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INFLUENCE OF INTERFACE CHARGES ON TRANSPORT MEASUREMENTS IN AMORPHOUS SILICON FILMS

I. SOLOMON, T. DIETL (*)
Laboratoire de Physique de la Matière Condensée, Ecole Polytechnique, 91128 Palaiseau Cedex, France

and

D. KAPLAN
L.C.R., Thomson-CSF, 91401 Orsay, France

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Résumé. — On montre que les grandes variations dans les mesures de transport effectuées sur des films minces de silicium amorphe à faible densité d'états peuvent être expliquées par l'effet d'une densité de charges variable à l'interface silicium-substrat. La quantité et le signe de ces charges ont pu être changés par application d'un champ électrique perpendiculaire au substrat.

L'effet de ces charges de surface peut être utilisé pour mesurer sans ambiguïté la densité d'états dans la bande interdite. On trouve ainsi des valeurs comprises entre 10^{16} et 10^{17} cm^{-3} eV^{-1}, significativement plus faibles que celles déduites des mesures d'effet de champ.

Abstract. — It is shown that the large variations found in transport measurements in thin films of low density-of-states amorphous silicon can be explained by the effect of a variable charge density at the film-substrate interface. The amount and even the sign of these charges have been varied by application of an electric field across the substrate.

These surface charges effects can be used to measure unambiguously the density of states in the gap. It is found to be between 10^{16} and 10^{17} cm^{-3} eV^{-1}, significantly smaller than values deduced from field-effect measurements.

It has been found that hydrogenated amorphous silicon prepared by plasma decomposition of silane SiH₄ [1], by sputtering in presence of hydrogen [2] or by evaporation with subsequent reaction with activated hydrogen [3] is a high resistivity, highly photoconductive, semiconductor. Most of the properties of this material can be explained by a comparatively low density of states in the gap of the order of 10^{17} cm^{-3} eV^{-1} [4].

Our conductivity measurements in thin films of this material, in particular activation energy and photoconductivity studies, are often plagued by apparent erratic variations which in some cases can be very large. It is found that the transport properties of the samples depend upon the thermal, optical and in some cases electrical history of the material.

A somewhat simplified, but striking example of this behaviour has been reported by Staebler and Wronski [5] for a 0.7 μm film of glow discharge amorphous silicon on a substrate of fused silica.

<table>
<thead>
<tr>
<th>State A</th>
<th>State B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Room temperature conductivity (Ω cm)^{-1}</td>
<td>10^{-6}</td>
</tr>
<tr>
<td>Activation energy Eₐ (eV)</td>
<td>0.57</td>
</tr>
<tr>
<td>Pre-exponential factor σ₀ (Ω cm)^{-1}</td>
<td>4 × 10³</td>
</tr>
<tr>
<td>Photoconductivity for 1 μW illumination/cm² (Ω cm)^{-1}</td>
<td>10^{-6}</td>
</tr>
<tr>
<td>Exponent γ of the photoconductivity versus light intensity σₚₑ ≈ Iγ</td>
<td>0.5</td>
</tr>
</tbody>
</table>

(*) On leave from Institute of Physics, Polish Academy of Sciences, 02668 Warsaw, Poland.

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They show that the sample has two completely different sets of properties after annealing above 150 °C (state A) or after intense illumination (state B), the variation induced between the two states being reversible. Their results are summarized in table I.

This behaviour is quite typical and shows the general trend but we find that, for our samples, it is much more complicated. An example of the departure from this simple picture: The annealed state shows a long term evolution in the dark at room temperature and it is quite usual to find a factor of 5 variation of the conductivity after a few weeks. Also, in many cases the different treatments (heat or light) do not give quite reversible results.

We show in this article that most of these effects can be explained by a strong bending of the bands at the surface of the films, due to a variable density of surface charges at the silicon-substrate interface. A particularly convincing argument is the reproduction of these effects by drifting charges in the substrate with a strong applied electric field.

1. Surface effects and apparent conductivity. — All the measurements are performed on hydrogenated amorphous silicon produced by glow discharge decomposition of silane in a reactor described elsewhere [6]. The films of thicknesses ranging from 0.6 to 3 μm are deposited on substrates of pure fused silica (suprasil of 0.25 mm thick) and the conductivity measurements are performed in a planar configuration (Fig. 1).

