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Three-body interaction and fluorite structure: Elastic properties of CaF_2, SrF_2, and BaF_2

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In spite of the fact that second-order elastic constants of some ionic crystals with fluorite structure can be fitted quite well by the shell model using the two-body interaction only, the situation is still unsatisfactory. In our previous work, it was therefore attempted to provide a satisfactory picture by introducing the three-body interaction. The importance of this interaction was estimated from a study of second-order elastic constants by using mainly the results of a first-principles calculation. In this paper, this investigation is extended to third-order elastic constants. In order to obtain explicit expressions for elastic constants, the terms arising from the Coulomb interaction and shell structure are determined from the work of Srinivasan and those arising from non-Coulomb two- and three-body interactions by the homogeneous deformation theory developed in the preceding paper. The results confirm the importance of the three-body interaction in the study of the elastic properties of CaF_2, SrF_2, and BaF_2.

I. INTRODUCTION

Ionic crystals with fluorite structure have been studied widely for a long time, primarily because they have one of the simplest crystal structures among those which lack a center of symmetry. It has been shown[1–3] that, although the elastic properties of some such crystals can be fitted quite well by the shell model without introducing the three-body interaction, the situation is still unclear. The contradictory conclusions of different authors while applying similar models to this class of materials and abnormally large estimates of some of the parameters of well-known interactions, are examples of the problems in the present state of our understanding. Regarding the role of the three-body interaction, the lack of clarification is amply described by Hayes and Stoneham,[2] who observe that the difference $c_{12}-c_{44}$ in Axe's calculation may be explained by a shell model without the three-body forces "although these contributions may be present." In this context we have previously tried to investigate[4] the importance of the three-body interaction, in the cohesion and the second-order elastic constants of these materials through a simple model, namely, the deformable shell model. The parameters of the model for calcium fluoride crystal were determined from the free-ion wave functions, so that an independent estimate of different interactions was possible. A detailed discussion of this point is given in the earlier work, which will be referred to as BSB. Encouraged by the results obtained therein, we thought it would be worthwhile to extend the calculations to the third-order elastic constants of these crystals. It is expected that since these constants involve still higher-order derivatives of the potential function, three-body interaction may play a more dominant role here.

The model to be used is the same as that in BSB. Each ion is polarizable; the shells interact among themselves through a two-body interaction (short-range and Coulomb) and a three-body interaction that arises due to their deformability; each core interacts through the usual spring-type and Coulomb interaction. In order to work out the explicit expressions for the third-order elastic constants, the non-Coulomb (two- and three-body) terms will be derived from the theory developed in the preceding paper[5] (called I hereafter), and the contributions of Coulomb interaction and polarizability of the ions will be taken from the work of Srinivasan.[3] The final agreement of the theoretical values with experiment and the fact that the inclusion of the three-body term improves the agreement in almost every case indicate the importance of three-body interaction for the anharmonic elastic properties of the materials under study.

In Sec. II we shall derive expressions for the elastic constants. First, they will be written (Sec. II A) in terms of the coefficients introduced in paper I; then the contributions of the non-Coulomb (Sec. II B) and Coulomb (Sec. II C) interactions will be obtained; finally, complete expressions will be written (Sec. II D) with a few words on their verification. In Sec. III we shall apply the theory to CaF_2, SrF_2, and BaF_2.

II. DERIVATION OF THE EXPRESSIONS FOR ELASTIC CONSTANTS FOR FLOURITE STRUCTURE

A. Expressions for elastic constants in terms of the coefficients

In the next section, we shall evaluate the contribution of non-Coulomb two-body and three-body interactions to the coefficients of I for the special case of fluorite structure using the general results obtained in I; the terms arising from Coulomb interaction and shell structure will not be derived explicitly. For our purpose, therefore, it is sufficient to work in the rigid-ion model. Thus the particles in a unit cell are Ca ($k=1$), F_1 ($k=2$), and F_2 ($k=3$), with the convention that Ca-F_1 sublattice has cubic ZnS structure. Also, we choose the internal strain of calcium ion to be zero,[6] $\vec{\mathrm{u}}(1)=0$.

