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Optical Materials Characterization.

Albert Feldman, Deane Horowitz, Roy M. Waxler [redacted] Marilyn J. Dodge

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Center for Materials Science

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OPTICAL MATERIALS CHARACTERIZATION

Abstract

The piezo-optic constants of CaF_2, BaF_2, and SrF_2 have been measured at 0.6328 μm and 1.15 μm. The temperature dependence of the refractive indices of CdF_2, MgF_2, and NaCl have been measured at several wavelengths in the infrared by the method of Fizeau interferometry. The linear thermal expansion coefficients of NaCl and CdF_2 as a function of temperature have also been measured.

OPTICAL MATERIALS CHARACTERIZATION

1. Technical Report Summary

1.1 Technical Problem

Windows subjected to high-average-power laser radiation will undergo optical and mechanical distortion due to absorptive heating. If the distortion becomes sufficiently severe, the windows become unusable. Theoretical calculations of optical distortion in laser windows depend on the following material parameters; absorption coefficient, refractive index, change of index with temperature, thermal expansion coefficient, stress-optical constants, elastic compliances, specific heat, thermal conductivity and density. Our program has been established to measure refractive indices, changes of index with temperature, stress-optical constants, elastic compliances, and thermal expansion coefficients of candidate laser window materials.

1.2 General Methodology

Laboratory experiments are conducted for measuring refractive indices, changes of index with temperature, stress-optical constants, elastic compliances, and thermal expansion coefficients.

The refractive indices of prismatic specimens are measured on precision spectrometers by using the method of minimum deviation. Two spectrometers are used. One instrument, which uses glass optics, is used for measuring refractive indices in the visible with an accuracy of several parts in 10^6. The other instrument, which uses mirror optics, is used for measuring refractive indices in the ultraviolet and the infrared to an accuracy of several parts in 10^5. Using both spectrometers we can measure refractive indices over the spectral region 0.2 µm to 50 µm.

We measure the coefficient of linear thermal expansion, α, by a method of Fizeau interferometry. The interferometer consists of a specially prepared specimen which separates two flat plates. Interference fringes are observed due to reflections from the plate surfaces in contact with the specimen. We obtain α by measuring the shift of these interference fringes as a function of temperature. We can measure α from −180 °C to 800 °C.

The change of refractive index with temperature, dn/dT, is measured by two methods. In the first method, we measure the refractive index with the precision spectrometers at two temperatures, 20 °C and 30 °C, by varying the temperature of the laboratory. This provides us with a measure of dn/dT at room temperature. The second method may be used for measuring dn/dT from 180 °C to 800 °C. We obtain dn/dT from a knowledge of the expansion coefficient and by measuring the shift of Fizeau fringes in a heated specimen as a function of temperature. The Fizeau fringes are due to interferences between reflections from the front and back surfaces of the specimens.

We measure piezo-optic coefficients and elastic compliances using a combination of Twyman-Green and Fizeau interferometers. From the shift of fringes in specimens subjected to uniaxial or hydrostatic compression, we obtain the necessary data for determining all the stress-optical constants and elastic compliances.

In materials with small piezo-optic constants or in materials that cannot withstand large stresses, we use interferometers designed to measure fractional fringe shifts. At 10.6 μm we use a modified Twyman-Green interferometer which has a sensitivity of 0.01λ. At 632.8 nm, we use a modified Dyson interferometer which has a sensitivity of 0.002λ. When using these interferometers to measure piezo-optic constants we must know the elastic constants of the material under test.

1.3 Technical Results

The temperature dependences of the thermo-optic coefficients of CdF_2, MgF_2 and NaCl have been measured over the temperature range -180 °C to 200 °C at discrete wavelengths in the infrared by the method of Fizeau interferometry. The linear thermal expansion coefficients of CdF_2 and NaCl were also measured over the same temperature range. (Section 2.1)

The piezo-optic constants q_{11}, q_{12}, and q_{44}, of CaF_2, BaF_2 and SrF_2 have been measured at 0.6328 μm and 1.15 μm. (Section 2.2)

1.4 Department of Defense Implications

The Department of Defense is currently constructing high-power laser systems. Criteria are needed for determining the suitability of different materials for use as windows in these systems. The measurements we are performing provide data that laser system designers can use for determining the optical performance of candidate window materials.

1.5 Implications for Further Research

We plan to measure the refractive indices of SrF_2 and MgF_2 from the ultraviolet into the infrared. Measurements of the thermo-optic coefficients of LiF, NaF_2, MgF_2, CdF_2, NaCl, Al_2O_3, CaF_2, BaF_2, SrF_2, KCl, and KBr are planned for the wavelengths 458 nm and 350 nm. Piezo-optic coefficient measurements are planned for SiO_2, CaF_2 and Al_2O_3 at 350 nm. All this work cannot be done by September 30, 1978, however, we will do as much as possible under the constraints of the funding available.

