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Lurie solution for spherical particle and spring layer model of interphases: Its application in analysis of effective properties of composites

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ABSTRACT

A new approach to the determination of equivalent inhomogeneity for spherical particles and the spring layer model of their interphases with the matrix material is developed. To validate this approach the effective properties of random composites containing spherical inhomogeneities surrounded by an interphase material of constant thickness are evaluated. The properties of equivalent inhomogeneity, incorporating only properties of the original inhomogeneity and its interphase, are determined employing a new approach based on the exact Lurie's solution for spheres. This constitutes the central aspect of the proposed approach being in contrast with some existing definitions of equivalent inhomogeneity whose properties dependent also on the properties of the matrix. With the equivalent inhomogeneity specified as proposed here, the effective properties of the material with interphases can be found using any method applicable to analysis of the materials with perfect interfaces (i.e., without interphases) and any properties of the matrix. In this work, the method of conditional moments is employed to this end. The choice of that method is motivated by the method's solid formal foundations, its potential applicability to inhomogeneities other than spheres and to anisotropic materials. The resulting effective properties of materials with randomly distributed spherical particles are presented in the closed-form and are in excellent agreement with values reported in technical literature, which are based on both formally exact and approximate methods.

1. Introduction

Connection between dissimilar materials is always accompanied by the presence of a layer, called *interphase*, whose properties are different than those of the adjacent bulk materials. Some problems involving such a layer (or many of them), e.g., single spherical inhomogeneity surrounded by layers of a different material, can be solved analytically using a formally exact approach (Lurie, 2005). However, most problems of that kind are too difficult (or too demanding) to lend themselves to exact analytical treatment. Consequently, over the past 40 years or so (Benveniste 1985; Hashin 1961, 1990, 1991; Lipton and Vernescu 1995; Luo and Weng 1987; Mal and Bose 1974; Walpole 1978; Zhong et al. 1997) various approaches have

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been proposed to approximately capture the influence of interphase layers on the quantities of interest, such as local stress and strain fields or effective properties of multi-constituent materials (composites).

The interphases are where some of the most important, complicated, and interesting phenomena in composite materials often occur. In composites with a high interphase-to-volume ratio, this may strongly affect the overall behavior of the material and requires appropriate models to capture such behavior analytically. In some situations, the interphase is well described by a mathematical surface, called *interface*. If both the displacements and tractions (stress vectors) can be assumed to be continuous across the interface, it is commonly called a "*perfect interface*". There exist situations, however, when it is more appropriate to use an "*imperfect interface*" model, i.e., an interface across which displacements, or tractions, or both, suffer suitably defined jumps (cf. Gurtin and Murdoch 1975; Gurtin et al. 1998; Benveniste and Miloh 2001; Hashin 1991, 2002a; Gu and He 2011; Gu et al. 2014).

In general, the interphase can be modeled by an interface (either perfect or imperfect) if the ratio of the interphase's thickness to a characteristic dimension of the composite (typically the size of the embedded inhomogeneities) is "sufficiently" small (Benveniste and Miloh 2001; Hashin 1991, 2002a; Gu and He 2011; Dong et al. 2014). The word "sufficiently" is typically understood in the asymptotic sense meaning that, treating this ratio as the only small parameter in the composite, the interphase can be adequately described by the leading term of the asymptotic expansion of the relevant equations. Depending on how other data of the problem are related to that ratio various interface conditions can be derived (Benveniste and Miloh 2001; see also Hashin 2002a; Rubin and Benveniste 2004; Dong et al. 2014).

Presence of interphases, or imperfect interfaces, significantly complicates evaluation of those properties, and the literature in that area is overwhelmingly numerical (Andrianov et al. 2007; Achenbach and Zhu 1989; Sangani and Mo 1997; Garboczi and Berryman 2001; McBride et al. 2012, among others). Even approaches based on simplified methods, such as self-consistent schemes, often do not lead to closed-form solutions and require numerical calculations, (Hashin 1991, 2002a). Treatment of perfect interfaces, although complex too, is more advanced, as can be gleaned from several good books (Christensen 1991; Mura 1987; Nemat-Nasser and Hori, 1999; Torquato 2002 among others). Effective (or homogenized) properties of composites with perfect interfaces are often given by closed-form formulas, which is a very attractive feature. Due to the complexity of the problem, those formulas are typically approximate; only for composites with regular arrangement of particles or fibers formally exact (typically numerical) results have been obtained (Andrianov et al. 2007; Sangani and Mo 1997; Garboczi and Berryman 2001, among others).

An interface model that has long been used in analysis of composites is the so called *spring layer model*. In that model tractions are continuous while displacements are allowed to experience a jump across the interface (Duan et al. 2007a,b; Hashin 1990, 1991; Sangani and Mo 1997). Thus, the continuum interphase is replaced by a layer of normal and tangential springs. This is an adequate approximation of reality if the interphase is thin and if it is sufficiently compliant in comparison with the stiffness of the surrounding materials. A slightly modified version of that model is also employed in this work, but it is used in the context of an entirely new approach to evaluate effective properties of composites with spherical inhomogeneities.

A difficulty that is associated with all interphase models, including the spring layer model, is determination of the parameters needed to describe them. Analyses of Hashin (2002a) and Benveniste and Miloh (2001) provide a rationale behind how the parameters describing various interface models should be related to properties of the interphase treated as elastic continuum. While theoretically insightful, this rationale is of limited practical utility as direct experimental determination of any interphase parameters, whether related to its continuum description or otherwise, is impossible. That can only be done via an inverse analysis, Lin et al. (2005); Wang et al. (2008), as done by Hashin and Monteiro (2002b). The results of that work will subsequently be used for validation of the methodology proposed herein.

A rare, quite complicated but "formally exact" three-dimensional numerical solution (employing series expansion in terms of spherical harmonics) for effective properties of a composite material containing spherical particles with spring layer interfaces was obtained by Sangani and Mo (1997). They tabulated the results for various volume fractions and various properties of the particles, as well as for various properties of the spring layer. These results are invaluable for comparisons and will be used for that purpose here.

In case of composites with interphases or imperfect interfaces and inhomogeneities in the form of particles or fibers, one viable and attractive way of evaluating their effective properties is to replace the inhomogeneity-interphase system by an "equivalent inhomogeneity" with suitably adjusted properties that represents both the original inhomogeneity and the interphase or imperfect interface. In that way, the problem of the material with imperfect interfaces is replaced by the problem with perfect interface, but with changed properties of the inhomogeneities. Consequently, using equivalent inhomogeneity, all the existing closed-form results for two-phase materials can be utilized to obtain the properties of the composites with imperfect interfaces or interphases. This approach has been pursued in several prior publications (Duan et al., 2007a; Hashin 2002a; Shen and Li 2005; Sevostianov and Kachanov 2007; Nazarenko et al. 2015a,b) (described in the sequel) and a new version of it is presented herein. The new version is an alternative to the energy-equivalent inhomogeneity, originally presented by Nazarenko et al., (2015) in the context of the Gurtin-Murdoch (1975) interphase model. In this work the "spring-layer" interphase model is considered instead (cf. Hashin 1990, 1991; Achenbach and Zhu 1989; Sangani and Mo 1997; Duan et al 2007a,b; Hashin 2002a; Gu et al. 2014 among others). The resulting properties of the equivalent inhomogeneity are used in conjunction with some previously obtained results for two-phase composites with perfect interfaces to determine the effective properties of random composites with spring-layer interfaces.

To the authors' best knowledge the possibility of using the equivalent properties of the inhomogeneity-interphase (or imperfect interface) system is first mentioned by Hashin, (1991). The author discusses an extension of this technique to problems involving multiple layers of different interphases Hashin (2002a). However, Hashin's discussion was restricted to the effective bulk modulus, as only for that case he was able to develop closed-form expression; a numerical approach was necessary for evaluating the effective shear modulus.

Prediction of effective moduli of multiphase composites based on the notion of equivalent inhomogeneity combined with the generalized self-consistent scheme was also presented in the two-part paper of Duan et al. (2007a,b) and Gu et al. (2014). Spherical particles or cylindrical fibers with various interface effects or interphases were considered, including the spring layer interface model dealt with herein. In both of the above contributions the unknown stiffness tensor of the equivalent inhomogeneity was determined using the Eshelby's formula for the change of elastic energy caused by insertion of an inhomogeneity in an infinite matrix (Eshelby, 1957). They assume that such change due to embedding the equivalent inhomogeneity is equal to the change caused by embedding the original inhomogeneity together with its interface. This approach yields properties of the equivalent particles/fibers dependent on the properties of the matrix material which is non-physical and it is in sharp contrast with the approach advocated by Hashin (2002a) and with the approach proposed in the present work. This is the main reason behind the new formulation of the problem presented in this work. Details of the procedure proposed here by which the original inhomogeneity and its interphase is replaced by an equivalent inhomogeneity, as well as comparison with the pertinent results obtained in the past, are presented in the second and third sections of the present article.

In summary, it is noted that the existing solutions explicitly accounting for presence of the spring layer model of interphase were obtained in the true closed-form only for the effective bulk modulus (Hashin 1990, 1991, 2002a; Duan et al. 2007a, Gu et al. 2014). Other effective properties (shear moduli) were given by a sequence of complicated formulas that still had to be evaluated numerically (Hashin 1990, 1991, 2002a; Duan et al. 2007a, Gu et al. 2014) or were obtained by numerical methods (Andrianov et al. 2007; Achenbach and Zhu 1989; Garboczi and Berryman 2001; Sangani and Mo 1997). The definition of equivalent inhomogeneity proposed here is significantly simpler and should enhance predictive capabilities of all approximate methods, irrespective of the material property of interest (bulk and shear). It is more direct than all of the several existing definitions, such as that based on the Mori–Tanaka method (Shen and Li 2005), Hashin–Shtrikman bounds (Sevostianov and Kachanov 2007) or Eshelby (1957) formula of the energy change (Duan et al. 2007a; Gu et al. 2014). Use of some of those approaches is likely to introduce a significant error already at the level of determining the properties of the equivalent inhomogeneity. If such an equivalent inhomogeneity is subsequently used to determine the effective properties of the composite using additional approximate schemes, an essential error can result for composites with high contrast in the component properties and for high volume fraction of particles.

