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Abstract

Thick-film resistors (TFRs) consist of a percolating network of conducting oxide nanoparticles dispersed in an insulating glassy matrix, whose resistive properties are dominated by quantum tunneling across insulating layers separating adjacent conducting grains. Tunneling processes are at the origin of the high sensitivity of the TFRs resistances to applied strains. We have measured transport and piezoresistive response for different RuO₂-based TFRs as a function of metallic concentration x and RuO₂ grain sizes. The conductivity is shown to vanish as x approaches a critical concentration x_c by following a power law with nonuniversal critical exponents, while the piezoresistivity diverges at the same critical concentration. We argue that nonuniversality and diverging piezoresistivity have the same origin and arise from the highly fluctuation inter-grain tunneling distances determined by the segregated microstructure of TFRs.

Transport nonuniversality

Transport properties of disordered insulator-conductor composites are characterized by the existence of a percolation critical value x_c of the conducting phase volume concentration x below which the system undergoes a metal-insulator transition [1]. Close to x_c the resistance R of the composite follows a power-law of the form:

$$R \approx R_0 (x - x_c)^{-t} \quad (1)$$

where R_0 is a prefactor and t is the transport critical exponent. **A vast class of materials displays the same value $t \approx 2.0$ regardless of the composition and of the microstructure.** Such universal behavior is due to the unimportance of the microscopic properties for transport which instead is governed by macroscopic quantities such as the percolating backbone [1].

In contrast to universal materials, various disordered systems follow Eq.(1) with critical exponents which depend on the particular material, usually taking values much larger than $t=2.0$. **The origin of such universality breakdown is still unclear.**

To investigate on the nature of transport nonuniversality, we have performed transport measurements on RuO₂-based thick-film resistors (TFRs). Two series with 40 nm and 400 nm mean RuO₂ grain size were fabricated by mixing the conducting powders with a lead borosilicate glass [PbO(75%wt)-B₂O₃(10%wt)-SiO₂(15%wt), temperature of fusion: $T_c=500$ °C]. The resistors were screen printed on Al₂O₃ substrates and fired at peak temperatures ranging from 525 °C to 600 °C.

The resulting resistance values as a function of RuO₂ volume fractions are reported in Fig.1 and Fig.2 for 400 nm and 40 nm RuO₂ grain sizes, respectively. All samples follow Eq.(1) with critical exponents values ranging from $t=1.47$ up to $t=4.2$. **We have therefore fabricated TFRs close to universality [Figs. 1(a) and 2(a)] and TFRs markedly nonuniversal [Fig. 1(b) and 2(b)].**

The problem is now to find the origin of nonuniversality. A way to solve this problem is the following: since nonuniversality means that the critical exponent t depends upon some (yet unknown) microscopic properties, then it could be possible to change it by applying some external perturbation to the system. The nature of the perturbation capable of changing the value of t will provide important informations upon the mechanism of universality breakdown

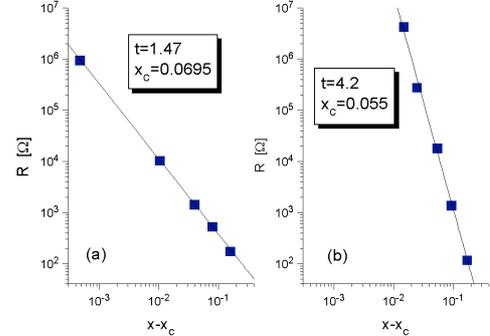


Fig.1 Sheet resistance of 400 nm RuO₂-glass TFRs. (a) firing temperature: 525 °C. (b) firing temperature: 600 °C.

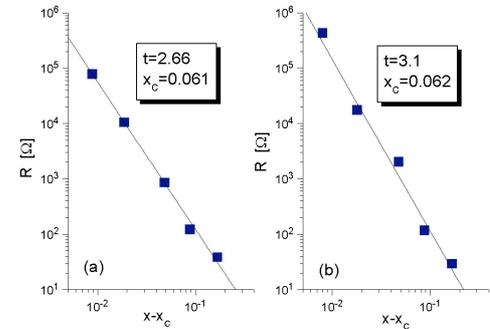


Fig.2 Sheet resistance of 40 nm RuO₂-glass TFRs. (a) firing temperature: 525 °C. (b) firing temperature: 550 °C.

