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Nonlinear elastic Landau coefficients in heterogeneous materials

S. GIORDANO, P. L. PALLA and L. COLOMBO^(a)

Department of Physics, University of Cagliari and Sardinian Laboratory for Computational Materials Science (SLACS, INFN-CNR) - Cittadella Universitaria, I-09042 Monserrato (CA), Italy, EU

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Abstract – We prove that the elastic fields within a generic nonlinear and anisotropic inhomogeneity embedded in a (linear and anisotropic) matrix are uniform. We apply this general result to the specific case of a dispersion of isotropic nonlinear spheres and we obtain a universal mixing scheme for the Landau coefficients. This scheme describes a complex scenario frequently found in material physics problems, where possible strong amplifications of the nonlinearities may arise in some given conditions.

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Introduction. – The central problem in predicting the elasticity of heterogeneous materials (like, *e.g.*, composite or nanostructured systems, mixtures, multi-defected or multi-cracked media) consists in the evaluation of their effective macroscopic elastic properties, still taking into account the actual microscale material features. This leads to the concept of homogenization, a coarse-graining approach addressed to determine the relationship between the microstructure and the effective elastic behavior.

Homogenization has been successfully developed for linear elastic properties (as well as for linear electric ones) [1]. The existence of upper and lower bounds for the effective elastic moduli has been proved, independently of the specific microstructure [2,3]. Alternatively, the effective elastic behavior of a heterogeneous system can be obtained by taking into account the spatial correlation among its constituents [4,5]. More in particular, dilute dispersions of inclusions in a homogeneous matrix have been widely studied both from the electrical [6] and the elastic [7] point of view, as well as for systems containing a distribution of cracks [8–11]. Finally, the iterative technique and the differential method are successfully applied for arbitrarily dense dispersions [12].

In heterogeneous or composite materials the nonlinear regime has been investigated only under specific conditions [13–18]. Nevertheless, the general nonlinear elastic features are relevant in many materials science problems. For instance, transient elastography has shown

its efficiency to map the linear and nonlinear properties of soft tissues and it is nowadays used as diagnostic technique [19,20]. In fact, it has been verified that malignant lesions tend to exhibit nonlinear elastic behavior contrary to normal tissues or to benign lesions. This point is explained by observing that malignant lesions alter the structure of the cellular network enhancing the nonlinear properties. The pathological nonlinear zones can be profitably described by the so-called Landau coefficients (see below), making feasible their localization through noninvasive imaging techniques (ultrasound and/or magnetic resonance) [21]. Another relevant example is offered by the engineering of semiconductor quantum dots, embedded in a confining solid matrix. The quantum dots growth, ordering and orientation (occurring during processing) are largely affected by elastic phenomena, even beyond the linear regime [22,23]. Finally, many problems of fracture mechanics in composite materials do contain nonlinear features like, *e.g.*, the interaction between the stress fields generated by a moving crack and a fiber (or, more generally, an inclusion) [24].

In this letter we prove a general property of the elastic strain field within any nonlinear and anisotropic inhomogeneity, embedded into a linear (but anisotropic) matrix. This general result is then applied to the more specific case of a dispersion of nonlinear isotropic spheres (see fig. 1), which paradigmatically represents most features of the above examples.

Nonlinearity can be introduced in the theory of elasticity by means of the exact relation for the Lagrangian

^(a)E-mail: luciano.colombo@dsf.unica.it

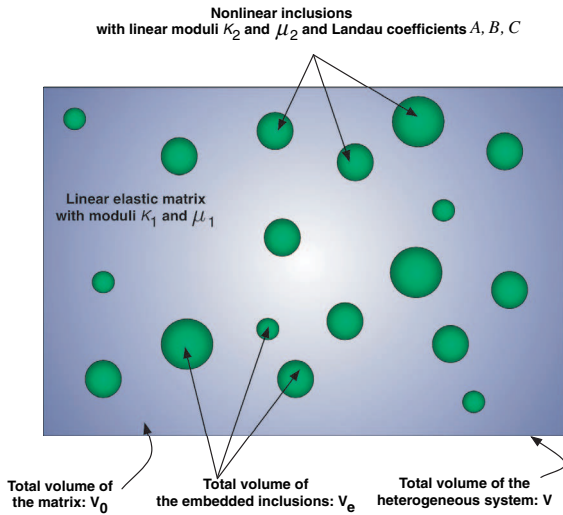


Fig. 1: Dispersion of nonlinear spheres in a linear matrix with volume fraction $c = V_e/V = V_e/(V_e + V_0)$.

