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Self-sustained oscillations in undoped a-Si:H

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Abstract. Self-sustained oscillations and fluctuations have been observed in undoped hydrogenated amorphous silicon (a-Si:H) thin films when small portions of the sample were biased using coplanar electrodes After subjecting the samples to very high voltages several times, the current flow becomes unstable. At constant voltage the current continuously increases ending in the irreversible formation of visible channels between the electrodes which exhibit an almost periodic structure. During this process of pattern formation random fluctuation as well as periodic single-frequency and mode-locked multiple-frequency current oscillations and random telegraph noise have been detected. These phenomena are attributed to microcracks in the sample which change their resistivity by the hopping of hydrogen between different binding configurations.

1. Introduction

Semiconductors represent highly nonlinear systems which are expressed via nonlinear current–voltage relations, negative differential conductivity, spontaneous formation of electric field domains and current filaments and self-sustained regular and chaotic oscillations. In the past, a bulk of work has been carried out on high-purity semiconductors which show instabilities due to impurity breakdown at low temperatures. The observed autonomous oscillations have been analysed in terms of the phenomenological theory of nonlinear dynamics and various routes to chaos could be recognised [1]. Nonlinearities in carrier transport as well as voltage or current fluctuations can reveal details of the impurities and are a highly sensitive probe for imperfections in the material [2].

Here we report on self-sustained oscillations in hydrogenated amorphous silicon (a-Si:H) which were observed at room temperature. A random switching of the conductivity, termed random telegraph noise (RTN), and random fluctuation with a $1/f$ power spectrum have been previously observed in a-Si:H and investigated in great detail [3, 4, 5, 6]. Random telegraph noise, first discovered in reversed biased pn junctions [7] usually occurs in small samples such as submicron silicon devices [8] and metal nano-bridges [9] where random charging and discharging of individual traps has a large effect on the resistance. In a-Si:H, RTN has also been observed in large samples with a coplanar geometry of contacts. The random switching of the resistance has been attributed to the presence of microchannels of small cross section which change their resistance due to the hopping of hydrogen atoms from one bonding configuration to another.

In the present study we show that in a-Si:H other well defined types of autonomous oscillations may also exist. When high voltages applied to coplanar contacts on the top of thin films, a slow run-away of the current at constant voltage occurs, which ends in the formation of irreversible channel-like structures in the material. During the formation of the channels large current fluctuations are observed which change their temporal character with time. Besides random telegraph noise and random fluctuations, the measurements revealed anharmonic oscillations with one fundamental frequency, frequency-locked oscillations with two characteristic frequencies, and intermittent breakdown of oscillations. In a qualitative discussion it is shown that these oscillations may result from hydrogen migration between adjacent current filaments which affects the current flow in a complex way.

At voltages that are not too high, where the current run-away is slow, a sequence of power spectra, phase portraits and Poincaré sections could be reconstructed as function of the current at constant voltage. The current flow through the sample is destabilized at low values of the current, showing first random fluctuations. With rising average current regular oscillations grow out from the fluctuations. These oscillations are low-dimensional deterministic as demonstrated by reconstructions of the phase space. The results show that the tendency in the course of the formation of a channel is from random fluctuations to periodic oscillations. The oscillations cease when the channel is formed with an almost periodic spatial pattern. This observation points to a spatio-temporal mechanism of pattern formation.

2. Experimental details

The films investigated here were undoped a-Si:H deposited by rf glow-discharge decomposition of silane (SiH_4) on fire-polished glass substrates. The substrate temperature was about 400 K. The films were of 0.35–0.4 μm thickness and had a hydrogen concentration of 18–20%. Coplanar non-injecting electrodes were prepared on top of the films by the evaporation of gold. The electrodes had the form of two parallel stripes. The distance between the contacts was 0.3 mm.

The samples were biased in series with a load resistor of between 1 and 5 MΩ by a controllable high-voltage source and, as a probe of the current, the voltage drop across the load resistor was measured. In order to analyse the spontaneous oscillations, time series of the voltage across the sample were digitally recorded with sampling intervals Δt ranging between 20 ns and 20 μs. Recorded signals were used to calculate power spectra, phase portraits and Poincaré sections. The measurements were carried out at ambient temperature.