The equivalent properties of the inhomogeneity-interphase system in this work are determined based on the Lurie's solution (Lurie 2005) for spheres. In contrast with that of Duan et al. (2007a) and Gu et al. (2014), they depend only on the properties of the original inhomogeneity and of the spring layer parameters (not on the properties of the matrix). Other than being non-physical, dependence of μ_{eq}developed by Duan et al. (2007a) and Gu et al. (2014) on the properties of the matrix seems to make it applicable only in combination with self-consistent method (which is employed in these papers). In contrast, μ_{eq}presented in our work, which is independent of the properties of the matrix, may be used in conjunction with *any method* (old or new) developed to evaluate the effective properties of composites without interphases, and with any matrix material. One small feature of the method discussed herein is that, as opposed to the previous spring layer models whose thickness was vanishingly small, a finite thickness of the spring layer is still retained. That feature is the reason for which the term "*spring layer interphase*" is used throughout this work. Finite interphase thickness has been included in other models, such as Cosserat model of Rubin and Benveniste (2004) and Dong et al. (2014), but to the authors knowledge, not in the spring layer models. Still, the main focus of this article is on the new definition of equivalent inhomogeneity and its validation through comparison of the effective properties based on that definition and the best analytical and experimental results available in the literature.

The basic assumptions and formulas behind the definition of the equivalent homogeneous representation of the inhomogeneity-interphase system are presented in Section 2. In Section 3 the probability-based technique, called the method of conditional moments, is briefly outlined and – in tandem with the results obtained in Section 2 – applied to evaluate effective properties of random composites with spring layer interphases. To validate the proposed approach some representative results are presented and compared to selected existing developments. Section 4 contains an overall discussion of the approach, presents some conclusions and evaluates potential for future extensions and applications of the approach. The paper also includes an appendix to which several details pertinent to the development included in Section 2 are relegated.

2. The properties of the equivalent inhomogeneity

2.1. A general view of the approach

The equivalent inhomogeneity is devised to represent the system consisting of the original inhomogeneity and surrounding interphase of thickness h. The interphase and the inhomogeneity are assumed elastic and have their own distinct properties. In the subsequent developments, it is assumed that the interphase is adequately represented by normal and tangentially oriented linear springs (spring layer).

The concept of the energy-equivalent inhomogeneity is essentially equivalent to a two-stage homogenization. In the first stage, using energy equivalence, the individual inhomogeneity and its interphase (spring layer) is replaced by an effective ("homogenized") inhomogeneity which combines the properties of both. In the second stage, the effective inhomogeneity is perfectly interfaced with the matrix (no interfacial jumps of any kind) and effective properties of the composite material evaluated. Any homogenization approach (numerical or analytical) can be used in the second stage.

The first stage of the process is simply a low-level homogenization step (or sub-homogenization), and like any homogenization procedure used in determining the effective properties of a system, requires the displacements on the boundary of the system (consisting of matrix and interphase, in this case) to be consistent with an average strain tensor of equivalent inhomogeneity ε_{eq}. At equilibrium these displacements cause attendant strain fields within the original inhomogeneity and displacement jumps across the interphase, both of which depend on the equivalent strains ε_{eq}. To solve for those quantities it is assumed that the original inhomogeneity undergoes a deformation described by a strain tensor ε associated with the Lurie solution for spheres (Lurie 2005). As a result, some stress components (tractions) at the boundary between the interphase and the matrix can be evaluated as a function of the properties of inhomogeneity, the properties of the interphase and the strain ε_{eq}. They can be used to evaluate the average stresses in the inhomogeneity-interphase system which leads to relations for effective properties of equivalent inhomogeneity $\mathbf{C}_{eq}$. It is emphasized, however, that if the overall (equivalent) properties of the composite material are sought, displacements at the matrix/interphase boundary are not necessarily constant – they depend on the homogenization technique used for this purpose.

2.2. Spring-layer model of the interphase

The spring-layer model of the interface has widely been used to describe the so called "soft interphase" (Hashin 1990, 1991; Achenbach and Zhu 1989; Sangani and Mo 1997; Duan et al. 2007a,b). In this model interface tractions remain continuous across the interface while displacements suffer a jump. With respect to a curvilinear orthogonal coordinate system on the sphere's surface the mathematical description of those properties is

$$[\![\sigma dS]\!]_S \cdot \mathbf{n} = [\sigma_2 dS_2 - \sigma_1 dS_1] \cdot \mathbf{n} = \mathbf{0}, \quad \mathbf{K} \cdot [\![\mathbf{u}]\!]_S = \sigma_1 \cdot \mathbf{n}. \tag{2.1}$$

The vector $\mathbf{n}$ is unit and normal to the interface between inhomogeneity and matrix. It is assumed that the normal $\mathbf{n}$ points away from the inhomogeneity. The double square brackets indicate the jump of field quantities across the interface, and the superscripts 1 and 2 indicate that the appropriate quantities are evaluated on the inhomogeneity or matrix side of the spring layer. $\mathbf{K} = K_n \mathbf{n} \otimes \mathbf{n} + K_s \mathbf{s} \otimes \mathbf{s} + K_t \mathbf{t} \otimes \mathbf{t}$ is a second-order tensor. K_n, K_s and K_t are the spring layer stiffness parameters in normal and tangential

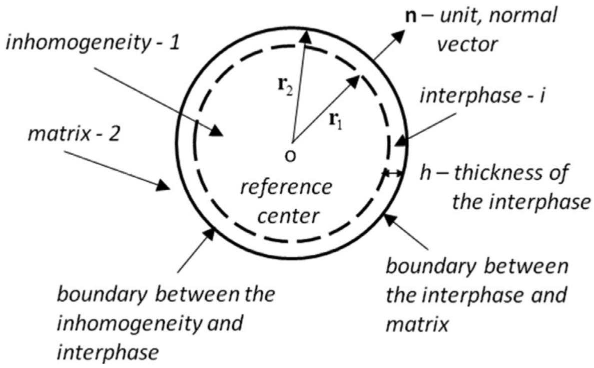

Fig. 1. Schematic illustration of inhomogeneity with interphase.

directions, respectively, and $\mathbf{s}$ and $\mathbf{t}$ represent two orthogonal unit vectors in the plane tangent to the interface.

The surface element dS introduced in Eq. $(2.1)_1$ is related to the fact that the finite thickness of the spring layer is retained in the present development. Given an infinitesimal area on the inhomogeneity side of the layer, bounded by a contour Γ, the area on the matrix side is outlined by the curve formed by intersections of the lines normal to the inhomogeneity along Γ with the matrix. Such modification of Eq. (2.1) guarantees that the equilibrium of the spring layer of finite thickness is maintained.

By Hashin (2002a), it was shown that thin and compliant interphase can accurately be modeled by a spring layer. The same conclusion was also drawn by Benveniste and Miloh (2001), and in both cases it was established that

$$K_n = \frac{\lambda_i + 2\mu_i}{h}, \quad K_t = K_s = \frac{\mu_i}{h}, \tag{2.2}$$

where λ_i and μ_i are Lame constants of the interphase. This idea is adopted in the present manuscript and properties of inhomogeneity/interphase system shown in Fig. 1 are determined using interface model described by Eq. (2.1).

In this case, the displacement jump is considered as the difference between the displacement on the interphase/matrix surface $\mathbf{u}_2$ and the one on the inhomogeneity/interface surface $\mathbf{u}_1$

$$[\![\mathbf{u}]\!] = \Delta\mathbf{u} = \mathbf{u}_2 - \mathbf{u}_1, \tag{2.3a}$$

where

$$\mathbf{u}_2 = \boldsymbol{\varepsilon}_{eq} \cdot [\mathbf{r}_1 + h\,\mathbf{n}]. \tag{2.3b}$$

$\mathbf{r}_1$ is the radius of the inhomogeneity, and h is the thickness of the interphase. The displacement vector $\mathbf{u}_1$ will be specified on the basis of Lurie solution for spheres in Sections 2.3 and 2.4. In view of the fact that the inhomogeneity is isotropic and the spring layer properties are constant (with $K_t = K_s$), the properties of the equivalent inhomogeneity will also be isotropic. Consequently, the bulk modulus and the shear modulus can be evaluated separately, which makes the analysis simpler and more efficient.

2.3. Moduli for the equivalent inhomogeneity

2.3.1. Bulk modulus

In order to evaluate the effective bulk modulus we consider hydrostatic average deformation for the inhomogeneity/spring layer system

$$\boldsymbol{\varepsilon}_{\mathrm{eq}} = \begin{bmatrix} \beta & 0 & 0 \\ 0 & \beta & 0 \\ 0 & 0 & \beta \end{bmatrix}, \tag{2.4}$$

with corresponding displacements on the interphase/matrix surface. In the spherical coordinate system (r,θ,φ), and for a spherical inhomogeneity of radius $r_1 = R$, this implies that

$$\mathbf{u}_2 = \boldsymbol{\varepsilon}_{\mathrm{eq}} \cdot \mathbf{r}|_{r=R+h} \;\Rightarrow\; u_{2[r]} = \beta[R+h], \quad u_{2[\theta]} = u_{2[\varphi]} = 0. \tag{2.5}$$

Due to complete rotational symmetry of the problem, the deformation within the original inhomogeneity is $u_r = u_r(r)$, $u_\theta = 0$ and $u_\varphi = 0$. Consequently, the displacement field vanishing at its center can be written as

$$u_r = F\frac{r}{R}, \;\; u_\theta = 0, \;\; u_\varphi = 0. \tag{2.6}$$

The constant F will be determined considering Eqs (2.1), (2.2), (2.3), (2.5) and noting that

$$u_{1[r]}|_{r=R} = F. \tag{2.7}$$

This, together with Eq. (2.5) permits to determine the displacement jumps in Eq. (2.1)

$$\Delta u_r = u_{2[r]} - u_{1[r]} = \beta\,[R+h] - F, \quad \Delta u_\theta = \Delta u_\varphi = 0. \tag{2.8}$$

In order to determine stresses within the original inhomogeneity, also entering this equation, it is noted that

$$\varepsilon_{rr} = \frac{\partial u_r}{\partial r} = \frac{F}{R}, \quad \varepsilon_{\theta\theta} = \frac{1}{r}\frac{\partial u_\theta}{\partial \theta} + \frac{u_r}{r} = \frac{F}{R}, \quad \varepsilon_{\varphi\varphi} = \frac{u_\theta}{r}\cot\theta + \frac{u_r}{r} = \frac{F}{R}, \tag{2.9}$$

and all remaining strain components vanish. Thus, as expected, the strain state within the inhomogeneity is purely volumetric, and the resulting stress state purely hydrostatic.

