Flicker noise in thin lipid films

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Nonlinear charge conduction in an organized assembly of lipid molecules has long been established. However, the exact mechanism of charge transport across them is still not very clear. Since resistance involves the second moment of the current distribution, while noise involves the fourth moment, the noise measurement should be more informative about the underlying mechanism of charge transport. Using this perspective, we have measured noise in thin solid films of lipid molecules over a wide range of voltages that covers linear as well as nonlinear regimes. The power spectra obtained in this system show an increase in the relative noise with bias. This can be explained by considering the system to be a random resistor network consisting of nonlinear current elements. [S0163-1829(97)05846-3]

INTRODUCTION

Power law correlations in space (fractal) or time (flicker noise) are manifest in a large-number of physical systems.[1] Noise amplitude in an inhomogeneous system is greater than that in a homogeneous system of comparable resistance, indicating that the microstructure determines the noise spectrum of the system. Thus the noise spectrum in inhomogeneous systems has been used as a probe to understand their microstructure and their complex conduction mechanism which is characterized by a nonlinearity at high field and distinct chordal resistance (V/I) and differential resistance (dV/dI) readings.[1–4]

Nonlinear conduction at high fields may arise either from a change in the existing charge conduction mechanism or due to some new charge conduction channels being created at high fields.[5] When the noise power is measured against applied voltage, an onset of nonlinearity occurs at a bias which is identical to the voltage responsible for nonlinearity in conduction, i.e., where the resistance deviates from its Ohmic value. This suggests a strong correlation between noise measurements and the conduction mechanism, and it is expected that the behavior of $1/f$ noise in the nonlinear range will be strongly motivated by the particular mode of conduction coming into play.[2]

A good example of nonlinear conduction at high fields is the charge transport across an organized lipid assembly.[6–8] Using a lipid planar membrane as a model for the more complex biological membrane, nonlinear charge conduction at high fields has been reported by several authors.[6,7] The mechanism of such nonlinearity has been attributed to the opening up of more water-filled pores in the otherwise poorly conducting lipid matrix through which charges can move.[6] However, as similar nonlinearity has also been reported in thin solid films of lipids, the creation of more of the water-filled pores at higher fields was the only factor to account for the observed nonlinearity may be ruled out.[8] One may expect in this particular case, that the existing charge conduction mechanism is affected by the high value of the applied bias.

Here we have reported our preliminary findings of the $1/f$ noise measurement in thin solid films of the lipid dipalmitoyl phosphatidyl choline (DPPC). This system shows nonlinear charge conduction at high applied bias.[8] The $1/f$ noise spectra have been analyzed and compared with those obtained with other systems, where the mechanism of charge conduction is well established. Noise measurement has also been carried on thin film of oxidized cholesterol (OC) and similar results are obtained.

EXPERIMENT

Cholesterol and DPPC were purchased from Sigma Chemical Co (St. Louis, MO). Cholesterol was oxidized and then recrystallized from *n*-octane.[9] AR-grade chemicals supplied by E. Merck Ltd. (Worli, Bombay 400 018) were used without further purification.

A thin film of lipid (OC or DPPC) was prepared by repeatedly dipping a tin-oxide-coated glass plate (courtesy of Professon E. Sackmann of Technische Universität Munchen, Germany) in chloroformic solution of the lipid (0.1 mol concentration). The film was kept in vacuum for 10–12 h to remove traces of solvent. The experimental sandwich cell consisted of this glass plate containing the thin film (2×2 cm) on which another tin-oxide-coated glass plate was placed with the lipid film lying between the two conducting faces of the glass plates. The cell was connected with the external circuit by using a pair of spring clips (please see Ref. 8 for the detailed experimental arrangement).

A constant current from a programmable constant current source (Keithley model 224) was passed through the sample. The corresponding voltage fluctuations measured using a

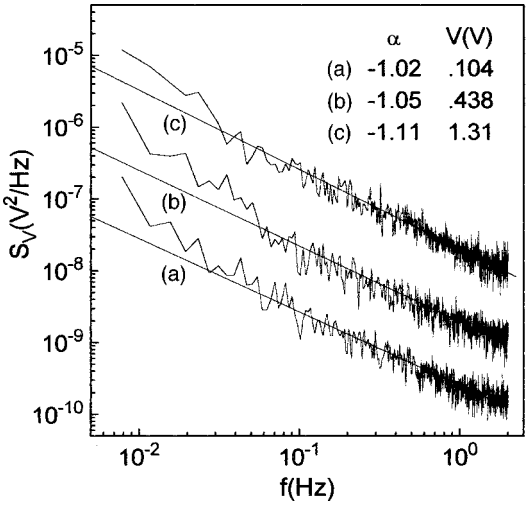

FIG. 1. Log-log plot of three noise spectral power $S_V(f)$ vs f for a thin solid film of lipid of resistance 5.16 kΩ. The solid lines denote a fit to a power law. The applied voltage and the corresponding value of the power law exponent are indicated in the figure.

