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## FIELD INDUCED ADSORPTION AT AN AMORPHOUS SILICON SURFACE STUDIED BY FIELD ION MICROSCOPY

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**Abstract** - Flickering and hopping of spots in the argon image of an a-Si:H layer on tungsten was investigated by studying the kinetics of the responsible adsorption and desorption processes as a function of tip temperature and imaging gas pressure. The localized binding energy of argon adsorbates on silicon is calculated using the isolated dipole model which yields a value of 0.20 eV. An increase of the gas pressure decreases the average residence times in both states which constitute a flickering or hopping phenomenon. Decreasing the tip temperature leads to opposite responses of the two residence times. These results do not support a model in which desorption is thermally activated. Both the adsorption and desorption rate are predominantly governed by the imaging gas pressure. A desorption process activated by image gas impact is suggested.

### 1. INTRODUCTION

The present FIM investigation was initiated in order to study the local atomic configuration in hydrogenated amorphous silicon (a-Si:H), a semiconducting material which is currently used in electronic devices.

It was found that it is possible to image an a-Si:H layer deposited onto tungsten by imaging with argon in the FIM. The images, however, showed insufficient detail to establish local atomic arrangements. The attention in this paper is focused on a marked feature, which was repeatedly and reproducibly observed in the FIM images of a-Si:H layers, the flickering and hopping of image spots.

These phenomena were reported by several authors [1-4] and are generally considered to originate from field adsorption and desorption events at the imaged site [5-7]. Adsorbed atoms greatly influence the ionization rate in the ionization zone just above their position [2,4,9,12-14].

The stability of the images enabled the investigation of these sites in terms of average residence times in the two alternative states as a function of tip temperature and imaging gas pressure. The present method requires residence times in the order of 1 second.

### 2. FIELD INDUCED ADSORPTION

It is well established that field ion emitter surfaces, prepared by low temperature field evaporation and kept at a constant high field in a FIM, are partly covered with field adsorbed imaging gas atoms [5-7]. The binding energy can be split in a long range and a short range contribution [8,10,11]. The long range contribution is equal to the field induced binding energy  $\frac{1}{2} \alpha_G F_0^2$ .  $\alpha_G$  is the electric polarizability of the gas atom,  $F_0$  is the nearly uniform field somewhat above the tip surface. The localized part is due to the attractive forces between the field induced dipoles of the adsorbate and the nearest surface atom. There are also secondary contributions due to surface charges and other polarized surface atoms, but these are considered to be of negligible importance.

For the adsorbed atom to desorb, only the localized part of the binding energy is of interest. It is the work needed to move an adsorbed atom from the adsorption site to a distant point where the dipole interaction is negligible but the applied field is still essentially  $F_0$ . The isolated dipole model [7] is applied in order to calculate the local electric fields at the adsorbate Ar and nearest surface atom (Si), respectively. The applied field for both the adsorbate and the surface atom can be taken the same, for the field penetration depth in silicon is much larger than the interatomic distance [7]. To find the localized binding energy, the difference in dipole energy between the adsorbed state and desorbed state should be taken into account for both the gas and the surface atom, i.e.:

$$Q_{\text{des}} = \frac{1}{2} \{ \alpha_s (f_s - 1) + \alpha_g (f_g - 1) \} F_0^2. \quad [1]$$

Substituting the appropriate values for silicon and argon ( $d(\text{Ar}, \text{Si}) = 0.308 \text{ nm}$ ,  $\alpha_{\text{Ar}} = 1.8 \cdot 10^{-40} \text{ F m}^2$ ,  $\alpha_{\text{Si}} = 2.4 \cdot 10^{-40} \text{ F m}^2$ ,  $F_0 = 22 \text{ V/nm}$ ) yields a localized binding energy of 0.20 eV. When this value is substituted in the Arrhenius rate equation, assuming an attempt rate of  $10^{12} \text{ s}^{-1}$  and a tip temperature of 80 Kelvin, the average adsorption residence time is in the correct order of magnitude to be measured. However, one should realize that the value is greatly influenced by the exact values of the binding energy and tip temperature. It merely indicates that the observed phenomena occur on the same time scale as the calculated value.

### 3. EXPERIMENTAL PROCEDURE

A thin layer of a-Si:H was deposited by plasma enhanced chemical vapour deposition onto tungsten tips prepared by the usual electrochemical method, followed by field evaporation in the FIM (Fig. 1). The background pressure was  $0.2 \mu\text{Pa}$ . The imaging gas pressure was varied between 0.1 and 1 mPa and tip temperatures between 65 and 100 Kelvin were used. The tip surface was cleared from unwanted adsorbates by field desorption at, or somewhat above, the best image field of argon. The desorption was carried out until a stable image was formed containing no apparent 'molecular' image spots, as are often observed in the case of contaminated tip surfaces.

