Low-frequency 1/f noise of RuO_2-glass thick resistive films

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Measurements of low-frequency noise in thick-film resistors at low temperatures are reported. Films were prepared in a standard "high temperature" process: 20 nm sized RuO_2 powder was mixed with 0.5 μm granular lead–borosilicate glass and organic solvent to give a paste, which was then screen printed onto alumina substrates and fired in a tunnel furnace. Measurements below liquid helium temperature reveal that the low frequency (1/f) noise increases with decreasing temperature, approximately as $T^{-\alpha}$, $\alpha=2.1\pm0.1$. Up to 4 T no dependence of noise intensity on magnetic field has been observed. Measurements of noise spectra in the range 4–300 K show that spectral and temperature slopes obey Dutta, Dimon, and Horn equation [Phys. Rev. Lett. **43**, 646 (1979)] only in the range $T>10$ K. Below this temperature a gap of constant width opens between noise exponent calculated from the spectral slope and from temperature dependence of noise magnitude. This gap occurs due to the change of noise coupling mechanism that takes place at ≈ 10 K. At higher temperatures this coupling is temperature independent. At lower temperatures coupling becomes temperature dependent. It is shown that data agree quantitatively with the concept that noise sources modulate energies for thermally activated hops in the percolation network. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2815677]

I. INTRODUCTION

The excellent performance of RuO_2-glass composite films as low-temperature thermometers[1] and bolometers[2] results from the appropriate sensitivity of their resistance to the temperature and its virtual independence to the magnetic field. Although, in such applications noise becomes a factor that limits the device resolution and/or detectivity, so far no papers deal with noise properties of RuO_2-glass films below liquid helium temperature and very few papers report[3–5] on noise versus temperature measurements in general. Our early measurements show that as $T\to 0$ the noise intensity of these films increases significantly.[6] As a result the resolution of RuO_2-based low-temperature sensors gets poorer.[7] As conduction mechanism in thick resistive films is still unknown no satisfactory explanation exists for the observed phenomenon. Experimental data presented in the article provide arguments in support or against existing models of conduction in these films. It is shown that the increase of low frequency noise in the lowest temperatures results from the enhanced sensitivity of thermally activated resistances that built up the percolation network to the fluctuations of their activation energies.

In Sec. II details of the experimental procedures both technological and measurement are described. Much attention is paid to prove that the main component of measured noise is the bulk noise of the films, and to thorough calibration of noise measurement setup. Thermal noise was used as a reference signal here. Results of low-frequency excess noise measurements are gathered in Sec. III. Discussion of the results in terms of existing theories of 1/f noise and conduction in RuO_2-glass films is provided in Sec. IV.

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II. EXPERIMENTS

A. Films fabrication

RuO_2-glass thick films were prepared by screen printing of a resistive paste on alumina substrate and firing in suitable temperature profile. The paste was composed of ruthenium dioxide and glass powders embedded in organic solvent. RuO_2 powder was prepared in a special heating process, which enables controlling the size of the particles.[8] Average RuO_2 grain diameter was estimated by x-ray peaks broadening, which gave the value of 13.7 nm, and by Brunauer-Emmet-Teller method (BET) or electron microscope which gave 15 or 25 nm, respectively. The glass frit consisted of 65% PbO, 10% B_2O_3, and 25% SiO_2 by weight. Glass particles were much larger, of 0.5 μm size. Pastes of volume fraction $v=0.10$, 0.12, and 0.16 of conducting component (RuO_2) were prepared. They were printed on substrates through 200 mesh screens to give films which were then fired in a flow furnace for 10 min. at peak temperature of 850 °C. The whole firing cycle with preheating and slow-cooling zones took in all 60 min. After firing the films gain the average thickness of ≈ 15.5 μm. Although, prior to the printing, the substrates were selected for its flatness, the roughness of both substrate and screen surfaces was the reason of significant nonuniformity in the film thickness. In the worst case for a 15 mm long sample the differences in the film thickness measured along its length got near 5 μm.

B. Film microstructure

Structural characteristics of the films were examined by transmission electron microscopy (TEM). The image taken for one of the films is shown in Fig. 1. Light glass areas and dark clusters of RuO_2 grains are visible. The latter appear

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FIG. 1. TEM picture of the film made of the paste containing $\upsilon=0.10$ of RuO_2.

due to the agglomeration of (very small) RuO_2 particles and segregation of these, still small, agglomerations on the surfaces of large glassy grains. An important role in electrical conduction plays the interface area between RuO_2 and glass. It was experimentally verified that ruthenium diffuses into a glass on the depth of 0.1–2 μm.[9–11] As a consequence a modified glass layer (reactive layer) forms around RuO_2 particle making its "effective size" much larger than its geometrical diameter. This makes conduction take place at very low concentrations of conducting constituent, well below the percolation threshold.

C. Samples

Electrical terminations to resistive layers were prepared in a similar printing-firing process prior to resistive film deposition. Conductive pastes used in this process contained either Au or PtAu composition. Such pastes were chosen in order to prevent the migration of the metal from the terminations, which could occur during firing of resistive film. Experiments report on enhanced dissolution of silver in glass in the presence of RuO_2.[12,13] On the contrary no diffusion of Au or Pt into glass was observed.[12] Metal migration from terminations can affect electrical properties of the contact-film interface. Diffusion of Ag is known to increase both the resistivity and noise.[14,15] In the case of gold and platinum noise was shown to remain not changed,[14] what is in favor of no diffusion of these metals into the glass.

