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Journal of the European Ceramic Society 24 (2004) 1893–1896

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Piezoresistive anisotropy of thick-film resistors

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Abstract

Numerous evidence suggests that thick-film resistors are close to a metal–insulator transition and that tunneling processes between metallic grains are the main source of resistance. By theoretical analysis of various percolative resistor network models we show that the piezoresistive response under imposed uniaxial strain is strongly dependent on the concentration of the conducting phase. In particular, the piezoresistive anisotropy is reduced as the system approaches its percolation threshold, following a power law in the critical region. We propose a simple relation between the conductance and the piezoresistive anisotropy valid in the critical region.

Keywords: Electrical conductivity; Nanocomposites; Sensors

1. Introduction

Transport properties of thick-film resistors (TFRs) are characterized by percolating behavior as a function of the metallic volume fraction and large strain sensitivity of conductance.[1,2] Together with their rather weak temperature dependence of transport, and high stability, these systems are successfully used in sensor applications such as pressure and force measuring devices.[2,3] An important aspect of TFRs properties of transport is identified in their microstructure, characterized by two phases, one conducting and the other insulating. In commercial TFRs, the conducting phase is constituted by RuO_2, or other Ru- or Ir-based oxides, grains which are embedded in a glassy matrix, i.e., the insulating phase. The space in between two neighboring metallic grains is usually a modified glass, resulting from the diffusion of nanosized RuO_2 clusters inside the original glass taking place during the firing process.[4]

Typical values of the piezoresistive gauge factor (GF) range from GF≈5 up to GF≈30, with little dependence on the direction of the external voltage drop with respect to the applied strain direction for the samples with higher GF values.[5] Here we point out that such quasi-isotropy of the piezoresistive response is an important element in investigating the role of microstructure in the transport of carriers in TFRs. We compute the piezoresistive response of different random resistor network models of TFRs as a function of site concentrations. We show that as the percolation threshold is approached from above, the piezoresistive anisotropy goes to zero by following an universal behavior governed by a power law. Our prediction could be easily experimentally tested, and could bring important informations on the topology of the percolative network and the critical point of TFRs.

2. Model

The large values of piezoresistance in TFRs are clear indications that tunneling between metallic grains or clusters is the main mechanism of transport. Hence, to model the piezoresistive behavior of TFRs, or of more general tunnelling–percolating systems, we consider a three-dimensional cubic random-resistor network whose bond conductances either zero or proportional to a tunneling exponential factor $\exp(-2d_i/\xi)$, where d_i is the tunneling distance between two neighbouring sites along the i direction and ξ the localization length, i.e., the linear size of electron localization.[6] In our model, the network bond directions are parallel to the orthogonal x, y, z axes (see Fig. 1 for a pictorial representation of a two-dimensional square lattice network), and in the

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doi:10.1016/S0955-2219(03)00466-7

absence of applied strains the tunneling distances are all equal: $d_i = d$ ($i = x,y,z$). In this situation, the network is isotropic and the total conductance $G_i = G$ depends only on the specific bond conductance distribution. To study the effect of an applied strain, let us assume that the resistor network is embedded in an homogeneous elastic medium and that the elastic coefficients of the network and the medium are equal. Hence, when a strain field ε_{ii} ($i = x,y,z$) is applied to the sample, the conductance changes to $G_i = G + \delta G_i$, where the variation δG_i can be expressed in terms of the (intrinsic) conductivity variation $\delta\sigma_i$ and a geometric (extrinsic) factor:[7]

$$\frac{\delta G_i}{G} = \frac{\delta\sigma_i}{\sigma} - \varepsilon_{ii} + \varepsilon_{jj} + \varepsilon_{kk} \tag{1}$$

where σ is the unstrained conductivity and the indexes i, j, k assume the values x, y, and z with cyclic permutations. In the above equation, the variation $\delta\sigma_i$ is due to the tunneling distance change $d \rightarrow d_i = d(1 + \varepsilon_{ii})$ which leads to $\exp(-2d/\xi) \rightarrow \exp(-2d/\xi)(1\text{-}2d\varepsilon_{ii}/\xi_i)$ for a conducting bond along the i direction. To express $\delta\sigma_i/\sigma$ as a function of a general ε_{ii}, let us first imagine that an uniaxial strain along the x direction ($\varepsilon_{xx} = \varepsilon$, $\varepsilon_{yy} = 0$, $\varepsilon_{zz} = 0$) is applied to the sample. In this situation, the conductivity along the x axis will in general be different from those along the y and the z axes which, by symmetry, are instead equal. Therefore, up to linear terms in ε, $\sigma_x = \sigma - \sigma\Gamma_\parallel\varepsilon$ and $\sigma_x = \sigma - \sigma\Gamma_\perp\varepsilon$, where