Around room temperature, and above, the conductivity $\sigma$ is thermally activated:

$$\sigma = \sigma_0 \exp\left(-\frac{E_a}{kT}\right). \quad (1)$$

The systematic variations of the activation energy $E_a$ with the conditions of preparation and the hydrogen content are described elsewhere in the framework of the hydrogen-silicon alloy picture [7].

We are concerned here with the effect of interface charges on the activation energy: because of the bending of the bands, the activation energy is a function $E_a(x)$ of the distance $x$ to the surface of the film (Fig. 2). We want to discuss first qualitatively the effect of this bending in typical cases, anticipating the quantitative results of the calculations of the next paragraph.

![Fig. 1. Planar configuration for conductivity and activation energy measurements of thin films of amorphous silicon.](image)

![Fig. 2. Band bending at the surface of an amorphous silicon film due to a surface density of charges $Q$.](image)

We show below that for quite a moderate density of positive charges $Q$ (of the order of a few $10^{11}$ cm$^{-2}$) near the substrate interface, the activation energy decreases by a value $eV_s = 0.2$ to 0.3 eV. The conductivity in the immediate vicinity of the interface is thus (Eq. (1)) some $10^3$ to $10^4$ times larger than the bulk conductivity. Consequently, although this large surface conductivity extends only in a range $\delta$, calculated below, of about 1/100 of the total thickness $d$ of the sample, the surface conductance is still much larger than that of the bulk.

In a planar configuration (Fig. 1) the conductance of the sample is that of the bulk in parallel with the conductance of the surface:

$$\sigma_{\text{app}} = \sigma_{\text{Bulk}} + \sigma_{\text{Surface}} \frac{\delta}{d}. \quad (2)$$

In many cases, the measured apparent conductivity $\sigma_{\text{app}}$ is dominated by the surface conductivity and all the information on the transport properties of the bulk are lost. The apparent activation energy is then mostly that of the surface and the pre-exponential factor has a reduced apparent value $\sigma_0 \delta/d$.

It is then quite clear that if the heat and light treatments vary the density of charges at the substrate-silicon interface, the apparent transport properties of the film will show large variations. For example, we find as in reference [5] that in general high temperature (150° to 200 °C) annealing tends to increase the conductivity and to decrease the activation energy and the pre-exponential factor. In our model, this is explained by an increase of the number of net positive charges at the interface, resulting in a lowering of the conduction band edge with respect to the Fermi level. The inverse effect is obtained by visible light illumination which tends to decrease the band bending. It is plausible to assume that the electrons, photo-created in the silicon, tunnel towards the positive charges in the substrate thus explaining a partial neutralization of these charges.
The model of a variable conductance due to interface charges is convincingly supported by the following electric field effects. Starting with an annealed sample, we apply for a few hours a strong electric field (5 x 10^4 V/cm) across the substrate maintained at a temperature near 150 °C. This is most simply done by putting the sample on a hot plate (Fig. 3), the silicon film down, electrically grounded by the plate, and then applying a high voltage on an electrode put on the top.

![Diagram](attachment:image.png)

**Fig. 3.** a) Experimental arrangement to vary the surface density of charges Q at the substrate-silicon interface. A positive or negative applied voltage \( V_A \) (\( V_A = + 2500 \) V or \( -2500 \) V) respectively increases or decreases the density of positive charges at the surface. b) When a large negative voltage is applied for a long time (~20 h at 150 °C), the charges Q at the interface can become negative and the bands are then bent as shown. For a large bending, the conductivity is carried by holes and the measured activation energy \( E' \) is that of the excitation of holes into the valence band. (State C.)

When the applied electric field is positive, we obtain after a few hours a sample which has a larger apparent conductivity and a lower activation energy than the annealed sample. Clearly, the electric field has been able to drift positive charges towards the silicon surface, thus increasing the band bending.

The application of a negative electric field shows some particularly interesting effects. During the first few hours, we obtain as expected a decrease of the conductivity and an increase of the activation energy, resulting after 8 hours in a state similar to that obtained by light irradiation. But if the experiment continues beyond 10 hours, the activation energy decreases again, resulting, after 20 hours, in a new state (that we could name state C) having a reduced activation energy as in the case of the annealed state (Fig. 3b). A straightforward explanation is the following. In the first phase, the applied electric field has drifted the positive charges out of the surface and/or negative charges towards the surface, resulting in a decrease of the band bending. After a long time the accumulation of negative charges is such that we have been able to bend the band upwards, the activation energy in state C being then the activation of holes towards the valence band. This is more than a conjecture: we have verified by the sign of a field effect measurement that the conductivity in state C is carried by holes. It is worth noting that in the process we have passed through the interesting flat band state.

