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Photo-elastic Behaviour of Ammonium Alum Crystals

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To test an earlier prediction by Bhagavantam, based on a group theoretical method, that the $T-23$ and $T_h-2/m\bar{3}$ classes of the cubic system need four independent constants, instead of three, for describing their photo-elastic properties, crystals of ammonium alum belonging to the $T_h-2/m\bar{3}$ class were studied completely. It was found that the crystal needs four independent constants and all the four stress-optical constants of alum have been determined. For sodium D light, they are

$$\begin{aligned} (q_{11}-q_{12}) &= -5.93 \times 10^{-13}, & (q_{11}-q_{13}) &= -5.25 \times 10^{-13}, & q_{44} &= -1.15 \times 10^{-13}, \\ q_{11} &= 5.5 \times 10^{-13}, & q_{12} &= 11.6 \times 10^{-13}, & q_{13} &= 10.9 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}. \end{aligned}$$

The values of the strain-optical constants are

$$\begin{aligned} (p_{11}-p_{12}) &= -0.0854, & (p_{11}-p_{13}) &= -0.0756, & p_{44} &= -0.0092, \\ p_{11} &= 0.38, & p_{12} &= 0.46, & p_{13} &= 0.45. \end{aligned}$$

1. Introduction

In the preceding paper in this journal (Bhagavantam & Suryanarayana, 1949) a group theoretical method is given of deriving the number of independent constants needed to describe any physical property in the 32 crystal classes. The numbers found in the case of photo-elasticity are at variance with those given by Pockels (1889, 1906, p. 460) and current in the literature for the classes

$$\begin{aligned} C_4-4, S_4-\bar{4}, C_{4h}-4/m; C_3-3, S_6-\bar{3}, C_{3h}-3/m, \\ C_6-6, C_{6h}-6/m; T-23 \text{ and } T_h-2/m\bar{3}. \end{aligned}$$

The non-vanishing stress-optical coefficients, when worked out directly for all the 32 classes, confirm the findings of the group-theoretical method. They show that in the case of photo-elasticity the 32 crystal classes are divided into eleven *Obergruppen*, and not nine as was formerly believed to be the case.

The present experimental investigation of the photo-elastic behaviour of certain crystals is undertaken to clear the existing discrepancy. While, according to Pockels, the cubic classes require only three constants, the new theory predicts four independent photo-elastic constants for the class $T_h-2/m\bar{3}$. Of the cubic crystals with which Pockels himself had worked, two, namely

potassium alum and ammonium alum, belong to this class (Wyckoff, 1931). The original paper of Pockels (1889) dealing with the alums shows that the orientations of his prisms were not suitable for deciding the present issue. We therefore made further measurements on alums, of which those on potassium alum have already been published (Bhagavantam & Suryanarayana, 1947). Results on ammonium alum crystals now obtained are reported here. They are in entire agreement with the findings of the group-theoretical method.

2. Theoretical considerations

The photo-elastic behaviour of the $T-23$ and $T_h-2/m\bar{3}$ classes of crystals is given by the equations

$$\left. \begin{aligned} B_{11}-B &= -(q_{11}P_{xx}+q_{12}P_{yy}+q_{13}P_{zz}), & B_{23} &= -q_{44}P_{yz}, \\ B_{22}-B &= -(q_{13}P_{xx}+q_{11}P_{yy}+q_{12}P_{zz}), & B_{31} &= -q_{44}P_{zx}, \\ B_{33}-B &= -(q_{12}P_{xx}+q_{13}P_{yy}+q_{11}P_{zz}), & B_{12} &= -q_{44}P_{xy}. \end{aligned} \right\} \quad (1)$$

In these equations $B=1/n^2$, $B_{11}^2=1/n_{11}^2$, etc., where n is the refractive index of the undeformed cubic crystal, n_{11} etc., are the components describing the Fresnel ellipsoid in the deformed condition, $P_{xx}\dots P_{xy}$ are the components of stress, and q_{11} , q_{12} , q_{13} and q_{44} are the

four independent stress-optical coefficients, all referred to the crystallographic axial system. According to Pockels $q_{12} = q_{13}$ and we then have only three constants.