2. Technical Report

2.1 Thermo-optic and Linear Thermal Expansion Coefficients

In addition to the previously reported [1] dn/dT data on CaF_2, BaF_2, KBr(RAP), KCl(RAP), LiF, NaF, SrF_2, ZnS(CVD), and ZnSe(CVD), dn/dT was measured on single crystals of CdF_2, MgF_2, and NaCl.

Figure 1 shows a plot of dn/dT as a function of temperature for CdF_2 at the two helium-neon laser wavelengths, 0.6328 µm and 3.39 µm, for the temperature range, -180 °C to 200 °C. The solid line curve represents a least squares, third order polynomial fit to the .6328 µm dn/dT data. Table 1 presents the results of this fit and a similar third order polynomial fit at 3.39 µm in a tabulated form with 20 °C temperature intervals. The errors in the table are the standard deviation of the experimental data to the least squares fit.

dn/dT as a function of temperature for NaCl is shown in Figure 2 for the three wavelengths, 0.6328 µm, 1.15 µm, and 3.39 µm. The results of a least squares third order polynomial fits for each wavelength are presented in Table 2.

MgF_2, which is an anisotropic crystal, was measured at 0.6328 µm and 3.39 µm with the electric field parallel to the c-axis to get dn_e/dT, and with the electric field perpendicular to the c-axis to get dn_o/dT. The birefringence as a function of temperature, $d(n_e-n_o)/dT$, was also measured at 0.6328 µm. The birefringence as a function of temperature was too small to be measured at 3.39 µm. The upper set of points in Figure 3 shows dn_o/dT as a function of temperature at 0.6328 µm and 3.39 µm, the middle set of points shows dn_e/dT as a function of temperature at 0.6328 µm and 3.39 µm, and the lower set of points shows the experimentally measured birefringence, $d(n_e-n_o)/dT$, at 0.6328 µm. The straight lines represent a linear least squares fit to the 0.6328 µm data in which there was the constraint that the difference in the dn_e/dT and dn_o/dT fits equals the birefringence fit, $d(n_e-n_o)/dT$. Table 3 shows the tabulated results for MgF_2.

The refractive indices, the specimen thicknesses, and references to the thermal expansion coefficient, all of which were used in the computation of the results for each of the materials, are given in Table 4.

The linear thermal expansion coefficients of NaCl and CdF_2 were also measured. Figure 4 gives the thermal expansion of NaCl in which the triangles are the experimental results and the circles are the AIP handbook values. The dashed line represents the fit to our data, which was used to calculate dn/dT for NaCl. Figure 5 gives the thermal expansion of CdF_2 as a function of temperature. Only one reference to the thermal expansion of CdF_2 was found.

2.2 Piezo-optic Constants

The piezo-optic constants have been treated amply in the literature [2] so that it is not necessary to describe them here. The rare-earth fluorides are cubic belonging to the crystal class m3m and have three piezo-optic coefficients, q_{11}, q_{12}, and q_{44}. These coefficients have been evaluated at two wavelengths, 0.6328 μm and 1.15 μm by measuring the changes in optical path length induced by compressive loading on specimens in the shape of rectangular prisms. Helium-neon laser sources were used at both wavelengths, and the optical path change was measured interferometrically by noting the shift in interference fringes. The fringes were detected by a silicon matrix vidicon camera and observed on a television monitor. Determinations were made for CaF_2, SrF_2 and BaF_2.

The specimens, which were obtained commercially, had been precision ground to the approximate dimensions, 38mm x 13mm x 13mm. The method of mounting and loading the specimens has been described earlier [3]. Two specimens of each material were fabricated. In the first specimen, the longest dimension was parallel to the <001> crystallographic direction and the light was propagated parallel to the <010> direction. In the second specimen the longest dimension was parallel to the <111> direction and the light was propagated along the <$\bar{1}$10> direction.

Two opposite long faces of each prism had been polished sufficiently flat and parallel so that about six localized, Fizeau-type interference fringes could be observed across the face when illuminated with collimated monochromatic light. At the wavelength, 0.6328 μm, q_{11} and q_{12} were determined by measuring the shift in these Fizeau fringes with load on the <001> specimens. The coefficient q_{44} was obtained from stress birefringence measurements on a <111> specimen. The optical set-up and equations relating the changes in refractive index with stress have been presented elsewhere [3-6].

