Conductivity and $1/f$-noise study of three-component random resistor networks

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Topological and electrical transport properties of three-component random resistor networks (RRN's) i.e., RRN's that contain three types of conductance $g=0$, $g=h_g \ll 1$, and $g=1$, are investigated. Such networks confirm the universality hypothesis but show a much faster increase of conductivity above, rather than inside, the critical region. This originates from the quasibicritical topology of a percolating cluster. Besides the classical percolation threshold in which the conducting infinite cluster is cut, there is also a metallic percolation threshold in which the metallic infinite subcluster (i.e., formed only from $g=1$ bonds) first appears. Another implication of the quasibicritical nature of the investigated RRN is the local peak in the S-versus-p relation, where S denotes the relative power spectrum of the $1/f$ noise and p is the concentration of occupied sites. The basic results obtained with the help of the node-link-blob picture of a percolation cluster were also confirmed by Monte Carlo small-cell real-space renormalization-group computations. The possibility and conditions of utilizing of three-component RRN's in the modeling of real metal-insulator composites are also discussed.

I. INTRODUCTION

The experimental relation

$$\sigma \sim (p-p_c)^t , \tag{1}$$

where σ is the conductivity of a metal-insulator composite, p the fraction of the metallic component, p_c its critical value, and t the conductivity exponent, can be explained by percolation theory.[1,2] In this approach, structural inhomogeneities of a real composite are modeled by a random resistor network (RRN), i.e., a lattice in which the bonds are either removed (with probability $1-p$) or occupied (with probability p) by the resistors of the same conductance g (e.g., $g=1$). For such a binary model the percolation theory predicts only one universal value of the exponent $t \cong 2$ (Ref. 1) for all three-dimensional (3D) lattices. However, this universal value of t has been observed only in a few 3D real composites. In practice, much larger values of t (up to 7) are measured.[3] To explain this, more complicated RRN's with a continuous distribution of bond conductance have been proposed. It was shown[4] that if conductances of all the occupied bonds are distributed according to the singular power law $P(g) \sim g^{-\alpha} (\alpha > 0)$ for $g \to 0$, then the resulting value of the exponent t becomes greater than its universal value. Arguments that the distribution of interparticle conductance with the low-conductance part (i.e., for $g \to 0$) described by a singular power law can take place in real percolating systems were also supplied.[5,6]

In a previous paper[7] we have followed suggestions of Carcia, Suna, and Childers[8] and presented simpler arguments leading also to an explanation of the large experimental values of the exponent t. We pointed out that values of t observed for the real composites are always derived from an approximation procedure in which experimental data are fitted with relation (1). The approximation is usually done over a broad range of the concentration p, not necessarily restricted to the critical region.[8-13] It results in a noncritical value of t that is affected not only by the low-conductance part of the interparticle conductance distribution, but also by the high-conductance one. To investigate how this high-conductance part of the bond-conductance distribution influences the value of t or, more generally, the σ-versus-p relation, we have performed[7] Monte Carlo (MC) calculations on RRN's in which bonds not removed were occupied by conductances according to a bond-conductance distribution containing the low- and high-conductance parts. For simplicity we have assumed single-δ-function representations for both of these parts. By taking into account the third δ function located at $g=0$ and associated with removed bonds, it is clear that we have analyzed the RRN with a three-point bond-conductance distribution. Such a network confirms the universality hypothesis, and $t \cong 2$ is observed in the critical region, i.e., for $p \to p_c$. It is because in this region the lattice conductance is dominated by the low-conductance bonds. In our case these bonds take $g=h_g \ll 1$. We have shown, however, that high-conductance bonds, characterized by $g=1$, though not relevant for the critical behavior, strongly reduce the width of the critical region. These bonds also cause a much steeper increase of the lattice conductivity above this region. The last two effects are, of course, of marginal theoretical interest. However, they result in t greater than 2, if it is extracted from fitting the lattice conductivity to the rule (1) in too wide a range of the concentration p. As we have mentioned above it is often done in the case of the real-composite data. The fact that the unknown value of the percolation threshold p_c is also extracted from the fitting procedure causes smoothing of the data and makes difficult a correct evaluation of the critical region and of the exponent t.[7,8,13]

It should be noted here that the above considerations can be easily generalized to more complicated RRN's: For example, for a RRN in which the low-conductance δ function $\delta(g-h_g)$ is replaced by a singular power-law distribution.

