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Monte Carlo simulation of the effect of atomic diagonal transition on cluster diffusion*

Xie Feng(谢 峰)[a], Wang Xiao-Ping(王晓平)[a)b)†],
Shi Qin-Wei(石勤伟)[b], and Zhao Te-Xiu(赵特秀)[a]

[a] *Department of Physics, University of Science and Technology of China, Hefei* 230026, *China*

[b] *Structure Research Laboratory, University of Science and Technology of China, Hefei* 230026, *China*

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The effect of atomic diagonal transition on the cluster diffusion and its size dependence is simulated by kinetic Monte Carlo method. The thresholds of atomic diagonal transition barriers E_{dt} are found to be 0.2eV and 0.4eV, corresponding to with and without evaporation and condensation mechanism, respectively. The results indicate that the cluster diffusion is controlled primarily by the atomic diagonal transition, and the cluster diffusion coefficient D decreases drastically with increasing E_d when $E_d < E_{dt}$. Moreover, D is also found to depend on the cluster size N. The size dependence of D can be described by $D \propto N^{-1.57\pm0.034}$ when E_d=0. This behaviour is very close to that of the perfect periphery diffusion. On the contrary, D is almost independent of E_d when $E_d > E_{dt}$, and the relationship between D and N changes into $D \propto N^{-1.08\pm0.027}$.

Keywords: cluster diffusion, Monte Carlo simulation, barrier
PACC: 6855, 6890, 6822, 7115Q

1. Introduction

The characteristics of adatom diffusion on the substrate during epitaxial growth have been widely studied. It is well known that the different adatom diffusion may result in different island densities as well as island morphologies, such as fractal or compact patterns.[1−6] In addition to the adatom diffusion, the diffusion of cluster, which contains from several tens to thousands atoms, can also directly affect the nucleation, growth and coarsening of islands in the submonolayer growth. Therefore, the behaviour of the cluster diffusion has drawn more and more attention, which would shed more light on how to control the morphology and the structure of epitaxial thin films.[7−9]

As to the cluster diffusion, it is important to explore how the cluster diffusion coefficient D depends on the cluster size N. Voter[10] investigated the problem by means of molecular dynamics and first found that, for the clusters containing more than 10 atoms ($N > 10$), the relationship between D and N could be described as,

$$D \propto N^{-\alpha}, \tag{1}$$

where α is the scaling exponent coefficient. Many researchers also obtain similar result through other analytic theories. Three kinds of mechanisms have been proposed,[11−13] which can dominate in the behaviours of the cluster diffusion and result in different α values. The first is the periphery diffusion (PD) mechanism, in which the cluster diffusion is caused by atomic motion along the cluster periphery, and α=1.5 is predicted. The second is the terrace diffusion (TD) mechanism, in which the cluster diffusion is controlled by the diffusion of cavities in clusters or atoms in vacancies, and α=1.0 should be required. The third is the evaporation and condensation (EC) mechanism, in which the cluster diffusion is dominated by the exchange process between the atoms in clusters and those in near environment, and α=0.5 should be observed. Note that if one atom evaporates from a cluster and reattaches to the cluster just after a few randomly walks, the process is called correlated evaporation and condensation (CEC) mechanism, and α=1.0 should be obtained.

Due to the simultaneous contributions of different mechanisms, the behaviour of the cluster diffusion may generally be influenced by many parameters, such as temperature, substrate, shape and size of clusters.

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†Corresponding author, email: xpwang@ustc.edu.cn

For example, the results of the ratio between various mechanisms show that, EC mechanism is more distinct at high temperatures while PD mechanism is dominant at low temperatures.[14]

As for the experimental studies, scanning tunnelling microscopy (STM) makes it possible to observe the cluster diffusion directly.[7−9] For instance, based on STM observations, Pai[8] reported α being respectively equal to 1.14 and 1.24 in Ag/Ag(100) and Cu/Cu(100) systems, and attributed the diversity of α values to the different perimeters of clusters in the two kinds of systems.

