Effective dielectric function of a random medium

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(Received 22 June 1999)

We derive an expression for the effective dielectric function of a multicomponent mixture with small heterogeneities. The result is generalized to the case of a continuous medium with fluctuating dielectric function. Besides, we obtain an original expression for the effective dielectric function of a dilute suspension of spherical beads. Our result differs from the previously known one and appears to be physically more correct at higher bead volume fractions. [S0163-1829/99/6006841-1]

I. INTRODUCTION

The dielectric function $\varepsilon(\omega)$ is one of the most important quantities describing the properties of a medium with respect to electromagnetic waves (light). In homogeneous media, the square root of the dielectric function determines the phase velocity $v_p$ and the extinction coefficient $\kappa$ of light of frequency $\omega$: $v_p(\omega) = c / \text{Re} \sqrt{\varepsilon(\omega)}$, $\kappa = (\omega/c) \text{Im} \sqrt{\varepsilon(\omega)}$, with $c$ the free space speed of light, and Re and Im denoting the real and imaginary parts of a complex argument, respectively.

In random media, the dielectric function is not solely a function of frequency $\omega$ of the wave but also a random function of the spatial coordinate $\mathbf{r}$: $\varepsilon = \varepsilon(\mathbf{r}, \omega)$. If the spatial scale $d$ of heterogeneities of the dielectric function is large ($d \gg \lambda$; $\lambda$ is the wavelength of light), the spatial dependence appears in the phase velocity and the extinction coefficient: $v_p = v_p(\mathbf{r}, \omega)$, $\kappa = \kappa(\mathbf{r}, \omega)$. It is the limit of geometrical optics: a photon path in the medium can be considered as consisting of consequent parts, each inside a domain of uniform $\varepsilon(\omega)$. In contrast, if $d$ is of the order of $\lambda$, the square root of $\varepsilon(\mathbf{r}, \omega)$ loses physical meaning and more sophisticated approaches are required to calculate the velocity\textsuperscript{2,3} and the extinction coefficient\textsuperscript{4,5}.

In the present paper we consider random media with typical size $d$ of heterogeneities of $\varepsilon(\mathbf{r}, \omega)$ small compared with the wavelength: $d \ll \lambda$. At the same time, $d$ is believed to be large enough to ensure that the volume $d^3$ is macroscopic (i.e., contains a large number of atoms or molecules). For such random media, the effective dielectric function $\varepsilon_{\text{eff}}(\omega)$ is commonly defined by\textsuperscript{1}

$$D(\mathbf{r}, \omega) = \varepsilon_{\text{eff}}(\omega) E(\mathbf{r}, \omega),$$

(1)

where $E$ and $D$ are the electric field and induction in a monochromatic electromagnetic wave of frequency $\omega$, respectively; an overbar denotes averaging over a volume $V$ such that $d^3 \ll V \ll \lambda^3$.

$$\langle \cdots \rangle = \frac{1}{V} \int_V \cdots d^3 \mathbf{r}.$$ (2)

The phase velocity and the extinction coefficient of light are then determined by the square root of $\varepsilon_{\text{eff}}$ in the same way as for homogeneous media: $v_p(\omega) = c / \text{Re} \sqrt{\varepsilon_{\text{eff}}(\omega)}$, $\kappa = (\omega/c) \text{Im} \sqrt{\varepsilon_{\text{eff}}(\omega)}$. To simplify the notation, we drop the $\omega$ dependence of $\varepsilon_{\text{eff}}$, $E$, and $D$ in the rest of the paper.