Although for our terminations no increase of noise due to metal migration is expected, the contacts can always be the source of undesired noise. Therefore the samples were designed in a form suitable for discrimination between bulk and contact components of the measured noise.[14,16] They were formed as multiterminal devices with several voltage legs along their sides. The distance between successive legs, as well as between first/last leg and adjacent current termination was 2.5 mm. The total length of the sample was $L=15$ mm. Apart from such large L samples also small S samples were manufactured. They were 6 mm long with two side contacts (2 mm apart) along both their edges. Both S and L samples were 1 mm wide.

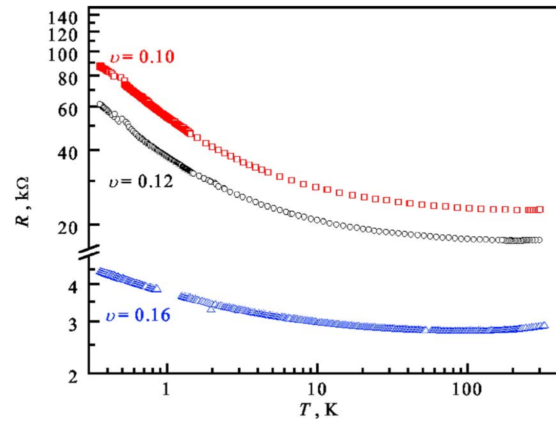

FIG. 2. (Color online) Resistance vs temperature for the S samples $\upsilon=0.10$, 0.12, and 0.16.

The resistance R of the S samples, for which most data are gathered, as a function of temperature is shown in Fig. 2. It was measured with low-excitation ac resistance bridge in a four wire arrangement of the samples connected in series. Voltage was measured on upper side contacts of the samples so the sheet resistance of the films can be calculated as 1/8 part of the measured resistance. It was 2.86, 2.14, and 0.36 kΩ/sq for $\upsilon=0.10$, 0.12, and 0.16, respectively.

D. Noise measurements

Noise measurements were made with either direct current (dc) or alternate current (ac) techniques[17] in a bridge configuration. Samples were paired to match their resistance $\approx\frac{1}{2}R$, placed in the bottom arms of the bridge and biased through load resistors of much larger resistance $\approx\frac{1}{2}R_B$ ($R_B \gg R$). The signal (voltage) from the bridge diagonal was amplified in 5186 low-noise preamp and in the case of ac excitation demodulated in 5105 lock-in amp (both from Signal Recovery). Power spectral densities $S_V(f)$ of voltage fluctuations were calculated in HP 35660A dynamic signal analyzer or PC-based acquisition system equipped with plug-in DAQ card from NI. Background noise $S_{V=0}$ was measured with no bias.

For ac bias the carrier frequency was 325 Hz. Spectra were calculated in the frequency range 0–25 Hz. In this frequency range the spectrum of background noise was flat. The values of $S_{V=0}$ averaged over the band $\Delta f=0.0625-3$ Hz as a function of temperature exactly agreed with the calculated components of Johnson noise coming from measured and load resistors and preamplifer noise. This agreement makes us sure that the measurement setup was calibrated very thoroughly.

Similar care was taken when calibrating dc setup. This technique enables recording noise spectra in much wider frequency range. As it requires larger biases it can be used only in higher temperatures. dc measurements were performed on pairs of L samples. Spectra were continuously measured and stored, whereas temperature changes slowly. Care was taken to ensure that drift component of f^{-2} did not change the

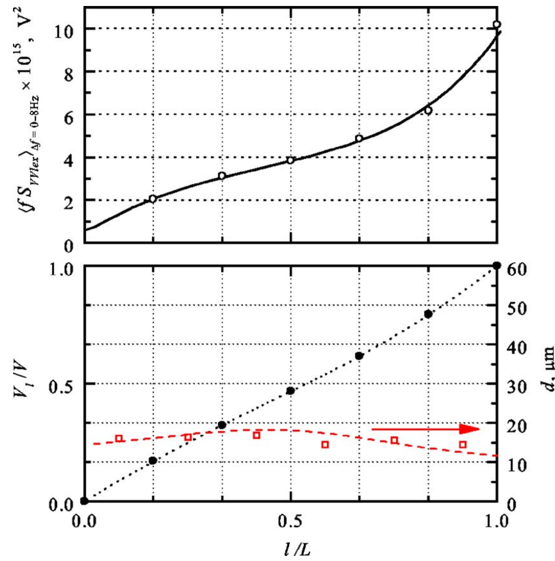

FIG. 3. (Color online) Noise magnitude $\langle fS_{VVl\mathrm{ex}}\rangle$ (open circles), voltage (filled circles), and film thickness d (squares) vs relative distance l/L for υ=0.12 L samples: #108 and #111 with Au terminations measured at T=0.94 K. The voltage across the whole sample was V=0.103 $\mathrm{V_{rms}}$. Lines are: the graph of Eq. (1) (solid, upper panel), and the function: d=15.5$\times V/L(dV_l/dl)^{-1}$ (μm) (dashed, lower panel).

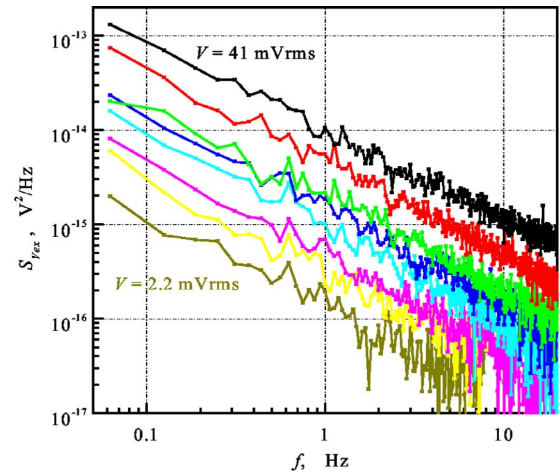

FIG. 4. (Color online) Power spectral densities of excess noise $S_{V\mathrm{ex}}$ measured at T=0.51 K for several voltages biasing a pair of υ=0.12 S samples.

spectra at low frequencies.[18] Independently of the method used, the excess noise was calculated as $S_{V_{\mathrm{ex}}}\equiv S_V-S_{V=0}$.

