D** and **E** must change accordingly; the connection with energy density U_E is given by the expression

$$U_E = \frac{1}{2}\mathbf{E} \cdot \mathbf{D} \tag{5}$$

Making use of equation 1 we get

$$U_E = \frac{\mu_r}{2\varepsilon_0}\left[\frac{D_x^2}{n_x^2} + \frac{D_y^2}{n_y^2} + \frac{D_z^2}{n_z^2}\right] \tag{6}$$

which on re-arranging gives

$$\frac{\mu_r}{2U_E\varepsilon_0}\left[\frac{D_x^2}{n_x^2} + \frac{D_y^2}{n_y^2} + \frac{D_z^2}{n_z^2}\right] = 1 \tag{7}$$

If we now make a variable substitution $x^2 \equiv \left(\mu_r D_x^2/2U_E\varepsilon_0\right)$ and recall that we are dealing with a uniaxial crystal then we arrive at the following equation for the index ellipsoid.

$$\frac{x^2 + y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1 \tag{8}$$

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

$$n_e(\theta) = \left\{\frac{\cos^2\theta}{n_0^2} + \frac{\sin^2\theta}{n_e^2}\right\}^{-1/2} \tag{9}$$

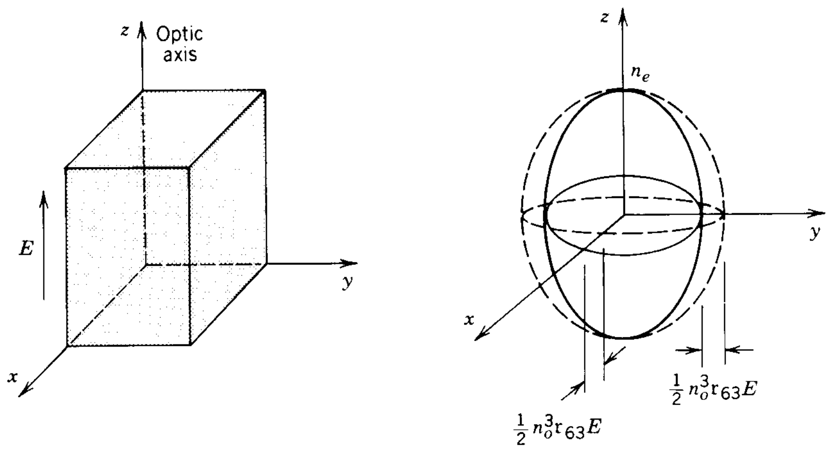

Figure 3: Index ellipsoid for a $\bar{4}$2m crystal

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,

$$P(\omega) = \varepsilon_0[\chi_{ij}^{(1)} + \chi_{ijk}^{(2)} E^0]E(\omega) \tag{10}$$

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.

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

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,

$$n(E) = n_0 + a_1E + \tfrac{1}{2}a_2E^2......$$

This introduces the connection between the linear electro-optic coefficients and the polarisation of the medium, see equation 10.

Linear Electro-optic Tensor

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

$$\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} + \frac{2yz}{n_4^2} + \frac{2xz}{n_5^2} + \frac{2xy}{n_6^2} = 1 \tag{11}$$

[2] By Taylor expanding the refractive index about $E = 0$ we can write
$n(E) = n_0 + a_1E + \frac{1}{2}a_2E^2$...
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 $\Delta\eta = (d\eta/dn)\Delta n = (-2/n^3)(-\frac{1}{2}rn^3E - \frac{1}{2}sn^3E^2..)$ with $\eta\,(E) = \eta + rE$

Clearly if the indices $1, 2, 3$,...are chosen to be coincident with the principal dielectric axes equation 11 must reduce to equation 8 in the absence of the electric field, *i.e.* that $1/n^2_{4,5,6} = 0$

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

$$\Delta\left(\frac{1}{n^2}\right) = \sum r_{ijk} E^0_k \tag{12}$$

This is a $3\times3\times3$ 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]

$j\downarrow$ 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

$$\begin{bmatrix} \Delta\left(\frac{1}{n^2}\right)_1 \\ \Delta\left(\frac{1}{n^2}\right)_2 \\ \Delta\left(\frac{1}{n^2}\right)_3 \\ \Delta\left(\frac{1}{n^2}\right)_4 \\ \Delta\left(\frac{1}{n^2}\right)_5 \\ \Delta\left(\frac{1}{n^2}\right)_6 \end{bmatrix} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{52} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \begin{bmatrix} E^0_1 \\ E^0_2 \\ E^0_3 \end{bmatrix} \tag{13}$$

