Renormalization-group approach to the calculation of the effective dielectric constant of a two-dimensional inhomogeneous system

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A renormalization-group procedure is proposed for calculating the effective dielectric constant of a two-dimensional inhomogeneous system (a square lattice whose nodes are occupied in a random way by particles). This new procedure is based on the circumstance that in each step of a renormalization-group transformation the electric fields produced by the old and new configurations of particles far from the surface of the lattice are approximately the same. Significantly, this approach incorporates the dipole-dipole interaction between particles. An asymptotic expression is derived for the dielectric constant. This expression has a singular term proportional to $|p - p_c|^{\nu}$.

1. INTRODUCTION

There is considerable interest in the problem of calculating the effective dielectric constant of composite materials.[1,2] A rapid development of research in this field has been stimulated not only by practical needs but also by the discovery of new physical phenomena in these materials.

Basically two approaches are presently being taken in the literature to describe the optical properties of composites. The first starts from the use of one of various versions of the effective-medium approximation.[3,4] This approach suffers from all the shortcomings of the mean-field method,[5] which become particularly serious for two-dimensional composite materials near percolation thresholds for one of the components. The second approach starts from a modeling of the composite medium by a lattice whose nodes (or links) are occupied in a random fashion by resistances and capacitances.[6-8]

The effective dielectric constant of such a medium is calculated on the basis of scaling representations or the transfer-matrix method.[6-8] That approach must apparently be abandoned in the optical frequency range, however, where the electromagnetic interactions between individual elements of the composite medium become important.

Berthier and Driss-Khodja[9] have recently attempted to take a renormalization-group approach to calculate the effective dielectric constant in the case with dipole-dipole interactions. However, the approximation of an effective medium was used in that paper in each step of the renormalization-group procedure. The ranges of applicability of the renormalization-group method and of the effective-medium method are different. The first is used near a percolation threshold, and the second far from it. The approach taken in Ref. 9 is thus internally contradictory.

In the present paper we propose another version of the renormalization-group procedure, in which the electromagnetic interactions between different structural elements are taken into account, and no use is made of the effective-medium approximation. This approach leads to the asymptotic behavior of the dielectric constant near the percolation threshold of a two-dimensional inhomogeneous system.

2. DESCRIPTION OF THE RENORMALIZATION-GROUP PROCEDURE

We consider a two-dimensional square lattice whose nodes are occupied in a random way with a probability p by spherical particles with a polarizability $\alpha(\omega)$. The electromagnetic field acting on each particle is the sum of the external field $\mathbf{E}_0$ and the induced field created by all the other particles. In calculating the effective dielectric constant of this inhomogeneous system, we restrict the discussion to the dipole-dipole interactions between particles. We use the following renormalization-group procedure.

1. The initial lattice is broken up into nonintersecting squares with a size equal to the lattice constant a (Fig. 1).

2. For each possible configuration of the particles on a selected quartet of nodes, an electrodynamic problem is solved. In other words, the field produced by the given configuration of particles in the external field $\mathbf{E}_0$ is calculated.

3. All possible configurations are put in one of two groups (Fig. 2). The first group contains those configurations in which there are occupied nodes along at least one diagonal of the square. The second group contains all other configurations. The probabilities for the realization of the first and second groups of configurations are p_1 and $1 - p_1$, respectively. In accordance with the particular way in which we have classified the configurations into groups, we have

$$p_1 = R(p) = p^2(2 - p^2). \tag{1}$$

Transformation R is the same as the transformation which was proposed in Ref. 10 in a renormalization-group study of the conductivity in a two-dimensional system near the percolation threshold p_c. The immobile unstable point of this transformation coincides fairly well with the percolation threshold on a square lattice. It was for this reason that we selected the method outlined above for classifying the configurations into groups.

