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Piezo-optic Birefringence in CsCl-Type Crystals

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Following the theoretical approach of Bansigir and Iyengar, the expression for p_{44} for the crystals of CsCl type is developed. Polarizabilities of some ions at 5890 Å are evaluated with experimental values of p_{44} and are found to be in good agreement with reported values. Polarizabilities of the ions in CsI at different wavelengths in the range 3000 to 5890 Å are also evaluated.

Introduction

Expressions for the change in refractive indices due to uniaxial stress along [100] in NaCl-type crystals were developed by Bansigir & Iyengar (1961*a*), and from these expressions the birefringence characterized by the elasto-optic constants $(p_{11}-p_{12})$ was obtained. The consequences of the theory were dealt with by Bansigir & Iyengar (1961*b*). Ethiraj & Bansigir (1973) extended the theory to the birefringence characterized by the elasto-optic coefficient p_{44} and the corresponding piezo-optic coefficient q_{44}.

Later Ethiraj, Krishna Murty & Bansigir (1973) developed the theory for CsCl-type crystals, but limited the discussion to observations made along [100] and birefringence characterized by $(p_{11}-p_{12})$. In this paper, an attempt is made to extend the theory to cover the birefringence characterized by p_{44}.

Theory and expression for p_{44}

Let a rectangular bar of crystal with orientation [1$\bar{1}$0], [001], [110] (to be called X', Y', Z' respectively) be stressed along the Z' direction. The induced refractive index changes are related to strain t' through the following expression based on the phenomenological theory of Pockels (1906):

$$n_{z'}-n_{x'}=-p_{44}\frac{n^3}{2}t', \qquad (1)$$

where n is the refractive index in the unstrained condition, $n_{z'}$ and $n_{x'}$ are the altered refractive indices for light vibrating in the Z' and X' directions.

The change in refractive index is caused by (i) change in density, *i.e.* change in the number of dispersing centres; and (ii) change in the polarizability of the ion.

If a unit volume of the crystal is stressed along the Z' direction, the change in the number of ions per unit volume is given by

$$-t'[1-(\sigma_{x'}+\sigma_{y'})]\,N_j$$

where $\sigma_{x'}$ and $\sigma_{y'}$ are the Poisson's ratios in the X' and Y' directions respectively and N_j is the number of ions per unit volume.

The factors affecting polarizability are (*a*) Lorentz–Lorenz anisotropy due to the material continuum, (*b*) Lorentz–Lorenz anisotropy due to dipoles within the cavity, and (*c*) Coulomb anisotropy due to presence of positive and negative charges within the cavity.

The Lorentz–Lorenz anisotropy due to the material continuum has been calculated by Mueller (1935). He has shown that for a medium strained in the Z' direction where the strain ellipsoid has an axial ratio of $1:1:1+t'$, the refractive index for light vibrating in the X' direction is given by

$$3(n_{x'}^2-1)=4\pi N_j\alpha_j[(n_{x'}^2+2)+K_{x'}(n^2-1)], \qquad (2)$$

with a similar expression for light vibrating in the Z' direction, where $K_{x'}=2t'/5$, $K_{z'}=-4t'/5$; α_j is the polarizability of the ion under consideration.

The expression for the field due to dipole anisotropy, if the incident field is polarized in the Z' direction, the centre of the cavity being occupied by a positive or negative ion, is given by

$$E_{jz'd}=D_{jkz'}(\mu_{kz'})\frac{t'}{\gamma^3}, \qquad (3a)$$

where j represents the ion occupying the centre of the cavity and k the ion the effect of which is to be calculated at the centre; d indicates that the field is dipole in nature and γ is the lattice constant.

For light polarized in the X' direction, the corresponding expression is

$$E_{jx'd} = D_{jkx'}\,(\mu_{kx'})\frac{t'}{\gamma^3}. \tag{3b}$$

$\mu_{kx'}$ and $\mu_{kz'}$ in the above expressions are dipole moments along X' and Z' respectively; $D_{jkx'}$ and $D_{jkz'}$ are the numerical values of finite lattice sums in the deformed lattice.

