1/f noise in nonlinear inhomogeneous systems

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$1/f$ noise has been measured in detail in a composite system of carbon-wax mixtures using a range of bias (V) that covered both linear and nonlinear regimes. S_V-V characteristics have been obtained and exhibit structures that can be correlated to those of conductance. It is found that the variation of $\mathcal{S}$ either by the conducting fraction or V is described by the same power law, R^{ω}, but with different exponents 1.7 ± 0.2 and ~3, respectively. Here $\mathcal{S}$ is the relative noise power and R is the chordal resistance. The noise data are consistent with results obtained earlier with I-V or dI/dV measurements. It is shown that the data can be explained by the addition of tunneling bonds as a result of applying voltage. Similar measurements have been carried out for comparison in samples of conducting polymer and V_2O_4 which follow Mott's variable-range hopping conduction and, thus, are different from mixtures. Differences in noise behavior with respect to the composites point to the usefulness of extending noise measurements to nonlinear regimes in other physical systems. [S0163-1829(96)01142-3]

I. INTRODUCTION

Resistance fluctuations have been measured in diverse physical systems, both homogeneous and inhomogeneous. A common property of this type of noise, irrespective of the nature of a given system, is that the power density varies as $1/f^{\lambda}$ in the frequency (f) domain where λ is order of unity (hence the name $1/f$ noise, also known as flicker noise). Apart from this $1/f^{\lambda}$ variation, the power spectrum is remarkably featureless. Homogeneous systems (e.g., pure metals or semiconductors) are ideal for studying fundamental properties of $1/f$ noise.[1,2] On the other hand, inhomogeneous samples provide oppurtunities for studying new features of $1/f$ noise in an enlarged space of additional variables like microstructural parameters, applied bias or current, etc., other than the usual frequency. Differences between the two kinds of systems with reference to noise have already been recognized.[3] Consider a generalized version of Hooge's empirical formula[1] for the relative noise power,

$$\mathcal{S}=\frac{S_R}{R^2}=\frac{S_V}{V^2}=\frac{S_I}{I^2}=\frac{V^{\gamma_o}}{f^{\lambda}}\mathcal{R}(R), \tag{1}$$

where $S_X=\langle\delta X^2\rangle$ is the spectral density when $X=R,V,I$ is the fluctuating variable. I is the current through sample when V is the applied dc voltage. R is the resistance defined as V/I. The value of the exponent γ_o is believed to be indicative of whether the noise is a driven phenomenon ($\gamma_o\neq0$) or a result of equilibrium resistance fluctuations ($\gamma_o=0$).[1] All the above relations are assumed to hold at a particular frequency. The form of the function $\mathcal{R}$ depends upon the particular system under consideration. In homogeneous samples, $\mathcal{S}$ is usually proportional to the resistance R so that

$$\mathcal{S}\sim\mathcal{R}\sim R. \tag{2}$$

The noise amplitude in a inhomogeneous system is known to exceed greatly that in a homogeneous system of comparable resistance. This has been seen in artificially fabricated films.[4] This means that the function $\mathcal{R}$ as given by Eq. (1) must contain quantities that strongly depend upon the details of microstructure of the system. For example, measurements[5] of noise in discontinuous films show

$$\mathcal{S}\sim\mathcal{R}\sim R^{\omega}, \tag{3}$$

where ω is close to 2. The physical origin of such different R dependence lies in the nature of the distribution of fields which are random in inhomogeneous samples due to random microgeometry, in contrast to being ordered in homogeneous samples.

Examples of studies of the flicker noise in inhomogeneous systems found in the literatures include composites of conductors and insulators,[6,7] solid-state devices,[8] sliding charge density wave (CDW) systems,[9,11] granular matalerials such as carbon resistors,[12] cermet thick films,[13] ZnO varistors,[14] and conducting polymers.[15] A strong motivation for all these studies has been the possibility of using $1/f$ noise measurements as a probe for further understanding of the underlying complex conduction mechanisms in those systems. A common feature of the inhomogeneous systems is nonlinearity in conduction at a sufficiently large bias where the resistance is no longer independent of the applied bias or current. Therefore, a study of electrical transport including noise, to be complete and more useful, should cover both linear and nonlinear regimes in inhomogeneous or disordered systems.

In this paper we will be primarily interested in the properties of the $1/f$ noise in nonlinear systems. We present detailed results obtained in carbon-wax composites.[16,17] Limited data obtained in the conducting polymer[18] (CP) and vanadium dioxide (V_2O_4) systems are also presented for comparision. All these samples show nonlinear conduction at room temperature as a function of the applied voltage. We pay special attention particularly to those features which are characteristics of noise in nonlinear regimes. Nonlinearities may arise in either of two possible ways: existing conduction mechanism being affected by the driving bias or new con-

duction channels coming into play as a result of the applied bias. Thus, it is expected that the behavior of the $1/f$ noise in the nonlinear range will be strongly influenced by the particular mode of the conduction mechanism that comes into play and, hence, its study should be useful to gain a better understanding of transport properties in general. Apart from nonlinear solid-state devices whose noise characteristics have been measured primarily for practical reasons, there are very few nonlinear physical systems whose noise properties have been studied systematically. CDW systems[9–11] provide an interesting example of nonlinear transport that has been studied by several groups. ZnO varistors have been also studied in nonlinear regime.[14] These studies point to several questions that relate specifically to description of the noise in nonlinear regimes: (1) In a nonlinear system the chordal resistance $R=V/I$ is generally different from the differential resistance $R=dV/dI$. Which R is relevant in the noise definition? (2) Is there any relation between $\mathcal{S}(V)$ and $R(V)$? (3) Do the fluctuations retain their statistical properties in the entire range of applied bias? We aim to address these issues in the discussion of our results except the third one which will be treated elsewhere.

Transport properties of composite systems have been the subject of many theoretical[19–22] and experimental investigations[23–27] for a variety of reasons (see Ref. 6 for further references). Apart from its practical values, a composite system is usually modeled by an ideal random resistor network (RRN) that offers an excellent opportunity for studying theoretically the current or voltage distribution leading to multifractality[6,19] in an inhomogeneous system. A mixture of conductors and insulators is characterized by the conductor fraction p. The length scale in such a percolative system is set by the correlation length having an exponent ν. The latter, together with the percolation probability exponent β, determines the geometrical properties of the conducting clusters.[28] Transport properties require different exponents t and κ. The resistance of a composite system tends to diverge as p approaches from above a critical value p_c, called the percolation threshold: $R\sim\Delta p^{-t}$ where $\Delta p=p-p_c$.[28] It has been found[19,20] that the relative noise amplitude $\mathcal{S}$ in a percolating system also diverges as the threshold is approached from the conducting side. Expressed in terms of experimentally more accessible resistance, the noise is given by

$$\mathcal{S}\sim\Delta p^{-\kappa}\sim R^{\omega}, \tag{4}$$

where $\omega=\kappa/t$. Numerical simulations[21] on discrete random lattices obtained $\omega\approx 0.87$ and 0.75 in $d=2$ and 3, respectively. However, it was soon realized[21,23,29] that to make comparision with experimental results meaningful required taking into account the continuum nature of real samples. This is due to the fact that the bond resistances in continuum models are given by a power-law distribution rather than a flat one in the discrete lattice model and that the noise is expected to be more sensitive to microstructure than macroscopic resistance.[30] Two models—random void and inverted random void—were considered. The random-void model consists of a conducting matrix with insulating holes and the inverted random-void model has an insulating matrix with conducting holes. Tremblay *et al.*[21] predicted that $\omega\approx 3.2$ and 2.1 in the random-void model, in two and three dimensions, respectively, while in the inverted random-void model $\omega\approx 0.87$ and 2.4 in two and three dimensions. On the experimental side, there seems to be substantial evidence (including present work) that $\mathcal{S}\sim R^{1.7-2.1}$ holds in *both* two (gold or silver films) (Ref. 5) and three dimensions (carbon-wax) (Ref. 25) in contradiction to the theoretical predictions. This value of ω is close to that in the random-void model in $d=3$ but is very different from the predicted values of two continuum or the discrete lattice models in $d=2$. Even in those cases where the values of ω have been found to be close to some predictions of the two models [$\omega=0.9$ and ≈ 3.4–6 in film,[5,26] $\omega\approx 3$ in AgPt-TFE (Ref. 23)], the physical basis for choosing one model over another is not clear. Several authors[5,25,27] have mentioned tunneling as a probable source for discrepencies between experimental and theoretical values of ω, although not always using convincing arguments.[2] Mantese *et al.*[27] suggested a two-component model that took into account explicitly an additional component from tunneling conduction in explaining the measured noise in metal-Al_2O_3. Recently, there have been suggestions[31] that discrepencies between experiments and theories may originate partly from the strong non-Gaussianity in samples particularly close to p_c. However, no quantitative results are available yet for estimation of effects of the non-Gaussianity on the noise exponents.

