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Theoretical multiple-scattering calculation of nonlinear elastic constants of disordered solids

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The multiple-scattering theory developed in various ways has significantly contributed to our understanding of the dielectric, mechanical, thermal, and other physical properties of disordered solids mainly in the linear-response regime. Its application has, however, been limited when both nonlinearity and disorder are present. In particular, very little work has been done to treat nonlinear elastic properties of disordered materials apart from the simple constant stress or strain averaging—an extreme approximation that is well known to lead to violation of equilibrium condition. Some of our earlier work goes beyond this simple approximation and provides a formal solution of the strain distribution including nonlinearity for an arbitrary disordered solid within the framework of the multiple-scattering theory. Following a similar approach, we propose in this work to develop a general theoretical framework and deduce explicit analytical expression for the third-order elastic constant, which is a generic quantity. As a specific application we have calculated the three independent third-order elastic constants for a particular type of disordered solid, namely, cubic polycrystals following two methods: a perturbative and an approximate self-consistent method developed in the present investigation. These methods have been applied to evaluate third-order elastic constants of eight different materials. The results seem to indicate that for low-anisotropy cases, the two methods give values that closely agree whereas for large anisotropy, they differ. In view of the scarcity of data on the third-order elastic constant of polycrystals, we have also calculated another important nonlinear parameter, namely, the pressure derivative of the second-order shear modulus, for which accurate measurements are available. In all cases, the calculated values are found to compare favorably with experiment. Lastly it may be mentioned that the method developed is quite general and may be adopted to treat nonlinearity in any tensor property of disordered materials.

I. INTRODUCTION

Recently, attention has been focused to study critically nonlinearities in the physical properties of different types of both crystalline and disordered materials. Although significant advancement of our understanding has been achieved with respect to disorder and nonlinearity separately, the situation becomes quite complex and interesting when both are important. However, progress in this field has been rather slow. This may be partly attributed to the lack of a suitable theoretical framework for analysis that is easy to apply and manipulate as well as to the inadequacy of experimental data in this area. Over the last few years, some works[1-3] on the nonlinear magnetic and dielectric susceptibilities of (mainly) composites have been reported. But so far as the nonlinear elastic behavior of such solids is concerned, there is little work[4] that attempts to go beyond simple averaging-type schemes. Following a similar but somewhat slightly different approach, we propose in the present investigation to develop a suitable theoretical framework to treat the nonlinear properties of disordered materials, of which the third-order elastic constant is the most important nonlinear mechanical susceptibility. In this section we outline the problem, indicate the method followed, and introduce the notation.

In what follows we shall consider the problem of the determination of effective elastic constants of an inhomogeneous material that consists of homogeneous phases. Under small deformation, stress (σ_{ij}) and strain (ε_{kl}) can be assumed to satisfy the nonlinear relation

$$\sigma_{ij}=C_{ijkl}\varepsilon_{kl}+\tfrac{1}{2}C_{ijklmn}\varepsilon_{kl}\varepsilon_{mn} , \tag{1}$$

where C_{ijkl} and C_{ijklmn} are the second-order elastic constant (SOEC) and the third-order elastic constant (TOEC), respectively. Now, if a similar relation holds for inhomogeneous materials as a whole in terms of averaged stress $\langle \sigma_{ij} \rangle$ and strain $\langle \varepsilon_{kl} \rangle$, namely,

$$\langle \sigma_{ij} \rangle = C^*_{ijkl}\langle \varepsilon_{kl} \rangle + \tfrac{1}{2}C^*_{ijklmn}\langle \varepsilon_{kl} \rangle \langle \varepsilon_{mn} \rangle , \tag{2}$$

then the problem is precisely to consider the effective SOEC C^*_{ijkl} and TOEC C^*_{ijklmn} of heterogeneous materials whose constituent's contribution from the nonlinear terms of Eq. (1) are not quite negligible.

Therefore, the problem essentially consists of the determination of the strain field in inhomogeneous materials under certain specified boundary conditions and then the performance of the averages. The problem of averaging is similar to the problem for inhomogeneous materials with contributions from linear terms only. But, in this case, the field-determining equilibrium equation

$$\frac{\partial \sigma_{ij}}{\partial r_j}=0 \tag{3}$$

is a nonlinear one and produces further difficulties.

Works done on the problem of estimating effective TOEC's of statistically homogeneous and isotropic random inhomogeneous materials in terms of the TOEC's and SOEC's of its constituent phases are mainly confined to use of a uniform strain or stress field,[5,6] which corresponds to Voigt- or Reuss-type averaging. However, these uniform solutions do not satisfy the equilibrium equation (3). Some authors[7,8] proposed, particularly for polycrystals, to equate three linear invariants of TOEC's of single crystals with that of a quasi-isotropic effective medium. This method also has the underlying assumption of homogeneous fields, and the results are identical with Voigt- (Reuss-) type averages for stiffness (compliance).

The problems of estimating the effective dielectric constant, electric conductivity, magnetic permeability, etc., are mathematically similar to this problem and are considerably simpler because they are all second-rank tensors for linear properties. Stroud and Hui[1] considered the problem of the dielectric constant for materials having a center of symmetry, in which, therefore, the first contribution beyond the linear term comes from cubic nonlinearity. In their work they simply neglected the contribution from any nonlinearities in the determination of electric fields and used the solution of linear problems to determine the effective nonlinear susceptibility. Zeng *et al.*[2] extended the method of Bergman[9] for the linear problem and made a representation of the effective linear dielectric constant and cubic susceptibility which effectively used a perturbation solution. Agarwal and Gupta[3] used a Green's-function approach and finally calculated linear and nonlinear susceptibilities in the single-grain *t*-matrix approximation.

The formal determination of the strain field in the presence of TOEC's has been considered by Middya, Basu, and Sengupta.[4] In this work we follow a similar but slightly different approach. We first present a brief outline of the Green's -function approach. In a true sense the problem is to be recast into another form so that it appears to be a problem of a linear medium with a contribution from nonlinear terms superposed as an effective force density. This approach has the same spirit of Zeller and Dederichs,[10] who include the inhomogeneity in the source terms, resulting in a form known variously as multiple-scattering theory, Green's-function approach, or effective-medium theory. Using this field solution, a perturbation-type expansion and close analog of the self-consistent (SC) effective elastic constant for cubic polycrystals have been derived. Section II contains the strain-field solution for an inhomogeneous medium with TOEC's using the Green's-function method. The determination of TOEC's using the perturbative approximation are presented for cubic polycrystals. Effective elastic constants up to third-order small terms have been calculated and are discussed in Sec. III. In Sec. IV an approximate SC *T*-matrix method of the solution has been provided. Useful relations between the pressure derivative (PD) of SOEC's and TOEC's for cubic materials presented in Sec. V are used to evaluate the effective pressure derivative of the SOEC in single-grain-scattering approximation. The results of the application of the above methods to different cubic polycrystals are discussed in Sec. VI, and the conclusions are summarized in Sec. VII.

II. FIELD SOLUTION IN THE GREEN'S-FUNCTION APPROACH

In an inhomogeneous material, SOEC $C_{ijkl}(\mathbf{r})$ and TOEC $C_{ijklmn}(\mathbf{r})$ vary from point to point and relate stress $\sigma_{ij}(\mathbf{r})$ to strain $\varepsilon_{ij}(\mathbf{r})$ at any point $\mathbf{r}$ by the relation

$$\sigma_{ij}(\mathbf{r})=C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})+\tfrac{1}{2}C_{ijklmn}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})\varepsilon_{mn}(\mathbf{r}) \,, \tag{4}$$

and effective elastic constants are defined through Eq. (2).

