Density of States and Extinction Mean Free Path of Waves in Random Media: Dispersion Relations and Sum Rules

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We establish a fundamental relationship between the averaged local density of states and the extinction mean free path of waves propagating in random media. From the principle of causality and the Kramers-Kronig relations, we show that both quantities are connected by dispersion relations and are constrained by a frequency sum rule. The results should be helpful in the analysis of wave transport through complex media and in the design of materials with specific transport properties.

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Fundamental questions in coherent transport of electromagnetic, electronic, or acoustic waves [1,2] as well as applications to imaging in complex media [3] have made wave propagation in random media a central issue in physics. Randomly or periodically structured materials allow one to design media or devices with unconventional properties [4–10].

The extinction mean free path (MFP) and the density of states (DOS) are fundamental concepts in coherent wave transport. The extinction MFP ℓ_e, defined by $\ell_e^{-1} = \ell_s^{-1} + \ell_a^{-1}$, with ℓ_s and ℓ_a the scattering and absorption mean free paths, respectively, describes the attenuation of the averaged (coherent) field. When absorption is negligible, the extinction MFP equals the scattering MFP. The latter is an important quantity since $k\ell_s$ is a measure of the strength of scattering, k being the wave number in the medium. In particular, the transition to Anderson localization in three dimensions is expected when $k\ell_s \lesssim 1$, according to the Ioffe-Regel criterion [1]. The DOS shapes many macroscopic transport properties [11]. The spectral behavior of ℓ_s and the DOS, and the influence of the DOS on the Ioffe-Regel criterion, were put forward in early studies of Anderson localization of light [12]. Since then, it has been often implicitly assumed that a relation between the DOS and MFP exists, such that a (local) minimum in the MFP should be somehow associated to a minimum in the DOS. The *local* density of states (LDOS) has also received increasing interest. The LDOS drives the spontaneous emission of light [13] and is at the root of novel imaging techniques based on field correlations [14]. Statistics of the LDOS characterize the transport regime [15], speckle patterns [16], or the local structure of a complex medium [17].

In this Letter, we establish a fundamental relationship between the averaged LDOS in a random medium and the extinction MFP. As a consequence of causality and the Kramers-Kronig relations, we show that both quantities are connected by dispersion relations and are constrained by a frequency sum rule. We focus the derivation on light propagation in scattering media, but the results should be applicable to any kind of waves.

Consider a scattering medium made of scatterers randomly distributed in free space (or in an otherwise homogeneous background medium). The (dyadic) Green function describes the response at point $\mathbf{r}$, and at a given frequency ω, to a point electric-dipole source $\mathbf{p}$ located at point $\mathbf{r}'$ through the relation $\mathbf{E}(\mathbf{r}) = \mu_0\omega^2\mathbf{G}(\mathbf{r},\mathbf{r}')\mathbf{p}$. For a given configuration of the random medium and for a point in vacuum (outside the scatterers), the LDOS at a point $\mathbf{r}$ is given by $\rho(\omega,\mathbf{r}) \equiv 2\omega/(\pi c^2)\mathrm{Im}\,\mathrm{Tr}\mathbf{G}(\mathbf{r},\mathbf{r})$, where c is the speed of light in vacuum and Tr denotes the trace of a tensor [11]. An extra term accounting for evanescent states associated with the magnetic field also exists for a point located in vacuum but in the near field of a surface [18]. The LDOS defined above, the quantity of interest in the present discussion, is the relevant part of the LDOS entering Fermi's "golden rule" in the calculation of the spontaneous decay rate of atoms [11,19,20]. The full LDOS, that would give the local energy density at equilibrium, contains an additional term describing the energy stored in the material degrees of freedom of the scatterers [outside the scatterers, the LDOS is entirely given by $\rho(\omega,\mathbf{r})$] [11,19].

