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Propagation in random media: Calculation of the effective dispersive permittivity by use of the replica method

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In this paper we use the replica method together with a variational method in order to study the propagation of a wave traveling in a disordered composite. We compute the effective permittivity tensor, which takes into account the effects of spatial dispersion. Our calculation is not perturbative in the frequency, and we obtain a self-consistent formula for the longitudinal and transverse parts of the permittivity tensor applicable to the whole range of frequency. Our result appears as a self-consistent version of the usual weak-disorder expansion and thus recovers all weak-disorder results. We have checked that the solution to the self-consistent equations is correct in the limits of high and low spatial frequency.

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I. INTRODUCTION

We study in this paper the propagation of an electromagnetic wave in a random composite. When the wavelength is of the order of (or smaller than) the typical size of the inhomogeneities, one cannot describe a heterogeneous material as quasihomogeneous. In order to use an effective medium concept, one has to introduce the notion of spatial dispersion, even if the microscopic permittivity is not dispersive. This can be done by describing the electromagnetic properties of the medium by an effective permittivity tensor that depends on both the frequency ω and position r. If the medium is described by a local random permittivity denoted by $\varepsilon(r,\omega)$, the definition of the effective response tensor ε^*_{ij} is (see, for example, [1])

$$\langle \varepsilon(r,\omega)E_i(r)\rangle = \int dr' \sum_{j=1}^{d} \varepsilon^*_{ij}(r-r',\omega)\langle E_j(r')\rangle \ , \quad (1)$$

where E_i is the ith component of the electric field (d is the space dimension) and where the brackets $\langle\ \rangle$ denote the average over the disorder. It should be emphasized that the effective dielectric tensor will be a nonlocal function with a correlation radius determined by the characteristic correlation length of the calculations of $\varepsilon(r,\omega)$. If this characteristic length is negligible compared to the other lengths in the problem (e.g., the wavelength λ), one obtains essentially the quasistatic limit. The main point is thus to introduce a size of the inhomogeneities. In order to do this we will suppose that the local permittivities are not independent from site to site but that they are correlated. We will deal with Gaussian distributed variables, so that it is just necessary to introduce the connected two-point correlation function $\langle \delta\varepsilon(r)\delta\varepsilon(r')\rangle = \langle \delta\varepsilon^2\rangle C(r-r')$. The function $C(r)$ has an extension of order l that is roughly the radius of the grain. In the following, we will specialize in the case where $C(r)$ is a function of the modulus of r only, so that the average medium is isotropic. It can be noted that in our previous paper [2(a)] we studied the quasistatic case, which thus corresponds here to $C(r) \propto \delta(r)$.

In contrast with most approaches to this problem, which are perturbative in the quantity l/λ (or the inverse) and thus lose the interesting regime where $\lambda \simeq l$, our calculation is variational. Moreover, these approaches necessitate the introduction of a reference permittivity that should disappear at the end of the calculation (as shown by Luck [3] in the quasistatic limit). This is usually not the case, and one has to determine this function *ad hoc* (for instance, by using a self-consistent argument). In our method, the self-consistency arises naturally, and the problem of this reference medium does not exist anymore.

We will use here an approach based on the use of the replica method [4]. This method is an alternative to the fermionic method, which was used in [5] in order to study propagation in random media. The replica method relies on an analytic continuation that cannot be proved, but it should be noted that in the fermionic representation there is also an ambiguity related to the sign of a determinant. Suppressing the absolute value of this determinant can lead to incorrect results (see, for example, [6]).

The use of the replica method allows us to perform disorder averages, but, as usual, introduces a coupling between different replicas. This coupling appears through a complicated effective Hamiltonian that is approximated by a variational method [7–10] relying on the Gibbs-Bogoliubov inequality. In addition, this variational approach is nonperturbative and allows us to compute the effective permittivity tensor for the whole range of fre-

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quency. Let us note that this method was successfully applied to the random resistor network problem in the quasistatic case [2(a)] and to the Hall effect in composite media [2(b)].

