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1995 J. Phys. A: Math. Gen. 28 693

(http://iopscience.iop.org/0305-4470/28/3/022)

J. Phys. A: Math. Gen. 28 (1995) 693–700. Printed in the UK

Effective permittivity of log-normal isotropic random media

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Received 8 August 1994

Abstract. A method of deriving the effective permittivity ε^{eff} of log-normal d-dimensional isotropic media is developed. The approach is based on applying the regularized Green function technique for the Laplace operator. The feasibility of exact determining of the effective permittivity is analysed by deriving ε^{eff} at third order in the log-permittivity variance. It is shown that the 3D effective permittivity is a functional of the spectrum of inhomogeneities and therefore cannot be expressed by a closed formula. In particular, the formula for the effective permittivity discussed previously by Landau and Lifshitz and Matheron deviates from the exact one in the third-order term.

An important concept in electrodynamics of randomly inhomogeneous media is the operator of effective permittivity $\hat{\Upsilon}_d^{\text{eff}}$. Consider a d-dimensional isotropic medium whose permittivity is regarded as a space random function $\varepsilon(\boldsymbol{x})$ of the coordinate vector $\boldsymbol{x} = (x_1, \ldots, x_d)$. The local induction $\boldsymbol{D}(\boldsymbol{x})$ and electric field $\boldsymbol{E}(\boldsymbol{x})$ obey the linear equation

$$\boldsymbol{D}(\boldsymbol{x}) = \varepsilon(\boldsymbol{x})\boldsymbol{E}(\boldsymbol{x}).$$

The effective permittivity operator relates the mean induction $\langle \boldsymbol{D}(\boldsymbol{x})\rangle$ and the mean electric field intensity $\langle \boldsymbol{E}(\boldsymbol{x})\rangle$ by the operator $\hat{\Upsilon}_d^{\text{eff}}$

$$\langle \boldsymbol{D}(\boldsymbol{x})\rangle = \hat{\Upsilon}_d^{\text{eff}} \cdot \langle \boldsymbol{E}(\boldsymbol{x})\rangle = \int \mathrm{d}\boldsymbol{x}'\, \hat{\varepsilon}_d^{\text{eff}}(\boldsymbol{x}, \boldsymbol{x}') \cdot \langle \boldsymbol{E}(\boldsymbol{x}')\rangle .$$

Here the tensor $\hat{\varepsilon}_d^{\text{eff}}(\boldsymbol{x}, \boldsymbol{x}')$ is the kernel of the operator $\hat{\Upsilon}_d^{\text{eff}}$. The purpose of $\hat{\Upsilon}_d^{\text{eff}}$ is to replace a basically heterogeneous medium by an equivalent homogeneous medium for further discussing the behaviour of average fields [1–10].

In a statistically homogeneous infinite medium $\hat{\Upsilon}_d^{\text{eff}}$ is an integral operator with a difference kernel

$$\hat{\varepsilon}_d^{\text{eff}}(\boldsymbol{x}, \boldsymbol{x}') = \hat{\varepsilon}_d^{\text{eff}}\left(\frac{\boldsymbol{x} - \boldsymbol{x}'}{l}\right) \tag{1}$$

where l is the characteristic spatial scale of inhomogeneities. In $\boldsymbol{k} = (k_1, \ldots, k_d)$ space the relation between Fourier transforms $\langle \boldsymbol{D}(\boldsymbol{k})\rangle$ and $\langle \boldsymbol{E}(\boldsymbol{k})\rangle$ becomes

$$\langle \boldsymbol{D}(\boldsymbol{k})\rangle = \hat{\varepsilon}_d^{\text{eff}}(\boldsymbol{k}) \cdot \langle \boldsymbol{E}(\boldsymbol{k})\rangle .$$

For isotropic permittivity fields $\hat{\varepsilon}_d^{\text{eff}}(\boldsymbol{k})$ is given by an isotropic tensor determining a spatial dispersion associated with random inhomogeneities in the medium [11, 12].