One striking result of the non-equivalence of q_{12} and q_{13} is that the crystal may become biaxial by a simple compression along a cube axis, say z axis, i.e. the crystal under stress in that direction becomes optically orthorhombic and not tetragonal. In the classes T_a-43m , $O-432$ and $O_h-4/m\bar{3}2/m$, where $q_{12} = q_{13}$, the crystal should become uniaxial for such compressions. This point could be settled by a direct observation of the path differences per unit pressure and unit length of the light beam in the crystal, between two beams, one vibrating along and the other perpendicular to, the direction of the pressure for light propagated along the other two cube axes, say x and y , i.e. when the observations are made along these axes. These two path differences should be different if the crystal has four independent constants. Again, for the same direction of pressure, the path differences should be identical when observations are made parallel to $[110]$ and $[\bar{1}10]$ directions.

The path differences for the various cases are expressed in Table 1. The derivations follow well-known principles, employed earlier by Pockels.

parallel beam passed through a Nicol mounted on a circular scale with vernier, the vibration direction of the Nicol being maintained at $+45^\circ$ or -45° to the vertical. The beam then passed through the crystal prism into a Babinet compensator, the principal axes of which were vertical and horizontal. After passing through the compensator the light was observed through an eye-piece containing a Nicol crossed with respect to the polarizer, and the usual Babinet fringes were obtained. When the crystal prism was compressed, the fringes shifted one way or the other and the magnitude of the shift was a measure of the path difference produced between the horizontally and vertically vibrating beams of light passing through the compressed crystal.

Compression was produced by a lever arrangement similar to that of Filon (1903-4). Details of the arrangement are described in an earlier paper by the authors (Bhagavantam & Suryanarayana, 1947).

Stresses not exceeding about 0.5 kg.mm.^{-2} were used in all cases, except in the $[111]$ direction. In this case, to obtain a displacement of the Babinet fringes which could be measured reasonably accurately, stresses up to 0.9 kg.mm.^{-2} were employed. Following Pockels, the displacement of the fringes was measured at the

Table 1. *Expressions for path differences*

No.	Direction of pressure	Direction of observation	Expression for path-difference for $T-23$ and $T_h-2/m\bar{3}$ classes	Expression according to Pockels' scheme	
1	[001]	[100]	$\frac{1}{2}n^3 (q_{11} - q_{12})$	} $\frac{1}{2}n^3 (q_{11} - q_{12})$	
2	[001]	[010]	$\frac{1}{2}n^3 (q_{11} - q_{13})$		
3	[001]	[110]	$\frac{1}{4}n^3 (2q_{11} - q_{12} - q_{13})$		
4	[001]	[$\bar{1}10$]	} $\frac{1}{2}n^3 (q_{44})$	} $\frac{1}{2}n^3 (q_{44})$	
5	[111]	[$\bar{2}11$]			
6	[111]	[011]			

If $q_{12} = q_{13}$, expressions in column 4 become identical with those given in column 5. An examination of the Table shows that two rectangular parallelepipeds with their lengths parallel to a cube axis, say [001], and the other two edges parallel (1) to the cube axes [100] and [010], and (2) to the [110] and [$\bar{1}10$] directions, will provide evidence in favour of the one or the other scheme, with additional internal checks. There is no difference between the two schemes if pressure is along [111].

3. Experimental arrangements

All the prisms needed were cut and polished from crystals grown in this laboratory by evaporation over a period of some months from aqueous solutions of Merck's pure ammonium alum. Single crystals with well-developed cube and octahedral faces and without flaws were selected. All the prisms of length 8 mm. and lateral dimensions 3 to 4 mm. had at least one of their ends terminated on a natural face. None of the prisms showed double refraction under crossed Nicols in an unstrained state.

Light from a sodium-vapour lamp was condensed on the slit of a collimator. The emergent horizontal

middle of the length of the prism at three places along its breadth, namely near the left edge, the middle and the right edge. Before each set of observations, a preliminary measurement of the fringe displacement at the three positions was made and only when the distribution of the load was fairly uniform were the final measurements made. In each of the three positions, the polarizing Nicol and the analysing Nicol of the compensator were turned through 90° , so that in the two positions the incident light was polarized at $+45^\circ$ and -45° to the vertical. Thus six measurements of the fringe displacement were made in all. For each of the six measurements, the initial position (without load) of the central Babinet fringe was taken twice before placing the load and twice after removing the load. The final position of the Babinet fringe (on load) was obtained by four settings of the crosswire on the edge of the fringe. Thus each of the six measurements was the mean difference of four settings of the initial and final positions of the fringe. In none of the prisms was there any noticeable consistent residual double refraction after unloading (as deduced from the shift of the initial position).

The prism was then turned through 90° and the measurements were repeated similarly with identical load.