In a similar way, effective elastic constants can be defined in terms of compliance tensors through the equations

$$\varepsilon_{ij}(\mathbf{r})=S_{ijkl}(\mathbf{r})\sigma_{kl}(\mathbf{r})+\tfrac{1}{2}S_{ijklmn}(\mathbf{r})\sigma_{kl}(\mathbf{r})\sigma_{mn}(\mathbf{r}) \,, \tag{5}$$

$$\langle \varepsilon_{ij}\rangle=S^{*}_{ijkl}\langle\sigma_{kl}\rangle+\tfrac{1}{2}S^{*}_{ijklmn}\langle\sigma_{kl}\rangle\langle\sigma_{mn}\rangle \,, \tag{6}$$

where S_{ijkl} and S_{ijklmn} are compliance constants of second and third order, respectively, and corresponding starred quantities are effective second-order compliance constants (SOCC's) and third-order compliance constants (TOCC's). Second- and third-order stiffness and compliance constants are related by

$$S_{ijkl}C_{klmn}=\tfrac{1}{2}(\delta_{im}\delta_{jn}+\delta_{in}\delta_{jm})=I_{ijmn} \tag{7}$$

and

$$S_{ijklmn}=-S_{ijpq}S_{klrs}S_{mnuv}C_{pqrsuv} \,, \tag{8}$$

and I is a unit operator given by

$$I_{ijkl}=\tfrac{1}{2}(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) \,.$$

A similar relation also holds for the starred quantities. This definition of effective TOEC's based on the nonlinear stress-strain relation may differ from the definition of effective TOEC's from a Taylor-series expansion of strain energy.[5] However, in this paper we shall use the definition of Eqs. (2) and (6) only. To evaluate the effective elastic constants from Eqs. (2) and (6), one needs to know the distribution of strain field $\varepsilon_{kl}(\mathbf{r})$ under a given surface displacement.

Under a certain surface displacement, the strain field within such an inhomogeneous nonlinear medium was obtained earlier by Middya, Basu, and Sengupta[4] in a Green's-function approach. The solution is of the form

$$\varepsilon=\varepsilon^0+G\,\delta C_s\,\varepsilon+\tfrac{1}{2}G\,\delta C_t\,\varepsilon\varepsilon \,. \tag{9}$$

In the above equation, the tensor indices are suppressed, operator notation has been used, and subscripts s and t are introduced to distinguish between second- and third-order quantities, respectively. Derivation of Eq. (9), highlighting the tricks of using the Green's-function method for nonlinear materials together with the meaning of operator notation, is given in Appendix A. ε^0 is the homogeneous strain field under surface displacement in a homogeneous and isotropic medium of SOEC C_s^0, and δC_s is the fluctuation on C_s^0 and is given by

$$C_s=C_s^0+\delta C_s \,. \tag{10}$$

G is the familiar Green's function[10] for a homogeneous and isotropic medium C_s^0 and, for an infinite medium, is given by

$$G_{ijkl}(\mathbf{r})=\tilde{G}_{ijkl}\delta(\mathbf{r})+\frac{1}{r^3}\hat{G}_{ijkl}(\mathbf{r}/r) \,. \tag{11}$$

$\hat{G}_{ijkl}(\mathbf{r}/r)$ is a direction-dependent tensor, and the integral of it over all direction vanishes. $\tilde{G}_{ijkl}$ is a constant tensor and is given by[10]

$$\tilde{G}_{ijkl}=\tilde{G}_{12}\delta_{ij}\delta_{kl}+\tilde{G}_{44}(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) \qquad (12)$$

and

$$\tilde{G}_{12}=\frac{1}{15\mu^0}\,\frac{(3K^0+\mu^0)}{(3K^0+4\mu^0)}\ ,$$

$$\tilde{G}_{44}=\frac{3}{10\mu^0}\,\frac{(K^0+2\mu^0)}{(3K^0+4\mu^0)}\ ,$$

where K^0 and μ^0 are the bulk and shear moduli of the medium C_s^0. Now we define two operators F_s and F_t, of which F_s is a fourth-rank and F_t is a sixth-rank tensor, through

$$\varepsilon=F_s\langle\varepsilon\rangle+\tfrac{1}{2}F_t\langle\varepsilon\rangle\langle\varepsilon\rangle\ . \qquad (13)$$

In spite of the variation of the elastic constant within the inhomogeneous material, the average strain $\langle\varepsilon\rangle$ depending on the surface displacement remains the same.

Therefore,

$$\langle F_S\rangle=I \text{ and } \langle F_t\rangle=0\ . \qquad (14)$$

Using the definitions of Eqs. (1) and (2) and the field solution ε in the form of Eq. (13), the effective SOEC and TOEC are obtained as

$$C_s^*=\langle C_sF_s\rangle\ , \qquad (15)$$

$$C_t^*=\langle C_sF_t+C_tF_sF_s\rangle\ . \qquad (16)$$

Equation (15) was used earlier for the determination of the effective SOEC in the perturbative approach.[10] Equations (9) and (13) give two implicit equations for the determination of F_s and F_t in terms of C_s and C_t:

$$F_s-I=(G\,\delta C_s\,F_s-\langle G\,\delta C_s\,F_s\rangle)\ , \qquad (17)$$

$$F_t-(GC_tF_sF_s-\langle GC_tF_sF_s\rangle)=G\,\delta C_sF_t-\langle G\,\delta C_s\,F_t\rangle\ . \qquad (18)$$

Instead of using the strain-field solution in the form of Eq. (13), we can iterate Eq. (9) and obtain an explicit solution for ε as

$$\varepsilon=\varepsilon^0+GT_s\varepsilon^0+\tfrac{1}{2}GT_t\varepsilon^0\varepsilon^0\ , \qquad (19)$$

with

$$T_s=\delta C_s+\delta C_sGT_s=\delta C_s(I-G\,\delta C_s)^{-1} \qquad (20)$$

and

$$T_t=(I+GT_s)C_t(I+GT_s)(I+GT_s)\ . \qquad (21)$$

With these equations of T_s and T_t, which are actually an infinite series of δC_s, we get two equations for the effective SOEC and TOEC:

$$C_s^*=C_s^0+\langle T_s\rangle(I+\langle GT_s\rangle)^{-1} \qquad (22)$$

and

$$C_t^*=[I-(C_s^*-C^0)G]\langle T_t\rangle\times(I+\langle GT_s\rangle)^{-1}(I+\langle GT_s\rangle)^{-1}\ . \qquad (23)$$

