The phenomenon of nonlinear optical birefringence in uniaxial crystals

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Abstract

Birefringence is a phenomenon that produces double – value nature of refractive indices in uniaxial crystals. This phenomenon gives rise to effects such as ordinary (o) and extraordinary waves (e) and Kerr effect which produces nonlinear coefficients called Kerr constants. In this work, twelve uniaxial crystals are considered. The minimum electric field intensity that is required to produce nonlinear birefringence is determined. The magnitudes of the nonvanishing dielectric tensor components and their corresponding critical angles have been evaluated for the twelve uniaxial crystals. The result shows that in negative uniaxial crystals, electromagnetic waves travel faster in the extraordinary axis than in the ordinary axes. The reverse is however obtained in positive uniaxial axis. The work has also provided the series to be employed when determining the Kerr constants for even order nonlinear coefficients. A motivation for this work is an attempt to make nonlinear optical phenomena accessible to physics undergraduate.

Keywords: Birefringence, uniaxial crystals, Kerr constants, electric field intensity and refractive indices.

Resumen

Birrefringencia es un fenómeno que produce el doble - valor natural de los índices de refracción en cristales uniaxiales. Este fenómeno da lugar a efectos como el ordinario (o) y las olas extraordinarias (e) y el efecto Kerr, que produce coeficientes lineales constantes llamados Kerr. En este trabajo, doce cristales uniaxiales son considerados. El mínimo de intensidad de campo eléctrico que se requiere para producir la birrefringencia lineal se determina. Las magnitudes de las componentes del tensor dieléctrico no nula y sus ángulos correspondientes críticos han sido evaluados para doce crsitales uniaxiales. El resultado muestra que en cristales uniaxiales negativos, las ondas electromagnéticas viajan más rápido en el eje extraordinario que en los ejes comunes. Sin embargo, es lo contrario obtenidos en un eje uniaxial positivo. El trabajo también ha proporcionado la serie que se emplea para determinar las constantes de los coeficientes de Kerr, incluso para no lineal. Una motivación para este trabajo es un intento de hacer los fenómenos ópticos no lineales de acceso a la licenciatura de Física.

Palabras clave: Birrefringencia, cristales uniaxiales, constants de Kerr, intensidad de campo eléctrico y los índices de refracción.

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I. INTRODUCTION

Crystals can exhibit a number of interesting optical properties such as double refraction, optical rotation or polarization effects [1]. Material in which two of the components of dielectric constants are equal is termed uniaxial crystal. This class includes trigonal, tetragonal and hexagonal crystals. The dielectric tensor can be written in matrix form as

$$\varepsilon = \begin{bmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{bmatrix}. \quad (1)$$

where ε_x, ε_y and ε_z are the nonvanishing dielectric tensor components.

Birefringence in anisotropic (uniaxial) medium has many interpretations in nonlinear optics. In [2] it is defined as a phenomenon that produces multi – value nature of refractive index, while [3] defines it as double refraction. However, its effect gives rise to many phenomena such as; ordinary and extraordinary waves, Kerr effect and nonlinear refractive indices. The phenomenon of birefringence was first discovered when it was observed that crystals of Iceland spar, a form of calcium, formed double images of objects seen through them [4].

The refractive index of a material is the factor by which electromagnetic radiation is slowed down (relative to vacuum) when it travels inside the material. Many transparent solids are optically isotropic, meaning that the index of refraction is equal in all directions throughout the crystalline lattice. All isotropic crystals (e.g. the cubic crystals) have equivalent axes that interact with light in a similar manner, regardless of the crystal orientation with respect to incident light waves [5].