There exists an extensive literature on the subject of the calculation of $\varepsilon_{\text{eff}}$ for random media with heterogeneities smaller than the wavelength (see, e.g., Refs. 6 and 7 and references therein). The problem has been approached by representing the effective dielectric function as a series in the dielectric contrast between various components of a random medium\textsuperscript{1,8,9} as well as in the volume fraction of one of the components (for two-component media).\textsuperscript{1} More general results have been obtained using the so-called renormalization technique.\textsuperscript{10} It is worth while to note that the methods applied for the calculation of the effective dielectric function of random media have a lot of common features with the techniques for the calculation of the conductivity of disordered metal samples\textsuperscript{11,12} and the elasticity of microheterogeneous media.\textsuperscript{13–15}

The purpose of this paper is to present and to discuss the following two important results. First, we find an original expression for the effective dielectric function of a multicomponent mixture. The expression is, to our knowledge, the first nonperturbative result for $\varepsilon_{\text{eff}}$. All previous results were series expansions in some small parameter and are only of use when the parameter is really small. We generalize our result to the case of a continuous medium with fluctuating dielectric function. Next, we provide a derivation of the effective dielectric function of a dilute suspension of small spherical beads. Our result differs from the commonly recognized one. Although we used the same level of approximations as in Ref. 1 and in many other papers, our expression for $\varepsilon_{\text{eff}}$ of a dilute suspension remains qualitatively correct even for high volume fractions of suspended particles.

II. DIELECTRIC FUNCTION OF A MIXTURE

Consider a random mixture of $N$ components characterized by dielectric functions $\varepsilon_i (i = 1, \cdots, N)$. We denote the volume fraction of the $i$th component by $\Phi_i$. The components are distributed in space in a way that the linear dimensions of regions filled by each of the components are all much smaller than the wavelength $\lambda$ of the electromagnetic wave (light) considered.

The electric field $E$ and the dielectric function $\varepsilon$ at a par-
ticular site \( \mathbf{r} \) can be represented as sums of mean values and deviations:

\[
\mathbf{E}(\mathbf{r}) = \bar{\mathbf{E}} + \delta \mathbf{E}(\mathbf{r}), \quad \varepsilon(\mathbf{r}) = \bar{\varepsilon} + \delta \varepsilon(\mathbf{r}).
\]

(3)

In Ref. 1, the calculation of the effective dielectric function of a mixture has been performed for \( |\varepsilon_1 - \bar{\varepsilon}| \ll \bar{\varepsilon} \). The following result has been obtained:

\[
\varepsilon_{\text{eff}} = \bar{\varepsilon} - \frac{(\delta \varepsilon)^2}{3 \bar{\varepsilon}}.
\]

(4)

This result is only valid in the second order of \( \delta \varepsilon / \bar{\varepsilon} \) and is therefore unacceptable for media with large fluctuations of the dielectric function. Below we present an alternative calculation of the effective dielectric function of a multicomponent disperse mixture. Our expression for \( \varepsilon_{\text{eff}} \) is valid for \( |\varepsilon_i - \bar{\varepsilon}| < \bar{\varepsilon} \) and coincides with Eq. (4) for \( |\varepsilon_i - \bar{\varepsilon}| \ll \bar{\varepsilon} \).

We start our derivation by averaging the relation \( \mathbf{D}(\mathbf{r}) = \varepsilon(\mathbf{r}) \mathbf{E}(\mathbf{r}) \) over a volume \( V \), \( dV \ll V \ll \lambda^3 \), as defined by Eq. (2). As the mean values of \( \delta \mathbf{E}(\mathbf{r}) \) and \( \delta \varepsilon(\mathbf{r}) \) are zeros by definition, we have

\[
\bar{\mathbf{D}} = \bar{\mathbf{E}} \bar{\varepsilon} + \bar{\varepsilon} \delta \mathbf{E}.
\]

(5)

As \( \varepsilon_{\text{eff}} \) represents a factor of proportionality between \( \bar{\mathbf{D}} \) and \( \bar{\mathbf{E}} \) [see Eq. (1)], in accordance with Eq. (5) we need to average \( \delta \varepsilon \) \( \delta \mathbf{E} \) in order to find \( \varepsilon_{\text{eff}} \). Let us first average \( \delta \mathbf{E} \) over a volume filled by one and the same mixture component. For this purpose, we note that one of the Maxwell equations, \( \text{div} \mathbf{D} = 0 \), with account for Eqs. (3) becomes

\[
(\bar{\varepsilon} + \delta \varepsilon) \text{div} \mathbf{E} + [\bar{\mathbf{E}} + \delta \mathbf{E}] \cdot \mathbf{V} \delta \varepsilon = 0.
\]

