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# Theory of strain derivatives of electronic and static dielectric constants of alkali halides and alkaline-earth fluorides

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An analysis of the first-, second-, and third-order strain derivatives of electronic and static dielectric constants of alkali halides and alkaline-earth fluorides has been performed within the framework of the Clausius-Mossotti theory. The strain derivatives of the electronic dielectric constant are evaluated using Ruffa's theory of electronic polarizabilities. The lattice contribution to the strain derivatives of the static dielectric constant has been calculated using two potential forms for the short-range repulsive energy showing the inverse power dependence and the exponential dependence upon the interionic separation. The contribution arising from the Van der Waals dipole-dipole and dipole-quadrupole interactions has also been considered. The calculated values of the strain derivatives of the dielectric constants are compared with the available experimental data. It is found that the inverse power law for the short-range repulsive potential is more consistent with experimental facts. This prediction is consistent with recent investigations on the elastic properties of ionic solids.

## I. INTRODUCTION

The Born theory of interionic forces[1] has been widely used to study the cohesive properties[2,3] surface energy,[4] defect energy,[5] and various other physical and chemical properties of ionic crystals.[6] In recent years the theory has been extended with considerable success to analyze the elastic constants and their pressure derivatives for alkali halides.[7-9] Recently Fontanella *et al.*[10] have measured the first-, second-, and third-order pressure derivatives of the static dielectric constant $\epsilon_s$ of alkali halides. In a subsequent paper Andeen *et al.*[11] have reported the first-, and second-order pressure derivatives of $\epsilon_s$ for $CaF_2$, $SrF_2$, and $BaF_2$ crystals. It seems therefore, desirable to make a comprehensive investigation of the applicability of the Born model to predict the variation of $\epsilon_s$ for ionic crystals under the effect of pressure.

There are two types of polarizations, electronic and ionic, contributing to the $\epsilon_s$ of ionic crystals. The strain derivatives of the electronic polarizabilities can be evaluated in an approximate manner following the method of Sharma *et al.*[12,13] based on Ruffa's theory of electronic polarizabilities.[14] Since the contribution of the strain derivatives of the electronic polarizabilities to the strain derivatives of $\epsilon_s$ for ionic solids is small,[15,16] the uncertainty in the evaluation of the former will not affect significantly the results for the latter. In the present paper we extend the method of Sharma *et al.*[12,13] to evaluate the third-order strain derivatives of the electronic dielectric constant $\epsilon_\infty$ for alkali halides and the second-order strain derivatives of $\epsilon_\infty$ for alkaline-earth fluorides.

The lattice contribution which arises from the relative displacement of ions can be evaluated from the Born model of interionic forces. Following the lines initially suggested by Bosman and Havinga,[15] Sharma *et al.*[13] have performed an analysis of the first- and second-order strain derivatives of $\epsilon_s$ and of the static polarizability $\alpha_s$ of alkali halides on the basis of the Born model. Sharma *et al.*[13] did not consider the contribution arising from the Van der Waals dipole-dipole and dipole-quadrupole interactions. However, recent studies[17-19] have revealed that these interactions contribute significantly to the short-range potential in ionic crystals. Since the higher-order strain derivatives of $\epsilon_s$ are very much sensitive to the short-range potential, the consideration of Van der Waals interactions is expected to lead to significant changes in the resulting values of the strain derivatives of $\epsilon_s$. We therefore evaluate the first- and higher-order strain derivatives of $\epsilon_s$ for NaCl-type alkali halides, taking into account the Van der Waals interactions. The first- and second-order derivatives of $\epsilon_s$ are also calculated for alkaline-earth fluorides. The calculations are preformed by adopting the inverse-power form as well as the exponential form for the short-range repulsive potential.