Anisotropic crystals on the other hand have crystallographically distinct axes and interact with light in a manner that is dependent upon the orientation of the crystalline lattice with respect to the incident light. According to [5], when an anisotropic crystal refracts light, the resulting rays are polarized and travel at different velocities. One of the rays travels with the same velocity in every direction through the crystal and is termed the ordinary ray, while the other travels with a velocity that is dependent upon the propagation direction within the crystal. This is referred to as extraordinary ray. The distance of separation between the ordinary and extraordinary rays increases with increasing crystal thickness. The two independent refractive indices of anisotropic crystals are quantified in terms of their birefringence. Thus, the birefringence B of the crystal as defined by [5] is given as:

$$B = \left| n_{high} - n_{low} \right| . \qquad (2)$$

where n_{high} is the largest refractive index and, n_{low} is the smallest refractive index. Eq. (2) agrees with the definition given by [6] that birefringence refers to the difference between the multiple refractive indices exhibited by an anisotropic material such as quartz.

In this work, the series to be employed in obtaining the Kerr constants for even order nonlinear susceptibility is proposed. The minimum value of the electric field intensity to cause birefringence in some uniaxial crystals is determined. The dielectric tensor components for the uniaxial crystals are also presented; their values reveal which of the axes the speed of light will be more retarded for the two classes of uniaxial crystals.

II. THE ORDINARY AND EXTRAORDINARY WAVES

In an anisotropic medium, there is a direction of propagation of the incident wave in which the refractive index is independent of the polarization. This direction is known as the optic axis. For uniaxial crystal, there is a single optic axis, while in biaxial crystal, there are two optic axes [7, 8]. This effect is illustrated in Fig.1.

According to [7], two types of waves can propagate in a uniaxial crystal;

(i) Waves linearly polarized perpendicular to the plane formed by the optic axis and the direction of incidence. This is called an ordinary axis (o). In this case, the field simply passes through the crystal in the expected or ordinary way, satisfying Snell's law. The ray corresponding to the wave whose refractive index is independent of the direction of propagation is called the ordinary ray.

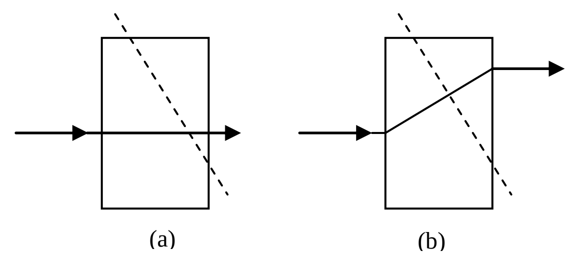


FIGURE 1. (a) ordinary and (b) extraordinary waves in a uniaxial birefringent crystal.

(ii) There are also waves that are linearly polarized parallel to the plane formed by the optic axis and the direction of incidence. These are referred to as extraordinary waves (e). The wave is deflected at the boundaries, and the rays emerging from the exit face are displaced with respect to the incident rays. The refractive index of this type of wave depends on the direction of propagation.

This effect of double refraction or birefringence is further demonstrated in Fig. 2 [8]. In Fig.2, subscripts 0 indicates incident wave, while, 1 and 2 indicate the refracted waves.

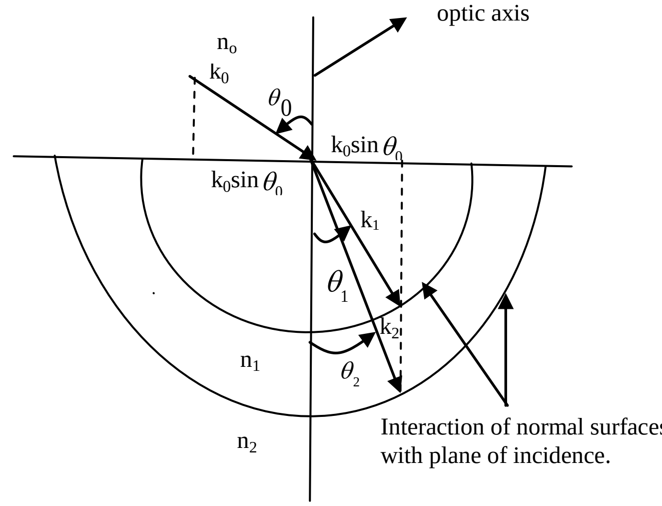


FIGURE 2. Double refraction at a boundary of ananisotropic medium.

