

## Non-Linear Piezo-optics\*

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Pockels's linear laws of photoelasticity which are valid for infinitesimal stresses and strains have been extended to the range of large stresses and finite strains. The various nonvanishing second-order photoelastic coefficients for all the crystal classes and the isotropic system have been evaluated. Finally the relevant equations involving the various combinations of the second-order photoelastic coefficients that occur in the conventional photoelastic measurements, have been derived for the isotropic system.

### Introduction

Recent measurements of the variation of the refractive index with hydrostatic pressure up to 7 kbar have revealed departures from the linear stress-optic law of Pockels (1889; see also Szivessy, 1929) in several materials (Vedam & Schmidt, 1965, 1966a; Vedam, Schmidt & Roy, 1966). However, the variation of the refractive index was found to be quite linear with volume strain, if the strain is computed using the non-linear theory of elasticity. But in the case of zinc blende (Vedam, Schmidt & Srinivasan, 1966; Vedam & Schmidt, 1966b), it is found that the change in refractive index is nonlinear even as a function of the volume strain. This indicates that the linear stress-optic and strain-optic relations of Pockels have to be modified for high pressures and finite strain, as shown below.

### Phenomenological equations of non-linear photoelasticity

The equation for the index ellipsoid in a general Cartesian system of coordinates is

$$a_{ij}x_i x_j = 1 \quad (i, j = 1, 2, 3). \quad (1)$$

Here the usual convention of summation over repeated indices is followed.  $a_{ij}$  are the components of dielectric impermeability tensor. Extending the Pockels theory of photoelasticity, we may write

$$\Delta a_{ij} = q_{ij,kl} t_{kl} + q_{ij,klmn} t_{kl} t_{mn} \quad (2)$$

and

$$\Delta a_{ij} = p_{ij,kl} \eta_{kl} + p_{ij,klmn} \eta_{kl} \eta_{mn}. \quad (3)$$

All the items in these equations are referred to the coordinate system in the undeformed state. Here the  $t_{kl}$  represent the components of the thermodynamic tensions (Thurston & Brugger, 1964) and  $\eta_{kl}$  the components of the Lagrangian strain tensor. These thermodynamic tensions  $t_{pq}$  are related to the applied stresses  $\tau_{ij}$  by

$$\tau_{ij} = \frac{\rho}{\rho_0} \frac{\delta x_i}{\delta a_p} \frac{\delta x_j}{\delta a_q} t_{pq} \quad (4)$$

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in the notation of Thurston & Brugger. The sign convention adopted by Nye (1960a) is used here.  $q_{ij,kl}$  and  $p_{ij,kl}$  are the usual first order piezo-optic and elasto-optic coefficients.  $q_{ij,klmn}$  and  $p_{ij,klmn}$  are the second order piezo-optic and elasto-optic coefficients. These latter coefficients are components of a sixth rank tensor. They are symmetric in the interchange  $kl \rightleftharpoons mn$  but not with respect to  $ij \rightleftharpoons kl$  or  $ij \rightleftharpoons mn$ . This is indicated by a *comma* after  $ij$ .

Using the following relation between  $t_{ij}$  and  $\eta_{kl}$  (Thurston & Brugger, 1964):

$$t_{ij} = C_{ijk} \eta_{kl} + C_{ijklmn} \eta_{kl} \eta_{mn} \quad (5)$$

we can relate  $q_{ij,klmn}$  and  $p_{ij,klmn}$  as follows:

$$p_{ij,klmn} = q_{ij,rs} C_{rsklmn} + q_{ij,rspq} C_{rskl} C_{pqmn}. \quad (6)$$

Here  $C_{ijkl}$  are the second order elastic constants and  $C_{ijklmn}$  are Brugger's (1964) third order elastic constants.