(6)

Inasmuch as due to isotropic nature of the mixture as a whole,

\[
\frac{\partial}{\partial x} \delta \mathbf{E}_x + \frac{\partial}{\partial y} \delta \mathbf{E}_y + \frac{\partial}{\partial z} \delta \mathbf{E}_z = \frac{1}{3} \text{div} \delta \mathbf{E},
\]

(7)

assuming that \( \bar{\mathbf{E}} + \delta \mathbf{E} \) is parallel to the \( x \) axis, we rewrite Eq. (6) as

\[
3(\bar{\varepsilon} + \delta \varepsilon) \frac{\partial}{\partial x} \delta \mathbf{E}_x + [\bar{\mathbf{E}}_x + \delta \mathbf{E}_x] \frac{\partial}{\partial x} \delta \varepsilon = 0.
\]

(8)

Up to this point, our analysis mimics the derivations of Ref. 1 except Eqs. (6) and (8) which we write without an assumption of small \( |\delta \varepsilon| \) and \( |\delta \mathbf{E}| \) compared to \( \bar{\varepsilon} \) and \( |\bar{\mathbf{E}}| \), respectively. We are not going to take advantage of such an additional assumption in the present paper. Instead, we look for \( \delta \mathbf{E}_x \) as a series:

\[
\delta \mathbf{E}_x = \sum_{n=1}^{\infty} \alpha_n(\delta \varepsilon)^n.
\]

(9)

The series expansion (9) only converges for sufficiently small \( \delta \varepsilon \). An estimate of its radius of convergence \( K \) may be found from Eq. (8). Indeed, substituting the expansion (9) into Eq. (8) and equating to zero the coefficients at all the powers of \( \delta \varepsilon \), we arrive at the following set of linear equations with respect to \( \alpha_n \) (\( n = 1, 2, \cdots \)):

\[
3\bar{\varepsilon} \alpha_1 + \mathbf{E}_x = 0,
\]

\[
6\bar{\varepsilon} \alpha_2 + 4\alpha_1 = 0,
\]

\[
\cdots
\]

\[
3n\bar{\varepsilon} \alpha_n + (3n - 2)\alpha_{n-1} = 0,
\]

\[
\cdots
\]

(10)

The solution of these equations is readily found:

\[
\alpha_1 = - \frac{\mathbf{E}_x}{3\bar{\varepsilon}},
\]

\[
\alpha_n = - \frac{\alpha_{n-1}}{n\bar{\varepsilon}}, \quad n = 2, 3, \ldots .
\]

(11)

The Cauchy test then yields

\[
R \geq \lim_{n \to \infty} \left| \frac{\alpha_n}{\alpha_{n+1}} \right| = \bar{\varepsilon}.
\]

(12)

Hence, expansion (9) is valid at least for \( |\delta \varepsilon| \ll \bar{\varepsilon} \).

As long as \( \alpha_n \sim \bar{\mathbf{E}}_x \) for any \( n \), the series (9) can be rewritten as

\[
\delta \mathbf{E}_x = \sum_{n=1}^{\infty} \beta_n(\delta \varepsilon)^n,
\]

(13)

where \( \beta_n = \alpha_n / \bar{\mathbf{E}}_x \). Applying an equality \( p(p + 1)(p + 2) \cdots (p + n - 1) = \Gamma(p + n) / \Gamma(p) \), which is true for any \( p > 0 \),\(^{16} \) we obtain from Eqs. (11)

\[
\beta_n = (-1)^n \frac{n!}{n!(\bar{\varepsilon})^n} \frac{1}{\Gamma\left(1 + \frac{1}{3}\right)}, \quad n = 1, 2, \ldots .
\]

