A qualitative theory of the effective conductivity of polycrystals

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(Submitted 29 June 1982)
Zh. Eksp. Teor. Fiz. **84**, 1756–1760 (May 1983)

The asymptotic regime of current flow in a randomly inhomogeneous anisotropic medium (a polycrystal) characterized by a conductivity tensor with principal values $\sigma_1 \gg \sigma_2 \gg \sigma_3$ is considered. A qualitative theory based on an analysis of the geometrical properties of the high-conductivity paths is constructed, and the effective conductivity of a polycrystal is found.

PACS numbers: 72.10.Bg

The problem of determining the effective conductivity of a randomly inhomogeneous medium and similar problems (of effective diffusion, thermal conductivity, static permeability) arise in various physical situations, and have continued to attract attention for over a hundred years now.[1]

These problems are of special theoretical interest in those cases in which the medium is markedly inhomogeneous, and the qualitative characteristics of the current-flow pattern (for definiteness we shall speak in the language of the problem of effective conductivity) and the order of magnitude of the effective conductivity are not apparent beforehand. In the case in which the conductivity is a scalar, the progress made in the understanding of the qualitative characteristics of the current-flow pattern is connected with the concept of percolation level,[2] while the success achieved in the estimation of the effective conductivity is connected with the investigation of the statistics of the geometrical properties characterizing infinite clusters at the percolation level.[3] This has revealed a significant inherent similarity between percolation theory and the theory of second-order phase transitions, since underlying both of them are geometrical critical phenomena.

Geometrical concepts are also important in the problem, which we shall consider here, of the determination of the effective conductivity of a statistically isotropic polycrystal whose crystallites are characterized by a highly anisotropic conductivity tensor with principal values σ_1, σ_2, and σ_3, though these concepts are different from those used in percolation theory. Specifically, a decisive role is played by the investigation of the structure of the integral curves of the family of directions of high conductivity, or, as we shall term it, high-conductivity lines. This structure is studied with the aid of methods similar to those used in the qualitative theory of ordinary differential equations.

Although the ordinary methods of solving the problems of the effective conductivity of highly inhomogeneous media (in particular, polycrystals) are not well developed, the two-dimensional problem has been solved exactly: the solution, as noted in Ref. 4, where it was first given, is possible owing to the peculiar symmetry of the equations for the current density (div $\mathbf{j} = 0$) and the electric field (curl $\mathbf{E} = 0$). Specifically, if the principal values of the two-dimensional conductivity tensor are equal to σ_1 and σ_2, then $\sigma^{\text{eff}} = (\sigma_1 \sigma_2)^{1/2}$. The existence of the exact answer allows us to test the qualitative theory developed below on the two-dimensional case. Therefore, we shall first set forth the principal ideas as applied to the two-dimensional case before going on to consider the three-dimensional case.[1)]

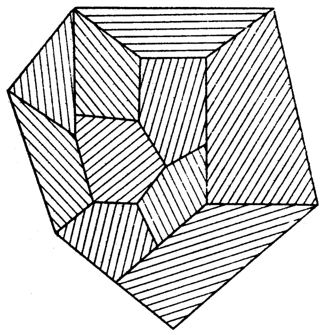

FIG. 1.

Let a plane be divided into two-dimensional crystallites, namely polygons bordering on each other along common sides (Fig. 1). Let the directions of high conductivity σ_1 be specified in each crystallite, and let the transverse direction be one of low conductivity σ_2 ($\sigma_1 \gg \sigma_2$). We shall assume that the situations in which the high-conductivity direction in a crystallite is parallel to one of its sides are degenerate, and we shall not consider them. In the nondegenerate case we can introduce the concept of high-conductivity lines: a polygonal line, each straight segment of which is parallel to the high conductivity direction of that crystallite in which that segment is located. In certain cases, if a straight segment of the polygonal line abuts against a vertex of the crystallite, the high conductivity line cannot be continued into the neighboring crystallites (or it can be continued, but nonuniquely). Except for these special cases, the high conductivity line can be continued from both ends. On the face of it, an ordinary high conductivity line bears a qualitative resemblance to a self-avoiding random-walk trajectory. But the high conductivity line, on getting into the region near the common vertex of three crystallites in which the directions of high conductivity intersect the sides converging at the vertex in question, winds around the vertex (Fig. 2). We shall say that such vertices are traps for the high conductivity lines. The property of a vertex's being a trap is a stable one (it does not disappear when the configuration is changed slightly), so that a finite fraction of all the vertices are traps. The "cross section for capture" by such a trap is of the order of the dimensions of a crystallite, and the probability of a high conductivity line eluding all the traps and going far off is exponentially small. Therefore, the typical asymptotic behavior of an ordinary high conductivity line is the winding

