

Electromagnetic response of systems with spatial fluctuations. I. General formalism

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We obtain closed-form expressions for the macroscopic dielectric response of systems with spatial fluctuations, taking into account the local-field effect. The macroscopic response is written, without reference to any specific representation, in terms of the microscopic dielectric operator and the projection operators that extract the average and the fluctuation components of the microscopic fields. The results are very general since few assumptions are made about the nature of the system and of its microscopic response, and very relaxed conditions are imposed on the average and fluctuation projectors. We put special emphasis on systems for which the length scale of the fluctuations is much smaller than the wavelength of light.

I. INTRODUCTION

The calculation of the optical properties of spatially inhomogeneous systems has attracted the attention of many workers since the early stages of development of the electromagnetic theory of light.¹ In this paper we develop a very general formalism to calculate the macroscopic dielectric response of a system, taking into account the effect of spatial fluctuations such as those due to the atomic structure of matter, to density fluctuations, or to any other kind of microscopic inhomogeneities. This effect is generally known as the local-field effect.² These calculations are usually devised in accordance with the specific characteristics of the system and to the nature of the spatial fluctuations. For example, the use of periodicity in the calculations of the local-field effect in crystals³ precludes their extension to nonperiodic structures. However, the formalism developed here offers a unified view and a systematic approach to the problem, and it shows an underlying structure in the macroscopic dielectric response of diverse spatially fluctuating systems.

We define the macroscopic dielectric operator $\hat{\epsilon}_M$ through the relation

$$\mathbf{D}_a = \hat{\epsilon}_M \mathbf{E}_a, \quad (1)$$

where \mathbf{D}_a and \mathbf{E}_a represent the macroscopic average of the displacement and electric field, respectively, and the caret of $\hat{\epsilon}_M$ refers to its operator character, which in the space-time representation takes the form

$$\mathbf{D}_a(\mathbf{r}, t) = \int dt' \int d^3r' \vec{\epsilon}_M(\mathbf{r}, \mathbf{r}'; t - t') \mathbf{E}_a(\mathbf{r}', t'), \quad (2)$$

where translational invariance in time has been assumed. We will restrict ourselves to nonmagnetic systems. Then the optical properties of the system can be expressed in terms of $\vec{\epsilon}_M$ through the solution of the macroscopic Maxwell's equations. By optical properties we understand the dispersion relations of the bulk electromagnetic modes in a boundless system and the reflection and transmission amplitudes, as well as the dispersion relations of surface electromagnetic modes in bound systems.

A well-accepted procedure to determine the macroscopic dielectric operator is to start with a model for the system expressed in terms of a Hamiltonian. Then, through the use of linear-response theory,⁴ one calculates what is known as the microscopic dielectric response $\hat{\epsilon}$ which relates the total displacement field \mathbf{D} to the total electric field \mathbf{E} as

$$\mathbf{D} = \hat{\epsilon} \mathbf{E}. \quad (3)$$

In general, both of these fields possess microscopic fluctuations, induced by the microscopic inhomogeneities of the system, which are coupled to the macroscopic fields by the fluctuations of $\hat{\epsilon}$.

Since the macroscopic operator $\hat{\epsilon}_M$ relates the average parts of the displacement and electric fields, one requires, as the next step, an averaging procedure to relate $\hat{\epsilon}$ to $\hat{\epsilon}_M$. In this procedure, the influence of the spatial fluctuations of $\hat{\epsilon}$ must be incorporated in the macroscopic dielectric operator. The construction of a general formalism to handle this averaging procedure is the central issue of this paper. We concentrate our attention on systems with spatial fluctuations whose characteristic length scale is much less than the wavelength of light.

The main advantages of the formalism developed here are its simplicity of structure and its generality. In order to make this generality explicit and to show how our formalism can be used in actual calculations, in the following paper of this series⁵ we obtain the macroscopic dielectric response of four different systems: the bulk crystal, the polarizable liquid, the semi-infinite crystal, and the rough surface.