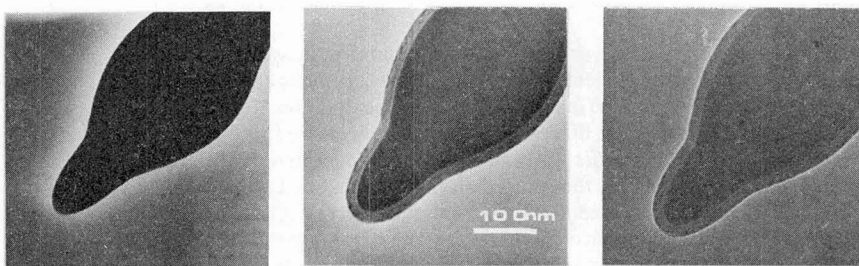


Fig. 1 (a) W-tip prepared by chemical polishing followed by field evaporation;  
(b) the same tip after deposition of an a-Si:H layer, before imaging in the FIM;  
and  
(c) after imaging in the FIM.

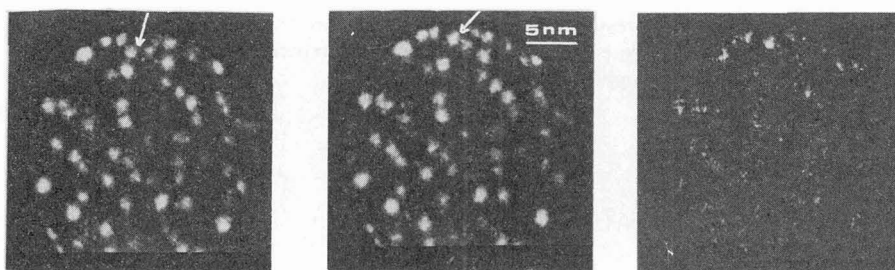


Fig. 2 (a,b) FIM images of a-Si:H, argon, 80 K, 4.4 kV;  
(c) digitized image generated by subtracting a from b.

The FIM image is shown in Fig. 2. The white arrows indicate the hopping spot. The last image is generated digitally by subtracting the first two images. The neighbouring spots seem to be repelled by the hopping spot.

Field evaporation of the a-Si:H layer could not be performed, as the adhesion of the layer to the tungsten tip was not sufficient to withstand the exerted stress, and the layer was stripped off as a whole prior to the onset of field evaporation. Fortunately, the deposited layers were extremely smooth as required for FIM imaging.

FIM relies on high fields in order to protect the surface from contamination by residual gases with low ionization thresholds. Argon as imaging gas has the disadvantage of having a low best image field which allows some contamination of the surface. The measurements to be described were carried out on a time scale where hardly any contamination occurred at the investigated location, as could be concluded from the stability of the image.

In order to measure the average residence time as a function of tip temperature, these measurements were carried out during cooling down the tip assembly. The actual time dependence of the tip temperature was determined beforehand by means of a platinum resistor mounted in the tip holder. The FIM images were recorded on video tape by means of a two-stage image intensifier and a video camera and could later be analyzed by placing a light detector setup in front of the monitor screen at the location of interest.

#### 4. RESULTS

Up to several hundreds of flickering or hopping events were observed per site. After correction of the trend in the average residence time, in case of the temperature decrease of the tip, the residence time distribution fitted well to an exponential probability density function, as is shown in Fig. 3. Residence times shorter than 20 msec could not be detected as a consequence of the utilized method, resulting in a deviation from the theoretical curve at short times.

Figure 4 depicts the average residence times in the two states A and B, as a function of time, during slowly cooling down the tip. Each point in the graph is the average of 20 subsequent residence times and the scatter from the straight line equals the theoretical value. The trends are of opposite signs. The absolute temperature changes with 1% during the experiment. Pressure variation could readily be performed by adjusting the inleak valve.

Fig. 5 shows the marked influence of a temporary pressure decrease on both two states. The number of residence times per point is 25. It should be noted that the experiments of temperature and pressure variation were performed on different hopping sites.

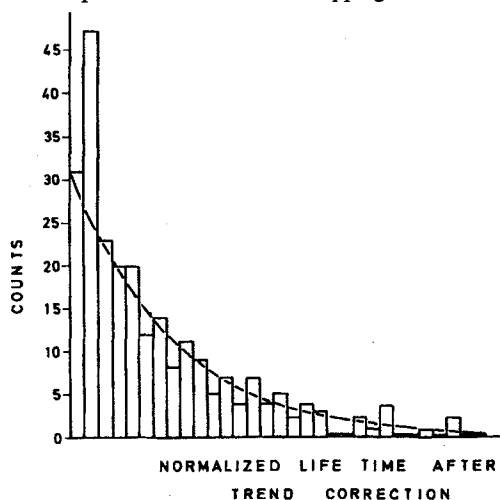


Fig. 3 Residence time distribution corrected for trend.