For each of these groups we calculate the mean field over the configurations at a distance much greater than the size of our square. The first and second groups are replaced by quartets of identical particles which do not interact with

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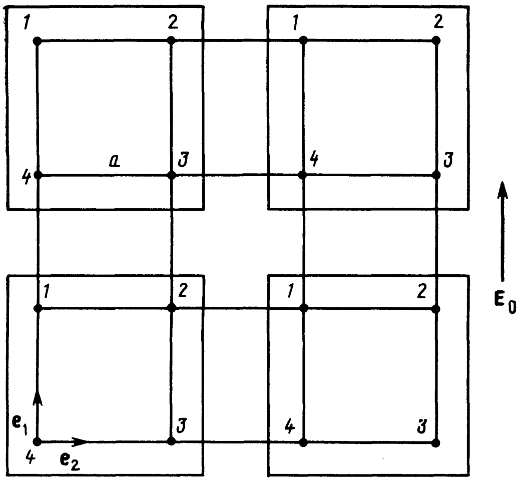

FIG. 1. First step of the renormalization-group procedure: the breakup of the lattice into nonintersecting squares. The arrow shows the direction of the polarization of the external field; $\mathbf{e}_1$ and $\mathbf{e}_2$ are unit vectors.

each other and which have polarizabilities $\alpha_1(\omega, p)$ and $\beta_1(\omega, p)$, respectively. These particles lie at the nodes of the original lattice. The new polarizabilities are found from the condition that the average field produced by the original configurations of the first and second groups far from the square under consideration is equal to the field produced by the quartet of noninteracting particles with the respective polarizabilities $\alpha_1(\omega, p)$ and $\beta_1(\omega, p)$.

4. The quartets of noninteracting particles which are found are treated as the unit elements of a new square lattice. The effective distance between these elements is $2a$. Consequently, the structure of the original lattice is restored after the first step of the renormalization-group procedure, but

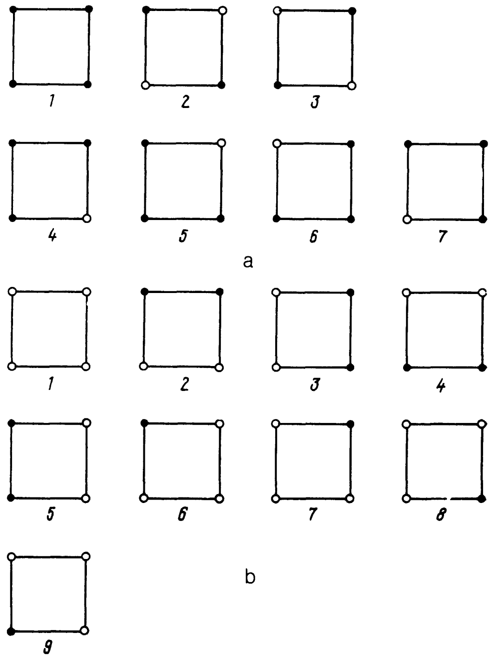

FIG. 2. Groups of configurations. a—First group; b—second group. ●) Occupied nodes; ○) vacant nodes.

the structure of an individual element has now become more complex. We wish to stress that this approach is based on the circumstance that the field produced by the four elements of the original structure and averaged over configurations is approximately equal to the field produced by a single element of the new structure.

5. This procedure is repeated until the distance between individual structural elements becomes comparable to the correlation length $\xi(p)$ in the inhomogeneous system. Near the percolation threshold, $\xi(p)$ is the sole characteristic length of the problem. The probability for the formation of clusters with a size greater than $\xi(p)$ is exponentially small,[11] so the discussion can be restricted to elements with dimensions smaller than or equal to $\xi(p)$.

After the nth step, an individual element consists of 4^n identical particles which do not interact with each other and which have polarizabilities $\alpha_n(\omega, p_{n-1})$ or $\beta_n(\omega, p_{n-1})$. The probabilities for the realizations of configurations with polarizabilities α_n and β_n are $p_n = R^n(p)$ and $1 - p_n$, respectively. The effective distance between the elements of such a structure is $2^n a$. After going through this procedure, we obtain a system of noninteracting particles at the nodes of a two-dimensional lattice whose effective dielectric constant can be described by

$$\varepsilon(\omega, p) = 1 + \frac{4\pi}{a^2 d} A_n(\omega, p), \qquad (2)$$

where

$$A_n(\omega, p) = p_n \alpha_n(\omega, p) + (1 - p_n)\beta_n(\omega, p),$$

and d is the effective thickness of the layer, equal in order of magnitude to the size of the particles.