There is a simple physical interpretation of our network. Two kinds of occupied bond may be associated with two distinct microscopic conduction mechanisms, observed between conductive grains. The two mechanisms can be metallic conduction for the sintered or touching grains and thermally activated tunneling for the nontouching ones[14,15] (or alternatively, enhanced interfacial ionic conduction and pure ionic conduction[16]). Now taking into account that the temperature influences the conductivity of different physical conduction mechanisms in different ways, a three-component RRN also becomes an useful tool for the explanation of more complex temperature dependencies showed by real metal-insulator composites with the help of percolation arguments.[17,18] It could be a way to identify composities for which the three-component RRN works well.

The purpose of this paper is to give a qualitative picture of the transport mechanisms in three-component RRN's and to investigate their $1/f$-noise properties. We believe that this network could be utilized in modeling of the experimental $1/f$-noise data especially of those composites to which such RRN's have previously been applied.[16-18]

II. $1/f$ NOISE OF PERCOLATING NETWORKS

In 1985 Rammal, Tannous, and Tremblay[19] gave a description of $1/f$ noise in RRN's. For a network with the binary distribution of bond conductance, they assumed that conductance g_α of any occupied bond α takes $g_\alpha = g + \delta g_\alpha$. Fluctuations δg_α were spatially uncorrelated, $\langle \delta g_\alpha \delta g_\beta \rangle = 0$ for $\alpha \neq \beta$, and their relative power spectra (the relative noise) were identical in all the occupied bonds, $\langle (\delta g_\alpha)^2 \rangle / g_\alpha^2 = s_\alpha = s$ for each occupied bond α. The fluctuations of bond conductance result in fluctuations δG of lattice conductance G. The RRN relative noise independent of the lattice size $S = L^3 \langle (\delta G)^2 \rangle / G^2$, where L denotes the lattice size, have been shown to diverge at the percolation threshold[19,20]

$$S \sim s(p-p_c)^{-\kappa} , \tag{2}$$

where the noise critical exponent $\kappa \cong 1.5$ for three-dimensional (3D) lattices.[21-23] More unexpected results have been observed for $1/f$ noise of a random resistor superconductor network (RRSN), i.e., the noiseless superconductor matrix in which the noisy resistors $g=1$, $s=1$, with concentration $1-p$, are embedded. For a RRSN the relative noise S diverges when the percolation threshold is approached from below,[21,24]

$$S \sim s(p_c-p)^{-\kappa'} , \tag{3}$$

despite the simultaneous increase of network conductivity,[25,26]

$$\sigma \sim g(p_c-p)^{-q} , \tag{4}$$

The exponent $q \simeq 0.75$ (Ref. 27) and only wide bounds $0.38 < \kappa' < 1.02$ for the exponent κ' (Ref. 21) have been found for 3D lattices. Investigations of the $1/f$ noise of the RRN's with a power-law distribution of bond conductance have also been carried out.[22,28,29] They led to the relation (2), but with a value of κ greater than its universal value $\kappa \cong 1.5$.

III. UNIVERSALITY OF THREE-COMPONENT RRN

To study the $1/f$ noise properties of the three-component RRN, we assume that all its bond conductances of the same type fluctuate with the same relative noise. Without a loss of generality, we may assume that conductances of the occupied bonds $g=1$ and $g=h_g$, fluctuate with relative noises $s=1$ and $s=h_s$, respectively. The fraction of the high-conductance bonds in all occupied bonds is given by a parameter b_1 $(0 \leq b_1 < 1)$, which can be called a "sintering coefficient." Properties of the RRN defined above can be qualitatively described with the help of the node-link-blob (NLB) picture of the percolating backbone.[1] In this model the conducting cluster is approximated by a superlattice with a lattice spacing equal to the correlation length ξ. All the nodes of this superlattice are connected by links, which in turn are composed of multidimensional small clusters called "blobs" and singly connected bonds (SCB's). The average number L_1 of SCB's within a link scales with ξ as $\xi^{1/\nu}$ (Ref. 30) where $\nu \cong 0.89$ is the correlation-length exponent defined by the relation $\xi \sim (p-p_c)^{-\nu}$.[1] In a three-component RRN the concentrations of the bonds $(g,s)=(1,1)$ and $(g,s)=(h_g,h_s)$ within the conducting infinite cluster are b_1 and $1-b_1$, respectively. This statistic also holds for any topological subset of the occupied bonds. Thus a set of SCB's within a link contains on average $L_1 b_1$ high-conductance bonds $(g,s)=(1,1)$ and $L_1(1-b_1)$ bonds of the type $(g,s)=(h_g,h_s)$. Of course, variations (from a link to a link) around these averages are observed. However, for $L_1(1-b_1) \gg 1$; i.e., for sufficiently long links, the probability of occurrence of a link without low-conductance SCB's is close to zero. That means that almost all links contain SCB's of the (h_g,h_s) type. These SCB's dominate link conductances (since $h_g \ll 1$) and thus the conductivity of the whole lattice. Hence the upper bound for the lattice conductivity can be obtained as follows:

