Finite coherence length of thermal noise in percolating systems

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(Received 27 March 2002; published 6 June 2002)

Noise has been measured in two types of conductor-insulator mixtures as a function of bias and composition. It was marked by a huge increase in magnitude as the resistance increased only slightly due to Joule heating. The noise (resistance) current scale $I_s(I_r)$ for nonlinearity were found to scale with the linear resistance R_0 as $I_s(I_r) \sim R_0^{-x_s(x_r)}$, where the exponent x_s is equal to 0.80 and 0.68 in carbon wax and carbon polyethylene, respectively, and $x_r \approx 0.5$. It is shown that the large increase of noise in nonohmic regime as well as the differences between the noise and resistance exponents are due to the finite-sized inequilibrium thermal fluctuations whose coherence length is same as the correlation length of the underlying percolating systems. A expression for x_s is derived.

The low-frequency resistance noise, also known as $1/f$ noise, is a very common phenomenon[1] in normal conductors and is increasingly being used as a tool for studying, particularly, disordered systems.[2] While its origin remains controversial, some of its features are well established. In particular, the correlation length of resistance fluctuations (ξ_s) is commonly assumed to be of the order of microscopic lengths.[1] This implies that the noise power should be inversely proportional to the number of fluctuators or the system volume L^d, d being the system dimensionality. But there are cases where relatively large increases in the noise levels under certain experimental conditions have been explained in terms of coarsening of the coherent volume with correlation lengths increasing to macroscopic scale. This, other things remaining the same, amounts to an increase in the noise level by a factor of $[L/(L/\xi_s)]^d = \xi_s^d$ with $\xi_s \leq L$. For example, the large increase in the noise at the onset of charge density wave motion in certain conductors upon application of an electric field has been ascribed to the finite coherence length of a charge density wave domain.[3] Analogous phenomena are the well-known critical opalescence in various systems[4] where the static correlation length diverges as a critical point is approached. Interestingly, in systems in nonequilibrium states but not necessarily near any critical point, the correlation length of fluctuations at equal time is also of macroscopic order.[5]

Earlier,[6] the resistance behavior of various composite samples (random mixtures of conductors and insulators) exhibiting substantial Joule heating was invesigated as a function of biasing currents. At low currents, the resistance $R = V/I$ had a bias-independent value R_0 but increased at high currents. It was found that the current I_r at which the sample resistance starts increasing scaled with R_0 as $I_r \sim R_0^{-x_r}$, where $x_r \approx 0.5$ is the onset exponent for resistance. In this paper, we report on systematic measurements of low-frequency resistance noise as a function of currents in two composites systems of carbon wax (C-W) and carbon polymer (C-HDPE). Samples under fixed currents were in nonequilibrium steady states (below breakdown) with temperature gradients. It was found that (i) the relative noise $\mathcal{S}$ (see below) behaved in a similar fashion as the resistance such

that the current scale I_s for onset of nonlinearity in noise[7] and the associated onset exponent x_s could be defined as before: $I_s \sim R_0^{-x_s}$. x_s was found to be larger than x_r:

$$x_s = \begin{cases} 0.80 \pm 0.03 & \text{(C-W)}, \\ 0.68 \pm 0.04 & \text{(C-HDPE)}. \end{cases} \tag{1}$$

(ii) The noise $\mathcal{S}$ of a sample as a function of resistance increased tremendously compared to that caused by varying static disorder (i.e., composition). It is argued that such an increase in noise results from additional resistance fluctuations induced by thermal fluctuations which are coherent over a length of the correlation length ξ of the underlying percolation structure i.e., $\xi_s \approx \xi$. It is shown that this assumption leads to an expression for x_s:

$$x_s = x_r + d\nu/4t - w_J/4, \tag{2}$$

where ν and t are percolation correlation and conductivity exponents, respectively,[8] and w_J is the noise exponent.[6] Using $d=3$, $\nu=0.9$ (Ref. 8) and experimental values of x_r, t, and w_J in Eq. (2) yields $x_s = 0.83 \pm 0.10$ and 0.67 ± 0.05 for C-W and C-HDPE, respectively. These values agree well with the experimental ones in Eq. (1).