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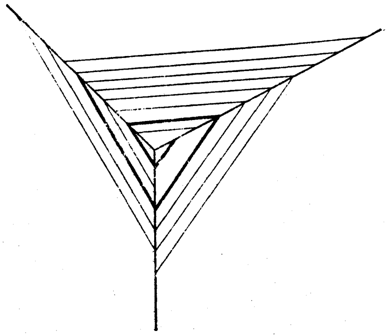

FIG. 2.

around a vertex trap (similar to the winding of a phase trajectory around a focus in the qualitative theory of smooth dynamical systems).

Another type of asymptotic behavior of high conductivity lines is their winding around a limit cycle—a closed high-conductivity line, see Fig. 3 (we classify it with the singular lines). In essence, the above-discussed cases exhaust the possible types of asymptotic behavior of high conductivity lines (in a plane).

Let us now introduce the concept of high conductivity bundles, by which we mean continuous groups of high conductivity lines having, in particular, identical asymptotic behavior (i.e., winding around one and the same focus or cycle) at both ends. The bundles border on each other along singular high-conductivity lines, as shown in Fig. 4.

Using the above-introduced geometrical forms, we can visualize the current flow pattern in a two-dimensional polycrystal in the case of extreme anisotropy as follows: the current flows largely along the bundles and the resistance, determined by the high conductivity σ_1, of the central part is small. As a bundle approaches a focus or a limit cycle, however, it narrows down, the current density increases, the resistance per unit length of the bundle rises, and, ultimately, the current overflows in the direction of low conductivity from one bundle to a neighboring bundle that winds around the same focus or limit cycle. In this case, because of the tapering of the bundles, the distance that the current has to get over in the direction of low conductivity is significantly smaller than the crystallite dimensions.

The foregoing can be formulated in the form of ideas about some equivalent network in which the good conductors (i.e., the central untapered parts of the bundles) are in contact with each other, forming a branched circuit in which the contact resistances are high, and, in the final analysis, determine the resistance of the circuit as a whole. Therefore, to determine the asymptotic dependence of σ^{eff} on σ_1 and σ_2

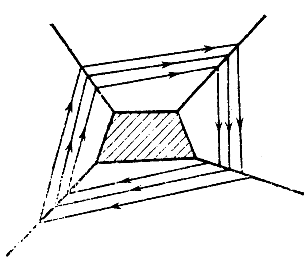

FIG. 3.

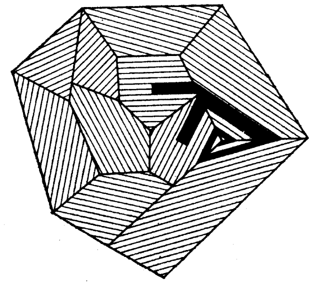

FIG. 4.

in the case when $\sigma_1/\sigma_2 \gg 1$, we must solve the problem of the determination of the asymptotic form of the resistance of a single typical contact. If we leave out the unimportant geometrical details, and spread out the bundles, we arrive at the situation depicted in Fig. 5a in the case of a contact at a focus and in Fig. 5b in the case of a contact on a limit cycle. Taking account of the spiral character of the winding of high conductivity lines around a focus or a limit cycle, we can take the dependence of the width h of a bundle on the distance l along the bundle in the form $h(l) = kl$ in the case of a focus and $h(l) = \mathrm{k}\exp(-l/l_0)$ in the case of a limit cycle. We can then easily estimate the contact resistance, using the variational principle (see Ref. 5) in the form

$$R_{\text{con}} I^2 = \inf_{\mathbf{j}} \left\{ \int_{s} \mathbf{j}\hat{\rho}\mathbf{j}\, dS \right\},$$

where the minimization is performed over all the $\mathbf{j}$ functions satisfying the conditions

$$\operatorname{div} \mathbf{j} = 0, \qquad \int \mathbf{j}\, d\mathbf{n} = I.$$

Hence for both a focus and a limit cycle we easily find that

$$R_{\text{con}} \sim c/(\sigma_1\sigma_2)^{1/2},$$

where the numerical factor c depends on the geometrical details. Since the resistances of all the contacts of the described equivalent network are of the same order of magnitude, and are proportional to $(\sigma_1\sigma_2)^{-1/2}$, the resistance of the entire network behaves similarly, i.e., the conductivity has the asymptotic form $\sigma^{\text{eff}} \approx (\sigma_1\sigma_2)^{1/2}$. It is impossible to determine the numerical factor in this formula with the aid of the above-expounded procedure. Clearly, the result obtained agrees with the exact expression for σ^{eff} (Ref. 4).