$$\sigma < \frac{h_g}{1-b_1}(p-p_c)^{\nu(d-2)+1} , \tag{5}$$

where the condition $h_g \ll 1$, and the relations $L_1 \sim \xi^{1/\nu}$ and $\xi \sim (p-p_c)^{-\nu}$ have been utilized to obtain Eq. (5). This relation leads us to the lower bound of the exponent t, $t > 1+\nu(d-2)$, the same as obtained for pure binary RRN.[31] Thus we conclude that our three-component RRN confirms the universality hypothesis. Similiarly, the relative noise S of the three-component RRN (with $h_g \ll 1$) can be also estimated:

$$S > \frac{h_s}{1-b_1}(p-p_c)^{-\nu(3d-4)+2t-1} . \tag{6}$$

In Eq. (6) we have utilized the formula of Rammal, Tannous, and Tremblay[19] to compute the link relative noise. As we can see the lower bound for the noise exponent $\kappa, \kappa > \nu(3d-4)-2t+1$, is again, the same as in the pure binary case:[21,22] the universality can be extended to the $1/f$ noise of a three-component RRN (see Fig. 2). As we have mentioned, the relations (5) and (6) hold under the assumption that $L_1(1-b_1) \gg 1$. It can be reached for sufficiently long links (i.e., for $p \to p_c$), for which the number L_1 of SCB's is sufficiently large.

IV. OFF-THRESHOLD PROPERTIES OF THE THREE-COMPONENT RRN

A larger p results in smaller ξ, thus in a smaller L_1. It means that, with p increasing, the concentration p_ξ of the links without low-conductance SCB's in the NLB superlattice also increases. The substantial increase of p_ξ occurs when $L_1(1-b_1)$ (the average number of low-conductance SCB's within a link) decreases below 1. In this case the NLB superlattice is formed from links that are either of high conductance with an average value of conductance equal to $1/L_1$ or of low conductance with an average value of conductance equal to h_g (they usually contain only one low-conductance SCB). The high-conductance links form separate clusters and if p continually increases, p_ξ can reach its threshold value $p_{\xi c}$, and then these separate clusters become a highly conducting infinite cluster connecting the opposite sides of the superlattice. At the scales much greater than ξ, the superlattice can be approximated by the RRSN for $p_\xi < p_{\xi c}$ and by the RRN for $p_\xi > p_{\xi c}$ (the highly conducting infinite cluster built from the links of $1/L_1$ conductance is not the ideal superconducting infinite cluster). In the vicinity of the percolation threshold $p_{\xi c}$, conductivity and the relative noise are described by the power-law equations of $(p_\xi - p_{\xi c})$, which can easily be transformed to

$$\sigma \sim h_g (p_c' - p)^{-q} A(p) , \tag{7}$$

$$S \sim h_s (p_c' - p)^{-\kappa'} B(p) , \tag{8}$$

for $p < p_c'$ and:

$$\sigma \sim 1 (p - p_c')^{t} A_1(p) , \tag{9}$$

$$S \sim 1 (p - p_c')^{-\kappa} B_1(p) , \tag{10}$$

for $p > p_c'$, where p_c' is the concentration of conducting bonds for which p_ξ reaches its threshold value $p_{\xi c}$ ($\cong 0.25$). In Eqs. (7)–(10) the functions $A(p)$, $B(p)$, $A_1(p)$, and $B_1(p)$ take finite values at $p = p_c'$. Taking into account the topological picture of three-component RRN and its physical interpretation, the second percolation threshold p_c' can be also called the metallic percolation threshold because for $p > p_c'$ the infinite metallic cluster first appears. This region of p can thus be called a metallic-conduction regime in contrast to a tunneling-conduction regime given by $p_c < p < p_c'$, where the low-conductance (i.e., tunneling) bonds dominate the transport properties of the lattice.