Noise has been studied extensively in many percolating systems such as composites both experimentally and theoretically but mostly in Ohmic regimes.[9] Recently, the bias-dependent behavior of noise near the percolation threshold has been measured.[10] A composite sample is primarily characterized by its conductor fraction p. Above a particular value p_c, called the percolation threshold, a continuous path is formed from one end of the sample to another, enabling current to flow through the system. A sample possesses a natural length scale which is the two-particle correlation length ξ given by $\xi \sim (p - p_c)^{-\nu}$. The noise (in the Ohmic regime) tends to diverge[11] as the conducting fraction (p) is reduced from large values above p_c. However, this divergence has been explained to originate entirely from the static geometrical disorder.[12] ξ_s remains of the order of microscopic scales and independent of $\xi(p)$. Electrical conductivity of a composite sample is characterized by the exponent t,
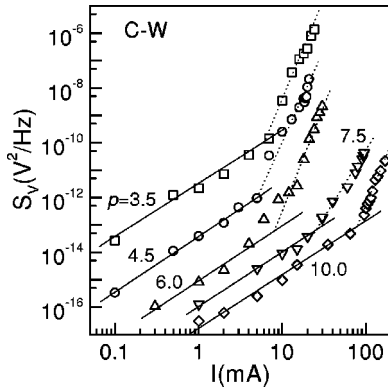
FIG. 1. Plots of noise power $S_V(0.5$ Hz$)$ vs dc current I in five carbon-wax samples with carbon percentage by volume (p) as marked. The solid lines are fits to a power law with all slopes equal to 2 and the dotted curves are only guide to the eye. For clarity, the data corresponding to $p=3.5$ and 4.5% have been shifted upward by factors of 100 and 10, respectively.

defined by $R_0 \sim (p-p_c)^{-t}$.[8] The resistance R_0 decreases with increasing p. The noise exponent w_J is defined by $\mathcal{S}(I \sim 0) \sim R_0^{w_J}$.

The carbon wax system is the same one as used earlier for noise measurements in the range $p \geqslant p_c$ (Ref. 10) but samples used in this work were in the Joule range $p > p_J$. The fraction $p_J > p_c$ is such that the resistance of a sample in response of an applied field increases if $p > p_J$. That the increase in resistance in this range of p is indeed due to the Joule heating has been confirmed by a number of observations[13] such as the sensitivity of measured values of resistances to the presence of a cooling fan near samples. For this system, $p_c = 0.76$% by volume, $p_J < 3$% and $t=2.05 \pm 0.15$.[14] C-W samples were all disk shaped, 10 mm in diameter, and 6 mm high. Connections to electrodes were such that current flowed parallel to the thickness of the disc. The preparation and characterization of C-HDPE samples have been described in Ref. 15. t has the value of 2.9 ± 0.1 in this system. These samples had dimensions $10 \times 4 \times 1$ mm^3. In this case, currents flowed parallel to the longest side. All measurements were done at room temperature at constant currents. For any constant current, sufficient time (more than 40 min) was allowed for the sample resistance to attain a steady value. Furthur details of noise measurements can be found in Ref. 10.