In Sec. II, we define the model and introduce the basic equations. In Sec. III, we introduce the replica method together with a variational method in order to obtain the self-consistent equations. In the discussion (Sec. IV) we will study the solution of these equations in the following limiting cases: the long and short wavelength limits (for which exact solutions are known) and also the weak-disorder solutions.

II. THE MODEL AND THE BASIC EQUATIONS

We first separate the local permittivity $\varepsilon(r,\omega)$ into a homogeneous part ε_0 (which can be frequency dependent) and a fluctuating part $\delta\varepsilon(r,\omega)$:

$$\varepsilon(r,\omega)=\varepsilon_0(\omega)+\delta\varepsilon(r,\omega) . \tag{2}$$

We choose here $\varepsilon_0(\omega)=\langle\varepsilon\rangle$ so that $\langle\delta\varepsilon\rangle=0$.

The fluctuating part at each point r (of a d-dimensional medium) is supposed to be a random variable distributed according to a Gaussian law:

$$p(\delta\varepsilon(r,\omega))\propto\exp\left[-\int dr\,dr'\delta\varepsilon(r)\times[\langle\delta\varepsilon^2\rangle C(r-r')]^{-1}\delta\varepsilon(r')\right] , \tag{3}$$

where the function $C(r)$ depends only on the modulus of r and roughly determines the size of the grain. The choice of Gaussian disorder is mainly dictated by technical considerations, but we believe that it will display the most important features of the dispersion in disordered media.

The electric field satisfies the propagation equation that is derived from Maxwell's equations:

$$\text{curl}\,\text{curl}\vec{E}-k_0^2\varepsilon(r,\omega)\vec{E}=\vec{0} , \tag{4}$$

where $k_0=\omega/c$.

$$\text{curl}\,\text{curl}\vec{E}-k_0^2\varepsilon_0\vec{E}=k_0^2\delta\varepsilon(r,\omega)\vec{E} , \tag{5}$$

or in integral form,

$$E_i(r)=E_{0i}(r)+\int dr'\sum_{j=1}^{d}G_{ij}(r-r')\delta\varepsilon(r',\omega)E_j(r') , \tag{6}$$

where $E_0(r)$ is the external applied field and G is the Green's function of the Helmholtz equation (5) for the homogeneous medium described by $\varepsilon_0(\omega)$. Its Fourier transform reads

$$G_{ij}(k)=-\frac{1}{\varepsilon_0(\omega)}P_{ij}(k)+\frac{k_0^2}{k^2-\varepsilon_0(\omega)k_0^2}Q_{ij}(k) , \tag{7}$$

where P is the projector along the direction of $\vec{k}$:

$$P_{ij}(k)=\frac{k_ik_j}{k^2} \tag{8}$$

and Q the orthogonal projector ($1=P+Q$). Let us make two remarks. First, at zero frequency the transverse part goes to zero and one retrieves the quasistatic limit:

$$G_{ij}(k)\underset{\omega=0}{\rightarrow}-\frac{1}{\varepsilon_0}P_{ij}(k) . \tag{9}$$

Second, all the Fourier transforms of the tensors that will appear (in particular, the effective permittivity) will have a unique decomposition over this basis (P,Q). For a statistically isotropic and homogeneous medium, one indeed has [11]

$$\varepsilon^*_{ij}(k)=\varepsilon^*_l(k)P_{ij}(k)+\varepsilon^*_t(k)Q_{ij}(k) , \tag{10}$$

where ε^*_l (ε^*_t) is the longitudinal (transverse) permittivity that depends only on the modulus of the vector k. In the long wavelength limit ($kl\ll 1$) these permittivities are equal and one finds

$$\varepsilon^*_{ij}=\varepsilon^*\delta_{ij} . \tag{11}$$

Equation (6) can be recast in the form

$$E_i(r)=\int dr'\sum_{j=1}^{d}M_{ij}^{-1}(r,r')E_{0j}(r') , \tag{12}$$

where

$$M_{ij}(r,r')=\delta_{ij}\delta(r-r')-G_{ij}(r-r')\delta\varepsilon(r',\omega) . \tag{13}$$

Averaging (6) and (12) and using Eq. (1), it can be easily shown that

$$\langle(1-G\delta\varepsilon)^{-1}\rangle_{ij}(k)=(1-G\delta\varepsilon^*)_{ij}^{-1}(k) . \tag{14}$$

Thus the problem is reduced to calculating the disorder average of the inverse (M^{-1}) of the random operator M.

