RECEIVED
APR 25 1996
OSTI

IS-T 1756

RHEED Studies of Ag/Si(111) Growth at Low Temperatures

by

Koehler, Ulrich

MS Thesis submitted to Iowa State University

Ames Laboratory, U.S. DOE

Iowa State University

Ames, Iowa 50011

Date Transmitted: January 2, 1996

PREPARED FOR THE U.S. DEPARTMENT OF ENERGY

UNDER CONTRACT NO. W-7405-Eng-82.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
plc

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

AVAILABILITY:

To DOE and DOE contractors: Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

prices available from: (615) 576-8401
FTS: 626-8401

To the public: National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

TABLE OF CONTENTS

ACKNOWLEDGMENTS	iii
CHAPTER 1. INTRODUCTORY CONSIDERATIONS	1
Ultrahigh Vacuum	1
Crystallography	2
Electron Diffraction	6
CHAPTER 2. EXPERIMENT	11
CHAPTER 3. RESULTS	14
CHAPTER 4. SUMMARY	21
BIBLIOGRAPHY	22
APPENDIX SPOT PROFILES	24

ACKNOWLEDGMENTS

Gaudeamus igitur,
juvenus dum sumus;
post iucundam juventutem,
post molestam senectutem
nos habebit humus.

Who would have thought of all this on that cold evening of November 9, 1989?
— I shall ever be thankful to the chances that came with it.

Whom else to thank? Where to start?

I would like to mention my major professor Michael Tringides, for the help he gave me in completing this work, and all the support that was necessary.

With the same gratitude I want to mention Senator J.W. Fulbright, whose idea in the first place made it possible for me to study in the United States of America. May his ideal of mutual understanding and tolerance grow, and may I have contributed towards this goal.

Where would I be without my family, without friends? It is impossible to mention them all, so I would simply like to remark my thankfulness to all their help, assistance, and support.

This work was performed at Ames Laboratory under Contract No. W-7405-Eng-82 with the U.S. Department of Energy. The United States government has assigned the DOE Report number IS-T 1756 to this thesis.

CHAPTER 1. INTRODUCTORY CONSIDERATIONS

It has always been interesting and rewarding to try to discover some of the laws that govern nature; and especially so with the immediate surface of macroscopic bodies, as well as the interactions of those surfaces with films. To a very large extent this has been hampered for a long time because of ever-present contaminations (cf. later), but also by a lack of adequate techniques. Among other techniques, RHEED — **R**eflection **H**igh-**E**nergy **E**lectron **D**iffraction — proved of tremendous value for controlling the growth process scientists aimed at.

Also it has always been the goal to grow films of better quality, i.e., films that are as smooth as possible, perhaps even on an atomic scale. This goes of course hand in hand with increasing knowledge of the basic physical laws.

The Ag/Si(111) system is well-suited for this work, since (a) the Si(111) plane with its structure and properties is relatively well-known, and (b) in difference to for example Si/Ge silver and silicon do not mix but rather conserve a sharp interface.

Ultrahigh Vacuum

One of the problems connected with research of surfaces is obviously contamination. Under normal circumstances, virtually every surface is saturated with various molecules of substances found in the air: water, carbon dioxide, oxygen, &c. The

Langmuir rule relates the exposure (in monolayers)

$$E = 10^6 \cdot p \cdot t,$$

with the pressure in Torr and time in seconds relatively well. Therefore, in order to probe the actual surface and exclude the influence of those contaminants on atomic growth, it is necessary to remove them permanently. Only in ultrahigh vacuum with $p < 10^{-9}$ Torr it is possible to establish these conditions.

Crystallography

It is probably appropriate to digress deeper into the matter of crystallography — since that's an always implied topic. For reasons of simplicity I would like to concentrate on the ideal case, i.e., neglect all possible faults.

The ideal three-dimensional crystal is thought to extend to infinity in all dimensions, in order to be able to neglect fringing effects. Its most dominant feature is periodicity: a unit, the so-called unit cell, repeats itself over and over. This smallest possible unit showcases all structural details found in the crystal. Using only the basis vectors ($\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$) that span this cell, one may reach every equivalent point anywhere in the crystal. The same approach can also be used for materials that are made up of more than one element, but exhibit a certain periodicity as well. Examples are the classic sodium chloride, and the semiconductor silicon carbide. Semiconductors of the fourth group in the periodical system of elements, which form covalent bonds, are mainly found in a certain type of crystal — face-centered cubic, or fcc for short (cf. Figure 1.1).

