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# Simulation of multilayer homoepitaxial growth on Cu (100) surface*

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The processes of multilayer thin Cu films grown on Cu (100) surfaces at elevated temperature (250–400 K) are simulated by mean of kinetic Monte Carlo (KMC) method, where the realistic growth model and physical parameters are used. The effects of small island (dimer and trimer) diffusion, edge diffusion along the islands, exchange of the adatom with an atom in the existing island, as well as mass transport between interlayers are included in the simulation model. Emphasis is placed on revealing the influence of the Ehrlich–Schwoebel (ES) barrier on growth mode and morphology during multilayer thin film growth. We present numerical evidence that the ES barrier does exist for the Cu/Cu(100) system and an ES barrier $E_{\rm B} > 0.125$ eV is estimated from a comparison of the KMC simulation with the realistic experimental images. The transitions of growth modes with growth conditions and the influence of exchange barrier on growth mode are also investigated.

**Keywords:** growth mode, ES barrier, multilayer growth, kinetic Monte Carlo simulation
**PACC:** 6855, 6840, 6890

## 1. Introduction

In recent years, much work has been devoted to describe the submonolayer (two-dimensional, 2D) growth of thin film, because of its importance to the development of subsequent film morphology and properties and the potential application in devices.[1−3] Apart from the experimental study, such as scanning tunnelling microscopy (STM), atomic force microscopy (AFM) observation, modelling and simulation of the island growth in epitaxy provide an effective tool to understand and manipulate epitaxial processes.[4−11] However, few works are presented to simulate the multilayer (three-dimensional, 3D) growth.[12−16] In fact, multilayer growth is common in the realistic experiments and the growth processes are more complex than submonolayer growth. Amar and Family presented the results of a realistic model for multilayer growth which includes a finite barrier to interlayer diffusion. A method for determining the Ehrlich-Schwoebel (ES) barrier based on a comparison of simulation with experimental results has been discussed.[17] On the other hand, Cu/Cu(100) provides a simple square lattice system, an ideal case for testing existing models without extra complexity, and its many aspects have been investigated.[18−24] Durr *et al* have determined the island morphologies and island separations from the two-dimensional diffraction patterns measured by high-resolution low-energy electron diffraction (LEED) during sub-monolayer homoepitaxy on the Cu(100) surface, from which the energy barrier for adatom diffusion and the binding energy for small islands were determined.[19] Zuo *et al* have given the scaling behaviour in the steady-state and island coalescence regimes of the Cu/Cu(100) system.[20] The evolution of mound morphology in reversible homoepitaxy on Cu(100) was also studied via spot profile analysis (SPA) LEED and STM.[24] However, details of multilayer Cu thin film growth on Cu(100) surface, dependence of growth mode on growth condition, especially the physical origin of the ES barrier for the Cu/Cu(100) system, are not fully understood and need further investigation. The value of ES barrier for the Cu/Cu(100) system is not yet obtained in previous works.[25−28]

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In this paper, a comprehensive kinetic Monte Carlo simulation model is presented to investigate the multilayer copper thin film growth on Cu (100) surface, where the effects of small island diffusion, edge diffusion along the islands, exchange of the adatom with an atom in the existing island, as well as mass transport between interlayers are studied. We concentrate on revealing the influence of the ES barrier on the growth mode during multilayer thin film formation and giving the evidence for the existence of the ES barrier in the Cu/Cu(100) system. Dependence of growth modes on surface temperature is also investigated.

## 2. Simulation model

The simulation model is described as follows:

1) Deposition. Monomers are deposited at randomly chosen positions of the square Cu (100) flat substrate with a flux $F$ (Monolayer/second, i. e. ML/s).

2) Terrace diffusion. The deposited monomers, dimers and trimers are allowed to diffuse on terrace with diffusion rate (Arrhenius form):

$$D = D_0 \exp\left\{-\left[E_{\rm d} + 0.04(i-1)\right]/k_{\rm B}T\right\}, \quad (i = 1, 2, 3), \tag{1}$$

where $i$ is the critical island size, $E_{\rm d}$ is the activation energy for monomers (for Cu/Cu(001), $E_{\rm d}$=0.42 eV[19]), $T$ is temperature and $k_{\rm B}$ is the Boltzmann constant. We assume that all the monomers, dimers and trimers diffuse with the same prefactor $D_0$=$10^{13}$ $\rm s^{-1}$.

