Fundamental Principles for Generalized Willis Metamaterials

René Pernas-Salomón and Gal Shmuel

Faculty of Mechanical Engineering, Technion-Israel Institute of Technology, Haifa 32000, Israel

(Received 10 August 2020; revised 4 October 2020; accepted 20 October 2020; published 1 December 2020)

Metamaterials that exhibit a constitutive coupling between their momentum and strain, show promise in wave manipulation for engineering purposes and are called Willis materials. They were discovered using an effective-medium theory, showing that their response is nonlocal in space and time. Recently, we generalized this theory to account for piezoelectricity, and demonstrated that the effective momentum can depend constitutively on the electric field, thereby enlarging the design space for metamaterials. Here, we develop the mathematical restrictions on the effective properties of such generalized Willis materials, owing to passivity, reciprocity, and causality. The establishment of these restrictions is of fundamental significance, as they test the validity of theoretical and experimental results—and applicational importance, since they provide elementary bounds for the maximal response that potential devices may achieve.

DOI: 10.1103/PhysRevApplied.14.064005

I. INTRODUCTION

The response of artificial composites with specially designed microstructures can fundamentally differ from the response of their constituents. Such composites are termed *meta*materials, and their features span various solid facets, including electromagnetic and mechanical properties [1-6].

A prominent thrust in metamaterial design is wave control [7-10], where some of the achievements thus far are wave suppressors, cloaking, negative refraction, and superlensing [11–24]. These phenomena are often manifestations of anomalous effective properties, such as negative refractive index and negative mass [25-29], which are analytically determined using homogenization (effectivemedium) theories [30-41]. Notably, Willis has developed an elastodynamic homogenization theory that predicts that the momentum and stress can be constitutively coupled to the strain and the velocity, respectively, by the now-termed Willis couplings [42-48]. These effective properties constitute additional degrees of freedom to manipulate waves, as has been demonstrated, e.g., to experimentally realize asymmetric reflection and scattering-free refraction [49-51].

Recently, Pernas-Salomón and Shmuel [52] have generalized the homogenization theory of Willis to account for constituents that linearly deform in response to nonmechanical fields, such as piezomagnetic and piezoelectric materials [53,54]. The main observation that the generalized theory delivers is the emergence of additional couplings of Willis type between the momentum and the velocity to the nonmechanical fields, as illustrated in Fig. 1. Accordingly, the momentum of piezoelectric (respectively, piezomagnetic) composites is coupled with the electric (respectively, magnetic) field, while the velocity is coupled with the electric displacement field (respectively, magnetic induction). We refer to metamaterials that exhibit these couplings as generalized Willis materials. The additional couplings not only enlarge the design space of metamaterial properties but also reflect a different mechanism to actively manipulate waves via nonmechanical stimuli.

Like all constitutive relations, those that describe Willis materials—standard and generalized—should respect basic physical principles. Srivastava [55] and Muhlestein *et al.* [56] have derived the mathematical restrictions that follow from reciprocity, passivity, and causality principles on standard Willis materials in the long-wavelength limit. Here, we continue their work by developing the restrictions that follow from these principles for *generalized* Willis materials.

Accordingly, in the development of the restrictions, we account for the coupling between the electric and mechanical governing equations, as well as the additional material properties in the constitutive relations. Furthermore, the analysis that we carry out for reciprocity—and to a certain extent for causality—goes beyond the long-wavelength limit, hence also providing insights into standard Willis materials, in addition to the long-wavelength results in Refs. [55,56].

In the sequel, we show that the obtained mathematical restrictions elucidate the physical nature of such generalized couplings. These restrictions are also of applicational importance, as pointed out in Refs. [55–57], since they

^{*}meshmuel@technion.ac.il



FIG. 1. The schematics of the cross-couplings reported by Pernas-Salomón and Shmuel [52] in composites with elasticity that is intrinsically coupled with other physics, such as piezoelectric and piezomagnetic materials.

provide means of testing the admissibility of experimental data and quantifying the maximal response that potential devices may achieve. For example, Quan *et al.* [58] have sought acoustic scatterers with maximum Willis coupling as follows from passivity and reciprocity, and then have employed their optimal structure to design metasurfaces for sound steering.

The paper is structured as follows. In the rest of this section, we recall relevant developments in Willis equations, discuss their uniqueness, and summarize our results before presenting derivations. Section II revisits the theory that has led to the generalized Willis equations and introduces a modified formulation, which is motivated by the analysis in Refs. [56,59,60] for the elastic case. Sections III–V develop the restrictions that passivity, reciprocity, and causality pose on the effective relations, respectively. Final comments conclude this paper in Sec. VI.

A. Relevant developments in Willis equations

Since this work is closely related to Willis equations, as it provides physical restrictions on their generalization, a more elaborated review of their relevant developments is in order. The topics discussed next do not constitute a complete review of the works in the field and aspects such as weighted averages [59,61], connections with asymptotic homogenization [62,63], etc. are not addressed here.

Willis has started to develop his formulation using a variational approach that extends the concepts of ensemble averaging and comparison media from elastostatics [42–44,64]. His effective relations exhibit two notable features, in addition to the emergence of the cross-couplings mentioned earlier. First, they are nonlocal in space—as known from elastostatics—*and in time*, even if the response of the original composite is history independent. (The nonlocal nature renders the effective relations nonunique, an issue that is discussed later.) Second, the kernel that describes the effective mass density is a second-order tensor.

More recently, Willis has developed a formulation that does not rely on a comparison medium, but rather on the Green function of the studied composite [46]. Importantly, he has resolved the lack of uniqueness in the effective properties, which occurs since the effective strain and velocity are derived from the same potential (displacement) field. This has been carried out by adapting the approach of Fietz and Shvets [65], which introduces an additional driving source using an eigenstrain, thereby forcing the effective relative strain and velocity to be independent. While it is questionable if such eigenstrains can be experimentally prescribed [66], their mathematical inclusion has the benefit of providing unique effective properties out of an equivalent class that exists when the eigenstrain vanishes. Source-driven homogenization has also been adopted later, in Refs. [67-69]. Having listed the main developments in Willis theory, we can now point out the common components with our theory for media that deform by nonmechanical stimuli: our theory also relies on ensemble averaging, incorporates eigenstrains as an additional driving source, and delivers unique effective properties based on the Green function of the original composite.

The recent interest in metamaterials [70] has disseminated to Willis effective relations, resulting in a bulk of papers that present experimental validation in the long-wavelength limit and analyze their structure [51,58, 60,71-81]. We list next some of the insights that are relevant to this paper. Milton *et al.* [82] have identified the similarity between Willis couplings and bianisotropy in electromagnetics (see also Refs. [56,58,69,77,83,84]). Sieck *et al.* [69] have provided a perceptive analysis on the source of the cross-couplings in periodic media, concluding that the their nonlocal part originates from multiple scattering and phase change at the mesoscale, while their local part originates from asymmetry in the unit cell. Similarly, Pernas-Salomón and Shmuel [85] have pointed out the analogy with the broken inversion symmetry in piezoelectric materials at the atomic scale, which leads to microscopic electroelastic coupling.

Spatially local couplings have been proposed by Milton et al. [82]. As pointed out in Ref. [84], the corresponding equations are the limiting case of the nonlocal equations, referred to as the Milton-Briane-Willis equations. According to Milton [66], the local form is more physical, owing to the difficulty in experimentally measuring unique nonlocal properties that include the cross-couplings. A local model has been developed by Milton [59], the stress of which depends on the acceleration rather than the velocity. Simpler spatially local models that report accelerationdependent stress have been given later in Refs. [56,60, 84,86]. These works suggest that the nonlocal nature of the operator conceals [87] a more physical constitutive description-one that employs the strain rate and acceleration as additional input functions. Here, we adapt and examine this suggestion to our settings, by introducing and analyzing a description that additionally includes the time derivative of the electric field as an input function, and find arguments that support the use of the alternative formulation.

B. Summary of our results

As discussed above, in Ref. [52], we have developed a dynamic homogenization theory for piezoelectric and piezomagnetic composites, which delivers nonlocal effective relations between suitably defined macroscopic fields. We have formally shown that additional couplings emerge in the effective relations between the macroscopic momentum and velocity to the nonmechanical fields. In the sequel, we develop the mathematical restrictions that the effective relations must satisfy in order to respect three principles.

The first principle that we analyze is passivity, which at the basic level means that the material does not generate energy. Formally, we require that the power supplied by external agents is always greater or equal to the rate of change of the energy stored by the material. This principle delivers inequalities for the skew-Hermitian and Hermitian parts of the Fourier transforms of the effective properties, as summarized in Table I. If the material exhibits major symmetries, then these inequalities apply to the imaginary and real parts of the transforms. If the material is passive and lossless, we find that the direct couplings—and combinations of cross-couplings—must be either Hermitian or skew-Hermitian.

The second principle that we employ is reciprocity, which refers to an equality between the power produced by conjugate fields of different problems. In the long-wavelength limit, it implies major symmetries for direct couplings and transpose relations between conjugate cross-coupling terms. This result includes the symmetries reported in Ref. [56] for the elastic properties in local Willis materials (i.e., Milton-Briane-Willis materials). Beyond the long-wavelength limit, we find that reciprocity requires the nonlocal operator to be self-adjoint with respect to the spatial variables. Technically, this translates to an interchange in the functional dependency in these variables, in addition to the transposition relations among the couplings (see Table I). From this analysis, we deduce that the formulation that does not use the time derivative of the velocity, strain, and electric field is unphysical, since it corresponds to imaginary properties in the time domain. By contrast, the modified formulation that is based on these rates leads to real properties in the time domain.

The last principle that we employ is causality, which means that an effect (e.g., momentum) cannot precede its cause (e.g., an electric field). This principle provides

TABLE I. The mathematical restrictions that the effective description of piezoelectric composites with generalized Willis couplings satisfy, owing to reciprocity and passivity. The restrictions that result from causality are of the Kramer-Kronig type, namely, $\hat{L}''(\omega) = -\frac{2}{\pi} \int_0^\infty \frac{\omega \hat{L}'(z)}{z^2 - \omega^2} dz$, and according to Landau and Lifshitz [89] should apply for any fixed value of κ .

Property	Real-space reciprocity	Fourier-space reciprocity	Passivity when $\kappa = 0$
Elasticity	$\tilde{\mathbf{C}}(\mathbf{x}, \mathbf{X}) = \tilde{\mathbf{C}}^{T}(\mathbf{X}, \mathbf{x})$	$\check{\mathbf{C}}(\boldsymbol{\kappa}) = \check{\mathbf{C}}^{T}(-\boldsymbol{\kappa})$	$i\check{\mathbf{C}}^{\mathrm{SH}}$ positive definite
Mass density	$\tilde{\boldsymbol{\rho}}(\mathbf{x}, \mathbf{X}) = \tilde{\boldsymbol{\rho}}^{T}(\mathbf{X}, \mathbf{x})$	$\check{\boldsymbol{\rho}}(\boldsymbol{\kappa}) = \check{\boldsymbol{\rho}}^{T}(-\boldsymbol{\kappa})$	$i\check{\rho}^{\rm SH}$ negative definite
Willis coupling	$\tilde{\mathbf{S}}^{\dagger}\left(\mathbf{x},\mathbf{X} ight)=\tilde{\mathbf{S}}^{T}\left(\mathbf{X},\mathbf{x} ight)$	$\check{\mathbf{S}}^{\dagger}(\boldsymbol{\kappa}) = \check{\mathbf{S}}^{T}(-\boldsymbol{\kappa})$	Bound for \check{S}^{QH}
Modified Willis coupling	$\widehat{\mathbf{S}}^{\dagger}\left(\mathbf{x},\mathbf{X} ight)=\widehat{\mathbf{S}}^{T}\left(\mathbf{X},\mathbf{x} ight)$	$\widehat{\mathbf{S}}^{\dagger}\left(\boldsymbol{\kappa}\right) = \widehat{\mathbf{S}}^{T}\left(-\boldsymbol{\kappa}\right)$	Bound for $\widehat{\mathbf{S}}^{ ext{QSH}}$
Permittivity	$\tilde{\mathbf{A}}\left(\mathbf{x},\mathbf{X}\right) = \tilde{\mathbf{A}}^{T}\left(\mathbf{X},\mathbf{x}\right)$	$\check{\mathbf{A}}(\boldsymbol{\kappa}) = \check{\mathbf{A}}^{T}(-\boldsymbol{\kappa})$	$i\check{\mathbf{A}}^{\mathrm{SH}}$ negative definite
Piezoelectric coupling	$\tilde{\mathbf{B}}^{\dagger}\left(\mathbf{x},\mathbf{X} ight)=\tilde{\mathbf{B}}^{T}\left(\mathbf{X},\mathbf{x} ight)$	$\check{\mathbf{B}}^{\dagger}\left(\boldsymbol{\kappa}\right)=\check{\mathbf{B}}^{T}\left(-\boldsymbol{\kappa}\right)$	Bound for $\check{\mathbf{B}}^{\text{QSH}}$
Electromomentum coupling	$\check{\mathbf{W}}^{\dagger}(\mathbf{x},\mathbf{X})=\check{\mathbf{W}}^{T}(\mathbf{X},\mathbf{x})$	$\check{\mathbf{W}}^{\dagger}(\boldsymbol{\kappa}) = \check{\mathbf{W}}^{T}(-\boldsymbol{\kappa})$	Bound for $\check{\mathbf{W}}^{\mathrm{QH}}$
Modified EM coupling	$\widehat{\mathbf{W}}^{\dagger}\left(\mathbf{x},\mathbf{X} ight)=\widehat{\mathbf{W}}^{T}\left(\mathbf{X},\mathbf{x} ight)$	$\widehat{\mathbf{W}}^{\dagger}\left(\boldsymbol{\kappa} ight)=\widehat{\mathbf{W}}^{T}\left(-\boldsymbol{\kappa} ight)$	Bound for $\widehat{\mathbf{W}}^{ extsf{QSH}}$

a connection between the real and imaginary parts of the (time) transforms of the couplings. The process that we employ is standard and straightforward and uses the Plemelj formulas to obtain relations of the Kramers-Kronig type for the generalized effective properties [88–91]. We clarify that our study of causality is restricted to the spatially local equations and note that the corresponding analysis supports the claim that the alternative formulation should be favored. Finally, we note that according to Landau and Lifshitz [89], the Plemelj formulas should hold for any fixed value of the wavevector.

