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BASIC RESEARCH

Theoretical Treatment of the Conductivity of Textured Inhomogeneous Materials

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Abstract—A theory is developed for the conductivity of textured inhomogeneous media. In the context of the effective medium approach, the conductivity's tensor components for a polycrystalline medium are calculated as functions of the conductivity's tensor components for the constituent crystallites, the degree of ordering of the crystallites in orientation, and the orientation of the sample in the laboratory system of coordinates. Some recommendations are given regarding the experimental determination of the basic characteristics of the sample, including the statistical spread of directions of the crystallite axes with respect to the texture axis.

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Inhomogeneous materials, among which are composite materials (including nanocomposites) and polycrystalline compounds, are of considerable importance for electronics [1]. The electrical and optical properties of such materials have attracted the attention of researchers for almost a century and a half, and the pioneer investigators in this field were Maxwell, Rayleigh, and Bruggeman. The classical studies, in which the main versions of the effective medium approach were suggested for the first time, were reviewed in [2, 3]. These versions were combined and generalized by Stroud [4], who treated the problem of conductivity in inhomogeneous media in detail. The effective medium approach developed in [4] is applicable to various inhomogeneous media that present conglomerates of grains (crystallites) statistically different in volume (much larger than the atomic volume), form, and orientation; in general, the crystallites can differ also in chemical composition.

Until the present time, in a large variety of studies concerned with the problem of conductivity of inhomogeneous materials, consideration was given to media consisting of homogeneous isotropic components. Among these are matrices and differently-shaped inclusions that are regularly or randomly distributed in the volume and oriented in a common direction or uniformly distributed directions, layered media, regular or random in structure, and polycrystalline media with no texture, in which orientations of the constituent crystallites are distributed uniformly [4]. A comprehensive review of the corresponding studies can be found in [5, 6].

In this study, we consider a textured random polycrystalline medium, i.e., a medium, in which the constituent crystallites exhibit some prevailing orientation defined by the texture's axis. The purpose of the study is to calculate the effective conductivity tensor and to analyze how the tensor depends on the parameters and orientation distribution of the crystallites. The crystallites are assumed to be ellipsoidal single-axis grains of the same conductivity type; however, the final calculation is executed for spherical crystallites. The problem is solved on the basis of one of the modified versions of the effective medium approach [4], specifically, the method of self-consistent solution (in accordance with the terminology accepted in [5]). As in [4], we assume that the contacts between the crystallites are ohmic.

We consider a sample of an inhomogeneous conducting medium of volume V in the external electric field $\mathbf{E}_0$. Then, the effective conductivity tensor $\boldsymbol{\sigma}_\mathbf{e}$ for the medium is defined by the equation $\langle \mathbf{J} \rangle = \boldsymbol{\sigma}_\mathbf{e}\langle \mathbf{E} \rangle$, where $\mathbf{J}$ is the current density and $\langle \mathbf{E} \rangle = \mathbf{E}_0$. Representing the conductivity tensor of the medium at the point $\mathbf{x}$ as $\boldsymbol{\sigma}(\mathbf{x}) = \boldsymbol{\sigma}_\mathbf{e} + \delta\boldsymbol{\sigma}(\mathbf{x})$ and taking into account the relation $\mathbf{J}(\mathbf{x}) = \boldsymbol{\sigma}(\mathbf{x})\mathbf{E}(\mathbf{x})$, we can turn from the equations of electrostatics $\nabla \mathbf{J} = 0$ and $\nabla \times \mathbf{E} = 0$ to the boundary's value problem of determining the electrostatic potential $\Phi(\mathbf{x})$:

$$\begin{cases} \nabla \boldsymbol{\sigma}_\mathbf{e} \nabla \Phi(\mathbf{x}) = -\nabla \delta\boldsymbol{\sigma} \nabla \Phi(\mathbf{x}). & \mathbf{x} \in V, \\ \Phi(\mathbf{x}) = \Phi_0(\mathbf{x}) \equiv -\mathbf{E}_0 \mathbf{x}, & \mathbf{x} \in S. \end{cases}$$