At 1.15 μm, the use of a <001> prism for the determination of q_{11} and q_{12} was found to be inadequate because of the small shift in interference fringes with load; instead, the <111> prism was used. Measurements were made of the shift in interference fringes for light polarized both vertically and horizontally. q_{11} and q_{12} were then evaluated by solving simultaneously the two equations

$$\Delta n_1 = n_o^3(q_{11} + 2q_{12} + 2q_{44})\,\frac{P}{6} \qquad (1)$$

and

$$\Delta n_2 = n_o^3(q_{11} + 2q_{12} - q_{44})\,\frac{P}{6} \qquad (2)$$

where Δn_1 and Δn_2 are respectively, the refractive index changes for light polarized vertically and horizontally, n_o is the initial refractive index, and P is the applied stress. To determine q_{11} and q_{12} from the above equations, it is necessary to know q_{44}, and this value was found by measuring the stress induced birefringence in the <111> specimen.

The results of the study are presented in Table 5. The data indicate that there is little dispersion between the determinations at 0.6328 μm and 1.15 μm. For comparison, data from the literature [7] taken at 0.6328 μm are also presented. Except for q_{11} and q_{12} in CaF_2, the disagreement of our data with the data in the literature is significant. We suspect that many of the deviations observed may be due to erroneous values of the elastic constants in the literature. These constants are used in the analysis of the interferometric data in order to obtain the piezo-optic coefficients. They are also used in the conversion of elasto-optic coefficients to piezo-optic coefficients.

An example of the difficulty with the elastic compliances arose in the measurement of the piezo-optic constants of SrF_2. It was found that the coefficients q_{11} and q_{12} obtained on the <001> specimen differed from the values obtained on the <111> specimen. The discrepancy was resolved by the performance of measurements on a Twyman-Green interferometer [8] in addition to the Fizeau interferometer measurements. Both sets of measurements permitted us to calculate elastic compliance components which differed from values in the literature. These values will be presented in a future report.

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Table 1. dn/dT of CdF_2 ($10^{-5}K^{-1}$)

Temperature (°C)	Wavelength (μm) 0.6328[a]	3.39[b]
-180	- 0.56	- 0.53
-160	- 0.64	- 0.64
-140	- 0.72	- 0.73
-120	- 0.78	- 0.81
-100	- 0.84	- 0.87
- 80	- 0.89	- 0.93
- 60	- 0.93	- 0.98
- 40	- 0.97	- 1.02
- 20	- 1.01	- 1.05
0	- 1.04	- 1.08
20	- 1.07	- 1.11
40	- 1.10	- 1.14
60	- 1.13	- 1.17
80	- 1.16	- 1.20
100	- 1.19	- 1.23
120	- 1.23	- 1.27
140	- 1.27	- 1.31
160	- 1.31	- 1.36
180	- 1.37	- 1.42
200	- 1.43	- 1.49

[a]Standard deviation from a third degree polynomial fit is 0.02

[b]Standard deviation from a third degree polynomial fit is 0.04

Table 2. dn/dT of NaCl ($10^{-5}K^{-1}$)

Temperature (°C)	Wavelength (μm) .6328[a]	1.15[a]	3.39[a]
-180	-2.16	-2.22	-2.24
-160	-2.40	-2.48	-2.49
-140	-2.61	-2.70	-2.70
-120	-2.79	-2.89	-2.89
-100	-2.96	-3.06	-3.05
- 80	-3.09	-3.20	-3.19
- 60	-3.21	-3.32	-3.31
- 40	-3.32	-3.42	-3.41
- 20	-3.40	-3.51	-3.49
0	-3.48	-3.58	-3.57
20	-3.54	-3.64	-3.63
40	-3.50	-3.70	-3.68
60	-3.65	-3.74	-3.73
80	-3.69	-3.79	-3.78
100	-3.74	-3.83	-3.83
120	-3.78	-3.88	-3.88
140	-3.83	-3.93	-3.94
160	-3.88	-3.99	-4.01
180	-3.94	-4.06	-4.09
200	-4.01	-4.14	-4.18

[a]Standard deviation from a third degree polynomial fit is 0.04

Table 3. dn/dT of MgF_2 ($10^{-6}K^{-1}$)

Temperature (°C)	Wavelength (μm) 0.6328[a] dn_e/dT	0.6328[a] dn_o/dT	3.39[b] dn_e/dT	3.39[b] dn_o/dT
-180	1.65	2.23	1.5	2.0
-160	1.54	2.12	1.4	2.0
-140	1.44	2.01	1.3	1.9
-120	1.33	1.90	1.2	1.8
-100	1.22	1.79	1.2	1.7
- 80	1.12	1.68	1.1	1.6
- 60	1.01	1.57	1.0	1.5
- 40	0.90	1.46	0.9	1.4
- 20	0.80	1.35	0.8	1.3
0	0.69	1.24	0.7	1.2
20	0.58	1.12	0.6	1.1
40	0.48	1.01	0.5	1.0
60	0.37	0.90	0.4	1.0
80	0.27	0.79	0.3	0.9
100	0.16	0.68	0.2	0.8
120	0.05	0.57	0.1	0.7
140	-0.05	0.46	0	0.6
160	-0.16	0.35	-0.1	0.5
180	-0.27	0.24	-0.2	0.4
200	-0.37	0.13	-0.3	0.3