strain (geometrical nonlinearity) and/or through a nonlinear stress-strain constitutive relation (non-Hookean physical nonlinearity) [25]. In this work, we adopt the physical nonlinearity standpoint, whereas the geometrical linearity is everywhere assumed: therefore, the balance equations are based on the small-strain tensor $\hat{\epsilon}$, on the symmetric Cauchy stress \hat{T} , and on an arbitrary (nonlinear) stress-strain relation.

A general theorem on the elastic behavior of a nonlinear anisotropic inhomogeneity. – The general property we are looking for sets out from the Eshelby theory, which solves the elasticity equations for a single anisotropic linear ellipsoid (stiffness $\hat{C}^{(2)}$) embedded into an anisotropic linear matrix (stiffness $\hat{C}^{(1)}$) [26]. Upon uniform remote loading $\hat{T}^\infty = \hat{C}^{(1)}\hat{\epsilon}^\infty$, it is proved [27] that the strain field $\hat{\epsilon}^s$ inside the ellipsoid is uniform and assumes the value

$$\hat{\epsilon}^s = \{\hat{I} - \hat{S}[\hat{I} - (\hat{C}^{(1)})^{-1}\hat{C}^{(2)}]\}^{-1}\hat{\epsilon}^\infty, \quad (1)$$

where the Eshelby tensor \hat{S} depends only on the geometry and on $\hat{C}^{(1)}$.

We can generalize this result to the case where the inhomogeneity is nonlinear, *i.e.* $\hat{T} = \hat{C}^{(2)}(\hat{\epsilon})\hat{\epsilon}$, where $\hat{C}^{(2)}(\hat{\epsilon})$ is any strain-dependent anisotropic stiffness tensor (see fig. 2). The energy balance is described by the Green formulation: for a given state of deformation, the stress power is absorbed into a strain energy function $U(\hat{\epsilon})$, leading to the constitutive equation $\hat{T}(\hat{\epsilon}) = \frac{\partial U(\hat{\epsilon})}{\partial \hat{\epsilon}}$ equivalent to $\hat{T} = \hat{C}^{(2)}(\hat{\epsilon})\hat{\epsilon}$ [25]. The strain energy function can be identified with the internal energy per unit volume in an isentropic process, or with the Helmholtz free-energy per unit volume in an isothermal process. In the present generalization nonlinear features are described by an arbitrary free-energy function $U(\hat{\epsilon})$. In order to cope with this

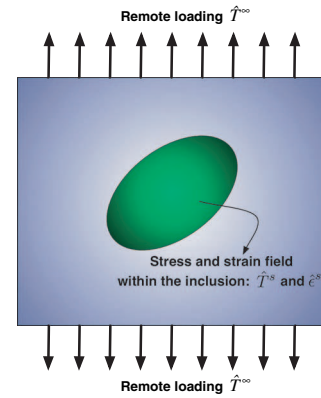


Fig. 2: Ellipsoidal inhomogeneity under remote load.

problem, we suppose to have found a solution for the equation

$$\hat{\epsilon}^s = \{\hat{I} - \hat{S}[\hat{I} - (\hat{C}^{(1)})^{-1}\hat{C}^{(2)}(\hat{\epsilon}^s)]\}^{-1}\hat{\epsilon}^\infty, \quad (2)$$

obtained from eq. (1) through the substitution $\hat{C}^{(2)} \rightarrow \hat{C}^{(2)}(\hat{\epsilon}^s)$. If such a solution $\hat{\epsilon}^s = \hat{\epsilon}^{s*}$ exists for a given $\hat{\epsilon}^\infty$, it means that the nonlinear inhomogeneity could be replaced by a linear one with constant stiffness $\hat{C}^{(2)} = \hat{C}^{(2)}(\hat{\epsilon}^{s*})$, without modifications of the elastic fields at any point. Therefore, if $\hat{\epsilon}^{s*}$ exists, then eq. (2) exactly describes, through self-consistency, the elastic behavior of the nonlinear anisotropic inclusion. This is not a trivial result: for instance, such a generalization of eq. (1) is not valid if a nonlinear behavior is assumed for material 1 (matrix).