Again, Eq. (22) is a known equation and is used extensively for the determination of the effective SOEC.[10] Equation (23) for the effective TOEC is new and resembles, in its appearance, the expression for the effective thermal expansion coefficient[11] in T-matrix form. Similarly, starting from Eqs. (5) and (6), we obtain the equilibrium stress solution under a given surface traction as

$$\sigma=\sigma^0+\Gamma\,\delta S_s\,\sigma+\tfrac{1}{2}\Gamma S_t\sigma\sigma\ , \qquad (24)$$

where σ^0 is the equilibrium stress under the same surface traction for a linear homogeneous medium of compliances S_s^0, and Γ is given by[10]

$$\Gamma=-C_s^0-C_s^0GC_s^0\ , \qquad (25)$$

where $C_s^0=(S_s^0)^{-1}$ and δS_s and S_s^0 are related to the SOCC $S_s(\mathbf{r})$ through

$$S_s(\mathbf{r})=S_s^0+\delta S_s(\mathbf{r})\ , \qquad (26)$$

of which S_s^0 is arbitrary within the limitation of positive definiteness. S_t is the TOCC. Similar to Eq. (13), the two operators E_s and E_t can be defined as

$$\sigma=E_s\langle\sigma\rangle+\tfrac{1}{2}E_t\langle\sigma\rangle\langle\sigma\rangle\ , \qquad (27)$$

where $\langle E_s\rangle=1$ and $\langle E_t\rangle=0$. E_s and E_t satisfy the equations similar to Eqs. (17) and (18) as

$$E_s-I=(\Gamma\,\delta S_s\,E_s-\langle\Gamma\,\delta S_s\,E_s\rangle)\ , \qquad (28)$$

$$E_t-(\Gamma S_tE_sE_s-\langle\Gamma S_tE_sE_s\rangle)=\Gamma\,\delta S_s\,E_t-\langle\Gamma\,\delta S_s\,E_t\rangle\ . \qquad (29)$$

The effective SOCC and TOCC come out to be

$$S_s^*=\langle S_sE_s\rangle\ , \qquad (30)$$

$$S_t^*=\langle S_sE_t+S_tE_sE_s\rangle\ . \qquad (31)$$

Direct iteration of Eq. (24) gives

$$\sigma=\sigma^0+\Gamma\,{}_rT_s\sigma^0+\tfrac{1}{2}\Gamma\,{}_rT_t\sigma^0\sigma^0\ , \qquad (32)$$

$${}_rT_s=\delta S_s+\delta S_s\,\Gamma\,{}_rT_s=\delta S_s(I-\Gamma\,\delta S_s)^{-1}\ , \qquad (33)$$

and

$${}_rT_t=(I+\Gamma\,{}_rT_s)S_t(I+\Gamma\,{}_rT_s)(I+\Gamma\,{}_rT_s)\ . \qquad (34)$$

Using this solution, two equations analogous to Eqs. (22) and (23) are obtained for the effective compliances:

$$S_s^*=S_s^0+\langle{}_rT_s\rangle(I+\langle\Gamma\,{}_rT_s\rangle)^{-1} \qquad (35)$$

and

$$S_t^*=[I-(S_s^*-S^0)\Gamma]\langle{}_rT_s\rangle\times(I+\langle\Gamma\,{}_rT_s\rangle)^{-1}(I+\langle\Gamma\,{}_rT_s\rangle)^{-1}\ . \qquad (36)$$

Equations (15), (16), (22), (23), (30), (31), (35), and (36) for the effective elastic constant, however, cannot be evaluated generally for random heterogeneous materials. In the next section, we evaluate the effective elastic constant under certain approximations.

III. EFFECTIVE ELASTIC CONSTANTS UP TO THIRD-ORDER SMALL TERMS

Iteration of Eqs. (17) and (18) yields a solution of F_s and F_t in the form of an infinite series involving δC_s and C_t. They are given by

$$F_s = I + G\,\delta C_s - \langle G\,\delta C_s\rangle + G\,\delta C_s\,G\,\delta C_s - \langle G\,\delta C_s\,G\,\delta C_s\rangle + \cdots \tag{37}$$

and

$$F_t = GC_t - G\langle C_t\rangle + 2GC_tG\,\delta C_s - 2G\langle C_tG\,\delta C_s\rangle + G\,\delta C_s\,GC_t - \langle G\,\delta C_s\,GC_t\rangle - G\,\delta C_s\,G\langle C_t\rangle \;. \tag{38}$$

We have considered here only up to the terms of second-order smallness in δC_s and C_t. Using these series (truncated after second-order terms), the strain-field solution is approximated by

$$\varepsilon = (I - G\,\delta C_s + G\,\delta C_s\,G\,\delta C_s)\varepsilon^0 + \tfrac{1}{2}(GC_t + G\,\delta C_s\,GC_t + 2GC_tG\,\delta C_s)\varepsilon^0\varepsilon^0 \;.$$

Using Eqs. (38) and (37) for F_s and F_t in Eqs. (15) and (16) and retaining terms up to third-order smallness in δC_s and C_t, we obtain the following expressions for the SOEC and TOEC:

$$C_s^* = \langle C_s\rangle + \langle \delta C_s\,G\,\delta C_s\rangle + \langle \delta C_s\,G\,\delta C_s\,G\,\delta C_s\rangle \tag{39}$$

and

$$C_t^* = \langle C_t\rangle + 3\langle \delta C_s\,GC_t\rangle + 6\langle \delta C_s\,G\,\delta C_sGC_t\rangle - 3\langle \delta C_s\,G\,\delta C_s\rangle G\langle C_t\rangle \;, \tag{40}$$

with $\delta C_s = C_s - \langle C_s\rangle$.

Equation (39) of the effective SOEC is a well-known perturbation expansion.[10] The terms of the right-hand side (RHS) of Eq. (40) are considered separately, and their expanded forms in terms of tensor indices are

$\langle C_t\rangle_{ijklmn}$ = Voigt average of third-order elastic constants,

$$3\langle \delta C_s\,GC_t\rangle_{ijklmn} = \langle (\delta C_s)_{ijop}G_{opqr}(C_t)_{qrklmn}\rangle + \langle (C_t)_{ijklop}G_{opqr}(\delta C_s)_{qrmn}\rangle + \langle (C_t)_{ijopmn}G_{opqr}(\delta C_s)_{qrkl}\rangle \;,$$

$$\begin{aligned} 6\langle \delta C_sG\,\delta C_sGC_t\rangle_{ijklmn} = {} & \langle (\delta C_sG)_{ijop}(\delta C_sG)_{opqr}(C_t)_{qrklmn}\rangle + \langle (C_t)_{ijklop}(G\,\delta C_s)_{opqr}(G\,\delta C_s)_{qrmn}\rangle \\ & + \langle (C_t)_{ijopmn}(G\,\delta C_s)_{opqr}(G\,\delta C_s)_{qrkl}\rangle + \langle (C_t)_{ijopqr}(G\,\delta C_s)_{opkl}(G\,\delta C_s)_{qrmn}\rangle \\ & + \langle (C_t)_{opqrmn}(G\,\delta C_s)_{opij}(G\,\delta C_s)_{qrkl}\rangle + \langle (\delta C_sG)_{ijop}(C_t)_{opklqr}(G\,\delta C_s)_{qrmn}\rangle \;, \end{aligned}$$

and

$$\begin{aligned} 3\langle \delta C_sG\,\delta C_s\rangle G\langle C_t\rangle_{ijklmn} = {} & \langle \delta C_sG\,\delta C_s\rangle_{ijop}G_{opqr}\langle C_t\rangle_{qrklmn} + \langle C_t\rangle_{ijklop}G_{opqr}\langle \delta C_sG\,\delta C_s\rangle_{qrmn} \\ & + \langle C_t\rangle_{ijopmn}G_{opqr}\langle \delta C_sG\,\delta C_s\rangle_{qrkl} \;. \end{aligned}$$

Evaluation of $\langle T_s\rangle$ and $\langle \mathbf{T}_t\rangle$ requires complete statistical information about $C_s(\mathbf{r})$ and $C_t(\mathbf{r})$. Therefore, with limited information it is not possible to use Eqs. (22) and (23) to compute the effective elastic constants.

