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Abstract. It is argued that the growth morphology of ultrathin metal films should fluctuate as a function of film thickness due to the quantum size effect. To verify this, the specularly reflected intensity of x-rays, electrons and He atoms has been measured during the growth of a thin Pb layer on top of an Si(7 × 7) substrate at 100 K. Curiously enough, the expected variation is observed in the He atom and electron scattering data, but not in the x-ray reflectivity. Our explanation is that the differences in the heat of formation for successive atomic layers have a strong effect on the step density, but not on the occupancy of the layers at the low growth temperatures necessary to obtain layer-by-layer growth. This is backed up by the results of a Monte Carlo simulation.

1. Introduction

For ultrathin metal films quantization of the electronic energy levels becomes a measurable quantity and leads to the so-called quantum size effects (QSEs). Several experiments have shown their importance. For instance, if electrons are injected into or scattered by a thin film and the absorption, transmission or reflection of these electrons is measured as a function of layer thickness, characteristic modulations in intensity are observed [1–5]. Quite recently, the existence of the quantized levels in thin metal films was also proved by photoelectron spectroscopic experiments [6].

In the present paper we will concentrate on the influence of quantization on the growth mechanism of thin metal films. Hinch *et al* [7] were the first to suggest such an effect to explain their He atom scattering data collected during the low-temperature epitaxial growth of thin Pb layers on a Cu(111) substrate. At higher coverages, the separation between maxima in the specularly reflected intensity did not correspond to a single monolayer but to a double and occasionally even to a triple layer. It is shown that maxima are observed if the layer thickness satisfies the condition

$$d = nd_0 \cong m\lambda_F/2 \tag{1}$$

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as closely as possible, where n and m are integers, d_0 is the interlayer spacing and λ_F is the Fermi wavelength of Pb. This condition corresponds to a situation in which the wavefunction of an electron at the Fermi energy matches the thickness of the layer. Filling in actual numbers for Pb ($d_0 = 2.84$ Å, $\lambda_F = 3.66$ Å), condition (1) is approximately met every second monolayer as is indeed experimentally observed. Similar conditions have been previously used to explain the oscillations in the conductivity [3–5], the specular RHEED intensity oscillations [3–5], and the oscillations in the intensity of emitted photoelectrons [6] for Pb films grown on Si(111). Note that only in the case of the conductivity measurements is the wavelength to be used in (1) the Fermi wavelength. In the other cases λ_F has to be replaced by the wavelength of the incoming electron beam and the wavelength of the outgoing photo-electron, respectively. The helium atom scattering result is quite peculiar, because electrons are not involved in the atom scattering process in a direct way. In their explanation of the effect, Hinch *et al* [7] assumed that a film is most stable if its thickness satisfies condition (1), leading to the simultaneous growth of two or occasionally three layers. Jałochowski *et al*, on the other hand, expressed their doubts about such a double-layer growth mechanism [4]. In our opinion, a severe weakness of the model is that a strong temperature dependence of the growth morphology is predicted, in contradiction with experimental observations [5, 8]. Nevertheless, it seems to us that the basic idea of the model, i.e. that the energy of formation of a metal layer should depend on its thickness because of the QSE, must be correct. Consequently, some influence on the initial growth morphology of metal layers may be expected.

To elucidate the nature and extent of the relationship between the quantization of the electronic energy levels and the morphology of ultrathin metal films, we have made a detailed study of the initial stages of growth for the Pb–Si(111)(7 × 7) system by measuring the intensity oscillations of specularly reflected electrons, x-rays and He atoms under virtually identical conditions. The three types of diffraction technique are expected to yield quite different and complementary information due to the different probing depth and the different nature of the scattering process.

In the next section we will first examine whether a measurable effect of the QSE on the morphology and structure of a thin metal film is expected from a theoretical point of view. In sections 3 and 4 the experimental details and the results of the experiments will be presented, respectively. For a proper understanding of the results it turns out to be very helpful to simulate the growth process using a kinetic Monte Carlo model (section 5). The implications of the comparison between the experimental result and the simulated data are discussed in the final section.