The paper is organized as follows: In Sec. II we obtain an exact expression for the macroscopic dielectric operator of nonmagnetic systems in terms of the microscopic dielectric response. For this purpose we discuss Maxwell's microscopic equations and the properties of the average and fluctuation projectors. In Sec. III we use the longitudinal and transverse projectors in order to obtain several approximate expressions for the macroscopic response of systems with small length-scale fluctuations.

The relationship between the macroscopic dielectric response and other useful microscopic response functions is discussed in Sec. IV, and Sec. V is devoted to conclusions.

II. FORMALISM

Maxwell's equations in a medium are

$$\begin{aligned} \nabla \cdot \mathbf{D} &= 4\pi\rho_{\text{ex}}, \quad \nabla \cdot \mathbf{B} = 0, \\ \nabla \times \mathbf{E} &= -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{j}_{\text{ex}} + \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, \end{aligned} \quad (4)$$

where ρ_{ex} and \mathbf{j}_{ex} are the external charge and current density, respectively, and all the other symbols have their usual meaning. It is usually assumed⁶ that these equations describe macroscopic fields that have been submitted to an averaging procedure which erases the microscopic fluctuations induced by the microscopic inhomogeneities of the system. Nevertheless, it is also possible to define \mathbf{D} and \mathbf{H} to include the microscopic fluctuations, and still obey Maxwell's equations as given in Eqs. (4). As an example, consider⁷

$$\begin{aligned} \mathbf{D}(\mathbf{r}, t) &\equiv \mathbf{E}(\mathbf{r}, t) + 4\pi \int_{-\infty}^t dt' \mathbf{j}(\mathbf{r}, t'), \\ \mathbf{H}(\mathbf{r}, t) &= \mathbf{B}(\mathbf{r}, t), \end{aligned} \quad (5)$$

where \mathbf{j} is the quantum expectation value of the induced current density. This definition might lead to some difficulties,⁸ such as a singularity of \mathbf{D} at the surface of magnetic materials due to the presence of surface currents. We will restrict ourselves to nonmagnetic materials, and so we will not deal with these difficulties.

The problem can now be stated as follows: Starting from a given microscopic dielectric response $\hat{\epsilon}$, derive an equation which relates only the average parts of the fields as in Eq. (1). There are different averaging procedures from which to choose the most adequate, according to the specific characteristics of the system under study as well as the particular length scales involved. Thus, we formally define two operators \hat{P}_a and $\hat{P}_f = \hat{1} - \hat{P}_a$ ($\hat{1}$ is the unit operator) which extract the average component $F_a = \hat{P}_a F$ and the fluctuation component $F_f = \hat{P}_f F$ of any function

$$F = F_a + F_f \rightarrow \begin{pmatrix} F_a \\ F_f \end{pmatrix}, \quad (6)$$

where we introduce the column-vector notation for later convenience. Since a second average cannot remove from F_a the fluctuations that were already removed from F by the first averaging, \hat{P}_a should be idempotent,

$$\hat{P}_a^2 = \hat{P}_a, \quad (7)$$

and thus

$$\hat{P}_f^2 = \hat{P}_f, \quad (8)$$

$$\hat{P}_a \hat{P}_f = \hat{P}_f \hat{P}_a = 0, \quad (9)$$

which simply means that \hat{P}_a and \hat{P}_f are projection operators.

Also, since the average part of the fields must obey the

macroscopic Maxwell's equations, we demand that \hat{P}_a commutes with the space and time differential operators.

As specific examples of averaging procedures, we mention ensemble-average⁹

$$F_a(\lambda) = \sum_c P_c F_c(\lambda), \quad (10)$$

spatial-average¹⁰

$$F_a(\mathbf{r}) = \int d^3r' P_a(\mathbf{r} - \mathbf{r}') F(\mathbf{r}'), \quad (11)$$

