Impact-driven effects in thin-film growth:
steering and transient mobility at the
Ag(110) surface

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Abstract
Low-energy atomic impacts on the Ag(110) surface are investigated by
molecular dynamics simulations based on reliable many-body semiempirical
potentials. Trajectory deflections (steering) caused by the atom–surface
interaction are observed, together with impact-following, transient-mobility
effects. Such processes are quantitatively analysed and their dependence on
the initial kinetic energy and on the impinging direction is discussed. A clear
influence of the surface anisotropy on both steering and transient mobility
effects is revealed by our simulations for the simple isolated-atom case and in
the submonolayer-growth regime. For the latter case, we illustrate how
steering and transient mobility affect the film morphology at the nanoscale.

1. Introduction

In 1989 Egelhoff and Jacob [1] reported a surprising result:
clear reflection high energy electron diffraction (RHEED)
oscillations were in fact detected by the authors while
monitoring metal on metal epitaxy on fcc(001) surfaces, at
temperatures as low as 77 K. This evidence was unexpected
since at temperatures at which adatom diffusion is supposed
to be totally frozen one would not have predicted to witness
ordered, layer by layer like growth. A possible explanation
was offered by the authors: the condensation energy gained
by bringing the impinging atom on the surface was somehow
transformed into kinetic energy of the adatom, allowing for fast
surface diffusion even when the substrate temperature was way
too low to overtake any of the relevant barriers. The intriguing
interpretation triggered several theoretical and computational
works aimed at shedding light on the phenomenon. Molecular
dynamics simulations demonstrated that the lateral transient
mobility acquired by the impinging atoms is usually limited
to a few atomic spacings, the condensation energy being
quickly distributed among the outermost substrate atoms [2, 3].
Nonetheless, a different kind of transient mobility was shown
to play a key role in determining a certain degree of ordering
in the growth. Newly deposited atoms hitting small three-
dimensional features (e.g. pyramids) eventually present on the
surface were shown to easily (the process being barrierless in
some cases) funnel down [2, 4] at the foot of the protrusion
until a stable site with the correct coordination was found. The
effect was demonstrated to produce a significant smoothing
of the growing film [4].

More recently a different kind of deposition-induced
mechanism was observed, leading, instead, to enhanced
roughening of the growing surface. A set of nice experimental
observations on copper growth on Cu(001) at low incidence
angles [5, 6] showed that the long-range interaction between
the incoming atoms and the surface can lead to a significant
increase in the roughness of the film and in the slope of the
typical mounds. Atomistic simulations [7, 8] confirmed this
steering effect, and showed that even under usual normal-
incidence conditions, the morphology of the film is affected
at the nanoscale. In fact, a 3D protrusion eventually formed
during the growth tends to attract incoming atoms, thus
enhancing its volume. Since a strong trajectory deflection
is possible only for incoming atoms with a small velocity
component in the direction perpendicular to the surface, steering is stronger when the initial kinetic energy and/or the incidence angle is smaller. Steering was also analysed, in a slightly different context, by Raible et al., nicely emphasizing the role played by this effect in inducing an instability during growth [9, 10]. Incoming-atom trajectory deflection has received further attention in the last few years, when a detailed theoretical analysis of the influence of both long-range and short-range atom–surface interaction on island shape and slope was presented [11–14], showing that the picture of funnelling-dominated low-temperature growth, due to steering, is not sufficient to capture the whole complexity of the physics involved.

In the present work we analyse transient mobility and steering effects in Ag/Ag(110). Our aim is to understand if the strong surface anisotropy can influence impact-following events, making it possible to tune some of the characteristics of the growing film at the nanoscale, and particularly the distribution of nanosized clusters on the surface, not only by changing the initial kinetic energy and the impact angle of the impinging atoms, but also their azimuthal angle. We recall that diffusion and growth at (110) fcc metal surfaces, including Ag(110), has been extensively investigated from both the experimental [15–24] and the theoretical point of view [25–35]. A detailed analysis of the impact-driven effects under different deposition conditions, however, is still lacking in the literature. Here we show that the knowledge of impact-driven effects may open new perspectives for thin-film nanostructuring.