3) Edge diffusion. In order to take into account island relaxation, the rate of diffusion for atoms along the edge of an island is also included by assuming that the rate of edge-diffusion is given by:

$$D_{\rm ed} = D_0 \exp(-E_{\rm ed}/k_{\rm B}T), \tag{2}$$

where $E_{\rm ed}$ is the edge-diffusion barrier ($E_{\rm ed}$ = 0.1 eV[19]).

4) Exchange diffusion. The adatom is allowed to replace an atom in the existing island with a rate

$$D_{\rm ex} = D_0 \exp(-E_{\rm ex}/k_{\rm B}T), \tag{3}$$

where $E_{\rm ex}$ is the exchange barrier ($E_{\rm ex}$ =0.20 eV [19]). It implies the growth of Cu/Cu(100) is reversible at not very low temperature.

5) Interlayer diffusion. When monomers diffuse to a step, they will attempt to go down the step with an extra step-barrier energy$E_{\rm B}$, i.e. the ES barrier[29,30] and the rate of interlayer diffusion is given by

$$D_{\rm B} = D_0 \exp(-E_{\rm B}/k_{\rm B}T). \tag{4}$$

The ES barrier is defined as the difference between the energy barrier for an adatom to hop from a terrace to the layer below and the energy barrier for diffusion on a flat terrace. The existence of this barrier has been shown to lead to a morphological instability.[31] Unfortunately, the ES barrier is still not known for a wide variety of systems, and is difficult to calculate from first principles.[32−34] We have put the ES barrier as a variable parameter appearing in the hopping rate in Eq.(4), for example in Ref.[35], where the authors successfully simulated the re-entrant layer-by-layer growth of Pt/Pt(111).

The critical island size is set to be $i$=3, i. e. islands having more than three atoms will not move any longer and form stable nuclei. All of the dimers or trimers will stick to an island with a unity probability and become parts of the island when they reach a nearest-neighbouring site of the existing island, while the monomers will diffuse along the island edge with a rate $D_{\rm ed}$ or exchange with an atom of the island with another rate $D_{\rm ex}$. The re-evaporation effect is negligible at not very high temperature. The periodic boundary condition is set up when mobile particles diffuse to the boundaries.

## 3. Results and discussions

Figure 1 shows the typical growth morphologies of 8.3 MLs Cu growth on Cu(001) surface, where the deposition flux $F$= 0.0208 ML/s, temperature $T$=299 K and the substrate size is set to be 100×100 lattice. From Figs.(a) to (d), the ES barrier is set to be 0, 0.075, 0.175 and 0.25 eV, respectively. If no ES barrier is considered, the growth mode is a perfect layer-by-layer growth. In this case, the deposited atoms diffuse thermally over the surface until they are captured by islands already present on the surface, or by other adatoms, in which case a new island may be formed. Adatoms which are deposited on an island can jump off the island easily and are incorporated at the island edge. Large islands coalesce, thereby forming a flat layer. A new layer starts to grow only after the underlying layer is completed. As a result, the growth mode performs a perfect layer-by-layer growth. On the other hand, for Cu atoms on Cu(100) surface the mobility along the island edge is faster than

on terraces ($E_{ed} < E_d$), so the islands tend to have compact and square-like shapes, as seen in Fig.1(a). When the ES barrier is increased to 0.075 eV, a few nucleation may occur on top of already existing islands before completion of the underlying layer. The growth mode is closely to layer-by-layer growth, as seen in Fig.1(b). With the increase of the ES barrier, the additional energy barrier may be so high that interlayer mass transport becomes difficult. More and more atoms will stay in the level where they were deposited originally. The transition of growth mode from layer-by-layer growth to 3D growth occurred, as shown in Figs.1(c) and (d). Meanwhile, the homoepitaxial experiments of Cu/Cu(100) under the same growth conditions show a complete 3D growth mode.[24] The 3D growth phenomenon in realistic experiments is evidence for the existence of the ES barrier during multilayer Cu growth on Cu(100) surface. Three-dimensional snapshots of close to layer-by-layer and 3D growth modes are shown in Fig.2, where the coverage is 10.3 MLs, $T$=299 K, $F$=0.0208 ML/s and the ES barrier is 0.075 and 0.25 eV, respectively. Pictures shown in Fig.1(d) and Fig.2(b) resemble the STM images of Cu/Cu(100) closely.[24]

