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An STM study of the step structure of Pb(110) and Pb(111)

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Abstract

The (110) and (111) surfaces of lead are investigated by scanning tunneling microscopy. They are atomically resolved with corrugations of 0.1 Å and 0.6 Å respectively. The STM images allow conclusions about the motion of the atoms at step edges and step-step interaction. At room temperature the steps on both surfaces are below the roughening transition. The influence of impurities and tip-surface interaction on the step fluctuation is discussed.

Keywords: Lead; Low index single crystal surfaces; Scanning Tunneling Microscopy; Surface structure

1. Introduction

Steps are important for many processes studied in surface science like growth, adsorption and roughening. Since the surface melting was discovered on Pb(110) [1], lead surfaces found widespread attention. The roughening temperature and the roughening mechanisms at atomic scale are not yet clear.

Steps on surfaces at elevated relative temperatures (T/T_m , where T_m is the melting temperature) cannot be imaged properly with STM. This is valid for low indexed as for vicinal metal and semiconductor surfaces [2–9]. The reasons are kink excitation, annihilation and movement processes in time intervals shorter than the time needed for one scan line. The undersampling in time of the step positions causes a mixture of temporal and spatial information. The problem of the evaluation of step shapes is therefore to find out

whether the step fluctuations are mainly spatial or temporal or both. If there are frizzes on a step of an STM topograph, there is no a priori or obvious information whether the whole step has been at that time at that position, or whether it was a moving single kink or a double kink. In the literature there is some controversy about that topic. For low relative temperatures the fluctuations are so slow that they are almost only spatial and the dependence is linear. At higher temperature, when the time constants for kink change processes are in the time range needed for one scan line, the fluctuations behave according to a power law of t .

Here we present STM-images of Pb(110) and Pb(111) and study the time dependence of the step fluctuations thereon.

2. Experimental

The experiments were performed in an UHV system with two chambers. One chamber is used for lock-in

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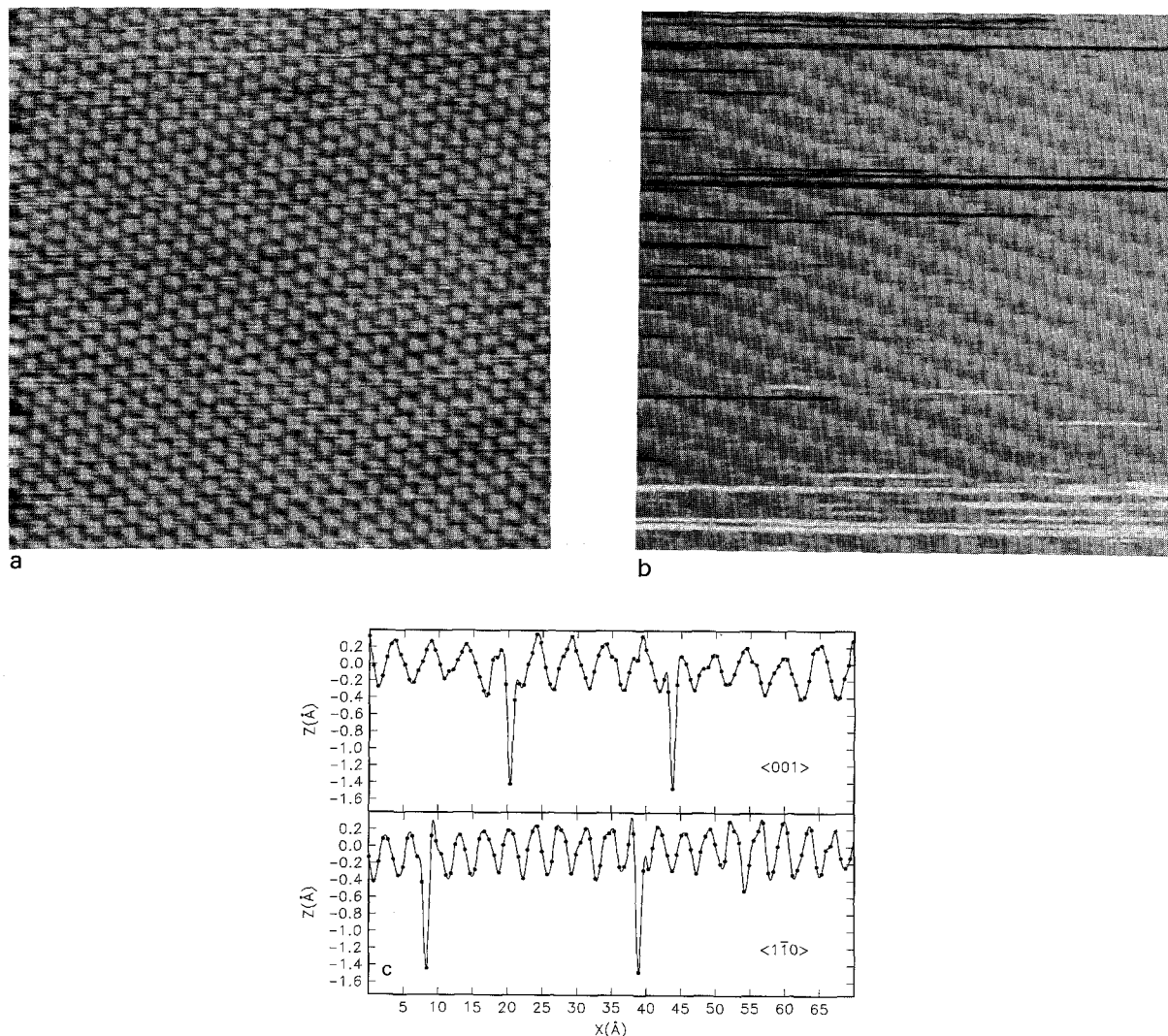