Now, for fluorite structure, the surrounding of any particle has at least the point-group symmetry T_d. Hence all second-rank tensors reduce to scalars, and third-rank tensors have only one independent element for which all the three Cartesian suffixes are different. In particular, we have

$$A\left(\begin{smallmatrix}k_1 & k_2\\ \alpha & \beta\end{smallmatrix}\right)=\delta_{\alpha\beta}A(k_1,k_2)\,,$$

$$A\left(\begin{smallmatrix}k_0\\ \alpha\end{smallmatrix}\beta\gamma\right)=\begin{cases}A(k_0) & \text{if } \alpha,\beta,\gamma \text{ are unequal}\\ 0 & \text{otherwise}\,,\end{cases}$$

and similarly for the coefficient G. Furthermore, since the surroundings of F_1 and F_2 ions are related by the operation of inversion, we have $A(2)=-A(3)$. Using these properties, one can solve easily Eq. (3) of I (where k_1,k_2 run over 2,3) and obtain

$$G\left(\begin{smallmatrix}2\\ \alpha\end{smallmatrix}\beta\gamma\right)=-G\left(\begin{smallmatrix}3\\ \alpha\end{smallmatrix}\beta\gamma\right)=A\left(\begin{smallmatrix}2\\ 1\end{smallmatrix}23\right)\Big/\left[A\left(\begin{smallmatrix}23\\ 11\end{smallmatrix}\right)-A\left(\begin{smallmatrix}22\\ 11\end{smallmatrix}\right)\right]\,, \tag{1}$$

where α,β,γ are unequal. Substituting for these two G coefficients in Eq. (3) of I and applying the symmetry operations for the fluorite structure, one obtains the following results:

$$\begin{aligned}
&c_{11}=A(1111),\quad c_{12}=A(1122),\quad c_{44}=A(2323)+2GA\left(\begin{smallmatrix}2\\ 1\end{smallmatrix}23\right)\,,\\
&c_{111}=B(111111),\quad c_{112}=B(111122),\quad c_{123}=B(112233)\,,\\
&c_{144}=B(112323)+4GB\left(\begin{smallmatrix}2\\ 1\end{smallmatrix}1123\right)+2G^2\left[B\left(\begin{smallmatrix}22\\ 11\end{smallmatrix}11\right)-B\left(\begin{smallmatrix}23\\ 11\end{smallmatrix}11\right)\right]\,,\\
&c_{155}=B(111313)+4GB\left(\begin{smallmatrix}2\\ 2\end{smallmatrix}1113\right)+2G^2\left[B\left(\begin{smallmatrix}22\\ 22\end{smallmatrix}11\right)-B\left(\begin{smallmatrix}23\\ 22\end{smallmatrix}11\right)\right]\,,\\
&c_{456}=B(231312)+6GB\left(\begin{smallmatrix}2\\ 1\end{smallmatrix}1312\right)+6G^2\left[B\left(\begin{smallmatrix}22\\ 12\end{smallmatrix}12\right)-B\left(\begin{smallmatrix}23\\ 12\end{smallmatrix}12\right)\right]+2G^3\left[B\left(\begin{smallmatrix}222\\ 123\end{smallmatrix}\right)-B\left(\begin{smallmatrix}223\\ 123\end{smallmatrix}\right)\right]\,.
\end{aligned} \tag{2}$$

Here, Voigt contracted notation has been used in the suffix of c and $G\left(\begin{smallmatrix}2\\ 1\end{smallmatrix}23\right)$ is abbreviated as G.

B. Coefficients for the non-Coulomb interaction

In the rigid-ion model, the (scalar) potential functions for the non-Coulomb two-body interaction are $\phi_1(R)$ between nearest-neighbor calcium and fluorine ions, and $\phi_2(R)$ between nearest-neighbor fluorine ions, where R is the separation between the interacting ions. Using the expressions given in Sec. III of paper I, one can obtain the coefficients that appear in Eq. (2) above, in terms of the derivatives of ϕ_1 and ϕ_2.