In particular, the radial stress within the inhomogeneity is

$$\sigma_{rr} = 2\mu_1\varepsilon_{rr} + \lambda_1 tr\boldsymbol{\varepsilon} = 2\mu_1\frac{F}{R} + 3\lambda_1\frac{F}{R} = 3K_1\frac{F}{R}, \tag{2.10}$$

where λ_1, μ_1 and K_1 are Lame constants and bulk modulus of the inhomogeneity.

The obtained results for displacement jumps and stresses are quite naturally used to determine the constant F and the effective bulk modulus. To this end, the interphase conditions of Eqs. (2.1) and (2.2) are now written in the following form

$$\sigma_{rr}|_{r=R} = K_{\mathrm{n}}\Delta u_r. \tag{2.11}$$

Accounting for (2.8) and (2.10) the above condition is

$$3K_1\frac{F}{R} = K_{\mathrm{n}}[\beta(R+h) - F]\,. \tag{2.12}$$

Introducing the dimensionless parameter $\delta = \frac{h}{R}$ and normalized spring stiffness in normal direction k_{n}, where $k_{\mathrm{n}} = RK_{\mathrm{n}}$, we determine the constant F from Eq. (2.12)

$$F = \frac{k_{\mathrm{n}}\beta R\,[1+\delta]}{3K_1 + k_{\mathrm{n}}}\,. \tag{2.13}$$

The surface element dS_1in Eq. $(2.1)_1$ is proportional to R^2 whereas dS_2 is proportional to $[R+h]^2$. Thus, combining Eqs. $(2.1)_1$, (2.10) and (2.13), one obtains

$$\sigma_2 = \frac{1}{[1+\delta]^2}\sigma_{rr}|_{r=R} = \frac{3K_1 k_{\mathrm{n}}\beta}{[1+\delta]\,[3K_1 + k_{\mathrm{n}}]}, \tag{2.14}$$

Consequently, the bulk modulus of equivalent inhomogeneity reads

$$\mathrm{K}_{\mathrm{eq}} \equiv \frac{\sigma_2}{3\beta} = \frac{K_1 k_{\mathrm{n}}}{[1+\delta]\,[3K_1 + k_{\mathrm{n}}]}. \tag{2.15}$$

Remark. In the limiting case, if the thickness of the interphase h is assumed to be negligibly small in comparison to the particle radius r (i.e., if$\delta \to 0$), the bulk modulus K_{eq} obtained here is identical with that of Hashin (1991), determined by composite assembly approach and with that of Duan et al., (2007a) computed on the basis of Eshelby solution.

2.3.2. Shear modulus

In order to evaluate effective shear modulus we consider a homogeneous deviatoric average deformation for the inhomogeneity/spring layer system. One possibility is to assume

$$\boldsymbol{\varepsilon}_{\mathrm{eq}} = \begin{bmatrix} -\beta & 0 & 0 \\ 0 & -\beta & 0 \\ 0 & 0 & 2\beta \end{bmatrix}. \tag{2.16}$$

In this case, the displacements on the interphase/matrix surface read

$$\mathbf{u}_2 = \boldsymbol{\varepsilon}_{\mathrm{eq}} \cdot \mathbf{r}|_{r=R+h} = \begin{bmatrix} -\beta\, x_2 \\ -\beta\, y_2 \\ 2\beta\, z_2 \end{bmatrix} = \begin{bmatrix} u_{2[x]} \\ u_{2[y]} \\ u_{2[z]} \end{bmatrix}, \tag{2.17}$$

with radial and tangential components

$$\begin{aligned} u_{2[r]} &= [u_{2[x]}\sin\theta + u_{2[z]}\cos\theta] \\ &= \beta\; r[2\cos^2\theta - \sin^2\theta]|_{r=R+h} \\ &= \beta\;[R+h]\,[3\cos^2\theta - 1], \\ u_{2[\theta]} &= [u_{2[x]}\cos\theta - u_{2[z]}\sin\theta] \\ &= -3\beta\; r\cos\theta\sin\theta|_{r=R+h} = -3\beta\;[R+h]\;\cos\theta\sin\theta, \\ u_{2[\varphi]} &= -[u_{2[x]}\sin\varphi + u_{2[z]}\cos\varphi] \\ &= \beta\; r[\cos\varphi\sin\varphi - \cos\varphi\sin\varphi]\,|_{r=R+h} = 0. \end{aligned} \tag{2.18}$$

Displacements on the inhomogeneity's surface for a homogeneous deviatoric deformation (accounting that the displacement at the particle center is zero) can be determined from Lurie's solution (Lurie 2005). For spherical inhomogeneity with radius R

$$u_r = \left[12\nu_1 A\frac{r^2}{R^2} + 2B\right] r\left[\frac{3}{2}\cos^2\theta - \frac{1}{2}\right],$$

$$u_\theta = -\left[(7-4\nu_1)A\frac{r^2}{R^2} + B\right] 3r\cos\theta\sin\theta, \;\; u_\varphi = 0. \tag{2.19}$$

On the surface of inhomogeneity ($r = R$) displacements are

$$u_{1[r]}|_{r=R} = [12\nu_1 A + 2B]\, R\left[\frac{3}{2}\cos^2\theta - \frac{1}{2}\right],$$
$$u_{1[\theta]}|_{r=R} = -[(7-4\nu_1)A + 2B]\, 3R\cos\theta\sin\theta, \tag{2.20}$$

and they result in the following jumps across the interphase

$$\Delta u_r = u_{2[r]} - u_{1[r]} = [2\beta(R+h) - (12\nu_1 A + 2B)R] \times \left[\frac{3}{2}\cos^2\theta - \frac{1}{2}\right],$$
$$\Delta u_\theta = u_{2[\theta]} - u_{1[\theta]} = 3[-\beta(R+h) + ((7-4\nu_1)A + B)R]\,\cos\theta\sin\theta. \tag{2.21}$$

Those jumps allow to evaluate one side of Eq. $(2.1)_2$. To evaluate the other side, the strains in the inhomogeneity are first determined to be

$$\varepsilon_{rr} = \frac{\partial u_r}{\partial r} = \left[18\nu_1 A\frac{r^2}{R^2} + B\right]\left[3\cos^2\theta - 1\right], \tag{2.22}$$

$$\varepsilon_{\theta\theta} = \frac{1}{r}\frac{\partial u_\theta}{\partial\theta} + \frac{u_r}{r} = -3\left[2(7-7\nu_1)A\frac{r^2}{R^2} + B\right]\cos^2\theta + \left[3(7-6\nu_1)A\frac{r^2}{R^2} + 2B\right], \tag{2.22}$$

$$\varepsilon_{\varphi\varphi} = \frac{u_\theta}{r}\cot\theta + \frac{u_r}{r} = \left[-3(7-10\nu_1)A\frac{r^2}{R^2}\right]\cos^2\theta - \left[6\nu_1 A\frac{r^2}{R^2} + B\right], \tag{2.22}$$

$$2\varepsilon_{r\theta} = \gamma_{r\theta} = \frac{1}{r}\frac{\partial u_r}{\partial\theta} + \frac{\partial u_\theta}{\partial r} - \frac{u_\theta}{r} = -6\left[(7+2\nu_1)A\frac{r^2}{R^2} + B\right]\cos\theta\sin\theta. \tag{2.22}$$

All remaining strain components vanish. The trace of the strain tensor is

$$tr\boldsymbol{\varepsilon} = \varepsilon_{rr} + \varepsilon_{\theta\theta} + \varepsilon_{\varphi\varphi} = A\frac{r^2}{R^2}[1-2\nu_1]\left[1 - 3\cos^2\theta\right], \tag{2.23}$$

which gives

$$\sigma_{rr} = 2\mu_1\varepsilon_{rr} + \lambda_1 tr\boldsymbol{\varepsilon} = 2\mu_1\left[18\nu_1 A\frac{r^2}{R^2} + B\right]\left[3\cos^2\theta - 1\right] - 21\lambda_1 A\frac{r^2}{R^2}[1-2\nu_1]\left[3\cos^2\theta - 1\right],$$
$$\sigma_{r\theta} = 2\mu_1\varepsilon_{r\theta} = -6\mu_1\left[(7+2\nu_1)A\frac{r^2}{R^2} + B\right]\cos\theta\sin\theta. \tag{2.24}$$

