Conductance-Noise Power Fluctuations in Hydrogenated Amorphous Silicon

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The power spectra of the $1/f$ noise in n-type-doped hydrogenated amorphous silicon (a-Si:H) are themselves time dependent. The noise power changes both in magnitude and as a function of frequency, reflecting a modulation of the properties of the fluctuators responsible for the current noise. Slow variations of the noise are strongly correlated over a broad range of frequencies. Spectral analysis of these noise power fluctuations, termed second spectra, also shows an approximate $1/f$ spectral slope, that is, the $1/f$ noise has $1/f$ noise. These results indicate that highly cooperative interactions exist between the fluctuators.

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The spectral density of resistance fluctuations in many semiconducting and metallic solids has an approximately $1/f$ dependence on frequency f which can be described as arising from a superposition of statistically independent Lorentzian power spectra with a broad distribution of relaxation times [1,2]. The individual Lorentzian spectra that comprise the $1/f$ noise can be resolved in sufficiently small-length-scale devices wherein the current is constrained to narrow regions leading to a sensitivity to the presence of a small number of defects (of order 1); one then observes switching noise in the resistance time traces [3–5]. The recent observation of random telegraph switching noise in coplanar measurements of n-type-doped hydrogenated amorphous silicon (a-Si:H) samples, with electrode separations as large as 1 mm, indicates that a few microscopic processes are dominating the electronic conduction on a macroscopic scale [6]. Possible explanations for these results are that the current paths are highly filamentary with weak links, or that fluctuators responsible for the switching and $1/f$ noise in a-Si:H interact through serial rather than parallel kinetics, or possibly both.

In this paper we report the first observation of higher-order correlations in the noise power of a-Si:H. The noise power of a-Si:H displays a complicated time dependence, with strong correlations in the variation of the noise between differing frequency octaves. Fourier transforms of the time-dependent variations in the noise power yield "second spectra" which themselves have a $1/f$ frequency dependence. These results are surprising given the large effective volumes ($\sim 10^{-7}$ cm^3) of the samples studied here if the individual fluctuators which contribute to the noise power are statistically independent of each other.

We have previously reported the average properties of the noise power of a-Si:H [7,8]. The spectral density of the coplanar current fluctuations has an $f^{-\gamma_1}$ frequency dependence from 1 Hz to 10 kHz with $0.9 < \gamma_1 < 1.1$ over the temperature range $190 < T < 450$ K [7]. The observation of random telegraph switching noise [6] is consistent with the current in a-Si:H being carried, at least in part, by inhomogeneous current paths, the conductance of which is time and temperature dependent [7,8]. The results described in this Letter clearly demonstrate that the noise power in a-Si:H is sensitive to a small number of effective fluctuators which exhibit cooperative dynamics.

The amorphous silicon films studied were synthesized in a rf glow-discharge deposition system. n-type doping is achieved by adding phosphine [10^3 ppm(volume)] to the silane; the substrate temperature was 520 K and the incident rf power was 2 W. The films are 1 μm thick, and were deposited onto Corning 7059 glass substrates. The a-Si:H film was patterned using photolithography and ion milling, with the area between the coplanar electrodes being 800 μm long by 200 μm wide, with an effective sample volume of 1.6×10^{-7} cm^3. All of the data reported here were obtained using voltages within the Ohmic regime. The sample is annealed at 450 K in the dark under a turbo-pumped vacuum to remove any surface adsorbates and effects of prior light exposure before the noise data are taken [9]. Details of the deposition technique and growth conditions [10], electronic [11], and noise properties [8] of these samples have been published previously. While this Letter describes noise power fluctuations for a single n-type a-Si:H film, similar results are observed for other samples deposited under comparable conditions [12].