Here, S is the surface area confining the volume V. With the Green's function $G(\mathbf{x}, \mathbf{x}')$ introduced through the conditions

$$\begin{cases} \nabla \boldsymbol{\sigma}_\mathbf{e} \nabla G(\mathbf{x}, \mathbf{x}') = -\delta(\mathbf{x} - \mathbf{x}'), & \mathbf{x}, \mathbf{x}' \in V, \\ G(\mathbf{x}, \mathbf{x}') = 0, & \mathbf{x}' \in S, \end{cases}$$

the potential $\Phi(\mathbf{x})$ can be represented as $\Phi(\mathbf{x}) = \Phi_0(\mathbf{x}) + \iiint_V G(\mathbf{x}, \mathbf{x}') \nabla' \delta\boldsymbol{\sigma}(\mathbf{x}') \nabla' \Phi(\mathbf{x}') d^3\mathbf{x}'$.

Let the crystallites be ellipsoids in shape. In this case, it is shown [4] that, if the tensor $\Gamma \equiv \Gamma_\mathbf{i}$ related to the ith crystallite is defined by the equality

$$\Gamma_i^{kl} = -\oint_{S_i'}\int \frac{\partial}{\partial x_k} G(\mathbf{x}, \mathbf{x}') n_l' d^2\mathbf{x}', \qquad (1)$$

where S_i' is the surface area of the ith crystallite, $\mathbf{n}'$ is the external normal surface S_i', we obtain the equation to determine the tensor $\boldsymbol{\sigma}_\mathbf{e}$:

$$\langle (\mathbf{I} - (\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e})\Gamma)^{-1} (\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e}) \rangle = 0. \qquad (2)$$

Averaging in (2) is over orientations of the crystallites in the coordinate system xyz associated with the texture's axis. The z axis is directed along the texture's axis; the x and y axes are orthogonal to the texture axis and to each other, otherwise being directed arbitrarily. In the calculation, we use also the two systems of coordinates, the laboratory system XYZ and the system $\xi\eta\zeta$ associated with a particular crystallite. The ζ axis is directed along the crystallite axis, and the directions of the ξ and η axes are chosen arbitrarily under the condition that these axes are orthogonal to the ζ axis and to each other. The orientation of the coordinate system $\xi\eta\zeta$ with respect to the system xyz (the matrix of rotation from xyz to $\xi\eta\zeta$) is denoted by $\mathbf{g}'(\psi', \vartheta', \varphi')$, where ψ', ϑ', and φ' are the Euler angles. The orientation of xyz with respect to XYZ is denoted by $\mathbf{G}(\Phi_0, \Theta_0, \Psi_0)$.

The distribution of orientations of the crystallites is assumed to be symmetric about the texture's axis; i.e., it is assumed that the distribution is taken as

$$\tilde{f}(\psi', \vartheta', \varphi') = \frac{1}{4\pi^2} f(\vartheta'),$$
$$0 \le \psi' < 2\pi, \quad 0 \le \vartheta' < \frac{\pi}{2}, \quad 0 \le \varphi' < 2\pi. \qquad (3)$$

Here, $f(\vartheta')$ is the density of the distribution of the angles ϑ' between the texture's axis and the crystallite's axes.

Let $\boldsymbol{\sigma}''$ and $\boldsymbol{\sigma}'$ be the matrices of the conductivity tensor of a crystallite in the systems $\xi\eta\zeta$ and xyz, respectively. Then, we have

$$\boldsymbol{\sigma}'' = \sigma_0 \begin{Vmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \alpha \end{Vmatrix}, \qquad (4)$$

$$\boldsymbol{\sigma}' = \sigma_0 \begin{Vmatrix} 1 + (\alpha - 1)\sin^2\vartheta'\sin^2\psi' & (1-\alpha)\sin^2\vartheta'\sin\psi'\cos\psi' & (\alpha-1)\sin\vartheta'\cos\vartheta'\sin\psi' \\ (1-\alpha)\sin^2\vartheta'\sin\psi'\cos\psi' & 1 + (\alpha-1)\sin^2\vartheta'\cos^2\psi' & (1-\alpha)\sin\vartheta'\cos\vartheta'\cos\psi' \\ (\alpha-1)\sin\vartheta'\cos\vartheta'\sin\psi' & (1-\alpha)\sin\vartheta'\cos\vartheta'\cos\psi' & 1 + (\alpha-1)\cos^2\vartheta' \end{Vmatrix}. \qquad (5)$$