2. Energy of formation of thin metal layers

One of the quantities which oscillates strongly with the layer thickness for ultrathin metal layers is the Fermi level. This is caused by the fact that the spacing between the electronic energy levels decreases with an increasing number of layers. Each time an additional level begins to be occupied the Fermi energy shows a cusplike structure. The easiest way to demonstrate the effect is to calculate the Fermi energy for noninteracting electrons in a square well potential [9]. As shown by Stratton [10], care should be taken to ensure conservation of charge by allowing for a small difference between the thickness of the slab and the width of the potential well. The calculation can be improved by including exchange and correlation effects in the density functional formalism [11]. The main difference is that the effective potential, which is kept constant in the square well model, is now calculated selfconsistently as well. From Schulte's calculations [11] it is evident that in fact the modulation of the Fermi

level is caused by a corresponding variation in the effective potential, whereas the variation in the kinetic energy with thickness is very small. This implies that the bulk contribution to the workfunction is small, but the surface contribution large. The explanation given by Schulte is that the wavefunction of each newly occupied level has a relatively large amplitude outside the surface. With increasing layer thickness, the strength of the surface dipole oscillates due to the periodic filling of new energy levels. This causes the average bulk potential to go up and down. Because the changes of the kinetic energy are small, the Fermi level rises and falls in parallel. As a result, the cohesive energy per electron will oscillate as well, because the average kinetic energy of an electron, which is part of the cohesive energy, is a notable fraction of the energy difference between the Fermi level and the bottom of the valence band [12]. The initial amplitude of the oscillation of the Fermi energy may be of the order of 1 eV according to Schulte. Therefore, the differences in cohesive energy for different layer thicknesses will be larger than kT up to rather thick films. Under equilibrium conditions, some layers will grow at the expense of others. This effect is the one-dimensional analogue of the formation of metal clusters with 'magic' numbers [13].

The above reasoning is the basis of the growth model proposed by Hinch *et al* [7]. However, we believe that this model cannot describe the low-temperature growth of metals properly, because the system is not allowed to thermally equilibrate during deposition. At temperatures as low as 100 K, it is very likely that surface diffusion is so slow that the atoms hardly move during the deposition of a monolayer. The distribution of the atoms is to a large extent determined by a random deposition process, modified by funnelling and transient motion of the atoms [14]. Under these circumstances, the influence of the QSE on the growth morphology is very much reduced because it is only felt by the system through the height of the barriers involved in lateral motion.

Although the information on the atomic distribution which is needed to determine the connection between the QSE and growth is in principle contained in the diffraction data we are going to present, it is not easily extracted. It is therefore necessary to compare the data with the results of a model calculation. We have chosen to simulate growth using a simple kinetic Monte Carlo calculation. An advantage of this approach is that not only the height distribution, but also the lateral distribution of the atoms within the toplayers, can be predicted. In view of the above discussion this aspect is important in connection with the interpretation of the He atom scattering data.

3. Experimental details

Three different diffraction experiments were carried out at three locations. The He atom diffraction experiment was performed at the University of Göttingen in Germany [8], the RHEED experiments were performed in Groningen in the Netherlands [15] and for the x-ray diffraction experiment use was made of the Synchrotron Radiation Source in Daresbury in the UK [16].

The preparation of the Si(111)(7 × 7) samples was carried out somewhat differently in the three experiments. For the atom diffraction, the sample was heated up to 1650 K, about 35 K below the melting point, and cooled down slowly afterwards. For the RHEED experiments a Shiraki cleaned sample was first heated at about 700 K overnight and then in an electron beam furnace up to about 1200 K for 5 min. In the case of the x-ray experiments the samples were flashed up to 1300 K and slowly cooled down to room temperature by electron beam heating. The different procedures may have resulted in a somewhat different roughness of the starting surface. However, in all cases carbide free and well developed

(7×7) diffraction patterns were obtained. The surfaces were also free of contamination within the detection limits of AES.

The growth experiments were performed in very much the same way. The clean Si(111)(7×7) samples were cooled down to about 100 K by clamping them to a liquid nitrogen cooled sample holder. The Pb was evaporated from a furnace at background pressures around 10^{-10} mbar. The growth rate was typically 1 ML min^{-1}.