(14)

Due to the arbitrariness in the choice of the \( x \) axis, a representation analogous to Eq. (13) is also true for \( \delta \mathbf{E} \):

\[
\delta \mathbf{E} = \sum_{n=1}^{\infty} \beta_n(\delta \varepsilon)^n.
\]

(15)

Multiplying Eq. (15) by \( \delta \varepsilon \) and averaging over all the mixture components, we find

\[
\delta \varepsilon \delta \mathbf{E} = \bar{\mathbf{E}} \sum_{n=1}^{\infty} \beta_n(\delta \varepsilon)^{n+1}.
\]

(16)

Substitution of this result into Eq. (5) with account for Eq. (1) gives

\[
\varepsilon_{\text{eff}} = \bar{\varepsilon} + \sum_{n=1}^{\infty} \beta_n(\delta \varepsilon)^{n+1}.
\]

(17)

For a mixture of \( N \) components,

\[
(\delta \varepsilon)^{n+1} = \sum_{i=1}^{N} \Phi_i(\varepsilon_i - \bar{\varepsilon})^{n+1},
\]

and hence Eq. (17) can be rewritten as
III. DIELECTRIC FUNCTION OF A FLUCTUATING MEDIUM

The main result of the previous section, Eq. (21), can be generalized to describe a continuous medium with fluctuating dielectric function $\varepsilon(r)$. Indeed, if we assume that $\Phi(\varepsilon)$ is the probability distribution for the dielectric function at a given site $r$ inside the medium to have a value $\varepsilon$, we obtain from Eq. (21) the following expression for the effective dielectric function of the medium:

$$
\varepsilon_{\text{eff}} = \bar{\varepsilon} + \sum_{i=1}^{N} \Phi_i(\varepsilon_i - \bar{\varepsilon})f\left(1 - \frac{\varepsilon_i}{\bar{\varepsilon}}\right),
$$

(19)

where we introduced

$$
f(x) = \sum_{n=1}^{\infty} \frac{\Gamma(n+1)}{n! \Gamma\left(\frac{2}{3}\right)} x^n.
$$

(20)

If $|x| \gg 1$, then the series (20) diverges and the expansions (9), (13), and (15) are unacceptable. If, however, $|x| < 1$, the series (20) converges,\(^6\) and $f(x) = (1 - x)^{-1/3} - 1$, and we finally find

$$
\varepsilon_{\text{eff}} = \bar{\varepsilon} + \sum_{i=1}^{N} \Phi_i u_i (1 + u_i)^{-1/3}, \quad |u_i| < 1,
$$

(21)

where $u_i = (\varepsilon_i - \bar{\varepsilon})/\bar{\varepsilon}$. The fact that the series (20) sums up for $|\varepsilon_i - \bar{\varepsilon}|/\bar{\varepsilon} < 1$ agrees with our estimate (12) of the radius of convergence of the expansion (9).

It is easy to convince oneself that $\varepsilon_{\text{eff}} \approx \bar{\varepsilon}$, and that Eq. (21) coincides with Eq. (4) in the limit of $u_i \ll 1$. In order to compare Eqs. (4) and (21) we normalize them by $\bar{\varepsilon}$ and plot in Fig. 1 for a particular case of a two-component mixture with $\varepsilon_2 / \varepsilon_1 = 2$. As seen from the figure, the behavior of both results is qualitatively similar, while they differ quantitatively. The ratio of the effective dielectric function, as given by our expression (21), to the average dielectric function $\bar{\varepsilon}$ reaches an absolute minimum at $\Phi_2 = 0.44$. We checked that the position of the minimum remains almost unchanged for $1 < \varepsilon_2 / \varepsilon_1 < 2$.

