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The discontinuity in the conductivity characteristics of undoped a-Si:H: statistical shift or metastability?

Pablo Jensen, Robert Meaudre and Mireille Meaudre

Département de Physique des Matériaux, Université Claude Bernard, Lyon I,
43 Boulevard du 11 Novembre 1918, 69622 Villeurbanne Cédex, France

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Abstract. We have tested two explanations for the discontinuity—or ‘kink’—in the DC conductivity versus $1/T$ characteristics of undoped a-Si:H. The statistical shift of the Fermi level leads one to predict the kink qualitatively but it is more difficult to see how it can be used in accounting for the data quantitatively. On the other hand, thermal equilibrium processes, although not quantitative, provide a general explanation for discontinuities in both doped and undoped a-Si:H. From the experimental tests carried out we infer that, although a contribution from the statistical shift cannot be excluded, the most likely cause of the kink is the metastability.

1. Introduction

It has been widely [1–3] observed that Arrhenius plots of the DC conductivity of undoped and doped a-Si:H show two linear regions intersecting at T_k , the kink temperature, which usually lies in the range 100–200 °C. Figure 1 shows a typical plot obtained with an undoped sample. Two attempts at providing an explanation for such results have been made for undoped material:

(i) Yang and Lee [1] cited an idea of Anderson and Paul [4] and suggested that heterogeneity is responsible for the kink;

(ii) Spear *et al* [2] supposed that there were two regimes of behaviour for the temperature dependence of $E_C - E_F$, and that E_C (the mobility edge energy) stops changing once it has reached (at $T = T_k$) a certain energy E_A , defined as ‘the onset of the band tail states’.

The disadvantage of these two explanations is that they introduce new assumptions. A more general explanation, which includes doped material, has been given by Overhof and Beyer [3, 5]. They studied the statistical shift of the Fermi level E_F extensively and concluded that it could explain the kinks. This model was generally accepted and some detailed calculations have been made in order to reproduce the kinks quantitatively [6, 7].

However, Overhof has admitted recently (see [8, 9]) that for doped a-Si:H the statistical shift of E_F cannot explain the kink and that the kink arises from thermal equilibrium processes, as demonstrated by the Xerox group [10, 11]. As thermal equilibrium processes are also present in undoped a-Si:H [12–15], the question arises as to

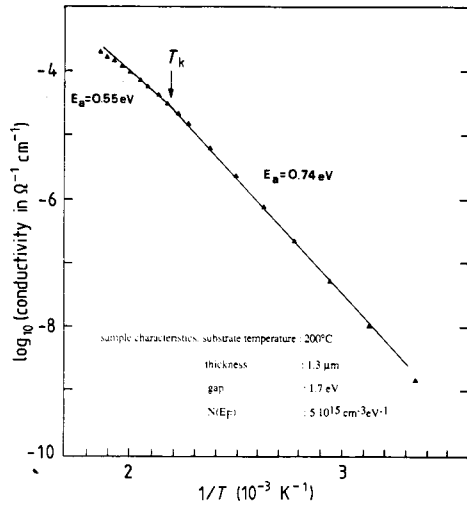


Figure 1. Full line: temperature dependence of the DC conductivity; \blacktriangle , calculated conductivity.

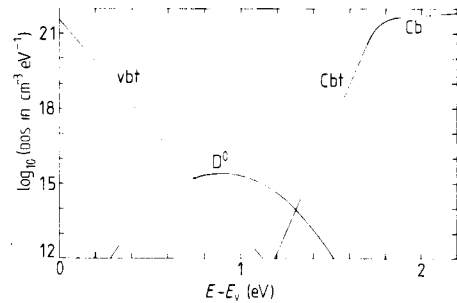


Figure 2. DOS model given in section 2.2.1. D^0 : dangling bond; C: conduction; v: valence; b: band; t: tail.

whether they do provide an acceptable explanation for undoped a-Si:H. In the following we shall investigate these two explanations; first the statistical shift, and then the metastability.

2. Statistical shift

In section 2.1 we show how the kink can be qualitatively explained in terms of the statistical shift. In section 2.2 we try, using a simple model of the density of states (DOS) to reproduce the kink quantitatively. Then we test the model and show that we meet with some difficulties in attempting to reproduce the data.

