Clausius-Mossotti-type approximation for elastic moduli of a cubic array of spheres

Israel Cohen and David J. Bergman
School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel
(Received 31 July 2002; revised manuscript received 9 February 2003; published 18 July 2003)

The method of elastostatic resonances is applied to the three-dimensional problem of nonoverlapping spherical inclusions arranged in a cubic array in order to calculate the effective elastic moduli. Explicit expressions, exact at least to order p^3 (where p is the volume fraction of the inclusions), are obtained for the bulk modulus and for the two shear moduli. The approximation used, which is the leading order in a systematic perturbation expansion of the appropriate modulus, is related to the Clausius-Mossotti approximation of electrostatics. Comparison with numerical calculations of the moduli and with previous work reveals that this approximation provides accurate results at low volume fractions of the inclusions and is a good estimate to the effective moduli at moderate volume fractions even when the contrast is high. Some of the expressions turn out to be identical to the Hashin-Shtrikman (HS) bounds.

DOI: 10.1103/PhysRevB.68.024104 PACS number(s): 62.20.Dc, 46.25.−y

I. INTRODUCTION

Kantor and Bergman[1] introduced an approach to the problem of calculating the effective elastic stiffness tensor $C^{(e)}$ of two-component composite materials with a specified microstructure. This approach, which was based on a calculation of elastostatic resonances of the system, was applied to composites in the form of regular two-dimensional (2D) arrays of circular-cylindrical inclusions of an isotropic elastic material with elastic stiffness tensor $C^{(1)}$ embedded in an isotropic host with elastic stiffness tensor $C^{(2)}$. For the cases of hexagonal and square arrays, they were able to obtain an explicit expansion of the 2D effective moduli $C^{(e)}$ in powers of the volume fraction p of the $C^{(1)}$ component up to a rather high order (e.g., the 2D bulk modulus κ_e was evaluated up to and including $O(p^{11})$ terms for an hexagonal array). For 3D microstructures, this approach was applied to a periodic array of differently oriented circular-cylindrical inclusions with cubic symmetry[2] in order to get expressions for the effective moduli which are exact at least up to terms of the order of p^2.

In this paper, we apply the same approach to a 3D model of cubic arrays of spherical inclusions of isotropic material $C^{(1)}(\kappa_1,\mu_1)$ (component 1) embedded in an isotropic host $C^{(2)}(\kappa_2,\mu_2)$ (component 2) (κ is the bulk modulus, μ is the shear modulus). This simple model is already a very difficult problem to solve, mainly because the eigenstates (i.e., elastostatic resonances) of an isolated spherical inclusion are not known. In fact, we have computed only a few of them—the dipole eigenstates. This turns out to be a useful exercise because these states are often responsible for the dominant part of the interaction between distortions of different inclusions, in analogy with electrostatic problems.

The results obtained for the macroscopic elastic moduli are in the form of simple algebraic expressions, similar both in form and in spirit to the Clausius-Mossotti (CM) expression for macroscopic dielectric constants of such composites. The expression for the macroscopic bulk modulus κ_e coincides with one of the Hashin-Shtrikman (HS) bounds,[3] which has often been used as an approximation to the exact value and is known to provide a good approximation for a wide range of relevant parameters. In contrast, the expressions for the two macroscopic shear moduli were not known previously.

To determine exactly the effective elastic stiffness tensor of 3D anisotropic composite materials is a very difficult task because of the difficulties and complications involved in solving the appropriate elastostatic equations in 3D systems. Most of the published numerical work deals with 2D problems because of these difficulties and complications, and also because of the fact that the practical problem of materials that are reinforced with parallel fibers is actually a 2D problem. The spatial cubic symmetry simplifies matters and enables one to solve numerically for the effective moduli essentially exactly in some special cases. Such is the work of Nunan and Keller,[4] who investigated the extreme contrast microstructure of a periodic cubic array of rigid spheres and who formulated an accurate numerical procedure to compute the elastic moduli for this case. Another numerical procedure to compute the effective elastic moduli is due to Nemat-Nasser *et al.*,[5,6] who investigated the opposite extreme contrast microstructure of a periodic cubic array of spherical voids. An important recent advance in determining the effective stiffness tensor of composite materials was made by Torquato,[7,8] who developed new perturbation expansions for the effective stiffness tensor which are absolutely convergent. These expansions involve n-point correlation functions which characterize the microstructure. Third-order explicit expressions were derived for the effective moduli of isotropic dispersions and for the bulk modulus of a composite with a cubically symmetric microstructure.[8] We have compared our results with all of this previous work and also with numerical calculations that we have performed. The simple algebraic expressions we have derived seem to provide accurate results for the effective moduli at low volume fractions of the inclusions even when the contrast is infinite; they continue to provide a good estimate at moderate volume fractions, and, as expected, they fail to do so when the volume fraction is high, especially when the volume fraction approaches its close-packing value (when the spheres begin to touch each other).

II. SUMMARY OF THE UNDERLYING THEORY

The approach (we will use a simplified form of the general theory, described in detail in Refs. 9 and 10) begins by introducing a somewhat generalized form of the original problem and replacing the $C^{(1)}$ material by a different material $C'^{(1)}(s)$, where

$$C'^{(1)}(s)=C^{(2)}-\frac{1}{s}\delta C=\frac{1}{s}C^{(1)}+\frac{s-1}{s}C^{(2)} \tag{2.1}$$

and $\delta C\equiv C^{(2)}-C^{(1)}$. This replacement also makes $C^{(e)}$ a function of the parameter s. When s lies in certain ranges the tensor $C'^{(1)}(s)$ becomes *unphysical*, i.e., it ceases to be positive definite. The original problem is retrieved by setting $s=1$.

The position-dependent local elastic tensor $C(\mathbf{r})$ of the system can now be written in the form

$$C(\mathbf{r})=C^{(2)}-\frac{1}{s}\theta_1(\mathbf{r})\delta C, \tag{2.2}$$

where $\theta_1(\mathbf{r})$ is the characteristic or indicator function:

$$\theta_1(\mathbf{r})=\begin{cases}1, & \mathbf{r} \text{ inside component } 1\\ 0 & \text{otherwise.}\end{cases} \tag{2.3}$$

The strain tensor $\varepsilon(\mathbf{r})$ in such a composite material, the boundaries of which undergo the displacement $u_i=\varepsilon_{ij}^{(0)}x_j$, is the solution of the operator equation

$$\varepsilon=\varepsilon^{(0)}+\frac{1}{s}\hat{\Gamma}\varepsilon, \tag{2.4}$$

where $\varepsilon_{ij}^{(0)}$ is any constant symmetric tensor and $\hat{\Gamma}$ is a linear integral operator defined in Eqs. (A5)–(A7) of Appendix A. In Refs. 1,9 and 10, it was shown that any effective elastic stiffness coefficient of a composite material which satisfies Eq. (2.2) can be written as a sum of simple poles in the following form:

$$\varepsilon^{(0)}C^{(2)}\varepsilon^{(0)}-\varepsilon^{(0)}C^{(e)}\varepsilon^{(0)}=\sum_n\frac{F_n}{s-s_n}, \tag{2.5}$$

where summation over tensorial indices is implied on the left-hand side, and the poles s_n and weights F_n are all real. Each pole is obtained as an eigenvalue of the operator $\hat{\Gamma}$: $\hat{\Gamma}|\varepsilon^{(n)}\rangle=s_n|\varepsilon^{(n)}\rangle$, where the eigenstate $|\varepsilon^{(n)}\rangle$ represents an elastostatic resonance of the sample, i.e., a state where the sample is internally deformed and strained even though the boundaries are undeformed. Obviously, such resonances can occur only at unphysical values of $C'^{(1)}(s)$.

A scalar product of two second-rank symmetric tensor fields ε,σ is defined by (* denotes complex conjugation)

$$\langle\varepsilon|\sigma\rangle\equiv\int dV\theta_1(\mathbf{r})\varepsilon_{ij}^{*}(\mathbf{r})\sigma_{ij}(\mathbf{r}). \tag{2.6}$$

Under this definition $\hat{\Gamma}$ is not Hermitian, and thus has different right and left eigenstates. However, if $|\varepsilon^{(n)}\rangle$ is a right eigenstate with eigenvalue s_n, then $\langle\varepsilon^{(n)}\delta C|\equiv\langle\tilde{\varepsilon}^{(n)}|$ is a left eigenstate with eigenvalue s_n^{*}, and they form a complete biorthogonal set (see, e.g., Ref. 11) inside phase 1,

$$\langle\tilde{\varepsilon}^{(n)}|\varepsilon^{(m)}\rangle=0 \quad \text{for} \quad n\neq m. \tag{2.7}$$

The weights F_n of Eq. (2.5) are given by

$$F_n=\frac{1}{V}|\langle\tilde{\varepsilon}^{(0)}|\varepsilon^{(n)}\rangle|^2/\langle\tilde{\varepsilon}^{(n)}|\varepsilon^{(n)}\rangle, \tag{2.8}$$

where V is the total volume occupied by the composite material. We define the "norm" of a state $\|\varepsilon\|$ to be

$$\|\varepsilon\|\equiv\langle\tilde{\varepsilon}|\varepsilon\rangle^{1/2}. \tag{2.9}$$

The question of whether the eigenstates are all normalizable, i.e., whether

$$\|\varepsilon^{(n)}\|^2=\int dV\theta_1(\mathbf{r})\varepsilon^{*}\delta C(\mathbf{r})\varepsilon(\mathbf{r})\neq 0 \quad \text{for all } n \tag{2.10}$$

is a nontrivial one because the fourth-rank tensor $\delta C\equiv C^{(2)}-C^{(1)}$ is not necessarily positive or negative definite. However, it was shown in Refs. 1 and 10 that all eigenstates with nonzero eigenvalues $s_n\neq 0$ are normalizable and that their eigenstates are real. Nevertheless, note that $\|\varepsilon^{(n)}\|^2$ can be either positive or negative, therefore $\|\varepsilon^{(n)}\|$ can be either real or imaginary.

