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Multiple scattering : a theoretic approach to nonlinear mechanical property of composites

Sudeshna Sarkar, Asis Kumar Ghorai[†] and T R Middya*

[†] Sripat Singh College, Jiaganj, Murshidabad, West Bengal, India
Condensed Matter Physics Research Centre, Department of Physics, Jadavpur University, Calcutta-700 032, India

E-mail trm@juphys.ernet.in

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Abstract In our previous work (Sudeshna Sarkar, T K Ballabh, T R Middya and A N Basu *Phys Rev* **B54**, 3926, 1996), the multiple scattering theory has been used to determine the effective third order elastic constants (TOEC) of polycrystals with constituent crystallites having noncubic symmetry The same theoretical approach has been utilised here to determine the effective nonlinear elastic constants of isotropic composites The results obtained have been applied to investigate the elastic percolation behaviour of a two-phase composite, one phase of which is void The behaviours of both second order and third order elastic constants and cross-over strain field near the percolation threshold have been studied

Keywords Multiple scattering theory, isotropic composites, elastic percolation behaviour

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1. Introduction

Study of the properties of inhomogeneous systems is an important branch both in physics and material science. Although the determination of the nonlinear tensorial properties of such systems has gathered much significance in recent years, limited works [1-5] have been reported in the case of nonlinear mechanical property and this may be due to the mathematical difficulty in treating higher rank elastic tensors.

In our previous work [6], the multiple scattering theory under the single-grain scattering approximation has been used to obtain the effective third order elastic constants (TOEC) of polycrystals having crystallites of noncubic symmetry. In this work, the same theoretical approach has been used to determine the effective TOEC of composites. It may be noted that unlike those of polycrystals, the phases of the composites are isotropic.

It is important to study the response of different physical behaviours of inhomogeneous media at and around the percolation threshold. In the case of linear properties, this behaviour is well studied but its extension in the nonlinear regime is rare. The present formalism has been utilised to obtain the percolation threshold both for linear and nonlinear elastic constants of a binary composite, one phase of which is void. The behaviours of second order elastic constants (SOEC) and TOEC near the percolation threshold have also been determined. In addition, the cross-over strain field near the percolation threshold has been studied.

In the next section, we give the analytical expressions for the effective SOEC and TOEC of a multi-phase composite. Section 3 deals with the percolation behaviour of effective SOEC and TOEC. The results obtained in Section 3 are used in Section 4 to study the crossover strain field near threshold. The conclusions of the present work are discussed in Section 5.

2. Outline of the theory

A composite medium consists of different isotropic phases and any physical property varies from one phase to another due to change in the nature of the material. Taking only first order nonlinearity, the local constitutive relation between stress $\sigma_{ij}(r)$ and strain $\varepsilon_{kl}(r)$ is

$$\sigma_{ij}(r) = C_{ijkl}(r)\varepsilon_{kl}(r) + \frac{1}{2} C_{ijklmn}(r)\varepsilon_{kl}(r)\varepsilon_{mn}(r), \quad (1)$$

* Corresponding Author.

where $C_{ijkl}(r)$ and $C_{ijklmn}(r)$ are the local SOEC and TOEC respectively. For the composite medium, the averaged stress and strain satisfy the relation

$$< \sigma_{ij}(r) > = C^*_{ijkl} < \varepsilon_{kl}(r) > + \frac{1}{2} C^*_{ijklmn} < \varepsilon_{kl}(r) > < \varepsilon_{mn}(r) >, \quad (2)$$

where C^*_{ijkl} and C^*_{ijklmn} are the effective SOEC and TOEC respectively and < > represents volume averaging. The problem is to determine C^*_{ijkl} and C^*_{ijklmn} in terms of the corresponding quantities of the constituent phases. Eqs. (1) and (2) imply that the strain field solution will be needed for this. The strain field is obtained from the equilibrium condition

$$\frac{\delta}{\delta r_i} \sigma_{ij} = 0. \quad (3)$$

The details of evaluation of the strain field are given in Ref. [5] and for brevity, we skip it here. In operator notation, the strain field can be written as [5]