After averaging over the positions of the scatterers and assuming statistical translational invariance, the averaged Green function obeys the Dyson equation [1,21]:

$$\langle\mathbf{G}(\mathbf{k})\rangle = \mathbf{G}_0(\mathbf{k}) + \mathbf{G}_0(\mathbf{k})\mathbf{\Sigma}(\mathbf{k})\langle\mathbf{G}(\mathbf{k})\rangle, \tag{1}$$

where $\mathbf{G}_0(\mathbf{k}) = [(k^2 - k_0^2)\mathbf{I} - \mathbf{k}\mathbf{k}]^{-1}$ is the Fourier transform of the free-space Green function, with $\mathbf{I}$ the unit tensor, $k_0 = \omega/c$, and $\mathbf{\Sigma}(\mathbf{k})$ is the self-energy containing the sum of all multiply connected scattering events [21]. From Eq. (1), the averaged Green function can be written as

$$\langle\mathbf{G}(\mathbf{k})\rangle = \frac{\mathbf{I}}{(k^2 - k_0^2)\mathbf{I} - \mathbf{k}\mathbf{k} - \mathbf{\Sigma}(\mathbf{k})}. \tag{2}$$

From this expression, one identifies the effective dielectric function $\epsilon_{\text{eff}}(\mathbf{k}) = \mathbf{I} + k_0^{-2}\boldsymbol{\Sigma}(\mathbf{k})$, which drives the propagation of the averaged field in the random medium. The effective dielectric function is, in general, a nonlocal and anisotropic dyadic response function. In practice, determining the effective dielectric function is a difficult problem, that can be solved only under some (sometimes severe) approximations [5,22]. In the present study, we do not need to refer to a specific model. Our arguments rely only on the existence of the effective dielectric function.

Under the following hypotheses, (i) the random medium is isotropic on average, and (ii) only field variations on scales larger than the size of the scatterers and the correlation distance between scatterers are accounted for, the dielectric function becomes a local and isotropic (scalar) function [1,22], i.e., $\epsilon_{\text{eff}}(\mathbf{k}) = \epsilon_{\text{eff}}\mathbf{I}$. We assume that these conditions are satisfied in the following. The averaged Green function can be decomposed into transverse and longitudinal parts $\langle \mathbf{G}\rangle = \langle \mathbf{G}^{\perp}\rangle + \langle \mathbf{G}^{\parallel}\rangle$, which in real space read as

$$\langle \mathbf{G}^{\perp}(\mathbf{R})\rangle = \left[\mathbf{I} + \frac{1}{k_{\text{eff}}^2}\boldsymbol{\nabla\nabla}\right]\frac{\exp(ik_{\text{eff}}R) - 1}{4\pi R}, \quad (3)$$

$$\langle \mathbf{G}^{\parallel}(\mathbf{R})\rangle = \text{P}\left[\mathbf{I} + \frac{1}{k_{\text{eff}}^2}\boldsymbol{\nabla\nabla}\right]\frac{1}{4\pi R} - \mathbf{L}\,\delta(\mathbf{R}), \quad (4)$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$ and $k_{\text{eff}} = n_{\text{eff}}k_0$, with $n_{\text{eff}} = \sqrt{\epsilon_{\text{eff}}}$ the effective (complex) refractive index of the random medium [23]. The symbol P stands for principal value, and $\mathbf{L}$ is a dyadic tensor that describes the singular behavior of the Green function at the origin. For a spherical exclusion volume, one has $\mathbf{L}(\mathbf{R}) = \mathbf{I}/(3k_{\text{eff}}^2)$ [24].

The singular term in the Green function contains an imaginary part (due to losses by scattering and/or absorption) so that $\text{Im}\,\text{Tr}\langle \mathbf{G}^{\parallel}(\mathbf{R})\rangle$ diverges at the origin. The study of the singularity problem, in the context of spontaneous emission in absorbing dielectrics, lead to an extension of the definition of LDOS: It is given by the imaginary part of the transverse Green function $\langle \mathbf{G}^{\perp}\rangle$ [25,26]. We shall use this definition in the present study:

$$\rho(\omega) = \frac{2\omega}{\pi c^2}\,\text{Im}\,\text{Tr}\langle \mathbf{G}^{\perp}(\mathbf{R} = \mathbf{0})\rangle. \quad (5)$$

Using Eq. (3), this leads to $\rho(\omega) = \rho_0(\omega)\text{Re}n_{\text{eff}}(\omega)$, with $\rho_0(\omega) = \omega^2/(\pi^2 c^3)$ the LDOS in free space [26]. The averaged LDOS is given by the real part of the effective refractive index.