From this starting point, surfaces are obtained simply by cutting. Usually Miller

Figure 1.1: Schematic of an fcc cell.

indices are being utilized to describe the planes and their orientation. They are simply defined as the reciprocals of the fraction of the basis vector where the plane cuts the unit cell (and since the unit cell is the smallest unit, there is no need to specify more than the orientation with respect to the unit cell). Frequently used planes in conjunction with Silicon are (100), (110), and (111). These planes are parallel to the y and z axes, the z axis, and cut through all of them at one lattice constant, respectively. Of course, a different orientation also implies different atom arrangement, and a different atom density in those planes (as shown in Figure 1.2). Unfortunately, this idealistic picture is especially untrue for Silicon. In order to lower the surface energy the atoms tend to relax — change bonding lengths along the surface normal — and reconstruct. The latter is probably readily perceptible, since it can be seen directly in changes of the (surface) unit cell. Especially the

Figure 1.2: Schematic of (a) a (110) plane and (b) a (111) plane atom arrangement (of an fcc lattice).

Si(111) plane reconstructs heavily in a very remarkable way. As of now, the model by Takayanagi *et al.* [1] — the so-called DAS model, for it incorporates **D**imers, **A**datoms, and **S**tacking faults, — is generally accepted as an accurate description of the reconstructed system. It involves three atomic layers and the forming of atomic pairs and stacking faults. As a result, the reconstruction unit cell incorporates an area of 49 original, bulk unit cells. The surface density of dangling bonds of the silicon atoms would also be reduced to 12 (from the original 49). An excellent perspective of the unit cell of the Si(111) 7×7 can be seen in Figure 1.3.

Figure 1.3: DAS model of the Si(111) 7×7 reconstruction (from [1]).

Electron Diffraction

Electron diffraction was first thought of when Louis de Broglie (see [2]) conceived the idea of "particle-wave duality" in quantum mechanics, in particular the relation between momentum, for instance of an electron, and wavelength:

$$\lambda = \frac{h}{p}.$$

This gave scientists the possibility to explore atomic structures which were previously inaccessible due to the fact that the maximum resolution is of the order of the used wavelength. In fact, first confirmation of de Broglie's theory came from Davisson and Germer, who discovered electron diffraction from a Nickel crystal. However, the use of Reflection High-Energy Electron Diffraction as done in this study requires some more considerations.

In the first place there is the question of penetration depth and surface sensitivity. Since we want to probe surface properties, it must be the ultimate goal to either prevent the electrons from entering too far into the substrate or effectively discriminate electrons with bulk information. Research into the problems of penetration depth and electron mean free paths has been made for instance by Seah and Dench, who phenomenologically determined the electron mean free path as a function of electron energies. Since the high energy (5keV in this case) electrons have a very large mean free path, and the discrimination is rather hard to perform, the most widely used way to overcome this is shifting to grazing incidence. This means that the electron strikes the surface under an angle of almost 90° to the surface normal. In our experiments, we vary the incident angle between ca. 0.5° and 4.5°.

The second thought should be made towards the formation of the diffraction

pattern (loosely based on [3]). For diffraction we consider elastic scattering only, i.e., the magnitudes of incident and outgoing wave are equal. Therefore the momentum transfer

$$\mathbf{S} = \mathbf{k} - \mathbf{k}_0, \quad (|\mathbf{k}| = \frac{2 \cdot \pi}{\lambda}),$$

and, as already mentioned, a two-dimensional lattice is described by

$$\mathbf{r}_i = m_1 \cdot \mathbf{a}_1 + m_2 \cdot \mathbf{a}_2$$

and the atomic positions within the unit cell by

$$\boldsymbol{\rho}_j = n_1 \cdot \mathbf{b}_1 + n_2 \cdot \mathbf{b}_2.$$

Likewise it is appropriate to define a reciprocal lattice (i.e., in k-space) as

$$\mathbf{a}_i \cdot \mathbf{a}_j^* = 2 \cdot \pi \cdot \delta_{ij}.$$

Thus on a two-dimensional surface without any z-periodicity the points in real space are represented by rods that extend in the third dimension. The unit cell in reciprocal space is called the Brillouin zone. With this (under the assumption of single scattering only — the first order Born approximation) a distant detector observes a scattering amplitude