These are the only stress components that enter Eq. $(2.1)_2$. The constants A and B are computed in terms of β from the interface conditions (2.1) and (2.2) (see details in Eqs. (A.8)–(A.10))

$$\begin{cases} A = \dfrac{(1+\delta)\beta}{M}[(2\mu_1 + k_t)\,k_n - [2\mu_1 + k_n]\,k_t] \\ B = \dfrac{(1+\delta)\beta}{M}[6\nu_1\,[k_n - \mu_1]\,k_t \\ \quad -[7\,(2\mu_1 + k_t) + 4\nu_1\,(\mu_1 - k_t)]\,k_n] \end{cases}, \tag{2.25}$$

and

$$M = -[4{\mu_1}^2[7+5\nu_1] + 2\mu_1[(7-4\nu_1)k_n + (7-\nu_1)k_t] + [7-10\nu_1]k_n k_t], \tag{2.26}$$

where $k_t = RK_t$. With the constants A and B defined, the average stresses within the inhomogeneity/interphase system can be evaluated and μ_{eq} can be determined from any of the following equations

$$2\mu_{eq} \equiv -\frac{\bar{S}_{xx}}{\beta} = -\frac{\bar{S}_{yy}}{\beta} = \frac{\bar{S}_{zz}}{2\beta}, \tag{2.27}$$

where $\bar{S}_{xx}$, $\bar{S}_{yy}$ and $\bar{S}_{zz}$ are the average deviatoric stresses. Considering that $\bar{S}_{xx} = \bar{S}_{yy} = -2\bar{S}_{zz}$ only $\bar{S}_{xx}$ will be evaluated:

$$\bar{S}_{xx} = \bar{\sigma}_{xx} - \frac{1}{3}tr\boldsymbol{\sigma} = \frac{1}{3}[\bar{\sigma}_{xx} - \bar{\sigma}_{zz}]. \tag{2.28}$$

The appropriate average stress formula for the inhomogeneity/interphase system is (Benveniste and Miloh 2001)

$$\bar{\sigma}_{ij} = \frac{1}{V}\int_{\partial V_2} t_i x_j dS, \tag{2.29}$$

where t_i is the traction and the integration is over the interphase/matrix boundary ∂V_2. The position vectors on the interphase/inhomogeneity and interphase/matrix surfaces have the following components respectively:

$$x_1 = R\sin\theta\,\cos\varphi,\ y_1 = R\sin\theta\,\sin\varphi,\ z_1 = R\cos\theta,$$
$$x_2 = [R+h]\sin\theta\,\cos\varphi,\ y_2 = [R+h]\sin\theta\,\sin\varphi,$$
$$z_2 = [R+h]\cos\theta. \tag{2.30}$$

Considering Eqs. $(2.1)_1$ and (2.29), the components $\bar{\sigma}_{xx}$ and $\bar{\sigma}_{zz}$needed in Eq. (2.28) are determined by

$$\bar{\sigma}_{xx} = \frac{1}{V_2}\int_{\partial V_2}\frac{R^2}{[R+h]^2}t_x x_2[R+h]^2\sin\theta\,d\theta\,d\varphi = \frac{1}{V_2}\int_{\partial V_2}R^2 t_x x_2\sin\theta\,d\theta\,d\varphi, \tag{2.31a}$$

$$\bar{\sigma}_{zz} = \frac{1}{V_2}\int_{\partial V_2}R^2 t_z z_2\sin\theta\,d\theta\,d\varphi\,. \tag{2.31b}$$

Taking into account that for r=R

$$\sigma_{rr} = t_r,\quad \sigma_{r\theta} = t_\theta, \tag{2.32}$$

t_x and t_z have the following form

$$\begin{aligned} t_x &= [\sigma_{rr}\sin\theta + \sigma_{r\theta}\cos\theta]\cos\varphi, \\ t_z &= [\sigma_{rr}\cos\theta - \sigma_{r\theta}\sin\theta]\,. \end{aligned} \tag{2.33}$$

Furthermore, from Eq. (2.24) σ_{rr} and $\sigma_{r\theta}$ evaluated at $r = R$ read

$$\begin{aligned}\sigma_{rr} &= 2\mu_1[18\nu_1 A + B]\left[3\cos^2\theta - 1\right] \\ &\quad + 21\cdot 2\mu_1\nu_1 A\left[3\cos^2\theta - 1\right] \\ &= 2\mu_1[-3\nu_1 A + B]\left[3\cos^2\theta - 1\right], \end{aligned} \tag{2.34a}$$

$$\sigma_{r\theta} = -6\mu_1[(7+2\nu_1)A + B]\ \cos\theta\sin\theta\ . \tag{2.34b}$$

Substituting Eq. (2.34) into Eq. (2.33) the tractions on the inhomogeneity's surface become

$$\begin{aligned} t_x &= [-6\mu_1 A(7+5\nu_1)\cos^2\theta\sin\theta \\ &\quad + \mu(6\nu_1 A - 2B)\sin\theta]\cos\varphi, \end{aligned} \tag{2.35a}$$

$$\begin{aligned} t_z &= 2\mu_1[-3\nu_1 A + B]\left[3\cos^2\theta - 1\right]\cos\theta \\ &\quad + 6\mu_1[(7+2\nu_1)A + B]\ \cos\theta\sin^2\theta. \end{aligned} \tag{2.35b}$$

Eqs. (2.35) and (2.30) substituted into Eq. (2.31) yield

$$\bar{\sigma}_{xx} = \frac{-2\mu_1}{5[1+\delta]^2}[21A + 5B], \tag{2.36a}$$

$$\bar{\sigma}_{zz} = \frac{4\mu_1}{5[1+\delta]^2}[21A + 5B]. \tag{2.36b}$$

Given that μ_{eq}resulting from Eqs. (2.27) and (2.28) is

$$\begin{aligned} 2\mu_{eq} &= -\frac{\bar{\sigma}_{xx} - \bar{\sigma}_{zz}}{3\beta} = \frac{21\mu_1}{5\beta[1+\delta]^2}[21A + 5B] \\ &= \frac{2\mu_1}{5\beta[1+\delta]^2}[21A + 5B], \end{aligned} \tag{2.37}$$

one gets

$$\begin{aligned} 2\mu_{eq} &= \frac{2\mu_1}{5[1+\delta]} \\ &\times \frac{4\mu_1[7+5\nu_1][2k_n + 3k_t] + 5[7-10\nu_1]k_n k_t}{\left[4\mu_1{}^2(7+5\nu_1) + 2\mu_1((7-4\nu_1)k_n + (7-\nu_1)k_t) + (7-10\nu_1)k_n k_t\right]}. \end{aligned} \tag{2.38}$$

It is noted that in the limit, if $\delta \to 0$, an equivalent inhomogeneity with imperfect interface is obtained. Furthermore,

(a) if $k_n \to \infty$ and $k_t \to \infty$

$$2\mu_{eq} = \frac{2\mu_1}{[1+\delta]}, \tag{2.39}$$

which reduces to $2\mu_{eq} = 2\mu_1$, if $\delta = 0$. This represents the perfect interface condition.

(b) if $k_n = k_t = 0$ $2\mu^{eq} = 0$, and the inhomogeneity behaves like a cavity,
(c) if $k_n \to \infty$ but k_t is finite (if $k_t = 0$, it is called "free sliding", Hashin (2002a); Duan et al. (2007b))

$$2\mu_{eq} = \frac{2\mu_1}{5[1+\delta]}\frac{8\mu_1[7+5\nu_1] + 5[7-10\nu_1]k_t}{[2\mu_1(7-4\nu_1) + (7-10\nu_1)k_t]}. \tag{2.40}$$

Remark. It should be noted that the concept of equivalent inhomogeneity presented here is quite different from the idea employed by Duan et al. (2007a) and Gu et al. (2014). In those papers the equivalent shear modulus μ_{eq} was determined on the basis of Eshelby solution by comparing the change in energy if the equivalent inhomogeneity is inserted in the infinite medium under far-field load and the change due to similar insertion of the original inhomogeneity together with the interface, (Eshelby 1957). As a result, μ_{eq} depends on the properties of that medium (matrix material). This is nonphysical, and in sharp contrast to the equivalent inhomogeneity approach advocated by, e.g., Hashin (2002a), and is explicitly excluded in the approach presented here.

The equivalent inhomogeneities described above may be used in conjunction with *any method* employed in evaluation of the effective properties of composites without interphases. The *method of choice* in this work is the Method of Conditional Moments (MCM) – a rigorous formal approach to analysis of random composites. The MCM is based on statistical analysis where the random structure of the material is entirely described by conditional probabilities, which – in comparison with the data specifying deterministic structures – is the only additional data. The main features of the MCM and some basic results associated with standard composites (without interphases) will be briefly outlined in the subsequent section.

3. Effective properties of random composites

3.1. The fundamentals of the method of conditional moments

We examine a representative macro-volume V of a linear elastic composite consisting of a matrix with randomly distributed particles. It is assumed that at each point of macro-volume $\mathbf{x}$ the Hooke's law is valid

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{C}(\mathbf{x}) : \boldsymbol{\varepsilon}(\mathbf{x}), \tag{3.1}$$

where $\boldsymbol{\sigma}(\mathbf{x})$ and $\varepsilon(\mathbf{x})$ are the stress and strain tensors, and $\mathbf{C}(\mathbf{x})$ is the material stiffness tensor. Here it is assumed that $\mathbf{C}(\mathbf{x})$ is statistically uniform random function of coordinates with a finite scale of correlation, whose one-point density distribution has the following form:

$$f(\mathbf{C}(\mathbf{x})) = \sum_{k=1}^{2} c_k \delta(\mathbf{C}(\mathbf{x}) - \mathbf{C}_k), \tag{3.2}$$

where $\delta(\bullet)$ denotes the Dirac delta function, c_1, c_2 and $\mathbf{C}_1$, $\mathbf{C}_2$ are volume fractions and stiffness tensors of the matrix and of the particles, respectively.

