Long-wavelength propagation in composite elastic media I. Spherical inclusions

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A self-consistent method of estimating effective macroscopic elastic constants for inhomogeneous materials with spherical inclusions is formulated based on elastic-wave scattering theory. The method for general ellipsoidal inclusions will be presented in the second part of this series. The case of spherical inclusions is particularly simple and therefore provides an elementary introduction to the general method. The self-consistent effective medium is determined by requiring the scattered, long-wavelength displacement field to vanish on the average. The resulting formulas are simpler to apply than previous self-consistent scattering theories due to the reduction from tensor to vector equations. In the limit of long wavelengths, our results for spherical inclusions agree with statically derived self-consistent moduli of Hill and Budiansky. Our self-consistent formulas are also compared both to the estimates of Kuster and Toksöz and to the rigorous Hashin–Shtrikman bounds. (For spherical inclusions and long wavelengths, the Kuster–Toksöz effective moduli are known to be identical to the Hashin–Shtrikman bounds.) A result of Hill for two-phase composites is generalized by proving that the self-consistent effective moduli always lie between the Hashin–Shtrikman bounds for n-phase composites. Numerical examples for a two-phase medium with viscous fluid and solid constituents show that the real part of our self-consistent moduli always lie between the rigorous bounds, in agreement with the analytical results. Some of the practical details in the numerical solution of the coupled, nonlinear self-consistency equations are discussed. Examples of velocities and attenuation coefficients estimated when the solid constituent possesses intrinsic absorption are also presented.

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INTRODUCTION

A new approach to the problem of estimating elastic constants of composite media has been proposed recently by the author.[1] In the present series of papers, the practical computational aspects of the method will be stressed and the results compared to known rigorous bounds on the elastic moduli. In this first paper in the series concerning spherical inclusions, the self-consistency equations determining the effective elastic moduli are given a simplified derivation which provides an elementary introduction to the general method. In the second paper, a more general derivation appropriate for ellipsoidal inclusions will be presented and further comparison to the results of other "self-consistent" methods for needle and disk inclusions will be made. The main goal of these papers is to provide convincing evidence of the usefulness of the method and to show that the numerical results are always consistent with the best known rigorous bounds on the elastic moduli. Another paper (which will be published elsewhere[2]) will treat the more theoretical aspects of the derivation from the point of view of multiple scattering theory for elastic waves.

The general background for the present method may be found in two recent review articles.[3,4] Watt, Davies, and O'Connell[3] discuss in detail the various methods which have been applied successfully to the study of elastic properties of composite materials. Included among the most notable methods are: (1) the variational method of Hashin and Shtrikman[5] for obtaining rigorous bounds on the elastic moduli and (2) the self-consistent approximation method of Hill[6] and Budiansky[7] for obtaining estimates of the effective elastic moduli. Elliott, Krumhansl, and Leath[4] review the application of the coherent potential approximation (CPA) to a wide variety of physical phenomena including the elastic properties of polycrystalline aggregates. The CPA is a general method based on multiple scattering theory which has several aliases including the effective medium theory (EMT) and the self-consistent method (SCM)—the particular choice of alias depending on the application. According to Bergman,[8] the earliest application of an effective medium theory was the one given by Bruggeman[9] for dielectrics. Recent formulations of the scattering theory method for elastic properties have been given by Zeller and Dederichs,[10] Korringa,[11] and Gubernatis and Krumhansl.[12]

The self-consistent approach which we will describe in the following pages has much in common with the self-consistent methods listed in the previous paragraph. The main point of departure of our scattering-theory approach is that we treat the displacement field **u** directly rather than treating the stress and strain tensors. Mal and Knopoff[13] have derived integral equations for the displacement field in the presence of a spherical inclusion. Kuster and Toksöz[14] used these integral equations to obtain estimates of the effective elastic moduli but their approach was not self-consistent. Our method follows easily from the approach of Kuster and Toksöz when self-consistency is required.

The outline of the paper is as follows: First, we quote the single-scatterer results of Yamakawa[15] and of Kuster and Toksöz.[14] Then we derive estimates of elastic constants using both the method of Kuster and Toksöz and the self-consistent method. Next, we compare these two estimates to the rigorous Hashin–Shtrikman bounds. In the static limit, we find that the Kuster–Toksöz estimates are identical to the Hashin–Shtrikman bounds and that the self-consistent estimates always lie between the bounds. In the third section, we present a series of sample calculations to illustrate typical results. The expected range of validity of the

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theory is discussed in the fourth section. The final section attempts to answer some criticisms which have been directed against effective medium theories in the past. Details concerning a singular point in the self-consistent equations are given special treatment in an Appendix.

I. EFFECTIVE ELASTIC CONSTANTS

Consider an isotropic and homogeneous spherical inclusion of radius a with bulk modulus K_i, shear modulus μ_i, and density ρ_i imbedded in an isotropic and homogeneous infinite matrix characterized by the constants K_m, μ_m, and ρ_m. The bulk and shear moduli may be real or complex. Suppose that a plane compressional wave with displacement $\mathbf{u}$ is incident along the x axis and that

$$\mathbf{u} = \hat{x}(ik)^{-1} \exp i(kx - \omega t). \tag{1}$$

In Eq. (1), k is the wavenumber and ω is the angular frequency. We have arbitrarily chosen the amplitude of the incident plane-wave equal to unity. Note that the factor $(ik)^{-1}$ is required to give $\mathbf{u}$ the dimensions of displacement and also to assure a real, finite strain tensor $\epsilon_{ij} = \partial u_i/\partial x_j + \partial u_j/\partial x_i$ in the limit $k \to 0$.

The spherical inclusion of type-i material will scatter the incident plane-wave producing scattered compressional and shear waves both inside and outside the sphere. When the wavelengths of interest are long so that $(ka) \ll 1$, the approximate solution of the resulting boundary value problem is straightforward but quite tedious and has been solved by Yamakawa[15] and also by Kuster and Toksöz.[14] These calculations need not be repeated here.

When the matrix is a solid $[\mathrm{Re}(\mu_m) \neq 0]$, the results for the radial and transverse components of the scattered wave from an inclusion of radius a_i centered at $\boldsymbol{\xi}_i$ are, respectively,

$$(u_r^s)_i = (ik)^{-1}(ka_i)^3 \exp i(kr_i - \omega t)/kr_i \times [B_0 - B_1\cos\theta - B_2(3\cos 2\theta + 1)/4], \tag{2}$$

and

$$(u_t^s)_i = (ik)^{-1}(sa_i)^3 \exp i(sr_1 - \omega t)/sr_i \times [B_1 \sin\theta + (3s/4k)B_2 \sin 2\theta], \tag{3}$$

where

$$B_0(K_m, K_i, \mu_m) = (K_m - K_i)/(3K_i + 4\mu_m), \tag{4}$$

$$B_1(\rho_m, \rho_i) = (\rho_m - \rho_i)/3\rho_m, \tag{5}$$

and

$$B_2(\mu_m, \mu_i, K_m) = \frac{20\mu_m(\mu_i - \mu_m)/3}{6\mu_i(K_m + 2\mu_m) + \mu_m(9K_m + 8\mu_m)}. \tag{6}$$

In (2) and (3), $r_i = |\mathbf{x} - \boldsymbol{\xi}_i|$, $k = \omega[\rho_m/(K_m + \frac{4}{3}\mu_m)]^{1/2}$, and $s = \omega(\rho_m/\mu_m)^{1/2}$—$k$ and s being the magnitudes of the wavevectors for compressional and shear waves in the matrix.