Fig. 1. (a) $(100 \text{ \AA})^2$ topograph of Pb(111), $V_t = -0.4 \text{ V}$, $I_t = 1.3 \text{ nA}$. (b) $(100 \text{ \AA})^2$ topograph of Pb(110), $V_t = -0.6 \text{ V}$, $I_t = 1.3 \text{ nA}$. (c) Sections along $[1\bar{1}0]$ and $[001]$ through a topograph of Pb(110) (lattice constant: $a_{\text{bulk}} = 4.95 \text{ \AA}$).

to UHV and sputtering. The second is equipped with a commercial STM (Omicron μ -STM) and an AES-system. The pressure in the main chamber is below 5×10^{-11} mbar. The tip was electrochemically etched from tungsten wire and cleaned by sputtering and voltage pulses up to 10 V while tunneling. The Pb sample surfaces were chemically etched and cleaned in situ by cycles of Ar ion sputtering until the contaminations were found to be below the detection limit of AES.

3. Results and discussion

Figs. 1a and 1b show $(100 \text{ \AA})^2$ topographs of the (111) and the (110) surface with atomic resolution. The corrugation amplitudes obtained are 0.5 and 0.1 \AA respectively. The images presented in this article contain only raw data. For the (110) surface a higher corrugation amplitude (0.7 \AA) was found occasionally, which may be caused by a special tip shape. In