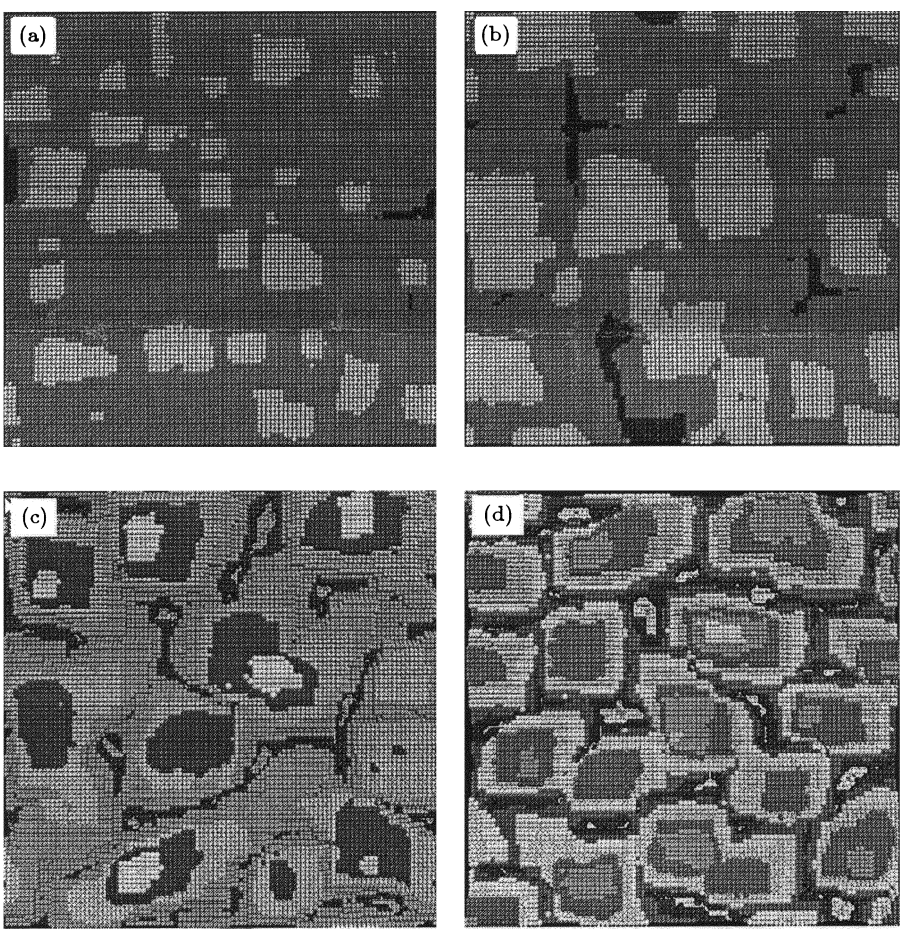



**Fig.1.** Morphologies obtained from KMC simulation at $T$=299 K, $F$=0.0208 ML/s and coverage $\theta$=8.3 MLs. From (a) to (d), the ES barrier is set to be 0, 0.075, 0.175 and 0.25 eV, respectively.