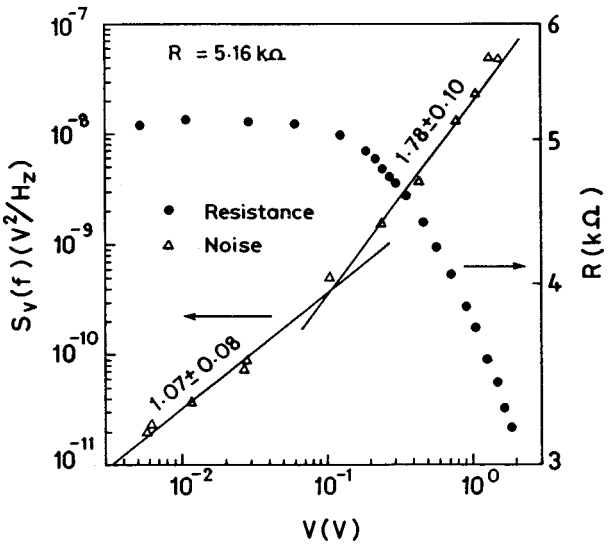

FIG. 2. Plot of the variation of resistance of a thin solid film of lipid ($R=5.16$ kΩ) with applied bias (solid circles) and the corresponding variation of S_V (open triangles).

digital multimeter were stored in a computer at an interval of 250 ms. Data were taken in sets of 1024 data points, which facilitated performing 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. To examine it as a function of applied bias, an average noise at 0.5 Hz was used throughout this work. The noise level of the electronic devices used in these experiments was found to be lower compared to the experimental noise spectrum by at least five orders of magnitude.

RESULTS AND DISCUSSION

Figure 1 shows the noise power spectrum $S_v(f)$ in a thin film of DPPC ($R_0=5.16$ kΩ) as a function of frequency at three different voltages. It is evident from the plot that each curve has $1/f$ character.

In Fig. 2 the voltage dependence of the noise power at 0.5 Hz (open symbols) and the chordal resistance ($R=V/I$, solid symbols) of the thin solid film of DPPC ($R=5.16$ kΩ) have been plotted. (The magnitude of the statistical error in both Figs. 1 and 2 is about the size of the symbol.) The structured noise curve of Fig. 2 has two distinct regimes, characterized by two slopes (1.07+0.08 and 1.78+0.10). The voltage V, where the variation of the slope starts, coincides with the onset of nonlinear conduction, i.e., where R deviates from its linear value. This indicates a close relation between the noise power and the conduction mechanism. Also, from the figure it is evident that while the resistance decreases in the nonlinear regime by a factor of 2 only, the noise amplitude increases by four orders of magnitude. Since noise involves the fourth moment of current distribution, while resistance involves the second moment, this result probably indicates that noise measurement will be more sensitive in detecting any change in the current distribution after the onset of nonlinearity.

A systematic study of the flicker noise in three different systems—namely, (A) carbon-wax composite, (B) conducting polymer (a powdered mixture of ferric chloride doped polypyrrole), and (C) vanadium dioxide—has been carried out by Nandi *et al.*[2] All three systems are disordered although (A) is a composite system, while the other two are one-component systems. The voltage dependence of the noise power is different in the linear and nonlinear regimes in each system and also different among various systems. This is due to the fact that the conduction mechanism is different in the three systems. Compared to the linear (i.e., low-bias) region, the noise increases at a faster rate in the nonlinear (i.e., high-bias) region for systems (B) and (C), whereas it increases at a slower rate for system (A). That the noise characteristics have a strong correlation with the resistance variation is apparent from the following observation. The resistance and slope of the relative noise power are con-

