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Determination of the Photoelastic Constants of NaCl, NaF, KCl, and KI by Means of Brillouin Scattering

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The photoelastic constants of NaCl, NaF, KCl, and KI have been determined by studying Brillouin scattering at 90° and 180°. The absolute intensities of the Brillouin doublets were obtained by comparing the Brillouin intensity with the intensity scattered by toluene, which has a well-known photoelastic coefficient. The results are NaCl: $p_{11} = 0.115 \pm 0.009$, $p_{12} = 0.161 \pm 0.008$, $p_{44} = -0.011 \pm 0.001$; NaF: $p_{12} = 0.123 \pm 0.009$; KCl: $p_{11} = 0.233 \pm 0.013$, $p_{12} = 0.169 \pm 0.008$, $p_{44} = -0.026 \pm 0.002$; and KI: $p_{11} = 0.208 \pm 0.012$, $p_{12} = 0.166 \pm 0.008$, $p_{44} = -0.011 \pm 0.001$.

I. INTRODUCTION

It has been pointed out by Vacher and Boyer[1] and others[2-5] that it is possible to determine the photoelastic constants of transparent crystals by means of Brillouin scattering. The advantages of this method over conventional ones are as follows: Surface effects may easily be avoided, the crystal remains essentially undisturbed, and it is easy to vary its orientation in order to obtain different combinations of the photoelastic constants without introducing any change in the scattering geometry.

A disadvantage is that for some crystals the cross section for Brillouin scattering is very small, making the observation of the Brillouin doublets difficult. In this series of experiments we have determined the photoelastic constants of NaCl, NaF, KCl, and KI by using a He-Ne laser and a Fabry-Perot spectrometer. The intensity calibration was performed with toluene which has a well-known photoelastic coefficient.[6]

II. THEORY

In the paper by Vacher and Boyer[1] it is shown that an analysis of the intensities of the Brillouin components for scattering at 90° and 180° will give enough information to make possible a calculation of the photoelastic constants of cubic crystals. Their expressions for the relative intensities of these components seem to be in error in some respects and we have therefore used the theory of Benedek and Fritsch[3] to calculate the intensities of the Brillouin doublets for the crystal orientations used in this experiment. These authors give the following expression for the intensity of the scattered field:

$$\langle |\vec{\mathrm{E}}'(\vec{\mathrm{K}}, t)|^2 \rangle = E_0^2 \left(\frac{\omega_0}{c}\right)^4 \frac{\epsilon_0^4}{R^2} \frac{V}{(4\pi)^2} \frac{kT}{\rho} \times \sum_{\mu=1}^{3} \frac{|\vec{\xi}^{\mu}|^2 K^2}{\omega_\mu^2(\vec{\mathrm{K}})} , \tag{1}$$

where

$$\vec{\xi}^{\mu} = \hat{l}_k \times (\hat{l}_k \times \vec{\zeta}^{\mu}) \tag{2}$$

and

$$\vec{\zeta}^{\mu} = p_{44}[\hat{\pi}^{\mu}(\hat{l}_K \cdot \hat{l}_{E_0}) + (\hat{\pi}^{\mu} \cdot \hat{l}_{E_0})\hat{l}_K] + p_{12}(\hat{\pi}^{\mu} \cdot \hat{l}_K)\hat{l}_{E_0} + (p_{11} - p_{12} - 2p_{44}) \times \sum_{l=1}^{3} (\hat{\pi}^{\mu})_l (\hat{l}_K)_l (\hat{l}_{E_0})_l \hat{l}_l . \tag{3}$$

E_0 is the amplitude and $\vec{\mathrm{k}}_0$ is the wave vector of the incident field; E is the amplitude and $\vec{\mathrm{k}}$ is the wave vector of the scattered field. R is the distance from the scattering volume V to the point of observation. $\vec{\mathrm{K}} = \vec{\mathrm{k}}_0 - \vec{\mathrm{k}}$ and ω_μ are the wave vector and frequency of the sound wave involved in the scattering process. $\hat{\pi}^{\mu}$ is a unit vector in the direction of the polarization of the sound wave. The components of $\hat{\pi}^{\mu}$ along the cube axes are $(\hat{\pi}^{\mu})_l$, $l = 1, 2, 3$. The unit vector $\hat{l}_K$ points the direction of propagation of the sound wave and has the components $(\hat{l}_K)_l$ along the cube axes. The unit vector $\hat{l}_k$ is parallel to $\vec{\mathrm{k}}$ and has components $(\hat{l}_k)_l$, and $\hat{l}_{E_0}$ is a unit vector in the direction of polarization of the incident light wave with the components $(\hat{l}_{E_0})_l$. $\hat{l}_l$ are unit vectors along the cube axes. The direction of $\vec{\xi}^{\mu}$ gives the polarization of the scattered field. Equation (3) differs from Eq. (36) in the paper by Benedek and Fritsch[3] by the factor of 2 multiplying p_{44}.

