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Colloquium: Materials that exceed classical thermodynamic bounds on properties

Roderic S. Lakes[*]

Department of Materials Science, Department of Engineering Physics,
and Department of Mechanical Engineering University of Wisconsin,
1500 Engineering Drive, Madison, Wisconsin 53706-1687, USA

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Bounds on properties are useful in guiding design of heterogeneous materials and in understanding the distinction between effects that are physically reasonable and those that are not. Several bounds on physical properties can be exceeded by an appropriate choice of material. The reason is that the “proofs” of bounds contain either unstated assumptions about the material or assumptions that are couched in language that appears to be that of incontrovertible mathematics but that actually entails assumptions about the material. If those assumptions are relaxed, limits or bounds can be exceeded, as is demonstrated by analysis and experiments. For example, heat capacity, compressibility, electrical capacitance, and refractive index can be negative. Thermal expansion in composites can be larger or smaller than that of any constituent and can be negative. Materials and systems are known that are non-Hermitian or nonreciprocal. Currently, active fields of endeavor have arisen from such conceptual sources. Research efforts have led to the development of new materials and new classes of materials.

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I. INTRODUCTION

Thermodynamic limits on physical properties of systems and materials are useful in helping to discriminate between properties that are physically attainable and those that are not physically reasonable. For example, physical properties such as dielectric permittivity, diffusivity (Nye, 1976), and elastic stiffness (Sokolnikoff, 1946; Nye, 1976) are normally considered to be positive. Bounds on properties of heterogeneous solids such as polycrystalline materials and composites can be useful because the microstructure can be sufficiently complicated that exact analysis of the properties is difficult. Moreover, in many experimental situations, there is incomplete knowledge of the microgeometry of the composite even

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[*] Contact author: rlakes@wisc.edu

if many sectional images are available. Bounds are also pertinent in structural optimization analyses.

The upper and lower bounds for physical properties of two-phase and multiphase composites have been obtained in terms of the volume fraction V and properties of the constituents indicated by subscripts 1 and 2. For elastic properties such as the shear modulus G, the Voigt relation for the upper bound is particularly simple, $G_c = G_1V_1 + G_2V_2$ (Hill, 1963). For the Reuss lower bound, the corresponding compliances are considered. Composites with simple laminated structures attain the Voigt and Reuss relations. Voigt and Reuss relations also provide bounds on the dielectric or conductivity behavior of a two-phase composite (Lurie and Cherkaev, 1986). Tighter bounds for isotropic materials have been obtained for elastic moduli (Hashin, 1962; Hashin and Shtrikman, 1963). Bounds for elasticity of polycrystalline solids were developed as well (Hill, 1952). These bounds are sufficiently useful that they are tabulated for polycrystalline materials along with single-crystal elastic properties (Simmons, 1965).

Limits and bounds often contain assumptions, sometimes unstated, about the material. If one relaxes the assumptions and uses an appropriate material, bounds on physical properties can be substantially exceeded. Materials that exhibit sufficient additional freedom may require further physical property constants or variables for a full description. To properly characterize such materials, all of them must be measured.

The question of possible violation of the fundamental laws of thermodynamics is a different field of study and is not explored in this Colloquium. For example, James Clerk Maxwell considered a microscopic agent called a demon that could sort atoms by their speed to alter the temperature of adjacent chambers of gas that were initially at the same temperature. By allowing only faster atoms to go into one chamber, that one would become hotter and the adjacent one would become cooler without any input of energy. That would violate the second law of thermodynamics. The concept has stimulated fundamental studies of thermodynamics and its relation to other fields such as information theory. A microscopic sorting agent may expend energy to measure the speed of atoms, thus neutralizing any benefit. In addition, measuring the speed of atoms generates information, and erasure of that information at the end of the process requires energy. Such a sorting agent has not been physically realized. Other kinds of one-way devices and systems have been envisaged and explored, but if entropy is reduced in one region, it is increased elsewhere, so the second law of thermodynamics has not been violated and no useful power is generated. Known one-way systems may not obey reciprocity, as reviewed in Sec. VI, but they do not offer any challenge to the second law. For example, electrical diodes that allow electric current in one direction are nonlinear. If they were ideal, one could rectify electrical noise from thermal fluctuations and extract power, but that is not possible, owing to the real behavior of diodes. Similarly, studies of the possibility of extracting power from the zero-point energy of the quantum vacuum via Casimir type effects have not resulted in any demonstrable violation of the first law of thermodynamics. By contrast, derivations of various bounds and constraints on physical properties of materials make use of thermodynamics in a secondary sense and entail assumptions about the kinds of materials. Such assumptions are not always stated. By envisaging a material that does not obey the assumptions, it may be possible to exceed the bounds on physical properties. That is the subject of this Colloquium.

Physical properties such as elastic constants and dielectric constants are defined in Eqs. (12)–(14); elastic moduli are defined in Eq. (9). For example, in elasticity Young's modulus E is defined as the inverse of a compliance S in Eq. (13) corresponding to stress-free lateral surfaces and constant temperature. Such properties are routinely used to predict responses in configurations other than those used in the original experiments. Stiffness and strength measurements by standard tests are used to predict the durability of electric power lines carrying considerable current. Testing configurations are generally not ideal. Often the nonideal aspect is inconsequential. For example, in solids adiabatic and isothermal compliances (Nye, 1976) typically differ by less than one part in 10^3. In actual experiments and applications, there may be an exchange of heat and even atoms with the environment, and hence an unintended power input. All materials exhibit coupled field effects (see Sec. V); systems are never fully closed. When such effects are large by intent, interesting effects may occur. One nevertheless speaks of properties such as elastic moduli, if one prefers, as effective properties.

Materials studied in the context of exceeding bounds may have an unusual or novel structure. That does not guarantee unusual physical properties. For example, quasicrystals were initially thought to be impossible based on geometrical assumptions regarding crystals. Their properties, however, do not exceed any bounds.

New materials and structures developed to achieve unusual or extreme properties tend to be heterogeneous. Some of these materials are composites with multiple constituents, and some are foams or lattices containing ribs or surfaces and void space. How fine must the structure size be in order to regard it as a material? Because people in different disciplines have pursued research in the areas under review, there is not necessarily agreement. For the purposes of this review, when continuum concepts of fields and material properties are used to describe an object, it will be referred to as a material.

Some terminology has emerged in connection with materials with extreme or unusual properties. For example, auxetic refers to a negative Poisson's ratio and metamaterial refers to a material with unusual or extreme properties, in particular, materials with structure at scales larger than the atomic scale or the size of crystallites in a polycrystalline solid. The first such materials and associated concepts considerably predate the neologisms, and many recent studies do not use neologisms in the title or abstract. Consequently, scholarship is impeded if one relies on such terminology. In this Colloquium descriptive terms are used rather than neologisms. Primary early references are cited to provide context for recent research and to help discern the degree of novelty. It is the purpose of this Colloquium to explore the conceptual underpinnings of limits or bounds on physical properties, crucial assumptions in obtaining the bounds, ways to exceed the limits, and the evolution into currently active fields of research.

This Colloquium is organized as follows. In Sec. II heat capacity is considered; the standard proof of positive heat capacity is discussed and negative heat capacity systems are presented. Section III deals with elastic and viscoelastic properties, including negative stiffness and moduli in Sec. III.A.2. Proof of symmetry of the stress tensor is presented and materials with an asymmetric stress tensor are considered in Sec. III.B.2. The assumption of parity invariance is discussed and chiral materials are reviewed in Sec. III.B.3, and proof of symmetry of the modulus matrix is discussed in Sec. III.B.4, with physical examples of materials with an asymmetric modulus matrix addressed in Sec. IV.A. Active and nonconservative materials are considered in Sec. IV. Such materials enable stable negative properties as well as "odd" effects associated with asymmetry of the modulus tensor and additional elastic constants. Non-Hermitian materials and systems are possible, as reviewed in Sec. IV.F. Electrical capacitance and permittivity including negative values are considered in Sec. IV.D. Coupled fields including thermal expansion, piezoelectricity, and the Hall effect are reviewed in Sec. V. Reciprocity with nonreciprocal examples are reviewed in Sec. VI. Properties that depend on resonance are reviewed in Sec. VII.

II. HEAT CAPACITY

A. Proof of positivity

When preparing coffee or tea, one applies heat under the kettle. One does not expect the water to get colder or to freeze solid. This experience is generalized and codified in the concept of heat capacity and in the classic demonstration in which is positive for constant volume (Wallace, 1972). The coffee expands as it warms, so it is not at a constant volume. It is not at constant pressure because atmospheric pressure changes with the weather. Even so, the coffee warms when heated, as expected.

The heat capacity is the ratio of change in heat Q to change in temperature T,

$$C_V = \frac{\partial Q}{\partial T}\bigg|_V, \tag{1}$$

in which the subscript V indicates constant volume.

This can be written in terms of the entropy S or the energy U as

$$C_V = T\frac{\partial S}{\partial T}\bigg|_V = \frac{\partial U}{\partial T}\bigg|_V. \tag{2}$$

Following Wallace (1972), the first law of thermodynamics is

$$dQ = dU + dW, \tag{3}$$

with dW the work done by the material. The quadratic form containing the internal energy U is positive definite,

$$\frac{\partial^2 U}{\partial S^2}\bigg|_V > 0, \tag{4}$$

with T the temperature and S the entropy.

Thus, the internal energy must be a minimum with respect to all variations, so $d^2U > 0$. Therefore,

$$\frac{\partial T}{\partial S}\bigg|_V = \frac{T}{C_V} > 0. \tag{5}$$

The temperature $T > 0$, so $C_V > 0$. The heat capacity is positive, which means that if heat is added, the temperature increases. In addition to the assumption of positive definite energy density, the system is assumed to be in equilibrium. It is also tacitly assumed that the material or system can be subdivided. In solids the heat capacity at constant volume differs from that at constant pressure or stress by 10^{-3} or less (Nye, 1976). For a monatomic ideal gas, $C_P/C_V = 5/3$.

B. Negative heat capacity

The notion of minimum internal energy is actually an assumption about the system or material. In some didactic analyses, the physical role of minimum internal energy as a criterion of stability is provided (Reichl, 2016); in many analyses it is not. Nevertheless, the analysis entails assumptions about the material.

Heat capacity is negative in stars (Lynden-Bell, Wood, and Royal, 1968; Lynden-Bell, 1999; Posch and Thirring, 2005), in which there is an internal source of power from nuclear fusion as well as a long-range gravitational interaction. If the fusion power increases in the star core, the star expands and is observed to become cooler. This feedback due to negative heat capacity stabilizes power production in stars like our Sun so that they shine steadily for eons (Posch and Thirring, 2005). The power within the star also increases as the star begins to complete its evolution and begins fusing helium in the core in addition to fusing hydrogen. The star then expands and cools to become a giant. Such cooler giant stars are prominent in the night sky, as seen by their warm yellow and orange colors. The progenitor stars were hotter and were yellow white, white, or blue white in color.

In star clusters the long-range gravitational interaction is accounted for in orbital dynamics via the laws of motion. Nevertheless, if more energy is introduced by a high velocity star entering the cluster, there is expansion against gravity and the average velocity of the stars; hence, the effective temperature of the cluster decreases. Systems with long-range forces such as gravity cannot be subdivided. The contrast with thermodynamic "proofs" has been explained (Lynden-Bell, 1999). The notion of negative heat capacity was appreciated by astronomers before it was by physicists (Lynden-Bell, 1999). Black holes constitute another gravitational example. The temperature of the Hawking radiation decreases as mass or energy is added, corresponding to a negative heat capacity.

