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Stable singular or negative stiffness systems in the presence of energy flux

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We report stable systems which exhibit quasistatic stiffness that can be negative or tend to infinity without external constraint. They are based on coupled fields in the non-equilibrium presence of energy flux that is modulated by force. They evade thermodynamic restrictions by relaxing a restrictive assumption: equilibrium. Negative values of physical properties, including compressibility and heat capacity, are considered forbidden in classical thermodynamics; such analyses provide bounds on the stiffness and other properties of multiphase materials. Stable negative and singular stiffness is demonstrated experimentally in a piezoelectric system and in a thermoelastic granular material. Coupled fields occur naturally under a wide range of conditions and form the basis for many forms of technology including sensors, actuators, and electric coolers. Because all materials exhibit at least one coupled field effect, the concept is broadly general and is applicable to attaining extreme values of any physical property e.g. stiffness, permittivity, piezoelectricity.

Keywords: elasticity; creep; thermodynamics; piezoelectrics; thermal expansion; viscoelasticity; coupled fields; negative properties

1. Introduction

If one stretches a rubber band (or other elastic material), the force applied gives rise to a stretch or deformation. Deformation divided by force is the structural compliance, positive if the deformation is in the same direction as the applied force. Taking into account the cross-section area and original length, strain divided by stress gives the material compliance, the inverse of which is the stiffness or elastic modulus. When pulled, the rubber band not only stretches, it changes in temperature. *All* materials exhibit at least one coupled field effect [1], for example coupling between mechanical and thermal variables: thermal expansion relates strain to temperature and the piezocaloric effect relates stress to temperature change.

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Such coupled fields have many ramifications. For example, the difference between adiabatic and isothermal compressibility forms the basis of heat engines and refrigerators. Materials exhibiting coupled field phenomena (Figure 1a) also include piezoelectric solids in which electric field and displacement are coupled to stress and strain, piezomagnetic and magnetostrictive [2] solids which couple stress and strain to magnetic field, and thermoelectric [3] solids that couple temperature changes to electric field; also, in fluid-filled porous materials such as rock, deformation is coupled to fluid pressure and flow. The coupling exhibited by such materials forms the basis of sensors, actuators, and related devices; enhancement of such coupling is consequently pertinent [1,3–5]. Coupled fields are well known and are subject to current study: damping via thermoelasticity [6,7], piezoelecticity [8,9], quantum dots [10]. Here, we use coupled fields to obtain stable negative stiffness and stable stiffness tending to infinity. The general expression for coupled fields is given as follows, then specific experimental embodiments are presented.

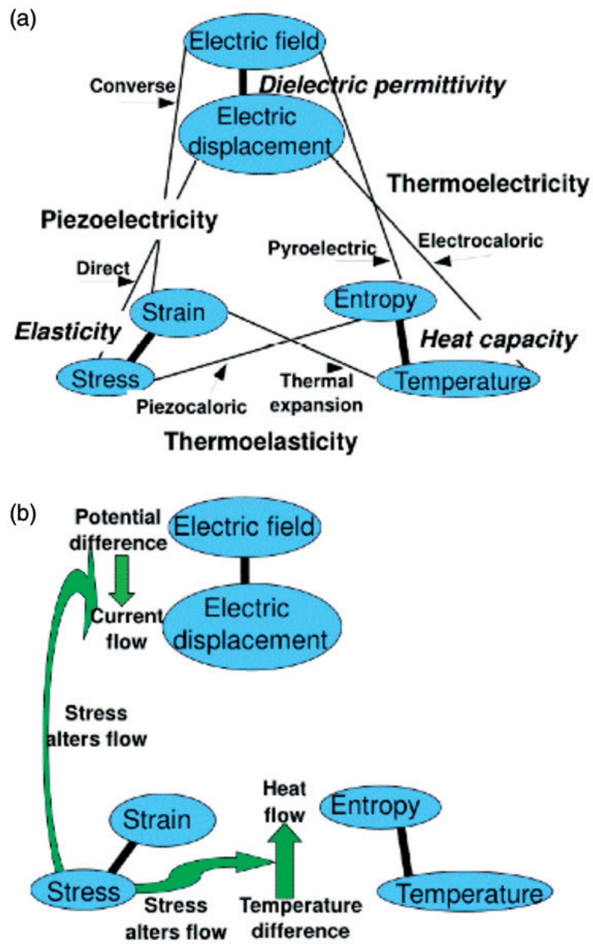

Figure 1. (a) Coupling paradigm adapted from Nye [1]. Thick lines represent primary cause-effect relations; thin lines represent coupling which depends on the material. (b) Stress-induced modulation (curved arrows) of flux to achieve extreme stiffness.

