Stress optical behaviour of ammonium pentaborate under linear and hydrostatic stresses

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Ammonium pentaborate (APB) is an orthorhombic crystal of mm2(C_{2v}) class. In the present investigation the authors present the results on the stress birefringence of APB under linear and hydrostatic stresses. It is shown that from stress birefringence measurements under linear stresses alone, the effect of hydrostatic pressure on the birefringences of an orthorhombic crystal can be obtained. A comparison is drawn between the natural birefringence and stress birefringence under linear and hydrostatic stresses.

1. Introduction

Ammonium pentaborate ($NH_4B_5O_8 \cdot 4H_2O$) is a biaxial crystal of the orthorhombic system and belongs to the class mm2(C_{2v}). It has twelve stress optical (q_{ij}) and twelve strain optical (p_{ij}) constants, Narasimhamurty [1]. The piezoelectric and dielectric properties of ammonium pentaborate (APB) and its isomorph potassium pentaborate (KPB) have been determined by Cook and Jaffe [2].

In the recent times a study of the elasto-optic properties of crystals has come into considerable prominence because of the innumerable technological applications of acousto-optic modulators and deflectors.

In the present communication we report stress optical studies on linearly stressed single crystals of APB for several orientations. From these studies we computed the effect of hydrostatic pressure on the double refraction of the crystal for observations in the three principal directions.

2. Theoretical considerations

The optical index ellipsoid of a crystalline medium in the undeformed condition when referred to an arbitrary rectangular coordinate system, is given by

$$B^0_{11}X^2 + B^0_{22}Y^2 + B^0_{33}Z^2 + 2B^0_{23}YZ + 2B^0_{31}ZX + 2B^0_{12}XY = 1 \qquad (1)$$

where B^0_{ij}, the optical parameters in the undeformed state, are given by

$$B^0_{11} = \frac{1}{(n^0_{11})^2}, \ldots, B^0_{23} = \frac{1}{(n^0_{23})^2} \ldots \text{etc.},$$

n^0_{ij} being the refractive indices.

In the undeformed state the index ellipsoid in the most general form is represented by

$$B_{11}X^2 + B_{22}Y^2 + B_{33}Z^2 + 2B_{23}YZ + 2B_{31}ZX + 2B_{12}XY = 1 \qquad (2)$$

where

$$B_{11} = \frac{1}{n^2_{11}}, \ldots, B_{23} = \frac{1}{n^2_{23}} \ldots \text{etc.}$$

According to Pockels' phenomenological theory of photoelasticity [1], in the region of perfect elasticity, where the generalized Hooke's law holds, the differences between B_{ij}, the optical parameters, in the deformed and undeformed states, are linear functions of the components of stress. Thus

$$B_{11} - B^0_{11} = -(q_{11}P_{xx} + q_{12}P_{yy} + q_{13}P_{zz} + q_{14}P_{yz} + q_{15}P_{zx} + q_{16}P_{xy})$$
$$B_{12} - B^0_{12} = -(q_{61}P_{xx} + q_{62}P_{yy} + q_{63}P_{zz} + q_{64}P_{yz} + q_{65}P_{zx} + q_{66}P_{xy}) \qquad (3)$$

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TABLE I Expressions for stress birefringence for a crystal of orthorhombic system

Direction of stress	Direction of observation	Stress birefringence per unit stress per unit thickness
[100]	[010]	$\frac{1}{2}(n_x^3 q_{11} - n_z^3 q_{31}) + S_{12}(n_z - n_x)$
[100]	[001]	$\frac{1}{2}(n_x^3 q_{11} - n_y^3 q_{21}) + S_{13}(n_y - n_x)$
[010]	[001]	$\frac{1}{2}(n_y^3 q_{22} - n_x^3 q_{12}) + S_{23}(n_x - n_y)$
[010]	[100]	$\frac{1}{2}(n_y^3 q_{22} - n_z^3 q_{32}) + S_{12}(n_z - n_y)$
[001]	[100]	$\frac{1}{2}(n_z^3 q_{33} - n_y^3 q_{23}) + S_{13}(n_y - n_z)$
[001]	[010]	$\frac{1}{2}(n_z^3 q_{33} - n_x^3 q_{13}) + S_{23}(n_x - n_z)$
[M]	[M′]	$\frac{1}{8}n_{yz}^3(q_{22} + q_{23} + q_{32} + q_{33} + 2q_{44}) - \frac{1}{4}n_x^3(q_{12} - q_{13})$ $- \frac{1}{4}(n_{yz} - n_x)(S_{22} + 2S_{23} + S_{33} - S_{44})$
[L]	[L′]	$\frac{1}{8}n_{zx}^3(q_{11} + q_{13} + q_{31} + q_{33} + 2q_{55}) - \frac{1}{4}n_y^3(q_{21} + q_{23})$ $- \frac{1}{4}(n_{zx} - n_y)(S_{11} + 2S_{13} + S_{33} - S_{55})$
[N]	[N′]	$\frac{1}{8}n_{xy}^3(q_{11} + q_{12} + q_{21} + q_{22} + 2q_{66}) - \frac{1}{4}n_z^3(q_{31} + q_{32})$ $- \frac{1}{4}(n_{xy} - n_z)(S_{11} + 2S_{12} + S_{22} - S_{66})$

