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Surface Science 377–379 (1997) 62–65

surface science

Step roughness on Ag(111) investigated by STM: a systematic study of tip influence

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Received 1 August 1996; accepted for publication 15 October 1996

Abstract

We have investigated monatomic steps on Ag(111) by STM at different temperatures. At room temperature, the rough appearance of these steps is usually attributed to thermal step fluctuations. We have investigated the influence of the tip systematically. Applying a new test, we demonstrate that even subtle influences can lead to wrong results in statistical analysis.

Keywords: Low index single crystal surfaces; Scanning tunneling microscopy; Silver; Surface diffusion

1. Introduction

In recent years, scanning tunneling microscopy (STM) has been used to investigate various dynamic processes at surfaces. In particular, the rough appearance of monatomic steps has been widely investigated on various metal surfaces in order to determine the microscopic mechanisms underlying mass transport [1–7]. For a given system, the dominating mechanism is expected to depend on temperature due to different activation energies [8]. However, the determination of thermodynamic properties from STM measurements requires that the influence of the tip can be neglected. This is not obvious considering the extreme conditions prevailing in the tunneling gap compared to the subtle balance between the activation energies and temperature. This situation has recently been illustrated by Li et al. [9]. While these authors report on significant tip-induced disturbances of the surface, in this paper we will concentrate on more subtle influences which are not obvious at first sight.

2. Experimental

The investigations described in this work have been performed in UHV, using a home-built variable-temperature STM [10]. We used electrochemically etched tungsten tips, prepared in situ by field emission. The samples were (111)-oriented Ag films prepared by thermal evaporation onto mica substrates kept at an elevated temperature during evaporation. These films typically display atomically flat terraces of several thousand ångströms [11] with occasional monatomic steps. The level of the impurities was below the detection limit of Auger electron spectroscopy.

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PII S0039-6028(96)01352-0

3. Results and discussion

Fig. 1 shows a series of STM images of monatomic steps imaged at different temperatures. Fig. 1a was recorded at 100 K. The step edge is absolutely straight, and no sign of thermally activated kinks or any atomic motion along the step is observable. At this temperature, steps can usually be imaged without tip disturbance for a wide range of tunneling parameters, even down to a tunneling resistance R_T of 1 MΩ, where the atomic lattice is routinely resolved (see Fig. 1a). Thus we can be sure that the observed structure is due to the intrinsic properties of the sample. Fig. 1b was recorded at 250 K. The step edge is essentially straight with a few individual kinks. The length of these kinks is determined to be 2.5 Å, as expected for single kinks on steps oriented along the close-packed[1$\bar{1}$0] direction on Ag(111). The position of these kinks changes from image to image, and it seems natural to attribute this behavior to the presence and motion of thermally activated kinks, i.e. again to the intrinsic properties of the sample. Finally, Fig. 1c was recorded at room temperature. The steps appear much rougher than in the previous images taken at lower temperatures. The details of the step roughness change from image to image, but the overall position and macroscopic shape of the step remain unchanged between many consecutive scans. We performed these measurements at different tunneling parameters: tunneling current and voltage were varied from 30 pA to 5 nA and from 10 mV to1.5 V (both polarities), respectively. In some rare cases we observe visible modifications of the surface even at moderate tunneling parameters ($R_T \approx 1$ GΩ) [12], similar to the findings of Li et al. [9]. However, we explicitly exclude these cases from our further analysis.