If the representative volume is under a uniform load, the resulting stresses and strains form statistically uniform random fields satisfying the property of ergodicity. This allows us to replace the averaging over representative volume by averaging over an ensemble of realizations (e.g., Gray 2009; Gnedenko 1962). In this case, the following relationship exists between macroscopic fields of stress $\bar{\boldsymbol{\sigma}}$ and strain $\bar{\boldsymbol{\varepsilon}}$:

$$\bar{\boldsymbol{\sigma}} = \mathbf{C}^* : \bar{\boldsymbol{\varepsilon}}, \tag{3.3}$$

where $\mathbf{C}*$ is the effective stiffness tensor, and the overbar $\bar{\bullet}$ denotes statistical averaging.

Having averaged Eq. (3.1) we obtain

$$\bar{\sigma} = \sum_{k=1}^{2} c_k \mathbf{C}_k : \bar{\varepsilon}_k, \quad \bar{\varepsilon} = \sum_{k=1}^{2} c_k \bar{\varepsilon}_k, \tag{3.4}$$

where $\bar{\varepsilon}_k = \langle \varepsilon(\mathbf{x})|_k^{(\mathbf{x})} \rangle$ is the expectation value of the strain tensor at point $\mathbf{x}$, provided that this point belong to the kth component.

It is evident by comparison of Eqs. (3.3) and (3.4) that in order to determine the effective stiffness tensor $\mathbf{C}*$ it is sufficient to find the relationship between the mean strain in the component $\bar{\varepsilon}_k$ (e.g., in the particles $\bar{\varepsilon}_1$) and the mean strain in the macroscopic volume $\bar{\varepsilon}$. In fact, if this relationship has the form:

$$\bar{\varepsilon}_1 = \mathbf{A} : \bar{\varepsilon}, \tag{3.5}$$

then, considering that $\bar{\varepsilon} = c_1 \bar{\varepsilon}_1 + c_2 \bar{\varepsilon}_2$ and using Eqs. (3.5) and (3.4), the relationship for the effective stiffness tensor reads

$$\mathbf{C}^* = \bar{\mathbf{C}} + c_1 \mathbf{C}_3 : \left[\mathbf{A} - \overset{4}{\mathbf{I}} \right]. \tag{3.6}$$

$\overset{4}{\mathbf{I}}$ is the fourth-order unit tensor, $\bar{\mathbf{C}}$ is the expectation value of the stiffness tensor

$$\bar{\mathbf{C}} = \sum_{k=1}^{2} c_k \mathbf{C}_k, \quad \mathbf{C}_3 = \mathbf{C}_1 - \mathbf{C}_2 . \tag{3.7}$$

To derive the formula for the rank four tensor $\mathbf{A}$ of Eq. (3.5), associated with a random material the following process is used in the MCM. First, the equilibrium equations for elastic medium and Eq. (3.1) lead to the following stochastic differential equation for the fluctuations $\mathbf{u}^0$of the random field of displacements (Khoroshun et al., 1993; Nazarenko et al. 2009):

$$div\left(\mathbf{C}_c : sym\left(\nabla \mathbf{u}^0(\mathbf{x})\right)\right) + div\left(\mathbf{C}^0(\mathbf{x}) : \varepsilon(\mathbf{x})\right) = \mathbf{0},$$
$$\mathbf{u}^0(\mathbf{x})\,|_\infty = \mathbf{0}, \tag{3.8}$$

where $\mathbf{C}^0(\mathbf{x}) = \mathbf{C}(\mathbf{x}) - \mathbf{C}_c$, $\mathbf{u}^0(\mathbf{x}) = \mathbf{u}(\mathbf{x}) - \bar{\varepsilon} \cdot \mathbf{x}$. Although the above equation is valid for any constant tensor $\mathbf{C}_c$, the accuracy of the MCM is enhanced if it is selected as follows (Khoroshun et al., 1993; Nazarenko et al. 2015):

$$\mathbf{C}_c = \begin{cases} \bar{\mathbf{C}}, & \text{if} \quad \mathbf{C}_1 \le \mathbf{C}_2 \\ \left(\overline{\mathbf{C}^{-1}}\right)^{-1}, & \text{if} \quad \mathbf{C}_1 \ge \mathbf{C}_2 \end{cases}. \tag{3.9}$$

Using the Green tensor, the solution of Eq. (3.9) is written in form of an integral over the infinite region V (see Nazarenko et al. 2009, 2014)

$$\mathbf{u}^0(\mathbf{x}) = \int_{V_y} \mathbf{G}(\mathbf{x}-\mathbf{y}) \cdot div\left(\mathbf{C}^0(\mathbf{y}) : \varepsilon(\mathbf{y}) - \boldsymbol{\beta}\right) dV_y, \tag{3.10}$$

where the Green tensor is the solution of the following system of differential equations:

$$div(\mathbf{C}_c : \nabla \mathbf{G}(\mathbf{x})) + \delta(\mathbf{x})\, \overset{2}{\mathbf{I}} = \mathbf{0}, \quad \mathbf{G}(\mathbf{x})\,|_\infty = \mathbf{0}. \tag{3.11}$$

Integration of Eq. (3.10) by parts and use of linear kinematic relations, leads to a stochastic integral equation for the random strain field

$$\varepsilon(\mathbf{x}) = \bar{\varepsilon} + \mathbf{K}(\mathbf{x}-\mathbf{y}) * \left[\mathbf{C}^0(\mathbf{y}) : \varepsilon(\mathbf{y})\right]. \tag{3.12}$$

The above operator notation for $\mathbf{K}(\mathbf{x}-\mathbf{y})$ has the following interpretation

$$\mathbf{K}(\mathbf{x}-\mathbf{y}) * \boldsymbol{\psi}(\mathbf{y}) = \int_V sym\left(\nabla_x(\nabla_x \mathbf{G}(\mathbf{x}-\mathbf{y}))\right) : \left(\boldsymbol{\psi}(\mathbf{y}) - \bar{\boldsymbol{\psi}}\right) dV_y, \tag{3.13}$$

where $\bar{\varepsilon}$ and $\bar{\boldsymbol{\psi}}$ are mean (expectation) values of $\varepsilon(\mathbf{x})$ and $\boldsymbol{\psi}(\mathbf{y})$.

Applying the technique of conditional averaging (see Khoroshun et al., 1993; Nazarenko et al. 2009) with respect to the multipoint conditional densities and limiting the process to a two-point approximation – which is tantamount to assuming identical strain distributions in all inhomogeneities the following linear algebraic equation, involving conditional one-point moments of the strain fields $\bar{\varepsilon}_\nu$, is obtained:

$$\bar{\varepsilon}_\nu = \bar{\varepsilon} + \sum_{k=1}^{2} \mathbf{K}^{\nu k} : \mathbf{C}_k^0 : \bar{\varepsilon}_k, \quad k, \nu \in \{1,\ 2\}, \tag{3.14}$$

where

$$\mathbf{K}^{\nu k} = \mathbf{K}(\mathbf{x}-\mathbf{y}) * p_{\nu k}(\mathbf{x}-\mathbf{y}). \tag{3.15}$$

The function $p_{\nu k}(\mathbf{x}-\mathbf{y})$ denotes the probability that point $\mathbf{x}$ belongs to the kth component, provided point $\mathbf{y}$ belongs to the νth component.

Clearly, Eq. (3.14) along with the previously used relationship $\bar{\varepsilon} = c_1 \bar{\varepsilon}_1 + c_2 \bar{\varepsilon}_2$, constitutes a system of two (tensorial) equations allowing to determine both $\bar{\varepsilon}_1$ and $\bar{\varepsilon}_2$ in terms of $\bar{\varepsilon}$. This defines tensor $\mathbf{A}$ of Eq. (3.5) and allows to evaluate the effective properties according to Eq. (3.6).

3.2. Closed-form formulas for effective properties of random composites with spherical particles and perfect interfaces

In order to specify Eq. (3.14) one must determine the two-point conditional probabilities$p_{\nu k}(\mathbf{x}-\mathbf{y})$ which characterize shape and arrangement of the inhomogeneities. This allows evaluating the convolution $\mathbf{K}(\mathbf{x}-\mathbf{y}) * p_{\nu k}(\mathbf{x}-\mathbf{y})$ of Eq. (3.15) following the general formula shown in Eq. (3.13).

Details of the evaluation of the effective stiffness tensor for composites with randomly distributed spherical particles are presented by Khoroshun et al., 1993; Nazarenko et al., 2014, 2015. Following the methodology described in those papers, the following expression for tensor $\mathbf{A}$ of Eq. (3.5) is obtained:

$$\mathbf{A} = \overset{4}{\mathbf{I}} + c_2 \left[\overset{4}{\mathbf{I}} - \mathbf{L} : \mathbf{C}'\right]^{-1} : \mathbf{L} : \mathbf{C}_3, \tag{3.16}$$

Its use in Eq. (3.6) yields

$$\mathbf{C}^* = \bar{\mathbf{C}} + c_1 \mathbf{C}_3 : \left[\overset{4}{\mathbf{I}} - \mathbf{L} : \mathbf{C}'\right]^{-1} : \left[c_2 \mathbf{L} : \mathbf{C}_3\right], \tag{3.17}$$

where $\bar{\mathbf{C}}$, $\mathbf{C}_3$ are determined in Eq. (3.7) and

$$\mathbf{C}' = c_1 \mathbf{C}_2 + c_2 \mathbf{C}_1 - \mathbf{C}_c. \tag{3.18}$$

Tensor $\mathbf{L}$ of Eq. (3.17) is an isotropic rank four tensor (the classical Hill tensor, see Mura 1987)

$$\mathbf{L} = 2b\, \overset{4}{\mathbf{I}} + a\, \overset{2}{\mathbf{I}} \otimes \overset{2}{\mathbf{I}}, \tag{3.19}$$

with

$$a = \frac{\lambda_c + \mu_c}{15\mu_c[\lambda_c + 2\mu_c]}, \quad b = -\frac{3\lambda_c + 8\mu_c}{30\mu_c[\lambda_c + 2\mu_c]}, \tag{3.20}$$

and with λ_c, μ_c being the Lamé constants of the reference medium defined according to Eq. (3.9).