To solve the problem of many inclusions inside a host material, one uses an approach similar to the tight-binding method from solid-state crystal electronics in order to expand the eigenstates (resonances) $|\varepsilon^{(n)}\rangle$ of the many inclusions problem in terms of the complete (biorthogonal) set of eigenstates $|\varepsilon^{(a\alpha)}\rangle$ of the individual inclusions, denoted by index a,

$$\theta_1|\varepsilon^{(n)}\rangle=\sum_{a\alpha}\frac{B_{a\alpha}^{(n)}\theta_a|\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(a\alpha)}\|}, \tag{2.11}$$

where $\theta_a(\mathbf{r})$ equals 1 only inside inclusion a and vanishes elsewhere. The detailed form of an eigenstate $|\varepsilon^{(a\alpha)}\rangle$ of the isolated inclusion a, as well as its eigenvalue $s_{a\alpha}$, depends on the shape of the inclusion, but not on its total size. In the same manner as for $|\varepsilon^{(n)}\rangle$, we can express the operator $\hat{\Gamma}$ of the many inclusions problem as a sum of the individual grain operators, $\hat{\Gamma}=\Sigma_a\hat{\Gamma}\theta_a\equiv\Sigma_a\hat{\Gamma}_a$, where $\hat{\Gamma}_a|\varepsilon^{(a\alpha)}\rangle=s_{a\alpha}|\varepsilon^{(a\alpha)}\rangle$. We then use these two expansions in the eigenvalue equation for $|\varepsilon^{(n)}\rangle$: $\hat{\Gamma}|\varepsilon^{(n)}\rangle=s_n|\varepsilon^{(n)}\rangle$, and after multiplying both sides of this equation with the state $\langle\tilde{\varepsilon}^{(b\beta)}|\theta_b$, we arrive at a matrix eigenvalue problem for the expansion coefficients $B_{a\alpha}^{(n)}$,

$$(s_n-s_\beta)\|\varepsilon^{(b\beta)}\|B_{b\beta}^{(n)}=\sum_{\substack{a\alpha\\ a\neq b}}\frac{\langle\tilde{\varepsilon}^{(b\beta)}\theta_b|\hat{\Gamma}|\theta_a\varepsilon^{(a\alpha)}\rangle B_{a\alpha}^{(n)}}{\|\varepsilon^{(a\alpha)}\|}. \tag{2.12}$$

This equation is general and holds for any composite material with nonoverlapping inclusions.

A great simplification occurs if the inclusions are identical and form a periodic array in space. In that case, Bloch's theorem immediately specifies the dependence of the eigenvectors on the inclusion index a,

$$B_{a\alpha}^{(n)}=\frac{1}{\sqrt{N}}B_{\alpha}^{(n)}e^{i\mathbf{k}\cdot\mathbf{r}_a}, \tag{2.13}$$

where N is the number of unit cells in the periodic sample and $\mathbf{r}_a$ represents the location of the inclusion a. By using Eqs. (2.11) and (2.13) in Eq. (2.8), together with the fact that $\langle\tilde{\varepsilon}^0|\theta_a\varepsilon^{(a\alpha)}\rangle$ and $\langle\tilde{\varepsilon}^{(a\alpha)}|\theta_a\varepsilon^{(a\alpha)}\rangle$ are independent of a, we obtain for the weights

$$F_n=\frac{N}{V}\delta_{\mathbf{k},\mathbf{0}}\left|\sum_{\alpha}\ \frac{B_{\alpha}^{(n)}(\mathbf{k})\langle\tilde{\varepsilon}^{(0)}|\theta_a\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(a\alpha)}\|}\right|^2\Bigg/\sum_{\alpha}\ |B_{\alpha}^{(n)}(\mathbf{k})|^2, \tag{2.14}$$

where there is no longer any summation over inclusion indices. Since only the $\mathbf{k}=\mathbf{0}$ Bloch states can have nonzero weights, only those states need to be considered. The eigenvalue problem (2.12) for the $\mathbf{k}=\mathbf{0}$ eigenstates becomes

$$(s_n-s_\beta)B_{\beta}^{(n)}=\sum_{\alpha}\ Q_{\beta\alpha}B_{\alpha}^{(n)}, \tag{2.15}$$

where the elements $Q_{\beta\alpha}$ can be expressed as sums of overlap integrals between the isolated inclusion eigenstates *of pairs of different inclusions*,

$$\begin{aligned}Q_{\beta\alpha}&=\sum_{\substack{a\\a\neq b}}\ \frac{\langle\tilde{\varepsilon}^{(b\beta)}\theta_b|\hat{\Gamma}|\theta_a\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(b\beta)}\|\ \|\varepsilon^{(a\alpha)}\|}\\&=\sum_{\substack{a\\a\neq b}}\ \frac{s_\alpha\int\theta_b\varepsilon^{(b\beta)*}\delta C\varepsilon^{(a\alpha)}dV}{\|\varepsilon^{(b\beta)}\|\ \|\varepsilon^{(a\alpha)}\|},\quad Q_{\beta\alpha}=Q_{\alpha\beta}^{*}.\end{aligned} \tag{2.16}$$

Note that the elements $Q_{\beta\alpha}$, as well as the elements $\langle\tilde{\varepsilon}^{(b\beta)}\theta_b|\hat{\Gamma}|\theta_a\varepsilon^{(a\alpha)}\rangle$, constitute a Hermitian matrix, even though the integral operator $\hat{\Gamma}$ is non-Hermitian. The appearance of the characteristic function $\theta_b(\mathbf{r})$ restricts the integration volume to the inside of inclusion b. Having found the eigenvectors $B_{\beta}^{(n)}$, the weights are given by the expression

$$F_n=\frac{1}{V_a}\left|\sum_{\alpha}\ \frac{B_{\alpha}^{(n)}\langle\tilde{\varepsilon}^{(0)}|\theta_a\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(a\alpha)}\|}\right|^2\Bigg/\sum_{\alpha}\ |B_{\alpha}^{(n)}|^2, \tag{2.17}$$

where V_a is the volume of a unit cell in the periodic composite structure.

III. CUBIC ARRAY OF SPHERES

Our system consists of a 3D cubic array of spherical inclusions of an isotropic material $C^{(1)}(\kappa_1,\mu_1)$ embedded in an isotropic host $C^{(2)}(\kappa_2,\mu_2)$. The two-body interaction between different spherical inclusions, which appears in the form of overlap integrals in Eq. (2.16), decreases with increasing distance between inclusions. In connection with this, the elements $Q_{\beta\alpha}$ always behave like some positive power of the volume fraction of the inclusions $p\equiv V_R/V_a$ when p is small. From the 2D elastostatic problem[1] of circular-cylindrical inclusions, and from the 3D electrostatic problem[10,12] of spherical inclusions, we expect that, in order to include the effects of these interactions to leading order in p, we only need to consider the dipole resonances. These states correspond to a strain field ε which is uniform inside the inclusion (a sphere) and decreases slowest with distance outside the inclusion. We *chose* these dipole resonances to be certain uniform strains inside the sphere and then followed the procedure described in Appendix B in order to find their shape outside the sphere and also the eigenvalues.

In order to form a complete biorthogonal set, there are six dipole (strain) eigenstates, as this is the number of constant linearly independent second-rank symmetric tensors in three dimensions. The first resonance corresponds to a pure, uniform compression inside the sphere of radius R—we call this state a ''compression dipole.'' Its right eigenstate is

$$\varepsilon_{ij}^{(1)}=\begin{cases}\dfrac{1}{3\sqrt{V_R}}\delta_{ij}, & r<R\\[2ex]\dfrac{1}{4\pi}\sqrt{V_R}\left(\delta_{ij}-\dfrac{3x_ix_j}{r^2}\right)\dfrac{1}{r^3}, & r>R,\end{cases} \tag{3.1}$$

where $V_R=4\pi R^3/3$ is the volume of the sphere, and its eigenvalue is

$$s_1=\frac{\delta\kappa}{\lambda+2\mu}\quad(\lambda=\kappa-\tfrac{2}{3}\mu). \tag{3.2}$$

Here and subsequently, we omit the subscript 2 from the elastic moduli of component 2. The other five resonances correspond to a pure shear strain inside the sphere and are all degenerate, with the same eigenvalue s_2. The forms of these strain states inside the sphere were chosen to be

$$\varepsilon_{ij}^{(2)}=I_{ij12},\quad \varepsilon_{ij}^{(3)}=I_{ij13},\quad \varepsilon_{ij}^{(4)}=I_{ij23}, \tag{3.3a}$$

$$\varepsilon_{ij}^{(5)}=\tfrac{1}{2}(I_{ij11}-I_{ij22}), \tag{3.3b}$$

$$\varepsilon_{ij}^{(6)}=(I_{ij11}+I_{ij22}-2I_{ij33}), \tag{3.3c}$$

where $I_{ijkl}=\frac{1}{2}(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})$. The common shear eigenvalue is

$$s_2=\frac{2(\kappa+2\mu)\delta\mu}{5(\lambda+2\mu)\mu}. \tag{3.4}$$

The forms of the shear resonances outside the sphere are hard to find but, having found one of them, we can obtain the others by appropriate rotations of the coordinate axes (see Appendix B). For example, $\varepsilon_{ij}^{(6)}$ corresponds to a displace-

ment field outside the sphere ($r>R$) of the form [$\mathbf{e}_r$, $\mathbf{e}_\theta$, $\mathbf{e}_\phi$ are unit vectors in the spherical coordinate system (r,θ,ϕ)]

$$\mathbf{u}^{(6)}=\left(\frac{3\lambda+5\mu}{\mu}P_2^{(0)}\frac{B}{r^2}+3P_2^{(0)}\frac{C}{r^4}\right)\mathbf{e}_r + \left(P_2^{(1)}\frac{C}{r^4}-P_2^{(1)}\frac{B}{r^2}\right)\mathbf{e}_\theta, \tag{3.5}$$