Let S_V denote the spectral power of voltage fluctuations at a constant current. Figure 1 shows plots of $S_V(0.5Hz)$ vs I for five C-W samples with p as indicated. At low currents (ohmic regime), S_V varied as I^2 (solid straight lines). At high currents, S_V varied faster than I^2 in the similar manner as V-I curves become nonlinear. Such concurrent nonlinear behavior of noise[7] and resistance becomes more evident in Fig. 2 where the normalized relative noise $\mathcal{S}$, defined by $\mathcal{S} = S_V/V^2$, (squares) and conductance $\sigma = 1/R$ (circles) are shown as functions of the current for the two C-W samples with $p=3.5$ (closed symbols) and 10% (open symbols). The conductance rather than resistance was used to avoid overlapping of data. The relative noise and the resistance of a sample have similar qualitative behavior in that both remain
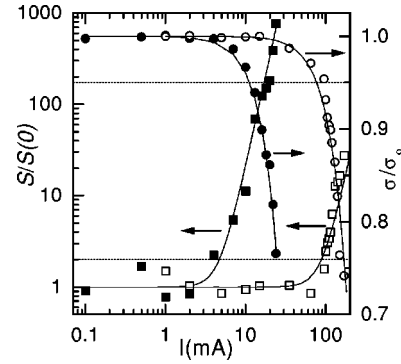

FIG. 2. Normalized relative noise power (squares) and conductance (circles) are shown as functions of I for two C-W samples with $p=3.5$% (closed symbols) and 10.0% (open symbols). The solid lines are fits to the functionals $\mathcal{S}/\mathcal{S}(0) = 1 + a_s I^4$ and $\sigma/\sigma_o = 1 - a_r I^2$ where $a_s(a_r) = 2.1 \times 10^9 (400)$ and 1.7×10^4 A^{-4} (8.5 A^{-2}) for $p=3.5$ and 10%, respectively. The dotted lines indicate the criteria for determining the current scales for onset of nonlinearity (see text).

close to their respective zero-bias values at small currents but appear to deviate from those values as the latter is increased. The currents I_s and I_r at which the onset of nonlinearity took place were determined by two independent methods. In the first method, they were determined by adopting the criteria that corresponded to the increase in noise by 100% and the decrease in conductance by 5%. These levels of changes are indicated by the dotted lines in Fig. 2. The criterion of 100% change of noise level was adopted to increase the accuracy of the determination of I_s as the noise increased steeply by several orders of magnitude while the conductance decreased only by an order of unity. As $p(R_0)$ increases (decreases), both I_s and I_r increase. Log-log plots of both I_r (circles) and I_s (squares) vs R_0 in the two systems are shown in Fig. 3. Straight lines indicate power-law fits with the slopes as indicated. In the second method, the currents for each sample was scaled with respect to I_s determined by trial and error such that the curves $\mathcal{S}(I)/\mathcal{S}(0)$ vs I/I_s of all samples of each
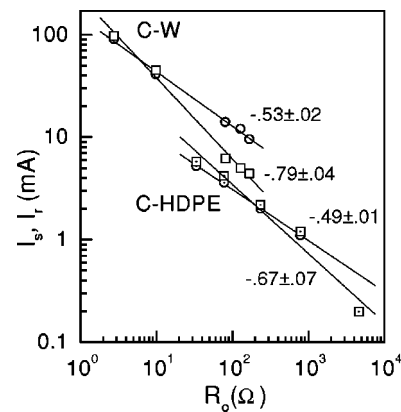

FIG. 3. Plots of nonlinearity current scales I_s (noise, squares) and I_r (conductance, circles) vs zero-field resistance R_0 for two systems. Data of C-HDPE samples have been shifted down by a factor of 5. The solid lines are the power law fits to the data with the exponents as indicated.
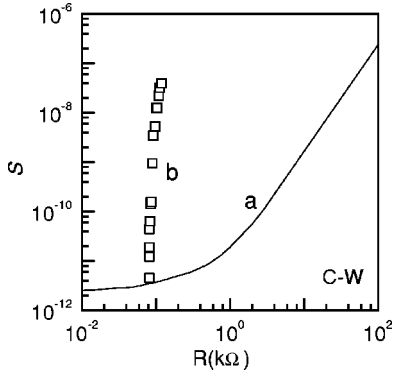
FIG. 4. Comparision of noise in the non-Ohmic regime with that in the Ohmic regime of a composite system. Relative noise is plotted against R for two cases: for curve (b), R is the bias-dependent resistance of a Joule sample ($p=10\%$) and for curve (a), R is the composition-dependent zero-bias resistance. High resistance data of curve (a) are taken from Ref. 10.