In addition to STM observation, kinetic Monte Carlo (KMC) simulation is also frequently used to study the problem of cluster diffusion. Many studies on the effects of substrate, temperature and cluster size on the diffusion coefficient D and its scaling behaviour have been reported. Various values of α, ranging from 0.6 to 1.6, are obtained.[14−18] For example, Pal[18] concentrated on how the cluster size N affected α, and found that $\alpha \approx 1.5$ for $N < 100$ but $\alpha \approx 0.5$ for $N > 100$. All these results indicate that the cluster diffusion is more complex than what one expects and more work is needed to elucidate this point.

Both KMC simulations and STM observations demonstrate that PD mechanism occurs most frequently in the cluster diffusion. When an atom diffuses along the cluster periphery, most of the contribution to the cluster diffusion comes from the transfer of atoms from one side of the cluster to the other side (side-transition), for little efficient diffusion would be produced if the atom moves only on the same side. Therefore, how the atom achieves the side-transition and how the barrier of side-transition influences D and α are open questions.

Many results show that the atoms could complete the side-transition via two mechanisms. One is the EC, and the other is the diagonal transition of an atom, in which the atom on one side of the cluster near the corner transfers to the next nearest neighbour vacant site on the other side. The diagonal transition process is named as the T mechanism in our model. Obviously, the barrier of diagonal transition is higher than that of diffusion to nearest-neighbour site (straight transition) with the same lateral coordination numbers. However, to our best knowledge until now, no systematic report has been presented concerning the effect of diagonal barrier on the behaviour of cluster diffusion. In this paper, an adjustable diagonal transition barrier E_d is introduced, and the effect of diagonal transition on the cluster diffusion coefficient D and scaling exponent α is studied. Our results show that D varies with E_d remarkably, and α is 1.08 for the case without diagonal transition, but changes to 1.57 for the case with free diagonal transition.

2. Model and simulation method

To study the cluster diffusion on a fcc(100) homogeneous substrate, the periodic square substrate with size 400×400 is employed in our KMC simulation. The various size clusters containing N atoms are substituted by randomly created compact clusters.

Based on the molecular dynamics,[10] the rate r of an atom hopping from one site to nearby vacant site is

$$r = \nu_0 \exp[-\Delta E(i,j)/k_{\mathrm{B}}T], \tag{2}$$

where ν_0, the vibration frequency of the atom, is given the value of $1\times10^{12}\mathrm{s}^{-1}$ in our simulation. T is the temperature of substrate (room temperature was used in our simulation), k_{B} is Boltzmann constant. $\Delta E(i,j)$ is the barrier for the atom hopping from site i to site j, and is expressed as[18]

$$\Delta E(i,j) = E_0 - \frac{J}{2}(n_j - n_i), \tag{3}$$

where E_0 is the barrier for free adatom hopping on the substrate, J is the lateral bond strength. E_0=0.52eV and J=$2E_0/5.5$ were presumed in the simulation.[18] n_i and n_j are the numbers of lateral nearest neighbours at site i and j, respectively. Though the model is simple as compared with the embedded atom method (EAM) and the effective medium theory (EMT),[14] its rationality has been demonstrated by some reports.[18,19] The advantage of this model is that N-fold way algorithm can be exploited in the simulation,[20] which can shorten the simulation time and supply more statistical samples.

In order to distinguish the difference between atom hopping to the next-nearest neighbour vacant site (diagonal hopping) and to the nearest neighbour vacant site (straight hopping), an additional diagonal hopping barrier E_d is presumed and its value can be adjusted between 0–0.7eV, corresponding to the homogeneous epitaxial growth. Therefore, the whole hopping barrier of the atom $\Delta E(i,j)$ is rewritten as

