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Generalized Lyddane-Sachs-Teller relation and disordered solids

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The realization that the Lyddane-Sachs-Teller (LST) relation can be presented in a particular second-moment representation with the use of only sum rules and causality provides a new way to characterize the electrodynamic response of disordered solids. In this paper we extend this idea in three different directions. (1) The individual second moments of the frequency-dependent transverse and longitudinal dielectric response functions are obtained for the case of multiple dispersion oscillators in high-symmetry crystals, and the generalized LST relation is recovered. (2) The fluctuation-dissipation theorem is used to show the connection between the second moments in ordered or disordered solids and the corresponding mean-square fluctuating polarization densities, thus relating these fluctuations with the generalized LST relation. (3) The moment representation is used to construct a wave-vector-dependent LST relation, applicable when the length scale of the disorder in an isotropic medium is smaller than that of the probing wavelength.

I. INTRODUCTION

Recently, it has been shown that the Lyddane-Sachs-Teller (LST) relation[1-3]

$$\epsilon_0/\epsilon_\infty=\omega_l^2/\omega_t^2 , \tag{1}$$

which describes the connection between ϵ_0, the dielectric constant at low frequencies, ϵ_∞, the dielectric constant at high frequencies, and the long-wavelength lattice-optical modes at ω_l and ω_t in a diatomic insulating crystalline solid, can be generalized to describe a corresponding connection for disordered solids.[4] The result makes use of a moment representation[5] to identify the important frequencies of the system. Let

$$\langle\omega^2\rangle_l\equiv\left[\int_0^\infty\frac{d\omega}{\omega}\omega^2\mathrm{Im}\left[\frac{-\epsilon_\infty}{\epsilon(\omega)}\right]\right]\times\left[\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{-\epsilon_\infty}{\epsilon(\omega)}\right]\right]^{-1} \tag{2}$$

and

$$\langle\omega^2\rangle_t\equiv\left[\int_0^\infty\frac{d\omega}{\omega}\omega^2\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]\right]\times\left[\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]\right]^{-1} \tag{3}$$

define these moments in terms of the appropriate longitudinal (irrotational) and transverse (solenoidal) linear-response functions, both of which are obtained from the dielectric function $\epsilon(\omega)$. In writing these equations we have inserted ϵ_∞, describing the constant high-frequency (e.g., static electronic) response, so that our expression is specifically appropriate to degrees of freedom that are well separated in frequency, such as the lattice response. In terms of these quantities the general expression valid in the long-wavelength limit is[4]

$$\frac{\langle\omega^2\rangle_l}{\langle\omega^2\rangle_t}=\frac{\epsilon_0}{\epsilon_\infty} . \tag{4}$$

Since this expression has been obtained using only sum rules and causality there is some interest in exploring the implications of this general result. In this paper we extend the analysis in three different directions.

It is known that for a high-symmetry crystal with N optically active modes the LST relation becomes[6,7]

$$\frac{\epsilon_0}{\epsilon_\infty}=\prod_{j=1}^{N}\frac{\omega_{lj}^2}{\omega_{tj}^2} , \tag{5}$$

even if the crystal is anharmonic.[8-10] Equations (4) and (5) necessarily require that

$$\frac{\langle\omega^2\rangle_l}{\langle\omega^2\rangle_t}=\prod_{j=1}^{N}\frac{\omega_{lj}^2}{\omega_{tj}^2} . \tag{6}$$

Section II of this paper is devoted to understanding and interpreting this result. For the individual moments defined by Eqs. (2) and (3), we obtain explicit expressions in terms of the poles of the corresponding response functions and show that the ratio of these expressions indeed reduces to Eq. (6).

By the fluctuation-dissipation theorem there is a clear connection between a fluctuating variable and the appropriate response function, and in Sec. III we show that at high temperatures the mean-square value of the fluctuating variable can be rewritten in terms of the second moments defined above, so that

$$\frac{\langle\omega^2\rangle_l}{\langle\omega^2\rangle_t}=\frac{\langle P_t^2\rangle}{\langle P_l^2\rangle}=\frac{\epsilon_0}{\epsilon_\infty} , \tag{7}$$

where P_l and P_t are the polarization densities associated with the longitudinal and transverse response.