III. REFRACTIVE INDEX DUE TO BIREFRINGENCE

In Fig.2, the refractive index for ordinary wave is denoted by n_o, and is independent of the direction of propagation. The refractive index for extraordinary wave is denoted by $n_e(\theta)$, and depends on the direction of propagation (θ) relative to the optic axis. This effect as earlier mentioned is known as birefringence.

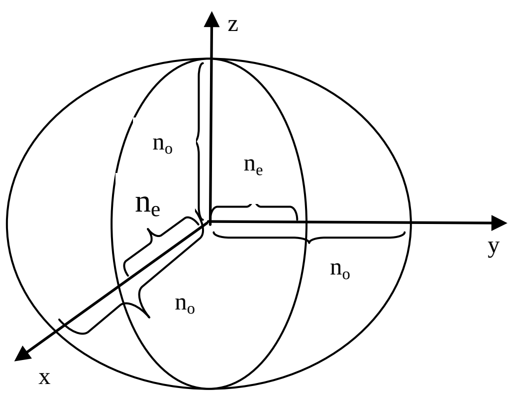
According to [9], the behaviour of refractive index is usually described in terms of refractive index surface, i.e. the index ellipsoid. In the case of the ordinary ray, it is a sphere; while for extraordinary ray, it is an ellipsoid, as explained in [8]. That is, in terms of ellipsoid, this effect becomes a three dimensional body with cylindrical symmetry. Two indices of refraction are then identical ($n_x = n_y$), so that the plane intersecting perpendicular to the optical axis forms a circle. If z - axis is considered as the axis of the cylindrical symmetry (the optical axis of a uniaxial crystal), then for uniaxial crystal, [10] defined the principal indices of refraction as

$$n_o^2 = \frac{\varepsilon_x}{\varepsilon_o} = \frac{\varepsilon_y}{\varepsilon_o} \quad \text{and} \quad n_e^2 = \frac{\varepsilon_z}{\varepsilon_o}, \tag{3}$$

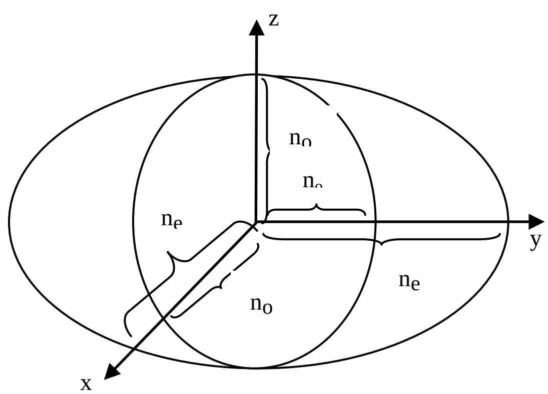
where ε_o is dielectric constant in free space (~ 8.85 x 10^{-12} F/m); ε_x , ε_y and ε_z are the dielectric constants along x, y and z – axes. For uniaxial crystals $\varepsilon_x = \varepsilon_y$. It is also a known fact that refractive index and critical angle of materials are related by

$$\sin c_o = \frac{1}{n_o} \text{ and } \sin c_e = \frac{1}{n_e}, \tag{4}$$

where c_o and c_e are the critical angles at the ordinary and extraordinary axes respectively. The critical angle of a material determines whether an internal ray will be reflected back into the material. As shown in Eq. (4), it is a function of the refractive index, and hence, the higher the refractive index the lower the critical angle.