4. Diffraction results

The main results have been summarized in figure 1. This shows the specularly reflected intensity of He atom, electron and x-ray beams during the growth of a Pb metal layer on top of an Si(111)(7×7) substrate at 100 K for scattering angles which approximately correspond to the anti-Bragg condition. The coverage scale was obtained by determining the deposition rate from the average oscillation period for coverages above 5 ML, assuming that this period corresponds to the deposition of a monolayer of bulk Pb. The latter assumption is only approximately true, because the oscillation period usually is somewhat smaller than 1 ML if the oscillations are damped due to roughening of the interface. From simulations of the growth process, however, the deviations are expected to be only a few per cent. The results are seen to be very different for the three techniques. As we will show below, this is caused by the different probing depth and the different scattering mechanism of the reflected particles.

The scattering cross section of x-rays is very small. X-rays, therefore penetrate deeply into the solid. One of the peculiarities of the x-ray results is the double-monolayer period of the oscillations. This is caused by a combination of two factors: (i) all the deposited Pb layers contribute equally to the diffraction intensity for the thin layers considered here and (ii) the contribution of the Si substrate is relatively small due to the large difference between the atomic scattering factors of Si and Pb. Consequently, in a good approximation the diffracted intensity may be calculated by considering a free-standing Pb film. Because beams reflected from two successive atomic layers destructively interfere at the anti-Bragg condition, the total intensity will be a maximum for an odd number of layers and minimum (zero for perfect layer-by-layer growth) for an even number of layers. This is precisely what is observed in the x-ray data.

In contrast to x-rays, thermal-energy atoms do not penetrate into the substrate at all. At the anti-Bragg condition the interference between the beams reflected from the topmost atoms is constructive for a complete layer and destructive for half a layer on top of a complete one. Oscillations with a single-monolayer period are expected in the case of perfect layer-by-layer growth. A very important additional factor in atom scattering is that the reflected intensity is strongly reduced by diffuse scattering from surface defects such as steps. All atoms scattered from a spot at the surface within a radius of about 10 Å are lost for the diffraction process. It is likely that the step density during low-temperature growth of metals is so high, even for nominally complete layers, that the reflected intensity is mainly determined by defect scattering. This assumption is borne out by the observation that very similar oscillation patterns are observed at the Bragg and anti-Bragg conditions [17]. Diffraction effects are apparently of minor importance and the observed oscillations are most likely mainly related to periodic variations in the step density during growth.

Because the mean free path of electrons in matter is small, beyond a few monolayers the influence of the substrate will be negligible and regular oscillations with a single-monolayer period are expected in the case of perfect layer-by-layer growth. However,
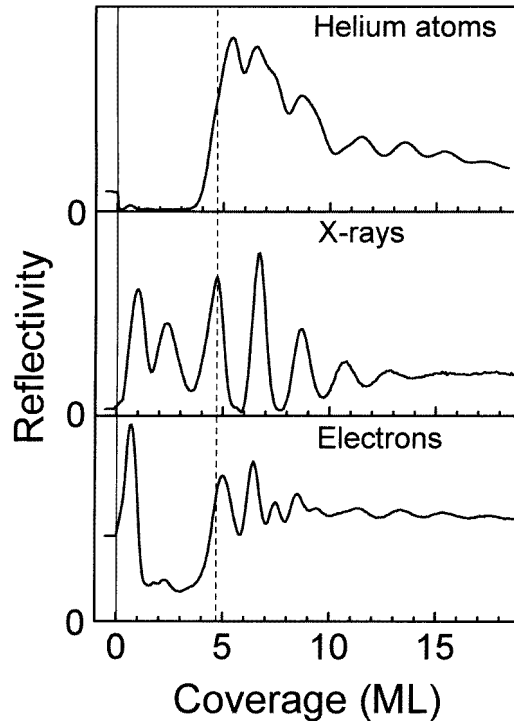
Figure 1. The intensity of specularly reflected beams of He atoms ($k = 5.3$ Å^{-1}, $\Theta_i = 45\,°$), electrons (15 kV, $\Theta_i = 1.24°$) and x-rays ($\lambda = 1$ Å, $\Theta_i = 5°$) during growth of a Pb film on top of an Si(111)(7 × 7) substrate at 100 K (k, λ and Θ_i are the wavevector, wavelength and angle of incidence of the incoming beam of particles, respectively).

multiple scattering makes quantification of the diffracted intensity a virtually impossible task. For that reason in the following we will concentrate on the x-ray and HAS results.