$$A(\mathbf{S}) = F \cdot \sum_i e^{i \cdot \mathbf{S} \cdot \mathbf{r}_i}$$

with the structure factor

$$F = \sum_j f_j \cdot e^{i \cdot \mathbf{S} \cdot \boldsymbol{\rho}_j}.$$

Therefore the diffraction intensity from a crystal of dimension $(N_1 \cdot \mathbf{a}_1 \times N_2 \cdot \mathbf{a}_2)$ can be written as

$$\begin{aligned} I(\mathbf{S}) &= A(\mathbf{S}) \cdot A^*(\mathbf{S}) \\ &= |F|^2 \cdot \frac{\sin^2(N_1 \cdot \mathbf{S} \cdot \mathbf{a}_1) \cdot \sin^2(N_2 \cdot \mathbf{S} \cdot \mathbf{a}_2)}{\sin^2(\mathbf{S} \cdot \mathbf{a}_1) \cdot \sin^2(\mathbf{S} \cdot \mathbf{a}_2)}. \end{aligned}$$

Clearly the maximum intensity is observed for the Laue conditions

$$\mathbf{S} = \mathbf{G}_{hkl},$$

i.e.,

$$\mathbf{S} \cdot \mathbf{a}_1 = 2 \cdot \pi \cdot h$$

$$\mathbf{S} \cdot \mathbf{a}_2 = 2 \cdot \pi \cdot k$$

for a two-dimensional array of scatterers.

A very popular tool to visualize these conditions is the Ewald sphere. The intersections of the sphere (of radius $|\mathbf{k}|$) with the rods of the two-dimensional reciprocal lattice then show the diffraction pattern. However, for several reasons the observed spots are not as sharp when using RHEED rather than with LEED, but more streaky:

- the absolute energy spread increases, leading to a higher blur of the sphere.
- the intersections are far from the normal,
- there is a certain modulation in the rods due to the periodicity in the third lattice dimension and the finite electron penetration depth.

Having this said, it looks like almost everything we can get from the use of RHEED is the information whether the desired surface has formed at all or not — by means of decyphering the observed pattern in search of the pattern corresponding to the unit cell of the desired film. Fortunately, this is not the case.

All the above considerations have been made under the assumption of infinitely extended, flat surface planes. However, the reality looks somewhat more grim. As direct-view techniques such as Scanning Tunneling Microscopy reveal, there is a

multitude of irregularities, the simplest ones of which can be addressed as follows in the limits of strictly kinetic approximation. The best approach to this would be to start with a homoepitactic two-level system. This means that at one time only two (successive) layers are exposed. It is obvious that in this case only the scattering from those two levels has to be considered — at least to first approximation. In addition, interference between the waves from the different levels will be observed. This results in a two-component profile, consisting of a diffuse background due to the existence of several layers and a sharp spike — the original spot — on top of it [4]:

$$I = 2 \cdot |f|^2 \cdot \theta \cdot (1 - \theta) \cdot [1 - \cos(\mathbf{q}_z \cdot \mathbf{c})] \\ + 2 \cdot \pi \cdot |f|^2 \delta(\mathbf{q})(1 - 2 \cdot \theta \cdot (1 - \theta) \cdot [1 - \cos(\mathbf{q}_z \cdot \mathbf{c})]).$$

It is clearly visible that the intensity varies considerably with $\mathbf{q}_z$, which in turn is, for a fixed system, determined by the incident angle. In general, this leads to distinguish in-phase condition

$$\mathbf{q}_z = \frac{2 \cdot n \cdot \pi}{\mathbf{c}}$$

and out-of-phase condition

$$\mathbf{q}_z = \frac{(2 \cdot n + 1) \cdot \pi}{\mathbf{c}}.$$

In the latter case, the central spike cannot be observed at $\theta = 0.5$ ML, while in the former the central spike does not change at all. For an increasing number of exposed layers the intensity of the central spike decreases dramatically, and basically can only be seen at the exact in-phase condition, if so at all.

In this — admittedly simplistic — view well-ordered growth can be characterized by pronounced oscillations, whose maxima do not decrease in intensity over time. Unfortunately, explanations to many problems are to date not very well developed,

for instance the influence of irregular steps — which might affect the form factor —, or the shift from heteroepitaxy to homoepitaxy (Ag/Ag(111) at higher coverages as opposed to Ag/Si(111) at zero coverage), and corrections from the application of dynamical scattering theory.