II. DYNAMIC HOMOGENIZATION OF PIEZOELECTRIC COMPOSITES

We consider a composite occupying the volume Ω , made of piezoelectric phases, driven by time-dependent body force density **f**, inelastic strain η , and free-charge density *q*. These sources generate in the composite stress σ , electric displacement **D**, and momentum density **p**, which satisfy the balance equations

$$\nabla \cdot \boldsymbol{\sigma} + \mathbf{f} - \dot{\mathbf{p}} = \mathbf{0},\tag{1}$$

and

$$\nabla \cdot \mathbf{D} = q, \tag{2}$$

where the superposed dot denotes a time derivative. At each material point **x**, these fields are related to the displacement gradient $\nabla \mathbf{u}$, velocity $\dot{\mathbf{u}}$, and electric potential gradient [92] $\nabla \phi$ through the constitutive equations of piezoelectricity [93], namely [94],

$$\begin{pmatrix} \boldsymbol{\sigma} \\ \mathbf{D} \\ \mathbf{p} \end{pmatrix} = \begin{pmatrix} \mathbf{C} & \mathbf{B}^{\mathsf{T}} & 0 \\ \mathbf{B} & -\mathbf{A} & 0 \\ 0 & 0 & \rho \end{pmatrix} \begin{pmatrix} \nabla \mathbf{u} - \boldsymbol{\eta} \\ \nabla \boldsymbol{\phi} \\ \dot{\mathbf{u}} \end{pmatrix}, \quad (3)$$

where ρ , **A**, **B**, and **C** are the spatially varying [95] local mass density, dielectric, piezoelectric, and elasticity tensor fields, respectively [96]. In coordinates, these tensors satisfy

$$A_{ij} = A_{ji}, \quad B_{ijk} = B_{jik}, \quad B_{ijk}^{\mathsf{I}} = B_{kij},$$

$$C_{ijkl} = C_{jikl} = C_{jilk} = C_{klij}, \quad \sigma_{ij} = \sigma_{ji}.$$
(4)

Pernas-Salomón and Shmuel [52] have proposed an effective description with constitutive equations for the composite by extending the approach of Willis [46]. This has been carried out by treating the composite as random, such that its properties are not only functions of **x** but also of the particular specimen that belongs to some sample space \mathscr{S} . The expectation value of any property, say ρ , is given by the ensemble average

$$\langle \rho \rangle (\mathbf{x}) = \int_{\mathscr{S}} \rho (\mathbf{x}, y) P(y) \, dy,$$
 (5)

where the parameter y is used to label the specimens and P is the probability-measure function over \mathscr{S} . The governing equations of our effective description are given by the following ensemble averages of Eqs. (1) and (2):

$$\nabla \cdot \langle \boldsymbol{\sigma} \rangle + \mathbf{f} - \langle \mathbf{p} \rangle = \mathbf{0}, \quad \nabla \cdot \langle \mathbf{D} \rangle = q, \tag{6}$$

in which $\langle \sigma \rangle$, $\langle \mathbf{D} \rangle$, and $\langle \mathbf{p} \rangle$ are the effective fields [97]. Based on the Green (tensor) function of the problem, Pernas-Salomón and Shmuel [52] have obtained constitutive equations for the effective fields in the form [98]

$$\begin{pmatrix} \langle \boldsymbol{\sigma} \rangle \\ \langle \mathbf{D} \rangle \\ \langle \mathbf{p} \rangle \end{pmatrix} = \begin{pmatrix} \mathcal{C} & \mathcal{B}^{\dagger} & \mathcal{S} \\ \mathcal{B} & -\mathcal{A} & \mathcal{W} \\ \mathcal{S}^{\dagger} & \mathcal{W}^{\dagger} & \mathcal{R} \end{pmatrix} \begin{pmatrix} \langle \nabla \mathbf{u} \rangle - \boldsymbol{\eta} \\ \langle \nabla \boldsymbol{\phi} \rangle \\ \langle \dot{\mathbf{u}} \rangle \end{pmatrix}, \quad (7)$$

where the matrix elements are now *nonlocal operators* in *time and space*. (At this point, we do not endow the couplings with superscript \dagger the meaning that this symbol usually designates and postpone it to Sec. IV.) We denote the column vectors on the left- and right-hand sides of Eq. (7) by $\langle h \rangle$ and $\langle g \rangle$ and put the latter statement into formal footing, namely,

where \mathcal{L} denotes the nonlocal effective constitutive operator and \tilde{L} is its kernel. In the sequel, we denote by \tilde{C} the kernel of C, by \tilde{S} the kernel of S, by $\tilde{\rho}$ the kernel of \mathcal{R} , etc. The effective operator exhibits three notable features, in addition to its spatiotemporal nonlocal nature. First, it couples $\langle \boldsymbol{\sigma} \rangle$ with $\langle \dot{\mathbf{u}} \rangle$, and $\langle \mathbf{p} \rangle$ with $\langle \nabla \mathbf{u} \rangle$, through the so-called Willis couplings S and S^{\dagger} . Second, (the kernel of) the effective mass density, $\tilde{\rho}$, is a second-order tensor. As mentioned, these two features-which are absent from the local constitutive equations and hence represent metamaterials—have been discovered by Willis [42–44] in his studies of purely elastic composites. The third distinctive feature reported by Pernas-Salomón and Shmuel [52] is the coupling \mathcal{W} between $\langle \mathbf{D} \rangle$ and $\langle \dot{\mathbf{u}} \rangle$, and the coupling \mathcal{W}^{\dagger} between $\langle \mathbf{p} \rangle$ and $\langle \nabla \phi \rangle$, which we term the electromomentum coupling. The transition to this effective description is schematically illustrated in Fig. 2. The kernel



FIG. 2. The body Ω is composed of different piezoelectric materials, the constitutive response of which is given by Eq. (3), as illustrated at the top of the sketch. Effectively, the response of the body is nonlocal with additional cross-couplings, as given by Eq. (7).

of \mathcal{L} is endowed with the minor symmetries

$$\tilde{C}_{ijkl} = \tilde{C}_{jikl}, \quad \tilde{C}_{ijkl} = \tilde{C}_{ijlk}, \quad \tilde{B}_{ijk} = \tilde{B}_{ikj}, \quad \tilde{B}^{\dagger}_{ijk} = \tilde{B}^{\dagger}_{jik}, \\
\tilde{S}_{ijk} = \tilde{S}_{jik}, \quad \tilde{S}^{\dagger}_{ikl} = \tilde{S}^{\dagger}_{ilk},$$
(9)

as they translate from the microscopic to the effective description, owing to the balance of angular momentum and independence from the antisymmetric part of $\nabla \mathbf{u}$. The major symmetries of the constitutive tensors in Eq. (4) induce additional symmetries between the effective tensors (and justify the superscript \dagger mentioned above), to be discussed in Sec. IV and the Appendix.

When the composite is statistically homogeneous, the constitutive operator becomes translation invariant, i.e., it depends only on the difference $\mathbf{x} - \boldsymbol{\chi}$; accordingly, Eq. (8) has the form of a convolution not only in time but also in space. Therefore, the Fourier transform with respect to both time and space yields constitutive relations in the form of simple products between the transforms of \tilde{L} and $\langle \mathbf{g} \rangle$. It follows that such an infinite medium admits plane waves in the form of (the real part of) $\langle \mathbf{u} \rangle = \mathbf{U}e^{i(\kappa_0 \cdot \mathbf{x} - \omega_0 t)}$ and $\langle \phi \rangle = \Phi e^{i(\kappa_0 \cdot \mathbf{x} - \omega_0 t)}$, for which the nonlocal constitutive equations are the simple products

$$\langle \mathbf{h} \rangle (\mathbf{x}, t) = \dot{\mathsf{L}} (-\boldsymbol{\kappa}_0, \omega_0) \langle \mathbf{g} \rangle (\mathbf{x}, t)$$
 (10)

in the (\mathbf{x}, t) space. (Again, the real part of the equation should be taken.) We emphasize that $\check{\mathsf{L}}(-\kappa_0, \omega_0)$ is the space-time Fourier transform of $\tilde{\mathsf{L}}$ according to the convention

$$\check{\mathsf{L}}(\boldsymbol{\kappa},\omega) = \int_{\Omega} d\mathbf{x} \int_{\mathbb{R}} dt \, \tilde{\mathsf{L}}(\mathbf{x},t) \, e^{i(\boldsymbol{\kappa}\cdot\mathbf{x}+\omega t)}, \qquad (11)$$

evaluated at $(-\kappa_0, \omega_0)$. In order not to introduce more notation to the already large set used here, we also use (\circ) for transforms that are applied only with respect to one of the two variables (time or space).

The objective of this work is to determine the mathematical restrictions imposed on relations (7)—and specifically on the electromomentum coupling—by the physical principles of reciprocity, passivity, and causality. In addition to form (7), we also analyze the form

$$\begin{pmatrix} \langle \boldsymbol{\sigma} \rangle \\ \langle \mathbf{D} \rangle \\ \langle \mathbf{p} \rangle \end{pmatrix} = \begin{pmatrix} \mathcal{C} & \mathcal{B}^{\dagger} & 0 \\ \mathcal{B} & -\mathcal{A} & 0 \\ 0 & 0 & \mathcal{R} \end{pmatrix} \begin{pmatrix} \langle \nabla \mathbf{u} \rangle - \boldsymbol{\eta} \\ \langle \nabla \phi \rangle \\ \langle \dot{\mathbf{u}} \rangle \end{pmatrix} + \begin{pmatrix} 0 & 0 & \widehat{\mathcal{S}} \\ 0 & 0 & \widehat{\mathcal{W}} \\ \widehat{\mathcal{S}}^{\dagger} & \widehat{\mathcal{W}}^{\dagger} & 0 \end{pmatrix} \begin{pmatrix} \langle \nabla \dot{\mathbf{u}} \rangle - \dot{\boldsymbol{\eta}} \\ \langle \nabla \dot{\boldsymbol{\phi}} \rangle \\ \langle \ddot{\mathbf{u}} \rangle \end{pmatrix},$$
(12)

where the kernel of the time Fourier transform of \widehat{S} is $-\check{S}/i\omega$, the kernel of the transform of \widehat{W} is $-\check{W}/i\omega$, etc. The motivation for this form is mentioned in Sec. I A and elaborated next. To this end, it is useful to note that the derivations that have led Willis [46] and Pernas-Salomón and Shmuel [52] to their nonlocal operators have been carried out after applying the Fourier transform with respect to time [99], where in the frequency domain the cross-coupling terms are products that include the term $-i\omega$. An ambiguity emerges when transforming back to the time domain: Should $-i\omega$ be identified with the kernel or with the time derivative of $\langle g \rangle$? The former leads to relations (7) and the latter to relations (12). The forthcoming analysis supports form (12), in agreement with Refs. [56,69].

Before we proceed, we note that a similar ambiguity exists when the transform is applied with respect to the spatial translation [69]. In this case, spatial derivatives turn to products with $i\kappa$ and the inverse transform has the same problem as with the inversion of products with $i\omega$. We can now highlight the motivation for introducing η : since it is not derived from a potential, there is no way to "pull outside" the gradient operator in order to obtain the effective displacement field and then mistake the effective velocity for the effective strain by multiplying and dividing by $i\omega$ [60]. Evidently, such operations lead to different sets of effective properties and, in particular, a set without Willis couplings [100]. Since, clearly, the velocity or strain cannot be derived from the electric potential, there is no need to introduce an "eigen electric field" in our theory [101]. Owing to the ambiguity associated with the nonlocal operator and the difficulty of measuring the nonlocal cross-coupling, Milton [66,84] has recently advocated either the use of the local cross-coupling or the use of a nonlocal operator that relates the displacement to the applied force. While we do not pursue this notion here, we note that the available experimental evidence for such cross-couplings has been obtained when nonlocal interactions are negligible (see, e.g., Refs. [75,78]), thereby supporting the advocacy of Milton for the local equations.

III. PASSIVITY

The term "passivity" has different uses in the literature. Here, it is interpreted as in Ref. [102], namely, a system is passive if there exists a positive-definite stored energy function for it, determined uniquely by its state variables, such that the power supplied to the system by external agents is always greater or equal to the rate of change of its stored energy. This requirement, in turn, poses restrictions on the constitutive parameters [55,103]. The implications of passivity have been employed in Refs. [55,56] to determine the restrictions on Willis materials. In this section, we extend the analysis to piezoelectric materials that exhibit electromomentum coupling, where by assuming passivity we derive restrictions on the constitutive tensors given in Eqs. (7) and (12).

We consider a piezoelectric solid of volume Ω that is surrounded by air. Across its boundary $\partial \Omega$, a surface charge density w_e and traction **t** are present, in addition to the volume densities q and **f**. For simplicity, eigenstrains are not considered here, bearing in mind that the effective properties to be used in the sequel are those identified using such eigenstrains. Assuming time-harmonic fields, we can express the complex rate of work done on the piezoelectric body by the mechanical and electrical sources, namely,

$$P_{c} = \oint_{\partial\Omega} \left(\frac{\mathbf{t} \cdot \dot{\mathbf{u}}^{*}}{2} + \frac{\phi \dot{w_{e}}^{*}}{2} \right) da + \int_{\Omega} \left(\frac{\mathbf{f} \cdot \dot{\mathbf{u}}^{*}}{2} + \frac{\phi \dot{q}^{*}}{2} \right) d\mathbf{x},$$
(13)

such that the real part of P_c is the time-average power delivered by the sources [93]. Using the connections $\mathbf{t} = \boldsymbol{\sigma} \cdot \mathbf{n}$ and $\mathbf{D} \cdot \mathbf{n} = -w_e$, where \mathbf{n} is a unit vector in the outward normal direction to $\partial \Omega$, we obtain a restatement of the complex Poynting's theorem for piezoelectric media in the settings of the quasielectrostatic approximation as [93]

$$P_{c} = \oint_{\partial\Omega} \left(\frac{\boldsymbol{\sigma} \cdot \dot{\mathbf{u}}^{*}}{2} - \frac{\boldsymbol{\phi} \dot{\mathbf{D}}^{*}}{2} \right) \cdot \mathbf{n} da + \int_{\Omega} \left(\frac{\mathbf{f} \cdot \dot{\mathbf{u}}^{*}}{2} + \frac{\boldsymbol{\phi} \dot{q}^{*}}{2} \right) d\mathbf{x}$$
$$= \int_{\Omega} \left(\frac{\boldsymbol{\sigma} : \nabla \dot{\mathbf{u}}^{*}}{2} + \frac{\dot{\mathbf{p}} \cdot \dot{\mathbf{u}}^{*}}{2} - \frac{\nabla \boldsymbol{\phi} \cdot \dot{\mathbf{D}}^{*}}{2} \right) d\mathbf{x}.$$
(14)

