Effective conductivity of strongly inhomogeneous media near the percolation threshold

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Transport phenomena in a two-phase strongly inhomogeneous composite below the percolation threshold ($p<p_c$) are considered. On the basis of a very simple model of percolation of current across intermediate layers connecting fragments of a metallic cluster, the influence of Joule heating on the effective electrical conductivity is found. It is shown that near the percolation threshold the dependence of the local conductivity on the temperature is appreciable, and allowance for this dependence can lead to a change in the critical index.

The great interest currently being shown in the description of transport phenomena in macroscopically inhomogeneous media is connected both with the possible practical applications and with the recently developed new methods of calculating effective transport coefficients of strongly inhomogeneous media. On the one hand, in the two-dimensional case a class of media has been found for which it is possible to obtain exact expressions for the effective transport coefficients (connecting, by definition, volume-averaged thermodynamic fluxes and forces),[1–3] and, on the other hand, the methods of percolation theory make it possible to find in the critical region (near the metal-insulator phase transition) the concentration dependence of the effective conductivity[4,5]; in particular,

$$\sigma^e\approx\sigma\tau^{-q},\quad \tau=(p_c-p)/p_c,\quad p<p_c, \tag{1}$$

where p is the concentration of the metallic phase (the good conductor), p_c is the critical concentration at which an infinite cluster appears in the medium, and $q>0$ is the critical exponent; in the three-dimensional case, $q\cong 0.98$.

Investigations of the distributions of the local electric-field intensity, current density, and Joule heating in strongly inhomogeneous media near the percolation threshold[6–8] point to their strong spatial nonuniformity. Explicit allowance for this fact can, in certain cases, change the behavior of the effective conductivity near the percolation threshold from that described by (1), which is based on purely geometrical considerations.

We consider here (qualitatively) the influence of Joule heating on the effective conductivity below the percolation threshold ($p<p_c$) and show that in certain cases this influence can become appreciable.

At $p>p_c$ there exists in the medium an infinite metallic cluster, through which percolation of current occurs. At $p<p_c$ the infinite cluster breaks down into finite fragments with characteristic size L (L is the correlation length).[4,5] The conduction in this case occurs through metallic-cluster fragments and thin intermediate layers of a poorly (in comparison with the metal) conducting medium between neighboring clusters. Henceforth we assume that the ratio of the conductivity σ_m of the good-conductor phase to the conductivity σ of the poorly conducting phase is such that the voltage drop across finite metallic fragments can be neglected in comparison with that across an intermediate layer.

In the first approximation we can assume that an intermediate layer has the shape of a cylinder with end faces of area s a distance l apart. As will be seen from the following, the characteristic size along the layer ($s^{1/2}$) is much greater than the distance across the layer (l), i.e., the layer is thin; thus, in the same approximation, the layer can be nonplanar (crumpled), and this will not affect its resistance (see also Fig. 1 in Ref. 9).

To calculate the effective conductivity it is sufficient to consider a volume L^3 in which there is one layer between metallic clusters. It is assumed that the overwhelming majority of the layers of which the main resistance of the medium is built up have equal resistances and are distributed randomly in the medium. We note that an analogous situation obtains in the percolation of current in a polycrystalline medium with crystallites having strongly anisotropic conductivity,[10] in which the main contribution to the resistance of the medium is built up at "traps" that are randomly distributed in the medium.

We consider now the characteristic volume L^3 for a strongly inhomogeneous medium. Its resistance ($R\cong L/\sigma^e S$, $S\cong L^2$ is easily estimated by considering an elementary circuit in which the resistances of the intermediate layer ($R_1\cong l/\sigma s$) and of the remaining part of the medium ($R_2\cong L/\sigma L^2$) are connected in parallel (the so-called two-zone model; see, e.g., Ref. 9):

$$\sigma^e\approx\sigma(1+s/\pi lL), \tag{2}$$

whence, if we take (1) into account,

$$s/Ll\sim\tau^{-q}. \tag{3}$$

For $p\to p_c$ we have $l\ll L$ and $s\ll S$ and we can assume the following asymptotic behavior:

$$l/L\sim\tau^r,\quad s/S\sim\tau^g,\quad r-g=q, \tag{4}$$

with $r>0$ and $g>0$; since $q>0$, we have $r>g$. Nothing is known about the numerical value of r, but, as will be seen below, these inequalities are sufficient.

