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ELECTRIC CONDUCTIVITY OF INHOMOGENEOUS TWO-COMPONENT MEDIA IN TWO DIMENSIONS

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The electric conductivity is calculated for regular inhomogeneous two-component isotropic medium in which droplets of one phase with conductivity σ_2 are embedded in another, with conductivity σ_1. An expression is formulated that can be used in many different situations and is of particular relevance in the case where the relative proportion of the components is temperature-dependent and varies over a wide range. Behavior of the effective conductivity depends on the spatial arrangements and the shape of the inclusions.

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1. INTRODUCTION

Determination of the effective conductivity σ_{eff} of spatially inhomogeneous heterophase systems is an old, but increasingly important problem of theoretical physics. With the advent of new nanoscale probes of condensed matter systems, it has become apparent that many very diverse systems that were previously thought to be homogeneous are in fact either statically or dynamically inhomogeneous. The effective conductivity in such cases cannot be dealt with in terms of homogeneous medium theory, is not trivial, and solutions are presently known only in some rather special cases. Different aspects of the theory and different limiting cases are extensively discussed in Ref. [1].

In this paper, we focus on the problem of calculating the effective conductivity of an inhomogeneous two-dimensional ($2D$) plane. The classical problem can be formulated as follows. We assume that a $2D$ system contains a mixture of N ($N \geq 2$) different phases or materials with different conductivities σ_i, $i = 1, 2, \ldots, N$. The arrangements of different phases can be random or regular. The question that we wish to address is how the effective conductivity of the plane depends on the conductivities of the phases, their concentration, and the spatial arrangements.

In the past, a number of different approaches have been used to tackle this problem. The exact result for the effective conductivity of a two-component system with a symmetric and isotropic distribution of components was obtained by Dykhne [2]. He found that the effective conductivity of the system is determined by the simple relation

$$\sigma_{eff} = \sqrt{\sigma_1 \sigma_2}.$$

A symmetric distribution used in this problem amounts to the case where the two components can be interchanged without changing the end result. Obviously, one requirement for a symmetric distribution is that the two components have equal proportions, but it also means that more general cases cannot be considered with this model.

Further investigations have shown that a more general duality relation is valid for $2D$ heterogeneous conductors than that initially considered by Keller and Dykhne [1]. More recently, it was shown that a more general relation for the effective conductivity tensor exists that is valid for multicomponent and anisotropic systems [3, 4]. The effective conductivity of several examples of ordered two-component systems was also calculated exactly [5–7]. It was shown in [5, 6] that for a chessboard plane and for a plane constructed of triangles, the relation derived by Dykhne is also valid.

A similar relation to the Dykhne formula for the effective conductivity of a system consisting of randomly distributed metallic and dielectric regions near a metal-to-insulator transition was derived by Efros