where $B=-5\mu R^3/(3\lambda+8\mu)$, $C=3(\lambda+\mu)R^5/(3\lambda+8\mu)$, and $P_l^{(m)}(\theta)$ is the associated Legendre function. The form of $\varepsilon_{ij}^{(6)}$ for $r>R$ is

$$\varepsilon_{11}=\left(\frac{3(x^2+z^2)}{r^2}-\frac{15x^2z^2}{r^4}-1\right)\frac{U(r)}{2r^3}+\left(\frac{x^2}{r^2}-\frac{5x^2z^2}{r^4}\right)\frac{3C}{r^5} + \left(\frac{z^2}{r^2}-\frac{5x^2z^2}{r^4}\right)\frac{3D(r)}{r^3}, \tag{3.6a}$$

$$\varepsilon_{22}=\text{same as } \varepsilon_{11}\text{with } x \text{ replaced by } y, \tag{3.6b}$$

$$\varepsilon_{33}=\left(\frac{12z^2}{r^2}-\frac{15z^4}{r^4}-1\right)\frac{U(r)}{2r^3}+\left(\frac{3z^2}{r^2}-\frac{5z^4}{r^4}\right)\frac{3C}{r^5} + \left(\frac{6z^2}{r^2}-\frac{5z^4}{r^4}-1\right)\frac{3D(r)}{r^3}, \tag{3.6c}$$

$$\varepsilon_{23}=\left(4-\frac{10z^2}{r^2}\right)\frac{3yz}{4r^5}U(r)+\left(2-\frac{5z^2}{r^2}\right)\frac{6yz}{r^7}C + \left(5-\frac{10z^2}{r^2}\right)\frac{3yz}{r^5}D(r), \tag{3.6d}$$

$$\varepsilon_{13}=\text{same as } \varepsilon_{23} \text{ with } y \text{ replaced by } x, \tag{3.6e}$$

$$\varepsilon_{12}=\left[\left(1-\frac{3z^2}{r^2}\right)\frac{3xy}{2r^2}-\frac{3xyz^2}{r^4}\right]\frac{1}{r^3}U(r) + \left(1-\frac{5z^2}{r^2}\right)\frac{3xy}{r^7}C-\frac{15xyz^2}{r^7}D(r), \tag{3.6f}$$

where $U(r)=[(3\lambda+5\mu)/\mu]B+(3/r^2)C$ and $D(r)=(C/r^2)-B$.

The effective elastic tensor of a composite material with cubic symmetry has the form

$$C_{ijkl}^{(e)}=\kappa_e\delta_{ij}\delta_{kl}+2\mu_e(I_{ijkl}-\delta_{ijkl})+2M_e\left(\delta_{ijkl}-\frac{1}{3}\delta_{ij}\delta_{kl}\right), \tag{3.7}$$

where κ is the bulk modulus and μ,M are two shear moduli which coincide in the isotropic case, and

$$\delta_{ijkl}\equiv\begin{cases}1, & i=j=k=l\\ 0 & \text{otherwise.}\end{cases} \tag{3.8}$$

In order to isolate only one of the above three effective elastic moduli κ_e,μ_e, or M_e in Eq. (2.5) we choose $\varepsilon^{(0)}$ to be one of the following:

$$\varepsilon_{ij}^{(0\kappa)}=\frac{1}{3}\delta_{ij}, \quad \varepsilon_{ij}^{(0\mu)}=I_{ij12}, \tag{3.9a}$$

$$\varepsilon_{ij}^{(0M)}=\frac{1}{\sqrt{12}}(I_{ij11}+I_{ij22}-2I_{ij33}). \tag{3.9b}$$

The fact that choices (3.9) are proportional to $\varepsilon^{(1)},\varepsilon^{(2)},\varepsilon^{(6)}$ respectively, together with the biorthogonality relation (2.7), implies that by choosing, e.g., $\varepsilon^{(0)}=\varepsilon^{(0\kappa)}$, the only contribution to the numerator in Eq. (2.17) comes from the $\varepsilon^{(1)}$ state.

In the dilute suspension limit, when $p\ll 1$, we can neglect the right-hand side of Eq. (2.15), so that the eigenstates are approximately equal to the isolated inclusion eigenstates. Thus we obtain from Eq. (2.5),

$$\kappa_2-\kappa_e\cong\frac{1}{V_a}\frac{|\langle\tilde{\varepsilon}^{(0\kappa)}|\varepsilon^{(1)}\rangle|^2/\langle\tilde{\varepsilon}^{(1)}|\varepsilon^{(1)}\rangle}{1-s_1}=\frac{1}{V_a}\frac{V_R\delta\kappa}{1-\dfrac{\delta\kappa}{\lambda+2\mu}} =\frac{p\,\delta\kappa}{1-\dfrac{\delta\kappa}{\lambda+2\mu}}. \tag{3.10}$$

In the same manner, by choosing $\varepsilon^{(0)}=\varepsilon^{(0\mu)}$ we obtain for the shear moduli

$$\mu_2-\mu_e\cong\frac{1}{V_a}\frac{|\langle\tilde{\varepsilon}^{(0\mu)}|\varepsilon^{(2)}\rangle|^2/\langle\tilde{\varepsilon}^{(2)}|\varepsilon^{(2)}\rangle}{1-s_2} =\frac{p\,\delta\mu}{1-\dfrac{2(\kappa+2\mu)\delta\mu}{5(\lambda+2\mu)\mu}}. \tag{3.11}$$

In this dilute limit, we get $M_e=\mu_e$. Expressions (3.10) and (3.11) can also be derived by considering the problem of a single spherical inclusion embedded in an infinite homogeneous isotropic elastic medium under the application of a uniform external strain field.[13]

In order to take the dipole-dipole interactions into account, we must calculate some elements of the matrix $Q_{\alpha\beta}$, namely, $\alpha,\beta=1,\ldots,6$. The overlap integrals of Eq. (2.16) which involve expressions such as (3.1) and (3.6) are difficult to calculate. But since the calculation of $Q_{\beta\alpha}$ involves summation of these integrals over all lattice sites, we can exploit the cubic symmetry to make replacements such as $x^2\to y^2$ (we choose the Cartesian axes to be the cubic symmetry axes of the lattice) in the integrands of Eq. (2.16) (see Appendix C). In this way, we find that $Q_{\beta\alpha}$ is a *diagonal* matrix. This is clearly a result of the cubic lattice symmetry.

A special problem arises in the summation of overlap integrals between two dipole states: there appear lattice sums of the terms of the form $P_l^{(0)}(\theta)/r^3$ [see Eq. (D12) of Appendix D], which are only semiconvergent. In these sums,

the distant contributions are as important as the nearby ones, and the series converges only thanks to the alternating signs of the angular functions. However, the isolated inclusion states, such as Eqs. (3.1) and (3.6), were calculated assuming that the isolated sphere was far away from the sample boundaries. Therefore, we can use the overlap integrals of these states only for the nearby lattice sites. The problem is solved by means of the concept of the local Lorentz field of the analogous problem in electrostatics (in fact, the components of $\varepsilon^{(1)}$ are proportional to the electric field created by a dipole at a distance r).

The treatment of dipole-dipole interactions in the ''language'' of resonances was originally introduced by Bergman[10,12] for the analogous problem of electrostatics, in order to obtain the CM approximation. It should be noted that (1) this is not the standard approach to obtain the CM approximation (for the standard approach, see, e.g., Ref. 14); (2) since the CM approximation involves summation of dipole interactions, it depends on the symmetry of the medium, i.e., whether the medium is isotropic, or has cubic or other kind of symmetry.

The CM-type approximation is obtained by breaking up the sum into near terms (evaluated exactly for a cubic array of spheres that are far away from the boundaries) and far terms (evaluated by replacing the discrete dipole array by a uniform continuous strain polarization, and taking into account the correct boundary conditions).[1] We start by writing the elastostatic equilibrium equation $\partial_j C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})=0$ for the dipole eigenstate $\varepsilon^{(a\alpha)}$ in terms of the local elastic tensor of Eq. (2.2). Doing so, we can find $\varepsilon^{(a\alpha)}(\mathbf{r})$ everywhere by solving the equation

$$\partial_j C^{(2)}_{ijkl}(\mathbf{r})\varepsilon^{(a\alpha)}_{kl}(\mathbf{r})=-\partial_j P_{ij}(\mathbf{r}), \tag{3.12}$$

where

$$P_{ij}(\mathbf{r})=\frac{1}{s_{a\alpha}}\theta_a(\mathbf{r})\,\delta C_{ijkl}\varepsilon^{(a\alpha)}_{kl}(\mathbf{r}) \tag{3.13}$$

is the elastic polarization density of the sphere, in analogy with the electrostatic case. The important properties of the dipole eigenstates $\varepsilon^{(a\alpha)}$ of an isolated inclusion are that they are constant inside the sphere, and that their eigenvalues are independent of the size of the sphere.