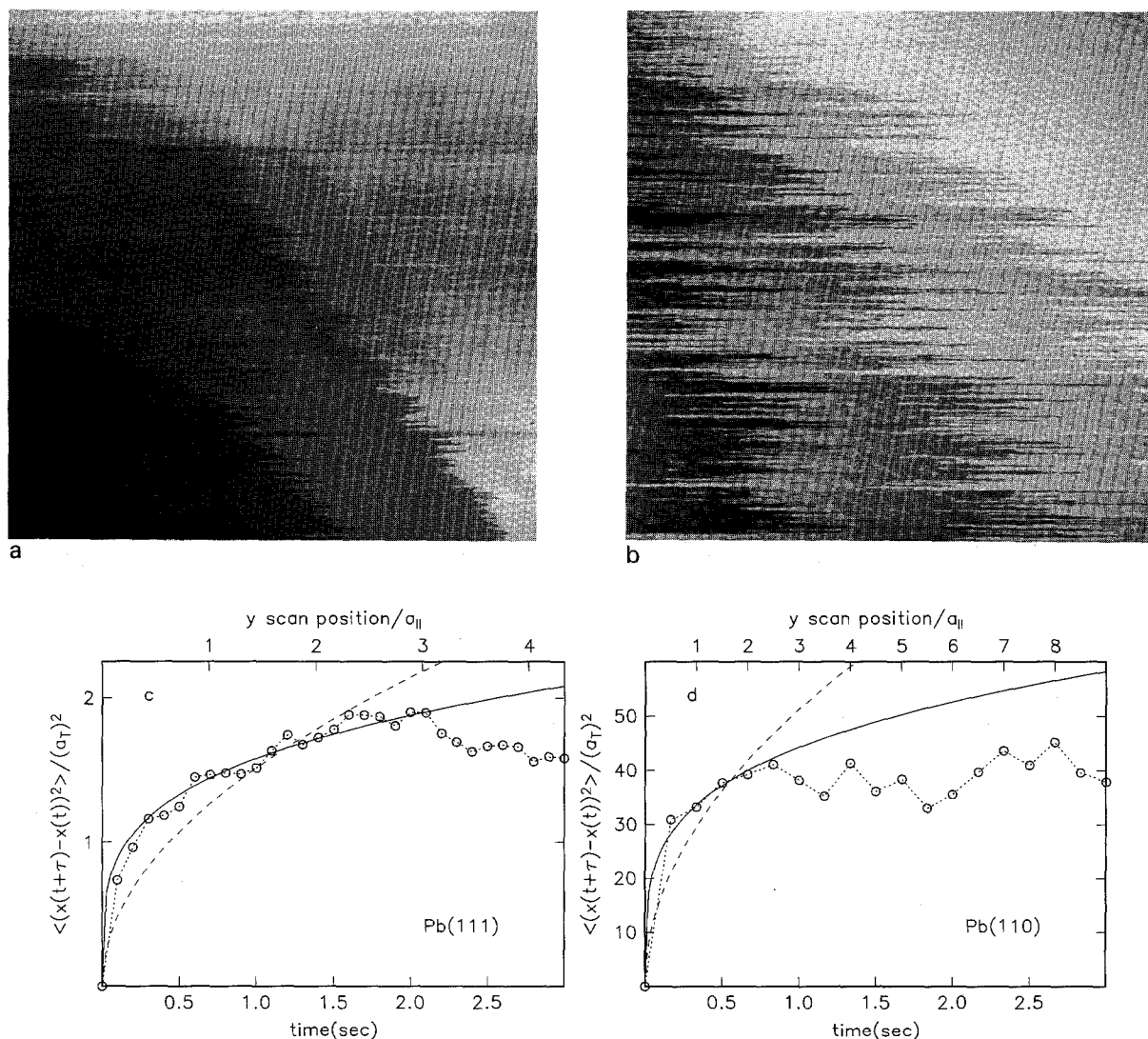


Fig. 2. (a), (b) $(200 \text{ \AA})^2$ topographs of Pb(111) and Pb(110), $V_t = -0.4 \text{ V}$, $I_t = 1.3 \text{ nA}$. (c), (d) Time dependence of the step fluctuations on Pb(111) and Pb(110). Circles: experimental values. The solid (dashed) line is a $t^{1/4}$ ($t^{1/2}$) fit of the small time data. (y scan position in units of the lattice constant along y; $F(\tau) = \langle (x(t+\tau) - x(t))^2 \rangle$ in square units of the lattice constant perpendicular to y).

comparison to the reconstructed surfaces of Au(110) and Pt(110) the $[1\bar{1}0]$ rows are better resolved here. It seems to be a general finding that the $[1\bar{1}0]$ rows can only be resolved properly on unreconstructed surfaces. A pure geometric consideration yields that a more isotropic arrangement of atoms would lead also to a more isotropic electron density around the Fermi level; but this should only result in a higher corruga-

tion amplitude for the direction perpendicular to the chains on the reconstructed surfaces. So the smoothing of the electron density in the $[1\bar{1}0]$ chains is supposed to be related to the tendency of a surface towards reconstruction.

The stripes in x -scan direction in Fig. 1b are from step movements. In the stripes the atoms are resolved too, with the expected shift between the layers. The

dark stripes arise, if the measured terrace just leaves the scanned area for a moment (< 0.1 s); the bright ones result from a neighboring terrace, the movements of which sometimes reach in the scanned area. A section through such an image along $[1\bar{1}0]$ and $[001]$ is given in Fig. 1c. Note that the height of the stripes almost equals 1.75 \AA , the monoatomic step height on that surface. The (111) surface topograph in Fig. 1a was recorded at equal conditions (11 s/picture), but does not show any stripes due to the lower step mobility.

Figs. 2a and 2b show $(200 \text{ \AA})^2$ topographs of Pb(111) and Pb(110). The atoms on the (111) surface and the $[1\bar{1}0]$ chains on the (110) surface are still visible. The steps appear frizzy, due to the under-sampling in time of the step position. The main part of the steps parallels low indexed directions. In Fig. 2a the step orientation is $[12\bar{3}]$ and $[11\bar{2}]$, although at the transition from one direction to another the step is rounded. If the scan direction is alongside a step there are no frizzes. This indicates that the kink concentration is rather low and the main part of the frizzes are time fluctuations. Therefore this is not the real step shape but whole parts of the step are at the actually measured position. On the (110) surface the frizzes are much longer ($\sigma_{f,(110)} = 30 \text{ \AA}$, $\sigma_{f,(111)} = 5 \text{ \AA}$) so that the fringes of one step sometimes reach to the neighboring step. The correlation of the absolute step position is lower than for (111) and there is no clear preference of low indexed directions.