0305-4470/95/030693+08$19.50

A number of works [1–9, 12] has been devoted to the investigation of small-scale asymptotics of the kernel (1)

$$\hat{\varepsilon}^{\text{eff}}(d) = \lim_{l\to 0} \hat{\varepsilon}_d^{\text{eff}}\left(\frac{x - x'}{l}\right).$$

For potential fields $E(x) = -\nabla\Psi(x)$ the problem reduces to calculating a scalar

$$\varepsilon^{\text{eff}}(d) = \lim_{k\to 0} \frac{k \cdot \hat{\varepsilon}_d^{\text{eff}}(k) \cdot k}{k^2}. \tag{2}$$

The effective permittivity $\varepsilon^{\text{eff}}(d)$ describes properties of the long-wavelength electrodynamics in d-dimensional heterogeneous dielectric. Mathematically equivalent problems have been encountered in many other branches of physics such as diffusion processes, heat transfer and transport in porous media. This also includes phenomena like transport problems in disordered media exhibiting critical behaviour, such as electron localization and hopping conduction in amorphous solids [13].

Although a huge body of literature is devoted to the problem of effective properties, there are only two exact closed formulas for the effective permittivity of continuous media. The first one, $\varepsilon^{\text{eff}} = (\langle\varepsilon^{-1}(x)\rangle)^{-1}$, is simply derived for a 1D medium. The second exact result, $\varepsilon^{\text{eff}} = \exp\langle\ln\varepsilon(x)\rangle$, was obtained for 2D isotropic medium using the theory of harmonic functions [14] and under more general assumptions by applying an elegant duality transformation [15, 16]. The latter result and its analog for discrete 2D media were used for calculating the percolation threshold, the fractal dimension of two-phase systems [10, 13] and the effective conductivity of a macroscopically isotropic 2D polycrystal [5, 9, 17]. Matheron [15] has shown that the geometric ponderation can be applied only to two-dimensional heterostructures possessing the specific property of symmetry and has listed several 2D counter examples [18].

In 3D heterostructures closed formulas for the effective permittivity similar to those for 1D and 2D are not known. Landau and Lifshitz [19] have mentioned that for isotropic media the expression $(\langle\varepsilon\rangle)^{2/3}(\langle\varepsilon^{-1}\rangle)^{-1/3}$ provides the first-order approximation of the effective permittivity for $d = 3$. Matheron [15] has discussed a similar conjecture for the effective permittivity in a space of any dimension

$$\varepsilon_{cj}^{\text{eff}}(d) = (\langle\varepsilon\rangle)^{(d-1)/d}(\langle\varepsilon^{-1}\rangle)^{-1/d}.$$

which for a log-normal isotropic medium has a simple analytical form:

$$\varepsilon_{cj}^{\text{eff}}(d) = \varepsilon_g \exp\left[\sigma^2\left(\frac{1}{2} - \frac{1}{d}\right)\right] \tag{3}$$

In (3) ε_g and σ^2 are the geometric average of the local permittivity and the variance of its logarithm, respectively. Note that the expression (3) implies independence of $\varepsilon^{\text{eff}}(d)$ on the shape of the permittivity correlation function.

The following properties of the conjecture (3) have caused considerable speculation [12, 20–22] as to its validity:

(i) the formula (3) leads to the exact results for d=1, 2 and $d = \infty$;

(ii) the conjecture provides an exact approximation of $\varepsilon^{\text{eff}}(d)$ up to σ^4-order for any dimension d [22];

(iii) numerical computations of $\varepsilon^{\text{eff}}(d)$ for $d = 3$ show good agreement with (3) up to $\sigma^2 = 7$ [20, 21].

The aforementioned properties of the conjecture (3) pose a fundamental question whether the effective permittivity of a d-dimensional isotropic medium can be expressed by a simple relationship such as (3), thus generalizing the 1D, 2D and $d = \infty$ exact results, or whether $\varepsilon^{\text{eff}}(d)$ is generally a functional of the heterogeneity spectrum.