A time record of the noise power of the resistance fluctuations of n-type a-Si:H at 300 K is shown in Fig. 1. A single power spectrum is obtained from a 1024-point time series using a HP 3561A spectrum analyzer using a fast Fourier transform (FFT) program. A large number of these power spectra (typically 10000) are calculated. To reduce the amount of data to be stored the power spectra are summed into seven frequency octaves. The lowest octave is 5–10 Hz, consisting of two power spectra points, and the highest octave, containing 128 power spectra points, ranges from 320 to 640 Hz. For $1/f$ noise each octave contains the same total noise power; consequently the curves in Fig. 1 are offset for clarity. In order to reduce the variance of each octave for ease of plotting, each data point in Fig. 1 is the result of averaging four FFT's. The fluctuations in the noise power in each octave are much larger than variations expected due to sampling of a random signal. There are large variations in the noise power magnitude as a function of time, with strong

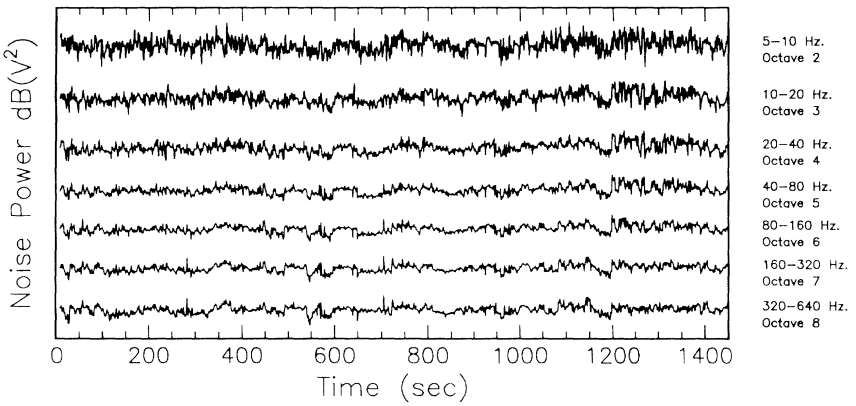

FIG. 1. Noise power as a function of time for an n-type-doped a-Si:H film (10^{-3} PH_3/SiH_4 gas-phase doping level) at 300 K for seven octaves. Each point is the result of averaging four FFT's. The noise is measured with Ohmic electrodes in a two-probe coplanar configuration with electrode width of 0.2 mm and separation of 0.8 mm. Identical results are obtained from four-probe measurements.

correlations of the noise power between differing octaves. Comparison of two-probe and four-probe measurements at several temperatures yields results identical to Fig. 1, confirming that the noise power fluctuations are due to the bulk a-Si:H film and are not contact effects.

The correlations between the differing octaves can be quantified by calculating the correlation coefficients, following the analysis of Restle, Weissman, and Black [13]. The correlation coefficient ρ_{ij} between octaves i and j is given by

$$\rho_{ij}=\sum_n (Q_i-\bar{Q}_i)(Q_j-\bar{Q}_j)/\sigma_i\sigma_j, \quad i\neq j, \tag{1}$$

where Q_j is the noise power for octave j, $\bar{Q}_j$ is the average noise power in that octave after averaging 10000 spectra, σ_j is the jth octave's standard deviation, and the sum is over the total number of noise power spectra measured. The ρ_{ij} values range from $+1$ to -1, where the former value corresponds to exactly positively correlated noise power and the latter value represents exactly inverse correlations between octaves. The ρ_{ij} between all pairs of octaves are calculated and all coefficients for an octave separation of 1 are averaged together. The ρ_{ij} for octave separations of 2 are similarly averaged, and so on.

The correlation coefficients as a function of octave separation for a-Si:H at 300 K are compared to the anticipated ρ_{ij}'s for a sum of statistically independent Lorentzian power spectra in Fig. 2. The continuous distribution of Lorentzian functions used in the calculation yields a $1/f$ frequency dependence for the total power spectrum, and the amplitude of each Lorentzian that contributes to the noise is independently randomly modulated [14]. The ρ_{ij} for the Lorentzian spectra are due to the overlap across octaves of the individual power spectra. As shown in Fig. 2, the ρ_{ij} for a-Si:H are much larger than for an ensemble of statistically independent fluctuators. The correlation coefficients decrease with increasing octave separation but even octave 1 (5–10 Hz) and octave 7 (320–640 Hz) have a correlation coefficient of 0.33 which is nearly a factor of 2 larger than for non-Gaussian noise from statistically independent Lorentzian spectra. Also shown in Fig. 2 are the values of the correlation coefficients calculated after ten FFT spectra are averaged together. Since each single FFT takes $\sim$1.5 sec to calculate, the net result of averaging ten spectra is to eliminate fast fluctuations which occur on time scales less than 15 sec. These averaged ρ_{ij} (open triangles in Fig. 2) are markedly larger for all octave separations, indicating that the correlations between the fluctuators become stronger on longer time scales. The ρ_{ij}'s growth with a quasilogarithmic dependence on the averaging time, as described in detail elsewhere [12]. The observation of switching noise in the resistance time traces indicates that for short times the noise power can be described by a Lorentzian; howev-