$$\varepsilon = \varepsilon^0 + G\delta C_S \varepsilon + \frac{1}{2} GC_t \varepsilon\varepsilon, \quad (4)$$

where $\delta C_S \equiv \delta C_{ijkl}(r)$ and $C_t \equiv C_{ijklmn}(r)$. ε^0 is the homogeneous strain field under given surface displacement in the linear homogeneous medium of elastic constant C^0_S and $\delta C_S(r)$ is the fluctuation on C^0_S such that $C_S(r) = C^0_S + \delta C_S(r)$. This fluctuation coupled with the nonlinearity C_t are assumed to be embedded as inhomogeneity in the linear medium C^0_S. G is the strain Green function [7] and assuming spherical grains, the explicit expression for G is given as [6]

$$G_{ijkl} = \frac{\left[\left(C^0_{12} + C^0_{44}\right)\delta_{ij}\delta_{kl} - \left(3C^0_{12} + 8C^0_{44}\right)I_{ijkl}\right]}{15C^0_{44}\left(C^0_{12} + 2C^0_{44}\right)}, \quad (5)$$

where $I_{ijkl} = \frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right)$.

Now eq. (4) can be cast in an alternative form as

$$\varepsilon = \varepsilon^0 + GT_S\varepsilon^0 + \frac{1}{2} GT_t \varepsilon^0 \varepsilon^0 \quad (6)$$

with T matrices

$$T_S = \delta C_S + \delta C_S G T_S = \delta C_S (I - G\delta C_S)^{-1} \quad (7)$$

and

$$T_t = (I + GT_S) C_t (I + GT_S)(I + GT_S). \quad (8)$$

Using eqs. (1), (2) and (6), C^*_S and C^*_t are given as

$$C^*_S = C^0_S + < T_S > (I + < GT_S >)^{-1} \quad (9)$$

and

$$C^*_t = \left[I - (C^*_S - C^0_S) G\right] < T_t > (I + < GT_S >)^{-1} (I + < GT_S >)^{-1}. \quad (10)$$

In eqs. (9) and (10), C^0_S can be chosen arbitrarily without violating the mechanical stability of the medium. We adopt a self consistent scheme of calculation in which C^0_S is chosen to be equal to the effective medium whose elastic property we are to determine *i.e.*

$$C^0_S = C^*_S. \quad (11)$$

Using eq. (11) in (9), we get

$$< T_S > = 0. \quad (12)$$

Thus, the chosen scattering medium has the characteristics that average scattering over the whole medium vanishes.

For TOEC the self-consistency is only approximate because in the homogeneous medium, nonlinearity is absent. The so called self-consistent solution for TOEC is

$$C^*_t = < T_t >. \quad (13)$$

From eq. (12) and using eq. (7), we get for a composite having isotropic components

$$\sum v_i \frac{\left(\mu^{(i)} - \mu^*\right)}{5\mu^*(3K^* + 4\mu^*) + 6(K^* + 2\mu^*)(\mu^{(i)} - \mu^*)} = 0 \quad (14)$$

and

$$\sum_i v_i \frac{K^{(i)} - K^*}{3K^{(i)} + 4\mu^*} = 0 \quad (15)$$

with $\mu^{(i)} = C^{(i)}_{44}$ and $K^{(i)} = \frac{1}{2}\left(C^{(i)}_{11} + 2C^{(i)}_{12}\right)$, where $\mu^{(i)}$ and $K^{(i)}$ are the shear modulus and bulk modulus respectively of the i-th phase and μ^* and K^* are the corresponding effective modulus of the composite. v_i denotes the volume fraction of the i-th phase. For composite, we have to do averaging over phase volume fraction because material property varies from phase to phase, Eqs. (14) and (15) give the self-consistent results for the effective SOEC.