## II. THEORY AND METHOD OF CALCULATION

The relevant expressions for the strain derivatives of $\epsilon_\infty$ can be obtained from the Lorentz-Lorenz relation

$$(\epsilon_\infty - 1)/(\epsilon_\infty + 2) = 4\pi\alpha_\infty/3V , \tag{1}$$

where $\alpha_\infty$ and $V$ are the electronic polarizability and volume per ion pair, respectively. In view of the additivity rule for electronic polarizabilities, we have[20]

$$\alpha_\infty = \alpha_+ + p\alpha_- , \qquad (2)$$

where $\alpha_+$ and $\alpha_-$ are the electronic polarizabilities of the cation and anion respectively. $p=1$, 2 for NaCl- and $CaF_2$-type crystals, respectively. The strain derivatives of $\alpha_+$ and $\alpha_-$ can be obtained by the method of Sharma *et al.*[12] which is based on the theory of crystalline polarizabilities proposed by Ruffa[14] and which makes use of a relationship between electronic polarizability and ionic radius. The final expressions for the derivatives of $\alpha_+$ and $\alpha_-$ obtained by the method of Sharma *et al.*[12] are

$$\frac{d\alpha_+}{dr} = -\frac{2\alpha_f E_f^2 (eV_m)}{(E_f - eV_m)^3 r} , \qquad (3)$$

$$\frac{d\alpha_-}{dr} = \frac{\alpha_-}{r_-}\left(1 - \frac{r_+}{\alpha_+}\frac{d\alpha_+}{dr}\right) , \qquad (4)$$

where $\alpha_f$ and $E_f$ are, respectively, the electronic polarizability and the excitation-energy parameter of the ion in the free state. $V_M$ is the Madelung potential existing at the cation site. $r_+$ and $r_-$ are the radii of the cation and anion, respectively. The expressions for the higher-order derivatives of $\alpha_+$ and $\alpha_-$ can be obtained from Eqs. (3) and (4) by the method of differentiation. The calculated values of the strain derivatives of $\epsilon_\infty$ are reported in Table I. Input data on ionic radii are taken from Tosi,[3] and electronic polarizabilities from Ruffa.[14]

The static dielectric constant arises from ionic as well as electronic polarizations. Following the Clausius-Mossotti relation, one can write

$$(\epsilon_s - 1)/(\epsilon_s + 2) = 4\pi\alpha_s/3V \qquad (5)$$

with

$$\alpha_s = \alpha_\infty + \alpha_i . \qquad (6)$$

$\alpha_i$, the ionic polarizability is given by

$$\alpha_i = (ZeS)^2/A , \qquad (7)$$

where $A$ is the short-range force constant coupling neighboring ions, $Z$ is the valence, and $S$ is the effective-charge parameter. The first- and higher-order straing derivatives of $\epsilon_s$ can be obtained from Eqs. (5)–(7). The force constant $A$, based on the theory of interatomic forces in ionic crystals, can be expressed as[21]

$$A = \tfrac{1}{3}[B^{II}(r) + (2/r)B^{I}(r)] , \qquad (8)$$

where $B^{I}(r) = \partial B(r)/\partial r$ and $B^{II}(r) = \partial^2 B(r)/\partial r^2$. $B(r)$ is the short-range potential energy. It can be expressed as the sum of overlap repulsive, $\phi(r)$, and Van der Waals energies

$$B(r) = \phi(r) - C/r^6 - D/r^8 , \qquad (9)$$

where $C$ and $D$ are, respectively, the Van der Waals dipole-dipole and dipole-quadrupole coefficients. The Born model makes no attempt to evaluate the repulsive energy in an ionic crystal from first principles. Instead, it assumes a sim-

TABLE I. Values of the strain derivatives of the electronic dielectric constant.

| | $V\frac{d\epsilon_\infty}{dV}$ | | $V^2\frac{d^2\epsilon_\infty}{dV^2}$ | $V^3\frac{d^3\epsilon_\infty}{dV^3}$ |
|---|---|---|---|---|
| Crystal | Calculated | Experimental | Calculated | Calculated |
| LiF | −0.62 | −0.37[a] | 0.99 | −2.37 |
| LiCl | −1.49 | | 3.00 | −9.23 |
| LiBr | −1.80 | | 3.47 | −10.15 |
| LiI | −2.54 | | 5.25 | −16.43 |
| NaF | −0.48 | −0.49[a] | 0.74 | −1.77 |
| NaCl | −0.95 | −0.85[a] | 1.61 | −4.13 |
| NaBr | −1.06 | −0.99[a] | 1.71 | −3.71 |
| NaI | −1.41 | | 0.92 | −0.78 |
| KF | −0.76 | | 1.61 | −5.35 |
| KCl | −0.88 | −0.93[a] | 1.55 | −4.28 |
| KBr | −0.89 | −1.10[a] | 1.54 | −3.37 |
| KI | −1.09 | −1.44[a] | 1.56 | −2.96 |
| RbF | −0.93 | | 2.19 | −7.83 |
| RbCl | −0.97 | −1.03[a] | 2.03 | −6.22 |
| RbBr | −0.97 | −1.22[a] | 2.01 | −5.54 |
| RbI | −1.09 | −1.43[a] | 1.96 | −4.67 |
| $CaF_2$ | −0.21 | −0.57[b] | −1.23 | |
| $SrF_2$ | −0.67 | −0.85[b] | 0.73 | |
| $BaF_2$ | −1.15 | −1.06[b] | 2.72 | |