In the process that led to Eq. (14), we have applied the divergence theorem and used the field equations (1) and (2) after the expansion of the divergence operator. The imaginary part of this volume integral relates to the total stored energy within Ω (elastic, kinetic, and electric energy) and its real part is the time-averaged power loss of the system. Since a passive material cannot generate energy, the inflow of power is always non-negative and hence $P'_c := \text{Re}P_c$ is non-negative too, where here and throughout the text we

use ' and " to denote the real and imaginary parts of any variable, respectively. This requirement imposes restrictions on the permitted values of the constitutive tensors, when P'_c is expressed using the generalized Willis relations. Invoking statistical homogeneity and considering plane-wave solutions (assuming that they are valid), we employ the form given in Eq. (10) to write the condition on P'_c as

$$P_{c}^{\prime} = \frac{1}{2} \operatorname{Re} \left\{ \int_{\Omega} \left(\dot{u}_{i,j}^{*} \check{C}_{ijkl} u_{k,l} + \dot{u}_{i,j}^{*} \check{B}_{ijk}^{\dagger} \phi_{,k} + \dot{u}_{i,j}^{*} \check{S}_{ijk} \dot{u}_{k} \right. \\ \left. - \phi_{,i}^{*} \check{B}_{ikl} \dot{u}_{k,l} + \phi_{,i}^{*} \check{A}_{ik} \dot{\phi}_{,k} - \phi_{,i}^{*} \check{W}_{ik} \ddot{u}_{k} \right. \\ \left. + \dot{u}_{i}^{*} \check{W}_{ik}^{\dagger} \dot{\phi}_{,k} + \dot{u}_{i}^{*} \check{S}_{ikl}^{\dagger} \dot{u}_{k,l} + \dot{u}_{i}^{*} \check{\rho}_{ik} \ddot{u}_{k} \right) d\mathbf{x} \right\} \ge 0; \quad (15)$$

here, we have used the fact that Re { $\nabla \phi \cdot \dot{\mathbf{D}}^*$ } = Re { $\nabla \phi^* \cdot \dot{\mathbf{D}}$ }. The components of $\check{\mathbf{L}}$ appearing in Eq. (15) are the transforms at ($-\kappa, \omega$) and we note that by linearity there is no loss of generality when considering a single κ vector. Equation (15) is simplified using the following relations. First, we introduce the skew-Hermitian parts of $\check{\rho}$, $\check{\mathbf{A}}$, and $\check{\mathbf{C}}$, namely,

$$\check{\rho}_{ik}^{\text{SH}} = \frac{1}{2} \left(\check{\rho}_{ik} - \check{\rho}_{ki}^{*} \right), \quad \check{A}_{ik}^{\text{SH}} = \frac{1}{2} \left(\check{A}_{ik} - \check{A}_{ki}^{*} \right),
\check{C}_{ijkl}^{\text{SH}} = \frac{1}{2} \left(\check{C}_{ijkl} - \check{C}_{klij}^{*} \right),$$
(16)

to rewrite the terms Re $\{\dot{u}_i^* \check{\rho}_{ik} \ddot{u}_k\}$, Re $\{\phi_{,i}^* \check{A}_{ik} \dot{\phi}_{,k}\}$, and Re $\{\dot{u}_{i,j}^* \check{C}_{ijkl} u_{k,l}\}$ as

$$\operatorname{Re}\left\{\dot{u}_{i}^{*}\check{\rho}_{ik}\ddot{u}_{k}\right\} = \frac{1}{2}\left(\dot{u}_{i}^{*}\check{\rho}_{ik}\ddot{u}_{k} + \dot{u}_{i}\check{\rho}_{ik}^{*}\ddot{u}_{k}^{*}\right)$$
$$= -i\omega\check{\rho}_{ik}^{\mathrm{SH}}\dot{u}_{i}^{*}\dot{u}_{k}, \qquad (17a)$$

$$\operatorname{Re}\left\{\phi_{,i}^{*}\check{A}_{ik}\dot{\phi}_{,k}\right\} = \frac{1}{2}\left(\phi_{,i}^{*}\check{A}_{ik}\dot{\phi}_{,k} + \phi_{,i}\check{A}_{ik}^{*}\dot{\phi}_{,k}^{*}\right)$$
$$= -i\omega\check{A}_{ik}^{\mathrm{SH}}\phi_{,i}^{*}\phi_{,k}, \qquad (17b)$$

$$\operatorname{Re}\left\{\dot{u}_{i,j}^{*}\check{C}_{ijkl}u_{k,l}\right\} = \frac{1}{2}\left(\dot{u}_{i,j}^{*}\check{C}_{ijkl}u_{k,l} + \dot{u}_{i,j}\check{C}_{ijkl}^{*}u_{k,l}^{*}\right)$$
$$= i\omega\check{C}_{ijkl}^{\mathrm{SH}}u_{i,j}^{*}u_{k,l}.$$
(17c)

We also note that *if*

$$\check{\rho}_{ik} = \check{\rho}_{ki}, \quad \check{A}_{ik} = \check{A}_{ki}, \quad \check{C}_{ijkl} = \check{C}_{klij}, \quad (18)$$

for all κ , then their skew-Hermitian part is equal to their imaginary part (and the Hermitian part is equal to the real part). The remaining terms can be written as

$$\operatorname{Re}\left\{\dot{u}_{i,j}^{*}\check{B}_{ijk}^{\dagger}\phi_{,k}-\phi_{,i}^{*}\check{B}_{ikl}\dot{u}_{k,l}\right\} = \frac{i\omega}{2}\left(\check{B}_{ijk}^{\dagger}-\check{B}_{kij}^{*}\right)\phi_{,k}u_{i,j}^{*} - \frac{i\omega}{2}\left(\check{B}_{ijk}^{\dagger}-\check{B}_{kij}\right)\phi_{,k}^{*}u_{i,j},$$
$$=\omega\operatorname{Re}\left\{i\left(\check{B}_{ijk}^{\dagger}-\check{B}_{kij}^{*}\right)\phi_{,k}u_{i,j}^{*}\right\} =: 2\omega\operatorname{Re}\left\{i\phi_{,k}\check{B}_{kij}^{\mathrm{QSH}}u_{i,j}^{*}\right\},$$
(19a)

$$\operatorname{Re}\left\{\dot{u}_{i,j}^{*}\check{S}_{ijk}\dot{u}_{k}+\dot{u}_{i}^{*}\check{S}_{ikl}^{\dagger}\dot{u}_{k,l}\right\}=\frac{i\omega}{2}\left(\check{S}_{ijk}+\check{S}_{kij}^{\dagger*}\right)\dot{u}_{k}u_{i,j}^{*}-\frac{i\omega}{2}\left(\check{S}_{ijk}^{*}+\check{S}_{kij}^{\dagger}\right)\dot{u}_{k}^{*}u_{i,j},$$
$$=\omega\operatorname{Re}\left\{-i\left(\check{S}_{ijk}^{*}+\check{S}_{kij}^{\dagger}\right)\dot{u}_{k}^{*}u_{i,j}\right\}=:2\omega\operatorname{Re}\left\{-iu_{i,j}\check{S}_{ijk}^{\operatorname{QH}}\dot{u}_{k}^{*}\right\},$$
$$=\omega\operatorname{Re}\left\{-i\left(\widehat{S}_{ijk}^{*}-\widehat{S}_{kij}^{\dagger}\right)\ddot{u}_{k}^{*}u_{i,j}\right\}=:2\omega\operatorname{Re}\left\{iu_{i,j}\widehat{S}_{ijk}^{\operatorname{QSH}}\ddot{u}_{k}^{*}\right\},$$
(19b)

$$\operatorname{Re}\left\{\dot{u}_{i}^{*}\check{W}_{ik}^{\dagger}\dot{\phi}_{,k}-\phi_{,i}^{*}\check{W}_{ik}\dot{u}_{k}\right\}=\frac{i\omega}{2}\left(\check{W}_{ik}^{\dagger*}+\check{W}_{ki}\right)\dot{u}_{i}\phi_{,k}^{*}-\frac{i\omega}{2}\left(\check{W}_{ik}^{\dagger}+\check{W}_{ki}^{*}\right)\dot{u}_{i}^{*}\phi_{,k},$$

$$=\omega\operatorname{Re}\left\{-i\left(\check{W}_{ki}^{*}+\check{W}_{ik}^{\dagger}\right)\dot{u}_{i}^{*}\phi_{,k}\right\}=:2\omega\operatorname{Re}\left\{-i\phi_{,k}\check{W}_{ki}^{\operatorname{QH}}\dot{u}_{i}^{*}\right\},$$

$$=\omega\operatorname{Re}\left\{-i\left(\widehat{W}_{ki}^{*}-\widehat{W}_{ik}^{\dagger}\right)\ddot{u}_{i}^{*}\phi_{,k}\right\}=:2\omega\operatorname{Re}\left\{i\phi_{,k}\widehat{W}_{ki}^{\operatorname{QSH}}\ddot{u}_{i}^{*}\right\}.$$
(19c)

If the symmetries

$$\check{B}_{ijk}^{\dagger} = \check{B}_{kij}, \quad \widehat{S}_{kij}^{\dagger} = \widehat{S}_{ijk}, \quad \widehat{W}_{ik}^{\dagger} = \widehat{W}_{ki}$$
(20)

hold *for all* κ , then $(\circ)^{\text{QSH}}$ is equivalent to the imaginary part of (\circ) , while $(\circ)^{\text{QH}}$ is equivalent to the real part. As we show in Sec. IV, symmetries (18) and (20) are compatible with reciprocity only in the long-wavelength limit. Using relations (19), Eq. (15) reads

$$P'_{c} = \frac{\omega}{2} \int_{\Omega} \left(u^{*}_{i,j} i\check{C}^{\text{SH}}_{ijkl} u_{k,l} + 2\text{Re}\left\{\phi_{,k} i\check{B}^{\text{QSH}}_{kij} u^{*}_{i,j}\right\} - 2\text{Re}\left\{u_{i,j} i\check{S}^{\text{QH}}_{ijk} \dot{u}^{*}_{k}\right\} - \dot{u}^{*}_{i} i\check{\rho}^{\text{SH}}_{ik} \dot{u}_{k} - \phi^{*}_{,i} i\check{A}^{\text{SH}}_{ik} \phi_{,k} - 2\text{Re}\left\{\phi_{,k} i\check{W}^{\text{QH}}_{ki} \dot{u}^{*}_{i}\right\} \right) d\mathbf{x} \ge 0.$$
(21)

To proceed, we follow the argument of Muhlestein *et al.* [56], which requires the restriction of subsequent analysis to the long-wavelength limit ($\kappa = 0$). In this limiting case, the strain, velocity, and electric fields in Eq. (21) can be prescribed arbitrarily and independently of each other through suitable sets of (boundary and volume) sources. Accordingly, we can first recover the conclusions of Srivastava [55] and Muhlestein *et al.* [56] in the limiting elastic case, by considering a configuration where the electric field vanishes, for which

$$P'_{c} = \frac{\omega}{2} \int_{\Omega} \left(u^{*}_{i,j} i \check{C}^{\text{SH}}_{ijkl} u_{k,l} - 2\text{Re} \left\{ u_{i,j} i \check{S}^{\text{QH}}_{ijk} \dot{u}^{*}_{k} \right\} - \dot{u}^{*}_{i} i \check{\rho}^{\text{SH}}_{ik} \dot{u}_{k} \right) d\mathbf{x} \ge 0,$$
(22)

and in terms of \widehat{S} ,

$$P_{c}' = \frac{\omega}{2} \int_{\Omega} \left(u_{i,j}^{*} i \check{C}_{ijkl}^{\text{SH}} u_{k,l} + 2\text{Re} \left\{ u_{i,j} i \widehat{S}_{ijk}^{\text{QSH}} \ddot{u}_{k}^{*} \right\} - \dot{u}_{i}^{*} i \check{\rho}_{ik}^{\text{SH}} \dot{u}_{k} \right) d\mathbf{x} \ge 0.$$
(23)

By setting the velocity to zero, we obtain

$$\int_{\Omega} u_{i,j}^* i \check{C}_{ijkl}^{\rm SH} u_{k,l} d\mathbf{x} \ge 0, \qquad (24)$$

where the case of a vanishing strain provides

$$\int_{\Omega} \dot{u}_i^* i \check{\rho}_{ik}^{\rm SH} \dot{u}_k d\mathbf{x} \le 0.$$
(25)

Eqs. (24)–(25) hold for arbitrary strain and velocity fields if and only if the Hermitian [104] forms $i\check{C}^{\rm SH}$ and $i\check{\rho}^{\rm SH}$ are positive and negative definite, respectively. If the medium is not only passive but also lossless, then the inequalities become equalities, which imply that \check{C} and $\check{\rho}$ are Hermitian; this agrees with the notion that Hermiticity implies energy conservation [105–107]. The equalities further imply that $\check{S}^{\rm QH}$ and $\widehat{S}^{\rm QSH}$ are null.

As mentioned, this analysis recovers the results of Srivastava [55] and Muhlestein *et al.* [56] for Milton-Briane-Willis materials (i.e., local Willis materials). To develop the restrictions on the couplings that arise in the electroe-lastic setting, we first assume a combination of sources for which the only nonvanishing field is the electric field. In

this setting, Eq. (21) provides

$$\int_{\Omega} \phi_{,i}^* i \check{A}_{ik}^{\mathrm{SH}} \phi_{,k} d\mathbf{x} \le 0.$$
(26)

Since $\nabla \phi$ is arbitrary, this condition holds if and only if the Hermitian form $i\check{A}^{SH}$ is negative definite and in the lossless case this implies that Å is Hermitian, again in agreement with the association of Hermiticity with energy conservation. If only the velocity vanishes, we have that

$$\int_{\Omega} \left(u_{i,j}^* i \check{C}_{ijkl}^{\mathrm{SH}} u_{k,l} + 2 \operatorname{Re} \left\{ \phi_{,k} i \check{B}_{kij}^{\mathrm{QSH}} u_{i,j}^* \right\} - \phi_{,i}^* i \check{A}_{ik}^{\mathrm{SH}} \phi_{,k} \right) d\mathbf{x} \ge 0.$$
(27)

If only the strain is zero,

$$\int_{\Omega} \left(-\dot{u}_{i}^{*} i \check{\rho}_{ik}^{\mathrm{SH}} \dot{u}_{k} - \phi_{,i}^{*} i \check{A}_{ik}^{\mathrm{SH}} \phi_{,k} - 2 \operatorname{Re} \left\{ \phi_{,k} i \check{W}_{ki}^{\mathrm{QH}} \dot{u}_{i}^{*} \right\} \right) d\mathbf{x} \ge 0,$$
(28)

from which we obtain

$$-2\operatorname{Re}\left\{\phi_{,k}i\check{B}_{kij}^{\mathrm{QSH}}u_{i,j}^{*}\right\} \leq u_{i,j}^{*}i\check{C}_{ijkl}^{\mathrm{SH}}u_{k,l} - \phi_{,i}^{*}i\check{A}_{ik}^{\mathrm{SH}}\phi_{,k}, \quad (29)$$

$$2\operatorname{Re}\left\{\phi_{,k}i\breve{W}_{ki}^{\operatorname{QH}}\dot{u}_{i}^{*}\right\} \leq -\dot{u}_{i}^{*}i\check{\rho}_{ik}^{\operatorname{SH}}\dot{u}_{k} - \phi_{,i}^{*}i\breve{A}_{ik}^{\operatorname{SH}}\phi_{,k},\qquad(30)$$

and the latter is replaced by

$$-2\operatorname{Re}\left\{\phi_{,k}i\widehat{W}_{ki}^{\mathrm{QSH}}\ddot{u}_{i}^{*}\right\} \leq -\dot{u}_{i}^{*}i\check{\rho}_{ik}^{\mathrm{SH}}\dot{u}_{k} - \phi_{,i}^{*}i\check{A}_{ik}^{\mathrm{SH}}\phi_{,k}, \quad (31)$$

when expressed in terms of $\widehat{\mathbf{W}}$. Equations (29)–(31) thus provide bounds for $\check{B}^{\text{QSH}},\check{W}^{\text{QH}}$ and \widehat{W}^{QSH} and in the lossless case imply that they are null.

IV. RECIPROCITY

Consider a time-invariant piezoelectric body and two arbitrary time-harmonic source distributions and denote these sources and the fields they excite by superscripts 1 and 2, respectively. The body is reciprocal if the power that distribution 1 produces together with the fields excited by distribution 2 is equal to the power that distribution 2 produces together with the fields excited by distribution 1. A schematic illustration of this property is given in Fig. 3.

