Homogeneous deformation theory with three-body interaction: Second- and third-order elastic constants of noncentrosymmetric crystals

Subinay Dasgupta
Physics Department, Jadavpur University, Calcutta 700032, India
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A general homogeneous deformation theory is developed for an arbitrary nonionic crystal structure, which lacks a center of inversion. The strain-energy density is expanded as a power series in macroscopic and internal strains. Expressions are derived for the second- and third-order elastic constants in terms of the coefficients of expansion, and for these coefficients in terms of the interaction potential. Two types of potentials are considered—a two-body central potential and a three-body potential that depends on two scalar distances between the interacting particles. The final expressions obtained are much simpler than those of previous work, in terms of force-constant matrix elements.

I. INTRODUCTION

The increasing importance of anharmonic properties of solids necessitates a thorough study of the third-order elastic constants. The theoretical calculation of these elastic constants from a microscopic point of view becomes complicated for those crystal structures where every lattice site is not a center of inversion, since in these cases a macroscopic strain gives rise to an internal strain.[1] Srinivasan[2] has given lattice theoretical expressions for third-order elastic constants of such crystals in terms of force-constant matrix elements, by using the method of homogeneous deformation as well as that of long waves. Unfortunately, these expressions are unusually complicated and make application in specific cases quite difficult, particularly when one aims at obtaining elastic constants in terms of interaction potential, and three-body interaction is important. This is primarily because one has to select the force constants in such a way that the well-known Born-Huang rotational and translational invariance conditions are satisfied. This is not at all easy when a large number of force constants have to be used. Moreover, the equilibrium condition (vanishing isotropic pressure) cannot be guaranteed, as isotropic stress cannot be written explicitly in terms of second-order force constants.

On the other hand, in a homogeneous deformation theory starting from a definite potential function for the interaction between the particles, all the above conditions are explicitly used. Hence this approach leads to much simpler final expressions for the elastic constants. Born and Huang[1] also have developed this theory for the second-order elastic constants considering two-body central interaction only. Our purpose in the present paper is to extend this method to the third-order elastic constants for any nonionic noncentrosymmetric crystal structure, considering both two-body central interaction and a class of three-body interaction. The theory will be applied to three crystals with the fluorite structure in the following paper.[3] For centrosymmetric crystals such an extension of homogeneous deformation theory has already been made by Sarkar and Sengupta.[4]

To develop the theory, we consider an infinite solid which undergoes homogeneous deformation described by macroscopic Lagrangian strain $\underline{s}$, and internal strain $\vec{u}(k)$ for atoms of type k. The strain energy per unit undeformed volume is then expanded as a power series in these two types of strains. Thus, up to terms third order in strain, one obtains [see Eq. (11.15) of Ref. 1]

$$E=\tfrac{1}{2}A(\alpha\beta\gamma\epsilon)s_{\alpha\beta}s_{\gamma\epsilon}+A\binom{k_0}{\alpha}{}_{\beta}\,{}_{\gamma})u_\alpha(k_0)s_{\beta\gamma}+\tfrac{1}{2}A\binom{k_1k_2}{\alpha\ \beta}u_\alpha(k_1)u_\beta(k_2)+\tfrac{1}{6}B(\alpha\beta\gamma\epsilon\zeta\eta)s_{\alpha\beta}s_{\gamma\epsilon}s_{\zeta\eta}$$
$$+\tfrac{1}{2}B\binom{k_0}{\alpha}{}_{\beta}\,{}_{\gamma}\,{}_{\epsilon}\,{}_{\zeta})u_\alpha(k_0)s_{\beta\gamma}s_{\epsilon\zeta}+\tfrac{1}{2}B\binom{k_1\ k_2}{\alpha\ \beta}{}_{\gamma}\,{}_{\epsilon})u_\alpha(k_1)u_\beta(k_2)s_{\gamma\epsilon}+\tfrac{1}{6}B\binom{k_1\ k_2\ k_3}{\alpha\ \beta\ \gamma})u_\alpha(k_1)u_\beta(k_2)u_\gamma(k_3)\ . \quad (1)$$