$$\Delta E(i,j) = E_0 - \frac{J}{2}(n_j - n_i) + E_d. \tag{4}$$

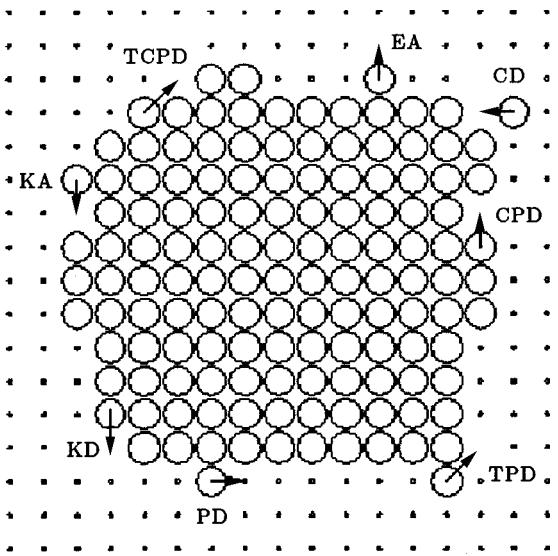


Fig.1. Illustration of different elementary diffusion mechanisms.

Figure 1 shows the schematic of different elementary diffusion mechanisms in a fcc(100) substrate. As seen from Fig.1, straight hopping of an atom along cluster periphery from a site with one nearest neighbour to a vacant site with one nearest neighbour is characterized as a PD event; hopping from a site with two nearest neighbours to a vacant site with two nearest neighbours is characterized as a CPD event, hopping from a site with one (two) nearest neighbours to a vacant site with two (one) nearest neighbours is characterized as a KA(KD) event. All the PD, CPD, KA and KD are generally classified as the PD mechanism. The process in which an atom evaporates from the cluster periphery and becomes a free atom is characterized as an EA event, and the inverse process is a CA event. A free atom hopping to a vacant site of the cluster periphery with two nearest neighbours is characterized as a CD event, and the inverse is an ED event. All the EA, ED, CA and CD are generally classified as EC mechanism. All the diagonal transitions, such as TPD and TCPD shown in Fig.1, are classified as a T mechanism.

According to Eqs.(2)–(4), each type of hopping has its intrinsic hopping rate. In the simulation, before any hopping, one type of possible hopping should be chosen based on its possibility P_k,

$$P_k = N_k r_k / \sum_i N_i r_i, \tag{5}$$

where N_i is the number of hopping of type i, and r_i is its hopping rate. Once a particular hopping is chosen and carried out, the atom positions are updated and the time is increased with Δt.

$$\Delta t = 1 / \sum_i N_i r_i. \tag{6}$$

The whole process described above is referred to one step. After n steps have been carried out, the displacement of the cluster mass centre, Δr_{cm}, can be obtained and the cluster diffusion coefficient D can be calculated from.

$$D = \lim_{t\to\infty} \frac{\langle \Delta r_{cm}^2(t) \rangle}{4t}. \tag{7}$$

To improve the accuracy of simulation, the relaxation process containing about 100–200 steps was first employed after the initial cluster generated, $2 - 3 \times 10^6$ Monte Carlo steps were then performed to obtain the diffusion coefficient D. Simulation for each cluster with a size N should be repeated at least 200 times in order to gain more accurate average value of D (N). For a good judgement of which mechanism is responsible for the cluster diffusion, the numbers and ratios of all types of hopping occurring during the simulation should be recorded, and the patterns of clusters with different diffusion time should also be recorded.

3. Results and discussion

3.1. Effect of diagonal transition barrier E_d on cluster diffusion coefficient D

The effect of diagonal transition barrier E_d on D can be directly explored based on Eq. (4). In order to avoid the fluctuation of D caused by size effect (see section 3.2) and have appropriate simulation time, the cluster with a size N=120 was chosen in this section. The value of E_d was adjustable from 0 to 0.7eV (E_d >0.7eV showed the same result as E_d ≈0.7eV). Both cases of EC permitted and EC forbidden were considered in the simulation.

Figure 2 shows the results of D varying with E_d for the cluster of N=120. Two features can be observed from Fig.2. First, D decreases apparently with increasing E_d, and it shows a saturated behaviour when E_d is larger than a threshold E_{dt}. The value of E_{dt} is about 0.2eV in the EC permitted case, but 0.4eV in the EC forbidden case. Second, when E_d <0.15eV, no distinct effect of E_d on D can be found for the two cases. However, when E_d is far larger than this, the value of D obtained in the EC permitted case is about one order of magnitude larger than that in the EC forbidden case.