Indeed, coupling of a thermodynamic system with gravity suffices to allow a negative heat capacity, as presented in an analysis of a thought experiment (Herrmann and Hauptmann, 1997). Specifically, in a simple model of negative heat capacity of a star, a gas in a cylinder is heated. As with many tutorials in thermodynamics, the gas is constrained by a piston. In this model the piston is connected by a stalk to a rotor disk that has a curved groove guide on which a string moves. The stalk on the piston engages the inner rotor shown

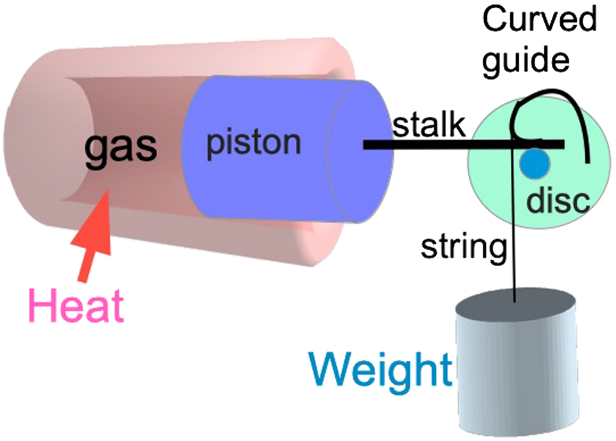

FIG. 1. Negative heat capacity system intended to model a star. A compressed gas in a cylinder is coupled to gravity via a string on a nonlinear curved guide. As heat is applied, the piston moves to the right as the gas expands, the disk rotates, and the gas temperature decreases. Adapted from Herrmann and Hauptmann, 1997.

as a dark circle and causes the disk to rotate quasistatically. The string supports a weight, as shown in Fig. 1. The arrangement provides a force law $F(x) = -C_2/x^a$, with C_2 a constant and a a constant such that $1 < a < \gamma$. The constant γ is an adiabatic exponent of the gas for which $PV^\gamma = \text{const}$, with P as pressure and V as volume. The system is stable if $a < \gamma$. The actual pressure-volume curve is neither isothermal nor adiabatic. As heat is added, the gas expands and cools. The heat capacity is negative via the nonlinear coupling with gravity. If one considers the gas and piston as the system with the stalk doing work on it, the heat capacity of the gas alone will be positive. That no longer models a star, in which gravity cannot be decoupled from the thermal aspects.

Negative heat capacity can also occur in materials, such as rapidly cooled amorphous materials near the glass transition temperature that are not in equilibrium (Bisquert, 2005). Negative heat capacity also occurs in the liquid-to-gas transition of finite systems such as clusters of ions (Gobet *et al.*, 2002) and has been studied in the context of energy fluctuations in the liquid-to-gas transition (Chomaz, Duflot, and Gulminelli, 2000) and in a vorticity model (Andersen and Lim, 2007).

The difference between extensive and intensive control parameters (Gross, 2006) has been pointed out in microcanonical and canonical analyses of negative heat capacity in phase transitions. Extensive properties are those that depend on the extent of a system (hence the size). They change as the system is scaled in size. Internal energy, entropy, and volume are extensive variables. Temperature, by contrast, is called intensive because it can vary with position. The assumption of energy as an extensive quantity fails for gravitating systems (Posch and Thirring, 2005) and for laboratory scale materials near phase transitions (Gross, 2006). The notion is also pertinent in macroscopic systems regardless of whether they have long- or short-range interactions. The restriction to homogeneous systems in the usual thermodynamic analyses is often unstated, but the restriction excludes phase separation and systems with few particles. Overcoming the thermodynamic restriction to positive heat capacity is consistent with conservation of total energy.

III. ELASTICITY AND VISCOELASTICITY

A. Stiffness and compressibility

1. Proof of positivity

In one dimension one expects a spring constant as a one-dimensional stiffness to be positive. Similarly, for compressibilityone expects an increase of pressure on a material to cause a decrease in volume. This perspective is codified as a thermodynamic proof, after Wallace (1972), as follows. The pressure P at constant entropy S as indicated by the subscript is, from the increment of work $dW = PdV$,

$$P = -\left.\frac{\partial U}{\partial V}\right|_S, \tag{6}$$

with V the volume. The quadratic form $\partial^2 U/\partial V^2$ containing the internal energy U is positive definite, so there must be a minimum with respect to all variations $d^2U > 0$,

$$d^2U = \left.\frac{\partial^2 U}{\partial S^2}\right|_V (dS)^2 + 2\left.\frac{\partial^2 U}{\partial S \partial V}\right|_{VS} (dS)^2 + \left.\frac{\partial^2 U}{\partial V^2}\right|_S (dV)^2. \tag{7}$$

The constant volume V and constant entropy S are indicated by subscripts. Thus, considering only variations in volume at constant entropy,

$$-\left.\frac{\partial P}{\partial V}\right|_S = \frac{1}{Vk_S} > 0, \tag{8}$$

with $k_S = (1/V)(\partial V/\partial P_S)$ the adiabatic compressibility. Thus, the compressibility [and hence the bulk modulus K (the inverse compressibility)] is positive.

2. Negative stiffness

The notion of minimum internal energy is actually an assumption about the system or material. Exceptions are as follows.

In one dimension negative spring constants can occur in buckled spring systems, or buckled bars or tubes in which energy is stored during the buckling process. Examples are shown in Fig. 2. Because energy is stored, the object is not in a minimum energy state as was assumed. A buckled object is unstable by itself but may be stabilized by an external constraint. In discrete (lumped) systems, it has long been known that negative stiffness is possible (Panovko, Gubanova, and Larrick, 1965). For example, if the input in the transverse direction for the buckled spring or strip is displacement controlled in Fig. 2, one can experimentally observe a region of negative stiffness. For the buckled tube, the controlled displacement is applied in the longitudinal (vertical) direction. The system is stabilized by displacement control; a regime of negative stiffness can be experimentally observed. If the force is controlled, the strip will exhibit snap through instability.

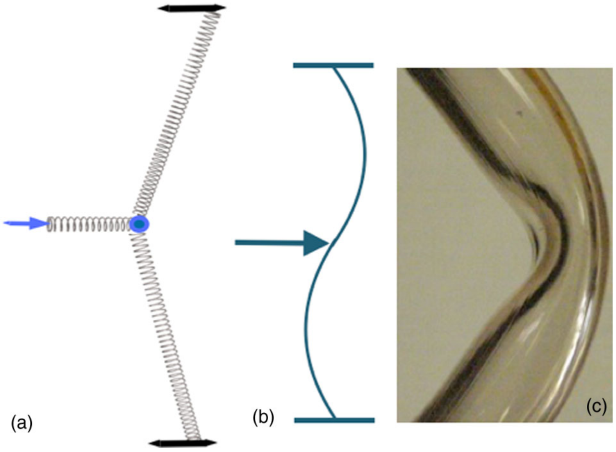

FIG. 2. Negative stiffness via buckling of (a) a spring system with constrained ends, (b) a flexible compressed strip with constrained ends, and (c) a flexible tube compressed vertically under displacement control. Arrows indicate an applied force or displacement. (c) Adapted from Lakes, 2020.

In three-dimensional solids, negative compressibility and negative moduli are possible. The usual assumption of a positive definite strain energy in elasticity (Sokolnikoff, 1946; Nye, 1976) implies that a block of material with free surfaces will be stable. If there is a constraint on the surfaces, the object can be stabilized even if it contains stored energy and does not manifest a positive definite strain energy.

There is more than one criterion of stability. The stability criterion of strong ellipticity also applies; it allows a wider range of properties than positive definiteness. The physical meaning is that waves in the material propagate with a real velocity. The bulk modulus (inverse compressibility) can be negative, but the shear modulus must still be positive.

A strongly elliptic solid with a negative bulk modulus can be stabilized provided that it is constrained at the surface. For flexible foams measurement of the negative bulk modulus was done constraining the specimen under volumetric prestrain (Moore *et al.*, 2006). In foams individual cells exhibit a negative incremental compressive modulus, but compression of such a foam reveals a band instability because the axial C elastic modulus must still be positive for stability (Lakes, Rosakis, and Ruina, 1993) even with a constraint; if it is negative, the material becomes unstable.

Negative elastic moduli in materials may be understood in the context of the Landau theory of phase transformations (Landau, 1965). Consider an energy function of strain and a control variable such as temperature, electric field, magnetic field, pressure, or composition, as illustrated in Fig. 3. For example, let the normalized temperature T be lowered from a level above the transformation temperature. The curve of energy versus strain at that temperature has a single minimum. This gradually flattens as temperature is lowered. The curve develops two minima or potential wells. The condition of zero strain becomes unstable. The crystal, if it is unconstrained, then changes shape, volume, or both corresponding to positive stiffness within a potential well at nonzero strain. If the strain is a shear strain corresponding to a shape change, the transformation is called martensitic. For example, nitinol (nickel titanium) alloys exhibit martensitic transformations and shape memory effects. If the strain is a hydrostatic strain, it is a volume change transformation.

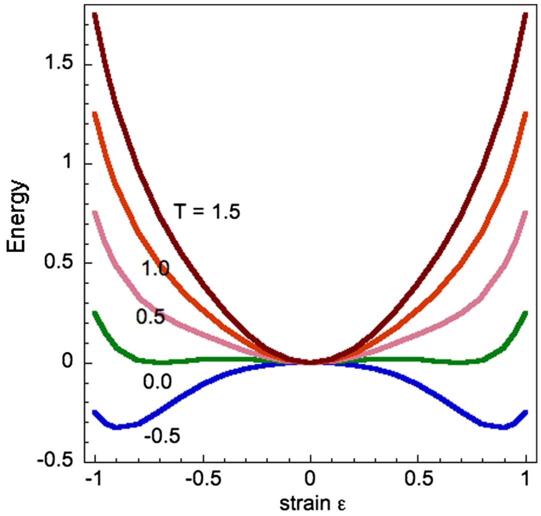

FIG. 3. Landau energy curves for a phase transformation. The normalized temperature is T. There are arbitrary units of energy and temperature. The curvature represents an elastic modulus. Adapted from Jaglinski *et al.*, 2007.

The curvature of the energy function represents an elastic modulus. Therefore, a flattening of the curve at the transformation corresponds to a softening of the modulus. One or more moduli will soften near a critical temperature T_c. If the bulk modulus softens more than the shear modulus, a minimum occurs in Poisson's ratio, with negative values possible (see Sec. III.D), as has been observed experimentally (Hirotsu, 1990; McKnight *et al.*, 2008). Below T_c the shape of the energy versus strain curve near the origin represents a negative modulus. If the crystal is unconstrained, the crystal will change shape, volume, or both rather than exhibit a negative modulus. If it is constrained, the negative modulus may be observed or consequences such as extreme damping or modulus in a composite may be observed.

3. Extreme stiffness

The stiffness of a composite is normally considered to be no larger than that of the stiffest constituent. The bounds on stiffness combined with the positivity of the moduli of the constituents based on the stability of an object with free surfaces provide that limit (Hill, 1963; Hashin, 1962; Hashin and Shtrikman, 1963). If one constituent is allowed to have a negative modulus or the material is supplied with external power, then the material does not obey the usual assumptions. An analysis and experiment showed that extremely high stiffness is possible (Lakes, 2001b).

Negative moduli in inclusions within a composite (Lakes, 2001b) have been used to attain arbitrarily large material damping, as observed in an experiment (Lakes *et al.*, 2001), and stiffness exceeding classical bounds. Composites with phase transforming inclusions in the negative stiffness regime can be stiffer than diamond over a range of temperature during a temperature scan (Jaglinski *et al.*, 2007). The experiments not only revealed extreme stiffness, they also admitted inference of the negative moduli of the inclusions. The negative stiffness is possible because, in the vicinity of a phase transformation, there is stored energy that can be released during subresonant oscillatory deformation. These materials are metastable. Solids containing inclusions of negative stiffness can be made stable with an appropriate choice of elastic constants (Drugan, 2007). Stable extreme effective dynamic stiffness can occur, but not static stiffness (Wojnar and Kochmann, 2014). Stiffness and damping of ferroelectric materials such as those used as inclusions can be tuned via the electric field as well as temperature (Wojnar, le Graverend, and Kochmann, 2014).

A source of power may be envisaged (Gold, 1948; Thompson, 1982; Martin, Mehta, and Hudspeth, 2000; Lakes, 2012a) rather than an internal energy, as discussed in Sec. IV.C. The extreme stiffness regime is stabilized via a source of power in an irreversible condition.

B. Symmetry and asymmetry

1. Symmetry of the material

The material need not be isotropic (properties independent of direction), though isotropy is often assumed in initial studies of elasticity. In classical elasticity (Sokolnikoff, 1946) stress σ_{ij} (force divided by area) is related to strain ϵ_{kl} (symmetric part of deformation gradient) via C_{ijkl} as the elastic modulus tensor,

$$\sigma_{ij} = \sum_{k=1}^{3}\sum_{l=1}^{3} C_{ijkl}\epsilon_{kl}. \tag{9}$$

This expression of Hooke's law [Eq. (9)] allows for anisotropy of the modulus or stiffness. There are 81 components of the elastic modulus or stiffness C_{ijkl}. The strain is symmetric by definition. The stress is symmetric in classical elasticity via a shear force balance argument. Then there are 36 independent components of the modulus tensor for the least amount of material symmetry corresponding to a triclinic crystal. It is expedient to write the modulus as a six-by-six matrix in two dimensions.