[a]Standard deviation to be determined

[b]Standard deviation from a linear fit is 0.2

Table 4. Data used in Computation of dn/dT

Material	Refractive index, n_o			t_o (mm)	α
	632.8 nm	1.15 μm	3.39 μm		
CdF_2	1.5735[a]		1.54[b]	7.33	c
MgF_2 n_e	1.3887[d]	1.384[e]	1.369[e]	13.40	f
MgF_2 n_o	1.3770[d]	1.373[e]	1.358[e]	13.40	f
NaCl	1.542[g]	1.5305[g]	1.5235[g]	14.08	h

[a]B. Krukoska-Fulde, T. Niemyski, J. Crystal Growth 1, 183-6 (1967).

[b]Estimated.

[c]See Figure 5.

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Table 5. Piezo-optic Constants of Three Alkaline Earth Fluorides

	λ = 0.6328 μm		λ = 1.15 μm
	NBS[a]	Literature[b]	NBS[a]
CaF_2			
q_{11}	-0.38±0.03	-0.41	-0.40±0.06
q_{12}	1.08±0.03	1.04	1.09±0.06
$(q_{11}-q_{12})$	-1.46±0.01	-1.45	-1.49±0.02
q_{44}	0.71±0.01	0.84	0.72±0.01
SrF_2			
q_{11}	-0.64±0.04	-0.58	-0.63±0.05
q_{12}	1.45±0.04	1.77	1.50±0.06
$(q_{11}-q_{12})$	-2.08±0.01	-2.35	-2.13±0.04
q_{44}	0.60±0.01	0.59	0.62±0.02
BaF_2			
q_{11}	-0.99±0.03	-0.62	-0.91±0.07
q_{12}	2.07±0.04	2.31	2.13±0.07
$(q_{11}-q_{12})$	-3.06±0.01	-2.93	-3.03±0.02
q_{44}	0.95±0.01	1.06	0.95±0.01

[a]The errors were calculated from the standard deviations of the experimental data.

[b]Reference (6), the data for SrF_2 were calculated from the values of p_{ij} and s_{ij} given in reference (6).

