June 2004 • NREL/SR-520-36095

Subcontractor Report

Experimental Studies of Light-Induced Changes in Long-Ranged Disorder in Amorphous Silicon

Final Subcontract Report
14 May 1999–31 August 2003

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University of Minnesota
Minneapolis, Minnesota

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National Renewable Energy Laboratory
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Office of Energy Efficiency and Renewable Energy
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Executive Summary

Project Objective: The principal objective of the proposed research is the experimental study and elucidation of the role that long ranged disorder plays in the light-induced metastable conductance changes in hydrogenated amorphous silicon (a-Si:H).

Motivation: The development of sources of low-cost renewable electrical energy is a high National priority. A consensus has developed that hydrogenated amorphous silicon (a-Si:H) is the material of choice for large-area photovoltaic devices. However, the efficiency of a-Si:H based solar cells decreases by nearly a factor of two following extended illumination, owing to the creation of light-induced defects resulting from the recombination of photo-excited charge carriers. If this light-induced defect formation could be reduced or eliminated, the practical conversion-efficiency of a-Si:H based solar cells would effectively double, yielding cost per kilowatt-hour values comparable to that of conventional fossil fuel generation of electricity. Recent experimental results and theoretical calculations indicate that associated with light-induced dangling bond creation there are alterations in the medium- and long-ranged disorder in the a-Si:H films. A determination of whether or not the long-ranged disorder is affected in the Staebler-Wronski effect is important, for if so, then even when local defect generation is prevented there could still remain metastable conductance changes that would affect photovoltaic device performance. Alternatively, if experimental studies clearly rule out light-induced changes in the long-range order in the a-Si:H film, then entire classes of models for the Staebler-Wronski effect can be eliminated from consideration.

Approach: Long range disorder (LRD), due to compositional modulations or potential fluctuations on length scales of 100 ~ 1000 Ångstroms, is believed to influence the current carrying states at and near the mobility edge in a-Si:H. Amorphous silicon films grown under varying deposition conditions, with altered microstructure, will be synthesized. Experimental studies of the thermopower/conductivity activation energy difference, along with the non-Gaussian statistical properties of the conductance fluctuations, will be employed to quantify the long range disorder in these films, providing a connection between the structural and electronic disorder. Changes in the LRD with light soaking and annealing treatments, if any, will be investigated.

Results: The main conclusion of this project is that there is no significant change in the electronic long-range order with light soaking. The results from this project indicate that the metatstable conductance changes associated with the Staebler-Wronski effect can be understood solely in terms of models that involve only local changes in bonding coordination. Attempts to understand or control this phenomenon by altering the long-ranged disorder will not be fruitful.

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Background and Motivation:

The conventional model for the Staebler-Wronski effect in hydrogenated amorphous silicon (a-Si:H) involves the breaking of strained Si-Si bonds due to the nonradiative recombination of photoexcited charge carriers, which create new dangling bond defects. The local motion of hydrogen is typically invoked to stabilize these newly created defects, upon annealing above 420 K sufficient hydrogen diffusion occurs to remove these excess dangling bonds. Recently, models for light induced conductivity degradation have been proposed which are intrinsically nonlocal in nature, involving changes in long-ranged potential fluctuations or strain fields associated with compositional morphology. A large number of recent experimental results suggest that in addition to creating additional dangling bonds, extended illumination yields significant changes in the disorder at the mobility edge, affecting the ability of the material to carry an electrical current. The nonlocal models for the Staebler-Wronski effect presume that light soaking increases or modifies the long range disorder already present in the film, believed to arise from nonuniform distributions of bonded hydrogen as well as potential fluctuations arising from randomly located charged defects. Consequently, even though the Fermi energy is uniform throughout the a-Si:H film, the conductivity will vary spatially, suggesting that electronic transport might best be described by a classical percolation process. However, since the inelastic (phase breaking) scattering length in a-Si:H is believed to be on the order of ~ 5 - 10 Å at room temperature, conventional transport measurements typically do not directly reveal information concerning disorder on much longer length scales.

Nevertheless, there are measurement techniques that are sensitive to long range inhomogeneities in amorphous silicon, such as comparisons of the temperature dependence of the dark conductivity and thermoelectric power (Beyer and Overhof's Q-function). The value of the energy separation between the Fermi energy and the mobility edge in the dark conductivity E_σ is larger than that of the thermopower E_S by as much as several hundred meV for a-Si:H. This activation energy difference is attributed to long-ranged disorder at the mobility edge, since in a conductivity measurement the charge carriers must overcome the highest potential barrier in their path, while in an open circuit thermopower measurement the charge carriers do not necessarily have to overcome the highest potential barrier. Hence in this model the activation energy difference $E_\Delta = E_\sigma - E_S$ is a measure of the amplitude of the long ranged fluctuations. Another experimental measurement that has been suggested to reflect long-ranged disorder in a-Si:H involves the non-Gaussian statistical nature of current fluctuations (1/f noise). Previous noise measurements on a-Si:H films have found that the coplanar current fluctuations have a spectral density which is well described by a 1/f frequency dependence for frequencies f ranging from $1 < f < 10^3$ Hz. Moreover, analysis of the non-Gaussian statistics which characterize the 1/f noise indicates the existence of cooperative interactions between the fluctuators. Increasing the structural disorder of the a-Si:H film by alloying with carbon or increasing the hydrogen microstructure by varying the deposition conditions under which the films are synthesized decreases the correlations of the noise power, suggesting that the non-Gaussian 1/f noise is sensitive to the long-ranged disorder.