Finally, in Sec. IV the possibility of extending the generalized LST relation to nonzero **k** is examined. In composite or disordered media the **k** vector in the medium is not a well-defined quantity. Because of the general validity of Eq. (4) in the long-wavelength limit, we suggest the appropriateness of defining an $\omega(\mathbf{k})$ dispersion curve in terms of the analogous second-moment expression involving the nonlocal dielectric function.

II. $\langle\omega^2\rangle_l$ AND $\langle\omega^2\rangle_t$ FOR MULTIPLE DISPERSION OSCILLATORS

To obtain explicit expressions for $\langle\omega^2\rangle_l$ and $\langle\omega^2\rangle_t$ for a high-symmetry crystal with N optically active modes,[11] we write the long-wavelength dielectric function in its product form:[9]

$$\frac{\epsilon(\omega)}{\epsilon_\infty}=\prod_{j=1}^{N}\left[\frac{\omega_{lj}^2-\omega^2-i\gamma\omega}{\omega_{tj}^2-\omega^2-i\gamma\omega}\right] . \tag{8}$$

For simplicity we are just working in the small-damping limit, in which case the $\{\omega_{tj}\}$ and $\{\omega_{lj}\}$ give the poles and zeros of $\epsilon(\omega)$. We recall that the equivalence of Eq. (8) to its perhaps more usual form,

$$\frac{\epsilon(\omega)}{\epsilon_\infty}=1+\sum_{j=1}^{N}\frac{S_j}{\omega_{tj}^2-\omega^2-i\gamma\omega} , \tag{9}$$

follows at once from the fact that the zeros of the latter function are the roots of a polynomial of degree N. For $\omega\to 0$, Eq. (8) reduces to Eq. (5).

Since $\epsilon(\omega)/\epsilon_\infty$ is a causal response function, it obeys the Kramers-Kronig (KK) relations.[12,13] In particular, the KK relation

$$\frac{2}{\pi}\int_0^\infty d\omega\,\omega\frac{\mathrm{Im}\left[\dfrac{\epsilon(\omega)}{\epsilon_\infty}\right]}{\omega^2-x^2}=\mathrm{Re}\left[\frac{\epsilon(x)}{\epsilon_\infty}-1\right] , \tag{10}$$

gives, for $x=0$,

$$\frac{2}{\pi}\int_0^\infty \frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]=\prod_{j=1}^{N}\frac{\omega_{lj}^2}{\omega_{tj}^2}-1 , \tag{11}$$

where Eq. (5) has been used. Moreover, the high-frequency limit of Eq. (8), namely

$$\frac{\epsilon(\omega)}{\epsilon_\infty}\approx 1-\frac{1}{\omega^2}\sum_{j=1}^{N}(\omega_{lj}^2-\omega_{tj}^2) , \tag{12}$$

combined with Eq. (10) in the same limit, leads directly to

$$\frac{2}{\pi}\int_0^\infty d\omega\,\omega\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]=\sum_{j=1}^{N}(\omega_{lj}^2-\omega_{tj}^2) , \tag{13}$$

which is just the well-known f-sum rule[13] for the present case. Hence $\langle\omega^2\rangle_t$ defined by Eq. (3) is given by the ratio of Eqs. (13) and (11),

$$\langle\omega^2\rangle_t=\left[\prod_{j=1}^{N}\omega_{tj}^2\right]\left[\frac{\sum_{j=1}^{N}(\omega_{lj}^2-\omega_{tj}^2)}{\prod_{j=1}^{N}\omega_{lj}^2-\prod_{j=1}^{N}\omega_{tj}^2}\right] . \tag{14}$$

For the $N=1$ case (e.g., rocksalt), Eq. (14) reduces to $\langle\omega^2\rangle_t=\omega_t^2$. This result was also obtained in Ref. 4, by direct integration of the $N=1$ version of Eq. (8) for $\epsilon(\omega)/\epsilon_\infty$.