In the spirit of the approach proposed here, the overall properties of the composite with the spring-layer model of interphases are obtained from Eqs. (3.17)–(3.20), valid for perfect interfaces, if the properties of the inhomogeneity ($\mathbf{C}_1$) are replaced with the properties of equivalent inhomogeneities ($\mathbf{C}_{\text{eq}}$) discussed in Sections 2.1 and 2.2. The effective bulk and shear moduli of the equivalent inhomogeneity are determined in Section 2.3. This leads to the following result

$$\mathbf{C}^* = \tilde{\tilde{\mathbf{C}}} + c_1\tilde{\mathbf{C}}_3 : \left[\overset{4}{\mathbf{I}} - \tilde{\mathbf{L}} : \tilde{\mathbf{C}}'\right]^{-1} : \left[c_2\tilde{\mathbf{L}} : \tilde{\mathbf{C}}_3\right], \tag{3.21}$$

where $\tilde{\tilde{\mathbf{C}}}$, $\tilde{\mathbf{C}}_3$, $\tilde{\mathbf{L}}$, and $\tilde{\mathbf{C}}'$ are determined in accordance with Eqs (3.7), (3.18)–(3.20) with $\mathbf{C}_1$ replaced by the stiffness tensor of the equivalent inhomogeneity $\mathbf{C}_{\text{eq}}$. As discussed earlier, the fourth order isotropic tensor $\mathbf{C}_{\text{eq}}$ is specified by bulk K_{eq} and shear μ_{eq} moduli of the equivalent inhomogeneity determined in Eqs. (2.15) and (2.40).

The scalar expression for the effective bulk and shear moduli of the composite can be extracted from a general tensorial formula (3.21) (cf. Nazarenko et al. 2014, 2015):

$$K^* = c_1\mathrm{K}_{\text{eq}} + c_2K_2 - \frac{c_1c_2[\mathrm{K}_{\text{eq}} - K_2]^2}{c_1K_2 + c_2\mathrm{K}_{\text{eq}} + 4/3\tilde{\mu}_c}, \tag{3.22}$$

$$\mu^* = c_1\mu_{\text{eq}} + c_2\mu_2 + \frac{4c_1c_2\tilde{b}[\mu_{\text{eq}} - \mu_2]^2}{1 - 4\tilde{b}[c_2\mu_{\text{eq}} + c_1\mu_2 - \tilde{\mu}_c]}, \tag{3.23}$$

where

$$\tilde{b} = -\frac{3\left[\tilde{K}_c + 2\tilde{\mu}_c\right]}{10\tilde{\mu}_c[3\tilde{K}_c + 4\tilde{\mu}_c]}, \tag{3.24}$$

and $\tilde{K}_c$, $\tilde{\mu}_c$ in above equations are specified as follows:

$$\tilde{K}_c = \begin{cases} c_1\mathrm{K}_{\text{eq}} + c_2K_2, & \text{if} \quad \mathrm{K}_{\text{eq}} \le K_2 \\ \left[c_1(\mathrm{K}_{\text{eq}})^{-1} + c_2(K_2)^{-1}\right]^{-1}, & \text{if} \quad \mathrm{K}_{\text{eq}} \ge K_2 \end{cases}, \tag{3.25}$$

$$\tilde{\mu}_c = \begin{cases} c_1\mu_{\text{eq}} + c_2\mu_2, & \text{if} \quad \mu_{\text{eq}} \le \mu_2 \\ \left[c_1(\mu_{\text{eq}})^{-1} + c_2(\mu_2)^{-1}\right]^{-1}, & \text{if} \quad \mu_{\text{eq}} \ge \mu_2 \end{cases}. \tag{3.25}$$

In the above formulasK_2 and μ_2 are the bulk and shear moduli of the matrix, whereas K_{eq} and μ_{eq} are determined in Eqs. (2.15) and (2.40).

3.3. Numerical results and comparisons

In order to illustrate the effect of imperfect interface we consider an epoxy matrix with the properties $E_2 = 3.45$ GPa and $\nu_2 = 0.35$, containing randomly distributed glass spheres with $E_1 = 72.4$ GPa and $\nu_1 = 0.2$, following Hashin (1991). The volume fraction of particles is $c_1 = 0.4$. The value of the parameter $\delta = h/R$ was taken to be $\delta =$ 0since the results obtained in this work are compared with those obtained assuming that values of δ.

As the first example the effect of degradation of interface properties, from perfect interface to total disbonding,

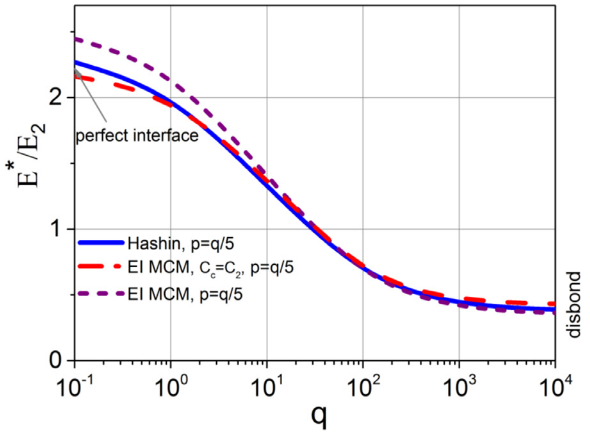

Fig. 2. Dependence of normalized Young's modulus $E*/E_2$ on tangential and normal bond parameters $q = 1/(RK_t)$ and $p = 1/(RK_n)$.

on the effective properties of the entire composite is considered and compared with the results obtained by Hashin (1991). In this example it is assumed that degradation of the normal (p) and tangential (q) interactions between the inclusion and the matrix is proportional ($p = q/5$), which is likely to be the most realistic scenario (although the coefficient of proportionality between p and q may be different than 5). Here, p and q are bond parameters adopted from Hashin (1991), where $p = 1/(RK_n)$, $q = 1/(RK_t)$ and K_n and K_t are previously introduced springs stiffness constants in the normal and tangential directions, cf. Eqs. (2.1) and (2.2).

The variation of the normalized effective Young's modulus $E*/E_2$ with the shear bond parameter q, and proportional variation of the normal parameters $p = q/5$, calculated by the MCM in combination with equivalent inhomogeneity approach – denoted by EI MCM – is shown in Fig. 2. In accordance to the remarks following Eq. (2.38), the results for the increasingly small and large p and q correspond to those calculated for particle with perfect interface and for voids, respectively. For comparison, the normalized Young's modulus for the same material obtained on the basis of the self-consistent scheme by Hashin (1991) is also shown in Fig. 2. The results obtained by MCM with equivalent inhomogeneity approach are clearly in good agreement with numerical results by Hashin (1991). Discrepancy is discernable for small values of the spring layer parameters, but even then it is within just a few percent. Moreover, in a particular case whereby the matrix is chosen as a reference medium (EI MCM, $\mathbf{C}_c = \mathbf{C}_2$), the results obtained by MCM are virtually identical with the results by Hashin, as shown in Fig. 2, even though the choice of the matrix as a reference medium is not optimal from the point of view of MCM. However, inclusion of the results obtained for precisely that selection of the reference medium in Figs 2 and 3, provides an additional illustration of some connection between the MCM and the self-consistent approach of Hashin (1991).

It is remarkable that all results in Figs. 2 and 3 are close to one another, in spite of rather fundamentals differences between the methods involved. The MCM is more

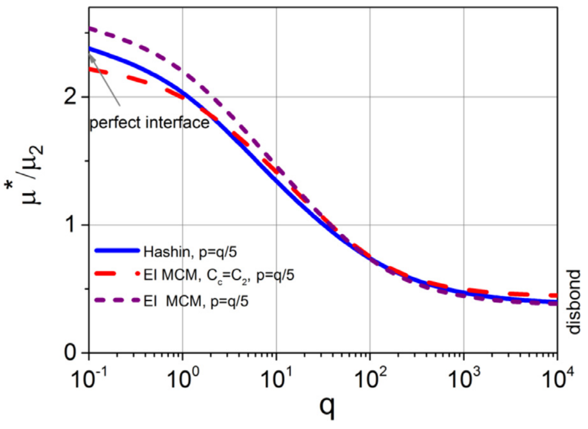

Fig. 3. Dependence of normalized shear modulus $\mu*/\mu_2$ on tangential and normal bond parameters $q = 1/(RK_t)$ and $p = 1/(RK_n)$.

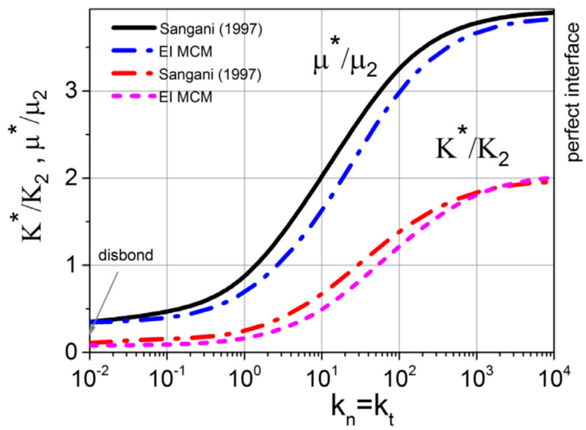

Fig. 4. Dependence of normalized bulk $K*/K_2$ and shear $\mu*/\mu_2$ moduli on tangential and normal bond parameters $k_t = RK_t$ and $k_n = RK_n$ if $k_n = k_t$.

formal and mathematical, and starts with the exact model to be analyzed, but in the solution process limits the number of conditional moments used in the analysis to two-point moments. The self-consistent method, on the other hand, is based on some intuitive arguments leading to an approximate model that needs to be analyzed.