Thus the application of an electric field on the heated (150 °C) substrate provides an unambiguous way to vary the band bending near the silicon surface and this technique has been applied for the quantitative comparison between theory and experiment of the next paragraph. However, it is not in practice as attractive as expected and the method is quite tedious for a precise control of the band bending. There are definite fatigue effects, and after a few cycles the drifting of the charges becomes more and more difficult, lengthy and irreproducible. It turns out, for a given sample, to be somewhat of a gamble to achieve a precise flat band condition which furthermore is quite unstable. A better control of the experimental conditions would require a more complete study of the motion of charges in silicon oxide [8] which is well beyond our field of interest.

2. **Surface band bending: quantitative study and comparison with experiment.** — A detailed study of the field effect in amorphous silicon has been made by Madan and co-workers [9] at the University of Dundee. Their approach has been to derive the density of states in the bulk from the field-effect response \( \delta \sigma / \delta E_T \), where \( E_T \) is an applied transverse field. Using a step-by-step approach one can derive the band bending for each value of \( E_T \) and thus construct the density-of-states function. A necessary assumption however is that the applied field \( E_T \) is not shielded by interface states. If this assumption is not valid, one is unable to relate accurately the band bending to \( E_T \) and the deduced density of states is overestimated.

Our approach consists in deducing the band bending from the activation energy observed in the temperature dependence of the resistance. This can be done even if \( E_T \) is unknown as is the case when it is due to uncontrolled fixed charges. We derive below the appropriate formulae (Fig. 2), limiting ourselves for simplicity to the case of a constant density of states in the vicinity of the Fermi level. In general, especially in the case of a large band bending, the variation of \( g(E) \) has to be taken into account and numerical calculations can be made to fit the density-of-state shape to the experimental data.

The bending of the band is given by the potential \( V(x) \) due to the influence of the density of charges \( Q \) at the surface. Taking the potential of the bulk as origin, \( V(x) \) is very simply calculated by solving Poisson's equation:

\[
\frac{\delta^2 V}{\delta x^2} = -\frac{\rho(x)}{\varepsilon \varepsilon_0}.
\]
The space charge density in the silicium, $\rho(x)$, being given, for a constant density of states $g$, by

$$\rho(x) = -e^2 g V(x) .$$ (4)

The solution is then simply

$$V(x) = V_s e^{-x/A}$$ (5)

where the Debye length $A$ has the value

$$A = \frac{1}{e} \sqrt{\frac{e \epsilon_0}{g}} .$$ (6)

The surface potential $V$ is obtained by the condition of total neutrality :

$$V_s = -\frac{Q}{\sqrt{\epsilon_0 e g}} .$$ (7)

Taking a density of states $g = 10^{17}$ cm$^{-3}$ eV$^{-1}$, typical for glow discharge amorphous silicon [4], this gives $A = 8.5 \times 10^{-2}$ $\mu$m. A surface potential of $|V_s| = 0.24$ V is obtained for a surface density of charges $Q/e = 2 \times 10^{11}$ cm$^{-2}$. This can be the actual density of surface charges, which is rather below the typical values found in silica substrates [8]. Or it can be the effect of a larger density of charges partially shielded by interface states. Thus for a rather modest effective density of charges, we predict quite a large effect on the surface conductivity.

Now the activation energy $E(x)$ varies along the normal to the surface and is given from equation (5) by :

$$E(x) = E_a - |eV_s| \exp(-x/A)$$ (8)

where $E_a$ is the activation energy of the bulk and we have explicitly considered the case of a decrease of the activation energy at the surface (effect of positive charges at the interface).