Besides allowing the derivation of the approximate formula for σ^{eff}, the foregoing analysis enables us to draw a conclusion about the highly inhomogeneous distribution of the Joule heat evolved during the flow of current in a polycrystal. Specifically, the major portion of the Joule heat will be evolved in that small part of the entire volume of the polycrystal which corresponds to the foci and limit cycles, where the overflow of the current from one bundle to another occurs; the ratio of this volume to the total volume is $\sim(\sigma_2/\sigma_1)^{1/2}$.

The three-dimensional case differs from the two-dimensional one in that it is characterized by a more complicated classification of the types of asymptotic behavior of the high conductivity lines and, consequently, of the types of electrical contacts between the bundles. Thus, instead of a single trap of the focus type, several types of traps are possible. The

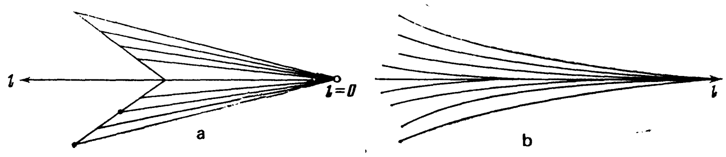

FIG. 5.

simplest of them is formed in the vicinity of an edge contact of three crystallites in which the high conductivity lines are such that their projections on a plane perpendicular to the edge under consideration form a two-dimensional focus trap. This means that a high conductivity line winds around the edge, as shown in Fig. 6. The electrical contact between two bundles that wind around the same edge can easily be estimated with the aid of the same variational principle. It turns out in this case that the resistance of such a contact (with $\sigma_1 \gg \sigma_2 \gg \sigma_3$) behaves asymptotically like $c(\sigma_1\sigma_2)^{-1/2}$, where σ_1 and σ_2 are the two leading—with respect to magnitude—principal values of the conductivity tensor. The constant c is determined by the geometry of the trap.

Certain other types of essentially three-dimensional traps that are formed at the junction of four crystallites and were investigated by us have a much higher resistance (of the order of $1/\sigma_2$) at a contact between bundles curling in them, which is due to a more rapid decrease of the cross-section area of the bundles as they curl in the essentially three-dimensional traps. We did not find electrical contacts with resistances that are asymptotically lower than $(\sigma_1\sigma_2)^{-1/2}$. If we assume that there are enough good contacts with resistances $\sim(\sigma_1\sigma_2)^{-1/2}$ to join the bundles into an infinite cluster (this seems to us to be likely, since the statistical weight of the local configurations that give rise to essentially three dimensional traps with high resistances is small), then we can in this case give an asymptotic estimate for the effective conductivity of a three-dimensional polycrystal:

$$\sigma^{eff} \sim (\sigma_1\sigma_2)^{1/2}.$$

It would be of interest to verify by computer simulation the assumption made above that the connectivity of the equivalent electrical circuit is due to the presence of edge traps.

It is worth noting that the self-consistent field method, which has, beginning with Maxwell, often been used to estimate the effective conductivity, and usually leads to qualitatively correct results,[6] yields for a polycrystal $\sigma^{eff} = (\sigma_1\sigma_2)^{1/2}$ in the two-dimensional case and $\sigma^{eff} \approx \frac{1}{2}(\sigma_1\sigma_2)^{1/2}$ in the three-dimensional case.

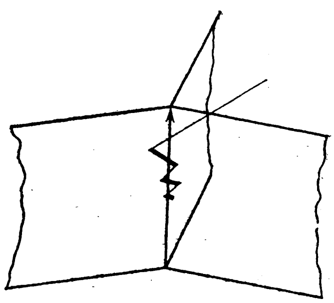

FIG. 6.

[1)] The point is not just that we do not know how to obtain the exact answer for σ^{eff} in the three-dimensional case, but also that the effective conductivity is not only a function of σ_1, σ_2, and σ_3, but depends further on the statistical properties of the ensemble of configurations corresponding to the mutual disposition and orientation of the individual crystallites in the polycrystal. The fact that in the two-dimensional case the answer does not depend on these properties (under the assumption that the configuration ensemble is statistically isotropic) is an exceptional property, which is all explained by the same symmetry of the equations.

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Translated by A. K. Agyei