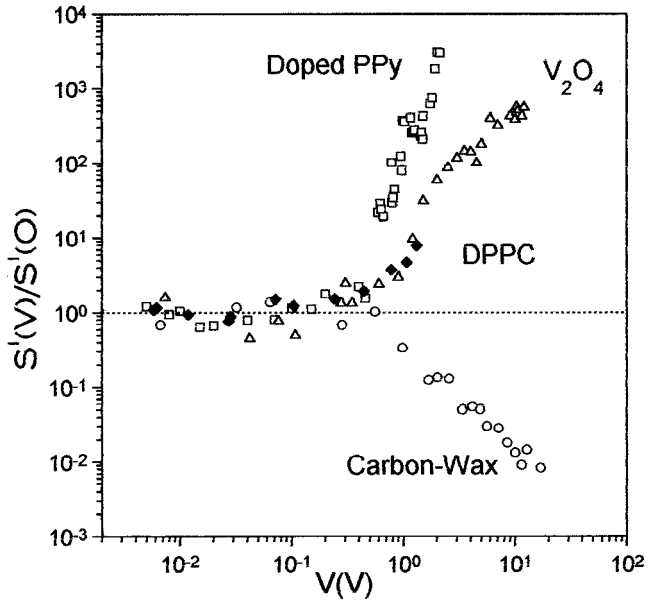

FIG. 3. Plots of normalized S' vs V of four types of disordered systems as indicated in the figure.

stant in the low-bias region, while the resistance changes with voltage in the high-bias region and the slope attains another constant value, this implies that the change in the conduction mechanism in the high-bias region influences the noise spectrum.

Comparison of the behavior of the noise data in thin lipid films with the other three systems shows that the former is similar to that of the conducting polymer and V_2O_4, where the noise power increases with the bias. A generalized relative noise power (S') at 0.5 Hz, normalized by its value in the linear range, has been plotted as a function of V in Fig. 3. While S' increases with bias for (B), (C), and thin lipid films by several orders of magnitude, it decreases in a similar way for (A). Had there been any field-induced increase in the number of parallel pathways in the thin lipid film, the amplitude of noise would have decreased with the bias as in case of (A). So the possibility of opening up of new parallel pathways can probably be ruled out here. But then how can one explain the enhancement in current with the increase in field, as evident from the resistance vs voltage curve (Fig. 2)? The only answer to this question probably lies in considering the possibility of a field-induced change in the conduction mechanism itself.

Hooge's empirical formula for the relative noise power is

$$S' = S_R/R^2 = S_V/V^2 = S_I/I^2 = [V^{t_0}/f^{\lambda}\mathcal{R}(R)], \tag{1}$$

where $S_x = \langle \delta x^2 \rangle$ is the spectral density and λ is of the order of unity. $x = R$, V, and I, and the value of t_0 determines whether noise is a driven phenomenon ($t_0 \neq 0$) or a result of equilibrium resistance fluctuations ($t_0 = 0$). The form of the function depends on the particular system under consideration.

Keeping in mind the S_V plot for the lipid film, we define a quantity t (Ref. 2) given by

$$2 + t = d(\ln S_V)/d(\ln V). \tag{2}$$

From Eq. (1) we have

$$d(\ln S_V)/d(\ln V) = 2 + t_0 + (V/\mathcal{R})(d\mathcal{R}/dR)(dR/dV). \tag{3}$$

From Eqs. (2) and (3), we get

$$t = t_0 + (V/\mathcal{R})(d\mathcal{R}/dR)(dR/dV). \tag{4}$$

For thin lipid films, $dR/dV < 0$ for $V > V_0$, where V_0 is the voltage at which nonlinearity sets in.

Thus, for $V > V_0$, the sign of $d\mathcal{R}/dR$ determines whether t will be less than or greater than t_0, but the functional form of $\mathcal{R}$ is determined by the microstructure of the system. Therefore, it can be inferred that the nature of the conduction mechanism will determine the behavior of the S_V-V curve after the onset of nonlinearity.

In order to understand the exact conduction mechanism involved in thin lipid films, let us analyze the different situations that will cause the noise to increase with bias. The addition of parallel conduction channels (as in Gefen's model for composites[5]) whose resistance is of the order of that of the regular bonds may cause the noise to increase, but as the field-induced tunneling bonds offer much higher resistance than regular bonds, this explanation for the observed nonlinearity in our system can be ruled out.

However, it is also possible to treat the system as being composed of nonlinear elements. It has been shown[2] that a random resistor network composed of such elements causes the noise to increase with bias. Thus, from the experimental results, we might explain the charge conduction in thin films of lipids at high bias are due to such constituent nonlinear elements.

The lipid molecules are an integral part of biological membranes, charge conduction through which is the key to all the major life-sustaining bioprocesses. While the organized lipid assemblies are extensively used to model the more complex biomembranes, the exact mechanism of charge transport through the biological membranes as well as through the model lipid membranes is still not very well understood. In this paper we have attempted to explain this charge conduction by a mechanism that treats the model membrane system of organized lipid assembly to be composed of nonlinear current elements. The nonlinear I-V characteristics and study of the noise spectrum on this system confirm the plausibility of our explanation.

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