We will now use this method to describe the properties of a disordered system near the percolation threshold. In accordance with the discussion above, we assume that the limiting value of n in expression (2) is

$$2^n a = \xi(p). \qquad (3)$$

We denote the value of A_n corresponding to this n by A.

3. CALCULATION OF THE DIELECTRIC CONSTANT

Let us use the procedure described above to calculate the response of this two-dimensional system to an external field. For definiteness we consider the case in which the external field is directed parallel to the plane of the lattice, along one of its links. For simplicity we restrict the calculations to the dipole-dipole interaction between nearest neighbors. In each step of the renormalization-group procedure in the calculation of the interaction between the four structural elements corresponding to this step, we model the field produced by an individual element as the field of a point dipole with a polarizability $4^{n-1}\alpha_{n-1}(\omega, p_{n-2})$ or $4^{n-1}\beta_{n-1}(\omega, p_{n-2})$, positioned at the center of the corresponding element (square). Under these assumptions, the field $\mathbf{E}_i$, $i = 1, 2, 3, 4$, acting on each of the four elements (Fig. 1) is found from the equations

$$\mathbf{E}_1 = \mathbf{E}_0 - \frac{1}{2^{n-1}a^3}[\chi_4\mathbf{E}_4 + \chi_2\mathbf{E}_2 - 3(\mathbf{e}_1\mathbf{E}_4)\chi_4\mathbf{e}_1 - 3(\mathbf{e}_2\mathbf{E}_2)\chi_2\mathbf{e}_2],$$

$$\mathbf{E}_2 = \mathbf{E}_0 - \frac{1}{2^{n-1}a^3}[\chi_1\mathbf{E}_1 + \chi_3\mathbf{E}_3 - 3(\mathbf{e}_2\mathbf{E}_1)\chi_1\mathbf{e}_2 - 3(\mathbf{e}_1\mathbf{E}_3)\chi_3\mathbf{e}_1],$$

$$E_3=E_0-\frac{1}{2^{n-1}a^3}[\chi_2E_2+\chi_4E_4-3(e_1E_2)\chi_2e_1-3(e_2E_4)\chi_4e_2],$$

$$E_4=E_0-\frac{1}{2^{n-1}a^3}[\chi_1E_1+\chi_3E_3-3(e_1E_1)\chi_1e_1-3(e_2E_3)\chi_3e_2]. \quad (4)$$

Here χ_i is the polarizability of element i, which is either $\alpha_{n-1}(\omega, p_{n-2})$ or $\beta_{n-1}(\omega, p_{n-2})$, depending on the particular configuration under consideration, $\mathbf{e}_1$ and $\mathbf{e}_2$ are unit vectors directed along the links of the square lattice, and $\mathbf{E}_0$ is the external electromagnetic field.

We wish to stress that the replacement of the 4^{n-1} dipoles in a square of side $2^{n-1}a$ by a single dipole—a replacement which was used in writing Eqs. (4)—can be justified approximately only if the wavelength of the electromagnetic field, λ, is much greater than $2^{n-1}a$. Accordingly, this calculation method can be used only under the condition that the correlation length $\xi(p)$ is shorter than the wavelength of the light:

$$\xi(p)<\lambda. \quad (5)$$

In accordance with the renormalization-group procedure described in Sec. 2, we find the following expressions for $\alpha_n(p_{n-1})$ and $\beta_n(p_{n-1})$ from Eqs. (4):

$$\alpha_n=\alpha_{n-1}\left(1+\frac{\alpha_{n-1}}{2^{n-1}a^3}\right)-(\alpha_{n-1}-\beta_{n-1})\left(1+\frac{2\alpha_{n-1}}{2^{n-1}a^3}\right)\frac{(1-p_{n-1})}{(2-p_{n-1}^2)},$$

$$\beta_n=\beta_{n-1}\left(1+\frac{\beta_{n-1}}{2^{n-1}a^3}\right)+(\alpha_{n-1}-\beta_{n-1})\left(1+\frac{2\beta_{n-1}}{2^{n-1}a^3}\right)\frac{p_{n-1}}{(1+p_{n-1})}. \quad (6)$$

We used an additional approximation in writing (6): We assumed $\alpha(\omega)/a^3<1$. Consequently, only the terms of first order in $\alpha_{n-1}/(2^{n-1}a^3)$ were retained in each step. This approximation is justified only if the frequency of the incident light is far from the resonant frequencies of resonant electromagnetic modes localized near individual particles. The optical properties of a random two-dimensional medium near resonant frequencies of local modes are discussed in Ref. 12.