In the experimental data two clearly separated regions can be distinguished. Below 5 ML, growth is not a very regular process. It is layer-by-layer-like, because there are clear oscillations in the x-ray reflectivity, but the finite value of the x-ray intensity for an odd number of monolayer indicates that the width of the growth front is quite large. The step density at the surface must in fact be very high, because the atom scattering intensity is virtually zero. In addition the reduced spacing between the extrema in the x-ray data in this initial region suggests that the Pb layer is severely strained with respect to bulk Pb. At 5 ML the layer suddenly becomes much more ordered. The period of the oscillation is more regular and the amplitude of the oscillations larger. The x-ray intensity minima go down to almost zero showing that the surface width becomes quite small. Also the strong increase of the HAS intensity indicates the development of a rather flat surface. These observations confirm the proposition made previously [15] that at the critical thickness of 5 ML a transition takes place from a strained to a relaxed phase.

We now come to the main issue of this paper. The HAS data above 5 ML very clearly show a superperiod which is related to the Fermi wavelength. The pattern is in fact very similar to the one observed for Pb on Cu(111) [7], i.e. instead of the single-monolayer period expected for simple layer-by-layer growth, roughly a double-monolayer period is observed jumping from odd to even layers every 10 ML. Although it is a rather dominating feature in
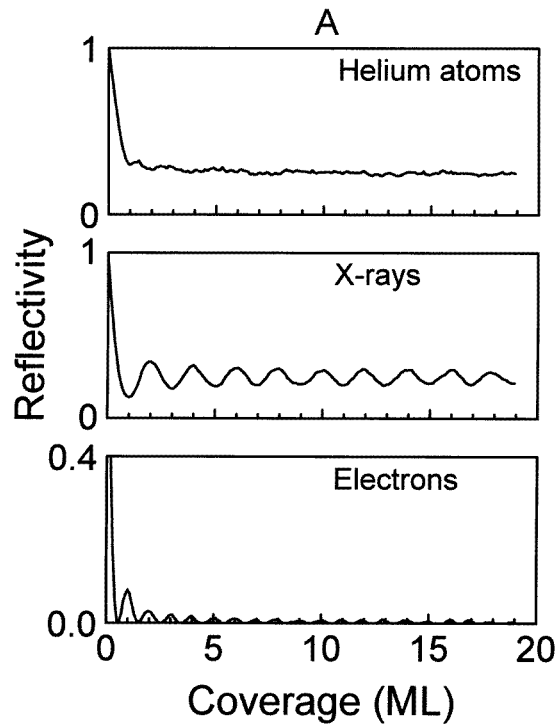
Figure 2. Specularly reflected intensity calculated using an MC growth model including funnelling only (case A).

HAS data, it does not seem to affect the x-ray data much. A growth mechanism as proposed by Hinch *et al* [7] would have a very strong effect on the x-ray data. During the period in which double-layer growth dominates, the intensity should stay more or less constant, either at a large value, if the top layer is odd, or a low value, if the toplayer is even. This is clearly not observed. We have also determined the partial coverages from scattering angle dependent ((0, 0)-rod) scans for 5, 6 and 7 ML [18]. Within 10–20% of a monolayer no evidence was found for such a double-layer growth mechanism. The double-layer growth hypothesis relies on the assumption that the HAS is predominantly a kinematic diffraction effect, because in that description the intensity is directly related to the fluctuation of the height distribution. As we have argued above, it is far more likely that the intensity is mainly determined by diffuse scattering from steps. The step density depends predominantly on the lateral distribution of the atoms. A possible explanation of the different influence of the QSE on the HAS and x-ray diffraction is that the variation of the surface energy has a very strong effect on the step density, but only a comparatively weak effect on the partial occupancy of successive layers. To verify this hypothesis we calculate the distribution of metal atoms in a simple kinetic MC growth model in the next section.