In the next section we shall describe the methodology used for the present study, while in section 3 results concerning isolated-atom deposition are discussed. If the effect of both steering and transient mobility are already well evident and easily identified for this simple case, it is in section 4 that their influence on growth is more directly shown. Submonolayer-growth simulations are indeed analysed, together with the statistics of deposition-dependent distributions of adatoms, addimers, and larger one-dimensional islands. Finally, a critical discussion of the simulations is offered in section 5, while the summary of the results and our conclusions are reported in section 6.

2. Details of the calculations

The whole set of results presented here was obtained by using classical molecular dynamics (MD) simulations. The Ag–Ag interaction was described by Rosato–Guillopè–Legrand (RGL) semiempirical potentials [36], characterized by a many-body form derived in the framework of the second-moment approximation of the tight-binding scheme. Although the RGL potentials in the present parameterization (the complete set of parameters is reported, for example in [27]) were not fitted on surface properties, they were shown to yield results in terms of surface diffusivities in nice agreement with experiments and/or ab initio calculations [27, 31, 37] not only for Ag, but also for Cu and Au. Surface reconstructions are also nicely predicted [38]. The simulation cell used to represent the pristine Ag(110) surface in the calculations described in section 3, dealing with isolated-atom deposition only, was composed of 768 atoms (16 layers, 48 atoms per layer), while for the submonolayer-growth simulations of section 4 an initial slab of 4320 atoms (20 layers, 216 atoms per layer) was used. Periodic boundary conditions were applied in the surface plane. In the MD simulations all of the atoms were allowed to move freely apart from the three bottom layers, which were kept frozen to bulk positions. Additional atoms were deposited only at the upper slab termination. A top view of the smaller simulation cell is given in figure 1.

The presence of channels in the [010] direction is evident. As is usual in the description of such a surface, we shall call in-channel (or, simply, x) the [010] direction and cross-channel (y) the [001] one. Deposition of additional atoms and system equilibration was simulated using the following procedure. The three layers directly above the three frozen ones were coupled to a Langevin thermostat (we used a friction term $\eta = 5 \times 10^{12} \text{ s}^{-1}$) imposing a desired temperature, while the dynamics of all the other atoms was simulated by using a standard velocity Verlet algorithm. The time step was set to 5 fs, a value that guarantees satisfactory energy conservation (in the absence of the thermostat). In the single-atom deposition simulations the slab was first equilibrated for 3 ps. A new atom with the desired impact parameters (kinetic energy, normal and azimuthal impact angles) was then introduced high above (right outside the cutoff radius) the surface. Due to the low impact-energy values considered here, 5 ps of evolution turned out to be sufficient to bring the atom to the surface, to have its excess energy dissipated into the simulation cell, and to equilibrate the whole system. In the submonolayer-growth simulation, after the first atom was deposited, additional ones were added one by one, with a rate of about 100 layers s$^{-1}$ (1 atom every 10 ps). Several independent simulations were carried out in order to collect a significant set of statistics. For post-processing and analysis purposes, right before inserting a new atom, configurations were saved and optimized using a standard steepest-descent algorithm. Further details of the simulations, and in particular on initial atom positioning, are given in the following two sections.
3. Results: isolated-atom deposition

In order to investigate steering and transient mobility effects we started by analysing the simplest case, i.e. the deposition of a single atom on the clean Ag(110) surface. For the normal-incidence case, the overall effect of trajectory deflections can be monitored by simply running several independent simulations where the initial impinging-atom (x, y) coordinates are randomly chosen within the surface unit cell, while the height z above the surface is kept fixed at the cutoff radius value (r = 5 Å with the present potentials’ parameterization). The spread of the atom positions after the impact across and out of the unit cell are then analysed and related to the deflections or to other atomic-scale mechanisms directly triggered by the impact. In the most general case where the impact is not normal and the azimuthal angle is different from zero, the (x, y) grid of starting coordinates, instead, was chosen over a displaced unit cell (the z coordinate, again, corresponding to the cutoff radius), built along the initial direction of impact. The procedure is schematically shown in figure 2.