CHAPTER 2. EXPERIMENT

Our experiments were carried out in a ultrahigh vacuum chamber with a base pressure of about $5 \cdot 10^{-11}$ Torr. We used a Bayard-Alpert type nude ion gauge for pressure measurements. The system is equipped with two turbomolecular pumps, a quadrupole mass spectrometer, an MBE-cell, and the RHEED system (gun and screen).

The samples were cleaned by heating them resistively to temperatures in excess of $T = 1200^\circ\text{C}$, thus eliminating possible carbon contamination. Care was taken that during this process the pressure did not exceed $5 \cdot 10^{-9}$ Torr, since we made bad experiences in previous experiments where the pressure had risen higher. The Si(111) 7×7 pattern did not show any traces of contaminants after the cleaning.

The elevated temperature was controlled with a pyrometer. Temperatures below room temperature were effectively monitored with a Chromel-Alumel thermocouple, which was spot-welded to one of the tantalum clips that held the sample. Before attaching it we checked its functionality by immersing it in liquid nitrogen and comparing the thermovoltage with the standard table (Omega). We estimate the error in the thermocouple reading to ± 10 K, because it is not directly touching the sample. The process of cooling down to our working temperature of $T = 190$ K took four hours (i.e., the thermocouple reading leveled off after that time); it was done in the

same way every time.

During deposition, pictures of the RHEED pattern were taken in preprogrammed time intervals. The camera is a manual gain high-sensitivity camera. The effusion cell is closed with a shutter that is equipped with an extra hole. This hole is of a smaller area than the opening in the cell crucible itself. It is in this way possible to switch between two different flux rates very quickly — without any need to change the cell temperature and wait for a new thermal equilibrium to establish. The only — though minor — drawback is that flux changes made in this fashion are limited to two fluxes with a fixed ratio.

A range of flux rates was calibrated using a Leybold Inficon quartz crystal monitor. From the drop of the crystal resonance frequency (because of the added mass of the deposited material) over time it is possible to determine the deposition rate directly as

$$R(\text{ML} \cdot \text{s}^{-1}) = \frac{T}{d \cdot t},$$

with the monolayer spacing d — $d = 2.36 \text{ \AA}$ for Ag(111) —, deposition time t and thickness

$$T = \frac{N_q \cdot d_q}{\pi \cdot d_f \cdot f_c \cdot Z} \cdot \arctan\left(Z \cdot \tan\left(\pi \cdot \frac{f_i - f_f}{f_i}\right)\right)$$

(for details see [5]).

For the MBE-cell setting (11.0V, 3.4A) that was used in this case, the calibration curves are shown in figure 2.1. It is clearly to be seen that the rates are very stable. They amount to

(11.0V, 3.4A) with shutter hole	$F = 673^{-1} \text{ML} \cdot \text{s}^{-1}$
(11.0V, 3.4A) shutter open	$F = 118^{-1} \text{ML} \cdot \text{s}^{-1}$

Figure 2.1: Flux rate calibration with MBE-cell @ (11.0V, 3.4A).

CHAPTER 3. RESULTS

In the course of this thesis a number of low-temperature deposition runs were made at different flux rates. They show some remarkable features.

First, it is very noticeable that in all runs, with all fluxes, the overall intensity of the specular spot increases dramatically in the first phase of the respective deposition. In fact, one might say that this initial increase represents the very first phase. Very clearly though, this phase is not of the same length for each experiment, and the amount of deposited silver — as shown in the overall intensity curves — varies between $\theta = 0.2$ ML and $\theta = 0.4$ ML. A look at the specular spot profiles (Appendix) and their comparison at $t = 0$ s and at the onset of the oscillation reveals that throughout this phase not only the height of the central spike increases, but the FWHM decreases as well — or at least stays the same. No broadening can be observed.

The likely explanation of this phenomenon is that we unfortunately observed a relatively poor, i.e., rough, surface to start with, at least on an intermediate scale. This roughness was somewhat smoothened out with the deposition of the very first silver atoms.

In agreement with this explanation would also be the fact that the actual onset of oscillation is different for every run, as already mentioned — although the conditions

Figure 3.1: Overall specular spot intensities as a function of coverage of the (0914951) and (0915951) depositions.

(temperature and flux) are the same.

These observations to all the deposition experiments that are going to be discussed here.