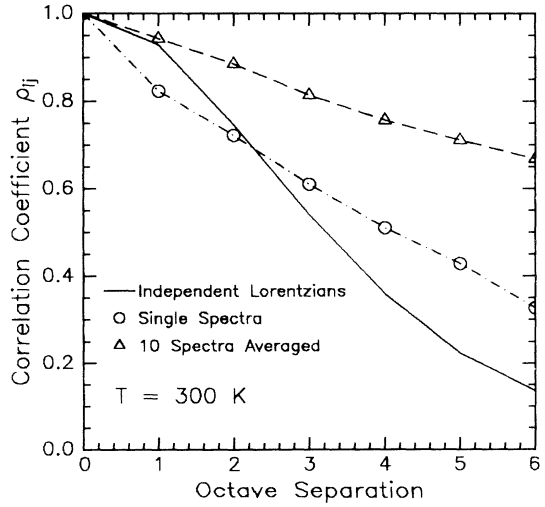

FIG. 2. Plot of the average correlation coefficient against octave separation for the sample in Fig. 1 at 300 K. △'s represent the correlation coefficients calculated after averaging ten FFT spectra and the solid line indicates the expected correlation coefficient for a continuous distribution of Lorentzian spectra whose amplitudes are independently modulated.

er, the large ρ_{ij}'s and their growth with averaging time indicate that the properties of these Lorentzians are not independently modulated but rather interact on long time scales through some form of cooperative dynamics [15].

The low-frequency components to the noise power fluctuations shown in Fig. 1 are clearly seen in the second spectra [14], as shown in Fig. 3. The second spectra are calculated from a time record of the noise power for a given octave, specified by f_1^k (the logarithmic average frequency for octave k). For each octave a power spectrum $S_2(f_2,f_1^k)$ is calculated at the frequency f_2, determined by the time scale of the fluctuations in Fig. 1. The second spectra are plotted by dividing the frequency f_2 by the center frequency of each octave f_1^k [16]. The second spectra in Fig. 3 are normalized so that if the noise is due to a Gaussian process (such as charge carrier trapping due to a large number of statistically independent defects) then S_2 is unity for all frequencies f_2. That is, higher-order correlations do not contain any additional information for Gaussian processes. Separate measurements of Johnson noise in a metal film resistor do indeed find "white" second spectra with a magnitude $S_2 \sim 1$. In contrast the second spectra for a-Si:H show a clear power-law frequency dependence with magnitudes well above unity.

The solid line in Fig. 3 represents a fit to the second-spectra data by the expression

$$S_2 = S_2(0) + \alpha_2 (f_2/f_1)^{-\gamma_2}, \quad (2)$$

where $S_2(0)$ and α_2 are constants and γ_2 is the S_2 spectral slope. The power-law exponent γ_2 is 1.13 at 300 K,

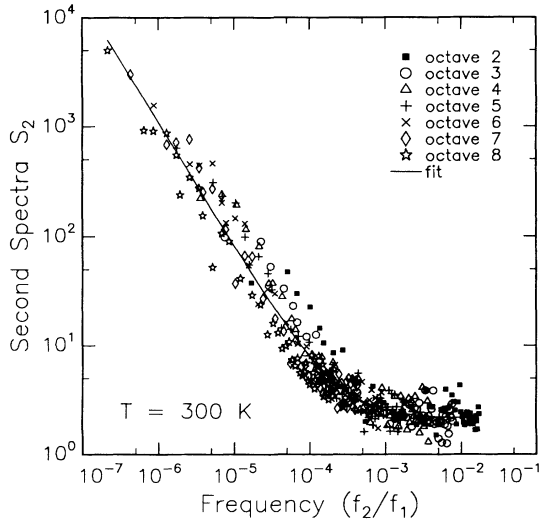

FIG. 3. Log-log plot of the second spectra, representing the Fourier transform of the data in Fig. 1, against f_2/f_1 at 300 K, where f_2 is the frequency of the noise power fluctuations, determined by the time scale of Fig. 1 and f_1 is the center frequency of each octave. The solid line represents a fit to the data of a power-law frequency dependence plus a frequency-independent constant.