E. Contact versus bulk noise

The influence of contacts on the noise was examined by scaling cross power spectra S_{VVl} between fluctuations δV_l on a side contact and fluctuations δV on the upper current tap versus the distance l between the side contact and the lower current tap.[16] In this experiment L samples were used. They were biased by ac current. Excess noise was calculated and, as it had 1/f spectrum, the product $fS_{VVl\mathrm{ex}}$ averaged $\langle\ \rangle$ over some frequency band, was used as a measure of noise magnitude. Results are shown in the upper panel of Fig. 3. Although, the data do not scale linearly with l it is still possible to prove that increase of the noise near film terminations is a pure bulk effect. Inspection of voltage distribution along the sample length reveals some increase of electric field near its terminations (see lower panel of Fig. 3). As migration of metal is not the reason,[14] this should be attributed to the decreasing thickness of the film near the contacts. This effect known as "film thinning" is caused by outflow of the glass from film to the contact and is reported in the subject literature.[19] Film thickness calculated as $d\propto(dV_l/dl)^{-1}$ (dashed line) and directly measured (squares, Vistronik C1) is also shown in Fig. 3. Increase of the noise near the terminations occurs because S_V is proportional both to reciprocal volume and electric field squared.[20] In all it gives d^{-3} dependence. Solid line in the upper panel of Fig. 3 was calculated as

$$\begin{aligned}\langle fS_{VVl\mathrm{ex}}\rangle &= S_{\mathrm{cont}}+S_{Vl(\mathrm{bulk})}\\ &=5.9\times10^{-16}\\ &\quad+8.4\times10^{-15}L^2/V^3\int_0^l(dV_l/dl)^3dl\ \ (\mathrm{V}^2),\end{aligned}\qquad(1)$$

assuming constant bulk noise intensity within the whole film to calculate $S_{Vl(\mathrm{bulk})}\propto\int(dV_l/dl)^3dl$ and an adjusting parameter S_{cont} for the contact noise. The result, $S_{VL(\mathrm{bulk})}$=9.3$\times10^{-15}$ V^2 is more than 90% of the total noise measured at current terminations (10.02×10^{-15} V^2). Similar measurements were performed for several L samples with both Au and PtAu contacts and at various (low) temperatures. In all cases contact noise was found not to exceed 10% of the total noise.

The solid line in Fig. 3 was calculated assuming that the source of the bulk component of excess noise are equilibrium resistance fluctuations. The agreement found proves that the assumption was correct. It was further confirmed by scaling noise magnitude against the bias.

III. RESULTS

A. ac measurements

Measurements were performed on S samples with Au terminations. Spectra of excess noise were measured for several (ac) voltages V across the samples at several fixed temperatures. As shown in Fig. 4 they are of 1/f shape. So, $\langle fS_{V_{\mathrm{ex}}}\rangle$ was used as a measure of noise magnitude for a given bias. Squared voltage scaling of $\langle fS_{V_{\mathrm{ex}}}\rangle$ was found only in $V\to0$ limit. Data in Fig. 5 show that at low temperature and for large voltages the dependence of $S_{V_{\mathrm{ex}}}$ on V^2 becomes sublinear. Although density of the driving current was as low as 10^{-2} A/cm^2 and power dissipated was kept below 100 nW, some heating of the samples takes place at larger bias voltages. The observed nonlinearity is due to nonuniform self-heating of the films and electrothermal feedback.[21,22]

In the low voltage limit the scaling $S_{V_{\mathrm{ex}}}\propto V^2$ is preserved so the noise is purely resistance. Its intensity $S\equiv\langle fS_{V_{\mathrm{ex}}}\rangle/V^2$ is plotted versus temperature in Fig. 6. In case of inhomogeneous samples parameter S should be used rather than Hooge parameter α_H (Ref. 20). Those two are trivially related α_H=SN, N is the number of charge carriers, only for homogeneous materials. In case of inhomogeneous media current-crowding effects involve the so called effective volume and make this relation more complex.[23] One can see that below 4

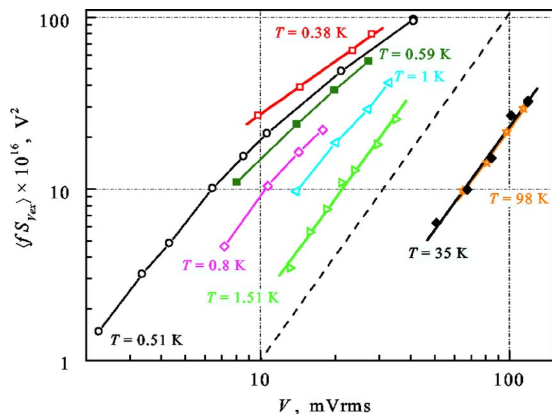

FIG. 5. (Color online) Dependence of noise magnitude $\langle fS_{V\mathrm{ex}}\rangle$ on voltage at selected temperatures measured for samples $\upsilon=0.12$. Dashed line has the slope of 2. Averaging $\langle\rangle$ was over the band $\Delta f=0.0625-3$ Hz.