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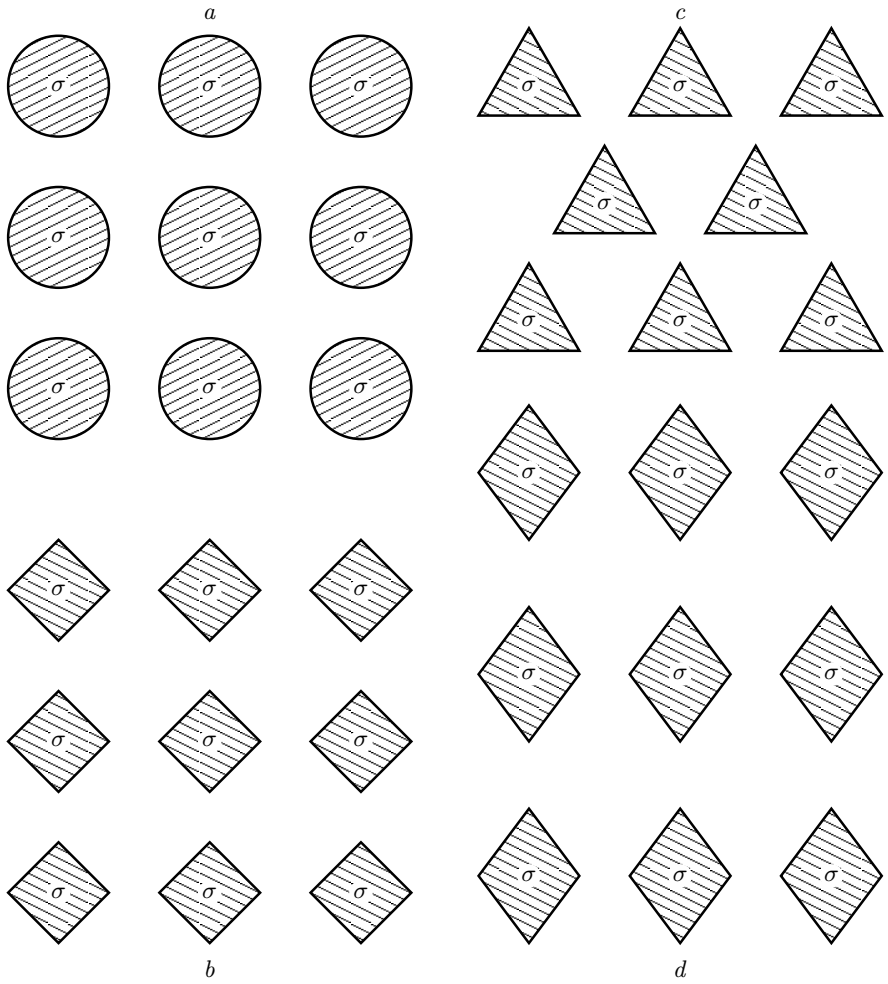

Fig. 1. Spatial arrangements of phases with conductivities $\sigma_1 = 1$ and $\sigma_2 = \sigma$ for four considered cases

and Shklovskii [8]. They generalized the expression of Dykhne on the basis of scaling arguments to the case of arbitrary concentrations of the two phases near a percolation threshold, such that the effective conductivity becomes

$$\sigma_{eff} = \sigma_1(\sigma_2/\sigma_1)^s, \tag{1}$$

where s is a universal scaling exponent. Critical exponents are also relatively well-known for this type of systems [9]. This relation is not applicable when the system is driven away from the percolation threshold and the general solution of the effective conductivity of an inhomogeneous medium thus remains an open problem.

Sen and Torquato [10] derived an expression that allows an explicit calculation of the effective conductivity tensor from the n-point probability functions $S_n(\mathbf{r}_1, \ldots, \mathbf{r}_n)$. These functions give the probability of the points at $\mathbf{r}_1, \ldots, \mathbf{r}_n$ to belong to the same phase, and are therefore uniquely determined by the spatial distribution of the phases. Unfortunately, the application of this method is limited because the computations with $n > 5$ are fairly time consuming.

Different expansions of the effective conductivity in terms of a small parameter have been used in the past [1, 11, 12]. In most cases, the low-order terms weakly depend on microgeometry. A diagrammatic expansion for the effective conductivity developed by Khalatnikov and Kamenshchik [13] promises to give more generally applicable results. The perturbative approach seems to be quite effective because it allows analyzing random and nonsymmetric distributions with different conductivities.

The problem was also discussed in the case where $N = 2$ and $N = 3$ on the basis of numerical calculations [3, 14]. It was shown that the effective conductiv-

ity for $N = 3$ is not universal and depends on the spatial arrangements of the phases. We have employed the boundary element method for efficient numerical treatment of two-dimensional multi-phase systems with an arbitrary arrangement of phases. More details on the method and its results can be found in [15].