The tendency observed in Fig. 2 for Young's modulus is also seen in Fig. 3 where variation of shear modulus is shown (the normal and tangential stiffness parameters vary proportionally and identically as in the case of Fig. 2). Good agreement between the results obtained by MCM in combination with equivalent inhomogeneity approach and the numerical results by Hashin (1991) is remarkable given the closed-form formulas derived in this work. The source of somewhat higher (but still surprisingly small) discrepancy for small values of those parameters is rather difficult to pinpoint, considering that both approaches are based on different set of approximations.

As a second numerical example a matrix (with $\nu_1 = 0.45$) containing randomly distributed rigid spherical particles ($K_1/K_2 = \infty$, $\mu_1/\mu_2 = \infty$) is considered. The volume fraction of particles is $c_1 = 0.45$. In this example the effects of interface degradation is also investigated but, for the sake of comparison with the results reported in the literature, the results are presented slightly differently. This problem is analyzed by Sangani and Mo (1997), who solved the governing differential equations of the problem using (truncated) series expansion. Accounting for the fact that the effect of weak interphase is most pronounced when the inhomogeneities are rigid, and considering that the approach of Sangani and Mo is formally exact, comparison of the result obtained here with those of Sangani and Mo is more meaningful than the comparison presented in the previous example.

Dependence of the effective normalized bulk and shear moduli on tangential and normal bond parameters are shown in Figs. 4– 6. In all cases, the effects of variation (degradation) of the interface properties (practically all the way up to total disbonding) on the effective properties of the entire composite are analyzed and presented along with those obtained by Sangani and Mo (1997).

Based on Fig. 4 it is concluded that the results obtained by MCM with equivalent inhomogeneity approach are in very good agreement with the numerical results by Sangani and Mo (1997), if the degradation of interphase properties in normal and tangential directions is proportional. Particularly good agreement is observed if the spring layer is either very soft or very stiff. In the limits of those two cases the effects of interphase vanish and the notion of equivalent inhomogeneity yields precise values of effective properties: cavity in the first case and the original inhomogeneity (with perfect interface) in the second (see comments following Eq. (2.38)). Thus, such an excellent agreement of the results in those two limiting situations confirms the high accuracy of the MCM used here (even for quite high volume fraction of rigid inhomogeneities), since the possible error associated with approximate definition of equivalent inhomogeneity is eliminated. This observation also indicates that the approximate inclusion of the interphase effects in the definition of equivalent inhomogeneity may be responsible for the error in the middle range of interphase parameters K_n and K_t. Other factors, alluded to at the end of this section, may also affect the discrepancy seen in Figs. 2 and 3.

Similarly good agreement for the effective normalized bulk and shear moduli is observed for variation of the tangential bond parameter from 10^{-2} to 10 if the normal bond parameter is equal to zero, Fig. 5, and for variation of the normal bond parameter from 10^{-2} to 10 if the tangential bond parameter is equal to zero, Fig. 6. Quite surprisingly the bulk modulus in Fig. 6, where $k_t = 0$, shows somewhat pronounced discrepancy, given its almost perfect agreement in Fig. 5, where $k_n = 0$.

Some general comments appear to be relevant regarding the comparison with the formally exact results of Sangani and Mo (1997). The first one relates to the fact that the values they report are obtained using a displacement-based approach. As such, the approach provides an upper bound of the bulk and shear moduli. With that in mind the fact that our results are generally below the values reported by Sangani and Mo is a good sign.

Table 1
Comparison of the effective bulk and shear moduli obtained by EI MCM with experimental data.

	Hashin Monteiro analysis	Hashin Monteiro analysis	Hashin Monteiro experiment	EI MCM	Discrepancy with experiment	Hashin Monteiro experiment	EI MCM	Discrepancy with experiment
c_1	μ_i/μ_2	ν_i	$K^*_{\exp}$ (Gpa)	$K^*_{\rm anl}$ (Gpa)	$\frac{K^*_{\exp}-K^*_{\rm anl}}{K^*_{\exp}}$	$\mu^*_{\exp}$ (Gpa)	$\mu^*_{\rm anl}$ (Gpa)	$\frac{\mu^*_{\exp}-\mu^*_{\rm anl}}{\mu^*_{\exp}}$
0.15	0.467	0.316	24.14	22.51	6.7 %	13.35	13.54	1%
0.27	0.475	0.400	26.81	24.18	9.8 %	14.87	15.21	2%
0.40	0.553	0.313	27.69	25.23	8.8 %	16.91	17.43	3%
0.52	0.612	0.311	29.96	26.42	11.2 %	19.26	19.89	3%

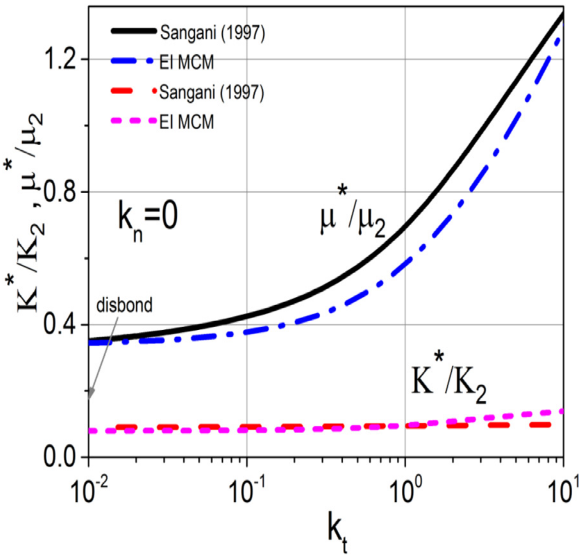

Fig. 5. Dependence of normalized bulk $K*/K_2$ and shear $\mu*/\mu_2$ moduli on tangential bond parameter k_t if $k_n = 0$.

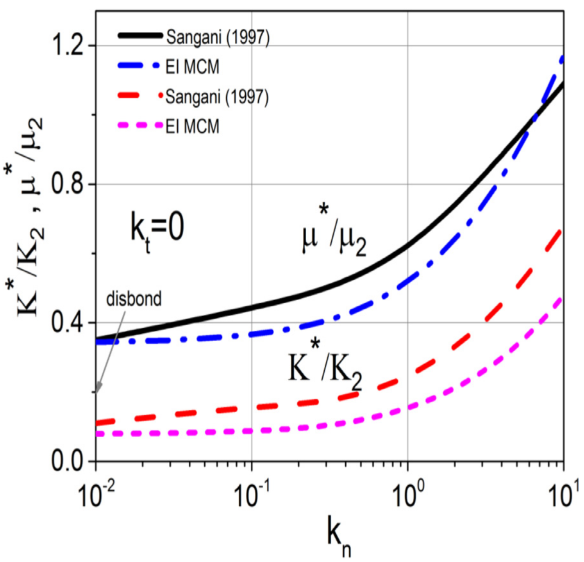

Fig. 6. Dependence of normalized bulk $K*/K_2$ and shear $\mu*/\mu_2$ moduli on normal bond parameter k_n if $k_t = 0$.

The second comment relates to the practical validity of the results obtained by varying one of the parameters k_t, k_n while the other completely vanishes. In this work, the analysis assuming the same variation of k_t, k_n was also followed for the comparison's sake. While such a choice is an illustration of the versatility of the approach used to solve the problem, its practical validity may be questioned. For example, assuming $k_t = 0$ everywhere is unrealistic because, under hydrostatic compression or under shear, zones of normal compression must exist within the interphase which, due to friction, would have to result in some tangential interactions. For the same reasons the assumption of $k_n = 0$ is not realistic. Realization of that, in conjunction with the first comment above, makes the comparison presented in Fig. 4 much more important in judging the quality of the results obtained herein. One can thus conclude that those results are very good, indeed.

Next, the results obtained by the equivalent inhomogeneity approach proposed herein in combination with the method of conditional moments (EI MCM) are compared with the experimental data presented by Hashin and Monteiro (2002b). The material used in that work is the cement paste matrix with $K_2 = 22.51$ GPa and $\mu_2 = 11.8$ GPa containing randomly distributed sand particles with $K_1 = 44$ GPa and $\mu_1 = 37$ GPa. There is an interphase layer around sand with elastic constant μ_i and ν_i presented in Table 1, which Hashin and Monteiro (2002b) obtained via inverse analysis for various volume fractions c_1 of sand particles. The averaged diameter of sand particles is 850 μm while interphase thickness was estimated at 25 μm.

In the inverse analysis of Hashin and Monteiro (2002b) the interphase properties $\mu_i$and ν_i were obtained by adjusting their values to match the analytically derived overall properties of the material with those determined experimentally. The analytical part was based on the generalized self-consistent scheme employing the exact Lurie solution for a sphere surrounded by the interphase layer of thickness h. The resulting properties of the interphase are about half of the properties of the cement paste. Thus, the spring layer appears to be adequate to model the cement based material considered by Hashin and Monteiro (2002b) provided the parameters (K_n, K_t) are evaluated according to Eq. (2.2), as suggested by Hashin (2002a) (cf. Benveniste and Miloh, 2001). So determined spring layer stiffness parameters are used herein to obtain the effective properties of the composite as specified in Eqs (2.15), (2.40), (3.22), (3.23).

It is seen in Table 1 that EI MCM predictions of the effective shear modulus are in good overall agreement with experimental values, even for high volume fraction of particles c_1. While it is hard to precisely pinpoint why the bulk modulus exhibits higher discrepancy, in general differences between predictions of the EI MCM approach and experiment were to be expected. One reason may be the transition from the continuous interphase model used by Hashin and Monteiro (2002b) to the spring layer model adopted here. In this context the formula of Hashin (2002a) reproduced in Eq. (2.2) may provide K_n and K_t with somewhat different level of accuracy, which may differently affect the bulk than the shear modulus. Another likely reason is the difference between the generalized self-consistent scheme used by Hashin and Monteiro (2002b) to identify the properties of the interphase and the MCM used here to evaluate the effective properties of the composite. Considering those differences it is believed that the results presented in Table 1 constitute an acceptable validation of the EI MCM approach advocated in this work.