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,

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix} \begin{bmatrix} E^2_x \\ E^2_y \\ E^2_z \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix} \tag{14}$$

If we take as the direction of the applied d.c. field $E^0 = E^0_z = E^0_3$ then the new index ellipsoid will given by

$$\frac{x^2}{n^2_0} + \frac{y^2}{n^2_0} + \frac{z^2}{n^2_e} + 2r_{63}xyE^0_z = 1 \tag{15}$$

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[3] The coefficients d and r are related as follows: $d = \dfrac{\varepsilon_0\chi^{(2)}}{2}$and $r \sim -\dfrac{4d}{\varepsilon_0 n^4}$ 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 $\omega_1 = \omega_2$

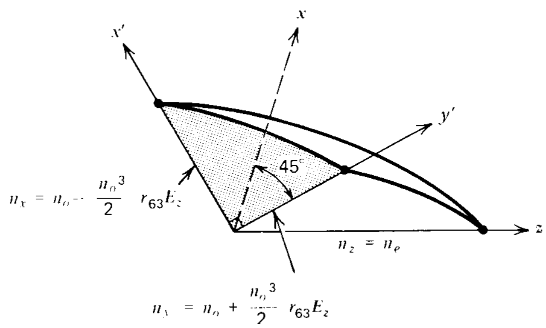

Figure 4: Rotation of axes by 45^0 about the optical axis.

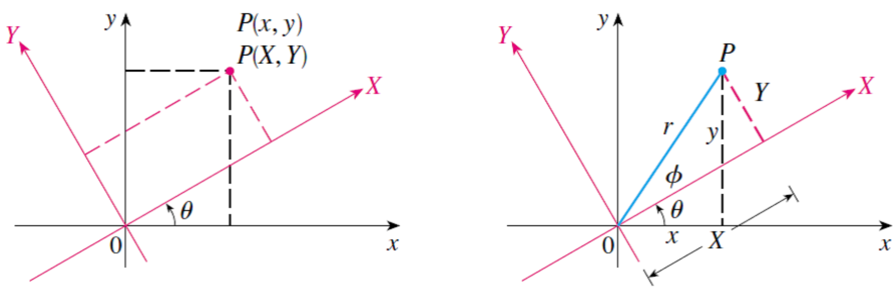

Figure 5: Rotation of Axes

A clockwise rotation take axes XY onto xy:(Equivalently a positive angle to the positive x-axis amounts to an anticlockwise rotation). The relationship between the different co-ordinate systems for a 45^0 rotation is given by simple trigonometry as follows:

$$\begin{pmatrix} x \\ y \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} X \\ Y \end{pmatrix} \tag{16}$$

or,

$$\begin{pmatrix} X \\ Y \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} x \\ y \end{pmatrix} \tag{17}$$

In the present case x, y represent the original axes which are transformed to x', y' ($\equiv X, Y$ in the figure) by an anticlockwise rotation. Thus inserting.

$$x = 1/\sqrt{2}\left(x' - y'\right) \text{ and } y = 1/\sqrt{2}\left(x' + y'\right) \tag{18}$$

into the equation for the ellipsoid 15 we have

$$\frac{(x' - y')^2}{2n_o^2} + \frac{(x' + y')^2}{2n_o^2} + \frac{2\left(x' - y'\right)\left(x' + y'\right)}{2} r_{63} E_z^0 + \frac{z^2}{n_e^2} = 1 \tag{19}$$

which when rearranged gives

$$\frac{x'^2}{n_o^2} + \frac{y'^2}{n_o^2} + \left(x'^2 - y'^2\right) r_{63} E_z^0 + \frac{z^2}{n_e^2} = 1 \tag{20}$$