As for the three-body interaction, the particles $(l'k')$ and $(l''k'')$ interact via a third particle (lk) through the potential

$$\psi=A_k\exp\left[-\frac{R+R'}{\rho}\right]\,,$$

provided that (lk) is a common nearest neighbor of $(l'k')$ and $(l''k'')$. Here A_k is a parameter, characteristic of the deformability of the electron cloud of the k ion, ρ is range parameter of overlap interaction between the nearest neighbors, $\vec{\mathrm{R}}$ stands for $\vec{\mathrm{R}}(l'k')-\vec{\mathrm{R}}(lk)$ and $\vec{\mathrm{R}}\,'$ for $\vec{\mathrm{R}}(l''k'')-\vec{\mathrm{R}}(lk)$. One can obtain the contribution of this interaction towards the coefficients of Eq. (2) by using the expressions of Sec. IV, paper I, through a laborious calculation, which can be simplified, however, by using the following observations.

(i) The equilibrium value of ψ and its derivatives depend on the index k only (through the term A_k).

(ii) The coefficients $B\left(\begin{smallmatrix}k_1 & k_2 & k_3\\ \alpha & \beta & \gamma\end{smallmatrix}\right)$ appears always in such a way that α,β,γ are all unequal and no two among $(l_1k_1),(l_2k_2),(l_3k_3)$ are nearest neighbors for any l_1,l_2,l_3. This implies that the summations where only one among k_1,k_2,k_3 is k will fall off.

(iii) For the coefficient $B\left(\begin{smallmatrix}k_1 & k_2\\ \alpha & \beta\end{smallmatrix}\gamma\epsilon\right)$ also, (l_1k_1) and (l_2k_2) are never nearest neighbors of each other.

C. Coefficients for the Coulomb interaction

As mentioned in I (Sec. V), for the Coulomb interaction some of the coefficients become indeterminate and the expressions of Sec. III of I cannot be used. In particular, each coefficient of the types $A\left(\begin{smallmatrix}k_1 & k_2\\ \alpha & \beta\end{smallmatrix}\right)$ and $B\left(\begin{smallmatrix}k_1 & k_2\\ \alpha & \beta\end{smallmatrix}\gamma\epsilon\right)$ that enters in Eq. (2) is indeterminate. However, the fluorite structure has the striking feature that, despite the indeterminacy, the combinations of the coefficients [e.g., $A\left(\begin{smallmatrix}23\\ 11\end{smallmatrix}\right)-A\left(\begin{smallmatrix}22\\ 11\end{smallmatrix}\right)$] which appear in Eq. (2) do converge, due to a cancellation of the parts which involve macroscopic electric field. Another feature of the fluorite structure is that although it is ionic and lacks in inversion symmetry at every particle position, it is not piezoelectric—the internal strain of the F_1 ions with respect to Ca ions is equal and opposite to that of the F_2 ions, so that the dipole moment of the $\frac{1}{2}$Ca-F_1 sublattice exactly balances that of the $\frac{1}{2}$Ca-F_2 sublattice.

To obtain the terms arising from Coulomb interaction in the expressions for elastic constants, we use the results of Srinivasan,[3] who has calculated the second- and third-order elastic constants assuming non-Coulomb interactions ϕ_1 and ϕ_2, in the framework of a both-ions—polarizable simple-shell model. However, since the calcium ions in the initial configuration occupy a position which is a center of symmetry, under a homogeneous deformation of the solid, this symmetry is maintained and these ions do not develop any dipole moment. Hence the only contribution to elastic constants comes from the polarizability of the fluorine ions. For infinitely large values of the spring constant k_2 for fluorine ions, his results reduce to those of a rigid-ion model. We compare these reduced results with our expressions applied to the special case when only two-body interaction is present. During comparison, however, care must be taken to make the first-order derivatives of ϕ_1 and ϕ_2 in Srinivasan's expressions identical with those in ours. This is because by the use of equilibrium condition [Eq. (3) below] one can express a combination of the two first-order derivatives in terms of a Coulomb term. Obtaining in this way the Coulomb contribution to each of the coefficients, we add the contribution of three-body interaction as computed in Sec. II B. This is possible because in each coefficient the contributions from different interactions are linearly additive. Now, since Srinivasan's expressions for finite values of k_2 show that the terms arising from shell structure in the expression for elastic constants come in additive form over the rigid ion values, we retain his evaluation of the contribution of the shell structure to the different elastic constants.