[a] Derived from the experimental data on the pressure dependence of the refractive index reported by Bendow *et al.* (Ref. 40).

[b] Cited in Ref. 11.

TABLE II. Values of the strain derivatives of the static dielelectric constant.

| | $V\left(\frac{d\epsilon_s}{dV}\right)$ | | | | | | $V^2\left(\frac{d^2\epsilon_s}{dV^2}\right)$ | | | $V^3\left(\frac{d^3\epsilon_s}{dV^3}\right)$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Experimental | | | | | | Experimental | | | Experimental |
| Crystal | a | b | c | d | e | f | a | b | f | a | b | f |
| LiF | 27.5 | 17.7 | 29.4 | 23.0 | 30.4 | 30.0 | 163 | 84 | 157 ± 7 | 1399 | 559 | −608 ± 1070 |
| LiCl | 38.3 | 26.0 | | 30.4 | 53.4 | | 279 | 158 | | 2787 | 1220 | |
| LiBr | 43.6 | 29.2 | | 40.4 | | | 329 | 185 | | 3464 | 1495 | |
| NaF | 10.8 | 7.80 | 12.0 | 12.5 | 12.2 | 12.7 | 48 | 31 | 50 ± 3 | 281 | 150 | −597 ± 390 |
| NaCl | 11.2 | 8.07 | 14.3 | 13.0 | 12.5 | 14.5 | 53 | 36 | 67 ± 3 | 315 | 177 | +37 ± 660 |
| NaBr | 11.5 | 8.08 | 15.5 | 14.9 | 16.0 | 15.9 | 55 | 36 | 99 ± 4 | 329 | 178 | +1081 ± 980 |
| NaI | 14.1 | 10.1 | | 16.6 | 12.4 | | 74 | 50 | | 486 | 273 | |
| KF | 13.3 | 9.77 | 8.93 | 12.8 | 11.3 | | 73 | 49 | | 516 | 285 | |
| KCl | 8.46 | 6.26 | 8.60 | 8.45 | 8.89 | 9.11 | 40 | 29 | 40 ± 2 | 225 | 136 | −296 ± 323 |
| KBr | 7.81 | 5.68 | 7.50, 8.93 | 8.35 | 8.21 | 8.92 | 36 | 25 | 44 ± 2 | 189 | 114 | +129 ± 431 |
| KI | 7.07 | 5.07 | | 8.16 | 6.58 | | 32 | 23 | | 159 | 113 | |
| RbF | 19.4 | 14.2 | | 14.5 | | | 133 | 85 | | 1215 | 632 | |
| RbCl | 9.17 | 6.91 | 8.73 | 7.79 | 8.22 | | 48 | 35 | | 294 | 185 | |
| RbBr | 8.27 | 6.23 | 7.96 | 7.46 | 7.37 | | 42 | 30 | | 238 | 146 | |
| RbI | 7.01 | 5.10 | | 6.94 | 6.39 | | 34 | 24 | | 174 | 107 | |
| $CaF_2$ | 19.9 | 15.3 | 19.0 | 22.1[g] | 20.4[h] | 18.7[i] | 105 | 66 | 112 ± 5[i] | | | |
| $SrF_2$ | 16.9 | 13.0 | | 21.8[g] | 16.0[h] | 16.0[i] | 94 | 72 | 85 ± 4[i] | | | |
| $BaF_2$ | 21.5 | 16.4 | 17.7 | 22.1[g] | 17.5[h] | 18.4[i] | 145 | 122 | 106 ± 5[i] | | | |

[a] Calculated from the inverse power form.
[b] Calculated from the exponential form.
[c] Reference 33.
[d] Reference 34.
[e] Reference 35.
[f] Reference 10.
[g] Reference 37.
[h] Reference 16.
[i] Reference 11.