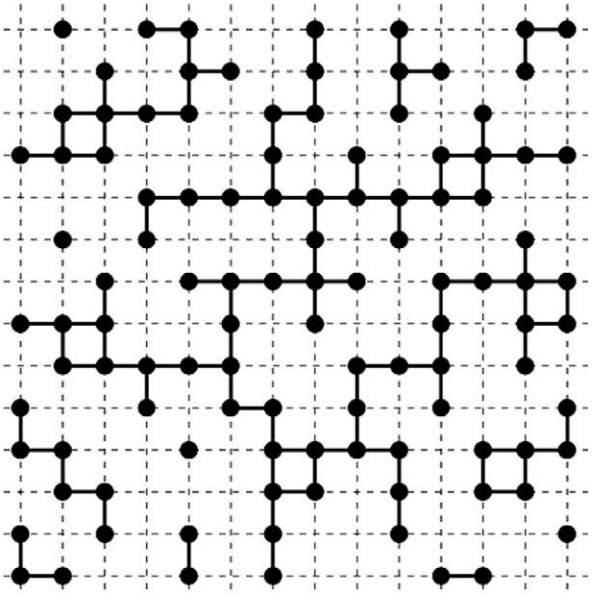

Fig. 1. Site-percolation model for a two-dimensional square lattice with a concentration x of sites (solid circles). Bonds connecting two adjacent sites have finite conductance $\exp(-2d/\xi)$ (thick solid segments). The remaining bonds have zero conductance (thin dashed segments).

$$\Gamma_\parallel = -\frac{\delta\sigma_x}{\varepsilon\sigma} \tag{2}$$

$$\Gamma_\perp = -\frac{\delta\sigma_y}{\varepsilon\sigma} = -\frac{\delta\sigma_z}{\varepsilon\sigma} \tag{3}$$

are the longitudinal and transverse piezoresistive coefficients, respectively. The above reasoning can be repeated for uniaxial strains along the y and the z axes and, since the problem is linear, for a general strain field ε_{ii} the conductivity variations $\delta\sigma_i$ reduce to:[8]

$$\frac{\delta\sigma_i}{\sigma} = -\Gamma_\parallel\varepsilon_{ii} - \Gamma_\perp\left(\varepsilon_{jj} + \varepsilon_{kk}\right) \tag{4}$$

The above equation permits to express all the piezoresistive coefficients $\Gamma_{ij} = \sigma\delta_i/\left(\varepsilon_{jj}\sigma\right)$ or, by using Eq.(1), the corresponding piezoresistive gauge factors $\mathrm{K}_{ij} = -\delta G_i/\left(\varepsilon_{jj}G\right)$ (that are the commonly measured quantities) in terms of $\Gamma_\parallel$ and $\Gamma_\perp$. For example, in a typical experiment with a thick cantilever beam having the main axis directed along the x direction, the strains are approximatively $\varepsilon_{xx} = \varepsilon$, $\varepsilon_{yy} = -\nu\varepsilon$, and $\varepsilon_{zz} = -\nu'\varepsilon$, where ν and ν' are the Poisson ratios of the cantilever and the resistive sample, respectively.[2,7] By using Eqs.(1) and (4), the longitudinal (K_L) and transverse (K_T) piezoresistive gauge factors are:

$$K_L \equiv K_{xx} = \left(1 + \Gamma_\parallel\right) + (1 - \Gamma_\perp)(\upsilon + \upsilon') \tag{5}$$

$$K_T \equiv K_{yx} = -\left(1 + \Gamma_\parallel\right)\upsilon - (1 - \Gamma_\perp)(1 - \upsilon') \tag{6}$$

Hence from a measurement of K_L and K_T it is possible to extract the intrinsic piezoresistive coefficients $\Gamma_\parallel$ and $\Gamma_\perp$. As we are going to show in the next section, the importance of $\Gamma_\parallel$ and $\Gamma_\perp$ resides on the fact that they permit to extract useful informations on the percolative nature of transport.

3. Results

We show in Fig. 2 the results of numerical Monte Carlo calculations of $\Gamma_\parallel$ and $\Gamma_\perp$ for a site percolation model in which a concentration of site x is removed at random (filled circles). For each missing site the bond conductances connecting the six neighboring sites are set equal to zero, while the remaining bonds have conductance $\exp(-2d/\xi)$ with $2d/\xi = 4$ when unstrained. Hence, a given bond has finite conductance only when two adjacent sites are present (see Fig. 1 for a two-dimensional case). We calculate the total conductance by solving numerically the Kirchhoff equations for all the nodes of the cubic network. In practice, we impose a unit voltage difference between two opposite sides of the network with periodic boundary conditions to the

remaining sides. The piezoresistive coefficient $\Gamma_{\parallel}$ ($\Gamma_{\perp}$) is then obtained by calculating, for a fixed configurations of bond resistors, the difference in conductance when $\varepsilon = 0$ and $\varepsilon = 0.01$ for strain directed parallel (orthogonal) to the imposed voltage drop direction.