Physically, the modulus element C_{1111} is a stiffness in the x direction that governs the speed of longitudinal waves in that direction with a wavelength much shorter than the object size, as in ultrasonic waves or seismic waves in Earth. Similarly, the modulus element C_{3333} is a stiffness in the z direction. The modulus element C_{2323} is a stiffness with respect to shear; it governs static shear properties as well as the velocity of shear waves. One can obtain all the modulus components by transmitting waves in different directions provided that the material is reasonably homogeneous on the scale of the wavelength.

For measurements of static stiffness, it is expedient to use the compliance formulation in which $\epsilon_{ij} = S_{ijkl}\sigma_{kl}$, with repeated indices summed over via the Einstein convention. The material may be isotropic or anisotropic. The compliance S_{ijkl} is also called J_{ijkl}. If one stretches or compresses a thin bar or rod, the stiffness expressed as Young's modulus E in the x direction is $E_1 = 1/S_{1111}$. The Poisson's ratio ν is given by the compliance element $S_{2211} = -\nu_{12}/E_1$.

2. Symmetry of the stress

Elastic solids undergo force per area or stress. Symmetry of the stress tensor was demonstrated by arguing that an unbalanced shear will cause a microelement of material to accelerate to high spin, contrary to the assumed equilibrium (Sokolnikoff, 1946; Nye, 1976).

The stress is symmetric provided that there is only force per area and no distributed moment per area or volume. That is the case provided that the size of the microstructure of the material is negligible compared to the size scale in the experiment or the homogenization analysis and provided that the material does not experience distributed torques on magnetic or electric dipoles. If there are such moments, they can balance an asymmetric stress.

If moment per area is allowed, the solid is Cosserat (Cosserat and Cosserat, 1909) [micropolar (Eringen, 1968)] elastic, not classically elastic. The moment per area is proportional to the gradient of a local rotation variable of points called the microrotation. Cosserat solids have a characteristic length scale that is non-negligible compared to the size scale of the observations. There are six elastic constants for the isotropic Cosserat solid in contrast to two elastic constants for an isotropic classical solid. The classical theory of elasticity has no length scale. By contrast, two characteristic lengths can be expressed in terms of the tensorial constants of the Cosserat solid. There is also an elastic constant that quantifies the degree of coupling between the microrotation and the rotation associated with the gradient of displacement of the points. These are continuum properties, as is the case with classical elastic constants; however, they can be related via homogenization analysis to physical length scales in the material under study. If the characteristic lengths are orders of magnitude smaller than the size scales in the experiment, the material can be viewed as classically elastic.

Many experiments on bone, foams (Rueger and Lakes, 2019), and 3D rib lattices (Rueger and Lakes, 2018) have been done in the context of Cosserat freedom. For isotropic materials experiments have determined all six elastic constants. Experimental protocols include measurements of nonclassical dependence of rigidity upon specimen thickness and full field determinations of nonclassical deformation fields. Full field methods have been used to demonstrate the asymmetry of the stress in such materials. Wave methods have also been used to study a granular ordered solid interpreted in the context of Cosserat elasticity (Merkel, Tournat, and Gusev, 2011).

For crystal lattices the predicted characteristic length scale is on the order of the interatomic bond length (Minagawa, Arakawa, and Yamada, 1980). In a 2D chiral lattice with a negative Poisson's ratio (Prall and Lakes, 1997), the characteristic length was found to be on the order of the cell size (Spadoni and Ruzzene, 2012).

There are other theories of elasticity with different degrees of freedom. A theory with less freedom than classical elasticity was studied during the development of elasticity theories. This theory, based on an analysis of forces between atoms in a material, predicted a Poisson's ratio of 1/4 for all isotropic materials and also fewer constants than classical elasticity for anisotropic materials. By contrast, classical elasticity, based on a tensor analysis of a continuum that undergoes strain in response to stress, predicts a range of Poisson's ratio from -1 to $1/2$ for isotropic materials. Early experiments revealed common materials to have Poisson's ratios ranging from 0.2 to 0.45, so the tensorial approach was chosen as the basis for what is now called classical elasticity. The early analysis of interatomic forces turned out to contain overly restrictive assumptions: it assumed that the forces were central forces and that the motion of the atoms was affine.

Moment per volume, in contrast to moment per area, occurs in a magnetic material with dipoles in the presence of a magnetic field (Dorfmann and Ogden, 2023) or in a ferroelectric material in an electric field. The stress in such materials can be asymmetric.

3. Symmetry of handedness: Chirality

Chiral materials distinguish right- versus left-handedness. Chirality is well known in the chemistry of biological materials such as sugar and DNA and also in mineral crystals such as quartz. The role of chirality is also well known in coupled field effects such as piezoelectricity and pyroelectricity, as well as in optical activity. Chirality is pertinent in this Colloquium because it is ignored in many analyses yet can give rise to interesting behavior and nonclassical response, including effects in non-Hermitian materials and systems, generalized elasticity, and Poisson's ratio beyond the accepted range.

As for designed chiral materials, chiral inclusions as in Fig. 4 can be incorporated into designed composites. Alternatively, lattices of ribs, plates, or surfaces can be made in chiral form.

Wood is chiral on macroscopic and microscopic scales. Some trees exhibit a substantially spiral grain (Leelavanichkul and Cherkaev, 2004). The resulting increase in flexibility may confer an advantage in windy environments. The chirality of wood on a microscopic scale has been observed and has been exploited to make high performance actuators that twist in response to a change in relative humidity (Plaza *et al.*, 2013). This is a coupled field effect; see Sec. V.

Physical properties describable by tensors of odd rank, for example, piezoelectricity and pyroelectricity, require materials to be chiral for nonzero effects. Chirality cannot be incorporated in classical elasticity because the fourth rank modulus or compliance tensor is invariant to the inversion of all three coordinate axes corresponding to chiral asymmetry.

Cosserat solids, which admit distributed moments and an asymmetric stress (see Sec. III.B.2), have sufficient freedom to accommodate elastic chirality. Phenomena such as coupling between squeezing and twist deformation can occur in chiral solids, as shown in an analysis (Lakes and Benedict, 1982) and experiments on bone (Lakes, 1981) and lattices (Reasa and Lakes, 2020). There are nine elastic constants for an isotropic chiral 3D solid. Chirality is also of interest in the context of active materials and non-Hermitian materials, as reviewed in Sec. IV.

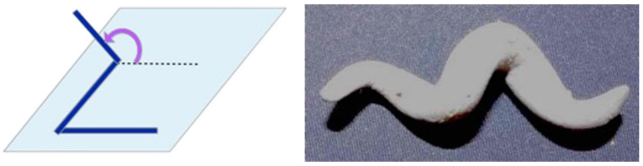

FIG. 4. Chiral inclusions. Left image: bent wire chiral inclusion. Adapted from Tsai *et al.*, 2005. Right image: twisted inclusion.

4. Symmetry of the modulus tensor

A material that is classically elastic, and possibly anisotropic, is describable by a conserved strain energy function $U = (1/2)C_{ijkl}\epsilon_{ij}\epsilon_{kl}$ in which the stress is $\sigma_{ij} = \partial U/\partial \epsilon_{ij}$ (Sokolnikoff, 1946). The strain is ϵ_{ij}. Then the modulus is symmetric with respect to the exchange of pairs of indices $C_{ijkl} = C_{klij}$, and the number of independent elastic constants is reduced from 36 to 21 for a triclinic material. Similarly, the compliance is symmetric ($S_{ijkl} = S_{klij}$). The number of classical independent constants is also reduced for other symmetries, for example, from twelve to nine for orthotropic and from six to five for hexagonal or axisymmetric. There are three moduli or compliances for cubic symmetry and two moduli for isotropic symmetry, regardless of whether the modulus or compliance tensor is symmetric (Rogers and Pipkin, 1963).

Not all materials are describable by a conserved strain energy, as presented in Secs. IV.A and III.A.2. Asymmetry of the modulus or compliance that may be called odd can occur, giving rise to additional constants (see Sec. IV.A), as discussed in Sec. IV.E.

C. Viscoelastic damping

1. Phenomena and positivity

Viscoelastic damping is manifest as a phase angle between stress and strain when deformation is sinusoidal in time in decay of amplitude of vibration of an object following an impact, as well as in the attenuation of waves. Damping can be represented as the imaginary part of a stiffness or modulus. In the time domain, strain increases in creep if the material is held under constant stress. All viscoelastic phenomena are nonequilibrium.

The damping has been proven to be positive by appealing to energy principles. Specifically, the assumption of a nonnegative rate of dissipation of energy implies positive viscoelastic damping (Christensen, 1972). Similarly, bounds have been obtained on properties of viscoelastic composites (Roscoe, 1969).

2. Extreme and negative viscoelastic damping

Energy principles entail an assumption about the type of material. Actual materials may be made with constituents with stored energy or there may be an external power source that allows amplification, which is effectively a negative damping.

As for composites with constituents with stored energy (see Sec. III.A.2), damping can be negative or can greatly exceed that of either constituent and can exceed predictions made using classical analysis. Specifically, composites with inclusions of negative stiffness can exhibit arbitrarily large material damping via analysis (Lakes, 2001b), via experiments with buckled tubes (Lakes, 2001a), and in experiments with a composite containing phase transforming inclusions (Lakes *et al.*, 2001). Transient negative damping was observed in these materials as well.

Negative damping also occurs in materials that are provided with a power source. This includes biological materials, which have a metabolic power source and are in certain semiconductors provided with an electric current. Such negative damping is not limited to transient effects. Details are provided in Sec. IV.B.

D. Poisson’s ratio

Poisson’s ratio refers to the ratio of lateral contraction strain to axial extension strain during stretching. Visualize a rubber band becoming thinner when stretched. The classical isotropic bounds on Poisson’s ratio (Sokolnikoff, 1946) are from −1 to 0.5, thus allowing negative values. The bounds on Poisson’s ratio are based on positivity of the bulk modulus (inverse compressibility) and the shear modulus. Negative Poisson’s ratio solids, while initially counterintuitive, have been with us for some time in two dimensions (Gibson *et al.*, 1982) and three dimensions (Lakes, 1987) and now constitute a popular field of study; some representative articles are cited later in the Colloquium. Negative Poisson’s ratios, though they do not necessarily violate any bounds, are still counterintuitive to many. The word *auxetic* was introduced for negative Poisson’s ratio materials well after they were first developed; the terminology is now broadly used. Achieving a negative Poisson’s ratio via a heterogeneous cellular structure does not depend on the chemistry of the solid material and does not depend on stored energy or metastability. Polymers and metals have been used to make these materials (Friis, Lakes, and Park, 1988).

A chiral 2D lattice (Prall and Lakes, 1997) exhibited a Poisson’s ratio of −1, regardless of strain. A 2D lattice with star shaped cells exhibited a Poisson’s ratio tunable to negative values by angle (Wang, Shen, and Liao, 2017).

Negative Poisson’s ratios have been inferred or observed in a variety of systems and materials. For example, a lattice of hinged rigid units or polygons exhibits a negative Poisson’s ratio (Wojciechowski, 1989; Grima, Alderson, and Evans, 2005). This kind of lattice also resists deformation due gradients in stress to a greater extent than known materials that exhibit an asymmetric stress; see Sec. III.B.2.

Poisson’s ratio can become negative in materials near a phase transformation (Hirotsu, 1990; McKnight *et al.*, 2008). Figure 5 shows the softening of the bulk modulus and a minimum in Poisson’s ratio for quartz near a phase transformation after McKnight *et al.* (2008). In such materials the bulk modulus (inverse compressibility), called K or B, softens more than the shear modulus. A negative bulk modulus is predicted via the Landau theory of phase transitions and is inferred in experiments with composites, as discussed in Sec. III.A.2, but violates the assumption of positive definite strain energy.

Hierarchical laminates can give rise to a negative Poisson’s ratio (Milton, 1992) in two dimensions, as illustrated in Fig. 6, and in three dimensions. There must be a sufficient contrast in modulus between the constituents for the Poisson’s ratio to be negative.

