Extreme Damping in Composite Materials with a Negative Stiffness Phase

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Composites with negative stiffness inclusions in a viscoelastic matrix are shown to have higher stiffness and mechanical damping $\tan\delta$ than that of either constituent and exceeding conventional bounds. The causal mechanism is a greater deformation in and near the inclusions than the composite as a whole. Though a block of negative stiffness is unstable, negative stiffness inclusions in a composite can be stabilized by the surrounding matrix. Such inclusions may be made from single domains of ferroelastic material below its phase transition temperature or from prebuckled lumped elements.

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Properties of a composite material depend upon the shape and properties of the heterogeneities, upon the volume fraction occupied by them, and upon the interface between the constituents [1,2]. Bounds for behavior of a composite of any structure have been developed assuming positive stiffness. For example, the composite stiffness cannot exceed the Voigt form $G_{c,u} = G_1V_1 + G_2V_2$, where G_1 and V_1, and G_2 and V_2, are the shear modulus and volume fraction of phases 1 and 2, respectively. The Hashin-Shtrikman bounding formulas [1] apply for isotropic composites; the lower bound for the shear modulus G_L of an *elastic* composite is

$$G_L = G_2 + \frac{V_1}{1/(G_1 - G_2) + [6(K_2 + 2G_2)V_2]/[5(3K_2 + 4G_2)G_2]}, \quad (1)$$

in which K_1, K_2, G_1, G_2, V_1, and V_2 are the bulk modulus, shear modulus, and volume fraction of phases 1 and 2, respectively. Positive stiffness values were tacitly assumed for this and for other bounds [3]. One attains the upper and lower Hashin-Shtrikman formulas for bulk modulus with a morphology in which the composite is filled with coated spheres of different size but an identical ratio of sphere size to coating thickness. The attainment is *exact* for the bulk modulus [4] and approximate for the shear modulus. The shear modulus formula is attained *exactly* by hierarchical laminates [5].

Dynamic viscoelastic properties are expressed in terms of the complex dynamic Young's modulus $E^* = E' + iE'' = E'(1 + i\tan\delta)$, with $E' = \mathrm{Re}\{E^*\}$ and $\tan\delta \equiv \mathrm{Im}\{E^*\}/\mathrm{Re}\{E^*\}$; δ is the phase angle between the stress and strain sinusoids. (The primes are conventional notation for the real and imaginary part, respectively, and do not represent derivatives.) The dynamic modulus is a function of frequency, and in composite materials it depends on constituent properties and morphology. A representative stiffness-loss map for several composites is shown in Fig. 1. The product $E'\tan\delta$ is a figure of merit [6] for the damping of structural vibration; however, most existing materials [7] exhibit maximum $E'\tan\delta < 0.6$ GPa. Some composites with higher values have been developed [8]. The purpose of this paper is to explore the consequences of negative stiffness constituents in achieving high damping in composites.

Lumped examples of negative stiffness include a column constrained in a buckled "S" shaped configuration [9]. If one presses laterally on the column, one can cause it to snap through. The negative stiffness condition is unstable. The column can be stabilized by a lateral constraint. Tubes following buckling offer decreasing force with an increase in deformation, hence, negative incremental stiffness [9]. Single cell tetrakaidecahedron models exhibit a compressive force-deformation relation that is not monotonic [10] under displacement control, hence, exhibit negative stiffness over a range of strain.

Distributed examples of negative stiffness include single domains of materials in the vicinity of certain phase

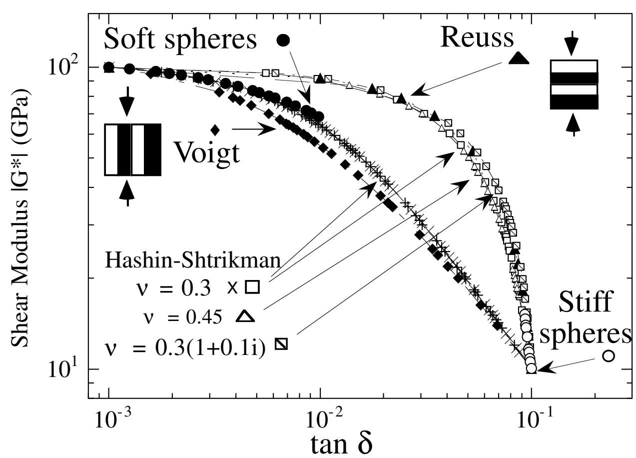

FIG. 1. Stiffness-loss map. Calculated behavior of several composites. Each point corresponds to a different volume fraction (adapted from Lakes [25]). One phase is stiff and low loss, and the other phase is more compliant and high loss.