The principle of reciprocity is independent of the level of isotropy and homogeneity of the body [108]; however, it requires that at each point the symmetry conditions

$$A_{ij}(\mathbf{x}) = A_{ji}(\mathbf{x}), \quad C_{ijkl}(\mathbf{x}) = C_{klij}(\mathbf{x})$$
 (32)

are satisfied [109]. In the homogenization process of a heterogeneous reciprocal body, it is thus required that the resultant effective properties also satisfy the reciprocity relation. Muhlestein et al. [56] have shown that



FIG. 3. The schematics of the reciprocity principle. The source distributions of problems 1 (cyan) and 2 (gray) are illustrated by darts. The resultant rates of the displacement and electric potential fields of problems 1 (blue) and 2 (black) are illustrated by arrows. The body is reciprocal if the power that distribution 1 produces together with the fields excited by distribution 2 (bottom left sketch) is equal to the power that distribution 2 produces together with the fields excited by distribution 1 (bottom right sketch).

Problem 1: $\mathbf{f}^{(1)}, a^{(1)}, \dot{\mathbf{u}}^{(1)}, \dot{\phi}^{(1)}$

this requirement imposes the following conditions on the effective properties of (spatially) *local* Willis materials:

$$\check{\rho}_{ik} = \check{\rho}_{ki}, \quad \check{S}^{\dagger}_{ijk} = \check{S}_{jki}, \quad \check{C}_{ijkl} = \check{C}_{klij}, \quad (33)$$

where the symmetry between \check{S}^{\dagger} and \check{S} is transmitted to the modified couplings, namely,

$$\widehat{S}_{ijk}^{\dagger} = \widehat{S}_{jki}.$$
(34)

Here, we first derive the generalization of these conditions to local materials exhibiting the electromomentum coupling and then analyze the general (nonlocal) case. Our departure point toward this end is the equations that govern the response of the body when subjected to two independent and arbitrary distributions of force, $\{f_i^{(1)}, f_i^{(2)}\}$, and charge densities $\{q^{(1)}, q^{(2)}\}$, namely,

$$\begin{pmatrix} \sigma_{ij,j}^{(1)} - \dot{p}_i^{(1)} \\ D_{j,j}^{(1)} \end{pmatrix} + \begin{pmatrix} f_i^{(1)} \\ -q^{(1)} \end{pmatrix} = \begin{pmatrix} 0_i \\ 0 \end{pmatrix}$$
(35)

and

$$\begin{pmatrix} \sigma_{ij,j}^{(2)} - \dot{p}_i^{(2)} \\ D_{j,j}^{(2)} \end{pmatrix} + \begin{pmatrix} f_i^{(2)} \\ -q^{(2)} \end{pmatrix} = \begin{pmatrix} 0_i \\ 0 \end{pmatrix}.$$
(36)

We denote the resultant displacement and electric potential fields by $\mathbf{w}^{(1)\mathsf{T}} := \left\{ u_i^{(1)}, \phi^{(1)} \right\}$ and $\mathbf{w}^{(2)\mathsf{T}} := \left\{ u_i^{(2)}, \phi^{(2)} \right\}$, respectively. Next, we left multiply Eqs. (35) and (36) by $\dot{\mathbf{w}}^{(2)\mathsf{T}}$ and $\dot{\mathbf{w}}^{(1)\mathsf{T}}$, respectively. The difference between the two products is

$$\sigma_{ij,j}^{(1)}\dot{u}_{i}^{(2)} + \dot{p}_{i}^{(2)}\dot{u}_{i}^{(1)} + \dot{u}_{i}^{(2)}f_{i}^{(1)} + \dot{\phi}^{(2)}D_{j,j}^{(1)} + \dot{\phi}^{(1)}q^{(2)} - \left(\sigma_{ij,j}^{(2)}\dot{u}_{i}^{(1)} + \dot{p}_{i}^{(1)}\dot{u}_{i}^{(2)} + \dot{u}_{i}^{(1)}f_{i}^{(2)} + \dot{\phi}^{(1)}D_{j,j}^{(2)} + \dot{\phi}^{(2)}q^{(1)}\right) = 0,$$
(37)

which can rearranged as

$$\dot{\phi}_{i}^{(1)}\dot{u}_{i}^{(2)} - \dot{p}_{i}^{(2)}\dot{u}_{i}^{(1)} + \sigma_{ij}^{(1)}\dot{u}_{i,j}^{(2)} - \sigma_{ij}^{(2)}\dot{u}_{i,j}^{(1)} + \dot{\phi}_{j}^{(2)}D_{j}^{(1)} - \dot{\phi}_{j}^{(1)}D_{j}^{(2)} = \Delta P, \qquad (38)$$

using the identities

Ì

$$\sigma_{ij,j}^{(1)}\dot{u}_{i}^{(2)} - \sigma_{ij,j}^{(2)}\dot{u}_{i}^{(1)} = \left\{\sigma_{ij}^{(1)}\dot{u}_{i}^{(2)} - \sigma_{ij}^{(2)}\dot{u}_{i}^{(1)}\right\}_{,j} - \left(\sigma_{ij}^{(1)}\dot{u}_{i,j}^{(2)} - \sigma_{ij}^{(2)}\dot{u}_{i,j}^{(1)}\right), \quad (39a)$$

$$\dot{\phi}^{(2)}D_{j,j}^{(1)} - \dot{\phi}^{(1)}D_{j,j}^{(2)} = \left\{\dot{\phi}^{(2)}D_{j}^{(1)} - \dot{\phi}^{(1)}D_{j}^{(2)}\right\}_{,j} - \left(\dot{\phi}_{,j}^{(2)}D_{j}^{(1)} - \dot{\phi}_{,j}^{(1)}D_{j}^{(2)}\right), \quad (39b)$$

where

$$\Delta P = \left\{ \sigma_{ij}^{(1)} \dot{u}_i^{(2)} + D_j^{(1)} \dot{\phi}^{(2)} \right\}_{,j} + f_i^{(1)} \dot{u}_i^{(2)} - q^{(1)} \dot{\phi}^{(2)} - \left\{ \sigma_{ij}^{(2)} \dot{u}_i^{(1)} + D_j^{(2)} \dot{\phi}^{(1)} \right\}_{,j} - \left(f_i^{(2)} \dot{u}_i^{(1)} - q^{(2)} \dot{\phi}^{(1)} \right).$$
(40)

The term ΔP is the differential form of the difference between the power that distribution 1 produces together with the fields excited by distribution 2 and the power that distribution 2 produces together with the fields excited by distribution 1 and hence vanishes if the body is reciprocal. The global form is obtained by volume integration, conversion of the first and third terms in the integral into surface integrals using the divergence theorem, and identification of the boundary sources $t_i^{(l)} = \sigma_{ij}^{(l)} n_j$ and $-w_e^{(l)} = D_j^{(l)} n_j$ of distribution *l*.

We now expand the terms on the left-hand side of Eq. (38) using the effective constitutive Eq. (10) in their spatially local form ($\kappa = 0$) to obtain

$$\begin{split} \left(\check{S}_{ijk} - \check{S}_{kij}^{\dagger}\right) \left(\dot{u}_{k}^{(1)}u_{i,j}^{(2)} - \dot{u}_{k}^{(2)}u_{i,j}^{(1)}\right) + \left(\check{\rho}_{ik} - \check{\rho}_{ki}\right)\dot{u}_{i}^{(2)}\dot{u}_{k}^{(1)} \\ &+ \left(\check{W}_{ik} - \check{W}_{ki}^{\dagger}\right) \left(\dot{u}_{k}^{(1)}\phi_{,i}^{(2)} - \dot{u}_{k}^{(2)}\phi_{,i}^{(1)}\right) \\ &+ \left(\check{A}_{ik} - \check{A}_{ki}\right)\phi_{,k}^{(2)}\phi_{,i}^{(1)} \\ &+ \left(\check{B}_{ijk}^{\dagger} - \check{B}_{kij}\right) \left(\phi_{,k}^{(1)}u_{i,j}^{(2)} - \phi_{,k}^{(2)}u_{i,j}^{(1)}\right) \\ &+ \left(\check{C}_{ijkl} - \check{C}_{klij}\right)u_{k,l}^{(1)}u_{i,j}^{(2)} = 0. \end{split}$$
(41)

The arbitrariness of the sources implies that the strain, electric, and velocity fields are also arbitrary. Accordingly, for Eq. (41) to hold for any $\nabla \mathbf{u}$, $\dot{\mathbf{u}}$, and $\nabla \phi$, the effective constitutive tensors must satisfy

$$\check{A}_{ik} = \check{A}_{ki}, \quad \check{B}_{ijk}^{\dagger} = \check{B}_{kij}, \quad \check{W}_{ki}^{\dagger} = \check{W}_{ik}, \tag{42}$$

in addition to the restrictions given in Eq. (33). It is clear that the modified couplings \widehat{W}^{\dagger} and \widehat{W} exhibit the same symmetry between \check{W}^{\dagger} and \check{W} , such that

$$\widehat{W}_{ki}^{\dagger} = \widehat{W}_{ik}.$$
(43)

In view of Eqs. (33) and (42), we can now revisit the conclusions in Sec. III and replace the conditions on the Hermitian and skew-Hermitian parts of the tensors *in the long-wavelength limit* by the conditions on their real and imaginary parts, respectively.

The foregoing analysis is obtained in the longwavelength limit. We next derive the general result for arbitrary wavelengths and show that the restrictions (33)and (42) are its specialization. This is carried out by showing that if the body is reciprocal, then the governing equations are self-adjoint and in turn so is the Green function, which renders the constitutive operator \mathcal{L} selfadjoint too. The latter property is remarked only in passing by Willis [46,48] and Pernas-Salomón and Shmuel [52] in their respective problems, perhaps because the notion that reciprocity and self-adjointness are closely related is somewhat known [110,111]. However, since the self-adjoint structure of the constitutive operator clearly depends on the definition of the effective description, it is discussed in more detail here.

To proceed, it is useful to employ the formulation of Barnett and Lothe [112], who have formulated the piezoelectric problem in a generalized space using the following definitions:

$$K_{\alpha i\beta j} = \begin{cases} C_{\alpha i\beta j}, & \alpha, \beta \in \{1, 2, 3\}, \\ B_{\alpha i j}^{\mathsf{T}}, & \beta = 4, \, \alpha \in \{1, 2, 3\}, \\ B_{i\beta j}, & \alpha = 4, \, \beta \in \{1, 2, 3\}, \\ -A_{i j}, & \alpha = \beta = 4, \end{cases}$$

$$\Lambda_{\alpha \beta} = \begin{cases} \delta_{\alpha \beta}, & \alpha, \beta \in \{1, 2, 3\}, \\ 0, & \alpha \text{ or } \beta = 4, \end{cases}$$

$$b_{\alpha} = \begin{cases} f_{\alpha}, & \alpha \in \{1, 2, 3\}, \\ -q, & \alpha = 4, \end{cases}$$
(44)

where the range of Latin subscripts is limited to $\{1, 2, 3\}$. The unified governing equations in terms of **K**, **A**, and **b** read, in index notation,

$$\left\{K_{\alpha i\beta j}w_{\beta,j}\right\}_{,i}+\rho\omega^2\Lambda_{\alpha\beta}w_{\beta}=-b_{\alpha},\qquad(45)$$

which defines the components $G_{\beta\gamma}(\mathbf{x}, \mathbf{X})$ of the Green matrix via

$$\left\{K_{\alpha i\beta j}G_{\beta \gamma,j}\right\}_{,i}+\rho\omega^{2}\Lambda_{\alpha\beta}G_{\beta\gamma}=-\delta_{\alpha\gamma}\delta\left(\mathbf{x}-\mathbf{X}\right),\quad(46)$$

where δ (**x** – **X**) is the Dirac delta. Equation (46) spells out explicitly the components of the symbolic Eq. (9) by Pernas-Salomón and Shmuel [52]. In the Appendix, we describe the standard procedure to obtain the adjoint equations and the corresponding adjoint Green tensor and verify the components of this tensor satisfy

$$G^{\dagger}_{\gamma\beta}\left(\mathbf{x},\mathbf{X}\right) = G^{*}_{\beta\gamma}\left(\mathbf{X},\mathbf{x}\right); \qquad (47)$$

we thus recover the known result that if the body satisfies $\rho^* = \rho$ and $K^{\dagger}_{\alpha i \beta j} = K_{\alpha i \beta j}$, where

$$K_{\alpha i\beta j}^{\dagger} = \begin{cases} C_{\alpha i\beta j}^{\mathsf{T}_{\ast}}, & \alpha, \beta \in \{1, 2, 3\}, \\ B_{\alpha i}^{\mathsf{T}_{\ast}}, & \beta = 4, \ \alpha \in \{1, 2, 3\}, \\ B_{i\beta j}^{\ast}, & \alpha = 4, \ \beta \in \{1, 2, 3\}, \\ -A_{ij}^{\mathsf{T}_{\ast}}, & \alpha = \beta = 4, \end{cases}$$
(48)

which is the case by virtue of Eq. (4), then the piezoelectric problem is self-adjoint [113,114]. As explained in the Appendix, in this case $G_{\gamma\beta}^{\dagger}(\mathbf{x}, \mathbf{X}) = G_{\gamma\beta}(\mathbf{x}, \mathbf{X})$, which, together with the previous result, implies that

$$G_{\gamma\beta}\left(\mathbf{x},\mathbf{X}\right) = G_{\beta\gamma}^{*}\left(\mathbf{X},\mathbf{x}\right).$$
(49)

We next recall the expression for the kernel \tilde{L} obtained by Pernas-Salomón and Shmuel [52], namely,

$$\tilde{\mathsf{L}} = \langle \mathsf{L} \rangle - \left\langle \mathsf{L}\mathsf{B}(\mathsf{B}\mathsf{G})^{\mathsf{T}}\mathsf{L} \right\rangle + \left\langle \mathsf{L}\mathsf{B}\mathsf{G}^{\mathsf{T}} \right\rangle \langle \mathsf{G} \rangle^{-\mathsf{T}} \left\langle \left(\mathsf{B}\mathsf{G}\right)^{\mathsf{T}}\mathsf{L} \right\rangle;$$
(50)

the symbolic matrix formulation for \tilde{L} translates to the following components:

$$\tilde{L}_{\alpha i \beta j} = \begin{cases} \tilde{C}_{\alpha i \beta j}, & \alpha, \beta \in \{1, 2, 3\}, \\ \tilde{B}_{\alpha i j}^{\dagger}, & \alpha \in \{1, 2, 3\}, \beta = 4, \\ \tilde{B}_{i \beta j}, & \alpha = 4, \beta \in \{1, 2, 3\}, \\ -\tilde{A}_{i j}, & \alpha = \beta = 4, \\ \tilde{S}_{i \beta j}, & \alpha \in \{1, 2, 3\}, \beta = 5, \\ \tilde{S}_{\alpha i j}^{\dagger}, & \alpha = 5, \beta \in \{1, 2, 3\}, \\ \tilde{W}_{i j}, & \alpha = 4, \beta = 5, \\ \tilde{W}_{i j}^{\dagger}, & \alpha = 5, \beta = 4, \\ \tilde{\rho}_{i j}, & \alpha = \beta = 5. \end{cases}$$
(51)

Through inspection of the components of Eq. (50) and employment of the symmetries of G and K and the fact that $L = L^*$, we verify the symmetry

$$\tilde{L}_{\alpha i\beta j}\left(\mathbf{x},\mathbf{X}\right)=\tilde{L}_{\beta j\,\alpha i}\left(\mathbf{X},\mathbf{x}\right),\tag{52}$$

where terms associated with the conventional couplings $\tilde{\mathbf{A}}$, $\tilde{\mathbf{B}}$, $\tilde{\mathbf{B}}^{\dagger}$, $\tilde{\mathbf{C}}$, and $\tilde{\boldsymbol{\rho}}$ and the modified couplings $\widehat{\mathbf{W}}$, $\widehat{\mathbf{W}}^{\dagger}$, $\widehat{\mathbf{S}}$, and $\widehat{\mathbf{S}}^{\dagger}$ also satisfy

$$\tilde{L}_{\alpha i\beta j}\left(\mathbf{x},\mathbf{X}\right) = \tilde{L}^{*}_{\beta j\,\alpha i}\left(\mathbf{X},\mathbf{x}\right),\tag{53}$$

while the couplings of Willis type in their original form satisfy

$$\tilde{L}_{\alpha i \beta j} (\mathbf{x}, \mathbf{X}) = -\tilde{L}^*_{\beta j \alpha i} (\mathbf{X}, \mathbf{x}),$$

 $\alpha \in \{1, 2, 3, 4\}, \ \beta = 5, \text{ and } \beta \in \{1, 2, 3, 4\}, \ \alpha = 5.$
(54)

It is important to note that the symmetry Eq. (52)—which delivers the self-adjoint property of \tilde{L} as explained later—originates from the symmetry $L = L^{T}$ and does not require the composite to be lossless; symmetries (53) and (54) originate from the assumption that L is also real.