5. Kinetic Monte Carlo growth model

The model to simulate low-temperature growth of metals was kept as simple as possible. We have assumed that surface diffusion is relatively unimportant. The justification of this
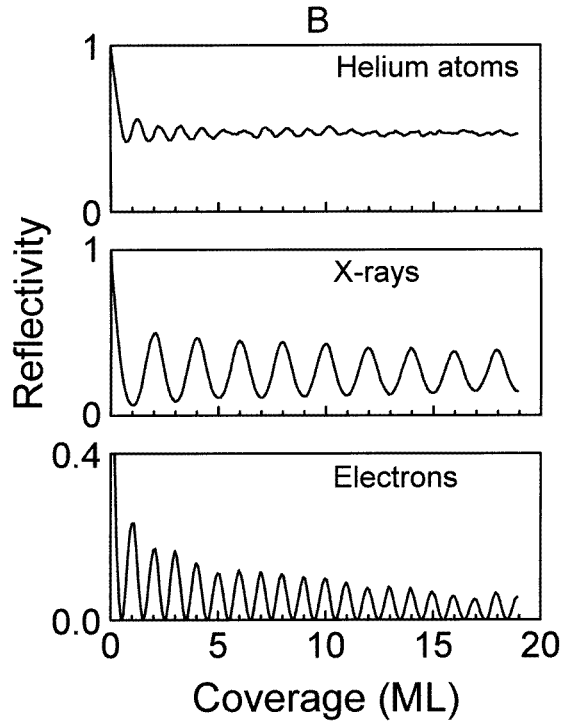
Figure 3. Specularly reflected intensity calculated using an MC growth model including funnelling and transient motion (case B).

assumption is based on the observation that the pattern of intensity oscillations for Pb on Si does not change very much on cooling from liquid nitrogen to liquid helium temperatures in either HAS [8] or RHEED [5]. Despite the absence of diffusion, layer-like growth is nevertheless possible at these very low temperatures, due to a number of low-temperature mechanisms, by which atoms are transferred from higher to lower levels [14].

Growth is started on a hexagonal lattice consisting of $3 \times 50 \times 50$ sites and subject to periodic boundary conditions. The atoms are dropped randomly onto this layer. If an atom is deposited at a site at which not all three atoms below are present, the atom is moved into one of the empty sites. This process, called funnelling, contributes to a layer-like growth because atoms are transferred from higher to lower levels. In addition, we will assume that the atoms which arrive near sites next to a step edge, i.e. sites for which there is at least one vacancy next to one of the three atoms below the site, may jump down to the lower level with finite probability. Physically the driving force of this process is the kinetic energy gained by the atom when it is attracted by the surface. A rough estimate of this energy can be obtained from the binding energy of a Pb atom, which is 2.03 eV in the bulk of a Pb crystal. Because a Pb adatom has only three nearest neighbours (nns) instead of twelve, the binding energy will be 0.5 eV if only nn interactions are assumed and relaxation effects are neglected. Part of this energy may be used to jump over the barrier at the step edge (transient mobility process) or to replace an edge atom, which is forced to jump to a neighbouring vacant site (replacement process). Although we believe that the replacement process is the most probable one, it is irrelevant for the MC model calculation which of these two mechanisms is actually happening. Because effectively the atom ends
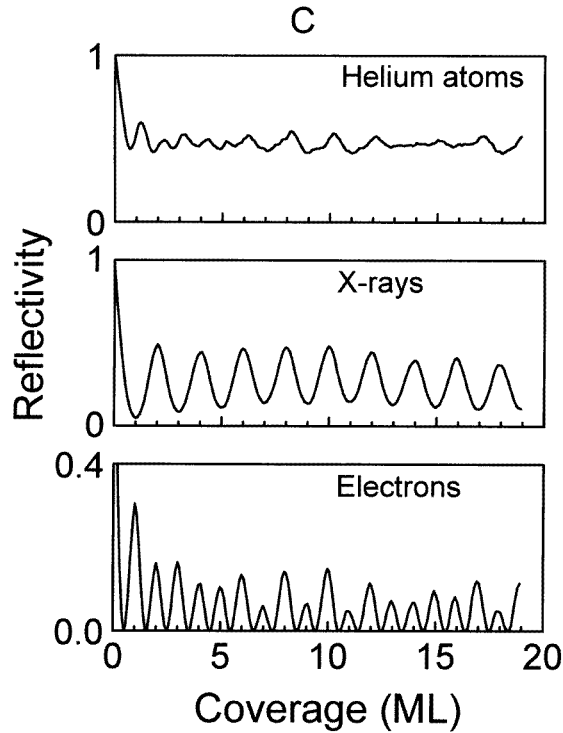
Figure 4. Specularly reflected intensity calculated using an MC growth model including funnelling and layer (QSE) dependent transient motion (case C).