The expression for the field created due to Coulomb anisotropy by charges present in the cavity for light polarized in the Z' direction is given by

$$E_{jz'c} = \left[\frac{\beta}{\gamma^3}\right] C_{jkz'} \left[\frac{\alpha_k}{f_k}\right] t' \tag{4a}$$

and for light polarized in the X' direction by

$$E_{jx'c} = \left[\frac{\beta}{\gamma^3}\right] C_{jkx'} \left[\frac{\alpha_k}{f_k}\right] t', \tag{4b}$$

where β is the number of valence electrons and α_k and f_k are the polarizability and the oscillator strength respectively of the positive or negative ion. $C_{jkz'}$ and $C_{jkx'}$ are numerical constants of the lattice. The Coulomb fields are created because under the influence of the incident field, the excess charges are displaced by amounts S_1, S_2 or S_k in general, giving rise to a dipole moment given by $f_k e^* S_k/(4\pi)^{1/2} = \mu_{kz'}$ or $\mu_{kx'}$, where e^* is the effective charge (Fröhlich, 1949).

The dipole moment of the jth ion in the presence of the resultant field (arising from continuum, dipole and Coulomb anisotropies) is given by

$$(\mu_{jz'})_{idcp} = \alpha_j F_{jz'} \text{ in the } Z' \text{ direction} \tag{5a}$$

$$(\mu_{jx'})_{idcp} = \alpha_j F_{jx'} \text{ in the } X' \text{ direction} \tag{5b}$$

where

$$F_{jz'} = E_{iz} + E_{jz'd} + E_{jz'c} + K_{z'} P_j \tag{6a}$$

$$F_{jx'} = E_{ix} + E_{jx'd} + E_{jx'c} + K_{x'} P_j. \tag{6b}$$

E_i is the incident field and P_j the polarization of the medium per unit volume.

Alternatively, the dipole moment in expression (6) could be due to the effective polarizabilities $\alpha_{jz'}$ or $\alpha_{jx'}$ in the Z' and X' directions under the incident field $E_{iz'}$ and $E_{ix'}$ according to

$$(\mu_{jz'})_{idcp} = \alpha_{jz'} E_{iz'} \tag{7a}$$

$$(\mu_{jx'})_{idcp} = \alpha_{jx'} E_{ix'}. \tag{7b}$$

It is assumed that the Lorentz–Lorenz equation for refractive index n in the form,

$$\frac{n^2-1}{n^2+2} = \sum \frac{n_j \alpha_j}{3} \tag{8}$$

holds good.

Equations (5), (6), (7) and (8) together yield

$$\alpha_{jz'} = \alpha_j \left[1 + \frac{(n^2+2)}{3}\left\{D_{jkz'}\frac{\alpha_k}{\gamma^3} + C_{jkz'}\frac{\alpha_k}{\gamma^3}\frac{\beta}{f_k} + \frac{Kz'}{3}(n^2-1)\right\} t'\right] \tag{9a}$$

$$\alpha_{jx'} = \alpha_j \left[1 + \frac{(n^2+2)}{3}\left\{D_{jkx'}\frac{\alpha_k}{\gamma^3} + C_{jkx'}\frac{\alpha_k}{\gamma^3}\frac{\beta}{f_k} + \frac{Kx'}{3}(n^2-1)\right\} t'\right]. \tag{9b}$$

The expressions for the D's and C's are evaluated by taking the radius of the cavity to be equal to $\frac{3}{2}\gamma$ and assuming that

$$x' = \frac{\gamma}{2}(1 - \sigma_{x'} t'),$$

$$y' = \frac{\gamma}{2}(1 - \sigma_{y'} t')$$

and

$$z' = \frac{\gamma}{2}(1 + t')$$

where t' = tensile strain along Z' and $\sigma_{x'} t'$ and $\sigma_{y'} t'$ are the corresponding strains along X' and Y' respectively. The higher powers of t', $\sigma_{x'} t'$ and $\sigma_{y'} t'$ are neglected in the evaluation of the numerical constants. The values thus obtained are given in Table 1.

