Def Sci J, Vol 40, No 1, January 1990, pp 1-14.

Piezooptics of Crystals

G.S. Ranganath and S. Ramaseshan
Raman Research Institute, Bangalore- 080

ABSTRACT

In this paper the effects of mechanical stress on the refractive, absorptive and the rotatory properties of crystals have been considered with particular emphasis on the role of crystal symmetry. Stress-induced optical activity and the photoelastic behaviour of transparent, weakly absorbing and metallic crystals have been discussed. Piezooptia of polycrystalline media has also been briefly dealt with.

1. INTRODUCTION

It gives us great pleasure in writing this article on piezooptics to a volume to remember Prof. S. Bhagavantam, who made so many important contributions to this field.

Stress-induced birefringence was discovered in 1816 by Brewster. The phenomenon of photoelasticity in crystals- wherein the changes of refractive properties under stress are considered - was studied thoroughly by Pockels[1]. A serious error he had made in the photoelastic properties of crystals having certain symmetries was first pointed out by Bhagavantam[2] almost four decades later. He also gave a simple experimental method of detecting the differences in the behaviour of two cubic photoelastic classes which Pockels had overlooked. Indeed it was this work which led to the revival of experimental and theoretical interest in this field. Stress-induced optical activity was discovered by Ranganath and Ramaseshan[3]. The photoelastic behaviour of metals and absorbing crystals led to piezoabsorption being used to understand the band structure of semiconductors. The photoelasticity of polycrystalline aggregates and composites was the subject of a series of studies. Then came the important advance made by Nelson and Lax[4] who pointed out that the body rotation

Received 8 January 1990

suffered by crystals under shear displacement alters some of the photoelastic coefficients in uniaxial and biaxial crystals - an effect particularly important in extracting photoelastic coefficients using Brillouin scattering techniques. Since 1980 the photoelastic constants of many semiconductors are being measured because such measurements may reveal the dimensionality of the covalent network in crystalline and amorphous materials. In this article, we shall briefly review some of these.

2. PIEZOREFRACTIONS

2.1 Photoelasticity

In a homogeneously deformed body, the effect of the deformation is to alter the parameters defining the laws of propagation of light in the medium. It is therefore necessary to choose a proper optical property that alters with stress or strain. In a transparent medium placed in an electric field E, the distribution of charges (in the atoms and molecules that constitute the medium) is altered, inducing an induction D, which is given, in a linear model, by

$$D = [\varepsilon]E \tag{1}$$

The components of [E], the dielectric tensor, are real and positive. Alternatively

$$E = [a]D \quad \text{with} \quad [a] = [\varepsilon]^{-1} \tag{2}$$

where [a] is the index tensor. Both [E] and $[a]$ are symmetric tensors of rank two whose tensor surfaces can be represented by general ellipsoids. Along any direction s in the index ellipsoid, two waves are propagated with their vibrations linearly polarized parallel to the principal axes of the central elliptic section drawn normal to the direction of propagation. The relative indices of these two waves are respectively equal to the corresponding semi-axes of the central elliptic section. When the crystal is stressed or strained the index ellipsoid deforms, but it continues to be an ellipsoid with changes in its dimensions and orientation, with respect to the index ellipsoid of the unstrained crystal. We shall denote the changes in components of the index tensor by Δa_{ij}. This may, in a first order theory be expressed as a homogeneous linear function of the components of stress or strain

$$\Delta a_{ij} = -q_{ijkl} X_{kl} \quad \text{or} \quad \Delta a_{ij} = p_{ijkl} x_{kl} \tag{3}$$

where X_{ij} and x_{ij} are stress and strain tensors of rank two. q_{ijkl} and p_{ijkl} are fourth rank tensor symmetric in i, j and k, *1.* These equations can also be written in the one index form as

$$\Delta a_i = -q_{ij} X_j \quad \text{or} \quad \Delta a_i = p_{ij} x_j \tag{4}$$

Unlike the elastic constant tensor, the tensors q_{ij} and p_{ij} are not symmetric in i and j. Consequently the number of independent constants may be described by a 6 x 6 matrix which in the triclinic class will have thirty-six independent components. This number would however be less for crystals containing various elements of symmetry. Pockels[1] classified the thirty-two point groups into nine classes, according to the number and the nature of surviving constants. This was shown to be erroneous by Bhagavantam[2] who showed that the thirty-two point groups fall into eleven classes, the so-called Laue symmetry groups, which are obtained if an additional inversion symmetry is introduced. **Bhagavantam**[5] later pointed out that it is possible to distinguish the different photoelastic classes by the study of tilt, if any, of the principal planes when stress acts along the principal axes of the index ellipsoid. One may easily distinguish the two photoelastic classes in the case of cubic crystals. For a stress along the cube axis in point groups 432, **m3m**, $\bar{4}3m$ cube the crystal becomes uniaxial, while in the point groups 23 and m3 it becomes biaxial. For a general direction of stress, the optic axial angle in the cubic crystals is independent of the magnitude of stress. While in other classes the optic axial angle depends on the magnitude of stress. A measurement of stress-induced birefringence in different geometries yields the photoelastic constants. The principle that photoelastic constants can be determined from the apparent decrease of magneto-optical rotation (Ramaseshan and **Chandrasekharan**[6]) was used to determine the photoelastic constants of the optically active crystals of $NaClO_3$ (point group 23) by Ramachandran and **Chandrasekharan**[7].