The purpose of this paper is to develop a method for deriving $\varepsilon^{\text{eff}}(d)$ of log-normal d-dimensional media. The problem is attacked by the regularized Green function technique [23] for the Laplace operator. The effective permittivity is obtained as a functional series in an expansion over the log-permittivity variance. The result is applied further to derive the explicit σ^6-order approximation of $\varepsilon^{\text{eff}}(d)$. It is shown that the σ^6-order term of the 3D effective permittivity depends on the choice of the correlation function and, hence, cannot be expressed by a closed formula like (3).

The governing equation for the electrostatic problem

$$\nabla \cdot \varepsilon(\boldsymbol{x})\nabla\Psi(\boldsymbol{x}) = -4\pi\rho^{\text{ext}}(\boldsymbol{x}) \tag{4}$$

allows one to determine $\varepsilon^{\text{eff}}(d)$ in terms of the mean potential

$$\varepsilon^{\text{eff}}(d) = \lim_{k\to 0}\frac{4\pi\rho^{\text{ext}}(\boldsymbol{k})}{k^2\langle\Psi(\boldsymbol{k})\rangle} \tag{5}$$

where $\rho^{\text{ext}}(\boldsymbol{x})$ is the charge density of the external field sources. The dielectric permittivity takes the form

$$\varepsilon(\boldsymbol{x}) = \varepsilon_g \exp[\phi(\boldsymbol{x})]$$

where the fluctuation $\phi(\boldsymbol{x})$ is a Gaussian homogeneous isotropic random field with $\langle\phi(\boldsymbol{x})\rangle = 0$, $\langle\phi^2(\boldsymbol{x})\rangle = \sigma^2$ and a spectrum $\Phi(k)$.

Applying the technique [23] of regularization of the d-dimensional Green tensor, equation (4) can be reduced to the stochastic integral equation in $\boldsymbol{k}$ space

$$\begin{aligned} \boldsymbol{F}(\boldsymbol{k}) &= \boldsymbol{F}^{(0)}(\boldsymbol{k}) + \hat{g}(\boldsymbol{k})\cdot\int\frac{\mathrm{d}\boldsymbol{q}}{(2\pi)^d}\xi(\boldsymbol{k}-\boldsymbol{q})\boldsymbol{F}(\boldsymbol{q}) \\ \boldsymbol{F}^{(0)}(\boldsymbol{k}) &= \mathrm{i}\boldsymbol{k}\frac{4\pi\rho^{\text{ext}}(\boldsymbol{k})}{\varepsilon_g k^2}. \end{aligned} \tag{6}$$

In (6) the new variables $\boldsymbol{F}(\boldsymbol{x})$ and $\xi(\boldsymbol{x})$ are defined by

$$\boldsymbol{F}(\boldsymbol{x}) = \frac{1+\exp[\phi(\boldsymbol{x})]}{2}\nabla\Psi(\boldsymbol{x}) \qquad \xi(\boldsymbol{x}) = \tanh\frac{\phi(\boldsymbol{x})}{2} \tag{7}$$

and the isotropic, orthogonal tensor

$$\hat{g}(\boldsymbol{k}) = \hat{I} - 2\frac{\boldsymbol{k}\boldsymbol{k}}{k^2} \tag{8}$$

is the Fourier transform of the regurilarized Green tensor ($\hat{I}$ being the identity tensor). Reformulation of the stochastic problem (4) in terms of the functions $\boldsymbol{F}(\boldsymbol{x})$ and $\xi(\boldsymbol{x})$ constitutes a basis for further analytical consideration. A remarkable property of the renormalized random field $\xi(\boldsymbol{x})$ is that it is bounded ($|\xi(\boldsymbol{x})| \leqslant 1$) with zero $(2n+1)$-variate correlators ($n = 0, 1, \ldots$).