The discussion in the last paragraph is actually valid for a linear network only. The literature contains references to discussions of the noise in at least two networks consisting of explicitly nonlinear elements.[32–34] Kenkel and Straley[32] considered a class of resistors whose I-V characteristics are given by

$$V=r|I|^{\alpha}\mathrm{sgn}(I), \tag{5}$$

where $\alpha>0$. Rammal and Tremblay[33] showed that the noise in such a network is determined essentially in the same manner as in the linear network except that κ (and other multifractal exponents) now become a function of α. Notice that in this case the chordal resistance is same as the differential resistance except for a constant factor. The analysis, however, used a generalized resistance defined as V/I^{α} which is proportional to the chordal resistance at a constant current. Another approach is to consider elements each one of which possesses a linear component and a cubic nonlinearity:[34]

$$i=\sigma_1 v+\sigma_3 v^3, \tag{6}$$

where i is the current flowing through a conducting bond and v is the applied bias accross the bond. σ_1 and σ_3 are coefficients of the linear and nonlinear terms, respectively. A smaller σ_3 corresponds to weak nonlinearity and a smaller σ_1 to strong nonlinearity. It was shown that the noise power in the linear regime in this model is related to the average value of the cubic (i.e., nonlinear) coefficient, $\langle\sigma_3\rangle$. But the problem of the noise in the nonlinear regime was not considered. One example of this model is the small nonlinearity caused by the joule heating in a disordered medium. An experiment[4] has been performed on an artficially generated random resistor network using this principle. Except for the heat-induced nonlinearity, we are not aware of any other

TABLE I. Parameters of carbon-wax samples. R_0 is the linear resistance. $2+\gamma_o$ is given by $S_V \sim V^{2+\gamma_o}$ at low bias. ω_V is given by $\mathcal{S} \sim R^{\omega_V}$ for a given sample. S_V and $\mathcal{S}$ are noise power at a constant current and relative noise, respectively. R is the chordal resistance.

$R_0(\mathrm{k}\Omega)$	$2+\gamma_o$	ω_V
40	1.7	5.9
65	2.0	3.2
92	1.7	1.7
200	1.8	2.8
450	1.5	2.1
600	1.7	3.9
700	1.3	4.5
900	2.0	4.6
1300	1.6	2.6
2000	1.8	2.0

experimental system which has been interpreted by either of the two nonlinear models mentioned just above.

A systematic study of the flicker noise in composites covering both linear and nonlinear regimes is presented here. For comparision, measurements were also carried out in two other disordered systems like conducting polymer and V_2O_4 which are known to have Mott's variable-range hopping conduction and offer examples having conduction mechanisms different from those in composites. After the present review of $1/f$ noise in inhomogeneous systems as currently available in the literature, we present experimental details in Sec. II and results in Sec. III. The results exhibit several pecularities of inhomogeneity. The noise continues to increase even though the resistance keeps on decreasing as a function of the applied bias. The behavior of noise as a function of the applied bias is closely related to that of the resistance. Section IV contains a discussion of the results. Comparisions with relevant previous works are made and existing models are critically judged and adapted to explain the qualitative features of the results. Finally an overview of some issues related to the noise in nonlinear regimes is made in Sec. V, particularly highlighting the unresolved ones.

II. EXPERIMENTAL

Low-frequency excess noise power was measured on disk-shaped (10 mm in diameter, 2 mm in height) carbon-wax samples under dc excitation by sending constant currents at room temperature. The samples used for noise measurements are similar to the ones as in Ref. 16 where further details can be found. This system has a low value of p_c of 0.76% by volume and an exponent t equal to 2.1. Resistances of the samples chosen in the present study ranged from a few tens of kΩ to MΩ (see Table I). The powdered mixture of ferric chloride (0.1 mol) doped polypyrrole was pelletized in disk-shaped samples under a pressure of 7 ton/cm^2. These conducting polymer samples exhibited variable-range hopping conduction.[18] Pellets of powdered vanadium dioxide of the same shape have also been prepared using the same technique. All measurements were made using pressed circular brass electrode contacts in a two-probe configuration to avoid capacitive coupling among different segments of the sample. These contacts have resistances typically less than 0.1% of the sample resistances used for noise measurements. To reduce extraneous noise sources due to thermal drift and electrical pickup, the sample holder was kept inside a properly grounded enclosure made of copper. A constant current from a programmable constant-current source (Keithley, model 224) was passed through the sample. The corresponding voltage fluctuations, measured using a digital multimeter with an integral time of 16.66 msec, were stored in a personal computer at an interval of 250 msec. Data were taken in sets of 1024 data points which facilitated performing of fast Fourier transforms. The resulting power spectra (512 points) were typically averaged over 10–20 sets at each current level. These steps were repeated for different currents (typically 20) for a particular sample. The noise spectral density was obtained in the frequency range from 7 mHz to 2 Hz and at dc voltages up to 25 V. To examine it as a function of the applied bias, an average noise at 0.5 Hz was used throughout this work. For this purpose, the noise power was first averaged over all data sets at a fixed current and then obtained as the average over a band of five frequencies (between 0.492 and 0.507 Hz, both inclusive) centered around 0.5 Hz. Samples were monitored to detect any possible (linear) resistance drift during an experiment by switching between the currently applied bias and a low bias. Such monitoring was, however, practicable only before a sample reached the ''saturated state.'' Otherwise, currents were increased in steps monotonically. After every change in current, sufficient time was allowed to pass for the sample to stabilize before data were acquired again. At a very high sample current a drift was observed. Results reported here were obtained with samples at voltages where there was no or negligible drift in the linear resistances.

It should be mentioned that aliasing can occur in any discrete sampling method such as the present one, thereby introducing some error in the spectrum power. The error at 0.5 Hz due to the power at 3.5 Hz is estimated to be about 7%. However, any antialiasing measure was considered to be unimportant in the context of the present work where the focus is on the variation of noise power with the applied bias rather than with frequency, and the effect of aliasing at a particular frequency is expected to be independent of the applied bias. The noise level of electronic devices used in these experiments was compared to that of the sample and found to be less by at least five orders of magnitude. The noise from the contacts was expected to be insignificant compared to that of the sample noise as the contact areas were large. The order of magnitude of the noise levels in a sample was same when measured by applying a constant voltage across the sample and recording the corresponding current fluctuations. Voltage fluctuations were also recorded by sending a constant current through the sample drawn from a battery and having a wire-wound ballast resistor connected in series, and the same level of noise power was observed.