We also have $\rho\omega^2(1/K^2) = \rho v^2$, where v is the phase velocity of the sound wave, which can be calculated together with $\hat{\pi}^{\mu}$ for phonons propagating in any direction in the crystal. This will not be done here but the results for the orientations used in this experiment will be given directly (For an account of these calculations, see, e.g., Gornall and Stoicheff.[2])

The scattered power for the orientations used in our experiments may now be calculated. Table I presents the results for 90° scattering. It should be noted that in the row labeled $\langle |\vec{\mathrm{E}}'|^2 \rangle$ we only present the quantities that are not constant for one

TABLE I. Scattering parameters and results for the 90° scattering experiments.

	$\hat{i}_K$	$(1/\sqrt{2})(-1,1,0)$	$(1/\sqrt{2})(-1,1,0)$	$(1/\sqrt{2})(-1,1,0)$	$(1,0,0)$	$(1,0,0)$	$(1,0,0)$	$(1,0,0)$
	$\hat{i}_k$	$(0,1,0)$	$(0,1,0)$	$(0,1,0)$	$(1/\sqrt{2})(1,-1,0)$	$(1/\sqrt{2})(1,-1,0)$	$(1/\sqrt{2})(1,-1,0)$	$(1/\sqrt{2})(1,-1,0)$
	$\hat{i}_{E_0}$	$(0,0,1)_\perp$	$(0,0,1)_\parallel$	$(0,1,0)_\perp$	$(0,0,1)_\perp$	$(0,0,1)_\perp$	$(1/\sqrt{2})(1,1,0)_\parallel$	$(1/\sqrt{2})(1,1,0)_\parallel$
	$\hat{\pi}^\mu$	$(1/\sqrt{2})(-1,1,0)_L$	$(0,0,1)_T$	$(0,0,1)_T$	$(1,0,0)_L$	$(0,0,1)_T$	$(1,0,0)_L$	$(0,0,1)_T$
	$\vec{\xi}^\mu$	$p_{12}(0,0,-1)$	$p_{44}(1/\sqrt{2})(-1,0,0)$	$p_{44}(1/\sqrt{2})(0,0,-1)$	$p_{12}(0,0,-1)$	$\frac{1}{2}p_{44}(-1,-10)$	$\frac{1}{2}(p_{11}-p_{12})(1/\sqrt{2})(-1,-1,0)$	$p_{44}(1/\sqrt{2})(0,0,-1)$
	ρv_c^2	$\frac{1}{2}(c_{11}+c_{12})+c_{44}$	c_{44}	c_{44}	c_{11}	c_{44}	c_{11}	c_{44}
	$\langle \lvert \vec{E}' \rvert^2 \rangle$	$2p_{12}^2/(c_{11}+c_{12}+2c_{44})$	$p_{44}^2/2c_{44}$	$p_{44}^2/2c_{44}$	p_{12}^2/c_{11}	$p_{44}^2/2c_{44}$	$(p_{11}-p_{12})^2/4c_{11}$	$p_{44}^2/2c_{44}$
	$\rho_c v_c^2$ (10^{+11} dyn/cm^2)[a]	4.32		1.26	4.87		4.87	1.26
	$10^2\, I_c/I_t$	1.05		0.0089	0.948		0.0195	0.0090
NaCl	$\lvert p_{12} \rvert$	0.160			0.161			
$n = 1.542$[b]	$\lvert p_{11}-p_{12} \rvert$						0.046	
	$\lvert p_{44} \rvert$			0.011				0.011
NaF	ρv^2 (10^{+11} dyn/cm^2)[c]	8.85			9.70			
$n = 1.325$[b]	$10^2\, I_c/I_t$	0.125			0.110			
	$\lvert p_{12} \rvert$	0.124			0.122			
	$\rho_c v_c^2$ (10^{+11} dyn/cm^2)[d]	3.01	0.63	0.63	4.06	0.63	4.06	0.63
	$10^2\, I_c/I_t$	1.36	0.077	0.076	1.01	0.080	0.036	0.082
KCl	$\lvert p_{12} \rvert$	0.169			0.169			
$n = 1.448$[b]	$\lvert p_{11}-p_{12} \rvert$						0.064	
	$\lvert p_{44} \rvert$		0.026	0.026		0.026		0.027
	$\rho_c v_c^2$ (10^{+11} dyn/cm^2)[d]	1.94	0.375	0.375	2.73		2.73	0.375
	$10^2\, I_c/I_t$	3.95	0.041	0.049	2.80		0.044	0.046
KI	$\lvert p_{12} \rvert$	0.166			0.166			
$n = 1.661$[b]	$\lvert p_{11}-p_{12} \rvert$						0.042	
	$\lvert p_{44} \rvert$		0.011	0.011				0.011