III. THE REPLICA METHOD AND THE VARIATIONAL TREATMENT

The problem is now reduced to calculating the disorder average of the inverse of a random operator. One can use the standard Gaussian formula and write M^{-1} using complex fields:

$$M_{ij}^{-1}(r,r')=\frac{\int\Pi_l\mathcal{D}(\phi_l^*,\phi_l)\phi_i^*(r)\phi_j(r')\exp\left[-\int dr\,dr'\sum_{ij}\phi_i^*(r)M_{ij}(r,r')\phi_j(r')\right]}{\int\Pi_l\mathcal{D}(\phi_l^*,\phi_l)\exp\left[-\int dr\,dr'\sum_{ij}\phi_i^*(r)M_{ij}(r,r')\phi_j(r')\right]} . \tag{15}$$

In order to compute the average of this quantity, we write the denominator as the limit when n goes to zero of

$$\left[\int \Pi_l \mathcal{D}(\phi_l^*, \phi_l) \exp \left[-\int dr\, dr' \sum_{ij} \phi_i^*(r) M_{ij}(r,r') \phi_j(r') \right] \right]^{n-1} . \tag{16}$$

We thus introduce n replicas of the fields ϕ_i^*, ϕ_i, which will be denoted by ϕ_i^{*a}, ϕ_i^a ($a = 1, \ldots, n$). We thus first consider n as an integer and then take the limit n going to zero. A similar approach was proposed by John and Stephen [12] for the problem of wave localization in random media. We can now write M^{-1} as

$$M_{ij}^{-1}(r,r') = \int \Pi_{i,a} \mathcal{D}(\phi_i^{*a}, \phi_i^a) \frac{\vec{\phi}_i^* \vec{\phi}_j}{n} \exp \left[-\int dr\, dr' \sum_{ij,a} \phi_i^{*a}(r) [\delta_{ij} \delta(r-r') - G_{ij}(r-r') \delta\varepsilon(r',\omega)] \phi_j^a(r') \right] , \tag{17}$$

where the limit $n \to 0$ is implicitly taken and where $\vec{\phi}_i = (\phi_i^1, \phi_i^2, \ldots, \phi_i^n)$ and $\vec{\phi}_i^* = (\phi_i^{*1}, \phi_i^{*2}, \ldots, \phi_i^{*n})$.

In the following, we give an outline of the calculation, which is very similar to the one performed in [2(a)]. We give full details of the calculation in the Appendix.

The average of Eq. (17) is now easy to compute, and since $\delta\varepsilon$ is a Gaussian variable, one obtains an effective quartic Hamiltonian $\mathcal{H}_{\text{eff}}$. The quantity $\langle M^{-1} \rangle$ is the propagator associated with this effective Hamiltonian. We replace this quartic Hamiltonian by the best Gaussian Hamiltonian in the sense of the variational principle. If this Hamiltonian has a kernel denoted by $K^{-1}(k)$, we immediately identify

$$\langle M^{-1} \rangle_{ij}(k) \approx K_{ij}(k) . \tag{18}$$

In terms of K, the variational equation reads

$$-K_{ij}^{-1} + \delta_{ij} - \sum_{l=1}^{d} G_{il}(k) \Lambda_{lj}(k) = 0 , \tag{19}$$

where

$$A_{ij}(k) = \langle \delta\varepsilon^2 \rangle \int dq\, C(q) [K(k-q) G(k-q)]_{ij} . \tag{20}$$