This research project involved measurements of the Q-function and the non-Gaussian current noise, as a function of light exposure for a series of n-type doped a-Si:H films grown at varying deposition temperatures, in order to elucidate the role, if any, that changing the long- and medium range order plays in light-induced defect creation.

Notable research accomplishments:

- **A series of undoped and n-type doped hydrogenated amorphous silicon films were synthesized in a plasma enhanced chemical vapor deposition system. The long-ranged disorder was varied by systematically changing the r.f. power during plasma deposition, the substrate temperature, the compensation ratio (that is, the ratio of n-type to p-type dopants simultaneously incorporated into the a-Si:H film) and the inclusion of silicon nanocrystallite within the amorphous silicon matrix.** The long-range disorder at the mobility edge of a-Si:H results both from the non-uniform distribution of bonded hydrogen as well as potential fluctuations caused by randomly located charged defects. The hydrogen microstructure, along with any impurity atoms, will result in spatial variations of the band gap throughout the amorphous material, as will the Coulombic potential fluctuations caused by charged dopant atoms and oppositely charged dangling bonds (see Figure 1). These fluctuations of the mobility edge occur on length scales of ~ 10 to 100 nm, based upon estimates of the hydrogen microvoid density and the average number of charged dopants and dangling bonds in moderately doped a-Si:H. Since the inelastic scattering length in a-Si:H is on the order of ~ 0.5 – 1.0 nm at room temperature, conventional transport measurements cannot be directly used to measure disorder on longer length scales.

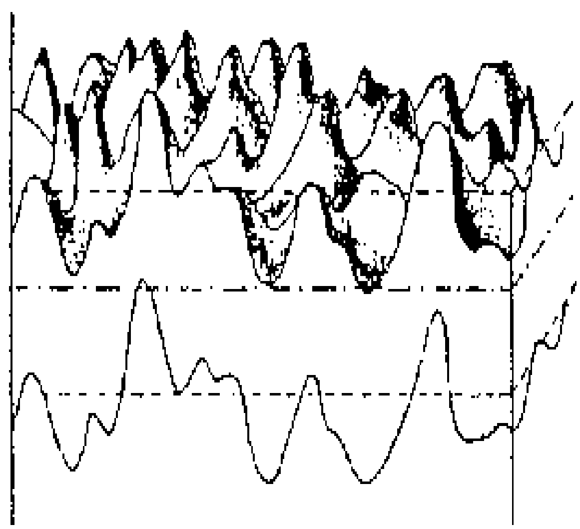

Figure 1: Illustration of the long-range disorder in the valence and conductance bands in hydrogenated amorphous silicon.

- The samples examined in this study were synthesized at the University of Minnesota in a capacitively coupled R.F. (13.56 MHz) system using plasma-enhanced chemical vapor deposition (PECVD) of silane (SiH_4) and phosphine (PH_3), for n-type doping. A uniform gas-phase doping ratio of $[PH_3]/[SiH_4] = 4x10^{-4}$ was used for all films in the first two series. In the first series of films the different samples were deposited at deposition temperatures of 60, 80, 150, and 250 °C, all with a constant R.F. power of 3 W (electrode area ~ 50 cm^2, corresponding to a deposition rate of ~ 2 Å/s). A second series of films was examined in which the films were deposited at R.F. powers of 3, 10, 20, 30 and 40 W (corresponding to deposition rates of ~ 2, 4, 6, 15, and 23 Å/s respectively) and the deposition temperature was held constant at 250 °C. All other deposition parameters were identical for both series of films. All films were deposited onto Corning 7059 glass substrates and range in thickness from 0.5 to 1.4 μm. In addition to the two film series described above, a series of compensated films (that is, counter-doped with both phosphine and diborane) were examined for which the ratio of phosphine (PH_3) to diborane (B_2H_6) was varied. The first of these films was deposited with a doping ratio of $[PH_3/B_2H_6] = 10^{-2}/3x10^{-3}$ at an R.F power of 2 W, the second with a doping ratio of $[PH_3/B_2H_6] = 10^{-2}/10^{-3}$ at an R.F. power of 3 W, and the third film with a doping ratio of $[PH_3/B_2H_6] = 10^{-2}/4x10^{-4}$ at an R.F. power of 10 W (this film was deposited with an electrode separation in the plasma chamber of about 3 inches, as opposed to approximately 1 inch for all the other films). All three films were deposited at a temperature of 250 °C. A series of films were synthesized in a different deposition chamber under conditions of high hydrogen dilution of silane at large gas chamber pressures, which yielded amorphous silicon films containing silicon nanocrystalline inclusions. These materials are undoped, and their higher resistance has made Q-function and 1/f noise measurements difficult. The electronic properties of these films are discussed in detail later in this section.