If the deformation produced by the stress  $\tau_{ij}$  does not involve any rotation, then  $\tau_{ij}$  can be written in terms of  $\eta_{ij}$ , according to Barsch (1963):

$$\tau_{ij} = C_{ijkl} \eta_{kl} + D_{ij,klmn} \eta_{kl} \eta_{mn}, \quad (7)$$

where

$$D_{ij,klmn} = \frac{1}{4} [C_{inkl} \delta_{jm} + C_{jnkl} \delta_{im} + C_{imkl} \delta_{jn} + C_{jmkl} \delta_{in} + C_{ilmn} \delta_{jk} + C_{jlmn} \delta_{ik} + C_{ikmn} \delta_{jl} + C_{jkmn} \delta_{il} - 2C_{ijkl} \delta_{mn} - 2C_{ijmn} \delta_{kl} + 2C_{ijklmn}]. \quad (8)$$

In such a case we can write the relations between  $\Delta a_{ij}$ , and  $\tau_{ij}$  analogous to equation (2), as

$$\Delta a_{ij} = q_{ij,kl} \tau_{kl} + q'_{ij,klmn} \tau_{kl} \tau_{mn} \quad (9)$$

and

$$p_{ij,klmn} = q_{ij,rs} D_{rs,klmn} + q'_{ij,rspq} C_{rskl} C_{pqmn}. \quad (10)$$

From the symmetry of the  $q_{ij,klmn}$  it can be seen that the total number of independent second order piezo-optic constants for a crystal of triclinic symmetry is 126. As the crystal symmetry increases this number decreases. Adopting the direct inspection method and the analytical methods (Nye, 1960b) wherever appropriate, all the independent non-vanishing second order piezo-optic coefficients have been derived for all the 32 crystal classes and the isotropic system. These coefficients are listed in Table 1. For the triclinic crystal

classes 1 and  $\bar{1}$ , all the constants listed in column 1 are present. The alphabetical symbols in Table 1 are abbreviations for the expressions given in Table 2.

The usual convention of replacing the index-pairs 11,22...12 by 1,2,...6 is adopted in the Tables. To avoid the excessive use of suffixes, the letter  $q$  is omitted from Tables 1 and 2; an entry 111 for example, stands for  $q_{111}$  and an entry  $\frac{1}{2}(3:116+126)$  for  $\frac{1}{2}(3q_{116}+q_{126})$ . These tables are completely valid for the components  $p'_{ij,klmn}$  and  $q'_{ij,klmn}$  as well.

The last row in Table 1 gives the total number of independent constants for each crystal class. These 'total' numbers are in agreement with those derived by group-theoretical methods by Bhagavantam & Srinanarayana (1949).

**Applications**

In the following we assume that the deformations produced by the stresses are 'pure' and that no rotations

are present. This appears to be a reasonable assumption in the case of pure tensions or compressions which are considered below. In the case of hydrostatic pressure however, the following relations are exact since there are no rotations here.

*(A) Isotropic system*

For an isotropic system there are four independent second order coefficients and they can be determined as follows:

*Case (i): uniaxial stress.* Let the stress be applied along the  $X$  axis. Then  $\tau_{11}=P$  and all the other stress components are zero. Then equation (2) becomes

$$\Delta a_1 = -2\Delta n_1/n^3 = q_{11}P + q'_{111}P^2 \tag{11}$$

and

$$\Delta a_2 = -2\Delta n_2/n^3 = q_{12}P + q'_{122}P^2 \tag{12}$$

Hence by studying the nonlinear variation of  $\Delta n$  with stress for light polarized parallel and perpendicular to