Obviously, the tensor $\boldsymbol{\sigma}_\mathbf{e}'$ is in the form

$$\boldsymbol{\sigma}_\mathbf{e}' = \begin{Vmatrix} \sigma_e^{xx} & 0 & 0 \\ 0 & \sigma_e^{xx} & 0 \\ 0 & 0 & \sigma_e^{xx} \end{Vmatrix}. \qquad (6)$$

In the system xyz, the nonzero components of the tensor Γ are only the diagonal components. In the case of spherical crystallites, these diagonal components can be calculated by the formulas

$$\Gamma^{xx} = \Gamma^{yy} = \frac{1}{2\sqrt{\sigma_e^{xx}\sigma_e^{zz}\varepsilon}}\left(\sqrt{1-\varepsilon} - \frac{\arcsin\sqrt{\varepsilon}}{\sqrt{\varepsilon}}\right),$$
$$\Gamma^{zz} = -\frac{1}{\sigma_e^{zz}\varepsilon}\left(1 - \sqrt{1-\varepsilon}\frac{\arcsin\sqrt{\varepsilon}}{\sqrt{\varepsilon}}\right) \qquad (7a)$$

at $\varepsilon > 0$, where $\varepsilon = 1 - \sigma_e^{xx}/\sigma_e^{zz}$ (this is the case at $\alpha > 1$), [4] and

$$\Gamma^{xx} = \Gamma^{yy} = \frac{\sqrt{\varepsilon^2 - \varepsilon} - \ln(\sqrt{-\varepsilon} + \sqrt{1-\varepsilon})}{2\sqrt{\sigma_e^{xx}\sigma_e^{zz}\varepsilon\sqrt{-\varepsilon}}},$$
$$\Gamma^{zz} = -\frac{1}{\sigma_e^{zz}\varepsilon}\left(1 - \sqrt{1-\varepsilon}\frac{\ln(\sqrt{-\varepsilon} + \sqrt{1-\varepsilon})}{\sqrt{-\varepsilon}}\right), \qquad (7b)$$

at $\varepsilon < 0$ ($\alpha < 1$).

The dependence of the components Γ^{kl} on the coordinate $\mathbf{x}$ is lacking, since at $V \longrightarrow \infty$, $G(\mathbf{x}, \mathbf{x}') \longrightarrow G(|\mathbf{x} - \mathbf{x}'|)$, and the integrals in (1) are independent of $\mathbf{x}$ [4].

Substituting (5) and (6) in (2) and performing some calculations, we obtain the system of equations

$$\begin{cases} \int\limits_0^{\frac{\pi}{2}} \dfrac{(\alpha-1)\left[u_x(v_x - v_z) + \dfrac{v_z}{2\sigma_0\Gamma^{xx}}\right]\cos^2\vartheta' + v_z\left[\dfrac{\alpha-1}{2}(u_x + v_x) + u_x v_x\right]}{(\alpha-1)(v_x - v_z)\cos^2\vartheta' + v_z((\alpha-1) + v_x)} f(\vartheta')d\vartheta' = 0, \\ \int\limits_0^{\frac{\pi}{2}} \dfrac{(\alpha-1)(v_x - u_z)\cos^2\vartheta' + u_z((\alpha-1) + v_x)}{(\alpha-1)(v_x - v_z)\cos^2\vartheta' + v_z((\alpha-1) + v_x)} f(\vartheta')d\vartheta' = 0. \end{cases} \quad (8)$$

Here,

$$u_x = 1 - \frac{\sigma_e^{xx}}{\sigma_0}, \quad u_z = 1 - \frac{\sigma_e^{zz}}{\sigma_0},$$
$$v_x = u_x - \frac{1}{\sigma_0\Gamma^{xx}}, \quad v_z = u_z - \frac{1}{\sigma_0\Gamma^{zz}}. \quad (9)$$

Thus, the problem of determining the conductivity tensor for the effective medium is reduced to the system of equations (8) with the two unknowns σ_e^{xx} and σ_e^{zz}. It is easy to verify that the system (8) gives solutions coincident with the results obtained previously in [4] or expected from physical considerations in the following three cases: (1) the crystallites are isotropic in conductivity ($\alpha = 1$); (2) all crystallite axes are parallel to the texture's axis ($f(\vartheta') = \delta(\vartheta')$); and (3) the distribution of directions of the crystallite axes is uniform.