3. Results

In figure 1 a set of current–voltage characteristics is shown which were recorded by repeatedly sweeping the bias voltage from zero to 1800 V. The sweep rate was 15 V s^{-1}. At the end of each scan the voltage was abruptly switched to zero and kept at zero for 30 seconds. On the first voltage scan applied to the untreated material, the film was practically insulating. With increasing numbers of scans the film got more and more conducting, yielding a superlinear increase of the current as function of the voltage. Finally breakdown occurred which is seen from the pronounced s-type current–voltage characteristics. After several scans (three to five) of the bias voltage the current flow lost stability. Keeping the bias voltage constant below breakdown led to a continuous increase of the average current (typically 3 μA min^{-1}) and the degrading of the sample. The final stage of the current increase was the creation of the visible channel between the contacts. As long as this state had not been reached the degraded samples partly restored their properties after exposition at zero bias voltage. This spatial pattern formation process is always accompanied by low-frequency excess noise, mostly of $1/f$ character, and self-generated oscillations which typically have fundamental frequencies of the order of 10 Hz to 100 Hz. The excess noise depends upon the measured current almost quadratically.

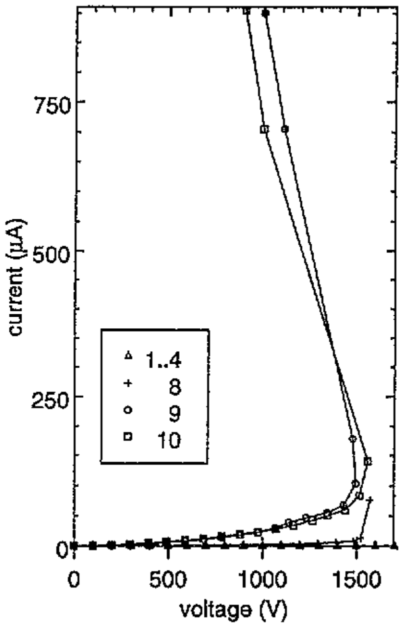

Figure 1. Current as a function of voltage for different scans of the voltage (numbers) applied to as-grown material.

Such channels, photographed through crossed polarizers using a microscope, are shown in figure 2. The channels have a complicated spatial structure. In general several channels are formed which may branch like lightning (figure 2, top panel). Each branch, in turn, consisted of a number of strata where a-Si:H was destroyed. The strata themselves showed an almost periodic spatial pattern (figure 2, bottom panel).

After several sweeps of the bias voltage an increase of the noise was observed which accompanied the recorded current. Above a threshold voltage but well below breakdown, the current started oscillating. Certain spots at the surface of the sample were much more efficient as regards the onset of oscillations as compared with other regions between the electrodes. In some cases the oscillations disappeared after several seconds. To make them more stable 'training' by the use of pulses that exceeded the value of the voltage monitoring the oscillatory process or by voltage pulses of reverse sign was necessary.

Illumination with intense band gap absorption light also stimulates the onset of almost periodic oscillations resembling those observed at higher temperatures in similar samples [10]. The frequency of these oscillations increases with rising irradiation intensity. The oscillations continued after the light was switched off and disappeared in a few seconds.

The main emphasis of this investigation is to characterize the self-sustained oscillations during the formation of the channels shown in figure 2. First we will show a set of different types of oscillations in figure 3 which were observed at different places in the current–voltage plane. The top plate of figure 3 shows a slightly noisy periodic oscillation. The next plate (figure 3(b)) displays an oscillation which begins with spikes whose period continuously decreases with rising current through the sample. The current as a function of time in figure 3(c) is typical for mode locking. The oscillation has two fundamental frequencies which lock

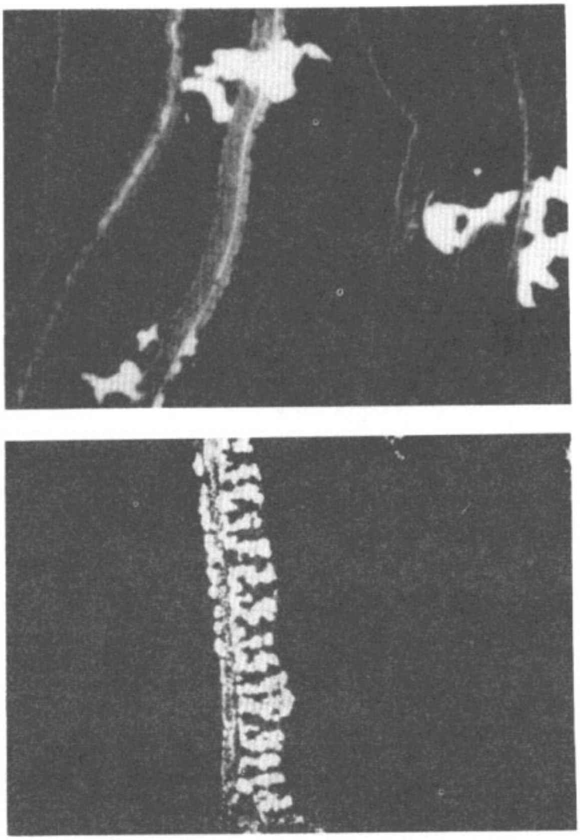

Figure 2. Photographs through crossed polarizers showing irreversible channels of destruction of a-Si:H. Top panel 156× magnified, bottom panel 450× magnified.

