

experiment.

- [1] A. A. Tyapkin, Vestn. AN SSSR, No. 12, 1964.
- [2] S. Fukui and S. Miyamoto, Nuovo cimento 11, 113 (1959).
- [3] G. A. Vorob'ev and N. S. Rudenko, PTE, No. 1, 109 (1965).

DEPENDENCE OF THE REFRACTIVE INDEX ON THE DENSITY OF THE SOLID AND LIQUID PHASES OF SHOCK-COMPRESSED IONIC CRYSTALS. RELAXATION TIME OF PHASE TRANSFORMATION UNDER SHOCK COMPRESSION.

S. B. Kormer, K. B. Yushko, and G. V. Kirshkevich  
 Submitted 1 November 1965  
 ZhETF Pis'ma 3, No. 2, 64-69, 15 January 1966

The methods described in [1] were used to investigate the refractive indices of shock-compressed alkali-halide compounds. For LiF, which remains transparent in the investigated range of pressures up to  $P \approx 700$  kbar, the refractive index was determined directly from the paths of the rays in the compressed matter. For NaCl, CsBr, KCl, and KBr crystals, which become opaque behind the shock-wave front <sup>1)</sup> the refractive indices were determined by Fresnel's formulas from the experimentally-measured coefficients of reflection of natural light incident on the front of the shock wave.

1. It turned out that the dependence of the refractive index on the degree of compression  $\sigma$  (where  $\sigma = \rho/\rho_0$  is the running density and  $\rho_0$  the density at  $T = 300^\circ\text{K}$  and  $P \approx 0$ ) for the crystals LiF, NaCl, and CsBr, which do not experience polymorphic transformations in the investigated range of pressures, can be represented schematically as shown in Fig. 1 for the region  $\sigma > 1$ . So long as the shock compressed crystal remains in the solid phase <sup>2)</sup>, the refractive index will change relatively little with the density. Comparison of the values of

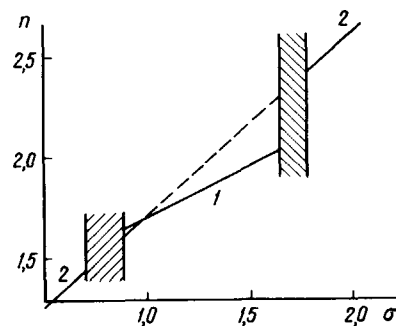


Fig. 1. Refractive index of ionic crystals vs. density in the solid and liquid phases

$$\frac{dn}{d\sigma} = \frac{n - n_0}{\sigma - 1} \quad (1)$$

obtained for this range of compressions ( $n_0$  = refractive index at  $P \approx 0$  and  $T = 300^\circ\text{K}$ ), with the values of  $(dn/d\sigma)_{\sigma=1}$  obtained [4-6] in investigations of the photoelastic properties of ionic crystals has shown that the function  $n(\sigma)$  is linear over a rather broad range (see fourth and fifth lines of the table) <sup>3)</sup>.

It turned out further that the refractive index increases appreciably when the melting is in a compressed state (see Fig. 1). The experimental points obtained for the liquid phase of the crystals CsBr, KCl, and KBr fit the relation (1) quite well, but only if  $dn/d\sigma$  is approximately 15 - 17 times larger than in the solid phase (see the table). We note that at normal pressure the change in density of the alkali halides in the liquid state changes the

Table

Crystal			LiF	KCl	NaCl	KBr	KI	CsBr
$n_0$			1.392	1.490	1.544	1.559	1.667	1.698
Solid phase	Present work	$\sigma^*$	1.30 - 1.48	1.68	1.45 - 1.68	1.70	-	1.63
		$dn/d\sigma$ , Eq. (1)	0.1	0.25***	0.24	0.35*	-	0.54
		$(dn/d\sigma)_{\sigma=1}$ , Ref. [4-6]	0.1	0.23	0.24	0.35	0.43	(0.48)**
		$dn/d\sigma$ , Eq. (2)	-	0.32	0.32	0.35	-	-
		$(dn/dT)_\rho \times 10^5$ , Ref. [7]	-0.26	-1.04	-0.78	0.0	-	-
Liquid phase	Present work	$\sigma^*$	-	1.83 - 1.98	-	1.88	-	1.8 - -2.07
		$dn/d\sigma$ , Eq. (1)	-	0.35	-	0.53	-	0.9
		$dn/d\sigma$ , Eq. (2)	-	0.446	0.520	0.517	-	(0.72)**

\* Investigated range of degrees of compression

\*\* Values obtained by extrapolating the data of [5] for the solid phase and analogous extrapolation of data obtained from [8,9] for the liquid phase.