We search for the analytical expressions for the components of the tensor $\boldsymbol{\sigma}_\mathbf{e}$. Let us assume that the condition

$$\frac{\sigma^{ij} - (\sigma_e)^{ij}}{\sigma_0} \ll 1, \quad i, j = 1, 2, 3, \quad (10)$$

is satisfied; i.e., the components of the tensor $\frac{1}{\sigma_0}(\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e})$ are small. Then, we have $(\mathbf{I} - (\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e})\boldsymbol{\Gamma})^{-1} \approx \mathbf{I} + (\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e})\boldsymbol{\Gamma}$, and equation (2) takes the form

$$\langle \boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e} \rangle + \langle (\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e})\boldsymbol{\Gamma}(\boldsymbol{\sigma} - \boldsymbol{\sigma}_\mathbf{e}) \rangle = 0. \quad (11)$$

It is easy to verify that, for nondiagonal elements, the tensor equation (11) is satisfied automatically and, for the elements $(\ )^{11}$ and $(\ )^{22}$, equations are the same; therefore, equation (11) is reduced to the system of two equations in u_x and u_z,

$$\begin{cases} (1 - I_1) + \dfrac{u_x}{\beta} + \sigma_0\beta\Gamma^{xx}\left[2(1 - 2I_1 + I_2) + 2\gamma(I_1 - I_2) + 2(1 - I_1)\dfrac{u_x}{\beta} + \left(\dfrac{u_x}{\beta}\right)^2\right] = 0, \\ 2I_1 + \dfrac{u_z}{\beta} + \sigma_0\beta\Gamma^{xx}\left[4(I_1 - I_2) + 4\gamma I_2 + 4\gamma I_1\dfrac{u_z}{\beta} + \gamma\left(\dfrac{u_z}{\beta}\right)^2\right] = 0, \end{cases} \quad (12)$$

where

$$\gamma = \frac{\Gamma^{zz}}{\Gamma^{xx}}, \quad \beta = \frac{\alpha - 1}{2}, \quad I_1 = \int\limits_0^{\frac{\pi}{2}} \cos^2\vartheta' f(\vartheta')d\vartheta',$$
$$I_2 = \int\limits_0^{\frac{\pi}{2}} \cos^4\vartheta' f(\vartheta')d\vartheta'. \quad (13)$$

We now consider two cases, when inequalities (10) are satisfied.

Case 1. If $|\alpha - 1| \ll 1$, i.e., the crystallites are slightly anisotropic, we have $|u_x| \ll 1$, $|u_z| \ll 1$, $|\varepsilon| \ll 1$, $\Gamma^{xx} \approx \Gamma^{zz} \approx -\frac{1}{3\sigma_0}$, and $\gamma \approx 1$. Substituting these relations in (12), from the resulting equations, we find the components of the tensor $\boldsymbol{\sigma}'_\mathbf{e}$ to the second-order approximation:

$$\begin{cases} \sigma_e^{xx} = \sigma_0\left[1 + \dfrac{1}{2}(\alpha - 1)(1 - I_1) - \dfrac{1}{12}(\alpha - 1)^2(1 - I_1^2)\right], \\ \sigma_e^{zz} = \sigma_0\left[1 + (\alpha - 1)I_1 - \dfrac{1}{3}(\alpha - 1)^2 I_1(1 - I_1)\right]. \end{cases} \quad (14)$$