with the ratios 1/3 and 1/4. The locking ratio randomly switches between these two values. The next recording (figure 3(d)) shows the characteristic behaviour of intermittency [15]; the amplitude of the periodic oscillation increases up to a threshold where the laminar phase is instantly interrupted. Finally, in the bottom plate a current trace is shown resembling random telegraph noise.

These oscillations suddenly occurred and disappeared or abruptly changed character after a few seconds due to the increase of the current. The value of the load resistor had no significant effect on the occurrence and the type of the oscillations. A direct comparison of time series recorded with different load resistors is not possible because the spontaneous changes occur different times for each scan of the bias voltage at. The observed oscillations, however, seem to be intrinsic and not circuit controlled [16]. Due to the rapid changes of the oscillations a complete analysis of the underlying scenarios was impossible. Only at low voltages was the rate of current increase so slow that the current did not substantially change within a few seconds. Such a time interval (about 10^3 periods) is necessary for a reasonable reconstruction of phase portraits and Poincaré sections. Figure 4 displays such an analysis carried out at a 450 V bias voltage. The plates are ordered with rising current from top to bottom. The figure shows from left to right the current through the sample as function of time and the corresponding power spectrum, the phase portrait and the Poincaré section. The current starts randomly fluctuating and proceeds into more and more regular oscillations

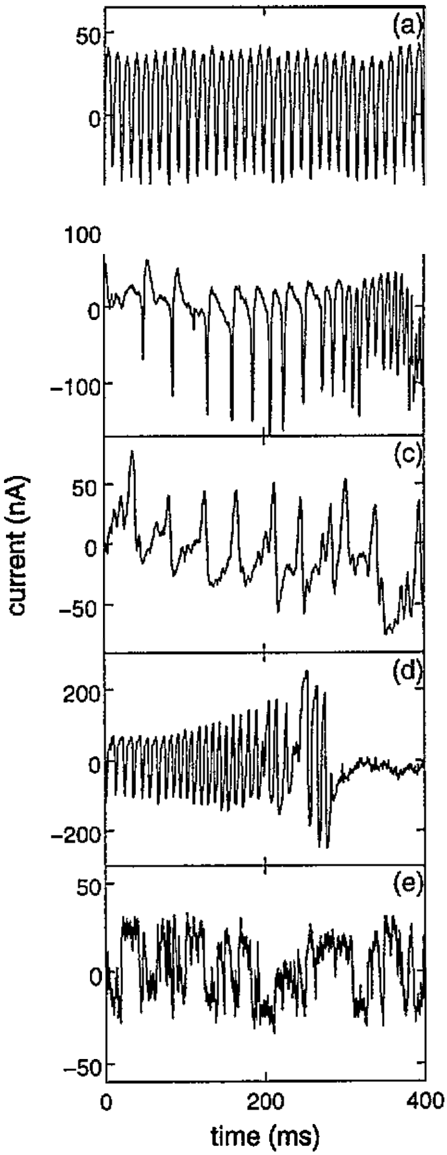

Figure 3. Different types of oscillations observed in a single sample. (a) Periodic oscillations with a single fundamental frequency, (b) spiking with continuously decreasing period, (c) frequency-locked oscillations with two fundamental frequencies, (d) intermittency, (e) random telegraph noise.

with increasing current through the sample. The power spectra display a fundamental in the range of 20–100 Hz and overtones of periodic oscillations with increasing signal-to-noise ratio. In the range of average current where overtones are observable, the noise is of f^{-1} character and the power density decreases somewhat faster; approximately like $f^{-\alpha}$ with $\alpha \simeq 2$. The power spectra, however, are not very conclusive due to the high noise level in the system. The transition from random fluctuations to a low-dimensional deterministic trajectory is most clearly observed in the phase space reconstructions. At high average current when the system is approaching the formation of channels, the phase portrait and the Poincaré section show a limit cycle unambiguously.