2. Coupled fields and extreme effective properties

Stress σ and strain ε are linked to coupled field variables [1] ξ and Ξ as follows, with J as material compliance for $\xi = 0$ and ψ, k, and K as constants:

$$\varepsilon = J\sigma + k\xi, \quad \Xi = K\sigma + \psi\xi \tag{1}$$

The field variables may be linked in a variety of ways. If ξ is temperature, then J is the isothermal compliance at long time; at time zero the adiabatic compliance differs. If ξ is electric field, J is the short-circuit compliance at long time; at time zero the open-circuit compliance differs. The difference between fast and slow response is due to stress-induced energy flux. A field variable that obeys a rate equation $\mathrm{d}\xi/\mathrm{d}t = -(\xi - \xi eq)/\tau$, with τ as a time constant and ξ_{eq} as an equilibrium value results in a macroscopic viscoelastic relaxation process. The coupled field may in this case represent an internal variable [11] such as dislocation density or concentration of vacancies or interstitial atoms. In technological application as an actuator, the user applies a specified value of a field variable, for example an electric signal to a piezoelectric material to obtain a motion or vibration.

To achieve negative stiffness or stiffness tending to infinity, it is sufficient that the coupled field variable ξ (e.g. electric field or temperature) increase with stress; it may be proportional to stress**.** The stress therefore modulates the corresponding flow (e.g. of electric current or of heat, Figure 1b). In the experiments described below, this is done with a contact condition [12] in which increased force results in increased contact area; this is best known in the classic Hertz solution for spheres but also occurs for all convex shapes. Hence greater flux occurs which causes expansion that neutralizes the deformation due to the force. It could also be done by material piezoresistance [13,14]. The total gradient in temperature or voltage is partitioned into a region of conductance that depends on force and a region of constant conductance so that the sensitive region's expansion is modulated by force. Neither rigid interfaces nor perfect bonds are assumed in the analysis or attempted in the experiments.

The total time-dependent effective creep compliance is $J(t) = c_1 + c_2\,\mathrm{e}^{-t/\tau}$ with c_1 and c_2 as constants that depend on coupling coefficients and applied gradient of ξ (e.g. electric field or temperature or other coupled variable) constant in time. The compliance can be positive, negative or zero (corresponding to infinite stiffness) depending on tuning as shown in Figure 2. Two experimental embodiments are presented in the following.

3. Experiments

The concept is illustrated experimentally in two systems, both of which entail a non-equilibrium flow of energy, and hence do not obey assumptions made in thermodynamic analyses of positive compressibility. The first contains a piezoelectric bimorph circular bender disc element (Digi-key 102-1170ND, 42 mm) supported at edges (Figure 3a) by a lens mount supported on an optical table. Deformation is related to force and voltage across the element. This coupling is achieved experimentally via a prism-shaped electrical contact (Tantone, type 633, Wabasha, MN) which modulates the electric current. Current was drained through a constant

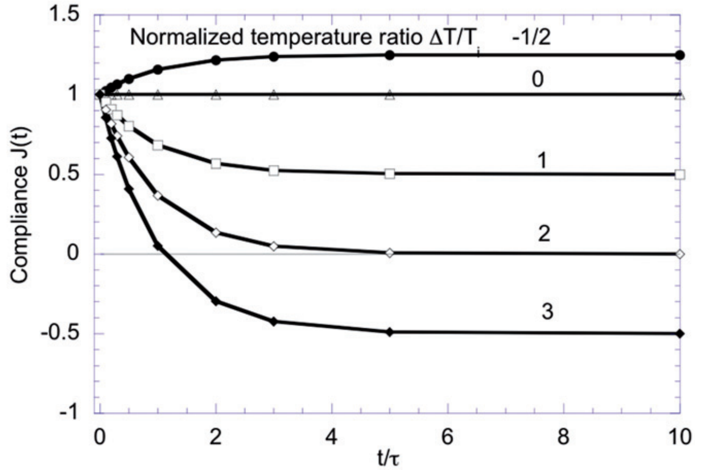

Figure 2. Analytical normalized compliance $J(t)$ as it depends on normalized time t/τ after application of force and normalized difference in field variable, e.g. temperature, constant in time.