If it is assumed that $\alpha = \alpha^- + \alpha^+$, the changes in the polarizabilities along the Z' and X' directions can be expressed as follows:

$$\delta\alpha_{z'} = \frac{t'}{4\pi\gamma^3}[A'(\alpha^2 - \alpha\alpha^-) + B'\alpha^2] \tag{10a}$$

$$\delta\alpha_{x'} = \frac{t'}{4\pi\gamma^3}[A''(\alpha^2 - \alpha\alpha^-) + B''\alpha^2] \tag{10b}$$

Table 1. *Expressions for the D's and C's*

$$D_{12z'} = D_{21z'} = \frac{t'}{4\pi\gamma^3}(-3{\cdot}5404 - 10{\cdot}6211\sigma_{x'} + 7{\cdot}0807\sigma_{y'})$$

$$D_{11z'} = D_{22z'} = \frac{t'}{4\pi\gamma^3}(2{\cdot}4701 + 7{\cdot}4087\sigma_{x'} - 4{\cdot}9395\sigma_{y'})$$

$$D_{12x'} = D_{21x'} = \frac{t'}{4\pi\gamma^3}(10{\cdot}6211 + 3{\cdot}5404\sigma_{x'} - 7{\cdot}0807\sigma_{y'})$$

$$D_{11x'} = D_{22x'} = \frac{t'}{4\pi\gamma^3}(-7{\cdot}4087 - 2{\cdot}4701\sigma_{x'} - 4{\cdot}9395\sigma_{y'})$$

$$C_{12z'} = C_{21z'} = \frac{t'}{4\pi\gamma^3}(14{\cdot}9187 - 4{\cdot}9575\sigma_{x'} - 5{\cdot}0269\sigma_{y'})\frac{S_{1\text{ or }2}}{f_{1\text{ or }2}}$$

$$C_{11z'} = C_{22z'} = \frac{t'}{4\pi\gamma^3}(7{\cdot}2698 - 2{\cdot}8887\sigma_{x'} - 2{\cdot}8885\sigma_{y'})\frac{S_{1\text{ or }2}}{f_{1\text{ or }2}}$$

$$C_{12x'} = C_{21x'} = \frac{t'}{4\pi\gamma^3}(4{\cdot}9575 - 14{\cdot}9187\sigma_{x'} - 5{\cdot}0269\sigma_{y'})\frac{S_{1\text{ or }2}}{f_{1\text{ or }2}}$$

$$C_{11x'} = C_{22x'} = \frac{t'}{4\pi\gamma^3}(2{\cdot}8887 - 7{\cdot}2698\sigma_{x'} - 2{\cdot}8885\sigma_{y'})\frac{S_{1\text{ or }2}}{f_{1\text{ or }2}}$$

where

$$A' = (12\cdot0210 + 36\cdot0596\sigma_{x'} - 24\cdot0404\sigma_{y'}) + \frac{(14\cdot5396 - 5\cdot7774\sigma_{x'} - 5\cdot7770\sigma_{y'})}{f_1} - \frac{(29\cdot8374 - 9\cdot9150\sigma_{x'} - 10\cdot0538\sigma_{y'})}{f_2}$$

$$A'' = (-36\cdot0596 - 12\cdot0210\sigma_{x'} - 24\cdot0404\sigma_{y'}) + \frac{(5\cdot7774 - 14\cdot5396\sigma_{x'} - 5\cdot7770\sigma_{y'})}{f_1} - \frac{(9\cdot9150 - 29\cdot8374\sigma_{x'} - 10\cdot0538\sigma_{y'})}{f_2}$$

$$B' = (2\cdot4701 + 7\cdot4087\sigma_{x'} - 4\cdot9395\sigma_{y'}) + \frac{(7\cdot2698 - 2\cdot8887\sigma_{x'} - 2\cdot8885\sigma_{y'})}{f_2}$$

$$B'' = (-7\cdot4087 - 2\cdot4701\sigma_{x'} - 4\cdot9395\sigma_{y'}) + \frac{(2\cdot8887 - 7\cdot2698\sigma_{x'} - 2\cdot8889\sigma_{y'})}{f_2}.$$

With all the anisotropies and the density change taken into consideration, the changes in the refractive indices along the Z' and X' directions are as follows:

$$dn_{z'} = \left[\frac{-\{1-(\sigma_{x'}+\sigma_{y'})\}(n^2-1)(n^2+2)}{6n} - \frac{2(n^2-1)^2}{15n} + \frac{A'N\gamma^3(n^2+2)^2}{226\cdot29n}u^2 - \frac{A'(n^2-1)(n^2+2)}{75\cdot43n}u + \frac{B'(n^2-1)^2}{25\cdot143nN\gamma^3}\right]t' \qquad (11a)$$