2.1. Qualitative analysis

To deduce the position of the Fermi level, one has to solve the neutrality equation, which should be written for undoped a-Si:H as

$$N^+ + p = N^- + n \quad (1)$$

where: p is the hole density in the valence band tail and below E_v ; $n = n_{BT} + n_C$ where n_{BT} is the electron density in the conduction band tail and n_C is the electron density above E_C ; and N^+ and N^- are the densities of charged defects (such as dangling bonds or impurities).

At room temperature, $n \approx 10^{11} \text{ cm}^{-3}$ and $p \approx 10^{10} \text{ cm}^{-3}$ ($E_C - E_F \approx 0.7 \text{ eV}$)—much smaller [5] than N^+ and N^- . Then (1) becomes $N^+ = N^-$ and $E_C - E_F$ is only weakly temperature dependent (we are not considering the red shift of the gap): $(E_C - E_F)(T) = (E_C - E_F)(0) + \gamma kT$ with $|\gamma| < 0.5$ (computer simulations using a dangling bond Gaussian peak and a charged impurity level, with both densities in the range 10^{15} – 10^{16} cm^{-3}).

But as the temperature rises, thermal excitation will increase n (and p , but $p < n$ as shown by our computer simulations) and the condition $n \ll N^+$, N^- will no longer hold. Then E_F will have to move towards the valence band edge ($|\gamma| \approx 4$ from the same computer simulations as before) in order to increase N^+ and reduce n . This generates the kink since the activation energy of the conductivity, given by the value $(E_C - E_F)(0)$ of $(E_C - E_F)(T)$ extrapolated to $T = 0$ K, is reduced. Thus our modelling predicts that the kink temperature, T_k , is the temperature at which n becomes of the same order of magnitude as N^+ and N^- .

2.2. Quantitative example

To illustrate and quantify the analysis given in section 2.1. we shall consider a simple model of the density of states (DOS).

2.2.1. Density of states (figure 2). We adopt a model of the DOS similar to that of Kocka *et al* [16]. This model assumes a Gaussian-shaped defect distribution between the exponential band tails. This distribution is attributed to the dangling bonds, the Gaussian shape arising from differences between the local environments of the Si atoms. The detailed composition of the DOS is now described.

(i) *The valence band tail.* For $E_C \geq E \geq E_v$ (we neglect the number of holes below E_v) we take an exponential valence band tail [17]: $g_v(E) = N_v \exp[-(E - E_v)/W_v]$; $N_v = 3.6 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$. $W_v = 0.051 \text{ eV}$ as obtained experimentally (section 2.2.3). We assume that W_v is given by the Urbach energy [17]. We note, however, that transport experiments often find slightly lower values for W_v . This detail is of no significance for our present discussion, as it will turn out that the hole density is always negligibly small.

(ii) *The conduction band and band tail [18].* For $E \geq E_C$: $g_C(E) = A_C(E - E_{C0})^{1/2}$; $A_C = 8.05 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-3/2}$; $E_{C0} = E_C - 0.2 \text{ eV}$.

For $E_C \geq E > E_C - 0.1 \text{ eV}$: $g_{BT}(E) = A_{BT}(E - E_C) + B_{BT}$; $A_{BT} = 3.2 \times 10^{22} \text{ cm}^{-3} \text{ eV}^{-2}$; $B_{BT} = 3.6 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$.

For $E \leq E_C - 0.1 \text{ eV}$: $g_{BT}(E) = A'_{BT} \exp[(E - E'_{C0})/W_C]$; $A'_{BT} = 4 \times 10^{20} \text{ cm}^{-3} \text{ eV}^{-1}$; $E'_{C0} = E_C - 0.1 \text{ eV}$; $W_C = 0.026 \text{ eV}$.

(iii) *Defects.* $g_D(E) = (N_0/W_0 \sqrt{\pi}) \exp\{[(E - E_0)/W_0]^2\}$; $E_C - E_0 \approx 0.9 \text{ eV}$ [16, 19]; $W_0 = 0.22 \text{ eV}$; $N_0 \sim 10^{15} \text{ cm}^{-3}$ and is adjusted to fit the data.