The existence and unicity of a solution $\hat{\epsilon}^{s*}$ for eq. (2) can be exactly proved under the sole hypothesis of convexity for the strain energy function $U(\hat{\epsilon})$. We rearrange eq. (2) as follows:

$$\begin{aligned} [\hat{I} - \hat{S}]\hat{\epsilon}^s + \hat{S}(\hat{C}^{(1)})^{-1}\frac{\partial U(\hat{\epsilon}^s)}{\partial \hat{\epsilon}^s} &= \hat{\epsilon}^\infty, \\ \hat{C}^{(1)}[\hat{S}^{-1} - \hat{I}]\hat{\epsilon}^s - \hat{C}^{(1)}\hat{S}^{-1}\hat{\epsilon}^\infty + \frac{\partial U(\hat{\epsilon}^s)}{\partial \hat{\epsilon}^s} &= 0, \end{aligned} \quad (3)$$

$$\frac{\partial}{\partial \hat{\epsilon}} \left\{ \frac{1}{2}\hat{\epsilon}\hat{C}^{(1)}[\hat{S}^{-1} - \hat{I}]\hat{\epsilon} - \hat{\epsilon}\hat{C}^{(1)}\hat{S}^{-1}\hat{\epsilon}^\infty + U(\hat{\epsilon}) \right\} = 0.$$

Now, the first term represents a symmetric (because of the Betti reciprocal theorem [28]) and positive definite (because of the minimum-potential-energy principle [29]) quadratic form in $\hat{\epsilon}$, while the second term is a linear function of $\hat{\epsilon}$. Therefore, the sum of these two terms is a convex functional with relative minimum at $[\hat{I} - \hat{S}]\hat{\epsilon}^\infty$. If $U(\hat{\epsilon})$ is a convex functional (with $U(0) = 0$) as well, the brackets in eq. (3) contain the sum of two convex terms: they result in an overall convex functional with a minimal extremum at $\hat{\epsilon}^s = \hat{\epsilon}^{s*}$. Therefore, a unique solution of eq. (2) exists under the convexity assumption for $U(\hat{\epsilon})$.

We have obtained an important new property of an arbitrary inhomogeneity: if the linear elastic matrix is subjected to remote uniform loading, then the stress and strain fields (\hat{T}^s and $\hat{\epsilon}^s$) inside the embedded inhomogeneity will be uniform, independently of the (nonlinear and

anisotropic) constitutive relation for the inhomogeneity itself.

Our theorem is based on the symmetric and the positive-definite character of the tensor $\hat{\mathcal{C}}^{(1)}[\hat{\mathcal{S}}^{-1} - \hat{I}]$. We have proved both properties, but for sake of brevity the cumbersome demonstrations will be published elsewhere.

Application of the general theorem to the case of a dispersion of spheres. – The general result stated in eq. (2) can be applied to fully homogenize any dispersion of arbitrarily nonlinear and anisotropic ellipsoidal inclusions. Let us now consider a more specific dispersion of nonlinear spherical isotropic inclusions. The density is described by the volume fraction c , defined as the ratio between the total volume of the embedded spheres and the total volume of the heterogeneous material (see fig. 1). The matrix is described by the linear constitutive equation $\hat{T} = 2\mu_1\hat{\epsilon} + (K_1 - \frac{2}{3}\mu_1)\text{Tr}(\hat{\epsilon})\hat{I}$, where K_1 and μ_1 are the bulk and shear moduli, respectively. To model the spherical inclusions, we adopt the most general isotropic nonlinear constitutive equation expanded up to the second order in the strain components: it follows that the function $U(\hat{\epsilon})$ can only depend upon the principal invariants of the strain tensor, *i.e.* $U = U(\text{Tr}(\hat{\epsilon}), \text{Tr}(\hat{\epsilon}^2), \text{Tr}(\hat{\epsilon}^3))$. Therefore, by expanding $U(\hat{\epsilon})$ up to the third order in the strain components, we obtain