Solving (6) by successive iterations

$$\boldsymbol{F}(\boldsymbol{k}) = \sum_{n=0}^{\infty}\boldsymbol{F}^{(n)}(\boldsymbol{k}) \qquad \boldsymbol{F}^{(n+1)}(\boldsymbol{k}) = \hat{g}(\boldsymbol{k})\cdot\int\frac{\mathrm{d}\boldsymbol{q}}{(2\pi)^d}\xi(\boldsymbol{k}-\boldsymbol{q})\boldsymbol{F}^{(n)}(\boldsymbol{q}) \qquad n = 0, 1, \ldots$$

and averaging the multiple scatter series for $\boldsymbol{F}(\boldsymbol{k})$ over ensemble of ξ-heterogeneities yield

$$\langle\boldsymbol{F}(\boldsymbol{k})\rangle = \hat{\Lambda}(\boldsymbol{k})\cdot\boldsymbol{F}^{(0)}(\boldsymbol{k}) \qquad \hat{\Lambda}(\boldsymbol{k}) = \hat{I} + \hat{g}(\boldsymbol{k})\cdot\hat{\lambda}(\boldsymbol{k}). \tag{9}$$

Here $\hat{\Lambda}(\boldsymbol{k})$ is the kernel of the mass operator of the Dyson equation for the mean field $\langle \boldsymbol{F}(\boldsymbol{k})\rangle$, $\hat{\lambda}(\boldsymbol{k}) = \sum_{n=1}^{\infty} \hat{\lambda}^{(n)}$ and

$$\hat{\lambda}^{(n)}(\boldsymbol{k}) = \int \frac{\mathrm{d}q_1}{(2\pi)^D} \cdots \frac{\mathrm{d}q_n}{(2\pi)^D} \hat{g}(q_1) \cdots \hat{g}(q_n) \Phi_\xi^{(n+1)}(\boldsymbol{k} - q_1, \ldots, q_{n-1} - q_n)$$
$$n = 1, 2, \ldots . \tag{10}$$

In (10) $\Phi_\xi^{(2n+1)} = 0$ and the multivariate $(2n+2)$-correlators of the homogeneous random field $\xi(\boldsymbol{k})$ are determined by

$$\langle \xi(\boldsymbol{k}_1) \ldots \xi(\boldsymbol{k}_{2n+2})\rangle = (2\pi)^d \delta(\boldsymbol{k}_1 + \cdots + \boldsymbol{k}_{2n+2}) \Phi_\xi^{(2n+2)}(\boldsymbol{k}_1, \ldots, \boldsymbol{k}_{2n+1}) \qquad n = 0, 1, \ldots$$

where $\Phi_\xi^{(2n+2)}(\boldsymbol{k}_1, \ldots, \boldsymbol{k}_{2n+1})$ is the spectrum of the $(2n+2)$-point correlation function of $\xi(\boldsymbol{x})$.

The effective permittivity results from (5), (7) and (9) as follows:

$$\varepsilon^{\mathrm{eff}}(d) = \frac{\varepsilon_g}{1 - 2\lambda(d)} \qquad \lambda(d) = \lim_{k \to 0} \frac{\boldsymbol{k} \cdot \hat{\lambda}(\boldsymbol{k}) \cdot \boldsymbol{k}}{k^2}. \tag{11}$$

Expanding even-order spectrum functions of the field $\xi(\boldsymbol{x})$ as a series of the variance of the Gaussian random function $\phi(\boldsymbol{x})$

$$\Phi_\xi^{(2n+2)}(\boldsymbol{k}_1, \ldots, \boldsymbol{k}_{2n+1}) = \sum_{m=n+1}^{\infty} \sigma^{2m} \Phi_m^{(2n+2)}(\boldsymbol{k}_1, \ldots, \boldsymbol{k}_{2n+1}) \qquad n = 0, 1, \ldots \tag{12}$$

and substituting (12) into (10) yield the following expression for the tensor $\hat{\lambda}(\boldsymbol{k})$:

$$\hat{\lambda}(\boldsymbol{k}) = \sum_{n=0}^{\infty} \sum_{m=n+1}^{\infty} \sigma^{2m} \hat{\lambda}_m^{(2n+1)}(\boldsymbol{k})$$

where

$$\hat{\lambda}_m^{(2n+1)}(\boldsymbol{k}) = \int \frac{\mathrm{d}\boldsymbol{k}_1}{(2\pi)^d} \cdots \frac{\mathrm{d}\boldsymbol{k}_{2n+1}}{(2\pi)^d} \Phi_m^{(2n+2)}(\boldsymbol{k}_1, \ldots, \boldsymbol{k}_{2n+1})$$
$$\times \hat{g}(\boldsymbol{k} + \boldsymbol{k}_1) \cdots \hat{g}(\boldsymbol{k} + \boldsymbol{k}_1 + \cdots + \boldsymbol{k}_{2n+1}). \tag{13}$$