up at a step or a kink instead of on top of a surface the total binding energy increases in the final configuration. This latter process is assumed to be sensitive to the QSE through layer thickness dependent barriers for atom transfer. Variations in the transfer probability to lower levels will undoubtedly have a strong effect on the growth morphology. The morphology found in the simulation is used to calculate the following quantities:

(i) the diffracted intensity of a free-standing metal film in the kinematic approximation at the anti-Bragg condition

$$I_X = \left(\sum_k (-1)^k \Theta_k \right)^2 \tag{2}$$

where Θ_k is the partial occupancy of layer k (the result will be compared with the x-ray diffraction data);

(ii) the diffracted intensity of the topmost atoms in the kinematic approximation at the anti-Bragg condition

$$I_e = \left(\sum_k (-1)^k [\Theta_{k-1} - \Theta_k] \right)^2 \tag{3}$$

(this quantity is assumed to be representative for the electron diffraction results);

(iii) the diffuse scattering of thermal energy He atoms by defects is assumed to be proportional to the step density. To keep the expression simple we ignore diffraction effects
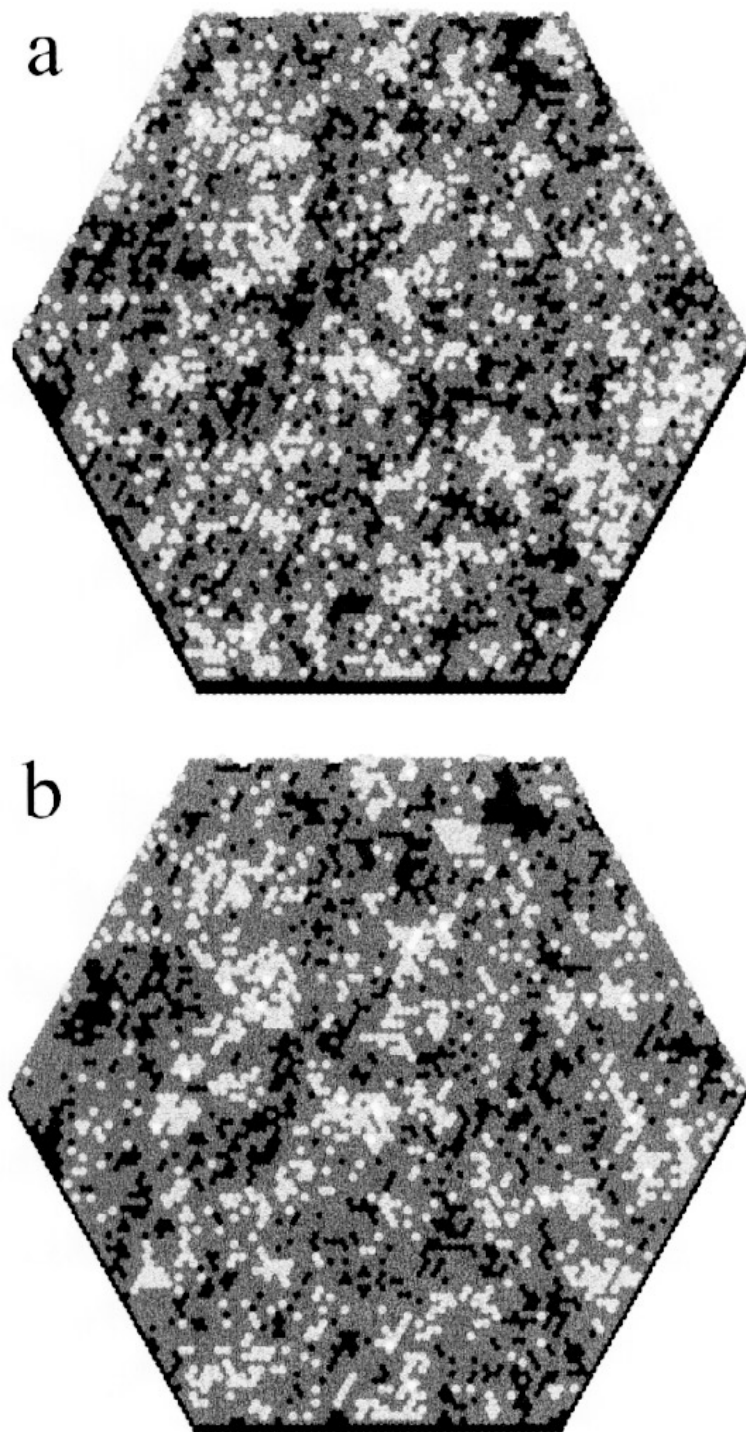
Figure 5. Atom distributions for the top layers of a metal film with a total coverage of (*a*) 10 ML and (*b*) 11 ML as obtained from a kinetic MC simulation of growth allowing for funnelling and QSE dependent transient mobility effects (case C).
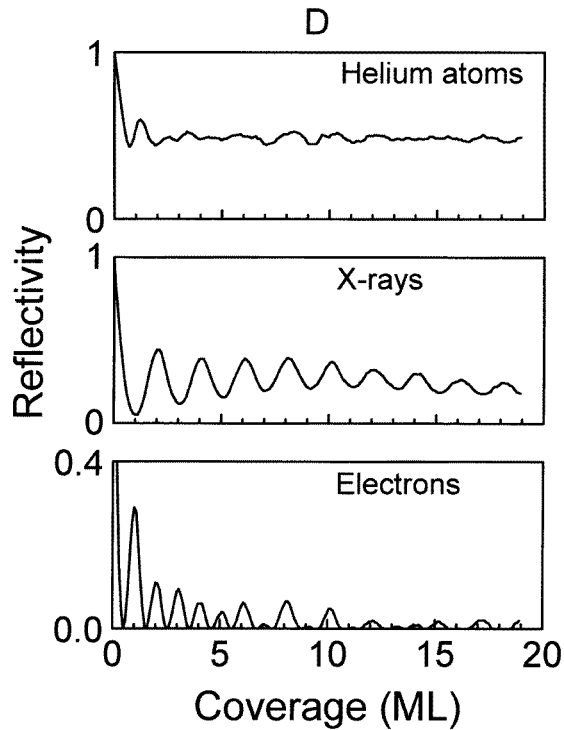