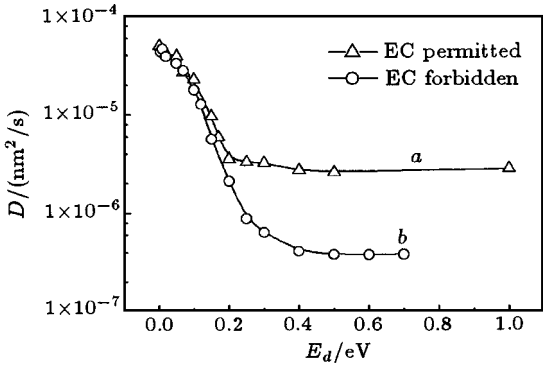


Fig.2. Cluster diffusion coefficient versus the diagonal transition barrier E_d for the cluster of N=120. Triangles (curve a) and circles (curve b) show the results for EC permitted and EC forbidden cases, respectively.

According to the model in section 2, the side-transition of atoms on the cluster periphery occurs dominantly via the T or CEC mechanism. If the diagonal transition occurs, an atom can move from one side to its adjacent side of the cluster. Consequently, the atom hopping will not be limited just on one side of the cluster, and the mass centre displacement of the cluster Δr_{cm} may have a more effective gain. For example, after an atom diffuses from one side to the opposite side along the periphery of the cluster with a gyration radius of R (two side-transition needed at least),[10] Δr_{cm} can be estimated by $\Delta r_{cm} = 2R/N \propto N^{-0.5}$. Hence, we can conclude from Fig.2 that a small E_d can enhance the diagonal transition, which may bring about the characters of the cluster diffusion similar to the pure PD mechanism. On the other hand, the side-transition process can also be accomplished via the CEC process, in which an atom near the corner of the cluster would evaporate first from one side and reattaches to the other adjacent side after several steps of free hopping.

Obviously, the contribution to cluster diffusion from the diagonal transition and the CEC processes is controlled by the barriers. When the diagonal transition barrier E_d is fairly small, the side-transition occurs dominantly via the T mechanism. As the curve for E_d <0.2eV in Fig.2 shows, D decreases drastically because the probability of diagonal transition decreases with increasing E_d.

Also as seen in Fig.2, D becomes almost independent of E_d for $E_d > E_{dt}$. This suggests that the side-transition is not dominated by the diagonal transition (T mechanism) any more. If there exists other mechanism in which the barrier is lower than E_{dt}, the side-transition will occur via this mechanism instead of the T mechanism. In the EC permitted case, the value of E_{dt} is considered to be the same as E_{CEC}, the barrier of side-transition is created via effective CEC mechanism. However, it is difficult to obtain the value of E_{CEC} directly. Therefore, the fractions of contribution from all the mechanisms for various E_d were recorded during the simulation instead, from which the effects of different mechanisms on the cluster diffusion can be explored in detail.

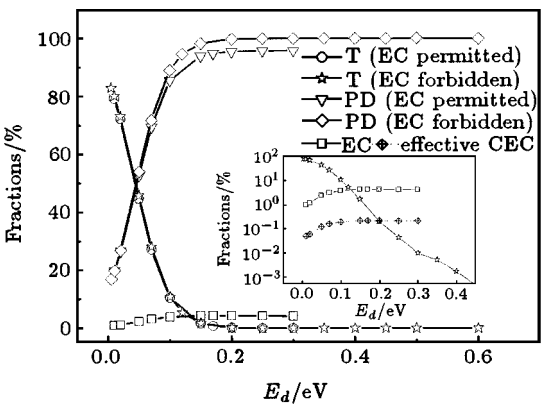


Fig.3. The fractions of contributions from different diffusion mechanisms versus E_d. Inset figure shows the small fraction by exponential axis.

As seen in Fig.3, when $E_d < E_{dt}$, the fraction from the T mechanism decreases rapidly with increasing E_d, no matter whether the EC process exists or not. The fractions from both EC and PD mechanisms increase with E_d. Moreover, the fraction from the EC mechanism is found to exceed that from the T mechanism when $E_d \approx$0.14eV. If 0.14eV is considered to be E_{dt}, its value is obviously smaller than 0.2eV obtained from Fig.2.