Turning to $\langle\omega^2\rangle_l$, we note from Eqs. (2) and (3) that its definition is the same as that for $\langle\omega^2\rangle_t$, except that $\epsilon(\omega)/\epsilon_\infty$ is replaced by $\epsilon_\infty/\epsilon(\omega)$. [The minus sign is irrelevant for the ratio in Eq. (2).] But from Eq. (8), one sees that this replacement is just equivalent to switching the l's and t's. Thus we have

$$\langle\omega^2\rangle_l=\left[\prod_{j=1}^{N}\omega_{lj}^2\right]\left[\frac{\sum_{j=1}^{N}(\omega_{tj}^2-\omega_{lj}^2)}{\prod_{j=1}^{N}\omega_{tj}^2-\prod_{j=1}^{N}\omega_{lj}^2}\right] . \tag{15}$$

When Eq. (15) is divided by Eq. (14) the rather complicated second factors on the right-hand sides are seen to cancel, yielding Eq. (6).

Bypassing the use of the KK transform by explicitly performing the integrals in Eqs. (2) and (3) using the dielectric function Eq. (8) leads to alternative, but equivalent formulas for $\langle\omega^2\rangle_l$ and $\langle\omega^2\rangle_t$ in terms of the $\{\omega_{lj}\}$ and $\{\omega_{tj}\}$. However, by $N=3$ the direct reduction of these more complicated expressions to Eqs. (14) and (15) is algebraically tedious.

III. RELATING THE MOMENTS TO THE FLUCTUATION-DISSIPATION VARIABLES

Equation (7) follows from a standard application of the Callen-Welton-Kubo fluctuation-dissipation theorem of linear-response theory.[14,15] Within the dipole approximation the Hamiltonian of a system in the presence of an applied external electric field may be written as $H=H_0-\mathbf{M}\cdot\mathbf{E}^{\mathrm{ext}}\exp(-i\omega t)\exp(\varepsilon t)$, where $\mathbf{M}$ is the system's electric dipole moment operator and $\varepsilon\to 0^+$. With the "external" susceptibility defined by the equation $\langle\mathbf{P}\rangle(t)=\langle\mathbf{M}\rangle(t)/V=\chi^{\mathrm{ext}}(\omega)\mathbf{E}^{\mathrm{ext}}\exp(-i\omega t)\exp(\varepsilon t)$, linear-response theory[15] gives

$$\chi^{\mathrm{ext}}(\omega)=-\frac{1}{3i\hbar V}\int_0^\infty dt\langle[\mathbf{M}^I(t),\mathbf{M}]\rangle_0 e^{i\omega t}e^{-\varepsilon t} , \tag{16}$$

where, as in Sec. II, we are working in the long-wavelength limit for isotropic media. In Eq. (16), the angle brackets denote an equilibrium thermal average over a canonical ensemble for H_0: $\langle\Theta\rangle_0\equiv Z^{-1}\mathrm{Tr}[\exp(-\beta H_0)\Theta]$, where $\beta=(k_BT)^{-1}$ and $Z=\mathrm{Tr}[\exp(-\beta H_0)]$. Also in Eq. (16), $\mathbf{M}^I(t)\equiv\exp(itH_0/\hbar)\mathbf{M}\exp(-itH_0/\hbar)$ is $\mathbf{M}$ in the interaction representation at time t, and the dot product is to be taken in the commutator. Using the fact that $\mathbf{M}$ is Hermitian, one easily obtains from Eq. (16) a standard fluctuation-dissipation theorem result

$$\frac{3\hbar}{\pi}\int_0^\infty d\omega[2n(\omega)+1]\mathrm{Im}\chi^{\mathrm{ext}}(\omega)=\langle \mathbf{M}\cdot\mathbf{M}\rangle_0 , \qquad (17)$$

where $n(\omega)$ is the Bose factor $[\exp(-\beta\hbar\omega)-1]^{-1}$. In obtaining this half-range integral form, one uses the identity $n(-\omega)=-[n(\omega)+1]$ and the fact that $\mathrm{Im}\chi^{\mathrm{ext}}(\omega)$ is odd in ω.