Mueller[8] gave an elegant method for determining the ratio of photoelastic constants in cubic crystals based on the **Raman-Nath** theory’ of light diffraction by ultrasonic waves. When ultrasonic waves are transmitted through cubic crystals, due to the strains, every volume element becomes birefringent and for light travelling perpendicular to the ultrasonic wave, the birefringence can be obtained from the index ellipse which is a section of the index ellipsoid normal to the direction of light propagation. Detailed analysis shows that for a plane wavefront of linearly polarized incident light at a general azimuth, the different orders of diffraction are again linearly polarized, but the direction of polarization is different in different orders. A measurement of the rotation of plane of polarization in the different orders with respect to that of the zeroth order gives the ratio of the photoelastic constants. This method has been extended to optically active cubic crystals by Vedam and **Ramachandran**[10].

2.1.1 Second Order Effects

Pockels’ linear laws of photoelasticity are valid only at small stresses and strains. Recent measurements of the variation of refractive index with hydrostatic pressure up to 7 k bar (Vedam, Schmidt and Roy”) have revealed departures from the linear law. The second order theory has been developed by Vedam and Srinivasan who worked out the number of non-vanishing second order constants for all the thirty-two point groups. There are 126 piezorefraction coefficients in a crystal of triclinic symmetry. As the crystal symmetry increases this number decreases.

2.1.2 Secondary Photoelasticity

We have so far discussed primary photoelasticity. But it must be remarked that in piezoelectric crystals there can be an indirect photoelastic effect due to the stresses and strains accompanying the electric field. This secondary effect is not always negligible. This phenomenon is also referred to as false photoelasticity.

2.2 Photoelasticity in Polycrystalliie Aggregates

2.2.1 Transmission of Light in a Stressed Aggregate

A polycrystalline aggregate consists of randomly oriented crystallites. The problem of light transmission through a polycrystalline aggregate has been considered by Raman and Viswanathan[12] and later by Ranganath and Ramaseshan[13]. The crystallite size is assumed to be larger than the wavelength of light. When a plane wavefront of linearly polarized light falls on such a medium, the incident light enters the first crystallite and as the light beam passes through the stack, the polarization state continuously changes. The emergent light from the polycrystal, therefore, consists of light beams polarized in different states; they are also incoherent as they travel through optically uncorrelated paths. Part of the incident intensity would be lost in intercrystalline boundary reflections.

A 'polycrystal' consisting of optically isotropic particles would also be optically isotropic. When this aggregate is stressed by a uniaxial load each of the constituent particles becomes optically uniaxial, with the axis of symmetry coinciding with the stress direction. Therefore, the polycrystal as a whole will behave as an optically uniaxial crystal with the axis of symmetry along the stress direction. The photoelastic birefringence introduced in the medium would be exactly same as that found in a single particle. On the other hand, one finds. an entirely different behaviour when particles are produced from a cubic crystal. Such particles though optically isotropic, are photoelastically anisotropic. We find the principal axes of the index ellipsoid to be generally tilted with respect to stress axis and this tilt varies from crystallite to crystallite.

As the stress-induced birefringence is small, we can justifiably. neglect, in this analysis, intensity loss due to intercrystalline reflections. A light beam of intensity ***I***, ellipticity (ω_0 = $\tan^{-1}$ b/a) and azimuth λ_0 is analytically represented by the four-component Stokes. vector

$$\sigma_0 = \begin{bmatrix} I \\ M_0 \\ C_0 \\ S_0 \end{bmatrix} \qquad \begin{array}{l} M_0 = I_0 \cos 2\omega_0 \cos 2\lambda_0 \\ C_0 = I_0 \cos 2\omega_0 \sin 2\lambda_0 \\ S_0 = I_0 \sin 2\omega_0 \end{array} \tag{5}$$

where I_0 is the intensity of the completely polarized part. Also $I \geqslant I_0$ depending upon whether the light beam is partially polarized (inequality sign) or completely polarized (equality sign). When the light beam passes through an optical system σ_0 changes to σ_0', the two being related by the equation

$$\sigma' = [m]\,\sigma_0 \tag{6}$$

where *[m]* is the 4 x 4 Mueller matrix. For a polycrystal, we find to a first order in δ the crystallite phase retardation.

$$[m] = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & -N\bar{\delta} \\ 0 & 0 & N\bar{\delta} & 1 \end{bmatrix} \tag{7}$$

where δ is the spatial average of δ_{ijk}. Therefore, the polycrystal is linearly birefringent with a phase retardation ϕ per unit thickness given by

$$\phi = \frac{2\pi}{\lambda} (\overline{\delta n_{ijk}}) \tag{8}$$

In other words, the photoelastic birefringence of the medium is the average of the stress-induced birefringences of the various crystallites. It is clear that this is equivalent to averaging the photoelastic tensor itself.