and wave-vector truncation¹¹

$$F(\mathbf{q}) \equiv P_a(\mathbf{q}) F(\mathbf{q}), \quad (12)$$

where P_c is the probability of finding the system in the configuration c of the ensemble, and $F_c(\lambda)$ is the value of a function $F(\lambda)$ in that configuration of arbitrary argument λ , $P_a(\mathbf{r})$ is a weight function, and $P_a(\mathbf{q})$ cuts off the high wave-vector components of the Fourier transform $F(\mathbf{q})$ of the function $F(\mathbf{r})$. There are still other useful procedures such as time averaging. The appropriate choice of which procedure to use depends on the nature of the system. For example, in crystalline solids an ensemble or time average does not erase the spatial fluctuations due to the lattice periodicity. We also want to point out that since \hat{P}_a is idempotent [Eq. (7)], in addition to being smooth,¹² $P_a(\mathbf{r})$ should obey

$$\int d^3r' P_a(\mathbf{r} - \mathbf{r}') P_a(\mathbf{r}') = P_a(\mathbf{r}), \quad (13)$$

which is not satisfied by any positive definite weight function. For the same reason, its Fourier transform should obey

$$P_a^2(\mathbf{q}) = P_a(\mathbf{q}), \quad (14)$$

which implies that $P_a(\mathbf{q})$ has the value 1 for small \mathbf{q} , and changes abruptly to 0 for large \mathbf{q} .

It seems to us that the requirement of idempotency has not been recognized previously in the literature, and nonidempotent averages—such as a spatial average with an arbitrary smooth weight function $P_a(\mathbf{r})$ (Ref. 12) or wave-vector truncation with a smooth cutoff $P_a(\mathbf{q})$ (Ref. 11)—have frequently been used. These averages may still be used in our formalism, but then our results would be restricted to the class of fields F for which

$$\hat{P}_a \hat{P}_a F = \hat{P}_a F. \quad (15)$$

It can be shown that when F has two very different length scales of variation, such as when light with wavelength λ shines on a crystal with lattice parameter $a \ll \lambda$, Eq. (15) is satisfied approximately by almost any smooth, normalized weight function.

Now, we rewrite Eq. (3), projecting it into the subspaces a and f , as

$$\begin{pmatrix} \mathbf{D}_a \\ \mathbf{D}_f \end{pmatrix} = \begin{pmatrix} \hat{\epsilon}_{aa} & \hat{\epsilon}_{af} \\ \hat{\epsilon}_{fa} & \hat{\epsilon}_{ff} \end{pmatrix} \begin{pmatrix} \mathbf{E}_a \\ \mathbf{E}_f \end{pmatrix}, \quad (16)$$

where we wrote

$$\hat{\epsilon} = \hat{\epsilon}_{aa} + \hat{\epsilon}_{af} + \hat{\epsilon}_{fa} + \hat{\epsilon}_{ff} \rightarrow \begin{pmatrix} \hat{\epsilon}_{aa} & \hat{\epsilon}_{af} \\ \hat{\epsilon}_{fa} & \hat{\epsilon}_{ff} \end{pmatrix} \quad (17)$$

in matrix form, and we define

$$\hat{O}_{\alpha\beta} \equiv \hat{P}_\alpha \hat{O} \hat{P}_\beta, \quad \alpha, \beta = a, f, \quad (18)$$

for any operator \hat{O} .

Thus the problem of calculating the macroscopic dielectric response [Eq. (1)] is to decouple \mathbf{D}_a and \mathbf{E}_a from Eq. (16), by finding a relationship between \mathbf{E}_f and \mathbf{E}_a . This we obtain from Maxwell's equations for the microscopic fields,

$$\nabla \times \nabla \times \mathbf{E} = \frac{4\pi i}{c^2} \mathbf{j}_{\text{ext}} + \frac{\omega^2}{c^2} \mathbf{D}. \quad (19)$$

If the external current has no fluctuations¹³ the fluctuating part of Eq. (19) can be written as

$$\left[\hat{\epsilon}_{ff} - \frac{c^2}{\omega^2} (\hat{\nabla} \times \hat{\nabla} \times)_{ff} \right] \mathbf{E}_f = -\hat{\epsilon}_{fa} \mathbf{E}_a, \quad (20)$$

where we used Eq. (16), and we put a caret over ∇ to emphasize its operator nature and to make our results representation independent; $\hat{\nabla}$ becomes $(\partial_x, \partial_y, \partial_z)$ in \mathbf{r} space, and it becomes $i\mathbf{q}$ in \mathbf{q} space. Solving Eq. (20) for \mathbf{E}_f , substituting back in Eq. (16) and comparing the result with Eq. (1) we obtain

$$\hat{\epsilon}_M = \hat{\epsilon}_{aa} - \hat{\epsilon}_{af} \left[\hat{\epsilon}_{ff} - \frac{c^2}{\omega^2} (\hat{\nabla} \times \hat{\nabla} \times)_{ff} \right]^{-1} \hat{\epsilon}_{fa}. \quad (21)$$