- **The materials properties of these films were characterized by measurements of their dark conductivity, photoconductivity, thermoelectric power, optical absorption spectrum obtained using Constant Photocurrent Measurements, both after annealing the films in the dark (state A) and following extended illumination with heat-filtered white light (state B) and by measurements of the infra-red absorption spectrum.** Figure 2 shows the infra-red absorption spectra for n-type a-Si:H films deposited at substrate temperatures of 250, 100 and 22 °C. The observed shift in the peak of the absorption curve from 2000 cm^{-1} to 2100 cm^{-1} as the substrate temperature during film growth is lowered is consistent with an increase in the concentration of $Si\text{-}H_2$ bonds, which are typically observed in material of lower electronic quality. All of the films investigated exhibited a light-induced decrease in the dark conductivity (the Staebler-Wronski effect) of varying magnitude, which was reversible upon annealing above 420 K.

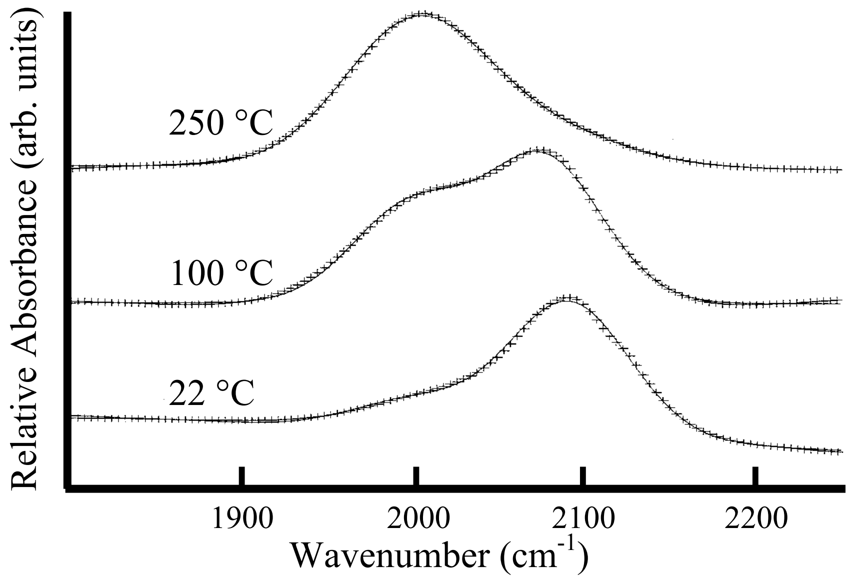

Figure 2: Infrared absorbance of the films deposited at temperatures of 22, 100, and 250 °C.

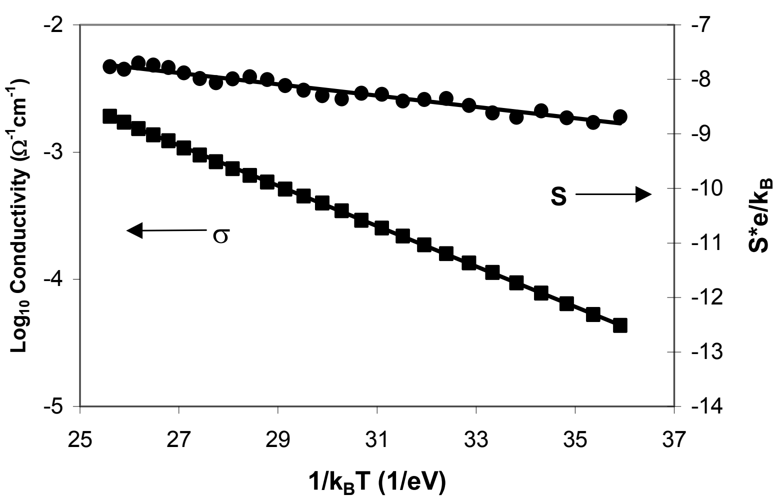

Figure 3: Arrhenius plot of the dark conductivity compared to the thermoelectric power against inverse temperature for an n-type doped a-Si:H film deposited at a substrate temperature of 80 °C. The differing slopes of the two plots indicate that the activation energy for the thermopower is less than that obtained from dark conductivity measurements, and is interpreted as reflecting the long-range disorder at the mobility edge.