[M] indicates a direction in the yz plane equally inclined to y and z axes.
[L] indicates a direction in the xz plane equally inclined to x and z axes.
[N] indicates a direction in xy plane equally inclined to x and y axes.
[M′], [L′], [N′] indicate directions perpendicular to [M], [L] and [N] in the yz, zx and xy planes, respectively.

where q_{ij} are the stress optical constants and P_{xx}, $P_{yy}, \ldots, P_{xy}$ are the stress components. The negative sign arises due to Pockels' assumption that a compressional stress is positive and that positive stress produces negative strain.

2.1. Effect of linear stress

Expressions for the photoelastic birefringence developed in a linearly stressed orthorhombic crystal for several of its orientations have been derived by Narasimhamurty [1] and therefore only the final expressions are reproduced in Table I. These expressions are for the total observed birefringence (being equivalent to the relative path retardations per unit stress per unit thickness) and include the contribution to the change in birefringence due to change in thickness of the birefringent crystal specimen when under linear stress. On separating the contribution to change in birefringence due to change in thickness, we obtain the pure photoelastic birefringence.

2.2. Effect of hydrostatic stress

In the case of hydrostatic stress, the stress components assume the values $P = P_{xx} = P_{yy} = P_{zz}$ and $P_{xy} = P_{zx} = P_{yz} = 0$, and so the equations for the change in the B_{ij} along the principal directions take the form

$$\left.\begin{aligned} B_{11} - B_{11}^0 &= -P(q_{11} + q_{12} + q_{13}) \\ B_{22} - B_{22}^0 &= -P(q_{21} + q_{22} + q_{23}) \\ B_{33} - B_{33}^0 &= -P(q_{31} + q_{32} + q_{33}) \end{aligned}\right\} \quad (4)$$

From the above relations, the change in double refraction due to hydrostatic pressure can be obtained for different directions of observations as follows.

2.2.1. Observation along z-axis

The change in double refraction of an orthorhombic crystal under hydrostatic pressure for an observation along z-axis is given by

$$\begin{aligned} (B_{11} - B_{22}) - (B_{11}^0 - B_{22}^0) &= (B_{11} - B_{11}^0) \\ - (B_{22} - B_{22}^0) &= -P(q_{11} + q_{12} + q_{13}) \\ &\quad + P(q_{21} + q_{22} + q_{23}) \end{aligned} \quad (5)$$

But

$$B_{11} - B_{11}^0 = \frac{-2\Delta n_{11}}{(n_{11}^0)^3} = -P(q_{11} + q_{12} + q_{13}) \quad (6)$$

assuming $(n_{11}^0 + n_{11}) = 2n_{11}^0$, $(n_{11}^0)^2(n_{11})^2 = (n_{11}^0)^4$ and $(n_{11} - n_{11}^0) = \Delta n_{11}$, as shown in [1].

Putting $\Delta n_{11} = \Delta n_x$ and $n_{11}^0 = n_x$ in Equation 6 we get,

$$B_{11} - B_{11}^0 = \frac{-2\Delta n_x}{n_x^3} = -P(q_{11} + q_{12} + q_{13})$$

From the above, we find

$$\Delta n_x = \tfrac{1}{2} n_x^3 P(q_{11} + q_{12} + q_{13}) \quad (7)$$

Similarly, we obtain

$$\Delta n_y = \tfrac{1}{2} n_y^3 P(q_{21} + q_{22} + q_{23}) \quad (8)$$

From Equations 7 and 8 we find the change in double refraction under hydrostatic pressure for observations along z-axis as

$$\Delta n_x - \Delta n_y = \frac{n_x^3}{2} P(q_{11} + q_{12} + q_{13}) - \frac{n_y^3}{2} P(q_{21} + q_{22} + q_{23}) \quad (9)$$

Equation 9 can now be rewritten in a form so as to express the terms on the right hand side as differences of q_{ij}, which can be obtained from experimental measurement of relative path retardations for different directions of linear stress and directions of observation. Thus

$$\Delta n_x - \Delta n_y = -\frac{P}{2}\Big[(n_y^3 q_{22} - n_x^3 q_{12}) - (n_x^3 q_{11} - n_y^3 q_{21}) - (n_z^3 q_{33} - n_y^3 q_{23}) + (n_z^3 q_{33} - n_x^3 q_{13})\Big] \quad (10)$$

Thus we see that from the observations on relative path retardations under linear stresses alone one can obtain the effect of hydrostatic pressure on the double refraction along the principal axial directions of an orthorhombic crystal, a result which is significant.

In a similar way, expressions for change in double refraction due to hydrostatic pressure can be obtained for observations along the x-axis and y-axis, respectively, and the same are given below.