For a two-component mixture with a small volume fraction of, say, the second component ($\Phi_2 \ll 1$, $\varepsilon_2 > \varepsilon_1$), condition $|u_i| < 1$ becomes $\Phi_2 > 0.5(\varepsilon_2 - 2\varepsilon_1)/(\varepsilon_2 - \varepsilon_1)$. In this case Eq. (21) simplifies:

$$
\varepsilon_{\text{eff}} = \varepsilon_1 + \Phi_2 (\varepsilon_2 - \varepsilon_1) (\varepsilon_1 / \varepsilon_2)^{1/3}.
$$

(22)

IV. DIELECTRIC FUNCTION OF A DILUTE SUSPENSION

In the present section we consider the important case of a dilute suspension of spherical particles with dielectric function $\varepsilon_2$ in a matrix medium with dielectric function $\varepsilon_1$. The particle volume fraction $\Phi_2$ is assumed to be much smaller than unity and the particle diameter $d$ is believed to be well below the wavelength $\lambda$ of light.

There are several ways to calculate the effective dielectric function of a dilute suspension of spherical particles. One of them is taken in Ref. 1 yielding

$$
\varepsilon_{\text{eff}} = \varepsilon_1 + 3\Phi_2 (\varepsilon_2 - \varepsilon_1) (2\varepsilon_1 + \varepsilon_2)^{-1}.
$$

(25)

Another possible way consists in using the scattering theory and leads to the very same result. Below we provide an original calculation of $\varepsilon_{\text{eff}}$ for a dilute suspension. Our derivation is, in our opinion, more transparent than the others. In addition, it leads to a result which differs from Eq. (25).

We start by averaging the electric field $\mathbf{E}$ over a macroscopic volume $V$ of the suspension (as above, we take $d^3 \ll V \ll \lambda^3$). In order to accomplish such an averaging, we first average $\mathbf{E}$ over the volume of the suspended particles (this yields $\mathbf{E}_2$) and (separately) over the volume of the rest of the medium (this yields $\mathbf{E}_1$). The final average is then

$$
\mathbf{E} = (1 - \Phi_2)\mathbf{E}_1 + \Phi_2 \mathbf{E}_2.
$$

(26)

The electric field $\mathbf{E}_2$ inside a dielectric sphere placed in an external electric field $\mathbf{E}_1$ is given by\(^1\)
As the same relation holds for the mean fields \( \bar{E}_1 \) and \( \bar{E}_2 \), with use of Eqs. (26) and (27) we can express \( \bar{E}_1 \) and \( \bar{E}_2 \) through \( \bar{E} \):

\[
\bar{E}_1 = \frac{2\varepsilon_1 + \varepsilon_2}{(2 + \Phi_2)\varepsilon_1 + (1 - \Phi_2)\varepsilon_2} \bar{E}.
\]

\[
\bar{E}_2 = \frac{3\varepsilon_1}{(2 + \Phi_2)\varepsilon_1 + (1 - \Phi_2)\varepsilon_2} \bar{E}.
\]

Now we average the electric induction \( \bar{D} = \varepsilon(r)\bar{E}(r) \) over the same volume \( V \) and get

\[
\bar{D} = (1 - \Phi_2)\varepsilon_1 \bar{E}_1 + \Phi_2\varepsilon_2 \bar{E}_2
\]

\[
= \varepsilon_1 \left[ 1 + 3\Phi_2 \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_1(2 + \Phi_2) + \varepsilon_2(1 - \Phi_2)} \right] \bar{E}.
\]

Comparing this result to Eq. (1) we find the effective dielectric function of a dilute suspension of small spherical particles:

\[
eff = \varepsilon_1 \left[ 1 + 3\Phi_2 \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_1(2 + \Phi_2) + \varepsilon_2(1 - \Phi_2)} \right].
\]

Evidently, this result differs from Eq. (25) although they coincide for \( \Phi_2(\varepsilon_2 - \varepsilon_1) / (2\varepsilon_1 + \varepsilon_2) \ll 1 \). A closer inspection of the derivation presented above reveals that our analysis would yield Eq. (25) if we assume the average field external with respect to a spherical particle to be \( \bar{E} \) instead of \( \bar{E}_1 \). The latter assumption, however, does not seem to be a correct one as the particles are surrounded by the matrix medium, not by the material of the particles themselves.

