The influence of tip-sample interaction on step fluctuations on Ag (111)

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Abstract

The fluctuations of the position of monatomic steps on Ag (111) are investigated by Scanning Tunneling Microscopy (STM). We analyze the influence of tip-sample interaction by varying the gap impedance over more than two orders of magnitude. For tunneling tips providing a weak tip-sample interaction, we show that the step position autocorrelation function remains essentially unaltered. In this unperturbed case, the kinetics of step fluctuations are found to be dominated by one-dimensional mass transport. For larger variations of tip-sample distance or for less favorable tip configurations we observe a tip-induced increase of the step fluctuations. Our measurements suggest that this effect is rather caused by short-range forces than by the electric field in the tunneling gap.

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

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1. Introduction

In recent years, the investigation of step fluctuations has attracted a great deal of attention [1-5]. There are two major interesting aspects of these experiments: First of all, miniaturization and the fabrication of nanoscopic structures requires a deeper understanding of atomic-scale diffusion processes which govern growth and equilibration phenomena at crystalline surfaces [6]. Using appropriate models [7], some of the relevant parameters can be accessed from the investigation of step fluctuations. Second, investigations of step fluctuations [1] were among the first dynamic measurements where diffusion processes are investigated online by scanning tunneling microscopy (STM). In this kind of experiments the samples are scanned continuously by the tunneling tip while the diffusing atoms perform a thermally activated hopping motion. In the past few years this method was applied to an increasing variety of systems [8, 9]. However, in some of these experiments, significant tip-induced perturbations were reported [9] (in the context of step fluctuations, see [3-5, 10]): It is well known that the closeness of tip and sample leads to considerable forces during STM operation [11], as exploited in many beautiful manipulation experiments at the atomic scale [12]. The same forces are suspected to significantly disturb the diffusion processes taking place while scanning, although the diffusion measurements are performed at larger gap impedance. In the case of step fluctuations, despite the large amount of work, this objection has not been debilitated convincingly, so far.

In a previous work [5] we already revealed a tip-induced disturbance in some of the experiments using a new scanning mode^{*}. Unfortunately, the

influence of this procedure and other similar tests involving scanning frequency and direction on the diffusing atoms is not yet understood [3-5, 13]. Therefore, we returned to the most direct test: In this work, we present

investigations of the fluctuations of an individual step at various tunneling currents and voltages and thereupon at various tip-sample distances. Our central findings are first that - presupposing a suitable tip - the observed step fluctuations remain essentially unaffected by a variation of the tunneling resistance over as much as two orders of magnitude corresponding to a variation of the tip-sample distance of 2.1 Å. Second, larger variations of the tip-sample distance or less favorable tip conditions do lead to significant tip-induced effects at small tip-sample separation.

2. Experimental

For our investigations, we used Ag (111) because the high mobility on this surface allows for convenient measurements at room temperature. The samples were oriented thin films on mica substrates grown by thermal evaporation, as described in the literature [14]. The substrates were cleaved in air, baked at 300°C overnight in UHV, and kept at the same temperature during evaporation. The films display atomically flat terraces of more than hundred nanometers with occasional monatomic steps. No impurities could be detected, neither by Auger electron spectroscopy nor directly by the STM^{*}. The measurements were performed using a homebuilt variable-temperature^{**} UHV-STM [15,

Ångstroms for the backward scan in order to produce an asymmetry of the scanning direction.

^{*} In contrast to the preparation of single crystals, no sputtering or high temperature annealing is required for the film preparation. Thus, many typical contamination problems are avoided.

^{**} All the experiments for this paper were carried out at room temperature.

^{*} In this "unidirectional scanning mode" the feedback loop is blocked and the tip is withdrawn by several

16]. We used electrochemically etched tungsten tips which were cleaned and sharpened in situ by field emission with a Ta counter electrode until an apparent work function of several eV was measured. Mostly, this procedure is followed by a final treatment with voltage pulses on the sample for further enhancement of the lateral resolution.