From (6) we find the following expression, which is valid at $n\geqslant 1$:

$$\alpha_n-\beta_n=\alpha(\omega)\prod_{k=0}^{n-1}\frac{1}{(2-p_k^2)(1+p_k)}\left[1+O\left(\frac{\alpha}{a^3}\right)\right], \quad (7)$$

where $p_0\equiv p$. Expression (7) shows that for any p the quantity $\alpha_n-\beta_n$ approaches zero with increasing n. More convenient than calculating α_n and β_n separately is to directly calculate the quantity A_n which appears in expression (2) for the effective dielectric constant. Instead of the two equations in (6) we find the relation

$$A_n=A_{n-1}+\frac{1}{2^{n-1}a^3}[A_{n-1}^2-(\alpha_{n-1}-\beta_{n-1})^2p_{n-1}^2(1-p_{n-1})^2]. \quad (8)$$

Recurrence relation (8) can be solved approximately. For this purpose we express A_n in (8) in terms of A_{n-2}. We find

$$A_n=A_{n-2}+A_{n-2}^2\left(\frac{1}{2^{n-1}a^3}+\frac{1}{2^{n-2}a^3}\right)-\sum_{i=1}^{2}\frac{1}{2^{n-i}a^3}(\alpha_{n-i}-\beta_{n-i})^2p_{n-i}^2(1-p_{n-i})^2$$
$$+\frac{1}{2^{n-1}a^3}\left\{\frac{2A_{n-2}}{2^{n-2}a^3}[A_{n-2}^2-(\alpha_{n-2}-\beta_{n-2})^2p_{n-2}^2(1-p_{n-2})^2]+\frac{1}{2^{2(n-2)}a^6}[A_{n-2}^2-(\alpha_{n-2}-\beta_{n-2})^2p_{n-2}^2(1-p_{n-2})^2]^2\right\}.$$

The expression in the curly brackets is small in comparison with the other terms, since it contains an additional small factor on the order of $(\alpha_{n-2}/2^{n-2}a^3)^2$. As above, we discard such terms. Repeating this procedure, we find the following expression for A_n:

$$A_n=\alpha p+(\alpha p)^2\frac{2}{a^3}\left(1-\frac{1}{2^n}\right)-\frac{1}{a^3}\sum_{i=0}^{n-1}\frac{1}{2^i}p_i^2(1-p_i)^2(\alpha_i-\beta_i)^2$$
$$\approx\alpha p\left\{1+\frac{2}{a^3}\alpha p\left[1-\frac{1}{2}(1-p)^2\right]\right\}-\frac{2}{a^3}(\alpha p)^2\frac{1}{2^n}. \quad (9)$$

The approximate equality in (9) was found by retaining only the first term in the sum over i. This simplification is justified by the circumstance that the quantity $(\alpha_i-\beta_i)^2$ falls off rapidly with increasing i [roughly as p_c^{4i}; see (7)].

Near the percolation threshold we have

$$\xi(p)=l_\pm a|p-p_c|^{-\nu}, \quad (10)$$

where $\nu=1.35$ is the critical exponent of the correlation length in the corresponding percolation problem,[13] and $l_\pm$ is a quantity on the order of unity (l_+ and l_- correspond to values $p>p_c$ and $p<p_c$). In the region in which condition (5) holds, we find the following expression for A from (3), (9), and (10):

$$A=\alpha p_c\left(1+\frac{2\alpha}{a^3}p_c\right)+\alpha(p-p_c)\left(1+\frac{4\alpha}{a^3}p_c\right)-\frac{2(\alpha p_c)^2}{a^3}l_\pm^{-1}|p-p_c|^\nu. \quad (11)$$