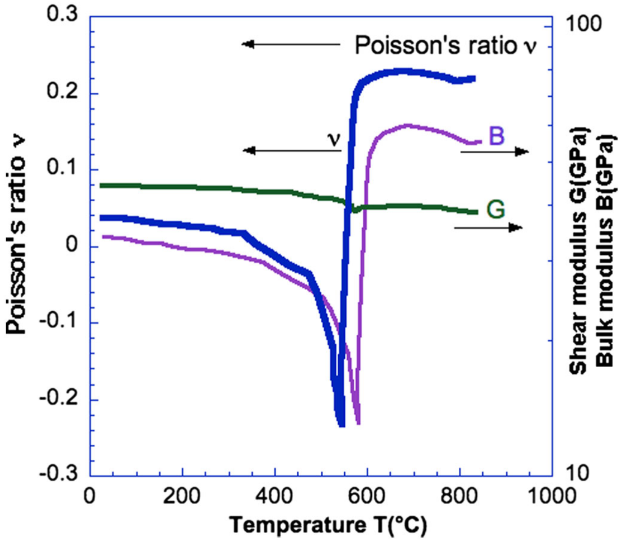

FIG. 5. Moduli and Poisson’s ratio ν for quartz near a phase transformation. The bulk modulus softens much more than the shear modulus near the transformation temperature. Adapted from McKnight *et al.*, 2008.

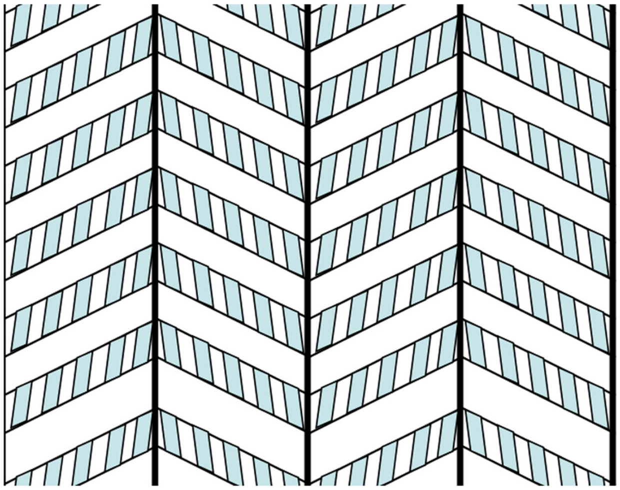

FIG. 6. Hierarchical laminate with a negative Poisson’s ratio. Adapted from Milton, 1992.

A 2D lattice with interlocking hexagonal subunits with embedded negative stiffness elements displays a Poisson’s ratio of −1 and tunable negative stiffness (Hewage *et al.*, 2016). The relationship between a negative Poisson’s ratio and negative stiffness is illustrated in Fig. 7. The region for which bulk modulus K and shear modulus G are positive is stable. Negative Poisson’s ratio is possible if the bulk modulus is sufficiently small. If the bulk modulus becomes somewhat negative, the material no longer obeys the criterion of positive definite strain energy. An unconstrained block is unstable; however, an external constraint can stabilize it provided that the solid obeys the condition of strong ellipticity. Strong ellipticity, which entails positive wave speeds, is a less rigorous condition than positive definiteness. If strong

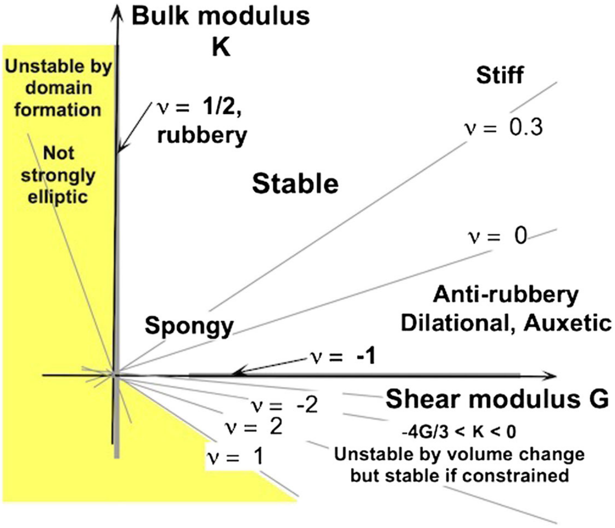

FIG. 7. Relation between the negative Poisson's ratio ν and negative stiffness. Shown is a map of bulk modulus K vs shear modulus G Negative moduli violate the assumption of positive definite strain energy, but a negative Poisson's ratio down to -1 is consistent with that assumption. Adapted from Wang and Lakes, 2005.

ellipticity is violated, the material becomes unstable with respect to the formation of bands of heterogeneity. Such bands are observed in some materials that undergo phase transformations and in open cell foams under compression.

In three dimensions, if elastic moduli are allowed to be negative, then the Poisson's ratio can attain values outside the classically allowable range (Wang and Lakes, 2005). Negative moduli entail instability. As we have seen, if a specimen of material is constrained at its surface, then it may be stabilized, thereby allowing negative moduli. While negative incremental bulk modulus has been observed in constrained triaxial compression experiments, it is not easy to measure Poisson's ratios under constrained conditions.

In directionally isotropic 3D chiral (Reasa and Lakes, 2020) materials with positive moduli, the classical bounds on Poisson's ratio can be exceeded. The reason for this is that the proof of the bounds assumes that changes in volume and changes in shape are independent. In chiral solids, even if they are directionally isotropic, squeeze and twist deformation are coupled via Cosserat freedom (Lakes and Benedict, 1982); see Sec. III.B.2. An analysis (Lakes and Benedict, 1982) and an experiment (Lakes and Huey, 2024) showed that if the specimen is not too large, this coupling allows Poisson's ratios outside the classically allowable range even for a positive shear and bulk modulus. Volume and shape changes are no longer independent in such materials, in contrast to the assumptions made in demonstrating a range of Poisson's ratio from assumed positivity of the moduli.

IV. ACTIVE AND NONCONSERVATIVE MATERIALS

Perfect elastic and dielectric materials are an idealization. Actual materials will dissipate energy (viscoelasticity or dielectric relaxation) and convert it into heat. Active materials are provided with a source of energy that is not subsumed in the elastic or dielectric analysis. Materials not usually classified as active also may release such energy during annealing or physical aging or in phase transitions. Such materials are nonconservative with respect to the field variables used (stress and strain for elastic materials, electric field and polarization for dielectrics). The total energy is conserved when we consider all energies.

A. Asymmetric modulus and compliance tensors

As an example of an effect of nonconservative material, consider stress strain relations (see Sec. III), including elasticity.

For materials that are nonconservative with respect to mechanical energy, description by a strain energy function is inapplicable. The modulus tensor is asymmetric with respect to the first and second pairs of indices, $C_{ijkl} \neq C_{klij}$, or for the compliance tensor (the inverse of the modulus tensor),

$$S_{ijkl} \neq S_{klij}. \tag{10}$$

This is the case for viscoelastic materials, in which modulus and compliance depend on frequency (Rogers and Pipkin, 1963; Day, 1971). There is no effect of viscoelasticity on the symmetry of the modulus tensor if the material is isotropic or cubic. If the material has less symmetry (for example, it is orthotropic or axisymmetric), the modulus or compliance can be asymmetric if there is dissipation or gain. Although dissipative materials and materials with gain are called nonconservative, the total energy is conserved. Only the elastic energy is nonconservative.

This sort of asymmetry gives rise to inequalities in Poisson's ratios that would be equal in the perfectly elastic case (Rogers and Pipkin, 1963). Specifically, $S_{2211} = -\nu_{12}/E_1$ and $S_{1122} = -\nu_{21}/E_2$. Suppose that the stiffness values are equal in a transversely isotropic solid: $E_1 = E_2$. The Poisson's ratios are unequal, with an unequal dependence on time or frequency; they can be measured to reveal the asymmetry associated with nonconservative damping or gain. For orthotropic materials the corresponding moduli or compliances as well as the Poisson's ratios need to be measured to obtain all the elastic constants.

The asymmetric compliance matrix for an orthotropic material may be represented as follows for a nonconservative material. The compliance elements are represented in terms of the technical constants the Young's modulus E, the shear modulus G, and the Poisson's ratio ν for physical insight. There are 12 independent compliance elements: three axial compliances $1/E$, three shear compliances $1/G$, and six more related to Poisson's ratio,

$$\mathbf{S} = \begin{pmatrix} \frac{1}{E_1} & \frac{-\nu_{12}}{E_2} & \frac{-\nu_{13}}{E_3} & 0 & 0 & 0 \\ \frac{-\nu_{21}}{E_1} & \frac{1}{E_2} & \frac{-\nu_{23}}{E_3} & 0 & 0 & 0 \\ \frac{-\nu_{31}}{E_1} & \frac{-\nu_{32}}{E_2} & \frac{1}{E_3} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{G_{23}} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{1}{G_{31}} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{G_{12}} \end{pmatrix}. \tag{11}$$

Wood, which is orthotropic, has long been known to exhibit an asymmetric compliance matrix (Neuhaus, 1983; Hering, Keunecke, and Niemz, 2012; Ozyhar, Hering, and Niemz, 2013) by virtue of its viscoelastic response. A similar asymmetry of the modulus and compliance is anticipated if a material is provided with an external power source so that it exhibits gain rather than loss. If the orthotropic material is elastic, there is a conserved strain energy density such that the matrix is symmetric and there are only three independent Poisson's ratios and a total of nine independent elastic constants.

B. Active materials with gain: Negative damping

At times, it has been expedient to hypothesize about abstract systems with active energy input to obtain odd effects as reviewed in Sec. IV.E or non-Hermitian effects as in Sec. IV.F. In contrast to such studies, materials and systems that are experimentally observed to be active or otherwise nonconservative are reviewed here.

1. Biological materials

Living tissues have a metabolic power source. They are the original active materials. Biological materials may in some contexts be interpreted using the continuum concepts that we have discussed. Living cells and tissues including cells in the hearing organ of the inner ear are active materials (Lau *et al.*, 2003). Ear cells can have negative damping (Gold, 1948; Kemp, 1978) and negative stiffness (Martin, Mehta, and Hudspeth, 2000; Hudspeth, 2014). Amplification of signals within the ear allows great sensitivity to sound. Amplification also neutralizes the damping associated with the fluidic environment in the tissue and permits the ear to discriminate frequencies that are close together. Musical perception requires such discrimination. The ear can also emit pure tones.

In muscle, which is active, the force depends on both the stretch and on control via the nerves of the animal or human. During contraction, muscle emits a low frequency sound (Oster and Jaffe, 1980).

2. Designed materials and systems

Among the most familiar human-made systems with gain are lasers, which have been with us for a long time (Maiman, 1961a, 1961b). Lasers provide a medium that amplifies light; the maser is a counterpart that amplifies microwaves (Schawlow and Townes, 1958). There are acoustic counterparts in which ultrasonic waves undergo amplification rather than attenuation. Such amplification occurs in semiconductors provided with an electric current (Hutson, McFee, and White, 1961; White, 1962; Toxen and Tansal, 1963). Sound or ultrasound will be emitted unless there is sufficient damping to compensate. Amplification of sound corresponds to a negative mechanical damping. More broadly, one may consider "excitable media" as materials in which a dissipation of energy is compensated for by a supply of power (Markus, Kloss, and Kusch, 1994).

C. Stabilization via energy flux

1. Stabilization of negative stiffness

To stabilize an object with negative stiffness (see Sec. III.A.2), a source of power rather than an internal energy can be envisaged. Negative spring constants can be stabilized in articulated groups of pipes carrying moving fluid (Thompson, 1982). Because a power source is used to move the fluid, an assumption of constant internal energy is not applicable. The flow of fluid in the pipes is nonequilibrium and irreversible.

2. Stabilization of extreme stiffness

Extremely high stiffness was achieved in composites with negative stiffness inclusions under metastable conditions, as discussed in Sec. III.A.3. Stored energy facilitated the negative stiffness. Stable singular stiffness was reported in a set of hinged pipes carrying moving fluid (Thompson, 1982). Extreme high stiffness is attainable in stable form in active systems with energy flux (Lakes, 2012b). The energy flow was modulated by the applied force in two experimental modalities: (i) electric current into a piezoelectric disk was modulated by contact force on a prismatic contact point and (ii) thermal flow into a chamber of granular bimaterial short helices was modulated by contact force on the granules. All materials exhibit at least one coupled field effect (see Sec. V), so the concept can be generalized.

D. Negative capacitance and permittivity

Electrical capacitance and permittivity are usually considered to be positive. As in elasticity, positive definiteness of the electrical energy implies that the capacitance is positive and that the dielectric permittivity is positive. The energy U stored in a capacitor of capacitance C with applied voltage v is $U = (1/2)Cv^2$.