This result is exact. The macroscopic dielectric response of a system is given by the average of its microscopic response, plus a correction due to the coupling between

the average and the fluctuating part of the fields. This correction is the local-field effect.

III. MICROSCOPIC SPATIAL FLUCTUATIONS

In this section we analyze our previous result in the cases in which the characteristic length scale of the spatial fluctuations is much less than the wavelength of light, what we call microscopic spatial fluctuations.

In order to carry out this analysis, it is convenient to introduce the longitudinal (L)

$$\hat{P}^L = \hat{\nabla} \hat{\nabla}^{-2} \hat{\nabla}, \quad (22)$$

and the transverse (T)

$$\hat{P}^T = -\hat{\nabla} \times \hat{\nabla}^{-2} \hat{\nabla} \times, \quad (23)$$

projection operators, such that $\mathbf{F}^L \equiv \hat{P}^L \mathbf{F}$ is the irrotational and $\mathbf{F}^T \equiv \hat{P}^T \mathbf{F}$ the solenoidal components of an arbitrary vector field $\mathbf{F} = \mathbf{F}^L + \mathbf{F}^T$. Here we introduced the inverse of the Laplacian operator $\hat{\nabla}^{-2}$ represented in \mathbf{r} space by the integral operator which acting on any function $G(\mathbf{r})$ yields

$$(\hat{\nabla}^{-2} G)(\mathbf{r}) = \frac{1}{4\pi} \int d^3 r' \frac{1}{|\mathbf{r} - \mathbf{r}'|} G(\mathbf{r}'), \quad (24)$$

and represented in \mathbf{q} space by $-1/q^2$. \hat{P}^L and \hat{P}^T satisfy

$$\begin{aligned} \hat{P}^L \hat{P}^L &= \hat{P}^L, & \hat{P}^T \hat{P}^T &= \hat{P}^T, \\ \hat{P}^L \hat{P}^T &= \hat{P}^T \hat{P}^L = 0, \\ \hat{P}^L + \hat{P}^T &= \hat{1}, \end{aligned} \quad (25)$$

and they commute with P_a and P_f .

Now we project Eq. (21) onto the longitudinal and transverse subspaces, and write the inverse of the operator within large parentheses:

$$\begin{aligned} [\hat{\epsilon}_{ff} - (\hat{\nabla} \times \hat{\nabla} \times)_{ff}]^{-1} &= \begin{pmatrix} \hat{\epsilon}_{ff}^{LL} & \hat{\epsilon}_{ff}^{LT} \\ \hat{\epsilon}_{ff}^{TL} & \hat{\epsilon}_{ff}^{TT} + \frac{c^2}{\omega^2} \hat{\nabla}^2 \hat{P}^T \hat{P}^T \end{pmatrix}^{-1} \\ &= \begin{pmatrix} (\hat{\epsilon}_{ff}^{LL})^{-1} & 0 \\ 0 & 0 \end{pmatrix} + \frac{\omega^2}{c^2} \begin{pmatrix} (\hat{\epsilon}_{ff}^{LL})^{-1} \hat{\epsilon}_{ff}^{LT} \hat{\nabla}^{-2} \hat{\epsilon}_{ff}^{TL} (\hat{\epsilon}_{ff}^{LL})^{-1} & -(\hat{\epsilon}_{ff}^{LL})^{-1} \hat{\epsilon}_{ff}^{LT} \hat{\nabla}^{-2} \\ -\hat{\nabla}^{-2} \hat{\epsilon}_{ff}^{TL} (\hat{\epsilon}_{ff}^{LL})^{-1} & \hat{\nabla}^{-2} \hat{P}^T \hat{P}^T \end{pmatrix} + \dots \end{aligned} \quad (26)$$