The expression for the response to a field directed perpendicular to the plane of the lattice is similar in structure. The asymptotic behavior of A and, correspondingly, of the dielectric constant near the percolation threshold, (11), is related in an important way to the large-scale cluster structure of this system. The nontrivial dependence of ε on p in (11) arises only when the interaction between particles is taken into account. The asymptotic behavior found for the effective dielectric constant differs markedly from the corresponding results found in models based on the use of Kirchhoff's equations.[7] In the first place, expression (11) differs from Ref. 7 in that it contains regular terms, which are linear in $p-p_c$ and which may dominate $\varepsilon(p)$. Second, the singular term in the $\varepsilon(p)$ dependence is proportional to $|p-p_c|^\nu$, while the conductivity $\sigma(p)$, in terms of which $\varepsilon(p)$ is expressed, is given by the following expressions in a two-dimensional system consisting of a poor conductor and a good conductor:[7]

$$\sigma(p) \propto \begin{cases} (p_c-p)^{-\nu}, & p<p_c, \\ (p-p_c)^{\nu}, & p>p_c. \end{cases} \quad (12)$$

The difference between the singular behavior in expression (11) and that in (12) at $p<p_c$ stems from the following circumstance. In the conductivity problem, the power-law increase in σ at $p<p_c$ occurs[7] under the condition that the conductivity of the system at the percolation threshold is much higher than the conductivity of the dielectric phase. In the system which we are considering in the present paper, the conductivity is zero for any p, since we are assuming that the particles which occupy the lattice nodes are not in contact with each other. A model of this sort makes it possible to determine the role played by the electromagnetic interaction between particles in the appearance of a singularity in the p dependence of ε. In the absence of an interaction, there the optical response will obviously contain no manifestation of the "geometric" phase transition in the system.

The singular behavior of expression (11) is the same as that of (12) at $p>p_c$. In Ref. 9, the exponent describing the behavior of the dielectric constant at $p>p_c$ was found to be 1.52. That value is higher than ν and does not agree with experimental data.

Analyzing the propagation of a light wave near the percolation threshold in a heterophase solid, Korzhenevskiĭ and Luzhkov[14] found an asymptotic behavior similar to the singular term in (11) for the effective refractive index n in the "short-wavelength" region:

$$n^2 \propto |p-p_c|^{2\alpha\nu}, \quad (13)$$

where $\alpha \approx 1/3$ and $p \to p_c$. We wish to stress that the asymptotic behavior for $\varepsilon(p)$ in (11) differs from that in (13) in that it is valid only under condition (5), i.e., only outside a certain neighborhood near the percolation threshold, whose size $|\Delta p| \approx (a/\lambda)^{1/\nu}$ depends on the wavelength of the light.

Finite clusters contribute substantially to the optical response in this problem, although such clusters are unimportant in the conductivity problem. The reason for this result is the presence of nonsingular terms in the expression for the effective dielectric constant. Using (2) and (11), we find an expression for the reflection coefficient $|r_s(p)|^2$ of s-polarized light incident normally on this system:

$$|r_s(p)|^2 = \frac{\omega^2 d^2}{4c^2} |1-\varepsilon(p,\omega)|^2$$

$$= |r_s(p_c)|^2 + \frac{8\pi^2\omega^2|\alpha(\omega)|^2 p_c}{c^2 a^4}(p-p_c)$$

$$\times \mathrm{Re}\left\{\left[1+\frac{2\alpha^*(\omega)}{a^3}p_c\right]\left[1+\frac{4\alpha(\omega)}{a^3}\right]\right\}. \quad (14)$$

Only the nonsingular terms which determine the linear dependence of the reflection coefficient on $p-p_c$ have been retained in (14). Gadenne *et al.*[15] have reported experimental results on the behavior of the optical reflection and transmission coefficients of an island gold film as a function of the occupation factor p, which was varied from zero to one. Near the percolation threshold, the reflection and transmission coefficients were found to be linear functions of $p-p_c$, in agreement with theoretical expression (14). Unfortunately, it is not possible to distinguish a singular component in the experimental data.

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