transformations. Bulk ferroelastic materials [11] in the vicinity of phase transformations exhibit a minimum stiffness at a transition temperature, and are unstable and revert to a banded domain structure below that transition temperature. Single domains [12] are expected to exhibit negative stiffness since the free energy in the Landau theory has a relative maximum, corresponding to unstable equilibrium, below a critical temperature [13]. One does not ordinarily speak of negative stiffness of ferroelastics or ferroelectrics because, owing to the instability, it is not observed in bulk multidomain samples. Ferroelastic materials are crystalline and anisotropic; however, the physical principles articulated here may be extended to such cases. Negative stiffness is also known in string theory [14]; however, we do not expect to make composites from strings.

Negative stiffness is to be distinguished from negative Poisson's ratio. Poisson's ratio, represented by ν, is defined as the negative lateral strain of a stretched or compressed body divided by its longitudinal strain. Most materials, stretched under axial tensile force, elongate longitudinally but also contract *laterally,* hence, have a positive Poisson's ratio. Poisson's ratio is dimensionless, and for most solids its value ranges between 0.25 and 0.33; the range for stability of isotropic solids is from -1 to 0.5; within that range all moduli are positive. Recently, Lakes and co-workers have conceptualized, fabricated, and studied negative Poisson's ratio foams [15,16] with ν as small as -0.8. These materials become fatter in cross section when they are stretched. The Poisson's ratio and moduli are within the range for stability. Negative stiffness, by contrast, refers to a situation in which a reaction force occurs in the same direction as imposed deformation.

Negative stiffness entails a reversal of the usual directional relationship between force and displacement in deformed objects. In ordinary positive stiffness elastic materials (such as a spring), the reaction force exerted by the material is in the opposite direction as the deformation. This corresponds to a restoring force. A material of negative stiffness exerts a reaction force in the same direction as the deformation, which tends to help the deformation proceed further. This is accomplished by a positive stored energy at unstable equilibrium.

To study viscoelastic composites, the dynamic elastic-viscoelastic correspondence principle [17,18] is applied to Eq. (1) so that all elastic constants become complex quantities [19] in Eq. (1) giving the complex viscoelastic shear modulus G_L^* of the lower composite. The correspondence principle is based on the fact that elastic and viscoelastic problems share the same boundary conditions and laws of motion; only the constitutive relation is different. Its validity depends on linearity and on the assumption that boundary conditions do not change nature from displacement to stress control, with time. No assumption of sign of stiffness is required. The equation for G_L^* is valid pointwise at each frequency; there is no need to make any assumptions about the frequency dependence. Time domain behavior, if desired, is obtained by Fourier transformation. Real and imaginary parts are separated to numerically prepare stiffness-loss maps and plots of viscoelastic properties vs constituent stiffness and volume fraction. The Hashin-Shtrikman formulas no longer represent bounds in the viscoelastic case; however, they are in most cases close to the true bounds [20], but that is immaterial in the present context for the following reason. Since these formulas are exactly attained via known microstructures in the elastic case, they are also exactly attained in the viscoelastic case by virtue of the correspondence principle. In the following development, we exceed bounds derived assuming positive stiffness constituents, on both stiffness and damping.

Composite damping $\tan\delta$ achieves a maximum when the inclusion stiffness is negative and comparable in magnitude to the matrix stiffness, as shown in Fig. 2. The maximum $\tan\delta$ exceeds 1. This is a very large damping, ordinarily seen only in solid polymers in the glass-rubber transition, and is much larger than the assumed damping, 0.1 for the matrix and 0 for the inclusions, of either constituent in the composite. The product $G' \tan\delta$ is large at the peak, corresponding to regions in the upper right region in Fig. 1. Moreover, a dilute concentration of inclusions of negative stiffness also has a substantial effect on the stiffness. The effect is larger than if the inclusions were rigid (much stiffer than the matrix) or if they were voids. In comparison with the $\sim$40% effect on stiffness shown in Fig. 2, 2% void inclusions reduces the composite stiffness by 3.7%, while 2% perfectly rigid (infinitely stiff) inclusions increases the composite stiffness by 4.3%. If the Poisson's ratio of the matrix is increased, the peak in Fig. 2 shifts to more negative values of inclusion stiffness.

