

ELASTIC CONSTANTS OF CRYSTALS

A New Method and Its Application to Pyrites and Galena

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1. Introduction

IN a recent note we reported¹ on a new method for the determination of the elastic constants of crystals. The purpose of the present paper is to explain in detail the new method and apply it to the cases of Pyrites and Galena which crystallise in the cubic system.

A review of the literature shows that there are a number of different experimental methods by which the elastic constants of crystals can be determined. The first of these is the well-known static method due to Voigt.² By bending and twisting crystal plates of different orientations, Voigt has obtained the complete set of elastic coefficients (the S's) that characterise the given crystal. The elastic constants (the C's) are then calculated from the relations that exist between the C's and the S's.

Bergmann and others³ have developed a dynamical method. The transparent crystal is usually taken in the form of a cube, cut and polished with one of its edges parallel to a crystallographic axis. This is then cemented to a quartz oscillator. The quartz is set to vibrate at one or the other of its overtones, and thus the crystal cube is thrown into resonant vibrations. This sets up a three-dimensional ultra-sonic grating in the crystal and a beam of light passing through it shows the cross-grating effects. Knowing one of the elastic constants, all the others can be calculated from these patterns. They have also developed a method which is applicable to opaque crystals. The patterns in such cases are obtained by reflection at a surface. Difficulties of interpretation have restricted the use of the latter method.

Atanasoff and Hart⁴ and Suryanarayana⁵ have developed another method which is applicable only to piezo-electric crystals. Plates of known orientation are cut from the mother crystal and are set into vibration piezo-electrically. The observed modes are identified with those required by Christoffel's theory and hence the elastic constants are calculated. This method yields results which are probably subject to various corrections as shown by Lawson.⁶

Baumgardt⁷ has developed a fourth method. Crystal plates of known orientation and having different thicknesses are placed in the path of a super-sonic beam and the acoustical energy which penetrates through the plates is measured by a torsion balance. The position of maximum transparency is thus located and used to calculate the elastic constants.

The method of composite piezo-electric oscillator developed by Balamuth⁸ has yielded very good results for single crystals of metals and other substances. To a rectangular plate of piezo-quartz is cemented a long specimen of the material of known orientation and having very nearly the same cross-section as the quartz. The composite body is suspended by fine threads. A loosely coupled oscillator is used to drive the composite block. Resonance frequencies are detected by using a thermo-micro-ammeter. Knowing the resonance frequency of the quartz alone and those of the composite block, the elastic constants of the specimen are calculated from observations on a requisite number of crystal plates. This method has also been used to determine the variation of the elastic constants with temperature.

2. Description of the Method Used in the Present Investigation

In these experiments a suitably cut and silvered quartz or tourmaline wedge is used as a piezo-electric vibrator. A series fed Hartley oscillator using a Mullard D. O. 24 tube serves as the source of excitation. As the frequency of the electrical circuit is varied, different points on the wedge are thrown into resonance and thus give a continuous ultrasonic spectrum. The range of the spectrum depends upon the angle of the wedge. Different wedges have been used in our investigations. A particular wedge of tourmaline covering a range of 2 to 10 MCs. has been found to be very useful.

The crystal plate is placed on an annular brass electrode. The wedge is laid on the crystal, using some liquid for securing good acoustical contact. The top of the wedge is touched lightly by a spring which keeps the wedge and the crystal pressed on to the brass electrode. This spring also functions as the second electrode. The mount is provided with levelling screws to facilitate accurate alignment. The arrangement is then dipped into a trough containing carbon tetrachloride. The ultrasonic beam from the wedge passes through the crystal and then enters the liquid. A beam of light with the usual optical arrangements is made to pass through the ultrasonic grating in the liquid and the Debye-Sears diffraction effects are observed. When the frequency of the wedge corresponds to the fundamental or an overtone of the thickness longitudinal vibration of the plate, the sound beam is best transmitted and the Debye-Sears pattern will have the maximum intensity.

It has been found that the setting for maximum transparency is very sharp and is capable of being reset to within 1 or 2%. The transmission frequency is then measured with a Philips Heterodyne Wavemeter.

With the observed value of the fundamental frequency f and the measured thickness d , the velocity v of propagation of longitudinal sound waves, in a direction parallel to the thickness of the plate, is calculated from the relation $v = 2df$. Using this value of v in Christoffel's equations, the effective elastic constant C'_{33} for the plate is calculated* from the relation $C'_{33} = v^2\rho$. ρ is the density of the material of the plate.

This method will not permit us to transmit thickness transverse waves and so has necessarily to be supplemented by at least one static torsion experiment if we desire to obtain the full set of elastic constants. For this purpose, the usual Searle's torsion apparatus for wires is modified for use with crystals in the present investigation. The twist is measured by using two mirrors mounted on the crystal plate, and a lamp and scale arrangement. A great degree of accuracy is obtained by measuring the shift of the light spot with a travelling microscope. Voigt has shown that when a crystal plate with its length parallel to a trigonal or higher order axis, or a digonal axis perpendicular to a second digonal axis, or perpendicular to a symmetry plane is twisted, the twist θ is given by

$$\theta = \frac{3 M l S'_{55}}{b d^3 16 (1 - 0.63 d/b)}$$

where l is the length, $2b$ is the breadth and $2d$ is the thickness of the plate. M is the twisting couple. In the case of plates of cubic crystals with lengths parallel to a cube edge $S'_{55} = S'_{44} = S_{44} = \frac{1}{C_{44}}$. To apply the above formula it is essential that $b > 3d$. In selecting plates, these facts are borne in mind.