The sum $Q_{\beta\alpha}$ of dipole-dipole interactions between all the other spheres and the one at the origin can be written as an overlap integral over the volume of that sphere, in which the integrand is the product of the strain field $\varepsilon^{(b\alpha)}$ of the central sphere, δC, and ε which is the strain field due to all other sphere dipoles, denoted by $\varepsilon^{\mathrm{loc}}$. This strain field, created at the central sphere by all the other spheres, is calculated by considering separately the contribution of the nearby spheres, i.e., those that lie within a sphere of radius L around the origin (this radius should be much larger than the sphere radii and the intergrain separations), and that of the distant spheres,

$$\varepsilon^{\mathrm{loc}}=\varepsilon^{\mathrm{loc}}_{\mathrm{near}}+\varepsilon^{\mathrm{loc}}_{\mathrm{far}}. \tag{3.14}$$

In order to calculate the contribution of the distant spheres (each one of them is uniformly polarized), we smear their polarization over space for $r>L$, replacing the actual inhomogeneous polarization density $P_{ij}(\mathbf{r})$ for $r>L$ by its (homogeneous) volume average. We denote the contribution of this macroscopic polarization by $\varepsilon^{\mathrm{macro}}_{\mathrm{far}}$. This field can be calculated by noticing that if we smear the polarization of all spheres (including the nearby ones), thereby creating a homogeneous polarization over all space, then the solution of Eq. (3.12) would simply be $\varepsilon=0$ (due to zero boundary conditions that an eigenstate must fulfill) and that this zero field is the result of two contributions: one due to the distant macroscopic polarization $\varepsilon^{\mathrm{macro}}_{\mathrm{far}}$ and the other due to the smeared nearby (homogeneous) polarization $\varepsilon^{\mathrm{macro}}_{\mathrm{near}}$. The vanishing of the overall strain means that $\varepsilon^{\mathrm{macro}}_{\mathrm{far}}=-\varepsilon^{\mathrm{macro}}_{\mathrm{near}}$. The near contribution $\varepsilon^{\mathrm{macro}}_{\mathrm{near}}$ is easy to calculate, since it is actually the strain field created by a homogeneously polarized sphere (a very large one—its radius is L) with polarization density $(1/s_{a\alpha})p\,\delta C\varepsilon^{(a\alpha)}$. This field is similar to the dipole eigenstate $\varepsilon^{(a\alpha)}$, which could also be viewed as resulting from a uniform polarization density $(1/s_{a\alpha})\,\delta C\varepsilon^{(a\alpha)}$ inside an isolated sphere. Consequently, we conclude that $\varepsilon^{\mathrm{macro}}_{\mathrm{near}}$ is the same as $\varepsilon^{(a\alpha)}$, except for the factor p (p is the volume fraction of the spheres),

$$\varepsilon^{\mathrm{macro}}_{\mathrm{far}}=-\varepsilon^{\mathrm{macro}}_{\mathrm{near}}=-p\varepsilon^{(a\alpha)}. \tag{3.15}$$

When this result is substituted for the sum of contributions to ε from all the dipoles whose distance from the origin is greater than L, we obtain

$$\begin{aligned}
Q_{\beta\alpha}&=\sum_{\substack{a\\ a\neq b}}\frac{\langle\tilde{\varepsilon}^{(b\beta)}\theta_b|\hat{\Gamma}|\theta_a\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(b\beta)}\|\ \|\varepsilon^{(a\alpha)}\|}\\
&=\sum_{\substack{a\\ 0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{\langle\tilde{\varepsilon}^{(b\beta)}\theta_b|\hat{\Gamma}|\theta_a\varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(b\beta)}\|\ \|\varepsilon^{(a\alpha)}\|}-ps_\alpha\delta_{\alpha\beta}\\
&=\sum_{\substack{a\\ 0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{s_\alpha\int\theta_b\varepsilon^{(b\beta)*}\delta C\varepsilon^{(a\alpha)}dV}{\|\varepsilon^{(b\beta)}\|\ \|\varepsilon^{(a\alpha)}\|}-ps_\alpha\delta_{\alpha\beta}.
\end{aligned} \tag{3.16}$$

In this equation, L must be large enough so that the use of average polarization density for $r>L$ is a good approximation. In practice, one sums the series over a set of spheres with larger and larger radius L until convergence is achieved.

Since $Q_{\beta\alpha}$ ($\alpha,\beta=1,\ldots,6$) is *diagonal* (see Appendix C), the effective moduli will have a similar form to those of a dilute system [see Eqs. (3.10) and (3.11)], but with shifted poles. If both states are compressional dipoles $\varepsilon^{(1)}$, then all the overlap integrals in Eq. (3.16) vanish, and the shifted pole of the whole system becomes

$$s^{(\kappa)}=s_1+Q_{11}=(1-p)s_1. \tag{3.17}$$

Thus, taking into account only the strongest (dipole-dipole) interactions, we obtain a CM-type approximation for κ_e:

$$\kappa_e \cong \kappa_2 - \frac{p\,\delta\kappa}{1-\dfrac{\delta\kappa(1-p)}{\lambda+2\mu}}$$

$$= \kappa_2 - \frac{p}{1/(\kappa_2-\kappa_1)-3(1-p)/(3\kappa_2+4\mu_2)}. \tag{3.18}$$

This expression for κ_e is identical to one of the HS bounds.[3]

Unlike the situation for κ_e, the overlap integrals between two $\varepsilon^{(2)}$ states (for μ_e) or between two $\varepsilon^{(6)}$ states (for M_e) do not vanish and must be summed numerically in Eq. (3.16). If we denote these sums by $s_2 G_\mu, s_2 G_M$ respectively, then

$$Q_{22} = s_2(G_\mu - p), \quad Q_{66} = s_2(G_M - p), \tag{3.19}$$

therefore, the shifted poles of the whole system are

$$s^{(\mu)} = s_2 + Q_{22} = (1-p+G_\mu)s_2, \tag{3.20}$$

$$s^{(M)} = s_2 + Q_{66} = (1-p+G_M)s_2. \tag{3.21}$$

The CM-type approximations for the two effective shear moduli are

$$\mu_e \cong \mu_2 - \frac{p\,\delta\mu}{1-(1-p+G_\mu)s_2}, \tag{3.22}$$

$$M_e \cong \mu_2 - \frac{p\,\delta\mu}{1-(1-p+G_M)s_2}. \tag{3.23}$$

The overlap integrals of Q_{22}, Q_{66}, which involve integrands of the form of Eqs. (3.6), are difficult to calculate. But since the calculation of $Q_{\beta\alpha}$ involves summation of these integrals over all lattice sites, we can exploit the cubic symmetry of the composite in order to make replacements such as $x^2 \to y^2$ in the integrands (see, e.g., Appendix C), and in this way notice that

$$3G_\mu = -2G_M. \tag{3.24}$$

The exact expressions for G_μ or G_M involve lattice sums which must be calculated numerically. The final result is

$$G_M = \frac{3\kappa+\mu}{\kappa+2\mu}(-0.929p + 1.142p^{5/3}). \tag{3.25}$$

A detailed calculation of G_M and an explanation of the numerical factors can be found in Appendix D.

In order to find higher-order corrections to these Clausius-Mossotti-type approximations, Eqs. (3.18), (3.22), and (3.23), one has to continue the perturbative treatment and calculate matrix elements $Q_{\beta\alpha}$ between the corresponding dipole eigenstates and nondipole eigenstates. Since nondipole eigenstates decrease with distance faster than $1/r^3$, these corrections will be of higher order in p. As was already stated, the fact that choices (3.9) of $\varepsilon^{(0)}$ are proportional to $\varepsilon^{(1)}, \varepsilon^{(2)}, \varepsilon^{(6)}$ respectively, together with the biorthogonality relation (2.7), implies that by choosing, e.g., $\varepsilon^{(0)} = \varepsilon^{(0\kappa)}$, the only contribution to the numerator in Eq. (2.17) comes from the $|\varepsilon^{(1)}\rangle$ state. Therefore, in order to calculate κ_e we need only consider those states which have nonvanishing matrix elements with the unperturbed compression dipole state $|\varepsilon^{(1)}\rangle$. Let $|\varepsilon^{(\eta)}\rangle$ denote a nondipole eigenstate (of the single inclusion) with eigenvalue s_η that has a nonvanishing matrix element with the compression dipole state $|\varepsilon^{(1)}\rangle$ (we will assume $s_1 \neq s_\eta$). We shall denote by a_0 the unit length of the simple cubic lattice, i.e., this is the shortest distance between inclusion centers. Since $|\varepsilon^{(\eta)}\rangle$ decreases with distance at least as $1/r^4$ outside the inclusion, then the overlap integrals between such a state $|\varepsilon^{(a\eta)}\rangle$ located at a distance $a \geqslant a_0$ from the origin and a compression state located at the origin will decrease at least as $1/a^4$, therefore $Q_{1\eta}$, which is the sum over a of all such overlap integrals and is dimensionless, will be at least of the order of $O[(R/a_0)^4] = O(p^{4/3})$. In order to find the new eigenstates of Eq. (2.15), we have to diagonalize the matrix subspace $Q_{\alpha\beta}$ with $\alpha,\beta = 1,\eta$; this way we obtain two new eigenstates with new eigenvalues. One of these states will be approximately $|\varepsilon^{(1)}\rangle$ with shifted eigenvalue. Using standard second-order perturbation theory, the correction to its eigenvalue $s^{(\kappa)}$ will be $|Q_{1\eta}|^2/(s_1 - s_\eta)$, therefore, the correction to κ_e will be of the order of $p|Q_{1\eta}|^2 = p^{11/3}$ [the additional factor p comes from the expression for the weight $F_n \propto (1/V_a)|\langle\tilde{\varepsilon}^{(0\kappa)}|\varepsilon^{(1)}\rangle|^2/\langle\tilde{\varepsilon}^{(1)}|\varepsilon^{(1)}\rangle$. The other new eigenstate will be approximately the unperturbed state $|\varepsilon^{(\eta)}\rangle$ but with an important correction to it $[Q_{1\eta}/(\eta - s_1)]|\varepsilon^{(1)}\rangle$: Only this correction has a nonvanishing scalar product with $\langle\varepsilon^{(0\kappa)}|$, therefore only this part will contribute to the weight F_n of $F(s)$. This contribution will be proportional to $p|Q_{1\eta}/(s_\eta - s_1)|^2$ [see Eq. (2.17)], i.e., also of the order of $p^{11/3}$. Similar considerations apply when we calculate the higher-order corrections to μ_e and M_e. Our general conclusion is that the next correction to the CM-type approximations derived here will begin at least at order $p^{11/3}$.