with a frequency-independent component of $S_2(0) \sim 2$ at scaled frequencies above 10^{-3} Hz. The white component of the second spectra at higher frequencies is similar to that expected for stationary independent Lorentzian functions [17], which is consistent with the smaller correlation coefficients observed at shorter time scales (Fig. 2). The spectral slope is in the range $0.65 < \gamma_2 < 1.1$ over the temperature range $190 < T < 450$ K with no apparent systematic trend. At a given temperature the second spectral slope γ_2 is always less than the spectral slope γ_1 of the first spectra (i.e., the conventional current noise power spectra). The frequency-independent term in Eq. (2) varied from 2 to 10, while α_2 ranged from 10^{-1} to 10^{-4} over this temperature regime, again with no systematic variation with temperature. Details of the temperature dependence of S_2 will be published separately [12].

The second spectra shown in Fig. 3 represent the absolute square of the Fourier transform of the noise power in a given octave k. We have also calculated the second-order cross sepctra [13] which are the product of the FFT of the noise power in octave i and the complex conjugate of the FFT of the noise power in octave j. Unlike the second spectra in Fig. 3, the cross spectra can consist of both real and imaginary terms. The imaginary component of the cross spectra can be considered as representing a phase difference between the changes in the noise power at one octave and the subsequent variations in the noise at another octave. Such a phase difference could arise if the experimental probe used to measure the noise spectral density, such as the dc current, actually induced the current fluctuations instead of simply passively monitoring variations in the bulk resistance of the sample [18]. The magnitude of the imaginary term of the cross spectra for a-Si:H is consistent with zero, while the real component has a power-law frequency dependence, $\propto 1/f_2$, over the entire temperature range investigated, indicating that the results described here reflect variations in the intrinsic properties of the a-Si:H film and are not an artifact of the measurement process.

The random telegraph switching noise [6] has been ascribed to the influence of inhomogeneous current paths in the a-Si:H film. Such current filaments could arise due to potential fluctuations from charged defects [19], the intrinsic disorder of the silicon network [20], or from the hydrogen microstructure [21]. The switching noise and the time dependence of the noise power described here indicate that the resistance of these postulated microchannels must change with time. We have previously suggested that the motion of bonded hydrogen, by changing defect bonding coordinations, could account for the variations of these filaments' resistance [8]. Recent nuclear magnetic resonance measurements [22] and supercell calculations [23] have found that metastable defect formation in a-Si:H, induced by the local change in a single defect's charge state, can involve the collective motion of hundreds of atoms. If hydrogen motion is responsible for

the variations in the properties of the fluctuators that give rise to the $1/f$ noise in a-Si:H, then the results reported here suggest that rearrangements of the hydrogen bonding configurations, which lead to changes in the electronic properties of a-Si:H, involve many hydrogen atoms moving collectively.

Similar indications of cooperative dynamics, reflected in the second spectra such as reported here have been previously observed in mesoscopically small CuMn spin glasses [16] and in simulations of kinetic Ising models [24]. In contrast the a-Si:H film studied here has an effective sample volume of $\sim 10^{-7}$ cm^3 with a coplanar electrode separation of 0.8 mm. We speculate that in highly disordered solids, such as heavily doped a-Si:H, the large-scale structural and electronic heterogeneities lead to the creation of a few inhomogeneous current paths. When studied with the appropriate experimental probe, such as noise measurements, these filaments reveal phenomena similar to that seen in mesoscopically small spin glasses [16].