ple functional dependence of the repulsive energy on the distance between the ions, and determining the intervening parameters from crystal data. In the early work on the cohesion of ionic solids, the Born repulsive energy was assumed to vary as an inverse power of the distance between the ions, with an exponent $\eta$ independent of the ion species. The repulsive potential energy has then the form

$$\phi(r) = \mu / r^{\eta} , \tag{10}$$

and the parameters $\mu$ and $\eta$ can be determined directly for each crystal from the equation of state and its volume derivative. In most applications of the Born model subsequent to 1930, the repulsive energy of two closed-shell ions has been assumed to vary exponentially with their distance, in keeping with the quantum-mechanical results.[22] The repulsive energy, as a function of the nearest-neighbor distance $r$, is then written in the form

$$\phi(r) = \lambda \exp(-r/\rho) , \tag{11}$$

and the parameters $\lambda$ and $\rho$ are derived similarly to $\mu$ and $\eta$.

In the present paper we adopt the inverse power as well as the exponential form to evaluate the lattice contribution to the strain derivatives of the static dielectric constant of alkali halides and alkali-earth fluorides. Taking account of the Van der Waals interactions, we can express the crystal energy $W$ as follows:

$$W = -\frac{\delta e^2}{r} + \phi(r) - \frac{C}{r^6} - \frac{D}{r^8} , \tag{12}$$

where $\delta$ is the Madelung constant. The two parameters appearing in each repulsive potential form can be evaluated from the following relations:

$$\frac{dW}{dr} = \frac{\delta e^2}{r^2} + \phi^{\mathrm{I}}(r) + \frac{6C}{r^7} + \frac{8D}{r^9} = 0 , \tag{13}$$

$$\frac{d^2W}{dr^2} = -\frac{2\delta e^2}{r^3} + \phi^{\mathrm{II}}(r) - \frac{42C}{r^8} - \frac{72D}{r^{10}} = \frac{9Kr}{\chi_T} , \tag{14}$$

where $\phi^{\mathrm{I}}(r) = d\phi/dr$ and $\phi^{\mathrm{II}}(r) = d^2\phi/dr^2$. $\chi_T$ is the isothermal compressibility. Values of the repulsive-potential parameters can be derived from Eqs. (13) and (14). The input data used in the calculations are as follows: $r$ from Tosi,[3] $\chi_T$ from Smith and Cain,[8] $C$ from Hajj,[23] and $D$ from Mayer.[24] Values of $A$ and its higher-order derivatives can be calculated from Eqs. (8)–(14). Using these, we can obtain the derivatives of $\alpha_i$, $\alpha_s$, and $\epsilon_s$. The first- and higher-order derivatives of $\epsilon_s$ calculated in the present study are given in Table II.

## III. RESULTS AND DISCUSSION

It is interesting to observe from Table I that $d\epsilon_\infty/dV$ is negative, $d^2\epsilon_\infty/dV^2$ is positive, and $d^3\epsilon_\infty/dV^3$ is again negative. The prediction that $d\epsilon_\infty/dV$ is negative for alkali halides and alkaline-earth fluorides is consistant with experimental data. Our calculated values of $Vd\epsilon_\infty/dV$ are in close agreement with experiment (Table I). The prediction that $V^2(d^2\epsilon_\infty/dV^2)$ is positive for alkali halides has been confirmed by an independent method[25] based on the Penn model.[26]

The evaluation of the strain derivaties of $\epsilon_s$ depends upon those of $\alpha_i$. It should be emphasized that the strain derivatives of $\alpha_i$ have been calculated with the help of Eq. (7). To reduce the computational labor we have evaluated quantities like $(1/\alpha_i)(d\alpha_i/dV)$, $(1/\alpha_i)(d^2\alpha_i/dV^2)$, etc. From Eq. (7) we find

$$\frac{1}{\alpha_i}\frac{d\alpha_i}{dV} = -\frac{1}{A}\frac{dA}{dV} , \tag{15}$$

which is clearly independent of the ionic charge or $S$. In deriving Eq. (15) it has been assumed that $S$ is independent of volume. Recent experimental and theoretical studies[27-32] support the approximate validity of this assumption. An equation similar to (15) was for the first time used by Bosman and Havinga.[15]

Values of $A$ and its derivatives sensitively depend upon the short-range energy $B(r)$ expressed by Eq. (9). It is seen from Table II that the strain derivatives of $\epsilon_s$ calculated using the inverse-power law are larger than the corresponding magnitudes obtained from the exponential law. The inclusion of Van der Waals dipole-dipole and dipole-quadrupole interactions significantly changes the magnitudes of the strain derivatives of $\epsilon_s$. This is why the results reported in this paper differ considerably from those obtained by Sharma *et al.*,[13] who use the Born model without Van der Waals interactions. In alkaline-earth fluorides we have not considered the Van der Waals interactions because of the scaricity of reliable information on these interactions. We have neglected also the second-neighbor repulsive interactions, as these do not influence the dielectric behavior of solids.