In the upper left hand part of picture 2b the absolute and the relative movements of the steps decrease more and more. This may be caused by a pinning center located above the scanned area. Often a pinning center is the origin of a step bundle. The impurity densities ($\rho_{(110)} \cong 10^{-5} \text{ \AA}^2$, $\rho_{(111)} \cong 10^{-6} / \text{ \AA}^2$) are taken from $(4000 \text{ \AA})^2$ scans by counting the protusions in each case. Between the impurities the steps have the possibility to move. At high step density, wandering or even movement of steps is damped. The point is that the arrangement of the steps is like a train: if one step moves, all other steps have to move accordingly otherwise overhangs would be created (entropic repulsion); overhangs are supposed to be unstable. There is also an effective step step repulsion (energetic repulsion) at lower relative temperature because the strain field at a step lowers the surface energy only, if there

is no other step nearby. In Fig. 2b the step arrangement is rather dense with respect to the length of the frizzes. Therefore there is a strong correlation between the steps. As a consequence of the repulsion between steps they can only move synchronously which leads to similar shapes of neighboring steps. The pair correlation coefficient for two steps depends on the terrace width. In Fig. 2b it is $+0.5$ for short Δy and a terrace width of 80 \AA .

All the measured step positions are regular lattice sites. From this one can conclude that a single atom hops at most once during the time needed for scanning one lattice constant (here: 3 ms). The step speed is faster than the y scan velocity (i.e. 5 \AA/s), because the autocorrelation of the step position is almost zero after only one scan line.

According to Ref. [6] for sufficient high temperatures there is no difference between the correlation function calculated from conventional or time images. This allows to convert the y coordinate of conventional images into a time coordinate. The evaluation of the mean square displacement ($F(\tau) = \langle (x(t+\tau) - x(t))^2 \rangle$) is shown in Figs. 2c and 2d. For the (111) as for the (110) surface steps the mean square displacement varies with $t^{1/4}$. This indicates that mass transport happens solely alongside the steps and that the steps are still below roughening. Three elementary processes for step movement are considered. (i) kink-diffusion-limited step motion, if mass exchange is only possible along steps; (ii) step/terrace-exchange-limited step motion, if diffusion via adatoms on the terraces is involved and (iii) terrace-diffusion-limited step motion, where a static concentration of adatoms is present on the terraces. The correlation function of the step position varies with $t^{1/4}$, $t^{1/2}$ or $t^{1/3}$, respectively [10]. Above the roughening temperature a logarithmically increasing correlation in space is expected [13]. An example for $t^{1/4}$ -step fluctuation is Cu(1 1 19) [6]; $t^{1/2}$ behaviour has been observed for Au(110) [4]. The $t^{1/4}$ behaviour on Pb(111) found here is in agreement with the results in Ref. [11]. At a certain time (or distance) the time correlation deviates from the power law (at 2 s in this study; at 0.1 s in Ref. [11]). This may be interpreted as influence from step pinning. The circles in Fig. 2c are calculated from the $[12\bar{3}]$ oriented step (B-type) in the right part of Fig. 2a. For Fig. 2d a $[1\bar{1}0]$ oriented step piece was used.

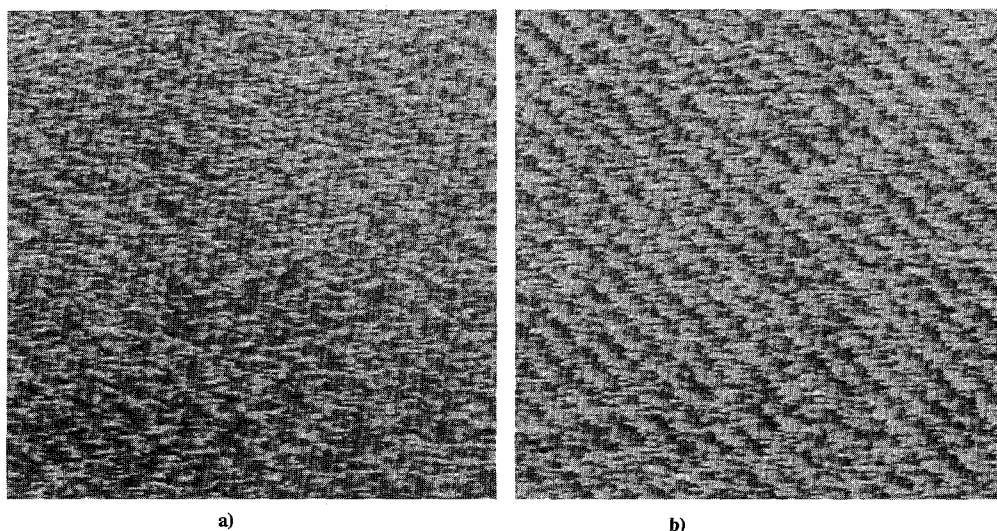


Fig. 3. $(100 \text{ \AA})^2$ topograph of Pb(110) at closer tunnel distance, $V_t = -0.010 \text{ V}$, $I_t = 1.2 \text{ nA}$. In the left picture the tip scans from right to left; in the right picture viceversa.

