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STM-imaging of nanostructure dynamics on Ag(111)-experimental challenges and solutions

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Abstract

We describe experimental problems arising with the continuous observation of nanostructure dynamics by STM. We discuss the necessity to use a high-speed STM, possibilities to deal with the thermal drift, and tests to rule out the influence of the scanning process on the observation.

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

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There are basically two approaches to investigate dynamic processes by scanning tunneling microscopy (STM). The one most widely used is the "quench and look" method. Structures that develop at higher temperature are frozen out by quenching to a lower temperature at which the STM-recording is performed. Therefore, the state of the system at a particular point in time can be investigated. This method is well known from field ion microscopy (FIM) [1]. In contrast to FIM-measurements, where the sample area is small, with an STM it is difficult to find again a specific structure in order to follow its evolution in time. Therefore, in order to observe the time evolution of single structures the recording has to take place while the process of interest proceeds. With increased interest in nanostructures and their development in time the second approach "on-site on-time" starts to become of interest (e.g., see Ref. [2]). In contrast to the snapshots of the "quench and look" method, with the "on-site on-time" method dynamics can be followed quasi-continuously. However, this approach poses new challenges for the experimentalist. Especially, an appropriate time resolution has to be chosen, not only to catch the process of interest, but also to deal with STM specific problems, i.e., finite scanning time and thermal drift. Further, the influence of the scanning process on the observation has to be ruled out. The aim of this paper is to discuss these problems.

In a recent letter we described the physics of vacancy island motion on Ag(111) at room temperature observed with the "on-site on-time" approach [3]. Before we use this example as a vehicle to demonstrate the method, we give a short account of the vacancy island motion. Monatomic deep vacancy islands perform a random motion on Ag(111) at room temperature. The diffusivity of the vacancy islands increases with decreasing island size. On an

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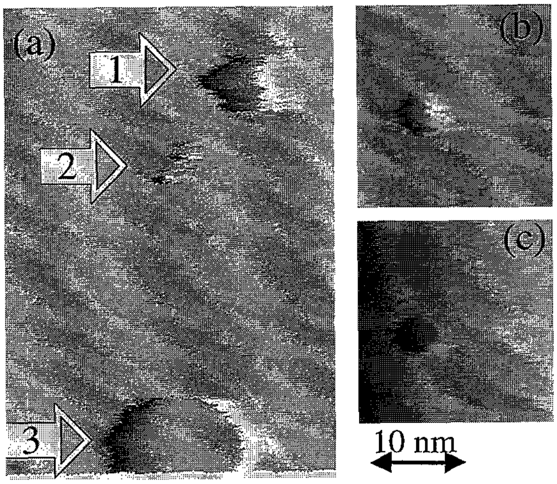

Fig. 1. STM-images. (a) Classical mode scanned within $t_i = 2.4$ min, $U_t = -0.5$ V, $I_t = 10$ nA. (b) Classical mode, $t_i = 160$ s, $U_t = -0.5$ V, $I_t = 5$ nA. (c) Modified mode, $t_i = 16$ s, $U_t = -0.5$ V, $I_t = 5$ nA.

atomistic scale the motion results from the diffusion of single Ag atoms from one part of the vacancy island boundary to another one, either by atoms that diffuse along the island boundary or by atoms, that detach from the boundary, diffuse over the terrace of the vacancy island and reattach to the boundary. The scaling laws of the vacancy island diffusivities reflect the different dimensionalities of the diffusion paths and allow to distinguish between the two atomic mechanisms. For vacancy islands on Ag(111) at room temperature with $5 < r < 15$ nm the island motion is caused by adatom diffusion over the terrace; their diffusivities scale with $1/r^2$.