III. RESULTS

Figure 1 shows typical noise power spectrums $S_V(f)$ in a carbon-wax sample with linear resistance $R_0 = 880$ kΩ as a function of frequency f at three different voltages. Each curve has a basic $1/f$ feature in that the slope marked on the curve is of the order of 1. The same is true for samples of

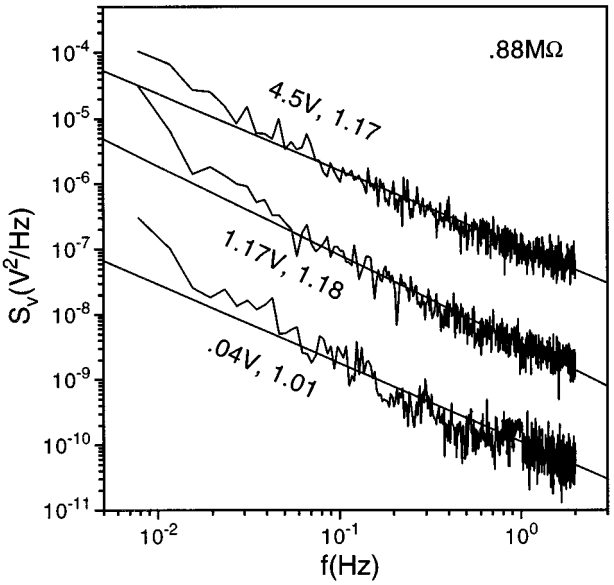

FIG. 1. Log-log plot of three noise spectral power $S_V(f)$ vs f for a sample of resistance 0.88 MΩ at three voltages (V). Each solid line is a fit to a power law. The applied voltage and the corresponding value of the power-law exponent are indicated on each curve.

different resistances (i.e., p). Slopes in various samples were within the range of 0.9–1.3. No apparent correlation between slopes and sample resistances or applied voltages in the range studied could be detected.

Figure 2(a) shows the voltage dependence of the excess noise power at 0.5 Hz (solid symbols) and the resistance ($R=V/I$, open symbols) of a carbon-wax sample with $R_0=92$ kΩ. The error in S_V may arise from two sources: The first one is statistical, the relative error being equal to $1/\sqrt{5N}$, where N is the number of data sets at a particular current and is between 10 and 20; the second one is current-to-current fluctuation in a given sample. The magnitude of the statistical error is about the size of a symbol in Fig. 2(a), whereas the second error could be guessed only from the data and is perhaps the dominant one in the present case. It is evident that the noise curve is quite structured. Thus, the determination of the structure will be limited by the finite number of voltages at which noise has been measured. The noise data seem to have three identifiable regimes indicated roughly by two labeled arrows and characterized by different slopes. In view of the nature of the S_V-V curves in Figs. 2–4 let us define a quantity γ given by

$$2+\gamma=\frac{d\ln S_V}{d\ln V}. \tag{7}$$

The first regime at voltages less than V_o represents a power-law relation, indicated by the solid line with a slope of 1.73 ($\gamma_o=-0.27$). Clearly, γ is equal to the constant γ_o in the first regime. The second regime between V_o and V_s is distinguished by the fact that the noise in this regime increases less rapidly than in the first regime. The voltage V_o at which the departure from the power-law behavior occurs is seen to be close to the onset of nonlinearity wherefrom R starts deviating rapidly from its linear value. The third regime at

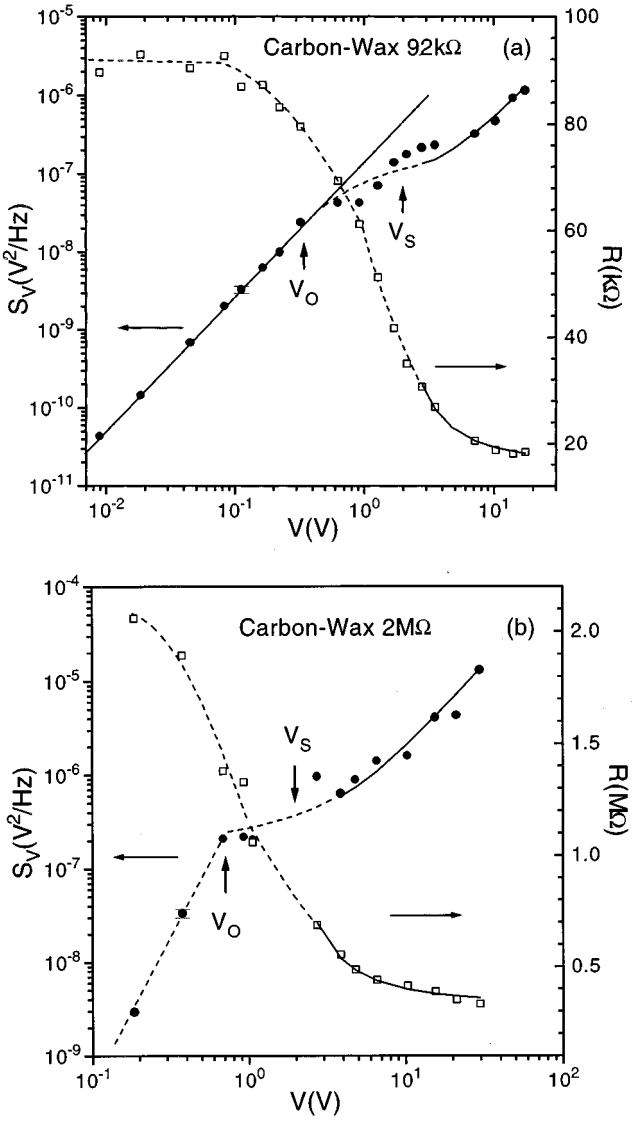

FIG. 2. Plots of S_V vs V for two samples of linear resistances as shown in (a) and (b). The arrows roughly delineate three regimes as discussed in the text. V_o and V_s denote onset and saturation voltages for the nonlinear conductance. The solid lines are fits as discussed in the text. Dashed curves are only guides to eyes. Typical statistical error in S_V is as indicated.

voltages greater than V_s is rather small in the figure but corresponds to one where the noise curve assumes an overall concave shape with the slope being larger than that in the second regime. The value of V_s is of the order of voltages at which dI/dV was found to approach saturation.[17] This suggests correlation of the noise power S_V with the nonlinear conductance. Let us denote γ in the third regime by γ_s which is clearly less than γ_o. Similar data as in Fig. 2(a) of an another sample with $R_0=2$ MΩ are shown in Fig. 2(b) where the first regime is less and the third regime is more extensive than those in Fig. 2(a). Here again the indication is that $\gamma_s<\gamma_o$. The limited range of the third regime and fluctuation in the data make it very unreliable to conclude from the data alone whether γ_s is constant (i.e., power law) or is a function of V. Later it is shown that γ_s is indeed a function of V. $2+\gamma_o$ along with R_0 of some samples used in the

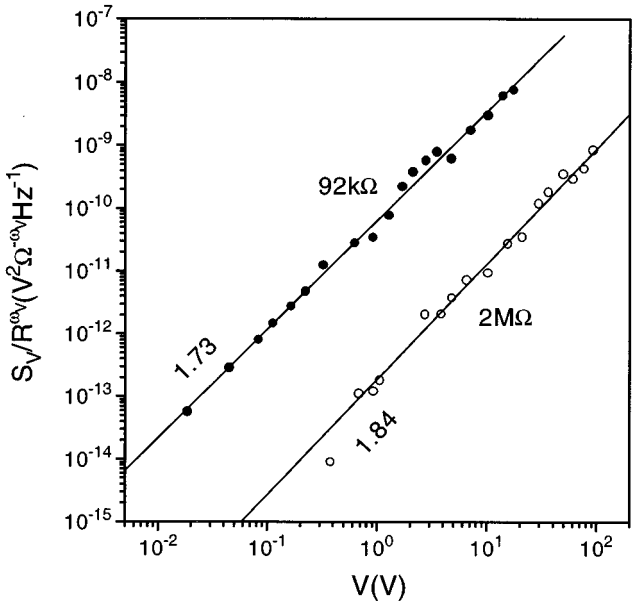

FIG. 3. Log-log plot of normalized noise power $S_V(f)/R^{\omega_V}$ vs V. Slopes are as indicated. Values of ω_V are given in Table I.