To avoid additional complications due to step-step interaction, isolated steps oriented along $[1\bar{1}0]$ direction were chosen. We cannot determine whether the microfacets are of (111)- or (100)-type. The next defects visible by STM were at least 400 Å away from the investigated site. To investigate the step fluctuations, we applied the line scan- or time image-technique used in many laboratories [1, 2]. Time images were recorded by scanning the same line perpendicular to the step edge (i.e. $[11\bar{2}]$ direction) 512 times in succession within 34 s with a density of roughly 7 pixels per atom. The drift of the STM amounted to 0.5 Å/min in lateral and 0.03 Å/min in vertical direction. For each set of parameters, 10 to 20 time images were recorded. All data were recorded in the constant current mode. The STM images shown throughout this paper are presented as unfiltered experimental raw data in a linear gray scale representation. Fig. 1 shows a typical time image. The discrete step positions are clearly resolved, as well as short excursions of the step position for just one scan line. The step position in units of pixels is determined from the experimental raw data using a simple threshold detection algorithm and subsequently digitized into units of atomic rows, as shown by the solid curve $x(t)$ in Fig. 1. From that curve, the step position autocorrelation function is calculated according to

$$G_x(\Delta t) = \left\langle \left(x(t + \Delta t) - x(t) \right)^2 \right\rangle \quad (1)$$

Here, the brackets denote an averaging over all possible time origins. Finally, the autocorrelation functions from the time images recorded under identical conditions are averaged.

3. Theoretical Background

At finite temperature, monatomic steps on crystalline surfaces are not static. Due to thermal fluctuations, their position perpendicular to their mean orientation becomes a function of time t : $x = x(t)$. The step stiffness provides the restoring force which tends to straighten the step. The dynamics are usually described in the Langevin formalism [7]. On the atomic scale, there are three competing diffusion scenarios: Atoms can detach from the step edge, diffuse on the adjacent terrace, and reattach at some other position. For fast terrace diffusion (evaporation-condensation scenario EC) the locations of detachment and reattachment are

uncorrelated whereas for slow terrace diffusion (terrace diffusion scenario TD) there is a correlation

Fig. 1. Time-image of a fluctuating step ($U_{tip} = 0.95$ V; $I = 1$ nA; size: 90 Å x 34 s). X gives the spatial coordinate along $[11\bar{2}]$ direction, t denotes time. The solid line shows the step position as determined from the experimental raw data.

due to a locally enhanced adatom density on the terrace in front of the desorption site. In the third case (periphery diffusion scenario PD), the individual atoms can only diffuse in one dimension along the step edge. For each of the three scenarios, an algebraic time-dependence of the autocorrelation function is expected:

$$G_x(\Delta t) = a \cdot \Delta t^b, \quad (2)$$

with an exponent b of $1/2$, $1/3$, or $1/4$ for (EC), (TD), and (PD) [17], respectively. The prefactor a depends on the dominant diffusion mechanism and contains information on the activation energies and diffusion barriers for the underlying atomic-scale processes.

4. Results

To obtain reliable information on the activation energies and diffusion barriers, the first task in the investigation of step fluctuations is, to see whether the value of b can be measured accurately without disturbance due to the tunneling tip. Otherwise, a possibly wrong identification of

the dominant diffusion mechanism renders any subsequent analysis worthless. Fig. 2a shows the step position autocorrelation function versus time recorded successively for three different values of gap impedance. The bias voltage was kept constant at $U_{\text{tip}} = 0.95$ V and the set point current was varied from 0.18 nA (curve (I)) to 5.0 nA (II) and back to 0.05 nA (III). Thereby, the tip-sample distance changed by -1.73 Å (I→II) and by $+2.02$ Å (II→III), respectively, as determined from the change in the piezo voltage. These values are in fair agreement with the independently measured apparent barrier height of 3.1 eV. As can be seen from the figure the experimental data essentially collapse. Obviously, the tunneling current and thus the tip-sample distance could be varied over quite a significant range without disturbing the step fluctuations. The fits of the experimental data to (eq. 2) with an upper cutoff time $\Delta t_{\text{max}} = 6$ s yield values for the exponent b of $0.25 (+0.01 / -0.02)$,

$0.27 (+0.01 / -0.01)$, and $0.24 (+0.02 / -0.01)$ for curve (I), (II), and (III), respectively. The error bars indicate the deviations when the somewhat arbitrary cutoff time is replaced by $\Delta t_{\text{max}} = 4$ s or 7.5 s. Fig. 2b reproduces the same kind of data obtained in another experiment. In this case, we kept the tunneling current constant at $I_t = 0.03$ nA. Curve (IV) was recorded at a tip bias voltage of $U_{\text{tip}} = -1$ V, curve (V) at $U_{\text{tip}} = -5$ mV. Upon reducing the bias voltage the tip-sample distance was reduced by 4 Å*. Although the absolute value of the tip-sample separation is not known, this should correspond to a reduction of roughly 50%. Simultaneously, the electric field within the tunneling gap *decreased* by two orders of magnitude. The fits yield values of $b = 0.24 (\pm 0.01)$ and $0.33 (\pm 0.01)$ for curve (IV) and (V), respectively. For this larger variation of the tip-sample distance a significant tip-induced disturbance of the step fluctuations is observed. Note, that the interpretation of the measured exponents b in terms of the diffusion models leads to different results.