However, a positive definite energy is an assumption about the system or material. Negative capacitance can occur if an internal source of energy or power is provided. Negative capacitance in feedback systems with amplification has long been known. Negative capacitance has been used to neutralize some of the positive capacitance of transducers (Forward, 1979) in order to improve their damping properties (Fukada *et al.*, 2004). A discrete feedback amplifier that provides negative capacitance is illustrated in Fig. 8. The capacitance at the input is $C = -C_0(R_2/R_1)$, with C_0 the feedback capacitance and R_1 and R_2 the resistances. The resistor R_0 serves to control the frequency response for the intended purpose of controlling damping of piezoelectric polymers. The system is stable provided that the absolute value of C is less than that of the source capacitance connected to the input (Fukada *et al.*, 2004). The amplifier is an active circuit that must be suppled with power from an external source, as indicated by the ±15 volts in Fig. 8.

Negative incremental capacitance was predicted to occur in ferroelectric materials in accordance with the Landau theory of phase transformations (Landau and Khalatnikov, 1954). Negative capacitance was inferred in ferroelectric devices (Salvatore, Rusu, and Ionescu, 2012) and in ferroelectrics

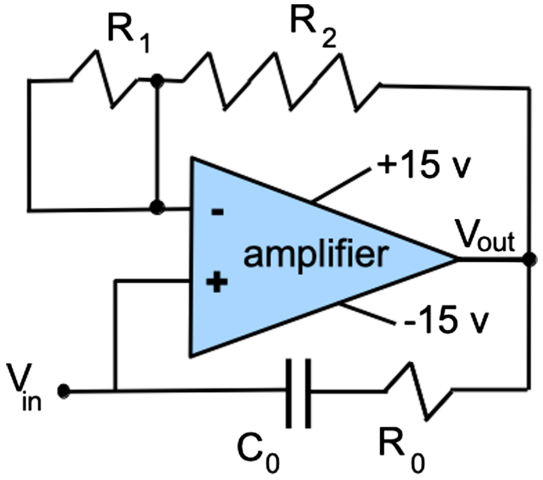

FIG. 8. Negative capacitance via a feedback amplifier.

(Appleby *et al.*, 2014). Negative capacitance was used in superlattices to achieve extremely large energy storage density combined with high power density in microcapacitors (Cheema *et al.*, 2024).

The refractive index depends on the dielectric permittivity and the magnetic permeability of the material. Negative refractive index has been achieved via resonating constituents in materials, as discussed in Sec. VII.

E. Odd material property tensors

Odd material property tensors are asymmetric. Tensors describing dielectric permittivity are symmetric if there is a conserved energy density function (Nye, 1976). Tensors describing coupled transport processes such as flow of heat and electricity are symmetric if the system obeys time-reversal invariance (Onsager, 1931a, 1931b). Materials and systems that do not obey these assumptions may exhibit asymmetric tensor properties.

Odd viscosity refers to an antisymmetric part of the viscosity tensor that is allowable in systems that are not invariant to time reversal (Avron, 1998). The Onsager relations imply that the symmetric part of the fourth rank viscosity tensor is even under time reversal and that the asymmetric part is odd (Onsager, 1931a, 1931b). To have nonzero odd terms, time-reversal invariance must be broken.

Avron (1998) demonstrated that odd effects cannot occur in 3D isotropic viscous or elastic materials but that they are possible in 2D isotropy. An analysis was adduced of possible odd effects in a quantum Hall fluid and in superfluid He^3. Moreover, experiments were suggested to demonstrate the effects. For example, a rotating object in a fluid experiences drag from the usual viscosity and normal pressure from the odd terms (if any). Asymmetry of the viscosity or modulus tensor can be an aspect of nonconservative systems, including those with a power source.

If internal degrees of freedom are excited by an external power source, time-reversal symmetry is broken. An antisymmetric diffusivity, denoted odd, emerges in a chiral random walk model in an active medium with an external supply of energy (Hargus, Epstein, and Mandadapu, 2021). The diffusivity tensor for such materials contains both a symmetric and an antisymmetric component. Molecular dynamics simulations of chiral active matter indicate that such effects can occur. Specifically, a directionally isotropic chiral active fluid composed of torqued dumbbells was considered. Since there is no rank-2 tensor in three dimensions that is both isotropic and antisymmetric, attention was given to two-dimensional diffusion. In a related study, shear and odd viscosity values were inferred in a model system consisting of actively torqued dumbbells using molecular dynamics simulations (Hargus *et al.*, 2020).

Odd elasticity refers to asymmetry of the modulus or compliance tensor and the appearance of additional material constants. As with odd viscosity, such asymmetry is permissible in nonconservative systems and materials.

Asymmetry in the compliance tensor [Eq. (10)] has been observed experimentally and has long been known in wood, which is orthotropic and viscoelastic. All 12 compliance components have been determined experimentally versus time for various woods (Neuhaus, 1983; Hering, Keunecke, and Niemz, 2012; Ozyhar, Hering, and Niemz, 2013). The substantially different time dependence of the nondiagonal elements of the compliance matrix [Eq. (11)] illustrates the odd nature of the viscoelastic character of wood though the terminology *odd* was not used. There are 12 independent compliances, in contrast to nine for orthotropic elastic solids governed by a conserved energy density.

A conceptual model of odd elasticity was presented with an analysis of hypothetical active solids as lattices with rib elements supplied with external power (Scheibner *et al.*, 2020). It was suggested that the dynamical matrix is non-Hermitian; see also Sec. IV.F.

The origin of additional stiffness constants in active solids as well as in dissipative solids differs from that of generalized continuum theories such as Cosserat elasticity, discussed in Sec. III.B.2, in which an internal energy function is assumed to exist. Asymmetry of the modulus or compliance tensor, if there is any, is independent of the asymmetry of the stress tensor.

In contrast to viscoelastic materials that dissipate energy, a source of energy is assumed to be provided in active materials. There is a hysteresis loop in cause versus effect for both viscoelastic materials that exhibit positive damping and materials with a power source that exhibit negative damping. The stress depends on the path of deformation for both dissipative and active materials. The hysteresis, whether dissipative or active, can be expressed as the imaginary part of a complex modulus. Because physical property functions are expected to be analytic, there must be frequency dependence of modulus via the Kramers-Kronig relations. The frequency dependence can be tuned for both kinds of materials.

Because elasticity by definition entails independence of the path of deformation, odd elasticity is really not elasticity. It nonetheless involves an asymmetry of the modulus tensor with respect to pairs of indices. Viscoelastic damping also suffices to provide an asymmetric modulus or compliance tensor, as has been observed.

The analysis of Rogers and Pipkin (1963) for asymmetry of the modulus in nonconservative materials does not distinguish

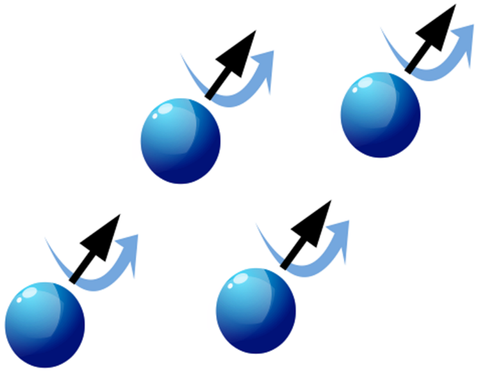

FIG. 9. Model system of fluid with odd viscosity based on inclusions given angular momentum via an external field. Adapted from Markovich and Lubensky, 2021.

gain from loss. Therefore, asymmetry of the modulus tensor in odd elasticity (or of the viscosity tensor) will not occur in isotropic or cubic materials. Because it is a tensor property, it is restricted to materials of axisymmetric or lower symmetry (Rogers and Pipkin, 1963; Avron, 1998). In that vein, isotropic examples include moments as well as forces (Scheibner *et al.*, 2020). Distributed moments are absent in classical elasticity even when the modulus is asymmetric owing to the absence of an energy function. In classical elasticity the stress is symmetric even if the modulus or compliance has odd components. Distributed moments and associated asymmetric stresses can occur in magnetic materials and in a variety of Cosserat solids, but not in classically elastic solids. In active materials the moments are nonetheless in contrast to the usual Cosserat elasticity in which an internal energy function is assumed to exist.

In a two-dimensional model material, Lin *et al.* (2023) showed that the odd moduli are proportional to the nonequilibrium force and dissipation coefficients. Results were interpreted in the context of the Onsager principle and nonreciprocity. Either positive or negative values of a driving torque variable will suffice to break time-reversal symmetry and allow odd moduli. A microscopic Hamiltonian theory, valid in both two and three dimensions (Markovich and Lubensky, 2021), showed that odd viscosity is present in any system with globally or locally aligned spinning components. Such a system is illustrated in Fig. 9. No dissipation is required to obtain the odd terms, but a material provided with external torques is active.

F. Non-Hermitian systems and materials

Non-Hermitian Hamiltonians were originally introduced in an abstract sense in quantum mechanics (Bender and Boettcher, 1998). A material describable by this sort of Hamiltonian can exhibit a phase transformation as it is tuned. A Hermitian matrix has the property that the transposed complex conjugate matrix is equal to the original matrix. In the integral representation of an operator F upon a function ψ, the operator is Hermitian if $\int (F\psi)^* \psi d\tau = \int \psi^* F \psi d\tau$.

It is believed that the Hamiltonian must be Hermitian in order to ensure that the energy spectrum of the eigenvalues of the Hamiltonian is real and that probability is conserved in time. This assumption is sufficient to guarantee these desired properties. It has been argued that it is not a necessary condition (Bender, Brody, and Jones, 2003) and that the weaker condition of parity-time ($\mathcal{PT}$) invariance is the actual necessary condition for a valid quantum mechanical theory. Parity refers to invariance under inversion of position and momentum. An open system or material with externally supplied energy does not necessarily entail proper non-Hermitian $\mathcal{PT}$ invariance.

To realize non-Hermitian materials physically, optical systems in one and two dimensions, with regions of gain and loss, that are describable by Schrödinger-like equations were analyzed (Makris *et al.*, 2008). To achieve gain, one requires a power source. Non-Hermitian materials can also be realized in systems that contain parts with different dissipation but no gain or amplification.

Eigenvalues as well as the corresponding modes of the system coalesce as a tuning parameter is varied. The coalescence of eigenvalues, which may correspond to frequencies, is called an exceptional point. Such behavior can be exploited in devices. A discrete non-Hermitian accelerometer with an improved signal-to-noise performance was developed and tested; eigenvalues and their corresponding eigenvectors coalesced (Kononchuk *et al.*, 2022). This device contains two coupled resonators with gain, loss, inductance, and capacitance balanced to achieve the required $\mathcal{PT}$ symmetry. Enhancement of actuation force in a discrete device was attained using a designed non-Hermitian system containing two coupled tuned mechanical resonators (Gupta *et al.*, 2023). A natural frequency splits into two frequencies as coupling is tuned.

G. Active chirality

As we saw in Sec. III.B.3, piezoelectricity, in which property coefficients comprise a third rank tensor, requires chirality, but classical elasticity in which the modulus or compliance is fourth rank cannot accommodate chirality. Even if the modulus or compliance is asymmetric with respect to pairs of indices ($S_{ijkl} \neq S_{klij}$), as in nonconservative materials, it is still fourth rank and cannot accommodate chirality. Chiral elastic effects do occur; they require a theory with more freedom than classical elasticity to be understood.

Because chirality entails noninvariance to inversion of all three coordinates, chiral materials are of interest in the context of non-Hermitian materials and systems. In addition, biological materials and systems are both active and chiral, so efforts to imitate them often make use of chirality.

A chiral granular material comprised of chiral wires (see Fig. 4) was made and studied (Tsai *et al.*, 2005). The granules rotated in a preferred direction when excited by vertical vibration. This is one of the few materials that allow angular momentum to be transmitted to the bulk of the material. Others include ferrofluids and liquid crystals subjected to a rotating magnetic field.

Groups of thousands of spinning and swimming starfish embryos spontaneously assemble into ordered patterns (Tan *et al.*, 2022). The organization is called a chiral crystal. The pattern can persist for tens of hours. Control is via the embryos' internal development. The embryos are chiral and spin about an axis. Self-sustained oscillations were observed. It was suggested that observed lattice defects and self-sustaining waves provide evidence of odd effects. It was pointed out that systems with nonreciprocal interactions allow energy to be extracted from quasistatic cycles.

A two-dimensional chiral liquid consisting of spinning colloidal magnets was made by Soni *et al.* (2019). When excited by an external magnetic field, surface flows suggest odd or Hall viscosity, which has been analyzed but has been experimentally elusive. In another ferromagnetic colloid, coarsening and phase separation were observed to be dependent on the frequency of the driving magnetic field (Massana-Cid *et al.*, 2021).