Figure 6. Specularly reflected intensity calculated using an MC growth model including funnelling, layer (QSE) dependent transient motion and thermal diffusion (case D).

by asuming that the in-phase diffraction condition is satisfied, so

$$I_{He} = 1 - \alpha \sum_k \rho_k. \tag{4}$$

The step density ρ_k is defined as the fraction of topmost atoms for which at least one nn site is vacant and α is a proportionality constant equal to the scattering cross section for small step densities. In the calculations presented below, α was taken to be unity. The basic difference between the second and last expression is that (3) depends on the height distribution in contrast to (4), which depends on the lateral distribution of atoms only.

The following cases have been considered:

Case A (figure 2). Random deposition including funnelling. As has been shown before [14] weak oscillations are observed in kinematic diffraction. The step density increases strongly up to 1 ML and remains constant afterwards. The atom scattering signal therefore is low and non-oscillating.

Case B (figure 3). Apart from funnelling, the step-edge transfer mechanism has been included with an equal transfer probability of 0.5 for all layers. The amplitude of the oscillations is increased and the damping reduced. Now also the step density oscillates. At the end of the growth run the partial occupancy of the layers is better than 99%, except for the last two layers. This result explains why rather perfect layer-by-layer growth is possible even at temperatures as low as 8 K [8, 5].