The plots of Fig. 2 are reminiscent of a resonance phenomenon. There are, however, no inertial terms: On the continuum scale, the representation, though time dependent, in quasistatic. In resonance, elastic terms can neutralize inertial terms at selected high frequencies; by contrast, in the present composites, elastic terms in the denominator can have opposite signs since one phase has a negative stiffness. This is illustrated in Eq. (2), in which the complex form of Eq. (1) is simplified by assuming a rubbery matrix, $K_2 \gg G_2$. As above, we allow $\mathrm{Re}(G_1) < 0$:

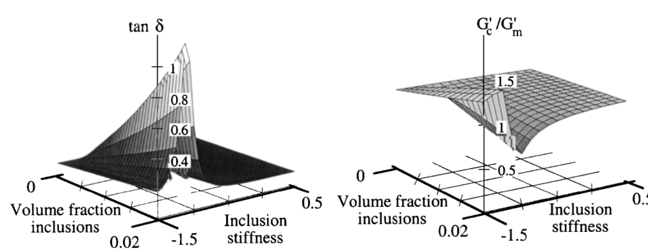

FIG. 2. Composite normalized stiffness G_c'/G_m' and mechanical damping $\tan\delta$ vs volume fraction and stiffness of elastic inclusions, normalized to matrix stiffness. Hashin-Shtrikman isotropic "lower" composite. Matrix damping, $\tan\delta = 0.1$; Poisson's ratio 0.3.

$$G_L^* = G_2^* + \frac{\frac{5}{3}V_1(G_1^* - G_2^*)}{1 + \frac{2}{3}V_1 + \frac{2}{3}(1 - V_1)\frac{G_1^*}{G_2^*}}. \quad (2)$$

As for stability, both the *composite* shear modulus G' and bulk modulus K' are positive over the ranges of variables considered; hence, the composite obeys the continuum stability criteria. However, negative composite stiffnesses, hence instability, may occur for certain combinations of sufficiently high inclusion concentration and sufficiently negative values of inclusion stiffness. As for local stability, the inclusions are stable [21] provided their stiffness is not too much less than $-G_m'$.

As matrix damping is reduced to 0.001, the composite damping peak becomes higher and narrower, as shown in Fig. 3. Even a minuscule concentration of inclusions in a low-damping matrix gives rise "homeopathic" effects: a substantive effect from a vanishingly small concentration of causal material. In physical systems, the concentration cannot tend to zero, owing to the nonzero size of atoms. The damping $\tan\delta = 0.001$ assumed in Fig. 3 is representative of structural metals; however, even smaller damping values are common in some alloys, e.g., aluminum alloy 6061, exhibits $\tan\delta \approx 3.6 \times 10^{-6}$ in torsion at room temperature [22]. As matrix damping tends to zero, behavior in the vicinity of $G_i' = -1.1G_m'$ becomes singular: Composite damping and stiffness become *unbounded*. So matrix damping has a stabilizing effect. The physical basis for the damping enhancement may be understood as follows. Consider a spherical elastic inclusion in an elastic matrix, under tension, after Goodier [23]. This solution contains no assumptions about constituent stiffness; here, we allow the inclusion stiffness to assume negative values. Such a sphere elongates the least for a stiff inclusion, more for an "inclusion" as stiff as the matrix, more for a cavity of zero stiffness, and yet more for an inclusion of negative stiffness G_i. The local deformation becomes unbounded as G_i, assumed real, tends to $-1.1G_m'$, as shown in Fig. 4. The value of G_i' for the transition becomes more negative as the matrix Poisson's ratio increases. For an inclusion which has a shear modulus which is negative, but much smaller in magnitude than that of the matrix, the inclusion is effectively under displacement constraint. As the inclusion stiffness approaches the matrix stiffness in magnitude, the deformation at its surface becomes much greater than the overall asymptotic deformation of the composite. Since local strain becomes large in an *elastic* composite of this type, the corresponding energy dissipation becomes large in a *viscoelastic* composite. Behavior for a viscoelastic inclusion, inferred via the correspondence principle, shows amelioration of the singularity seen in the *elastic* case, as shown in Fig. 4.

