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HAL Id: jpa-00226806
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Submitted on 1 Jan 1987

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THERMAL ROUGHENING OF TUNGSTEN SURFACES STUDIED BY FIELD EMISSION

Y.M. Gong* and R. Gomer
The James Franck Institute, The University of Chicago, Chicago, IL 60637, U.S.A.

Abstract.- Surface self-diffusion measurements of tungsten atoms and other defects were carried out by the field emission fluctuation method at various positions along the zones (011)-(112) and (011)-(001). In the first zone, consisting of (011) terraces and closepacked steps 2-dimensional diffusion attributed to atoms on terraces and 1-dimensional diffusion attributed to kinks diffusing along steps was found. At 875 K kink diffusion decreases sharply, then increases again and become 2-dimensional at 950 K, suggesting disorder and eventual disappearance of steps. This is accompanied by a decrease in atom diffusion and eventual merging of both regimes above 1000 K. On the zone (011)-(001) steps are not closepacked and only 1-dimensional was seen, although two regimes were also found. The lack of 2-dimensional diffusion is attributed to the inability of atoms to climb none closepacked steps. Evidence for roughening was also found in most locations of this second zone but at much lower temperatures than along (011)-(112).

I- INTRODUCTION

Roughening transitions on stepped metal surfaces are of theoretical interest but have received relatively little experimental attention. Engel and coworkers have recently look at stepped Ni surfaces and found evidence of roughening from He diffraction studies/1/. We show here that changes in the surface self-diffusion of W atoms and other defects can give an indication of roughening. The regions chosen for investigation lie in the zone (011)-(112) and the zone (011)-(001). Fig. 1 gives the probed planes in two zones. A very preliminary report of this work has been given/2/ and a detailed account is to be published elsewhere/3/.

II- PRINCIPLE OF METHOD

The method used here is an extension of the field emission fluctuation technique, described in detail previously/4,5/. As normally used a field emitter is uniformly covered with adsorbate and current from a small region, 50-100 Angstroms in radius is measured by means of a probehole. At temperatures where the adsorbate is mobile there are fluctuations in the number of adsorbed particles in the probe and this gives rise to fluctuations in emission current. The time autocorrelation function of these fluctuations is nearly proportional to that of the particle number, \( f_n(t) \)

\[
f_n(t) = \frac{<n^2>}{A} \int d^2 x \int d^2 x' \frac{e^{-|x-x'|^2/4Dt}}{(4\pi Dt)^{3/2}}
\]

(1)

*Permanent address: Department of Radio-Electronics, Peking University, Beijing, The People’s Republic of China.
where \( \langle (\delta n)^2 \rangle \) is the mean square number fluctuation in the probed area \( A \), \( D \) the diffusion coefficient, \( t \) the time and \( \gamma \) the dimensionality of diffusion. It can be seen from Eq.(1) that \( f_n(t) \propto t^{-1} \) for 2-dimensional and \( \propto t^{-1/2} \) for 1-dimensional diffusion at long times. It can be shown that \( f_n(t) \) can be expressed as a function \( t/t_0 \) where

\[
\tau_0 = r_0^2/4D
\]  

(2)

where \( r_0 \) is the radius of the probed area \( A \). So that comparison of the experimental and theoretical curves yields \( \tau_0 \) and hence \( D \).

In the present case the diffusing entities are not absorbed species at fixed average concentration but "loose" W atoms and other defects, created thermally. It can be shown that this need not affect the form of the correlation function if the lifetime of the diffusing species is comparable to \( \tau_0 \).

III - EXPERIMENTAL PROCEDURE

The detail of the experimental procedure have been given previously. The final pressure in the system was \( 3-4 \times 10^{-11} \) torr. The experiments consisted of finding the field emission current correlation function \( f_1(t) \) for a number of probe-hole locations along the zone (011)-(112) and along the zone (011)-(001), using the electrostatic deflection electrodes to move the emission pattern. The emitter radius was \( r_e = 1880 \) Angstroms and the probe radius was \( r_0 \sim 60 \) Angstroms. Since Fowler-Nordheim parameters for W atoms and other defects are not known the experimental correlation function is then compared to a theoretical one based on Eq.(1) couched in terms of \( t/t_0 \). The activation energy \( E \) for surface self-diffusion can be got from the plots of \( \log D \) vs. \( 1/T \). This is based on the assumed form.
Fig. (2) - Work function $\phi$ vs. $T$ for (257), (123) and (235).

Some work function measurements were also made. Fig. (2) gives work function vs. temperature.