as a series in which each consecutive term is smaller than the previous one by a factor $||(\omega^2/c^2) \hat{\nabla}^{-2} \hat{P}^T|| \sim l^2/\lambda^2$, where l is the length scale of variation of the fluctuations of the field, and $\lambda = c/2\pi\omega$ is the free-space wavelength. Here we defined

$$\hat{O}^{AB} = \hat{P}^A \hat{O} \hat{P}^B, \quad A, B = L, T \quad (27)$$

and used a notation similar to that of Eq. (17). In the cases in which $l \ll \lambda$, we can retain only the first term in Eq. (26), and write (21) as

$$\hat{\epsilon}_M = \hat{\epsilon}_{aa} - \hat{\epsilon}_{af} (\hat{\epsilon}_{ff}^{LL})^{-1} \hat{\epsilon}_{fa}. \quad (28)$$

This is our main result for systems with microscopic spatial fluctuations.

To make the expansion (26), and therefore to obtain the approximate result (28), we assumed $\lambda^2/l^2 \gg ||\hat{\epsilon}||$. If the system has an undamped resonance, this condition may be violated in its immediate vicinity. The validity of macroscopic theories when the wavelength of light inside the medium $\lambda/||\hat{\epsilon}_M||^{1/2}$ is of the order of atomic dimensions has been questioned and discussed by Sipe and Van Kranendonk.¹⁴

Note that if we choose a spatial average or a wave-vector truncation, then $l \ll \lambda$ is a condition imposed on the averaging procedure \hat{P}_a , i.e., which wave vectors

should be considered macroscopic. On the other hand, if we choose an ensemble average, then $l \ll \lambda$ is a condition imposed on the physical system itself, i.e., on the length scale of its thermodynamic fluctuations. For this reason, the macroscopic response of systems such as a fluid near its critical point, for which there are fluctuations on all length scales, cannot be obtained from Eqs. (28).¹⁵

It can be shown that our neglect of all terms of order l^2/λ^2 and higher in Eq. (26) is equivalent to the neglect of the transverse fluctuating electric field. That it is negligible can be seen by considering a system of polarized molecules. The part of the electric field which fluctuates with a length scale l is produced by molecules at a distance $\sim l$ from the observation point. If the molecules have a dipole moment p , the Coulomb (longitudinal) part of this field is proportional to p/l^3 , while its radiation (transverse) part is proportional to $\omega^2 p/c^2 l$, so it is l^2/λ^2 times smaller (for a more general discussion, see Refs. 16 and 17). Therefore, the effects of light scattering, such as a complex ϵ_M for systems with no true absorption,¹⁵ are not taken into account by Eq. (28). However, we believe that the vast amount of systems with spatial fluctuations of microscopic length scales^{5,17-19} and whose macroscopic response can be obtained from Eq. (28) following a unified approach, makes ours a very attractive formalism. For systems with significant fluctuations of long length scales, the exact result, Eq. (21), may be used.²⁰

In a similar way, starting from the microscopic equation

$$\mathbf{E} = \hat{\epsilon}^{-1} \mathbf{D}, \quad (29)$$

we calculate the macroscopic inverse dielectric operator defined by

$$\mathbf{E}_a = \hat{\epsilon}_M^{-1} \mathbf{D}_a, \quad (30)$$

and we obtain

$$\hat{\epsilon}_M^{-1} = (\hat{\epsilon}^{-1})_{aa} - (\hat{\epsilon}^{-1})_{af} [(\hat{\epsilon}^{-1})_{ff}^{\text{TT}}]^{-1} (\hat{\epsilon}^{-1})_{fa}, \quad (31)$$

where

$$[(\hat{\epsilon}^{-1})_{ff}^{\text{TT}}]^{-1} (\hat{\epsilon}^{-1})_{ff}^{\text{TT}} = \hat{P}_f \hat{P}^{\text{T}}. \quad (32)$$

It can easily be checked that the expressions given by Eqs. (28) and (31) obey

$$\hat{\epsilon}_M^{-1} \hat{\epsilon}_M = \hat{\epsilon}_M \hat{\epsilon}_M^{-1} = \hat{P}_a. \quad (33)$$

Note that \hat{P}_a is the same as the identity operator when restricted to the nonfluctuating fields on which the macroscopic response ought to operate.