In the single-grain scattering approximation, it is assumed that the contribution from terms involving the properties of two different grains is zero on the average. However, we shall neglect the contribution of these terms altogether. Equations (22) and (23) take the form

$$C_s^* = C_s^0 + \langle t_s\rangle(I + \langle Gt_s\rangle)^{-1} \tag{41}$$

and

$$C_t^* = [I - (C_s^* - C^0)G]\langle t_t\rangle(I + \langle Gt_s\rangle)^{-1} \times (I + \langle Gt_s\rangle)^{-1} \;, \tag{42}$$

where

$$t_s = \delta C_s + \delta C_sGt_s = \delta C_s(I - G\delta C_s)^{-1} \tag{43}$$

and

$$t_t = (I + Gt_s)C_t(I + Gt_s)(I + Gt_s) \;. \tag{44}$$

t_s and t_t thus defined for each grain are analogous to T_s and T_t within a grain, but zero outside it. This approximation physically corresponds to the neglect of intergranular scattering (interaction). Expansion of Eq. (42) up to third-order small terms in δC_s and C_t reproduces the same Eq. (40). The expression for effective compliances are similar to Eqs. (39)–(44); only G is to be replaced by Γ, δC_s by δS_s, and C_t by S_t. In the following we shall calculate explicitly the effective stiffness constants for cubic polycrystals.

The SOEC and TOEC of cubic crystals, whose orientation with respect to a reference frame is described by rotation matrix a_{ij}, can be expressed as

$$C_{ijkl} = C_{12}\delta_{ij}\delta_{kl} + C_{44}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + d\,a_{iu}a_{ju}a_{ku}a_{lu} \;, \tag{45}$$

$$C_t = C_{123}\delta^1 + C_{144}\delta^2 + C_{456}\delta^3 + d_1A^1 + d_2A^2 + d_3A^3 \;, \tag{46}$$

where δ^1, δ^2, and δ^3 are isotropic base tensors and A^1, A^2, and A^3 are sixth-rank base tensors, having Voigt symmetry, involving rotation matrix. Explicit expressions for δ's and A's are given in Appendix B. d is the anisotropy coefficient for the SOEC. d_1, d_2, and d_3 are the three anisotropy coefficients for the TOEC. For isotropic materials all these anisotropic coefficients are zero; they are given by

$$d = C_{11} - C_{12} - 2C_{44} , \tag{47}$$

$$d_1 = C_{111} - 3C_{112} + 2C_{123} + 12C_{144} - 12C_{166} + 16C_{456} , \tag{48}$$

$$d_2 = C_{112} - C_{123} - 2C_{144} , \tag{49}$$

$$d_3 = C_{166} - C_{144} - 2C_{456} . \tag{50}$$

The orientation averages of A^1, A^2, and A^3 for random orientation over all possible angles are simply

$$\langle A^1 \rangle_{ijklmn} = \tfrac{1}{35}(\delta^1 + \delta^2 + \delta^3)_{ijklmn} , \tag{51}$$

$$\langle A^2 \rangle_{ijklmn} = \tfrac{1}{5}(3\delta^1 + \delta^2)_{ijklmn} , \tag{52}$$

$$\langle A^3 \rangle_{ijklmn} = \tfrac{1}{5}(2\delta^2 + 3\delta^3)_{ijklmn} . \tag{53}$$

Using these results for averaging, we evaluate the terms of the RHS of Eq. (40) in terms of six independent constants of cubic crystals:

$$\langle C_t \rangle = C_{123}^V \delta^1 + C_{144}^V \delta^2 + C_{456}^V \delta^3 , \tag{54}$$

$$C_{123}^V = C_{123} + \frac{d_1}{35} + \frac{3d_2}{5} , \tag{55}$$

$$C_{144}^V = C_{144} + \frac{d_1}{35} + \frac{d_2}{5} + \frac{2d_3}{5} , \tag{56}$$

$$C_{456}^V = C_{456} + \frac{d_1}{35} + \frac{3d_3}{5} , \tag{57}$$

$$3\langle \delta C_s G C_t \rangle = 6\tilde{G}_{44} d (A_1 \delta^1 + A_2 \delta^2 + A_3 \delta^3) , \tag{58}$$

$$A_1 = -4(d_1 + 7d_2 + 4d_3)/175 , \tag{59}$$

$$A_2 = (2d_1 + 42d_2 - 20d_3)/525 , \tag{60}$$

$$A_3 = (d_1 + 11d_3)/175 , \tag{61}$$

and

$$6\langle \delta C_s G\, \delta C_s G C_t \rangle - 3\langle \delta C_s G\, \delta C_s \rangle G \langle C_t \rangle = 12(\tilde{G}_{44})^2 d^2 (B_1 \delta^1 + B_2 \delta^2 + B_3 \delta^3) , \tag{62}$$

$$B_1 = 2(15C_{144}^V - 17C_{144})/25 + 8(7C_{456}^V - 9C_{456})/175 , \tag{63}$$

$$B_2 = (5C_{144}^V - 3C_{144})/25 + 4(21C_{456}^V - 19C_{456})/525 - \frac{8d_1}{525} - \frac{102d_3}{525} , \tag{64}$$

$$B_3 = 2(15C_{144}^V - 17C_{144})/25 + 2d_1/175 + 3d_3/525 . \tag{65}$$

To evaluate Eqs. (62)–(65), grain shapes are assumed to be spherical, for simplicity. It is to be noted that the second-order small terms (A_1, A_2, A_3) in Eqs. (59)–(61) are functions of the anisotropy coefficients d, d_1, d_2, and d_3 only, whereas the third-order small terms contain C_{144} and C_{456} beside d_1, d_2, and d_3.

Exactly similar results are obtained for effective compliances, only we have to replace

d by $d' = S_{11} - S_{12} - 2S_{44}$,

d_1 by $d_1' = S_{112} - 3S_{123} + 12S_{144} + 2S_{123} - 12S_{166} + 16S_{456}$,

d_2 by $d_2' = S_{112} - S_{123} - 2S_{144}$,

d_3 by $d_3' = S_{166} - S_{144} - 2S_{456}$,

and C and G by S, and Γ, respectively.

Now it is easy to calculate the effective compliances, and inverting these values according to Eq. (8), we get the values for the effective stiffness constants.

Single-crystal SOEC's and TOEC's for some cubic crystals are collected in Table I. Using Eq. (40), C_t^* is calculated with these single-crystal elastic constants and its analog is derived for effective compliances S_t^* and are presented in Tables II and III, respectively. In the last inversion of S_t^* by Eq. (8), the value of the effective SOEC C_s^* is assumed to be the value under the same field solutions, which are, in general, found to be quite close to experimental results.

TABLE I. Single-crystal SOEC's and TOEC's for some cubic crystals (in 10^{11} dyn cm^{-2}). Data in this table are collected from Barsch (Ref. 5) and Simmons and Wang (Ref. 14).