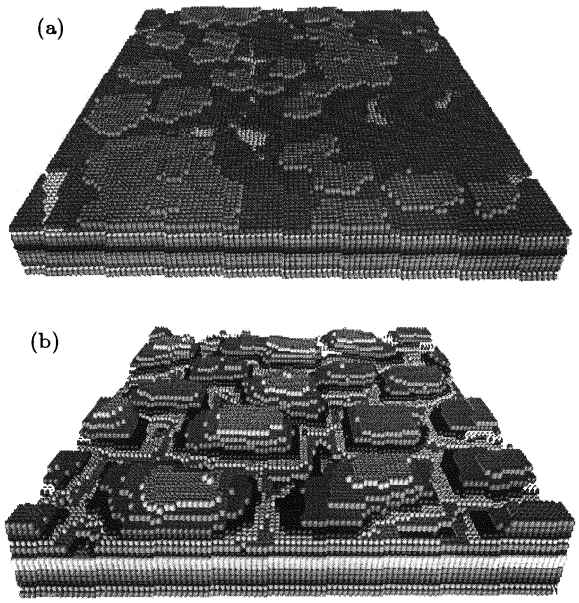



**Fig.2.** Close to layer-by-layer (a) and 3D (b) growth pictures from 3D KMC simulation at $T$=299 K, $\theta$=10.3 MLs and $F$=0.0208 ML/s, where $E_B$ =0.075 and 0.25 eV, respectively.

In order to accurately determine the growth mode during multilayer Cu thin film growth, the surface roughness as a function of coverage is calculated. The surface roughness is defined as[36]

$$w = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(h(x_i) - \bar{h})^2}, \qquad (5)$$

where $n$ is the total number of deposited atoms, $h(x_i)$ is the height of thin film at the position of $x_i$ and $\bar{h}$ is the average height of thin films, which is given by $\bar{h} = (1/n)\sum_{i=1}^{n} h(x_i)$. Figure 3 shows the coverage dependence of surface roughness for various $E_B$ at 299 K and $F$=0.0208ML/s. It is clear that when $E_B$ <0.075 eV, the growth mode is a complete layer-by-layer growth, while the transition from layer-by-layer growth to 2D–3D Stranski–Krastanov (S-K) growth occurs when the ES barrier is increased to 0.125 eV. Furthermore, when $E_B$ >0.175 eV, the growth mode performs a complete 3D growth. From a rough comparison of KMC simulation with experimental results,[24] an ES barrier $E_B$ >0.125 eV can be estimated. Of course, the accurate value of the ES barrier cannot be obtained from the simple simulation model, due to lack of the detailed experimental data, such as the root-mean-square surface width and the kinematic reflection high-energy electron diffraction (RHEED) intensity. Further study is necessary.

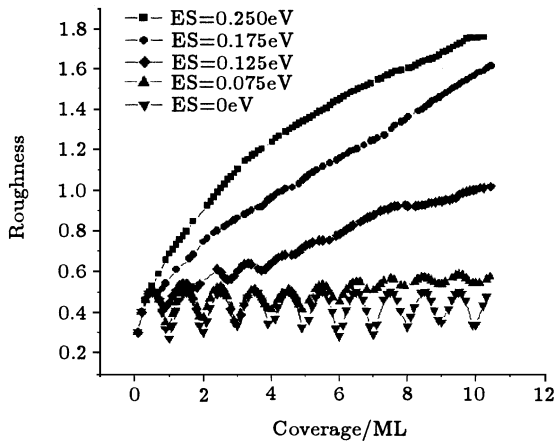



**Fig.3.** Coverage dependence of the surface roughness for various $E_B$ at $T$=299 K and $F$=0.0208ML/s.

During the metal-on-metal growth process, the surface temperature plays a crucial role on the mobility of adatoms and consequently affects the growth mode of multilayer thin film formation. Figure 4 displays the morphologies for various temperatures from KMC simulations, where the ES barrier $E_B$=0.175 eV, $F$=0.0208 ML/s and coverage $\theta$=6.0 MLs. At a relative lower temperature ($T$=250 K), the mobility of deposited atoms is slow, resulting in multiple islands formed on the surface and thus higher surface roughness. The growth mode is a perfect 3D growth, as shown in Fig.4(a). With increasing temperature, the mobility of adatoms will become so fast and the mobility of small island will increase so that coalescence effect enhances significantly, leading to the occurrence of layer-by-layer growth mode, as shown in Figs.4(c) and 4(d). Figure 5 shows the surface roughness for various temperatures, where $F$=0.0208 ML/s, coverage $\theta$=6.0 MLs and the ES barrier $E_B$ =0.175 eV. Clearly, when the temperature is lower than 300 K, the growth mode is a perfect 3D growth, while at higher temperatures ($T$ >300K), the growth mode will change to layer-by-layer growth at low coverage ($\theta$ < 4ML).