In fact, the above difference can be attributed to the fact that only a little part of EC process could result in an effective side-transition, because the atom evaporated from one side of the cluster has much more opportunity to reattach to the cluster at the same side than at the other side. For estimating the ratio of effective CEC to EC, each hopping of the atom has been recorded, and the statistical analysed results indicate that the effective CEC process occupies only 5% of EC for various E_d, as shown in the inset of Fig.3. Assuming that E_{dt} is equal to the value of E_d at which the fraction from the effective CEC mechanism just exceeds that from the T mechanism, E_{dt} can be found to be ~0.2eV, being consistent with the result of Fig.2.

As for the EC forbidden case, E_{dt} is found to be ~0.4eV from Fig.2. The result can also be attributed to the fact that there exist some complicated compound mechanisms, by which the side-transition of

atoms can also be achieved. These mechanisms may be composed of several PD, KA, and KD processes, and one of the possible mechanisms is shown in Fig.4.

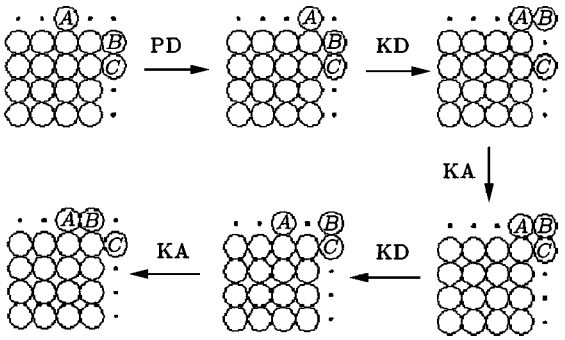


Fig.4. Schematic of one possible compound mechanism for side-transition.

Note that the probability of the compound processes is very small during the random atom diffusion. For example, the probability of the process shown in Fig.4 can be expressed approximately as:

$$\gamma = \left(\frac{n}{L}\right)^2 \cdot \frac{K_{11}}{2K_{11}+K_{21}} \times \frac{K_{21}}{2K_{11}+K_{21}} \cdot \frac{K_{12}}{K_{21}+2K_{12}+K_{11}} \times \frac{K_{21}}{2K_{21}} \cdot \frac{K_{12}}{K_{11}+K_{21}+2K_{12}}, \quad (8)$$

where K_{11}, K_{12}, and K_{21} are the hopping rates of PD, KA, and KD processes respectively, n is the number of mobile atoms (the atoms with less than three lateral nearest-neighbours in our model) along the cluster periphery. L is the perimeter of the cluster. $(n/L)^2$ in Eq.(8) indicates that atom B needs assistance of atoms A and C to complete the side-transition process (see Fig.4). As for the cluster with N=120, the statistical value of n is about 12. Therefore, the probability γ can be estimated to be about 5×10^{-5}. In the simulation, other compound processes have also been considered, and the corresponding probabilities are found to be about the same value stated above or smaller sometimes. As seen in the inset of Fig.3, when $E_d \approx$0.4eV, the fraction from the T mechanism drops to about 10^{-5}. Compared with γ, the average barrier of the compound mechanism can be estimated to be ~0.4eV, which is also in agreement with the value deduced from Fig.2. This indicates that, in the EC forbidden case, the side-transition can still occur via the compound processes although the T mechanism has been much suppressed due to a rather large value of E_d.

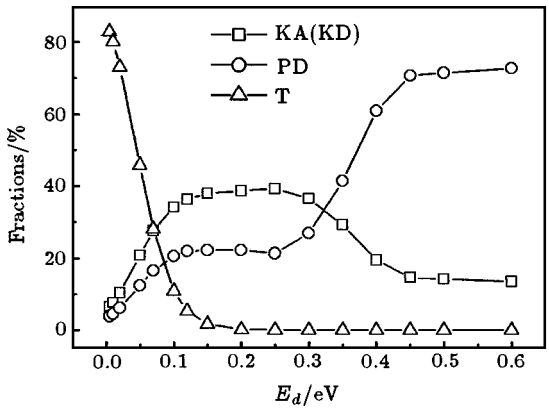


Fig.5. Fractions of contribution from all the mechanisms in the EC forbidden case.