Material	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
Ag	12.22	9.07	4.54	−84.30	−52.90	18.90	5.60	−62.60	8.30
Au	19.29	16.38	4.15	−172.90	−92.20	−23.30	−1.30	−64.80	−1.20
Cu	16.61	11.99	7.56	−127.10	−81.40	−5.00	−0.30	−78.00	−9.50
MgO	29.71	9.54	15.61	−489.50	−9.50	−6.90	11.30	−65.90	14.70
Nb	2.46	1.39	0.29	−25.30	−11.60	−4.79	−3.20	−1.77	1.27
Si (1)	16.58	6.39	7.96	−82.50	−45.10	−6.40	1.20	−31.00	−7.00
Si (2)	16.58	6.39	7.96	−74.40	−41.80	0.20	2.90	−31.50	−7.00
Columbium[a]	2.47	1.33	0.28	−25.64	−11.40	−4.67	−3.43	−1.68	1.37
Al (1)	1.07	0.61	0.28	−10.76	−3.15	0.36	−0.23	−3.40	−0.30
Al (2)	1.07	0.61	0.28	−12.24	−3.73	0.25	−0.64	−3.68	−0.27

[a] Columbium and niobium are the same material. We use the two different names to indicate the two different sources of data.

TABLE II. Effective TOEC (in 10^{11} dyn cm^{-2}) and the PD of the SOEC for cubic polycrystals (from stiffness).

						PD of shear modulus		
Material	Order	C_{44}^*	C_{123}^*	C_{144}^*	C_{456}^*	Present calc.	Calc. of Ref. 12	Expt. of Ref. 15
Ag	1	3.35	−0.17	−14.59	−12.45	1.97	1.42	1.40
	2	3.06	−4.40	−13.71	−9.69	1.53		
	3	3.04	0.38	−16.95	−7.78	1.60		
Au	1	3.07	−40.23	−16.15	−15.01	1.02	1.09	1.05
	2	2.82	−42.64	−16.06	−12.48	0.83		
	3	2.80	−38.72	−18.55	−11.27	0.88		
Cu	1	5.46	−25.12	−13.58	−19.36	1.78	1.36	1.35
	2	4.89	−28.91	−14.19	−13.74	1.28		
	3	4.83	−22.91	−18.18	−11.49	1.36		
MgO	1	13.40	−2.40	−16.76	−29.64	2.19	2.45	2.42
	2	13.12	−3.60	−13.85	−34.84	2.43		
	3	13.11	−2.81	−14.02	−35.33	2.49		
Nb	1	0.39	−4.95	−3.64	0.69	0.48		
	2	0.37	−5.03	−3.66	0.82	0.39		
	3	0.37	−4.91	−3.71	0.82	0.43		
Si (1)	1	6.81	−21.80	−5.52	−8.78	0.53	0.46	0.45
	2	6.67	−21.21	−6.35	−7.60	0.46		
	3	6.66	−20.62	−6.72	−7.41	0.47		
Si (2)	1	6.81	−18.42	−4.76	−9.18	0.50		
	2	6.67	−17.50	−5.82	−7.81	0.43		
	3	6.66	−16.86	−6.24	−7.59	0.44		
Cb	1	0.40	−4.59	−3.80	0.78	0.53		
	2	0.37	−4.65	−3.84	0.96	0.43		
	3	0.37	−4.49	−3.92	0.95	0.48		
Al (1)	1	0.26	−0.54	−0.94	−0.91	1.70	1.82	1.80
	2	0.26	−0.54	−0.94	−0.89	1.67		
	3	0.26	−0.53	−0.95	−0.89	1.68		
Al (2)	1	0.26	−0.47	−1.28	−0.87	2.07		
	2	0.26	−0.48	−1.28	−0.85	2.04		
	3	0.26	−0.47	−1.28	−0.85	2.05		

IV. APPROXIMATE SELF-CONSISTENT SOLUTION

A close analog of the SC calculation for SOEC's can be obtained for nonlinear elasticity by explicitly imposing the constraint $C_s^0=C_s^*$ on the choice of C_s^0. It must be mentioned that in the present form a proper SC approach cannot be done, as one must restrict the choice of the scattering medium to be a linear one so that the field solution in the Green's-function approach may be possible.

In this approximation the constraint of equality of the linear part of the scattering medium,

$$C_s^0=C_s^* \ , \tag{66}$$

together with Eqs. (22) and (23) produce the following two equations for C_s^* and C_t^*:

$$\langle T_s \rangle = 0 \ , \tag{67}$$

an implicit equation for the determination of C_s^*, and

$$C_t^* = \langle T_t \rangle \ , \tag{68}$$

an explicit equation for C_t^*.

Calculations of C_s^* and C_t^* are presented in Table IV for cubic polycrystals using the following expressions:

$$T_t = (I+GT_s)C_t(I+GT_s)(I+GT_s) \ , \tag{69}$$

$$(I+GT_s)_{ijkl} = X\,\delta_{ij}\delta_{kl} + Y(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) + Z a_{iu}a_{ju}a_{ku}a_{lu}) \ . \tag{70}$$

a_{ij} is the rotation matrix describing the orientation of the crystallite with respect to a reference frame. X, Y, and Z are defined as

TABLE III. Effective TOEC (in 10^{11} dyn cm^{-2}) and the PD of the SOEC for cubic polycrystals (from compliances).

Material	Order	C^*_{44}	C^*_{123}	C^*_{144}	C^*_{456}	PD of shear modulus
Ag	1	2.59	21.71	−33.35	5.13	1.53
	2	3.05	−24.53	−0.05	−17.78	1.24
	3	2.99	8.70	−22.87	−3.82	1.66
Au	1	2.38	−18.08	−33.13	−1.71	0.99
	2	2.82	−60.80	−4.70	−17.61	0.57
	3	2.76	−31.84	−23.09	−8.83	0.96
Cu	1	3.96	5.57	−40.05	5.67	1.30
	2	4.86	−59.52	6.33	−25.47	0.92
	3	4.72	−9.58	−27.63	−5.25	1.44
MgO	1	12.80	−1.90	−11.23	−42.63	2.92
	2	13.12	−2.98	−14.50	−34.06	2.42
	3	13.11	−2.97	−13.76	−35.74	2.51
Nb	1	0.36	−4.65	−3.91	0.96	0.43
	2	0.37	−5.05	−3.64	0.81	0.39
	3	0.37	−4.93	−3.70	0.82	0.42
Si (1)	1	6.50	−17.83	−8.94	−5.56	0.45
	2	6.67	−21.58	−6.11	−7.71	0.45
	3	6.67	−20.45	−6.84	−7.35	0.47
Si (2)	1	6.50	−13.58	−8.83	−5.45	0.42
	2	6.67	−17.91	−5.56	−7.94	0.42
	3	6.67	−16.67	−6.37	−7.51	0.45
Cb	1	0.35	−4.13	−4.18	1.13	0.49
	2	0.37	−4.69	−3.82	0.94	0.43
	3	0.37	−4.53	−3.90	0.96	0.46
Al (1)	1	0.26	−0.50	0.98	−0.86	1.66
	2	0.26	−0.55	−0.94	−0.89	1.67
	3	0.26	−0.53	−0.95	−0.89	1.68
Al (2)	1	0.26	−0.45	−1.31	−0.82	2.03
	2	0.26	−0.49	−1.27	−0.85	2.04
	3	0.26	−0.47	−1.28	−0.85	2.05

TABLE IV. Effective TOEC (in 10^{11} dyn cm^{-2}) and the PD of the SOEC for cubic polycrystals in an approximate self-consistent calculation ($C_s^* = C_s^0$).