Relations (7) and (9) describe the dependence of lattice conductivity σ on the fraction p of the metallic component above the critical region. They both show a faster increase of σ when p increases above, rather than within the critical region, where conductivity is described by Eq. (5). The whole dependence [described by Eqs. (5), (7), and (9)] coincides with the MC results presented in Fig. 1. As we have mentioned above, this leads to the relatively large value of the exponent t when it is extracted from the data lying in a broad range of p. The NLB picture also helps us to explain how the high-conductance part of the bond-conductance distribution influences the width of the critical regime: The greater the fraction b_1, the greater p_ξ (for fixed p) and the smaller value of $p = p_c'$ is necessary to reach $p_\xi = p_{\xi c}$ $[(p_c' - p_c) \sim (1-b_1)/(1 - p_{\xi c})]$. Thus the critical region is reduced if the concentration b_1 of the high-conductance bonds in the whole population of the conducting bonds increases. This again agrees well with the results of the numerical simulations of the three-component RRN (Ref. 7) (see also Fig. 1).

Now consider the $1/f$ noise of a three-component RRN. The S-versus-p dependence is described by Eq. (6) in the critical regime and by Eqs. (8) and (10) above this region. Equations (6) and (10) show a decrease in S as p

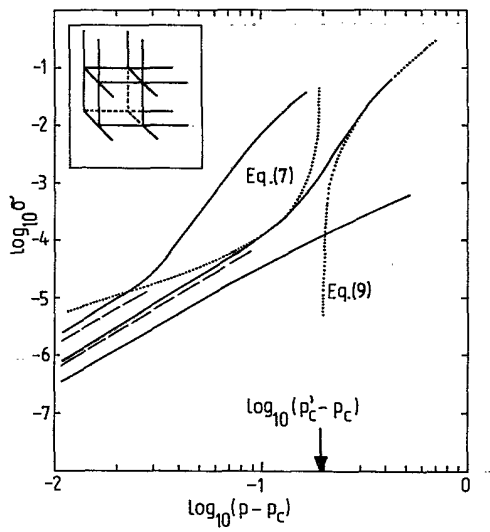

FIG. 1. Conductivity σ vs $p - p_c$ of the three-component RRN (p is the fraction of the sites present, p_c is the percolation threshold), obtained by the MC small-cell RSRG. The lower solid line refers to the classical binary RRN in which all the occupied bonds take low-conductance $g = h_g = 0.001$. The two upper lines refer to the case when the fraction $b_1 = 0.5$ (middle solid line) or 0.8 (upper solid line) of low-conductance bonds are replaced by the high-conductance ones (i.e., with $g = 1$). For $p \to p_c$ all the solid lines indicate the same slope $t = 2.16 \pm 0.05$, which is very close to the universal value $t \simeq 2$. The broken lines drawn for $p \to p_c$ arise from shifting the lowest solid line by $-\log_{10}(1-b_1)$ [see Eq. (5)]. The arrow indicates the location of the metallic percolation threshold p_c' for RRN with $b_1 = 0.5$. For this RRN Eqs. (7) and (9) are placed into the plot as the dotted lines. In the inset the cell utilized in the RSRG computations is shown.