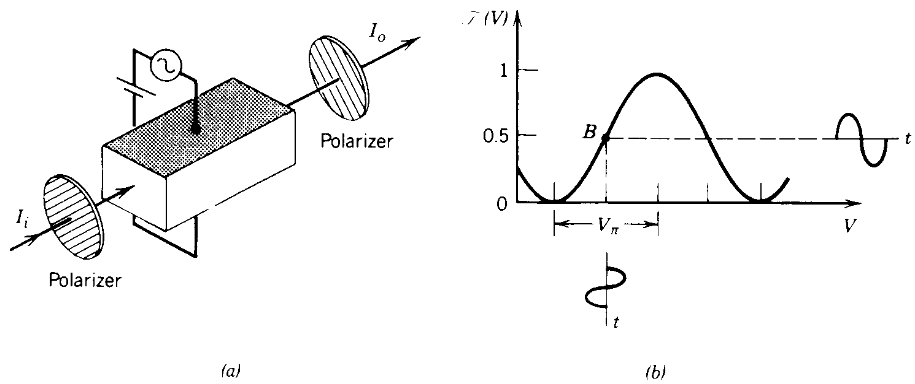

Figure 6: Electro-optic modulator used as an intensity modulator.

leading to

$$\frac{x'^2}{n_o^2}\left(1+n_o^2 r_{63} E_z^0\right)+\frac{y'^2}{n_o^2}\left(1-n_o^2 r_{63} E_z^0\right)+\frac{z^2}{n_e^2}=1 \tag{21}$$

This identifies

$$\frac{1}{n_{x'}^2}=\frac{\left(1+n_o^2 r_{63} E_z^0\right)}{n_o^2} \tag{22}$$

Or equivalently

$$n_{x'}^2=\frac{n_o^2}{(1+n_o^2 r_{63} E_z^0)} \tag{23}$$

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

$$\Delta n = |n_{x'} - n_{y'}| = n_0^3 r_{63} E_z^0 \tag{24}$$

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

$$\phi = \frac{2\pi}{\lambda}\Delta n d = \pi \tag{25}$$

and the half-wave voltage is

$$V_\pi = \frac{\lambda}{2n_0^3 r_{63}} \tag{26}$$

Appendix

Wave-vector surface

We consider here the form of the wave equation for the propagation of a plane wave in transparent, lossless, crystalline material. We start by noting the relationship between the displacement vector D and the electric field E which are connected by the tensorial permittivity $\bar{\bar{\varepsilon}}$;. for simplicity we take the principal axis system such that,

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \begin{bmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \tag{27}$$

where $n_i^2 \equiv (\varepsilon_{ii}/\varepsilon_0)$ are the principal refractive indices. By convention for a uniaxial crystal $n_x = n_y$; this index is referred to as the ordinary refractive index while n_z is referred to as the extraordinary index. If $n_z > n_0$ $(= n_x = n_y)$ the crystal is said to be a positive uniaxial and for the converse the crystal is called a negative uniaxial.

To solve the wave equation in this more general case we assume a plane monochromatic wave of form

$$E = E_0 \exp i\left(\mathbf{k}.\mathbf{r} - \omega t\right) \tag{28}$$

The operators ∇ and $(\partial/\partial t)$ in the wave equation act on the fields such that

$$\nabla \rightarrow i\mathbf{k} \tag{29}$$

$$\partial/\partial t \rightarrow -i\omega \tag{30}$$

furthermore the wavevector can be written as

$$\mathbf{k} = \frac{n\omega}{c}\widehat{k} \tag{31}$$

where the index of refraction n depends on the orientation of the material and the polarisation state of the electric field. With such monochromatic waves Maxwell's equations in terms of the complex amplitudes become

$$\nabla.\mathbf{D} = \mathbf{0} \Rightarrow \mathbf{k}.\mathbf{D} = \mathbf{0} \tag{32}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \Rightarrow \frac{n}{c}\widehat{k}\times\mathbf{E} = \mathbf{B} \tag{33}$$

$$\nabla.\mathbf{B} = \mathbf{0} \Rightarrow \mathbf{k}.\mathbf{B} = \mathbf{0} \tag{34}$$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} \Rightarrow \frac{n}{c}\widehat{k}\times\mathbf{H} = -\mathbf{D} \tag{35}$$

where free charges ρ_f and free currents j_f are taken to be zero.