Material	C^*_{44}	C^*_{123}	C^*_{144}	C^*_{456}	PD of shear modulus
Ag	3.02	1.50	−17.77	−7.19	1.60
Au	2.79	−37.83	−19.16	−10.91	0.89
Cu	4.79	−21.19	−19.45	−10.58	1.36
MgO	13.11	−2.83	−13.95	−35.48	2.50
Nb	0.37	−4.92	−3.71	0.82	0.43
Si (1)	6.66	−20.56	−6.77	−7.39	0.47
Si (2)	6.66	−16.80	−6.28	−7.57	0.45
Cb[a]	0.37	−4.51	−3.91	0.95	0.47
		(−4.85±1.2)[b]	(−3.70±0.02)[b]	(0.75±0.005)[b]	
Al (1)[c]	0.26	−0.53	−0.95	−0.89	1.68
Al (2)[c]	0.26	−0.47	−1.28	−0.85	2.05
		(−3.89±0.168)[b]	(−1.24±0.93)[b]	(−0.863±0.011)[b]	

[a] Data for columbium are taken from Reddy (Ref. 16).

[b] Data within parentheses are experimental values.

[c] Data for aluminum are taken from Graham, Naddler, and Chang (Ref. 17).

$$X=(I+GT_s)_{12}=-\frac{2\tilde{G}_{44}[d+2(C_{44}-C_{44}^*)]}{1-2\tilde{G}_{44}[d+2(C_{44}-C_{44}^*)]}\ , \tag{71}$$

$$Y=(I+GT_s)_{44}=-\frac{1}{1-4\tilde{G}_{44}(C_{44}-C_{44}^*)}\ , \tag{72}$$

$$\begin{aligned}Z&=(I+GT_s)_{11}-(I+GT_s)_{12}-2(I+GT_s)_{44}\\&=2d\tilde{G}_{44}/\{[1-4\tilde{G}_{44}(C_{44}-C_{44}^*)-2d\tilde{G}_{44}][1-4\tilde{G}_{44}(C_{44}-C_{44}^*)]\}\ .\end{aligned} \tag{73}$$

T_t can be written as

$$\begin{aligned}T_t=&[(2Y+Z)^3C_{123}+3X(2Y+Z)(3C_{123}+4C_{144}+2d_2+3(9C_{123}+18C_{144}+8C_{456}+d_1+9d_2+12d_3)\\&+(X+2Y+Z)]\delta^1+4Y^2[(2Y+Z)C_{144}+X(3C_{123}+4C_{144}+2d_2)]\delta^2\\&+8Y^3\delta^3[8Y^3d_1+12Y^2Z(d_1+8d_3)+Z^2(Z+6Y)]+(d_1+12d_3+C_{456})A^1\\&\quad+[8Y^3d_2+4Y^2X(d_1+3d_2+8d_3)+(6C_{144}+8C_{456}+d_1+3d_2+12d_3)XZ(Z+4Y)\\&\qquad+4Y^2Z(3d_2+4C_{144})+(d_2+2C_{144})Z^2(Z+6Y)]A^2+[8Y^3d_3+4Y^2Z(d_3+2C_{456})]A^3\ .\end{aligned} \tag{74}$$

Using the average values of A^1, A^2, and A^3 in Eq. (74), one can easily find the effective third-order elastic constants.

V. EFFECTIVE PRESSURE DERIVATIVE OF THE SOEC

In this section we consider the estimation of another important nonlinear parameter for cubic polycrystals, namely, the effective PD of the SOEC, which involves a certain combination of TOEC's and SOEC's. The individual values of the TOEC obtained from the application of the methods developed in the previous sections are used to calculate the PD's. As there exist quite accurate measurements for these quantities, they provide an indirect test of the predictions according to theories. As the bulk modulus of cubic polycrystals is an invariant, there exists only one independent pressure derivative, the PD of the shear modulus, which we now evaluate.

PD's of the SOEC are related to SOEC's and some combination of TOEC's. Barch[5] gave a relation for cubic symmetry as

$$\frac{\delta C_{ijkl}}{\delta p}=-[(C_{ijkl}+C_{ijklmm})]/3K+D_{ijkl}\ , \tag{75}$$

where

$$D_{ijkl}=\delta_{ij}\delta_{kl}-\delta_{ik}\delta_{jl}-\delta_{il}\delta_{jk}\ . \tag{76}$$

K is the bulk modulus, which is an invariant for cubic polycrystals. So the PD of the bulk modulus would also be an invariant, and the particular contraction

$$C_{iikkmm}=(3C_{111}+6C_{112}+2C_{123}) \tag{77}$$

should be invariant, which is simply equal to the Voigt-averaged $C_t^V=\langle C_t\rangle$ and also to the Reuss-averaged $C_t^R=\langle S_t\rangle^{-1}$. Together with C_t^V and C_t^R, we have also calculated

$$\frac{\delta C_{44}}{\delta p}=C'_{44}=-(C_{44}+3C_{144}+4C_{456})/3K-1\ ,$$

in Table II. The experimental values are provided for comparison wherever available. Also, the result of C'_{44} is compared with the computation of Paul, Middya, and Basu[12] from direct differentiation of Eq. (22) in the self-consistent approximation.

In Tables II and III we have presented the calculations starting from Voigt- and Reuss-type averages, respectively. Order 1 represents Voigt- (Reuss-) type averages and order 2 and 3 are the calculations including second- and third-order correction terms.

Before commenting on the numerical results, we note that the A's of Eqs. (59)–(61) representing the second-order small terms depend only on the anisotropy coefficients d, d_1, d_2, and d_3, whereas in the third-order small terms C_{144} and C_{456} appear explicitly along with the anisotropy coefficients d_1, d_2, and d_3.

VI. APPLICATION AND RESULTS

As an application of perturbative and approximate self-consistent method, we have calculated the three independent third-order elastic constants and pressure derivative of the shear modulus of eight polycrystals, namely Ag, Au, Cu, MgO, SiO_2, Nb, Al, and Columbium. The SOEC and TOEC of the corresponding single crystals are given in Table I.

First, we discuss the results of the perturbation method. The three independent effective TOEC's namely, C_{123}^*, C_{144}^*, and C_{456}^* have been calculated. The other three may be derived in terms of these constants. In Tables II and III two sets of values starting with Voigt and Reuss initializations are presented. The contributions up to a different order of terms are indicated in different rows. As a check of the calculations, the invariant combination, namely, $C_{iijjkk}=(3C_{111}+6C+2C_{123})$ that appears in the pressure derivative of the bulk modulus for cubic polycrystal is computed and is found to remain constant with each order of perturbation. In Tables II and III order 1 represents Voigt- and Reuss-type averages of the TOEC. Orders 2 and 3 are values of the effective SOEC and TOEC including second- and third-order corrections on them. The two sets of results clearly indicate that for materials with low anisotropy [defined according to Barsch[5] as $\alpha=(|V-R|)/H$, V, R, and H denoting Voigt, Reuss, and Hill averages for a particular third-order elastic constant], namely, MgO to Al(2), both the calculations agree. But for the third-order elastic constants with high anisotropy ($\alpha > 1$), in particular for Cu, Ag, and Au, the two results are widely different. However, it must be recalled that for the effective TOEC, unlike the SOEC, the Voigt and Reuss averages do not represent any bounds on the actual values. Still, the results of the present perturbation calculation seem to indicate an empirical correlation between the magnitude of anisotropy and closeness of end results up to third order of perturbation, which will be a helpful indicator for the analysis of the data.