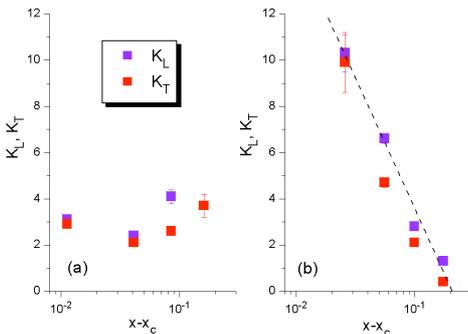


Fig.3 Longitudinal (K_L) and transverse (K_T) piezoresistance response of 400 nm RuO₂-glass TFRs. (a) firing temperature: 525 °C. (b) firing temperature: 550 °C.

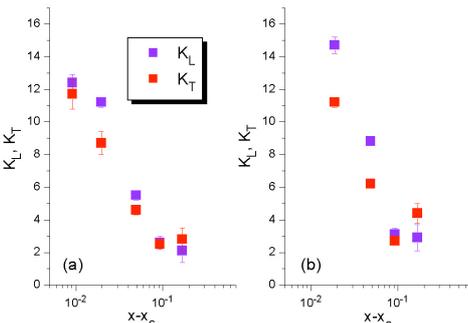


Fig.4 Longitudinal (K_L) and transverse (K_T) piezoresistance response of 40 nm RuO₂-glass TFRs. (a) firing temperature: 525 °C. (b) firing temperature: 550 °C.

Piezoresistance response and origin of nonuniversality

We have identified in applied mechanical stress a perturbation capable of changing the transport critical exponent. We have recorded the change of resistance under applied strain ϵ in cantilever bar measurements. The resulting piezoresistance factor

$$K = \frac{\Delta R}{\epsilon R} \quad (2)$$

has been recorded as a function of different RuO₂ concentrations x and for different strain-voltage drop orientations: the longitudinal (transverse) piezoresistance K_L (K_T) has been obtained for main strain applied parallel (orthogonal) to the direction of the current.

K_L and K_T are reported in Figs.3 and 4 for the same TFRs of Figs.1 and 2. Both piezoresistance responses have similar behavior indicating a closeness to the perfectly isotropic limit postulated to occur at the critical percolation threshold x_c [2]. In Fig. 3(a), K_L and K_T do not display a clear x dependence, while for the other cases the piezoresistance diverges at x_c by following approximately a logarithmic behavior of the form:

$$K = K_0 + B \ln\left(\frac{1}{x - x_c}\right) \quad (3)$$

A logarithmic divergence of K can be understood only by allowing a strain dependence of the transport critical exponent t . In fact the above equation with $K_0 = \Delta R_0 / \epsilon R_0$ and $B = dt/d\epsilon$ follows by direct differentiation of Eq.(1) with respect to ϵ . On the contrary, when t is universal $dt/d\epsilon = 0$ and the piezoresistance is simply equal to K_0 and does not depend on the volume fraction, as in Fig. 3(a). This last result is compatible with the (quasi) universal transport of Fig. 1(a).

The strain dependence of the transport critical exponent is compatible with the theory of Balberg [3] on the universality breakdown induced by quantum tunneling between conducting grains with large dispersion of tunneling distances. According to this theory, the critical exponent is [3,4,5]

$$t = \begin{cases} 2.0 & \text{if } \nu + 2a / \xi < 2.0 \\ \nu + 2a / \xi & \text{if } \nu + 2a / \xi > 2.0 \end{cases} \quad (4)$$

where $\nu=0.88$ is the correlation length exponent, a is the mean tunneling distance and ξ is the tunneling decay length. Upon an applied strain ϵ , the mean tunneling distance changes to $a(1+\epsilon)$, while both ν and ξ remain unchanged. Hence the critical exponent acquires an ϵ -dependence and $B=dt/d\epsilon \neq 0$.

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