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Variation of the Refractive Indices of Calcite, with Pressure to 7 kbar[1]

By

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The variation of the ordinary and extraordinary refractive indices of calcite for $\lambda = 5893$ Å with hydrostatic pressure to 7 kbar has been measured from the shift of the localized interference fringes across the sample which was kept in a fluid pressure medium. The change in the thickness of the sample under pressure was considered by utilizing the non-linear theory of elasticity. The ordinary and the extraordinary refractive indices were found to increase linearly with pressure with slopes of 0.50×10^{-3}/kbar and 0.44×10^{-3}/kbar respectively in the entire region investigated. The results are compared with the theoretical values evaluated from Bragg's theory, and are also interpreted according to Mueller's theory of photoelasticity.

Die Abhängigkeit des ordentlichen und außerordentlichen Brechungsindex von Kalkspat vom hydrostatischen Druck wurde bis 7 kbar für $\lambda = 5893$ Å durch die Verschiebung der Interferenzringe gemessen. Die Kristalle wurden durch ein flüssiges Einbettmedium beobachtet. Die durch den Druck entstandene Veränderung der Dicke der Probe wurde durch die nichtlineare Theorie der Elastizität berücksichtigt. Es wurde festgestellt, daß der ordentliche, sowie der außerordentliche Brechungsindex linear mit einem Anstieg von $0{,}50 \times 10^{-3}$/kbar, bzw. $0{,}44 \times 10^{-3}$/kbar mit dem Druck ansteigt. Die Resultate werden mit den theoretischen Werten der Braggschen Theorie verglichen und nach der Muellerschen Theorie der Photoelastizität ausgewertet.

1. Introduction

It is of interest to examine the relationship between pressure and the refractive indices of calcite in order to study the effect of varying interatomic distance upon the polarizability of the ions. The only complete investigation of this property to date is that of Pockels [1] whose measurements were confined to uniaxial stress, as were all photoelastic investigations until recently. Thus all of these measurements were limited to low stress levels and hence susceptible to possible large errors for reasons given by Vedam and Schmidt [2].

Bragg [3] has made a theoretical calculation of the indices of refraction for calcite at standard room temperature and pressure. These calculations can be made at various pressures and thus extended to give a theoretical value of $\mathrm{d}n/\mathrm{d}P$.

The present paper describes measurements made using hydrostatic pressure to about seven kilobars and reports the piezo-optic behavior of $CaCO_3$ at these pressures. These results are compared with the theoretical values evaluated from Bragg's theory and finally interpreted in the light of Mueller's theory of photoelasticity.

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2. Experimental Method

The experimental arrangement used is essentially the same as has been described earlier [4, 5] in connection with the measurement of dn/dP of α-quartz and vitreous silica. In brief, the high pressure vessel was of conventional design with an alumina window, and sovasol, an optically transparent fluid marketed by Socony Mobil Oil Company was used as the pressure medium. Natural calcite specimens of very good optical quality were cut, ground and polished such that the localized Newtonian interference fringes could be easily observed through a telemicroscope. As the crystal was subjected to hydrostatic pressure, both the thickness and the refractive indices changed, with a resulting shift of the fringe pattern across a fiducial mark on the crystal. The change in the refractive index was evaluated from the formula

$$\Delta n = \frac{p\,\lambda - 2\,n\,\Delta t}{2\,t_0}\,, \tag{1}$$

where p is the number of fringes shifted, t_0 is the initial thickness of the crystal, Δt is the change in thickness of the crystal under pressure and λ the wavelength of the light employed.

3. Results

Table 1 lists the variation of the refractive indices n_0 and n_e for Calcite at $\lambda = 5893$ Å for various pressures up to about 7 kbar, as well as the observed fringe shift at these pressures, for one of the specimens used. Since the pressure employed was quite large, the change in the thickness of the sample and the corresponding volume strain were calculated from the non-linear theory of elasticity developed by Murnaghan [6], Hearmon [7] and others [8]. The elastic constants data of Voigt [9] were used in these calculations.

Bridgman [10] has measured the linear compressibility perpendicular to the c-axis and the volume compressibility of calcite. Using his values we calculated the linear and volume strains and evaluated the corresponding changes in the thickness of the sample and refractive indices. The latter are given in Table 1. The uncertainty in Δn is ± 3 in the third figure.

4. Discussion

The values of strain computed from the elastic constants data of Voigt were found to be consistently larger than the values computed from the compressibility data of Bridgman. This difference is about 15%. This is understandable since a calculation of the volume compressibility from the elastic data gives a value about 15% higher than that reported by Bridgman. This occurs even though both the elastic constants as determined by Voigt and the compressibility measurements as obtained by Bridgman were carried out under isothermal conditions. In our following discussion we shall be concerned with the values computed from Bridgman's data because he includes a nonlinear term as well in his strain data.

Fig. 1 and 2 represent respectively the variation of the ordinary and the extraordinary refractive indices of Calcite with pressure and volume strain. They are both seen to be linear with pressure and volume strain in the region investigated. The slopes are 0.50×10^{-3}/kbar and 0.44×10^{-3}/kbar respectively.