We can find a combination of the two shear moduli, which is independent of the numerical factors G_μ, G_M. Using relation (3.24), along with the fact that G_μ, G_M are at least of the order of $O(p)$, we find [by Taylor expansion of Eqs. (3.22) and (3.23) in powers of p] that the following combination is exact at least up to order $O(p^3)$:

$$\frac{3}{5}\mu_e + \frac{2}{5}M_e \cong \mu_2 - \frac{p\,\delta\mu}{1-(1-p)s_2}$$

$$= \mu_2 - \frac{p}{\dfrac{1}{\mu_2-\mu_1} - \dfrac{6(\kappa_2+2\mu_2)(1-p)}{5(3\kappa_2+4\mu_2)\mu_2}}. \tag{3.26}$$

This expression is identical to one of the HS bounds for μ_e in the case of a composite with an isotropic microstructure,[3] and provides a similar bound for the combination $(3\mu_e + 2M_e)/5$ in the case of a cubic microstructure.[9]

IV. SUMMARY AND DISCUSSION

Expressions (3.18), (3.22), (3.23), and (3.26) provide good estimates of the effective moduli under a wide range of

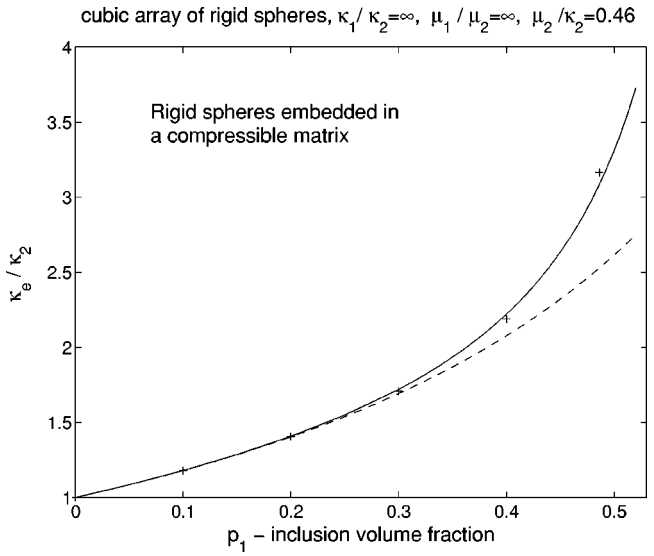

FIG. 1. Normalized effective bulk modulus κ_e/κ_2 vs inclusion volume fraction. Rigid spheres (component 1) embedded in a compressible matrix (component 2): - -, CM approximation; —, third-order approximation of Torquato (Ref. 8); +, numerical data of Nunan and Keller (Ref. 4).

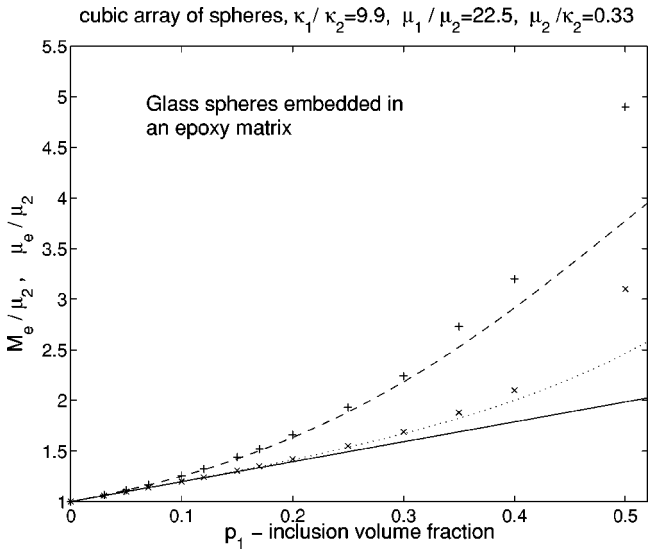

FIG. 2. Normalized effective shear moduli vs inclusion volume fraction for glass spheres (component 1) embedded in an epoxy matrix (component 2): ···, CM approximation for μ_e/μ_2; - -, CM approximation for M_e/μ_2; ×, numerical results (Ref. 16) for μ_e/μ_2; +, numerical results (Ref. 16) for M_e/μ_2; —, dilute approximation.

conditions. Expression (3.18) for the bulk modulus is well known in elastostatics. However, it is known as a HS bound and not as a CM-type approximation for the effective bulk modulus. Nemat-Nasser *et al.*,[6] who investigated the special case of a periodic cubic array of spherical voids, have noticed that their approximation for the effective bulk modulus, the so-called ''simplest approximation,'' overshoots the upper HS bound by about 1%, possibly due to truncation errors in the numerical calculation. They have further claimed that if a particular relation [Eq. (7.7) in Ref. 6] for an infinite series is to hold (we can prove this relation to be exact), then their approximation is identical to the upper HS bound. In view of our results, it seems that the simplest approximation, which involves replacing a nonuniform strain field $\boldsymbol{\varepsilon}(\mathbf{r})$ inside the inclusion by its average value, is the analog of the CM-type approximation. By comparing it with a complete numerical solution, Nemat-Nasser *et al.*[5] concluded that this approximation yields adequate results up to inclusion volume fractions of about 30%. Nunan and Keller,[4] who investigated the opposite extreme case of a periodic cubic array of rigid spheres, formulated a procedure to compute the elastic moduli for this case numerically. By comparing their accurate numerical results for the elastic moduli with the simplest approximation of Nemat-Nasser *et al.*, they found that for low and moderate concentrations, the simplest approximation agrees well with their exact results. In Fig. 1, we compare expression (3.18) for the effective bulk modulus with the results obtained by Nunan and Keller for a simple cubic array of rigid spheres [see Table V in Ref. 4: In terms of the notations used by Nunan and Keller $\kappa_e/\kappa_2=1+(\gamma+2\alpha/3)\mu_2/\kappa_2$], and with the third-order approximation of Torquato [Eq. (4.15) in Ref. 8, where we used the tabulations of the three-point parameter ζ_2 obtained by McPhedran and Milton[15]]. This comparison shows that (a) the results are indistinguishable up to $p=0.2$; (b) at $p=0.3$, the deviation is about 1%; (c) at $p=0.4$, it is about 6%; (d) at higher volume fractions the deviation becomes considerable, especially when the volume fraction approaches its close-packing value $p_c=\pi/6\simeq 0.52$, at which the spheres begin to overlap. The third-order approximation of Torquato, which incorporates third-order correlation functions of the microstructure, actually goes beyond the CM-type approximation (which can be considered as a second-order approximation) and is in good agreement with the numerical results of Nunan and Keller even at high volume fractions. The comparison with two different extreme cases, namely spherical voids and rigid spheres, supports our belief that Eq. (3.18) is exact up to higher orders in p than those stated above and can provide a good approximation for the bulk modulus over a wide range of parameters. Equation (3.26), though less known, was also derived in the past as a bound for this particular combination of the two shear moduli of a cubic array of spheres.[9]

Equations (3.22) and (3.23) for the two shear moduli μ and M were not obtained previously, as far as we know, and also provide good estimates of the effective properties under a wide range of conditions. In Figs. 2 and 3, we compare these CM-type approximations for μ and M with exact numerical calculations we have performed[16] for two cases: (a) the inclusions are stiffer than the host medium (the matrix) by at least one order of magnitude; (b) the inclusions are softer than the matrix by similar factors. We also compare the CM approximation with the low-order dilute approximation, Eq. (3.11) (note that within this approximation $\mu_e=M_e$). As can be seen, the CM approximation is in excellent agreement with the numerical results up to volume fraction, $p\approx 0.2$. Even at higher and moderate volume fractions, it still provides good approximations: In case (a), where the contrast (i.e., the ratio between the elastic moduli of the matrix and that of the inclusions) is greater than 1, this approxi-

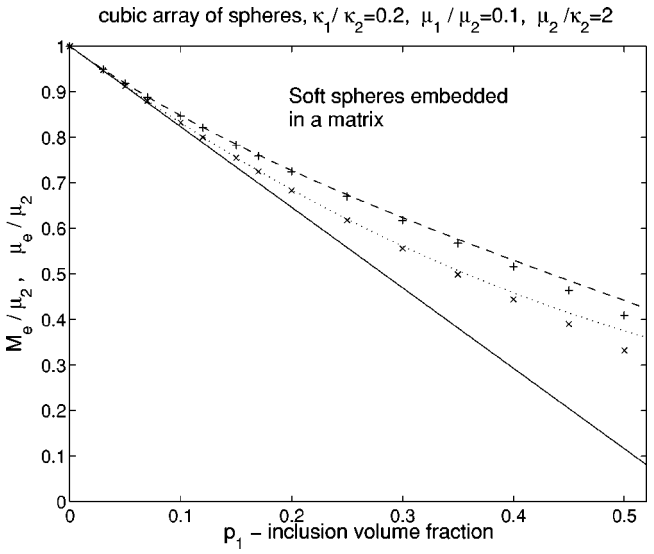

FIG. 3. Normalized effective shear moduli vs inclusion volume fraction. Inclusions (component 1) are softer than the matrix (component 2): ⋯, CM approximation for μ_e/μ_2; - -, CM approximation for M_e/μ_2; ×, numerical results (Ref. 16) for μ_e/μ_2; +, numerical results (Ref. 16) for M_e/μ_2; —, dilute approximation.