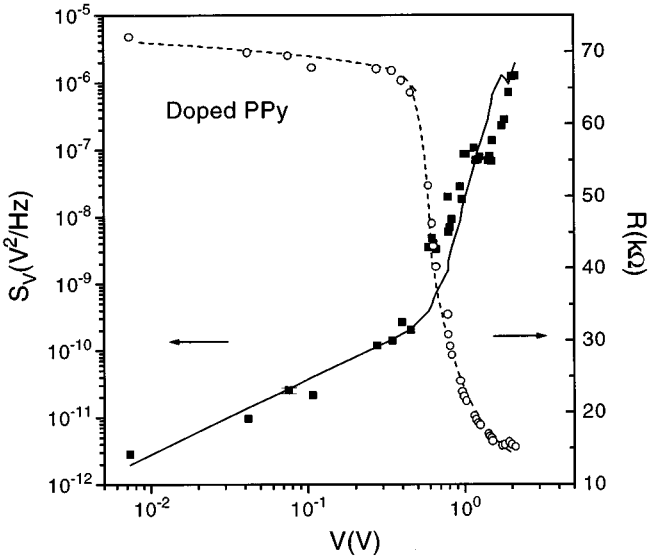

FIG. 4. Log-log plot of S_V vs V for conducting polymer of linear resistance as shown. The solid line is a fit as discussed in the text. The dashed curve is only a guide to eyes. Typical statistical error in S_V is as indicated.

present work is presented in Table I. $2+\gamma_o$ has an average of 1.7 ± 0.2.

In Fig. 2 it is seen that even while the resistance continues to decrease as the applied bias is increased beyond the onset voltage, the absolute noise power keeps on increasing, albeit slowly. This behavior is contrary to that in homogeneous systems. This gives rise to the possibility that the noise suitably normalized by some function of the varying resistance may be described by a single function. That it is indeed so is shown in Fig. 3 where the same S_V of Fig. 2 is plotted after being normalized by R^{ω_V}. Values of ω_V used for these two samples as well as for others are given in Table I. The distinct structures in Fig. 2 are no longer there and the data of each sample fall in a single line. This means that for composites, the noise of a sample at a *fixed* p as a function of the bias can be written as

$$S_V \sim V^{2+\gamma_o} R^{\omega_V}. \tag{8}$$

Equation (8) holds in *both* linear and nonlinear regimes. In practice, in cases where γ_o could be reliably determined as in Fig. 2(a), $S_V/V^{2+\gamma_o}$ was fitted to R^{ω_V} by treating the power ω_V as a fitting parameter. This ensured that the log-log plot of S_V/R^{ω_V} vs V has the same slope as $2+\gamma_o$. In other cases [e.g., the sample with $R_0 = 2$ MΩ in Fig. 2(b)] where γ_o cannot be reliably determined, the power ω_V was found by trial and error. In such cases $2+\gamma_o$ was assumed to be equal to the slope of a plot such as shown in Fig. 3. Fits for most of the samples were good but values of ω_V were found to have some degree of scatter as evident in Table I. ω_V has an average of ~ 3 (excluding the value of 5.9). Generally, ω_V is larger than $2+\gamma_o$.

Figure 4 shows similar data as in Fig. 2 but obtained in a conducting polymer sample. The noise data are seen to have only two distinct regimes characterized by two different slopes. However, in this case the noise at large bias increases more rapidly than at lower bias in contrast to the situation in carbon-wax. However, as in the latter, the voltage at which this change in slope occurs coincides with the onset of nonlinearity. The data after the onset of nonlinearity cover more than four orders of noise power and are tentatively assumed to be governed by a power law. We have $\gamma_o \approx -0.9$ and $\gamma_s \approx 1$. The data in vanadium dioxide are similar to those in conducting polymer. In this case γ_o and γ_s are about -0.9 and 0.5, respectively. Significant deviation from the quadratic dependence of the noise power on the applied voltage is thus found even at low voltages (linear regimes). The dashed curve represents the best fit of the entire S_V-V to a function

$$S_V \sim V^{2+\gamma_o} \exp[a(R_0/R)^b], \tag{9}$$

where $a \sim 0.6$ and $b \sim 1.8$. The particular choice of the fitting function in Eq. (9) was prompted by the fact that while the resistance decreased by a factor of 5 only, the noise amplitude increased by more than four orders of magnitude. It is worthwhile to remember that with the fitting function involving an exponential, the fitting parameters a and b are very susceptible to any noise in the data. Hence the values should be treated with some caution. For the purpose of comparing noise behavior of the three systems, we plot the generalized relative noise power $\mathcal{S}' = \mathcal{S}/V^{\gamma_o}$ at 0.5 Hz normalized by its value in the linear range as a function of V as shown in Fig. 5. Differences and similarities among the three systems in the nonlinear range are clearly brought about in this particular plot.

Figure 6 shows plots of S_V vs V for several carbon-wax samples of different resistances (i.e., p). It is evident that the variation of the noise with R in the nonlinear regime is much less than that in the linear regime. The log-log plots of the noise spectral density S_V vs R_0 are shown in Fig. 7 for various samples at 0.1 and 5 V as marked. Over a wide range of linear resistances R_0, the data at 0.1 V (open circles) are fitted by a power law with the exponent $\omega = 1.7 \pm 0.2(4)$. Since $\omega = \kappa/t$ and $t = 2.1$,[16] $\kappa \approx 3.6$. Corresponding values in

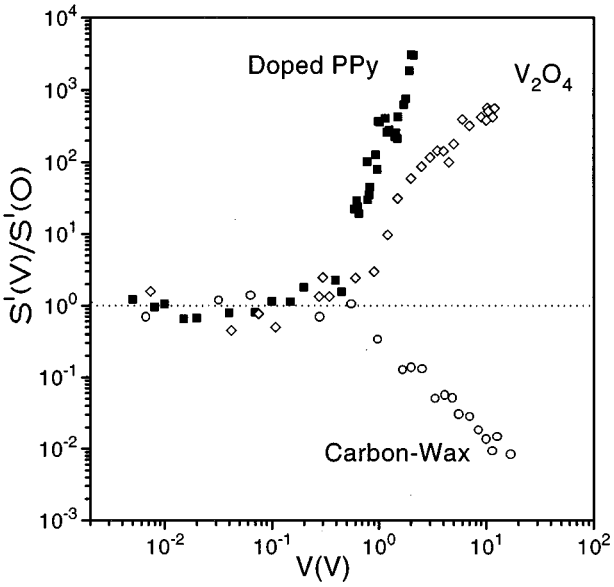

FIG. 5. Plots of normalized $\mathcal{S}'(=\mathcal{S}/V^{\gamma_0})$ vs V of three types of disordered systems.

the saturated state are $\omega_s \approx 0.5$ and $\kappa_s \approx 1.1$. It may be mentioned that no substantial difference was noticed using the noise power at another frequency such as 1 Hz.

IV. DISCUSSION

A. General

Data presented so far indicate quite unambiguously that while the noise spectral density in the frequency domain remains insensitive to a change in the conduction behavior (Fig. 1), it as a function of the applied bias does show a

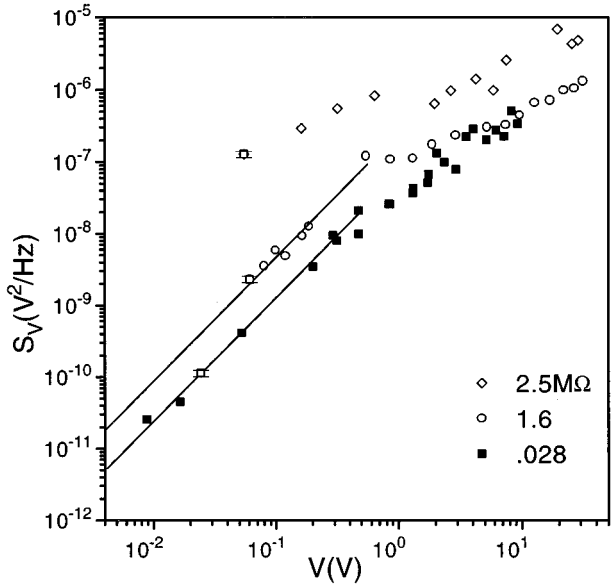

FIG. 6. Plots of S_V vs V of three carbon-wax composites of linear resistances as shown against each curve. The solid lines indicate the power-law fits up to the onset voltage. Typical statistical error in S_V is as indicated.