Quite generally, the conductivity $\sigma(x)$ at different locations of the film is given from equation (1) :

$$\sigma(x) = \sigma(\text{bulk}) \exp(eV(x)/kT) .$$ (9)

For the simple case of a constant density of states, $V(x)$ is given explicitly by equation (5), and $\sigma(x)$ has the form :

$$\sigma(x) = \sigma(\text{bulk}) \exp \left( \frac{eV_s}{kT} \exp(-x/A) \right) .$$ (10)

In a planar configuration the conductivity of a film of thickness $d$ is obtained by summing the currents at the different $x$'s :

$$\sigma_{\text{app}} = \frac{1}{d} \int_0^d \sigma(x) \, dx .$$ (11)

This integral has to be performed numerically. However for the discussion it is useful to remark that the integral

$$f(A) = \int_0^d \exp[A \exp(-x/A)] \, dx$$ (12)

is very well approximated, for $d \gg A$, by the function

$$f(A) \approx d + \frac{A}{A-1} e^A .$$ (13)

The error is less than 3 % for $A \geq 4$. Thus, for a decrease of the activation energy at the surface larger than about $4kT$, the apparent conductance can be explicitly obtained from equations (10), (11) and (13). It is convenient to write the result in the form :

$$\sigma_{\text{app}} = \sigma_B + \frac{\delta}{d} \sigma_s$$ (14)

where the bulk conductivity $\sigma_B$, the surface conductivity $\sigma_s$ and the effective surface thickness $\delta$ are respectively

$$\sigma_B = \sigma_0 \exp(-E_a/kT)$$

$$\sigma_s = \sigma_0 \exp \left( -\frac{E_a - |eV_s|}{kT} \right) ,$$ (15)

$$\delta = \frac{|eV_s|}{kT} - 1 .$$

The form of the apparent conductivity $\sigma_{\text{app}}$ (eq. (14)) shows that a transport experiment in the planar configuration measures the conductivity of the bulk in parallel with a sheet of thickness $\delta$ having in general the much larger surface conductivity $\sigma_s$. When the surface conductance (2nd term of eq. (14)) is much larger than that of the bulk, the activation energy is that of the surface :

$${\begin{array}{c}
\sigma_{\text{app}} \sim \sigma_0 \exp \left( -\frac{E_a - |eV_s|}{kT} \right) , \\
\sigma_{\text{app}} \sim \sigma_0 \delta/d , \\
\sigma_{\text{bulk}} \gg \delta \sigma_0 , \end{array}}$$ (16)

When $|eV_s|$ is not too large, at a certain temperature surface and bulk conductance become of the same order and the log $\sigma$ versus $1/T$ curve departs from a straight line : the low temperature part gives the surface activation energy, whereas the high temperature lines trends toward the bulk activation energy.

With these formulae, we can give a coherent picture of the effects described by Staebler and Wronski [5] and quantitatively explain their results (see Table I and Fig. 4). The high activation energy state (state B) obtained after a long illumination is probably very near the flat band condition so that, for that sample :

$$E_a = 0.87 \text{ eV} \quad \text{and} \quad \sigma_0 = 10^5 \text{ (} \Omega \text{ cm)}^{-1} .$$
After annealing, the surface bending of the bands is restored (state A), the surface activation energy is lowered and the measurements are dominated by the surface conductance. In that state, the experiment gives \( \sigma_{\text{app}} = 4 \times 10^{2} \, (\Omega \text{ cm})^{-1} \) and
\[
| e V_s | = 0.87 - 0.57 = 0.3 \, \text{eV}.
\]
Using equations (6), (7) and (15), we deduce:

- the effective surface thickness:
  \( \delta = 2.8 \times 10^{-2} \, \text{µm} \),
- the Debye length:
  \( \Delta = 0.295 \, \text{µm} \),
- the density of states:
  \( g \approx 10^{16} \, \text{cm}^{-3} \, \text{eV}^{-1} \),
- the surface density of positive charges in state A:
  \( Q_e \approx 8 \times 10^{10} \, \text{cm}^{-2} \).

It is interesting to notice that this picture explains also the results of the photocconductivity measurements without further adjustable parameter. According to the photocconductivity study of Anderson and Spear [10], the photocconductivity of a low activation energy material is much larger than that of a high activation energy material, the turning point being at about \( E_a \approx 0.64 \, \text{eV} \). Extrapolated to 1 µW/cm² of incident light, the ratio is about 10^4. Now according to our picture, in state A, with a Debye length of 0.295 µm, only about 1/10 of the sample has an activation energy below the limiting value of 0.64 eV, so that the apparent photocconductivity in that state is reduced by a factor of 10 (Fig. 4). This explains a photocconductivity ratio of 10^4 between the two states instead of the larger ratio of 10^4 expected from their activation energies of 0.57 and 0.87 eV.