Another interesting phenomenon which should be pointed out is that the growth mode is also coverage-dependence at higher temperatures, as shown by the curve at $T$=400 K in Fig.5. In fact, with increasing coverage, more and more defects will appear on the surface. The completion of this kind of defects will become difficult, i.e. the surface wetting is poor in the case of higher ES barrier, giving rise to the transition from layer-by-layer growth to 3D growth. Of course, in the case of lower ES barrier ($E_B$ <0.075 eV), the surface wetting effect is sufficient and the growth

mode will remain layer-by-layer growth with increasing coverage, i.e. independent of the coverage, as can be seen in Fig.3.

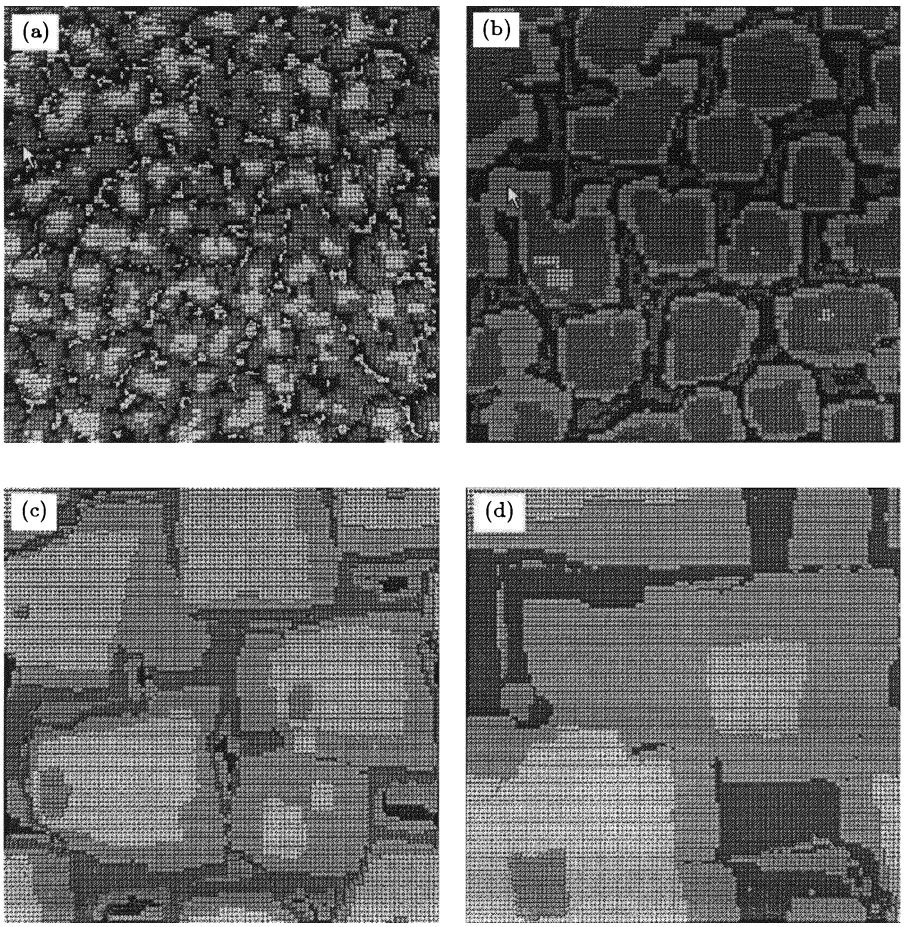



**Fig.4.** Morphologies obtained from KMC simulation at $F$=0.0208 ML/s and coverage $\theta$=6.0 MLs, where the ES barrier $E_{\mathrm{B}}$ =0.175 eV. From (a) to (d), the temperature is set to be 250, 300, 350 and 400 K, respectively.