In the context of spontaneous decay rate calculations in dielectrics, one usually includes the so-called local field corrections [20,25]. It has been shown that these corrections are a consequence of the local correlations in the scatterer positions induced by the nonoverlapping conditions [19]. To keep consistency with assumptions (i) and (ii) used previously, we will disregard the influence of such correlations and keep expression (3) for the averaged transverse Green function.

The second important quantity in our discussion is the extinction MFP ℓ_e, defined as the decay length of the intensity of the averaged field by scattering and absorption [1], given by the imaginary part of the effective refractive index [22,23]: $\ell_e(\omega) = c/[2\omega\text{Im}n_{\text{eff}}(\omega)]$.

We now establish the relationships between the averaged LDOS and MFP. From the principle of causality, one can derive the Kramers-Kronig (KK) relations that connect the real and imaginary parts of the susceptibility of any linear material. Regarding the optical response, the KK relations are usually written in terms of the dielectric function [27]. It can be shown that the refractive index in passive materials is also a quantity that satisfies the KK relations [28]. A straightforward application of the KK relations to the effective refractive index, using the expressions of $\rho(\omega)$ and $\ell_e(\omega)$ given above, leads to

$$\frac{\rho(\omega)}{\rho_0(\omega)} = 1 + \frac{c}{\pi}\text{P}\int_0^{\infty}\frac{[\ell_e(\omega')]^{-1}}{\omega'^2 - \omega^2}d\omega', \quad (6)$$

$$\frac{1}{\ell_e(\omega)} = -4\pi\omega^2 c^2\text{P}\int_0^{\infty}\frac{\rho(\omega') - \rho_0(\omega')}{\omega'^2(\omega'^2 - \omega^2)}d\omega'. \quad (7)$$

These dispersion relations demonstrate that the averaged LDOS and the extinction MFP are not independent and provide a method to determine one quantity from the spectrum of the other one. In particular, Eq. (6) shows that, from an extinction spectrum (a natural measurement in spectroscopy), one can deduce the averaged LDOS.

From the KK relations, sum rules for the dielectric constant and the refractive index can be obtained [29]. In particular, it is well established that the refractive index of any passive and causal medium satisfies $\int_0^{\infty}\omega\text{Im}n_{\text{eff}}(\omega)\times[\text{Re}n_{\text{eff}}(\omega) - 1]d\omega = 0$ and $\int_0^{\infty}[\text{Re}n_{\text{eff}}(\omega) - 1]d\omega = 0$ [29]. Beyond the principle of causality, the derivation of these sum rules relies on the assumption of a material behaving as a free-electron gas in the high frequency limit: $\epsilon_{\text{eff}}(\omega) \sim 1 - \omega_p^2/\omega^2$ when $\omega \to \infty$, where ω_p is an effective plasma frequency. Note that this high frequency behavior is expected as soon as the frequency is much larger than the resonance frequencies of the effective medium. The sum rules for the effective refractive index can be translated into new sum rules involving the averaged macroscopic DOS and the extinction MFP. In particular, we obtain

$$\int_0^{\infty}\frac{\rho(\omega) - \rho_0(\omega)}{\omega^2\ell_e(\omega)}d\omega = 0. \quad (8)$$

This relation is the main result of this Letter. It demonstrates that the spectra of the averaged LDOS and of the extinction MFP are intimately connected and constrained by a simple sum rule. This sum involves the extinction mean free path and applies to media in which extinction is provided by scattering and/or absorption. The simplicity and the generality of this relation are striking. Let us remind that this sum rule and the dispersion relations (6) and (7) are valid under three conditions: (i) The medium is passive and causal, (ii) the medium behaves as a free-electron gas in the high frequency limit, and (iii) the effective medium is described by an isotropic and local dielectric function. The second sum rule for the refractive index leads to a relation involving the averaged LDOS only: $\int_0^\infty [\rho(\omega) - \rho_0(\omega)]/\omega^2 d\omega = 0$. This sum rule was established previously in the context of spontaneous emission in dielectric media [30]. Regarding wave propagation in random media, it establishes a constraint on the potential modifications of the averaged LDOS. In particular, it shows that $\rho(\omega)$ is necessarily lower than the LDOS in free space $\rho_0(\omega)$ in a spectral range and greater than $\rho_0(\omega)$ in another spectral range [the numerator $\rho(\omega) - \rho_0(\omega)$ has to change sign for the integral to vanish]. This behavior is also given by Eq. (8) because $\ell_e(\omega) > 0$ in a passive medium.