K, S increases significantly with decreasing temperature. This is quite an important result as few experimental studies devoted to temperature dependence of low frequency noise in thick-film resistors explore quite different temperature range.[3–5] Chen *et al.*[4] measured noise between 77 and 425 K. Masoero *et al.*[3] and Pellegrini *et al.*[5] studied temperature dependence of $1/f$ noise and resistance in a range 77 −750 K. Conductive phase of their films was either bismuth ruthenate ($Bi_2Ru_2O_7$) (Ref. 3) or IrO_2 (Ref. 5). Noise of carbon resistors was measured by Black *et al.*[24] near room temperature and by Fleetwood *et al.*[25] in the range 77 – 450 K. As for other materials, the continuous increase of low frequency resistance noise with decreasing temperature has been observed for metal films,[26,27] doped semiconductors,[21,28–30] and metal–insulator composites.[31,32] The phenomenon is well understood for metallic conduction and at MIT where it is connected with universal conductance fluctuations (UCF).

Measurements in magnetic field were performed for S samples $\upsilon=0.12$ and $\upsilon=0.16$. They are illustrated in Fig. 7. As one can see S does not depend on magnetic field B. This conclusion holds at least in the range from −4 to +1 T, in which the measurements were performed. Not shown in any figure is that the whole S_V vs V dependence, including nonlinear effects at larger voltages is B independent.

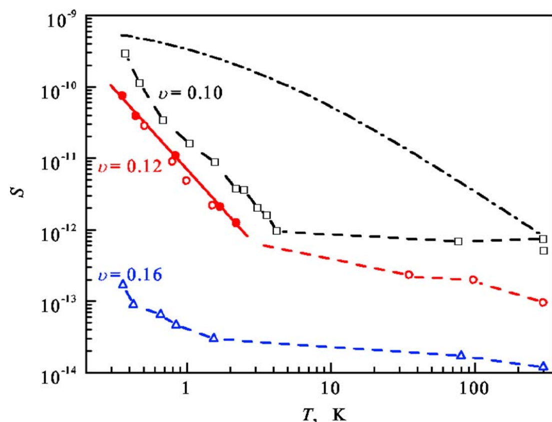

FIG. 6. (Color online) Normalized noise S as a function of temperature. Filled and empty symbols for $\upsilon=0.12$ series refer to two measurement sessions (performed with six month delay) with different values of load resistance R_B. Line drawn through $\upsilon=0.12$ data has the slope $-\alpha=-2.2$. Dashed–dotted line is the plot of $\propto(d\ln R/d\ln T)^2$.

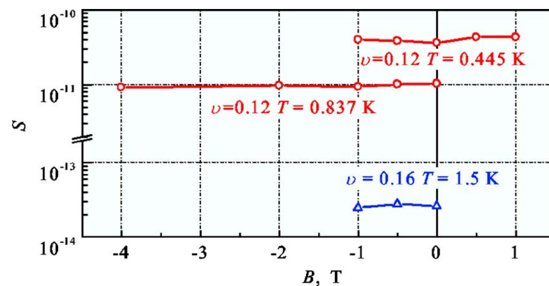

FIG. 7. (Color online) Normalized noise S as a function of magnetic field at selected temperatures measured for samples $\upsilon=0.12$ and 0.16.

B. dc measurements

Reported measurements were performed on $\upsilon=0.12$ L samples with PtAu terminations. Noise spectra were measured in the range 0.5−5000 Hz. Data gathered in a single “thermal run” were displayed in the form of two-dimensional map of the product $fS_{V\mathrm{ex}}(f)/V^2$ versus frequency and reciprocal temperature. Two such maps are shown as an example in Fig. 8. Streaks on the maps show migration of the local maxima with temperature. Lines drawn along the streaks prove that for each line $f_{\max}$ depends on $1/T$ linearly and so thermally activated random processes are responsible for the

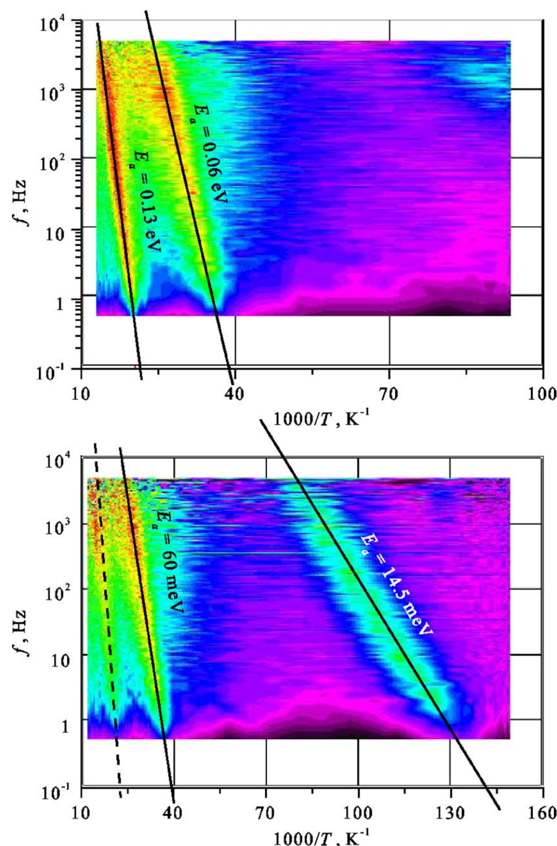

FIG. 8. (Color online) Sample maps of $fS_{V\mathrm{ex}}(f)/V^2$ vs frequency and reciprocal temperature measured with dc technique for samples #102, #103 ($\upsilon=0.12$, PtAu contacts) in two experimental runs.