system collapsed on a single curve.[13] Similar procedures were followed with resistance data for I_r (see Fig. 3 of Ref. 6). This method yielded $x_r = 0.53 \pm 0.02$ and 0.45 ± 0.02 and $x_s = 0.81 \pm 0.03$ and 0.68 ± 0.04 in C-W and C-HDPE, respectively. Averages of x_s from the two methods are shown in Eq. (1) and averages of x_r were used in Eq. (2). w_J was obtained from the relative noise in Ohmic regimes $\mathcal{S}_0$. Its value in C-HDPE was found to be 0.13 ± 0.06.[6] But, the data of C-W had large scatter. As a consequence, a power-law fit yielded $w_J = 0.11 \pm 0.21$.[13] These values in Joule samples, significantly smaller than those near p_c, were used in Eq. (2) to obtain finally the predicted values as mentioned above.

The most significant feature of the nonlinear noise in the Joule regime is, of course, the huge increase in magnitude compared to the small change in sample resistance. This is further emphasized by plotting (curve b) the relative noise in a Joule sample (C-W, $p=10\%$) against the bias-dependent resistance R in Fig. 4 which also shows the variation (curve a) of the relative noise in C-W with static disorder characterized by the linear resistance $R \equiv R_0(p)$. Let us note that all measurements in this work were carried out in the steady and reversible states *below* breakdown[6] and hence, no change in the topology of percolating networks is expected. It is then clear that small changes in resistances of the network elements alone cannot account for the large increase of the noise level. Indeed we show below that temperature fluctuations which modulate resistances are responsible for this phenomenon. The local rise in temperature results in a change in the local resistance r determined by the temperature coefficient of resistivity $\beta = (1/r)dr/dT$. The final state under a constant current condition and a positive β is determined by the balance between the heat generated and the loss of heat to the environment per unit time.

The electrical response to Joule heating in percolating networks has been treated in detail by several authors.[6,16,17] In such a network, the changed resistance due to Joule heating, in the first approximation, is given by[17]

$$R = R_0 + a\beta h R_0^2 \mathcal{S}_0 I^2. \tag{3}$$

Here a is a simple constant. h is the heat transfer coefficient, defined as the ratio between temperature rise and power generated in a conducting element. $\mathcal{S}_0 = \mathcal{S}(0)$ is the relative noise power in the Ohmic regime (Figs. 1 and 2). Each solid line through conductance data in Fig. 2 is a fit to the expression $1 - a_r I^2$ according to Eq. (3). Here, $a_r = a\beta h R_0 \mathcal{S}_0$. The current scale I_r for the onset of nonlinearity in resistance is given by $I_r \sim (a\beta h R_0 \mathcal{S}_0)^{-1/2} = a_r^{-1/2}$.[6] It may be noted that the quantity h, as defined above, strives to provide, in the spirit of mean-field approach, a reasonable description of heat conduction in inhomogeneous media, which otherwise poses a formidable problem. Ultimately, it governs the process of exchange of heat between the sample and the environment[16] and hence, is quite susceptible to fluctuations.

Equation (3) formed the basis for obtaining the *linear* noise by the so called third harmonic measurements.[16] It will now be used to calculate the *nonlinear* noise, i.e., noise in the non-Ohmic regime. A small variation δR is given by $\delta R/R = (1 + a_r I^2 - 2a_r^2 I^4)\delta R_0/R_0 + a_r I^2(1 - a_r I^2)\delta h/h$ after neglecting variation of $\mathcal{S}_0$. Using this and assuming that fluctuations in the Ohmic regime R_0 are uncorrelated with those in h (or thermal fluctuations), i.e., $\langle \delta h \delta R_0 \rangle = 0$, it could be easily verified that $\mathcal{S} = \langle \delta R^2 \rangle / R^2$ is given by