Interestingly, the modified cross-couplings $\widehat{\mathbf{W}}$, $\widehat{\mathbf{W}}^{\dagger}$, $\widehat{\mathbf{S}}$, and $\widehat{\mathbf{S}}^{\dagger}$ are related via same symmetry as the conventional couplings, i.e., Eq. (53). The combination of Eqs. (52)–(54) implies that the conventional couplings and the modified cross-couplings are real, while those of Willis type are pure imaginary. Together with fact that according to Eq. (46) the Green tensor is an even function of ω [115], this result implies that in the space-time domain the conventional couplings $\tilde{\mathbf{A}}$, $\tilde{\mathbf{B}}$, $\tilde{\mathbf{B}}^{\dagger}$, and $\tilde{\mathbf{C}}$, $\tilde{\boldsymbol{\rho}}$ and the coupling of Willis type $\widehat{\mathbf{S}}, \widehat{\mathbf{S}}^{\dagger}, \widehat{\mathbf{W}}$, and $\widehat{\mathbf{W}}^{\dagger}$ are real—as they should be, since they relate real physical quantities. By contrast, in the space-time domain, the cross-couplings \tilde{S} , \tilde{S}^{\dagger} , \tilde{W} , and \mathbf{W}^{\dagger} are pure imaginary—an unphysical result. This observation agrees with the analysis of Norris et al. [68] in the purely elastic case, who have shown that $\tilde{\mathbf{C}}$ and $\tilde{\boldsymbol{\rho}}$ are real in the space-time domain, while \tilde{S} and \tilde{S}^{\dagger} are pure imaginary.

For statically homogeneous media, we can employ the Fourier transform with respect to the translation $\mathbf{x} - \mathbf{X}$ and write these symmetries using indices in the transformed domain as

$$\check{C}_{klij}^{*}\left(\boldsymbol{\kappa},\omega\right)=\check{C}_{ijkl}\left(\boldsymbol{\kappa},\omega\right)=\check{C}_{klij}\left(-\boldsymbol{\kappa},\omega\right),\qquad(55a)$$

$$\check{B}_{kij}^{*}\left(\boldsymbol{\kappa},\omega\right)=\check{B}_{ijk}^{\dagger}\left(\boldsymbol{\kappa},\omega\right)=\check{B}_{kij}\left(-\boldsymbol{\kappa},\omega\right),$$
(55b)

$$\check{A}_{ji}^{*}(\boldsymbol{\kappa},\omega) = \check{A}_{ij}(\boldsymbol{\kappa},\omega) = \check{A}_{ji}(-\boldsymbol{\kappa},\omega), \qquad (55c)$$

$$\check{\rho}_{ji}^{*}(\boldsymbol{\kappa},\omega) = \check{\rho}_{ij}(\boldsymbol{\kappa},\omega) = \check{\rho}_{ji}(-\boldsymbol{\kappa},\omega), \qquad (55d)$$

for terms associated with conventional couplings; the modified cross-couplings terms satisfy the same form of symmetries, such that

$$\widehat{S}_{jki}^{*}(\boldsymbol{\kappa},\omega) = \widehat{S}_{ijk}^{\dagger}(\boldsymbol{\kappa},\omega) = \widehat{S}_{jki}(-\boldsymbol{\kappa},\omega), \qquad (56a)$$

$$\widehat{W}_{ji}^{*}(\boldsymbol{\kappa},\omega) = \widehat{W}_{ji}^{\dagger}(\boldsymbol{\kappa},\omega) = \widehat{W}_{ji}(-\boldsymbol{\kappa},\omega), \qquad (56b)$$

while when they are in their original form, they satisfy

$$-\check{S}_{jki}^{*}(\boldsymbol{\kappa},\omega)=\check{S}_{ijk}^{\dagger}(\boldsymbol{\kappa},\omega)=\check{S}_{jki}\left(-\boldsymbol{\kappa},\omega\right),\qquad(57a)$$

$$-\check{W}_{ji}^{*}(\boldsymbol{\kappa},\omega)=\check{W}_{ij}^{\dagger}(\boldsymbol{\kappa},\omega)=\check{W}_{ji}(-\boldsymbol{\kappa},\omega).$$
 (57b)

The symmetry between $\tilde{\mathbf{W}}$ and $\tilde{\mathbf{W}}^{\dagger}$ is shown in detail in the Appendix. It is clear that in the limit $\kappa = \mathbf{0}$, the symmetries (55) and (57) recover symmetries (33) and (42). Equations (55)–(57) also endow the adjoint notion to the

symbol \dagger for the nonlocal operators \mathcal{S}^{\dagger} and $\mathcal{W}^{\dagger},$ since these symmetries imply that

$$\int_{\Omega} \mathcal{S} \left(\dot{\mathbf{u}} \left(\mathbf{X} \right) \right) : \nabla \mathbf{u} \left(\mathbf{x} \right) d\mathbf{x} = \int_{\Omega} \dot{\mathbf{u}} \left(\mathbf{x} \right) \cdot \mathcal{S}^{\dagger} \left(\nabla \mathbf{u} \left(\mathbf{X} \right) \right) d\mathbf{x},$$
(58a)

$$\int_{\Omega} \mathcal{W}(\dot{\mathbf{u}}(\mathbf{X})) \cdot \nabla \phi(\mathbf{x}) d\mathbf{x} = \int_{\Omega} \dot{\mathbf{u}}(\mathbf{x}) \cdot \mathcal{W}^{\dagger}(\nabla \phi(\mathbf{X})) d\mathbf{x},$$
(58b)

as well as for \mathcal{B}^{\dagger} , which satisfies

$$\int_{\Omega} \mathcal{B} \left(\nabla \mathbf{u} \left(\mathbf{X} \right) \right) \cdot \nabla \phi \left(\mathbf{x} \right) d\mathbf{x} = \int_{\Omega} \nabla \mathbf{u} \left(\mathbf{x} \right) \cdot \mathcal{B}^{\dagger} \left(\nabla \phi \left(\mathbf{X} \right) \right) d\mathbf{x};$$
(59)

the nonlocal operators \mathcal{R} , \mathcal{A} , and \mathcal{C} are self-adjoint in the above sense. At the cost of repetition, we clarify that the equality between the middle and rightmost terms in Eqs. (55)–(57) is obtained by relying only on the fact that $L = L^{T}$. This property leads to a self-adjoint effective operator \mathcal{L} in the above sense and, specifically, renders \mathcal{S}^{\dagger} and \mathcal{W}^{\dagger} (respectively, $\widehat{\mathcal{S}}^{\dagger}$ and $\widehat{\mathcal{W}}^{\dagger}$) the adjoints of \mathcal{S} and \mathcal{W} (respectively, $\widehat{\mathcal{S}}$ and $\widehat{\mathcal{W}}$); the same goes for \mathcal{B}^{\dagger} and \mathcal{B} via Eq. (59). Otherwise, it would by justified to replace the \dagger notation to distinguish them from the adjoint operators.

We further clarify that the equality between the lefthand and middle terms in Eqs. (55)–(57) relies only on the fact that in the frequency domain, the properties of the composite satisfy $\check{L} = \check{L}^{T*}$, or in other words they are Hermitian. This *does not* necessarily imply that L is symmetric (although it can be) and it immediately satisfies Eqs. (24)–(26) as equalities. A case in which $\check{L} = \check{L}^{T*}$ and $\check{L} \neq \check{L}^{T}$ corresponds to a nonreciprocal medium, the losses of which are compensated by the energy it generates, such that on average the material is lossless and passive (no energy loss or gain).

V. CAUSALITY

The principle of causality states that an effect must follow its cause. This principle implies the analyticity of the response functions of linear systems and vice versa, namely, analyticity implies causality [116,117]. With the interpretation of the constitutive properties of linear materials as response functions, causality through analyticity provides relations between the real and imaginary parts of their (time) Fourier transforms. These relations were first obtained in electromagnetics for the permeability and permittivity tensors, where they are known as the Kramers-Kronig relations [88–91]. This concept was applied later on in other branches of physics—and specifically in mechanics—to obtain conditions on the pertinent constitutive properties [55,118]. Alù [57,67] has shown that in certain cases the bianisotropic tensor is essential for respecting causality in passive media. (We clarify that the model Alù considered is local.) Indeed, some of the electromagnetic homogenization schemes from which this cross-coupling tensor is absent violate causality in such media [1,119,120]. Analogously, Sieck *et al.* [69] have recognized the need in Willis coupling to satisfy causality by the effective constitutive properties in elastodynamics. We clarify that the information from the Kramers-Kronig relations is limited for active media, since it is possible to realize anomalous responses for real frequencies (such as antiresonance) using suitable causal polynomials (see, e.g., Ref. [121]). To get useful results, it is thus necessary to couple causality with passivity [122].

In this section, we develop the restrictions placed by causality on the effective properties (3), i.e., when, microscopically, the medium exhibits the intrinsic piezoelectric effect, and macroscopically also exhibits the effective electromomentum coupling. The framework developed in Ref. [52] constitutes a platform to carry out this task with respect to the effective operator \tilde{L} , which we recall is non-local both in space and time. To facilitate the analysis, here we focus on the long-wavelength limit $\kappa = 0$ and neglect spatially nonlocal effects on causality [123]. Nevertheless, we recall that according to Landau and Lifshitz [89], the resultant equations should apply for any fixed value of κ . Accordingly, we omit the spatial dependency of the fields in Eq. (8) and rewrite it as

$$\mathbf{h}(t) = \int_{-\infty}^{t} \tilde{\mathbf{L}}(t-T) \mathbf{g}(T) dT, \qquad (60)$$

bearing in mind that it holds at each material point. Let $\tau = t - T$; causality implies that \tilde{L} must satisfy

$$L(\tau) = 0, \text{ for } \tau < 0.$$
 (61)

The approach taken here to relate the real and imaginary parts of its Fourier transform is standard and employs the Plemelj formulas, see, e.g., Ref. [124]. This approach is summarized next to provide a self-contained analysis. As discussed by Nussenzveig [124], certain assumptions regarding $\tilde{L}(t)$ are required in order for $\check{L}'(\omega)$ and $\check{L}''(\omega)$ to be related, where we recall that ' and " denote the real and imaginary parts of a variable, respectively. In view of Eq. (61), Eq. (11) takes the form

$$\check{\mathsf{L}}(\omega) = \int_0^\infty \tilde{\mathsf{L}}(\tau) e^{i\omega\tau} d\tau, \qquad (62)$$

where the integral is only over \mathbb{R}_+ , implying that $\check{\mathsf{L}}(\omega)$ has an analytic continuation in the upper half of the complex plane. If we further assume at first that $\check{\mathsf{L}}(t)$ is square integrable, then through the Parseval-Plancherel theorem, we have that $\check{L}(\omega)$ is square integrable along any line in the upper half of the complex plane that is parallel to the real axis, such that [124]

$$\lim_{\alpha \to \pm \infty} \check{\mathsf{L}} \left(\omega' + i\omega'' \right) = 0, \quad \omega'' \ge 0.$$
 (63)

This property is employed in the application of Cauchy's integral formula to a closed curve Γ about an arbitrary point ω_0 in the upper half of the complex plane, $(\omega_0'' > 0)$

$$\check{\mathsf{L}}(\omega_0) = \frac{1}{2\pi i} \oint_{\Gamma} \frac{\check{\mathsf{L}}(z)}{z - \omega_0} dz, \tag{64}$$

in order to show that it reduces to an integration along the real axis:

$$\check{\mathsf{L}}(\omega_0) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{\check{\mathsf{L}}(z)}{z - \omega_0} dz, \quad z'' = 0.$$
(65)

The case of real ω_0 is obtained using a closed contour that avoids ω_0 by a semicircle of radius ϵ and, taking the limit as $\epsilon \to 0$, to show that

$$\check{\mathsf{L}}(\omega_0) = \frac{1}{i\pi} \int_{-\infty}^{\infty} \frac{\check{\mathsf{L}}(z)}{z - \omega_0} dz, \quad z'' = \omega_0'' = 0, \qquad (66)$$

where

$$\int_{-\infty}^{\infty} = \lim_{\epsilon \to 0} \left(\int_{-\infty}^{\omega - \epsilon} + \int_{\omega + \epsilon}^{\infty} \right)$$
(67)

denotes the Cauchy principal value. The real and imaginary parts of Eq. (66) provide the following relations:

$$\check{\mathsf{L}}'(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\check{\mathsf{L}}''(z)}{z - \omega} dz, \qquad (68a)$$

$$\check{\mathsf{L}}''(\omega) = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\check{\mathsf{L}}'(z)}{z-\omega} dz$$
(68b)

between the $\check{L}'(\omega)$ and $\check{L}''(\omega)$ at any real frequency ω . An alternative form of the relations given in Eq. (68) is obtained using the fact that $\tilde{L}(t)$ is real, and hence

$$\check{\mathsf{L}}^*(\omega) = \left(\int_0^\infty \tilde{\mathsf{L}}(t) \, e^{i\omega t} dt\right)^* = \int_0^\infty \tilde{\mathsf{L}}(t) \, e^{-i\omega t} dt = \check{\mathsf{L}}(-\omega),$$
(69)

leading to

$$\check{\mathsf{L}}'(\omega) = \check{\mathsf{L}}'(-\omega), \qquad (70a)$$

$$\check{\mathsf{L}}''(\omega) = -\check{\mathsf{L}}''(-\omega). \tag{70b}$$

The employment of these symmetries leads to

$$\check{\mathsf{L}}'(\omega) = \frac{2}{\pi} \int_0^\infty \frac{z \check{\mathsf{L}}''(z)}{z^2 - \omega^2} dz,$$
(71a)

$$\check{\mathsf{L}}''(\omega) = -\frac{2}{\pi} \int_0^\infty \frac{\omega \check{\mathsf{L}}'(z)}{z^2 - \omega^2} dz.$$
(71b)