[a] Ultrasonic data from Ref. 8, supported by our Brillouin shifts.

[b] See Ref. 7.

[c] Ultransonic data from Ref. 9.

[d] Hypersonic data from Ref. 3.

TABLE II. Scattering parameters and results for 180° scattering.

	$I_{c\perp}$	$I_{c\parallel}$		$I_{c\parallel}/I_{c\perp}$	$(I_{c\parallel}/I_{c\perp})^{1/2}\cdot 2\,\lvert p_{12}\rvert$
$\hat{l}_K$	$(1/\sqrt{2})(-1,1,0)$	$(1/\sqrt{2})(-1,1,0)$	NaCl	0.864	0.298
$\hat{l}_k$	$(1/\sqrt{2})(-1,1,0)$	$(1/\sqrt{2})(-1,1,0)$	NaF	0.48	0.170
$\hat{l}_{E_0}$	$(0,0,1)$	$(1/\sqrt{2})(1,1,0)$	KCl	1.78	0.451
$\hat{\pi}^{\mu}$	$(1/\sqrt{2})(-1,1,0)_L$	$(1/\sqrt{2})(-1,1,0)_L$	KI	1.29	0.377
$\vec{\xi}^{\mu}$	$p_{12}(0,0,-1)$	$\frac{1}{2}(p_{11}+p_{12}-2\,p_{44})(1/\sqrt{2})(-1,-1,0)$			
ρV^2	$\frac{1}{2}(c_{11}+c_{12})+c_{44}$	$\frac{1}{2}(c_{11}+c_{12})+c_{44}$			
$\langle\lvert\vec{E}'\rvert^2\rangle$	$\dfrac{2p_{12}^2}{c_{11}+c_{12}+2\,c_{44}}$	$\dfrac{(p_{11}+p_{12}-2\,p_{44})^2}{2(c_{11}+c_{12})+4\,c_{44}}$			

and the same crystal. The symbols ⊥ and ∥ in the row $\hat{l}_{E_0}$ indicate whether the incident light is polarized in a direction perpendicular to or parallel to the scattering plane and the subscripts L and T in the row $\hat{\pi}^{\mu}$ indicate whether the sound wave involved in the scattering process is a longitudinal or transverse one. The results for 180° scattering are given in Table II.

Our values for $\langle\lvert\vec{E}'\rvert^2\rangle$ in Table I are in accordance with those given by Fabelinskii[10] and also with those given by Vacher and Boyer[1] except for $\hat{l}_K=(1,0,0)$, $\hat{l}_k=(1/\sqrt{2})(1,-1,0)$, $\hat{\pi}^{\mu}=(1,0,0)_L$, and $\rho v^2=c_{11}$. Here we obtain $\langle\lvert\vec{E}'\rvert^2\rangle=(p_{11}-p_{12})^2/4c_{11}$ compared with their value $(p_{11}-p_{12})^2/c_{11}$. For 180° scattering our values agree with those of Vacher and Boyer.[1]

III. EXPERIMENTAL TECHNIQUE

The light source used in these experiments was a Siemens He-Ne laser with an output of about 8