4. Conclusions and discussion

The concept of equivalent inhomogeneity formulated differently in the past by several other authors, including recent energy-based approach by these authors (Nazarenko et al. 2015) in the context of Gurtin and Murdoch (1975) interface conditions, has been reformulated herein to include the spring layer model of interphases. This model is particularly suitable for thin compliant interphases where displacement jumps are significant and traction jumps are small. In the presented developments, the spring layer is characterized by a finite thickness. Evaluation of the properties of the equivalent inhomogeneity consisting of an original inhomogeneity and interphase is based on the Lurie solution for spheres.

The equivalent inhomogeneity approach developed here aims at expanding predictive capabilities of the existing methods used to determine effective properties of composites with perfect interfaces (i.e. without interphases), with both deterministic and random microstructures. In particular, it significantly expands the range of applicability of the method of conditional moments (MCM) used herein. MCM is a rigorous statistical homogenization method, but without the idea of equivalent inhomogeneity it has been applicable only to materials with perfect interfaces, just like other statistical methods (cf. Torquato 2002). The combination of equivalent inhomogeneity and MCM accommodates interphases and leads to closed-form expressions for the effective bulk and shear moduli of random composites with spherical particles. Both of those expressions exhibit good engineering accuracy. Thus, this work constitutes a step towards expanding the range of applicability of the methods originally conceived to analyze problems without interphases. In this work that was demonstrated in the context of MCM, but the notion of the equivalent inhomogeneity presented here is just as easily applicable to other methods (e.g., self-consistent schemes, or even numerical methods).

The principal goal in this work was development and validation of the notion of equivalent inhomogeneity for the spring layer model of interphase, that would be capable to provide all material properties of the composite and lead to a closed-form solution with good engineering accuracy. That has been (and could only be) achieved by comparisons with the existing accurate analytic solutions and with the experimental data. To this end only spherical particles are considered, since only such particles have been considered in the existing three-dimensional solutions (existing two-dimensional analyses of circular fibers are analogous to spheres in three dimensions) and in evaluation of the experimental data. For the same reason only three different composite materials were analyzed. One of those materials was the epoxy matrix reinforced by spherical glass particles, analyzed by Hashin (1991) using the self-consistent scheme. The second material (matrix containing rigid particles) was the one investigated by Sangani and Mo (1997), employing a “formally exact” solution of the governing differential equations approach via expansion of the unknown fields in a series of spherical harmonics. The third material consisted of cement paste matrix, spherical sand particles and an interphase; this material was evaluated experimentally and used in inverse analysis to determine the properties of the interphase.

The conclusion one can fairly draw from the numerical evaluations is that, if the normal and tangential spring layer parameters vary proportionally, the results developed in this work are in remarkably good agreement with both those based on the self-consistent method and those based on formally exact solution of the governing differential equations. This fact validates the proposed approach and is particularly noteworthy considering that only the results presented herein are given by closed-form expressions. This is very advantageous in the preliminary stage of analysis (or design) of composite materials and structures, when fast but sufficiently accurate estimates are very helpful in the decision making process.

It can also be observed that, if either normal or tangential component of the spring layer stiffness vanishes, the discrepancy between the estimates presented here and those available in the literature somewhat increases. While it has to be acknowledged that any set of data, including that with vanishing normal or tangential spring layer parameters, can be legitimately used for evaluation of the solution method, one has to keep in mind that not all sets of data are justifiable on the physical ground, and some sets of data (particularly those approaching certain limits) may demand different solution methods than other sets. A more detailed discussion of that is presented at the end of Section 3, which is shortly paraphrased here. If the normal stiffness parameter of the spring layer vanishes, for example, the interaction between the matrix and the inhomogeneity should be modeled as a contact problem, as done by Achenbach and Zhu (1989). None of the results presented in Fig. 5 includes contact conditions, and for that reason their value in analysis of materials whose interphases posses vanishing normal stiffness is not of any practical importance. On the other hand, the assumption that tangential stiffness parameter vanishes everywhere irrespectively of the character of normal interaction is, for solids, in contradiction with the law of friction.

One result obtained here, which is of particular value for the EI MCM approach advocated in this work, is acceptable level of agreement with the experimental results obtained for a cement-based composite material with compliant interphases. Even though for this problem the bulk modulus obtained by the EI MCM method displays somewhat higher discrepancy with the experiment than does the shear modulus, the fact that the data used in this work was obtained using an inverse analysis based on quite different theoretical (and approximate) model makes that level of discrepancy understandable and, thus, acceptable.

Extension of the notion of equivalent inhomogeneity including the spring layer interphase for other particle shapes is conceptually possible. For example, equivalent three-dimensional inhomogeneity for cylindrical or spheroidal particles can be defined energetically, in a manner akin to that adopted by Nazarenko et al. (2015). This will be undertaken in the future and it is likely to further expand the practical utility of the equivalent inhomogeneity approach.

In conclusion, even though additional evaluations and comparisons would be definitely beneficial, the fact that a good engineering-level agreement with the best existing results can be encapsulated in a simple closed-form formula (not requiring hours of powerful computer time, cf. Sangani and Mo (1997)) is reassuring. Those existing results used for validation of the presented approach are numerical or experimental and, to the best knowledge of the authors, the only ones currently available. The comparisons performed to validate the proposed approach indicate that the combination of the equivalent inhomogeneity introduced in this work and of the MCM used to evaluate the effective properties of the investigated composites is capable of providing simple and reliable results. It is believed, however, that the proposed definition of the equivalent inhomogeneity will be equally effective when used jointly with other methods of micromechanics.

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Appendix A. Evaluation of constants A and B in Lurie solution

The constants A and B are computed in terms of β from the following conditions for $r = R$

$$\sigma_{rr}|_{r=R} = K_n \Delta u_r, \quad \sigma_{r\theta}|_{r=R} = K_t \Delta u_\theta. \tag{A.1}$$

Considering Eqs. (2.21) and (2.34a), the first of the above conditions is

$$\begin{aligned} &2\mu_1[18\nu_1 A + B]\left[3\cos^2\theta - 1\right] \\ &-21\lambda_1 A[1-2\nu_1]\left[3\cos^2\theta - 1\right] \\ &= K_n[\beta(R+h) - (6\nu_1 A + B)R]\left[3\cos^2\theta - 1\right]. \end{aligned} \tag{A.2}$$

For the above equation to be satisfied for every θ one must have

$$\begin{aligned} &2\mu_1[18\nu_1 A + B] \ - 21\lambda_1 A[1-2\nu_1] \\ &= K_n[\beta\,[R+h] - [6\nu_1 A + B]R]\,. \end{aligned} \tag{A.3}$$

The second equation is obtained from Eq. $(A.1)_2$ (as well as Eqs. (2.21) and (2.34a)) which yields

$$\begin{aligned} &-2\mu_1[(7+2\nu_1)A + B] \\ &= K_t[-\beta\,[R+h] + [(7-4\nu_1)A + B]\,R]\,. \end{aligned} \tag{A.4}$$

The two equations for A and B are conveniently rewritten in the form

$$\begin{aligned} &[36\mu_1\nu_1 - 21\,[1-2\nu_1]\,\lambda_1 + 6\nu_1 k_n]A + [2\mu_1 + k_n]B \\ &= k_n[1+\delta]\,\beta, \end{aligned} \tag{A.5a}$$

$$\begin{aligned} &[2\mu_1\,[7+2\nu_1] + [7-4\nu_1]\,k_t\,]A + [2\mu_1 + k_t]B \\ &= k_t[1+\delta]\,\beta, \end{aligned} \tag{A.5b}$$

where $\delta = h/R$ (cf. Eq. (2.13)) and

$$k_n = RK_n \text{ and } k_t = RK_t. \tag{A.6}$$

A more compact form of Eqs (A.5a,b) is:

$$\begin{aligned} M_{11}A + M_{12}B &= k_n[1+\delta]\,\beta, \\ M_{21}A + M_{22}B &= k_t[1+\delta]\,\beta, \end{aligned} \tag{A.7}$$

with

$$\begin{aligned} M_{11} &= 36\mu_1\nu_1 - 21\cdot 2\mu_1\nu_1 + 6\nu_1 k_n = 6\nu_1\,[k_n - \mu_1], \\ M_{12} &= 2\mu_1 + k_n, \\ M_{21} &= 2\mu_1\,[7+2\nu_1] + [7-4\nu_1]\,k_t = 7\,[2\mu_1 + k_t] \\ &\quad + 4\nu_1\,[\mu_1 - k_t], \\ M_{22} &= 2\mu_1 + k_t. \end{aligned} \tag{A.8}$$

Denoting

$$\begin{aligned} M &= \det\begin{vmatrix} M_{11} M_{12} \\ M_{21} M_{22} \end{vmatrix} = 6\nu_1\,[k_n - \mu_1]\,[2\mu_1 + k_t] \\ &\quad - [2\mu_1 + k_n]\,[7(2\mu_1 + k_t) + 4\nu_1\,(\mu_1 - k_t)k_n] \\ &= -[4\mu_1{}^2[7+5\nu_1] + 2\mu_1\,[(7-4\nu_1)\,k_n + (7-\nu_1)\,k_t] \\ &\quad + [7-10\nu_1]\,k_n k_t] \end{aligned} \tag{A.9}$$

the solution of Eqs. (A.7) is given by the formulas

$$\begin{cases} A = \dfrac{(1+\delta)\beta}{M}[M_{22}k_n \ - M_{12}\ k_t] \\ B = \dfrac{(1+\delta)\beta}{M}[M_{11}k_t \ - M_{21}\ k_n] \end{cases}. \tag{A.10}$$

In a somewhat expanded form this last result is reproduced in Eq. (2.25).

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