Table 1. *Second order photoelastic coefficients*

Crystal class	2	222	3	3m	6	6m2	4	4mm	23	43m	Iso-tropic
	m	2mm	3	32	6	6mm	4	42m	23	432	
const	2/m	222/mmm	3	3 2/m	6/m	622/mmm	4/m	422/mmm	23	432 3 2/m	
111	111	111	111	111	111	111	111	111	111	111	111
112	112	112	112	112	112	112	112	112	112	112	112
113	113	113	113	113	113	113	113	113	113	113	113
114	0	0	114	114	0	0	0	0	0	0	0
115	115	0	115	0	0	0	0	0	0	0	0
116	0	0	116	0	116	0	116	0	0	0	0
122	122	122	a	a	a	a	122	122	122	122	122
123	123	123	123	123	123	123	123	123	123	123	123
124	0	0	124	124	0	0	0	0	0	0	0
125	125	0	125	0	0	0	0	0	0	0	0
126	0	0	126	0	126	0	126	0	0	0	0
133	133	133	133	133	133	133	133	133	133	122	122
134	0	0	134	134	0	0	0	0	0	0	0
135	135	0	135	0	0	0	0	0	0	0	0
136	0	0	136	0	136	0	136	0	0	0	0
144	144	144	144	144	144	144	144	144	144	144	x
145	0	0	145	0	145	0	145	0	0	0	0
146	146	0	b	0	0	0	0	0	0	0	0
155	155	155	155	155	155	155	155	155	155	155	y
156	0	0	c	c	0	0	0	0	0	0	0
166	166	166	d	d	d	d	166	166	166	155	y
211	211	211	211	211	211	211	122	122	133	122	122
212	212	212	a'	a'	a'	a'	112	112	113	112	112
213	213	213	123	123	123	123	123	123	123	123	123
214	0	0	214	214	0	0	0	0	0	0	0
215	215	0	215	0	0	0	0	0	0	0	0
216	0	0	-116	0	-116	0	-126	0	0	0	0
222	222	222	222	222	222	222	111	111	111	111	111
223	223	223	113	113	113	113	113	113	112	112	112
224	0	0	e	e	0	0	0	0	0	0	0
225	225	0	e'	0	0	0	0	0	0	0	0
226	0	0	-126	0	-126	0	-116	0	0	0	0
233	233	233	133	133	133	133	133	122	122	122	122
234	0	0	-134	-134	0	0	0	0	0	0	0
235	235	0	-135	0	0	0	0	0	0	0	0
236	0	0	-136	0	-136	0	-136	0	0	0	0
244	244	244	155	155	155	155	155	155	166	155	y
245	0	0	-145	0	-145	0	-145	0	0	0	0
246	246	0	f'	0	0	0	0	0	0	0	0
255	255	255	144	144	144	144	144	144	144	144	x
256	0	0	f	0	0	0	0	0	0	0	0
266	266	266	d'	d'	d'	d'	166	166	155	155	y
311	311	311	311	311	311	311	311	311	122	122	122
312	312	312	312	312	312	312	312	312	123	123	123
313	313	313	313	313	313	313	313	313	112	112	112
314	0	0	314	314	0	0	0	0	0	0	0
315	315	0	315	0	0	0	0	0	0	0	0

Table 1 (cont.)