When the matrix is an inviscid fluid ($\mu_m = 0$), the result[14] for the radial component of the scattered wave is

$$(u_r^s)_i = (ik)^{-1}(ka_i)^3 \exp i(kr_i - \omega t)/kr_i \times (B_0 - B_1\cos\theta), \tag{7}$$

where

$$B_0(K_m, K_i, 0) = (K_m - K_i)/3K_i, \tag{8}$$

and for potential flow

$$B_1(\rho_m, \rho_i) = \frac{\rho_m - \rho_i}{\rho_m + 2\rho_i} = \left(\frac{3\rho_m}{\rho_m + 2\rho_i}\right)\left(\frac{\rho_m - \rho_i}{3\rho_m}\right). \tag{9}$$

Of course, no transverse scattered wave is possible.

In both cases, terms of order $(ka_i)^5$ have been neglected. The main difference between the solid and fluid matrix cases is the inertial terms (5) and (9). This difference is a consequence of the fact that the virtual mass of a sphere oscillating in an ideal fluid is the mass of the sphere plus one half the mass of the displaced fluid (induced mass effect,[16] see Appendix B).

Given these single-scatterer results, we will now discuss two methods of estimating the effective elastic constants of a composite material.

A. Kuster-Toksöz estimates

Kuster and Toksöz[14] used the following method to estimate effective elastic constants. Consider a sphere of radius a of the composite material (with $i = 1, m = 2$) imbedded in an infinite matrix of type-2 material (see Fig. 1). If we do a hypothetical scattering experiment in this medium, the net scattering from all type-1 spheres (neglecting multiple scattering and still assuming $ka \ll 1$) will depend only on the relative volume concentration c_1 of type-1 material in the composite. At low concentrations of inclusion material, this scattering must be equivalent to that obtained by replacing the

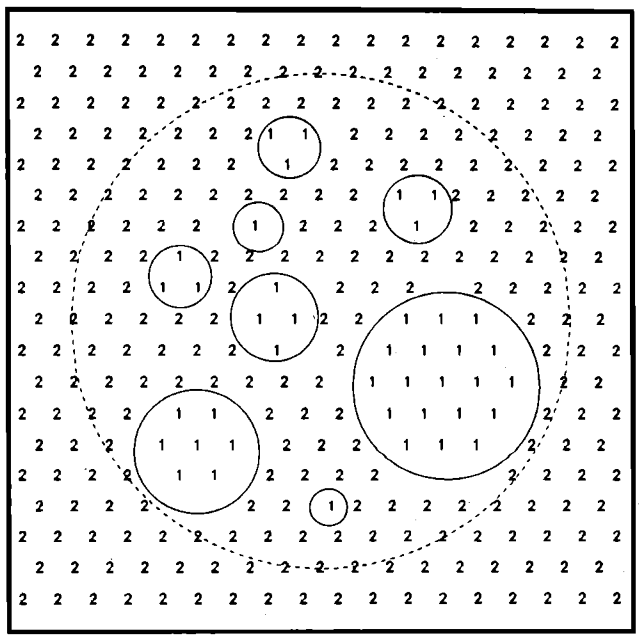

FIG. 1. Schematic diagram of the model composite used by Kuster and Toksöz to derive Eq. (11). The true composite is replaced by a large sphere (dashed line) containing spherical inclusions of type-1 material. Matrix is composed of type-2 material.

composite sphere by an homogeneous sphere with the same effective elastic constants (K^*, μ^*, ρ^*) as the composite, i.e., replace the large sphere in Fig. 1 by a sphere of type-* material.

Thus, for N scatterers in a sphere of radius a centered at the origin, we find

$$(u_r^s)^* \cong \sum_{i=1}^{N} (u_r^s)_i, \quad (u_t^s)^* \cong \sum_{i=1}^{N} (u_t^s)_i. \tag{10}$$

Substituting (2) and (3) into (10) and using the approximation for large $r = |\mathbf{x}|$ that $r_i \cong r - \mathbf{n}\cdot\boldsymbol{\zeta}_i$, where $\mathbf{n} = \mathbf{x}/r$, Eq. (10) for the radial component reduces to

$$\begin{aligned} &a^3[B_0(K_2, K^*, \mu_2) - B_1(\rho_2, \rho^*)\cos\theta - B_2(\mu_2, \mu^*, K_2)(3\cos 2\theta + 1)/4] \\ &\cong \sum_{i=1}^{N} a_i^3 \exp(-ik\mathbf{n}\cdot\boldsymbol{\zeta}_i)[B_0(K_2, K_1, \mu_2) - B_1(\rho_2, \rho_1)\cos\theta - B_2(\mu_2, \mu_1, K_2)(3\cos 2\theta + 1)/4] \end{aligned} \tag{11}$$

and a similar expression may be written for the transverse component. The only dependence on k in (11) is in the phase factor on the rhs. We note that by assumption $k|\mathbf{n}\cdot\boldsymbol{\zeta}_i| < ka \ll 1$ so that for very long wavelengths the phase factor may be replaced by unity. Finally, if the total volume of the inclusions is chosen properly so that $c_1 = \sum a_i^3/a^3$, we obtain from (11)

$$\begin{aligned} B_0(K_2, K^*, \mu_2) &= c_1 B_0(K_2, K_1, \mu_2), \\ B_1(\rho_2, \rho^*) &= c_1 B_1(\rho_2, \rho_1), \\ B_2(\mu_2, \mu^*, K_2) &= c_1 B_2(\mu_2, \mu_1, K_2). \end{aligned} \tag{12}$$

The equations for the transverse scattered wave reproduce the last two equations in (12).

For a solid matrix, the Kuster–Toksöz estimates are therefore given by

$$(K_2 - K^*)/(3K^* + 4\mu_2) = c_1[(K_2 - K_1)/(3K_1 + 4\mu_2)], \tag{13}$$

$$\rho_2 - \rho^* = c_1(\rho_2 - \rho_1), \tag{14}$$

$$(\mu^* - \mu_2)/(\mu^* + F_2) = c_1[(\mu_1 - \mu_2)/(\mu_1 + F_2)], \tag{15}$$

where

$$F_i = (\mu_i/6)[(9K_i + 8\mu_i)/(K_i + 2\mu_i)], \tag{16}$$

and

$i = 2$ in (15).

For an inviscid fluid matrix, a similar derivation yields

$$(K_2 - K^*)/K^* = c_1(K_2 - K_1)/K_1, \tag{17}$$

$$(\rho_2 - \rho^*)/(\rho_2 + 2\rho^*) = c_1(\rho_2 - \rho_1)/(\rho_2 + 2\rho_1). \tag{18}$$

Two comments should be made at this point. First, note that, if instead we treat the type-1 constituent as the matrix and type-2 as the inclusion, we can use the method of Kuster and Toksöz to obtain a second set of formulas to estimate the effective elastic constants. These formulas are obtained from (13)–(18) by interchanging the roles of 1 and 2, i.e., $1 \leftrightarrow 2$. Except for (14), the resulting formulas are different from the original so we see that the Kuster–Toksöz estimates are not symmetric. Second, note that Kuster–Toksöz estimates may be given for an arbitrary number of constituents with volume concentrations c_i, where $\sum_{i=1}^{n} c_i = 1$. If the matrix material is type-1, the Kuster–Toksöz estimates for a solid matrix take the form

$$1/(K^* + \tfrac{4}{3}\mu_1) = \sum_{i=1}^{n} c_i/(K_i + \tfrac{4}{3}\mu_1), \tag{19}$$

$$\rho^* = \sum_{i=1}^{n} c_i \rho_i, \tag{20}$$

and

$$1/(\mu^* + F_1) = \sum_{i=1}^{n} c_i/(\mu_i + F_1), \tag{21}$$

where F_1 is given by (16) for $i = 1$. For an inviscid fluid matrix, the equivalent results are

$$1/K^* = \sum_{i=1}^{n} c_i/K_i, \tag{22}$$

$$\rho^*/(\rho_1 + 2\rho^*) = \sum_{i=1}^{n} c_i\rho_i/(\rho_1 + 2\rho_i), \tag{23}$$

and of course $\mu^* = 0$.

B. Self-consistent estimates

The concept used by Kuster and Toksöz to estimate effective elastic constants is valid at low inclusion concentrations where single scattering effects dominate. However, at higher concentrations a different concept is generally required.