Thus the wave equation in the crystal may be reduced to the following in a similar way, *i.e.*

$$\nabla \times (\nabla \times \mathbf{E}) = -\frac{\partial}{\partial t}(\nabla \times \mathbf{B}) \tag{36}$$

which becomes,

$$\frac{n^2}{c^2}\widehat{k}\times\left(\widehat{k}\times\mathbf{E}\right) = -\mu\mathbf{D} \tag{37}$$

On using the well known vector identity for a triple cross product this becomes

$$\frac{n^2}{c^2}\left[\left(\widehat{k}\cdot\mathbf{E}\right)\widehat{k}-\mathbf{E}\right]=-\mu\mathbf{D} \tag{38}$$

Now writing out explicitly the $k-$vector in terms of a unit vector as

$$\widehat{k}=\widehat{\mathbf{x}}s_x+\widehat{\mathbf{y}}s_y+\widehat{\mathbf{z}}s_z \tag{39}$$

which gives for equation 38.

$$D_i=\frac{n^2}{\mu c^2}\left[E_i-\left(\mathbf{k}\cdot\mathbf{E}\right)s_i\right] \tag{40}$$

Writing this as the inverse given that the co-ordinate system chosen was one in which ε_{ij} is diagonal we have

$$\mathbf{E}=\frac{\mu_r}{\varepsilon_0}\begin{bmatrix}\frac{1}{n_x^2} & 0 & 0\\ 0 & \frac{1}{n_y^2} & 0\\ 0 & 0 & \frac{1}{n_z^2}\end{bmatrix}\mathbf{D} \tag{41}$$

substituting the relevant E_i component into equation 40 then gives

$$D_i=\frac{n^2}{\mu c^2}\left[\frac{\mu_r D_i}{\varepsilon_0 n_i^2}-\left(\mathbf{k}\cdot\mathbf{E}\right)s_i\right] \tag{42}$$

Finally, solving for D_i

$$D_i=\frac{\left(\widehat{k}\cdot\mathbf{E}\right)s_i}{\mu c^2\left[(1/n_i^2)-(1/n^2)\right]} \tag{43}$$

which with the help of the first Maxwell equation, $\mathbf{k}\cdot\mathbf{D}=\mathbf{0}$ yields,

$$\frac{s_x^2}{(1/n_x^2)-(1/n^2)}+\frac{s_y^2}{(1/n_y^2)-(1/n^2)}+\frac{s_z^2}{(1/n_z^2)-(1/n^2)}=0 \tag{44}$$

This is known as *Fresnel's equation* and must hold for the original assumption of a plane-wave solution in the crystal.

This may be re-written as follows

$$\left(\frac{1}{n_y^2}-\frac{1}{n^2}\right)\left(\frac{1}{n_z^2}-\frac{1}{n^2}\right)s_x^2+\left(\frac{1}{n_x^2}-\frac{1}{n^2}\right)\left(\frac{1}{n_z^2}-\frac{1}{n^2}\right)s_y^2+\left(\frac{1}{n_x^2}-\frac{1}{n^2}\right)\left(\frac{1}{n_y^2}-\frac{1}{n^2}\right)s_z^2=0 \tag{45}$$

As an example let's take one principal plane at a time, *e.g.* the k_x-k_y plane where $s_z=0$; this yields

$$\left(\frac{1}{n_z^2}-\frac{1}{n^2}\right)\left[\left(\frac{1}{n_y^2}-\frac{1}{n^2}\right)s_x^2+\left(\frac{1}{n_x^2}-\frac{1}{n^2}\right)s_y^2\right]=0 \tag{46}$$