D. Final expressions for elastic constants

We give below the final expressions for Brugger elastic constants as obtained from above. New notations introduced are as follows: $-e$ is the total charge of the ion at fluorine site, Y_2e the shell charge of that ion, k_2 the spring constant of that ion, a is half the lattice constant,

$$S_1=\frac{e^4}{k_2a^7}(1+Y_2)^2,\quad S_2=\frac{e^2}{a^3}\frac{1+Y_2}{k_2}\ ,$$

$t_{11}=(1/a)\phi_1'$, $t_{12}=a\phi_1''$, $t_{13}=a^3\phi_1'''$, and similarly, $t_{21}=(1/a)\phi_2'$, etc., where the primes indicate derivatives with respect to R^2 evaluated at equilibrium. In addition,

$$B_1=A_1\exp(-\sqrt{3}a/\rho),\quad B_2=A_2\exp(-\sqrt{3}a/\rho)\ ,$$

$$M_1=\left[\frac{2}{\sqrt{3}\rho a^2}+\frac{2}{\rho^2a}\right]\left[\frac{7}{3}B_1+B_2\right],$$

$$M_2=\left[\frac{14}{3\sqrt{3}\rho a^2}+\frac{2}{\rho^2a}\right]B_1+\left[\frac{2}{\sqrt{3}\rho a^2}+\frac{2}{3\rho^2a}\right]B_2\ ,$$

$$M_3=\left[\frac{2}{\sqrt{3}\rho a^2}+\frac{2}{\rho^2a}+\frac{2}{\sqrt{3}\rho^3}\right]\left[\frac{7}{3}B_1+B_2\right],$$

$$M_4=\left[\frac{14}{3\sqrt{3}\rho a^2}+\frac{26}{9\rho^2a}+\frac{2}{\sqrt{3}\rho^3}\right]B_1+\left[\frac{2}{\sqrt{3}\rho a^2}+\frac{10}{9\rho^2a}+\frac{2}{3\sqrt{3}\rho^3}\right]B_2\ ,$$

$$g=G/a=-\left[t_{12}+M_2-2.5144\frac{e^2}{a^4}\right]\Bigg/\left[2t_{12}+8t_{22}+4t_{11}+12t_{21}-\left[\frac{56}{3\sqrt{3}\rho a^2}-\frac{4}{\rho^2a}\right]B_1-\left[\frac{8}{\sqrt{3}\rho a^2}-\frac{4}{3\rho^2a}\right]B_2\right].$$

The final expressions are

$$c_{11}=t_{12}+4t_{22}+M_1+0.5559\frac{e^2}{a^4}\ ,$$

$$c_{12}=t_{12}+M_1-1.7325\frac{e^2}{a^4}\ ,$$

$$c_{44}=t_{12}+M_2-1.7325\frac{e^2}{a^4}+2g\left[t_{12}+M_2-2.5144\frac{e^2}{a^4}\right]-25.29S_1\ ,$$

$$c_{111}=\tfrac{1}{2}t_{13}+8t_{23}-M_3-5.9627\frac{e^2}{a^4}\ ,$$

$$c_{112}=\tfrac{1}{2}t_{13}-M_3+1.5917\frac{e^2}{a^4}\ ,$$

$$c_{123}=\tfrac{1}{2}t_{13}-M_3+5.4791\frac{e^2}{a^4}\ ,$$

$$c_{144}=\tfrac{1}{2}t_{13}-M_4+5.4791\frac{e^2}{a^4}$$
$$+2g\left[t_{13}+2t_{12}-\left[\frac{16}{9\rho^2 a}+\frac{4}{\sqrt{3}\rho^3}\right]B_1-\left[\frac{8}{9\rho^2 a}+\frac{4}{3\sqrt{3}\rho^3}\right]B_2+3.3526\frac{e^2}{a^4}\right]$$
$$+g^2\left[2t_{13}+32t_{23}+12t_{12}+48t_{22}+8M_4-\frac{8\sqrt{3}}{\rho^3}(B_1+\tfrac{1}{3}B_2)-36.3362\frac{e^2}{a^4}\right]$$
$$+S_1(33.7191-365.4555g-918.9000S_2-3000.9143Y_2S_2)\,,$$