To make contact with the generalized LST relation, one must first reexpress Eq. (17) in terms of the appropriate *macroscopic* response functions. Recall that the macroscopic susceptibility $\chi(\omega)$, and the dielectric function $\epsilon(\omega)\equiv 1+4\pi\chi(\omega)$, are defined to give the local ratio between the macroscopic polarization density $\mathbf{P}$, or electric displacement $\mathbf{D}$, and the macroscopic $\mathbf{E}$: $\mathbf{P}=\chi\mathbf{E}$, $\mathbf{D}=\epsilon\mathbf{E}$. For $\mathbf{k}\to\mathbf{0}$ there is no distinction between "longitudinal" and "transverse" χ or ϵ. However, χ^{ext} in the preceding was defined by $\mathbf{P}=\chi^{\mathrm{ext}}\mathbf{E}^{\mathrm{ext}}$, and χ^{ext} will be different for irrotational (i.e., longitudinal) or solenoidal (i.e., transverse) external fields, even for $\mathbf{k}\to\mathbf{0}$, since the induced polarizations will differ in each case. Thus we will henceforth write $\chi_{\mathrm{l}}^{\mathrm{ext}}(\omega)$ and $\chi_{\mathrm{t}}^{\mathrm{ext}}(\omega)$. We recall that in an infinite medium a long-wavelength longitudinal polarization density $\mathbf{P}_{\mathrm{l}}$ produces a macroscopic electric field $-4\pi\mathbf{P}_{\mathrm{l}}$, whereas a long-wavelength transverse polarization density produces no macroscopic electric field[16] (we are working in the electrostatic approximation here, so that polariton effects[17] are not included). Hence in the presence of a longitudinal external electric field, the macroscopic electric field is $\mathbf{E}_{\mathrm{l}}=\mathbf{E}_{\mathrm{l}}^{\mathrm{ext}}-4\pi\mathbf{P}_{\mathrm{l}}$, whereas for a transverse external field one has $\mathbf{E}_{\mathrm{t}}=\mathbf{E}_{\mathrm{t}}^{\mathrm{ext}}$. The latter case immediately gives $\chi_{\mathrm{t}}^{\mathrm{ext}}=\chi(\omega)=[\epsilon(\omega)-1]/4\pi$. For the longitudinal case, one has $\mathbf{E}_{\mathrm{l}}^{\mathrm{ext}}=\mathbf{E}_{\mathrm{l}}+4\pi\mathbf{P}_{\mathrm{l}}=\mathbf{D}_{\mathrm{l}}=\epsilon\mathbf{E}_{\mathrm{l}}$, yielding the familiar "screening" result $\mathbf{E}_{\mathrm{l}}=\mathbf{E}_{\mathrm{l}}^{\mathrm{ext}}/\epsilon$. This gives $\chi_{\mathrm{l}}^{\mathrm{ext}}(\omega)=\chi(\omega)/\epsilon(\omega)=[1-1/\epsilon(\omega)]/4\pi$. Taking imaginary parts, we have $\mathrm{Im}\chi_{\mathrm{t}}^{\mathrm{ext}}(\omega)=(4\pi)^{-1}\mathrm{Im}\epsilon(\omega)$ and $\mathrm{Im}\chi_{\mathrm{l}}^{\mathrm{ext}}(\omega)=(4\pi)^{-1}\mathrm{Im}[-1/\epsilon(\omega)]$. Since $\mathbf{E}^{\mathrm{ext}}$ couples to the system via the term $-\mathbf{M}\cdot\mathbf{E}^{\mathrm{ext}}$, we see that $\chi_{\mathrm{t}}^{\mathrm{ext}}(\omega)$ and $\chi_{\mathrm{l}}^{\mathrm{ext}}(\omega)$ involve just the corresponding components of $\mathbf{M}$ or $\mathbf{P}$. Hence Eq. (17) above leads to the ratio