We now tackle the problem of selecting the appropriate time scale to observe such a motion. The experiments were performed in a UHV-chamber. After several ion sputtering and electron bombardment heating cycles vacancy islands of different sizes are created by ion sputtering [4]. The sample is then transferred in situ to an STM of the beetle-type [5]. In the classical operating mode the image recording is started manually and an image of 512 lines is recorded in several minutes. Fig. 1 demonstrates a necessity to speed-up this recording for dynamic observations. The vacancy island "1" on Fig. 1a seems to have a shape that differs considerably from the expected equilibrium shape on Ag(111) at room temperature (hexagon with rounded corners); the larger island "3" resembles it more. The strange shape of island "1" in Fig. 1a is due to the fact that the island has moved markedly during the scanning time. The larger island "3" moved slower ($D \alpha 1/r^2$) and has thus been imaged more correctly. This issue is further illustrated by the dark spots "2" in Fig. 1a: they result from a single very small island which moving very rapidly and normally to the scanning direction has been imaged several times. The improvement achieved by reducing the scanning time by one order of magnitude is obvious in Figs. 1b and 1c. The image of the same island recorded with higher speed (Fig. 1c) approaches the expected equilibrium shape. Thus a high-speed STM is necessary for a correct shape imaging of fast moving structures, which is a prerequisite to determine the center-of-mass position of islands. We modified the classical arrangement to allow us a recording of one image of 128 lines within 0.8 s and an automatic imaging at fixed time intervals between 1 s and 150 min. We could have reached in principle the same goal by slowing down the process by lowering the temperature. However, for a reliable statistical treatment up to 600 images had to be taken in a row. Slowing down the process would enhance the recording time of such a movie beyond reasonable experimenting times. The thermal drift (discussed below) during hours of measurement would prevent the proper pursuit of a single structure, and surface contamination to which diffusion is very sensitive would have impaired the results.

To illustrate the performance of the high-speed STM Fig. 2 shows a large vacancy island. The entire scans were recorded in 0.8 s; the image shown is only one quarter of the total area scanned. The island shape changes considerably with time and a resulting center-of-mass displacement is observed.

Another time problem arising while scanning repeatedly is indicated in Fig. 3. We attempt to scan exactly the same spot of the sample on all images, but the thermal drift causes a slight shift. The black–white line marks the path of a large vacancy island of about 60 atoms in diameter. The mean square displacement of such a large island is due to its random motion only about 0.9 nm^2/min [3]. Hence, this island can be used to demonstrate the thermal drift towards the lower right in Fig. 3. Even

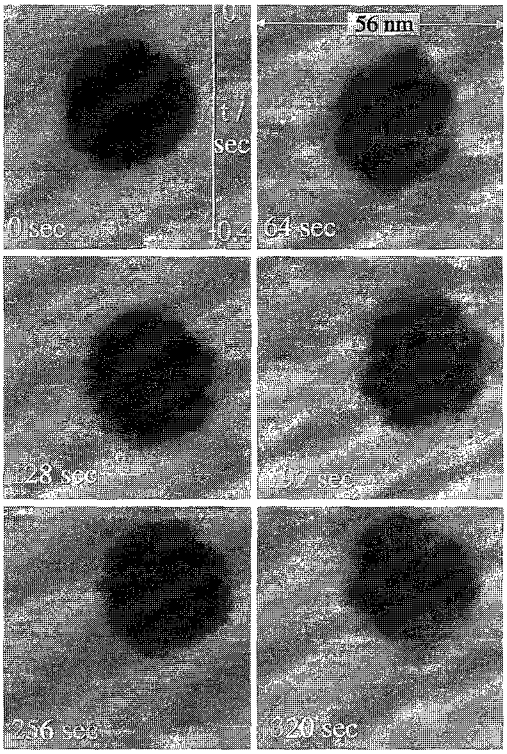

Fig. 2. Step edge fluctuations of a vacancy island; causing a motion of its center-of-mass; $U_t = -0.5$ V, $I_t = 10$ nA.

worse, the drift may vary in size and direction over larger periods. If there was a dislocation or an impurity on the surface, it would be possible to use this as a reference point. However, these structures might influence the motion and fortunately there were none. We therefore chose a different approach and separated the motion of the vacancy islands from the motion of the microscope by taking the relative motion of two about equally sized vacancy islands. As shown in [3], the formalism of a random motion can easily be modified to describe the relative motion of a vacancy island pair, which is also random, if both islands move randomly. In Fig. 4 there is a drift to the lower right corner on the movie, but the relative motion of the two vacancy islands is obviously random (Fig. 4b).

