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)

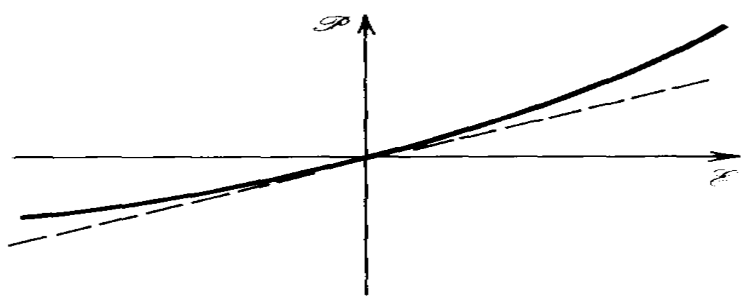

Figure 1: Linear versus nonlinear electric field effects

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.

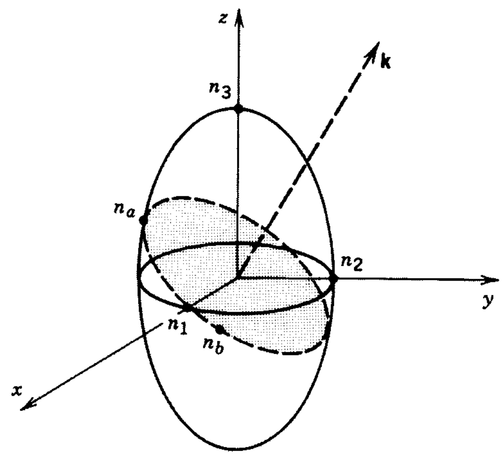

Figure 2: Index ellipsoid

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.

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

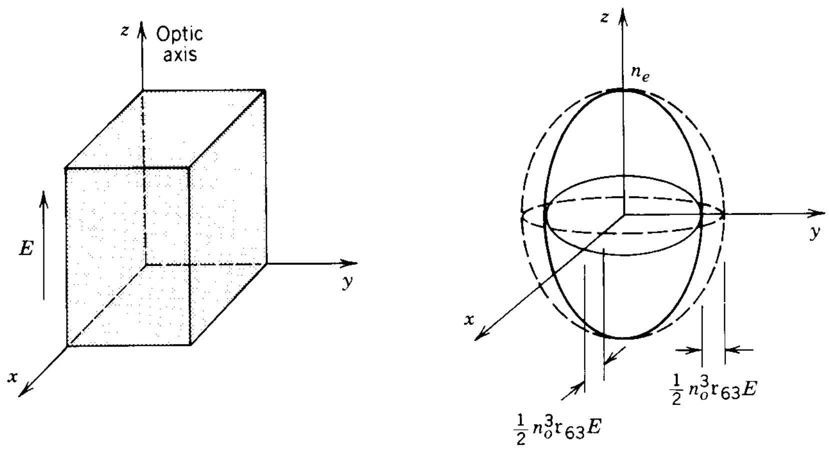

Figure 3: Index ellipsoid for a $\bar{4}$2m crystal

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 $\Delta\eta = (d\eta/dn)\Delta n = (-2/n^3)(-\frac{1}{2} r n^3 E - \frac{1}{2} s n^3 E^2..)$.

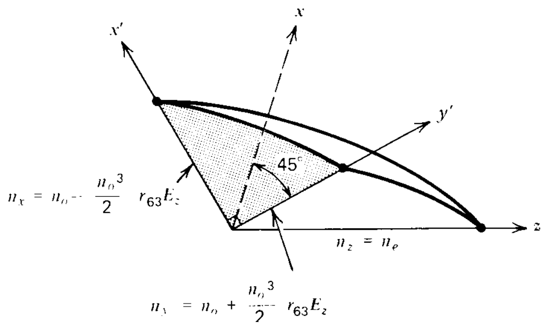

Figure 4: Rotation of axes by 45^0 about the optical axis.

$j \downarrow$ 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

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

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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 $\omega_1 = \omega_2$

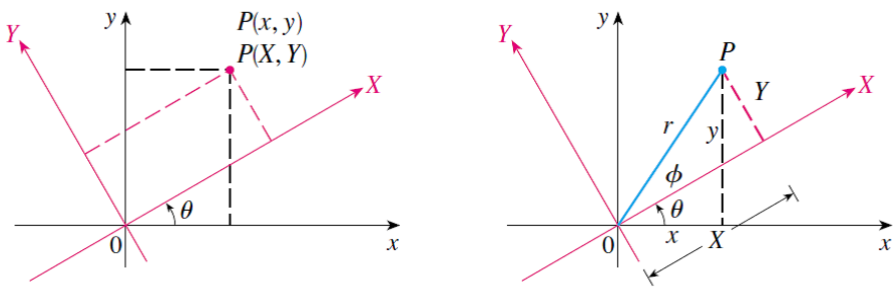

Figure 5: Rotation of Axes

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

$$\begin{pmatrix} x \\ y \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} X \\ Y \end{pmatrix} \tag{13}$$

or,

$$\begin{pmatrix} X \\ Y \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} x \\ y \end{pmatrix} \tag{14}$$