In this paper, we consider the conductivity of a two-phase system in two dimensions for a wide range of concentrations and conductivities. One phase is assumed to be composed of droplets (of different shapes) with conductivity σ_2 embedded within a medium of conductivity σ_1 (see Fig. 1). We begin with calculating the effective conductivity σ_{eff} using a perturbation theory approach with the two phases having the respective volume fractions $(1-\nu)$ and ν. Because the problem is linear, we can introduce a dimensionless conductivity σ, measured in units of $\sigma_1 = 1$, and the effective conductivity σ_{eff} is a function of $\sigma = \sigma_2/\sigma_1 = \sigma_2$ and ν. The volume-averaged conductivity

$$\bar{\sigma} = \frac{1}{V}\int \sigma\, dV$$

is given by

$$\bar{\sigma} = (1-\nu) + \nu\sigma. \tag{2}$$

If the conductivities of the two phases are not vastly different,

$$|\sigma - 1| \ll 1,$$

the effective conductivity can be calculated by perturbation theory [13]. To apply perturbation theory, we rewrite the spatial dependence of the conductivity as

$$\sigma(\mathbf{r}) = \bar{\sigma}(1 - \alpha(\mathbf{r})), \tag{3}$$

where

$$\alpha(\mathbf{r}) = \frac{\sigma(\mathbf{r}) - \bar{\sigma}}{\bar{\sigma}}.$$

Assuming that the spatial distribution of conductivity is uncorrelated, we then obtain

$$\int d\mathbf{r}\, \alpha(\mathbf{r})\alpha(\mathbf{r}+\mathbf{r}') = \frac{(\sigma-1)^2\nu(1-\nu)}{\bar{\sigma}^2}\delta(\mathbf{r}'). \tag{4}$$

A straightforward calculation shows that up to the second order in α, the conductivity is given by

$$\sigma_{eff} = \bar{\sigma} - \frac{(\sigma-1)^2\nu(1-\nu)}{2\bar{\sigma}}. \tag{5}$$

This result has been known for many years and was derived for the dielectric function of dielectric mixtures [16]. In Refs. [11, 12], it was also derived using a systematic perturbative expansion, which showed it to be exact to the second order in α. The second term in Eq. (5) represents the first nonvanishing contribution due to the inhomogeneity of the distribution of the phases. In the case where $\nu = 0.5$, the result coincides with the expansion of the exact expression for the conductivity up to the second order in $(\sigma_2 - \sigma_1)$ [2]:

$$\sigma_{eff} = \sqrt{\sigma_1\sigma_2}. \tag{6}$$

2. CONDUCTIVITY OF A REGULAR ISOTROPIC TWO-COMPONENT SYSTEM IN TWO DIMENSIONS

Next, we exactly calculate the effective conductivity of the plane with different regular isotropic distributions. As before, we consider a $2D$ plane constructed from two different phases with different conductivities $\sigma_1 = 1$ and $\sigma_2 = \sigma$. The regions with the conductivity σ_2 have a circular shape with radius R and form a regular square lattice with the period a as shown in Fig. 1*a*. Changing the radius R from 0 to $a/2$, we can change the volume fraction of the second phase from $\nu = 0$ to the critical concentration $\nu_c = 0.785$, whereafter the regions with the conductivity σ_2 start to overlap and the percolation threshold is reached. In the case of metallic droplets, the total charge density must be zero, while a finite charge density can accumulate on the surface between different phases. This allows us to formulate the integral equation for the surface charge density [5, 6]. We define the surface charge density by the relation

$$\rho(\theta)R\, d\theta = d\rho(\theta),$$

where $d\rho(\theta)$ is the charge on a small part of the surface between the two components with the length

$$dl = R\, d\theta.$$

We recall that the scalar potential at the point $\mathbf{r}$ is determined by the relation

$$\phi = E_0 x - 2\int d^2r' \ln|\mathbf{r}-\mathbf{r}'|\rho(\mathbf{r}'), \tag{7}$$

where $\ln|\mathbf{r}-\mathbf{r}'|/2\pi$ is the $2D$ Green's function. The boundary conditions on the surface between two phases are [16]