In conclusion, the statistical properties of resistance fluctuations in doped a-Si:H reveal a sensitivity to a small number of fluctuation processes. There are strong correlations of the noise power over several frequency octaves, indicating the presence of strong interactions over differing time scales. The second spectra of the time-dependent noise power also have an approximate $1/f$ frequency dependence, reflecting the interactions between the fluctuators responsible for the current noise. Further studies of the second spectra as the materials properties of the a-Si:H are systematically varied will help elucidate the microscopic mechanism responsible for the noise power fluctuations, determine whether the cooperative dynamics are hierarchically constrained [16], and also improve our understanding of transport in amorphous semiconductors.

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[1] P. Dutta and P. M. Horn, Rev. Mod. Phys. **53**, 497 (1981).
[2] M. B. Weissman, Rev. Mod. Phys. **60**, 537 (1988).
[3] C. T. Rogers and R. A. Buhrman, Phys. Rev. Lett. **53**, 1272 (1984).
[4] K. S. Ralls, W. J. Skocpol, L. D. Jackel, R. E. Howard, L. A. Fetter, R. W. Epworth, and D. M. Tennent, Phys. Rev. Lett. **52**, 228 (1984).
[5] C. T. Rogers, R. A. Buhrman, H. Kroger, and L. N. Smith, Appl. Phys. Lett. **49**, 1107 (1986).
[6] C. E. Parman, N. E. Israeloff, and J. Kakalios, Phys. Rev. B **44**, 8389 (1991).
[7] C. Parman and K. Kakalios, Phys. Rev. Lett. **67**, 2529 (1991).
[8] C. E. Parman, N. E. Israeloff, and J. Kakalios (to be published).
[9] M. Tanielian, Philos. Mag. B **45**, 435 (1982); D. L. Staebler and C. R. Wronski, Appl. Phys. Lett. **31**, 292 (1976).
[10] R. A. Street, J. C. Knights, and D. K. Biegelsen, Phys. Rev. B **18**, 1880 (1978).
[11] J. Kakalios and R. A. Street, Phys. Rev. B **34**, 6014 (1986).
[12] C. E. Parman, N. E. Israeloff, and J. Kakalios (to be published).
[13] P. J. Restle, M. B. Weissman, and R. D. Black, J. Appl. Phys. **54**, 5844 (1983).
[14] P. J. Restle, M. B. Weissman, G. A. Garfunkle, P. Pearah, and H. Morkoc, Phys. Rev. B **34**, 4419 (1986).
[15] While a small number of independent Lorentzians could account for the large ρ_{ij}'s, they cannot explain the growth of the correlation coefficients with averaging time. See also Ref. [12].
[16] N. E. Israeloff, G. B. Alers, and M. B. Weissman, Phys. Rev. B **44**, 12613 (1991).
[17] G. A. Garfunkle, G. B. Alers, M. B. Weissman, and N. E. Israeloff, Phys. Rev. B **40**, 8049 (1989); G. A. Garfunkle, G. B. Alers, and M. B. Weissman, Phys. Rev. B **41**, 4901 (1990).
[18] K. P. O'Brien and M. B. Weissman, Bull. Am. Phys. Soc. **37**, 321 (1992).
[19] R. A. Street, J. Kakalios, and M. Hack, Phys. Rev. B **38**, 5603 (1988); H. Overhof and W. Beyer, Philos. Mag. B **43**, 433 (1981).
[20] C. Cloude, W. E. Spear, P. G. LeComber, A. C. Hourd, Philos. Mag. B **54**, L113 (1986).
[21] J. A. Reimer, R. W. Vaughan, and J. C. Knights, Phys. Rev. Lett. **44**, 193 (1980).
[22] R. E. Norberg, J. Bodart, R. Corey, P. A. Fedders, W. Paul, W. Turner, and S. Jones, MRS Symposia Proceedings No. 258 (Materials Research Society, Pittburgh, to be published).
[23] P. A. Fedders, Y. Fu, and D. A. Drabold, Phys. Rev. Lett. **68**, 1888 (1992).
[24] G. B. Alers, M. B. Weissman, A. B. Kinzig, and N. E. Israeloff, Phys. Rev. B **36**, 8429 (1987).