The dispersion relations and sum rules involve frequency summations up to infinity. One can wonder whether there is any frequency range in which, on the one hand, dispersion phenomena are important but, on the other hand, the assumption of a local effective dielectric function stills holds. For homogeneous media, this issue has been discussed in Ref. [27]. For random media, frequency dispersion phenomena are important in the vicinity of a resonance frequency ω_0 of the effective dielectric function $\epsilon_{\rm eff}$. At frequencies $\omega \gg \omega_0$, $\epsilon_{\rm eff}$ tends to unity and the integrand in the KK relations vanishes. If ℓ_c denotes the correlation length of the random medium (ℓ_c equals at least the size of the particles), the condition $\lambda_0 \gg \ell_c$ must be satisfied for our theory to be valid, where $\lambda_0 = 2\pi c/\omega_0$. In this case, in the frequency region where the integrand in the KK contributes, the condition of locality of the effective dielectric function is satisfied. This condition can be satisfied, e.g., for a system of semiconductor or metallic particles with size a, that can exhibit a (phonon or plasmon) resonance with $\lambda_0 \gg a$, or with a weakly correlated atomic system in which λ_0 is fixed by the atomic transition and can be much greater than the mechanically induced correlations. The condition might be more difficult to satisfy for Mie resonances, for which $\lambda_0 \lesssim a$.

In the following, we illustrate the general behavior induced by relations (6)–(8) in a particular case. Let us consider a random scattering medium with an effective dielectric function exhibiting a resonance at a particular frequency ω_0. The resonance can be induced by an internal resonance of the scatterers or of purely geometric origin (or both). We choose a Lorentz model of the form

$$\epsilon_{\rm eff}(\omega) = 1 + \frac{\mathcal{F}\omega_0^2}{\omega_0^2 - \omega^2 - i\omega\Gamma}, \tag{9}$$

where the parameter $\mathcal{F}$ is an effective oscillator strength and Γ is the linewidth. From this expression, the effective index $n_{\rm eff} = \sqrt{\epsilon_{\rm eff}}$ is readily obtained numerically, as well as the spectra of the averaged DOS $\rho(\omega)$ and the extinction MFP $\ell_e(\omega)$. A regime of strong scattering is identified when ${\rm Im}n_{\rm eff}(\omega) > {\rm Re}n_{\rm eff}(\omega)$, which corresponds to $k\ell_e < 1/2$, where $k = {\rm Re}k_{\rm eff}$ is the wave number in the medium [11]. In this regime, the effective medium satisfies ${\rm Re}\epsilon_{\rm eff}(\omega) < 0$, and the averaged field is strongly damped (the effective medium has a metallic character). In the regime $k\ell_e > 1/2$, one has ${\rm Re}\epsilon_{\rm eff}(\omega) > 0$, and the effective medium has a dielectric character. The transition between these two regimes is driven by the parameter $\mathcal{P} = \mathcal{F}\omega_0/\Gamma$. For $\mathcal{P} < 2$, one has ${\rm Re}\epsilon_{\rm eff}(\omega) > 0$ at all frequencies. For $\mathcal{P} > 2$, there is a frequency range for which ${\rm Re}\epsilon_{\rm eff}(\omega) < 0$ or, equivalently, $k\ell_e < 1/2$.

We show in Fig. 1 the spectra of the averaged LDOS in the case of an effective medium with $\mathcal{P} = 1$ (dashed line) and $\mathcal{P} = 20$ (solid line). The corresponding spectra for the MFP are shown in Fig. 2. The averaged LDOS for $\mathcal{P} = 1$ (dashed line in Fig. 1) shows a typical dispersion behavior around the resonance frequency (corresponding to $\delta = 0$). At the resonance frequency, the MFP (dashed line in Fig. 2) exhibits a minimum. This is a classical behavior of quantities connected by KK dispersion relations. For $\mathcal{P} = 20$, the averaged LDOS (solid line in Fig. 1) exhibits a region with a low value (pseudogap), corresponding to the spectral region for which ${\rm Re}\epsilon_{\rm eff}(\omega) < 0$ or $k\ell_e < 1/2$. Close to the lower pseudogap band edge, the LDOS exhibits a strong oscillation that is a feature of the underlying dispersion relation. In the same spectral range, the extinction MFP exhibits a region with a low value. This behavior, illustrated here in a particular case, is dictated by the dispersion relations and the frequency sum rule. These relations give a rigorous and quantitative basis to a behavior that is implicitly assumed in many discussions on wave transport in random media.