$$E_n^1 - E_n^2 = 4\pi\rho(\theta), \tag{8}$$

$$\sigma_1 E_n^1 = \sigma_2 E_n^2. \tag{9}$$

Substituting

$$\mathbf{r}' = \mathbf{i}(ma + R\cos\theta') + \mathbf{j}(na + R\sin\theta'),$$

$$\mathbf{r} = \mathbf{i}R\cos\theta + \mathbf{j}R\sin\theta$$

in Eqs. (7)–(9), we obtain an integral equation for the surface charge density in the form

$$\rho(\theta) = \frac{\kappa}{2\pi}\left[E_0\cos\theta + 2r\sum_{n,m=-\infty}^{\infty}\int_{-\pi}^{\pi} d\theta'\rho(\theta')\,\mathrm{Re}\left\{\frac{\exp(i\theta)}{m + r(\cos\theta' - \cos\theta) + i(n + r(\sin\theta' - \sin\theta))}\right\}\right], \tag{10}$$

where

$$r = \frac{R}{a}, \quad \kappa = \frac{1-\sigma}{1+\sigma}.$$

As shown in the Appendix, the sum over m can be calculated exactly and the integral equation for the surface charge density is reduced to the form

$$\rho(\theta) = \frac{\kappa}{2\pi}\left[E_0\cos\theta + 2r\sum_{n=-\infty}^{\infty}\int_{-\pi}^{\pi} d\theta' K(n,\theta,\theta')\rho(\theta')\right], \tag{11}$$

where

$$K(n,\theta,\theta') = \pi\frac{\cos\theta\sin\left(2\pi r(\cos\theta' - \cos\theta)\right) + \sin\theta\,\mathrm{sh}\left[2\pi(n + r(\sin\theta' - \sin\theta))\right]}{\mathrm{ch}\left[2\pi(n + r(\sin\theta' - \sin\theta))\right] - \cos\left(2\pi r(\cos\theta' - \cos\theta)\right)}. \tag{12}$$

Expanding the surface density $\rho(\theta)$ in terms of Legendre polynomials $P_l(\cos\theta)$ and taking into account that

$$\rho(-\theta) = \rho(\theta), \quad \rho(\pi - \theta) = -\rho(\theta),$$

where

$$\rho(\theta) = \sum_{l=1}^{\infty} c_{2l-1}P_{2l-1}(\cos\theta), \tag{13}$$

we obtain the linear set of algebraic equations for the coefficients c_{2l-1},

$$\frac{2c_{2l-1}}{4l-1} = \frac{\kappa}{2\pi}\left[\frac{2}{3}E_0\delta_{l,1} + 2r\sum_{k=1}^{\infty} c_{2k-1}K_{l,k}\right], \tag{14}$$

where

$$K_{l,k} = \sum_{n=-\infty}^{\infty}\int_0^{\pi} d\theta'\int_0^{\pi} d\theta\, K(n,\theta,\theta')\sin\theta P_{2l-1}(\cos\theta)P_{2k-1}(\cos\theta'). \tag{15}$$

Solving Eq. (14) with a finite number of Legendre polynomials taken into account, we obtain the surface charge density in Eq. (13). As a result, the effective conductivity is evaluated by calculating the total current

$$j = \sigma_1 E_n = E_n$$

through the semicircular surface with the radius $R' = a/2$ (see Fig. 1*a*). Calculations similar to that of Eq. (12) lead to the expression for the effective conductivity

$$\sigma_{eff} = \frac{\kappa}{4\pi}\int_{-\pi/2}^{\pi/2} d\theta\left[\cos\theta + \frac{2r}{E_0}\sum_{n=-\infty}^{\infty}\int_{-\pi}^{\pi} d\theta' K'(n,\theta,\theta')\rho(\theta')\right], \tag{16}$$

where

$$K'(n,\theta,\theta') = \pi\frac{\cos\theta\sin(2\pi(r\cos\theta' - (1/2)\cos\theta)) + \sin\theta\,\mathrm{sh}\left(2\pi(n + r\sin\theta' - (1/2)\sin\theta)\right)}{\mathrm{ch}\left(2\pi(n + r\sin\theta' - (1/2)\sin\theta)\right) - \cos\left(2\pi(r\cos\theta' - (1/2)\cos\theta)\right)}. \tag{17}$$