From eq. (13), the general expression for self-consistent C^*_t in terms of the effective SOEC and TOEC of the phases is given as

$$C^*_t = \Sigma_i\, v_i \left[\left\{3(X^{(i)})^2\, (X + 2Y)^{(i)} (3C_1 + 2C_2)^{(i)}\right.\right.$$

$$+12X^{(i)}(Y^{(i)})^2 C_1^{(i)}\big\}\delta^{(1)} + 4X^{(i)}(Y^{(i)})^2 C_2^{(i)}\delta^{(2)}$$

$$+8(Y^{(i)})^3 C_t^{(i)}\big], \tag{16}$$

where

$$X^{(i)} = \frac{3K^* + 4\mu^*}{3K^{(i)} + 4\mu^*}\left[\frac{2(K^* + 2\mu^*)(\mu^{(i)} - \mu^*) - 5\mu^*(K^{(i)} - K^*)}{5\mu^*(3K^* + 4\mu^*) + 6(K^* + 2\mu^*)(\mu^{(i)} - \mu^*)}\right] \tag{17}$$

$$Y^{(i)} = \frac{5\mu^*(3K^* + 4\mu^*)}{2\{5\mu^*(3K^* + 4\mu^*) + 6(K^* + 2\mu^*)(\mu^{(i)} - \mu^*)\}}, \tag{18}$$

$$C_1^{(i)} = 3C_{123}^{(i)} + 4C_{144}^{(i)}, \tag{19}$$

$$C_2^{(i)} = 3C_{144}^{(i)} + 4C_{456}^{(i)}, \tag{20}$$

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

and

$$\delta_{ijklmn}^{(2)} = 2\left(\delta_{ij}I_{klmn} + \delta_{kl}I_{ijmn} + \delta_{mn}I_{ijkl}\right).$$

Using appropriate tensor indices, the three effective TOEC *i.e.* C_{123}^*, C_{144}^* and C_{456}^* can be obtained from eq. (16). Thus from eqs. (14)-(16), we can obtain the effective SOEC and TOEC of a composite under the self-consistent scheme.

3. SOEC and TOEC of a percolative system

The elasticity of percolative systems (*e.g.* a two-phase composite of which one phase is void) is a very active field of research. The first study of the elastic behaviour of such a system near percolation threshold was probably due to P. G. de Gennes [8]. After him, several studies [9-12] on 2-d lattice models have been reported. The elastic constants are shown to go to zero as *p i.e.* the volume fraction of voids tends to p_c, the percolation threshold.

Two classes of elastic network models can be defined depending on the kind of elastic properties the individual bonds have. Firstly, we may mention a model of which the bonds that transmit all the elastic information and this includes the bond bending model [9]. A different model falling into the same class is the beam lattice model [13]. In these models, if the probability of having bonds present falls below the percolation threshold, the elastic moduli will be zero.

The second type includes bonds that transmit only partial elastic information [14]. Elasticity of central force systems [9] falls in this class. The variation of different elastic moduli near percolation threshold has been studied for this type of network.

Later Arbabi and Sahimi [15, 16] used Monte Carlo simulation to study bond and site percolation on 2-d and 3-d elastic and superelastic percolation networks with central and bond-bending forces. They have proposed an accurate method for determining the elastic percolation threshold.

Benguigui [17] performed an experiment to study the elastic properties of a 2-d percolating system. He used a metal foil with holes randomly punched on it. Both electrical and elastic properties of such a system were studied. It was observed that both elastic modulus and conductivity go to zero near their respective thresholds with different exponents. The elasticity exponent $T = 3.5 \pm 0.4$ is found to be larger than the conductivity exponent $t = 1.2 \pm 0.1$. The experimental results of Benguigui are found to agree with the theoretical prediction of Bergman [12].

Effective medium theory (EMT) has been applied to treat percolation problems [18-19] like how the elastic moduli go to zero with $p_c - p$ or whether the ratio of the moduli are universal near p_c. EMT has been used to determine the effective elastic moduli of a 2-d continuum system containing randomly oriented elliptical inclusions [20]. The cases of void and rigid reinforcements have been considered and the elastic percolation thresholds are determined from both symmetric and asymmetric self-consistent methods. Although EMT is not very accurate near the threshold, EMT is known to work quite well for a large range of concentrations almost upto the threshold [19].