To further clarify the above conclusion, the fractions from all the mechanisms during cluster diffusion in the EC forbidden case were recorded. As seen from Fig.5, the fractions from both PD and KA (KD) mechanisms increase while that from the T mechanism decreases for $0 < E_d < 0.15$eV. With further increase of E_d, all the fractions become invariable until $E_d \approx$0.3eV. Moreover, the fraction of PD increases rapidly while that of KA (KD) decreases correspondingly for 0.3eV$< E_d <$0.45eV. The behaviours indicate that the opportunity of diagonal transition decreases with increasing E_d, which may result from the reduction of the kink density along the cluster periphery. Consequently, more and longer straight edges of the cluster should be expected, which were confirmed by the observation on the recorded patterns of clusters in the simulation. It is worth noting that the fraction from KD(KA), which is the key in the compound process, reaches a minimum and becomes invariable for $E_d >$0.45eV, and D reaches correspondingly a saturation at this point (Fig.2, curve *b*).

3.2. Size dependence of cluster diffusion coefficient D (N)

Based on the results and discussion of the above section, the size dependence of D is investigated with two different diagonal transition barriers, E_d=0 and E_d=0.7eV. Figure 6 demonstrates a variety of D obtained for different cluster sizes (15$< N <$3000). As shown in Fig.6, D decreases with increasing N, and the scaling behaviour $D \propto N^{-\alpha}$ is well presented. The value of the scaling coefficient α is found to be 1.08±0.027 for E_d=0.7eV and 1.57±0.034 for E_d=0, respectively.

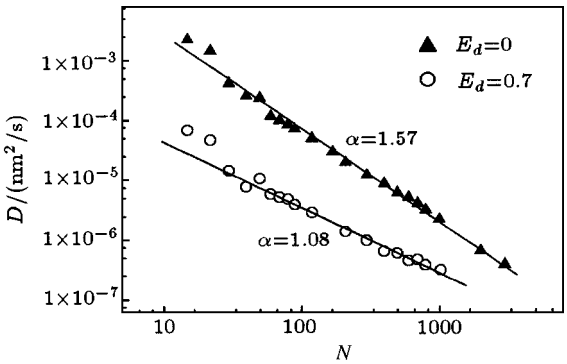


Fig.6. The cluster diffusion coefficient D versus cluster size N. Solid and open symbols are respectively corresponding to the cases of E_d=0 and E_d=0.7eV.

Also from Fig.6, we can see that, for E_d=0.7eV (most of the diagonal transition are inhibited in this case), the power-law character of D-N is not obvious in the region for small cluster sizes (N <50). The behaviour, similar to many reported results, may come from the fluctuation effect of cluster size (up to ±10 atoms). It has been pointed out that the fluctuation could affect the value of D notably and bring about the simulation results to deviate from the predicted ones.[17] In fact, the fluctuation of D presented in small size region is mainly caused by the fact that the diffusion of small size clusters may be readily influenced by their shapes. For example, square-like clusters on fcc(100) substrate are more stable than clusters with other shapes, consequently, the value D (n^2) of square-like cluster is expected to be smaller than D (n^2+1).[15] To avoid such a fluctuation effect on the scaling behaviour, the scaling exponent α is fitted only for the region of $N > 40$ in our discussion.

Since the TD mechanism has not been involved in our simulation, the result of $\alpha \approx 1$ in the case of E_d=0.7eV indicates that CEC should be the dominant mechanism in the cluster diffusion process. In the simulation, the fraction of contribution from the EC mechanism $P_{\rm EC}$ is also found to decrease with increasing cluster size N, and the relation of $P_{\rm EC} \propto N^{-0.26\pm0.014}$ can be obtained. More investigations are needed to elucidate whether there is any connection between $P_{\rm EC} \propto N^{-0.26\pm0.014}$ and $D \propto N^{-1.08}$.