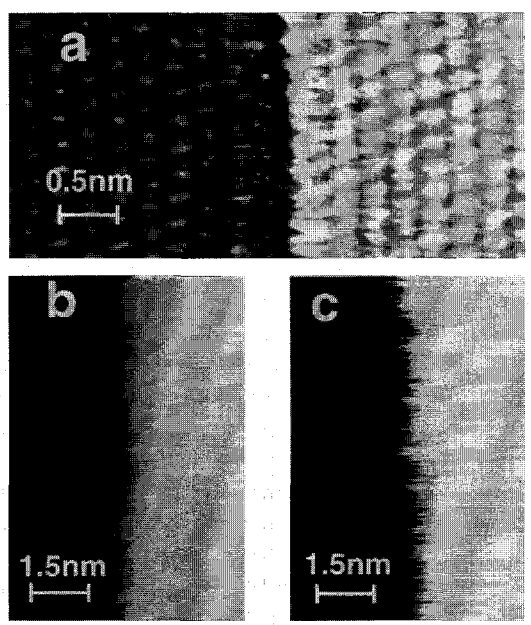

Fig. 1. Monatomic steps at different temperatures: (a) 100 K, (b) 250 K, (c) 295 K.

Let us adopt the usual point of view that the apparent step roughness is due to thermodynamic equilibrium fluctuations when the surface is not visibly modified by the STM. Stronger tests of this assumption will be described later in this work. In the following, we concentrate on isolated steps oriented parallel to the close-packed[1$\bar{1}$0] direction. Usual STM images contain a complicated mixture of time and space information and therefore, in order to investigate the apparent step roughness in more detail, we proceeded by repeatedly scanning the same line perpendicular to the step [2]. The essential physics of step fluctuations can be described by a model which was worked out by Bartelt et al. [8] on the basis of a Langevin formalism. The most important finding of this model is that one can determine the dominant microscopic mechanism underlying mass transport at the step edge from the step position $x(t)$ measured as a function of time. In principle, there are two different mechanisms: (i) atoms can either move via the diffusion of thermally activated kinks along the steps, or (ii) they can desorb from the step edge into the two-dimensional (2D) adatom gas on the adjacent terrace. In the latter case, an equilibrium will establish between atoms desorbing from the step and readsorbing from the 2D gas. In this situation, one can distinguish two different regimes, where either desorption/adsorption from the step (ii) or diffusion of the adatoms on the terrace (iii) is the rate-limiting process. The model predicts a power-law divergence of the step-position autocorrelation function

$$G_x(\Delta t) = \langle (x(t+\Delta t) - x(t))^2 \rangle \propto \Delta t^b,$$

with an exponent b of 1/4, 1/2 or 1/3 in case (i), (ii) and (iii), respectively. Fig. 2 presents the result of this analysis for our measurements. The correlation function G_x shows a power-law divergence with an exponent of $b = 0.32 \pm 0.03$. $G_x(\Delta t)$ is calculated from a total of 80 000 linescans recorded on different steps at different tunneling parameters within the range mentioned above. No systematic variation of the exponent could be observed. The observed value of b is consistent with scenario (iii) described above, where the step fluctuations are dominated by the exchange of atoms with the 2D gas of adatoms, with terrace diffusion being the rate-limiting process. In view of the statistical (!) error, our result is incompatible with the two other mechanisms. Our result is in agreement with a recent study by Morgenstern et al. [13], who investigated the decay of 2D Ag adatom islands on Ag(111).

The above analysis leads to a relatively consistent picture. However, we still felt uncomfortable regarding the general assumption underlying this analysis, i.e. that the statistical properties of thermal equilibrium fluctuations remain unaffected by the STM tip. Considering the dramatic surface modifications which can occur accidentally even at a tunneling resistance of the order of 1 GΩ, we strongly believe that this assumption has to be

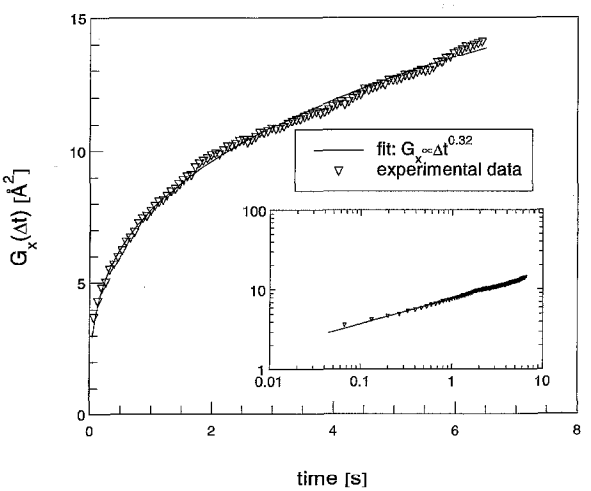

Fig. 2. Step-position autocorrelation function versus time. The solid line is a power-law fit $G_x(\Delta t) \propto t^{0.32}$. The inset shows the same data in a log–log plot.