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.

$$x = 1/\sqrt{2}\left(x' - y'\right) \text{ and } y = 1/\sqrt{2}\left(x' + y'\right) \tag{15}$$

into the equation for the ellipsoid 12 we have

$$\frac{(x'-y')^2}{2n_o^2} + \frac{(x'+y')^2}{2n_o^2} + \frac{2\left(x'-y'\right)\left(x'+y'\right)}{2} r_{63} E_z^0 + \frac{z^2}{n_e^2} = 1 \tag{16}$$

which when rearranged gives

$$\frac{x'^2}{n_o^2} + \frac{y'^2}{n_o^2} + \left(x'^2 - y'^2\right) r_{63} E_z^0 + \frac{z^2}{n_e^2} = 1 \tag{17}$$

leading to

$$\frac{x'^2}{n_o^2}\left(1 + n_o^2 r_{63} E_z^0\right) + \frac{y'^2}{n_o^2}\left(1 - n_o^2 r_{63} E_z^0\right) + \frac{z^2}{n_e^2} = 1 \tag{18}$$

This identifies

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

Or equivalently

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

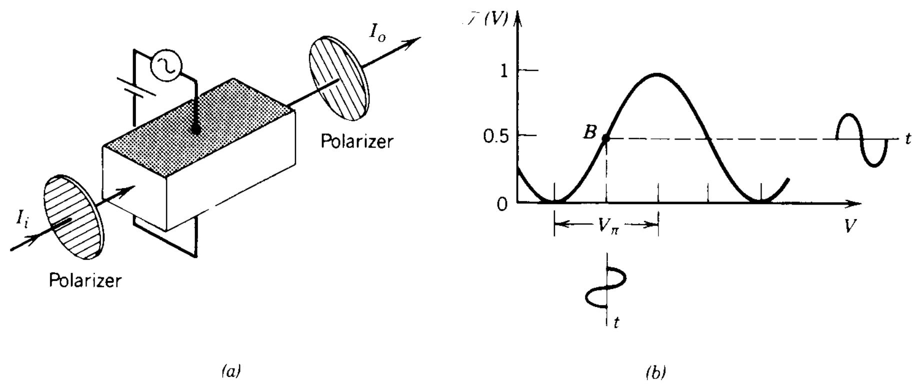

Figure 6: Electro-optic modulator used as an intensity modulator.

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

$$\Delta n = |n_{x'} - n_{y'}| = n_0^3 r_{63} E_z^0 \tag{21}$$

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

$$\phi = \frac{2\pi}{\lambda} \Delta n d = \pi \tag{22}$$

and the half-wave voltage is

$$V_\pi = \frac{\lambda}{2 n_0^3 r_{63}} \tag{23}$$

Appendix

Wave-vector surface

Linear but tensorial χ

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \tag{24}$$

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

$$\boldsymbol{\nabla}\times(\boldsymbol{\nabla}\times\mathbf{E}) + \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = -\frac{1}{c^2}\boldsymbol{\chi}\frac{\partial^2\mathbf{E}}{\partial t^2} \tag{25}$$

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

$$\mathbf{k}\times(\mathbf{k}\times\mathbf{E}) + \frac{\omega^2}{c^2}\mathbf{E} = -\frac{\omega^2}{c^2}\boldsymbol{\chi}\mathbf{E} \tag{26}$$

The cartesian components of this equation are thus

$$\left(-k_y^2 - k_z^2 + \frac{\omega^2}{c^2}\right)E_x + k_x k_y E_y + k_x k_z E_z = -\frac{\omega^2}{c^2}\chi_{11}E_x \tag{27}$$

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

$$\left(-k^2 + \frac{\omega^2}{c^2}\right)E_x = -\frac{\omega^2}{c^2}\chi_{11}E_x \tag{28}$$

$$\left(-k^2 + \frac{\omega^2}{c^2}\right)E_y = -\frac{\omega^2}{c^2}\chi_{22}E_y \tag{29}$$

$$\frac{\omega^2}{c^2}E_z = -\frac{\omega^2}{c^2}\chi_{33}E_z \tag{30}$$

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

$$k = \frac{\omega}{c}\sqrt{1+\chi_{11}} = \frac{\omega}{c}\sqrt{K_{11}} = \frac{\omega}{c}n_1 \tag{31}$$

$$k = \frac{\omega}{c}\sqrt{1+\chi_{22}} = \frac{\omega}{c}\sqrt{K_{22}} = \frac{\omega}{c}n_2 \tag{32}$$

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

Now the equations for the components [27] lead to the following condition for non-trivial solution for the field components not to vanish, *i.e.*

$$\begin{vmatrix} (\frac{n_1\omega}{c})^2 - k_y^2 - k_z^2 & k_x k_y & k_x k_z \\ k_y k_x & (\frac{n_2\omega}{c})^2 - k_x^2 - k_z^2 & k_y k_z \\ k_z k_x & k_z k_y & (\frac{n_3\omega}{c})^2 - k_x^2 - k_y^2 \end{vmatrix} = 0 \tag{33}$$