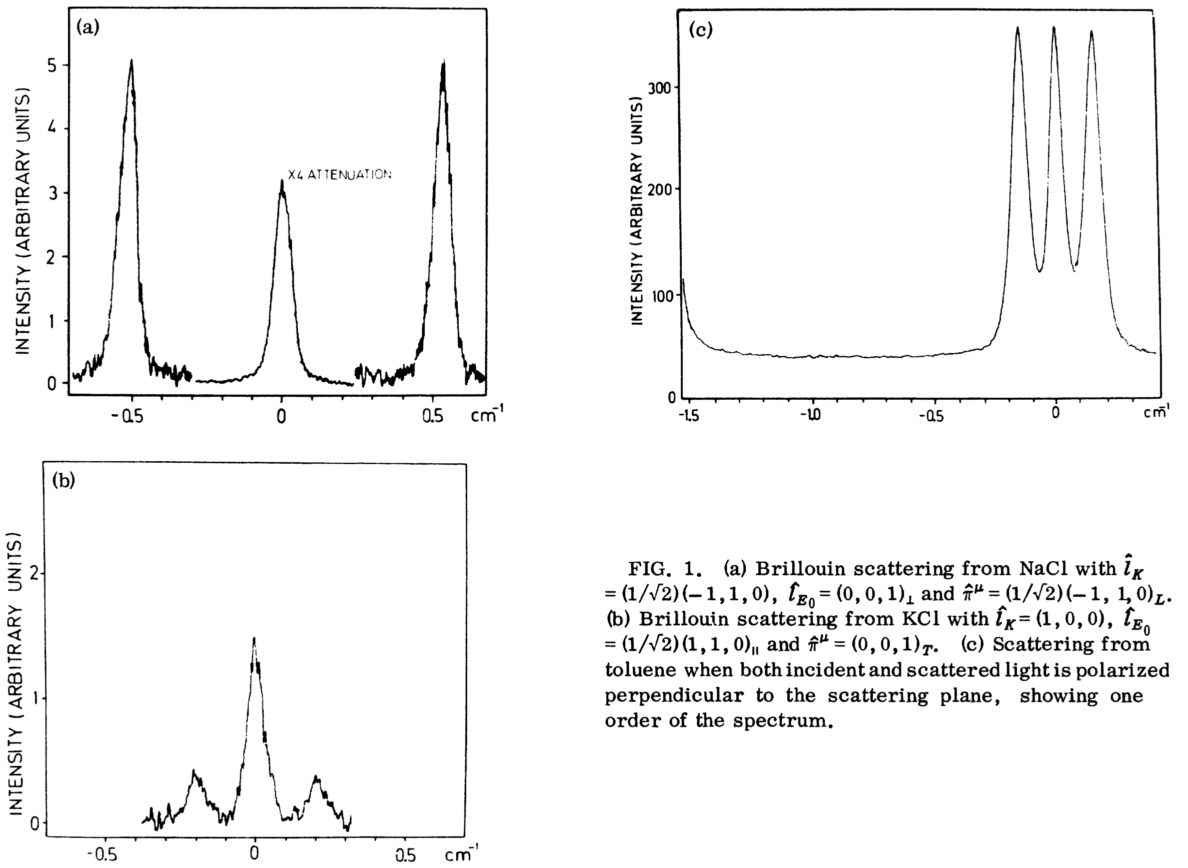

FIG. 1. (a) Brillouin scattering from NaCl with $\hat{l}_K=(1/\sqrt{2})(-1,1,0)$, $\hat{l}_{E_0}=(0,0,1)_\perp$ and $\hat{\pi}^{\mu}=(1/\sqrt{2})(-1,1,0)_L$. (b) Brillouin scattering from KCl with $\hat{l}_K=(1,0,0)$, $\hat{l}_{E_0}=(1/\sqrt{2})(1,1,0)_\parallel$ and $\hat{\pi}^{\mu}=(0,0,1)_T$. (c) Scattering from toluene when both incident and scattered light is polarized perpendicular to the scattering plane, showing one order of the spectrum.