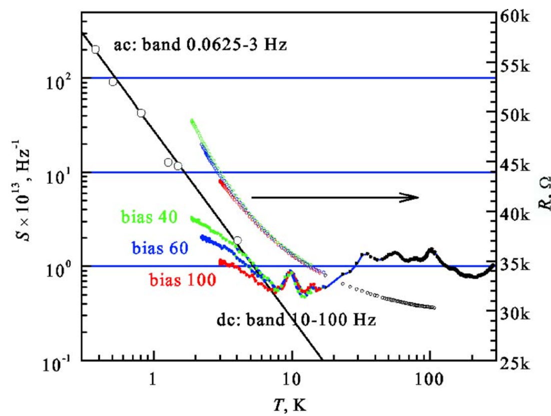

FIG. 9. (Color online) Normalized excess noise (left-hand axis) and resistance (right-hand axis) vs temperature measured with dc (small points) or ac (large circles) bias for samples #102 and #103. Line is the function graph of $\propto T^{-2}$. dc measurements were performed for three different voltages 40, 60, and 100 V across the measurement bridge. Voltage across the samples changes during experiment and was in the range 0.6−0.85, 0.9−1.2, and 1.5−1.9 V, respectively.

features appearing in the noise spectra. Activation energies can be calculated from the slope of the lines. For samples #102 and #103 we have found E_a=0.2, 0.13, 0.06, and 0.0145 eV and the corresponding prefactors τ_0=0.3, 0.018, 0.99, and 32 ps. Experiments with another pair of samples, #104 and #106, show that although generally the maps remain similar, the details: number of maxima and values of activation energies can be different.

Increase of noise intensity in the lowest temperatures, known from ac experiments, cannot be seen in the maps of Fig. 8 but it is still possible to observe it in dc measurements when the voltage used to bias the samples gets smaller and lower temperatures become accessible. In Fig. 9 normalized noise S, calculated by averaging $fS_{V\mathrm{ex}}/V^2$ over the band 10 −100 Hz, is plotted versus T. In this frequency range streaks on the maps of Fig. 8 produce local maxima at 55, 33, and 10 K. Below 7 K noise starts increasing but curves for different bias voltages split. This split is due to self-heating of the samples and was observed also in ac measurements. Note that split of noise curves is associated with rather little splitting of resistance versus temperature curves. The latter were measured simultaneously with noise spectra and are displayed in Fig. 9 with respect to the right axis. As mentioned in Sec. III A this virtual inconsistency is due to nonuniform heating and electrothermal feedback effect.

For the lowest dc bias of 40 V noise is not suppressed down to T=4 K. This can be concluded upon supplemental ac measurements which, as made with much lower excitation, can give the value of noise not subjected to the heating effects. As shown in Fig. 9 both ac and dc values measured for 40 V bias follow the same line when $T>4$ K. This line drawn through all ac results shows that as $T\rightarrow 0$ normalized noise increases in a power-law manner and the exponent found is $\alpha\approx 2$. This value is slightly smaller than the value $\alpha\approx 2.2$ found for υ=0.12 S samples of Fig. 6. The difference gives the view on sample-to-sample variation of this exponent and/or the order of errors made in experiments.

Data in Fig. 9 are suitable for comparison with other experimental reports on temperature dependence of noise in thick-film resistors. The authors of Ref. 4 have found a minimum in $S(T)$ at 280 K. At the same time $R(T)$ had the minimum at 350 K. They concluded that noise and resistance were caused by different mechanisms. In Ref. 3 a minimum in $S(T)$ shifted relative to the minimum in $R(T)$ to slightly lower temperatures has been found. The authors concluded that there is more than one physical source generating excess noise in their films. Eventually in Ref. 5 measurements similar to ours made in temperature range 80–800 K undoubted confirmed the existence of thermally activated noise source with activation energy of $E_a\approx 1.52$ eV. Thermally activated origin of the noise was also confirmed for carbon resistors.[24,25]

IV. DISCUSSION

Conduction mechanism in RuO_2-glass thick films is still unknown. Thermally activated tunneling,[33] fluctuation induced tunneling,[34,35] conduction in a narrow band,[36] variable range hopping (VRH),[37,38] space charge limited transport,[8] emissions over graded barriers,[35] weak localization,[39] are considered transport mechanisms. Noise data presented in this article provide arguments in favor or against some of them.

A. Diffusion noise

Thermally activated processes that produce features in the noise spectra could be electron scattering from diffusing defects. This mechanism is worth to be considered as activation energy of 230 meV was found for H^+ diffusion in Nb metallic films.[40] On the other hand, the prefactors of relaxation times in the range $10^{-5}<\tau_0<10^{-3}$ s found for this process were much larger when compared to our values of $\tau_0<10^{-10}$ s. The hypothesis can be verified for each activation energy by the trial fit of the data with noise diffusion equation[40]

$$-\frac{\partial \ln S}{\partial \ln f}\equiv\gamma=1+\frac{kT}{E_a}\frac{\partial \ln S}{\partial \ln T},\tag{2}$$

which relates spectral slope γ with temperature dependence of the noise at frequency f. Equation (2) for a given activation energy contains no free parameters what makes the test definite. In Fig. 10 data of Figs. 8 and 9 are analyzed in the framework of noise diffusion equation. Noise exponent γ calculated as the slope of the noise spectra at $f=(10\times 100)^{1/2}=31.7$ Hz is compared with that predicted from the right-hand side (rhs) of Eq. (2) using temperature dependence of Fig. 9 measured under 40 V dc bias and values of activation energy that produce maxima at T=33, 55, and 100 K. For each of these values the variation of noise exponent predicted from temperature dependence is much smaller than calculated from the spectral slope. Moreover for T=33 and 55 K an offset between the two sides of Eq. (2) becomes apparent. Thermally activated diffusion as a source of the noise seems unlikely, also because another model of $1/f$ noise fits the data in Fig. 10 much better.