The self-consistent solutions as obtained in Section 2 are utilised to obtain the elastic constants near the threshold p_c for a 3-d composite whose first phase is void. The behaviour of both effective SOEC and TOEC has been investigated.

The first phase being void, all its elastic constants are zero. We denote by p the volume fraction of the first phase. Then in eqs. (13) and (14), taking $K^{(1)} = \mu^{(1)} = 0$, $v_1 = p$ and $v_2 = 1 - p$, we get a quadratic equation in μ^* as

$$8\mu^{*2} - \mu^*\left[4\mu^{(2)}(2 - 5p) + 3K^{(2)}(p - 3)\right]$$

$$+9K^{(2)}\mu^{(2)}(2p - 1) = 0. \tag{21}$$

Now, at the threshold, the void phase forms a spanning cluster in the system and hence the effective elastic constants also vanish. Putting $\mu^* = 0$ in eq. (21), the threshold is obtained as

$$p_c = 0.5. \tag{22}$$

Since at threshold, the system breaks down, we want to study the behaviour of the system as p_c is approached from below *i.e.* for $p < p_c$. In eq. (21), taking $p = p_c - \delta$, where δ is a very small quantity and thus neglecting higher powers of δ, the behaviour of μ^* close to threshold comes out to be

$$\frac{36K^{(2)}\mu^{(2)}}{4\mu^{(2)} + 15K^{(2)}}(p_c - p). \tag{23}$$

Now in eq. (14), taking $K^{(1)} = 0$, K^* is given by

$$K^* = \frac{4(1-p)\mu^* K^{(2)}}{4\mu^* + 3pK^{(2)}} \tag{24}$$

Using eq. (22) in eq. (23), behaviour of K^* near threshold is

$$K^* = \frac{48K^{(2)}\mu^{(2)}}{4\mu^{(2)} + 15K^{(2)}}(p_c - p). \tag{25}$$

In eqs. (23) and (25), p_c is 0.5 and the critical exponent is 1. The value of p_c is in agreement with that obtained from the self-consistent solution of Watt *et al* [21]. Further, both the critical exponent and percolation threshold match with those obtained by Bergman and Kantor [18] from a generalization of EMA results. It is also observed that using eqs. (23) and (25) as $p \to p_c$

$$\frac{K^*}{\mu^*} \tag{26}$$

that is the ratio of bulk to shear moduli approaches a universal value. Identical result has been obtained by Bergman and Kantor [18].

Uptil now, as it has already been mentioned, the nonlinear elastic properties of percolative systems have received little attention. In what follows, we focus our attention on the nonlinear part of the problem. To obtain the behaviour of effective TOEC near threshold, we start from eq. (16) which is further simplified now, because the first phase being void, its SOEC and TOEC are zero. Since the effective TOEC are dependent on effective SOEC, the percolation threshold is found to be the same in two cases, however, the exponents differ. We give below the expressions for the effective TOEC near threshold. The details of the calculation are given in the Appendix. Thus,

$$C^*_{123} = (1-p)\Big\{3A_1^2(A_1 + 2A_2)\left(9C^{(2)}_{123} + 18C^{(2)}_{144} + 8C^{(2)}_{456}\right) + 8A_2^3 C^{(2)}_{123} + 12A_1A_2^2\left(3C^{(2)}_{123} + 4C^{(2)}_{144}\right)\Big\}(p_c - p)^3, \tag{27}$$

$$C^*_{144} = (1-p)\Big\{4A_1A_2^2\left(3C^{(2)}_{144} + 4C^{(2)}_{456}\right) + 8A_2^3C^{(2)}_{144}\Big\}(p_c - p)^3 \tag{28}$$