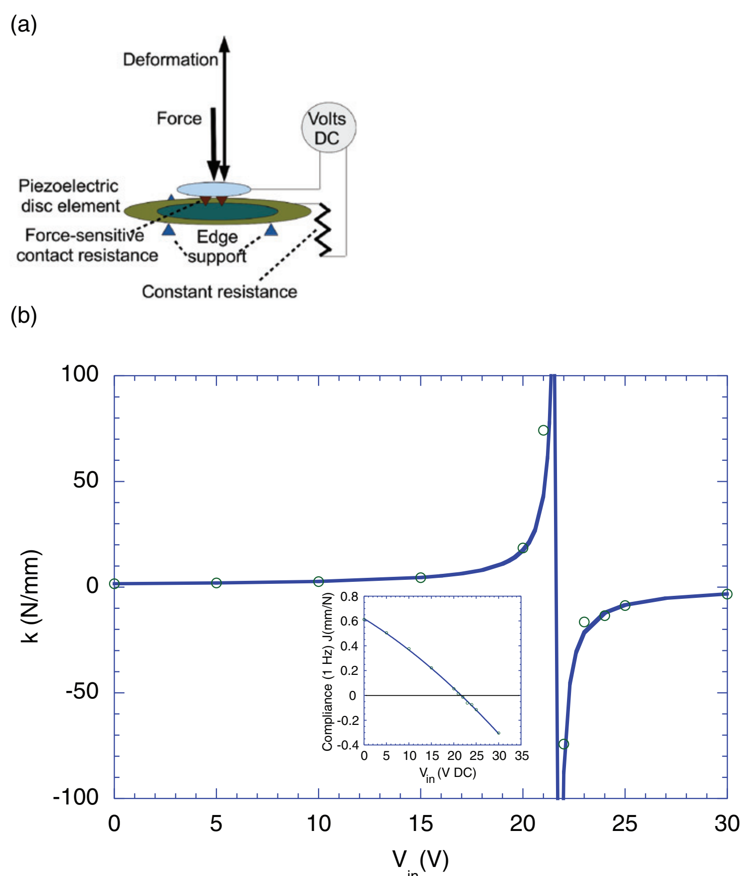

Figure 3. Measured stiffness at 1 Hz of a piezoelectric bender disc (a) vs. input DC voltage (b). Inset: compliance as a spring constant.

resistor so that the voltage across the piezoelectric element was force dependent and consequently the electrical conductivity contained a force-dependent portion and a force-independent portion as assumed in the analysis. The results reported were undertaken with a force applied by action of a coil upon a permanent magnet 3 mm in diameter, 1 mm thick; calibration was achieved with an analytical balance. The electric current to the coil was prescribed as a 1 Hz sinusoid from a Stanford Research SRS DS345 function generator. As the force increases, the contact resistance decreases, resulting in an increased voltage across the piezoelectric element. The deformation response was converted to an electrical signal with a fiber optic sensor, type MTI 2000; its output was measured via an Ithaco 3961B lock in amplifier. The fiber optic sensor was calibrated with a precision micrometer. By tuning the DC input voltage, the observed compliance is positive, negative or zero. Zero compliance corresponds to infinite stiffness. The force-dependent voltage entails a flow of current through a resistor, and hence an energy flux. The frequency (1 Hz) was well below the natural frequency and the frequency associated with RC delay. As shown in Figure 3b, the compliance at low frequency 1 Hz (corresponding to long time) decreases with applied voltage, crosses zero, and becomes negative. The system is stable without constraint. The structural stiffness (inverse compliance) tends to infinity.

A thermoelastic system is presented as a second experimental system. The coupled field variables are temperature and entropy (Figure 1a). Deformation depends on force via the elastic compliance and depends on temperature via thermal expansion. A cylindrical assembly of bimetallic helix segments of diameter and length about 2.5 mm consisting of about one and a half turn of strip with thickness 0.15 mm was chosen to obtain large expansion and moderate compliance. This granular system was pressed with a flat-ended indenter and held within a tube (Figure 4a). The force was abruptly applied in a step function in time via a controlled electric current through a coil acting upon a magnet. For a temperature difference of 79°C across the helices, there was a 3°C difference between the base and ambient; consequently the thermal conductivity contained a force-dependent portion and a force-independent portion as assumed in the analysis. Deformation was measured using a Trans Tek 240-000 LVDT and calibration was made with a precision micrometer. The force was applied by action of a coil upon a permanent magnet 12 mm in diameter and 3.2 mm thick; calibration was achieved with an analytical balance. The electric current to the coil was prescribed as a step function in time from a Matsusada R4K-80 controlled power supply. The coil and magnet were separated from the helix segments and the supporting optical table by a tubular stalk. Waveforms for force and deformation were captured with a Tektronix TDS 420A digital oscilloscope.