2.2.2 The Average Photoelastic Tensor

To find the average photoelastic tensor, we must know the stresses acting on the crystallite. But the elastic response of a polycrystal depends crucially on stress or strain continuity across the grains. As early as 1889, Voigt[14] calculated the elastic constants of polycrystalline media assuming strain continuity with discontinuous stress. Reuss”, on the other hand, computed the elastic constants of the aggregate assuming stress to be continuous. It may be remarked that both the Voigt (strain continuity) and Reuss (stress continuity) conditions have their innate deficiencies. In the former, the forces between the grains will not be in equilibrium, while in the latter, the grains will not fit together (Hill[16]). This means that the Voigt condition leads to intrinsic instabilities while the Reuss condition leads to voids in the aggregate. In both these procedures crystallites are assumed to be randomly oriented, (i.e., with no preferred orientation) and data on single crystals are used to compute those of the polycrystals (see also Bhagavantam”).

Since the stress optical constants of the aggregate can be measured when the crystallites are transparent, it appeared to us that this may be yet another approach to this problem of stress or strain continuity. One has to work out both the elastic and the photoelastic tensors under the Voigt and the Reuss limits. One gets the interesting result that the photoelastic constants are very much more sensitive to stress or strain continuity than the elastic constants are.

For the Voigt condition x_{kl} is assumed to be same for all crystallites and one has to average c_{ijkl} to get the average stress. $(\bar{p}_{ijkl})$ gives the average birefringence in this case. On the other hand for the Reuss condition x_{kl} is assumed to be the same for all crystallites so that the average strain and birefringence are obtained from the averages of (s_{ijkl}) and (q_{ijkl}).

Though for a single crystal $(p_{ij}) = (q_{im})(c_{mj})$ and $(q_{ij}) = (p_{im})(s_{mj})$, these relations do not hold, however, for a polycrystal. Again we know that $(\bar{q}_{ij})$ and $(\bar{c}'_{ij})$ $[= (\bar{s}_{ij})^{-1}]$ correspond to stress continuity in the medium while $(\bar{p}_{ij})$ and $(\bar{s}'_{ij})$ $[=(\bar{c}_{ij})^{-1}]$ correspond to strain continuity in the medium. Hence $\bar{q}_{im}\,\bar{c}'_{mj} = \bar{p}'_{ij}$ are the strain optical constants for stress continuity and $\bar{p}_{im}\,\bar{s}'_{mj} = \bar{q}'_{ij}$ are the stress optical constants for strain continuity.

We have calculated the elastic and photoelastic constants of randomly oriented polycrystalline aggregate of alkali halides from the experimental values for single crystals. As these are cubic crystals, the equations get further simplified. Ranganath and **Ramaseshan**[18] found

$$\bar{p}_{11} > \bar{p}'_{11}; \quad \bar{p}_{12} < \bar{p}'_{12} \quad \text{and} \quad \bar{p}_{44} > p'_{44} \tag{9}$$

Interestingly from the average elastic tensor of the polycrystalline aggregate, it is known that

$$\bar{s}_{11} < s'_{11}; \quad (\bar{s}_{11} + 2\bar{s}_{12}) < (\bar{s}'_{11} + 2\bar{s}'_{12}) \text{ and } \bar{s}_{44} < \bar{s}'_{44} \tag{10}$$

This result has been theoretically established by **Hill**[16]. It would therefore be important to find a theoretical basis for the inequality found in the photoelastic behaviour of the polycrystalline aggregates.

Experimentally, the quantity $(\bar{p}_{11} - \bar{p}_{12})$ or $(\bar{q}_{11} - \bar{q}_{12})$ (i.e., $\bar{p}_{44}/2$ or $\bar{q}_{44}$ as the medium is isotropic) can be easily obtained rather than the individual components. To determine the sensitivity of the elastic and the photoelastic constants to stress or strain continuity, we define a parameter f given by

$$f = 2(\bar{A}_{ij} - \bar{A}'_{ij})/(\bar{A}_{ij} + \bar{A}'_{ij}) \tag{11}$$

where $\bar{A}_{ij}$ and $\bar{A}'_{ij}$ are the ijth components of elastic or photoelastic matrix for strain or stress continuity. For the elastic constant $[c_{44}]$, the maximum value of f is 0.5. However for the photoelastic constant $[q_{44}]$, the highest value of f is 8.0. Therefore, we conclude that the photoelastic birefringence is very much more sensitive to stress or strain continuity than the elastic constants are.

It must be remarked that a similar study by Flannery and **Marburger**[19], in the limit of the crystallite size-being much less than the wavelength of tight, has also shown some interesting piezooptical effects. They show that the polycrystalline photoelastic constants not only deviate appreciably from the single crystal value, but $(\bar{p}_{11} - \bar{p}_{12})$ may also change sign relative to the single crystal value.

From the simple model presented here it can be argued that, to a fair approximation, in a polycrystalline aggregate consisting of crystallites immersed in an isotropic medium like glass or plastic, the net photoelastic constant of the aggregate can be obtained from a simple mixture rule given by

$$\bar{p}_{ij} = x\,\bar{p}'_{ij} + (1-x)\,\bar{p}''_{ij}$$

where x is the fraction of the volume occupied by the crystallites and $\bar{p}'_{ij}$ and $\bar{p}''_{ij}$ are the photoelastic constants of the crystal and the matrix respectively.