(a)



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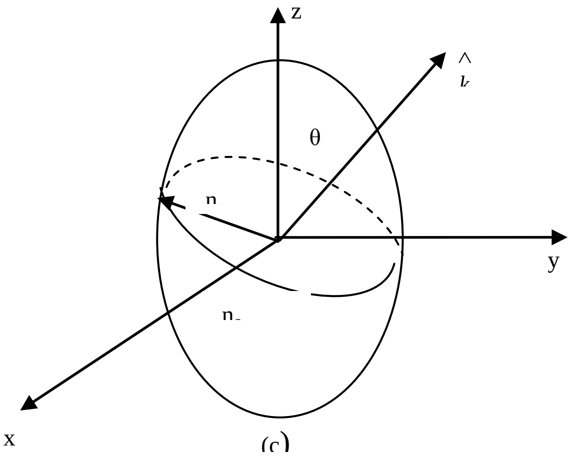

(c)

FIGURE 3. Index ellipsoid of (a) negative uniaxial cristal ($n_e < n_o$); (b) positive uniaxial crystal ($n_e > n_o$) and (c) uniaxial crystal with z – axis as the rotational symmetric axis.

In Fig. 3 (a), the refractive index for the ordinary ray n_o is greater than that for the extraordinary ray n_e. In Fig. 3 (b), the refractive index for the ordinary ray n_o is less than that for the extraordinary ray n_e. In Fig. 3 (c), the polarization of the ordinary and extraordinary waves can be determined using the index ellipsoid [10]. For uniaxial crystal, the refractive indices in the x – and y – directions can be represented by n_o.

The ellipsoid has the z - axis as the rotational symmetric axis, with refractive index n_e. If the ellipsoid is deformed and tilted through a rotation in space with respect to the original ellipsoid, the original index ellipsoid in the principal coordinate axes is given as [8, 10].

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1. \tag{5}$$

For uniaxial crystals, two of the refractive indices are identical ($n_x = n_y = n_o$), and $n_z = n_e$, and, that this particular ellipsoid can be characterized by two values of refractive indices; n_o (the ordinary index), which corresponds to n_x or n_y, and n_e (extraordinary index) corresponding to n_z [2]. Eq. (5) can therefore be written as

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1. \tag{6}$$

The projection of the ellipsoid can further be depicted on the *y – z* plane as shown in Fig. 4. The polarization of the ordinary wave points perpendicular to the *y - z* plane, [11].

In Fig.4, the polarization of the extraordinary wave is along the vector OA, and the index of refraction is thus $n_e(\theta)$. It follows therefore that for any angle θ

$$z = n_e(\theta)\sin\theta, \qquad 7(a)$$

$$y = n_e(\theta)\cos\theta. \qquad 7(b)$$

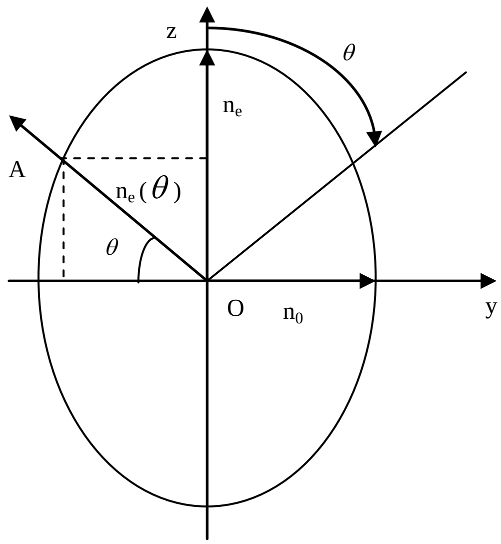

FIGURE 4. Projection of ellipsoid along the $y-z$ plane.

The equation of the ellipse (projection of the ellipsoid with $x = 0$) is thus

$$\frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1. \qquad (8)$$

Eq. (8) is the equation of an ellipsoid of revolution [12]. From Eqs. (7) and (8), one obtains

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}. \qquad (9)$$

The index of refraction varies from $n_e(\theta) = n_o$ for $\theta = 0^o$ to $n(\theta) = n_e$ for $\theta = 90^o$ [13]. Eq. (9) shows that the index is independent of the direction of propagation of the wave vector. Chin (1989), [10], classified the uniaxial crystals into two; crystals with $n_e > n_o$ are referred to as positive uniaxial crystals, while those with $n_e < n_o$ are called negative uniaxial crystals. Table I shows the refractive índices of some uniaxial crystals.