Consider a sphere of radius a of the composite material imbedded in an infinite matrix of type-* material. For now, the elastic properties of this matrix are arbitrary. Next replace the true composite sphere with a sphere whose matrix is also of type-* material containing spherical inclusions of both type-1 and type-2 material in the same relative proportions as in the original composite (see Fig. 2). For hypothetical scattering experiments in this medium, the net scattering from all type-1 and type-2 spheres may be estimated again [using (2)–(6)] by neglecting multiple scattering effects.[1,2] The concept we use to determine the effective elastic constants is that of impedance matching. We choose the imbedding medium (type-*) so that the net scattering (as estimated above) from all the spheres vanishes identically. In other words, we imbed the composite sphere in a medium whose parameters we are free to vary. Then we adjust those parameters until the lowest order scattering vanishes. When the impedances are matched, the inclusion becomes transparent to the incident plane wave. The resulting estimate is still not exact because multiple scattering effects have been neglected. Nevertheless, we expect the self-consistent estimate to be superior to that of Kuster and Toksöz for large concentrations of inclusion material. The complete justification of this approach follows well-

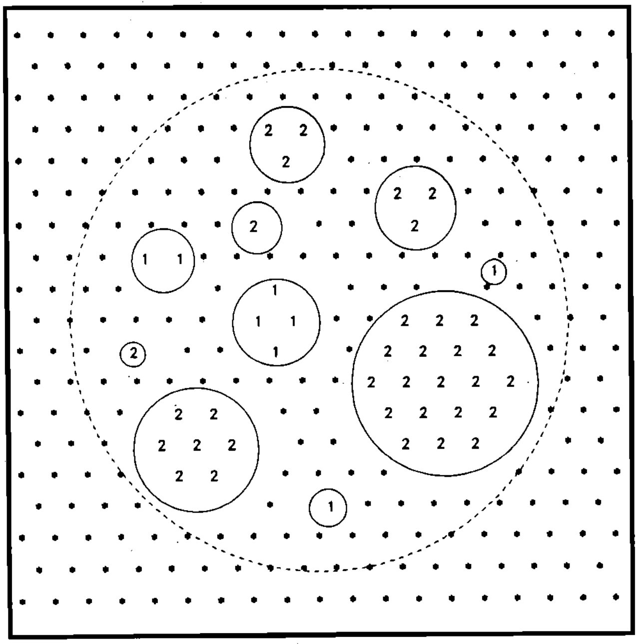

FIG. 2. Schematic diagram of the model composite used in the self-consistent theory to derive Eq. (25). The true composite is replaced by a large sphere (dashed line) containing spherical inclusions of both type-1 and type-2 material in the proper relative proportions. Matrix is composed of the effective material (type-*) to be determined.

known lines of argument[1,17] and will be presented elsewhere.[2]

The arguments of the preceding paragraph translate easily into formulas for the effective elastic constants. The formulas corresponding to (10) are therefore

$$\sum_{i=1}^{n} (u_r^s)_i = 0 , \quad \sum_{i=1}^{n} (u_t^s)_i = 0 , \tag{24}$$

and (11) is replaced by

$$0 = \sum_{i=1}^{n} a_i^3 \exp(-ik\mathbf{n}\cdot\boldsymbol{\xi}_i)[B_0(K^*, K_i, \mu^*) - B_1(\rho^*, \rho_i)\cos\theta - B_2(\mu^*, \mu_i, K^*)(3\cos 2\theta + 1)/4] . \tag{25}$$

Replacing the phase factor by unity and choosing

$$c_i = a_i^3 \Big/ \sum_{j=1}^{n} a_j^3 , \tag{26}$$

we find that (12) is replaced by

$$\sum_{i=1}^{n} c_i B_0(K^*, K_i, \mu^*) = 0 , \quad \sum_{i=1}^{n} c_i B_1(\rho^*, \rho_i) = 0 , \quad \sum_{i=1}^{n} c_i B_2(\mu^*, \mu_i, K^*) = 0 . \tag{27}$$

After substituting for the B's in (27), the resulting equations may be rewritten in the canonical forms

$$\frac{1}{K^* + \frac{4}{3}\mu^*} = \sum_{i=1}^{n} \frac{c_i}{K_i + \frac{4}{3}\mu^*} , \tag{28}$$

$$\rho^* = \sum_{i=1}^{n} c_i \rho_i , \tag{29}$$

and two possible formulas for μ^* follow from (27)

$$\frac{1}{\mu^* + F^*} = \sum_{i=1}^{n} \frac{c_i}{\mu_i + F^*} , \tag{30a}$$

or

$$\mu^* = 0 . \tag{30b}$$

The nonuniqueness of the formula for μ^* is a new feature of the self-consistent theory obtained through our derivation which was not present in the formulations of Hill[6] and Budiansky.[7]

Observe first that (28)–(30) are symmetric under interchange of constituent labels. This fact follows immediately from the general functional dependence exhibited in (27). Second observe that, whereas (19) and (21) provide explicit formulas for the Kuster–Toksöz estimates of K^* and μ^*, Eqs. (28) and (30) are implicit formulas for the self-consistent estimates of K^* and μ^* which must be solved by an iterative process.

Next observe that, if (7)–(9) are substituted into (24), we find for the inviscid fluid case that

$$\frac{1}{K^*} = \sum_{i=1}^{n} \frac{c_i}{K_i} , \tag{31}$$

$$\frac{1}{\rho^* + \frac{1}{2}\rho^*} = \sum_{i=1}^{n} \frac{c_i}{\rho_i + \frac{1}{2}\rho^*} , \tag{32}$$

and by assumption $\mu^* = 0$ [cf. (22) and (23)]. Equations (31) and (32) provide the self-consistent estimates of the elastic moduli and density for a fluid suspension. Note that (31) is identical to (22) which in turn is identical to the lower Hashin–Shtrikman bound for this case (see Sec. II). Thus, in our numerical examples (Sec. III), we will present only one curve on the figures labeled "SELF-CONSISTENT" and two on those labeled "KUSTER-TOKSÖZ." The lower curve on these plots is always valid both for the Kuster–Toksöz and for the self-consistent theory. It should be emphasized that this equivalence of the two theories occurs only when the matrix of the true composite is an inviscid fluid.

Note that if none of the shear moduli vanish, the solution $\mu^* = 0$ in (30b) is spurious. If some shear modulus vanishes and we make the choice $\mu^* = 0$ in (30b), then (28) reduces to (31). Also note that, if any $\mu_i = 0$ in (30a), then one solution (more than one solution exists) of (30a) is $\mu^* = 0$. If not all μ_is vanish, then at least two solutions of (30a) exist and at least one of those solutions is $\mu^* = 0$. More discussion of special cases with one $\mu_i = 0$ is given in Appendix A.

The fact that inviscid fluids must receive special treatment is discussed in Part II of this series. Here we wish to emphasize that the results for the effective density of fluid suspensions quoted in (9), (23), and (32) are valid when the fluid matrix is inviscid and the flow around the spherical inclusions is potential flow. The density (32) resulting from the induced-mass effect is also pertinent in special circumstances at high frequencies as is shown in Appendix B. In all other cases, the self-consistent formulas (28)–(30a) apply.

II. HASHIN-SHTRIKMAN BOUNDS

Using a new variational principle Hashin and Shtrikman[5,3] derived rigorous upper and lower bounds on the effective elastic moduli. These bounds are the best bounds known which depend only on the constituent moduli and volume concentrations. The bounds for K^* coincide if all bulk moduli are equal—independent of the values of the shear moduli; the bounds for μ^* coincide if and only if all shear moduli are equal. If all the shear moduli are equal, $K^+=K^-=K^*$ and the formulas agree with Hill's exact result for this case.[18]

A. Kuster-Toksöz moduli

It turns out that the Kuster–Toksöz estimates for real effective moduli in the presence of spherical inclusions are identical to the Hashin–Shtrikman bounds. Since this fact has been noted previously,[19] we will not repeat the proof here.