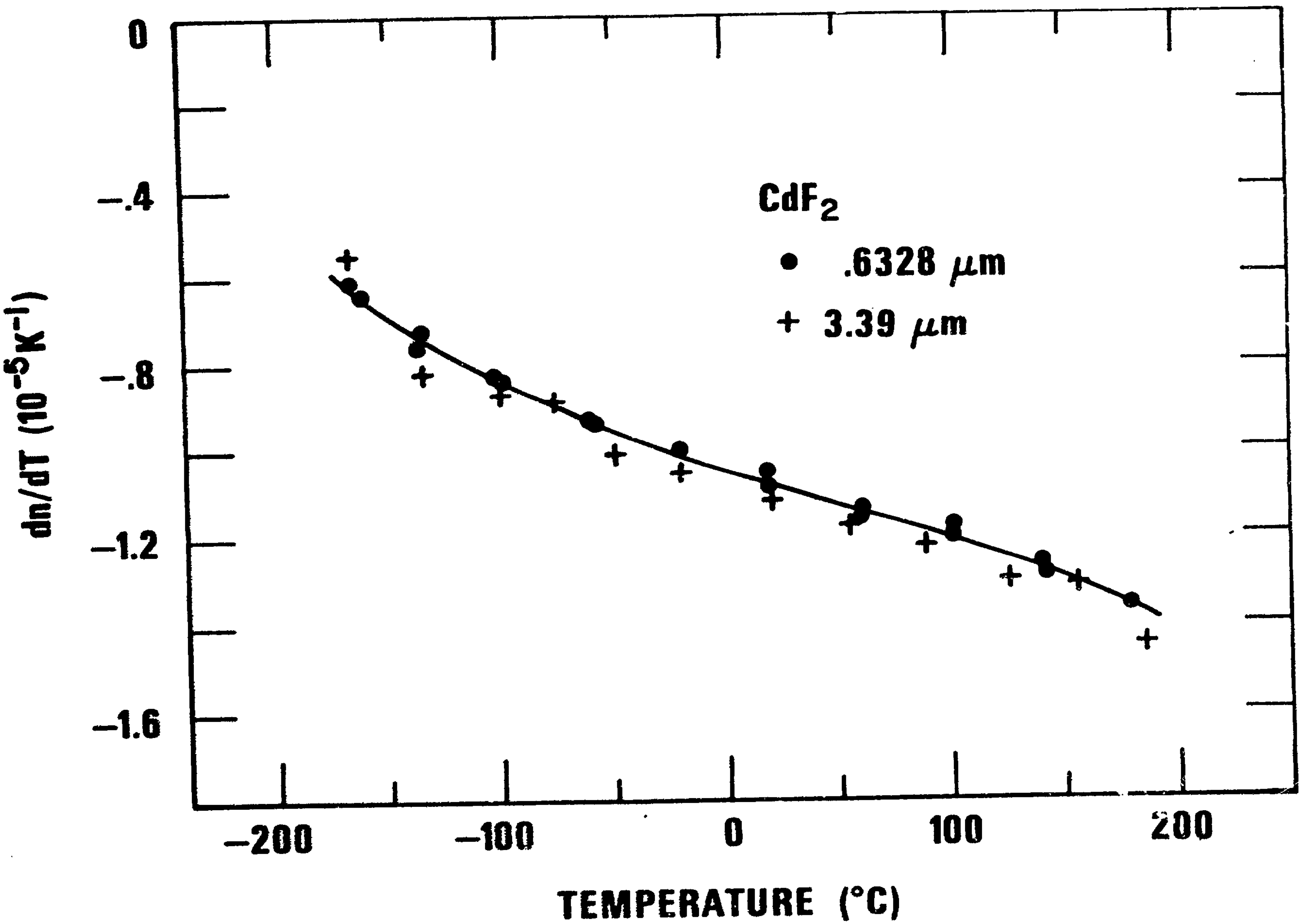

Fig. 1 dn/dT of CdF_2 as a function of temperature.