The field-effect measurements give a very small response \( d\sigma/dE_t \) showing that in our samples a large density of surface states exists at the silicon-substrate interface. The effect is 10 to 100 times smaller than expected if there was no shielding by the surface states and thus cannot be used to vary significantly the band bending. However, the sign of the field effect has been used to give unambiguously the type of carriers responsible for the surface conductivity.

The experimental points are well explained by a density of states \( g(E) \) of about \( 5 \times 10^{16} \, \text{cm}^{-3} \, \text{eV}^{-1} \) at 0.5 eV below the conduction band edge and one order of magnitude larger for \( E_a = 1 \, \text{eV} \) (see insert of Fig. 5). This is substantially lower than the estimates of the Dundee group [9], which exceed \( 2 \times 10^{17} \, \text{cm}^{-3} \, \text{eV}^{-1} \) throughout this range. We remark that most of the experimental points, for a sample having had a normal treatment, are scattered from 0.65 to 0.72 eV, and 10^{-10} to 8 \times 10^{-10} \, (\Omega \text{ cm})^{-1}. It is then tempting, as it is usually done, to settle for an average value, for example in this case \( E_a = 0.69 \, \text{eV} \) and \( \sigma = 2 \times 10^{-10} \, (\Omega \text{ cm})^{-1} \) whereas our analysis gives what we believe are the actual bulk values:

\( E_a = 0.77 \, \text{eV} \) \( \sigma (\text{bulk}) = 3 \times 10^{-11} \, (\Omega \text{ cm})^{-1} \).

Figure 5 shows the compiled data for one of our samples deposited at 395 °C and of thickness \( d = 0.62 \, \text{µm} \). The low activation energy points (below 0.6 eV) have been obtained by application of a large electric field, positive or negative, across the substrate. The other points have been obtained either by the same method or by annealing (150 °C to 170 °C) or visible light illumination.

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\( E_a = 0.77 \, \text{eV} \) \( \sigma (\text{bulk}) = 3 \times 10^{-11} \, (\Omega \text{ cm})^{-1} \).
3. Conclusion. — Transport properties are important in the characterization of the amorphous silicon films prepared by different methods. Unfortunately, large scattering is found in this type of measurement, and, even worst, the scattering for a given sample is often of the same order as the scattering between different samples, making correlation studies rather difficult and unprecise.

We believe that most of these variations can be explained by the surface effects described in this article. The result of our analysis is that, very often, the measured activation energy for undoped hydrogenated amorphous silicon films is lower than that of the actual value of the bulk. We suspect also that some of the very low values of the pre-exponential factors $\alpha_0$ found in the literature are due to the same effects.

A by-product of this study is a determination of the density of states in the material. We find, especially in the region of 0.5 eV below the conduction band a rather small value (between $10^{16}$ and $10^{17}$ eV$^{-1}$) which correlates well with magnetic resonance studies [11, 12]. This value is substantially smaller than the values found by Madan and coworkers, which always exceed $2 \times 10^{17}$ cm$^{-3}$ eV$^{-1}$ [9].

The type of measurements shown in figure 5, although quite tedious, could be used for an unambiguous determination of the density of states of the different materials. Further work is in progress to try to make this method more practical by a better control of the drifted surface density of charges, and to extend the range of energies in the gap by using doped material. Preliminary experiments show that, in general, the band bending on the free surface of the films is much less than at the silicon-substrate interface. But further studies on the effect of different surface treatments are required for a good control of the free-surface states.

The effect of surface states in insulating substrates could be overcome by using a sandwich configuration for the transport measurements, at the price, however, of other difficulties that we were not able to overcome: non-negligible contact resistances which may be non-ohmic, possibility of injecting electrodes, etc. The present study was prompted by an interest in spin-dependent photoconductivity [13] and magnetoresistance studies of photoconductivity, for which a planar configuration is best adapted because of the required combination of microwave and light irradiation. We had found also in these experiments some erratic results similar to that of transport studies. It is quite obvious that the same uncontrolled band bending at the surface is responsible for these variations. The techniques described in this article are being used for our spin-dependent studies [14], resulting now in a good control of the experimental reproducibility.

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