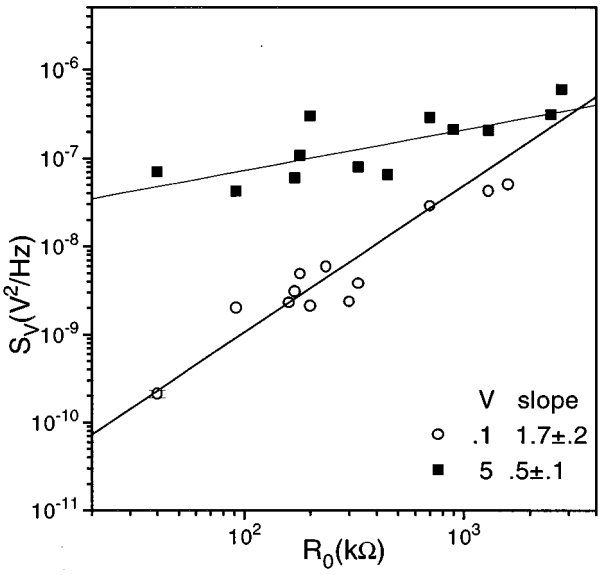

FIG. 7. Log-log plot of noise power S_V(f) at 0.5 Hz vs initial sample resistance R_0 for $V=0.1$ and 5 V. The straight lines indicate power laws with exponents as indicated. Typical statistical error in S_V is as indicated. However, sample-to-sample fluctuation is much larger than the statistical error in a single sample.

characteristic dependence (Figs. 2–5). Dependence of S_V on V, on the one hand, is different in linear and nonlinear regimes of a given sample and, on the other, is also different among various systems with different conduction mechanisms. This supports the expectation that the noise in the whole range of applied bias could be useful as a probe of transport properties in disordered materials. Let us first consider the conclusions that follow from general considerations. Note that the identities in Eq. (1) still remain valid provided R is the chordal resistance, V/I. As long as a system is in linear regime (i.e., low V), R and, hence, the generalized relative noise $\mathcal{S}'$ are constant, i.e., independent of applied bias. In real samples γ_o may have a nonzero value. When $\gamma_o=0$, $\mathcal{S}'=\mathcal{S}$. Both $\mathcal{R}$ and R are expected to be smooth functions of their arguments, namely, R and V, respectively. Then, it follows from Eq. (1) that initially at low voltages S_V will be dominated by the strongly varying factor of $V^{2+\gamma_o}$ (provided $2+\gamma_o$ is not small). As the applied bias is sufficiently increased, nonlinearity in conduction will be more pronounced. Naturally, the mechanisms that are responsible for the nonlinear effects will also cause noise to deviate from the power-law behavior in the linear regime. Hence, correlation of the noise with the onset of nonlinearity as seen in Figs. 2 and 4 appears quite natural. Generally, $S_V(V)$ will have characteristics closely related to those of $R(V)$. This has been amply demonstrated in systems like CDW conductors such as TaS_3 (Ref. 10) and organic conductors.[35] From Eqs. (7) and (1) we can obtain a general expression of γ:

$$\gamma=\gamma_o+\frac{V}{\mathcal{R}}\frac{d\mathcal{R}}{dR}\frac{dR}{dV}. \tag{10}$$

At low bias $\gamma \approx \gamma_o$. In all our samples, $dR/dV<0$; i.e., the resistance decreases with bias. Thus, for $V>V_o$, depending on the sign of $d\mathcal{R}/dR$, γ_s will be less or greater than γ_o.

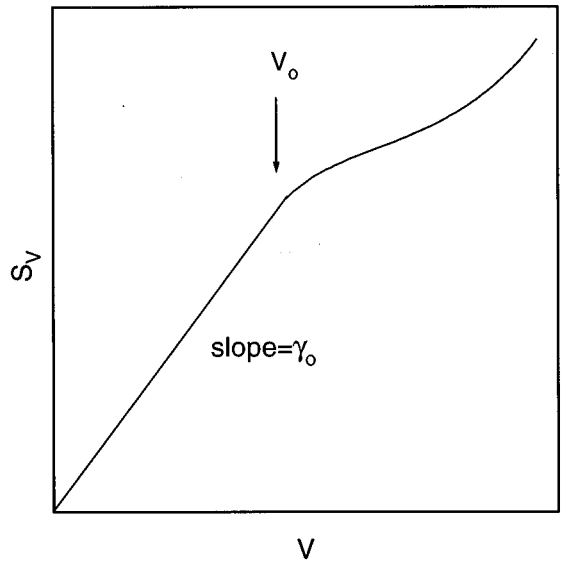

FIG. 8. Schematic plot of a type of S_V-V curve whose slope decreases at the onset voltage V_o. See text for further discussion.

γ_s will be, in general, a function of V. Figure 8 shows schematically a situation where γ decreases after the onset of nonlinearity ($V \sim V_o$) and then keeps increasing, giving the S_V-V curve a concave appearance for $V > V_o$. Obviously, it is the nature of the conduction mechanism that will determine the exact nature of the curve after the onset of nonlinearity. Differences between composites and other two systems as seen in Figs. 2, 4, and 5 are indeed due to different conduction mechanisms in these systems. Composites are prototypes of a percolation phenomenon.[28] It is generally believed that nonlinear conduction in composites results from tunneling between dangling clusters.[36,16,37] In contrast, conduction mechanisms in the other two systems are described by variable-range hopping,[18,38] but the mechanism for nonlinearity remains ambiguous.

From Eq. (1) we have $S_V \sim V^{2+\gamma_o}$ in the linear regime. On the average, γ_o is found to be ~ -0.3 in carbon-wax and ~ -0.9 in the CP and V_2O_4 samples. The value of γ_o in carbon-wax is small and agrees with those obtained in several composites earlier,[23,24] signifying that noise can be ascribed to equilibrium resistance fluctuations. But it is evident from Table I that γ_o tends to be definitely less than 0. Values of γ_o in the other two systems show that equilibrium resistance fluctuation alone cannot account for the noise even in the linear regime. γ_o was earlier found to lie between -0.5 and -0.3 in polypyrrole thin-film resistors.[15] It was suggested that a shot noise component which varies linearly with the current or voltage may be responsible for negative values of γ_o. It may be noted that most of the inhomogeneous systems found in the literatures have had noise measurements performed only in the linear region of the applied bias although not always stated so explicitly. Next we consider the results of the carbon-wax system in detail and a model to explain qualitatively those results. The other two systems will be considered later.

B. Carbon-wax: S_V-V curves

Studies of nonlinearity in gold films[36] and carbon-wax mixtures[16,17,37] in the last several years led to increased understanding of its behavior. Initially, the conduction is Ohmic at small (dc) bias. When the bias is increased beyond a certain value V_o called the onset bias, the conductance starts increasing from its linear value. As the bias is further increased, the differential conductance tends to saturate at a higher level. Thus, the nonlinear behavior of a composite system is governed by two voltage scales: the onset voltage V_o and the saturation voltage V_s, which characterize the approach to saturation. Both V_o and V_s scale with the linear conductance Σ_o as $V_o \sim \Sigma_o^{x_o}$ and $V_s \sim \Sigma_o^{x_s}$ where $x_o \approx 0.4$ and $x_s \approx 0.06$. The saturated state at high voltages ($> V_s$) has the transport exponent t_s different from the corresponding value in the linear regime, t. In fact, $t_s = ct$ where $c = 0.76 \pm 0.07$.[17] Such a state has been called an ''altered percolation system.''