The result above applies to the case of a uniform distribution of circular droplets within the plane. To see how the effective conductivity depends on the shape of the regions with the conductivity σ_2, we have performed calculations in the case where circular droplets were replaced with squares, triangles, and rhombuses with the ratio of diagonals $\operatorname{tg}\alpha = a/b$, where a and b are translation vectors along x and y axes respectively (see Fig. 1 *b, c, d*). In all these cases, Eqs. (10)–(17) are slightly modified because in a polar coordinate system, $r(\theta)$ is a function of the angle. Unlike in the case of circles, the percolation threshold for cases b, c, and d is $\nu_c = 0.5$. We note that in the case of rhombuses, the lattice is anisotropic and $\sigma_{eff}^{11} \neq \sigma_{eff}^{22}$.

3. DISCUSSION

The results of the calculations of the effective conductivity are presented in Fig. 2, as a function of σ^ν for different values of the volume fraction ν. It is easy to check that the results satisfy the generalized duality relation [3, 4]

$$\sigma_{eff}^{11}(\sigma_1, \sigma_2)\sigma_{eff}^{22}(1/\sigma_1, 1/\sigma_2) = 1. \tag{18}$$

For circles, squares, and triangles,

$$\sigma^{22} = \sigma^{11}.$$

For rhombuses,

$$\sigma^{22}(\alpha) = \sigma^{11}(\pi/2 - \alpha).$$

Figure 2 *a, b, c, d* shows that for small κ, the perturbation theory [11–13] (Eq. (5)) gives the correct result independent of the geometry.

3.1. Approximate expression for the effective conductivity

Although the predictions in Fig. 2 represent the results of a precise numerical calculation, they are not very tractable when it comes to comparing with experimental data, being the result of numerical calculations. It is therefore helpful to try to obtain a functional form for describing the behavior predicted in Fig. 2, which also includes all the relevant parameters, such as the volume fraction ν and the two conductivities σ_1 and σ_2. Such an expression can then be used in a wide range of problems, provided the validity range is taken into account. We describe the properties of such a heuristically determined function and determine its validity range in terms of the parameters ν, σ_1, and σ_2.

As can be seen from Fig. 2, the dependence of the effective conductivity on σ shows similar behavior independently of the particular geometry of the phases. First, we observe that when κ is small, all the curves are linear in σ^ν with the same slope. In the relatively wide interval $0.1 < \sigma < 10$, the effective conductivity is determined by the equation

$$\sigma_{eff}(\sigma) = \sigma_1^{(1-\nu)}\sigma_2^\nu. \tag{19}$$

The range of applicability of this formula becomes wider as we approach the percolation threshold ν_c. When $\sigma = \sigma_2/\sigma_1 \gg 1$, the effective conductivity saturates at σ_{sat}. The value of σ_{sat} is not universal and depends on the geometry. It was pointed out recently that in the case of circles with $\nu < 0.5$ in the entire range of σ, the effective conductivity may be approximated by the formula [18]

$$\sigma_{eff}(\kappa) = \frac{1 - \nu\kappa}{1 + \nu\kappa}. \tag{20}$$

To derive an approximate expression for the effective conductivity, we assume that Eq. (20) remains correct if we replace ν with the effective volume fraction $\nu_{eff}(\kappa, \nu)$. We require that

$$\nu_{eff}(\kappa, \nu) \approx \nu \quad \text{as} \quad \kappa \to 0$$

or

$$\nu \to 0$$

and

$$\nu_{eff}(\kappa, \nu) \approx \frac{1}{\kappa}\frac{1 - \sigma^{\nu_c}}{1 + \sigma^{\nu_c}} \quad \text{as} \quad \nu \to \nu_c$$

to satisfy Eq. (19), which is valid at $\nu = \nu_c$. It is easy to see that the function