V. COUPLED FIELDS

Most materials exhibit coupling between multiple field variables. Coupling can occur between mechanical, electrical, thermal, magnetic, and humidity variables, depending on the material. Coupled fields are of interest in the context of basic science and for their role in sensors, actuators, and energy conversion devices. Equilibrium thermodynamic analysis is appropriate for thermal expansion, piezoelectricity, and pyroelectricity when studied under ideal conditions (Nye, 1976).

In actual materials coupled fields give rise to nonequilibrium viscoelastic dissipation. Thermoelastic coupling causes temperature variation from deformation. Heat flows between the object and the environment or between crystals and other heterogeneities in the material, dissipating energy (Zener, 1948; Nowick and Berry, 1972). In piezoelectric materials electric conduction dissipates energy from deformation induced polarization. Deformation also influences the diffusion of atoms. Coupling with the chemical potential has been predicted (Larché and Cahn, 1985) and observed (Shi, Markmann, and Weissmüller, 2018).

Energy relations between dissimilar atoms in alloys can give rise to uphill diffusion such as that occurring in spinodal decomposition (Krishna, 2015). For example, fluctuations in composition in an initial single-phase state of an alloy such as ZnAl grow to form regions of segregated phases (Mainville *et al.*, 1997). Such materials exhibit viscoelasticity (Nowick, 1951). Alloy from rapidly cooled melt retains the liquid's mix of atoms. This solid is metastable. Warming the solid triggers segregation of Zn-rich and Al-rich phases while releasing heat. Internal and external coupled field variables can give rise to dissipative response or metastability.

A. Symmetry of even-rank tensors

The thermal expansion tensor is symmetric because the strain is symmetric by definition. Symmetry of even-rank tensors that govern permittivity, elasticity, and magnetic permeability follows from the assumption of a conserved energy density that pertains to reversible processes between equilibrium states (Nye, 1976). As we have seen, materials with stored energy and materials that dissipate energy are not describable by a conserved energy density.

Conduction of heat or electricity and other transport processes require nonequilibrium thermodynamics. Tensors that govern coupled processes such as heat and electrical flux obey reciprocity via the Onsager relations (Onsager, 1931a, 1931b) owing to time-reversal symmetry. The Onsager relations also require transport coefficient tensors such as thermal conductivity and diffusion to be symmetric, but the inference is more indirect (Casimir, 1945; Nye, 1976). The system or material may in fact not obey time-reversal symmetry as in active materials, materials exhibiting the Faraday effect, and other materials and systems.

B. Thermal expansion

Materials ordinarily expand when heated. The second rank thermal expansion tensor is the ratio of expansion strain to temperature change (Nye, 1976). The physical cause is a slight anharmonicity or nonlinearity in the force between atoms in the material. Contraction of a material on heating seems counterintuitive because one might expect atoms to occupy more space as their vibration amplitudes increase with heating (Barrera *et al.*, 2005). That view is sensible in the context of an analysis of the effect of vibration on the mean interatomic bond length in the presence of a slight nonlinearity. Negative thermal expansion, though counterintuitive, is allowable within thermodynamic bounds.

Exceptions with negative expansion are known, such as zirconium tungstate (Mary *et al.*, 1996), in which the unit cell has a complex structure. This material is structurally cubic and has an isotropic thermal expansion tensor. Negative expansion occurs in other crystalline solids (Evans, 1999; Barrera *et al.*, 2005), including those with a complex unit cell. Further materials include Si and Ge and other tetrahedrally bonded crystals at low temperatures, β quartz at high temperatures, and some ceramics and zeolites with framework structures. Negative expansion can occur near phase transitions (Evans, 1999). For example, negative expansion over a wide range of temperature occurs in the ferroelectric lead titanate $\mathrm{PbTiO_3}$ and attains a peak of large magnitude near the phase transition temperature (Mikhaleva *et al.*, 2012). Phase transitions also allow negative stiffness (see Sec. III.A.2), and other negative properties not permissible in standard thermodynamic analyses.

In some materials relative motion orthogonal to the interatomic bonds considered can cause thermal contraction. Thermal expansion can be anisotropic in some single crystals and composites. For example, single crystals of $\mathrm{Ag_3Co(CN)_6}$ and $\mathrm{Ag_3Fe(CN)_6}$ have a large positive expansion in one direction and a large magnitude of negative expansion in another direction (Goodwin and Kepert, 2005).

As for composite materials, classical bounds on the expansion of a composite have been derived (Cribb, 1968). The theoretical expansion can be no higher than that of the constituent with the highest expansion and no lower than that of the constituent with the lowest expansion. It is assumed that the constituents have a perfect fit. It is tacitly assumed that the system is initially in a state of minimum energy.

These bounds for composites can be exceeded if either assumption is relaxed. Expansion can be arbitrarily large positive or negative (Lakes, 1996), tunable by geometry in rib lattices that contain ribs with two materials with different expansions (Lakes, 2007). In contrast to the assumptions made in the bounds, the constituents are not bonded on all

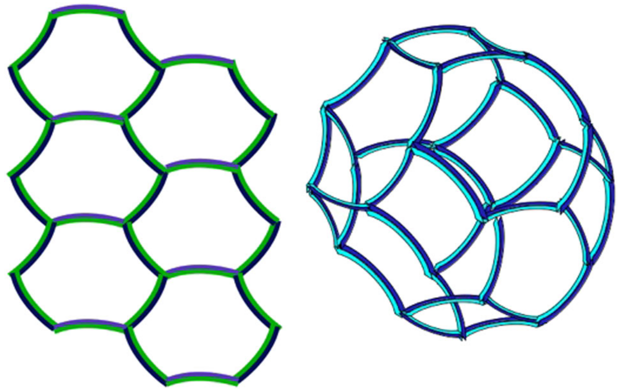

FIG. 10. Lattices with bimaterial ribs for controllable thermal expansion or controllable piezoelectric sensitivity. Left sketch: 2D hexagonal lattice. Right sketch: 3D lattice cell. Adapted from Lakes, 2007.

surfaces. No special chemical composition is required, just a dissimilarity in expansion of the two layers in each rib. The lattices can be dense in two or three dimensions. Rib lattices with bimaterial ribs are illustrated in Fig. 10.

Lattices can be made with multiple extreme properties. For example, one can obtain a controllable extreme thermal expansion as well as a negative Poisson's ratio, as illustrated in Fig. 11. This lattice also exhibits Cosserat elastic properties and hence provides a reduction in stress concentration around holes and other defects. Materials with multiple designed properties are called multifunctional.

Zero expansion is of considerable practical interest. For such purposes some alloys, such as Invar and glasses, are known with near zero expansion. The tunable rib lattices can be designed with zero expansion, but they are compliant. Rib lattices with zero expansion incorporating cells with two kinds of ribs, each of which is made with a different material, have been developed (Steeves *et al.*, 2007). These lattices offer improved stiffness combined with near zero expansion. In composites with particulate inclusions, extreme positive or negative thermal expansions can be achieved provided that the inclusions have negative stiffness, which is possible if there is stored energy (Wang and Lakes, 2001) or a power source.

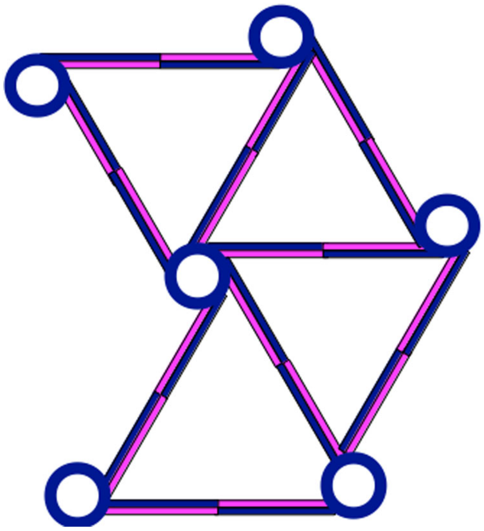

FIG. 11. Lattice with bimaterial ribs for controllable thermal expansion or controllable piezoelectric sensitivity. A chiral lattice that also has a Poisson's ratio −1 (Prall and Lakes, 1997) and is Cosserat elastic (Spadoni and Ruzzene, 2012) is displayed.

C. Piezoelectricity and pyroelectricity

Piezoelectric materials generate an electric polarization when subjected to a mechanical stress and undergo a strain (deformation per length) when subjected to an electric field (Nye, 1976). The effect is due to an asymmetric distribution of electric charge in the material.

In piezoelectricity the direct piezoelectric effect represents sensitivity to stress σ_{ij}, giving rise to an electric displacement vector $\mathcal{D}_i$ and hence a charge density via the third rank direct effect sensitivity tensor d^d_{ijk} (in pC/N); see Eq. (12). The Einstein summation convention for repeated indices is used. The electric displacement is also related to the electric field $\mathcal{E}_j$ via the dielectric tensor K_{ij}. The converse piezoelectric effect sensitivity is the ratio of strain ϵ_{ij} to the imposed electric field $\mathcal{E}_k$, or the ratio of deformation displacement to imposed voltage, in pm/V via Eq. (13),

$$\mathcal{D}_i = d^d_{ijk}\sigma_{jk} + K_{ij}\mathcal{E}_j + p^{\text{pyr}}_i \Delta T, \tag{12}$$

$$\epsilon_{ij} = S^{T,\mathcal{E}}_{ijkl}\sigma_{kl} + d^c_{kij}\mathcal{E}_k + \alpha^{\text{exp}}_{ij}\Delta T, \tag{13}$$

$$\Delta\mathcal{S} = \alpha^{\text{pzcal}}_{ij}\sigma_{ij} + p^{\text{electr}}_i\mathcal{E}_i + (C^{\sigma,\mathcal{E}}/T)\Delta T, \tag{14}$$

in which α^{exp}_{ij} is the thermal expansion, $\alpha^{\text{pzcal}}_{ij}$ is the piezocaloric effect, and p^{electr}_i is the electrocaloric effect.

For some materials the electric displacement also depends on the temperature change ΔT via the pyroelectric effect p^{pyr}_i in Eq. (12). The pyroelectric effect was known to the ancient Greeks. Heated rocks containing tourmaline crystals were observed to attract dust and other debris.

The strain ϵ_{ij} also depends upon the stress σ_{kl} via the elastic compliance $S^{T,\mathcal{E}}_{ijkl}$ at constant temperature T and electric field $\mathcal{E}$, and upon the temperature change ΔT via the thermal expansion α^{exp}_{ij} (Nye, 1976). The heat capacity per volume at constant stress and electric field $\mathcal{E}$ is $C^{\sigma,\mathcal{E}}$ in Eq. (14), with $\Delta\mathcal{S}$ the entropy change.

Bounds on properties of piezoelectric composites are known, but they can be exceeded in these materials, which do not obey the assumptions made in the bound analysis (Bisegna and Luciano, 1996). For example, giant piezoelectric effects, as with giant thermal expansion, can be obtained in lattices of bimaterial strips (Lakes, 2014) and in composites with inclusions of negative stiffness (Wang and Lakes, 2001). Large piezoelectric effects can also be obtained in structures with discrete negative stiffness elements (Kalathur and Lakes, 2016). Lattices as well as structures with flexible bender elements are, however, far more compliant than the usual piezoelectric ceramics. A giant piezoelectric response was obtained in systems provided with an external power source (Lakes, 2012a). In this experiment flow of heat was modulated by variable contact in a granular layer. By this process electric polarization of pyroelectric origin contributes to stress-generated electric polarization and hence a piezoelectric response. It has long been known that some piezoelectric materials are sensitive to the gradient of stress, as well as to stress itself (Bursian and Zaikovskii, 1968; Bursian and Trunov, 1974),

$$\mathcal{D}_i = d^d_{ijk}\sigma_{jk} + d_{ijkl}\frac{\partial \sigma_{jk}}{\partial x_l}. \tag{15}$$

These effects involve a fourth rank coupling tensor term d_{ijkl} as in Eq. (15), in addition to the previously considered effect of a tensor of third rank. The effects can be substantial in ferroelectric materials and in materials near a phase transition. In recent parlance materials exhibiting gradient piezoelectricity have been called flexoelectric. This class of materials is of interest in the context of the prediction of large effects in BN sheets (Naumov, Bratkovsky, and Ranjan, 2009). Potential applications in nanogenerators could generate power from ambient disturbances. Gradient sensitivity in piezoelectricity is related to gradient sensitivity in generalized elasticity such as Cosserat elasticity, which is discussed in Sec. III.B.2. In both cases there is a characteristic length in the continuum theory associated with a physical length scale in the microstructure.