3. PIEZOROTATION

If one discards the assumption that the components of the tensor relating $\boldsymbol{D}$ and $\boldsymbol{E}$ are real then

$$E = [a]D - i[G]D \tag{12}$$

If there is no dissipation of energy, then [G] is an antisymmetric second rank tensor. Mathematically $[G]$ can be replaced by $\Gamma \times$, a vector operator. Therefore

$$E = [a]D - i\Gamma \times D \tag{13}$$

This implies a rotation in the plane of polarization, which would therefore, depend upon the direction of propagation in an anisotropic crystal, and it could be expressed as a linear vector function of s the wave normal.

$$\Gamma = [v]s \tag{14}$$

[v] may be called the fundamental optical activity tensor, which need not be symmetric, i.e.,

$$v_{ij} \neq v_{ji} \tag{15}$$

And the optical rotation ρ can be written as follows :

$$\rho = g_{ij} s_i s_j \tag{16}$$

where $[g]$ is symmetric and

$$g_{ij} = \frac{\pi n_m^3}{\lambda} \left(\frac{v_{ij} + v_{ji}}{2} \right) \tag{17}$$

where n_m is mean refractive index.

The variation of optical activity with hydrostatic stress has been observed in quartz, benzil, and $NaClO_3$ (Myers and Vedam[20]), but the components of the tensor describing the phenomena was not obtained. In the first order phenomenological theory of piezorotation, change in g_{ij}, that is Δg_{ij}, is a linear function of stress or strain.

$$\Delta g_{ij} = -R_{ijkl} X_{kl} \quad \text{or} \quad \Delta g_{ij} = S_{ijkl} x_{kl} \tag{18}$$

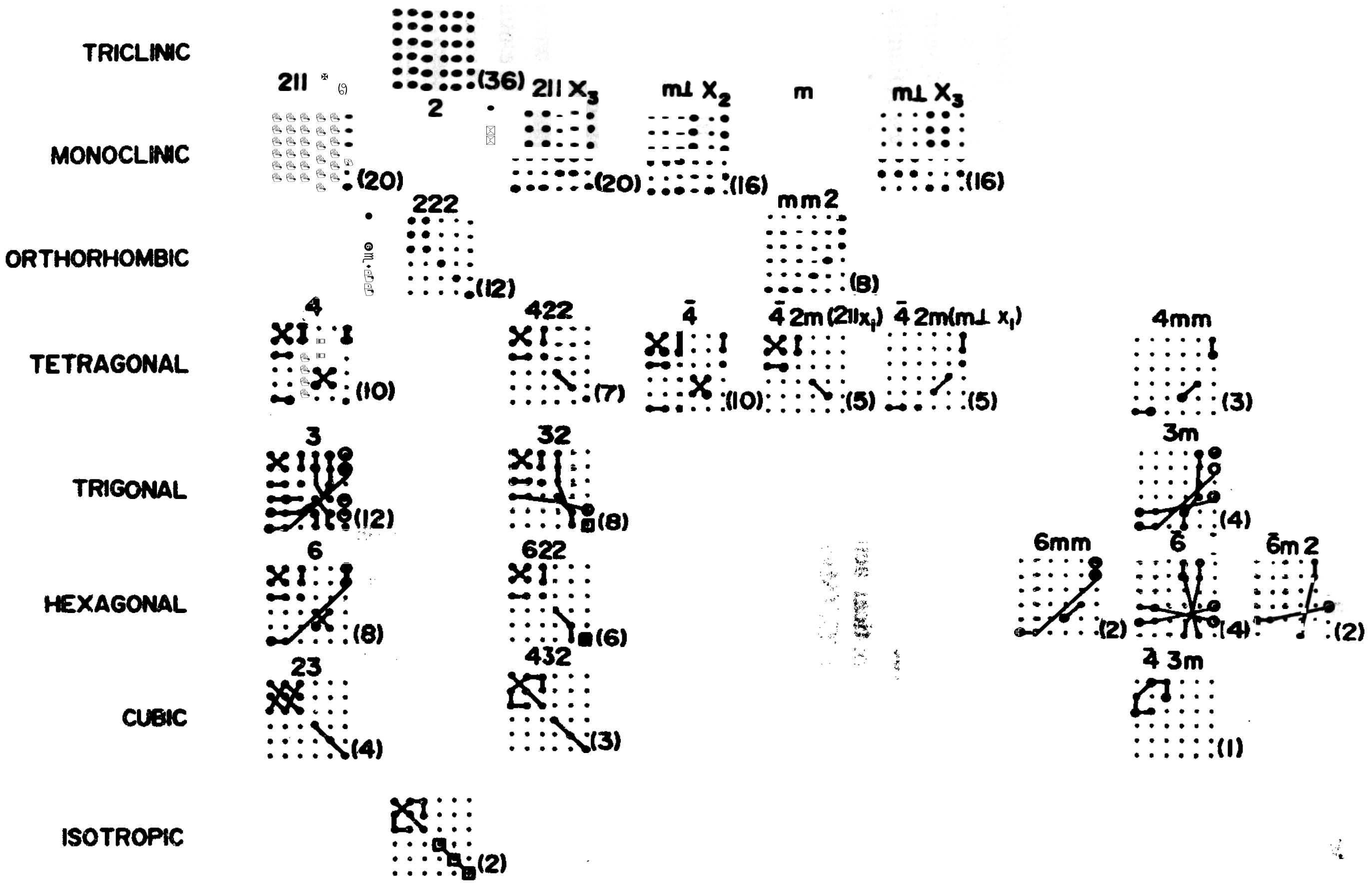
R_{ijkl} and S_{ijkl} are symmetric in i, j and k, 1. Δg_{ij} is a second rank axial tensor while X_{kl} and x_{kl} are second rank polar tensors. It can be easily shown that R_{ijkl} and S_{ijkl} are fourth rank axial tensors. They exist only in non-centrosymmetric classes. The tensor components may be called piezorotatory coefficients.