$$\nu_{eff}(\kappa, \nu) = \nu + \frac{1}{\kappa} \times \\ \times \frac{1 - \left(\dfrac{1 - (1 - p(\nu))\kappa}{1 + (1 - p(\nu))\kappa}\right)^{\nu_c}}{1 + \left(\dfrac{1 - (1 - p(\nu))\kappa}{1 + (1 - p(\nu))\kappa}\right)^{\nu_c}} - (1 - p(\nu))\nu_c, \tag{21}$$

where $p(\nu) \to 0$ as $\nu \to \nu_c$ and $p(\nu) \to 1$ as $\nu \to 0$, satisfies all the above requirements. The function $p(\nu)$ is not universal and depends on the geometric shape of the region with conductivity σ and on the particular arrangement of these inclusions in the $2D$ plane. In Fig. 3, we plot $p(\nu)$ as a function of $1 - \nu/\nu_c$ in cases a, b, and c. Case d is different because the effective conductivity is anisotropic. As is clearly seen from Fig. 3, the behavior of the function $p(\nu)$ for circles (case a) is different from the cases of squares and triangles (b and c). On the other hand, in cases b and c, $p(\nu)$ shows similar behavior.

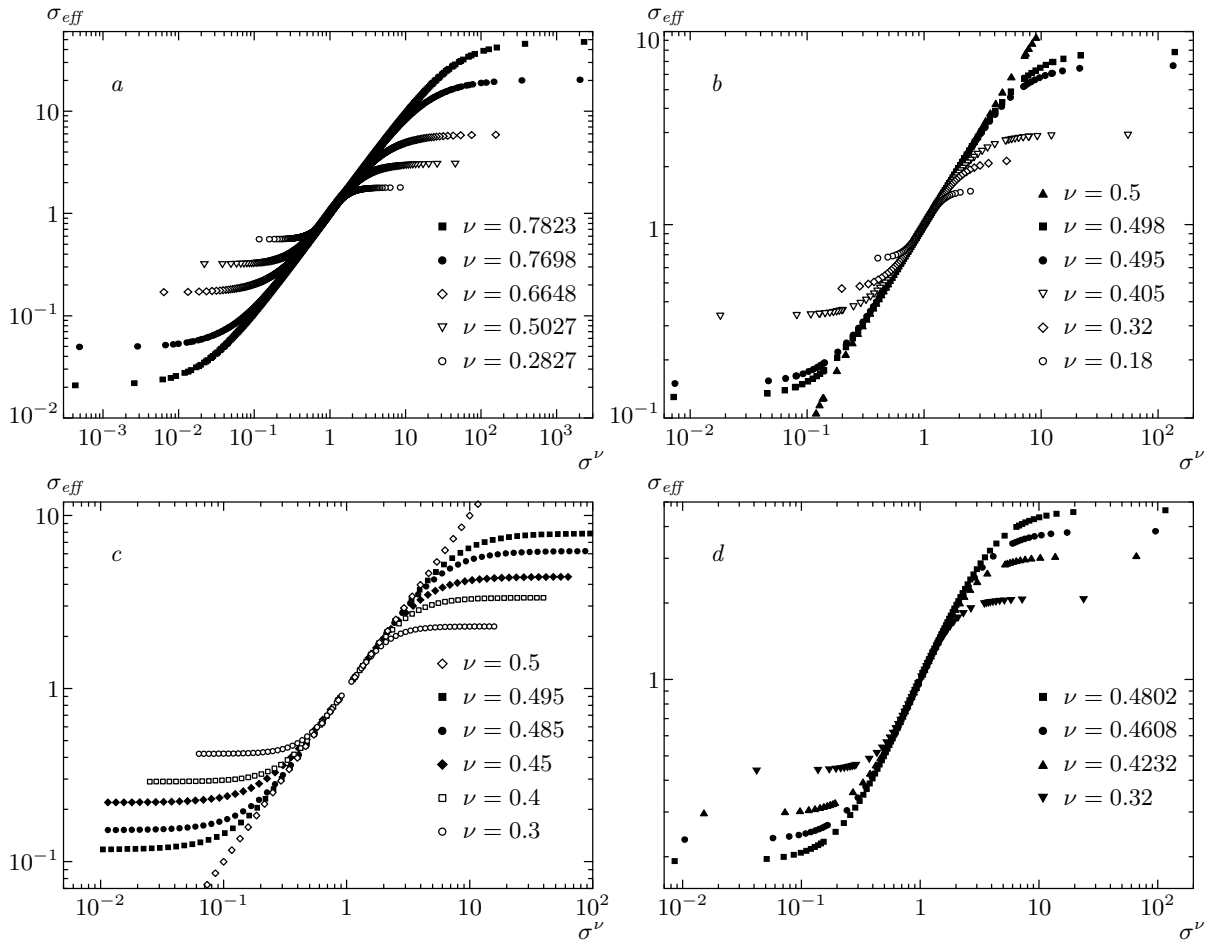