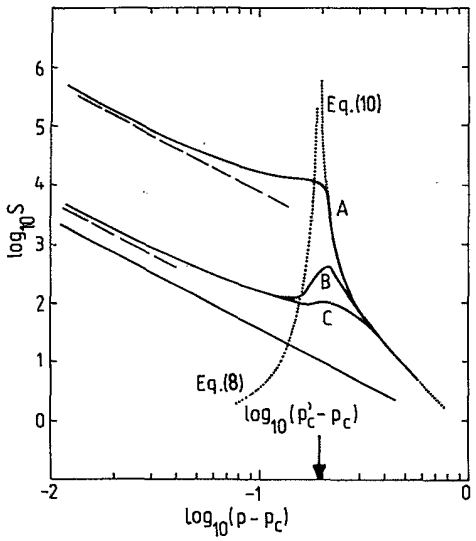

FIG. 2. Relative noise S vs $p-p_c$ of the three-component RRN obtained using RSRG method (see text). The lower solid line refers to the classical binary RRN in which all the occupied bonds take the conductance $g=h_g$ and the relative noise $s=h_s=1$. The upper solid lines refer to the situations in which the equal fractions $b_1=0.5$ and $1-b_1=0.5$ of occupied bonds take the high-conductance $g=1$ with the relative noise $s=1$ and the low-conductance $g=h_g$ with the relative noise $s=h_s$; line A, $h_s=100$, $h_g=0.0001$; line B, $h_s=1$, $h_g=0.0001$; line C, $h_s=1$, $h_g=0.001$. All the lines show the same slope $-\kappa\simeq-1.87\pm0.05$ as p approaches p_c, which is our RSRG estimate of the universal value of $\kappa\cong1.5$. The broken lines drawn for $p\to p_c$ arise from shifting the lowest solid line by $\log_{10}[h_s/(1-b_1)]$ [see Eq. (6)]. The dotted lines refer to the theoretical NLB predictions given by Eqs. (8) and (10).

increases. Equation (8), however, predicts increasing S as p increases (see Fig. 2). This indicates that the local maximum on S versus p at the second percolation threshold p_c' could be observed. However, this maximum can only occur for the relatively small values of h_s. A large value of h_s shifts the critical-region part of S versus p [Eq. (6)] upwards, which masks the maximum. In this case Eq. (8) manifests itself by decreasing of the magnitude of the $S(p)$ slope as p approaches p_c' from below (see Fig. 2, line A). This results in much steeper decrease in S after p exceeds the percolation threshold p_c'. This feature basically differs the S-versus-p dependence from the σ-versus-p one. In the latter case the transition between the tunneling and the metallic regions is smooth because of the same type of the monotonicity of Eqs. (5), (7), and (9). So, if we look at the noise data of a three-component RRN, the separation between the tunneling- and metallic-conduction regimes is more readily made than if we look at the conductivity data. One should keep this conclusion in mind when analyzing the $1/f$-noise measurements of real metal-insulator composites.

V. IS A LOCAL MAXIMUM ON THE S-VERSUS-p RELATION POSSIBLE?

In the previous chapter the local maximum on the S-versus-p dependence for a three-component RRN has been theoretically predicted. This maximum occurs at $p=p_c'$ if the magnitude of the relative noise S originated from the critical behavior described by Eq. (6) is smaller than the magnitude of the relative noise originated from the off-threshold behavior. The latter one is described by Eqs. (8) and (10) in the vicinity of p_c', while exactly at p_c' it is approximated by[32]

$$S\sim C1h_g^{-\kappa/(q+t)}+Dh_sh_g^{-\kappa'/(q+t)}\ , \tag{11}$$

where C and D are constants. The smaller h_g for fixed h_s results in the greater relative noise given by Eq. (11) and does not influence S given by Eq. (6). This leads to the peak on $S(p)$ (see Fig. 2, lines B and C). On the other hand, increase of h_s with h_g held constant results in two noise enhancements given by Eqs. (6) and (11). However, the first enhancement is greater than the second one. In this situation the maximum on $S(p)$ will be masked (see Fig. 2, lines A and C). To verify our topological NLB analysis and to identify the h_g and h_s values for which the nonmonotonic behavior of S versus p is observed, we have performed the numerical simulations of three-component RRN. To estimate lattice conductivity σ and relative noise S, we have utilized the MC small-cell real-space renormalization-group (RSRG) method.[33,34] We have chosen the 3D cell of size $b=2$ shown in the inset of Fig. 1. To enhance the accuracy at the calculations, we have introduced two improvements. First, to rescale the probability p, we have applied a two-parameter transformation, which is simply a 3D version of the Nakanishi-Reynolds[35] two-parameter 2D transformation. Second, we have rescaled the whole joint distribution of lattice conductivity and the relative noise $P(\sigma,S)$ [the untruncated transformation of $P(\sigma,S)$], not only its average values.[33,34] The results are shown in Figs. 1 and 2. On this basis we can reproduce all the properties of the three-component RRN which we have previously extracted from our NLB analysis.