$$U(\hat{\epsilon}) = \mu_2\text{Tr}(\hat{\epsilon}^2) + \frac{1}{2}\left(K_2 - \frac{2}{3}\mu_2\right)[\text{Tr}(\hat{\epsilon})]^2 + \frac{A}{3}\text{Tr}(\hat{\epsilon}^3) + B\text{Tr}(\hat{\epsilon})\text{Tr}(\hat{\epsilon}^2) + \frac{C}{3}[\text{Tr}(\hat{\epsilon})]^3 \quad (4)$$

and deriving the stress, we get

$$\hat{T} = 2\mu_2\hat{\epsilon} + \left(K_2 - \frac{2}{3}\mu_2\right)\text{Tr}(\hat{\epsilon})\hat{I} + A\hat{\epsilon}^2 + B\{\text{Tr}(\hat{\epsilon}^2)\hat{I} + 2\hat{\epsilon}\text{Tr}(\hat{\epsilon})\} + C[\text{Tr}(\hat{\epsilon})]^2\hat{I} \quad (5)$$

for the material corresponding to the spherical inclusions. The parameters A , B , and C are the Landau moduli [25] and they represent the deviation from the standard linearity. The explicit expression of the Eshelby tensor for a sphere is reported below [27]:

$$\mathcal{S}_{ijkh} = \frac{1}{15(1-\nu_1)}[(\delta_{ik}\delta_{jh} + \delta_{ih}\delta_{jk})(4-5\nu_1) + \delta_{kh}\delta_{ij}(5\nu_1-1)], \quad (6)$$

where ν_1 is the Poisson ratio of the matrix. We can evaluate the effect of \mathcal{S}_{ijkh} over an arbitrary strain ϵ_{kh}^s , getting

$$\hat{\mathcal{S}}\hat{\epsilon}^s = \frac{6}{5}\frac{K_1+2\mu_1}{3K_1+4\mu_1}\hat{\epsilon}^s + \frac{1}{5}\frac{3K_1-4\mu_1}{3K_1+4\mu_1}\text{Tr}(\hat{\epsilon}^s)\hat{I}. \quad (7)$$

By inserting eqs. (5) and (7) into eq. (2), it can be proved that

$$L\hat{\epsilon}^s + M\text{Tr}(\hat{\epsilon}^s)\hat{I} + N(\hat{\epsilon}^s)^2 + O\hat{\epsilon}^s\text{Tr}(\hat{\epsilon}^s) + P\text{Tr}[(\hat{\epsilon}^s)^2]\hat{I} + Q[\text{Tr}(\hat{\epsilon}^s)]^2\hat{I} = \hat{\epsilon}^\infty, \quad (8)$$

defining the explicit relation between the internal strain $\hat{\epsilon}^s$ and the remote deformation $\hat{\epsilon}^\infty$, for a single nonlinear spherical inhomogeneity. The parameters

$$L = 1 + \frac{6}{5}\frac{K_1+2\mu_1}{3K_1+4\mu_1}\left(\frac{\mu_2}{\mu_1}-1\right), \quad (9)$$

$$M = \frac{5K_2 - K_1\left(3 + 2\frac{\mu_2}{\mu_1}\right) - 4(\mu_2 - \mu_1)}{5(3K_1 + 4\mu_1)}, \quad (10)$$

$$N = \frac{3}{5}\frac{A}{\mu_1}\frac{K_1+2\mu_1}{3K_1+4\mu_1}, \quad (11)$$

$$O = \frac{6}{5}\frac{B}{\mu_1}\frac{K_1+2\mu_1}{3K_1+4\mu_1}, \quad (12)$$

$$P = \frac{1}{15(3K_1+4\mu_1)}\left[15B - A\left(1 + 3\frac{K_1}{\mu_1}\right)\right], \quad (13)$$

$$Q = \frac{1}{15(3K_1+4\mu_1)}\left[15C - 2B\left(1 + 3\frac{K_1}{\mu_1}\right)\right] \quad (14)$$

depend on both linear and nonlinear moduli.