This fact has two important consequences:

(1) First, this equivalence implies that the Kuster–Toksöz estimates may not be particularly good estimates for large concentrations of inclusions. Of course, this equivalence also guarantees that in the special cases[18] when the Hashin–Shtrikman bounds are exact so are the Kuster–Toksöz estimates. But in more general circumstances, we expect the self-consistent estimates to be preferred since they always lie between the Hashin–Shtrikman bounds (see Sec. II B).

(2) The second consequence is that the approach of Kuster and Toksöz provides a new derivation of the Hashin–Shtrikman bounds. This derivation is in fact closely related to the results of Hashin[20] who shows that for two-phase media the bounds on K^* are exact results for a "composite spheres assemblage" where an aggregate is filled with nonoverlapping composite spheres of varying diameter. Each composite sphere consists of an inner sphere of type-1 whose radius is a_i and a concentric shell of type-2 whose outer radius is b_i such that the total volume concentration of type-1 material is $c_1=(a_i/b_i)^3$ as desired. The relationship between the Hashin–Shtrikman bounds on μ^* for the "composite spheres assemblage" and other bounds on μ^* obtained in Ref. 20 is discussed elsewhere by Hashin.[21] We note that Hashin was not able to determine the exact μ^* for the "composite spheres assemblage" using his method. Christensen and Lo[22] have recently studied this problem using a self-consistent imbedding method.

B. Self-consistent moduli

Hill[6] has shown that the self-consistent shear moduli for two-phase composites with spherical inclusions always satisfy the Hashin–Shtrikman bounds, i.e., $\mu^-\leq\mu^*\leq\mu^+$, whenever $(\mu_1-\mu_2)(K_1-K_2)\geq 0$. Thus, the self-consistent moduli have the desirable property that they are always consistent with these known rigorous results.

We will now prove that the self-consistent formulas are contained between the Hashin–Shtrikman bounds for real n-phase composites. Our proof is of necessity quite different from the one given by Hill. Consider the following expressions for the HS bounds obtained easily from (19), (21), and the equivalence of the Kuster–Toksöz estimates:

$$\frac{1}{K^+_{HS}+\frac{4}{3}\mu_n}=\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu_n}, \tag{33}$$

$$\frac{1}{K^-_{HS}+\frac{4}{3}\mu_1}=\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu_1}, \tag{34}$$

$$\frac{1}{\mu^+_{HS}+F_n}=\sum_{i=1}^{n}\frac{c_i}{\mu_i+F_n}, \tag{35}$$

and

$$\frac{1}{\mu^-_{HS}+F_1}=\sum_{i=1}^{n}\frac{c_i}{\mu_i+F_1}, \tag{36}$$

assuming $K_1\leq K_i\leq K_n$ and $\mu_1\leq\mu_i\leq\mu_n$. Equations (33)–(36) should be compared to (28) and (30a) for the self-consistent moduli. We take it for granted that solutions of (28) and (30a) exist and that the solutions of interest satisfy $K_1\leq K^*\leq K_n$ and $\mu_1\leq\mu^*\leq\mu_n$.

Now it will prove convenient to define two auxiliary functionals $K(\mu)$ and $\mu(F)$ given by

$$K(\mu)=\left[\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu}\right]^{-1}-\frac{4}{3}\mu \tag{37}$$

and

$$\mu(F)=\left[\sum_{i=1}^{n}\frac{c_i}{\mu_i+F}\right]^{-1}-F. \tag{38}$$

Then we find easily that

$$\frac{dK}{d\mu}=\frac{4}{3}\left[\sum_{i=1}^{n}\frac{c_i}{(K_i+\frac{4}{3}\mu)^2}-\left(\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu}\right)^2\right]\times\left[\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu}\right]^{-2}\geq 0, \tag{39}$$

and

$$\frac{d\mu}{dF}=\left[\sum_{i=1}^{n}\frac{c_i}{(\mu_i+F)^2}-\left(\sum_{i=1}^{n}\frac{c_i}{\mu_i+F}\right)^2\right]\left[\sum_{i=1}^{n}\frac{c_i}{\mu_i+F}\right]^{-2}\geq 0. \tag{40}$$

The inequalities in (39) and (40) follow easily upon applying the Schwarz inequality for sums to the numerators in brackets. The equality in (39) occurs if and only if $K_1=K_i=K_n$ and similarly in (40) if and only if $\mu_1=\mu_i=\mu_n$. Thus, except for these trivial cases, $K(\mu)$ and $\mu(F)$ are strictly increasing functions of their arguments. In particular, if $\mu_1\leq\mu^*\leq\mu_n$ and $F_1\leq F^*\leq F_n$,

$$K^-_{HS}=K(\mu_1)\leq K^*=K(\mu^*)\leq K^+_{HS}=K(\mu_n), \tag{41}$$

and

$$\mu^-_{HS}=\mu(F_1)\leq\mu^*=\mu(F^*)\leq\mu^+_{HS}=\mu(F_n). \tag{42}$$

To complete the proof, we must show that the quantity

$$F(\mu)=(\mu/6)\{[9K(\mu)+8\mu]/[K(\mu)+2\mu]\}, \tag{43}$$

is also a monotonic function of its argument. We find easily that

$$6\frac{dF}{d\mu}=\left(9K^2+16\mu K+16\mu^2+10\mu^2\frac{dK}{d\mu}\right)(K+2\mu)^{-2}\geq 0, \tag{44}$$

where the inequality in (44) follows from (39) and the requirements for dynamically stable materials that $K\geq 0$ and $\mu\geq 0$. This completes our proof that the self-consistent moduli for n-phase composites always lie between the Hashin–Shtrikman bounds, but never outside the bounds.

To conclude this section, we should also remark that the self-consistent elastic moduli agree with all the exact results which are known. For example, Hill's result[18] that

$$\frac{1}{K^*+\frac{4}{3}\mu^*}=\sum_{i=1}^{n}\frac{c_i}{K_i+\frac{4}{3}\mu^*} \tag{45}$$

when $\mu^*=\mu_1=\mu_i=\mu_n$ follows immediately from (28) and (30a). If all bulk moduli are equal, $K^*=K_1$ independent of the values of the μ_i's and μ^* as it should. For two-phase media, the theory always gives the proper values and slopes for K^* and μ^* as a function of concentration at the extreme points $c_1=0$ and $c_1=1$.

III. EXAMPLES

This section of the paper is devoted to various examples illustrating the practical application of the theory for two-phase composites. Although the ideal case with no intrinsic absorption is of great theoretical interest, the case when the composite's constituents possess intrinsic absorption is of more practical importance. For the sake of brevity, the discussion will be limited to this case.

Any real elastic material will possess some intrinsic absorption for waves of finite wavelength. Various mechanisms for intrinsic absorption have been discussed by Kuster and Toksöz[23] and more recently by Johnston, Toksöz, and Timur.[24] In this paper, we treat the intrinsic absorption as a phenomenologically determined parameter. Such intrinsic absorption leads to complex elastic moduli of the form

$$K=K_R(1+i\tan\delta_K)\,,\quad \mu=\mu_R(1+i\tan\delta_\mu) \tag{46}$$

where $\tan\delta$ is the loss tangent and K_R, μ_R are the real parts of the bulk and shear moduli. The loss tangent may be determined experimentally with laboratory samples or, in the case of ocean sediments, phenomenologically. With this generalization of the elastic moduli, it is straightforward to apply the self-consistent method and the Kuster–Toksöz method. The calculations require complex arithmetic. However, this modification is trivial.