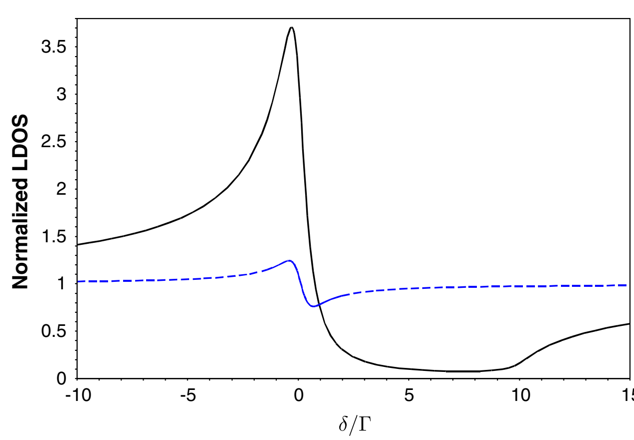

FIG. 1 (color online). Normalized averaged LDOS $\rho(\omega)/\rho_0(\omega)$ versus the normalized detuning from resonance $\delta/\Gamma = (\omega - \omega_0)/\Gamma$. The effective dielectric function $\epsilon_{\rm eff}(\omega)$ is given by Eq. (9), with $\omega_0 = 10^{15}$ Hz, $\Gamma = 10^9$ Hz, and $\mathcal{P} = 1$ (dashed line) or $\mathcal{P} = 20$ (solid line).

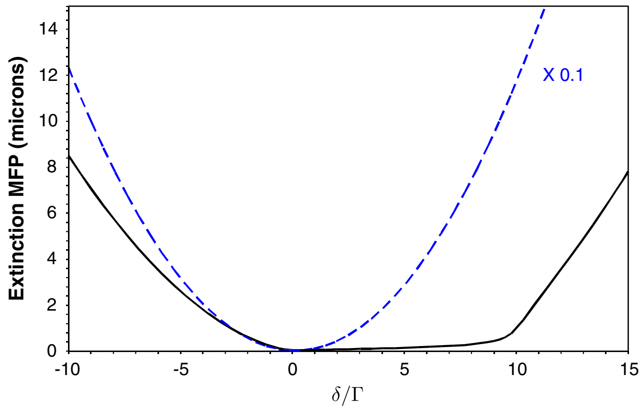

FIG. 2 (color online). Extinction MFP $\ell_e(\omega)$ in micrometers versus the normalized detuning from resonance $\delta/\Gamma = (\omega - \omega_0)/\Gamma$. The parameters are as in Fig. 1. For clarity, the curve corresponding to $\mathcal{P} = 1$ has been scaled by a factor of 0.1.

We also note that if the effective dielectric constant is known analytically, one has $\mathrm{Im}\epsilon_{\mathrm{eff}} = 2\mathrm{Im}n_{\mathrm{eff}}\mathrm{Re}n_{\mathrm{eff}}$ at each frequency. This leads to $\mathrm{Im}\epsilon_{\mathrm{eff}}(\omega) = c/[\omega\ell_e(\omega)]\rho(\omega)/\rho_0(\omega)$. This explicit relation is useful when an analytical model of ϵ_{eff} is available, a situation that occurs only in a few specific cases.

In summary, from the principle of causality, we have established dispersion relations and a frequency sum rule that constrain the spectral variations of the averaged LDOS and the extinction MFP in a random scattering medium and sustain general features of their spectral behavior close to a resonance of the effective medium. Our results are strictly valid in media with negligible correlations in the scatterer positions. Experimental deviations from KK relations could then be a signature of microscopic correlations.

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