The effective permittivity (11) is obtained as a functional series in terms of the moments of the random field $\phi(\boldsymbol{x})$

$$\lambda(d) = \sum_{m=1}^{\infty} \sigma^{2m} \lambda^{(m)}(d) \tag{14a}$$

$$\lambda^{(m)}(d) = \lim_{k \to 0} \sum_{n=0}^{m-1} \frac{\boldsymbol{k} \cdot \hat{\lambda}_m^{(2n+1)}(\boldsymbol{k}) \cdot \boldsymbol{k}}{k^2}. \tag{14b}$$

In the remainder of the paper the three first terms of the series (14) are derived and are compared in parallel with the conjecture (3). The latter can be expanded in σ^2-series using the same representation (11)

$$\lambda_{cj}(d) = \sum_{m=1}^{\infty} \sigma^{2m} \lambda_{cj}^{(m)}(d) \tag{15a}$$

$$\lambda_{cj}^{(m)}(d) = \frac{(-1)^{m+1}}{2m!} \left(\frac{1}{2} - \frac{1}{d}\right)^m . \tag{15b}$$

σ^2-order term (m = 1). This is obtained from (13) and (14)

$$\lambda^{(1)}(d) = \lim_{k\to 0} \frac{\boldsymbol{k}\cdot\hat{\lambda}_1^{(1)}(\boldsymbol{k})\cdot\boldsymbol{k}}{k^2} \tag{16}$$

$$\lim_{k\to 0} \hat{\lambda}^{(1)}(\boldsymbol{k}) = \int \frac{d\boldsymbol{k}_1}{(2\pi)^d} \hat{g}(\boldsymbol{k}_1)\Phi_1^{(2)}(\boldsymbol{k}_1). \tag{17}$$

With the use of the relationship $\Phi_1^{(2)}(\boldsymbol{k}_1) = \Phi(k_1)/4$ following from (12), the integral in (17) is easily calculated:

$$\int \frac{\mathrm{d}\boldsymbol{k}_1}{(2\pi)^d} \hat{g}(k_1)\Phi(k_1) = \hat{I}\ \frac{\mathrm{Tr}\,\hat{g}}{d} \int \frac{\mathrm{d}\boldsymbol{k}_1}{(2\pi)^d} \Phi(k_1) = \hat{I}\ \frac{d-2}{d}. \tag{18}$$

Substituting (18) into (16) leads to

$$\lambda^{(1)}(d) = \lambda_{cj}^{(1)}(d) = \frac{1}{2}\left(\frac{1}{2} - \frac{1}{d}\right). \tag{19}$$

σ^4-order term (m = 2). This is given by

$$\lambda^{(2)}(d) = \lim_{k\to 0} \frac{\boldsymbol{k}\cdot[\hat{\lambda}_2^{(1)}(\boldsymbol{k}) + \hat{\lambda}_2^{(3)}(\boldsymbol{k})]\cdot\boldsymbol{k}}{k^2}. \tag{20}$$

It follows from (12) that

$$\Phi_2^{(2)}(\boldsymbol{k}_1) = -\frac{1}{2^3}\Phi(k_1) \tag{21a}$$

and

$$\Phi_2^{(4)}(\boldsymbol{k}_1, \boldsymbol{k}_2, \boldsymbol{k}_3) = \frac{(2\pi)^d}{2^4}[\delta(\boldsymbol{k}_1 + \boldsymbol{k}_2)\Phi(k_1)\Phi(k_3) + \delta(\boldsymbol{k}_1 + \boldsymbol{k}_3)\Phi(k_1)\Phi(k_2)$$
$$+\delta(\boldsymbol{k}_2 + \boldsymbol{k}_3)\Phi(k_1)\Phi(k_2)]. \tag{21b}$$