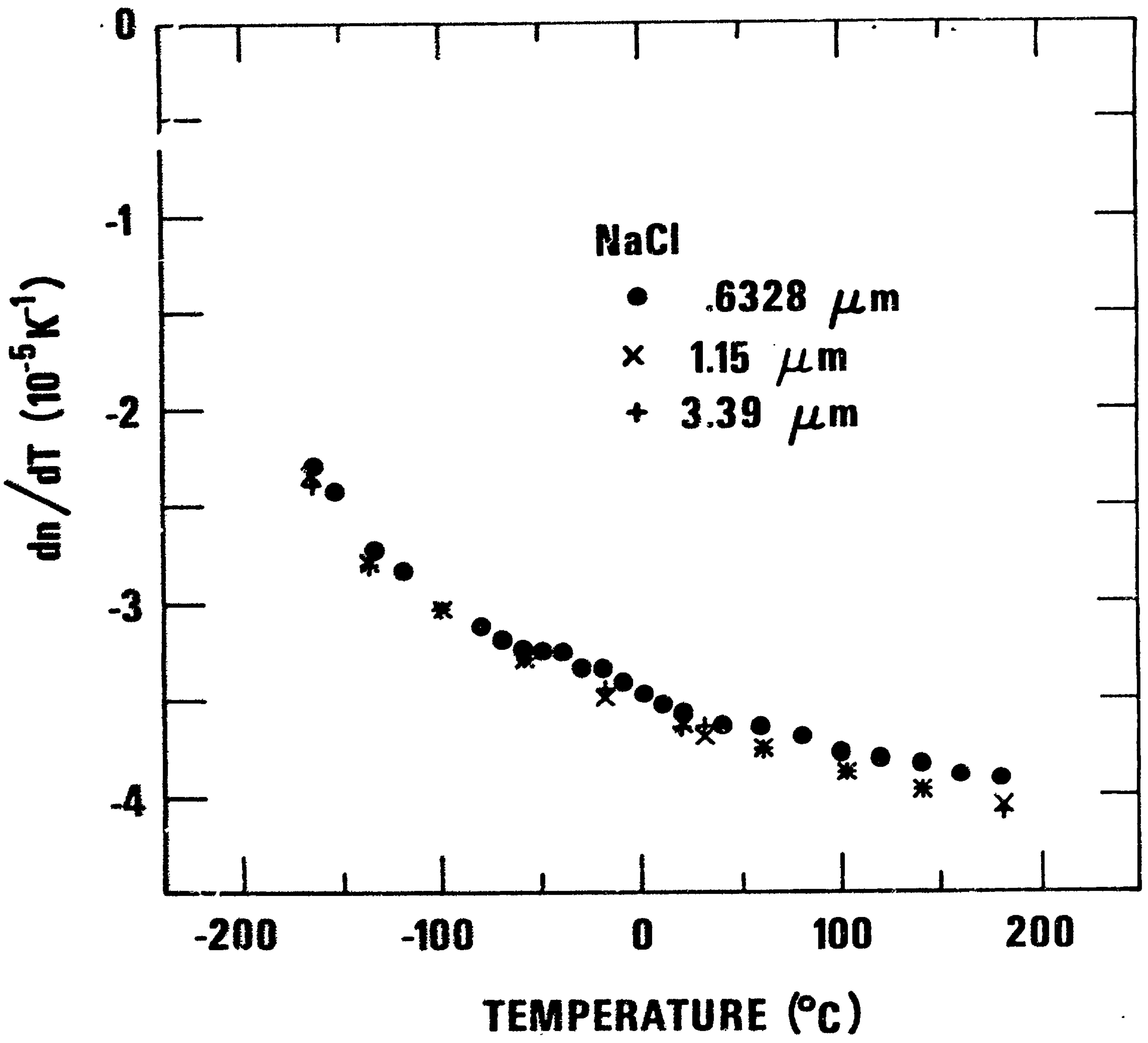

Fig. 2 dn/dT of NaCl as a function of temperature.

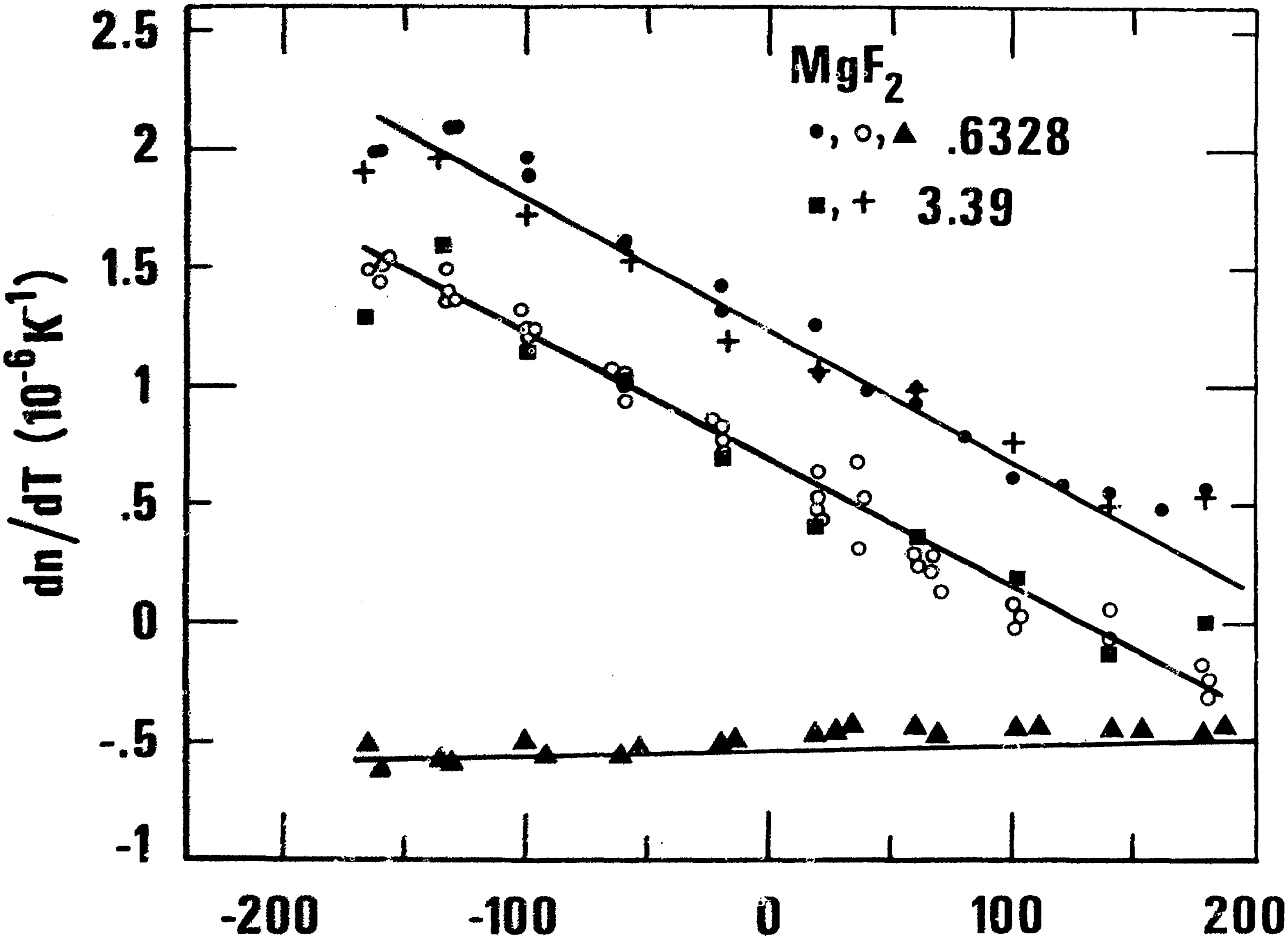

Fig. 3 dn/dT of MgF_2 as a function of temperature. The upper curve is for dn_o/dT, the middle curve is for dn_e/dT, and the bottom curve is the birefringence, $d(n_e-n_o)/dT$.