When there are no missing sites ($x = 1$, $p = 1$), the current flows exclusively along paths directed along the direction of the voltage drop. In this case, the longitudinal piezoresistive coefficient is $\Gamma_{\parallel} = 2d/\xi = 4$ while the transverse one $\Gamma_{\perp}$ is zero [see Eqs.(2) and (3)]. When sites are removed ($x < 1$), $\Gamma_{\parallel}$ gets reduced and at the same time the transverse coefficient is enhanced in such a way that $\Gamma_{\parallel} > \Gamma_{\perp}$. This behavior is due to the fact that as sites are removed from the network, the missing bonds force the current to flow also along directions perpendicular to the voltage drop (see Fig. 1). Hence, when the strain is along the voltage drop, the current visits also regions where instead the bonds are unstrained (reduction of $\Gamma_{\parallel}$) while when the voltage drop is perpendicular to the direction of ε, the current is influenced also by regions where the bonds are strained (enhancement of $\Gamma_{\perp}$). This trend gets amplified as x is further reduced and at the percolation threshold ($p_c = x_c^2 \approx 0.098$) $\Gamma_{\perp} \rightarrow \Gamma_{\parallel}$. Qualitatively, this behavior is observed also when the bond conductances have some distribution or when different statistics like that of bond percolation models are considered. For example, the open symbols in Fig. 2 are the results of a random resistor network having an uniform distribution of the tunneling exponents ($2 \leqslant 2d/\xi \leqslant 6$). Hence, as a general rule, the longitudinal and transverse piezoresistive coefficients become equal as the random resistor network reaches its percolation threshold. This behavior is better analyzed in Fig. 3 where we plot the piezoresistive anisotropy factor $(\Gamma_{\parallel} - \Gamma_{\perp})/\Gamma_{\parallel}$ obtained from the data of Fig. 2 as a function of $p - p_c$. For both site percolation models with (solid symbols) or without (open symbols) distribution of $2d/\xi$ values, the anisotropy factor monotonically goes to zero as $p \rightarrow p_c$ and in the critical region $|p - p_c| \ll 1$ it follows a power law behavior:

$$\frac{\Gamma_{\parallel} - \Gamma_{\perp}}{\Gamma_{\parallel}} \approx (p - p_c)^{\lambda} \tag{7}$$

This specific power law dependence is made more clear in the inset of Fig. 3 where the data are plotted in a log–log scale. A fit of the numerical data to Eq. (7) leads to $\lambda = 0.44 \pm 0.07$ (solid symbols: $2d/\xi = 4$) and $\lambda = 0.4 \pm 0.2$ (open symbols: $2 \leqslant 2d/\xi \leqslant 4$). The power law behavior of Eq. (7) can also be inferred from the transport properties of anisotropic bond percolation models studied some time ago.[9–13] In those works, percolating networks with random bond conductances were defined in such a way that the conducting bonds were equal along two directions, for example, y and z, but different from those along the third direction, that is x. Topological considerations,[9] renormalization group analysis,[10] and numerical calculations,[11–13] showed that the quantity $1 - \sigma_y/\sigma_x = \alpha(p)$ has a power law behavior $\alpha(p) \approx (p - p_c)^{\lambda'}$ close to the percolation threshold, independently of the microscopic bond anisotropy.[10] In our model, bond anisotropy is induced by the effect of an applied strain, so that the two situations are equivalent. In fact, by neglecting the geometrical factors, it is easy to see from Eqs. (2) and (3) that in our piezoresistive model $1 - \sigma_y/\sigma_x \approx \varepsilon(\Gamma_{\parallel} - \Gamma_{\perp})$, so that the two exponents λ and λ' are equal.

Eq. (7) suggests a possible experimental route to characterize transport properties in TFRs or other