- **In order to quantify the long-range disorder of these films, the temperature dependence of the dark conductivity and thermoelectric power were measured.** While for crystalline semiconductors these two measurements yield identical values for the activation energy, in amorphous semiconductors the conductivity activation energy is always larger than that obtained from thermopower (see fig. 3). This difference has been interpreted as reflecting the long-range disorder in these films. We confirmed that a-Si:H synthesized under non-optimal conditions, associated with increased long-ranged disorder, do indeed display larger Q-function values. A convenient means with which to compare these two measurements is the Q-function proposed by Overhof and Beyer,

$$Q = \frac{e}{k_B}|S| + \ln(\sigma(\Omega cm)) = Q_0 - \frac{E_Q}{k_B T} \quad (1)$$

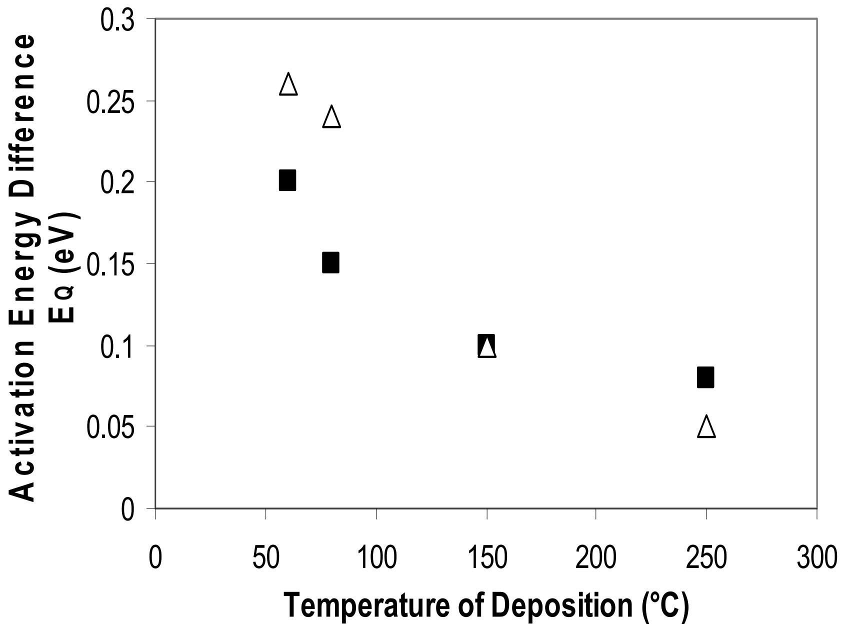

Figure 4: Difference between conductivity and thermopower activation energies (E_Q) as a function of deposition temperature (the squares and triangles represent two sets of measured data).

where E_Q is defined as (E_σ - E_S). In a-Si:H the magnitude of E_Q has been interpreted as a measure of the magnitude of the long-range disorder [5,6] and E_Q is expected to increase as the electronic quality of a sample degrades (as reflected in a lower dark conductivity and higher dangling bond defect density). Figure 4 shows that E_Q increases when the deposition conditions under which the a-Si:H films are grown are varied, such as lowering the deposition temperature, leading to higher hydrogen content and lower electronic quality. An increase in E_Q in n-type doped a-Si:H following light exposure would indicate a shift of the conduction band to higher energies, providing asymmetrical shifts in E_σ and E_S.

- **For all of the materials investigated under this contract, the value of the Q-function, reflecting the electronic long-range disorder, was unchanged following light-induced defect creation** (see figures 5 and 6). This is consistent with the results of Quicker and co-workers (D. Quicker, P. W. West and J. Kakalios, Mat. Res. Soc. Proc. (Materials Research Society, Pittsburgh, PA) **467**, 73 (1997)) who reported no change in the Q-function in films which exhibited either a Staebler-Wronski effect or a persistent photoconductivity effect. However, Hauschidlt and co-workers (D. Hauschildt, W. Fuhs and H. Mell, Phys. Stat. Sol. (B) **111**, 171 (1982)) have reported a large increase in E_Q for a 10^{-4} PH_3 doped a-Si:H film following 60 hours of illumination. To determine whether changes in long ranged disorder appear only after extended light soaking, the Q-function of an n-type a-Si:H film, deposited at 250 C, was measured following 100 hours of continuous light exposure; no change in E_Q was observed.