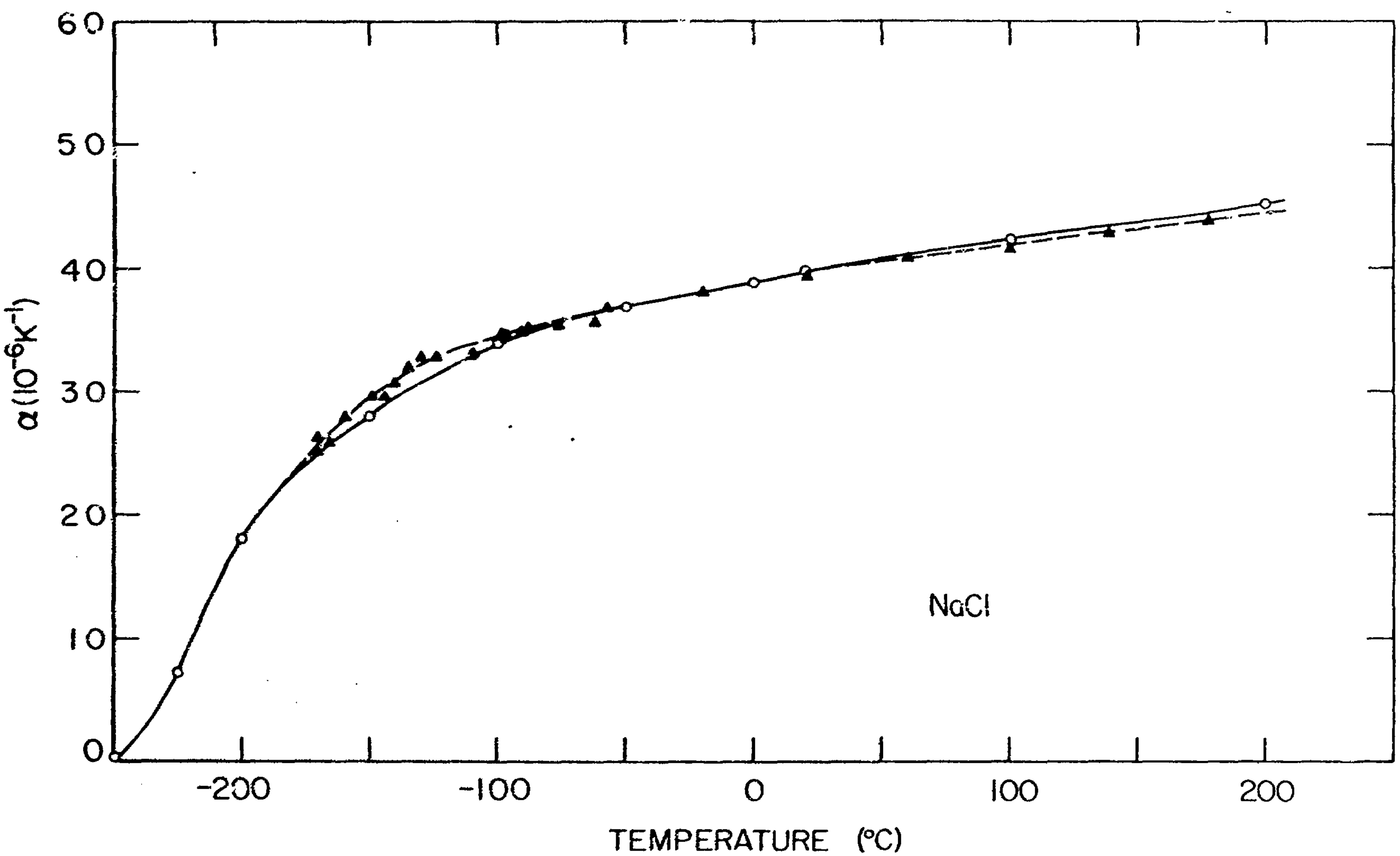

Fig. 4 Thermal expansion coefficient for NaCl as a function of temperature. The triangles are our experimental points and the circles are obtained from the AIP Handbook.