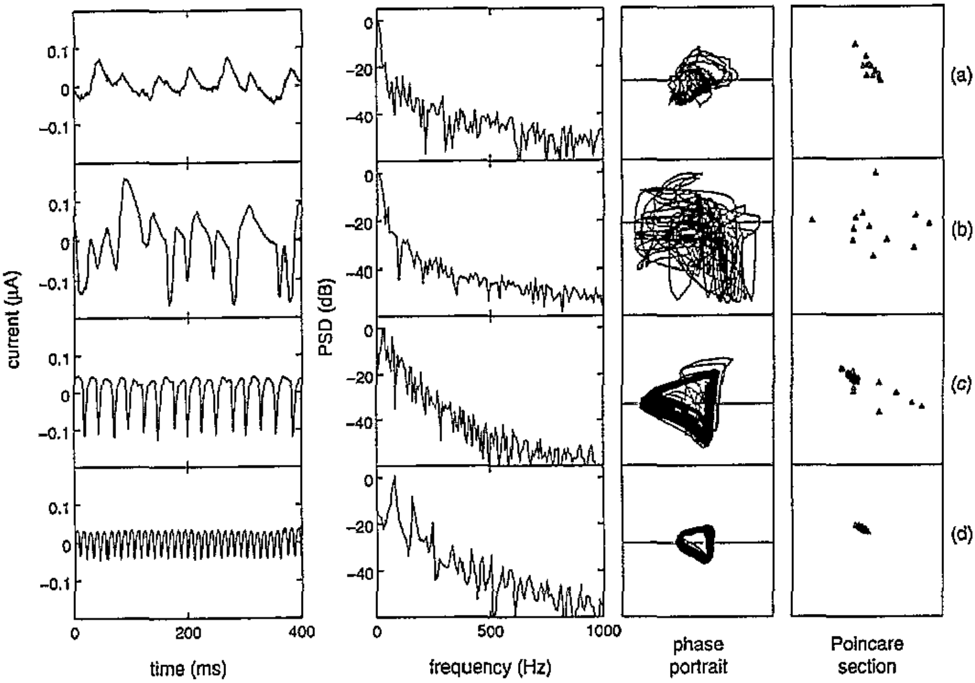

Figure 4. Temporal structure of oscillations during formation of channels observed at 450 V bias voltage after five repeated scans of the voltage. The current increases from top to bottom from 6 μA to 17 μA. From left to right: current as function of time, spectral power density, phase portrait, Poincaré section.

4. Discussion

There is strong evidence that the charge flow in the amorphous hydrogenated silicon is inhomogeneous. This was directly revealed in [11] by scanning a-Si:H films with a small-size mercury and mercury-laser probes. The presence of current filaments was also proved in [5] and used to explain random telegraph noise in a-Si:H structures with coplanar contacts. The most probable explanation for the existence of filaments is the inhomogeneous hydrogen distribution in the volume of the films investigated. The value of the a-Si:H mobility gap is strongly dependent upon the hydrogen content. Thus the energy barriers for charge transport are significantly lower at places with decreased densities of hydrogen atoms. These regions exist at the surfaces of pores and microcracks which are supposed to be the dominant defects in amorphous semiconductors [12]. It should be mentioned that the existence of leakage channels does not mean that the hydrogen content in the regions adjacent to the walls of pores and cracks must be lower compared with the mean concentration in corresponding samples. On the contrary, the hydrogen contents are usually higher due to the passivation of large numbers of dangling bonds in the process of a-Si:H fabrication [10]. Thus it is very probable that leakage channels are located very close spatially to the regions with increased values of mobility gaps.

In the case of coplanar geometry of [11, 5] single leakage channels could be registered. For the coplanar geometry of this paper these channels form a percolation net which carries the main amount of current between the metal electrodes on the surface of a-Si:H (compare with [4], where this percolation net was responsible for the current leading to random telegraph noise, while the total resistance was determined by n-doped regions of a-Si:H films). The observed random telegraph noise was attributed by the authors of [4] to random changes occurring in the percolation net of leakage channels. The connectivity of this net depends on the detailed atomic bonding along the percolation cluster. Consequently, the motion of a single defect or a group of defects (e.g. hydrogen atoms) could decrease the conductivity of the corresponding filament or even close one of the highly conductive paths for the electron transport. Fluctuations of the conductivity of the percolation cluster explain the peculiarities of random telegraph noise. We argue that they can also be responsible for the creation of the self-generated oscillations observed in the present case. The proposed model is based on the following assumptions:

(i) The changes of current in the process of oscillations are due to hops of hydrogen atoms over small distances between the conductive channels of the percolation net.