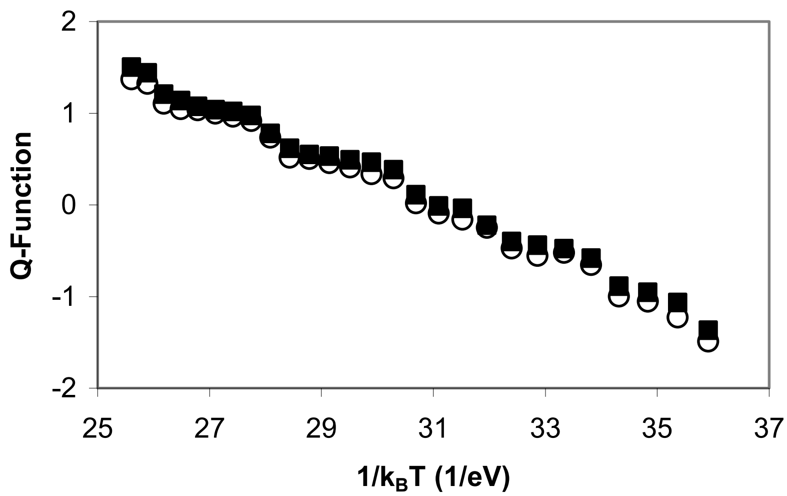

Figure 5: Plot of the Q-function against inverse temperature for an a-Si:H film deposited at 80 °C in the annealed state A (open circles) and after light induced defect creation (filled squares).

- The conductance fluctuations of these films were also investigated. The current fluctuations exhibit a spectral density with an inverse frequency dependence, commonly referred to as "1/f noise". Gaussian noise results from an ensemble of statistically independent fluctuators, and is characterized by a frequency independent second spectra (the spectral density of the time dependent variations in the noise power). In contrast, non-Gaussian 1/f noise typically exhibits a power-law frequency dependence, due to non-linear interactions between the noise

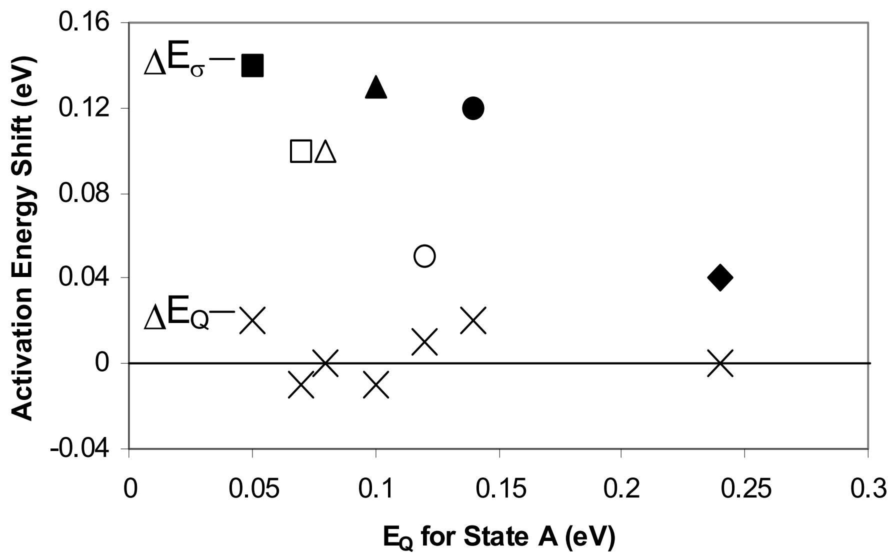

Figure 6: Plot of the shift in activation energy for the conductivity and Q-function with respect to the E_Q value in State A. The solid symbols are the values of $\Delta E_\square$ for the films in the temperature deposition series described in the text. The square, triangle, circle and diamond represent the 250, 150, 100, 80 °C films respectively. The open triangle and open square are for the films deposited at 1 and 10 W respectively with a doping of 4 x 10^{-4}, while the open circle is for the film deposited at 3 W with a doping of 4 x 10^{-3} (all deposited at 250 °C). The value of ΔE_Q for a film is given by the X plotted directly beneath its value for ΔE_σ.

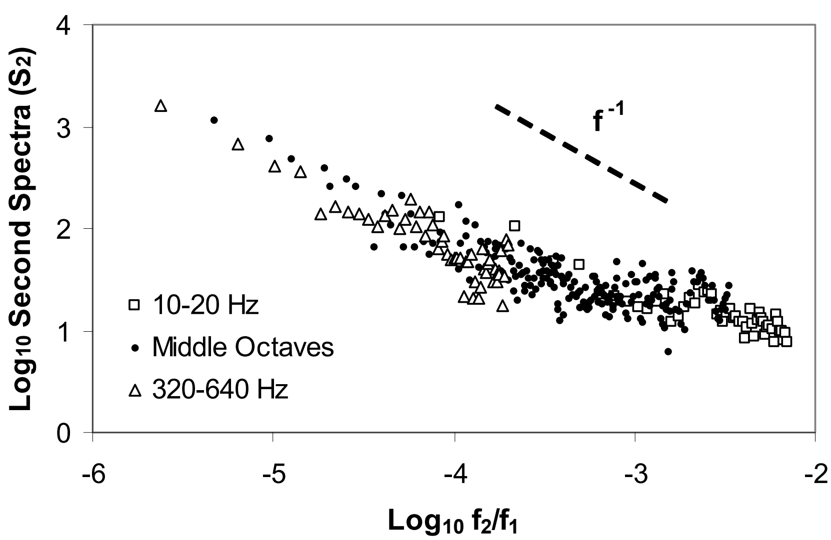

Figure 7: Log-log plot of the scaled second spectra plotted against f_2 over f_1 (the center frequency of the given octave) for the film deposited at 150 °C, measured at 370 °K.