D. Elastocaloric, barocaloric, and electrocaloric effects

The elastocaloric effect (or piezocaloric effect) is the coupling between stress and entropy via $\alpha^{\text{pzcal}}_{ij}$ in Eq. (14). It is the converse of thermal expansion. If the stress is applied sufficiently rapidly, the temperature changes. Similarly, the barocaloric effect is coupling between pressure and entropy. The electrocaloric effect gives rise to an entropy change from an electric field via p^{electr}_i in Eq. (14). It is the converse of the pyroelectric effect. These effects have long been known. Recently, there have been many studies of these effects motivated by their potential in solid-state refrigeration. Large values for these quantities are found when the material is in the vicinity of a phase transition (Mañosa, Planes, and Acet, 2013). Materials include ferroelectrics such as $BaTiO_3$, LaFeSi compounds, and shape memory alloys. Giant electrocaloric effects were observed in thin film lead zirconate titanate $PbZr_{0.95}Ti_{0.05}O_3$ at temperatures near the Curie phase transition (Mischenko *et al.*, 2006). Although properties can be extremely high, they do not directly violate any thermodynamic bounds.

E. Hall effect

Apply an electric current through a conductor. Then impose a magnetic field orthogonal to the current. A transverse voltage appears across the conductor orthogonal to both the current and the magnetic field. That is the Hall effect (Hall, 1879). It is caused by deflection of the charge carriers in the current by the magnetic field. The Hall effect may be represented via an asymmetric second rank conductivity tensor. The sign of the transverse voltage allows inference of the sign of the charge carrier. The magnitude of the voltage determines the number n of charge carriers of charge q per volume. The Hall coefficient is $R_H = \mathcal{E}_y / j_x B_z$, in which the current density is j, the applied magnetic field is B, and the resulting transverse electric field is $\mathcal{E}_y$ (Lovett, 1989; Preston and Dietz, 1991). The Hall effect is first order in magnetic field; the magnetoresistance is second order (Lovett, 1989). The Hall coefficient may be written as $R_H = 1/nq$. This is an exact result with no apparent freedom. However, it is assumed that the material is homogeneous.

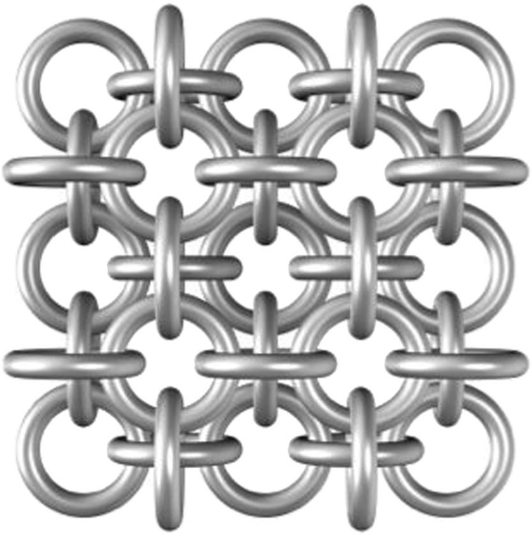

FIG. 12. Chain mail structure for control of the Hall effect. From Whyte, 1998.

If that assumption is relaxed, it is possible to envisage heterogeneous structures in which the sign of the Hall coefficient is reversed in two dimensions (Briane, Manceau, and Milton, 2008) and three dimensions (Briane and Milton, 2009b). The structures consist of interlocking rings, as illustrated in Fig. 12. They resemble chain mail used in ancient armor.

Giant Hall effects of unbounded magnitude are possible in hierarchical laminated composites with multiple length scales (Briane and Milton, 2009a). The laminate in two dimensions consists of columnar elements that act as batteries in series. The laminate in three dimensions is of rank 3 with three length scales, and it can provide isotropic properties. If one phase tends to zero conductivity, it becomes void space. The rank-3 laminate is based on a laminate used in the analysis of bounds on the conductivity of polycrystalline materials (Schulgasser, 1977). Laminates that themselves contain laminates were originally conceptualized by Maxwell (1873) for an analysis of conductivity. Such a laminate is illustrated in Fig. 13.

F. Faraday effect

The Faraday effect is the rotation of linearly polarized light in the plane of polarization due to the action of an external magnetic field on the material through which the light passes. The effect can be interpreted via coupling of electrical permittivity to a magnetic field in materials. This magneto-optic effect can be incorporated into an asymmetric second

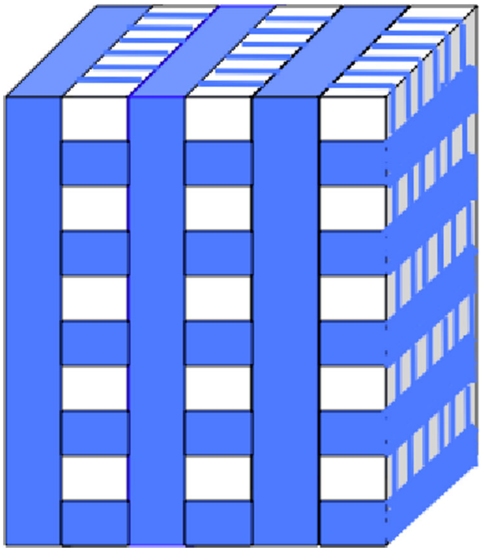

FIG. 13. Hierarchical laminate after Maxwell (1873).

rank permittivity tensor (Pershan, 1967) with complex off-diagonal terms [Eq. (16)] proportional to the applied magnetic field,

$$\epsilon_{ij} = \begin{pmatrix} \epsilon_1 & -i\epsilon_g & 0 \\ i\epsilon_g & \epsilon_1 & 0 \\ 0 & 0 & \epsilon_1 \end{pmatrix}. \tag{16}$$

The magnetic field provides a violation of time-reversal invariance such that the asymmetry in the properties can occur (Onsager, 1931a, 1931b). The materials are called gyrotropic.

As for designed heterogeneous materials, magneto-optic effects can be obtained via an array of toroid coils (Marinov and Fedotov, 2023). Gyrotropic materials are discussed further in Sec. VI.C.

VI. RECIPROCITY AND NONRECIPROCAL MATERIALS

There are several kinds of reciprocity. For waves reciprocity states that the frequency response functions between any two points remain the same after source and receiver are exchanged. Reciprocity in acoustics can be traced to a 1878 treatise on sound by Rayleigh. If sound waves are generated at a point A, the resulting velocity potential at another point B is the same in both magnitude and phase, as it would have been at A had B been the source of sound (Potton, 2004). More simply stated, if I can hear you, then you can hear me. Demonstration of reciprocity makes use of assumptions of linearity and time-reversal invariance.

Nonreciprocal materials and systems can be developed if one introduces chirality or an asymmetric dielectric tensor as in the Faraday effect (Potton, 2004), or nonlinearity or time dependent constitutive properties (Nassar *et al.*, 2020). A one-way mirror may come to mind in this context. Actually, such mirrors are not truly one-way. They are partially reflective mirrors that are fully reciprocal and have equal partial reflectivity in both directions. They appear to be one-way because they are set up with one room brightly illuminated and the observer situated behind the mirror in another room that is darkened. Some aspects of reciprocity are now discussed.

A. Reciprocity in coupled fields

Reciprocity in coupled field effects such as thermal expansion and piezoelectricity refers to equality between coefficients that describe different phenomena (Nye, 1976). For example, the coefficient of thermal expansion α_{ij}^{exp}, which quantifies the ratio of expansion strain to applied temperature change in Eq. (13), and the coefficient $\alpha_{ij}^{\text{pzcal}}$ of piezocaloric effect in Eq. (14), which expresses the ratio of entropy change to applied stress, refer to different phenomena. The same symbol α_{ij} is usually used for both because of the assumption of reciprocity. The piezocaloric effect may be demonstrated by rapidly (adiabatically, $\Delta\mathcal{S} = 0$) applying a stress to a material. Before there has been time for heat flow to occur, one observes that the material has changed temperature in response. Reciprocity means the coefficients for these different phenomena are the same.

For piezoelectric materials reciprocity refers to equality between the coefficients for the direct effect [Eq. (12)] and those for the converse effect [Eq. (13)]: $d^c_{kij} = d^d_{kij}$. Reciprocity of piezoelectric coefficients is so universally accepted that the same symbol d is used for both sensitivity tensors. Reciprocity is shown (Nye, 1976) using the assumptions of equilibrium and the existence of an energy function.

Assumption of a conserved function of energy is inapplicable in viscoelastic materials that dissipate energy or in active materials for which a power source is provided. Some piezoelectric materials, such as quartz, exhibit minimal damping, but others, such as lead metaniobate, exhibit substantial damping.

If an external source of energy flux is provided, large nonreciprocal effects in piezoelectricity occur (Faust and Lakes, 2015). In this experiment thermal flux was modulated by the applied force. Nonreciprocal effects gave rise to an enhancement of factor of 5 in sensitivity.

Energy flux entails a nonequilibrium condition, necessitating nonequilibrium thermodynamics, including the Onsager relations, for analysis. The Onsager reciprocity relations stem from the principle of microscopic reversibility (Onsager, 1931a, 1931b). An example is given of coupled flow of heat and electricity in which cross terms that represent thermoelectricity obey reciprocity relations. It is acknowledged that microscopic reversibility is less general than the fundamental laws of thermodynamics. The principle of dynamical reversibility does not apply in the presence of external magnetic fields or Coriolis forces. The Onsager reciprocal relations then break down.

B. Reciprocity in elasticity

In elasticity, we have the Maxwell-Betti reciprocity relation (Maxwell, 1864) (Betti, 1872). Force applied to the elastic solid at point a is called F_a, and u_{ba} is the displacement at point a caused by a force at point b. Force applied to the

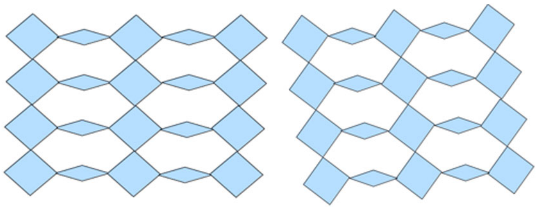

FIG. 14. Nonreciprocal lattice based on nonlinear deformation. Left image: undeformed. Right image: deformed. Adapted from Coulais, Sounas, and Alu, 2017.

elastic solid at point b is F_b, and u_{ab} is the displacement at point b caused by a force at point a. The reciprocity condition is $F_a u_{ba} = F_b u_{ab}$. This form of reciprocity can be regarded as intuitive: if one presses an object on side A, the other side B moves a certain amount. One expects that if one presses side B with the same force, then side A will move the same amount (Coulais, Sounas, and Alu, 2017). Elastic reciprocity is highly pertinent in the analysis and design of many mechanical systems. The assumptions required to demonstrate elastic reciprocity include time-reversal invariance and reversibility. Linear elasticity is assumed. Elasticity entails independence of time. If one assumes linearity, energy arguments can be used to demonstrate reciprocity (Sokolnikoff, 1946).

Nonreciprocal effects occur in lattice materials of ribs or of connected polygon nodes that are in a nonlinear regime on the threshold of instability (Coulais, Sounas, and Alu, 2017). Near an instability there will be irreversibility and nonlinear hysteresis, so nonreciprocal effects are admitted. The lattice, which resembles tilting square lattices with negative Poisson's ratio, is illustrated in Fig. 14. By analogy with electromagnetic nonreciprocal devices such as isolators and circulators (which depend on resonance), static mechanical nonreciprocity may form the basis for future devices (Potton, 2004).

A composite hydrogel was reported to have mechanical nonreciprocity owing to direction dependent buckling of nanofillers made of graphene oxide sheets (Wang *et al.*, 2023). The sheets were unidirectionally oriented in a direction to encourage buckling. The material, via its asymmetric deformation, had the ability to induce the directional transport of a variety of objects including small worms.

C. Gyrators and gyrotropic materials

The earliest gyrotropic systems were discrete electric circuit elements (Tellegen, 1948). The gyrator gyrates a current i into a voltage v, and vice versa. One can make a gyrator by converting electrical oscillations to mechanical and back or using ferromagnetic materials. For an electrical transformer, $i_1 = -ui_2$ and $v_2 = uv_1$, but for a gyrator $v_1 = -si_2$ and $v_2 = si_1$, with u and s constants representing the system. Governing equations for gyrators resemble those for mechanical gyroscopes. Ferromagnetic materials have been used to make gyrators (Hogan, 1952). A gyrator can also be realized by means of mechanically coupled piezoelectric and electromagnetic transducers, by means of the Hall effect in a square plate of bismuth, or via electrical-electrical coupling through a gyroscopic link. The gyrator violates reciprocity.