Figures 3–6 are examples of the elastic moduli, velocities, and attenuation factors (Q^{-1}) computed using the theoretical results of the previous sections and the definitions

$$k=\frac{\omega}{v_c}\left(1+\frac{i}{2Q_c}\right)=\omega\left(\frac{\rho^*}{K^*+\frac{4}{3}\mu^*}\right)^{1/2}. \tag{47}$$

and

$$s=\frac{\omega}{v_s}\left(1+\frac{i}{2Q_s}\right)=\omega\left(\frac{\rho^*}{\mu^*}\right)^{1/2}. \tag{48}$$

The choices of parameters were made following Kuster and Toksöz.[14] Our type-1 constituent is a solid (sandstone or quartz-rich crystalline rock) with $\rho_1=2.70$ g/cc, $K_1=0.44$ Mb, $\mu_1=0.37$ Mb, and $\tan\delta_K=0.004$. Our type-2 constituent is water with $\rho_2=1.00$ g/cc, $K_2=0.022$ Mb, $\mu_2=i\omega\eta/\rho_2$, where $\omega=2\pi\times10$ Hz and η

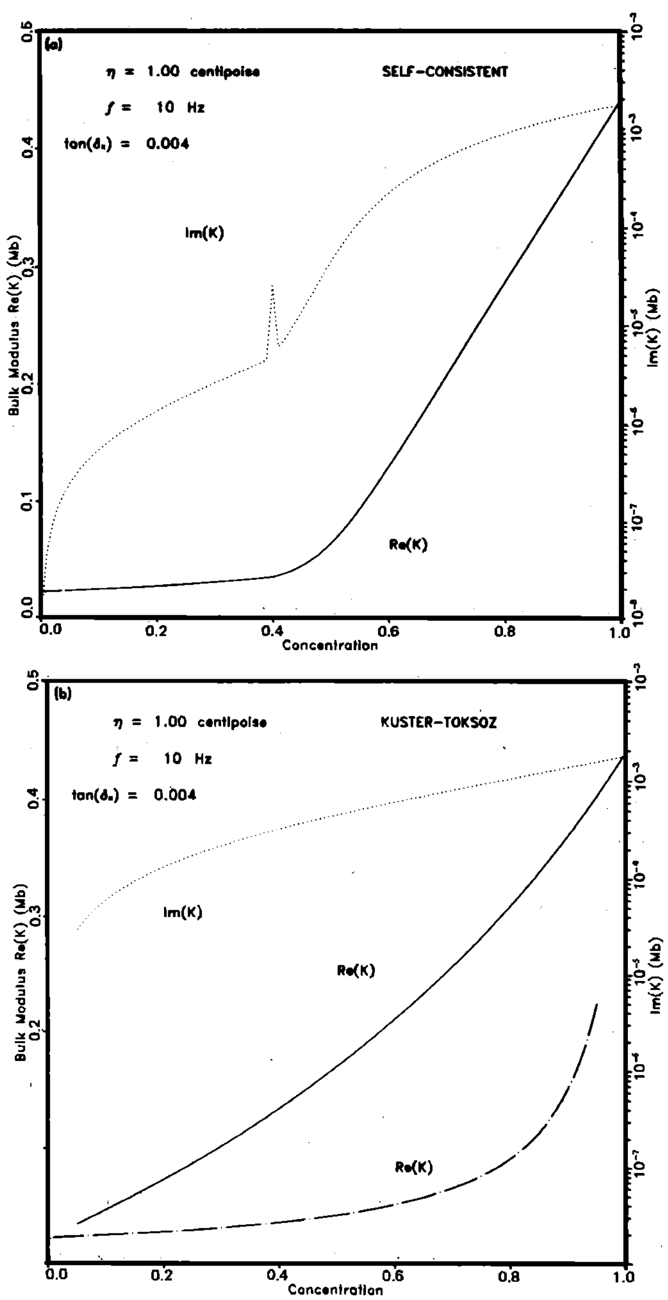

FIG. 3. Real and imaginary parts of the bulk modulus as a function of concentration computed using (a) the self-consistent theory and (b) the Kuster–Toksöz theory. The constituents have moduli $K_1=0.44\times(1+i0.004)$Mb, $\mu_1=0.37$Mb, $K_2=0.022$Mb, and $\mu_2=i\omega\eta/\rho_2=i6.28\times10^{-9}$ Mb.

$=1.00$ centipoise. The figures are plotted as the concentration of solid varies from $c_1=0$ (pure water) to $c_1=1$ (pure rock).

Figures 3 and 4 compare the bulk (Fig. 3) and shear (Fig. 4) moduli obtained with the self-consistent [Figs. 3(a) and 4(a)] and Kuster–Toksöz [Figs. 3(b) and 4(b)] estimates. The computations were done on Bell Labs' IBM 370 in double precision. Computations were made at 101 equally spaced points along the abscissa including the end points. No iteration is required for the Kuster–Toksöz estimates. The self-consistent estimates must be obtained by iteration. Typically ten iterations or less are required to attain successive iterates which

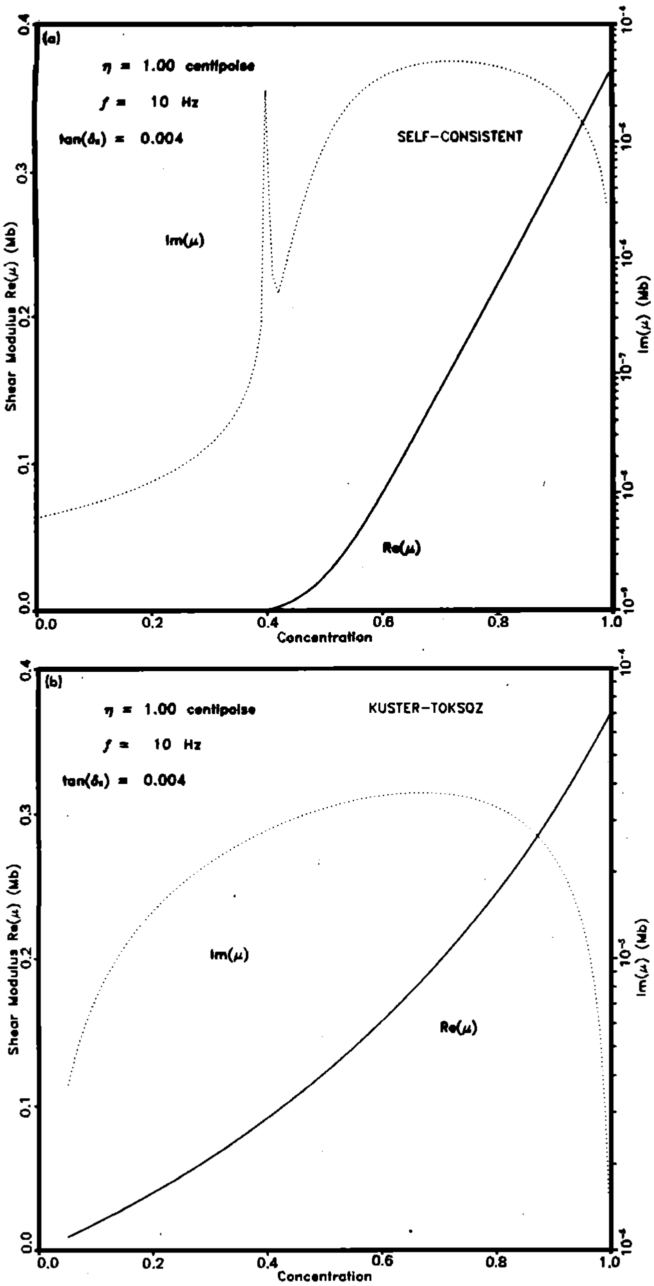

FIG. 4. Real and imaginary parts of the shear modulus as a function of concentration computed using (a) the self-consistent theory and (b) the Kuster–Toksöz theory. Same constituents as Fig. 3.