Here and below, the Greek letters α,β, etc., denote Cartesian components and the summation is implied over repeated Cartesian and and other indices. From this expression one can immediately obtain the symmetry properties of the coefficients, which arise because the strain tensor $\underline{s}$ is symmetric, and because the coefficients are really the derivatives of the strain energy with respect to internal and macroscopic strain components, where the order of differentiation is immaterial. The detailed expressions for the coefficients as lattice sums involving derivatives of the two-body and the three-body potential functions will be given in Secs. III and IV. In Sec. II we shall derive general expressions for the second- and third-order elastic constants in terms of these coefficients. The concluding section is devoted to general discussions on the theory.

II. EXPRESSIONS FOR ELASTIC CONSTANTS

Under homogeneous deformation the kth particle in the lth cell will be displaced from the original position $\vec{r}(lk)$ to[1]

$$\vec{R}(lk)=\vec{r}(lk)+\underline{v}\,\vec{r}(lk)+\vec{u}(k)\ , \tag{2a}$$

where $\underline{v}$ is the symmetric infinitesimal strain tensor, related to the Lagrangian strain tensor $\underline{s}$ by the equation

$$\underline{s}=\underline{v}+\tfrac{1}{2}\underline{v}\,\underline{v} \tag{2b}$$

or

$$\underline{v}=\underline{s}-\tfrac{1}{2}\underline{s}\,\underline{s}+\cdots\ , \tag{2c}$$

where $\cdots$ represents terms of higher order in $\underline{s}$. The internal strain can be expressed in terms of Lagrangian strain following Srinivasan,[2] by the use of the equilibrium condition $\partial E/\partial u_\alpha(k)=0$. There it has been proved that also for third-order elastic constants, it suffices to know internal strain up to the first order in Lagrangian strain. Thus one has to solve the equation

$$A\binom{k}{\alpha}\binom{k_2}{\beta}u_\beta(k_2)+A\binom{k}{\alpha}\binom{}{\beta\gamma}s_{\beta\gamma}=0\ ,$$

and obtain the solution in the form

$$u_\alpha(k)=G\binom{k}{\alpha}\binom{}{\beta\gamma}s_{\beta\gamma}\ .$$

We now substitute this solution in Eq. (1) and compare the resulting expression with the equation[5]

$$E=\tfrac{1}{2}c_{\alpha\beta\gamma\epsilon}s_{\alpha\beta}s_{\gamma\epsilon}+\tfrac{1}{6}c_{\alpha\beta\gamma\epsilon\zeta\eta}s_{\alpha\beta}s_{\gamma\epsilon}s_{\zeta\eta}\ ,$$

which defines the second- and third-order Brugger elastic constants c. The final result is

$$c_{\alpha\beta\gamma\epsilon}=A(\alpha\beta\gamma\epsilon)+\left[A\binom{k_0}{\theta}\binom{}{\alpha\beta}G\binom{k_0}{\theta}\binom{}{\gamma\epsilon}\right]_{\alpha\beta,\gamma\epsilon}\ ,$$

$$c_{\alpha\beta\gamma\epsilon\zeta\eta}=B(\alpha\beta\gamma\epsilon\zeta\eta)+3\left[B\binom{k_0}{\theta}\binom{}{\gamma\epsilon\zeta\eta}G\binom{k_0}{\theta}\binom{}{\alpha\beta}+B\binom{k_1k_2}{\theta\lambda}\binom{}{\alpha\beta}G\binom{k_1}{\theta}\binom{}{\gamma\epsilon}G\binom{k_2}{\lambda}\binom{}{\zeta\eta}\right]_{\alpha\beta,\gamma\epsilon,\zeta\eta}$$
$$+B\binom{k_1k_2k_3}{\theta\lambda\mu}G\binom{k_1}{\theta}\binom{}{\alpha\beta}G\binom{k_2}{\lambda}\binom{}{\gamma\epsilon}G\binom{k_3}{\mu}\binom{}{\zeta\eta}\ . \tag{3}$$

Here (and below) subscripts to thick square brackets mean that the expression within brackets is to be symmetrized with respect to interchanges between the suffix indices, including proper numerical factor. Thus,

$$[f(m,n,p)]_{m,n,p}\equiv\tfrac{1}{3}[f(m,n,p)+f(n,m,p)+f(p,n,m)]\ ,$$

where m,n,p may stand either for a single index or for a pair of indices, and f is a function involving them.