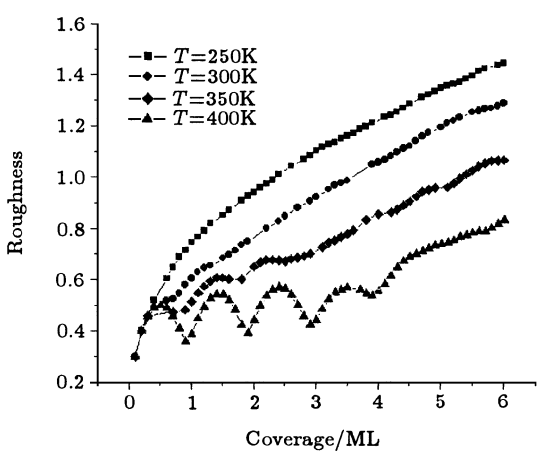



**Fig.5.** The surface roughness as a function of coverage for various temperatures, at $F$=0.0208 ML/s, coverage $\theta$=6.0 MLs and the ES barrier $E_{\mathrm{B}}$ =0.175 eV.

Finally, we have attempted to reveal the influence of the exchange barrier on the growth morphology or growth mode. Figure 6 displays the coverage dependence of the surface roughness at different exchange barriers and ES barriers. The dark small dot indicates the results in which no exchange process is included in the simulations, while the dark square shows the results at $E_{\mathrm{ex}}$=0.20 eV, where the ES barrier keeps the same value $E_{\mathrm{B}}$=0.175 eV. It can be seen clearly that there are almost no difference between different exchange barriers but at the same ES barrier. It is illustrated that the microscopic exchange diffusion process cannot be reflected in the surface roughness. In other words, the exchange barrier does not affect the growth mode or growth morphology. In fact, for homoepitaxial growth, it is difficult to identify the adatoms and the replaced atoms, as well as to identify the island shapes before and after the exchange process finishes. It is also unlikely to describe the exchange process by the observation of island morphologies or calculation of island density. Nevertheless, it is important to note

that the exchange diffusion process is very difficult to treat theoretically because it requires an accurate description of both the localized adatom–substrate interactions and the harmonic and anharmonic distortions of the substrate. Further experimental and/or more accurate theoretical calculations are needed to investigate this microscopic mechanism.

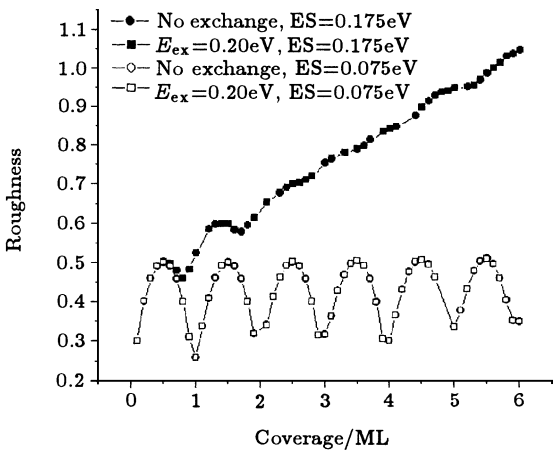



**Fig.6.** Coverage dependence of the surface roughness for different exchange barriers and ES barriers, at $T$=299K and $F$=0.0208ML/s.

# 4. Conclusions

In summary, a comprehensive kinetic Monte Carlo simulation model has been presented to investigate the multilayer copper thin film growth on Cu (100) surface, where the effects of small island diffusion, edge diffusion along the islands, exchange of the adatom with an atom in the existing island, as well as mass transport between interlayers are studied. Three kinds of growth modes, such as layer-by-layer growth, S-K growth and 3D growth are given under different growth conditions. Our simulation results indicate that:

1. The ES barrier does exist for the Cu/Cu(100) system. An ES barrier $E_{\rm B}$ >0.125 eV is estimated from a comparison of the KMC simulation with the realistic experimental images.

2. The growth mode depends strongly on the substrate temperature. The transition from layer-by-layer mode to S-K mode and 3D mode will occur with the decrease of temperature.

3. The exchange barrier does not affect the growth mode or growth morphology for the homoepitaxial growth.

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