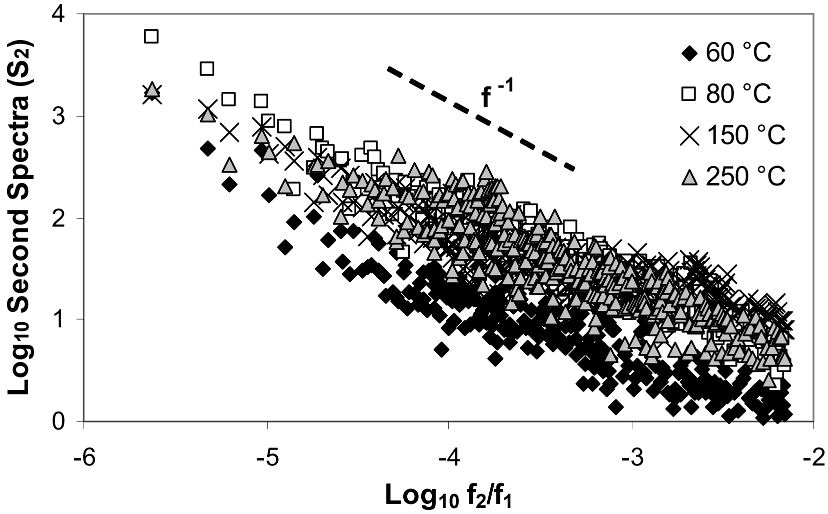

Figure 8: Log-log plot of the second spectra for the films in the temperature deposition series plotted against f_2 over f_1, measured at 370 °K (60 °C film measured at 420 °K).

sources. The statistics that characterize the conductance fluctuations in a-Si:H are found to be non-Gaussian (fig. 7), indicating that the noise results from a small number of interacting fluctuators. The systematic studies carried out as part of this project found that the non-Gaussian character of the 1/f noise was insensitive to variations in the long-range disorder, as varied by altering the deposition conditions of the a-Si:H films described above (fig. 8). **A secondary result of this research project was the determination that the non-Gaussian 1/f noise is insensitive to changes in the long-range disorder in a-Si:H induced by varying the deposition conditions under which the films were grown.**

Methods to alter the medium- and long-range order in a-Si:H through extensive modifications of the deposition process were also investigated. Films deposited under heavy hydrogen dilution and at high plasma gas chamber pressures were obtained, from NREL, USSC and the University of Minnesota in collaboration with faculty in the Prof. U. Kortshagen in the University of Minnesota Mechanical Engineering department. Transmission electron microscopy measurements were performed by Prof. C. Barry Carter of the Department of Chemical Engineering and Materials Science at the University of Minnesota and confirm that these films contain silicon nanocrystalline inclusions uniformly dispersed throughout the amorphous silicon matrix (see figures 9 and 10). These materials exhibited dark conductivity and mid-gap defect densities comparable to a-Si:H films deposited under nominally optimal conditions. **Thin film amorphous silicon containing the nanocrystallite inclusions displayed higher photosensitivities (ratio of photo-to-dark conductivity) and exhibited slightly slower light-induced degradation.** As shown in fig. 11, in some cases the ratio of photoconductivity to dark conductivity was larger in after extended light soaking for these films than the initial state A values for pure a-Si:H. These preliminary results, obtained during the no-cost extension period of the subcontract, are encouraging and seem to warrant further investigation.