3. Results of Check Experiments on Aluminium

In a series of check experiments, plates of commercial aluminium cut and ground have been used. For a plate of thickness 1.58 mm. maximum transmission occurred at 2.092 MCs. which gives a value of 11.80×10^{11} dynes per sq.cm.† for the elastic constant C_{11} of aluminium. It may be remarked here that transmissions at overtone frequencies of 4.183 MCs. and 6.25 MCs. have also been observed for the same plate. For the static experiments on torsion, a plate about 2 cm. long, 4 mm. broad and 1 mm.

* For an account of Christoffel's theory and the values of C'_{33} in specific cases, see a following paper by Bhagavantam and Suryanarayana published in this number.

† In this and subsequent papers the elastic constants will be given in units of 10^{11} dynes per sq.cm.

thick was used and an average value of 2.55 was obtained for C_{44} . Treating the plate as an isotropic substance, and using the relation between the Young's modulus and the constants C_{11} and C_{44} , a value of 6.95 is obtained for the Young's modulus of aluminium. This may be compared with 7.05 which is the usually accepted value.

4. Elastic Constants of Pyrites

Pyrites crystallises in the cubic system and belongs to the class T_h . Its elastic constants have already been measured by Voigt by static methods. According to Voigt this crystal has a negative C_{12} . This has been attributed by some workers⁹ to a possible existence of twinning in the plates used by him.

Flawless single cubes of Pyrites obtained from Nepal and kindly supplied to us by Sir. C. V. Raman have been used in these investigations. Sections parallel to (100) and (110) faces have been cut and ground. A (100) plate of 0.96 mm. thickness showed a transmission fundamental at 4.42 MCs. giving an effective constant $C_{33}' = C_{11} = 36.2$. A transmission on (110) of 0.98 mm. thickness showed a fundamental at 3.69 MCs. giving an effective elastic constant $C_{33}' = \frac{1}{2}(C_{11} + C_{12} + 2C_{44}) = 26.3$. In both these cases, the results represent averages of a number of readings taken. In the static torsion experiment with a (100) plate, C_{44} has been obtained as 10.4. Combining these results, we get C_{12} as -4.4 . The elastic constants thus obtained by us are compared in Table I with those of Voigt.

TABLE I

Elastic Constants of Pyrites

	Authors	Voigt
C_{11}	36.2	36.1
C_{12}	-4.4	-4.74
C_{44}	10.4	10.54

5. Elastic Constants of Galena

Galena belongs to the O_h class of the cubic system. A single cube obtained from Cherokee County, Kansas, U.S.A., has been used in these investigations. Elastic constants of Galena have not been measured so far by anyone. Hence a large number of sections have been prepared and used during the present investigations and fuller details of the results are given.

TABLE II
Measurements on Galena

Plate	Thickness in mm.	Density	Transmission fundamental frequency in MCs.	Effective Elastic Constant C_{33}'	
				Value	Expression
(100)	0.41	7.564	2.265	2.61	C_{11}
(100)	1.96	7.564	0.486	2.75	C_{11}
(100)	2.44	7.564	0.379	2.59	C_{11}
(110)	1.56	7.560	1.124	9.29	$\frac{1}{2}(C_{11} + C_{12} + 2C_{44})$
(111)	1.48	7.565	1.328	11.69	$(1/3)(C_{11} + 2C_{12} + 4C_{44})$
(210)	1.47	7.563	1.033	6.98	$(1/25)(17C_{11} + 8C_{12} + 16C_{44})$

For the static torsion experiments, a (100) plate 1.98 mm. thick and 6.23 mm. broad and about 7 mm. long was used. The length of the plate is parallel to a cube edge. The bobbin used for applying the couple had a diameter of 1.93 cm. When the effective length under torsion was 5.20 mm. the twists observed for different loads are given below.

TABLE III
Twisting of a Galena Plate

Load in gm.	20	40	60
Twist in 10^{-5} radians	3.48	6.52	10.43

Average twist for 20 gms. load is 3.41×10^{-5} radians. This gives us a value of $C_{44} = 4.39$. When the effective length under torsion was 5.25 mm. and the load was 50 gms. the twist was 8.48×10^{-5} radians giving a value of $C_{44} = 4.54$. The average of these two gives a C_{44} of 4.47.

Measurements on (100) give an average value of 2.65 for C_{11} (Table II). Combining with the observations on (110), we get a value of 6.99 for C_{12} . The three elastic constants are therefore obtained as $C_{11} = 2.65$; $C_{12} = 6.99$; $C_{44} = 4.47$.

The transmissions on (111) and (210) are used as checks. The effective constants C_{33}' calculated by assuming the above values come out as 11.50 and 6.88 for the (111) and (210) plates respectively. These compare very well with the observed figures 11.69 and 6.98.

6. Discussion of Results

So far as the results on Pyrites are concerned, we confirm the observations of Voigt. The negative value of C_{12} cannot presumably be

attributed to twinning. Another interesting feature of Pyrites is the complete breakdown of Cauchy's relations.

In the case of Galena, there are again two noteworthy features. The first is the breakdown of Cauchy's relations and the second is the surprisingly low value of C_{11} , which is perhaps connected with the easy cleavage along (100).

The observed values of the elastic constants enable us to calculate the bulk modulus $K = (\frac{1}{3})(C_{11} + 2C_{12})$ for Galena which comes out as 5.44 comparing very well with 5.11 due to Madelung and Fuchs.¹⁰

7. Summary

A new method for determining the elastic constants of crystals based on the transmission of longitudinal sound waves is given. The novelty of the method consists in the use of a continuous ultrasonic spectrum and the employment of the Debye-Sears effect as a delicate test for detecting maximum transmission. The method needs to be supplemented by at least one static torsion experiment to obtain the complete system of elastic constants. It has been applied to the cases of Pyrites and Galena. Voigt's results on Pyrites are confirmed and values for the elastic constants of Galena are given for the first time.

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