The monitoring of the relative motion of two vacancy islands [6] asks also for high-speed and fast repetition imaging because the two islands are scanned at different times t_1 and t_2 (see Fig. 4). As a result the following two points have to be considered, when choosing the relative motion approach. First, the islands move while scanning. Therefore, the time intervals Δt_1 and Δt_2 between the rescanning of the first and the second island, respectively, differ slightly from the time interval Δt between the start of consecutive images. We have to choose scanning speeds so that the error based on this effect is negligible compared to the statistical error, and hence, $\Delta t_1 \approx \Delta t_2 \approx \Delta t$ can be assumed for data analysis. Asking for an error less than 10% of the statistical error and assuming that for an island with diffusivity D moving primarily in y-direction (worst

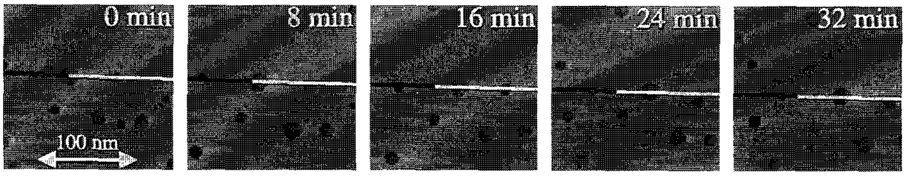

Fig. 3. Thermal drift: vacancy island motion on Ag(111) with an additional drift of the microscope; line marks the center-of-mass of the largest vacancy island; black part has the same length on all images; $U_t = -0.5$ V, $I_t = 5$ nA.

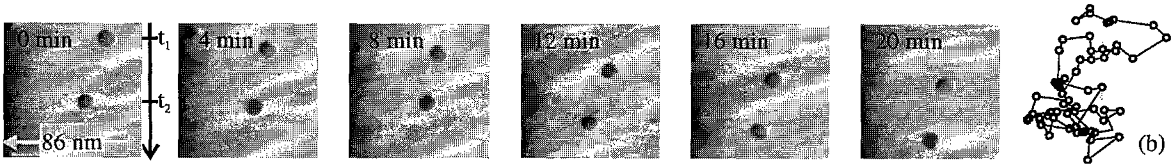

Fig. 4. Relative motion of two vacancy islands of about same size; $U_t = -0.49$ V, $I_t = 9.5$ nA. (b) Relative displacement for all 56 images of the movie enlarged by a factor of 4; $\Delta t = 30$ s.

case) the displacement during the deviation from Δt is less than four times the mean displacement, $\sqrt{(D)/s^2\Delta t} < 0.1$ has to be fulfilled, where s is the scanning speed. Second, one has to scan fast enough, so that the *change* in thermal drift during $\Delta t^{(1,2)} = t_1 - t_2$ is negligible. The most extreme change we ever encountered was from 0.4 to -0.3 nm/min within 100 min. With a scanning speed of up to 1 image/min the change during $\Delta t^{(1,2)} < 1$ min is less than 0.007 nm/min. Hence, by analysing the relative motion the error caused by the thermal drift is reduced to its derivative and so becomes negligible.

Finally we discuss another important point. The motion of phosphoric vacancies on GaP(110) [7] was scanning induced. However, we are interested in intrinsic surface dynamics and want to rule out any effect of the scanning. To show that the observed motion is not caused or influenced by the scanning process scanning parameters have to be varied and the experimental results compared. We varied tunneling current, tunneling voltage (magnitude and polarity), scanning speed, scanning direction. Further, the time of tip–sample interaction was enhanced by scanning the sample several times in between the analysed images. We demonstrate the independence of the motion from the scanning by this example. For a scanning speed of 12 s/image one image was taken every 240 s and the island displacement between the images was measured. Then the spot was scanned every 15 s with the same scanning speed. Here the displacement between every 16th image was measured only, so that the time difference was 240 s again. The second procedure was done twice. The comparison of the three resulting displacement distributions in Fig. 5 shows that the deviation is negligible within the statistical variation. Hence, the scanning process does not influence the diffusion of the vacancy islands.

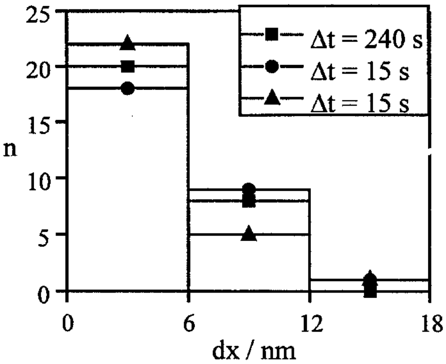

Fig. 5. Displacement distribution; scanning speed: 12 s/image; recording frequency: 1/240 and 1/15 Hz, respectively.

In conclusion we have shown, that, if choosing the "on-time on-site" approach in order to study surface dynamics by STM, it is crucial to select the right scanning speed, to eliminate the thermal drift and to make sure that the physical process is not influenced by the scanning.

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