The red shift of the band gap with rising temperature is simulated by a temperature dependent $E_C - E_0$ given by [7]:

$$(E_C - E_0)(T) = (E_C - E_0)(0) - 2.5 kT$$

where $(E_C - E_0)(0) = 0.92 \text{ eV}$, to fit the data in figure 1.

The correlation energy for double occupation of dangling bond states has been assumed to be 0.4 eV and we adopt the correlated-spin statistics given by Okamoto *et al* [20].

This simple DOS implies [21] that at low temperatures, when the neutrality equation becomes $N(D^-) = N(D^+)$ (D^+ and D^- are charged dangling bonds), E_F is pinned at $E_0 + U/2$.

2.2.2. Computer simulation. Our program calculates $E_F(T)$ using the neutrality equation (1) and the DOS described in section 2.2.1. Once the position of E_F has been obtained,

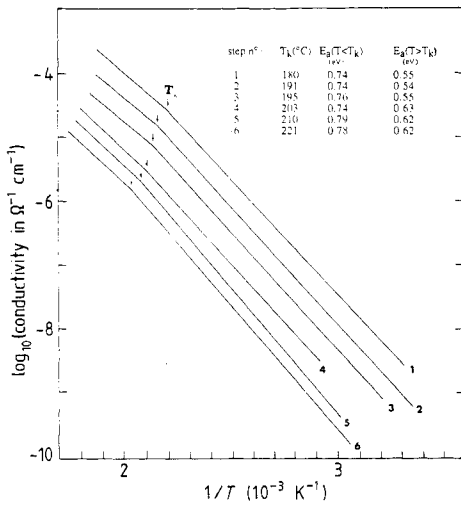


Figure 3. Temperature dependence of the DC conductivity after subsequent annealing steps: 1, 250 °C, 10 min; 2, 300 °C, 5 min; 3, 350 °C, 5 min; 4, 350 °C, 3 h; 5, 350 °C, 3 h; 6, 350 °C, 4 h. To facilitate comparison, each curve has been shifted from the preceding one by a factor of 2: for step i we have plotted $\sigma_i/2^{i-1}$.

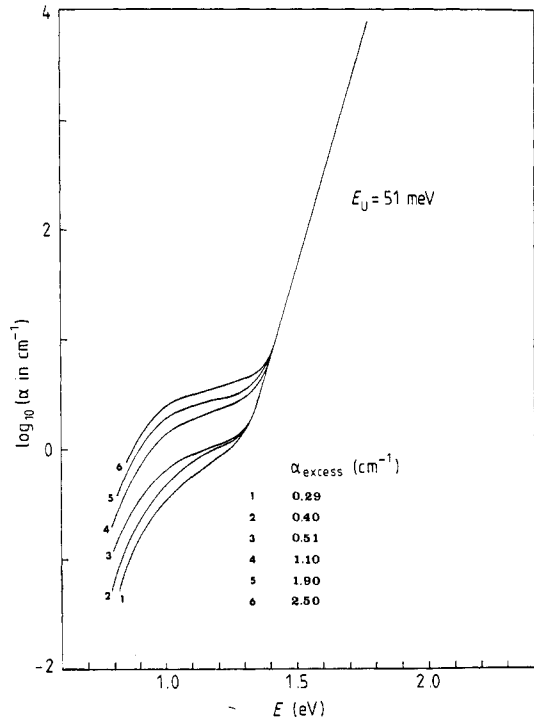


Figure 4. Absorption measured by CPM after subsequent annealing steps (see figure 3). E_U : Urbach tail.

the DC conductivity is easily deduced. It is assumed that the conductivity is given by $\sigma = e\mu n_C$ where μ is the free carrier mobility ($\mu = 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) [10]. Figure 1 shows the calculated temperature dependence of σ . We can see that, as expected, the calculated conductivity shows a downward kink. Moreover, we checked (table 1) that the kink occurs when n becomes of the same order of magnitude as $N(D^+) + N(D^-)$.