It has long been known that the Faraday rotation of the plane of polarization in optics, which is represented by off-diagonal terms in the permittivity, is antireciprocal. The off-diagonal terms in the permittivity tensor are small in comparison with the diagonal terms in most materials. Recently, efforts have been made to design materials that exhibit stronger effects. For example, heterogeneous gyrotropic structures were designed to enhance the effective nonreciprocal behavior by reducing the contribution of the diagonal susceptibilities using negative dielectric effects, which are possible, as discussed in Sec. IV.D. The suggested implementation is in the midinfrared region (Katsantonis *et al.*, 2023). A microwave gyrotropic lattice made of circuit elements was presented by Wang *et al.* (2012). It does not require an external magnetic bias. Strong Faraday-like effects have been observed in simulations and experiments. The effective medium was nonreciprocal.

D. Toroids

Toroidal windings of wire, which are illustrated in Fig. 15, have long been used in electronics. They confine the magnetic field resulting from current in the winding wire to the interior of the torus and provide electromagnetic inductance. The inductance can be increased by providing a magnetic core within the toroid. Time varying current in a toroidal winding does generate an electric field outside the torus via Maxwell's equations.

Electromagnetic fields associated with a wire helix (solenoid) bent in a ring (toroid) were considered as a classical analog to an "anapole" interaction that violates parity conservation in particle physics (Zel'Dovich, 1958). *Anapole* refers to zero pole or without poles because the interaction is not present in standard multipole expansions and cannot be represented as a superposition of multipole terms. An anapole moment in the nucleus of cesium leads to parity nonconservation that was recognized in the context of electroweak field unification (Wood *et al.*, 1997). Parity nonconservation entails the absence of mirror reflection symmetry; it is displayed by objects with left- or right-handedness. Parity refers to invariance under an inversion of position and momentum. Optical spectroscopy experiments were done to reveal the effect in cesium atoms.

Toroids have been of recent interest by virtue of their unusual electromagnetic properties. For example, the strength of their interaction with electric and magnetic fields depends on the time derivatives of the fields rather than the field amplitude alone. Toroidal multipoles are not incorporated into

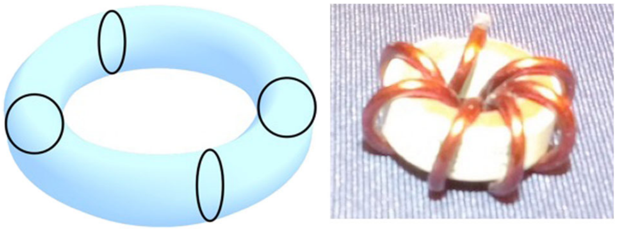

FIG. 15. Left image: toroid with wire loops. Right image: physical toroid with core.

the usual multipole expansion. Solid materials that contain toroidal magnetic order have been called ferrotoroids. Piezoelectric materials do not obey spatial inversion symmetry, and ferromagnetic materials do not obey time-reversal symmetry; materials with magnetic toroidal order disobey both time-reversal and spatial inversion symmetry (Papasimakis *et al.*, 2016). Toroidal domains were observed in an olivine crystal and have been explored in other materials.

Recently, it has been recognized that interactions between toroidal currents could violate reciprocity. An experimental study of optical activity in a chiral planar array of toroid coils disclosed resonant effects at microwave frequencies and off-diagonal gyrotropic terms in the permittivity (Papasimakis *et al.*, 2009). An array of toroid coils was analytically found to behave as a reciprocal gyrotropic birefringent effective medium (Marinov and Fedotov, 2023). Toroids in the subwavelength regime exhibit a gyrotropic response resembling optical activity. The winding of the wire around the toroid can be done with a spiral character, but it need not be done that way. The winding can be also done as an array of current loops or as two spirals with the opposite sense. Chiral (unbiased) gyrotropy is time-reversal symmetric and is therefore reciprocal (Caloz *et al.*, 2018).

With toroids, optical activity can be achieved in which the plane of polarization is rotated. Analysis has shown that negative indices of refraction can be achieved in arrays of toroids in a dielectric matrix (Marinov *et al.*, 2007). In a study of toroidal response (Kaelberer *et al.*, 2010), the experimental configuration made use of a 22-by-22 array of elements consisting of four rectangular, electrically disconnected metal wire loops embedded in a dielectric slab. The rationale compared with an actual toroid was to avoid a dipole moment from helical windings. Toroid winding could have been made to avoid that with windings of opposite helicity. Toroidal resonant effects were observed at microwave frequencies of 14–17 GHz.

VII. RESONANCE AND WAVES

The simplest example of a negative property due to dynamic effects is a discrete single degree of freedom system with an inertia term and a restoring force term. Such systems are studied in the context of differential equations. Physical examples include a discrete mechanical system with a spring, a mass, and a damper and a discrete electrical system with a capacitor, an inductor, and a resistor. The phase angle between cause and effect at frequencies above resonance approaches 180°, which corresponds to a negative ratio between effect and cause. At resonance the system is compliant and the phase angle is 90°. The compliance (effect divided by cause) at resonance increases as the dissipation is reduced. Systems with multiple degrees of freedom can exhibit antiresonances corresponding to small values of compliance. Resonance at acoustic and ultrasonic frequencies requires structure at scales much larger than the atomic or molecular.

Negative properties have been reported in electromagnetic materials that exhibit local resonances. Such properties have been used to make lenses with unusual refractive properties including negative refraction. Predictions based on a quasistatic nonresonant condition do not apply.

A. Negative elastic properties

If a composite is envisaged with an array of mass-spring resonators in cavities, effective mass density can be negative (Milton and Willis, 2007). A negative effective elastic modulus is also possible in composites with resonating constituents such as spherical inclusions containing a dense lead core surrounded by a shell of rubber (Sheng *et al.*, 2003), as well as an array of resonators based on cavities containing fluid and an outflow channel (Fang *et al.*, 2006). In contrast to a negative modulus via stored energy or power supply (see Sec. III.A.2), effects via resonance appear only at sufficiently high frequency. Composites of this type have been considered for acoustic sound shielding (Calius *et al.*, 2009) and cloaking (Milton, Briane, and Willis, 2006).

B. Negative refractive index

The refractive index governs the speed of waves and the behavior of waves that interact with interfaces between materials. Analyses of the behavior of lenses, windows, and dielectric mirrors make use of the refractive index.

The concept of negative refractive index was presented by Veselago (1967). Such materials have negative dielectric permittivity and magnetic permeability. Because the square of the refractive index n is given by the product of the permittivity and permeability $n^2 = \epsilon\mu$, it appears that for real values the refractive index is not affected by simultaneously negative permittivity and permeability. Refractive effects do occur in relations that contain permittivity and permeability separately.

It was anticipated that the effects could occur in plasmas and gyrotropic materials in which the permittivity and permeability are tensors, not scalars. Consequences of negative electric and magnetic properties in the refraction of lenses were studied. For example, a flat plate made of negative material is predicted to be capable of focusing waves. These negative effects require dispersion (frequency dependence) of wave speed. Negative refractive index was predicted to enable improved lenses with resolution superior to the accepted diffraction limit to be made (Pendry, 2000).

As for early physical embodiments, three-dimensional electromagnetic lattices of intersecting straight wires developed by Bracewell (1954) and Rotman (1962) provided an effective medium for waves with a wavelength that was sufficiently long compared with the lattice spacing. Properties were similar to those of neutral plasma. Such lattices were used to interpret the propagation of microwaves in the ionosphere. Plasma exhibits dispersion of waves and a characteristic resonant frequency called the plasma frequency.

Resonance of microstructure in composites made of coated cylinders is predicted to rise to extreme dielectric properties provided that the outer layer dielectric constant is the negative of either the core dielectric constant or the matrix dielectric constant (Nicorovici, McPhedran, and Milton, 1994). Effects occur even if the inclusions have a small concentration.

Periodic lattice composites containing copper split ring resonators exhibit negative refraction (Smith *et al.*, 2000). Similarly, a periodic lattice of split ring resonators was predicted to exhibit negative electric permittivity and magnetic permeability at microwave frequencies in the vicinity of 5 GHz (Shelby *et al.*, 2001). Negative refraction was observed experimentally in a lattice consisting of square copper split ring resonators and copper wire strips assembled from circuit board elements (Shelby, Smith, and Schultz, 2001). Arrays of toroids have been also used as previously described. Chiral structures can be used to obtain negative refraction (Pendry, 2004). Chiral resonance leads to negative refraction via a single resonance rather than resonance in both permittivity and permeability. Negative refraction of light was obtained using a waveguide exhibiting a surface plasmon polariton mode (Lezec, Dionne, and Atwater, 2007).

Lattices of laminated platelike elements exhibited a negative refractive index over a range of infrared wavelengths (Valentine *et al.*, 2008). These lattices of fishnet structure were made on a multilayer metal-dielectric stack via using focused ion-beam milling. The metal was silver and the dielectric was magnesium fluoride. This method has sufficient resolution to cut nanometer size ribs and plates.

Negative index materials have been used to make super lenses that focus visible light to a resolution of 70 nm (Smolyaninov, Hung, and Davis, 2007). This resolution exceeds the usual diffraction limit and is superior to that of a standard optical microscope. The lenses were made with alternating layers of positive and negative refractive index. Negative index was achieved by depositing fine scale concentric rings of polymer on a gold thin film. Composites and lattices that depend on resonance share the common aspect that they exhibit dispersion (frequency dependence) of properties.

C. Reciprocity in waves

For waves reciprocity states that the frequency response functions between any two points are the same after exchange of the source and receiver (Nassar *et al.*, 2020). If reciprocity applies to a material, one cannot adjust wave transmission to be different in opposite directions. In particular, reciprocal materials do not admit acoustic or optical diodes. Reciprocity for waves can fail in active materials with external energy input. Such materials violate time-reversal invariance. Reciprocity can fail in nonlinear materials. For example, electrical diodes exhibit a nonlinear relation between voltage and current. Bifurcations allow nonreciprocal behavior and hence unidirectional propagation of waves. A well-known bifurcation occurs in the buckling of a column that can diverge into any of several buckled states. This is a nonlinear phenomenon. If a material is made with ribs near a buckling transition, it can exhibit nonreciprocal response. The goals of the study of nonreciprocal media are to advance basic science and enable new devices that allow the unidirectional propagation of waves and control of the direction of waves.

VIII. DISCUSSION

Limits and bounds via thermodynamics on physical properties are useful in guiding design with materials, but it has been found that many such bounds are overly restrictive. Some limits are derived using assumptions, not always stated, about the system or material. If a material is chosen or designed that does not obey those assumptions, then the limits and bounds can be exceeded. Doing so not only manifests the freedom of the researcher but also expands the regime of performance that can be attained in practical uses of materials. Many new materials and classes of materials have been made in the context of achieving extremal physical properties, exceeding accepted bounds on physical properties, or exhibiting a response that is not anticipated in the classical theories. Many of these materials have a larger scale structure. The properties can depend strongly on the nature of this structure. Thus, the researcher or designer is not restricted by the chemical composition of the materials used as much as would be the case if homogeneous materials were used.

Living tissue and certain designed materials are active in that a power source is provided. Not even the mineral kingdom is passive or at equilibrium. Annealing (physical aging) occurs as energy stored in the formation of the material is gradually released. Earth itself, though of great age, exhibits dynamic activity powered by a dilute concentration of long-lived radioactive elements. Matter formed from minerals via human technology is not fully passive either. Annealing occurs and physical properties change with time. Energy is liberated slowly during annealing. Therefore, neither minerals nor materials made by humans are as passive or dead as is usually assumed in equilibrium thermodynamic derivations.

Materials may exhibit counterintuitive properties, giant values of properties, or both and yet be consistent with classical thermodynamic limits. Such materials are nonetheless interesting and of potential practical significance.

Materials with additional freedom can be explored at one level by demonstrating the existence of that freedom. For a full characterization of the material, one needs all the physical constants or material property functions. This is an effort in progress for many of the materials discussed.

New materials and new classes of materials have been developed in the context of seeking extreme or unusual properties, in particular, materials with negative or extreme properties based on cellular or lattice structure or on constrained metastability, as well as nonreciprocal materials. This can be a creative process that contrasts with the methods of investigation of materials found in nature. For future endeavors one may study carefully implicit and explicit assumptions in theories that limit freedom, but to facilitate creativity, not to overemphasize strategy.

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