This choice allowed us to directly compare the normal and out-of-normal deposition cases in terms of the final atomic positions, keeping the same surface unit cell as reference. Let us now analyse the results, which were obtained for a substrate temperature T = 100 K. We shall comment further on the meaning of the substrate temperature in our MD simulations in section 5. We performed simulations at normal and grazing (α = 12.5° with respect to figure 2) incidence, both along the in-channel and cross-channel directions, with an initial kinetic energy of 0.1 and 1.0 eV. In each case we performed 2000 MD runs, and analysed the final, optimized position of the impinging atom, gathering statistical data on the frequency of the various possible configurations; moreover, exploiting the translational symmetry of the clean surface (if a trajectory starting at (x, y) ends up on the right-hand neighbour of the target cell, it would have on average ended up on the target cell if it had started at (x - a, y), where a is the lattice parameter) one can calculate the capture area of a cell, i.e. the function p(x, y) returning the probability that a trajectory ‘geometrically aimed’ at the position (x, y) with respect to the centre of a given cell will land up at the equilibrium position over that cell. One could estimate p(x, y) by dividing the surface with a dense mesh, and counting how many of the trajectories pointed to a given interval (x_i, x_{i+1}) x (y_j, y_{j+1}) + 1) finish over the target cell; the number of trajectories one should accumulate to obtain smooth results over a fine grid is enormous: we have instead calculated

\[ p(x) \approx \frac{1}{\sum_{i} \sum_{j} \delta(x - x_i) \delta(y - y_j)} \]

where the delta functions have been replaced by normalized Gaussians

\[ g_{\sigma}(x) = \frac{1}{\sigma \sqrt{2\pi}} e^{-x^2/\sigma^2} \]

with the width \( \sigma = 0.3 \) Å, chosen in order to obtain a smooth p(x). Although the resulting capture probability is heavily smeared and a quantitative analysis would require more intensive sampling, the qualitative features of p(x) are conserved, and they highlight geometrical effects responsible for the capture processes. The normal-incidence simulations (figure 3) show no surprises: most of the trajectories finish with the impinging atom in the equilibrium position of the target cell.

Trajectories aimed on the borders of the target cell may occasionally get captured on the nearest neighbouring cell, and there is a small probability for exchange processes, with the impinging atom taking the place of a surface atom, which becomes an adatom in the nearby row; as clearly shown in the contour plots in figure 3, there is a well defined zone for which the falling atom has a non-vanishing probability (reaching 10% for the 1.0 eV case) to ‘bounce’ into the nearby cell on the same row. For the exchange and rebound processes we may speak of transient mobility, because the impact energy activates...
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The cross-channel grazing incidence results are depicted in figure 6; here we see again steering effects that cause the impinging atom to fall systematically short: since the barriers for cross-channel diffusion are much higher than those for in-channel diffusion, the transient mobility effects are here much less evident than in the previous case, and the differences between the two initial kinetic energies are negligible, but for a slight enhancement of exchange event probability.

4. Results: submonolayer growth

If the single-atom case already revealed non-trivial impact-following events, it is natural to predict that after one or more adatoms are already present on the surface, such events become more pronounced and complex at the same time. Indeed, even a single adatom can be seen as a small protrusion extending out of the surface and, as such, steering trajectories of newly incoming material towards it. At the same time, the presence of an adatom within the channel can limit the range of transient-mobility diffusion. In order to look at similar effects, in this section we analyse simulations where, every 10 ps, a new atom is deposited on the surface until a 0.5 coverage is reached. To allow for some degrees of relaxation (see section 5), the substrate temperature was set to $T = 300$ K. We performed simulations for the same six sets of impact parameters used in section 3, but we selected for further analysis only the two in-channel grazing configurations and the 0.1 eV normal-incidence ones, because they better display the combined effects of steering and transient mobility. In order to single out the effects of steering on growth, we compare the results of our present MD simulations with those obtained by a kinetic Monte Carlo (KMC) model [32] at low temperature (100 K and below). The KMC model incorporates all diffusive processes and random deposition with downward funnelling, but does not take into account the steering.