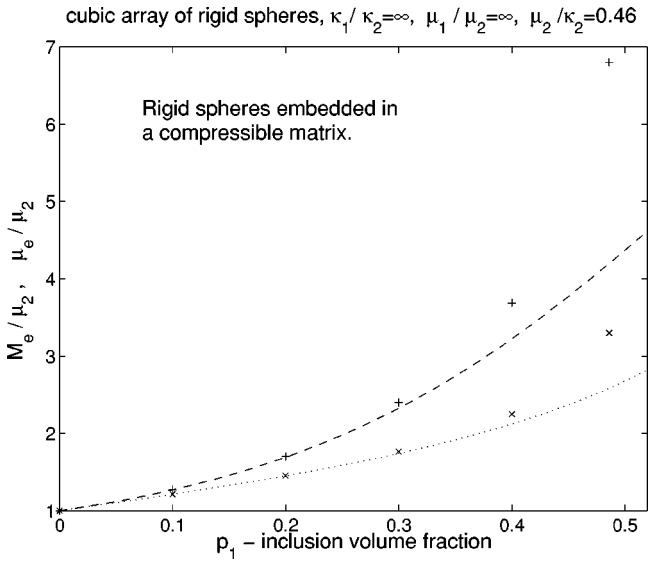

FIG. 4. Normalized effective shear moduli vs inclusion volume fraction. Rigid spheres (component 1) embedded in a compressible matrix (component 2): ⋯, CM approximation for μ_e/μ_2; - -, CM approximation for M_e/μ_2; ×, numerical data of Nunan and Keller (Ref. 4) for μ_e/μ_2; +, numerical data of Nunan and Keller (Ref. 4) for M_e/μ_2.

mation is only about 2% off the exact result at $p=0.3$, but the deviation becomes considerable at high volume fractions—it is about 10% off the exact result at $p=0.4$, beyond which Eqs. (3.22) and (3.23) cease to be good approximations. In case (b), where the contrast is less than 1, the CM-type approximation is only about 3% off the exact result at $p=0.4$. This is very good, especially if we remember that the spheres begin to overlap at $p_c \simeq 0.52$. In Fig. 4, we compare expressions (3.22) and (3.23) with those of Table V of Ref. 4 for the extreme case of rigid spheres. (In terms of the notations used by Nunan and Keller, $\mu_e/\mu_2 = 1+\beta$, $M_e/\mu_2 = 1+\alpha$.) The results are indistinguishable up to volume fraction $p=0.2$. At $p=0.3$ the deviation is about 3% and at $p=0.4$ it becomes considerable (about 10%). As the volume fraction p of the spherical inclusions increases and the distance between the spheres decreases, the interaction between the distortion fields of different inclusions becomes stronger and it no longer suffices to consider only the dipole-dipole interaction, as is done in the CM-type approximation. In order to describe this strong interaction more accurately, one has to consider also the nondipole eigenstates, i.e., the higher multipole distortion fields.

Corrections to the above CM-type expressions begin at order that is not less than $p^{11/3}$, in analogy with the 2D problem of a square lattice of cylindrical inclusions,[1] where it was found that the corrections to the CM approximation begin with order p^5 for the bulk modulus, and with order p^4 for the shear moduli. We note that in the analogous 3D electrostatic problem[10,12] of a cubic array of spheres, the correction to the CM expression actually begins at order $p^{13/3}$, since the correction of the order of $p^{11/3}$ vanishes due to the cubic lattice symmetry. From the 2D problem, we know that many of the matrix elements $Q_{\alpha\beta}$ of higher orders in p vanish because (a) overlap integrals between some types of states vanish (due to the spherical symmetry of the grains, e.g., the vanishing of overlap integrals between any two compression dipoles in our 3D case); (b) many sums of interactions vanish because of the lattice symmetry. Therefore, we expect our CM-type approximations to be exact up to higher orders in p than those stated above, and to provide good estimations of the elastic moduli even for cases where either the inclusion volume fraction is high, provided that the contrast is not too different from 1 (e.g., 0.2, 5), or the contrast is much greater than 1 (e.g., rigid spheres) or much less than 1 (e.g., spherical voids) but the volume fraction is low or moderate. These expectations are supported by previous work and by our numerical calculations.[4–6,8,16] When this is not the case, one must invoke the more accurate methods cited above. For very extreme cases, such as rigid spheres which almost touch each other, yet other approaches must be used, such as asymptotic analysis.[4]

For nonperiodic (e.g., random) systems of nonoverlapping spheres, the original secular equation (2.12) must be used instead of its simplified form (2.15). Even for such cases we can claim, without performing any further calculations, that the CM approximation (3.18) for κ_e is exact at least up to $O(p^2)$. This results from the vanishing of the overlap integrals between any two compression dipoles (see Appendix B), which is a consequence only of the inclusion shape. Therefore, the subspace of the secular equation (2.12), which includes only the compression dipole interactions, will be diagonal. The interaction $Q_{1\alpha}$ between any compression dipole and a shear dipole is of the order of $O(R^3/a^3) = O(p)$, and we can use perturbation treatment, which will give corrections of the order $p|Q_{1\alpha}|^2 = p^3$. The solution will, therefore, differ from Eq. (3.18) only in that order. This conclusion is also supported by the perturbation expansion for the effective elastic moduli developed by Torquato.[7,8] For the

special case of isotropic dispersions, third-order relations for the effective elastic moduli were obtained by using three-point correlation functions of the microgeometry.[8] It was shown there that the series expansion can be regarded as expressing the effects of a perturbation around the optimal structures which realize the HS bounds. Thus, the second-order approximation is actually Eq. (3.18). Expression (3.18) was also derived in the past for the case of isotropic distributions of spheres as a mean-field approximation which may be corrected on the basis of cluster expansions,[17] but it is difficult to calculate the next correction by this approach. Therefore, an estimation of the next correction in powers of p, which involves the solution of a two-sphere problem, was not obtained there.

ACKNOWLEDGMENTS

This research was supported in part by grants from the US-Israel Binational Science Foundation and the Israel Science Foundation.

APPENDIX A: THE OPERATOR $\hat{\Gamma}$

The local displacement field $u_l(\mathbf{r})$ in a composite material whose elastic stiffness tensor is described by Eq. (2.2) must, in principle, be found by solving the differential equations of equilibrium elasticity

$$C^{(2)}_{ijkl}\partial_j\partial_k u_l = \frac{1}{s}\delta C_{ijkl}\partial_j[\theta_1(\mathbf{r})\partial_k u_l], \tag{A1}$$

together with the boundary conditions $u_i = u_i^{(0)} \equiv \varepsilon^{(0)}_{ij}x_j$. We can treat the right-hand side as a source term. Using the tensor Green function $g_{lm}(\mathbf{r},\mathbf{r}';C^{(2)})$ for the problem (d is the dimensionality)

$$C^{(2)}_{ijkl}\partial_j\partial_k g_{lm}(\mathbf{r},\mathbf{r}';C^{(2)}) = -\delta_{im}\delta^d(\mathbf{r}-\mathbf{r}'), \tag{A2}$$

$$g_{lm}(\mathbf{r},\mathbf{r}';C^{(2)}) = 0, \quad \mathbf{r} \text{ on the boundary}, \tag{A3}$$

we obtain for u_l

$$\begin{aligned} u_l(\mathbf{r}) &= u_l^{(0)} - \frac{1}{s}\int dV' g_{lm}(\mathbf{r},\mathbf{r}';C^{(2)})\delta C_{jmkn}\partial'_j[\theta_1(\mathbf{r}')\partial'_k u'_n] \\ &= u_l^{(0)} + \frac{1}{s}\int dV' \theta_1(\mathbf{r}')\partial'_j g_{lm}(\mathbf{r},\mathbf{r}';C^{(2)})\delta C_{jmkn}\partial'_k u'_n, \end{aligned} \tag{A4}$$

where we used integration by parts and the boundary conditions (A3). Taking derivatives of Eq. (A4) and using the symmetries of the elastic stiffness tensor, together with the symmetry of the Green tensor $g_{lm}(\mathbf{r},\mathbf{r}') = g_{ml}(\mathbf{r}',\mathbf{r})$, which follows from reciprocity, we finally obtain an integral equation for the local strain tensor $\varepsilon_{ij}(\mathbf{r}) = \frac{1}{2}(\partial_i u_j + \partial_j u_i)$:

$$\begin{aligned} \varepsilon_{ij}(\mathbf{r}) &= \varepsilon^{(0)}_{ij} \\ &\quad + \frac{1}{s}\int dV' \theta_1(\mathbf{r}')G_{ijkl}(\mathbf{r},\mathbf{r}';C^{(2)})\delta C_{klmn}\varepsilon_{mn}(\mathbf{r}'), \end{aligned} \tag{A5}$$

where

$$G_{ijkl} = \tfrac{1}{4}(\partial_i\partial'_l g_{jk} + \partial_i\partial'_k g_{jl} + \partial_j\partial'_l g_{ik} + \partial_j\partial'_k g_{il}). \tag{A6}$$

Equation (A5) can be written in symbolic form

$$\varepsilon = \varepsilon^{(0)} + \frac{1}{s}\hat{\Gamma}\varepsilon, \tag{A7}$$

thus defining the linear integral operator $\hat{\Gamma}$. Clearly, $\hat{\Gamma}$ depends on the microstructure [through $\theta_1(\mathbf{r})$] and on $C^{(2)}$, but is independent of $C^{(1)}$.

APPENDIX B: FINDING THE DIPOLE EIGENSTATES

The strain eigenstates of the single grain problem are solutions of the equilibrium equations of elastostatics (in the absence of body forces): $\partial_j\sigma_{ij} = 0$, where σ is the stress tensor, related to the strain tensor by the linear local relation $\sigma_{ij}(\mathbf{r}) = C_{ijkl}(\mathbf{r})\varepsilon_{kl}(\mathbf{r})$. In terms of the displacement $\mathbf{u}$, and for uniform isotropic components, these equations become

$$\mu\nabla^2\mathbf{u} + (\lambda+\mu)\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\mathbf{u}) = \mathbf{0}, \tag{B1}$$

where λ, μ are the Lamé coefficients, related to the bulk modulus by the relation (in three dimensions) $\kappa = \lambda + 2\mu/3$. We solve these equations inside each component of the composite material [i.e., a spherical isotropic inclusion $C'^{(1)}(s)$ embedded in an isotropic host $C^{(2)}$] separately, then we impose continuity of both the displacement and the traction at the surface of the sphere. Since the eigenstates represent a situation where the sample is internally deformed and strained but the external boundaries are undeformed, the strain must decrease to zero for $|\mathbf{r}| \to \infty$ and should also be nonsingular everywhere else. This requirement restricts the number of allowed strain states which can appear in a linear combination for $|\varepsilon^{(\alpha\alpha)}\rangle$. From the continuity conditions, we get a set of homogeneous equations for the coefficients of the linear combination. Setting the determinant equal to zero leads to special allowed values which the effective elastic tensor $C'^{(1)}$can take inside the sphere, and therefore determines the eigenvalues s_α by Eq. (2.1).