To determine the absolute value of the stress-optical constants, a measure of the absolute change in the refractive index for vertically or horizontally polarized beam of light was needed. For this purpose, the crystal itself was used as an interferometer by forming localized fringes in the crystal, as has been done recently by Ramachandran (1947) in his determination of the photo-elastic constants of diamond.

The procedure adopted was to work a pair of surfaces of the prism so as to obtain reasonably widely separated localized fringes, i.e. curves of equal thickness, with reflected light. If light is reflected at the usual 45° angle at a glass plate the observation of the shift of the fringes is not possible when the light is vibrating horizontally, because of the very low intensity. Light was therefore reflected at an angle of about 10° on to a pair of faces of the prism, and the localized fringes, formed by reflection of the beam at the two surfaces, were observed through a microscope. In obtaining the fringes, thicknesses of the crystal which gave rise to destructive interference between the D_1 and D_2 lines had to be avoided. The microscope was focused on a reference mark on the crystal itself and the movement of the fringes on loading was observed against this mark. The shift of the fringes is the combined result of the change in the thickness of the crystal and the change in the refractive index. Because of the low value of the elastic constants, a shift of one complete fringe could be produced in alums by stresses of the order of 0.3 kg.mm.^{-2} . The load producing a shift of one or two complete fringes was noted for rays polarized vertically and horizontally. The sign of the shift, namely, whether the order of interference is decreasing or increasing at the point under observation, settled the sign of the stress-optical constants unambiguously.

A description of the three prisms employed in the investigation is given in Table 2. The orientations are accurate to within 1° .

4. Results

(a) Determination of $(q_{11} - q_{12})$, $(q_{11} - q_{13})$ and q_{44}

The observations with the Babinet compensator for the three prisms are given in detail in Table 3.

The fringe width of the Babinet compensator was 144.6 divisions, which corresponded to a path retardation of one wave-length of sodium light. The stress-optical constants were calculated as follows. From the mean shift of the Babinet fringe, the path difference δ corresponding to unit stress (1 dyne cm.^{-2}) and unit length (1 cm.) of light beam in the stressed crystal were calculated, noting that the path difference is $5.893 \times 10^{-5} \text{ cm.}$ for a shift of 144.6 divisions. The stress-optical constant in the corresponding direction of observation is $2\delta/n^3$, where δ is the path difference ($n_x - n_z$) between the horizontally and vertically vibrating light beams. In the above experiments, the shift was always in the same direction as for common glass, i.e. the crystal becomes negative uniaxial when compressed along the trigonal axis. Hence, the sign of the constants q_{44} , $(q_{11} - q_{12})$ and $(q_{11} - q_{13})$ is negative.

An examination of Table 3 reveals the following facts:

(1) In prism I, stress-optical constants in the x and y directions differ from each other by 12%, which is much more than any possible experimental error.

(2) The two values bracketed together in prism II are the values of one and the same constant when observed along two directions, and differ by 1.5%. The mean (5.57) of these values differs by 2% from the mean (5.68) of the two constants from prism I, which difference is of the order of the experimental error for the first two prisms.

(3) The two values for prism III are for the same constant q_{44} and differ by 3.5%, which experimental error is higher in this case because of the smaller shift of the Babinet fringe measured.

In view of the above, it can be stated with confidence, that there is a real difference between the quantities $(q_{11} - q_{12})$ and $(q_{11} - q_{13})$ of about 12%. Hence the

Table 2. Description of the prisms employed

No. of prism	Length		Breadth		Thickness	
	cm.	Parallel to	cm.	Parallel to	cm.	Parallel to
I	0.82	[001]	0.406	[100]	0.406	[010]
II	0.82	[001]	0.409	[110]	0.410	[110]
III	0.82	[111]	0.398	[211]	0.334	[011]

Table 3. Photo-elastic constants of ammonium alum: observations with compensator

Prism	Observation parallel to Direction	cm.	Load in kg.	Mechanical advantage of lever	Shift of the Babinet fringe in divisions of head scale						Mean	Stress-optical constant (in $10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$)	Expression for the constant
					Left		Middle		Right				
					$+45^\circ$	-45°	$+45^\circ$	-45°	$+45^\circ$	-45°			
I	[100]	0.406	2	3.973	45.62	45.5	44.5	45.0	42.12	41.5	44.04	6.02	$(q_{11} - q_{12})$
	[010]	0.406	2	3.973	41.32	40.22	41.25	40.38	35.25	36.2	39.10	5.34	$(q_{11} - q_{13})$
II	[110]	0.409	2	3.973	41.25	40.88	40.6	40.5	40.38	40.5	40.68	5.61	$\frac{1}{2}(2q_{11} - q_{12} - q_{13})$
	[110]	0.410	2	3.973	41.75	41.0	38.88	40.88	39.4	39.25	40.19	5.53	
III	[211]	0.398	3	3.973	15.88	15.38	13.12	14.88	15.5	16.0	15.13	1.13	q_{44}
	[011]	0.334	3	3.973	13.12	13.75	13.0	13.88	12.62	12.62	13.16	1.17	

description of the photo-elastic behaviour of ammonium alum needs four independent constants, and not three, because $q_{12} \neq q_{13}$.