On the (110) surface one has to choose carefully the steps for a statistical analysis. Because σ_f is higher, the criterion that the steps do not interact is broken earlier. The closer the steps, the more their motion is damped by the repulsion. Therefore $F(\tau)$ of narrow terraces saturates fastly. But the fluctuations at really free steps on Pb(110) (Fig. 2d) can also be classified according to $t^{1/4}$, at least for a certain time. A quantitative analysis will be given in Ref. [12].

Frizziness can also be caused by tip surface interaction. It is known that the force applied at a surface is in the range 10^{-9} to 10^{-10} N. In Figs. 3a and 3b the scan lines in each frame were taken alternating while the tip moves from right to left and viceversa. In comparison to the other topographs, a very low tunnel voltage (0.01 V) was applied here. The frizzes belong to the $[1\bar{1}0]$ rows. They always point in the direction of the tip movement; this shows that there is an attractive interaction between tip and sample surface atoms at low tunnel distances.

Because of the danger of misinterpreting the data they have to be checked for the possibility that the frizzes at steps arise from pulling along the atoms by the tip. One could recognize a tip surface interaction by a difference in the fringes' root mean square displacement in pictures where the scan lines have been

taken alternating in different scan directions. Because of the limited scan range STM pictures usually contain either only descending or ascending steps. It is more probable to detach a step atom and carry it on the terrace at the same level than to lift it to the upper terrace. Thus if scanning transverse to a step, an interaction would cause an asymmetry of the fringes' length distribution. In Ref. [7] frizziness induced by tip surface interaction on copper has been reported. This was concluded from the increase of the fringe length during successive pictures of the same area and also from coincidence of tip changes with changes in the average fringe length. A stronger case of tip surface interaction are short circuits between sample and tip observed above room temperature on lead [14]. Neither short circuits nor changes in the length distribution of the frizzes have been observed in our measurements. Although the above mentioned tests lead to a negative result, the authors cannot exclude contributions from tip surface interaction completely. On the other hand the genuine existence of the frizzes on Pb(110) implies surprisingly large mass transport.

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References

- [1] J.W.M. Frenken and J.F. van der Veen, *Phys. Rev. Lett.* 54 (1985) 134.
- [2] M. Poensgen, J.F. Wolf, J. Frohn, M. Giesen and H. Ibach, *Surf. Sci.* 274 (1992) 430.
- [3] J.F. Wolf, B. Vicenzi and H. Ibach, *Surf. Sci.* 249 (1991) 233.
- [4] L. Kuipers, M.S. Hoogeman and J.W.M. Frenken, *Phys. Rev. Lett.* 71 (1993) 3517.
- [5] J.W.M. Frenken, R.J. Hamers and J.E. Demuth, *J. Vac. Sci. Technol. A* 8 (1990) 293.
- [6] M. Giesen-Seibert, R. Jentjens, M. Poensgen and H. Ibach, *Phys. Rev. Lett.* 71 (1993) 3521.
- [7] F. Thibaudau and J. Cousty, *Ultramicroscopy* 42–44 (1992) 511.
- [8] S. Rousset, S. Gauthier, O. Sibolet, J.C. Girard, S. de Cheveigné, M. Huerta-Garnica, W. Sacks, M. Belin and J. Klein, *Ultramicroscopy* 42–44 (1992) 515.
- [9] J.C. Girard, S. Gauthier, S. Rousset, W. Sacks, S. de Cheveigné and J. Klein, *Surf. Sci.* 301 (1994) 245.
- [10] E.D. Williams, *Surf. Sci.* 299/300(1994) 502, and references therein.
- [11] L. Kuipers, PhD thesis, University of Amsterdam, 1994.
- [12] S. Speller, W. Heiland, A. Biedermann, E. Platzgummer, C. Nagl, M. Schmid and P. Varga, to be published.
- [13] J. Villain, D.R. Grempel and J. Lapujoulade, *J. Phys. F* 15 (1985) 809.
- [14] L. Kuipers and J.W.M. Frenken, *Phys. Rev. Lett.* 70 (1993) 3907.