The special case of dipole eigenstates corresponds to a strain field $\varepsilon_{ij}(\mathbf{r})$ which is uniform inside the inclusion (a sphere) and decreases the most slowly with distance outside the inclusion. We *chose* these dipole resonances to be certain uniform strains inside the sphere [see Eqs. (3.1) and (3.3)] and then followed the procedure described above in order to find their shape outside the sphere, along with their eigenvalues. Since the microstructure is spherically symmetric, it is best to express the equilibrium equations in spherical coordinates (r,θ,ϕ), where $\mathbf{e}_r$, $\mathbf{e}_\theta$, $\mathbf{e}_\phi$ will denote the appropriate unit vectors.

Only in rare instances can an interesting problem in 3D elasticity be solved by elementary means. One such case is the deformation of a spherical shell by uniform internal and external pressures (see, e.g., Refs. 18 and 19). Finding the compression dipole eigenstate is a similar problem, since the displacement vector which corresponds to it has only one component in the **r** direction which is spherically symmetric. Imposing on this solution the additional requirements that an eigenstate must fulfill and the continuity conditions, as specified above, we arrive at expressions (3.1) and (3.2) for the compression eigenstate and eigenvalue.

The other five dipole eigenstates correspond to pure shear distortions inside the sphere. Since these states lack spherical symmetry, they are very hard to find. The simplest shear eigenstate to be found should be $\varepsilon_{ij}^{(6)}$, since it corresponds to a displacement field inside the sphere which has azimuthal symmetry

$$\mathbf{u}^{(6)}(\mathbf{r}) = r[-2P_2(\cos\theta)\mathbf{e}_r + P_2^{(1)}(\theta)\mathbf{e}_\theta], \tag{B2}$$

where $P_n(\cos\theta)$ is the Legendre polynomial and $P_n^{(m)}(\theta)$ is the associated Legendre function. In order to find the solution outside the sphere, we used the set of solutions of the elastostatic equilibrium equations found in Ref. 20. Outside the sphere these solutions are $\mathbf{M}_{nm}^2, \mathbf{N}_{nm}^2, \mathbf{E}_{nm}^2$, where $\mathbf{M}_{nm}^2, \mathbf{N}_{nm}^2$ are two sets of transverse solutions and $\mathbf{E}_{nm}^2$ is the set of longitudinal solutions. (In the notation of Ref. 20, $\mathbf{u}^{(6)} = -\mathbf{N}_{01}^1$ inside the sphere.) The solution outside the sphere should have the same azimuthal symmetry and in order to satisfy continuity conditions everywhere on the surface of the sphere it must also have the same dependence on θ. The only solutions which fulfill these requirements are $\mathbf{N}_{02}^2, \mathbf{E}_{02}^2$, therefore outside the sphere $\mathbf{u}^{(6)}(\mathbf{r})$ should be a linear combination of these solutions only. Following the procedure described above to find the eigenvalues [imposing continuity of both the displacement field (u_r, u_θ) and the tractions $(\sigma_{rr}, \sigma_{r\theta})$], we obtain Eqs. (3.4)–(3.6). Note that the strain eigenstate is written in Eq. (3.6) in Cartesian coordinates, not in spherical coordinates, in order to exploit the cubic symmetry of the composite in a more obvious fashion when calculating lattice sums for the matrix elements $Q_{\alpha\beta}$ (see Appendix C).

The form of the other four shear dipole eigenstates outside the sphere can be obtained from $\varepsilon_{ij}^{(6)}$ by appropriate symmetry rotations. Since the elastostatic equilibrium equations and the integral operator $\hat{\Gamma}$ are invariant under such rotations, any state obtained from $\varepsilon_{ij}^{(6)}$ by performing such rotations will still be an eigenstate with the same eigenvalue, and the same is true for any linear combination of such states. Therefore, we just have to find appropriate rotations (and linear combinations of such rotations) which transform $\varepsilon_{ij}^{(6)}$ inside the sphere into one of the other shear eigenstates of Eqs. (3.3) and apply the same transformations to the $\varepsilon_{ij}^{(6)}$ outside the sphere; in this way we obtain the forms of all the shear eigenstates outside the sphere.

APPENDIX C: EVALUATION OF DIPOLE-DIPOLE INTERACTIONS $Q_{\alpha\beta}$

The main difficulty after finding the dipole eigenstates is to calculate the matrix elements $Q_{\alpha\beta}$ $(\alpha,\beta=1,\ldots,6)$ of the eigenvalue problem (2.15). These elements are given by Eq. (3.16) $(Q_{\beta\alpha}=Q_{\alpha\beta}^*)$:

$$\begin{aligned} Q_{\beta\alpha} &= \lim_{L\to\infty} \sum_{\substack{a \\ 0<|\mathbf{r}_b-\mathbf{r}_a|<L}} \frac{\langle \tilde{\varepsilon}^{(b\beta)}\theta_b | \hat{\Gamma} | \theta_a \varepsilon^{(a\alpha)}\rangle}{\|\varepsilon^{(b\beta)}\| \, \|\varepsilon^{(a\alpha)}\|} - ps_\alpha \delta_{\alpha\beta} \\ &= \lim_{L\to\infty} \sum_{\substack{a \\ 0<|\mathbf{r}_b-\mathbf{r}_a|<L}} \frac{s_\alpha \int \theta_b \varepsilon^{(b\beta)*} \delta C \varepsilon^{(a\alpha)} dV}{\|\varepsilon^{(b\beta)}\| \, \|\varepsilon^{(a\alpha)}\|} - ps_\alpha \delta_{\alpha\beta}. \end{aligned} \tag{C1}$$

In order to evaluate the overlap integrals, we locate the origin of our coordinate axes at the center of the sphere *b*, while the location of the center of the sphere *a* will be denoted by $\mathbf{r}_a$. With this choice, the overlap integrals in Eq. (C1) are given by

$$\langle \tilde{\varepsilon}^{(b\beta)}\theta_b | \varepsilon^{(a\alpha)}\rangle = \int dV \theta_b(\mathbf{r}) \tilde{\varepsilon}^{(\beta)*}(\mathbf{r}) \varepsilon^{(\alpha)}(\mathbf{r}-\mathbf{r}_a). \tag{C2}$$

Note that summation over tensorial indices is implied on the right-hand side, and that an expression for $|\mathbf{r}|<R$ is to be used for $\tilde{\varepsilon}^{(\beta)*}(\mathbf{r})$, while an expression for $|\mathbf{r}-\mathbf{r}_a|>R$ is to be used for $\varepsilon^{(\alpha)}(\mathbf{r}-\mathbf{r}_a)$. The fact that the dipole states have a uniform value of $\tilde{\varepsilon}^{(\beta)}$ for $r<R$ along with the fact that for these states $\tilde{\varepsilon}^{(\beta)} \propto \varepsilon^{(\beta)}$ for $r<R$ simplify the integrand of Eq. (C2). Thus, using the fact that $\varepsilon_{ii}^{(1)}=0$ for $|\mathbf{r}-\mathbf{r}_a|>R$ [see Eq. (3.1)], we immediately obtain that the interaction between any two compression dipole eigenstates of different inclusions vanishes:

$$\langle \tilde{\varepsilon}^{(b1)}\theta_b | \varepsilon^{(a1)}\rangle \propto \int dV \theta_b \varepsilon_{ii}^{(1)}(\mathbf{r}-\mathbf{r}_a) = 0, \tag{C3}$$

and therefore $Q_{11} = -ps_1$.

The same procedure can be applied in order to calculate the interactions between the compression dipole and any of the shear dipoles $\varepsilon^{(2)}, \ldots, \varepsilon^{(6)}$. In these cases, however, the overlap integrals do not vanish, but due to the cubic symmetry of the lattice, it can be shown without explicit calculation that $Q_{1\alpha}=0$ for $\alpha\neq 1$. For instance, the overlap integral with the $\varepsilon^{(5)}$ state is

$$\langle \tilde{\varepsilon}^{(b5)}\theta_b | \varepsilon^{(a1)}\rangle \propto \int dV \theta_b(\mathbf{r}) [\varepsilon_{11}^{(1)}(\mathbf{r}-\mathbf{r}_a) - \varepsilon_{22}^{(1)}(\mathbf{r}-\mathbf{r}_a)]. \tag{C4}$$

By Eq. (3.1)

$$\varepsilon_{11}^{(1)}(\mathbf{r}) - \varepsilon_{22}^{(1)}(\mathbf{r}) \propto \frac{1}{r^5}(x^2-y^2). \tag{C5}$$

Since this expression is to be expanded around $\mathbf{r}_a=(a_x,a_y,a_z)$, therefore the roles of x^2,y^2 are not equivalent in the integrand of Eq. (C4). But since, eventually, we have to sum these integrals over all lattice sites a that lie inside a sphere of radius L, then, as far as $Q_{\beta\alpha}$ is concerned, the roles of x^2,y^2 are equivalent (we choose the Cartesian axes to be the cubic symmetry axes of the lattice), and they can replace each other in a lattice with cubic symmetry. Therefore, we can make replacements such as $x^2\rightarrow y^2$ in the integrand of Eq. (C2), and especially we obtain $Q_{15}=0$.