2.2.2. Observations along x-axis

$$\Delta n_y - \Delta n_z = -\frac{P}{2}\left[(n_x^3 q_{11} - n_y^3 q_{21}) + (n_z^3 q_{33} - n_y^3 q_{23}) - (n_x^3 q_{11} - n_z^3 q_{31}) - (n_y^3 q_{22} - n_z^3 q_{32}^3)\right] \quad (11)$$

2.2.3. Observations along y-axis

$$\Delta n_z - \Delta n_x = -\frac{P}{2}\left[(n_x^3 q_{11} - n_z^3 q_{31}) + (n_y^3 q_{22} - n_z^3 q_{32}) - (n_y^3 q_{22} - n_x^3 q_{12}) - (n_z^3 q_{33} - n_x^3 q_{13})\right] \quad (12)$$

3. Experimental details and results

The crystals for the present series of experiments were grown in our laboratory by the method of slow evaporation of an aqueous solution of APB. Seeding was found necessary for the growth of large sized crystals. However, every attempt of ours resulted in a crop of single crystals overlapping on one another. Nevertheless, it was possible to isolate optically good quality type portions of single crystal and to obtain differently oriented crystal specimens with dimensions ranging from 5 to 8 mm.

The stress birefringence measurements over the entire visible region have been made on each of the orientations using the birefringent compensator method of Veerabhadra Rao and Narasimhamurty [3] and the results have been checked at $\lambda = 589$ nm using a Babinet compensator. The observed relative optical path retardations are corrected for the effect due to change in the thickness due to the elastic strains in the linearly compressed crystal specimens. The observed and final values of stress birefringence for different orientations are collected in Table II.

The principal refractive indices of APB that are involved in the computation of q_{ij} have been taken from Cook and Jaffe [2] and are as follows:

$$n_x = 1.427; n_y = 1.431; n_z = 1.486.$$

The complete set of elastic constants of APB, as given below, has been experimentally determined for the first time by the authors [4]

$$\left.\begin{aligned} C_{11} &= 2.54; C_{44} = 0.21; \\ C_{12} &= 2.17 \\ C_{22} &= 3.03; C_{55} = 0.29; \\ C_{13} &= 0.31 \\ C_{33} &= 5.18; C_{66} = 1.33; \\ C_{23} &= 0.56 \end{aligned}\right\} \text{(all in units of } 10^{11}\text{ dyn cm}^{-2}\text{)}$$

$$\left.\begin{aligned} S_{11} &= 101.62; S_{44} = 475.06; \\ S_{12} &= -73.21 \\ S_{22} &= 86.44; S_{55} = 346.38; \\ S_{13} &= +1.95 \\ S_{33} &= 19.73; S_{66} = 75.06; \\ S_{23} &= -5.05 \end{aligned}\right\} \text{(all in units of } 10^{-13}\text{ cm}^2\text{ dyn}^{-1}\text{)}$$

TABLE II Stress birefringence under linear stress in comparison with the natural birefringence of ammonium pentaborate (APB) single crystal at $\lambda = 589$ nm and at room temperature

Direction of stress	Direction of observation	Natural birefringence (Unstressed crystal)	Stress birefringence under linear stress	
			Total observed stress birefringence (per unit stress per unit thickness) ($\times 10^{13}$)	Pure photoelastic birefringence (per unit stress per unit thickness) ($\times 10^{13}$)
[100]	[010]	−0.059	−18.90	−14.58
[100]	[001]	−0.004	−26.21	−26.22
[010]	[001]	+0.004	−19.78	−19.80
[010]	[100]	−0.055	−12.35	− 8.32
[001]	[100]	+0.055	+ 8.00	+ 8.11
[001]	[010]	+0.059	+20.43	+20.13
[M]	[M′]	+0.031	−26.47	−29.38
[L]	[L′]	+0.025	+ 6.05	+ 4.69
[N]	[N′]	−0.057	− 2.61	− 2.13

[M], [L], [N] and [M′], [L′], [N′] denote the same direction as described in Table I.

These values of the elastic constants have been used to evaluate the contribution to the change in birefringence due to the change in thickness of the crystal for different directions of stresses and directions of observation using expressions in Table I.

In Table II a comparison is drawn between the natural birefringence and the stress birefringence for different directions of linear stresses and directions of observation in APB single crystal at $\lambda =$ 589 nm and at room temperature.

The effect of hydrostatic pressure on the birefringence of APB crystal along its principal axial directions as deduced from Equations 10, 11 and 12 is shown in Table III; the degree of anisotropy in the photoelastic birefringence under the hydrostatic pressure can easily be seen therein.

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TABLE III Stress birefringence under hydrostatic stress in comparison with the natural birefringence of ammonium pentaborate single crystal at $\lambda = 589$ nm and at room temperature

Direction observation	Natural birefringence	Change in birefringence under hydrostatic stress (P) ($\times 10^{13}$)
[100]	$n_y - n_z = -0.055$	$\Delta n_y - \Delta n_z = -\ 4.79 \times P$
[010]	$n_z - n_x = +0.059$	$\Delta n_z - \Delta n_x = +23.34 \times P$
[001]	$n_x - n_y = -0.004$	$\Delta n_x - \Delta n_y = -18.44 \times P$