We now show that the above geometrical structure (3), (4) makes it possible to obtain the concentration dependence of the thermopower α^e. First we consider the case when there is a large difference in the thermal conductivities of the phases: $\varkappa_m \gg \varkappa$. Then the temperature difference ΔT, specified at the boundaries of the volume L^3 (the average temperature gradient is $\sim \Delta T/L$), is concentrated across the intermediate layer (here the temperature gradient is $\sim \Delta T/l$). Considering an elementary circuit with the emf's connected in parallel ($\varepsilon_1 \cong \varepsilon^2 \cong \Delta\alpha\Delta T$), we obtain

$$\alpha^e \approx \frac{\varepsilon^e}{\Delta T} = \frac{\varepsilon_1+\varepsilon_2 R_1/R_2}{1+R_1/R_2}\frac{1}{\Delta T} = \Delta\alpha, \tag{5}$$

whence it follows that for $p \to p_c$ the thermopower α^e does not depend on the concentration.

The second case is that of a small difference in the thermal conductivities ($\varkappa_m \cong \varkappa$); here $\varepsilon_1 \cong \Delta\alpha\Delta T l/T$, $\varepsilon_2 \cong \Delta\alpha\Delta T$, and

$$\alpha^e \approx \Delta\alpha\,(l/L+\tau^q)/(1+\tau^q), \tag{6}$$

whence, taking into account the concentration behavior (4) of l/L as $p \to p_c$, we obtain

$$\alpha^e \approx \Delta\alpha\tau^q. \tag{7}$$

Thus, for $\varkappa_m \cong \varkappa$, as $p \to p_c$ the thermopower α^e decreases in accordance with the law (7).

The concentration dependence of the thermopower near the percolation threshold was considered earlier in Refs. 3 and 11. There it was assumed that the concentration dependences of the effective electrical conductivity and thermal conductivity were known ($\varkappa^2 \cong \varkappa\tau^q$ for $\varkappa_m \gg \varkappa$, and $\varkappa^e$ does not depend on p for $\varkappa_m \cong \varkappa$), and the behavior of α^e followed from the relation

$$\alpha^e = [\alpha_1\sigma_1\varkappa_2 - \alpha_2\sigma_2\varkappa_1 - \sigma_1\sigma_2(\alpha_1-\alpha_2)\varkappa^e/\sigma^e]\,(\sigma_1\varkappa_2-\sigma_2\varkappa_1)^{-1}, \tag{8}$$

which was obtained in Refs. 3 and 12 and relates σ^e, α^e, and $\varkappa^e$ in two-phase media. In (8) terms with $(\alpha^e)^2$ have been discarded; in comparison with the other terms they are small—of order ZT ($Z = \sigma\alpha^2/\varkappa$ is the thermoelectric quality factor). We note that the relation (8) is valid only in the approximation linear in the gradient, i.e., with neglect of the Joule heating and the temperature dependences of the local transport coefficients.

The fact that the calculation of α^e based on the use of the relation (8) agrees with a calculation based only on geometrical considerations points, on the one hand, to the consistency of the proposed geometrical structure (3), (4), and, on the other hand, to those approximations in which the concentration dependence of α^e can be considered.

Analogously, it can be shown that the proposed geometrical structure (3), (4) makes it possible to obtain the dependence of the Hall coefficient on concentration and field in the two-dimensional case (the three-dimensional situation is more complicated and will not be considered here).

We shall assume for simplicity that the metallic-cluster fragments are perfectly conducting; this makes it possible to confine ourselves to considering the fields and currents only in the poorly conducting phase, while specifying the condition $E_\tau = 0$ on the boundary with the metal (E_τ is the tangential component of the electric-field intensity).[13]

In the two-dimensional case the intermediate layer is a thin strip of thickness l, separating metallic-cluster fragments of width b (b is the size of the cluster contacts with the intermediate layer). We choose the coordinate system in such a way that the X axis is in the direction of the average field $\langle E\rangle$, and consider a characteristic "volume" L^2, in which a cluster is also oriented, on the average, along the X axis (precisely such volumes L^2 contribute to the critical behavior of the effective transport coefficients). The averaged Ohm's law in this case has the form

$$\langle j_x\rangle = \sigma_s^e\langle E\rangle, \quad \langle j_y\rangle = -\sigma_a^e\langle E\rangle. \tag{9}$$

Local currents can be considered as a system of two mutually perpendicular currents flowing, on the average, along the X and Y axes. The former currents are connected with the diagonal component $\sigma_s = \sigma(1+\beta^2)^{-1}$ of the conductivity tensor, and the latter currents (the Hall currents) are connected with the nondiagonal component $\sigma_a = \sigma\beta(1+\beta^2)^{-1}$ (β is the dimensionless magnetic field).