$$\frac{\displaystyle\int_0^\infty d\omega[2n(\omega)+1]\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]}{\displaystyle\int_0^\infty d\omega[2n(\omega)+1]\mathrm{Im}\left[\frac{-\epsilon_\infty}{\epsilon(\omega)}\right]}=\frac{\langle \mathbf{P}_{\mathrm{t}}\cdot\mathbf{P}_{\mathrm{t}}\rangle_0}{\langle \mathbf{P}_{\mathrm{l}}\cdot\mathbf{P}_{\mathrm{l}}\rangle_0} , \qquad (18)$$

valid for arbitrary temperatures, within the framework of linear-response theory. For high temperatures, $n(\omega)\approx k_B T/\hbar\omega$, so that Eq. (18) becomes

$$\frac{\displaystyle\int_0^\infty \frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon(\omega)}{\epsilon_\infty}\right]}{\displaystyle\int_0^\infty \frac{d\omega}{\omega}\mathrm{Im}\left[\frac{-\epsilon_\infty}{\epsilon(\omega)}\right]}\approx\frac{\langle \mathbf{P}_{\mathrm{t}}\cdot\mathbf{P}_{\mathrm{t}}\rangle_0}{\langle \mathbf{P}_{\mathrm{l}}\cdot\mathbf{P}_{\mathrm{l}}\rangle_0} . \qquad (19)$$

By comparison with Eqs. (2) and (3), and use of the fact that the numerators of these equations are equal,[13] we see that the left-hand side of Eq. (19) is just $\langle\omega^2\rangle_{\mathrm{l}}/\langle\omega^2\rangle_{\mathrm{t}}$. This observation together with Eq. (4) establishes Eq. (7). One can now see that ferroelectric behavior, which the "crystalline" LST relation connects with specific modes going "soft," is more generally connected with a large temperature-dependent increase in the thermal-equilibrium transverse polarization fluctuations, even for such highly disordered or overdamped systems that well-defined collective modes do not exist.

In Ref. 4, the generalized LST relation given above in Eq. (4) was derived from the KK relations (and sum rules derived from them). Because the real and imaginary parts of $\chi^{\mathrm{ext}}(\omega)$ of Eq. (16) satisfy the KK relations for all temperatures, the left-hand side of Eq. (19), which is just $\langle\omega^2\rangle_{\mathrm{l}}/\langle\omega^2\rangle_{\mathrm{t}}$, is equal to $\epsilon_0/\epsilon_\infty$ for all temperatures, within the present context of linear-response theory.[18] Hence it is interesting to note that the ratios constituting the left-hand sides of Eqs. (18) and (19) above provide two different exact characterizations of the dynamical behavior of disordered (or ordered) media in terms of integrals over the temperature-dependent dielectric function: the ratio on the left-hand side of Eq. (18) is equal to $\langle \mathbf{P}_{\mathrm{t}}\cdot\mathbf{P}_{\mathrm{t}}\rangle_0/\langle \mathbf{P}_{\mathrm{l}}\cdot\mathbf{P}_{\mathrm{l}}\rangle_0$ for all temperatures, while the ratio on the left-hand side of Eq. (19) is $\epsilon_0/\epsilon_\infty$ for all temperatures. In the high-temperature classical regime $k_B T\gg\hbar\omega$, these separate equalities become equal, as we have just seen.

IV. EXTENSION OF THE GENERALIZED LST TO NONZERO k VECTOR

As long as the length scale of the disorder in a solid is much less than that of the probing wavelength, it is useful to identify an effective wave vector, $\mathbf{k}$, in the medium. The generalized LST relation suggests that the second-moment representation offers a natural way to accomplish this, provided that Eqs. (2) and (3) can be extended to finite wave vector.