We next examine the case in which $\check{L}(\omega)$ is not square integrable but a bounded function. In this case, property (63), which is needed to obtain Eq. (65), no longer holds and consequently the relation between $\check{L}'(\omega)$ and $\check{L}''(\omega)$ can be determined only up to an arbitrary real constant. To determine this constant, a knowledge of the value of $\check{L}(\omega)$ at some real frequency is needed. Let us say that $\check{L}(\omega)$ is differentiable and known at ω_0 ; then, we can repeat the procedure that led to Eqs. (68) and (71), except that now we replace $\check{L}(\omega)$ by the function

$$\frac{\Delta \check{\mathbf{L}}}{\Delta \omega}(\omega) := \frac{\check{\mathbf{L}}(\omega) - \check{\mathbf{L}}(\omega_0)}{\omega - \omega_0},\tag{72}$$

since it is bounded for $\omega \to \omega_0$, analytic in the upper half of the complex plane *and square integrable*. If we further assume that $\omega_0 \to \infty$, the end result can be put in the form

$$\check{\mathsf{L}}'(\omega) = \frac{2}{\pi} \int_0^\infty \frac{z \check{\mathsf{L}}''(z)}{z^2 - \omega^2} dz + \check{\mathsf{L}}'(\infty), \qquad (73a)$$

$$\check{\mathsf{L}}''(\omega) = -\frac{2}{\pi} \int_0^\infty \frac{\omega \check{\mathsf{L}}'(z)}{z^2 - \omega^2} dz, \qquad (73b)$$

cf. Eq. (4.7) in Ref. [56]. Equation (73) should hold for all the tensor-valued (history) functions that comprise $\check{L}(\omega)$, i.e., \tilde{A} , \tilde{B} , \tilde{B}^{\dagger} , and \tilde{C} , as well as \tilde{S} , \tilde{W} , their adjoints, and their modified versions \hat{S} and \hat{W} . This conclusion thus generalizes previous works by requiring that the electromomentum tensor is also subjected to relations of the Kramer-Kronig type, as expected. We recall that in Sec. IV show that the Fourier transforms of the constitutive tensors of reciprocal, passive, and lossless media are all real, including the modified cross-couplings \hat{S} , \hat{S}^{\dagger} , and \hat{W} and \hat{W}^{\dagger} , and excluding \check{S} , \tilde{S}^{\dagger} , \check{W} , and \check{W}^{\dagger} , which are pure imaginary. When this conclusion is combined with conditions (73), we obtain in the long-wavelength limit of reciprocal, passive, and lossless media that

$$\check{\mathbf{A}}(\omega) = \check{\mathbf{A}}'(\infty), \, \check{\mathbf{B}}(\omega) = \check{\mathbf{B}}'(\infty),
\check{\mathbf{B}}^{\dagger}(\omega) = \check{\mathbf{B}}^{\dagger\prime}(\infty), \, \check{\mathbf{C}}(\omega) = \check{\mathbf{C}}'(\infty),
\check{\boldsymbol{\rho}}(\omega) = \check{\boldsymbol{\rho}}'(\infty),$$
(74)

and, similarly, for the modified cross-couplings $\widehat{S},\,\widehat{S}^{\dagger},\,\widehat{W},$ and $\widehat{W}^{\dagger},$

$$\widehat{\mathbf{S}}(\omega) = \widehat{\mathbf{S}}'(\infty), \quad \widehat{\mathbf{S}}^{\dagger}(\omega) = \widehat{\mathbf{S}}^{\dagger\prime}(\infty),
\widehat{\mathbf{W}}(\omega) = \widehat{\mathbf{W}}'(\infty), \quad \widehat{\mathbf{W}}^{\dagger}(\omega) = \widehat{\mathbf{W}}^{\dagger\prime}(\infty),$$
(75)

where notably, the couplings of Willis type in their original representation must be null. This is clear from Eq. (73b), as the integrand on the right-hand side is identically zero since these couplings are pure imaginary, which then implies that the left-hand side—which is their imaginary part—also vanishes.

VI. CLOSURE

Piezoelectric and piezomagnetic materials exhibit intrinsic coupling with nonmechanical fields. Recently, it has been shown that the effective response of composites made of such constituents exhibits additional cross-couplings that are absent from the response of the constituents and are of Willis type [52]. The recent development of such generalized Willis materials comes with the question: What are the mathematical restrictions that their constitutive relations should satisfy in order to respect passivity, reciprocity, and causality? In this paper, we address this question by adapting standard methodologies used in electromagnetics, elastodynamics, and mathematics [55,56, 105,124].

We arrive at the following findings. From passivity, we obtain several inequality conditions on the skew-Hermitian and Hermitian parts of the Fourier transforms of the effective properties. From reciprocity, we find certain symmetry and adjoint relations that the effective operator satisfies. These conditions generalize the conditions in Refs. [55,56] for the Milton-Briane-Willis equations (i.e., the long-wavelength limit of the Willis equations), not only by accounting for the electromomentum coupling but also for nonlocal interactions, leading to wave-vector-dependent conditions. Finally, from causality, we obtain relations of the Kramer-Kronig type between the real and imaginary parts of the operator in the frequency domain. A summary of the mathematical restrictions is given in Table I. One implication that follows these restrictions is that the additional cross-couplings in the time domain are not with the electric field and velocity but with their time derivative. This insight is analogous to the insights in Refs. [56,59,60,69], suggesting acceleration-dependent stress and strain rate-dependent momentum formulation in the elastic case.

We conclude this paper by highlighting the applicational impact of our results. Our conclusions assess how energy is converted in such metamaterials and, in turn, what is the efficiency that devices based on these cross-couplings can achieve. Indeed, the counterpart of our conclusions in the acoustic setting [55–57] has guided Quan *et al.* [58] in the design of metasurfaces with maximum Willis coupling for sound steering. Similarly, we expect this work to promote the design of devices that exploit the electromomentum coupling to efficiently manipulate mechanical waves.

ACKNOWLEDGMENTS

We thank Graeme Milton for enriching discussions on nonlocality and uniqueness, his insights on causality, and for useful references. We thank Doron Shilo for his observation regarding the similarity with the broken inversion symmetry at the atomic scale in piezoelectric materials. This research was supported by the Israel Science Foundation, funded by the Israel Academy of Sciences and Humanities (Grant No. 2061/20), the United States–Israel Binational Science Foundation (Grant No. 2014358), and the Ministry of Science and Technology (Grant No. 880011).

APPENDIX: DERIVATIONS RELATED TO THE ADJOINT OPERATOR AND GREEN TENSOR

The left-hand side of Eq. (45) defines the action of an operator \mathcal{M} on the vector field w. Accordingly, the left-hand side of Eq. (45) is the α component of the action of \mathcal{M} on the vector field $G_{.(\gamma)}$, the components of which are $G_{\beta(\gamma)}, \beta = 1, 2, 3, 4$. The adjoint operator \mathcal{M}^{\dagger} is defined via the Green identity

$$\left\langle \mathcal{M}\left(\mathbf{w}\right),\mathbf{v}\right\rangle _{\Omega}-\left\langle \mathbf{w},\mathcal{M}^{\dagger}\left(\mathbf{v}\right)\right\rangle _{\Omega}=\mathsf{B}.\mathsf{T}.,$$
 (A1)

where

$$\left\langle \mathcal{M}\left(\mathbf{w}\right),\mathbf{v}\right\rangle _{\Omega}:=\int_{\Omega}\mathcal{M}\left(\mathbf{w}\right)\cdot\mathbf{v}^{*}d\mathbf{x}=\int_{\Omega}\left\{ \mathcal{M}\left(\mathbf{w}\right)\right\} _{\alpha}v_{\alpha}^{*}d\mathbf{x}$$
(A2)

and B.T. denotes boundary terms associated with a surface integral of some bilinear function of w, v, and their derivatives, where the superscript * denotes the complex-conjugate operation. Setting $w = w^{(1)}$ and $v = w^{(2)}$, we obtain

$$\begin{split} \left\langle \mathcal{M}\left(\mathbf{w}\right),\mathbf{v}\right\rangle_{\Omega} \\ &= \int_{\Omega} \left(\left\{ C_{ijkl} u_{k,l}^{(1)} + B_{ijk}^{\mathsf{T}} \phi_{,k}^{(1)} \right\}_{j} u_{i}^{(2)*} + \left\{ B_{ijk} u_{k,l}^{(1)} - A_{ij} \phi_{,j}^{(1)} \right\}_{,i} \phi^{(2)*} + \omega^{2} \rho u_{i}^{(1)} u_{i}^{(2)*} \right) d\mathbf{x} \\ &= \int_{\Omega} \left(u_{k}^{(1)} \left\{ C_{ijkl} u_{i,j}^{(2)*} + B_{kji}^{\mathsf{T}} \phi_{,i}^{(2)*} \right\}_{,j} + \phi^{(1)} \left\{ B_{ijk} u_{j,k}^{(2)*} - A_{ij} \phi_{,i}^{(2)*} \right\}_{,i} + u_{i}^{(1)} \omega^{2} \rho u_{i}^{(2)*} \right) d\mathbf{x} \\ &= \int_{\Omega} \left(u_{k}^{(1)} \left\{ C_{klij}^{\mathsf{T}} u_{i,j}^{(2)*} + B_{kji}^{\mathsf{T}} \phi_{,i}^{(2)*} \right\}_{,j} + \phi^{(1)} \left\{ B_{ijk} u_{j,k}^{(2)*} - A_{ji}^{\mathsf{T}} \phi_{,i}^{(2)*} \right\}_{,i} + u_{i}^{(1)} \omega^{2} \rho u_{i}^{(2)*} \right) d\mathbf{x} \\ &= \int_{\Omega} \left(u_{k}^{(1)} \left\{ C_{klij}^{\mathsf{T}*} u_{i,j}^{(2)} + B_{kji}^{\mathsf{T}*} \phi_{,i}^{(2)} \right\}_{,j}^{*} + \phi^{(1)} \left\{ B_{ijk}^{*} u_{j,k}^{(2)} - A_{ji}^{\mathsf{T}*} \phi_{,i}^{(2)} \right\}_{,i}^{*} + u_{i}^{(1)} \left\{ \omega^{2} \rho^{*} u_{i}^{(2)} \right\}^{*} \right) d\mathbf{x} \\ &= \left\langle \mathbf{w}, \mathcal{M}^{\dagger} \left(\mathbf{v} \right) \right\rangle_{\Omega}, \end{split}$$
(A3)

using integration by parts, where the boundary terms that result in the process are indeed bilinear functions of w and v and are omitted from Eq. (A3) for brevity. This identifies \mathcal{M}^{\dagger} with the adjoint equations

$$\left\{K_{\alpha i\beta j}^{\dagger}w_{\beta,j}\right\}_{,i} + \rho^{*}\omega^{2}\Lambda_{\alpha\beta}w_{\beta} = m_{\alpha},\tag{A4}$$

$$K_{\alpha i \beta j}^{\dagger} = \begin{cases} C_{\alpha i \beta j}^{\mathsf{T}_{*}}, & \alpha, \beta \in \{1, 2, 3\}, \\ B_{\alpha i \beta}^{\mathsf{T}_{*}}, & \beta = 4, \ \alpha \in \{1, 2, 3\}, \\ B_{i \beta j}^{*}, & \alpha = 4, \ \beta \in \{1, 2, 3\}, \\ -A_{i j}^{\mathsf{T}_{*}}, & \alpha = \beta = 4. \end{cases}$$
(A5)

Accordingly, the components of the adjoint Green matrix $G^{\dagger}_{\beta_{Y}}(\mathbf{x}, \mathbf{X})$ are defined by

$$\left\{K_{\alpha i\beta j}^{\dagger}G_{\beta \gamma, j}^{\dagger}\right\}_{,i} + \rho^* \omega^2 \Lambda_{\alpha \beta}G_{\beta \gamma}^{\dagger} = -\delta_{\alpha \gamma}\delta\left(\mathbf{x} - \mathbf{X}\right).$$
(A6)

Following the standard procedure, we set $w_{\alpha}(\boldsymbol{\chi}, \mathbf{x}) = G_{\alpha(\gamma)}(\boldsymbol{\chi}, \mathbf{x})$ and $v_{\alpha}(\boldsymbol{\chi}, \mathbf{X}) = G_{\alpha(\beta)}^{\dagger}(\boldsymbol{\chi}, \mathbf{X})$, and employ Eqs. (45)–(A1), (A4) and (A6) to show that

$$\begin{split} \left\langle \mathcal{M} \left(\mathbf{w} \right), \mathbf{v} \right\rangle_{\Omega} &- \left\langle \mathbf{w}, \mathcal{M}^{\dagger} \left(\mathbf{v} \right) \right\rangle_{\Omega} \\ &= \int_{\Omega} \left\{ \mathcal{M} G_{.(\gamma)} \left(\mathbf{\chi}, \mathbf{x} \right) \right\}_{\alpha} G_{\alpha(\beta)}^{\dagger *} \left(\mathbf{\chi}, \mathbf{X} \right) d\mathbf{\chi} \\ &- \int_{\Omega} G_{\alpha(\gamma)} \left(\mathbf{\chi}, \mathbf{x} \right) \left\{ \mathcal{M}^{\dagger} G_{\alpha(\beta)}^{\dagger} \left(\mathbf{\chi}, \mathbf{X} \right) \right\}^{*} d\mathbf{\chi} \\ &= \int_{\Omega} \delta_{\alpha\gamma} \delta \left(\mathbf{\chi} - \mathbf{x} \right) G_{\alpha(\beta)}^{\dagger *} \left(\mathbf{\chi}, \mathbf{X} \right) d\mathbf{\chi} \\ &- \int_{\Omega} G_{\alpha(\gamma)} \left(\mathbf{\chi}, \mathbf{x} \right) \delta_{\alpha\beta} \delta \left(\mathbf{\chi} - \mathbf{X} \right) d\mathbf{\chi} \\ &= G_{\gamma(\beta)}^{\dagger *} \left(\mathbf{x}, \mathbf{X} \right) - G_{\beta(\gamma)} \left(\mathbf{X}, \mathbf{x} \right) = 0, \end{split}$$
(A7)

and hence $G_{\gamma\beta}^{\dagger}(\mathbf{x}, \mathbf{X}) = G_{\beta\gamma}^{*}(\mathbf{X}, \mathbf{x})$. If the body satisfies $K_{\alpha i\beta j}^{\dagger} = K_{\alpha i\beta j}$ and $\rho^{*} = \rho$ —which is the case by virtue of Eq. (4)—then the problem is self-adjoint, as Eq. (45) is identical to Eq. (A4). In this case $\mathcal{M} = \mathcal{M}^{\dagger}$, hence $G_{\gamma\beta}^{\dagger}(\mathbf{x}, \mathbf{X}) = G_{\gamma\beta}(\mathbf{x}, \mathbf{X})$, which, together with the previous result, implies that

$$G_{\gamma\beta}\left(\mathbf{x},\mathbf{X}\right) = G^{*}_{\beta\gamma}\left(\mathbf{X},\mathbf{x}\right). \tag{A8}$$