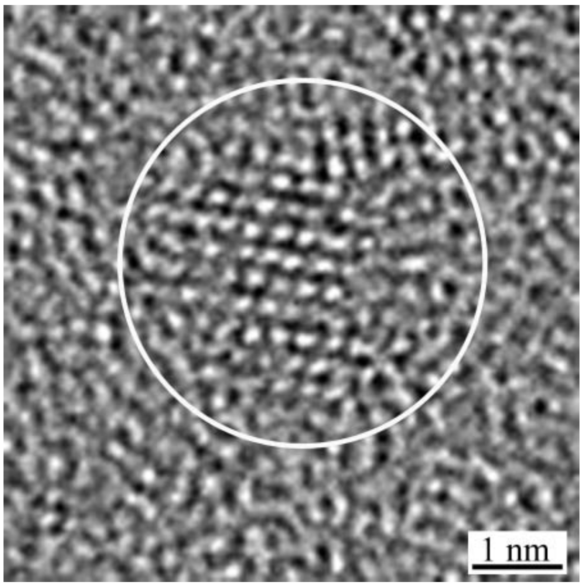

Figure 9: High resolution transmission electron micrograph of a a/nc-Si:H film deposited with a H_2/SiH_4 ratio of 50 and a gas chamber pressure of 1450 mTorr. A nanocrystalline silicon inclusion with an average diameter of 3 nm is clearly indicated within the white circle in the figure. TEM image obtained by Prof. C. Barry Carter of the University of Minnesota.

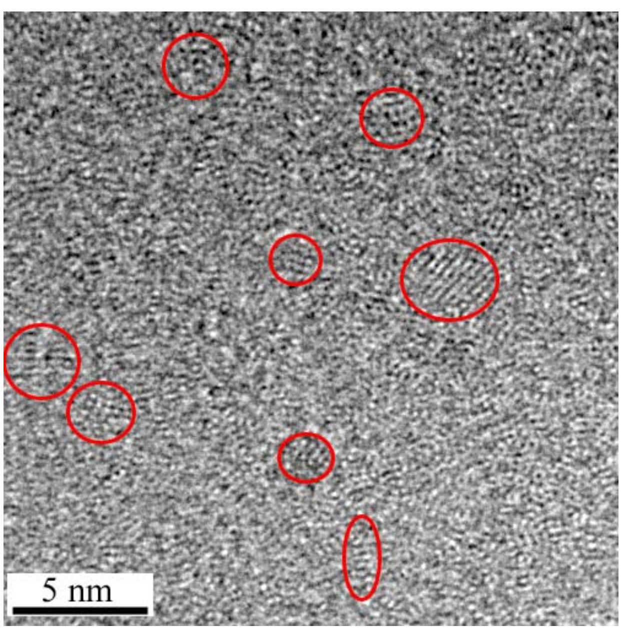

Figure 10: Low magnification Transmission Electron Micrograph of the a/nc-Si:H film shown in fig. 9. Several silicon nanocrystalline inclusions, with an average diameter of 3 – 4 nm, are indicated by the red circles. TEM image obtained by Prof. C. Barry Carter of the University of Minnesota.

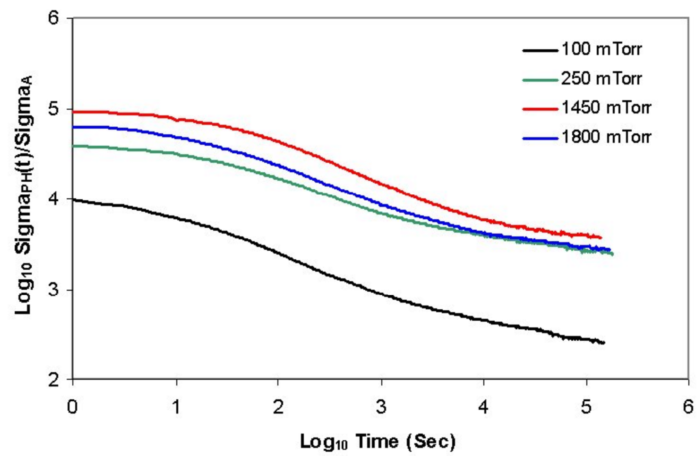

Figure 11: Decay of the ratio of the photoconductivity to the annealed state A dark conductivity for a series of a/nc-Si:H films deposited at high gas chamber pressures (red, blue and green lines) compared to an a-Si:H film (black line) as a function of exposure time due to metatstable defect formation (the Staebler-Wronski effect).

Conclusions:

None of the a-Si:H films studied here, representing a range of structural and electronic quality, displayed any significant change in the Q-function despite appreciable light-induced changes in the dark conductivity. If the Q-function is indeed a measure of the long ranged disorder, then these data indicate that the light induced degradation of the dark conductivity and thermopower results solely from a shift of the Fermi energy deeper into the mobility gap, away from the conduction band edge. We can therefore exclude all models for the Staebler-Wronski effect that involve a shift of the conduction band edge to higher energies, due to modulations in the medium or long-range disorder of the amorphous semiconductor. The results of this research program have narrowed the "phase space" which one must investigate in order to elucidate the mechanisms underlying the light-induced conductance degradation effect.

A secondary result of this research project is the thorough characterization of the non-Gaussian statistics which characterize the 1/f noise observed in a-Si:H as the structural and electronic properties of the films are systematically varied. Contrary to expectations, the second spectra which describes the time dependent fluctuations of the noise power in a-Si:H is found to be fairly insensitive to changes in the deposition conditions under which the films are grown. These results challenge our understanding of the mechanisms responsible for the 1/f noise in a-Si:H, and further investigations are underway to improve our understanding of this phenomenon.