2.2.3. Model tests; experimental procedure. The main consequence of our modelling is that T_k depends on N_0 , the total defect density. Increasing this latter quantity should also increase T_k , if nothing else is changed. Therefore we can test our model, provided that we are able to increase N_0 . We notice that Toth *et al* [22] were able to eliminate the kink by holding their samples at up to 350 °C for ≈ 3 h. In addition, their conductivity characteristics were not changed much by this treatment. So we decided to hold our samples at high temperatures ($T \geq 300$ °C) and to see whether T_k increased and whether this increase was correlated with an increase in the defect density or not.

The experimental procedure was the following. The samples prepared by glow discharge under standard conditions [15] were given chromium coplanar electrodes. Then they were heated up to a fixed temperature and held there for a certain time. σ -plots were obtained as the samples were cooled slowly from this temperature down to

Table 1. The number of electrons and charged dangling bonds calculated using the model of the DOS given in section 2.2.1. T_k is the kink temperature as determined by the σ -plot.

T (K)	$n_c + n_{BT}$ (10^{14} cm^{-3})	$N(D^+) + N(D^-)$ (10^{14} cm^{-3})
300	1.3×10^{-3}	9.0
360	2.7×10^{-2}	9.0
400	0.17	9.1
440	0.74	9.2
450 (T_k)	1.0	9.2
460	1.3	9.2
470	1.7	9.3
500	3.2	9.7
550	6.6	11

Table 2. The defect density N_0 has been calculated using $N_0 = C_0 \alpha_{\text{excess}}$ with C_0 adjusted to fit $T_k(\text{exp.})$ of curve 1. This gives $C_0 = 1.7 \times 10^{16} \text{ cm}^{-2} \text{ eV}^{-1}$, comparing well with the value given by Smith *et al* [13]: $1.9 \times 10^{16} \text{ cm}^{-2} \text{ eV}^{-1}$. N_0 is given in cm^{-3} and T_k in $^\circ\text{C}$.

Step No	1	2	3	4	5	6
N_0	5×10^{15}	6.9×10^{15}	8.8×10^{15}	1.9×10^{16}	3.3×10^{16}	4.3×10^{16}
$T_k(\text{calc.})$	≈ 180	189	196	220	238	247
$T_k(\text{exp.})$	180	191	195	203	210	221

room temperature and there the sub-band-gap absorption was measured by CPM [23]. The sub-band-gap absorption is known to be related to the total defect density by $N_0 = C_0 \int \alpha \, dE$ where C_0 is a constant and $\int \alpha \, dE$ the ‘excess’ absorption [24]. All thermal treatments and conductivity measurements were made in the dark, in vacuum (10^{-6} Torr).

Figure 3 shows an example of the σ -characteristics obtained after the different thermal treatments. We can see that, as expected, T_k increases significantly (180 $^\circ\text{C}$ to 220 $^\circ\text{C}$). But does the defect density increase? Figure 4 shows that it does indeed: in fact, an increase of the sub-band-gap absorption indicates the introduction of new defect states [13].

2.2.4. Discussion. First of all we stress the fact that neither the conductivity characteristics nor the CPM spectra were entirely changed by these treatments. Thus, as a first approximation and to remain within our simple model of the DOS, we will suppose that we always have a Gaussian dangling bond peak with the same W_0 but a higher N_0 .

Coming back to the results, we see that we have checked the prediction of our model: the increase of T_k is correlated with that of N_0 . Table 2 shows a quantitative comparison of the calculated and experimental values of T_k . The agreement is quite good except for for the highest N_0 -values.

There is, however, a more hidden but more important disagreement: the values of the kink conductivities σ_k . First we demonstrate that, according to this model, these kink conductivities measure the total charged defect density. In fact, at the kink, $n_{BT} + n_c$

Table 3. The ratio $r = n_{\text{BT}}/n_{\text{C}}$ at the kink for different T and DOS: DOS 1 is the DOS given in section 2.2.1; DOS 2 is the DOS given in section 2.2.1, except that $W_{\text{C}} = 0.040$ eV.