In Fig. 2 we plot Eqs. (25) and (31) normalized by \( \bar{E} \). The plots are only meaningful for \( \Phi_2 < 0.64 \) since a higher volume fraction cannot be achieved for hard spheres. The figure shows that Eqs. (25) and (31) give very close results for \( \Phi_2 < 0.1 \). Although the higher volume fractions are beyond the region of validity of both equations (\( \Phi_2 \ll 1 \) is assumed during their derivations), Eq. (31) exhibits a physically more correct behavior as it tends to \( \varepsilon_2 \) for \( \Phi_2 \to 1 \). Thus we conclude that our Eq. (31) could be used for interpretation of experimental data at higher volume fractions than Eq. (25), although both equations require \( \Phi_2 \ll 1 \).

V. CONCLUSIONS

The present paper deals with the effective dielectric function \( \varepsilon_{\text{eff}}(\omega) \) of random media in which the heterogeneities of the dielectric function \( \varepsilon_{\text{eff}}(r, \omega) \) are much smaller than the wavelength \( \lambda \) of the electromagnetic wave (light) under consideration. Two significant results are reported.

First, we present a derivation of \( \varepsilon_{\text{eff}} \) of a random \( N \)-component disperse mixture. Our result is only valid for \( |\varepsilon_i - \bar{\varepsilon}| \ll \bar{\varepsilon} \ (i = 1, \cdots, N) \), where \( \varepsilon_i \) is the dielectric function of the \( i \)-th component, and \( \bar{\varepsilon} \) is the average dielectric function of the mixture. The analysis of the obtained expression for the effective dielectric function reveals that the ratio of \( \varepsilon_{\text{eff}} \) to \( \bar{\varepsilon} \) is always less than unity. For a two-component mixture, \( \varepsilon_{\text{eff}} / \bar{\varepsilon} \) exhibits an absolute minimum at \( \Phi_2 \) somewhat below 0.5, with \( \Phi_2 \) a volume fraction of the optically more dense component (only real-valued positive dielectric functions of the components have been explored for the time being). The expression for \( \varepsilon_{\text{eff}} \) of a multicomponent mixture is generalized to the case of a continuous medium with fluctuating dielectric function.

Second, we derive an original expression for the effective dielectric function of a dilute suspension of spherical beads of diameter \( d \ll \lambda \). Our expression differs from the one obtained in Ref. 1, although both results coincide in the first order of the bead volume fraction \( \Phi_2 \). Despite the fact that our expression is derived within the same approximation of \( \Phi_2 \ll 1 \) as the one of Ref. 1, it exhibits physically more correct behavior at higher volume fractions. Thus we expect that it should be more appropriate for the interpretation of experiments. Similarly to the case of a two-component mixture, the ratio of the effective dielectric constant of a dilute suspension to \( \bar{\varepsilon} \) exhibits an absolute minimum at \( \Phi_2 \) somewhat below 0.5. This therefore seems to be a common feature for systems of the considered type.

The reported results leave space for further investigations. First of all, the effect of light absorption in one (or several) suspension (mixture) components on the effective dielectric constant of the whole medium could be studied by using our expressions (21) and (31) with complex-valued dielectric
functions $\varepsilon_i$ of the components. Next, the laser action in mixtures (suspensions) could be investigated in a similar manner. Finally, more sophisticated theories dealing with the most exciting case of heterogeneities of the dielectric function which size is of the order of $\lambda$ could use our expressions to check their results in the limit of infinitely small heterogeneity size.

ACKNOWLEDGMENTS

This work is partially supported by the Russian Foundation for Basic Research (Grant RFBR No. 98-02-17112), International Center for Fundamental Physics in Moscow (through the INTAS 96-0457 project), and Samsung Electronics Co., Ltd., Moscow Research Center.

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