Piezoresistive anisotropy of percolative granular metals

C. Grimaldi\textsuperscript{1}, P. Ryser\textsuperscript{1}, and S. Strässler\textsuperscript{1,2}

\textsuperscript{1} Institut de Production et Robotique, LPM, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland
\textsuperscript{2} Sensile Technologies SA, PSE, CH-1015 Lausanne, Switzerland

The piezoresistive response of granular metals under uniaxial strain is strongly dependent on the concentration of the conducting phase. Here we show that the piezoresistive anisotropy is reduced as the system approaches its percolation threshold, following a power law behavior in the critical region. We propose a simple relation between the conductance and the piezoresistive anisotropy which could be used in relation to real materials and notably to the thick film resistors. PACS numbers: 72.20.Fr, 72.60.+g, 72.80.Ng

Disordered systems constituted by random dispersions of metallic and insulating phases, also commonly referred to as granular metals, show transport properties strongly dependent on the concentration $x$ of the metallic phase in the composite. For $x$ larger than a critical concentration $x_c$, transport is metallic-like while for $x \rightarrow x_c$ the system undergoes a metal-to-insulator transition. In the critical region $(x - x_c) \ll 1$, the conductance $G$ follows a power-law behavior $G \sim (x - x_c)^{t}$ where $t$ is the critical exponent. The minimal model of a theoretical description of transport in granular metals is based on extensions of percolation theory to random-resistors network models. Numerical simulations of such percolative resistors show that as $x$ approaches $x_c$ from above, fluctuations of the microscopic (bond) currents get enhanced. It has been shown recently that this phenomenon can be spoiled to get more accurate experimental determinations of $x_c$ compared to common conductance measurements.

The enhancement of bond current fluctuations is basically also at the origin of the peculiar behavior of the piezoresistance effect as $x \rightarrow x_c$. Under an uniaxial imposed strain, in fact, the piezoresistive response is highly anisotropic when $x \simeq 1$, being maximum for external electric fields applied along the direction of the imposed strain (longitudinal response) and minimum along the orthogonal directions (transverse response). This is due to the fact that for $x \simeq 1$ the microscopic bond currents are basically all in the direction of the external electric field. Instead, lower values of $x$ force the bond currents to have components also along directions perpendicular to the external fields so that the anisotropy of the piezoresistive response is gradually reduced until at $x = x_c$ the longitudinal and transverse responses become equal. Hence, at the percolation threshold the piezoresistive effect is perfectly isotropic.

The transport properties of granular metals under uniaxial strain could in principle help to characterize the vicinity to the percolation threshold with an higher accuracy compared to that typical of common conductance measurements. The interest on the piezoresistive response of granular metals however concerns also more applicative problems. In fact, a particular class of granular metals made of RuO$_2$, IrO$_2$ or other metal-oxide granules embedded in a glassy matrix, also commonly known as thick-film resistors (TFRs), are characterized by a quite large piezoresistive response, which is used in fabricating highly sensitive pressure and force sensors. It is thought that intergrain tunneling processes are responsible for the TFRs high strain sensibility. A characteristic common to most of the TFRs is that their piezoresistive response is quasi-isotropic, indicating that the values and directions of the microscopic currents are highly fluctuating functions and that the TFRs are quite close to the percolation threshold. This is confirmed also by the observation of power-law dependence of $G$ upon RuO$_2$ concentrations.

In this communication, we compute the piezoresistive response of different random resistor network models as a function of bond or site concentrations. We show that close to the percolation threshold, the piezoresistive anisotropy follows an universal behavior governed by an exponential 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.

To model the piezoresistive behavior of TFRs, or more generally of percolative granular metals, 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 neighboring sites along the $i$ direction and $\xi$ the localization length. In our model, the network bond directions are parallel to the orthogonal $x, y, z$ axes, and in the 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 $\varepsilon_{ii} (i = x, y, z)$ is applied to the sample, the conductance changes to $G_i = G + \delta G_i$, where the variation $\delta G_i$ can be expressed in terms of the (intrinsic) conductivity varia-
FIG. 1. Monte Carlo calculations of longitudinal (Γ∥) and transverse (Γ⊥) piezoresistive coefficients of bond percolation (a) and site percolation (b) cubic network models. The calculations have been performed for networks of L^3 sites with L = 30 and L = 40 by numerically solving iteratively the Kirchhoff node equations (for more details see Ref. [3]). Filled symbols refer to bond conductances with tunneling exponent 2d/ξ = 4, while open symbols are the results of uniform random distribution 2 ≤ 2d/ξ ≤ 6. In (a), Γ∥ and Γ⊥ become equal at the critical bond concentration p = p_c ≃ 0.248. In (b), piezoresistive isotropy is reached at the critical site concentration x = x_c ≃ 0.314 (which corresponds to p_c = x_c^2 ≃ 0.098).