	Before thermal treatment ($T_{\text{k}} = 453$ K)	After thermal treatment ($T_{\text{k}} = 494$ K)
DOS 1	6.6	4.9
DOS 2		12.9

is supposed to be equal to a fixed fraction of the charged defects: let us say $n_{\text{BT}} + n_{\text{C}} = 0.1(N(\text{D}^+) + N(\text{D}^-))$ (table 1). As the ratio $n_{\text{BT}}/n_{\text{C}}$ is easy to calculate if one assumes a conduction band DOS, from the knowledge of $\sigma_{\text{k}} (\equiv e\mu n_{\text{C}}$ at the kink) one can infer a rough measure of the total charged defect density:

$$N(\text{D}^+) + N(\text{D}^-) \approx (10/e\mu)(1 + r)\sigma_{\text{k}} \quad (2)$$

where $r = n_{\text{BT}}/n_{\text{C}}$. The disagreement arises then from the value of $(\sigma_{\text{k}})_{\text{after}}$ measured after all the thermal treatments as compared to the value of $(\sigma_{\text{k}})_{\text{before}}$ measured before them. In fact, the total defect density has risen by a factor of 10 (table 2) and so has the charged one since we have supposed that the shape of the dangling bond peak has not changed and, according to our model, E_{F} always lies at $E_0 + U/2$. Then (2) implies that $(\sigma_{\text{k}})_{\text{after}}$ is roughly $10 \times (1 + r)_{\text{before}}/(1 + r)_{\text{after}} = 13$ times larger than $(\sigma_{\text{k}})_{\text{before}}$ (cf the first line of table 3). Comparing this to the actual increase $4.7 \times 10^{-5}/2.6 \times 10^{-5} = 1.8$ shows that there is some difficulty in using this simple model to explain the data quantitatively.

2.3. Another model of the DOS?

We have seen in section 2.1 that the statistical shift could be used to explain the kink for undoped a-Si:H qualitatively, but in the last section that it cannot be used to reproduce it quantitatively if we use the ‘dangling-bond-only’ DOS. Nevertheless, the problem of the quantitative disagreement could be resolved by slightly adjusting the model and allowing a widening of the conduction band while the temperature is held at 350 °C, which would increase $(1 + r)_{\text{after}}$ (table 3) and/or a *charged* defect density increase of less than a factor 10 due to a change of the defect peak shape for example. These two hypotheses do not seem too unlikely, but we do not have any experimental evidence to support them.

One might also be tempted to change the DOS by introducing, for example, charged impurities [25] to get better quantitative agreement, but the kink conductivity still sets a problem: we have observed, for most samples, kink conductivities of about $10^{-5} \Omega^{-1} \text{cm}^{-1}$. This would imply a *charged* defect density (equation (2)) below 10^{15}cm^{-3} which seems too small [25, 26]. However, the question of the charged defect density in undoped a-Si:H is still open and more experimental evidence is needed.

3. Metastability

The basic process of metastability is the following: for temperatures below the equilibrium temperature T_{E} the DOS is frozen (the time to reach equilibrium is long compared

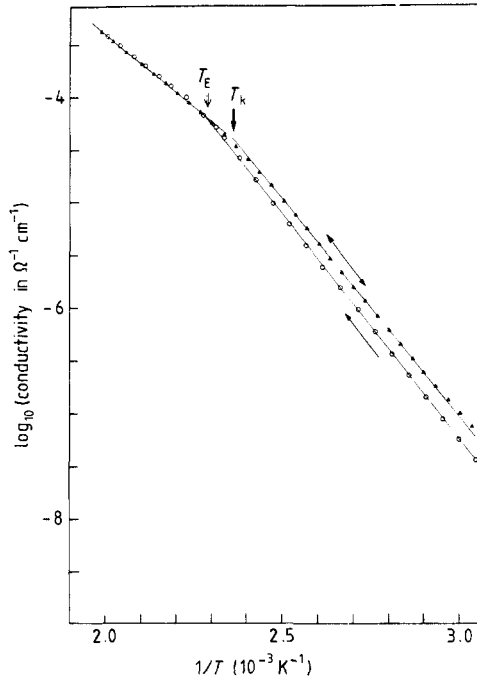


Figure 5. Kink and equilibrium temperature determination: \blacktriangle , slow cooling; \circ , slow heating after a quench.