The reason for the wide difference between the two approximations, in particular for C^*_{123}, is not difficult to understand. The difference between the initial values, i.e., Voigt and Reuss averages, for these materials is large and the perturbation up to third order is not sufficient to bring about convergence. For better results for these constants, one has to extend the perturbation calculation to still higher orders. Alternatively, one may try a somewhat better initialization by choosing some suitable combination of Voigt and Reuss values, say, a Hill-type average, which may hopefully yield a better result. However, we do not proceed in this direction in this preliminary attempt. It may be noted here that the effective pressure derivatives of the shear modulus for the above three polycrystals calculated with two different initializations are much closer. It is because of the fact that in the PD of C^*_{44} TOEC's occur through a combination of the form $3C_{144}+4C_{456}$, which varies little even if the individual variation is strong.

Next, we discuss the results of the approximate self-consistent method, which is given in Table IV. As a check, we have calculated (not presented here) two sets of values starting from the stiffness constants as well as the compliance constants. The results are identical up to two decimal places. A comparison of the results in Tables II–IV clearly shows that for low anisotropy constants for all the polycrystals considered in the present investigation the perturbative calculation and approximate self-consistent calculation give results that compare closely. In the absence of any other guiding factors, this finding is important about the reliability of the predictions made, at least for the low-anisotropy cases. Finally, we discuss the different calculations in relation to experiment.

As was already mentioned, since we are unable to locate the range for any effective third-order elastic constant in the absence of any bound, it is difficult to make any comments on the different theoretical predictions made except insofar as how they directly compare with actual observation. From Tables II–IV it is quite clear that for the effective pressure derivative of the shear modulus for which data are available, in five cases the perturbative and self-consistent methods give results that agree quite closely with each other and also with observation. It may be noted in this connection that the present results are almost identical with those obtained by Paul, Middya, and Basu,[12] who have employed a totally different method for the calculation of the effective PD. Their approach consists in a direct differentiation of the effective SOEC [Eq. (39)] using experimental values of the PD of the individual second-order crystal elastic constants. This agreement provides an independent justification of the present methods developed and also indicates the reliability of the results obtained. Again, so far as the individual effective TOEC is concerned, the experimental data are available only for two cases, Columbium and Nb. In both cases the results obtained by the two methods agree quite favorably with each other and also with observation, and the predictions are well within experimental error. However, the range of error in observation in these two cases and the closeness of the predicted results, as the anisotropy is small, by the two methods do not indicate preference for either.

In the light of the results of the present investigation, the cases of Ag and Cu polycrystals are interesting. The Voigt and Reuss average calculations for the effective C_{123} and C_{456} of Ag and Cu suggest opposite signs (see Tables II and III). Again, both the perturbative and self-consistent approaches in the present work suggest $+ve$ sign for the effective C_{123} of Ag and $-ve$ sign for the effective C_{456} of both Ag and Cu. But the values predicted by the two methods for the C_{123} of Ag and Cu are widely different. In view of the lack of sufficient convergence in the case of C_{123} of Ag and Cu, it seems that the approximate self-consistent result is probably more accurate. However, it poses an interesting problem, and it will be highly instructive to perform measurements on the effective TOEC of Ag and Cu polycrystals which will ultimately decide between theories for such high-anisotropy cases.

VII. CONCLUSION

Finally, we summarize the main conclusions. In this work we have found the strain distribution including nonlinearity for a disordered material and, utilizing the same, have provided analytical expressions for the effective TOEC for cubic polycrystals that expressly go beyond simple constant stress- or strain-type averaging procedures. For eight cubic polycrystals, explicit numerical calculations have been done following two different methods of solution, namely, a perturbative and an approximate self-consistent approach, and the results obtained are satisfactory for low-anisotropy cases, but there remains problems with high-anisotropy cases. Further investigations, both experimental and theoretical, are necessary for resolving them. Last, it may be mentioned that the present method, although it has been developed with respect to the elastic property only for the TOEC, is quite straightforward for extending the method to treat nonlinearity in any other tensor property and in other disordered systems such as noncubic polycrystals and in multiphase composites, etc., as well as to compute still higher-order nonlinearities, some of which are planned to be considered in a future publication.

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APPENDIX A: DERIVATION OF EQUILIBRIUM STRAIN-FIELD SOLUTION

Let us consider a linear, homogeneous, and isotropic medium of the SOEC C^0_{ijkl}. An external force per unit volume $f_i(\mathbf{r})$ is applied at every point $\mathbf{r}$, and it is deformed under a specified surface displacement at its boundary.

Stress and strain within the medium are related by

$$\sigma_{ij}=C^0_{ijkl}\varepsilon_{kl}\ . \tag{A1}$$

Equilibrium strain distribution is governed by the equation

$$\frac{\partial\sigma_{ij}(\mathbf{r})}{\partial r_j}+f_i(\mathbf{r})=0\ . \tag{A2}$$

In terms of displacement $\mathbf{u}$, this equation appears to be

$$c^0_{ijkl}\frac{\partial^2 u_k}{\partial r_i\partial r_j}+f_i(\mathbf{r})=0\ . \tag{A3}$$

The solution for $u_k(\mathbf{r})$ can be written simply as

$$u_k(\mathbf{r})=u^0_k(\mathbf{r})+\int g_{kl}(\mathbf{r},\mathbf{r}')f_l(\mathbf{r}')d\mathbf{r}'\ , \tag{A4}$$

where $u^0_k(\mathbf{r})$ is the solution of (A3) with $f_i(\mathbf{r})=0$, under the given surface displacement. $g_{kl}(\mathbf{r},\mathbf{r}')$ is the displacement Green's function and satisfies the equation

$$C^0_{ijkl}\frac{\partial^2 g_{kp}}{\partial r_i\partial r_j}+\delta_{ip}\delta(\mathbf{r}-\mathbf{r}')=0\ , \tag{A5}$$

$$g_{kp}(\mathbf{r},\mathbf{r}')=g_{kp}(\mathbf{r}-\mathbf{r}')=g_{kp}(\mathbf{R})\ .$$

For infinite material and the boundary condition that $g_{kp}(\mathbf{R})$ vanishes at surface, $g_{kp}(\mathbf{R})$ is given by[10]

$$g_{kp}(\mathbf{R})=\frac{1}{8\pi\mu^0 R}\left[\frac{3K^0+7\mu^0}{3K^0+r\mu^0}\delta_{kp}+\frac{3K^0+\mu^0}{3K^0+4\mu^0}R_kR_p\right]\ , \tag{A6}$$

where $K^0=(C^0_{11}+2C^0_{12})/3$; $\mu^0=C^0_{44}$ are bulk and shear moduli of the medium C^0.