Gefen *et al.*[36] proposed a model that, in spite of some limitations particularly at high bias,[16,17] gives a reasonable description of nonlinear conduction at low bias in a composite and correctly predicts the scaling of the onset bias. This model, called a ''dynamic random resistor network'' (DRRN) model, considers a network consisting of conducting and insulating bonds with the conducting fraction $p > p_c$. An insulating bond has the property that if the voltage across it is larger than a certain critical voltage it becomes conducting. For sufficiently low applied voltage across the network the current flows only through the backbone of the percolating system so that the conduction is linear. As the applied bias is increased, some insulating bonds will have voltages accross them exceeding the critical voltage and, hence, will start conducting, thereby causing macroscopic conduction to be nonlinear. In real samples, there will be conducting clusters branching off the backbone (dangling clusters), separated by insulating regions of very small widths. It is expected that tunneling or hopping conduction will take place through such regions, thus providing extra paths for electrons. It must be clearly understood that the tunneling in the *nonlinear* regime just described is different from the one (i.e., two-component fluctuation) considered by several authors[27,6,39] in the *linear* regime. The latter is of no relevance here.

Upon comparison of Fig. 8 with the data of carbon-wax composites in Fig. 2, one observes considerable similarity between the two in that the noise curves indeed appear to be a power law at small voltages and are generally concave after the onset of nonlinearity. However, the noise curves in real samples have plateaus in the second regimes just after the onset. In view of the behavior of the conductance described above, this is not surprising. There are two characteristic voltages V_o and V_s. The smaller one V_o has been already identified with the onset voltage. The voltage [~ 0.3 V in Fig. 2(a)] at which a noise curve changes its slope has been plotted as a function of the initial conductance ($1/R_0$) of different samples. The slope of the log-log plot is found to be ≈ 0.4 which is in very good agreement with the value of the onset voltage exponent determined earlier from I-V (Ref. 16) and dI/dV (Ref. 37) measurements at the room temperature. The value of V_s is of the order of voltages at which dI/dV were found to approach saturation.[17] This confirms the correlation of the noise power, S_V with the nonlinear conductance. Interestingly, Mantese *et al.*[24] observed in Ni-Al_2O_3

composites that initially the noise amplitude increased quadratically with the applied voltage V. As the latter exceeded a certain value, the noise amplitude increased only linearly. Both these variations were reported to have taken place in the Ohmic regime, somewhat contrary to the observations in the present work. However, no explanation was given. The fact that the variation at higher voltages was less than that at low voltages is consistent with $\gamma_s < \gamma_o$ found in this work (see Fig. 2). Kusy and Kleinpenning[14] measured noise in ZnO varistors mostly in the linear range. But some measurements extended to the nonlinear range as well. It was found that $\mathcal{S}$ in this case also decreased in the nonlinear regime as in composites. Since a varistor sample can be thought of a network of ZnO grains and thin insulating layers separating the grains, a model involving tunneling channels as in composites may explain the data.

S_V-V curves are structurally similar to I-V curves. But the question of an exact relationship still remains open. An empirical answer is provided by Eq. (8) which, as recast below, may be considered a generalized version of Eq. (3):

$$S_V = A V^{2+\gamma_o} R_0^{\omega} (R/R_0)^{\omega_V}, \tag{11}$$

where A is a constant. To emphasize, we restate again that $\omega_V (\sim 3)$ is different from $\omega (\sim 1.7)$. Equation (11) gives the noise amplitude in a closed form as a function of V at a fixed p and, thus, is useful in describing the noise in the whole range of V. An explanation of (11) should provide an additional challenge for modeling the nonlinear state, in general, and the saturated (altered) state, in particular. The result may be compared with the bias dependence of the noise found in CDW systems where the latter was given by IdR/dV.[10] Using the knowledge of conduction in large bias[17] it is possible to derive from Eq. (11) some relations among various quantities. For large $V > V_s$ in the saturated state, the dV/dI approaches the asymptotic value $R_\infty(p)$ so that

$$R \approx R_\infty V/(V-\lambda), \tag{12}$$

where λ is a quantity depending on p only. Thus, using Eq. (12) in Eq. (11), S_V at large V could be rewritten as

$$S_V \sim V^{2+\gamma_o} R_0^{\omega-(1-c)\omega_V} \left(\frac{V}{V-\lambda} \right)^{\omega_V}. \tag{13}$$

In the above we have used the fact that $R_\infty \sim {R_0}^c$, where c is same as defined earlier.[17] Using Eq. (13) in Eq. (7) one obtains an expression for γ_s in composites for $V > V_s$:

$$\gamma_s = \gamma_o - \frac{\omega_V}{(V/\lambda - 1)}. \tag{14}$$

Equation (14) confirms that $\gamma_s < \gamma_o$. The relation also shows that γ_s increases with V and asymptotically approaches γ_o. Consequently, S_V should be a concave function of V for $V \geqslant V_s$ as shown schematically in Fig. 8 and Fig. 2. The solid lines in Fig. 2 are fits of resistance and noise power for $V > V_s$ to Eqs. (12) and (13), respectively. The values of λ used were 1.3 and 1.4 for $R_0 = 92$ kΩ and 2 MΩ respectively. Excellence of the fits indicates how well two independent transport measurements (i.e., dI/dV and noise) corroborate each other.

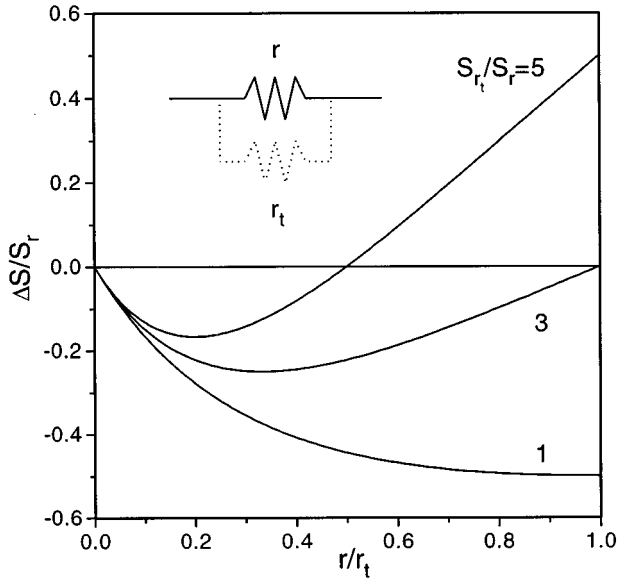

FIG. 9. Plot to indicate the effect of adding a parallel resistor of resistance r_t (inset, dotted line) to a single resistor of resistance r (inset, solid line) on relative noise. $\Delta\mathcal{S}$ is the change in the relative noise. S_r and S_{r_t} are relative noise powers of the resistors r and r_t, respectively.