For isotropic nongyrotropic media, the nonlocal dielectric tensor may be written as[19]

$$\epsilon_{\alpha\beta}(\mathbf{k},\omega)=\epsilon_{\mathrm{l}}(k,\omega)\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}+\epsilon_t(k,\omega)\left[\delta_{\alpha\beta}-\frac{k_\alpha k_\beta}{|\mathbf{k}|^2}\right] , \qquad (20)$$

where $\epsilon_{\mathrm{l}}(k,\omega)$ and $\epsilon_t(k,\omega)$ are the dielectric functions for longitudinal and transverse probes. These quantities are functions of just the magnitude of $\mathbf{k}$, and for long wavelengths they are *macroscopic* dielectric functions, analogous to the discussion given in Sec. III.

Generalizing Eqs. (2) and (3), we define the weighted second moments of the longitudinal and transverse response functions as

$$\langle\omega^2(k)\rangle_{\mathrm{t}}\equiv\left[\int_0^\infty\frac{d\omega}{\omega}\omega^2\mathrm{Im}\left[\frac{\epsilon_{\mathrm{t}}(k,\omega)}{\epsilon_{\mathrm{t}}(k,\infty)}\right]\right]\times\left[\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon_{\mathrm{t}}(k,\omega)}{\epsilon_{\mathrm{t}}(k,\infty)}\right]\right]^{-1} \qquad (21)$$

and

$$\langle\omega^2(k)\rangle_{\mathrm{l}}\equiv\left[\int_0^\infty\frac{d\omega}{\omega}\omega^2\mathrm{Im}\left[\frac{-\epsilon_{\mathrm{l}}(k,\infty)}{\epsilon_{\mathrm{l}}(k,\omega)}\right]\right]\times\left[\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{-\epsilon_{\mathrm{l}}(k,\infty)}{\epsilon_{\mathrm{l}}(k,\omega)}\right]\right]^{-1} , \qquad (22)$$

where again we have assumed that the specific degrees of freedom under investigation are well separated in frequency so that the appropriate high-frequency response $\epsilon(k,\infty)$ is independent of ω.

The analytical properties of the nonlocal dielectric response functions $\epsilon_t(k,\omega)$ and $\epsilon_l^{-1}(k,\omega)$ are such that these functions generally satisfy the KK relations and associated sum rules[20] for fixed $\mathbf{k}$, in which case the derivation given in Ref. 4 for the generalized LST relation (4) goes through for each value of k. In particular, the equality of the "f sum rules"

$$\int_0^\infty d\omega\,\omega\,\mathrm{Im}\left[\frac{-\epsilon_l(k,\infty)}{\epsilon_l(k,\omega)}\right]=\int_0^\infty d\omega\,\omega\,\mathrm{Im}\left[\frac{\epsilon_t(k,\omega)}{\epsilon_t(k,\infty)}\right] \tag{23}$$

is obtained from Eqs. (7.1) and (7.13) of Ref. 20, when we include the high-frequency response. The inclusion of Eq. (23) into the ratio of Eqs. (21) and (22) yields

$$\frac{\langle\omega^2(k)\rangle_l}{\langle\omega^2(k)\rangle_t}=\frac{\displaystyle\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon_t(k,\omega)}{\epsilon_t(k,\infty)}\right]}{\displaystyle\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{-\epsilon_l(k,\infty)}{\epsilon_l(k,\omega)}\right]}\ , \tag{24}$$

and for $k\neq0$ the Kramers-Kronig relations give for zero frequency

$$\frac{2}{\pi}\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{\epsilon_t(k,\omega)}{\epsilon_t(k,\infty)}\right]=\frac{\epsilon_t(k,0)}{\epsilon_t(k,\infty)}-1 \tag{25}$$

and

$$\frac{2}{\pi}\int_0^\infty\frac{d\omega}{\omega}\mathrm{Im}\left[\frac{-\epsilon_l(k,\infty)}{\epsilon_l(k,\omega)}\right]=1-\frac{\epsilon_l(k,\infty)}{\epsilon_l(k,0)} \tag{26}$$