Let us now move to the actual case of a dilute dispersion of spheres (see fig. 1). Under the hypothesis of a small c , the average value of the strain in the overall system is given by $\langle\hat{\epsilon}\rangle = c\hat{\epsilon}^s + (1-c)\hat{\epsilon}^\infty$. Similarly, the average value of the stress is $\langle\hat{T}\rangle = \hat{\mathcal{C}}^{(1)}\langle\hat{\epsilon}\rangle + c\hat{T}^s - c\hat{\mathcal{C}}^{(1)}\hat{\epsilon}^s$. The average fields $\langle\hat{T}\rangle$ and $\langle\hat{\epsilon}\rangle$, combined through eq. (8), determine the effective constitutive equation for the heterogeneous system. Basically, it is written as eq. (5) where, however, the effective linear and nonlinear elastic moduli μ_{eff} , K_{eff} , A_{eff} , B_{eff} and C_{eff} must be introduced. As for the linear elastic coefficients, we obtain

$$\mu_{eff} = \mu_1 + c\frac{\mu_2 - \mu_1}{c + (1-c)\left[1 + \frac{6}{5}\left(\frac{\mu_2}{\mu_1} - 1\right)\frac{K_1+2\mu_1}{3K_1+4\mu_1}\right]}, \quad (15)$$

$$K_{eff} = K_1 + \frac{(3K_1+4\mu_1)(K_2-K_1)c}{3K_2+4\mu_1-3c(K_2-K_1)}. \quad (16)$$

We observe that K_{eff} and μ_{eff} only depend upon their linear counterparts for materials 1 and 2. Interesting enough, the effective Landau coefficients show a more complicated structure

$$A_{eff} = c\frac{A}{L'^2} - 2c\frac{N'(\mu_2 - \mu_1)}{L'^3}, \quad (17)$$

$$B_{eff} = 2c\frac{(N'M' - L'P')(\mu_2 - \mu_1)}{L'^3(L' + 3M')} - c\frac{(N' + 3P')\left[K_2 - K_1 - \frac{2}{3}(\mu_2 - \mu_1)\right]}{L'^2(L' + 3M')} + c\frac{B}{L'^2}. \quad (18)$$

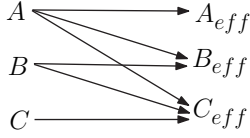


Fig. 3: Mixing scheme for the nonlinear Landau coefficients.

$$\begin{aligned}
C_{eff} = & \frac{1}{9} \frac{c(9C + 9B + A)}{(L' + 3M')^2} + \frac{1}{9} \frac{c(A - 3B)}{L'^2} \\
& + \frac{1}{9} \frac{c(4N' + 6O')(\mu_2 - \mu_1)}{L'^2(L' + 3M')} - \frac{2}{9} \frac{c(3B + A)}{L'(L' + 3M')} \\
& + \frac{1}{9} \frac{c(3N' + 9P')(K_2 - K_1)}{L'^2(L' + 3M')} - \frac{4}{9} \frac{N'(\mu_2 - \mu_1)c}{L'^3} \\
& - \frac{1}{3} \frac{c(9Q' + 3O' + 3P' + N')(K_2 - K_1)}{(L' + 3M')^3}, \quad (19)
\end{aligned}$$

where we have introduced the parameters $L' = c + (1 - c)L$, $M' = (1 - c)M$, $N' = (1 - c)N$, $O' = (1 - c)O$, $P' = (1 - c)P$, and $Q' = (1 - c)Q$. We note that eqs. (15)–(19) hold even in the limiting case of $c = 1$, falling beyond the adopted hypothesis of small volume fraction.