tested more thoroughly. Therefore, we have performed two more standard tests and a third test, which to our knowledge has not so far been described in the literature. First of all, we simply reduced the scanning frequency by a factor of ten. No significant influence on the correlation function could be observed in our measurements. However, this is not a very rigorous test, because it only probes the influence of the number of events in which the tip crosses the step. But, one could easily imagine that the influence of the tip depends on the total time it spends in the vicinity of the step, which remains unaffected by this test. Another test, which is usually performed, is to analyze forward and backward scans of the STM separately, i.e. step-up and step-down scans. Again, our data do not show any significant dependence on the scanning direction. However, in the usual operation mode of STM, both forward and backward scans are performed alternately. Therefore, a possible asymmetry of the tip influence is not necessarily observable. In order to circumvent this problem, we have modified the scanning mode of our STM: first, we scan across a step in one direction. Then we block the STM-feedback loop and withdraw the tip 6 Å from the surface by applying an additional voltage to the z-electrode of the piezo. Afterwards we move the tip back to its starting position, where we switch on the feedback again and restart the same cycle [14]. In this way we maximize a possible asymmetric tip influence. The intriguing result is shown in Fig. 3. The correlation function is plotted as a function of time for data recorded in the unidirectional mode, for downward-scanning and for upward scanning, separately. The data for downward scanning agree well with those recorded in the standard scanning mode. In contrast, scanning upwards leads to a significantly weaker time dependence of the autocorrelation function. This discrepancy is observed frequently, although it is not always as drastic as in Fig. 3. It is important to point out that this holds explicitly for data which passed the standard tests described above. Fig. 3 clearly demonstrates the influence of the tip. More details regarding this dependence on the tunneling parameters will be reported in a forthcoming publication [15]. Therefore, the step roughness on Ag(111) cannot

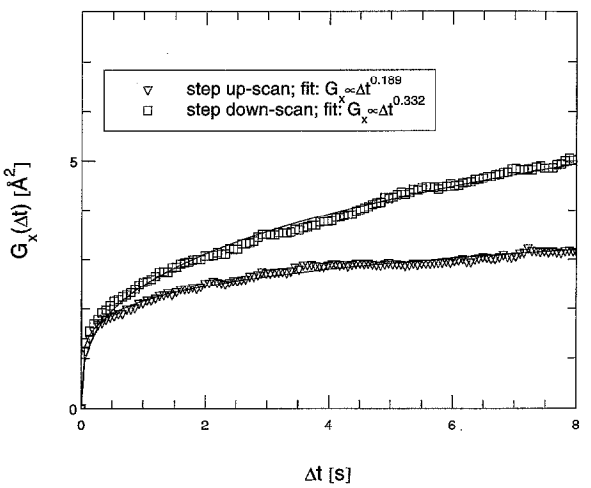

Fig. 3. Step-position autocorrelation function versus time for separate upward and downward scanning in the unidirectional scanning mode. $I_t = 1$ nA, $U_{tip} = 0.9$ V.

simply be interpreted in terms of equilibrium fluctuations.

4. Summary

We have investigated the rough appearance of monatomic steps on Ag(111) by STM. A statistical analysis of our data yields a power-law divergence of the step correlation function with time, which is usually interpreted in terms of equilibrium step fluctuations. However, more thorough tests than those usually reported in the literature show that the details of this divergence are influenced by the presence of the STM tip. Therefore, we believe that the extraction of thermodynamic parameters from STM investigations of step roughness is not reliable unless the perturbation due to the tip is studied in more detail.

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