Similar expressions for elastic constants have also been obtained by Fuller and Naimon (Ref. 6, Appendix A); they have used, however, the quantity $u_\alpha(k)+v_{\alpha\beta}u_\beta(k)$ in place of $u_\alpha(k)$ [see Eq. (B4) of Ref. 6].

III. EXPRESSIONS FOR THE COEFFICIENTS FOR TWO-BODY INTERACTION

In this section we shall obtain expressions for the coefficients in terms of two-body central interaction, following Born and Huang.[1] We switch over to a new variable $y=R^2$, where $\vec{R}=\vec{R}(l'k')-\vec{R}(lk)=\vec{R}(l'k',lk)$ (for example), which, according to Eqs. (2a) and (2b), changes due to homogeneous deformation by

$$\Delta y=2r_\alpha r_\beta s_{\alpha\beta}+2r_\alpha v_{\alpha\beta}[u_\beta(k')-u_\beta(k)]$$
$$+2r_\alpha[u_\alpha(k')-u_\alpha(k)]+[u_\alpha(k')-u_\alpha(k)]^2\ , \tag{4}$$

where $\vec{r}$ stands for $\vec{r}(l'k',lk)$. The right-hand side can be written in terms of Lagrangian and internal strain only, by expressing $\underline{v}$ in terms of $\underline{s}$ from Eq. (2c).

The atom-atom interaction energy per unit volume is $(1/2v_0)\sum_{kl'k'}\phi$, where v_0 is the volume of unstrained unit lattice cell, and ϕ is the interaction potential between the particles (lk) and $(l'k')$. The Taylor expansion of strain-energy density (two-body part) about the initial configuration gives

$$E=\frac{1}{2v_0}\sum_{kl'k'}[\phi'\Delta y+\tfrac{1}{2}\phi''(\Delta y)^2+\tfrac{1}{6}\phi'''(\Delta y)^3]\ , \tag{5}$$

where the primes over ϕ indicate derivatives with respect to y, evaluated at initial configuration. The self-term $(lk)=(l'k')$ is to be omitted in the summation of Eq. (5) and in every similar case hereafter.

The procedure is now straightforward, although some lengthy calculation is necessary. Substituting for Δy from Eq. (4) in Eq. (5), one can express E in powers of $\underline{s}$ and $\vec{u}$. The coefficients of the first power of $\underline{s}$ and $\vec{u}$ should be zero becauses of the equilibrium condition, and then we obtain an expression for E of the form of Eq. (1). Comparing the two expressions one can immediately write down the coefficients in terms of the potential derivatives. The coefficients A are the same as those given by Born and Huang,[1] and the remaining ones are given below:

$$B(\alpha\beta\gamma\epsilon\zeta\eta)=(4/v_0)\sum_{kl'k'} r_\alpha r_\beta r_\gamma r_\epsilon r_\zeta r_\eta \phi''' ,$$

$$B(\overset{k}{\alpha}\beta\gamma\epsilon\zeta)=-(8/v_0)\sum_{l'k'}(r_\alpha r_\beta r_\gamma r_\epsilon r_\zeta \phi'''+[[\delta_{\alpha\beta} r_\gamma r_\epsilon r_\zeta \phi'']_{\beta,\gamma}]_{\beta\gamma,\epsilon\zeta}) ,$$

$$B(\overset{k_1\ k_2}{\alpha\ \beta}\gamma\epsilon)=(8/v_0)\left[\delta_{k_1k_2}\sum_{\substack{l'k'\\ k=k_1}} - \sum_{\substack{l'\\ k=k_1\\ k'=k_2}}\right](r_\alpha r_\beta r_\gamma r_\epsilon \phi'''+\tfrac{1}{2}\delta_{\alpha\beta} r_\gamma r_\epsilon \phi''+[[\delta_{\alpha\gamma} r_\beta r_\epsilon \phi'']_{\gamma,\epsilon}]_{\alpha,\beta}) ,$$