For each set of parameters we performed 30 independent simulations up to 0.5 coverage, and we postprocessed the optimized structures after every impact to estimate some averages, representative of the growing layer morphology, such as the average number of $n$-islands at $\theta$ coverage, $a_n(\theta)$ or simply the average length of islands,

$$l(\theta) = \sum_n n a_n(\theta) / \sum_n a_n(\theta).$$
\[ T_1 0 0 K \]

channel 1.0 eV

A

channel 0.1 eV

A

A

A

A

3.0

4.0

Figure 6. Figures showing the final position of the impinging atom ((a) and (c)) and the capture probability function ((b) and (d)) for cross-channel grazing incidence (\( \alpha = 12.5^\circ \), along x) simulations; impact energies are 0.1 eV ((a) and (b)) and at 1.0 eV ((c) and (d)). The percentage of the trajectories ending up at each site is indicated, and \( p(x, y) \) is drawn as a contour plot as in figure 3. The target unit cell is three cells to the top ((a) and (c)) or to the bottom ((b) and (d)) of the reference cell, marked with a dashed outline.

Figure 7. Average length of islands as a function of the coverage; results are averaged over 30 independent simulations.

\( I(\theta) \) is plotted in figure 7 and \( a_n(\theta) \) from single adatoms up to 5-islands are reported in figure 8; the film as grown under the different conditions has quite different structures: the normal-incidence case compares very well with the KMC model, but for the tendency to form three-dimensional structures, which is enhanced in the MD simulations.

This shows that steering is effective in increasing the surface roughness at normal-incidence deposition, similarly to what was observed in [8] for the (001) surface. The average island length is 30% longer for 1.0 eV in-channel grazing incidence, while the 0.1 eV in-channel case is positioned halfway; in fact, \( a_n(\theta) \) is a much more sensitive index of the film structure, and displays for example numerous \( \geq 4 \) islands in the in-channel 1.0 eV configuration, high abundance of dimers and tendency towards 3D growth in the 0.1 eV in-channel case. Explanation for this variety in the in-channel trajectories again requires the mutual influence of transient mobility and steering: grazing incidence causes the impinging atom to interact for a longer distance with the surface, feeling the effects of already deposited atoms, while momentum parallel to the channels will determine if the impinging atom will be steered to fall right beside another adatom (this explaining the high concentration of dimers in the 0.1 eV case) or diffuse into the channel until it collides with a growing island (this explaining the longer islands registered at 1.0 eV).

Our results show that surface roughness is smaller for high-energy grazing incidence than for low-energy grazing incidence. This observation can be ascribed to two different phenomena: three-dimensional growth is partly due to overhanging-site occupation (see figure 9), corresponding to shallow minima from which the system can escape with the aid of transient mobility; moreover, at high impact energy atoms landing on short islands have sufficient transient mobility to reach the border of the island, and eventually surmount the low in-channel Ehrlich–Schwoebel barrier [32] to fall down on the
lower terrace, while at 0.1 eV two adjacent dimers in parallel channels are sufficient to trap the impinging atom on the upper layer.

A careful calibration of the impact angle and energy would probably permit one to fine-tune the morphology, enhancing or depressing selectively the presence of the various types of islands. The possibility to tune the three-dimensional growth is also promising as it should have an influence on the defectivity of the film.

5. Critical discussion: simulated substrate temperature

MD simulations of Ag/Ag(110) growth in the submonolayer regime were carried out at a substrate temperature $T = 300$ K. This temperature value should not be regarded as representative of an actual experimental temperature. As explained below, the present simulations are an attempt to model some features of growth at very low temperatures. From the theoretical point of view there are two main phenomena which need to be simulated when trying to model crystal growth: deposition and diffusion between subsequent depositions. While the atom–surface impact can be modelled directly by MD, since the time scale involved is of only a few picoseconds, the real challenge is to describe the evolution of the system before the next impinging-atom landing. Here the difference between MD accessible time scales and the experimental ones is huge: real deposition rates can be slower than 1 ML s$^{-1}$, while MD ones are closer to $10^7$–$10^9$ ML s$^{-1}$ (depending on the system size). Only if the temperature is so low that virtually no activated events are possible between subsequent depositions, is the morphology of the growing film directly determined by the sequence of fast impacts, since atoms remain frozen to the final position reached after the impact energy is dissipated. For the present system, the barrier for adatom diffusion along the fast, in-channel direction was calculated to be $\sim 0.28$ eV [31]. Dimers and chains diffuse much more slowly (barriers of 0.45 eV or more). Estimating the average residence time $\tau$ in a given site by the Arrhenius relation