\*\*\* Value for the second phase

refractive index by about 1.5 times more than in the solid state (compare the sixth and tenth lines of the table). The values of  $(dn/d\sigma)_{\sigma=1}$  for the liquid phase of ionic crystals at normal pressure, listed in the table, were determined from the relation <sup>4)</sup>

$$dn/d\sigma = \rho_0 (\partial n / \partial T)_P / (\partial \rho / \partial T)_P, \quad (2)$$

where  $(\partial n / \partial T)_P$  was taken from [8] and  $(\partial \rho / \partial T)_P$  from [9].

From the results in the table it follows that for each of the phases (solid and liquid separately) we can assume, albeit approximately, identical relations of the type (1) for  $n(\sigma)$  <sup>5)</sup>, with  $(dn/d\sigma)_{liq} > (dn/d\sigma)_{sol}$ , in a wide range of densities. These relations have a common point at  $\sigma \approx 1$  (see Fig. 1). This explains the apparent contradiction between the fact that melting decreases the refractive index at normal pressure but increases it in the compressed state (see Fig. 1).

The relatively slow variation of  $n(\sigma)$  in the solid phase is apparently due to the increase in the repulsion forces with decrease in the interionic distances, and to the decreased deformability of the ion shells under the influence of the incident light. In other words, compression reduces the electronic polarizability. Similar ideas were advanced by Burstein

and Smith [10] to explain the relatively small values of  $(dn/d\sigma)_{\sigma=1}$  obtained in investigations of the photoelastic properties of ionic crystals. Following these qualitative ideas, we can relate the difference between  $dn/d\sigma$  in the solid and liquid phases to the change in the character of the bond upon melting. This question, however, needs a detailed theoretical analysis.

2. In investigating the reflectivity of the shock-wave front in the solid phase of KCl and KBr it was noted that at  $P \approx 140$  kbar the reflection coefficient is 2 - 3 times smaller than that corresponding to a relation of the type (1) for  $n(\sigma)$ , with values of  $dn/d\sigma$  that follow from [4-6], whereas at  $P \approx 200$  kbar for KCl and 260 kbar for KBr the obtained results are close to those expected.

We recall (see Sec. 1 and the table) that for other crystals the results of our measurements are in satisfactory agreement with the data of the cited papers. It is natural to relate the indicated difference with the polymorphic transformation of KCl and KBr into the CsCl structure, which occurs at  $P \approx 20$  kbar [11,12]. If we assume that up to  $P \leq 140$  kbar the phase transformation of KCl and KBr occurs after a time  $\tau > 10^{-11}$  sec, the light will be reflected from a layer of matter situated on the front of the shock wave in a metastable state (point 1, Fig. 2), corresponding to the dynamic adiabat of the first phase<sup>6)</sup>. Since the latter is steeper than the adiabat of the second phase, a smaller density jump on the shock-wave front corresponds also to a smaller refractive index. The non-equilibrium states of the first phase of KCl and KBr (point A, Fig. 2), determined from the shock-wave velocity, from the dependence (1) with  $dn/d\sigma$  as given in the table, and from the measured reflection coefficient, are shown in Fig. 2. For KCl the point obtained lies somewhat to the left of the first-phase adiabat calculated from the equation of state in [3] (see Fig. 2).

With increasing pressure, the temperature increases (for KCl,  $T = 1300^\circ\text{K}$  at  $P = 136$  kbar and  $T = 2100^\circ\text{K}$  at  $P = 200$  kbar [3]), the relaxation time decreases, and the phase transformation takes place in a layer thinner than  $\lambda/2\pi$  ( $\lambda$  = wavelength of the incident light). In this case the refractive index will correspond to the total jump in volume behind the front of the shock wave (point 2', Fig. 2). Taking account of the fact that the values of  $dn/d\sigma$  obtained for phase II turned out to be close to the values in [4-6] for phase I (see the table), the measured reflection coefficients turned out to be close to those expected. It follows from the foregoing that upon shock compression with  $P = 200 - 260$  kbar, the polymorphic transition in KCl and KBr takes place within a time  $\tau < 10^{-11}$  sec. (It is interesting that the polymorphic transition has no effect on the  $n(\sigma)$  dependence of these ionic crystals.) The same time is