Substituting (21) into (13) yields

$$\lim_{k\to 0} \hat{\lambda}_2^{(1)}(\boldsymbol{k}) = -\hat{I}\ \frac{d-2}{2^3 d}$$
$$\lim_{k\to 0} \hat{\lambda}_2^{(3)}(\boldsymbol{k}) = \frac{1}{2^4} \int \frac{\mathrm{d}\boldsymbol{k}_1}{(2\pi)^d} \frac{\mathrm{d}\boldsymbol{k}_2}{(2\pi)^d} \Phi(k_1)\Phi(k_2) \tag{22}$$
$$\times \left\{\hat{g}(\boldsymbol{k}_1)\cdot\hat{g}(\boldsymbol{k}_2)\cdot[\hat{g}(\boldsymbol{k}) + \hat{g}(\boldsymbol{k}_1 + \boldsymbol{k}_2)] + \hat{g}(\boldsymbol{k}_1 + \boldsymbol{k}_2)\right\}.$$

By employing (8) and the obvious relationship $\mathrm{Tr}\,\hat{g}(\boldsymbol{k}_1)\cdot\hat{g}(\boldsymbol{k}_1 + \boldsymbol{k}_2)\cdot\hat{g}(\boldsymbol{k}_2) = d - 2$ the last integral is straightforwardly calculated:

$$\lim_{k\to 0} \hat{\lambda}_2^{(3)}(\boldsymbol{k}) = \frac{d-2}{2^3 d}\left[\hat{I} + \frac{d-2}{2d}\hat{g}(\boldsymbol{k})\right]. \tag{23}$$

The expressions (20), (22) and (23) recover the result [22]

$$\lambda^{(2)}(d) = \lambda_{cj}^{(2)}(d) = -\frac{1}{2\ 2!}\left(\frac{1}{2} - \frac{1}{d}\right)^2. \tag{24}$$

σ^6-order term (m = 3). This is obtained from (14) as follows:

$$\lambda^{(3)}(d) = \lim_{k\to 0} \frac{k \cdot [\hat{\lambda}_3^{(1)}(k) + \hat{\lambda}_3^{(3)}(k) + \hat{\lambda}_3^{(5)}(k)] \cdot k}{k^2}.$$

Explicit expressions of functions $\Phi_3^{(2)}(k_1)$, $\Phi_3^{(4)}(k_1, k_2, k_3)$ and $\Phi_3^{(6)}(k_1, k_2, k_3, k_4, k_5)$ result from (12) in the form

$$\Phi_3^{(2)}(k_1) = \frac{1}{2^6}\left[\Phi(k_1) + \tfrac{1}{3}\int \frac{dq_1}{(2\pi)^D}\frac{dq_2}{(2\pi)^D}\Phi(k_1 - q_1)\Phi(q_1 - q_2)\Phi(q_2)\right] \tag{25a}$$

$$\begin{aligned}\Phi_3^{(4)}(k_1, k_2, k_3) = -\frac{1}{2^5}\{&2(2\pi)^D[\delta(k_1 + k_2)\Phi(k_1)\Phi(k_3) + \delta(k_1\\ &+k_3)\Phi(k_1)\Phi(k_2) + \delta(k_2 + k_3)\Phi(k_1)\Phi(k_2)] + \Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\Phi(k_1)\Phi(k_2)\Phi(-k_1 - k_2 - k_3) + \Phi(k_1)\Phi(k_2)\Phi(-k_1 - k_2 - k_3)\\ &+\Phi(k_1)\Phi(k_3)\Phi(-k_1 - k_2 - k_3)\}.\end{aligned} \tag{25b}$$