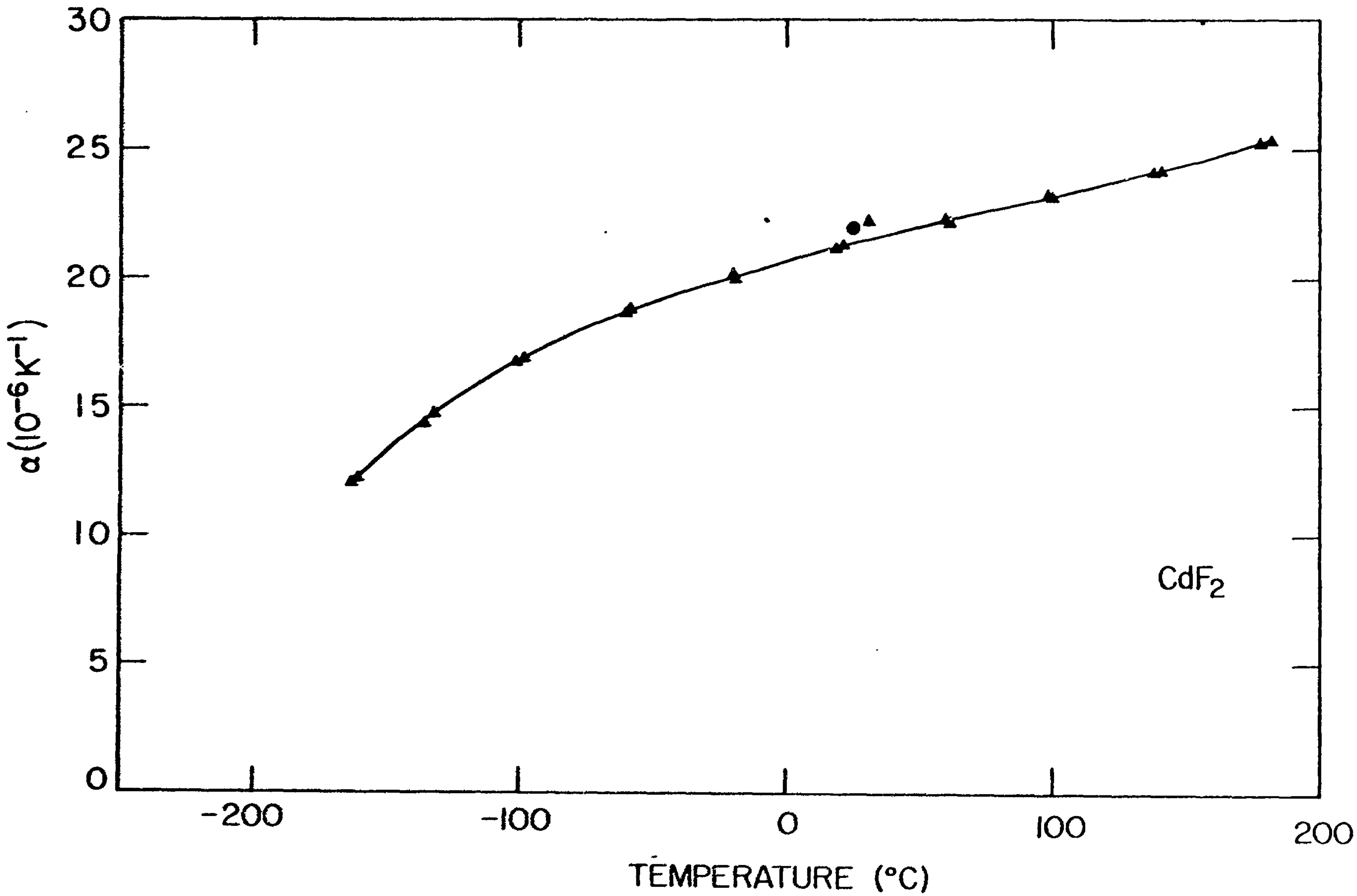

Fig. 5 The thermal expansion coefficient for CdF_2 as a function temperature. The triangles show our experimental data and the single circle is from S. S. Ballard and J. S. Browder, Appl. Optics 5, 1873 (1966).

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16. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)*

The piezo-optic constants of CaF_2, BaF_2, and SrF_2 have been measured at 0.6328 μm and 1.15 μm. The temperature dependence of the refractive indices of CdF_2, MgF_2, and NaCl have been measured at several wavelengths in the infrared by the method of Fizeau interferometry. The linear thermal expansion coefficients of NaCl and CdF_2 as a function of temperature are also presented.

17. KEY WORDS *(six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)*

Thermal coefficient of refractive index; thermo-optic constant; linear thermal expansion; piezo-optic constants; photo-elastic constants, CaF_2; BaF_2; SrF_2; CdF_2; MgF_2; NaCl.

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