$$dn_{x'} = \left[\frac{-\{1-(\sigma_{x'}+\sigma_{y'})\}(n^2-1)(n^2+2)}{6n} - \frac{(n^2-1)^2}{15n} + \frac{A''N\gamma^3(n^2+2)^2}{226\cdot29n}u^2 - \frac{A''(n^2-1)(n^2+2)}{75\cdot43n}u + \frac{B''(n^2-1)^2}{25\cdot143nN\gamma^3}\right]t', \qquad (11b)$$

where

$$u = \frac{\alpha^- \text{ or } \alpha^+}{\gamma^3}.$$

Therefore

$$p_{44} = \frac{-C_{44}(1+\sigma_{x'})}{E'}\frac{2}{n^3}\left[\frac{(n^2-1)^2}{5n} + \frac{(A''-A')N\gamma^3(n^2+2)^2}{226\cdot29n}u^2 - \frac{(A''-A')(n^2-1)(n^2+2)}{75\cdot429n}u + \frac{(B''-B')(n^2-1)^2}{25\cdot143n}\right]. \qquad (12)$$

The expressions for $\sigma_{x'}$, $\sigma_{y'}$ and Young's modulus E' in terms of the elastic constants are:

$$E' = \frac{4(C_{11}-C_{12})(C_{11}+2C_{12})C_{44}}{C}$$

$$\sigma_{x'} = \frac{2C_{11}C_{44} - (C_{11}-C_{12})(C_{11}+2C_{12})}{C}$$

$$\sigma_{y'} = \frac{2C_{12}C_{44}}{C},$$

where $C = 2C_{11}C_{44} + (C_{11}-C_{12})(C_{11}+2C_{12})$.

Table 2. *Polarizabilities* α_j (cm^3 × 10^{-24}) *at* 5890 Å

Ion	Calculated value	Fajans & Joos (1924)	Born & Heisenberg (1924)	Pauling (1927)	Shockley (1946)	Shockley, Tessman & Khan (Kittel, 1953)
Cl	3·65 NH_4Cl 4·72 CsCl	3·53	3·05	3·66	2·92	2·97
I	7·04 CsI	7·55	6·28	7·10	6·41	6·44
Cs	2·07 CsCl 2·52 CsI	–	–	2·42	–	3·02
NH_4	1·33 NH_4Cl	–	–	–	–	–

1. Ionic polarizabilities as calculated from the present theory

Using experimental values of p_{44} and assuming the oscillator strength $f_1 = f_2 = 1$, we calculated the polarizabilities of the various ions for wavelength 5890 Å and compared them with the values obtained by other workers. The results are given in Table 2.

2. Polarizabilities of ions at different wavelengths

Equation (12) is used to calculate the polarizabilities of Cs^+ and I^- ions at different wavelengths in the range 3000–5890 Å. The data on the stress–optic dispersion are taken from Laiho & Korpela (1968) and refractive indices from Rodney (1955). The results are given in Table 3. The total polarizability is seen to increase towards shorter wavelength, while the polarizability of the positive ion is almost constant over the entire range.

Table 3. *Variation of polarizabilities of* Cs^+ *and* I^- *with wavelength*

Wavelength (Å)	α_{Cs}	α_I	$\alpha = \alpha_{Cs} + \alpha_I$
3022	2·56	8·56	11·12
3466	2·53	8·00	10·53
4047	2·52	7·59	10·11
4358	2·52	7·43	9·95
5085	2·52	7·27	9·79
5460	2·52	7·11	9·63
5790	2·52	7·06	9·58
5890	2·52	7·04	9·56

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Primary Extinction for Finite Crystals. Square-Section Parallelepiped

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The results are given for the calculation of the profile function of the scattering curve and the calculated primary extinction factor for a crystal in the form of a square-section parallelepiped as a function of its size τ, expressed in extinction length units. The calculations are based on the equations of the dynamical theory of diffraction. Asymmetry of the scattering curve and a shift of its principal maximum to larger angles, with τ and the Bragg angle increasing, are found. Approximate expressions for calculating the primary extinction factor as a function of τ are given.

1. Introduction

As is shown, the existing approximations of the theory of X-ray diffraction in mosaic crystals are usually based on the supposition of primary and secondary extinction effects. The primary extinction is connected with the effect of coherent scattering in a single mosaic block. Being an incoherent effect of scattering, the secondary extinction is calculated by the Darwin–Zachariasen transfer equations. These equations have been used in estimating primary extinction in finite crystals (Zachariasen, 1967; Becker & Coppens, 1974), though they do not really apply to coherent scattering effects.