and

$$C^*_{456} = 8(1-p)\,A_2^3 C^{(2)}_{456}(p_c - p)^3, \tag{29}$$

where superscript (2) refers to the second phase and

$$A_1 = \frac{8\left(4\mu^{(2)} + 15K^{(2)}\right)\left(4\mu^{(2)} - 3K^{(2)}\right)}{\left(15K^{(2)} - 20\mu^{(2)} + 48\mu^{(2)}p\right)\left(4\mu^{(2)} - 3K^{(2)} + 36K^{(2)}p\right)}, \tag{30}$$

and $$A_2 = \frac{36K^{(2)}}{36K^{(2)}p + 4\mu^{(2)} - 3K^{(2)}}. \tag{31}$$

From eqs. (27) – (29), we find that in the nonlinear case, the exponent is 3. Hence, the thresholds for effective SOEC and TOEC are same but the exponents differ. To our knowledge, the present study involving TOEC of percolative system is the first of its kind.

Earlier, EMA has been used by some authors [22–24] to treat the percolation behaviour with respect to nonlinear dielectric property which being a lower rank tensorial property than TOEC, is easier to handle.

4. Cross-over strain field in percolative system

The results obtained in the previous section will now be utilised to determine the cross-over strain field in a percolating solid-void system. The cross-over strain field is defined as that field at which the linear response and nonlinear response of the composite become comparable. The concept of cross-over field originated from the work of Blumenfeld and Bergman [22] who used the result of Stroud and Hui [23] on the conductivity of a weakly nonlinear medium to obtain the scaling behaviour of critical current I_c which denotes the transition from linear to nonlinear behaviour. Hui [24] attempted to calculate the cross-over electric field in percolating perfect conductor-nonlinear normal metal composite within the framework of the effective medium theory. The exponent of cross-over field was found to be 1/2 for all spatial dimensions. According to the definition of the cross-over field, in the elastic case, the following condition must be satisfied

$$C^*_{ijkl} < \varepsilon_{kl} > = \frac{1}{2} C^*_{ijklmn} < \varepsilon_{kl} >< \varepsilon_{mn} >. \tag{32}$$

Now, for the isotropic material, there are two types of strain – the bulk and the shear strain. Thus,

$$< \varepsilon_{11} > = < \varepsilon_{22} > = < \varepsilon_{33} > = \varepsilon_1(say) \tag{33}$$

which denotes the bulk strain and

$$< \varepsilon_{23} > = < \varepsilon_{13} > = < \varepsilon_{12} > = \varepsilon_4(say) \tag{34}$$

denotes the shear strain. Then from eq. (32), we get

$$\left(3C^*_{12} + 2C^*_{44}\right)\varepsilon_1 = \frac{9}{2}C^*_{123}\varepsilon_1^2 + C^*_{144}\left(9\varepsilon_1^2 + 3\varepsilon_4^2\right) + 4C^*_{456}\left(\varepsilon_1^2 + 2\varepsilon_4^2\right) \tag{35}$$

and

$$\varepsilon_4 = \frac{C^*_{44} - \varepsilon_1(3C^*_{144} + 4C^*_{456})}{2C^*_{456}} \tag{36}$$

Substituting eq. (36) in eq. (35), we get a quadratic equation in ε_1 as

$$A\varepsilon_1^2 - 2B\varepsilon_1 + C = 0, \tag{37}$$

where

$$A=\left(3C_{144}^{*}+4C_{456}^{*}\right)^{2}\left(3C_{144}^{*}+8C_{456}^{*}\right)$$
$$+2C_{456}^{*2}\left(9C_{123}^{*}+18C_{144}^{*}+8C_{456}^{*}\right), \tag{38}$$

$$B=\left(3C_{144}^{*}+4C_{456}^{*}\right)\left(3C_{144}^{*}+8C_{456}^{*}\right)C_{44}^{*}$$
$$+2C_{456}^{*2}\left(3C_{12}^{*}+2C_{44}^{*}\right) \tag{39}$$

and

$$C=\left(3C_{144}^{*}+8C_{456}^{*}\right)C_{44}^{*2}. \tag{40}$$

Using the results of the previous section *i.e.* the behaviour of the effective SOEC and TOEC near threshold, we get $A\sim(p_c-p)^9$, $B\sim(p_c-p)^7$ and $C\sim(p_c-p)^5$.