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

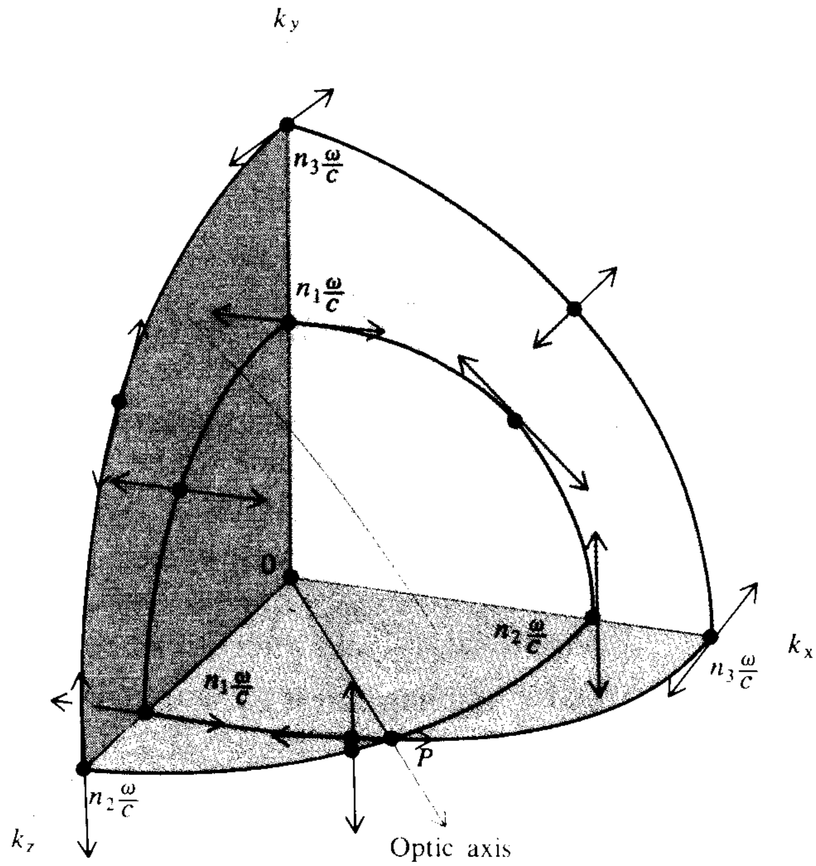

Figure 7: Wave-vector surface for an anisotropic crystal

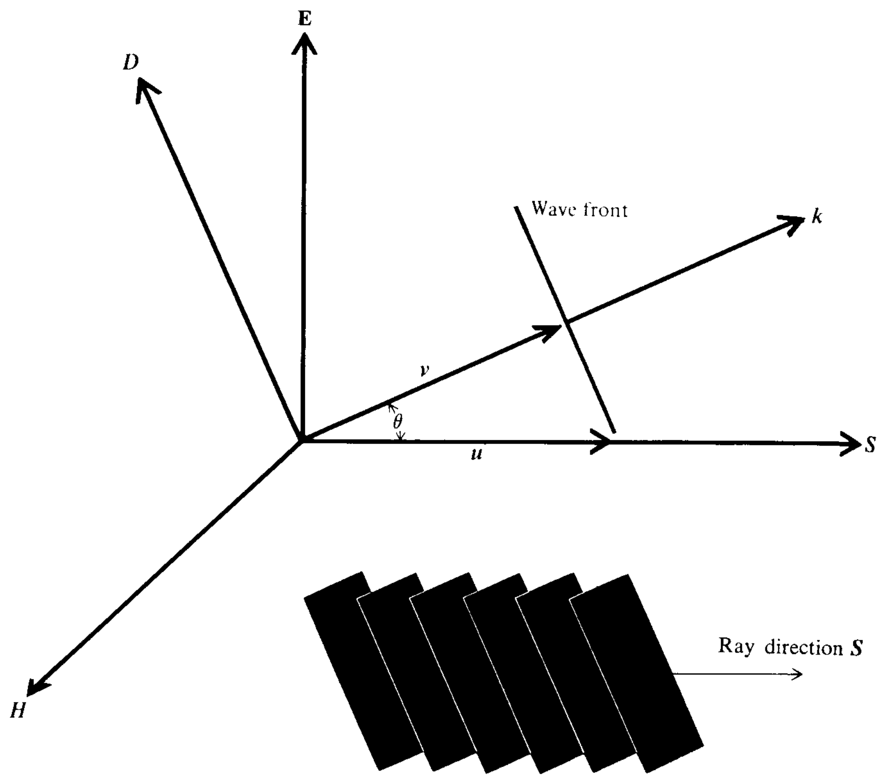

Figure 8: Walk off. Poynting's vector and k are no longer collinear

$$k_x^2 + k_y^2 = \left(\frac{n_3\omega}{c}\right)^2 \tag{34}$$

$$\frac{k_x^2}{(n_2\omega/c)^2} + \frac{k_y^2}{(n_1\omega/c)^2} = 1 \tag{35}$$

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

$$u = \frac{v}{\cos\theta} \tag{36}$$

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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