(ii) The hops of hydrogen are stimulated by the energy released in the process of nonequilibrium electron–hole recombination.

(iii) Nonequilibrium charge carriers are generated in the high resistance regions of closely located conductive filaments.

Suppose we have two filaments which carry the main amount of the current between the electrodes as it is shown in figure 5, and the filaments pass through a high resistance region with high hydrogen content. This region can be associated with a group of small voids or developed microcracks which initially existed in the samples studied. The presence of filaments is confirmed by observations of the samples destroyed and by the fact of local sensitivity of the area under investigation to light. It is natural to assume that the total resistance of both filaments is determined by the joint region with high a hydrogen content. Nonequilibrium electrons and holes are injected into the high-resistance region from the neighbouring low-resistance regions. In the case where the current through one of the filaments dominates, the low-activation-energy processes of hydrogen migration will be more pronounced in this channel. This is a typical manifestation of the Staebler–Wronski effect [13], but the changes of conductivity are more pronounced because of the local character of the current flow that enables hydrogen atoms to leave the cross section of the corresponding filament. This effect results in a decrease of the hydrogen content in one of the channels while the amount of hydrogen in the second channel increases. Then the process is repeated. The system considered has two symmetrical quasistable states. Switching between these two states is known to occur in the case of strongly nonequilibrium conditions. A starting influence is necessary to initiate the self-sustained oscillatory process. Training by voltage pulses is supposed to take the role of this influence. The incident light can also be efficient for this purpose because it results in a conductivity change. This also explains the increase of the frequency as the hydrogen migration process becomes faster. Once the oscillations have started, they continue for some time after turning off the light but the frequency gradually decreases.

Let us mention once more that the observed oscillations can hardly be attributed to thermal effects in the films studied. Even in the case of photoconductivity fluctuations at increased temperatures, with typical periods of the order of tens of seconds, the authors of [10] met serious difficulties when they tried to explain the process via the thermal diffusion of hydrogen. Unreasonably high values of atomic hydrogen diffusion coefficients were necessary for this explanation. In our case, with lower temperatures and much higher frequencies, thermal mechanisms are even more improbable.

The superlinear current–voltage curves are easily explained in the proposed model by the existence of high-resistance regions where charge injection occurs. The gradual increase

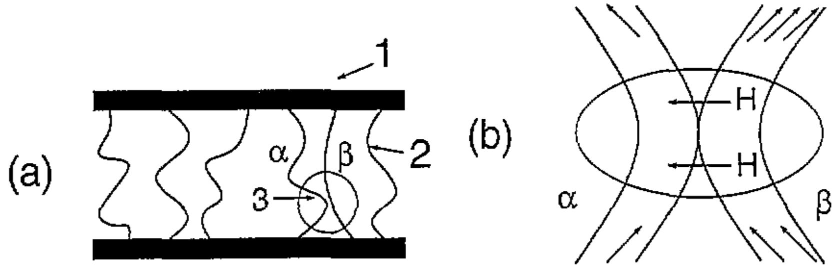

Figure 5. Hydrogen migration leading to current oscillations. (a): (1) Coplanar metal electrodes, (2) a-Si:H film, (3) region with increased hydrogen content, (4) α, β—closely located current filaments passing through (3). (b) Migration of H atoms from β to α.

of conductivity is attributed to losses of hydrogen in the filaments. Similar considerations were applied to explain the increase of currents in the coplanar structures degraded by passing through large currents [14]. The abrupt increase of current and the formation of visible channels can be explained by avalanche breakdown of high-resistance regions. The fact that more than one region of this sort can exist within the length of the filament can account for the observed structure of the channel destroyed and the presence of a number of characteristic oscillation frequencies.

5. Conclusion

In summary, the current flow at high voltages in a-Si:H thin films is destabilized after scanning the voltage several times to values slightly below but close to breakdown. At constant voltage the current gradually increases and finally a visible channel between the coplanar electrodes is formed where the properties of the material are irreversibly changed. In the channels spatially almost periodic strata are created. During the formation of these spatial patterns, the current through the sample shows random fluctuations and regular oscillations of different character. At low average current random fluctuations generally set in yielding an increased noise level. These random fluctuations proceed into more and more periodic oscillation as the point of channel formation is approached. These observations indicate a spatio-temporal pattern formation mechanism in a-Si:H and turbulence in current flow. On a microscopic level this mechanism is attributed to hydrogen migration between adjacent current filaments which grow out of microcracks in the sample.

Acknowledgments

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