Using Eqs. (14) and (18), we identify Λ as $\delta\varepsilon^*$. Moreover, using Eqs. (7) and (19), one can show that

$$KG(k) = -\frac{1}{\varepsilon_l^*(k,\omega)} P(k) + \frac{k_0^2}{k^2 - \varepsilon_t^*(k,\omega) k_0^2} Q(k) . \tag{21}$$

We therefore see that KG is in fact the dipolar tensor computed in the dispersive effective medium determined by $\varepsilon^*(k,\omega)$:

$$KG(k) \equiv G^*(k) . \tag{22}$$

The final self-consistent equation can thus be rewritten as

$$\delta\varepsilon_{ij}^*(k) = \langle \delta\varepsilon^2 \rangle \int dq\, C(q) G_{ij}^*(k-q) , \tag{23}$$

whose explicit expression is

$$\begin{aligned} \varepsilon_l^*(k,\omega) &= \varepsilon_0 + \langle \delta\varepsilon^2 \rangle \int \frac{d^d q}{(2\pi)^d} C(k-q) \left[\beta + (\alpha - \beta) \frac{(k \cdot q)^2}{k^2 q^2} \right] , \\ \varepsilon_t^*(k,\omega) &= \varepsilon_0 + \langle \delta\varepsilon^2 \rangle \int \frac{d^d q}{(2\pi)^d} C(k-q) \left[\beta + \frac{1}{d-1} (\alpha - \beta) \left[1 - \frac{(k \cdot q)^2}{k^2 q^2} \right] \right] , \end{aligned} \tag{24}$$

where

$$\begin{aligned} \alpha(q) &= -\frac{1}{\varepsilon_l^*(q,\omega)} , \\ \beta(q) &= \frac{k_0^2}{q^2 - \varepsilon_t^*(q,\omega) k_0^2} . \end{aligned} \tag{25}$$

Equation (23) [and (24)] is the main result of our paper. These equations determine in a self-consistent way the functions $\varepsilon_t^*(k,\omega)$ and $\varepsilon_l^*(k,\omega)$. In the discussion we will mostly use Eq. (23).

IV. DISCUSSION

Equation (23) is the generalization in the finite frequency (and wavelength) case of Hori's equations [13,14] [which we also obtained by means of the same method [2(a)]. First, it seems useful to make the connection between expression (23) for $\varepsilon^*(k,\omega)$ and the usual weak-disorder result [15]. Let us recall that, from Eq. (6), to lowest order in $\delta\varepsilon$, we have

$$\begin{aligned} &\langle E(r) \rangle \approx E_0(r) , \\ &\langle \delta\varepsilon(r) E_i(r) \rangle \\ &\quad \approx \langle \delta\varepsilon^2 \rangle \int dr' \sum_{j=1}^{d} G_{ij}(r-r') C(r-r') E_{0j}(r') , \end{aligned} \tag{26}$$

and from (1) we obtain

$$\delta\varepsilon_{ij}^*(r) \approx \langle \delta\varepsilon^2 \rangle C(r) G_{ij}(r) . \tag{27}$$

Our result (23) thus appears as a natural self-consistent generalization of Eq. (27), where we replace G by G^*. We would like to stress that self-consistency appears naturally in our treatment. Of course, Eq. (23) is equivalent to (27) to first order and, therefore, reproduces the usual weak-disorder case, especially the dispersion relations for longitudinal and transverse modes [15]. It should also be noted that, since our treatment is variational (and not perturbative), we believe formula (23) to be as accurate for a strong permittivity contrast as in the quasistatic case [2(a)].