The compliance, shown in Figure 4b, approximates a step response indicating elastic behavior, when the indenter is at ambient temperature. The compliance in response to the heated indenter decreases with time, crossing zero and becoming negative. The time of zero crossing, and hence the attainment of infinite stiffness, depends on the temperature difference (Figure 2). The heat flow, and hence the thermal expansion, is modulated by a force-induced change in thermal contact at the surface and between each helix segment. The time delay results from thermal conduction lag. The response is stable with no constraint on the motion.

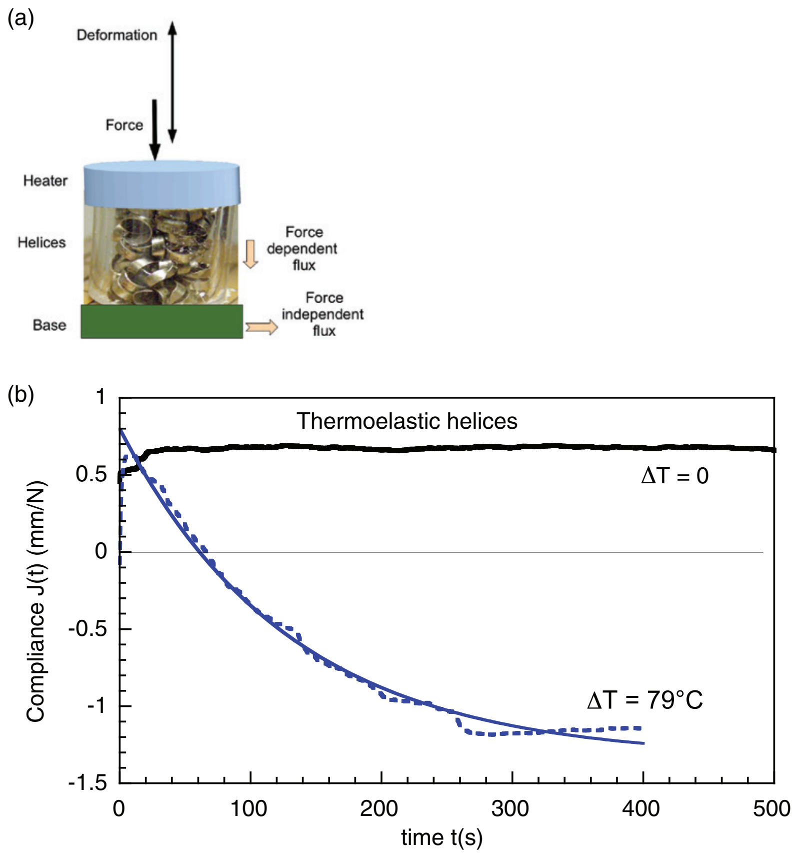

Figure 4. Measured time-dependent compliance of a thermoelastic granular material (a) of bimetallic helix segments with and without a temperature gradient (b). Solid-curve fit is exponential.

The reversed time dependence (creep) that is observed is classically forbidden [15]. However that analysis assumes a system with no external energy input and does not apply to the present system. *Any* granular material subject to sufficient thermal gradient has the potential for such effects because contact area, and hence thermal conduction, increases with force.

4. Analysis

Stress σ (force/area) and strain ε (deformation per length) are linked to coupled field variables ξ and Ξ as follows, with J as a material compliance and ψ, k, and K as constants.

$$\varepsilon = J\sigma + k\xi, \quad \Xi = K\sigma + \psi\xi \tag{2a, b}$$

For a thermoelastic material with α as thermal expansion, T as temperature, Equation (2a) is

$$\varepsilon = J\sigma + \alpha\Delta T \tag{3}$$

Suppose two segments in contact with thermal conductivity k, length L, and area A; for each define $\Phi = kA/L$. The total difference in temperature $(T_2 - T_1)$ is assumed to be constant in time. The heat flow due to a difference in temperature is, with T_b as the temperature at the interface,

$$\mathrm{d}Q/\mathrm{d}T = \boldsymbol{\Phi}_2(T_2 - T_b) = \boldsymbol{\Phi}_1(T_b - T_1) \tag{4}$$

To determine the transient response, we relate the flow rate to the heat capacity,

$$\begin{gathered}\mathrm{d}Q/\mathrm{d}T = C_p, \text{ so since } (\mathrm{d}Q/\mathrm{d}T)(\mathrm{d}T/\mathrm{d}t) = C_p\, \mathrm{d}T/\mathrm{d}t,\\ C_p\mathrm{d}T_b/\mathrm{d}t = \boldsymbol{\Phi}_2 T_2 - \boldsymbol{\Phi}_2 T_b - \boldsymbol{\Phi}_1 T_b - \boldsymbol{\Phi}_1 T_1.\end{gathered} \tag{5}$$