Group theoretical methods may profitably be applied to determine the number of independent piezorotatory coefficients in any crystal system. It is found that piezorotatory coefficients are non-vanishing in non-centrosymmetric classes, while they vanish in centrosymmetric classes. A most unexpected result obtained, is that the groups **4mm, 3m,** 6, **6m2, 6mm, $\bar{4}$3m** which do not show optical activity have piezorotation coefficients. The physical significance is that stress induces optical activity in these classes (Ranganath and Ramaseshan[3]). Another interesting result is that crystals belonging to the same photoelastic class get further classified by piezorotation.

The non-vanishing coefficients for all the twenty-one non-centrosymmetric classes are given in the Table 1. For point groups in column 1 $[R_{ijkl}]$ tensor is of the same form as $[q_{ijkl}]$ the piezorefractive tensor. Column 2 gives cases of optically active classes where $[R_{ijkl}]$ and $[q_{ijkl}]$ differ. Column 3 refers to cases which are normally optically inactive.

Even in the most symmetric of the point groups **$\bar{4}$3m,** this stress induced optical activity is not easy to observe, since we cannot escape from stress-induced linear birefringence. Thus piezorotation can be seen only along the optic axis of the stressed crystal. Careful analysis shows that the stress should not act along any symmetry

Table 1. Forms of the piezorotation matrics



Key :• Zero component, ● non-zero component, ●–● ai componeni san ;n, ●–○ equal mponents opposite sign. In (q)
matrices: ◉ a component equal to +2 times the ● to which joined, ◎ a pon qual to –2 tim the ● to which it is joined, ▣
= $q_{11} - q_{12}$. In (p) matrices: ◉ a component equal to the ● t which it is joi , ◎ nponent equal the minu: f the ● to which it is
joined, ▣ = 1/2 ($p_{11} - p_{12}$).

direction. Only then one may reveal the effect along the optic axis. Generally photoelastic birefringence is small and sometimes it even vanishes at a characteristic wavelength. In these systems at such wavelengths, stress-induced optical activity can be more easily observed. The effect can even be demonstrated beautifully in another way. We subject the $\bar{4}3m$ crystal to an ultrasonic stress wave propagating along **(100)** direction. Then at the wavelength of zero photoelastic birefringence, an incident circularly polarized plane wavefront **travelling** along (010) emerges as a sinusoidally corrugated wavefront. This results in the familiar Raman-Nath diffraction pattern which will have totally different intensities for right and left circular waves. This is attributable to stress-induced optical activity. On the other hand, a plane wavefront of linearly polarized light gets split into two differently corrugated wavefronts of opposite circular states. This results in a Raman-Nath diffraction with the different order elliptically polarized. This is in striking contrast to diffraction when only photoelasticity is present, where the different orders are linearly polarised.

4. PIEZOABSORPTION

If [a] is complex but the system absorbs light, then

$$E = [\hat{a}]D = ([a] + i[b])D \tag{19}$$

It is quite easy to show that *[b]* represents the absorption property of the solid if it is a second rank symmetric tensor. This may in general be again represented by an ellipsoid. If one is to describe the propagation of light in an absorbing crystal one has to consider the central elliptic sections of the index and absorption ellipsoids, normal to the direction of propagation.

In uniaxial crystals the two ellipses have their principal axes parallel; but this is nor necessarily so in the general biaxial case. When absorption is present refractive index is complex given by A = $n - ik$ or $\hat{\varepsilon} = \varepsilon - i\eta$ then $\varepsilon = n^2 - k^2$ and $\eta = 2nk$. In a general crystal

$$\varepsilon_{ij} = n_{ip}\, n_{pi} - k_{ip}\, k_{pi},\ \eta_{ij} = 2n_{ip}\, k_{pi}$$

The effect of stress or strain on an absorbing crystal is to distort the index and absorption ellipsoids. In the first order theory we can describe these effects by

$$\Delta \hat{a}_{ij} = \hat{q}_{ijkl} X_{kl} \quad \text{or} \quad \Delta\, \hat{a}_{ij} = \hat{p}_{ijkl}\, x_{kl} \tag{21}$$

where $[\hat{p}]$ and $[\hat{q}]$ are fourth rank photoelastic tensors with complex components.

It may be mentioned that in 23 and m3 cubic class with stress along (110) and direction of observation (100) both the elliptic sections are tilted with respect to the direction of stress. Thus we have two photoelastic classes in absorbing cubic crystals as well. The tilt of the absorption ellipsoid is different from that of the index ellipsoid **(Fig.1).** The method of measuring the piezorefractive and piezoabsorption coefficients requires a measurement of birefringence and dichroism (Ramaseshan, et *al.*[21]).