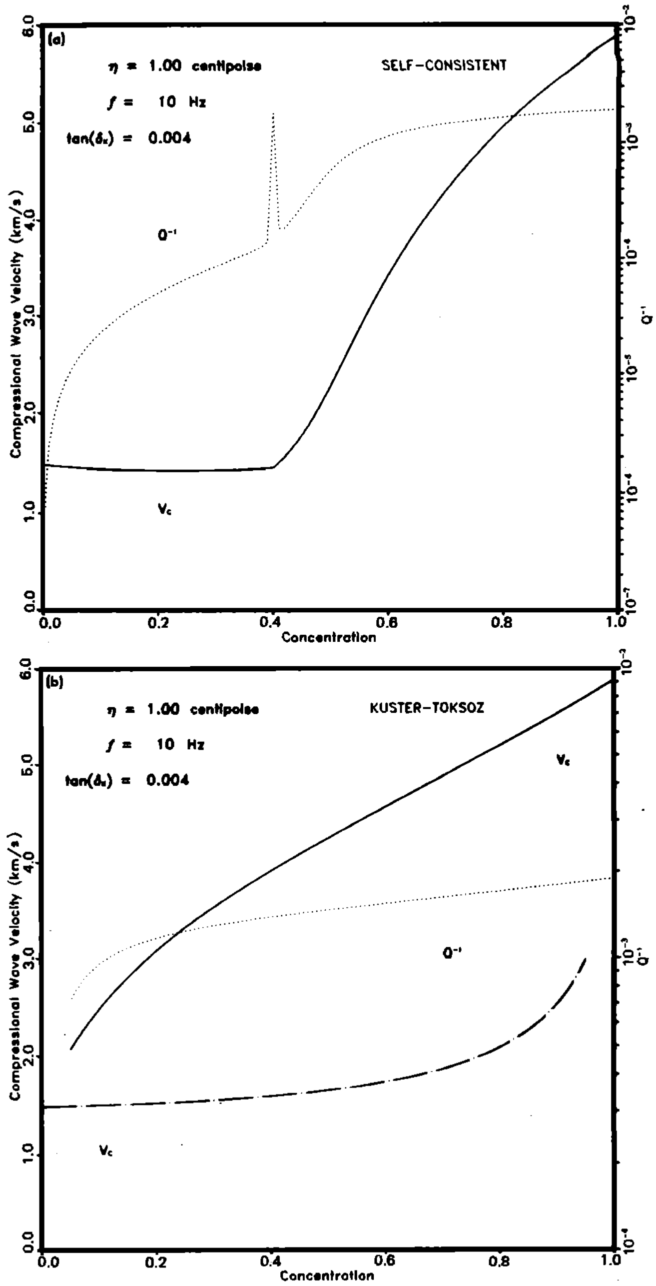

FIG. 5. Velocity and attenuation factor (Q^{-1}) for compressional waves as a function of concentration computed using (a) the self-consistent theory and (b) the Kuster–Toksöz theory. Same constituents as Fig. 3 with $\rho_1 = 2.70$ g/cc and $\rho_2 = 1.00$ g/cc.

satisfy the accuracy criterion that the sum of the absolute values of the relative errors for bulk and shear moduli is less than 2×10^{-6}. As the concentration approaches $c_1 \to 0.4$ (the singular point, see Appendix A), the number of iterations required for convergence rises gradually to 200 or more until $c_1 = 0.4$ at which point the iteration process does not converge (if one constituent is a fluid). At the singular point, we substitute the analytical result given in Appendix A and then proceed with the iteration process at the next concentration grid point. As we see in Figs. 3 and 4, the results obtained with this procedure are highly satisfactory.

In Figs. 3(a) and 4(a), we see that the self-consistent results for the bulk and shear moduli start at the fluid values and remain close to the fluid values until c_1 reaches the singular point. As c_1 increases beyond $c_1 = 0.4$, the moduli increase almost linearly towards values for the pure solid. The most distinctive feature of both plots is the peak in the imaginary part at $c_1 = 0.4$ which, of course, characterizes the singular point.

The two estimates of K^* in Fig. 3(b) were obtained using the alternate Kuster–Toksöz estimates. The lower estimate assumes the matrix of the composite remains a fluid for all concentrations, indeed the second estimate of $\mathrm{Re}(\mu^*)$ not shown in Fig. 4(b) is $\mathrm{Re}(\mu^*) = 0$. (Recall that this estimate is also a valid alternate solution of the self-consistency equations.) The upper estimate assumes a solid matrix for all concentrations, in-

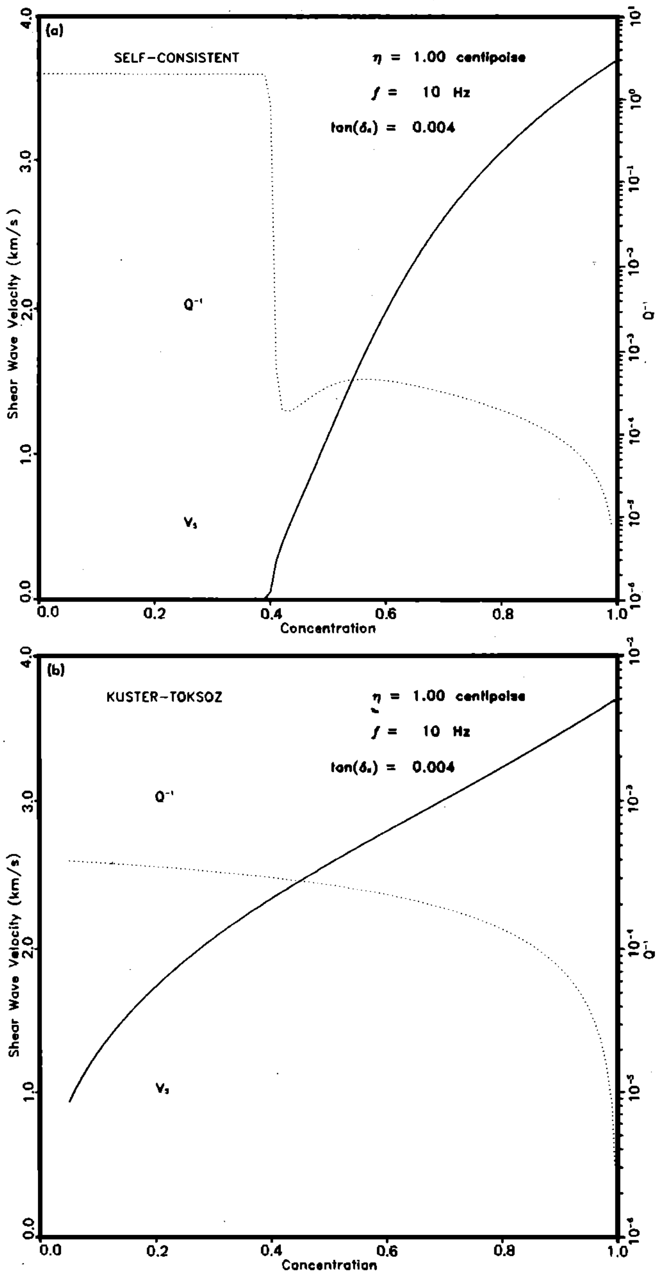

FIG. 6. Same as Fig. 5 for shear waves.

deed the estimate of $\mathrm{Re}(\mu^*)$ shown in Fig. 4(b) is nonzero for all concentrations $c_1 > 0$. By contrast, the estimates in Figs. 3(a) and 4(a) show the composite is a fluid $[\mathrm{Re}(\mu^*) = 0]$ for $c_1 < 0.4$ and a solid $[\mathrm{Re}(\mu^*) \neq 0]$ for $c_1 > 0.4$. No transition is possible for the Kuster–Toksöz estimates. Note that, although K^* cannot obtain an imaginary part for the Kuster–Toksöz estimates in the absence of intrinsic absorption, a finite value persists for all concentrations in Fig. 3(b) in the presence of intrinsic absorption.