316	0	0	0	0	0	0	316	0	0	0	0
322	322	322	311	311	311	311	311	311	133	122	122
323	323	323	313	313	313	313	313	313	113	112	112
324	0	0	-314	-314	0	0	0	0	0	0	0
325	325	0	-315	0	0	0	0	0	0	0	0
326	0	0	0	0	0	0	-316	0	0	0	0
333	333	333	333	333	333	333	333	333	111	111	111
334	0	0	0	0	0	0	0	0	0	0	0
335	335	0	0	0	0	0	0	0	0	0	0
336	0	0	0	0	0	0	0	0	0	0	0
344	344	344	344	344	344	344	344	344	155	155	y
345	0	0	0	0	0	0	0	0	0	0	0
346	346	0	-315	0	0	0	0	0	0	0	0
355	355	355	344	344	344	344	344	344	166	155	y
356	0	0	314	314	0	0	0	0	0	0	0
366	366	366	g	g	g	g	366	366	144	144	x
411	0	0	411	411	0	0	0	0	0	0	0
412	0	0	412	412	0	0	0	0	0	0	0
413	0	0	413	413	0	0	0	0	0	0	0
414	414	414	414	414	414	414	414	414	414	414	x'
415	0	0	415	0	415	0	415	0	0	0	0
416	416	0	h'	0	0	0	0	0	0	0	0
422	0	0	i	i	0	0	0	0	0	0	0
423	0	0	-413	-413	0	0	0	0	0	0	0
424	424	424	424	424	424	424	424	424	424	424	y'
425	0	0	425	0	425	0	425	0	0	0	0
426	426	0	j	0	0	0	0	0	0	0	0
433	0	0	0	0	0	0	0	0	0	0	0
434	434	434	434	434	434	434	434	434	434	424	y'
435	0	0	435	0	435	0	435	0	0	0	0
436	436	0	436	0	0	0	0	0	0	0	0
444	0	0	444	444	0	0	0	0	0	0	0
445	445	0	-555	0	0	0	0	0	0	0	0
446	0	0	k	0	k	0	446	0	0	0	0
455	0	0	-444	-444	0	0	0	0	0	0	0
456	456	456	l	l	l	l	456	456	456	456	w
466	0	0	412	412	0	0	0	0	0	0	0
511	511	0	511	0	0	0	0	0	0	0	0
512	512	0	512	0	0	0	0	0	0	0	0
513	513	0	-436	0	0	0	0	0	0	0	0
514	0	0	-425	0	-425	0	-425	0	0	0	0
515	515	515	424	424	424	424	424	424	434	424	y'
516	0	0	h	h	0	0	0	0	0	0	0
522	522	0	i'	0	0	0	0	0	0	0	0
523	523	0	436	0	0	0	0	0	0	0	0
524	0	0	-415	0	-415	0	-415	0	0	0	0
525	525	525	414	414	414	414	414	414	414	414	x'
526	0	0	j	j	0	0	0	0	0	0	0
533	533	0	0	0	0	0	0	0	0	0	0
534	0	0	-435	0	-435	0	-435	0	0	0	0
535	535	535	434	434	434	434	434	434	424	424	y'
536	0	0	413	413	0	0	0	0	0	0	0
544	544	0	-555	0	0	0	0	0	0	0	0
545	0	0	-444	-444	0	0	0	0	0	0	0
546	546	546	l	l	l	l	456	456	456	456	w
555	555	0	555	0	0	0	0	0	0	0	0
556	0	0	-k	0	-k	0	-446	0	0	0	0
566	566	0	512	0	0	0	0	0	0	0	0
611	0	0	m	0	m	0	611	0	0	0	0
612	0	0	n	0	n	0	0	0	0	0	0
613	0	0	-136	0	-136	0	613	0	0	0	0
614	614	0	b'	0	0	0	0	0	0	0	0
615	0	0	e'	e'	0	0	0	0	0	0	0
616	616	616	q	q	q	q	616	616	424	424	y'
622	0	0	m'	0	m'	0	-611	0	0	0	0
623	0	0	136	0	136	0	-613	0	0	0	0
624	624	0	r'	0	0	0	0	0	0	0	0
625	0	0	r	r	0	0	0	0	0	0	0
626	626	626	s	s	s	s	616	616	434	424	y'
633	0	0	0	0	0	0	0	0	0	0	0
634	634	0	-135	0	0	0	0	0	0	0	0
635	0	0	134	134	0	0	0	0	0	0	0
636	636	636	t	t	t	t	636	636	414	414	x'
644	0	0	145	0	145	0	644	0	0	0	0
645	645	645	u	u	u	u	645	645	456	456	w
646	0	0	v	v	0	0	0	0	0	0	0
655	0	0	-145	0	-145	0	-644	0	0	0	0
656	656	0	v'	0	0	0	0	0	0	0	0
666	0	0	n	0	n	0	0	0	0	0	0

No. of 126  
independent  
const.

68 39 42 26 24 17 34 22 13 9 4

the direction of stress, one can evaluate  $q'_{111}$  and  $q'_{122}$  from these two equations.