$$\begin{aligned}\Phi_3^{(6)}(k_1, k_2, k_3, k_4, k_5) = \frac{(2\pi)^{2D}}{2^6}[&\delta(k_1 + k_2)\delta(k_3 + k_4)\Phi(k_1)\Phi(k_3)\Phi(k_5)\\ &+\delta(k_1 + k_2)\delta(k_3 + k_5)\Phi(k_1)\Phi(k_3)\Phi(k_4)\\ &+\delta(k_1 + k_2)\delta(k_4 + k_5)\Phi(k_1)\Phi(k_3)\Phi(k_4)\\ &+\delta(k_1 + k_3)\delta(k_2 + k_4)\Phi(k_1)\Phi(k_2)\Phi(k_5)\\ &+\delta(k_1 + k_3)\delta(k_2 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_4)\\ &+\delta(k_1 + k_3)\delta(k_4 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_4)\\ &+\delta(k_1 + k_4)\delta(k_2 + k_3)\Phi(k_1)\Phi(k_2)\Phi(k_5)\\ &+\delta(k_1 + k_4)\delta(k_2 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\delta(k_1 + k_4)\delta(k_3 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\delta(k_1 + k_5)\delta(k_2 + k_3)\Phi(k_1)\Phi(k_2)\Phi(k_4)\\ &+\delta(k_1 + k_5)\delta(k_2 + k_4)\Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\delta(k_1 + k_5)\delta(k_3 + k_4)\Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\delta(k_2 + k_3)\delta(k_4 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_4)\\ &+\delta(k_2 + k_4)\delta(k_3 + k_5)\Phi(k_1)\Phi(k_2)\Phi(k_3)\\ &+\delta(k_2 + k_5)\delta(k_3 + k_4)\Phi(k_1)\Phi(k_2)\Phi(k_3)].\end{aligned} \tag{25c}$$

Using (25) one obtains after cumbersome transformations

$$\lambda^{(3)}(d) = \lambda_{cj}^{(3)}(d) + \frac{1}{2d}\left[\beta(d) + \frac{1}{6d^2}\right]. \tag{26}$$

where

$$\beta(d) = \int \frac{dk_1}{(2\pi)^d}\frac{dk_2}{(2\pi)^d}\frac{dk_3}{(2\pi)^d}\,\theta_d(k_1, k_2, k_3)\Phi(k_1)\Phi(k_2)\Phi(k_3) \tag{27}$$

and

$$\theta_d(\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3) = \frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_2\,(k_1^2+\boldsymbol{k}_1\cdot\boldsymbol{k}_2)}{k_1^2(\boldsymbol{k}_1+\boldsymbol{k}_2)^2}\left[\frac{1}{d} - \frac{\boldsymbol{k}_2\cdot\boldsymbol{k}_3\,(k_3^2+\boldsymbol{k}_1\cdot\boldsymbol{k}_3+\boldsymbol{k}_2\cdot\boldsymbol{k}_3)}{k_2^2(\boldsymbol{k}_1+\boldsymbol{k}_2+\boldsymbol{k}_3)^2}\right]$$

$$+\frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_2\,\boldsymbol{k}_2\cdot\boldsymbol{k}_3}{k_1^2(\boldsymbol{k}_1+\boldsymbol{k}_2)^2(\boldsymbol{k}_2+\boldsymbol{k}_3)^2}\left[\left(\boldsymbol{k}_1\cdot\boldsymbol{k}_3 - \frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_2\,\boldsymbol{k}_2\cdot\boldsymbol{k}_3}{k_2^2}\right)\left(\frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_3}{k_3^2}-1\right)\right.$$

$$-\left(\frac{k_1^2}{k_2^2}+2\frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_2}{k_2^2}+\frac{\boldsymbol{k}_1\cdot\boldsymbol{k}_3}{k_3^2}\right)$$

$$\left.\times\frac{(k_1^2+k_2^2+2\boldsymbol{k}_1\cdot\boldsymbol{k}_2+\boldsymbol{k}_1\cdot\boldsymbol{k}_3+\boldsymbol{k}_2\cdot\boldsymbol{k}_3)(k_2^2+k_3^2+2\boldsymbol{k}_2\cdot\boldsymbol{k}_3+\boldsymbol{k}_1\cdot\boldsymbol{k}_2+\boldsymbol{k}_1\cdot\boldsymbol{k}_3)}{(\boldsymbol{k}_1+\boldsymbol{k}_2+\boldsymbol{k}_3)^2}\right].$$