We can similarly think of piezoabsorptive rotation or piezorotatory dichroism when g_{ij} is made complex. Here also we find stress to induce circular dichroism in the six point groups which exhibit stress-induced optical rotation.

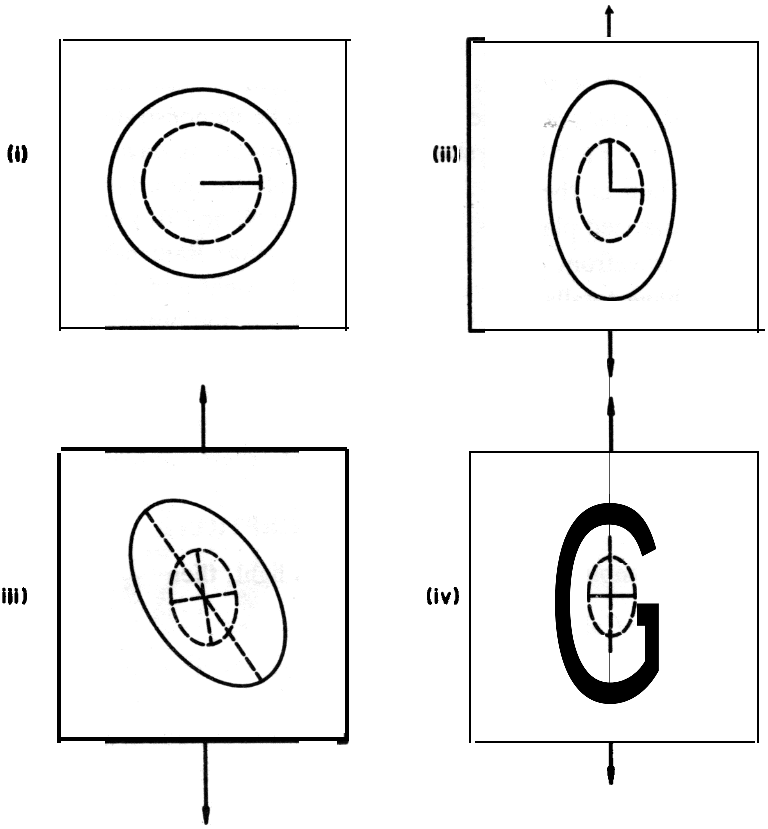


Figure 1. Sections of the index **ellipsoid** (full line) and absorption ellipsoid (dotted line). (i) before deformation; (ii) after deformation in isotropic and all cubic crystals with stress along (111) and (100); (ii) Tand T_h class with stress along (110) and direction of observation (100). Note the tilt of both the elliptic sections with respect to direction of stress; and (iv) same as (iii) but in T_d, O and O_h classes with no tilt in principal planes.

5. PIEZO-FARADAY ROTATION

Skaggs and Broersma have found stress to alter Faraday rotation in crystals. This rotation tensor given by f_{ij} is asymmetric unlike the classical optical rotation tensor g_{ij} which is symmetric. This results in transverse Faraday rotation, i.e., magneto optic rotation perpendicular to the magnetic field, in the point groups 1, $\bar{1}$, 2, m, 2/m, 3, $\bar{3}$, 4, $\bar{4}$, 4/m, 6 $\bar{6}$, and 6/m (Ranganath”). Under stress f_{ij} changes and the phenomenon can be described by a fourth rank polar tensor T_{ijkl} which is not symmetric in i, j. Also we can expect transverse Piezo-Faraday rotation in classes exhibiting only classical Faraday rotation in the unstressed state. A problem closely connected to this phenomenon was studied by Bhagavantam, the effect of stress on the optical properties of magnetic crystals. Magnetic crystals in general show a Faraday-type of rotation. Interestingly, in certain magnetic point groups, the magnetic symmetry forbids this effect. Bhagavantam has found the surprising result, that in these point groups, Faraday rotation is induced under stress. This effect has been predicted in cubic crystals belonging to m3 and m3m.

6. PIEZOROTATION IN POLYCRYSTALS

The extraction of Piezorotation and Piezo-Faraday rotation components would be generally difficult due to the Piezooptic birefringence that always accompanies the measurements. The optical rotation has to be measured along the optic axis of the stressed crystal. In view of this limitation, only a few of the tensor components can be extracted. However, as we have already seen a polycrystalline aggregate has a simple photoelastic behaviour which can be exploited to get more tensor components. A measurement of rotation along the stress direction yields an average of tensor components which can be used as additional data.