Hence Eq. (24) reduces to the generalized LST relation for finite k, which is

$$\frac{\langle\omega^2(k)\rangle_l}{\langle\omega^2(k)\rangle_t}=\frac{\epsilon_l(k,0)}{\epsilon_t(k,\infty)}\frac{\epsilon_t(k,0)-\epsilon_t(k,\infty)}{\epsilon_l(k,0)-\epsilon_l(k,\infty)}\ . \tag{27}$$

In the limit $k\to0$ the longitudinal and transverse dielectric functions become identical,[19] so that $\epsilon_t(0,0)=\epsilon_l(0,0)=\epsilon_0$ and $\epsilon_t(0,\infty)=\epsilon_l(0,\infty)=\epsilon_\infty$. Hence Eq. (27) simplifies and Eq. (4), the LST relation in the second-moment representation, is recovered. Equation (27) shows that for $k\neq0$ the dynamical properties are still directly related to the high-frequency and static response of the system at that particular k value. The general nature of this result suggests that a particularly useful single frequency with which to characterize the response of a disordered system to a particular probe (l,t) of wave vector k is $[\langle\omega^2(k)\rangle_{l,t}]^{1/2}$. The effective dispersion curves constructed in this manner would allow one to make contact with the well-known theoretical description of the long-wavelength behavior in single crystals.[16]

V. CONCLUSIONS

By focusing our attention on the electromagnetic response of solids we have explored some of the ramifications of a particular moment representation for characterizing the dynamics. We have found that the weighted second-moment descriptions of the transverse and longitudinal responses for the case of multiple dispersion oscillators in single crystals are rather complicated; however, the complexity is the same for both types of response and drops out when ratio of the two moments is considered. For disordered solids we have found through the fluctuation-dissipation theorem that the ratio of the transverse and longitudinal mean-square polarization fluctuations is asymptotically equal to $\langle\omega^2\rangle_l/\langle\omega^2\rangle_t$ in the high-temperature limit. Hence the temperature variation of the ratio of these dynamical quantities in the classical limit is described uniquely by the temperature dependence of the dc dielectric constant. Finally, we have found that as long as the length scale of the disorder in an isotropic medium is small compared to the wavelength of the transverse and longitudinal probes, then in the moment representation a nonlocal LST relation can be constructed for finite wave vector. This relation involves just the static and high-frequency response for that wave vector. Although our presentation has tended to emphasize the lattice dynamics context since that is where the LST relation was discovered, the results connect equally well the optical and static properties of other degrees of freedom such as the electronic interband transitions of ordered or disordered nonconductors. The long-wavelength response of superlattice structures with tetragonal symmetry can be included in this group, since as long as the wave vector is along the tetragonal symmetry axis our analysis in terms of scalar equations follows.

Because the derivations presented in this paper are general and rely only on the KK relations and their associated sum rules, or on the fluctuation-dissipation theorem, this approach need not be restricted to the analysis of the long-wavelength electromagnetic properties. The extension of our methods to other kinds of probes (such as acoustic waves) in composite media and disordered solids should be straightforward. More generally, if the response of an arbitrary system can be described by a vector field whose irrotational and solenoidal components are readily probed, then the weighted second-moment representation provides a useful characterization of the dynamics independent of the applicability of a mode picture. Such a characterization preserves the intimate connection between the dynamic and static responses.

Note added in proof. We have recently become aware of an interesting earlier paper in which the generalized LST relation of Noh and Sievers,[4] given here by Eq. (4), was obtained for the explicit case of a classical harmonic disordered solid [M. F. Thorpe and S. W. de Leeuw, Phys. Rev. B **33**, 8490 (1986)]. It should be noted that the moments $\langle\omega^{-2}\rangle_x$ defined in that paper are the reciprocals of the moments $\langle\omega^2\rangle_x$ defined here in Eqs. (2) and (3), with $x=l,t$. The work of both the present paper and that of Noh and Sievers[4] is more general than that of Thorpe and de Leeuw, since it concerns any system described by a linear causal isotropic dielectric response function satisfying the KK relations and associated sum rules, as we have stressed in Sec. V.

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