As evident from (14), the components of the tensor $\boldsymbol{\sigma}'_e$ depend both on the parameters defining the conductivity tensors of individual crystallites (σ_0, α) and on the distribution of orientations of the crystallites; in (14), the last-mentioned dependence is expressed in terms of the integral I_1. Let us assume that the directing vector $\mathbf{k}'$ of the crystallite's principal axis is characterized by the Beltrami coordinates μ, ν normally distributed with the zero mathematical expectation and the dispersion s^2 (μ, ν are the coordinates of the point of intersection of the line continuing the vector $\mathbf{k}'$ with the plane tangent to the unit hemisphere at its pole [7]); i.e.,

$$\hat{f}(\mu, \nu) = \frac{1}{2\pi s^2}\exp\left(-\frac{\mu^2+\nu^2}{2s^2}\right). \quad (15)$$

Then, for the density of distribution of the angles ϑ' of the texture's axis with the crystallite's axes, we obtain

$$f(\vartheta') = \frac{1}{s^2}\frac{\tan\vartheta'}{\cos^2\vartheta'}\exp\left(-\frac{\tan^2\vartheta'}{2s^2}\right), \quad 0 \le \vartheta' \le \frac{\pi}{2}.$$

Then,

$$I_1 = -\frac{1}{2s^2}\exp\left(\frac{1}{2s^2}\right)\mathrm{Ei}\left(-\frac{1}{2s^2}\right), \quad (16)$$

where $\mathrm{Ei}(x) = \int_{-\infty}^{x}\frac{e^t}{t}dt$ is the integral's exponential function [8].

Case 2. If the crystallites have a small spread in orientation, i.e., if the dispersion s^2 is small, we have $\sigma_e^{xx} \approx \sigma_0$, $\sigma_e^{zz} \approx \alpha\sigma_0$, $|u_x| \ll 1$, and $u_z = 1 - \alpha + \alpha\delta_z$, where δ_z is of the same order of smallness as u_x and s^2. In calculating the integrals I_1 and I_2 at the small dispersion, the normal distribution can be approximated with a quadratic distribution with the same dispersion; therefore, for the density of distribution of the coordinates μ, ν, we can take, instead of (15), the expression

$$\hat{f}(\mu, \nu) = \begin{cases} \frac{1}{3\pi s^2}\left(1 - \frac{\mu^2+\nu^2}{6s^2}\right), & \mu^2+\nu^2 \le 6s^2, \\ 0, & \mu^2+\nu^2 > 6s^2. \end{cases}$$

Then,

$$f(\vartheta') = \begin{cases} \frac{2}{3s^2}\left(1 - \frac{\tan^2\vartheta'}{6s^2}\right)\frac{\tan\vartheta'}{\cos^2\vartheta'}, \\ \vartheta' \in [0, \arctan\sqrt{6}s], \\ 0, \quad \vartheta' > \arctan\sqrt{6}s, \end{cases}$$

and for the integrals I_1 and I_2, we obtain

$$I_1 \approx 1 - 2s^2, \; I_2 \approx 1 - 4s^2. \quad (17)$$

Linearizing Eq. (12) in u_x and δ_z and taking into account (17), we obtain

$$\begin{cases} \sigma_e^{xx} = \sigma_0\left(1 + \sqrt{\alpha-1}\arcsin\sqrt{\frac{\alpha-1}{\alpha}}s^2\right) \\ \sigma_e^{zz} = \sigma_0\left[\alpha - (\alpha-1)\left(1 + \frac{\alpha}{\sqrt{\alpha-1}}\arcsin\sqrt{\frac{\alpha-1}{\alpha}}\right)s^2\right], \end{cases} \quad (18a)$$

at $\alpha > 1$,

and

$$\begin{cases} \sigma_e^{xx} = \sigma_0\left(1 - \sqrt{1-\alpha}\ln\left(\frac{1+\sqrt{1-\alpha}}{\sqrt{\alpha}}\right)s^2\right) \\ \sigma_e^{zz} = \sigma_0\left[\alpha + (1-\alpha)\left(1 + \frac{\alpha}{\sqrt{1-\alpha}}\ln\left(\frac{1+\sqrt{1-\alpha}}{\sqrt{\alpha}}\right)\right)s^2\right], \end{cases} \quad (18b)$$

at $\alpha < 1$.