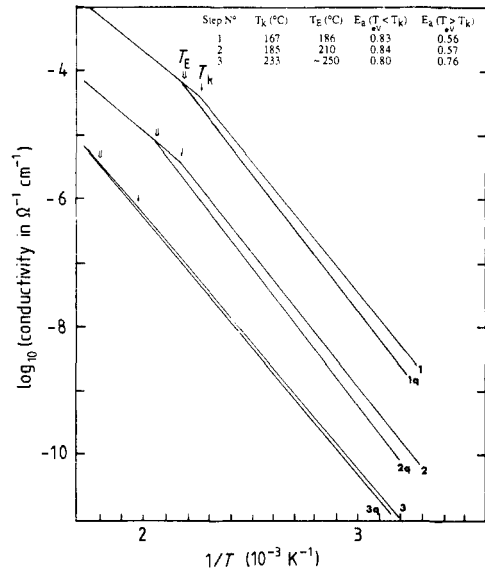


Figure 6. Temperature dependence of the DC conductivity after subsequent treatments (q: quenched state). 1, 1q: 250 °C, 30 min; 2, 2q: 350 °C, 9 min; 3, 3q: helium-ion bombardment. To facilitate comparison, each curve has been shifted from the preceding one by a factor of 10: for step i we have plotted $\sigma_i/10^{i-1}$.

with experimental time scales) but above T_E the DOS is allowed to equilibrate and thus changes with the temperature [11]. As the Fermi level is mainly determined by the DOS (and of course the temperature), it is easy to understand that at T_E the conductivity could show some anomaly—let us say some kink. That is exactly what is observed in n-doped a-Si:H¹⁰.

3.1. Experimental details

The conductivity of standard undoped a-Si:H samples [15] was studied in the coplanar configuration. The samples were first annealed at 200–250 °C (the substrate temperatures during deposition) for 30 min to eliminate the effects of any adsorbed gases. Figure 5 shows an example of determination of T_E and T_k . The sample was quenched from 250 °C at a rate of about 10 °C s⁻¹ (more details about the quenching and the quenched state are given elsewhere [15]). Then it was slowly warmed up to 250 °C (lower curve, heating rate 1 °C min⁻¹). Finally the sample was slowly cooled from 250 °C down to room temperature (upper curve, cooling rate 0.02 °C s⁻¹). It is worth noting that the slow-cool curve could also be obtained by slowly heating the sample after a slow cool. T_E and T_k are determined as follows (figure 5): the low-temperature lines are produced

Table 4. A comparison of the kink and equilibrium temperatures for a variety of undoped standard samples.

Sample	T_k (°C)	T_E (°C)
1	177	186
2	170	190
3	180	200
4	200	219
5	154	176
6	173	188

until they cross the high-temperature one (for T_k the high-temperature σ -line has also to be produced): the crossing points determine the equilibrium and kink temperatures.

3.2. The relationship between T_E and T_k

To discuss whether the metastability is responsible for the kink in undoped material, we should first check whether the kink and equilibrium temperatures are close. Table 4 shows that in a variety of standard samples this is indeed the case. To be more precise we see that, roughly, $T_E = T_k + 20$ °C. This can be easily understood in the context of the hydrogen glass model [11]. In fact, in this model, T_E and T_k should be interpreted as the glass transition temperatures (T_g) for the quenched and slow-cooled states respectively. In glasses it is well known that the higher the quench rate the higher T_g [27]. Then we see that the experimental data confirm that T_k can be interpreted as the *freezing temperature for a slowly cooled sample*.

3.3. Model test

A much more stringent test of the metastability explanation of the kink would be, on the same sample, to change T_k (as in section 2.2.3) and to see whether T_E changes or not. Clearly, if T_k increases when T_E remains constant our model will be conclusively contradicted.

3.3.1. Experimental details. T_E and T_k were determined as detailed in section 3.1. We used two subsequent methods to change T_k : the first was the heating to 350 °C as described in section 2.2.3 and the second was helium-ion bombardment carried out in our laboratory. A total dose of 10^{16} ions cm^{-2} was implanted at an energy $E = 1.5$ MeV, on samples held at room temperature. These implantation conditions were chosen to increase the defect density by an order of magnitude [21] uniformly throughout the sample.