Unfortunately, the favorable situation encountered in Fig. 2a depends critically on the configuration of the tunneling tip. To illustrate this, we show an example of a spontaneous tip change (see Fig. 3), where the tip-sample interaction increased dramatically although, as concluded from the vertical tip position, only few atoms were involved in the event. The tip change lead to an initial extension of the z -piezo of 2.1 Å and relaxed to 1.1 Å a few scan lines later. The z -levels of the terraces on both sides of the step appear reduced by the latter amount in the lower part of the image [17]. After the tip change, the step looked much rougher and the resolution was enhanced, as can be seen from the appearance of the corrugation of the atomic rows. Although the origin of such minor and spontaneous tip changes is never known in detail - for instance, we cannot exclude that an impurity atom was involved in this event - this example shows that they can alter tip-interaction and apparent step roughness dramatically. From events similar to the one shown here, we infer that atomic resolution typically coincides with a rather strong tip-sample interaction. This experience is in agreement with both the well known experimental fact that atomic resolution on close packed metal surfaces typically involves small tip-sample separation and with theoretical arguments given by C. J. Chen [19] which relate the Bardeen tunneling matrix element to the Heisenberg resonance energy on the ground of tip-sample wavefunction overlap.

Fig. 2. Step position autocorrelation function versus time for various tunneling resistances. For clarity, only every fourth point is plotted. a) Data sets for three different tunneling current recorded subsequently at $U_{\text{tip}} = 0.95$ V. Inset: logarithmic plot of curve (II) and (III). b) Two data sets recorded subsequently at $I = 0.03$ nA. The solid line are fit curves (see text).

* This relatively large value is due to a slight non-linearity in the I - V -characteristic of that particular configuration of the tunneling tip near zero bias voltage.

Fig. 3. Time-image ($U_{\text{tip}} = 1.15$ V; $I = 1$ nA; size: $90 \text{ \AA} \times 34$ s) with a tip change (see arrow) leading to a rougher appearance of the step edge and the emergence of the atomic rows. Note that in a line scan along $[11\bar{2}]$ direction, usually only every second atomic row is imaged.

Of course, drastic effects like in Fig. 3 can be easily eliminated when performing step fluctuation measurements routinely. But what about the variations of tip-sample interaction for various stable but different tip configurations? To provide a broader statistical basis, we repeated the experiment several times for various values of setpoint current and bias voltage for each new tip and sample. To compare the results, it would be desirable to present the data as a function of the absolute tip-sample distance. However, in STM, measuring the latter usually leads to severe tip damage. Instead, we chose to plot the exponent b of the autocorrelation function versus the tunneling resistance (Fig. 4) which gives a reasonable measure of the absolute tunneling distance. For low gap impedance, significant scattering of the data is observed with values of b up to ≈ 0.33 . In limit of high gap impedance, we find a decrease towards $b \approx 0.25$. Note that these two values point to different diffusion scenarios. Obviously, tip-sample interaction increases the step fluctuations at small tip-sample separation. In most of the experiments, that means for most of the tips in these measurements, the dependence is much stronger

Fig. 4. Exponent b of the step position autocorrelation function versus gap impedance derived from three independent experiments A, B, and C. The solid horizontal lines indicate the predictions of the diffusion scenarios (TD) and (PD), respectively.

than in Fig. 2a. Unfortunately, tip characterization by $I(V)$ - or $I(z)$ -spectroscopy was not found to suitable in order to predict whether a particular tip configuration would allow for undisturbed measurements.

5. Discussion

One important result of this work is that - presupposing a favorable tip configuration - the autocorrelation function does not significantly depend on the tip-sample separation over the broad range investigated in Fig. 2a. Considering the strong distance-dependence typically encountered in atom manipulation experiments [12, 20], it is hard to imagine that the tip causes an essentially constant disturbance of the step fluctuations over such a large range. When an even larger range of tip-sample separation is investigated tip-induced effects become apparent, as was shown in Fig. 2b. Therefore, as a first answer to the persistent discussion [3-5, 10] on the reliability of this kind of measurements, we conclude that it is in principle possible to measure step fluctuations on Ag (111) at room temperature with negligible tip influence.

To determine the dominant diffusion mechanism, the exponent b of the autocorrelation function has to be analyzed. For all the data presented in Fig. 2a, they fall close to 0.25, the model prediction for the one-dimensional diffusion scenario (PD). The high gap impedance- and thus weak tip-sample interaction-limit of Fig. 4 also

indicates that the intrinsic step fluctuations on Ag (111) at room temperature are dominated by the one-dimensional diffusion of atoms along the step edge.