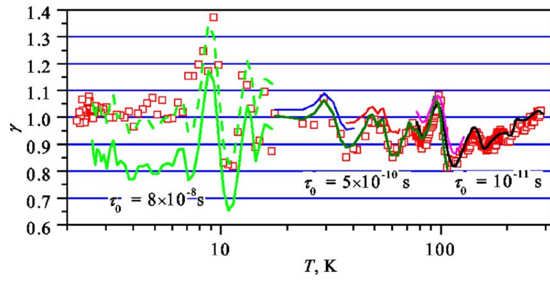

FIG. 10. (Color online) Noise exponent calculated from spectral slope (points) at $f=31.7$ Hz compared with the values calculated from temperature dependence of noise (lines) through rhs of Eq. (2) (thin solid) and activation energies $E_a=0.2$ (most right), 0.13, and 0.06 eV (most left) or Eq. (3) (thick solid) or Eq. (5) (dashed).

B. DDH analysis

Dutta, Dimon, and Horn (DDH) derived $1/f$ spectrum from the presence of thermally activated transitions between the states of approximately equal energy.[41] Their theory relates frequency and temperature dependencies of noise intensity by

$$\gamma = 1 - \frac{1}{\ln(\pi\tau_0 f)}\left(\frac{\partial \ln S}{\partial \ln T} - 1\right), \tag{3}$$

where τ_0 is attempt time to make a transition. In Fig. 10 spectral exponent γ is compared with that predicted from the rhs of Eq. (3). In the range $T>15$ K the agreement is excellent although not all assumptions of DDH model are exactly met: two different values of τ_0 were used to achieve such good obeying of the DDH equation. Both these values fall within the range of $10^{-11}>\tau_0>10^{-14}$ s, acceptable for condensed matter.[42] Spread and relatively large value of this time suggest that two-state systems are of various sizes and they are big objects rather than point like defects. The nature of these systems is not known, but as states of roughly equal energy and continues distribution of activation energies are unlikely in a pure bulk crystal in thermal equilibrium, the most probable candidates of noise sources in our thick resistive films are grain boundaries and states in the glass.

DDH theory assumes that random process couples to resistivity through temperature independent mechanism. The excellent agreement found for $T>15$ K confirms that in this range the coupling mechanism is indeed temperature independent. The conclusion is worthy to mention as conduction mechanism in thick-film resistors is still the subject of controversy. Most of the proposed approaches assume that thermally activated tunneling either between metallic grain[33,35,37] or through dopant states in the glassy matrix[35–39] is the dominant transport mechanism. In both approaches elementary transitions are described by local resistances $r_{ij} \propto \exp(2s_{ij}/a + E_{ij}/kT)$. Assuming that fluctuations in two-state systems influence local resistivity via modulation of activation energy E_{ij} and tunneling rate (e.g., via modulation of barrier height and so localization length a) we get

$$\delta r_{ij} = r_{ij}\left(\frac{\delta r_0}{r_0} + \frac{\delta E_{ij}}{kT}\right), \tag{4}$$

where all the fluctuations in the tunneling process were compacted into δr_0. At higher temperatures the second term in parentheses of Eq. (4) is small and $\delta r_{ij}/r_{ij} \approx \delta r_0/r_0$ is indeed temperature independent. Our data are thus consistent with the concept that tunneling is the transport mechanism in thick-film resistors in the considered range of temperatures, $T>15$ K. For lower temperatures in Fig. 10 a gap opens between the two sides of Eq. (3). Solid line in the figure depicts the rhs of this equation and is drawn for $\tau_0=8\times10^{-8}$ s. This value was assumed in order to make temperature based dependence (line) able to reproduce the variation of ≈ 0.5 in the spectral data (points) at 10 K. Smaller values of τ_0 can reduce the gap (approximately twice for $\tau_0=10^{-11}$ s) but simultaneously make the mentioned variation smaller (≈ 0.3). Possible and most probable reason for opening the gap in Fig. 10 for $T<10$ K is that coupling mechanism in this temperature range is no longer temperature independent. For temperature dependent coupling mechanism the DDH relation takes the form[25]

$$\gamma = 1 - \frac{1}{\ln(\pi\tau_0 f)}\left(\frac{\partial \ln S}{\partial \ln T} - \frac{\partial \ln(\overline{[\langle\delta r_{ij}^2\rangle]}/\overline{r_{ij}}^2)}{\partial \ln T} - 1\right), \tag{5}$$

where the inner averaging ([]) is over all two-state systems able to produce fluctuations of r_{ij} and the outer one ($\bar{\ }$) involves current-crowding weighting factors appearing due to inhomogeneous current flow.[43] First, rough approximation is to neglect current-crowding and replace both averages with $\langle\delta r_{ij}^2\rangle/r_{ij}^2$. At low temperature Eq. (4) predicts $\delta r_{ij}/r_{ij} \approx \delta E_{ij}/kT$. Assuming that mean square fluctuation of activation energy $\langle\delta E_{ij}^2\rangle$ caused by transitions in a single two-state system is temperature independent we get $\partial \ln(\langle\delta r_{ij}^2\rangle/r_{ij}^2)/\partial \ln T=-2$. With this approximation the rhs of Eqs. (3) and (5) differ by temperature independent factor of $-2/\ln(\pi\tau_0 f)$, what is observed in our experiment. The line in Fig. 10 derived from temperature dependence of noise intensity through Eq. (5) fits experimental data in the range $T<10$ K much better than those calculated using Eq. (3).