$$c_{155}=\tfrac{1}{2}t_{13}-M_4+1.5917\frac{e^2}{a^4}+2g\left[t_{13}-2M_4+8.3814\frac{e^2}{a^4}\right]$$
$$+g^2\left[2t_{13}+4t_{12}+16t_{22}+\left[\frac{64}{9\rho^2 a}-\frac{8}{\sqrt{3}\rho^3}\right]B_1+\left[\frac{32}{9\rho^2 a}-\frac{8}{3\sqrt{3}\rho^3}\right]B_2+18.1681\frac{e^2}{a^4}\right]$$
$$+S_1(84.2968+152.5547g+459.4500S_2+150.4584Y_2S_2)\,,$$

$$c_{456}=\tfrac{1}{2}t_{13}-M_2-\frac{2}{3\sqrt{3}\rho^3}B_1+5.4791\frac{e^2}{a^4}+3g\left[t_{13}+2t_{12}-\frac{4}{3\sqrt{3}\rho^3}B_1+3.3526\frac{e^2}{a^4}\right]$$
$$+3g^2\left[2t_{13}+4t_{12}+16t_{22}-\frac{8}{3\sqrt{3}\rho^3}B_1+18.1681\frac{e^2}{a^4}\right]+g^3\left[4t_{13}-8M_2-\frac{16}{3\sqrt{3}\rho^3}B_1+38.4058\frac{e^2}{a^4}\right]$$
$$+S_1(50.5787+548.1824g+579.4045g^2+1378.3499S_2+451.3727Y_2S_2+2913.7094gS_2+4884.1539S_2^2)\,.$$

The cohesive energy per unit cell is

$$U=8\phi_1+6\phi_2+(28A_1+12A_2)\exp\left[-\frac{\sqrt{3}a}{\rho}\right]-5.8182\frac{e^2}{a}\,.$$

The equilibrium condition which ensures the vanishing of isotropic pressure in initial configuration is

$$t_{11}+t_{21}+0.4848\frac{e^2}{a^4}-\frac{1}{3\sqrt{3}\rho a^2}(\tfrac{7}{3}B_1+B_2)=0\,. \qquad (3)$$

We have verified that the expressions for the second-order elastic constants given above agree with those obtained otherwise in BSB, after using the equilibrium condition. As a partial check of the expressions for the third-order elastic constants, we have verified the following equations, which in fact describe the change of second-order elastic constants under bulk stress:[7]

$$a\frac{dc_{11}}{da}=c_{11}+c_{111}+2c_{112}\,,$$
$$a\frac{dc_{12}}{da}=c_{12}+c_{123}+2c_{112}\,, \qquad (4)$$
$$a\frac{dc_{44}}{da}=c_{44}+c_{144}+2c_{155}\,.$$

TABLE I. Values of parameters.

Material	a (10^{-1} nm)	b (10^{-16} J)	ρ (10^{-2} nm)	Y_2	k_2 (10^3 N/m)	A_2 (10^{-13} J)	b'^{a} (10^{-10} J/m)	σ^{a} (10^{-1} nm)	ξ^{a} (10 nm^{-1})
CaF_2	2.717[b]	5.7919	2.535	−2.35	1.230	2.38	−1.577	2.883	1.988
SrF_2	2.885[b]	2.5222	3.000	−2.35	1.228	0.28			
BaF_2	3.088	4.1266	3.000	−2.35	1.232	0.71			

[a] Same for all materials.

[b] Harmonic value.