The first deposition was made at $T = 190$ K at a flux rate $F = 673^{-1} \text{ML} \cdot \text{s}^{-1}$, i.e., the lower rate. It is shown (Figure 3.1) together with a deposition (0915951) at the higher flux $F = 118^{-1} \text{ML} \cdot \text{s}^{-1}$, but the same temperature.

The overall intensity starts after the initial phase with a nice decrease as expected from ordered growth, and an according increase again. However, following this maximum, we observe basically a monotonous intensity decay without the slightest hint whatsoever to subsequent oscillations. Furthermore, this visible oscillation is even

more prolonged than expected, i.e., it takes more than one monolayer silver deposited to complete one monolayer coverage. Also the maximum does not quite reach the initial intensity again.

Comparing these observations with the diffraction theory discussed in the preceding paragraph, a fast conclusion would be that we see all signs of multilayer growth. This appears to be the case at least for $\theta > 1.5$. Not quite as unambiguous as this statement sounds is the situation when studying the spot profiles. At this place, however, I should mention that all almost profiles suffer from a more or less pronounced feature which we are at the moment unable to explain. It is a spot that appears during deposition very close to the specular spot, and brightens throughout the deposition. It is a very singular phenomenon, and, being without a counterpart, we cannot put it into a certain diffraction pattern. Tentatively we ascribe it to a possible contamination of the sample — although we have no idea of what would cause it. This unwelcome fact necessitates some imagination. It appears strange, but one cannot neglect that the FWHM of the central spike increases from the start of the oscillation to its minimum, only to decrease again until the oscillation maximum. Additionally, the intensity drops and rises again.

What can we infer from this observation? First, we see the growth of relatively small islands, starting with a relatively well-defined two-layer system. Towards the completion of the layer however, the number of exposed layers, or surface roughness Δ , increases. The growing diffuse part of the spot shows that clearly. Second, the existing islands — at the close of the layer — are relatively large, so that we may infer a lower nucleation density at the start of the subsequent deposition. Therefore, we should not be surprised to find only multilayer growth at higher coverages, as the

central spike shows, which basically becomes buried in the diffuse part.

This gives rise to the question how a higher nucleation density — a higher number of relatively small islands — can be achieved, or produced, or preserved, since it seems in agreement with nucleation theory to promote a better two-dimensional growth. One way to achieve it is suggested by Rosenfeld *et al.* [6], who show that depositing a small amount of silver at low temperatures and proceeding with the deposition at substantially higher temperatures yields the desired effect.

In our experiment we tried to bypass the inherent problem of additional contamination of the interface during the crucial warm up. We utilized the shutter, thus being able to change the flux rate in a matter of seconds. In effect we expected to see the same result: enhanced growth when depositing first at a vastly larger flux rate $F = 118^{-1} \text{ML} \cdot \text{s}^{-1}$, resulting in a high nucleation density, followed by the original lower flux $F = 673^{-1} \text{ML} \cdot \text{s}^{-1}$.

The deposition in question, (0922951), accordingly shows the same characteristics as expected. From the onset of oscillation until its minimum, the height of the central spike decreases while its FWHM increases. In turn, towards the completion of the layer the FWHM decreases almost to its initial value, while the intensity increases. Remarkably, the spike – diffuse part ratio is in terms of improved two-dimensional growth better than the aforementioned run. Even at higher coverages, the central spike is still quite visible and not hidden in the diffuse part.

Thus so far our results agree with Rosenfeld's results and nucleation theory.

In addition to these depositions, we performed a deposition experiment (0926951) with reciprocal approach. We deposited first about half a monolayer silver at $F = 673^{-1} \text{ML} \cdot \text{s}^{-1}$, switching subsequently to the higher flux rate. Presumably

Figure 3.2: Overall specular spot intensities as a function of coverage of the (0922951) and (0926951) depositions.

this procedure does not work in the desired way, since theoretically there is no way to induce a high nucleation density with a low flux.

Surprisingly, this method works very well. The graph of the overall specular spot intensity of this particular deposition experiment scales extraordinarily well with one of the aforementioned experiments, as can be seen in Figure 3.2. Moreover, this particular graph approaches the ideal cosine-like intensity variation even better, while the saturation intensity maintains a higher level as well.