As discussed in Sec. IV, the symmetries of G are required in showing that \tilde{L} —which is a function of G—satisfies the symmetries that are given by Eqs. (52)–(54), i.e., it is selfadjoint. The components of \tilde{L} involve lengthy expressions, which we omit here. We choose, however, to provide an expression for \check{W} and \check{W}^{\dagger} and show the symmetry that we report in the body of the paper. We begin by writing the result for \check{W}^{\dagger} from Eq. (50) as

$$\tilde{\mathbf{W}}^{\dagger}(\mathbf{x}, \mathbf{X}) = -\boldsymbol{\alpha}_{32} + \boldsymbol{\gamma}_{32}, \tag{A9}$$

where $\boldsymbol{\alpha}_{32}$ and $\boldsymbol{\gamma}_{32}$ are the (3,2) entries of the symbolic 3×3 block matrices $\boldsymbol{\alpha} = \langle LB(BG)^{T}L \rangle$ and $\boldsymbol{\gamma} = \langle LBG^{T} \rangle \langle G \rangle^{-T} \langle (BG)^{T}L \rangle$, which read

$$\boldsymbol{\alpha}_{32}(\mathbf{x}, \mathbf{X}) = s \left\langle \rho(\mathbf{X}) \left(\nabla_{\mathbf{X}} \mathbf{G}_{11}^{\mathsf{T}} \right) \mathbf{B}^{\mathsf{T}}(\mathbf{X}) \right\rangle - s \left\langle \rho(\mathbf{x}) \left(\nabla_{\mathbf{X}} \mathbf{G}_{21}^{\mathsf{T}} \right) \mathbf{A}(\mathbf{X}) \right\rangle, \qquad (A10)$$

$$\begin{split} \boldsymbol{\gamma}_{32}(\mathbf{x}, \mathbf{X}) &= s \left\langle \rho(\mathbf{x}) \mathbf{G}_{11}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{11}^{\mathsf{T}} \right) \mathbf{B}^{\mathsf{T}}(\mathbf{X}) \right\rangle - s \left\langle \rho(\mathbf{x}) \mathbf{G}_{11}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{21}^{\mathsf{T}} \right) \mathbf{A}(\mathbf{X}) \right\rangle \\ &+ s \left\langle \rho(\mathbf{x}) \mathbf{G}_{21}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{12}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{11}^{\mathsf{T}} \right) \mathbf{B}^{\mathsf{T}}(\mathbf{X}) \right\rangle - s \left\langle \rho(\mathbf{x}) \mathbf{G}_{21}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{12}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{21}^{\mathsf{T}} \right) \mathbf{A}(\mathbf{X}) \right\rangle \\ &+ s \left\langle \rho(\mathbf{x}) \mathbf{G}_{11}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{21}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{12}^{\mathsf{T}} \right) \mathbf{B}^{\mathsf{T}}(\mathbf{X}) \right\rangle - s \left\langle \rho(\mathbf{x}) \mathbf{G}_{11}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{21}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{22}^{\mathsf{T}} \right) \mathbf{A}(\mathbf{X}) \right\rangle \\ &+ s \left\langle \rho(\mathbf{x}) \mathbf{G}_{21}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{22}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{12}^{\mathsf{T}} \right) \mathbf{B}^{\mathsf{T}}(\mathbf{X}) \right\rangle - s \left\langle \rho(\mathbf{x}) \mathbf{G}_{21}^{\mathsf{T}} \right\rangle \left\langle \mathbf{V}_{22}^{\mathsf{T}} \right\rangle \left\langle \left(\nabla_{\mathbf{X}} \mathbf{G}_{22} \right) \mathbf{A}(\mathbf{X}) \right\rangle, \end{split}$$
(A11)

where

$$\mathbf{V} \equiv \mathbf{G}^{-1} = \left(\begin{array}{cc} \mathbf{V}_{11} & \mathbf{V}_{12} \\ \mathbf{V}_{21} & \mathbf{V}_{22} \end{array}\right). \tag{A12}$$

Note that if G satisfies the symmetry $G^{T*}(\mathbf{x}, \mathbf{X}) = G(\mathbf{X}, \mathbf{x})$, then $G^{T*}(\mathbf{x}, \mathbf{X})^{-1} = G(\mathbf{X}, \mathbf{x})^{-1}$ and, consequently, $V^{T*}(\mathbf{x}, \mathbf{X}) = V(\mathbf{X}, \mathbf{x})$, so

$$\mathsf{V}_{pq}^{\mathsf{T}*}(\mathbf{x}, \mathbf{X}) = \mathsf{V}_{qp}(\mathbf{X}, \mathbf{x}). \tag{A13}$$

On the other hand, from Eq. (50) we have

$$\tilde{\mathbf{W}}^{\mathsf{T}*}(\mathbf{X}, \mathbf{x}) = -\boldsymbol{\alpha}_{23}^{\mathsf{T}*}(\mathbf{X}, \mathbf{x}) + \boldsymbol{\gamma}_{23}^{\mathsf{T}*}(\mathbf{X}, \mathbf{x}), \qquad (A14)$$

where

$$\boldsymbol{\alpha}_{23}^{\mathsf{T}*}(\mathbf{X}, \mathbf{x}) = s^* \left\langle \rho^{\mathsf{T}*}(\mathbf{x}) \left(\nabla_{\mathbf{x}'} \mathbf{G}_{11}^* \right) \mathbf{B}^{\mathsf{T}*}(\mathbf{X}) \right\rangle - s^* \left\langle \rho^{\mathsf{T}*}(\mathbf{x}) \left(\nabla_{\mathbf{x}'} \mathbf{G}_{12}^* \right) \mathbf{A}^{\mathsf{T}*}(\mathbf{X}) \right\rangle, \quad (A15)$$

$$\boldsymbol{\gamma}_{23}^{\mathsf{T*}}(\mathbf{X}, \mathbf{x}) = s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{11}^* \rangle \langle \mathsf{V}_{11}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{11}^*) \, \mathbf{B}^{\mathsf{T*}}(\mathbf{X}) \rangle - s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{11}^* \rangle \langle \mathsf{V}_{11}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{12}^*) \, \mathbf{A}^{\mathsf{T*}}(\mathbf{X}) \rangle + s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{11}^* \rangle \langle \mathsf{V}_{12}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{21}^*) \, \mathbf{B}^{\mathsf{T*}}(\mathbf{X}) \rangle - s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{11}^* \rangle \langle \mathsf{V}_{12}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{22}^*) \, \mathbf{A}^{\mathsf{T*}}(\mathbf{X}) \rangle + s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{12}^* \rangle \langle \mathsf{V}_{21}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{11}^*) \, \mathbf{B}^{\mathsf{T*}}(\mathbf{X}) \rangle - s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{12}^* \rangle \langle \mathsf{V}_{21}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{12}^*) \, \mathbf{A}^{\mathsf{T*}}(\mathbf{X}) \rangle + s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{12}^* \rangle \langle \mathsf{V}_{22}^* \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{21}^*) \, \mathbf{B}^{\mathsf{T*}}(\mathbf{X}) \rangle - s^* \langle \rho^{\mathsf{T*}}(\mathbf{x}) \mathbf{G}_{12}^* \rangle \langle \mathsf{V}_{22} \rangle \langle (\nabla_{\mathbf{x}'} \mathbf{G}_{22}^*) \, \mathbf{A}^{\mathsf{T*}}(\mathbf{X}) \rangle .$$
 (A16)

Note that if $L^{T*} = L$, since $G^{T*}(\mathbf{X}, \mathbf{x}) = G(\mathbf{x}, \mathbf{X})$, Eq. (A13) holds and $\alpha_{32}(\mathbf{x}, \mathbf{X}) = -\alpha_{23}^{T*}(\mathbf{X}, \mathbf{x})$, $\gamma_{32}(\mathbf{x}, \mathbf{X}) = -\gamma_{23}^{T*}$ (**X**, **x**), for $s = -i\omega$, indicating that $\tilde{\mathbf{W}}^{\dagger}(\mathbf{x}, \mathbf{X}) = -\tilde{\mathbf{W}}^{T*}$ (**X**, **x**). We assume statistically homogeneous media, so $\tilde{\mathbf{W}}^{\dagger}(\mathbf{x} - \mathbf{X}) = -\tilde{\mathbf{W}}^{T*}(\mathbf{X} - \mathbf{x})$ and its Fourier transform leads to the relation $\tilde{\mathbf{W}}^{\dagger}(\kappa) = -\tilde{\mathbf{W}}^{T*}(\kappa)$ or

$$\check{\mathbf{W}}^{\dagger}(\boldsymbol{\kappa},\omega) = -\check{\mathbf{W}}^{\mathsf{T}*}(\boldsymbol{\kappa},\omega), \qquad (A17)$$

which is the first relation in Eq. (57b). If L is real, from the Fourier transform of Eq. (52) we have

$$\check{\mathbf{W}}^{\dagger}(\boldsymbol{\kappa},\omega) = \check{\mathbf{W}}^{\mathsf{T}}(-\boldsymbol{\kappa},\omega). \tag{A18}$$

Finally, from Eqs. (A17) and (A18) we obtain the relation

$$\dot{\mathbf{W}}(\boldsymbol{\kappa},\omega) = -\dot{\mathbf{W}}^*(-\boldsymbol{\kappa},\omega). \tag{A19}$$

- C. R. Simovski, Material parameters of metamaterials (a review), Opt. Spectrosc. 107, 726 (2009).
- [2] C. É. Kriegler, M. S. Rill, S. Linden, and M. Wegener, Bianisotropic photonic metamaterials, J. Sel. Top. Quantum Electron. 16, 367 (2010).
- [3] M. Kadic, T. Bückmann, R. Schittny, and M. Wegener, Metamaterials beyond electromagnetism, Rep. Prog. Phys. 76, 126501 (2013).
- [4] M. Wegener, Metamaterials beyond optics, Science 342, 939 (2013).
- [5] K. Bertoldi, V. Vitelli, J. Christensen, and M. V. Hecke, Flexible mechanical metamaterials, Nat. Rev. Mater. 2, 17066 (2017).
- [6] M. Kadic, G. W. Milton, M. V. Hecke, and M. Wegener, 3D metamaterials, Nat. Rev. Phys. 1, 198 (2019).
- [7] J. Hao, Y. Yuan, L. Ran, T. Jiang, J. A. Kong, C. T. Chan, and L. Zhou, Manipulating Electromagnetic Wave Polarizations by Anisotropic Metamaterials, Phys. Rev. Lett. 99, 063908 (2007).
- [8] P. Celli and S. Gonella, Manipulating waves with LEGO®bricks: A versatile experimental platform for metamaterial architectures, Appl. Phys. Lett. 107, 081901 (2015).
- [9] S. A. Cummer, J. Christensen, and A. Alù, Controlling sound with acoustic metamaterials, Nat. Rev. Mater. 1, 16001 (2016).

- [10] C. Mo, J. Singh, J. R. Raney, and P. Purohit, Cnoidal wave propagation in an elastic metamaterial, Phys. Rev. E 100, 013001 (2019).
- [11] J. B. Pendry, Negative Refraction Makes a Perfect Lens, Phys. Rev. Lett. 85, 3966 (2000).
- [12] D. Schurig, J. J. Mock, B. J. Justice, S. A. Cummer, J. B. Pendry, A. F. Starr, and D. R. Smith, Metamaterial electromagnetic cloak at microwave frequencies, Science 314, 977 (2006).
- [13] R. V. Craster and S. Guenneau, Acoustic Metamaterials: Negative Refraction, Imaging, Lensing and Cloaking (Springer Science & Business Media, Netherlands, 2012), Vol. 166.
- [14] W. J. Parnell and T. Shearer, Antiplane elastic wave cloaking using metamaterials, homogenization and hyperelasticity, Wave Motion 50, 1140 (2013).
- [15] D. Bigoni, S. Guenneau, A. B. Movchan, and M. Brun, Elastic metamaterials with inertial locally resonant structures: Application to lensing and localization, Phys. Rev. B 87, 174303 (2013).
- [16] A. Srivastava, Metamaterial properties of periodic laminates, J. Mech. Phys. Solids 96, 252 (2016).
- [17] H. Ge, M. Yang, C. Ma, M. H. Lu, Y. F. Chen, N. Fang, and P. Sheng, Breaking the barriers: Advances in acoustic functional materials, Natl. Sci. Rev. 5, 159 (2018).
- [18] J. H. Oh, S. Qi, Y. Y. Kim, and B. Assouar, Elastic Metamaterial Insulator for Broadband Low-Frequency Flexural Vibration Shielding, Phys. Rev. Appl. 8, 054034 (2017).
- [19] R. Getz and G. Shmuel, Band gap tunability in deformable dielectric composite plates, Int. J. Solids Struct. 128, 11 (2017).
- [20] Z. Hou, H. Ni, and B. Assouar, Pt-Symmetry for Elastic Negative Refraction, Phys. Rev. Appl. 10, 44071 (2018).
- [21] E. Bortot, O. Amir, and G. Shmuel, Topology optimization of dielectric elastomers for wide tunable band gaps, Int. J. Solids Struct. 143, 262 (2018).
- [22] L. Morini, Y. Eyzat, and M. Gei, Negative refraction in quasicrystalline multilayered metamaterials, J. Mech. Phys. Solids **124**, 282 (2019).
- [23] B. Lustig, G. Elbaz, A. Muhafra, and G. Shmuel, Anomalous energy transport in laminates with exceptional points, J. Mech. Phys. Solids 133, 103719 (2019).
- [24] D. Psiachos and M. M. Sigalas, Tailoring one-dimensional layered metamaterials to achieve unidirectional transmission and reflection, Phys. Rev. B 99, 184110 (2019).
- [25] R. A. Shelby, D. R. Smith, and S. Schultz, Experimental verification of a negative index of refraction, Science 292, 77 (2001).

- [26] Y. Ding, Z. Liu, C. Qiu, and J. Shi, Metamaterial with Simultaneously Negative Bulk Modulus and Mass Density, Phys. Rev. Lett. 99, 093904 (2007).
- [27] Y. Cheng, J. Y. Xu, and X. J. Liu, One-dimensional structured ultrasonic metamaterials with simultaneously negative dynamic density and modulus, Phys. Rev. B 77, 045134 (2008).
- [28] S. H. Lee, C. M. Park, Y. M. Seo, Z. G. Wang, and C. K. Kim, Composite Acoustic Medium with Simultaneously Negative Density and Modulus, Phys. Rev. Lett. 104, 054301 (2010).
- [29] X. N. Liu, G. K. Hu, G. L. Huang, and C. T. Sun, An elastic metamaterial with simultaneously negative mass density and bulk modulus, Appl. Phys. Lett. 98, 251907 (2011).
- [30] G. W. Milton, *The Theory of Composites* (Cambridge University Press, Cambridge, 2002), Vol. 6.
- [31] R. V. Craster, J. Kaplunov, and A. V. Pichugin, Highfrequency homogenization for periodic media, Proc. Math. Phys. Eng. Sci. 466, 2341 (2010).
- [32] D. Torrent and J. S. Dehesa, Multiple scattering formulation of two-dimensional acoustic and electromagnetic metamaterials, New J. Phys. 13, 093018 (2011).
- [33] A. Srivastava and S. N. Nasser, Overall dynamic properties of three-dimensional periodic elastic composites, Proc. Math. Phys. Eng. Sci. 468, 269 (2012).
- [34] D. Torrent, Y. Pennec, and B. D. Rouhani, Effective medium theory for elastic metamaterials in thin elastic plates, Phys. Rev. B 90, 104110 (2014).
- [35] T. Antonakakis, R. V. Craster, and S. Guenneau, Asymptotics for metamaterials and photonic crystals, Proc. Math. Phys. Eng. Sci. 469, 20120533 (2013).
- [36] T. Antonakakis, R. V. Craster, and S. Guenneau, Homogenisation for elastic photonic crystals and dynamic anisotropy, J. Mech. Phys. Solids 71, 84 (2014).
- [37] A. Srivastava, Elastic metamaterials and dynamic homogenization: A review, Int. J. Smart. Nano Mater. 6, 41 (2015).
- [38] M. Caleap and B. W. Drinkwater, Metamaterials: Supraclassical dynamic homogenization, New J. Phys. 17, 123022 (2015).
- [39] A. V. Amirkhizi, Homogenization of layered media based on scattering response and field integration, Mech. Mater. 114, 76 (2017).
- [40] R. P. Pernas-Salomón and G. Shmuel, Dynamic homogenization of composite and locally resonant flexural systems, J. Mech. Phys. Solids 119, 43 (2018).
- [41] A. Sridhar, V. G. Kouznetsova, and M. G. D. Geers, A general multiscale framework for the emergent effective elastodynamics of metamaterials, J. Mech. Phys. Solids 111, 414 (2018).
- [42] J. R. Willis, Polarization approach to the scattering of elastic waves—I. Scattering by a single inclusion, J. Mech. Phys. Solids 28, 287 (1980).
- [43] J. R. Willis, Variational principles for dynamic problems for inhomogeneous elastic media, Wave Motion 3, 1 (1981).
- [44] J. R. Willis, The nonlocal influence of density variations in a composite, Int. J. Solids Struct. 21, 805 (1985).