This result agrees with the recently improved interpretation of the experiments by Morgenstern et al. [21, 22]. Very early investigations of step fluctuations by Poensgen et al. also excluded diffusion according to the evaporation-condensation scenario [1]. Having determined the dominant diffusion mechanism, one can start to analyze the prefactor a from the fits. From all the measurements presented here, we obtained a value of $a = 0.7 \pm 0.1$, measured in units of (atomic rows)². In the absence of temperature dependent measurements, we cannot give an accurate number for the activation energy of the microscopic processes from our data. However, a rough estimate using reasonable assumptions for the attempt frequency (see [16]) shows that it is in reasonable agreement with the expectations from theoretical calculations by Stoltze [23].

Let us now return to the issue of tip-sample interaction. Fig. 4 shows that - as a second answer to the major question addressed in this work - for the majority of tip configurations, a significant perturbation is observed in this system. From the scattering of the data, we conclude that the amount of the tip-induced disturbance varies from tip to tip. This finding is in accordance with a recent theoretical study by Bouju et al. [24]. In their simulations of atom manipulation experiments, these authors calculated the dispersion energy between the tip and a weakly bound adsorbate atom for three different configurations of the tip apex. Depending on the tip configuration, they find different modes of adatom manipulation and different threshold values for the tip-sample distance at which manipulation sets in. Notwithstanding the differences between this system and ours, their conclusion that "the tip apex structure ... plays an important role in the atomic manipulation process" should hold for our experiments, too.

It is interesting to compare this result with our own previous experiments [5] where we obtained a value of $b = 0.32 \pm 0.03$ for the exponent of the correlation function. These earlier data were recorded at various tunneling resistances around 1 G Ω or below. We anticipated already in [5] that $b \approx 0.32$ is not reliable. These results are, however, in agreement with Fig. 4 and the assumption of a tip-induced increase of the step fluctuations at low gap impedance.

The disclosure of a tip-induced disturbance of the atomic motion was recently described in the literature for semiconductors [9] as well as for metallic systems [10, 25]: Li et al. [10] reported on tip-assisted diffusion on Ag (110). They find that the tip can significantly affect adatom diffusion at ambient temperature and at sub-nA tunneling

currents. Bott et al. investigated the diffusion of individual adatoms on Pt (111). They found that even at low temperature the mean square displacement of adatoms increased when the tip-sample distance was reduced [26]. From a comparison of their growth studies to kinetic Monte Carlo simulations, they obtain a diffusion barrier of 0.27 eV for the adatom migration. For both systems, Ag (110) and Pt (111), the diffusion barriers are much higher (> 0.29 eV for Ag (110); [23]) than both the experimental (0.097 eV; [27]) and the calculated value (0.064 eV; [23]) for adatom migration on Ag (111). In view of the lower diffusion barriers, it is not surprising to find tip-induced perturbations on Ag (111), too.

The following scenario gives a tentative explanation: The slow speed of the tip compared to adatom diffusion together with an attractive interaction with individual adatoms might result in a „cloud“ of enhanced adatom density underneath the tip. Scanning across the step would then lead to an increased material exchange between the step edge and the adatom gas. Thus a combination of tip-sample interaction and the low self-diffusion barrier on Ag (111) would explain the observed tip-induced effects. This concept is in analogy to the one proposed by Li et al. [10].

Based on the data presented in Fig. 2, we further suggest that the electric field within the tunneling gap plays a minor role for the observed tip-induced disturbance at small gap impedance. The significant decrease of the tip-sample distance between curve (IV) and curve (V) in Fig. 2b together with the simultaneous reduction of the electric field lead us to conclude that short-range forces rather than the electric field are responsible for the increase of the step fluctuations observed in curve (V). Furthermore, we did not find any effect of the polarity of the bias voltage. For instance, curve (V) and curves (I), (II), and (III) were recorded at roughly the opposite electric fields. At least for static dipole moments one would expect to see a polarity dependence which was indeed observed earlier in experiments with Cs atoms on GaAs (110) [28].

6. Conclusions

We analyzed step fluctuations on Ag (111) as a function of tunneling current and bias voltage. In most of the experiments, we observe a tip-induced increase of the step fluctuations at low gap impedance. The amount of tip-induced perturbation depends strongly on the tip.

From the high gap impedance limit we conclude that the kinetics of step fluctuations on Ag (111) at room temperature is dominated by one-dimensional mass transport along the step edges. We found that the only reliable procedure to eliminate the tip influence is to vary the gap impedance over a range

as large as possible. Finally, we argue that short range forces rather than the electric field in the tunneling gap lead to the disturbances observed at low gap impedance.

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