$$B(\overset{k_1\ k_2\ k_3}{\alpha\ \beta\ \gamma})=(12/v_0)\left[\left[\delta_{k_2k_3}\sum_{\substack{l'\\ k=k_3,k'=k_1}}\right]_{k_1,k_2,k_3} - \tfrac{1}{3}\delta_{k_1k_2}\delta_{k_2k_3}\sum_{\substack{l'k'\\ k=k_1}}\right](2r_\alpha r_\beta r_\gamma \phi'''+[3\delta_{\beta\gamma} r_\alpha \phi'']_{\alpha,\beta,\gamma}) .$$

IV. CONTRIBUTION TO THE COEFFICIENTS FROM THREE-BODY INTERACTION

Effective three-body interactions are of considerable importance in the study of many ionic solids. Examples of some very widely studied forms of three-body interaction for the case of ionic solids, are the one due to Lundqvist[7]

$$\psi=z'z''[(1/R)f(R')+(1/R')f(R)] ,$$

and the one obtained from the deformable shell model[8]

$$\psi=Df(R,R') .$$

Here the notation is as follows. ψ is the interaction potential between the particles $(l'k')$ and $(l''k'')$ mediated by a third particle (lk); f is a short-range overlap function; z',z'' are ionic charges of the particles k',k'', respectively; D is a constant, characteristic of the particle k, and $\vec{\mathrm{R}}'$ is $\vec{\mathrm{R}}(l''k'',lk)$. One should note that both of these interactions are covered by a general form $\psi=\psi(R,R')$, which is symmetric under R,R' exchange. It is this type of three-body interaction only, to which we shall be confined hereafter. We thus exclude, for example, the Keating potential used in covalent crystals, which is a function, also of the angle between $\vec{\mathrm{R}}$ and $\vec{\mathrm{R}}'$:

$$\psi=K(\vec{\mathrm{R}}\cdot\vec{\mathrm{R}}'-\vec{\mathrm{r}}\cdot\vec{\mathrm{r}}')^2 ,$$

where K is a constant and $\vec{\mathrm{r}}'=\vec{\mathrm{r}}(l''k'',lk)$.

The procedure to be followed now is almost similar to that of the preceding section. The contribution of three-body interaction to the strain-energy density is

$$E^{(3)}=\frac{1}{2v_0}\sum_{kl'k'l''k''}[\psi(R,R')-\psi(r,r')] .$$

By Taylor-series expansion one obtains

$$E^{(3)}=\frac{1}{2v_0}\sum_{kl'k'l''k''}[2\psi_1\Delta y+\psi_{11}(\Delta y)^2+\psi_{12}(\Delta y)(\Delta y')+\tfrac{1}{3}\psi_{111}(\Delta y)^3+\psi_{112}(\Delta y)^2(\Delta y')] ,$$

where $y'=R'^2$, and the subscripts 1 and 2 to ψ indicate derivatives with respect to y and y', respectively.

The equilibrium condition when both two-body interaction ϕ and three-body interaction ψ are present will be

$$\sum_{\substack{l'k'\\ k=k_0}} r_\alpha\phi'+\left[\sum_{\substack{l'k'l''k''\\ k=k_0}} - \sum_{\substack{lkl''k''\\ k'=k_0}}\right]r_\alpha\psi_1=0 \quad \text{for all } k_0,\alpha ,$$

and

$$\sum_{kl'k'} r_\alpha r_\beta\phi'+\sum_{kl'k'l''k''} 2r_\alpha r_\beta\psi_1=0 \quad \text{for all } \alpha,\beta .$$

We get a total of $3(n-1)$ independent equations from the first condition, and six from the second. These determine the shape of the unit cell and the relative position of the n particles in the unit cell for the equilibrium configuration.