How does this compare with the specular spot profiles? Again, the height of the

spike reaches at completion of the oscillation the same value as at the onset. The FWHM behaves accordingly, also achieving the original value at completion of the oscillation again. Additionally, the diffuse part is appreciably low, even at higher coverages. Therefore the surface roughness δ must be relatively small, even at higher coverages. Presumably in this steady state regime at $\theta \geq 1.5$ we observe at growth similar to stepflow, however with a number of islands growing and extending their edges.

What is the best way to understand or explain these results? — The main obstacle is clearly the result of the (0926951) experiment. It is in a way even prohibited: if high nucleation density caused well-ordered growth, as detailed in [6], and perhaps as demonstrated by the (0922951) deposition, how can a high nucleation density be achieved by means of depositing with a low flux rate? How does a steady state at higher coverages (which clearly is attained) be explained?

In short: it seems that there is need to explain experimental results which are not covered by theory as of now.

One possible way may be to look at it in the light of the results of Roos *et al.* ([7]), who report flux independence of Ag/Si(111) growth. That would make it fairly simple and readily perceptible. However, there are two points that cannot be accounted for in this way: no statement is made as to what happened when the flux rate is changed during deposition, and secondly why not all depositions show the same tendency. In particular, if both flux rates yield the same result (behaviour), why cannot a similar intensity graph be recorded for the all-low-flux deposition (0914951)? If flux rates did not carry any influence it should be expected to scale also. Another, though rather hazy way of thinking of it would be that upon imping-

ing on the surface the adatoms have a certain characteristic time, during which they dissipate their excess energy — much like condensation in a finite, albeit short, time frame. This characteristic time was connected with the flux rate —, i.e., the mean time between arrival of adatoms. However, this interplay is strongly disturbed upon changing the flux rate in such a way that the growth is greatly enhanced.

CHAPTER 4. SUMMARY

In this thesis it could be shown that it is possible to achieve well-ordered growth at low temperatures when changing fluxes during the course of the deposition.

It could additionally be demonstrated that nucleation theory fails to predict or explain at least part of the results, in particular when deposition takes place at an initially low rate, with presumably a relatively low nucleation density, followed by a change to a high flux rate.

This points at an inherent lack in nucleation theory as used for example in [6].

Alternative explanations were presented based on flux-independent growth as reported by Roos *et al.* ([7]).

BIBLIOGRAPHY

- [1] K. Takayanagi, Y. Tanishiro, S. Takahashi, and M. Takahashi: *Structure Analysis of Si(111)7x7 Reconstructed Surface by Transmission Electron Diffraction. Surf. Sci.* **164** (1985) 367.
- [2] L. de Broglie: *A Tentative Theory of Light Quanta*. In *Philosophical Magazine and Journal of Science*, vol. 47, sixth series. London: Taylor and Francis, 1924.
- [3] M.G. Lagally: *Diffraction Techniques*. In *Methods of Experimental Physics, Vol.22: Solid State Physics: Surfaces*, R.L. Park, and M.G. Lagally (Eds.). Orlando, FL: Academic Press, 1985.
- [4] M.G. Lagally, D.E. Savage, and M.C. Tringides: *Diffraction from Disordered Surfaces: An Overview*. In *Reflection High-Energy Electron Diffraction and Reflection Electron Imaging of Surfaces*, P.K. Larsen, and P.J. Dobson (Eds.). New York, NY: Plenum Press, 1988.
- [5] *XTC Thin Film Thickness and Rate Monitor Technical Manual*. Leybold Inficon Inc., East Syracuse, NY, 1990

- [6] G. Rosenfeld, R. Servaty, C. Teichert, B. Poelsema, and G. Comsa: *Layer-by-Layer Growth of Ag on Ag(111) Induced by Enhanced Nucleation: A Model Study for Surfactant-Mediated Growth. Phys. Rev. Lett.* **71** (1993) 895.
- [7] K.R. Roos, and M.C. Tringides: *Low-temperature, flux-independent epitaxy in Ag/Si(111). Surf. Sci.* **302** (1994) 37.

APPENDIX SPOT PROFILES

For reasons of better comparability the profiles are chosen to represent the same deposition stages for every run:

- at the start of deposition
- at the onset of the first oscillation
- at the first minimum
- at the first maximum
- after deposition of several monolayers

0914951.00

0914951.02

0914951.08

0914951.17

0914951.80

0915951.01

0915951.05

0915951.12

0915951.60

0922951.01

09222951.02

0922951.19

0922951.50

0926951.22

0926951.48

0926951.75