This procedure can be applied in order to calculate any element $Q_{\beta\alpha}$ $(\alpha,\beta=1,\ldots,6)$. In this way, we find that $Q_{\beta\alpha}$ is a *diagonal* matrix. This is clearly a result of the cubic lattice symmetry.

APPENDIX D: CALCULATION OF G_M

By its definition, Eq. (3.21), G_M is given by the following expression:

$$G_M=\lim_{L\rightarrow\infty}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{\int dV\theta_b\varepsilon^{(b6)*}\delta C\varepsilon^{(a6)}}{\|\varepsilon^{(b6)}\|\ \|\varepsilon^{(a6)}\|}. \quad \text{(D1)}$$

In order to evaluate the overlap integrals, we locate the origin of our coordinate axes at the center of the sphere b, while the location of the center of the sphere a will be denoted by $\mathbf{r}_a$. With this choice, the overlap integrals in Eq. (D1) are given by

$$\int dV\theta_b\varepsilon^{(b6)*}\delta C\varepsilon^{(a6)}=\int_{V_R}dV\varepsilon^{(6)*}(\mathbf{r})\delta C\varepsilon^{(6)}(\mathbf{r}-\mathbf{r}_a), \quad \text{(D2)}$$

where the integration is performed inside a sphere of radius R and volume V_R located at the origin. Note that summation over tensorial indices is implied in the integrand, and that expression (3.3c) (for $|\mathbf{r}|<R$) is to be used for $\varepsilon^{(6)}(\mathbf{r})$, while expression (3.6) (for $|\mathbf{r}-\mathbf{r}_a|>R$) is to be used for $\varepsilon^{(6)}(\mathbf{r}-\mathbf{r}_a)$. Thus we find

$$G_M=\frac{1}{6V_R}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\int_{V_R}dV[\varepsilon_{11}^{(6)}(\mathbf{r}-\mathbf{r}_a)+\varepsilon_{22}^{(6)}(\mathbf{r}-\mathbf{r}_a)-2\varepsilon_{33}^{(6)}(\mathbf{r}-\mathbf{r}_a)]. \quad \text{(D3)}$$

In order to simplify things, we will use the fact that due to the cubic symmetry $Q_{16}=0$ (see Appendix C), therefore also $Q_{61}=Q_{16}^*=0$. Using the explicit form of $\varepsilon^{(1)}$ for $|\mathbf{r}|<R$, Eq. (3.1), this implies that

$$0=\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\int_{V_R}dV[\varepsilon_{11}^{(6)}(\mathbf{r}-\mathbf{r}_a)+\varepsilon_{22}^{(6)}(\mathbf{r}-\mathbf{r}_a)+\varepsilon_{33}^{(6)}(\mathbf{r}-\mathbf{r}_a)]. \quad \text{(D4)}$$

Using this result in Eq. (D3) we obtain a simplified expression for G_M:

$$G_M=-\frac{1}{2V_R}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\int_{V_R}dV\varepsilon_{33}^{(6)}(\mathbf{r}-\mathbf{r}_a). \quad \text{(D5)}$$

When substituting expression (3.6c) for $\varepsilon_{33}^{(6)}(\mathbf{r})$, we can exploit the cubic symmetry in order to make replacements such as $x^2\rightarrow y^2$. At first sight, one may think that, since this expression is to be expanded around $\mathbf{r}_a=(a_x,a_y,a_z)$, therefore the roles of x^2,y^2 are not equivalent in the integrand of Eq. (D5). But since, eventually, we have to sum these integrals over all lattice sites a that lie inside a sphere of radius L, then, as far as G_M is concerned, the roles of x^2,y^2,z^2 are equivalent (we choose the Cartesian axes to be the cubic symmetry axes of the lattice), and they can replace each other in a lattice with cubic symmetry. Therefore, we can make replacements such as $x^2\rightarrow y^2$ in the integrand of Eq. (D5), and in particular, we can replace z^2/r^2 by 1/3.

After making such replacements and collecting terms, we arrive at the following expression for G_M:

$$G_M=-\frac{1}{2V_R}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\int_{V_R}dVf(\mathbf{r}-\mathbf{r}_a), \quad \text{(D6)}$$

where

$$\begin{aligned}f(\mathbf{r})&=-\frac{9}{2}\frac{\lambda+\mu}{3\lambda+8\mu}\left(5R^3\frac{1-5\cos^4\theta}{r^3}-7R^5\frac{1-5\cos^4\theta}{r^5}\right)\\&=\frac{18}{7}\frac{\lambda+\mu}{3\lambda+8\mu}\left(5R^3\frac{5P_2(\cos\theta)+2P_4(\cos\theta)}{r^3}-7R^5\frac{5P_2(\cos\theta)+2P_4(\cos\theta)}{r^5}\right).\end{aligned} \quad \text{(D7)}$$

Note that λ,μ refer to the elastic moduli of component 2, $P_n(\cos\theta)$ is a Legendre polynomial. Using the same argument as before, because of the cubic lattice symmetry of our composite material, the contribution of the term $P_2(\cos\theta)=(3z^2/r^2-1)/2$ to G_M will be canceled, therefore we need only consider the function

$$f(\mathbf{r})=\frac{36}{7}\frac{\lambda+\mu}{3\lambda+8\mu}\left(5R^3\frac{P_4(\cos\theta)}{r^3}-7R^5\frac{P_4(\cos\theta)}{r^5}\right). \quad \text{(D8)}$$

In order to continue further, we need to expand $f(\mathbf{r})$ around the lattice point a, which will be denoted by the lattice vector $\mathbf{r}_a=(a,\theta_a,\phi_a)$. For this purpose, we use expansion for solutions of Laplace's equation found in Refs. 10 and 12 in spherical harmonics centered around the origin,

$$\frac{Y_{l'}^{(m')}(\Omega_{\mathbf{r}-\mathbf{r}_a})}{|\mathbf{r}-\mathbf{r}_a|^{l'+1}}=\sum_{n,m}(-1)^{l'+m'}[4\pi(2l'+2n+1)(2l'+1)(2n+1)]^{1/2}\begin{pmatrix} l'+n & l' & n \\ m'-m & -m' & m\end{pmatrix}$$

$$\times\frac{(2l'+2n-1)!!}{(2n+1)!!(2l'-1)!!}r^n Y_n^{(m)}(\Omega_{\mathbf{r}})\frac{Y_{l'+n}^{(m'-m)}(\Omega_{\mathbf{r}_a})}{a^{l'+n+1}}, \tag{D9}$$

where the explicit expression for the $3-j$ symbol for $M=m_1+m_2$, $J=j_1+j_2$ is

$$\begin{pmatrix} J & j_1 & j_2 \\ -M & m_1 & m_2\end{pmatrix}=(-1)^{j_1-j_2+M}\left[\frac{(2l_1)!(2l_2)!}{(2J+1)!}\frac{(J+M)!(J-M)!}{(l_1+m_1)!(l_1-m_1)!(l_2+m_2)!(l_2-m_2)!}\right]^{1/2}. \tag{D10}$$

We can now use Eq. (D9) in order to expand the term $P_4(\cos\theta)/r^5$ of Eq. (D8) around $\mathbf{r}_a$. As for the other term $P_4(\cos\theta)/r^3$, we write it as $[P_4(\cos\theta)/r^5]r^2$ and expand r^2 around $\mathbf{r}_a$, $|\mathbf{r}-\mathbf{r}_a|^2=r^2-2ar\cos\gamma+a^2$, where γ is the angle between $\mathbf{r}$ and $\mathbf{r}_a$, then use the addition theorem for spherical harmonics in order to express $\cos\gamma=P_1(\cos\gamma)$ in terms of spherical harmonics of the 3D angles $\Omega_{\mathbf{r}},\Omega_{\mathbf{r}_a}$: $P_1(\cos\gamma)=(4\pi/3)\Sigma_{m=-1}^{1}Y_1^{(m)}(\Omega_{\mathbf{r}_a})Y_1^{(m)*}(\Omega_{\mathbf{r}})$. After performing the above expansions, we use the orthogonality of spherical harmonics in order to perform the integration of $f(\mathbf{r}-\mathbf{r}_a)$ inside a sphere of radius R located at the origin to obtain

$$G_M=-\frac{1}{2V_R}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}36\left(\frac{\lambda+\mu}{3\lambda+8\mu}\right)$$

$$\times\left(\frac{5}{7}V_R\left(\frac{R}{a}\right)^3P_4(\cos\theta_a)-2V_R\left(\frac{R}{a}\right)^5P_4(\cos\theta_a)\right)$$

$$=\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{27}{2\pi}\left(\frac{\lambda+\mu}{3\lambda+8\mu}\right)\left(-\frac{5}{7}\frac{P_4(\cos\theta_a)}{(a/a_0)^3}p\right.$$

$$\left.+2\left(\frac{3}{4\pi}\right)^{2/3}\frac{P_4(\cos\theta_a)}{(a/a_0)^5}p^{5/3}\right), \tag{D11}$$

where $p=(4\pi/3)R^3/a_0^3$ is the volume fraction of the spherical inclusions. The lattice sums must be performed numerically. Note that the sum of $P_4(\cos\theta_a)/(a/a_0)^3$ is only semiconvergent: It depends crucially on the particular type of summation, in our case all the cubic lattice points inside a sphere of radius L. In contrast with that, the sum of $P_4(\cos\theta_a)/(a/a_0)^5$ is absolutely convergent. The numerical results for a simple cubic lattice are

$$\lim_{L\to\infty}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{P_4(\cos\theta_a)}{(a/a_0)^3}=2.724, \tag{D12}$$

$$\lim_{L\to\infty}\sum_{\substack{a\\0<|\mathbf{r}_b-\mathbf{r}_a|<L}}\frac{P_4(\cos\theta_a)}{(a/a_0)^5}=3.108. \tag{D13}$$

Inserting these numerical values into Eq. (D11) and using the relation $\kappa=\lambda+2\mu/3$, we obtain Eq. (3.25).

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