If $f_l(\mathbf{r})$ is now given in the form

$$f_l(\mathbf{r}')=\frac{\partial F_{ln}(\mathbf{r}')}{\partial r'_n}\ , \tag{A7}$$

then the solution of the field is obtained by differentiating (A4) and then integrating by parts in the form

$$\varepsilon_{ki}(\mathbf{r})=\varepsilon^0_{ki}+\int G_{kiln}(\mathbf{r},\mathbf{r}')F_{ln}(\mathbf{r}')d\mathbf{r}'\ , \tag{A8}$$

$$G_{kilm}(\mathbf{r},\mathbf{r}')=-\frac{1}{2}\left[\frac{\partial^2 g_{kl}(\mathbf{r},\mathbf{r}')}{\partial r_i\partial r'_m}+\frac{\partial^2 g_{il}(\mathbf{r},\mathbf{r}')}{\partial r_k\partial r'_m}\right]\ . \tag{A9}$$

In a nonlinear medium the superposition theorem is not valid, so that the Green's-function method cannot be applied directly. We consider an inhomogeneous nonlinear elastic material to be a linear homogeneous medium of the elastic constant C^0_{ijkl}, on which the inhomogeneity δC_{ijkl} and nonlinearity C_{ijklmn} are superposed such that

$$C_{ijkl}(\mathbf{r})=C^0_{ijkl}+\delta C_{ijkl}(\mathbf{r})\ . \tag{A10}$$

Now the stress-strain relation in this type of material is given by

$$\sigma_{ij}(\mathbf{r})=C^0_{ijkl}\varepsilon_{kl}(\mathbf{r})+\delta C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})+\tfrac{1}{2}C_{ijklmn}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})\varepsilon_{mn}(\mathbf{r})\ . \tag{A11}$$

The equilibrium equation (A2) with $f_i(\mathbf{r})=0$ now looks like

$$C^0_{ijkl}\frac{\partial^2 u_k}{\partial r_i\partial r_j}+\frac{\partial}{\partial r_j}\left[\delta C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})+\tfrac{1}{2}C_{ijklmn}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})\varepsilon_{mn}(\mathbf{r})\right]=0\ . \tag{A12}$$

Equation (A12) shows that we are now considering a medium of the elastic constant C^0_{ijkl} together with a force per unit volume appearing as a result of the inhomogeneity δC_{ijkl} and nonlinearity C_{ijklmn} of the material. Comparing (A3), (A7), and (A12), we can identify $F_{ij}(\mathbf{r})$ as

$$F_{ij}(\mathbf{r})=\delta C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})+\tfrac{1}{2}C_{ijklmn}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})\varepsilon_{mn}(\mathbf{r})\ . \tag{A13}$$

Therefore, the solution for the strain field is obtained as

$$\varepsilon_{kl}(\mathbf{r})=\varepsilon^0_{kl}+\int G_{klmn}(\mathbf{r},\mathbf{r}')[\delta C_{mnop}(\mathbf{r}')\varepsilon_{op}(\mathbf{r}')+\tfrac{1}{2}C_{ijklmn}(\mathbf{r}')\varepsilon_{kl}(\mathbf{r}')\varepsilon_{mn}(\mathbf{r}')]d\mathbf{r}'\ . \tag{A14}$$

This equation can be in the operator form

$$\varepsilon=\varepsilon^0+G\,\delta C_s\varepsilon+\tfrac{1}{2}GC_t\varepsilon\varepsilon\ . \tag{A15}$$

The subscripts s and t are introduced to distinguish between second- and third-order quantities. The product of the two operators X and Y should be understood as

$$(XY)_{ijmn}(\mathbf{r},\mathbf{r}')=\int X_{ijkl}(\mathbf{r},\mathbf{r}'')Y_{klmn}(\mathbf{r}'',\mathbf{r}')dr''\ . \tag{A16}$$

A similar meaning is associated with the product of operators of different rank. In Eq. (A15) the operators δC_s and C_t are obtained from the material properties δC_{ijkl} and C_{ijklmn} in the following prescription:

$$\delta C_{ijkl}(\mathbf{r},\mathbf{r}')=\delta C_{ijkl}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\ , \tag{A17}$$

$$C_{ijklmn}(\mathbf{r},\mathbf{r}')=C_{ijklmn}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\ . \tag{A18}$$

Physically, this implies that the nonlocality of C in the scale of inhomogeneity is negligibly small. The reverse prescription for returning from operator to material property is provided by relations of the type

$$\delta C_{ijkl}(\mathbf{r})=\int \delta C_{ijkl}(\mathbf{r},\mathbf{r}')d\mathbf{r}' \, , \tag{A19}$$

$$C_{ijklmn}(\mathbf{r})=\int C_{ijklmn}(\mathbf{r},\mathbf{r}')d\mathbf{r}' \, . \tag{A20}$$

APPENDIX B

In Eq. (46) a short notation has been used for base tensors. In the following we write this explicitly:

$$\delta^{(1)}_{ijklmn}=\delta_{ij}\delta_{kl}\delta_{mn} \, ,$$

$$\delta^{(2)}_{ijklmn}=2(\delta_{ij}I_{klmn}+\delta_{kl}I_{ijmn}+\delta_{mn}I_{ijkl}) \, ,$$

where

$$I_{klmn}=\tfrac{1}{2}(\delta_{km}\delta_{ln}+\delta_{kn}\delta_{lm}) \, ,$$

$$\delta^{(3)}_{ijklmn}=2(\delta_{ik}I_{jlmn}+\delta_{il}I_{jkmn}+\delta_{im}I_{jnkl}+\delta_{in}I_{jmkl}) \, ,$$

$$A^{(1)}_{ijklmn}=a_{iu}a_{ju}a_{ku}a_{lu}a_{mu}a_{nu} \, ,$$

$$A^{(2)}_{ijklmn}=\delta_{ij}a_{ku}a_{lu}a_{mu}a_{nu}+\delta_{kl}a_{iu}a_{ju}a_{mu}a_{nu}+\delta_{mn}a_{iu}a_{ju}a_{ku}a_{lu} \, ,$$

$$\begin{aligned}A^{(3)}_{ijklmn}=&\,\delta_{ik}a_{ju}a_{lu}a_{mu}a_{nu}+\delta_{il}a_{ju}a_{ku}a_{mu}a_{nu}+\delta_{im}a_{ju}a_{ku}a_{lu}a_{nu}+\delta_{in}a_{ju}a_{ku}a_{lu}a_{mu}\\&+\delta_{jk}a_{iu}a_{lu}a_{mu}a_{nu}+\delta_{jl}a_{iu}a_{ku}a_{mu}a_{nu}+\delta_{jm}a_{iu}a_{ku}a_{lu}a_{nu}+\delta_{jn}a_{iu}a_{ku}a_{lu}a_{mu}\\&+\delta_{km}a_{iu}a_{ju}a_{lu}a_{nu}+\delta_{kn}a_{iu}a_{ju}a_{lu}a_{mu}+\delta_{lm}a_{iu}a_{ju}a_{ku}a_{nu}+\delta_{ln}a_{iu}a_{ju}a_{ku}a_{lu} \, .\end{aligned}$$

The orientation average of $A^{(1)}$ can be obtained from Wagniere[13] and is given by

$$\langle A^{(1)}_{ijklmn}\rangle=\tfrac{1}{35}(\delta^{(1)}_{ijklmn}+\delta^{(2)}_{ijklmn}+\delta^{(3)}_{ijklmn}) \, .$$

$\langle A^{(2)}\rangle$ and $\langle A^{(3)}\rangle$ [Eqs. (52) and (53)] can be obtained similarly by noting that

$$\begin{aligned}\langle a_{iu}a_{ju}a_{ku}a_{lu}\rangle&=\tfrac{1}{5}(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\\&=\tfrac{1}{5}\delta_{ij}\delta_{kl}+\tfrac{2}{5}I_{ijkl} \, .\end{aligned}$$

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