Along the X axis the current flows in a manner analogous to that in the case considered above with $\beta = 0$. Thus,

$$\sigma_s^e \approx \sigma_s\tau^{-q}, \tag{10}$$

and in the two-dimensional case, in place of (3), we shall have $b/l \propto \tau^{-q}$. Along the axis outside the layer the current density $j_y = -\sigma_a\langle E\rangle$; in the layer, the local field $E \cong \langle E\rangle L/l$, and, correspondingly, $j_y' \cong -\sigma_a\langle E\rangle L/l$. We note that the entire Hall current approaching a metallic cluster passes through the intermediate layer ($j_y L \cong j_y' l$), as is entirely natural, since the metallic cluster is assumed to be ideal ($E_\tau = 0$). Thus, the average Hall current is equal to $\langle j_y\rangle = -\sigma_a\langle E\rangle$, whence, according to (9),

$$\sigma_a^e \approx \sigma_a. \tag{11}$$

Knowing σ_s^e and σ_a^e, one can easily find the field and concentration dependences of the components ρ_s^e and ρ_a^e of the resistivity tensor (and hence the field and concentration dependences of the Hall coefficient $R_e = \rho_a^e/H$):

$$\rho_s^e \approx \frac{1}{\sigma}\frac{1+\beta^2}{\beta^2+\tau^{-2q}}\tau^{-q}, \quad \rho_a^e \approx \frac{\beta}{\sigma}\frac{1+\beta^2}{\beta^2+\tau^{-2q}}. \tag{12}$$

In the investigation of ρ_s^e and ρ_a^e we can distinguish three regions of fields:

$$\beta \ll 1, \qquad \rho_s^e \approx \rho\tau^q, \qquad \rho_a^e \approx \rho\tau^{2q}\beta, \tag{13}$$

$$1 \leqslant \beta \leqslant \tau^{-q}, \qquad \rho_s^e \approx \rho\tau^q\beta^2, \qquad \rho_a^e \approx \rho\tau^{2q}\beta^3, \tag{14}$$

$$\beta \gg \tau^{-q}, \qquad \rho_s^e \approx \rho\tau^{-q}, \qquad \rho_a^e \approx \rho\beta. \tag{15}$$

The concentration and field dependences (12)–(15) of the components of the resistivity tensor coincide fully with those from Ref. 13. It is necessary to note that in the presently considered model of the intermediate layer the region (15) cannot be realized, for if $\beta > \tau^{-q}$ the current flowing out of one end face of a metallic cluster is deflected so strongly by the magnetic field that it does not arrive at the second face, and the picture considered up to now of the percolation current through the metallic clusters ceases to be valid.[1)]

We now return to the question of the choice of the model to be used. The shape of an intermediate layer between two metallic-cluster fragments is not obvious in advance. The simplest alternative (if we dismiss exotic shapes whose probability, by definition, is negligible) is that the ends of the metallic cluster are not "blunt," but "sharp." In the two-dimensional case this situation was considered in Ref. 14, where it was shown that the effective conductivity in this case has a logarithmic dependence ($\sigma^e \cong -\sigma \ln\tau$) on the concentration, rather than a power dependence. In Ref. 14 the metallic inclusions, in the shape of squares, do not change their shape as they come together ($p \to p_c$). It is clear, however, that by means of a special change of shape upon change of the concentration it is possible to specify any dependence $\sigma^e = \sigma^e(p)$, including a power dependence. Thus, if as $p \to p_c$ the intermediate layer does not change its shape, the simplest layer shape giving a power-law behavior will evidently be a thin cylinder. These arguments, and also the regular concentration and field dependences of α^e, $\varkappa^e$, ρ_s^e, and ρ_a^e, permit one to hope that the proposed layer shape correctly reflects the principal properties of the medium near the percolation threshold.