Giving a weight of 2 to the mean value of $(2q_{11} - q_{12} - q_{13})$ from prism II, and unit weight to the values of $(q_{11} - q_{12})$ and $(q_{11} - q_{13})$ from prism I, the most probable values of $(q_{11} - q_{12})$ and $(q_{11} - q_{13})$ are deduced by forming normal equations from the above observationalequations. This gives $(q_{11} - q_{12}) = -5.93 \times 10^{-3}$ and

$$(q_{11} - q_{13}) = -5.25 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}.$$

It is from these values that $(p_{11} - p_{12})$ and $(p_{11} - p_{13})$ are calculated in a later paragraph. The mean value of q_{44} is $-1.15 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$.

(b) *Determination of the absolute stress-optical coefficients q_{11} , q_{12} and q_{13}*

For this purpose, prism I was used. The dimension parallel to [010] was reduced from 0.406 cm. to 0.293 cm. and a set of distorted elliptical fringes was obtained in that direction. The prism was acting like a double convex lens of very large focal length in the direction of observation. A suitable mark near the middle of the observed crystal face was chosen and the loads producing a shift of two complete fringes at the mark, for light vibrating vertically and horizontally, were noted. The spacing of the fringes at the point of observation was about $\frac{1}{2}$ mm. and the dark bands themselves, for well-known reasons, were not very sharp. On this account, any shift due to a change of about 50 g. in a load of 1500 g. just escaped notice. With these limitations, the loads for a shift of two complete fringes for vertically and horizontally vibrating beams of light were 2000 and 1500 g. respectively, the mechanical advantage of loading being 3.973. The shift was away from the centre of the fringe system and hence, at the point of observation, the order of interference was increasing.

The absolute values of the constants were calculated from the equations

$$n - n_x = \frac{\lambda}{2t_2} \left[\frac{2n}{\lambda} (t_2 - t_1) - \delta n \right],$$

$$q_{11} = (n - n_x) \frac{2}{n^3 P_{zz}}.$$

Here n is the refractive index of the undeformed crystal, n_x is the refractive index in the stressed crystal for light with vibration direction vertical, $(t_2 - t_1)$ is the increase in thickness of the crystal along the direction of observation for the corresponding load, λ is the wavelength of light, δn is the increase in the order of interference at the point of observation, and P_{zz} is the stress producing the shift. In our experiments $\delta n = +2$ and $n = 1.459$

(Landolt & Börnstein, 1931, p. 917). Similar equations for light vibrating horizontally are

$$n - n_x = \frac{\lambda}{2t_3} \left[\frac{2n}{\lambda} (t_3 - t_1) - \delta n \right],$$

$$q_{13} = (n - n_x) \frac{2}{n^3 P_{zz}}.$$

The value of P_{zz} here is different from the P_{zz} given earlier.

The elastic constants of ammonium alum have been determined recently in this laboratory by Sundara Rao (1948). They are

$$C_{11} = 2.50 \times 10^{11}, \quad C_{12} = 1.06 \times 10^{11}, \\ C_{44} = 0.80 \times 10^{11} \text{ dyne cm.}^{-2}.$$

Employing the data given above, we get

$$q_{11} = 4.9 \times 10^{-13} \text{ and } q_{13} = 11.5 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1},$$

giving a difference of 6.6 compared with the most probable value of 5.25 given earlier. It must be pointed out that a value of 1900 g., for example, instead of 2000 g. for the load used brings q_{11} to 5.9×10^{-13} from the present $4.9 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$. Hence, in view of the facts stated earlier about the fringe displacements, the values of q_{11} and q_{13} can be regarded as giving the order of magnitude only. Provisionally the values may be taken as

$$q_{11} = 5.5 \times 10^{-13}, \quad q_{13} = 10.9 \times 10^{-13},$$

and $q_{12} = 11.6 \times 10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$,

as obtained by forming normal equations after giving a weight of 4 for the Babinet observations relative to the observations on absolute values.