Additionally, both the fractions of contribution from KA (KD) and CPD are found to increase with increasing N in our simulation, which are similar to the results found by Pal and Fichthorn.[18] Obviously, the variation of fraction of KA (KD) is related to the number of kinks on the cluster periphery, i.e., the more the kinks, the more the KA (KD) hoppings are expected to occur. Therefore, more kinks should be found on the periphery of the large cluster as compared with the small one. It is considered that more kinks on the cluster periphery will make the shape of cluster approach a circle rather than a square. Figure 7 shows two typical shapes of clusters with different sizes. As is seen, the shape of the cluster with N=500 looks more circle-like than that with N=40. Moreover, the diffusion behaviour of the circle-like cluster is found to be less influenced by n^2 than that of square-like one. Consequently, the fluctuation of D can be depressed for large size clusters, and the power-law dependence will be well improved.

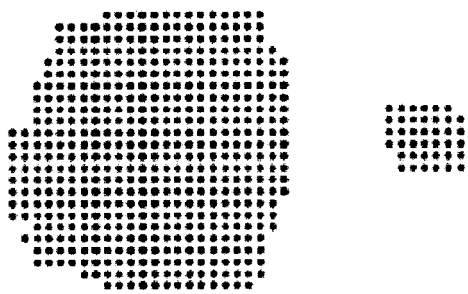


Fig.7. The typical shapes for large size (N=500) and small size (N=40) clusters.

In the case of E_d=0, the barrier of diagonal transition is, in fact, only controlled by the variation of lateral neighbours (see Eq.4). Therefore, the T mechanism is equivalent to the PD mechanism in this case. As discussed in section 3.1, the cluster diffusion is dominantly controlled by the T mechanism under the condition of small E_d. Accordingly, the cluster diffusion can be considered as dominated by the PD mechanism and α should be about 1.5. This prediction is consistent very well with our simulation result $\alpha = 1.57 \pm 0.034$. Furthermore, the fluctuation of D for small size clusters is also weaker than that in the case of E_d=0.7eV (see Fig.6). In our simulation, the fractions of all the mechanisms with various cluster sizes are also recorded when E_d=0. The results show that the fraction of the T mechanism is about 82%–85%, the PD mechanism is about 15%, and the EC mechanism is almost zero(see Fig.3), and these fractions are nearly independent of the cluster size. All the above results also support our explanation.

Note that in Figs 6 and 2, the cluster diffusion coefficient D for E_d=0.7eV just ranges in 10^{-20}–10^{-18}cm^2/s for different cluster sizes, which is rather smaller than that of typical experimental data ($\sim 10^{-17}$cm^2/s).[7] However, under the same condition, the value of D for E_d=0 is about 20 times larger than that for E_d=0.7eV. From this point, one

can conclude that the diagonal transition is important in the experimental observation for the cluster diffusion. Moreover, α values between 1–1.5 obtained from the experiments also indicate the existence of diagonal transition in the cluster diffusion.[8,9]

4. Conclusions

The effect of diagonal transition on the cluster diffusion coefficient and its scaling behaviour has been studied with kinetic Monte Carlo simulation on the fcc(100) substrate. It is found that a threshold value E_{dt} exists for the diagonal transition barrier E_d, and E_{dt} is about 0.2eV for the EC permitted case but about 0.4eV for the EC forbidden case. Our simulation indicates that E_{dt} ~0.2eV results from CEC dominated diffusion process, and E_{dt} ~0.4eV from the compound process composed of PD, KA, and CPD hoppings. When $E_d < E_{dt}$, the cluster diffusion is mainly controlled by the T mechanism, and the diffusion coefficient D decreases drastically with increasing E_d. On the contrary, when $E_d > E_{dt}$, the cluster diffusion is mainly dominated by CEC mechanism or the compound process, and D is almost independent of E_d. Furthermore, the cluster size dependence of D can be described by a power-law, $D \propto N^{-\alpha}$. The scaling exponent α is found to be 1.57 ± 0.034 in E_d=0 case, in which the behaviour of the process approaches that caused by the PD mechanism. On the other hand, α is changed to 1.08 ± 0.027 for E_{dt}=0.7eV, which is consistent with the prediction of the CEC mechanism.

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