The final expressions for the coefficients are given below. The self-terms for which any two of (lk), $(l'k')$, $(l''k'')$ are the same should be excluded from every lattice sum, as usual. We have

$$A(\alpha\beta\gamma\epsilon)=(4/v_0)\sum_{kl'k'l''k''}(r_\alpha r_\beta r_\gamma r_\epsilon\psi_{11}+r_\alpha r_\beta r'_\gamma r'_\epsilon\psi_{12}) ,$$

$$A(\overset{k_0}{\alpha}\beta\gamma)=(4/v_0)\left[\sum_{\substack{lkl''k''\\ k'=k_0}} - \sum_{\substack{l'k'l''k''\\ k=k_0}}\right](r_\alpha r_\beta r_\gamma\psi_{11}+r_\alpha r'_\beta r'_\gamma\psi_{12}) ,$$

$$A(\overset{k_1\ k_2}{\alpha\ \beta})=(4/v_0)\{[\mathscr{A}]_{k,k'}(\delta_{\alpha\beta}\psi_1+2r_\alpha r_\beta\psi_{11})+2\mathscr{B} r_\alpha r'_\beta\psi_{12}\} ,$$

where

$$\mathscr{A}=\delta_{k_1k_2}\left[\sum_{\substack{l'k'l''k''\\k=k_1}}-\sum_{\substack{l'l''k''\\k=k_1,k'=k_2}}\right],$$

and

$$\mathscr{B}=\mathscr{A}+\sum_{\substack{lkl''\\k'=k_1\\k''=k_2}}-\sum_{\substack{l'k'l''\\k=k_2\\k''=k_1}},$$

$$B(\alpha\beta\gamma\epsilon\zeta\eta)=(8/v_0)\sum_{kl'k'l''k''}\{r_\alpha r_\beta r_\gamma r_\epsilon r_\zeta r_\eta\psi_{111}+[3r'_\alpha r'_\beta r_\gamma r_\epsilon r_\zeta r_\eta\psi_{112}]_{\alpha\beta,\gamma\epsilon,\zeta,\eta}\},$$

$$B(\overset{k_0}{\alpha}\beta\gamma\epsilon\zeta)=(8/v_0)\left[\sum_{\substack{lkl''k''\\k'=k_0}}-\sum_{\substack{l'k'l''k''\\k=k_0}}\right]\{r_\alpha r_\beta r_\gamma r_\epsilon r_\zeta\psi_{111}+(r_\alpha r'_\beta r'_\gamma r'_\epsilon r'_\zeta+r_\alpha r_\beta r_\gamma r'_\epsilon r'_\zeta+r_\alpha r'_\beta r'_\gamma r_\epsilon r_\zeta)\psi_{112}$$
$$+[[\delta_{\alpha\beta}(r_\gamma r_\epsilon r_\zeta\psi_{11}+r_\gamma r'_\epsilon r'_\zeta\psi_{12})]_{\beta,\gamma}]_{\beta\gamma,\epsilon\zeta}\},$$

$$B(\overset{k_1\,k_2}{\alpha\;\beta}\gamma\epsilon)=(8/v_0)([\mathscr{A}]_{k,k'}\{2r_\alpha r_\beta r_\gamma r_\epsilon\psi_{111}+2r_\alpha r_\beta r'_\gamma r'_\epsilon\psi_{112}+[[2\delta_{\beta\epsilon}r_\alpha r_\gamma\psi_{11}]_{\alpha,\beta}]_{\gamma,\epsilon}$$
$$+\delta_{\alpha\beta}(r_\gamma r_\epsilon\psi_{11}+r'_\gamma r'_\epsilon\psi_{12})\}+[\mathscr{C}\{2r_\alpha r'_\beta r_\gamma r_\epsilon\psi_{112}+[\delta_{\beta\epsilon}r_\alpha r'_\gamma\psi_{12}]_{\gamma,\epsilon}\}]_{(\overset{k_1}{\alpha}),(\overset{k_2}{\beta})}),$$

where

$$\mathscr{C}=\delta_{k_1k_2}\sum_{\substack{l'k'l''k''\\k=k_1}}+\sum_{\substack{lkl'\\k'=k_1\\k''=k_2}}-\sum_{\substack{l'k'l''\\k=k_1\\k''=k_2}}-\sum_{\substack{l'l''k''\\k=k_2\\k'=k_1}},$$