As for geometry dependence, Reuss composites for which stiffness is $1/G_c = V_1/G_1 + V_2/G_2$, as well as composites with a dilute concentration of spheres [2] for which stiffness is

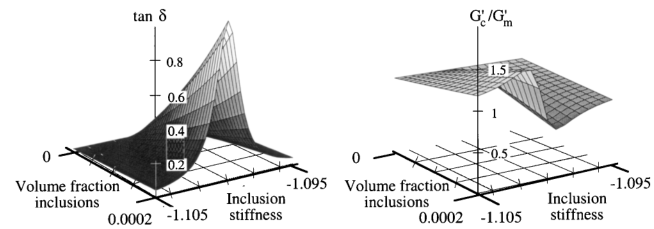

FIG. 3. Illustration of "homeopathic" effects of a minuscule concentration of inclusions in a low-damping matrix. Composite normalized stiffness G_c'/G_m' and mechanical damping $\tan\delta$ vs volume fraction and stiffness of inclusions, normalized to matrix stiffness. Hashin-Shtrikman isotropic "lower" composite. Matrix damping, $\tan\delta = 0.001$; Poisson's ratio 0.3.

$$G_{cmp} = G_2 + (G_1 - G_2) \times \frac{5(3B_2 + 4G_2)}{[9B_2 + 8G_2 + 6(B_2 + 2G_2)(G_1/G_2)]} V_1, \quad (3)$$

exhibit similar response to negative stiffness phases as the Hashin-Shtrikman composite. The predicted anomalies are therefore robust with respect to the details of the assumed geometry.

Viscoelastic composites with inclusions of negative stiffness may have several uses as follows. They may be used in studying properties of single domains of ferroelastic, ferroelectric, shape memory martensite, or ferromagnetic materials. A dilute concentration is sufficient to obtain substantial effects, particularly if the matrix chosen has a small mechanical damping. Effects can be seen with a minuscule amount of sample material. Some materials of interest cannot be easily prepared as large single crystals; polycrystalline arrays may be brittle.

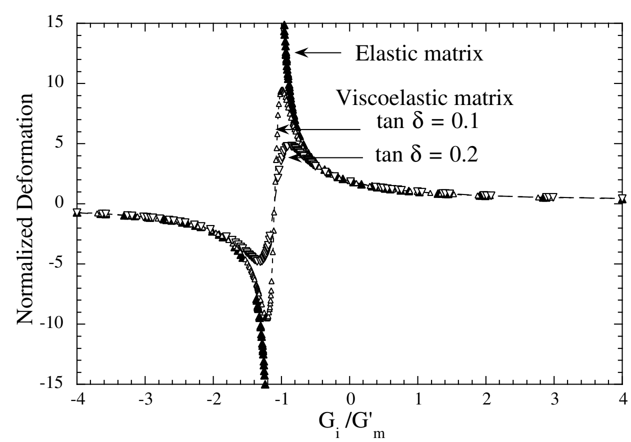

FIG. 4. Deformation at the surface of a spherical inclusion vs inclusion stiffness based on analytical solution of Goodier [23]. Solid symbols, elastic inclusion. Open symbols, viscoelastic inclusion via the correspondence principle. Deformation is normalized to deformation far from the inclusion. Inclusion stiffness is normalized to the matrix stiffness. Poisson's ratio of matrix is 0.3. Inclusions are assumed to have a bulk modulus 100 times that of the matrix.

Such composites also find applications in which high stiffness and damping is needed, as well as in high performance sensors and actuators based on thermoelastic or piezoelectric coupling. Inclusions need not be temperature sensitive ferroelastics. Prestressed or prebuckled elements may be used as inclusions. Indeed, recent experiments [24] have disclosed giant damping effects in two systems: a macroscopic system with buckled compliant tubes as negative stiffness elements, and a dilute particulate composite with ferroelastic inclusions just below the transition temperature as negative stiffness elements.

Composite materials of unbounded mechanical damping $\tan\delta$ are possible if the inclusion phase has negative stiffness. Development of high-damping composites built of conventional materials in novel geometrical arrangement could optimize many existing technologies in which stiffness and damping are both important.

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