(c) *Evaluation of the strain-optical coefficients*

The four strain-optical coefficients were calculated from the equations

$$p_{11} = q_{11} C_{11} + (q_{12} + q_{13}) C_{12}, \quad p_{44} = q_{44} C_{44},$$

$$p_{12} = q_{12} C_{11} + (q_{11} + q_{13}) C_{12},$$

$$p_{11} - p_{12} = (q_{11} - q_{12}) (C_{11} - C_{12}),$$

$$p_{13} = q_{13} C_{11} + (q_{11} + q_{12}) C_{12},$$

$$(p_{11} - p_{13}) = (q_{11} - q_{13}) (C_{11} - C_{12}).$$

They are cited in Table 4.

5. Discussion of the results

The photo-elastic constants of potassium and ammonium alums are cited in Table 4.* The values of Pockels (Sziessy, 1929, p. 855) cannot be compared directly

* The stress-optical constants calculated from the compensator observations of potassium alum published earlier by the authors are all 4% higher on account of a small error in the conversion factors.

Table 4. *Photo-elastic constants of potassium and ammonium alums*
Stress-optical constants (in $10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$)

	Stress-optical constants (in $10^{-13} \text{ cm.}^2 \text{ dyne}^{-1}$)						Strain-optical constants (pure numbers)					
	$q_{11} - q_{12}$	$q_{11} - q_{13}$	q_{44}	q_{11}	q_{12}	q_{13}	$p_{11} - p_{12}$	$p_{11} - p_{13}$	p_{44}	p_{11}	p_{12}	p_{13}
Potassium alum	-5.21	-4.64	-0.63	3.7	9.1	8.5	-0.0758	-0.0675	-0.0054	0.27 ₅	0.35 ₄	0.34 ₅
Ammonium alum	-5.93	-5.25	-1.15	5.5	11.6	10.9	-0.0854	-0.0756	-0.0092	0.38	0.46	0.45

with these values, because they were derived from prisms of orientations which involve all the four constants, but without taking such a fact into account.

It may be noted that the stress-optical constants for ammonium alum are the largest known of all cubic crystals so far studied.

Four independent constants are required for describing the photo-elastic behaviour in class $T_h-2/m\bar{3}$, because the cube axes are only digonal and not tetragonal. It can now be stated as a general rule applicable to all cubic crystals, that pressure along any axis of trigonal or tetragonal symmetry makes the crystal optically uniaxial, whereas pressure along any digonal axis, or in a general direction, makes the crystal biaxial.

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Note on the Bhagavantam-Suryanarayana Method of Enumerating the Physical Constants of Crystals

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A method alternative to that of Bhagavantam and Suryanarayana is given for enumerating, by group theory, the number of independent constants for any physical property of crystals in the 32 classes; the method consists of finding first the explicit form of the representation in question for the full group of all rotations and reflexions and then obtaining the form for the individual crystal classes by specialization.

In this journal Bhagavantam & Suryanarayana (1949) have described a group-theoretical method of determining the number of independent constants describing various physical properties of crystals in each of the 32 crystal classes, their method being a variant of a method developed by the present author (Jahn, 1937) in a paper on the enumeration of the elastic constants of crystals. It is the purpose of this note to show that the results of Bhagavantam & Suryanarayana may be obtained in a different manner which adheres more closely to the original method of the writer.

Bhagavantam & Suryanarayana consider a number of different types of physical properties (relations between tensors) for which they list the character of the appropriate representation. It may be verified that the representation in each case is expressible in terms of

that of a polar vector as shown in the accompanying Table I.†

In Table I, V denotes the representation of a polar vector and the notation of Tisza (1933) is used for the symmetrical product, $[V^2]$, of V with itself and higher

† *Notation for irreducible representations.* In this paper, the standard notation used in molecular spectroscopy for the irreducible representations of the symmetry groups is adhered to: thus A or B denote always one-dimensional representations, E two-dimensional, F three-dimensional; different representations being distinguished by different suffixes. The $(2L+1)$ -dimensional representation of the group (R_∞) of all rotations is denoted by D_L . g and u distinguish representations which are even or odd with respect to inversion, whilst a single or a double prime ('') is used to distinguish representations which are even or odd with respect to a plane of symmetry. For a detailed account of this notation, which goes back to Tisza (1933), and for a full account of the algebra of irreducible representations, the reader is referred to the book by Herzberg (1945).

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