7. PIEZOOPTICS DUE TO BODY TORQUES

So far we have expressed the deformations in the index, absorption or gyration surfaces as a linear function of the stress or strain components. However, what we really impose on a crystal is the displacement gradient $u_{i,j}$. If we express deformation as a linear function of this parameter, then

$$\Delta a_{ij} = p_{ijkl}\, u_{k,l}$$

This can be rewritten as

$$\Delta a_{ij} = p_{ijkl}\, x_{kl} + p'_{ijkl}\, e_{kl}$$

where x_{kl} and e_{kl} are the symmetric and the antisymmetric strain tensors, i.e.

$$x_{kl} = 1/2(u_{k,l} + u_{l,k})$$

$$e_{kl} = 1/2\,(u_{k,l} - u_{l,k}) \tag{25}$$

and p_{ijkl} is the piezooptic tensor already discussed. The effect of e_{kl} is to bodily rotate the crystal. When we compute the contribution of this body rotation to Δa_{ij}, we get

$$p'_{ijkl} = 1/2\,[a_{il}\,\delta_{kj} + a_{lj}\,\delta_{ik} - a_{ik}\,\delta_{lj} - a_{kj}\,\delta_{ij}] \tag{26}$$

As is to be expected, p'_{ijkl} is antisymmetric in $k \rightleftarrows l$ interchange of indices. In a static experiment however, this is a very trivial effect which can be taken care off. Nelson and **Lax**[4] have pointed out for the first time, that the effects of such body rotation cannot be ignored when the shear strain is inhomogeneous as in an acoustic wave.

It is easy to see that p'_{ijkl} does not exist in isotropic and cubic symmetries. In lower symmetries we find some of the piezooptic constants to change. For example, in uniaxial crystals it changes p_{44}, while in the orthorhombic symmetry p_{44}, p_{55} and p_{66} get altered. It is also clear that these corrections will depend on a_{ij}. Hence it would be very important in highly birefringent crystals.

Nelson and **Lazay**[23] have experimentally demonstrated that these corrections to the classical piezooptic constants in the case of photoelasticity are measurable through Brillouin scattering. It is easy to show that in the case of an uniaxial crystals with the incident light propagating along the crystallographic x-axis and with the electric field polarized along the unique z-axis, the scattered intensity as seen in the x-direction for the y-polarization state and in the y-direction for the z-polarization state, are entirely different, due to this rotation effect of the crystallites.

8. PHOTOELASTIC PHASE-MATCHING IN HARMONIC GENERATION

It is well-known (Schubert and Wilhelmi[24]) that in optically nonlinear media for an efficient generation of a higher harmonic, phase-matching is a very important criterion. For example, for second harmonic generation in uniaxial crystals, the ordinary (or the extraordinary) refractive index for the fundamental wave should be equal to the extraordinary (or ordinary) refractive index for the second harmonic. This is generally achieved by sending light along a particular direction of propagation. In uniaxial crystals that have just missed the phase-matching conditions, one can, in principle, phase-match the waves by applying a hydrostatic stress or a uniaxial stress. Stress changes the ordinary and extraordinary indices and may even alter them by the right amounts. Incidentally, from the practical point of view, it is desirable to have this direction of phase-matching perpendicular to the optic axis. There are possibilities that an application of stress may even change the direction of phase-matching to this orientation. Measurements of photoelastic constants together with their frequency dispersion are therefore very valuable for these applications.

9. LIGHT SCATTERING AND PIEZOOPTICS

Generally piezooptical constants are measured using a transmission technique, i.e., by studying the light beam coming out of the material. It is well-known (Weller[25] and Chandrasekharan[26]) that stress birefringence can be detected by scattering techniques. This has been applied to the study of photoelastic stress (Srinath[27]) as well. Here one observes light scattered perpendicular to direction of the main beam which is usually very intense. This method can be used to demonstrate the differences between the two photoelastic classes in cubic crystals. For example, let the incident light be a circular wave travelling along the cube edge and the crystals are compressed along the cube edge. In one case the crystal becomes uniaxial about the stress direction so that the main wave remains in the same polarization state all along its path. Thus scattered light will be uniformly in the same polarization state. On the other hand, in the second cubic class, the crystal becomes orthorhombic with the cube edge(s) as the symmetry direction(s). In this case the incident wave undergoes changes in polarization state as it travels through the material, so that the scattered light will exhibit a fringe pattern which will be particularly pronounced when observed along the face diagonal (110). This is so because for this direction of transverse viewing, periodically at certain depths the polarization is linear with the electric vibrations normal to the direction of viewing. Separation between the fringes gives photoelectric birefringence. Thus the presence or otherwise of this fringe pattern can distinguish the two cubic nhotoelastic classes.

10. PIEZOOPTICS OF *1D* AND *2D* CONDUCTORS

Crystals that show nearly metallic conduction along a direction or a plane have been reported in recent times. The piezooptics of such systems do not appear to have been discussed in literature. We shall briefly consider this phenomenon in the case of uniaxial crystals.

In a simple phenomenological theory we can approximate these systems to have a negative dielectric constant ε (since k will be generally very much larger than n) along the directions of metallic conduction. It then immediately follows that the dielectric or the index tensor cannot be any more represented by a closed surface. It will be in general a hyperboloid of one sheet in 1*D* and of two sheets in 2*D* metallic systems. In these symmetries even the absorption tensor *[b]* cannot be represented by a closed surface. It will be a cylinder for 2*D* and a pair of parallel planes for 1*D* metallic conduction in crystals of uniaxial symmetry.

In piezooptics we look at deformations of such tensor surfaces. The symmetry of the index and the absorption surfaces indicate that not all the components of $[\hat{p}]$ and $[\hat{q}]$ need be complex. The piezooptic coefficients that represent certain deformations of these surfaces will be complex. For example, in the hexagonal system p_{33} or q_{33} and p_{31} or q_{31} will be complex for a 1*D* conducting crystal, while the same coefficients will be real for 2*D* conducting crystals, which will have p_{11} or q_{11} and p_{12} or q_{12} as complex quantities. Details of this phenomenon are being worked and will be published elsewhere.