- [45] J. R. Willis, Continuum Micromechanics. (Springer-Verlag New York, Inc., New York, NY, USA, 1997), p. 265.
- [46] J. R. Willis, Effective constitutive relations for waves in composites and metamaterials, Proc. Math. Phys. Eng. Sci. 467, 1865 (2011).
- [47] J. R. Willis, A comparison of two formulations for effective relations for waves in a composite, Mech. Mater. 47, 51 (2012).
- [48] J. R. Willis, The construction of effective relations for waves in a composite, C. R. Mécanique 340, 181 (2012).
- [49] S. Koo, C. Cho, J. Jeong, and N. Park, Acoustic omni meta-atom for decoupled access to all octants of a wave parameter space, Nat. Commun. 7, 13012 (2016).
- [50] A. Merkel, V. R. García, J. P. Groby, J. Li, and J. Christensen, Unidirectional zero sonic reflection in passive \mathcal{PT} -symmetric Willis media, Phys. Rev. B **98**, 201102(R) (2018).
- [51] Y. Liu, Z. Liang, J. Zhu, L. Xia, O. M. Monval, T. Brunet, A. Alù, and J. Li, Willis Metamaterial on a Structured Beam, Phys. Rev. X 9, 011040 (2019).
- [52] R. P. Salomón and G. Shmuel, Symmetry breaking creates electro-momentum coupling in piezoelectric metamaterials, J. Mech. Phys. Solids 134, 103770 (2020).
- [53] W. P. Mason, Piezoelectric Crystals and Their Application to Ultrasonics (Van Nostrand, New York, 1950).
- [54] A. Regaz, L. Boukezzi, B. Zegnini, and D. Mahi, in 2015 4th International Conference on Electrical Engineering (ICEE) (IEEE, Boumerdes, 2015), p. 1.
- [55] A. Srivastava, Causality and passivity in elastodynamics, Proc. Math. Phys. Eng. Sci. 471, 20150256 (2015).
- [56] M. B. Muhlestein, C. F. Sieck, A. Alù, and M. R. Haberman, Reciprocity, passivity and causality in Willis materials, Proc. Math. Phys. Eng. Sci. 472, 20160604 (2016).
- [57] A. Alù, Restoring the physical meaning of metamaterial constitutive parameters, Phys. Rev. B 83, 081102(R) (2011).
- [58] L. Quan, Y. Ra'di, D. L. Sounas, and A. Alù, Maximum Willis Coupling in Acoustic Scatterers, Phys. Rev. Lett. 120, 254301 (2018).
- [59] G. W. Milton, New metamaterials with macroscopic behavior outside that of continuum elastodynamics, New J. Phys. 9, 359 (2007).
- [60] H. Nassar, Q. C. He, and N. Auffray, Willis elastodynamic homogenization theory revisited for periodic media, J. Mech. Phys. Solids 77, 158 (2015).
- [61] J. R. Willis, Exact effective relations for dynamics of a laminated body, Mech. Mater. 41, 385 (2009).
- [62] H. Nassar, Q.-C. He, and N. Auffray, On asymptotic elastodynamic homogenization approaches for periodic media, J. Mech. Phys. Solids 88, 274 (2016).
- [63] S. Meng and B. B. Guzina, On the dynamic homogenization of periodic media: Willis' approach versus two-scale paradigm, Proc. Math. Phys. Eng. Sci. 474, 20170638 (2018).
- [64] M. Willatzen and J. Christensen, Acoustic gain in piezoelectric semiconductors at ϵ -near-zero response, Phys. Rev. B **89**, 041201(R) (2014).

- [65] C. Fietz and G. Shvets, Current-driven metamaterial homogenization, Phys. B Condens. Matter 405, 2930 (2010).
- [66] G. W. Milton, A unifying perspective on linear continuum equations prevalent in physics. Part IV: Canonical forms for equations involving higher order gradients, arXiv:2006.03161, 2020.
- [67] A. Alù, First-principles homogenization theory for periodic metamaterials, Phys. Rev. B 84, 075153 (2011).
- [68] A. N. Norris, A. L. Shuvalov, and A. A. Kutsenko, Analytical formulation of three-dimensional dynamic homogenization for periodic elastic systems, Proc. Math. Phys. Eng. Sci. 468, 1629 (2012).
- [69] C. F. Sieck, A. Alù, and M. R. Haberman, Origins of Willis coupling and acoustic bianisotropy in acoustic metamaterials through source-driven homogenization, Phys. Rev. B 96, 104303 (2017).
- [70] Metamaterials actually have a long history before they earned this name: see the enlightening summary by Milton [84].
- [71] S. Nemat-Nasser, J. R. Willis, A. Srivastava, and A. V. Amirkhizi, Homogenization of periodic elastic composites and locally resonant sonic materials, Phys. Rev. B 83, 104103 (2011).
- [72] S. Nemat-Nasser and A. Srivastava, Overall dynamic constitutive relations of layered elastic composites, J. Mech. Phys. Solids 59, 1953 (2011).
- [73] A. L. Shuvalov, A. A. Kutsenko, A. N. Norris, and O. Poncelet, Effective Willis constitutive equations for periodically stratified anisotropic elastic media, Proc. Math. Phys. Eng. Sci. 467, 1749 (2011).
- [74] Z. H. Xiang and R. W. Yao, Realizing the Willis equations with pre-stresses, J. Mech. Phys. Solids 87, 1 (2016).
- [75] M. B. Muhlestein, C. F. Sieck, P. S. Wilson, and M. R. Haberman, Experimental evidence of Willis coupling in a one-dimensional effective material element, Nat. Commun. 8, 15625 (2017).
- [76] H. Nassar, X. C. Xu, A. N. Norris, and G. L. Huang, Modulated phononic crystals: Non-reciprocal wave propagation and Willis materials, J. Mech. Phys. Solids 101, 10 (2017).
- [77] X. Su and A. N. Norris, Retrieval method for the bianisotropic polarizability tensor of Willis acoustic scatterers, Phys. Rev. B 98, 174305 (2018).
- [78] A. Melnikov, Y. K. Chiang, L. Quan, S. Oberst, A. Alù, S. Marburg, and D. Powell, Acoustic meta-atom with experimentally verified maximum Willis coupling, Nat. Commun. 10, 3148 (2019).
- [79] Y. Zhai, H.-S. Kwon, and B.-I. Popa, Active Willis metamaterials for ultracompact nonreciprocal linear acoustic devices, Phys. Rev. B 99, 220301(R) (2019).
- [80] J. Lau, S. T. Tang, M. Yang, and Z. Yang, Coupled Decorated Membrane Resonators with Large Willis Coupling, Phys. Rev. Appl. 12, 014032 (2019).
- [81] Y. Chen, X. Li, G. Hu, M. R. Haberman, and G. Huang, An active mechanical Willis meta-layer with asymmetric polarizabilities, Nat. Commun. 11, 3681 (2020).
- [82] G. W. Milton, M. Briane, and J. R. Willis, On cloaking for elasticity and physical equations with a transformation invariant form, New J. Phys. 8, 248 (2006).

- [83] G. W. Milton and J. R. Willis, On modifications of Newton's second law and linear continuum elastodynamics, Proc. Math. Phys. Eng. Sci. 463, 855 (2007).
- [84] G. W. Milton, A unifying perspective on linear continuum equations prevalent in physics. Part II: Canonical forms for time-harmonic equations. arXiv:2006.02433, 2020.
- [85] R. P. Salomón and G. Shmuel, Symmetry breaking creates electro-momentum coupling in piezoelectric metamaterials, J. Mech. Phys. Solids 134, 103770 (2020).
- [86] G. W. Milton, Extending the theory of composites to other areas of science, J. Appl. Mech. 84, 036501 (2017).
- [87] The ambiguity associated with the nonlocal operator is explained in Sec. II.
- [88] R. de L. Kronig, On the theory of dispersion of x-rays, J. Opt. Soc. Am. 12, 547 (1926).
- [89] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, Course of Theoretical Physics, Vol. 8 (Pergamon Press LTD., Oxford, second edition, 1984).
- [90] B. Nistad and J. Skaar, Causality and electromagnetic properties of active media, Phys. Rev. E 78, 036603 (2008).
- [91] M. G. Silveirinha, Examining the validity of Kramers-Kronig relations for the magnetic permeability, Phys. Rev. B 83, 165119 (2011).
- [92] In this way, the Faraday equation for the electric field in the quasielectrostatic approximation is identically satisfied. This approximation means that the difference between time scales of the mechanical and electromagnetic effects allows to neglect time derivatives and magnetic fields in the Maxwell equations.
- [93] B. A. Auld, *Acoustic Fields and Waves in Solids* (Wiley-Interscience, New York, 1973).
- [94] Equation 3 is to be interpreted using the proper products of the matrix elements and the column vector, e.g., $\mathbf{C} : \boldsymbol{\eta}$ is a double contraction, which reads in components $C_{ijkl}\eta_{kl}$, $\mathbf{B}^{\mathsf{T}} \cdot \nabla \phi$ is a single contraction, etc..
- [95] The dependency in \mathbf{x} is omitted from Eq. (3) for brevity.
- [96] This is a local material, since the resultant fields at a certain point and time depend only on the variables in that point and time. If the response of the constituents depend on the history, e.g., they are viscoelastic, then the material is temporally nonlocal and the constitutive relations become convolutions with respect to time.
- [97] Note that $\langle \mathbf{f} \rangle = \mathbf{f}$, $\langle q \rangle = q$ and $\langle \eta \rangle = \eta$, since \mathbf{f} , q, and η are taken as sure.
- [98] Note that in Eq. (3), we amend our notation for the coupling between (σ) and (∇φ) in Salomón and Shmuel [52], by changing the superscript T to †.
- [99] More precisely, the Laplace transform with the variable *s* is used, which is connected to the Fourier transform via $s = -i\omega$.
- [100] The problem goes beyond ambiguity between velocity and strain, since it also follows that even in nonlocal elastostatics, the compatibility of the effective strain with the effective displacement field implies that there are infinitely many kernels that equivalently relate the stress and the strain [48].
- [101] This is only partially correct, since in view of the previous footnote, without such an eigen electric field there

are infinitely many kernels that equivalently relate the electric displacement field and electric field in nonlocal electrostatics [66].

- [102] A. Fettweis and S. Basu, Multidimensional causality and passivity of linear and nonlinear systems arising from physics, Multidimens. Syst. Signal Process. 22, 5 (2011).
- [103] B. Banerjee, An Introduction to Metamaterials and Waves in Composites (CRC Press, Boca Raton, 2011).
- [104] The product of i and a skew-Hermitian form is Hermitian.
- [105] W. C. Chew, A new look at reciprocity and energy conservation theorems in electromagnetics, IEEE Trans. Antennas Propag. 56, 970 (2008).
- [106] N. Moiseyev, Non-Hermitian Quantum Mechanics (Cambridge University Press, Cambridge, 2011).
- [107] G. Shmuel and N. Moiseyev, Linking Scalar Elastodynamics and Non-Hermitian Quantum Mechanics, Phys. Rev. Appl. 13, 024074 (2020).
- [108] The real form employed next also applies if the material is active or lossy; however, such materials do not satisfy the complex reciprocity relation [93], unless gain and loss are balanced.
- [109] J. D. Achenbach, *Reciprocity in Elastodynamics* (Cambridge University Press, Cambridge, 2003).
- [110] L. Knopoff and A. F. Gangi, Seismic reciprocity, Geophysics 24, 681 (1959).
- [111] A. F. Gangi, Constitutive equations and reciprocity, Geophys. J. Int. 143, 311 (2000).
- [112] D. M. Barnett and J. Lothe, Dislocations and line charges in anisotropic piezoelectric insulators, Phys. Status Solidi (b) 67, 105 (1975).
- [113] A. D. Yaghjian, Generalized or adjoint reciprocity relations for electroacoustic transducers, J. Acoust. Soc. Am. 53, 341 (1973).

- [114] J. Yang, Vibration of piezoelectric crystal (World Scientific, New Jersey, 2013).
- [115] Assuming that the composite has time-independent properties.
- [116] J. S. Toll, Causality and the dispersion relation: Logical foundations, Phys. Rev. 104, 1760 (1956).
- [117] S. R. De Groot and P. Mazur, *Non-Equilibrium Thermodynamics* (Dover, New York, 1984).
- [118] J. M. Carcione, F. Cavallini, J. Ba, W. Cheng, and A. N. Qadrouh, On the Kramers-Kronig relations, Rheol. Acta 58, 21 (2019).
- [119] R. A. Depine and A. Lakhtakia, Comment I on "Resonant and antiresonant frequency dependence of the effective parameters of metamaterials", Phys. Rev. E 70, 048601 (2004).
- [120] A. L. Efros, Comment II on "Resonant and antiresonant frequency dependence of the effective parameters of metamaterials", Phys. Rev. E 70, 048602 (2004).
- [121] F. G. Vasquez, G. W. Milton, and D. Onofrei, Active Exterior Cloaking for the 2D Laplace and Helmholtz Equations, Phys. Rev. Lett. 103, 073901 (2009).
- [122] Graeme Milton, Private communication. See also Ref. [125], where bounds on the dispersion relations are obtained using causality in passive media. Remarkably, these bounds are for finite ranges of frequencies.
- [123] M. Ringbauer, C. Giarmatzi, R. Chaves, F. Costa, A. G. White, and A. Fedrizzi, Experimental test of nonlocal causality, Sci. Adv. 2, e1600162 (2016).
- [124] H. M. Nussenzveig, Causality and Dispersion Relations (Academic, New York, 1972).
- [125] G. W. Milton, D. J. Eyre, and J. V. Mantese, Finite Frequency Range Kramers-Kronig Relations: Bounds on the Dispersion, Phys. Rev. Lett. **79**, 3062 (1997).