C. Modeling of noise in nonlinear composites

In conductor-insulator mixtures such as the carbon-wax system the relative noise decreases with V after the onset of nonlinearity (Fig. 5). This is essentially due to the increase in the number of fluctuators, which generally results in the decrease in the noise amplitude. It is recalled that the nonlinearity in the DRRN model arises from the appearance of extra parallel conduction channels with the increase in bias. In the context of noise this means that the nonlinearity is accompanied by an increase in the total number of fluctuators or, equivalently, the system size. We now show that addition of parallel resistors to an existing network, under certain conditions, does lead to a decrease in the total noise of the network. For this purpose, it is sufficient to consider the result of the addition of a single resistor in parallel to a given network. Consider a single (ohmic) resistor of resistance r and relative noise $\mathcal{S}_r$ (inset of Fig. 9). Let us connect an another (ohmic) resistor of resistance r_t and relative noise $\mathcal{S}_{r_t}$ to it in parallel and denote the noise of the parallel combination by $\mathcal{S}_{\rm par}$. Using composition rules based on Cohn's theorem[20] the change in noise, $\Delta\mathcal{S} = \mathcal{S}_{\rm par} - \mathcal{S}_r$, can be written as

$$\Delta\mathcal{S} = -\mathcal{S}_r x \left[\frac{x(1-y)+2}{(1+x)^2} \right], \tag{15}$$

where $x = r/r_t$ and $y = \mathcal{S}_{r_t}/\mathcal{S}_r$. $\Delta\mathcal{S}$ is plotted in Fig. 9 as a function of x for different values of y. It is seen that for small x or large r_t the noise decreases irrespective of the noise level of the parallel resistor. However, if the latter is too noisy (i.e., $y > 1$), the noise may increase ($\Delta\mathcal{S} > 0$) for large x. Figure 10 shows two more examples of a modified star triangle (upper) and a simple fractal (lower) in the form

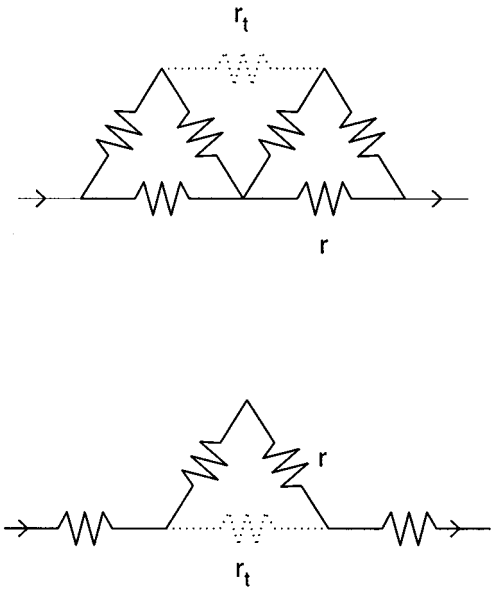

FIG. 10. Two examples of simple networks. The upper one is a star triangle and the lower one is a one-dimensional Koch curve of stage 1. Dotted bonds represent additional parallel bonds.

of a one-dimensional Koch curve of stage 1 where the added resistors are shown with dotted lines. The change in noise for the modified star triangle is

$$\Delta\mathcal{S}=-\frac{\mathcal{S}_r x}{4}\left[\frac{8x^2+13x-2xy+6}{(4x^2+7x+3)^2}\right] \tag{16}$$

and that for the Koch curve of stage 1 is

$$\Delta\mathcal{S}=-\frac{S_r x}{2}\left[\frac{2x^3+2x^2+(4y-1)x-1}{(2x^2+3x+1)^2}\right]. \tag{17}$$

$\Delta\mathcal{S}$ in both cases shows same qualitative behavior as in Fig. 9.

The decrease in the noise of a network due to the addition of resistors in parallel as examplified by the above results appears quite plausible when we consider the general expression of the noise in a network:[22]

$$\mathcal{S}=\mathcal{S}_e\sum_\alpha\left(\frac{g_\alpha}{G}\right)^2\left(\frac{V_\alpha}{V}\right)^4, \tag{18}$$

where $\mathcal{S}_e$ is the relative noise of a conducting bond. g_α is conductance of the bond α and V_α is the voltage accross the bond. The addition of a resistor in parallel is equivalent to changing the conductance of that particular bond from 0 to a small value g_β. This leads to an increase in the network conductance G by $\delta G=g_\beta V_\beta/V^2$ where V_β is the voltage accoss the bond β when the network is held at a fixed voltage V.[22] For a change of conductance of the bond β, the change in the total noise, $\Delta\mathcal{S}$, can be determined using Eq. (18) to first order in g_β as in the following:

$$\Delta\mathcal{S}/\mathcal{S}_e=\sum_\alpha 4\left(\frac{g_\alpha}{G}\right)^2\left(\frac{V_\alpha}{V}\right)^3\left(\frac{\delta V_\alpha}{V}\right)-\sum_\alpha 2\left(\frac{g_\alpha}{G}\right)^2\left(\frac{V_\alpha}{V}\right)^4\frac{\delta G}{G}. \tag{19}$$

The second term on right-hand side is negative since $\delta G>0$ and is of the order of g_β. The first term may be written as

$$\sum_\alpha 4\left(\frac{g_\alpha V_\alpha{}^2}{GV^2}\right)\left(\frac{g_\alpha V_\alpha\delta V_\alpha}{V^2}\right).$$

For bonds far from the bond β, $\delta V_\alpha\approx 0$. Furthermore, $\Sigma_\alpha g_\alpha V_\alpha\delta V_\alpha=0$ as a consequence of Kirchoff's law.[22] Even if the weight factor of fractional power dissipation makes this sum nonzero, the first term is expected to be less than the second term so that noise should decrease. The condition that g_β be small should be easily met in real samples as tunneling bonds are expected to be of much higher resistances than those of regular bonds.

D. Disordered materials

Now we discuss noise in two other disordered systems, namely, conducting polymer and V_2O_4, which are essentially a one-component system and, hence, different from mixtures. Data from another such system (ZnO) were obtained before[14] in both linear and nonlinear regimes but were presented without highlighting S_V-V characteristics as done in Figs. 4 and 5. A comparision of the two systems with Fig. 8 shows that the noise curves change very rapidly from a power-law behavior at $V<V_o$ to another apparent power-law behavior (constant γ_s) for $V>V_o$. There is also no indication of any second voltage scale as in composites. In this case we can derive an empirical expression for the I-V curve at high voltages using Eq. (9). Assuming that $S_V\sim V^{2+\gamma_s}$ for $V>V_o$, we have $V^{2+\gamma_s}\sim V^{2+\gamma_o}\exp[a(R_0/R)^b]$ so that

$$I\sim\frac{V}{R_0}\left(\frac{\gamma_s-\gamma_o}{a}\ln V\right)^{1/b} \tag{20}$$

for $V>V_o$ and $I=V/R_0$ for $V<V_o$.

The most important difference between composites (Fig. 2) and conducting polymer (Fig. 4) is that γ_s is negative in the former but positive in the latter. In other words, the noise increases at a faster rate with V in the nonlinear regime than in the linear regime in the two disordered systems. The analysis of noise data in disordered materials is hindered by a credible theory to explain the experimental data on nonlinearity in such systems.[38] Thus, in the absence of a guiding theory, it may be instructive to consider different situations that will cause the relative noise to increase with the bias. One possibility is the addition of parallel conduction channels as in the DRRN model for composites. It is seen in Fig. 9 that in a certain parameter range (e.g., $r/r_t\sim 1$ and $\mathcal{S}_{r_t}/\mathcal{S}_r\sim 1$), $\Delta\mathcal{S}$ can become positive; i.e., the relative noise *increases* with the addition of a parallel resistor. To see whether such a possibility may exist in the case of systems exhibiting variable-range hopping conduction, we note that the electronic transport in such systems may be mapped into an equivalent random resistor network.[40] Conduction in the hopping mechanism, which is a phonon-assisted quantum-mechanical tunneling of an electron from one localized state to another, is influenced by an external field. The probability of an electron transfer between the sites randomly distributed in space and energy depends exponentially on the external field. With the increase in field, the hopping probability of

the electrons increases and the electrons can hop to more and larger distant sites. This will lead to a rearrangement of the equivalent network and, hence, the equivalent resistance of the network changes. This fact can be pictured as follows: After some critical field extra paths are generated in the system which are similar to the original paths and the resistance of the system decreases as a result of these extra parallel paths.