Consider an exponential time dependence

$$T_b(t) = p + q\ \exp\{-t/\tau\}. \tag{6}$$

Substituting and solving for p, q, τ,

$$p = T_{b\,final} = (\boldsymbol{\Phi}_2 T_2 + \boldsymbol{\Phi}_1\ T_1)/(\boldsymbol{\Phi}_2 + \boldsymbol{\Phi}_1) \tag{7}$$

$$q = T_1 - p \tag{8}$$

$$\tau = C_p/(\boldsymbol{\Phi}_2 + \boldsymbol{\Phi}_1) \tag{9}$$

The compliance is $J(t) = \varepsilon(t)/\sigma$, so from Equation (3), considering for simplicity that the thermal expansion of one of the series segments is sufficiently larger than that of the other,

$$J(t) = J + (\alpha/\sigma)\,[T_b(t) - T_1] \tag{10}$$

The zero of this time scale is the time at which the load is applied.

Therefore depending on the sign of the stress (compression is negative) and the direction of the thermal gradient, the time-dependent compliance $J(t)$ can increase with time as in the usual kind of creep, or can decrease with time, even to zero or negative values. A similar analysis applies to other coupled fields.

5. Discussion

Thermodynamic proofs [16,17] of positive properties make use of assumptions that may be stated in mathematical form but which entail restrictions on the type of material or physical system [18]. For example [16,19], a positive definite matrix is assumed. This allows negative cross properties that have no associated energy density, such as Poisson's ratio [20], piezoelectricity and thermal expansion [21] but requires positive stiffness or modulus. Positive definite entails a system with no stored energy and hence does not apply to composites [22] with pre-strain or materials undergoing phase transformation. Moreover, there is a tacit assumption of equilibrium. That does not apply to the systems considered here, which undergo energy flux. Energy flux occurs in all natural systems as well as in technological systems. For example, jointed pipes with flowing fluid can exhibit unusual mechanical effects [23]. Servo-controlled machines can maintain an effective zero compliance or other programmed response [24]. The range of materials suggested theoretically, as well as the lumped system and granular material studied experimentally, do not make use of such machines or control electronics.

Energy flux is minimized only in specific laboratory environments; for example in isothermal measurements of physical properties. Experiments that are inadvertently done in the presence of a gradient or flux typically return an average of intrinsic properties over the volume of the sample. They do not return extremely high or reversed properties. Such effects require design of the material as presented above, or a rather unusual set of incidental conditions.

The present systems exhibit a stable *compliance* crossing zero, which corresponds to singular stiffness tending to infinity. This contrasts with negative effects from resonance [25] of metamaterials via different principles. This is also in contrast to *moduli* that can soften to zero, then become negative according to Landau theory [26] in the vicinity of phase transformations, Such transformations do not lead directly to singular stiffness, though extremely high stiffness [27] (not proven stable) or viscoelastic damping [28] can be attained in composites via balance [29] between positive and negative stiffness. This can make use of a series (Reuss) morphology for which the compliance is $J_c = J_1 V_1 + J_2 V_2$ with V as volume fraction and the subscript indicating the constituent or a Hashin-Shtrikman coated-sphere morphology. In the series model the sum of a positive and negative compliance can give a small or zero compliance and hence a large stiffness tending to infinity. A parallel (Voigt) model has a stiffness $E_c = E_1 V_1 + E_2 V_2$ for which positive and negative contributions sum to a small or zero result. Negative moduli from phase transformations are unstable; indeed the transformation is the result of the instability. Negative modulus [30] or stiffness is observable provided an experimentally realisable external constraint is applied. Negative compressibility is also associated with internal constraints as in the van der Waals analysis of condensation of gases [31]. Negative compressibility also occurs in analysis of collisions in a two-dimensional system of infinitely many interacting hard discs constrained in a strip shaped region of fixed area and shape [32,33]. The systems explored here are stable without constraint and can be tuned from positive to negative compliance through a region of singular stiffness. The coupled field concept is quite general and is applicable to achieving extreme effective properties in all physical systems.

6. Conclusion

Stable systems are demonstrated that exhibit quasistatic stiffness that can be negative or tend to infinity without external constraint. They are based on coupled fields in the non-equilibrium presence of energy flux.

Acknowledgments

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