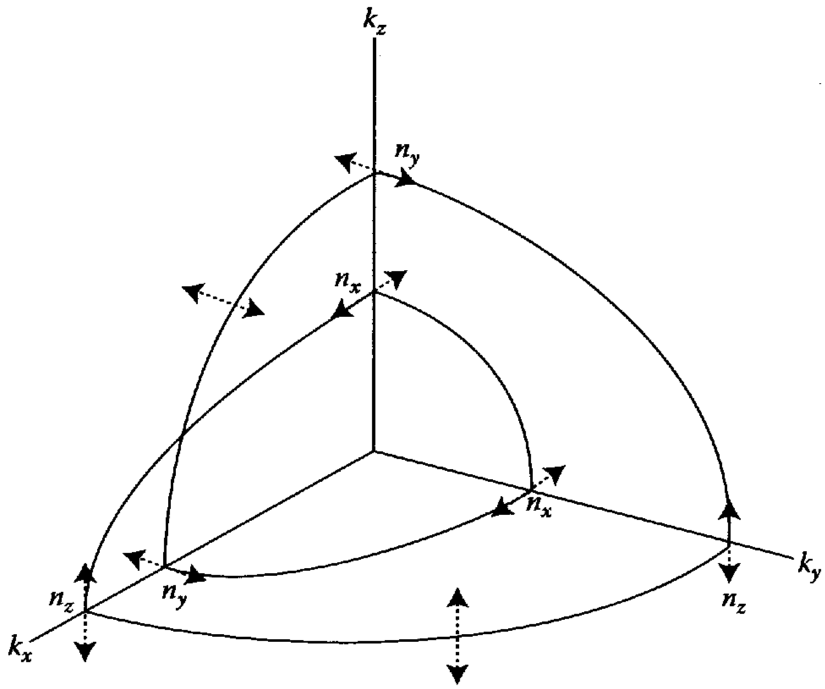

Figure 7: Wave-vector surface

Noting that the unit vector condition requires $s_x^2 + s_y^2 = 1$ then either

$$n_z = n \tag{47}$$

or

$$\left[\frac{s_x^2}{n_y^2} + \frac{s_y^2}{n_x^2} - \frac{1}{n^2}\right] = 0 \tag{48}$$

In the former case we have simply a constant value of the refractive index ($n = n_0$) or in the latter a refractive index which varies being given by the equation of an ellipse. In this case the unit vector $\widehat{k} = \widehat{\mathbf{x}} \sin\phi + \widehat{\mathbf{y}} \cos\phi$ so that

$$\frac{1}{n^2(\phi)} = \frac{\sin^2\phi}{n_y^2} + \frac{\cos^2\phi}{n_x^2} \tag{49}$$

Similarly results follow for both the $k_y - k_z$ and.$k_x - k_z$ planes: there are two solutions one which is independent of the direction of k called the *ordinary* wave, and the other which changes with the k-vector direction and with a value given by the equation of an ellipse, called the *extraordinary* wave. The intersection of the two curves in the $k_x - k_z$ plane corresponds to the crystal's optical axis. For extraordinary waves, vectors **k** and **D** are not perpendicular means that Poynting's vector **S** is no longer parallel with k; the angle between them ρ is known as the *walk off angle;* this is also the same as the angle between **D** and **E** It is given by

$$\tan(\rho + \theta) = \frac{n_o^2}{n_e^2} \tan\theta \tag{50}$$

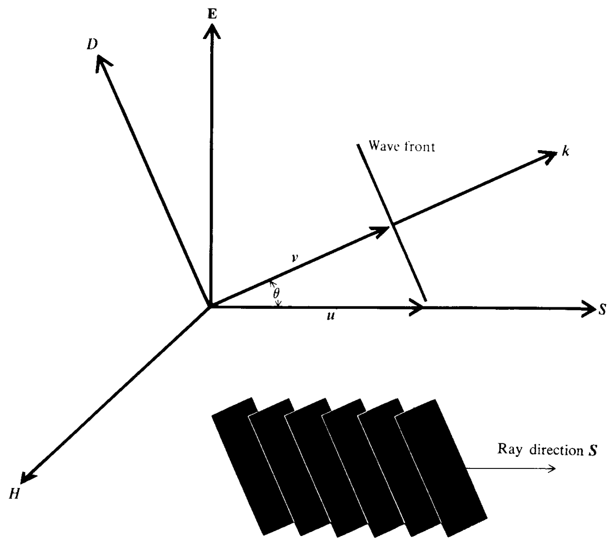

Figure 8: Walk off. Poynting's vector and k are no longer collinear

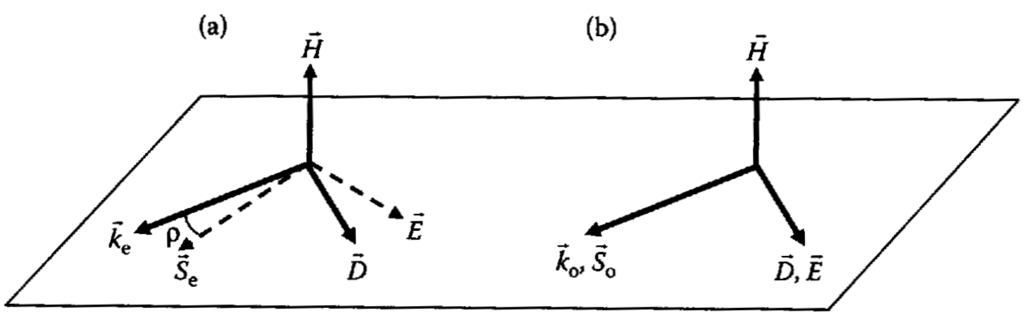

Figure 9: Walf off between S and k

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