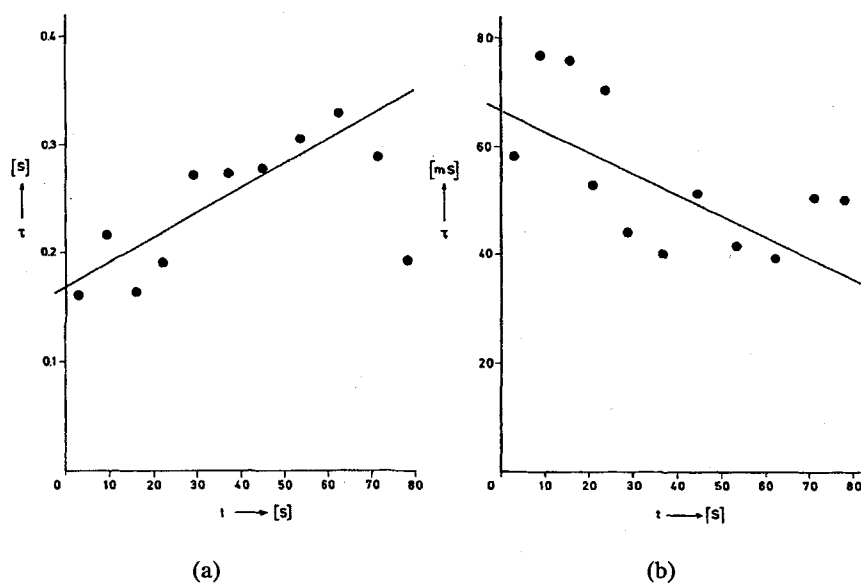


Fig. 4 Residence times of two states (A: Fig. 4a; B: Fig. 4b) as a function of temperature  $T = 96.5 - 0.016 t$  ( $T$  in K,  $t$  in s).

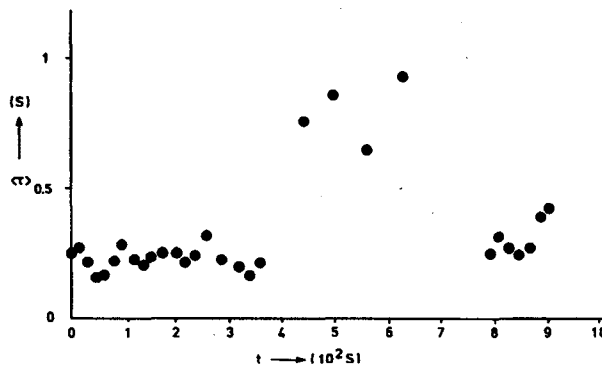


Fig. 5a Residence times of state A as a function of pressure.  $p = 11$  n Pa at  $400 \text{ s} < t < 700 \text{ s}$ ,  $p = 38$  n Pa elsewhere.

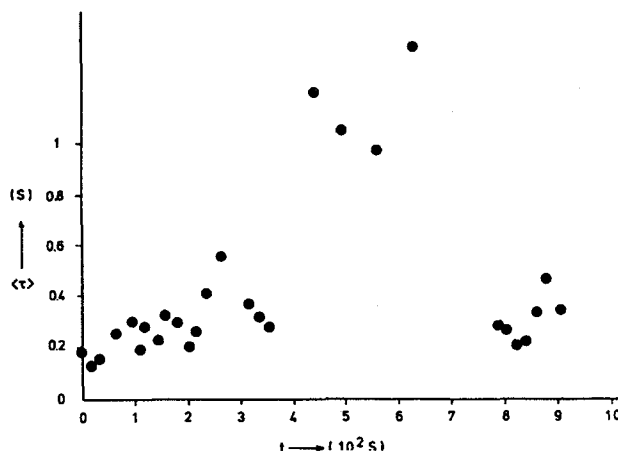


Fig. 5b Residence times of state B as a function of pressure  $p = 11$  n Pa at  $400 \text{ s} < t < 700 \text{ s}$ ,  $p = 38$  n Pa elsewhere.

## 5. DISCUSSION

The residence times in state A as well as in state B are gas pressure dependent. The processes responsible for the transitions from state A to B and B to A, respectively, must have this pressure dependence. Image gas adsorption and desorption activated by impact of polarized argon gas atoms can explain this dependence as well as the observed cyclic behaviour. The energy of an incoming argon dipole (0.2 eV) is sufficient to break the dipole-dipole bond.

At about 90 K, the thermally activated desorption rate is much lower than the rate resulting from dipole impact as is seen from the pressure dependence. The binding energy of the adsorbate is therefore larger than 0.20 eV. Kreuzer [15] has questioned the validity of equation [1] and proposed field induced chemisorption to occur, possibly resulting in higher binding energies. The apparent opposite response to a small temperature variation cannot be accounted for by our model.

Further experiments with laser irradiation in order to vary the tip temperature more rapidly than we could perform with the existing tip assembly might give more conclusive arguments to discover the nature of the phenomenon.

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