Figures 5 and 6 plot the velocities and attenuations derived from Figs. 3 and 4 using the definitions in Eqs. (47) and (48). For the self-consistent estimates in Figs. 5(a) and 6(a), the ρ^* of (29) is used for the entire curve. The plateau in Q^{-1} for $c_1 < 0.4$ in Fig. 6(a) is easily explained. When $\mu^* = i\mu_I$ is pure imaginary, the definition of Q_s shows that

$$Q_s^{-1} = 2\left[\mathrm{Im}\left(\frac{1}{\mu^*}\right)^{1/2} \Big/ \mathrm{Re}\left(\frac{1}{\mu^*}\right)^{1/2}\right] = 2\,. \qquad (49)$$

Thus, when the self-consistent composite material is a viscous fluid $[\mathrm{Re}(\mu^*) = 0]$, the self-consistent estimate gives $Q_s^{-1} = 2$. This result agrees with the value plotted in Fig. 6(a).

For the Kuster–Toksöz estimates in Figs. 5(b) and 6(b), the ρ^* of (14) is used for the upper (solid) curve and ρ^* of (18) is used for the lower (fluid) curve. The attenuation factor Q^{-1} appearing in Fig. 5(b) is associated with the upper curve; the lower curve has no attenuation ($Q = \infty$).

IV. DISCUSSION AND CONCLUSIONS

In previous sections, we have derived the self-consistent formulas for the elastic constants of composite media, compared these formulas to the Hashin–Shtrikman bounds, and demonstrated that the formulas can be solved to yield useful results. In this section, we will consider the range of applicability of the method.

As the frequency increases and the wavelength of the incident plane-wave becomes comparable to the size of the inclusions, the theories described in this paper become inapplicable. Multiple scattering effects become important when $ka = 0(1) = sa$, so a different approach is required.

The self-consistent method was derived under the assumption that the wavevectors k and s satisfy $ka \ll 1$ and $sa \ll 1$ where a is the dimension of a typical inclusion. In fact, our formulas (28) and (30a) do not depend explicitly on the length a. Rather the values of k and s we compute from (47) and (48) fix the relevant length scales and determine whether or not the theory is valid in a particular application. For example, consider Fig. 5(a). At $c_1 = 0.8$, we find $v_c \cong 5.0$ km/s and $Q^{-1} \cong 2 \times 10^{-3}$. Thus, $|k|^{-1} \cong 80$ m at 10 Hz or 0.8 m at 1 kHz while a typical e-folding distance is of order $2Q|k|^{-1} \cong 40$ km at 10 Hz or 0.4 km at 1 kHz. Therefore, we expect the theory to be quite reliable for low frequencies if a typical spherical inclusion in the composite has diameter $a \ll 0.8$ m. In addition, there would be little attenuation over most scales of experimental interest. For extreme contrast, consider Fig. 6(a). At $c_1 = 0.4$, we find $v_s \cong 0.05$ km/s and $Q^{-1} = 0.85$. Thus $|s|^{-1} \cong 80$ cm at 10 Hz or 8 mm at 1 kHz while a typical e-folding distance is of order $2Q|s|^{-1} \cong 2$ m at 10 Hz or 2 cm at 1 kHz. We see that the theory is much less reliable in this range of frequencies at $c_1 = 0.4$ than it is at $c_1 = 0.8$. We conclude that the self-consistent theory should be reliable for frequencies less than 1 kHz as long as the volume concentration of solid inclusions is not too close to the singular point $c_1 = 0.4$. To improve upon the self-consistent theory, we must include multiple scattering effects.[2]

Results of our self-consistent theory should be compared to experimental findings. (A number of favorable theoretical comparisons may be found in the review by Watt, Davies, and O'Connell.[3]) In their experimental paper, Kuster and Toksöz[23] compare six theoretical models to their experimental results on spherical inclusions in a viscous fluid (say water) matrix. They found that their own theory and the Reuss average[3] (for this particular case the Reuss average is identical to the Kuster–Toksöz estimate) were in best agreement with the experimental results. Because the matrix and the composite were fluids, it turns out that the appropriate self-consistent result to compare is $\mu^* = 0$ (30b). But as has been explained previously, the Kuster–Toksöz estimate and the self-consistent estimate are identical in this case. Kuster and Toksöz use the density formula (18) [or (23)] when calculating the compressional wave speed. This choice is appropriate in the experimental range of frequencies (~500 kHz) because the parameter $\zeta^{-1} \approx 5 \times 10^{-3}$ with $a \approx 0.14$ mm [see (B2)]. However, since the inclusions are nearly neutrally buoyant in all cases studied, the distinction between Eqs. (23) and (20) or between Eq. (32) and (29) is negligible (less than one part in 10^5). Thus, with the use of any of these density estimates, the good agreement between theory and experiment found by Kuster–Toksöz for velocity estimates in fluid suspensions also applies to our self-consistent theory.

We conclude that the self-consistent theory provides useful estimates of the elastic properties of composite media. The resulting estimates for the bulk and shear moduli always satisfy the rigorous Hashin–Shtrikman bounds and reduce to the exact results in those cases where exact results are known. The estimates are unique unless one of the constituents has vanishing rigidity. The estimate for the shear modulus possesses a threshold of rigidity (at $c = 0.4$) when one of the constituents is a fluid (in agreement with physical expectations) but the precise location of this threshold is probably in error. The theory can also be used to predict effective elastic constants for complex composites containing constituents with phenomenologically determined intrinsic absorption.

V. CRITICISM OF EFFECTIVE MEDIUM THEORIES

Although the theory of composites presented here and in the following paper (Part II) differs in some details from the effective medium theories which preceded it, the theory nevertheless shares many of the characteristics of the earlier theories. Since these earlier theories have been discussed and criticized by various authors during the last 15 years, the present author feels obligated to provide answers here to some of the more recurrent criticisms of these theories.

Hashin[21] has argued that, since mathematically rigorous bounding methods are available for the effective elastic constants of composites and since the effective medium theories are inherently nonrigorous and approximate, no certain knowledge is to be gained by using such methods. Hashin concludes that effective medium theories should therefore be avoided whenever possible in preference to the rigorous bounding methods. It is difficult to refute an argument such as this when the conclusion is inescapable given the premise; however, we may criticize the premise. Since the known rigorous bounds are often far from each other in cases of most interest, it is certainly desirable to have additional theoretical estimates of the effective elastic constants for comparison with experiment. If the theoretical estimate does not agree with experiment, then we have an additional constraint on theories of elastic composites. By studying the implications of such constraints, we can hope to learn something more about the physics of composites. If we limit our research to those matters about which we can hope to gain certain knowledge, we limit ourselves to questions of mathematics only and unnecessarily exclude physics.

Many critics[19,25,26] believe that a major (or even fatal) flaw of the self-consistent theory is its prediction of a threshold of rigidity at a finite concentration of solid material (e.g., $c = 0.4$) in a fluid. In the case of cracks in a solid matrix, O'Connell and Budiansky[27] and Bruner[25] have argued the pros and cons respectively of an effective medium theory. Many of these arguments are equally applicable to the present theory. We will not repeat these arguments but merely state that the present author finds the arguments of O'Connell and Budiansky[27] to be more convincing.

For similar reasons, Christensen[26] takes a strong stand against the self-consistent theory for multiphase media. Basing his argument on the "very strange behavior" of the self-consistent theory in extreme examples with either voids or rigid inclusions, he dismisses the self-consistent approach altogether as "merely a convenient and appealing operational scheme, which, unfortunately, gives erroneous results." But later, in his discussion of single phase media, Christensen admits that "in application to polycrystalline materials [the self-consistent method] appears to be a very reasonable approach." To this last comment, we would add that Part II of this series establishes the fact that our self-consistent scheme also gives very reasonable results for multiphase media when all constituents have finite shear and bulk moduli which do not differ by orders of magnitude. The method admittedly has some problems at present in those cases when the constituents' moduli do differ by orders of magnitude as in Christensen's examples. However, we believe the proper attitude towards these problems is first to be aware of them and then to seek methods of correcting the deficiencies—rather than just discarding the whole theory, the good along with the bad. Instead of assuming an attitude of hopelessness, we should assume that the theory is merely incomplete and strive to complete it in the future.