Let us now consider the two extreme limits ($kl \ll 1$ and $kl \gg 1$), for which exact results are known. We will first study the long wavelength limit, i.e., $kl=0$. In this case Eq. (23) reads in real space

$$\delta\varepsilon^*_{ij} \approx v\langle \delta\varepsilon^2\rangle G^*_{ij}(r=0) = -\frac{\delta_{ij}\langle\delta\varepsilon^2\rangle}{d\varepsilon^*_l} \tag{28}$$

(where v is the volume of the grain proportional to l^3). This implies that

$$\varepsilon^*_l = \varepsilon_0 - \frac{\langle\delta\varepsilon^2\rangle}{d\varepsilon^*_l}\ , \quad \varepsilon^*_t = \varepsilon_0 - \frac{\langle\delta\varepsilon^2\rangle}{d\varepsilon^*_l}\ . \tag{29}$$

We thus observe that $\varepsilon^*_l = \varepsilon^*_t$, as expected in this limit. We also note that these equations are the same as Hori's one (for a Gaussian disorder) in the long wavelength limit. These equations were studied in [13,14].

In the case of very high (spatial) frequency, where kl is tending towards infinity, we obtain

$$\delta\varepsilon^*_{ij}(k) \approx \langle\delta\varepsilon^2\rangle G^*_{ij}(k)\ , \tag{30}$$

leading us to

$$\varepsilon^*_l = \varepsilon_0 - \frac{\langle\delta\varepsilon^2\rangle}{\varepsilon^*_l}\ , \quad \varepsilon^*_t = \varepsilon_0 + O\left[\frac{k_0^2}{k^2}\right]\ . \tag{31}$$

The expression for ε^*_l is the effective permittivity for a one-dimensional medium within this approximation, as can be seen for Eq. (29) with $d=1$, and ε^*_t is the average value (corresponding to the case where d is infinite).

For this limit (kl infinite) the exact result is known and can be demonstrated as follows. For $|r-r'|$ less than the disorder correlation length, the permittivity is constant and equal to $\delta\varepsilon(r)$. The second term of the right-hand side of Eq. (6) can thus be rewritten as (omitting all inessential indices)

$$\begin{aligned}\int dr' G(r-r')\delta\varepsilon(r')E(r') &\approx \delta\varepsilon(r)\int_{|r-r'|<l} dr' G(r-r')E(r') \\ &+ \int_{|r-r'|>l} dr' G(r-r')\delta\varepsilon(r')E(r')\ .\end{aligned} \tag{32}$$

The limit $kl \to \infty$ may also be physically realized with k fixed and l going to infinity. The second integral of Eq. (32) tends towards zero in this limit (since it is a converging integral). The electric field at point r is thus given by

$$E(r) = E_0(r) + \delta\varepsilon(r)\int dr' G(r-r')E(r')\ , \tag{33}$$

which can be rewritten as

$$E(r) = \left[\frac{\varepsilon_0}{\varepsilon(r)}P + Q\right]E_0(r)\ , \tag{34}$$

where P and Q are the usual projector (here in real space). From the expression of $\langle\varepsilon(r)E(r)\rangle$ and $\langle E(r)\rangle$ deduced from (34), we obtain the effective permittivity:

$$\varepsilon^*_l = \left\langle\frac{1}{\varepsilon}\right\rangle^{-1}\ , \quad \varepsilon^*_t = \langle\varepsilon\rangle\ . \tag{35}$$

This is consistent with (31) since ε_l is the exact result for one-dimensional systems.

V. CONCLUSION

In this paper we have used the replica method together with a variational method in order to study the propagation of a wave traveling in a disordered composite. To do this, we compute the effective permittivity tensor, which takes into account the effects of spatial dispersion. Our calculation is not perturbative in the frequency, and we obtain self-consistent formulas for the longitudinal and transverse parts of the permittivity tensor applicable to the whole range of frequency.

We have studied the case of correlated Gaussian disorder and obtained a self-consistent equation for the permittivity tensor. This equation is a natural self-consistent generalization of the weak-disorder expansion and should therefore hold even for the strong disorder case. We also checked that our solution is correct in the limits of high and low spatial frequency where exact results are known.

The complete study of our equations (in particular the interesting case when the wavelength is of order the size of inhomogeneities) will be presented in a forthcoming paper. We are also currently investigating the possibility of applying this method to the case of binary correlated disorder, which is substantially more complicated technically.