Despite the assumptions adopted to work out the above equations, the following properties define a general scenario valid for any nonlinear heterogeneous system having reference to the geometry of fig. 1. First of all, we have proved that A , B , and C determine the effective nonlinear moduli of the heterogeneous material by following the universal mixing scheme shown in fig. 3.

Furthermore, if the linear elastic moduli of the matrix and of the spheres are the very same (*i.e.* $K_1 = K_2$ and $\mu_1 = \mu_2$), we simply obtain $K_{eff} = K_1$, $\mu_{eff} = \mu_1$ and $A_{eff} = cA$, $B_{eff} = cB$, $C_{eff} = cC$. In other words, the effective Landau coefficients are simply rescaled by the volume fraction c .

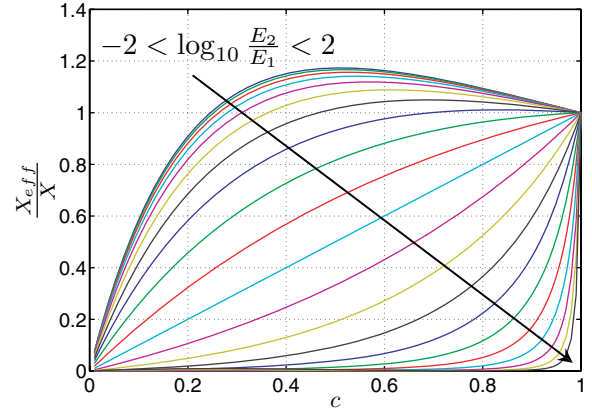
Then, if we consider the special case of identical Poisson ratios $\nu_1 = \nu_2 = 1/5$ (and different Young moduli $E_1 \neq E_2$), we obtain $\nu_{eff} = 1/5$, and

$$E_{eff} = \frac{E_1(1 - c) + E_2(1 + c)}{E_1(1 + c) + E_2(1 - c)} E_1. \quad (20)$$

The Landau coefficients can be calculated as follows:

$$X_{eff} = \frac{8E_1^3 c}{[E_1(1 + c) + E_2(1 - c)]^3} X, \quad (21)$$

where $X = A$, B or C . Therefore, $\nu_1 = \nu_2 = 1/5$ is the only situation where the mixing scheme of fig. 3 is simplified, generating a direct correspondence among the nonlinear moduli of the spheres and the effective nonlinear moduli of the heterogeneous system. We deduce from fig. 4 that the effective modulus X_{eff} can be as large as the modulus X (also for small volume fraction c) if $E_2/E_1 \ll 1$. The special value $1/5$ for the Poisson ratio is not uncommon. For linear porous materials (with spherical pores) and for linear dispersions of rigid spheres, the following property

Fig. 4: Nonlinear Landau coefficients given in eq. (21) for the special case of identical Poisson ratios $\nu_1 = \nu_2 = 1/5$, for a varying ratio between Young moduli E_2 and E_1 .

holds: if $\nu_1 = 1/5$, then we have $\nu_{eff} = 1/5$ for any volume fraction of pores or rigid spheres [12]. Moreover, in both the above systems, in the limit of $c \rightarrow 1$, the effective Poisson ratio converges to the fixed value $\nu_{eff} = 1/5$, irrespective of the matrix Poisson ratio [12].

Finally, we consider dispersed spheres made of a nonlinear isotropic and incompressible material (*i.e.* $K_2 \rightarrow \infty$ in eq. (5)). The corresponding constitutive equation is obtained as follows:

$$\begin{aligned}
\hat{\epsilon} = & \frac{1}{2\mu_2} \hat{T} + \frac{A}{12\mu_2^3} \hat{T} \text{Tr}(\hat{T}) + \frac{A}{24\mu_2^3} \text{Tr}[(\hat{T})^2] \hat{I} \\
& - \frac{1}{6\mu_2} \text{Tr}(\hat{T}) \hat{I} - \frac{A}{8\mu_2^3} (\hat{T})^2 - \frac{A}{36\mu_2^3} [\text{Tr}(\hat{T})]^2 \hat{I}, \quad (22)
\end{aligned}$$