Finally, it has been pointed out by Toksöz and Cheng[19] (among others) that the experimental evidence of Walsh, Brace, and England[28] shows a nonzero bulk modulus for porous glass at porosities higher (up to 70% porosity) than the predicted location of vanishing bulk modulus at ~50% porosity for spherical voids in the symmetric self-consistent theory. The fact that the effective medium theory is not consistent with this experimental result is therefore quoted as a major failing of the theory. If this example were a valid application of the self-consistent theory, then it would indeed be a point against the theory. However, two clarifying comments should be made: (1) To the author's knowledge, the effective medium theories are the only ones that exhibit a finite threshold of rigidity. Yet, physical intuition strongly suggests that a threshold of rigidity *must exist* at some finite concentration when solid material is added to a fluid. Thus, the proponents of any theory which does not exhibit this threshold (i.e., all noneffective-medium theories) should admit that there is this point against their proposed theory. (2) For the case of the data on porous glass, we would argue that using the symmetric self-consistent theory for spherical inclusions is simply a misapplication of the theory. For low porosity, Walsh *et al.*,[28] note that the bubbles in the glass will tend to be spherically shaped so assuming spherical voids is appropriate in this case. For high porosity, Walsh *et al.*,[28] note that the bubbles need not be perfectly spherical. Furthermore, for high porosity, it is *clearly inappropriate* to represent the porous glass by a distribution of *a few spherical glass beads somehow suspended in a matrix of voids*: yet this is the model used by the critics when comparing the self-consistent theory to the experimental data. A sensible comparison between theory and experiment could be achieved by assuming instead that the glass frame consists of thin filaments of solid material and that these filaments may be represented approximately by small-aspect-ratio prolate spheroids or by needle-shaped inclusions in the void (see Part II). This approach has been used with considerable success to explain[29] the experimentally observed wave speeds in a water-saturated porous glass structure.[30]

In conclusion, it should be stressed that the present author does *not* claim that our new theory is the final word on elastic composites; the theory is obviously unsatisfactory in several situations and more work will be required to improve the theory in these applications. On the other hand, the author does claim that the theory has many appealing features and that no other contemporary theory, when considered *in toto*, has yet been conclusively demonstrated to be better on either theoretical or experimental grounds.

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APPENDIX A: SINGULAR POINT

When one of the phases of a two-phase composite is a fluid (e.g. $\mu_2=0$) or has shear modulus much less than the other constituent ($\mu_2/\mu_1 \ll 1$), a singular point arises at 40% concentration of the solid constituent ($c_1 = 0.4$). To show this, consider

$$\frac{1}{\mu^*+F^*} = \frac{c_1}{\mu_1+F^*} + \frac{c_2}{\mu_2+F^*}, \tag{A1}$$

and suppose $\mu^*/K^* \ll 1$. Then to lowest order in μ^*, we have

$$F^* = \frac{\mu^*}{6}\left(\frac{9K^*+8\mu^*}{K^*+2\mu^*}\right) \cong \frac{3}{2}\mu^*. \tag{A2}$$

For $\mu^* \approx \mu_2 \cong 0$, the first term on the rhs of (A1) may be neglected compared to the second. We find

$$\mu^* \cong \mu_2/(1-\tfrac{5}{2}c_1) \quad \text{for} \quad c_1 < 0.4. \tag{A3}$$

For $\mu_2/\mu^* \ll 1$, we have

$$\frac{1}{\mu^*+F^*} \cong \frac{c_1}{\mu_1+F^*} + \frac{c_2}{F^*}. \tag{A4}$$

Now note that, for all $0 \le \mu^*/K^* \le \infty$, $F^* = \alpha\mu^*$ where $\frac{2}{3} \le \alpha \le \frac{3}{2}$. Then, by substituting for F^* and rearranging terms, we find

$$\mu^* \cong [1-c_2(1+1/\alpha)]\mu_1 = (c_1 - c_2/\alpha)\mu_1. \tag{A5}$$

As $c_1 \to 0.4$, we know μ^*/K^* becomes small so $\alpha \to \frac{3}{2}$ and (A5) becomes (for c_1 close to 0.4)

$$\mu^* \cong (5c_1 - 2)\mu_1/3 \quad \text{for} \quad c_1 > 0.4. \tag{A6}$$

If $\mu_2 \equiv 0$, then (A3) and (A6) are consistent at $c_1 = 0.4$ and $\mu^* \to 0$ from both sides. However, if $\mu_2 \neq 0$ but $\mu_2/\mu_1 \ll 1$, then (A3) and (A6) are incompatible. In this case, we must use some other method to evaluate μ^* at $c_1 = 0.4$. (The iterative method does not converge.) Consider rearranging (A1) such that

$$F^* = \mu_1\mu_2[(\mu^*\langle 1/\mu\rangle - 1)/(\langle\mu\rangle - \mu^*)], \tag{A7}$$

where $\langle\mu\rangle = c_1\mu_1 + c_2\mu_2$ and $\langle 1/\mu\rangle = c_1/\mu_1 + c_2/\mu_2$. Since (A6) implies that μ^* is decreasing rapidly from the right and (A3) implies μ^* is increasing rapidly from the left, we expect μ^* to attain some intermediate value between μ_1 and μ_2 such that $\mu_2 \ll \mu^* \ll \mu_1$. Since neither the arithmetic mean ($\langle\mu\rangle \cong c_1\mu_1$) nor the harmonic mean ($\langle 1/\mu\rangle^{-1} \cong \mu_2/c_2$) is intermediate in this sense, we will try as an ansatz the geometric mean of the two, namely for $c_1 = \frac{2}{5}$

$$\mu^* \cong (\langle\mu\rangle\langle 1/\mu\rangle^{-1})^{1/2} \cong (\tfrac{2}{3}\mu_1\mu_2)^{1/2}. \tag{A8}$$

Substituting (A8) into (A7), we find

$$F^* \cong \mu_1\mu_2/\mu^* \cong \tfrac{3}{2}\mu^* \tag{A9}$$

which is consistent with $\mu^*/K^* \ll 1$. Thus, our ansatz (A8) provides a good estimate of the shear modulus at the singular point.

APPENDIX B: INDUCED-MASS EFFECT FOR VISCOUS FLUIDS

Landau and Lifshitz[16] calculate the drag on a sphere oscillating in a fluid. With the substitution $v = -\dot{v}/i\omega$, their result is

$$F = \tfrac{1}{2}\rho_f[1 + \tfrac{9}{2}\zeta^{-1} + i\tfrac{9}{2}\zeta^{-1}(1+\zeta^{-1})](4\pi a^3/3)\dot{v}, \tag{B1}$$

where v is the velocity of the sphere in the direction of oscillation, a is the radius of the sphere, ρ_f is the fluid density, ω is the angular frequency of oscillation, and

$$\zeta = (a^2\omega\rho_f/2\eta)^{1/2}. \tag{B2}$$

The additional induced-mass ρ' of the sphere due to drag is

$$\rho' = \tfrac{1}{2}\rho_f[1 + \tfrac{9}{2}\zeta^{-1} + i\tfrac{9}{2}\zeta^{-1}(1+\zeta^{-1})]. \tag{B3}$$

In the inviscid limit, $\eta \to 0$ and $\zeta^{-1} \to 0$ and $\rho' = \frac{1}{2}\rho_f$ as expected. When $\eta \neq 0$, (B3) again approaches $\rho' = \frac{1}{2}\rho_f$ if $\omega \to \infty$. Thus, the induced-mass $\rho' = \frac{1}{2}\rho_f$ is valid for an inviscid fluid or for viscous fluids at high frequencies of oscillation.

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