IV - RESULTS AND DISCUSSION

The regions examined were stepped surfaces along the zone (011)-(112) and along the zone (011)-(001). In the first case terraces and steps have $<110>$ orientation, steps are closepacked and run along $<111>$ directions. In the second zone terraces are initially $<110>$ and steps $<100>$, with terraces becoming $<100>$ and steps $<110>$ as (001) is approached. The steps are not closepacked and run along $<100>$ directions.

For the zone (011)-(112) results were very similar on all stepped surfaces and showed two distinct types of diffusion. This could be seen by using different time scales for the correlation. The more rapidly decaying function showed $t^{-1}$ behavior at long times, the slower $t^{-1/2}$ behavior at long times. This suggests that the fast regime corresponds to atoms diffusing on and across terraces; this requires atoms to climb and descend steps. The slower 1-dimensional process suggests diffusion along steps, and probably corresponds to diffusion of negative kinks, or positive-negative kink combinations. Fig. (3) shows log $D$ vs. $1/T$ for both types for a typical case. Evidence for a roughening transition comes from the following observations: At 875 K, $D_{\text{slow}}$ decreases sharply and then increases at 925 K. Up to this temperature slow diffusion is still 1-dimensional but at 950 K becomes 2-dimensional. This suggests that steps contain some kinks which can diffuse, but that at $T > 875$ K steps become sufficiently disordered to impede kink diffusion and at 950 K become so disordered that kinks run in every direction and diffusion is 2-dimensional. The fast, single atom diffusion increases up to

\[ D = D_0 \exp(-E/kT) \]  \hspace{1cm} (3)
Fig. (3) - log $D$ vs. $1/T$ for slow and fast diffusion regimes on (257). Values of activation energies $E$ and prefactors $D_0$ are indicated on the figure. The initially faster diffusion is 2-dimensional, the initially slow regime 1-dimensional below 950 K.

900 K and then decreases, because the increased step disorder and decreasing terrace widths impede this process. Above 1000 K only a single diffusion regime is discernible. Thus we conclude that on surfaces containing closepacked steps there is a gradual but well defined increase in disorder leading eventually to enough roughening for steps to lose their identity.

On the zone (011)-(001) where steps are not closepacked and terraces switch orientation the situation seems more complicated. Two diffusion regimes are seen on (023) and (017), which consist of (011) and (001) terraces respectively, but both regimes are 1-dimensional. On (023) (Fig.(4)) the fast diffusion decreases sharply at 650 K and merges with the slow one at 714 K; above this temperature $D$ increases again but become 2-dimensional only at 900 K. However, no sharp decrease in the slow regime, presumably corresponding to kink diffusion is seen. On (017) the fast diffusion drops at 650 K and merges with the slow diffusion at 770 K; the latter increases with only a change in slope at 700 K (Fig.(5)). Diffusion becomes 2-dimensional at 900 K. On (013) no real evidence for a transition is seen, while (012) is similar to (013). Thus roughening seems less well defined, or at least is less clearly seen from diffusion changes, when steps are not closepacked. It seems clear, however, that some roughening occurs but at much lower temperatures than along the zone (011)-(112).

Experiments on (011), (112) and (111) were also made. On (011) no significant signal was seen and it shows that the population of free atoms there is negligible. On (112) a single diffusion regime was seen and the observed diffusion is essentially 2-dimensional. On (111) a single 2-dimensional regime was seen but at $T = 750$ K $D$ decreased, then increased again, indicating a surface roughening.
Both regimes of $D$ vs. $1/T$ for $(023)$, (x) fast, (o) slow are 1-dimensional below 900 K.

Fig. (4) - log $D$ vs. $1/T$ for $(023)$. (x) fast, (o) slow regime. Both regimes are 1-dimensional below 900 K.

Both regimes are 1-dimensional below 900 K.

Fig. (5) - log $D$ vs. $1/T$ for $(017)$. (x) fast, (o) slow regime. Both regimes are 1-dimensional below 900 K.
It is necessary to point that in the experimental procedures the presence of the electric field does not cause the thermal-field build up of W atoms in the range of the temperatures.

V - CONCLUSION

The experimental results have indicated that self-diffusion on stepped surface can be used to investigate roughening by the field emission fluctuation method. It is rather clear that roughening on well defined stepped surfaces proceed via partial disordering of steps, followed by more global disorder.

VI - ACKNOWLEDGMENT

This work was supported by NSF Grant 86-00320. We have also benefited from the MRL of the National Science Foundation at the University of Chicago.

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