$$\mathcal{S}/\mathcal{S}_0 \simeq 1 + 2a_r I^2 + (\langle \delta h^2 \rangle / h^2 \mathcal{S}_0 - 3)a_r^2 I^4 \tag{4}$$

after keeping terms up to the order of I^4. According to Eq. (3), $a_r I^2$ is equal to the fractional change of a sample resistance at current I which is of the order of 1 (see Refs. 6 and 18 for its maximum possible values). Since the relative noise, in contrast, increases in the same range of currents by several orders of magnitude (see Fig. 2) we must have $\langle \delta h^2 \rangle / h^2 \gg \mathcal{S}_0$ so that Eq. (4) reduces to

$$\mathcal{S}/\mathcal{S}_0 \simeq 1 + (\langle \delta h^2 \rangle / h^2 \mathcal{S}_0)a_r^2 I^4. \tag{5}$$

Fits to the noise data according to Eq. (5) with $a_s = (\langle \delta h^2 \rangle / h^2 \mathcal{S}_0)a_r^2$ are shown in Fig. 2. The values of a_s and a_r given in Fig. 2 yield $\langle \delta h^2 \rangle / h^2 \mathcal{S}_0 \sim 10^4$ to 10^2 in the range of 3.5–10% of p, much greater than 1. Goodness of fittings proves the earlier assertion that it is the secondary (i.e., thermal) source of noise that is responsible for a huge increase in noise level in the nonlinear regimes in Joule samples. The primary source is, of course, the one responsible for $\mathcal{S}_0$. Let us now consider the possible origin of such large magnitude of the thermal noise.

The inhomogeneous nature of percolating networks give rise to randomly occuring "hot spots" which are the regions carrying high current densities and get heated most. Consequently, thermal gradients result in steady heat flows away from those spots, thus setting up nonequilibrium steady states. A percolating system according to the "Node-Link-Blob" picture[8] could be viewed as a homogeneous system on the scale of the correlation length $\xi(p)$. Links, i.e., singly connected bonds in this picture are the hot spots. With such multiple sources within the length $\xi(p)$ and a length scale $\xi(p)$, one has to deal with the question of a multitude of correlation lengths[19] that the noise due to thermal diffusion normally possesses. We assume that a volume of ξ^d will fluctuate thermally in a coherent manner, i.e., thermal ξ_s will

simply snap onto ξ. This means that $\langle \delta h^2 \rangle / h^2 \sim \xi^d$. Such coarsening of coherent length in inhomogeneous systems explains the large increase in the thermal noise. It is quite instructive to compare thermal fluctuation $\langle \delta T^2 \rangle / T^2$ in the Joule regime with that in equilibrium. The latter at room temperature is roughly inversely proportional to the number of particles in a sample, i.e., 10^{-20} whereas the former is approximately equal to $\langle \delta h^2 \rangle / h^2 \sim 10^{-8}$ to 10^{-10} with $\mathcal{S}_0 \approx 10^{-12}$ at 0.5 Hz. From Eq. (5), the current scale for non-linear noise is given by $I_s^{-1} \sim (\langle \delta h^2 \rangle / h^2 \mathcal{S}_0)^{1/4} a_r^{1/2}$, which, upon using $\xi \sim R_0^{\nu/t}$ and $a_r = I_r^{-2}$, yields the relation (2). Note that the latter predicts $x_s - x_r$ to have theoretical values of 0.34 and 0.5 in 3D and 2D, respectively, ignoring w_J and hence, the exponents to be more divergent in two dimension.

In conclusion, we extended experimental study of noise to two unusual conditions of non-Ohmic (without any threshold) conduction and inequilibrium states in a disordered system of composites. The noise was shown to involve two distinct sources with different coherence lengths. In the present case, the secondary thermal noise was induced by the Joule heating and became dominant in the non-Ohmic regime due to amplification by coarsening of the coherence length. This points to possible further use of noise study as a tool in systems such as manganites[20] which exhibit multiple phases having very likely different fluctuation properties.

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