The exact expressions are too big and so are not given here explicitly. From the solution of the quadratic equation and using the variation of A, B and C with (p_c-p), the strain field ε_1 is found to behave as

$$\varepsilon_1\sim(p_c-p)^{-2}. \tag{41}$$

Using eq. (41) in eq. (36), we get

$$\varepsilon_4\sim(p_c-p)^{\,2}. \tag{42}$$

Thus within EMA, the exponent for cross-over strain field is 2. The linear regime broadens near the threshold since from eqs (41) and (42), it appears that close to p_c, very strong field will be needed to observe the cross-over behaviour. Hui [24] suggested that if one phase of the composite is a hard material (elastic constant very large), the linear response regime shrinks. So it is expected that reverse will happen in the case where one phase is void, and our theory reveals the same fact.

5. Conclusions

Some interesting aspects of the nonlinear mechanical property of composites have been studied in this work. The percolation study for nonlinear elastic property and the cross-over strain field have been investigated for the first time. EMA may not give accurate results near the percolation transition, however, it can be used as a first step to treat percolation behaviour of nonlinear properties. These studies will become more interesting if the shape of the grains or phases can be taken into consideration. The study is in progress and will be reported in a future communication.

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Appendix

We give here the scheme of calculation starting from eq. (16) to obtain the equations for effective TOEC near percolation threshold.

For the first phase *i.e.* void, all elastic constants vanish. Thus,

$$K^{(1)}=\mu^{(1)}=0$$

and

$$C_{123}^{(1)}=C_{144}^{(1)}=C_{456}^{(1)}=0.$$

Then eq. (16) simplifies as

$$C_i^{*}=(1-p)\Big[\Big\{3(X^{(2)})^2(X+2Y)^{(2)}(3C_1+2C_2)^{(2)}$$
$$+12X^{(2)}(Y^{(2)})^2C_1^{(2)}\Big\}\delta^{(1)}+4X^{(2)}(Y^{(2)})^2C_2^{(2)}\delta^{(2)}$$
$$+8(Y^{(2)})^3C_i^{(2)}\Big] \tag{A1}$$

where

$$X^{(2)} = \frac{3K^{*}+4\mu^{*}}{3K^{(2)}+4\mu^{*}}$$

$$\frac{2(K^{*}+2\mu^{*})(\mu^{(2)}-\mu^{*})-5\mu^{*}(K^{(2)}-K^{*})}{5\mu^{*}(3K^{*}+4\mu^{*})+6(K^{*}+2\mu^{*})(\mu^{(2)}-\mu^{*})} \tag{A2}$$

$$Y^{(2)} = \frac{5\mu^{*}(3K^{*}+4\mu^{*})}{2\left\{5\mu^{*}(3K^{*}+4\mu^{*})+6(K^{*}+2\mu^{*})(\mu^{(2)}-\mu^{*})\right\}}, \tag{A3}$$

$$C_{1}^{(2)} = 3C_{123}^{(2)} + 4C_{144}^{(2)}, \tag{A4}$$

$$C_{2}^{(2)} = 3C_{144}^{(2)} + 4C_{456}^{(2)}. \tag{A5}$$

Using eqs. (23) and (25) which give the behaviour of μ^{*} and K^{*} respectively near threshold, $X^{(2)}$ and $Y^{(2)}$ take the form

$$X^{(2)} = -\frac{8(4\mu^{(2)}+15K^{(2)})(4\mu^{(2)}-3K^{(2)})}{(4\mu^{(2)}-3K^{(2)}+36K^{(2)}p)(15K^{(2)}-20\mu^{(2)}+48\mu^{(2)}p)}(p_{c}-p) \tag{A6}$$

and

$$Y^{(2)} = \frac{36K^{(2)}}{36K^{(2)}p+4\mu^{(2)}-3K^{(2)}}(p_{c}-p). \tag{A7}$$

In eq. (A1) putting appropriate tensor indices and using eqs (A1) - (A7), we get eqs. (27)–(29).