11. THEORY OF PIEZOOPTIC PHENOMENA

At present there is no satisfactory theory explaining the various piezooptic phenomena in crystals. Attempts have been mainly focussed on understanding photoelasticity. Here too only in ionic crystals has any progress been made (Mueller[28], Ramaseshan and Sivaramakrishnan[29], Srinivasan[30] and Ranganath, et *al.*[31]) . Measurement of the photoelastic constant at various wavelengths in alkali halides has revealed many interesting features. For example, sodium halides have negative and potassium halides have positive photoelastic birefringence and in the latter systems it changes over to negative values at shorter wavelengths becoming optically isotropic at a wavelength characteristic of the material. In the photoelastic birefringence there appears to be, a contribution not only from the lattice deformation, but also from the splitting of the absorption bands of the crystal.

However, in covalent systems it has been argued (Weinstein, et *al.*[32]) that a measurement of the following parameter

$$\chi = \frac{\Sigma_{ij} p_{ij} x_j \varepsilon_i^2/(\varepsilon_i - 1)}{3 \Sigma_i x_i} \qquad (27)$$

indicates the dimensionality of the covalent network. They found $\chi < 0$ to imply a 3-D network. However, $\chi > 1$ implies a dimensionality less than 3-D. This is particularly interesting since for Ge family $\chi < 0$, while for chalcogen-based molecular solids $\chi > 1$. For compounds with IV-VI group elements $0 < \chi < 1$. This situation is at present beyond the scope of the proposed model.

REFERENCES

1. Pockels, F., Lehrbuch der Kristalloptic, (Teubner, Berlin), 1906, p.480.
2. Bhagvantam, S., *Proc. Ind. Acad. Sci.,* A16 (1942), 359.
3. Ranganath, G.S. & Ramaseshan, S., *J. Opt. Soc. Am., 59* (1969).
4. Nelson, D.F. & Lax, M., *Phys. Rev. Letts., 24* (1970), 379.
5. Bhagavantam, S., *Proc. Ind. Acad. Sci., A35* (1950), 129.
6. Ramaseshan, S. & Chandrasekharan, V., *Current Science, 20* (1951), 150.
7. Ramachandran, G.N. & Chandrasekharan, V., *Proc. Znd. Acad. Sci.,* A33 (1952), 199.
8. Mueller, H., *Z. Kristolloger,* A99 (1938), 122.
9. Raman, C.V. & Nath, N., *Proc. Ind. Acad. Sci., A2* (1935), 406.
10. Vedam, K. & Ramachandran, G.N., *Proc. Znd. Acad. Sci., A34* (1951), 240.
11. Vedam, K., Schmidt, E.D.D. & Roy, R., *J. Am. Ceram. Soc.,* 49 (1966), 532.
12. Raman, C.V. & Viswanathan, K.S., *Proc. Ind. Acad. Sci.,* A41 (1955), 37.
13. Ranganath, G.S. & Ramaseshan, S., *Opt. Acta.,* 19 (1972), 781.
14. Voight, W., *Ann. Phys. Leipzig, 38* (1889), 573.
15. Reuss, A., *Z. Angew Math. Mech., 9* (1929), 49.
16. Hill, R., *Proc. Phys. Soc. Lond., Sec.* A65 (1952), 349.
17. Bhagavantam, S., *Proc. Ind. Acad. Sci., A37* (1953), 585.
18. Ranganath, G.S. & Ramaseshan, S., *Pramana,* 1 (1973), 78.
19. Flannery, M. & Marburger, J., *Appl. Phys. Rev. Letts., 23* (1976), 600.
20. Myers, M.B. & Vedam, K., *J. Opt. Soc. Am., 55* (1965), 1180.
21. Ramaseshan, S., Ranganath, G.S., Ramesh, T.G. & Rajagopalan, S.R., *Mater. Rev. Bull., 4* (1969), 535.
22. Ranganath, G.S., *Proc. Ind. Acad. Sci.,* A75 (1972), 237.
23. Nelson, D.F. & Lazay, P.D., *Phys. Rev. Letts., 25* (1970), 1187.
24. Schubert, M. & Wilhelmi, B., Nonlinear Optics and Quantum Electronics, (John Wiley, New York), 1986.
25. Weller, R., *J. Appl. Phys., 10* (1939), 266.
26. Chandrasekharan, V., *Proc. Ind. Acad. Sci.,* A26 (1947), 110.
27. Srinath, L.S., Scattered Light Photoelasticity, (Tata McGraw Hill, New Delhi), 1983.
28. Mueller, H., *Physics, 6* (1935), 179.
29. Ramaseshan, S. & Shivaramakrishnan, V., *Current Science,* 25 (1956), 246.
30. Srinivasan, R., *Z. Physik,* 155 (1959), 281.
31. Ranganath, G.S., Rajagopalan, S.R. & Ramaseshan, S., *J. Radiation, 4* (1970), 313.
32. Weinstein, B.A., Jallen, R. & Slade, M.L., *Phys. Rev.,* B24 (1981), 4652.