Case C. This is like case B, but now the transition probability has been chosen to depend on thickness d as

$$p = p_0 + p_1 \cos[2\pi(2d/\lambda_F)] \tag{5}$$

to simulate the effect of the QSE. Note that the transition probability p is largest if condition (1) is satisfied. The results of figure 4 were obtained for $p_0 = 0.5$ and $p_1 = 0.2$. Except for the first few monolayers, the step density varies with a double- or occasionally a triple-monolayer period. The kinematic scattering of the topmost atoms (the electron diffraction case) shows a similar modulation. The fluctuation in peak height now obviously reflects variations in the occupancies of the outer layers and not the distribution within the layers. In the XRD results, the only change with respect to case B is an additional low-amplitude, long-wavelength variation of the overall intensity. These variations are caused by small ($< 5\%$) fluctuations in the partial occupancies of the outermost layers, if the total coverage is an odd or even number of layers. The simulated patterns are qualitatively in agreement with the experimental results. In figure 5 the simulated distribution of atoms over the outer layers is depicted for total coverages of 10 and 11 ML. The higher step density in the 10 ML case is apparent from these pictures.

Case D (figure 6). This is like case C, but in addition some thermal diffusion has been introduced to see how the results change at finite temperature. It is seen that the main effect is that the oscillations become much more damped. The reason is that diffusion will increase the size of the islands and the distance between them, so that chances become larger that an atom stays on a terrace. As a consequence, the surface width will increase more rapidly with coverage at finite temperature. As in case C, the partial occupancies are quite similar for an odd and an even number of monolayers.

6. Discussion

The above analysis shows that at temperatures which are so low that normal surface diffusion cannot contribute to equilibrate the surface morphology rather perfect layer-by-layer growth is nevertheless possible by funnelling and transient mobility processes. Although we believe that in the early stages of growth the binding energy of metal atoms is very dependent on the QSE , the slow kinetics will prevent the formation of islands with magic thicknesses, i.e. thicknesses satisfying (1). The only source of a nonuniform distribution over the different levels is the transient mobility process, which allows the transfer of edge atoms to a lower level by a non-thermal excitation over a (layer dependent) barrier. The simulation shows that this process only causes minor deviations of the partial occupancies in favour of the more stable layers. The influence on the step density, however, is quite large. In our opinion, it is this effect which causes the dominant influence of the QSE on the He atom scattering data. The inherent assumption underlying this statement is that the oscillations of the reflected intensity of He atoms are mainly caused by periodic variations of the diffuse scattering by steps at the surface.

Basically our explanation is similar to the one given by Hinch *et al* [7], i.e. the growth of metal films is influenced by the thickness dependent cohesive energy of a thin metal layer due to the QSE. In their explanation, however, thermal equilibrium is assumed, leading to double- or triple-layer growth. In our view, the morphology of the surface depends on the QSE through transient mobility effects. The observation that the phenomena are virtually temperature independent [8, 5] is in agreement with this conclusion.

For completeness we have to consider an alternative explanation for the dominant contribution of the QSE to the HAS data, suggested by Jałochowski *et al* [4]. Due to

the QSE, the exponential tail of the electron density above the surface will vary with the thickness of the film. This will influence the position of the turning point for the scattered atoms and make the effective interlayer distance dependent on the number of layers. Using Schulte's calculations [9], however, one can estimate that this effect is too small to be of importance.

7. Conclusions

It was found that the relationship between QSE and the growth morpology of ultrathin films is a very subtle one. Following a suggestion made previously [7], it was assumed that the QSE enters the problem through a thickness dependent energy of formation of the metal layer. At temperatures at which surface diffusion is important this would lead to an oscillation in the height distribution function, having the QSE periodicity. At low growth temperatures, on the other hand, the only processes which are affected by the thickness dependent variations in the formation energy are transient mobility processes. Kinetic MC calculations show that layer dependent transient motion hardly affects the distribution over the layers, but does have a strong affect on the step density. This explains the apparently conflicting experimental result that the QSE dominates the growth oscillations measured using He atoms, but is virtually absent from those using an x-ray beam. These rather speculative conclusions could be verified by performing low-temperature STM studies on ultrathin metal layers.

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