C. Percolation

More detailed treatment of the inhomogeneous media is not easy. In the framework of DDH theory the overall normalized noise is the sum of local terms[43]

$$S = \frac{\sum_{ij}(i_{ij}/I)^4[\langle\delta r_{ij}^2\rangle]kTD_{ij}(E)/f}{(\sum_{ij}(i_{ij}/I)^2 r_{ij})^2}, \tag{6}$$

where $E=-kT\ln(\pi f\tau_0)$ is the activation energy of a two-state system, which modulate resistance r_{ij} of bond ij carrying the fraction i_{ij}/I of the total current I, and $D_{ij}(E)$ is the distribution function of E. The sum in the denominator is just the total resistance R of the samples. Equation (5) can be obtained from Eq. (6) when distribution of activation energies is not a local property, i.e., $D_{ij}(E)=D(E)$. In general this is not true and for our samples was verified experimentally through direct measurements of noise maps for different re-

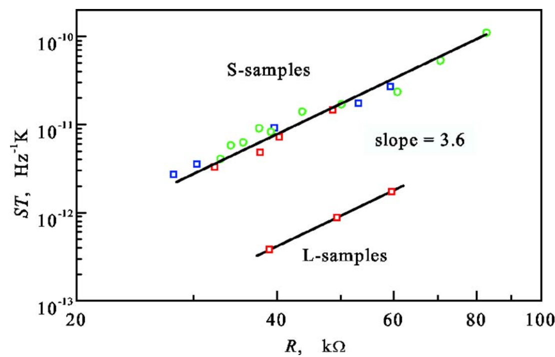

FIG. 11. (Color online) Data from Figs. 9 and 6 for films υ=0.12 (squares) and υ=0.1 (circles) redrawn as ST vs R. Lines show the fit with percolation power law.

gions of the samples.[44] As discussed earlier in the case of thermally activated resistances r_{ij} their relative mean square fluctuations could also be considered as position independent $\langle \delta r_{ij}^2\rangle / r_{ij}^2 = \langle \delta E_{ij}^2\rangle/(kT)^2 \propto (kT)^{-2}$, which is further simplification. Eventually, the term

$$\frac{M_4}{M_2^2} \equiv \frac{\sum_{ij}(i_{ij}/I)^4 r_{ij}^2}{\left(\sum_{ij}(i_{ij}/I)^2 r_{ij}\right)^2} = \frac{\sum_{ij}(i_{ij}/I)^4 r_{ij}^2}{R^2} \tag{7}$$

could be considered as temperature independent. Dashed line in Fig. 10 was calculated for all these simplifications. One step backward needs the estimation of the term in Eq. (7). Percolation theory predicts $M_4 \propto M_2^{2+w} \propto R^{2+w}$, w is the exponent[43] but this relation reflects only topological inhomogeneity. For the samples in question increase of total resistance R with lowering temperature is caused by the (inhomogeneous) change of local resistances r_{ij} and topological redistribution of local currents induced by this change. Both effects make the fourth moment M_4 of current distribution depend on R more stronger (larger values of w) and introduce additional, temperature dependent term into Eq. (6). The precise form of this term depends on type and degree of samples inhomogeneity. For the data of Figs. 9 and 6 we estimate it as $\propto T^{1-\alpha}$ so as the dependence $S \propto T^{-\alpha}$ observed in the low temperature limit could be restored.

On the theoretical side of the problem we can refer to various models of inhomogeneity. Percolation theory predicts $w \approx 0.87$ for percolation on a regular lattices[43] or $w \approx 2.1$ or 2.4 for Swiss-cheese models of percolation in continuous space.[45] The values of exponent w may be higher when systems contain nonrandom nonuniformities in the concentration (υ) of metallic component.[46] Thick-film resistors are usually highly segregated structures in which large glassy grains produce "voids" in the percolation network (see Fig. 1 and e.g., Ref. 47). Films are thus nonuniform and larger values of w are likely to be observed. Our data for samples υ=0.12 and υ=0.10 redrawn in ST vs R (see Fig. 11) coordinates give the value $w \approx 3.6$, which are considerably larger than theoretical estimates cited previously but in the range acceptable for nonuniform samples. Besides, one must remember that percolation equations take into account only "thinning" of the percolation cluster but not simultaneous change of local resistances. When both (effects) are considered exponent w must change. Quantitative analysis is possible in the extreme case when resistances r_{ij} that built up the network take on the values from exponentially wide range. This is the case of VRH, which is discussed in Sec. IV D.

The concept that noise increases with decreasing temperature due to percolation-like scenario was originally proposed for granular metals.[31] Mantese *et al.* postulated that the Arrhenius factors involved in intergrain conductance froze out conduction paths and made noise intensity increase in direct agreement with the percolation power law $S \propto M_4/M_2^2 \propto R^w$. By fitting their experimental data with this equation they have found the values of exponent $w \approx 4.22$ for Pt-Al_2O_3 and $w \approx 2.39$ for Mo-Al_2O_3 composites. The concept has been further explored by Carter *et al.*[48] for carbon black/polymer composites. In their experiment the noise and resistivity occurred to increase simultaneously with *increasing* temperature. The value of exponent w=0.7 was found from the scaling of noise versus resistance.