TABLE II. Cohesive energy (in 10^6 J/mol) and elastic constants (in 10^{11} N/m^2) of CaF_2, SrF_2, BaF_2. The experimental values of second-order elastic constants are the harmonic ones, those of third-order elastic constants are at 295 K, and those of cohesive energy, at 0 K.

	$-U$	c_{11}	c_{12}	c_{44}	$-c_{111}$	$-c_{112}$	$-c_{123}$	$-c_{144}$	$-c_{155}$	$-c_{456}$
CaF_2										
Experiment	2.59[a]	1.759[b]	0.684[b]	0.360[b]	12.46±0.91[c]	4.00±0.30[c]	2.54±0.29[c]	1.24±0.15[c]	2.14±0.09[c]	0.748±0.038[c]
Calculation	2.69	2.038	0.656	0.292	12.69	5.36	3.72	2.05	2.87	0.97
Calculation without three-body interaction	2.70	1.789	0.407	0.219	10.98	3.65	2.00	1.93	2.74	1.29
SrF_2										
Experiment	2.44[a]	1.302[d]	0.484[d]	0.353[d]	8.21±0.11[e]	3.09±0.05[e]	1.81±0.12[e]	0.951±0.066[e]	1.75±0.03[e]	0.421±0.028[e]
Calculation	2.50	1.427	0.430	0.218	8.42	3.34	2.04	1.24	2.05	0.65
Calculation without three-body interaction	2.51	1.296	0.299	0.177	7.60	2.52	1.22	1.18	1.98	0.82
BaF_2										
Experiment	2.30[a]	0.981[f]	0.448[f]	0.254[f]	5.84±0.15[g]	2.99±0.14[g]	2.06±0.11[g]	1.21±0.03[g]	0.889±0.019[g]	0.271±0.001[g]
Calculation	2.35	1.092	0.398	0.232	6.29	2.96	1.97	0.92	1.57	0.44
Calculation without three-body interaction	2.36	0.995	0.301	0.209	5.65	2.31	1.33	0.88	1.57	0.58

[a] Reference 8.
[b] Reference 9.
[c] Reference 10.
[d] Reference 11.
[e] Reference 12.
[f] Reference 13.
[g] Reference 14.

III. APPLICATION TO SPECIFIC MATERIALS

In this section, the model developed so far will be applied to the single crystals CaF_2, SrF_2, and BaF_2. The specific forms of nearest-neighbor and second-nearest-neighbor interaction that will be used here are the same as those in BSB:

$$\phi_1(r)=b\exp(-r/\rho)\ ,$$

$$\phi_2(r)=b'(r-\sigma)\exp(\sigma\xi-r\xi)\ .$$

Since the strongly bound valence-electron cloud of the cation will be deformed much less than the loosely bound one of the anion, the parameter A_1 has been neglected in comparison to A_2. The values of all the parameters (Table I) are taken from BSB. Cohesive energy and elastic constants calculated by using these values are displayed in Table II.[15]

The results for the ten properties (cohesive energy and the nine elastic constants) of the three materials show up the prominent feature that, in each case (except c_{11} for CaF_2) inclusion of three-body interaction introduces a correction in the right direction. This striking observation indicates strongly that three-body interaction is indeed important for the study of elastic properties of the materials concerned. For CaF_2 the result is more impressive, because all the parameters (except Y_2 and k_2) are fixed without using any crystal property (see BSB). For SrF_2 and BaF_2, however, two parameters were fixed otherwise. Table II shows furthermore that, for each of the ten properties calculated there, the relative contribution of three-body interaction is of the same order of magnitude for the three crystals, as one would expect normally. The relative contribution of three-body interaction to second- and third-order elastic constants range up to 38% and 46%, respectively. Lastly, despite the rather crude way of fixing the parameters, the agreement between the theory and experiment is quite well.

The objective of the present investigation has been to estimate the importance of three-body interaction. In this preliminary application, no scope remains for an emphasis on the agreement between the theoretical and experimental values and we have not allowed the parameter values to vary arbitrarily for obtaining a best fit with the experiment.

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[15] Some calculational mistakes in BSB have been corrected in Tables I and II.