where only the nonlinear coefficient A appears. Such a relationship imposes $\text{Tr}(\hat{\epsilon}^s) = 0$, as requested by the incompressibility. As for the corresponding effective linear moduli, eq. (15) for μ_{eff} remains unchanged, while eq. (16) leads to $K_{eff} = K_1 + (K_1 + \frac{4}{3}\mu_1) \frac{c}{1-c}$. On the other hand, the Landau coefficients have been eventually found to be

$$A_{eff} = 125A\theta, \quad B_{eff} = -\frac{125}{3}A\theta, \quad C_{eff} = \frac{250}{9}A\theta, \quad (23)$$

where

$$\theta = \frac{c(3K_1 + 4\mu_1)^3 \mu_1^3}{\{6(K_1 + 2\mu_1)[c\mu_1 + (1 - c)\mu_2] + \mu_1(9K_1 + 8\mu_1)\}^3}. \quad (24)$$

One can observe that the effective nonlinear elastic moduli depend only on the modulus A , fully describing the nonlinearity of the spheres. This is an example of mixing where a single nonlinear modulus affects all of the nonlinear Landau coefficients of the mixture.

In order to show the most important achievements of our theory, we performed a numerical implementation of eqs. (15)–(19). Under the hypothesis of $K_1 \gg K_2$, we always observe a sizeable amplification of the nonlinear effective modulus C_{eff} . In fig. 5 we have plotted the

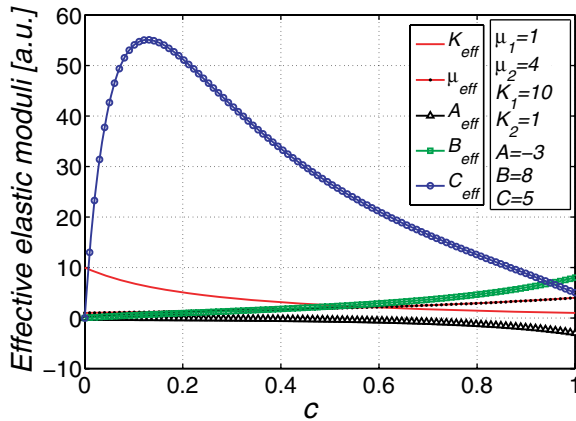


Fig. 5: Linear and nonlinear effective elastic moduli in terms of the volume fraction c .

effective moduli *vs.* the volume fraction c , considering the mixture parameters $\mu_1 = 1, \mu_2 = 4, K_1 = 10, K_2 = 1, A = -3, B = 8, C = 5$ in arbitrary units. The enhancement of C_{eff} is remarkable; we underline that such an intriguing feature is obtained for any set of parameters, provided that the matrix is much more incompressible than the inclusions. We point out that this feature occurs well within the range of validity of the present theory, namely for small values of c (see fig. 5). Our numerical results prove that the Landau moduli are not limited by some given bounds, at variance with the case of their linear counterparts, which are constricted by the classical Hashin-Shtrikman limitations [2,3].

More in general, the enhancement of Landau moduli suggests that the nonlinear effective properties can be strongly affected by the linear moduli of the constituents of the heterogeneous material. For example, the ratio C_{eff}/C is sensibly modulated by the ratio K_1/K_2 . This point could be relevant in analyzing normal or pathological tissues in biomechanics and in studying functionalized materials with specific nonlinear properties. In fact, the relationship here established between microstructural features and effective properties can be used for designing and improving materials or, conversely, for interpreting experimental data in terms of the elastic behavior of their constituents.

Conclusions. – In conclusion, we have developed a complete homogenization theory for a dilute dispersion of nonlinear spheres into a linear matrix. A fully analytical set of equations describing the effective elastic behavior is worked out. The present theoretical device is properly suited to recognize complex unusual mixing phenomena exhibited by a heterogeneous material. In particular, we proved that each effective Landau coefficient only depends upon its counterpart in the nonlinear inclusions for special values of the Poisson ratio. Finally, we obtained a sizeable enhancement of some nonlinearities when $K_1 \gg K_2$.

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