Now we determined the matrix of the conductivity tensor $\boldsymbol{\sigma}_e$ for the effective medium in the laboratory coordinate system XYZ. We rewrite $\boldsymbol{\sigma}'_e$ as $\boldsymbol{\sigma}'_e = \sigma_e^{xx}\left\|\begin{matrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \alpha_e \end{matrix}\right\|$, where $\alpha_e = \frac{\sigma_e^{zz}}{\sigma_e^{xx}}$. Then, similar to (5), we obtain (with the notation $\beta_e = \frac{\alpha_e - 1}{2}$ introduced for brevity)

$$\boldsymbol{\sigma}_e = \sigma_e^{xx}\left\|\begin{matrix} 1 + 2\beta_e\sin^2\Theta_0\sin^2\Phi_0 & -\beta_e\sin^2\Theta_0\sin2\Phi_0 & \beta_e\sin2\Theta_0\sin\Phi_0 \\ -\beta_e\sin^2\Theta_0\sin2\Phi_0 & 1 + 2\beta_e\sin^2\Theta_0\cos^2\Phi_0 & -\beta_e\sin2\Theta_0\cos\Phi_0 \\ \beta_e\sin2\Theta_0\sin\Phi_0 & -\beta_e\sin2\Theta_0\cos\Phi_0 & 1 + 2\beta_e\cos^2\Theta_0 \end{matrix}\right\|. \quad (19)$$

To the first-order approximation, the values of σ_e^{xx} and β_e are determined from the expressions given below.

In case 1 (the crystallites are slightly anisotropic, $|\alpha - 1| \ll 1$), we have

$$\sigma_e^{xx} \approx \sigma_0\left[1 + \frac{1}{2}(\alpha - 1)(1 - I_1)\right],$$

$$\beta_e \approx \frac{\alpha - 1}{4}(3I_1 - 1),$$

where I_1 is determined from (16).

In case 2 (the dispersion of crystallite orientations is small, $s^2 \ll 1$), σ_e^{xx} is determined from (18a) and $\beta_e = \frac{\alpha-1}{2}\left(1 - s^2\left[1 + \frac{\alpha+1}{\sqrt{\alpha-1}}\arcsin\sqrt{\frac{\alpha-1}{\alpha}}\right]\right)$ at $\alpha > 1$ and σ_e^{xx} is determined from (18b) and $\beta_e = \frac{\alpha-1}{2}\left(1 - s^2\left[1 + \frac{\alpha+1}{\sqrt{1-\alpha}}\ln\left(\frac{1+\sqrt{1-\alpha}}{\sqrt{\alpha}}\right)\right]\right)$ at $\alpha < 1$.

Expressions (14), (18a), and (18b) give the solution of the direct problem in an explicit form (in the above-considered special cases): the components of the effective conductivity tensor of a textured polycrystalline medium is calculated in relation to the parameters σ_0 and α characterizing the conductivity tensor for an individual crystallite and to the density of distribution of directions of the crystallite's axes about the texture's axis. The same expressions as (14), (18a), and (18b) can be used to solve the inverse problem, i.e., the problem of estimating the parameter s^2 defining the statistical spread of directions of the crystallite's axes about the texture's axis. According to (19), the effective conductivity tensor $\boldsymbol{\sigma}_\mathbf{e}$ in the laboratory system is symmetric. Thus, measuring six components of the tensor $\boldsymbol{\sigma}_\mathbf{e}$, e.g., σ_e^{XX}, σ_e^{YY}, σ_e^{ZZ}, σ_e^{XY}, σ_e^{XZ}, and σ_e^{YZ}, one can determine the quantities σ_e^{xx} and σ_e^{zz} that define the effective conductivity tensor in the system associated with the texture; then, using (14), (18a), and (18b), one can estimate the parameter s^2 of the orientation distribution of crystallites.

It should be noted that the system of equations (8) gives the solution of the problem in the general case. In fact, if the density of orientation distribution of crystallites is known, the problem can be solved by numerical methods. In addition, the system of equations (8) can be used to derive an analytical solution of the problem in some other special cases, e.g., in the case of a slightly nonuniform distribution of crystallite orientations.

The model treated in this study can serve as one of the starting points for the development of the conductivity theory for more complex materials (nanocomposites) and stimulate more comprehensive experimental investigations of the electric properties of polycrystalline materials.

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