Using the geometrical structure considered, we find the influence of the Joule heating on the effective electrical conductivity. Since almost the entire current passes through the intermediate layer, the heat liberated in it is

$$Q_1 = \int \rho j^2\, dV \approx (\Delta\varphi)^2 \sigma s/l, \tag{16}$$

where $\Delta\varphi$ is the potential difference applied to the boundaries (the average field $\langle E \rangle \cong \Delta\varphi/L$).

In the stationary regime all the heat should move away to the boundaries of the volume L^3 (we assume that the average temperature of the sample is T). It is possible to find the heat flux Q_2 moving out of the layer by making use of the geometry of the layer—from (3) and (4) it follows that

$$l/s^{1/2} \approx \tau^{r-g/2}, \quad 2r-g>0, \tag{17}$$

and also making use of an analogy with electrostatics (the capacitance of an ellipsoid with two equal axes that are much greater than the third[15]):

$$Q_2 \approx \varkappa s^{1/2} \Delta T, \tag{18}$$

where ΔT is the amount by which the temperature of the layer exceeds that of the medium; we have selected the case $\varkappa_m \cong \varkappa$ and have omitted the unimportant factor $\pi^{2/3}$.

Equating $Q_1 = Q_2$, we obtain

$$\Delta T \approx \tau^{-g/2} I \Delta\varphi / K, \tag{19}$$

where I is the current through the volume L^3 and $K = \varkappa L^2/L$ is the thermal conductance of this volume. Thus, the closer to the percolation threshold, the stronger the heating of the layer. It is clear that the actual existence of this heating is not connected with the specific shape of the layer. We shall consider the situation $I\Delta\varphi = \text{const}$, when the Joule heat liberated in volume L^3 in unit time does not depend on the concentration. It follows from (19) that for an arbitrarily small but finite value of $I\Delta\varphi$ there is a concentration above which the heating in the intermediate layer can become large: $\Delta T/T \gg 1$. In this case allowance for the temperature dependence of the local transport coefficients will introduce an important correction into the values of the effective coefficients.

In the study of the effective properties of macroscopically inhomogeneous media the temperature dependence of the local transport coefficients is, as a rule, neglected. There are, apparently, two reasons for this. A weak dependence of the local transport coefficients has a weak influence on the effective properties; for a strong dependence the medium becomes nonuniform on the average, and the effective transport coefficients cease to be self-averaging quantities—a correct determination of them in the present case is problematic.

Near the percolation threshold allowance for the local dependence of the coefficients is important even in the absence of an average temperature gradient. Since the layers, and hence the places of local heating, are distributed throughout the medium in a random manner, the medium remains uniform on the average even when the temperature dependence is taken into account. Thus, in the situation under consideration, correct (qualitative) allowance for the temperature dependence of the local and effective conductivity is possible. With $\sigma = \sigma(T)$, instead of (1) we obtain

$$\sigma^e \approx \sigma(T+\Delta T)\tau^{-q}, \tag{20}$$

where ΔT is determined from (19).

As a very simple illustration we can consider a linear dependence of the local conductivity on the temperature: $\sigma(T) = a + bT$. In this case, at a definite concentration p_0 ($aK\tau_0^{g/2}/bI\Delta\varphi \ll 1$), the critical exponent of $\sigma^e(p)$ changes:

$$\sigma^e(p) \approx b(I\Delta\varphi/K)\tau^{-(q+g/2)}. \tag{21}$$

In inhomogeneous media, transport processes leading to a nonlinear current-voltage characteristic have recently been under active study (see, e.g., Refs. 16–18). We note one further fact that follows from (20): Allowance for the temperature dependence leads to a nonlinear current-voltage characteristic of the medium as a whole.

At a sufficiently large electric-field intensity $\Delta\varphi/l$ in the layer, the dependence of the local conductivity on the field can become appreciable. In this case we can no longer neglect the possible roughness of the faces of the layer—a protrusion on the base of the cylinder can lead to the appearance of a narrow channel[19] in which the current density is higher than in the rest of the layer. The change to a new mode of current flow leads to nonlinearity of the current-voltage characteristic as a whole. Pinching (contraction) of the current can also be due to another cause—a sufficiently strong dependence of the local conductivity on the temperature.[20–22] The current flow in a medium consisting of nonlinear conductors was studied in Ref. 23.

It is necessary to note that the model under consideration works below the percolation threshold ($p < p_c$) but outside the region of smearing,[5] in which, evidently, the medium has a substantially more complicated fractal structure.

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