$$B(\overset{k_1\,k_2\,k_3}{\alpha\;\beta\;\gamma})=(24/v_0)(\mathscr{D}\{\tfrac{1}{3}r_\alpha r_\beta r_\gamma\psi_{111}+\tfrac{1}{2}[\delta_{\beta\gamma}r_\alpha\psi_{11}]_{\alpha,\beta,\gamma}\}+[\mathscr{E}(r'_\alpha r_\beta r_\gamma\psi_{112}+\tfrac{1}{2}\delta_{\beta\gamma}r'_\alpha\psi_{12})]_{(\overset{k_1}{\alpha}),(\overset{k_2}{\beta}),(\overset{k_3}{\gamma})}),$$

where

$$\mathscr{D}=\delta_{k_1k_2}\delta_{k_2k_3}\left[\sum_{\substack{lkl''k''\\k'=k_1}}-\sum_{\substack{l'k'l''k''\\k=k_1}}\right]+\left[3\delta_{k_2k_3}\left[\sum_{\substack{l'l''k''\\k=k_2\\k'=k_1}}-\sum_{\substack{l'l''k''\\k=k_1\\k'=k_2}}\right]\right]_{k_1,k_2,k_3},$$

and

$$\mathscr{E}=-\delta_{k_1k_2}\delta_{k_2k_3}\sum_{\substack{l'k'l''k''\\k=k_1}}+\delta_{k_1k_2}\sum_{\substack{l'l''k''\\k=k_1\\k'=k_3}}+(\delta_{k_1k_3}-\delta_{k_2k_3})\sum_{\substack{l'l''k''\\k=k_1\\k'=k_2}}+2\left[\delta_{k_2k_3}\sum_{\substack{lkl'\\k'=k_2\\k''=k_1}}-\sum_{\substack{l'l''\\k=k_3\\k'=k_2\\k''=k_1}}\right]_{k,k'}.$$

V. DISCUSSIONS

Following Born and Huang,[1] one can immediately obtain some identities and symmetry relations that the coefficients satisfy. For the three-body interaction $A(\overset{k_0}{\alpha}\beta\gamma)$, and $B(\overset{k_0}{\alpha}\beta\gamma\epsilon\zeta)$ give zero when summed over k_0, whereas

$$A(\overset{k_1\,k_2}{\alpha\;\beta}),\ B(\overset{k_1\,k_2}{\alpha\;\beta}\gamma\epsilon),\ B(\overset{k_1\,k_2\,k_3}{\alpha\;\beta\;\gamma})$$

give zero when summed over two k_i indices. For two-body interaction, however, all the above coefficients give zero when summed over any one of the indices k_i. Furthermore, although in every coefficient we have guaranteed the symmetries mentioned just below Eq. (1) only, in the case of two-body interaction, they are automatically symmetric under the exchange of any two Cartesian or k_i indices. An exception is $B(\overset{k_1\,k_2}{\alpha\;\beta}\gamma\epsilon)$ which is symmetric in k_1,k_2, in α,β, and in γ,ϵ, but not in α,γ, etc. The three-body coefficients, however, do not have any such extra symmetry.

The expressions for the coefficients that we have obtained here, could also be worked out from the results of Srinivasan,[2] by expressing the force-constant matrix elements in terms of interatomic potential and imposing the equilibrium condition. This method will, however, be more laborious than the one followed here. The final expressions for the elastic constants as obtained here are quite different in form from those obtained by Srinivasan. Therefore a direct comparison between them is not easy for an arbitrary crystal structure.

We have not considered Coulomb interaction so far. For this interaction, some of the coefficients become indeterminate[1] and therefore depend on the surface for a finite crystal. To develop the theory consistently in this case, one has to write them in terms of the macroscopic electric field and the macroscopic strain, with surface independent coefficients. For the second-order elastic constant, this has been done by Dasgupta and Sengupta.[9] For the convergent coefficients one can use of course, the expressions deduced in Sec. III. In order to obtain the electrostatic contribution to elastic constants, one can also tackle the problem of indeterminacy by using the Ewald theta transformation, as has been done by Fuller and Naimon.[6]

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