mW when operating in the TEM_{00} mode. A $\frac{1}{2}\lambda$ plate in front of the laser was used to rotate the plane of polarization of the light beam. We used two identical scattering cells, both made by Hellma GmbH, West Germany, equipped with black-anodized aluminum inserts to reduce stray light. One was filled with toluene of spectroscopic grade for calibration purposes, while the other one contained the crystal to be investigated. The NaCl, KCl, and KI single crystals were of the best quality obtainable from the Harshaw Chemical Co. These samples were cubic in shape, 2 cm on an edge, and the faces were all chosen to be (100) faces. The NaF single crystal was obtained from Chalmers University of Technology, Sweden and was approximately cylindrical with diameter ~2 cm. This sample was oriented by x-ray techniques. The orientation of the crystals relative to the incident light beam was accurate to within ±1° during the experiments. To make it possible to change the orientation of a crystal without disturbing the light path through the sample cell, the cell was filled with a liquid having the same refractive index as the crystal.[3] The liquids used were mixtures of methanol, toluene, and di-iodomethane. The scattered light was first passed through a polarization analyzer for selection of a particular Brillouin doublet and then through a pressure-scanned Fabry-Perot interferometer, which had a free spectral range of about 1.7 cm^{-1}. The scanning was accomplished by an excess pressure of nitrogen inside the interferometer which was allowed to leak through an adjustable valve. This pressure was measured by a strain-gauge transducer and the signal was applied to the x input of an x-y chart recorder. An EMI 9558A photomultiplier equipped with magnetic defocusing by means of a permanent-magnet ring[11] served as detector. Since the light beam was chopped, the output of the photomultiplier could be analyzed by lock in techniques and presented on the y axis of the recorder. We used an integration time constant of 1–10 sec during the recording of our spectra and the time required to record one order of a spectrum was 30–120 min.

The geometrical solid angle of the detecting system was 0.008 sr for 90° scattering and 0.020 sr for 180°.

To obtain the absolute values for the photoelastic constants we compared the scattered-light intensity from the crystals with that from toluene using the same geometry (90° scattering) and the same laser-power input. The photoelastic coefficient of toluene is known to be $n^4p = 1.60$.[6] Since both the input and output polarizations have to be perpendicular to the scattering plane for toluene, while for the crystals the polarizations can be parallel and/or perpendicular, it was checked that the detection system was insensitive to the state of polarization.

TABLE III. Results obtained for the photoelastic constants. The values of $(I_{c\parallel}/I_{c\perp})^{1/2}\cdot 2\,|p_{12}|$ from Table II are also compared with $(p_{11}+p_{12}-2p_{44})$ obtained from the values listed here. The errors indicated in this case are the contributions from I_c and I_t only.

	NaCl			NaF	KCl			KI	
p_{11}	0.115 ± 0.009	0.110[a]	0.137[b]		0.233 ± 0.013	0.170[a]	0.215[c]	0.208 ± 0.012	0.210[a]
p_{12}	0.161 ± 0.008	0.153[a]	0.178[b]	0.123 ± 0.009	0.169 ± 0.008	0.124[a]	0.159[c]	0.166 ± 0.008	0.169[a]
p_{44}	−0.011 ± 0.001	−0.010[a]	−0.0108[b]		−0.026 ± 0.002	−0.024[c]	−0.031[d]	−0.011 ± 0.001	
$(I_{c\parallel}/I_{c\perp})^{1/2}\cdot 2\,\lvert p_{12}\rvert$	0.298 ± 0.009				0.451 ± 0.014			0.377 ± 0.018	
$\lvert p_{11}+p_{12}-2\,p_{44}\rvert$	0.298 ± 0.010				0.454 ± 0.010			0.396 ± 0.022	

[a] Reference 16.

[b] Reference 7, p. 441.

[c] Reference 17.

[d] Reference 18.

TABLE IV. Brillouin cross sections σ_B in cm^{-1} for incident light polarized perpendicular to the scattering plane.

	NaCl		KCl		KI	
$\sigma_B \times 10^8$ cm^{-1} $\hat{l}_K = (1, 0, 0)$	2.1 ± 0.2	$2.23 \pm 10\%$[a]	2.1 ± 0.2	$2.57 \pm 10\%$[a]	7.3 ± 0.7	$12.1 \pm 10\%$[a]
$\sigma_B \times 10^8$ cm^{-1} $\hat{l}_K = (1/\sqrt{2})(-1, 1, 0)$	2.4 ± 0.2	$2.15 \pm 10\%$[a]	2.9 ± 0.3		10.3 ± 1.0	

[a] Reference 5.