APPENDIX: VARIATIONAL PRINCIPLE

We have to average the operator M^{-1} over the disorder. We thus have to compute the quantity

$$\mathcal{J} = \left\langle \exp\left[\int dr\, dr' \sum_{ij,a} \phi_i^{*a}(r) G_{ij}(r-r')\delta\varepsilon(r',\omega)\phi_j^a(r')\right]\right\rangle\ . \tag{A1}$$

This average can easily be performed, and we can write the average of M^{-1} as

$$\langle M^{-1}\rangle_{ij}(r-r')=\int \Pi_{i,a}\mathcal{D}(\phi_i^{*a},\phi_i^a)\frac{\vec{\phi}_i^*\cdot\vec{\phi}_j}{n}e^{-\mathcal{H}_{\text{eff}}} , \tag{A2}$$

where the effective Hamiltonian is

$$\mathcal{H}_{\text{eff}}=\int dr \sum_{i,a}|\phi_i^a(r)|^2-\tfrac{1}{2}\int dr_1 dr_2 C(r_1-r_2)-\sum_{iji'j'ab}\int dr\, dr' G_{ij}(r-r_1)G_{i'j'}(r'-r_2)\phi_i^{*a}(r)\phi_j^a(r_1)\phi_{i'}^{*b}(r')\phi_{j'}^b(r_2) . \tag{A3}$$

As usual in the replica method, the average over the disorder introduces a coupling between different replicas. In order to study this Hamiltonian, we will use a variational method.

In order to simplify the presentation, we shall assume a replica symmetric ansatz. We shall not address the problem of replica symmetry breaking since we believe that, as in the quasistatic case, there is no replica symmetry breaking [1]. This probably corresponds to the self-averaging property of the effective permittivity. We therefore assume a Gaussian ansatz with kernel K^{-1} diagonal in replica space, but we shall make no further assumption concerning other indices:

$$Z_0=\int \Pi_{ia}\mathcal{D}(\phi_i^{*a},\phi_i^a)\exp\left[-\int dr\, dr' \sum_{i,j,a}\phi_i^{*a}(r)K_{ij}^{-1}(r-r')\phi_j^a(r')\right] . \tag{A4}$$

The contractions are thus given by

$$\langle \phi_i^{*a}(r)\phi_j^b(r')\rangle_0=\delta^{ab}K_{ij}(r-r') , \quad \langle \phi_i^{*a}(r)\phi_j^{*b}(r')\rangle_0=\langle \phi_i^a(r)\phi_j^b(r)\rangle_0=0 , \tag{A5}$$

where $\langle\ \rangle_0$ denotes an average using Z_0.

Using this ansatz in place of $\mathcal{H}_{\text{eff}}$ in (A2), we immediately get

$$\langle M^{-1}\rangle_{ij}(k)\approx\langle \phi_i\phi_j\rangle_0(k)=K_{ij}(k) . \tag{A6}$$

The quantity K minimizes the variational free energy (given by the Gibbs-Bogoliubov inequality)

$$\mathcal{F}[K]=\text{const}+\langle \mathcal{H}_{\text{eff}}\rangle_0+\mathcal{F}_0 , \tag{A7}$$

where $\mathcal{F}_0=-\ln Z_0$.

The variational free energy (per unit volume and per replica) can be written [using Wick's theorem together with formulas (A5)] as

$$f=-\int\frac{d^dk}{(2\pi)^d}\text{Tr}\ln K(k)+\int\frac{d^dk}{(2\pi)^d}\text{Tr}K(k)-\tfrac{1}{2}\langle\delta\varepsilon^2\rangle\int dk_1dk_2C(k_1)\text{Tr}\{K(k_2)G(k_1+k_2)K(k_1+k_2)G(k_2)\}, \tag{A8}$$

where $K(k)$ and $G(k)$ are the Fourier transform of $K(r)$ and $G(r)$. The minimization of the free energy [with respect to $K(k)$] yields Eqs. (19) and (20) of the text.

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