D. VRH

Theories of resistance noise in VRH are not consistent.[30,49–51] For those which predict increase of noise with decreasing temperature, the expected dependence is either exponential[49] or of power law, $S \propto T^{-\alpha}$. The divergence in Fig. 6 is obviously weaker than exponential. Also, in contradiction to the theories,[49] in our experiment in $T \to 0$ limit the noise exponent $\gamma \approx 1$ (see Fig. 10) and no saturation of the spectra for $f \to 0$ is observed. For power-law dependencies theoretical predictions for exponent α are ≈ 3.4 for Efros–Shklovskii hopping or $\alpha \approx 1.7$ for Mott hopping in three-dimensional (3D), both when two-state systems interact with hopping network via dipolar potential.[30,50] The values $\alpha \approx 2$ or 2.2 found for our films are different from both cited earlier. Much better agreement can be found when states interact with the hopping network via Coulomb potential. Exponent $\alpha \approx 2.3$ specific for Mott hopping is very close to our values. The fundamental objection arises however (general for thick-film resistors) when plotting $\ln R$ vs $(T_0/T)^{1/n}$, namely that the quantities extracted from T_0 are unphysical and temperature range in which VRH relation holds does not satisfy the condition $T \ll T_0$ (Ref. 35).

E. UCF

The main objection against UCF as a source of the noise is that they require 1/2 noise suppression by the magnetic field[27] which was not observed (see Fig. 7). Minor arguments come from the analysis of temperature dependence of noise intensity. Data in Fig. 10 for $T < 7$ K seem too rough to estimate reliably the value of exponent γ but they indicate $\gamma \approx 1$. Spectra of Fig. 4 measured with higher accuracy give $\gamma = 1 \pm 0.4$. DDH theory predicts then that a factor which accounts for the shift of characteristic times with temperature, $D(\ln f, T)$ depends on temperature as $\propto T^{0\pm 0.4}$. Keeping all remaining assumptions of Ref. 32 the same it is expected for the noise caused by (unsaturated) UCF, that $S\sigma^2 \propto T^{-0.4\pm 0.4}$, σ is conductivity. This is much weaker divergence than for thick-film resistors as our data clearly exhibit the scaling

$S/R^2 \propto T^{-1.42}$. Disagreement is even more pronounced when UCF are in the saturated regime. Then the scaling $S\sigma^2 \propto T^0$ is expected.

F. Temperature fluctuations

The case when resistance fluctuations are caused by temperature fluctuations can be easily eliminated by means of elementary considerations. Assuming samples homogeneity the relative noise can be estimated from the variance of temperature fluctuations, $\langle(\Delta T)^2\rangle = kT^2/C_V$ (C_V is the heat capacity of the samples), as[52]

$$S \propto \frac{\langle(\Delta V)^2\rangle}{V^2} = \beta^2\langle(\Delta T)^2\rangle \propto \left(\frac{d\ln R}{d\ln T}\right)^2 C_V^{-1}, \quad (8)$$

where $\beta \equiv (1/R)dR/dT$ is the temperature coefficient of resistance. In Fig. 6 the squared logarithmic derivative of resistance for S samples $\upsilon = 0.1$ is drawn. The curve flattens as temperature decreases so that the derivative in Eq. (8) acquires a constant value as $T \to 0$. In this limit temperature dependence of noise intensity is then described by the temperature dependence of specific heat. The latter for the components of the resistive pastes scale as $\propto T^{1.3}$ for a glass[53] and as $\propto T$ for RuO_2 (Ref. 54). The expected value of exponent $1 \leq \alpha \leq 1.3$ is then far from those found experimentally. Also the absolute value of noise intensity calculated by the use of Clarke and Voss formula of Ref. 52 is many orders of magnitude smaller than measured in experiments.

V. CONCLUSIONS

In summary, our observation is that at ≈ 10 K there is a change of a mechanism through which fluctuations couple to resistivity. At higher temperatures this mechanism is temperature independent. One possibility is that noise sources modulate rates of tunneling transition and thus produce fluctuations of local resistances and of the flowing current. Below 10 K the coupling becomes temperature dependent. Here data agree quantitatively with the concept that noise sources modulate energies of thermally activated hops in the percolation network. In this scenario the observable noise is attributed to fluctuations in mobility rather than in the number of carriers.

Activation energy E_{ij} of local conductance can be estimated from Arrhenius plot of the sample resistance. E_{ij} ranges from a few tens of microelectron volts at 1 K to several hundreds of microelectron volts at room temperature. These values are typical for thick resistive films and are too small to be interpreted as arising from charging the grains. On the other hand the well-known fact is that Ru (RuO_2) diffuses into the glass and large density of dopants exists in the barrier insulator.[9–11,33,36] This wipes out the grain boundaries and makes Coulomb blockade unimportant. Then E_{ij} has the meaning of energy required to activate a trap-assisted-tunneling channel. As described in Ref. 50 noise sources may couple to E_{ij} electrostatically, e.g., via Coulomb or dipolar potential. These sources are localized in a glassy matrix or grain boundaries. One possibility is that they are two-level systems, which exist in glasses and are responsible for anomalous behavior of their specific heat.[53] Interestingly, the existence of two-level systems in a glassy matrix of thick resistive films was postulated by Skrbek *et al.*[55] who found them responsible for resistance relaxation observed in carbon and RuO_2 thick-film low-temperature thermometers. Another possibility is that energies of dopants that belong to percolation network fluctuate due to changing occupation of the dopants at the peripherals of the percolation cluster. In this picture fluctuations of dopants occupation trigger fluctuations of dopants energies. Note, eventually, that percolation-like transport involving tunneling and thermally activated transitions can account for low magnetoresistance of the samples and magnetic field independent noise reported in Sec. III. As pointed out in Ref. 56 the resistance of these transport mechanisms is expected to be magnetic field independent, as opposed to weak localization and hopping which strongly depend on magnetic field and are expected to produce B-dependent resistance fluctuations.

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