The absolute values for $\vec{\xi}^{\mu}$ may now be calculated from

$$|\vec{\xi}^{\mu}| = \left(\frac{I_c}{I_t}\,\frac{\rho_c v_c^2}{\rho_t v_t^2}\right)^{1/2} \frac{1.60}{n_c^4}\left(\frac{n_c}{n_t}\right) , \qquad (4)$$

where I_c is the intensity of a Brillouin doublet corresponding to a particular $\vec{\xi}^{\mu}$ in Table I and I_t is the Brillouin intensity of toluene. The hypersonic velocities in the crystals and toluene are denoted v_c and v_t, respectively, and ρ_c and ρ_t are the densities. $n_c = \sqrt{\epsilon}$ is the refractive index of the crystal and n_t that of toluene. The factor (n_c/n_t) corrects for the variation of the solid angle of the detector with the refractive index in the scattering cell.

The scattering in the backward direction was analyzed in the following manner because of the difficulty of maintaining the same scattering volume for toluene and the crystals in this case. When $\hat{l}_k = (1/\sqrt{2})(1, 1, 0)$ and $\hat{l}_{E_0} = (0, 0, 1)$ the Brillouin intensity is denoted $I_{c\perp}$ and this intensity gives us information about $|p_{12}|$. When $\hat{l}_{E_0} = (1/\sqrt{2})(1, -1, 0)$ we have the scattered intensity $I_{c\parallel}$ which contains information about $|p_{11} + p_{12} - 2p_{44}|$. By comparing the expressions for $\langle |\vec{E}'|^2 \rangle$ in these two cases, we obtain

$$|p_{11} + p_{12} - 2p_{44}| = (I_{c\parallel}/I_{c\perp})^{1/2}\, 2\, |p_{12}| . \qquad (5)$$

$|p_{12}|$ and $|p_{44}|$ are known from the previous measurements at 90°. The Rayleigh and Brillouin peaks of toluene were resolved graphically, and all the intensities were then obtained by measuring the area of the spectral peaks.

IV. RESULTS

Three typical recorder tracings are shown in Fig 1, and the results of our measurements are presented in Tables I and II. When evaluating the numerical values for the $|\vec{\xi}^{\mu}|$ we have, apart from the constants given in Table I, used $\rho_t v_t^2 = 1.52 \times 10^{10}$ dyn/cm^2 and $n_t = 1.493$.[12] $\rho_t v_t^2$ was obtained from $\rho_t = 0.8669$ g/cm^3 [13] and $v_t = 1326$ m/sec.[14] All the values quoted refer to room temperature, which was the temperature of our samples, $(22 \pm 0.5\,°\mathrm{C})$.

From the results given above it is possible to calculate absolute values and relative signs for p_{11}, p_{12}, and p_{44}, except for NaF for which the scattering off the transverse modes was too weak to be detected. As the Brillouin intensity always is a measure of $|\vec{\xi}^{\mu}|^2$ we have used the relation $p_{11} + 2p_{12} > 0$, based on the Clausius-Mossotti relation,[15] to obtain the absolute signs. Our results are given in Table III together with the results of earlier measurements. When estimating the errors in the p_{ij} we have used, for $\rho_c v_c^2$ and $\rho_t v_t^2$, the data given in the references. For $\rho_c v_c^2$ they lie in the range 0.4–4% and for $\rho_t v_t^2$ we have the error 2%. The value $n^4 p = 1.60$ for toluene is estimated to have the error ± 0.03. The accuracy of I_c/I_t (3–12%) was deduced from several experiments with the same crystal and from the variation of the intensity of the Stokes and anti-Stokes components. The contribution from the refractive indices n_c and n_t is negligible ($< 0.2\%$)

It is seen from Table III that the results for the photoelastic constants obtained by the standard methods used earlier agree very well with our results. The Brillouin cross sections for NaCl, KCl, and KI for scattering off longitudinal sound waves have also been measured earlier by Schoen and Cummins[5] using another method of calibration. Their results are given in Table IV, where we also present our values, which are readily calculated from our photoelastic constant p_{12}. The agreement here is obviously less satisfactory.

There has also been some theoretical work on the photoelastic constants. Aggarwal and Szigeti have calculated these constants for NaCl, KCl, and KI using Clausius-Mossotti principles,[15] but the results are not very satisfactory, especially not for p_{11}. They obtained, for example, $p_{11} = 0.61$, $p_{12} = 0.22$, and $p_{44} = -0.031$ for KCl.

This work shows that it is possible, by means of Brillouin scattering, to determine the photoelastic constants for cubic crystals with reasonable accuracy. Using a laser of higher power and shorter wavelength it should be possible to further increase the accuracy, especially for p_{11} and p_{44}.

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