Switching to spherical variables in the integral (27) yields

$$\beta(d) = \Omega_d^3\int_0^\infty \frac{\mathrm{d}k_1}{(2\pi)^d}\frac{\mathrm{d}k_2}{(2\pi)^d}\frac{\mathrm{d}k_3}{(2\pi)^d}\vartheta_d(k_1,k_2,k_3)k_1^{d-1}\Phi(k_1)k_2^{d-1}\Phi(k_2)k_3^{d-1}\Phi(k_3) \qquad (28)$$

where $\Omega_d = 2\pi^{d/2}/\Gamma(d/2)$ and

$$\vartheta_d(k_1,k_2,k_3) = \int\frac{\mathrm{d}^3\Omega(\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3)}{\Omega_d^3}\theta_d(\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3) \qquad (29)$$

The expressions (26)–(28) determine the third-order correction of the effective permittivity (11), (14). However, unlike the first- and second-order corrections (19), (24) the third-order term depends on the spectrum $\Phi(k)$ of the log-permittivity $\phi(\boldsymbol{x})$. Indeed, if the σ^6-order term is invariant with respect to the spectrum then $\beta(d)$ (28) has to be constant for any Φ. In particular, it follows from (26) that the conjecture (3) is valid only if the condition

$$\beta(d) = -1/6d^2 \qquad (30)$$

is fulfilled for any functions $\Phi(k)$.

Employing a standard variation procedure the necessary and sufficient condition for the functional $\beta(d)$ be invariant with respect to the functions $\Phi(k)$ and satisfy (30) is

$$\vartheta_d^{(sm)}(k_1,k_2,k_3) = \text{constant} = -\frac{1}{d^2} \qquad (31)$$

where

$$\vartheta_d^{(sm)}(k_1,k_2,k_3) = \sum_{\substack{l,p,t=1\\ l\neq p\neq t}}^{3}\vartheta_d(k_l,k_p,k_t)$$

where the function $\vartheta_d(k_1,k_2,k_3)$ is symmetrized over all arguments.

Calculations of the integral (29) for $d=1$ and $d=2$ show agreement with the conjecture (31) and reproduce 1D and 2D exact results

$$\vartheta_1^{(sm)} = -1 \qquad \vartheta_2^{(sm)}(k_1,k_2,k_3) = -\tfrac{1}{4}.$$

In contrast, $\vartheta_3^{(sm)}(k_1,k_2,k_3) \neq$ constant. Indeed

$$\lim_{k_l\to 0}\vartheta_3^{(sm)}(k_1,k_2,k_3) = \lim_{k_l\to\infty}\vartheta_3^{(sm)}(k_1,k_2,k_3) = -\tfrac{1}{9} \qquad l=1,2,3 \qquad (32a)$$

and

$$\vartheta_3^{(sm)}(k,k,k) = \tfrac{3}{16}\,Li_2(1/3)+\tfrac{1}{64}\pi^2+\tfrac{3}{16}\ln 3\left(\tfrac{87}{70}+\tfrac{1}{2}\ln 3-\ln 2\right)-\tfrac{527}{840}\approx -0.18 \qquad (32b)$$

where

$$Li_p(z) = \frac{z}{\Gamma(p)} \int_0^\infty \mathrm{d}t \, \frac{t^{p-1}}{\mathrm{e}^t - z} \qquad |z| < 1 \quad \mathrm{Re}\, p > 0.$$

is the polylogarithm of order p [24]. The results (32) show that $\beta(d)$ is a functional of the correlation function and therefore the 3D conjecture (3) deviates from the correct value (26) in σ^6-order term.

In conclusion, the effective permittivity of d-dimensional media is derived as a functional series in terms of the log-permittivity moments. The explicit expression of the σ^6-order approximation of $\varepsilon^{\mathrm{eff}}$ is obtained as a functional of the heterogeneity spectrum. It is shown that the third-order term in the expansion of the effective permittivity is invariant with respect to the spectrum for $d = 1$ and $d = 2$ only and becomes a functional of the heterogeneity spectrum in 3D. This proves that the effective permittivity of a 3D isotropic heterogeneous medium is a functional of the heterogeneities spectrum and is not expressed in simple manner as in (3).

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