In collaboration with faculty in the Mechanical Engineering Dept. and the Chemical Engineering and Materials Science Dept. at the University of Minnesota, preliminary results have also been obtained in the synthesis and characterization of thin film hydrogenated amorphous silicon that contains silicon nanocrystalline inclusions. These films are grown in a PECVD reactor chamber with the silane gas heavily diluted with hydrogen, and at gas chamber pressures above 1 Torr. We have confirmed, via direct observation using high resolution transmission electron microscopy, that these materials contain nanocrystallite with diameters ranging from 1 to 20 nm, depending on the deposition conditions. Despite the extreme conditions under which these materials were deposited, the electronic conductivity and mid-gap defect density is comparable or better than found for typical a-Si:H films deposited under optimal conditions. While these materials with nanocrystalline inclusions still exhibit a Staebler-Wronski effect, the initial photosensitivities (that is, that ratio of photoconductivity to dark conductivity in State A) are much higher, so that even after extended light soaking their values are still larger than the initial state A values fro pure a-Si:H. Further investigation of this new class of thin film amorphous semiconductors may yield material with superior characteristics for photovoltaic applications.

Publications Supported, in whole or in part, by Contract:

"Long Range Disorder and Metastability in Amorphous Silicon", D. Quicker, P. W. West and J. Kakalios, J. Non-Cryst. Solids, **266-269**, 397 (2000).

"The Staebler-Wronski Effect and 1/f Noise in Amorphous Silicon", T. J. Belich and J. Kakalios, Materials Research Society Symposia Proceedings (Materials Research Society, Pittsburgh, PA) **664**, A14.5 (2001).

"Metastability and Long-Ranged Disorder in Amorphous Silicon", T. J. Belich, J. Huston and J. Kakalios, Electrochemical Society Meeting Proceedings, (Electrochemical Society, Pennington, New Jersey) (2001).

"Medium Range Order and 1/f Noise in Hydrogenated Silicon Thin Films", T. J. Belich and J. Kakalios, Materials Research Society Symposia Proceedings (Materials Research Society, Pittsburgh, PA) **715**, 287 (2002).

"Long Range Disorder and the Staebler-Wronski Effect in n-type Amorphous Silicon", T. J. Belich and J. Kakalios, Phys. Rev. B **66**, 195212 (2002).

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"Hydrogenated Amorphous Silicon Thin Films with Nanocrystalline Silicon Inclusions", T. J. Belich, S. Thompson, C. R. Perrey, U. Kortshagen, C. B. Carter and J. Kakalios, Materials Research Society Symposia Proceedings (Materials Research Society, Pittsburgh, PA) (in press).

"Non-Gaussian 1/f Noise as a Probe of Long-Range Structural and Electronic Disorder in Amorphous Silicon", T. J. Belich, Z. Shen, S. Campbell and J. Kakalios, Proceedings of SPIE vol.

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13. ABSTRACT (Maximum 200 words): The principal objective of the proposed research is the experimental study and elucidation of the role that long-ranged disorder plays in the light-induced metastable conductance changes in hydrogenated amorphous silicon (a-Si:H). The development of sources of low-cost renewable electrical energy is a high national priority. A consensus has developed that a-Si:H is the material of choice for large-area photovoltaic devices. However, the efficiency of a-Si:H-based solar cells decreases by nearly a factor of two following extended illumination, owing to the creation of light-induced defects resulting from the recombination of photo-excited charge carriers. If this light-induced defect formation could be reduced or eliminated, the practical conversion-efficiency of a-Si:H-based solar cells would effectively double, yielding cost per kilowatt-hour values comparable to that of conventional fossil fuel generation of electricity. Recent experimental results and theoretical calculations indicate that associated with light-induced dangling-bond creation, there are alterations in the medium- and long-ranged disorder in the a-Si:H films. A determination of whether or not the long-ranged disorder is affected in the Staebler-Wronski effect is important, for if so, then even when local defect generation is prevented, there could still remain metastable conductance changes that would affect photovoltaic device performance. Alternatively, if experimental studies clearly rule out light-induced changes in the long-ranged order in the a-Si:H film, then entire classes of models for the Staebler-Wronski effect can be eliminated from consideration. The results from this project indicate that the metatstable conductance changes associated with the Staebler-Wronski effect can be understood solely in terms of models that involve only local changes in bonding coordination. Attempts to understand or control this phenomenon by altering the long-ranged disorder will not be fruitful.

14. SUBJECT TERMS: PV; light-induced; metastable conductance; hydrogenated amorphous silicon (a-Si:H); device; solar cells; Staebler-Wronski effect; thermopower activation energy; transmission electron micrograph; photoconductivity; thin film

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