The above equation permits to express all the piezoresistive coefficients Γ_{ij} = −δσ_i/ε_{jj}σ or, by using Eq. (3), the corresponding piezoresistive gauge factors K_{ij} = −δG_i/ε_{jj}G (that are the commonly measured quantities) in terms of Γ∥ and Γ⊥. For example, in a typical experiment with a thick cantilever beam having the main axis directed along the x direction, the strains are approximately ε_{xx} = ε, ε_{yy} = −νε, and ε_{zz} = −ν'ε, where ν and ν' are the Poisson ratios of the cantilever and the resistive sample, respectively. By using Eqs. (4, 5), the longitudinal (K_L) and transverse (K_T) piezoresistive gauge factors are:

\[ K_L \equiv K_{xx} = (1 + \Gamma_\parallel) + (1 - \Gamma_\perp)(\nu + \nu'), \]

\[ K_T \equiv K_{yx} = -(1 + \Gamma_\parallel)\nu - (1 - \Gamma_\perp)(1 - \nu'). \]

Hence from a measurement of K_{xx} and K_{yx} it is possible to extract the intrinsic piezoresistive coefficients Γ∥ and Γ⊥. As we are going to show below, the knowledge of Γ∥ and Γ⊥ permits to characterize the nature of transport of percolative piezoresistors.

As already pointed out in the introduction, the piezoresistive response depends on a non trivial way on the vicinity of the resistor network to its percolation critical point. Consider in fact Fig. 1a in which we report numerical Monte Carlo calculations of Γ∥ and Γ⊥ for bond percolating random resistor networks. In this model, a fraction p of bonds distributed at random has conductance exp(−2d/ξ), while the remaining fraction 1 − p has zero conductance. In the figure, filled symbols refer to a model in which the tunneling exponent 2d/ξ is set equal to 4, while the open circles are obtained by treating ξ as a random variable in such a way that 2 ≤ 2d/ξ ≤ 6.

When there are no missing bonds (p = 1) the piezoresistive response is anisotropic with the longitudinal coefficient much larger than the transverse one, reflecting the fact that the current flows mainly along paths parallel to the direction of the voltage drop. This is more evident for the 2d/ξ = 4 data (filled circles) for which Γ∥ = 2d/ξ = 4 and Γ⊥ = 0 at p = 1. If fluctuations in the values of 2d/ξ are allowed (empty circles) even at p = 1 the microscopic currents develop a component orthogonal to the voltage drop direction leading to Γ∥ < 2d/ξ and Γ⊥ > 0. When bonds are removed (p < 1), the longitudinal response gets reduced and at the same time the transverse piezoresistive coefficient is enhanced. This trend persists all the way down to the percolation threshold (p = p_c ≃ 0.2488) where Γ∥ equals Γ⊥ and the system shows perfect piezoresistive isotropy. As shown in Fig.1b, the same qualitative behavior holds true also for site percolation models in which a concentration x of sites is removed at random with remaining conducting bonds having 2d/ξ = 4 (filled squares) or 2 ≤ 2d/ξ ≤ 6 (open squares).

From the results of both bond percolation and site percolation models of Fig.1 we can infer that as a general rule Γ∥ → Γ⊥ when the resistor network approaches its
to see from Eqs. (8) that in our piezoresistive model \( \sigma_y/\sigma_x - 1 \sim (\Gamma_\parallel - \Gamma_\perp)/\epsilon \), so that the two exponents \( \lambda \) and \( \lambda' \) are equal.

In summary, we have shown numerically that in the critical regime of percolative resistor networks the piezoresistive anisotropy has a power law behavior. This result suggests a possible experimental route to characterize transport properties in TFRs or other 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. 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. (8) or equivalently:

\[
G \sim \left( \frac{\Gamma_\parallel - \Gamma_\perp}{\Gamma_\parallel} \right)^{\lambda/t},
\]

where \( t \) is the critical exponent of the unstrained conductance \( G \sim (p - p_c)^t \). We are not aware of published data from which it is possible to infer \( G \) and \( (\Gamma_\parallel - \Gamma_\perp)/\Gamma_\parallel \) as a function of the metallic concentration in a controlled way, so Eq. (8) cannot yet be tested. However, 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 in qualitative accord therefore with Eq. (8). We encourage acquisition of additional experimental data in TFRs to verify our predictions.

1. S. Kirkpatrick, Rev. Mod. Phys. 45, 574 (1973).
2. M.-C. Chan, A. B. Pakhomov, and Z.-Q. Zhang, J. Appl. Phys. 87, 1584 (2000).
3. C. Grimaldi, P. Ryser, and S. Strässler, Phys. Rev. B 64, 064201 (2001).
4. N. M. White and J. D. Turner, Meas. Sci. Technol. 8, 1 (1997).
5. M. Prudenziati, Handbook of Sensors and Actuators (Elsevier, Amsterdam, 1994), p.189.
6. G. E. Pike and C. H. Seager, J. Appl. Phys. 48, 5152 (1977).
7. C. Canali, D. Malavasi, B. Morten, M. Prudenziati, and A. Taroni, J. Appl. Phys. 51, 3282 (1980).
8. M. Hrovat, J. Holc, D. Belavič, and S. Šoba, J. Mater. Sci. Lett. 13, 992 (1994)
9. A. Kusy, Physica B 240, 226 (1997).
10. B. Morten, L. Pirozzi, M. Prudenziati, and A. Taroni, J. Phys. D: Appl. Phys. 12, L51 (1979).
11. B. I. Shklovskii, Phys. Status Solidi (b) 85, K111 (1978).
12. C. J. Lobb, D. J. Frank, and M. Tinkham, Phys. Rev. B 23, 2262 (1981).
13. A. K. Sarychev and A. P. Vinogradoff, J. Phys. C: Solid State Phys. 12, L681 (1979); ibid. 16, L1073 (1983).