*Case (ii): biaxial stress.* Let a stress  $P_1$  be applied along the  $X$  axis and a stress  $P_2$  along the  $Y$  axis. The relevant equations for the variation of refractive index are

$$\Delta a_1 = -2\Delta n_1/n^3 = q_{11}P_1 + q_{12}P_2 + q'_{111}P_1^2 + q'_{122}P_2^2 + 2q'_{112}P_1P_2 \quad (13)$$

$$\Delta a_2 = -2\Delta n_2/n^3 = q_{12}P_1 + q_{11}P_2 + q'_{122}P_1^2 + q'_{111}P_2^2 + 2q'_{112}P_1P_2 \quad (14)$$

$$\Delta a_3 = -2\Delta n_3/n^3 = q_{12}(P_1 + P_2) + q'_{122}(P_1^2 + P_2^2) + 2q'_{123}P_1P_2. \quad (15)$$

Thus by studying the nonlinear variation of refractive index with stress, for light polarized along  $Z$  and  $X$  or  $Y$  axes, the second order coefficients  $q_{123}$  and  $q_{112}$  can be determined.

*Case (iii): hydrostatic pressure.* In this case  $\tau_{11} = \tau_{22} = \tau_{33} = -P$  and all the other stress components are zero. Then the change in refractive index is given by

$$\Delta a_1 = \Delta a_2 = \Delta a_3 = -2\Delta n/n^3 = -(q_{11} + 2q_{12})P + (q'_{111} + 4q'_{112} + 2q'_{122} + 2q'_{123})P^2. \quad (16)$$

Thus measurements with hydrostatic pressure provide an additional relation among the second order coefficients which can be used to check the results of the previous two cases.

#### (B) Cubic crystals of $\bar{4}3m$ , $432$ and $4/m\bar{3}2/m$ classes

Following a similar reasoning to that used in the isotropic case we can easily work out the equations for the three cases mentioned above. Further since the directions of stress and directions of observation could be along the  $[100]$ ,  $[110]$ ,  $[111]$  and  $[\bar{2}11]$  directions we can get a more than sufficient number of equations to determine the nine second order photoelastic coefficients.

Finally a few remarks about the feasibility of such experiments will be in order. With present day techniques, hydrostatic pressure studies can be carried out to about 15 kbar without any complication and nonlinear variations of refractive index can be measured. As regards uniaxial pressure measurements extensive investigations of Stokes & Li (1963) on magnesium oxide reveal that with careful crystal preparation techniques like proper annealing, chemical polishing and careful handling, one can apply tensile stresses greater than 11 kbar without plastic deformation. It may be mentioned that this limit of 11 kbar was set by the failure of the grips rather than the onset of plastic deformation. Employing similar techniques with other crystals one could hopefully extend the upper limit of uniaxial stress measurements.

As regards the biaxial stress measurements, the authors are not aware of any previous photoelastic measurements on solids employing biaxial stresses. However, the authors do not foresee any major difficulties in carrying out such measurements if one adopts a biaxial system somewhat similar to that employed by Buessem, Cross & Goswami (1966) for dielectric measurements. Of course the usual precautions with uniaxial stress measurements mentioned above have to be taken with the biaxial stress measurements.

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Table 2. Key to alphabetical symbols used in Table 1