$$\tau = \frac{1}{v_0} \exp \left( \frac{E}{k_B T} \right),$$

where $E$ is the activation energy of the mechanism bringing the system out of the site and $k_B$ is the Boltzmann constant, and by setting the frequency prefactor $v_0$ equal to the standard value $v_0 = 10^{13}$ s$^{-1}$, we can easily estimate that for $T \lesssim 100$ K, $\tau \gtrsim 10$ s, a value for which surface diffusion can be declared frozen (unless extremely slow-deposition experiments are carried out). In this sense, our submonolayer-growth simulations should be regarded as representative of real substrate temperatures lower than 100 K. This is confirmed by the comparison with the KMC simulations [32], which show that all diffusive processes are practically frozen at 100 K and below.

However, when we tried to use in the simulations a temperature comparable with this estimated threshold, the ultra-fast deposition rate occasionally induced the formation of artificial metastable structures (a problem which did not appear in the single-adatom deposition case), with the whole system trapped in ultra-shallow minima which would suddenly disappear if enough time were available for it to evolve. It was sufficient to raise the temperature to $T = 300$ K in order to strongly reduce this problem, without introducing any real diffusive event$^4$. Let us emphasize once again that a realistic modelling of growth at $T = 300$ K requires one to leave the simple MD approach. Ideally, one should introduce the impact-driven mechanisms presented here within a simulation scheme able to reach extended time scales. Kinetic Monte Carlo, or accelerated MD techniques [39] could be the right choice for a significant extension of our work. In spite of the limitations illustrated above, we stress that all of the observations on the effects of steering and transient mobility, and on their dependence on deposition parameters remain valid, since the whole set of simulations was run under the same high-rate conditions. Also, we would like to stress that the actual effect of steering in real Ag(110) is expected to be stronger than the one reported here, due to the limitations in trajectory deflection induced by the relatively short cutoff used in the simulations.

6. Conclusions

In this paper we have analysed the effects of steering and transient mobility on deposition and low-temperature

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Figure 9. Snapshots of the surface at $\theta = 0.5$ coverage, representative of the layer morphology. The images represent some of the outcomes of our simulation, chosen for the best compliance with the average island size distribution. (a) is from a normal incidence, 0.1 eV simulation, (b) and (c) from grazing, in-channel simulations, respectively at 0.1 and 1.0 eV. The different shades code the island lengths, and arrows point to overhanging sites.

(This figure is in colour only in the electronic version)
submonolayer growth of Ag/Ag(110). In general, both steering and transient mobility have significant effects on the morphology of nanosize aggregates on the surface. By tuning the initial energy and the angle of incidence, one may control some features of surface nanostructuring.

For deposition at normal incidence on a flat surface, the simulations have not revealed any significant transient mobility of the adatom, which stops after impacting the target cell in about 90% of cases, even when the initial energy is 1 eV. The incoming kinetic energy is dissipated in the substrate, without giving rise to any directed motion in the surface plane. In contrast, for grazing incidence along the in-channel direction, there is an interplay between steering and transient mobility, which is now possible because the initial velocity already has a component on the surface plane. Due to steering, the incoming atom tends to fall shorter with respect to the target cell, while transient mobility has the opposite effect, pushing the atom to travel further by a few hopping events that occur before thermalization.

Steering and transient mobility also have significant effects on submonolayer growth. Normal incidence deposition causes some increase of the roughness of the film, because incoming atoms are attracted by already existing protrusions [7]. However, this increased roughness effect is much more important when depositing at grazing incidence with low initial kinetic energy. In this case, the incoming atom feels a much stronger steering effect, but without presenting significant transient mobility after impact. This enhances the probability of forming dimers and small aggregates, on which a second layer can nucleate. For high-energy grazing incidence, the effects of transient mobility come into play. Now, longer chains are formed on the surface at the expense of dimers. Moreover, the roughness decreases because there is a smaller probability of nucleating a second layer, due to transient mobility causing interlayer downward processes.

Our results have thus shown the possibility of tuning growth morphologies by changing the initial energy and the angle of incidence of incoming atoms already in the submonolayer regime. We expect that these effects would be even more important in multilayer growth, especially for grazing incidence deposition, due to the appearance of shadowing effects besides steering and transient mobility. Work is in progress on this subject.

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