It can also be easily shown, using the partitioning theorem,²¹ that the longitudinal-longitudinal projection of Eq. (28) and the transvers-transverse projection of Eq. (31) can be written simply as

$$(\hat{\epsilon}_M^{\text{LL}})^{-1} = (\hat{\epsilon}^{\text{LL}})_{aa}^{-1}, \quad (34)$$

and

$$[(\hat{\epsilon}_M^{-1})^{\text{TT}}]^{-1} = [(\hat{\epsilon}^{-1})^{\text{TT}}]_{aa}^{-1}. \quad (35)$$

In these equations the a - f coupling is taken into account by first inverting and then taking the average.

We recall that in the absence of longitudinal-transverse (LT) coupling, as in the case of an isotropic system or a system with cubic symmetry, the full macroscopic description of the dielectric response is given solely in terms of $\hat{\epsilon}_M^{\text{LL}}$ and $(\hat{\epsilon}_M^{-1})^{\text{TT}}$.

To simplify the inversion procedures which appear in Eqs. (28), (31), (34), and (35) we have found it convenient to use the relations

$$\begin{aligned} [(\hat{\epsilon}^{-1})^{\text{TT}}]^{-1} &= \hat{\epsilon}^{\text{TT}} - \hat{\epsilon}^{\text{TL}} (\hat{\epsilon}^{\text{LL}})^{-1} \hat{\epsilon}^{\text{LT}} \\ &= \hat{\epsilon} - \hat{\epsilon} (\hat{\epsilon}^{\text{LL}})^{-1} \hat{\epsilon} \end{aligned} \quad (36)$$

and

$$\begin{aligned} (\hat{\epsilon}^{\text{LL}})^{-1} &= \hat{\nabla} \hat{\nabla}^{-2} (\hat{\epsilon}^{\text{cc}})^{-1} \hat{\nabla} \\ &= \hat{\nabla} (\hat{\epsilon}^{\phi\phi})^{-1} \hat{\nabla}^{-2} \hat{\nabla}, \end{aligned} \quad (37)$$

where we introduced the scalar operators $(\hat{\epsilon}^{\text{cc}})^{-1}$ and $(\hat{\epsilon}^{\phi\phi})^{-1}$ which we call the charge-charge and the potential-potential inverse dielectric responses. For example, Eq. (34) can then be simply written as

$$\begin{aligned} (\hat{\epsilon}_M^{\text{LL}})^{-1} &= \hat{\nabla} \hat{\nabla}^{-2} (\hat{\epsilon}^{\text{cc}})_{aa}^{-1} \hat{\nabla} \\ &= \hat{\nabla} (\hat{\epsilon}^{\phi\phi})_{aa}^{-1} \hat{\nabla}^{-2} \hat{\nabla}, \end{aligned} \quad (38)$$

where we only have to invert a scalar operator.

Even this type of inversion might not be necessary if we rely on the following interpretation of $(\epsilon^{\text{cc}})^{-1}$ and $(\epsilon^{\phi\phi})^{-1}$: In the absence of transverse electric fields, the external and total charge densities are related by

$$\rho = (\hat{\epsilon}^{\text{cc}})^{-1} \rho_{\text{ext}}, \quad (39)$$

and the external and total Coulomb potentials are related by

$$\phi = (\hat{\epsilon}^{\phi\phi})^{-1} \phi_{\text{ext}}. \quad (40)$$

Thus $(\hat{\epsilon}^{\text{cc}})^{-1}$ and $(\epsilon^{\phi\phi})^{-1}$ can be calculated directly from the response of the system to an external perturbation by neglecting retardation effects.

If we now write ρ_{ext} in terms of an external polarization field, $\rho_{\text{ext}} = -\nabla \cdot \mathbf{P}_{\text{ext}}$, and then use Eq. (37), we can write

$$\mathbf{E}^{\text{L}} = -4\pi (\hat{\epsilon}^{\text{LL}})^{-1} \mathbf{P}_{\text{ext}}. \quad (41)$$

Then $(\hat{\epsilon}^{\text{LL}})^{-1}$ is the external-polarization—electric-field response function in the absence of retardation.