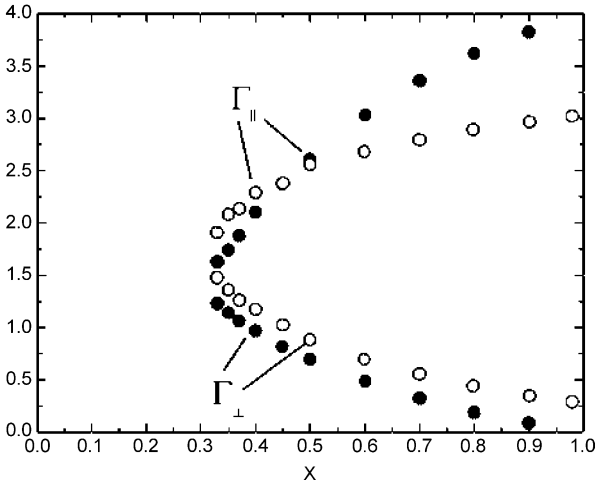

Fig. 2. Monte Carlo calculations of longitudinal ($\Gamma_{\parallel}$) and transverse ($\Gamma_{\perp}$) piezoresistive coefficients of a site percolation cubic network. The calculations have been performed for networks of N^3 sites with N up to $N = 40$ by numerically solving the Kirchhoff node equations. Filled symbols refer to bond conductances with tunneling exponent $2d/\xi = 4$, while open symbols are the results of uniform random distribution $2 \leqslant 2d/\xi \leqslant 6$. Piezoresistivity isotropy is reached at the critical site concentration $x = x_c \approx 0.314$ (which corresponds to the critical bond concentration $p_c = x_c^2 \approx 0.098$).

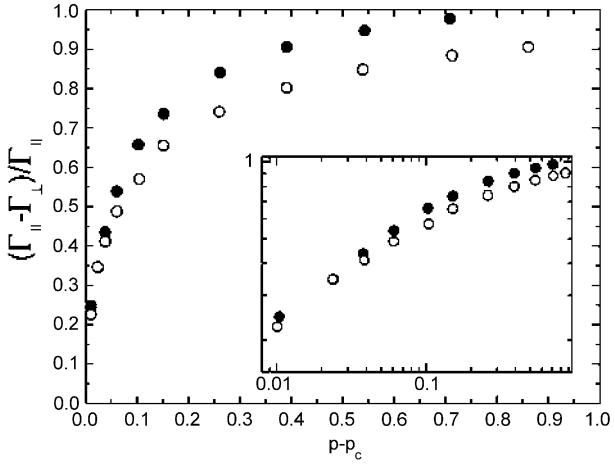

Fig. 3. Piezoresistive anisotropy factor as a function of $p - p_c$ for the site percolation data of Fig. 1. In the inset the same data of the main figure are plotted in a log–log scale.

piezoresistive granular compounds. In fact, as pointed out in the introduction, typical TFRs show a power law behavior of the conductance G as a function of metallic concentration.[1] Hence, we predict that if this behavior is due to the closeness to a percolative critical point, then the piezoresistive anisotropy factor should follow Eq. (7) or equivalently:

$$G \approx \left(\frac{\Gamma_{\parallel} - \Gamma_{\perp}}{\Gamma_{\parallel}}\right)^{\lambda/t} \tag{8}$$

where t is the critical exponent of the unstrained conductance $G \approx (p - p_c)^t$. There is evidence that the piezoresistive gauge factors anisotropy $(K_L - K_T)/K_L$ in some commercial TFRs is lower for higher resistive samples,[5] in qualitative accord therefore with Eq. (8).

The critical behavior of the piezoresistive anisotropy factor is a robust feature which we have tested to survive also in more general models. For example, by allowing bonds to have random orientations with respect to the applied strain, it is possible to show that the longitudinal and transverse piezoresistive coefficients reduce to $\Gamma_{\parallel}^* = (3\Gamma_{\parallel} + 2\Gamma_{\perp})/5$ and $\Gamma_{\perp}^* = (\Gamma_{\parallel} + 4\Gamma_{\perp})/5$, where $\Gamma_{\parallel}$ and $\Gamma_{\perp}$ are the corresponding coefficents for the ordered bond network.[8] Hence, close to the percolation threshold, the new effective piezoresistive anisotropy factor $(\Gamma_{\parallel}^* - \Gamma_{\perp}^*)/\Gamma_{\parallel}^*$ is proportional to $(p - p_c)^{\lambda}$ just as found for a regular lattice.

4. Conclusions

In summary, we have shown how to extract the intrinsic pietoresistive responses $\Gamma_{\parallel}$ and $\Gamma_{\perp}$ of a TFR or other tunnelling–percolating systems from the knowledge of the gauge factors and the values of applied strains. The so-obtained longitudinal and transverse coefficients permit to define the piezoresistive anisotropy factor $(\Gamma_{\parallel} - \Gamma_{\perp})/\Gamma_{\parallel}$ which has a power law behavior as a function of the metallic volume fraction if the system is sufficiently close to the percolation threshold. The power law behavior of $(\Gamma_{\parallel} - \Gamma_{\perp})/\Gamma_{\parallel}$ indicates criticality of the piezoresistive response, and it could be a useful tool to locate the metal–insulator transition in TFRs. We would like also to stress that the measurement of the piezoresistive anisotropy is an alternative and, in principle, more practical way to study the role of transport anisotropy of disordered materials, since anisotropy can be externally controlled by an applied strain rather being given fixed by intrinsic material properties.

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