The three-component RRN proposed as a model of a class of the metal-insulator composites is a simple idealization of the current-carrying physical network formed in a real composite. In real structures, distributions of grain sizes, constriction diameters, tunneling distances, and areas, etc., all result in distributions of conductance and relative noise of the sintered and nonsintered conducting grains. These distributions are not simple δ functions. It seems, however, that they cluster around some average values (g_C,s_C) and (g_B,s_B), where g_C and g_B differ by several orders of magnitude. It is also possible that the distribution of the tunneling conductance is singular at $g=0$.[6] As already mentioned, all these effects, if taken into account in our RRN model, do not fundamentally change the σ-versus-p and S-versus-p dependencies described in the previous sections. However, they make even a rough estimation of the values of g_C, g_B, and, therefore, of $h_g=g_B/g_C$ very difficult, especially if this estimation depends on the parameters of the microscopic conduction model. Instead, a macroscopically based indirect identification of the parameter h_g is possible. Namely, one can compare variations of the conductivities of the real composite and those of the three-point

RRN with different values of parameter h_g in respective ranges of concentration p. For example, looking at the data in Ref. 13, we find for the RuO_2-based thick-film cermets a factor of about 4×10^6 in variation of the conductivity when the metallic filling fraction passes from 0.02 to 0.3. In the respective range of the concentration p conductivity of three-component RRN varies by about 6 orders of magnitude for $h_g=0.001$ (see Fig. 1) or by about 7 orders of magnitude for $h_g=0.0001$. Thus we identify for this composite $0.0001<h_g<0.001$. It should be noted here that the calculations of the conductivity of the model sintered and nonsintered contacts done by Vest[36] for the RuO_2-based thick-film cermets led him to the value $h_g\simeq0.001$, which agrees well with our estimate. Because of our special interest in RuO_2-based thick-film cermets,[3,7,11,17,18,37–39] we have performed our numerical calculations for $h_g=0.001$ and 0.0001 (see Figs. 1 and 2).

To answer the question "is the local maximum on the S versus p characteristic of any real composite possible?," an estimation of acceptable values of h_s is still needed. It is much more difficult than the h_g estimation. Microscopic mechanisms generating the $1/f$ noise are still not explained, and the calculations of the relative noise of the model intergrain contacts are possible only to the qualitative accuracy. For this reason, now we cannot answer the question whether the h_s value of order of 1, for which three-component RRN shows the local maximum on the S-versus-p relation, is physically acceptable or not. At this moment we would like to refer to some $1/f$-noise measurements in which nonmonotonic S-versus-p dependencies were observed. In 1979 one of us observed[39] such a dependence for the bismuth ruthenate thick resistive films; the identification procedure led us to the value $h_s<1$. Inokuma, Taketa, and Haradome[40] reported data for RuO_2 cermets indicating the nonmonotonic behavior of the $1/f$-noise versus conducting component concentration. Recently Bobran[13] has also measured the relative noise of the RuO_2-based thick resistive films. His measurements show the local peak on the S-versus-p dependence and we identify in this case $h_s\sim30$. It seems, however, that the latter data do not always confirm the inverse proportionality of the measured relative noise to the volume of the sample.

On the other hand, much larger values of h_s have recently been estimated.[32,41] Tremblay and Fourcade[41] and Morozovsky and Snarsky[32] considered a binary RRN composed of the bonds of the type (1,1) and (h_g,h_s). Assuming $h_s\sim h_g^{-1}$, they found[32] a value of the noise critical exponent, $\kappa\simeq6.4$. As an experimental confirmation of this result, Morozovsky and Snarsky[32] referred to the measurements on the carbon-wax model composites for which $\kappa=5\pm1$ have been found.[42] As we have mentioned above, we now refrain from any arguments that could distinguish between these two different estimations of parameter h_s. It is possible that the values of h_s, which are different for different composites, are not restricted to within such narrow bounds.

VI. CONCLUSIONS

We have performed an analysis of the topology and the electrical transport properties of three-component RRN. Such a network has already been proposed as a model for the metal-insulator composites[7,8,17,18,37] in which tunneling- and metallic-conduction regimes can be detected (e.g., via measurements of temperature coefficient of resistance). We have explained in a qualitative manner a basic feature of the three-component RRN, which has been reported earlier.[7,8] This feature is an evident increase of the conductivity exponent t if it is extracted from data lying over a broad range of the metallic concentration p. This increase is caused by the quasibicritical nature of the RRN under study. Furthermore, we have described in terms of the NLB picture of the percolating cluster the dependence of the $1/f$-noise intensity S on concentration p. Our NLB analysis indicates the possibility of a nonmonotonic dependence of S on p. We have found that a local peak on the S-versus-p dependence can occur if h_s (the ratio of the relative noise of poor and good intergrain contacts) is of order 1. We have not found any convincing physical arguments which can reject this value of h_s as being physically unacceptable. Thus we conclude that the three-component RRN could be useful in an explanation of the S-versus-p dependences which show a nonmonotonic increase of S as p approaches the percolation threshold p_c.

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