Fig. 2. Effective conductivity of the plane as a function of σ^ν for different volume fractions and four considered geometries

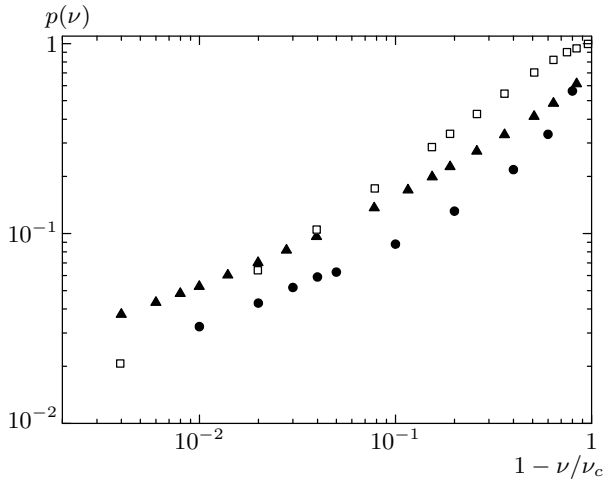

Fig. 3. Dependence of the function $p(\nu)$ on $1 - \nu/\nu_c$ for cases a (circles), b (squares), and c (triangles)

3.2. Shape dependence of the effective conductivity

The function $p(\nu)$ is related to the value of

$$\sigma_{sat} = \frac{1 + \nu_{eff}(\kappa = -1, \nu)}{1 - \nu_{eff}(\kappa = -1, \nu)}.$$

Therefore, the behavior of $p(\nu)$ close to the percolation threshold should be different for different geometries. In Fig. 4, we plot the value of σ_{sat} as a function of $(1 - \nu/\nu_c)$ in the case of circles, squares, and triangles. There is an important difference between these two cases. In the case of circles, σ_{sat} has a power-like divergence $(1 - \nu/\nu_c)^{-k}$ $(k \approx 0.5)$. For squares and triangles, this behavior is logarithmic. In both cases, close to percolation threshold, σ_{sat} is proportional to the average inverse distance between boundaries of the neighboring circles or squares,