Another possibility is to consider a network consisting of elements which are assumed to possess the following simple nonlinear I-V characteristics:

$$V = r_1 I + r_2 I^{\alpha}, \tag{21}$$

where r_1, r_2 are two constants and $\alpha > 1$. For $r_2 < 0$ the network resistance will decrease with the applied bias. Such elemental I-V characteristics have been considered earlier[36] for composites but their prediction regarding the onset current exponent x_o was found to be at variance with the experimental observations. In fact, the same is true also in case of the conducting polymer.[38] Nevertheless, the calculation of $\mathcal{S}$ for such I-V as in Eq. (21) serves to bring out several features of noise in nonlinear regimes. Obviously it is not possible to calculate analytically $\mathcal{S}$ for an arbitrary network. Instead we focus on the noise of a single element and attempt to draw conclusions for a general network. Let us define $\mathcal{S}_1 = \langle \delta r_1^2 / r_1^2 \rangle, \mathcal{S}_2 = \langle \delta r_2^2 / r_2^2 \rangle$, and $\mathcal{S}_{12} = \langle \delta r_1 \delta r_2 / r_1 r_2 \rangle$. Here the notation is as usual. In terms of these definitions we have the relative noise given by

$$\mathcal{S} = (\mathcal{S}_1 r_1^2 I^2 + \mathcal{S}_2 r_2^2 I^{2\alpha} + 2\mathcal{S}_{12} r_1 r_2 I^{\alpha+1}) / V^2. \tag{22}$$

This equation illustrates the complexity when the noise is characterized by more than a parameter. From Eq. (22) it follows that if *either* of r_1 and r_2 is zero, the noise is independent of bias. In particular, we obtain an unexpected but interesting result that a network of nonlinear elements described by such a strong nonlinear relation as Eq. (5) fails to exhibit the voltage-dependent noise. It may be recalled that the noise data in ZnO-based varistors which are thought to be described by a relation like Eq. (5) were explained by invoking contribution from tunneling. To examine the variation of $\mathcal{S}$ with bias, we calculate the derivative of Eq. (22) with respect to current:

$$\frac{d\mathcal{S}}{dI} = 2(\alpha - 1) r_1 r_2 I^{\alpha} \left[\frac{r_2(\mathcal{S}_2 - \mathcal{S}_{12}) I^{\alpha-1} - r_1(\mathcal{S}_1 - \mathcal{S}_{12})}{V^3} \right]. \tag{23}$$

Assuming that $\mathcal{S}_1 > \mathcal{S}_{12}$ and $\mathcal{S}_2 > \mathcal{S}_{12}$ it follows from Eq. (23) that $d\mathcal{S}/dI$ is always positive for $r_2 < 0$ and small I. In other words, a random resistor network consisting of nonlinear elements such as given by Eq. (21) allows an increase in the relative noise with the bias as shown in Figs. 4 and 5. However, whether an arbitrary network will exhibit (an apparent) power law for $\mathcal{S}$-V in the nonlinear range remains to be verified.

E. Noise exponents in composites

Let us now discuss variation of the noise amplitude due to microstructural factors in composites. The later are characterized by the conducting fraction p which determines the (linear) resistance. In two other systems, p is not a relevant parameter to change the resistance which is usually changed by varying the temperature.[14] In the linear range (i.e., low V) the value of the exponent ω is 1.7 (Fig. 7) which is same as that found by Chen and Chou[25] also in carbon wax. However, it has been already noted[2] that the explanation in terms of number fluctuation of the charge carriers in tunneling put forward by the authors is untenable. As discussed in the Introduction the value of the exponent cannot be explained at present satisfactorily by any existing theory.

At high voltage, this system enters into a saturated state characterized by a different conductivity exponent t_s which is less than the usual conductivity exponent t. In this state at $V = 5$ V, the noise exponent ω_s is found to be 0.5 (Fig. 7) which is less than ω in the linear range, i.e., 1.7, and even less than unity [Eq. (2)] in a homogeneous sample. That ω_s is less than ω may be rationalized by the fact that a sample tends to be more homogeneous in terms of the conducting bond distribution as the bias is increased. Following this logic, one would then expect that as a sample approaches the saturated state the noise exponent would monotonically decrease (from 1.7) towards unity. Thus, the fact that the value of ω_s is less than 1 cannot be reconciled by this argument. Almost same results are obtained at $V = 10$ and 20 V. The two exponents ω_s and ω are, however, related. From Eq. (13) it follows easily that

$$\omega_s = \omega - (1 - c)\omega_V. \tag{24}$$

The above relation is interesting in the sense that two quantities ω and ω_s by definitions must involve different samples with different p's whereas ω_V is obtained basically with only one sample at a time. Large uncertainities in the quantities in Eq. (24), however, make the relation rather less useful. Putting $\omega = 1.7$ and $\omega_V \sim 3$ in Eq. (24) gives $\omega_s \sim (1 \pm 0.6)$ compared to 0.5 ± 0.1 determined directly.

V. CONCLUSIONS

It is quite evident from the previous discussion that S_V-V characteristics of inhomogeneous or disordered systems can be quite structured and are somewhat analogous to I-V characteristics in carrying information on transport properties of these systems. Not only should measurements of these two characteristics be useful in understanding especially the nonlinear conduction, but it could be also used for verifying the consistency of a transport model as has been done in the case of composites. We described how results obtained from independent measurements of dI/dV in composites were useful in interpreting the noise data in a consistent manner. In the same way, it is hoped that the noise data would prove useful in building a suitable model of nonlinearity in the disordered systems. It is well known that the noise involves the fourth moment of the current distribution whereas the resistance involves the second moment. Hence the noise is expected to be more sensitive to any change in the current distribution as a result of the onset of nonlinearity than the resistance. For example, not only is the sign of change in the relative noise power after the onset of nonlinearity different from that of the resistance in Mott systems, but also the noise increased by more than four orders of magnitude while the resistance decreased by a factor of 5 only.

It is significant that the functional dependence of $\mathcal{S}$ on the resistance varied by either p at a $fixed\ V$ or V at a $fixed\ p$ is given by the same power law although the exponent ω_V at a $fixed\ V$ is different and larger than the exponent ω at a $fixed\ p$. This once again highlights the difference between the usual random and the correlated percolation problems. The nonlinear states of the composites are examples of a correlated dynamical percolation system.[17] The linear states correspond to the usual random percolation problem. The saturated states at high voltage are where dI/dV becomes constant and $\mathcal{S}'$ also tends to a constant value [Eq. (13)]. These states have both transport exponents t_s and ω_s different from their values in the linear regime. A proper model of this state is still lacking. This is also emphasized by the fact that ω_s is less than unity. The reasons for the relatively large sample to sample scatter in the values of ω_V are also not clear.

Through the introduction of the function $\mathcal{R}$ we have tried to suggest a framework to analyze the noise data in the entire range of applied bias. Generally speaking, the form of the function $\mathcal{R}$ should be a characteristic of the particular nature of inhomogeneity or disorder in a given physical system. We see that whether R of a composite system is varied by either p or V, $\mathcal{R}$ remained a power law. In a conducting polymer or V_2O_4, $\mathcal{R}$ is, tentatively, of the form $\exp[a(R_0/R)^b]$ when R is varied by changing the bias. It will be interesting to find out what happens to these functional forms as R_0 is varied by changing the temperature which is expected to alter bond resistances in a system differently than the bias. Temperature, unlike in homogeneous systems, plays a strong role in the systems under investigation. In ZnO-based varistors, R_0 was varied using temperature.[14] It was observed that $\mathcal{R}\sim R_0$ but has a functional dependence on V qualititatively similar to that of a composite.

An unresolved issue concerns the value of γ_0. It has a small negative value in composites but is significantly different from zero in disordered materials.

Note added in proof. We came to know of the following references which are relevant to the present work. A. A. Snarskii, A. E. Morozovsky, A. Kolek, and A. Kusy, Phys. Rev. B **53**, 5596 (1996); A. E. Morozovsky and A. A. Snarskii, Int. J. Electron. **73**, 925 (1992); Sov. Phys. JETP **68**(5), 1066 (1989).

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