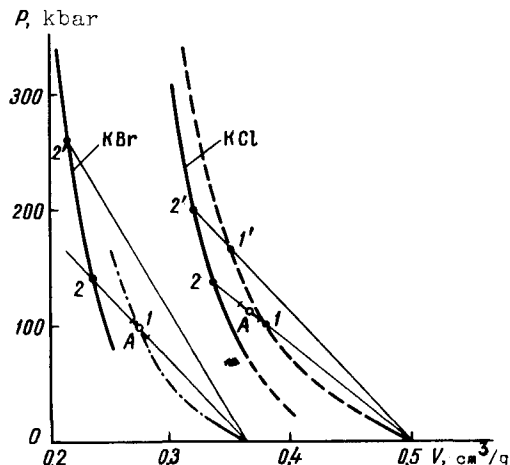


Fig. 2. Equilibrium and non-equilibrium dynamic adiabats of KCl and KBr. ——— calculated after [12,3], ----- calculated after [3], -.-.- interpolation, -o-o-o present experiments.

characteristic also of melting in the shock-wave front, since the refractive index (reflection coefficient) experiences a jump at pressures corresponding to the transition of the solid phase into liquid (see Fig. 1).

- [1] Ya. B. Zel'dovich, S. B. Kormer, M. V. Sinitsyn, and K. B. Yushko, DAN SSSR 138, 1333 (1961), Soviet Phys. Doklady 6, 494 (1961).
- [2] S. B. Kormer, M. V. Sinitsyn, G. A. Kirillov, and L. T. Popova, JETP 49, 135 (1965), Soviet Phys. JETP 22, 97 (1966).
- [3] S. B. Kormer, M. V. Sinitsyn, G. A. Kirillov, and V. D. Urlin, JETP 48, 1033 (1965), Soviet Phys. JETP 21, 689 (1965).
- [4] H. Mueller, Phys. Rev. 47, 947 (1935).
- [5] E. Burstein and P. L. Smith, *ibid.* 74, 229 (1948).
- [6] P. S. Krishnan, Progress in Crystal Physics, Interscience Publishers, N.Y., 1958.
- [7] A. J. Bosman and E. E. Havinga, Phys. Rev. 129, 1593 (1963).
- [8] Y. Laszycki and F. Naudin, Comptes rendus 256, 1282 (1963).
- [9] International Critical Tables 3, 24.
- [10] E. Burstein and P. L. Smith, Proc. Ind. Acad. Sci. A28, 377 (1948).
- [11] P. Bridgman, Phys. Rev. 57, 237 (1940).
- [12] L. V. Al'tshuler, M. N. Pavlovskii, L. V. Kuleshova, and G. V. Simakov, FTT 5, 279 (1963), Soviet Phys. Solid State 5, 203 (1963).
- [13] S. B. Kormer, M. V. Sinitsyn, A. I. Funtikov, V. D. Urlin, and A. V. Blinov, JETP 47, 1202 (1964), Soviet Phys. JETP 20, 811 (1965).

1) See [2] concerning the nature of opaqueness of shock-compressed crystals.

2) The melting limits of shock-compressed NaCl and KCl were determined experimentally in [3]. The authors of the same reference obtained also melting regions of LiF, CsBr, and KBr.

3) It must be borne in mind here that the error in our measurements is quite large and amounts to  $\Delta n/n = \pm 1\%$ .

4) In formula (2) we neglect the term  $(\partial n/\partial T)_{\rho_0}$ . If we assume the same values as for the solid phase [7] (see the table) for the liquid phase, for which  $(\partial n/\partial T)_{\rho}$  is not known, then the change in  $dn/d\sigma$  will not exceed 10%.

5) Allowance for the temperature dependence of the refractive index does not lead to appreciable errors, at least for the solid phase. Thus, for KCl, for which  $(\partial n/\partial T)_{\rho}$  has the largest value (see the table), the error in  $dn/d\sigma$  due to neglecting the temperature correction does not exceed  $\approx 2\%$  for  $\sigma = 1.68$  and  $T = 3800^\circ\text{K}$ .

6) The development of the phenomenon in a relaxation phase transformation of ionic crystals on the front of a shock wave was considered in [13].