The results we obtained in this and the previous sections are very general although they are written in a quite abstract notation. In the next paper of this series⁵ we illustrate their use in actual calculations of the dielectric and optical properties of several systems of physical interest.

IV. FURTHER DEVELOPMENTS

Our starting point in the previous sections was the microscopic dielectric response. A very important problem we did not consider there is the calculation of this response from the Hamiltonian operator of the system. This is usually done by dividing the Hamiltonian H into a nonperturbed part H_0 and a time-dependent perturbation H_1 , and the time-dependent polarization induced by this perturbation is calculated using linear-response theory.

The nature of the microscopic response function obtained using this scheme depends on how H_0 and H_1 are chosen.

For example, in the random-phase approximation (RPA),²² H_0 contains no electron-electron interaction and H_1 contains the interaction between the electrons and the total electric field. Thus in the RPA one obtains an approximation to the microscopic susceptibility $\hat{\chi} = (\hat{\epsilon} - \hat{1})/4\pi$ from which the macroscopic dielectric response could be calculated using the results of this paper. However, if we keep in H_0 the full electron-electron interaction, H_1 would contain the interaction with the external field only and we would obtain an external susceptibility $\hat{\chi}_{\text{ext}}$ defined by

$$\mathbf{P} = \hat{\chi}_{\text{ext}} \mathbf{E}_{\text{ext}}. \quad (42)$$

Similarly, if we keep in H_0 the longitudinal electron-electron interaction and take account of the transverse interaction by including in H_1 the interaction with the polarization field²³

$$\mathbf{E}_P \equiv \mathbf{E}_{\text{ext}} + \mathbf{E}_{\text{ind}}^T, \quad (43)$$

where $\mathbf{E}_{\text{ind}}^T$ is the transverse induced field, we would obtain an approximation to the pseudosusceptibility $\hat{\chi}_P$ defined by

$$\mathbf{P} = \hat{\chi}_P \mathbf{E}_P. \quad (44)$$

In this case, the results derived previously are not immediately applicable. However, our formalism can be easily extended²³ so that, using the same approximations as before, we obtain the following expressions for the macroscopic dielectric response in terms of $\hat{\chi}_{\text{ext}}$ and $\hat{\chi}_P$:

$$\hat{\epsilon}_M = \hat{1} + 4\pi(\hat{\chi}_{\text{ext}})_{aa} \left[\hat{1} - 4\pi \left[\hat{P}^L + \frac{\omega^2}{c^2} \hat{\square}^{-1} \hat{P}^T \right] (\hat{\chi}_{\text{ext}})_{aa} \right]^{-1}, \quad (45)$$

$$\hat{\epsilon}_M = \hat{1} + 4\pi(\hat{\chi}_P)_{aa} [\hat{1} - 4\pi \hat{P}^L (\hat{\chi}_P)_{aa}]^{-1}, \quad (46)$$

where $\hat{\square}^{-1}$ is the inverse of the D'Alembertian operator $\hat{\square} \equiv \nabla^2 + \omega^2/c^2$. This expression will be the object of further research.

V. CONCLUSIONS

We have obtained very general expressions [Eqs. (28), (31), (34), and (35)] for the macroscopic dielectric response of systems with microscopic spatial fluctuations in terms of their microscopic dielectric response. The only approximation made in the derivation of our main results was the neglect of the transverse component of the microscopic fluctuations of the electric field. This approximation is appropriate whenever the microscopic scale of variation is much smaller than the free-space wavelength of the fields involved: this is the case for most systems which can be described from a macroscopic point of view.

Our results can be applied to a very wide range of systems. The reason for this is that in their derivation we avoided particular representations of the fields and operators involved. We also introduced average and fluctuation projectors which were left almost unspecified; only their most important properties were required. In the next paper of this series⁵ we obtain from these results in a unified way expressions for the dielectric and the optical properties of systems of the most diverse nature.

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