$$\sigma_{sat} \propto \int \frac{dy}{1 - 2f(y)},$$

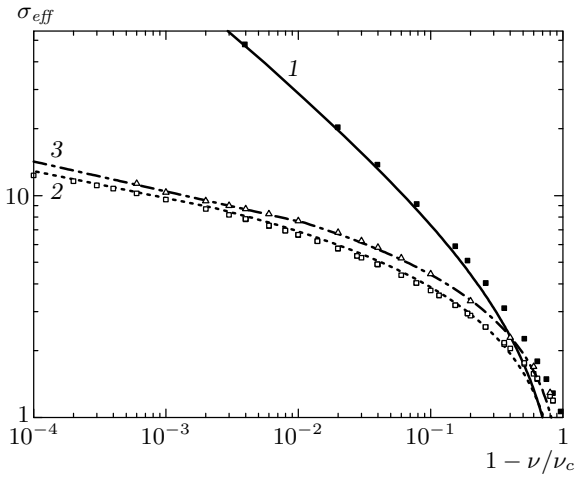

Fig. 4. Saturated effective conductivity as $\sigma \to \infty$ for cases a, b, and c. Full, dotted, and dashed lines show different analytic asymptotic behavior for these cases: 1 — $(\pi/2 + \arcsin((\nu/\nu_c)^{1/2}))/(1-\nu/\nu_c)^{1/2} - \pi/2$ (case a), 2 — $1.3\ln(1/(1-(\nu/\nu_c)^{1/2}))$ (case b), 3 — $\sqrt{2}\ln(2/\sqrt{3}(1-(\nu/\nu_c)^{1/2}))$ (case c)

where

$$f(y) = \sqrt{r^2 - y^2}$$

for circles and

$$f(y) = r - |y|$$

for squares. We here assume that the period of the system is 1, and the dimensionless size of the circle and the square is r. Direct integration leads to the following results:

$$\sigma_{sat} \propto \frac{\pi/2 - \arccos((1-\nu/\nu_c)^{1/2}}{(1-\nu/\nu_c)^{1/2}} - \pi/2 \qquad (22)$$

for circles and

$$\sigma_{sat} \propto -\ln\left(1-(\nu/\nu_c)^{1/2}\right) \qquad (23)$$

for squares (Fig. 4). For triangles (c), the asymptotic formula is similar to Eq. (23) with different numeric coefficients. Interestingly, this observation suggests that behavior of the function $p(\nu)$ is different depending on the curvature of the embedded regions.

4. CONCLUSION

From calculations of the effective conductivity of inhomogeneous two-phase systems in two dimensions, we find that the results of precise numerical calculations can be approximated by a universal function for σ_{eff}, Eqs. (20) and (21), where the function $p(\nu)$ depends on the spatial arrangements of the $2D$ plane and on the shape of the inclusions with conductivity σ. It is shown that in a large interval of the conductivity σ, the effective conductivity σ_{eff} is determined by the spatial average of the logarithm of individual conductivities. The closer the system is to the percolation threshold, the larger the validity range of this result. For large values of the conductivity σ, σ_{eff} saturates at a value σ_{sat}. The value of σ_{sat} near the percolation threshold is determined by the average inverse distance between boundaries of neighboring regions with the conductivity σ in the direction of the field (Eqs. (22) and (23)).

The model that we have developed is quite generally applicable and can be applied in some interesting situations, such as cuprates and other two-dimensional complex transition metal oxides that exist near a phase-separation threshold. Importantly, there appears to be a significant amount of experimental evidence that many anomalous properties of oxides are associated with the coexistence of two or more phases. The application of the presented model may help understanding the transport properties of such systems.

APPENDIX

Here, we show how the sum over m in Eq. (10) can be calculated exactly. We represent the sum as

$$S = \sum_{m=-\infty}^{\infty} \mathrm{Re}\left\{\frac{\exp(i\theta)}{m+\beta+i\alpha}\right\},$$

where

$$\beta = r(\cos\theta' - \cos\theta), \quad \alpha = n + r(\sin\theta' - \sin\theta).$$

The sum over m is calculated using the definition of the digamma function. As a result, we express the sum as

$$S = \mathrm{Re}\left\{\exp(i\theta)[\psi(-\beta - i\alpha) - \psi(1+\beta+i\alpha)]\right\} = \\ = \pi\,\mathrm{Re}\left\{\exp(i\theta)\,\mathrm{ctg}(\pi(\beta+i\alpha))\right\}.$$

Calculating the imaginary part of the previous equation, we obtain the result in Eq. (12),

$$S = \pi\frac{\cos\theta\sin(2\pi\beta) + \sin\theta\,\mathrm{sh}(2\pi\alpha)}{\mathrm{ch}(2\pi\alpha) - \cos(2\pi\beta)}.$$

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