3.3.2. Results. Figure 6 shows the σ -plots for one sample in the initial state, after 9 h at 350 °C and after helium-ion bombardment. It can be seen that both T_E and T_k increase in the same way suggesting that T_k does indeed correspond to the freezing temperature after a slow cool.

3.4. Discussion

In section 2.2 we have seen that the ‘statistical shift model’ can be used to explain why an increase of the defect density induces an increase of T_k . But how can we explain the

increase of the freezing temperature with the 'metastability model'? We shall see that this can be qualitatively understood in terms of the hydrogen glass model [11]. In fact, according to that model, T_g (to simplify we replace T_E and T_k by T_g , the glass transition temperature) is the temperature at which the hydrogen diffusion coefficient is equal to $10^{20} \text{ cm}^2 \text{ s}^{-1}$. Now to increase T_g we would have to slow down the hydrogen diffusion, and that is exactly what is expected for heating at 350°C for two reasons. First, it has recently been found [28, 29] that the diffusion of hydrogen is reduced by an increase of the defect density. The second reason has to do with dispersive hydrogen diffusion [30]. At 350°C , the hydrogen moves fast and then a significant fraction gets trapped in deep traps from which escape is difficult at lower temperatures. Then heating the sample to 350°C irreversibly reduces hydrogen diffusion. For the ion bombardment we would also expect an increase of T_g , because of the increase of the defect density.

As a first conclusion, we see that the metastability model can in principle explain the kink, and qualitatively agrees with the experimental results found in section 3.2.

4. General discussion

We shall discuss some features of the conductivity plots that indicate that the metastability is closely related to the kink.

It is interesting to note that there is a perfect correlation between the kink curvature and the change in conductivity after a quench. More precisely we see that for doped a-Si:H there is an increase in activation energy for $T > T_k$ (upward kink) and that the quenched conductivity is higher than that from slow cooling [11]. For undoped a-Si:H we observe a decrease in slope for $T > T_k$ (downward kink) and a smaller quenched conductivity. Now an upward kink means that E_F is closer to the appropriate mobility edge (E_C for undoped and n-doped a-Si:H, E_v for p-doped a-Si:H) than it would be without the kink. Everything happens as if, after a quench, the sample partially 'remembered' this position of E_F and had a higher conductivity. The same reasoning could be applied to the downward kink. This is easy to understand in terms of the glass model and seems an argument in favour of the explanation of the kink by this model: if the kink resulted only from a statistical shift, why would the sample 'remember' the position of E_F at higher temperature? Electronic relaxation is very rapid and would not give quenching effects [14].

The other argument concerns the kink magnitude. We see in figure 6 that before any treatment the kink is quite pronounced and so is the metastability. On the other hand the kink after heating to 350°C and irradiation is very weak and so is the metastability effect. This indicates that the magnitude of the kink, which could be quantified for example in terms of the difference in activation energies, determines the magnitude of the metastable effects which could be quantified in terms of the ratio $\sigma(\text{after a quench})/\sigma(\text{after a slow cool})$ at room temperature for example. More experimental evidence is needed to verify this argument.

To preclude a metastability explanation of the kink it would be sufficient to find an undoped sample with a kink and no quenching effects after quenching from temperatures above T_k , or with quenching effects and no kink. Another possibility is to change T_k without affecting T_E . Until now we have never observed such effects.

5. Conclusions

We have seen that the statistical shift of E_F could be used to explain the kink qualitatively but that we face several quantitative problems. In particular, the observed increase of the conductivity measured at the kink is smaller than expected from the increase in the defect density. On the other hand, the metastability model is more general because it explains the effect in doped and undoped material. Otherwise the perfect correlation of T_k and T_E observed in a variety of undoped samples and, furthermore, the perfect correlation of their changes in the same sample would be difficult to explain. Even if a contribution to the kink from the statistical shift of E_F cannot be totally excluded, the observed facts suggest metastability as a more natural and preferable explanation.

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