IV. THE KERR EFFECT

In [14] the Kerr effect is defined as either light induced double refraction, or an intensity – dependent refractive index. In nonlinear optics, strong field of high intensity (such as output of a laser) can cause a medium's refractive index to vary as the light passes through it. If the refractive index varies quadratically with the field (linearly with the intensity), it is called optical Kerr effect. According to [15], Kerr effect is a manifestation of an electric field on a material. [7] defined the Kerr effect as thus the difference in the refractive indices ($n_e - n_o$) for light polarized parallel and perpendicular to the optic axis is proportional to the square of the applied field E. In [10] it is expressed as

TABLE I. Indices of refraction of some uniaxial crystals.

Crystal	n_o	n_e
Calcite ($CaCO_3$)	1.658	1.486
Lithium niobate ($LiNbO_3$)	2.286	2.200
Lithium tantalite ($LiTaO_3$)	2.176	2.180
Quartz (SiO_2)	1.544	1.553
Rutile (TiO_2)	2.616	2.903
Beryl (Be_3Al_2)	1.602	1.557
Calomel (Hg_2Cl_2)	1.973	2.656
Magnesium Fluoride (MgF_2)	1.380	1.385
Sapphire (Al_2O_3)	1.768	1.760
Sodium Nitrate ($NaNO_3$)	1.587	1.336
Peridot (Mg, Fe) SiO_4	1.690	1.654
Ice (H_2O)	1.309	1.313

$$n_e - n_o = K\lambda E^2 \qquad (10)$$

where λ is the vacuum wavelength of light, and K is known as the Kerr constant (K – constant). The Kerr – constant is a numerical factor (a coefficient) defined by [16] for odd number nonlinear susceptibilities as

$$K = \frac{n!2^{1-n}}{\left(\frac{n-1}{2}\right)!\left(\frac{n+1}{2}\right)!}. \qquad (11)$$

From Eq. (10) and Table II, the minimum electric field **E** that is required to cause birefringence in these classes of uniaxial crystals can easily be calculated. This is shown in Table III, and it involves the values of second order and the fourth order Kerr constants. The value of λ that was used comes in as a result of considering the corresponding minimum value of the THz frequency (1THz), and the speed of light (~ 3 x $10^8 m/s^2$). It is important to note that of the two classes of crystals (i.e. isotropic and anisotropic), nonlinear effects such as Second Harmonic Generation (SHG), and other higher even order nonlinear effects that arise as a result of the interaction of THz radiation can only be observed in anisotropic crystals, out of which uniaxial crystals are examples [9]. It is therefore appropriate to use the minimum value in the THz range in the evaluation process in Table III. Using Eqs. (1) and (3), the nonvanishing dielectric tensor components for the twelve (12) uniaxial crystals have been calculated (see Table III).

TABLE II. Birefringence and Critical angle of some uniaxial crystals.

Crystal	n_o	n_e	$c_o(^o)$	$c_e(^o)$	B
Calcite ($CaCO_3$)	1.658	1.486	37.1	42.3	- 0.172
Lithium niobate ($LiNbO_3$)	2.286	2.200	25.9	27.0	- 0.086
Lithium tantalite ($LiTaO_3$)	2.176	2.180	27.4	27.3	0.004
Quartz (SiO_2)	1.544	1.553	40.4	40.1	0.009
Rutile (TiO_2)	2.616	2.903	24.5	20.2	0.287
Beryl (Be_3Al_2)	1.602	1.557	38.6	40.0	- 0.045
Calomel (Hg_2Cl_2)	1.973	2.656	30.5	22.1	0.683
Magnesium Fluoride (MgF_2)	1.380	1.385	46.4	46.2	0.006
Sapphire (Al_2O_3)	1.768	1.760	34.4	34.6	- 0.008
Sodium Nitrate ($NaNO_3$)	1.587	1.336	39.1	48.5	- 0.251
Peridot (Mg, Fe) SiO_4	1.690	1.654	36.3	37.2	- 0.036
Ice (H_2O)	1.309	1.313	49.8	49.6	0.004