In recent times there have been various experimental investigations[10,11,15,16,33-37] on the measurements of the pressure derivates of $\epsilon_s$ for alkali halides and alkaline-earth fluorides. These measurements, in general, are based on the three-terminal geometrical-capactiance method which has distinguishable merits[38] over the old immersion method.[39] The three-terminal geometrical method is better than the latter in two respects. First, it is a quicker method, and second, it does not use high-dielectric-constant liquids which can prove troublesome. In fact, the liquid-solid surface interactions produce significant changes in the dielectric constant measured by the immersion method. For alkali halides the measurements performed by Fontanella *et al.*[10] are highly accurate as these authors have assigned an uncertainty of only 0.2% to the first-order pressure derivatives of $\epsilon_s$, whereas the uncertainty reported by other investigators[34,35] is of the order of ±5%. The results of Fontanella *et al.* have subsequently been corroborated by Bartels and Smith.[36] The high accuracy of the results obtained by Fontanella *et al.* is due in part to the use of higher-order terms to describe the variation of $\epsilon_s$ with pressure and in part to the improved experimental techniques. Andeen *et al.*[11] have measured the pressure derivatives of $\epsilon_s$ for alkaline-earth fluorides adopting a similar experimental method to that of Fontanella *et al.* Measurements reported by Andeen *et al.* are accurate within 0.1% and are in close agreement with those published by Jones[33] and Samara.[16] The second- and third-order pressure derivatives of $\epsilon_s$ for six alkali halides, viz. LiF, NaF, NaCl, NaBr, KCl, and KBr have been determined by Fontanella *et al.* to an accuracy of about 2% and 20%, respectively. The second-order pressure derivatives of $\epsilon_s$ for $CaF_2$, $SrF_2$, and $BaF_2$ have been measured by Andeen *et al.* to an accuracy of about 2%.

Values of the strain derivatives of $\epsilon_s$ based on the experimental data are included in Table II for the sake of comparison. We note from Table II that the experimental errors are very large for $V^3(d^3\epsilon_s/dV^3)$. In contrast, $V(d\epsilon_s/dV)$ and $V^2(d^2\epsilon_s/dV^2)$ appear to be quite accurate. Our calculated values of $V(d\epsilon_s/dV)$ and $V^2(d^2\epsilon_s/dV^2)$ present good agreement with experimental data (Table II). It is also encouraging to note that our calculated values of $V^3d^3\epsilon_s/dV^3$ for LiF, NaCl, NaBr, and KBr agree with experimental data within the experimental uncertainties. Obviously there is need for more accurate measurments of the third-order pressure derivatives of $\epsilon_s$ for a larger number of crystals.

The first- and second-order strain derivatives of $\epsilon_s$ calculated from the inverse-power law are, in general, closer to the experimental values than those evaluated from the exponential law. It is pertinent to mention here that Smith and co-workers[8] have recently calculated the pressure dependence of the isothermal bulk modulus for alkali halides adopting the exponential and the inverse-power forms for the short-range energy and using their ultrasonically measured data. These investigators also found that the inverse-power law yields better agreement with experiment. Thus the present study and the investigations of Smith and co-workers reveal that the inverse-power law is more consistent with the experimental data on the pressure derivatives of dielectric and elastic constants. Moreover, our results, like those of Smith and co-workers, predict that the exponential law yields values lower than those obtained from the inverse-power law.

Finally, it should be mentioned that the strain derivatives of $\epsilon_\infty$ are much smaller in magnitude than the corresponding derivatives of $\epsilon_s$. This is because the dominant contribution to the strain derivatives of $\epsilon_s$ arises from the relative displacement of the ions. A similar conclusion has been drawn by previous investigators.[15,16]

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