Symbol	Expression	Symbol	Expression
$a$	$(111 + 211 - 222)$	$a'$	$(111 + 112 - 222)$
$b$	$-\frac{1}{2}(115 + 125 + 2:215)$	$b'$	$-\frac{1}{2}(115 + 2:125 + 215)$
$c$	$\frac{1}{2}(114 + 124 + 2:214)$	$c'$	$\frac{1}{2}(114 + 2:124 + 214)$
$d$	$-\frac{1}{4}(2:111 + 2:112 - 211 - 3:222)$	$d'$	$\frac{1}{4}(2:111 - 2:112 + 211 - 222)$
$e$	$-(114 + 124 + 214)$	$e'$	$-(115 + 125 + 215)$
$f$	$\frac{1}{2}(114 - 124)$	$f'$	$-\frac{1}{2}(115 - 125)$
$g$	$\frac{1}{2}(311 - 312)$	$h'$	$-\frac{1}{2}(511 + 3:512)$
$h$	$\frac{1}{2}(411 + 3:412)$	$i'$	$-(511 + 2:512)$
$i$	$-(411 + 2:412)$	$j'$	$-\frac{1}{2}(511 - 512)$
$j$	$\frac{1}{2}(411 - 412)$		
$k$	$\frac{1}{2}(415 - 425)$	$m'$	$\frac{1}{2}(3:116 + 126)$
$l$	$\frac{1}{2}(424 - 414)$		
$m$	$-\frac{1}{2}(116 + 3:126)$	$r'$	$-\frac{1}{2}(115 - 215)$
$n$	$-\frac{1}{2}(116 - 126)$		
$q$	$-\frac{1}{4}(2:111 + 211 - 3:222)$		
$r$	$\frac{1}{2}(114 - 214)$		
$s$	$\frac{1}{4}(2:111 - 211 - 222)$		
$t$	$\frac{1}{2}(113 - 123)$		
$u$	$-\frac{1}{2}(144 - 155)$		
$v$	$\frac{1}{2}(124 + 214)$	$v'$	$\frac{1}{2}(125 + 215)$
$w$	$\frac{1}{8}(111 - 2:112 - 122 + 2:123)$		
$x$	$\frac{1}{2}(122 - 123)$	$x'$	$\frac{1}{2}(112 - 123)$
$y$	$\frac{1}{2}(111 - 2:112 + 122)$	$y'$	$\frac{1}{2}(111 - 122)$

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## Some Comments on Scattering Phenomena

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It is well known that in the calculation of wave functions very good results can be obtained for the molecular or atomic energy, namely 99% of the experimental value or better. Some other physical properties do not agree as well with experimental data; these include dissociation energies and ionization potentials. In the case of the physical properties dependent for their calculation upon electron charge densities a better agreement is obtained, perhaps because the first order correction to the Hartree-Fock charge distribution is zero. The good agreement obtained for total energies may come from compensated errors in various parts of their distributions, and cannot give sufficient information concerning the other properties. It is now reasonable to believe that very accurate calculations and experiments on scattering phenomena may be used together to study electronic distribution in various scatterers. It is the aim of this paper to show that scattered intensities and spectral studies of the modified radiation may give complementary information.

Several calculations made for light atoms and molecules are discussed and compared with experimental data when possible. Some results obtained are discussed in relation to chemical bonding for which accurate atomic and molecular electronic densities are required.

### Introduction

During recent years a large number of calculations have been done on scattering factors and intensities and on the shape of the Compton profiles. Generally made by different authors, these calculations have never been definitely compared. This comparison now seems very useful with the advent of high resolution spectrometers and more accurate wave functions. It is only recently that Kilby (1965) has shown that the Compton profile, to a first approximation, can be deduced from the Waller & Hartree (1929) theory; when the results obtained with the help of this theory are compared with experimental data, it is shown that the method is very convenient for studying the effects of scattering as well as the Compton profiles. Although these methods are first approximations to more rigorous theories, they give good results as far as incident beams of high energy are concerned.

Important contributions to this work have recently been published particularly by Bonham & Iijima (1963), Bonham (1965a, b, c, d, e); Tavard, Roux & Cornille (1962, 1963), Tavard, Roux & Rouault (1964), Tavard & Roux (1965); Tavard (1966); Kilby (1963, 1965), and Bartell & Gavin (1964). Important sets of references can be found in these articles.

### Waller and Hartree's theory

It is easy to show with the use of Waller & Hartree's theory that scattered intensities, in the case of incident X-ray of high energy, can be written as a sum of terms in the form:

$$I_{0m} = I_{cl} \left| \frac{\nu'}{\nu} \right|^3 |D_{0m}|^2 \quad (1)$$

where  $\nu$  and  $\nu'$  are the frequencies of the incident and scattered radiations, and