From Eqs. (12) and (13), the nonlinear susceptibility can be written as a series of terms of increasing order of even nonlinearity as

$$\bar{\chi}(\omega;E)=\frac{1}{4}\chi^{(2)}(-\omega;\omega,-\omega,\omega)\left|E\right|+ \\ +\frac{3}{16}\chi^{(4)}(-\omega;\omega,-\omega,\omega;-\omega,\omega)\left|E\right|^{3}+... \\ +\frac{(n-1)!2^{-n}}{\left(\frac{n-2}{2}\right)!\left(\frac{n}{2}\right)!}\chi^{(n)}(-\omega;\omega,...-\omega;\omega)\left|E\right|^{n-1} \quad (14)$$

V. DISCUSSIONS

From Table II, one can easily identify a positive and a negative uniaxial crystal. However, the positive uniaxial crystals tend to have higher critical angles than the negative uniaxial crystals. Except for Rutile (TiO_2) and Calomel ((Hg_2Cl_2) that have relatively higher birefringence, and hence lower values of the critical angles. This means that in physical applications, Rutile and Calomel are capable of producing fuzzy pictures (out of focus), since materials that exhibit strong birefringence produce fuzzy pictures [6].

Similarly, in negative uniaxial crystals, the values of the critical angles are higher in the extraordinary axis than the ordinary axes. This shows that electromagnetic wave will travel faster in the extraordinary axis than in the ordinary axes. However, one obtains the reverse in positive uniaxial crystals. This could also be observed in the nonvanishing dielectric components (see Table III).

TABLE III. Electric field required to Produce Birefringence in some uniaxial crystals.

Crystal	Electric field Intensity (V/m)		Nonzero dielectric Tensor Components	
	Second order $K=\frac{1}{2}$	Fourth order $K=\frac{3}{16}$	$\varepsilon_x=\varepsilon_y$ $(\times 10^{-11})$	ε_z $(\times 10^{-11})$
Calcite ($CaCO_3$)	1.153×10^2	1.749×10^2	2.433	1.954
Lithium niobate ($LiNbO_3$)	1.071×10^2	2.142×10^2	4.652	4.283
Lithium tantalite ($LiTaO_3$)	2.309×10^1	2.667×10^1	4.190	4.206
Quartz (SiO_2)	3.464×10^1	4.000×10^1	2.110	2.134
Rutile (TiO_2)	1.956×10^2	2.259×10^2	6.056	7.458
Beryl (Be_3Al_2)	1.732×10^1	2.828×10^1	2.271	2.145
Calomel (Hg_2Cl_2)	6.748×10^1	1.100×10^2	3.445	6.243
Magnesium Fluoride (MgF_2)	6.324×10^0	1.033×10^1	1.685	1.698
Sapphire (Al_2O_3)	7.303×10^0	1.193×10^1	2.766	2.741
Sodium Nitrate ($NaNO_3$)	4.090×10^1	6.680×10^1	2.230	1.580
Peridot (Mg, Fe) SiO_4	1.549×10^1	2.530×10^1	2.525	2.412^1
Ice (H_2O)	5.164×10^0	8.433×10^0	1.516	1.526

VI. CONCLUSION

This work has brought out the different effects that can be produced as a result of nonlinear optical birefringence in twelve uniaxial crystals. These include the values of the electric field necessary to produce birefringence; the critical angles for the two different axes in uniaxial crystals, the dielectric tensor components for the different axes of the crystals, and the series that is required to give the necessary Kerr constants for even nonlinear coefficients.

The work has also shown that in negative uniaxial crystals, the speed of light travels faster in the extraordinary axis than the ordinary axes. The reverse of this result is however obtained in positive uniaxial crystals.

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