Simulation of Depositions of a Lennard-Jones Cluster on a Crystalline Surface

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Depositions of amorphous Lennard-Jones clusters on a crystalline surface are numerically investigated. From the results of the molecular dynamics simulation, we found that the deposited clusters exhibit a transition from multilayered adsorption to monolayered adsorption at a critical incident speed. Employing the energy conservation law, we can explain the behavior of the ratio of the number of atoms adsorbed on the substrate to the cluster size. The boundary shape of the deposited cluster depends strongly on the incident speed, and some unstable modes grow during the spread of the deposited cluster on the substrate. We also discuss the wettability between different Lennard-Jones atoms.

Subject Index: 023, 331, 335, 545

§1. Introduction

A nanocluster containing $10^{-10,000}$ molecules exhibits intermediate properties between bulk materials and individual molecules. Recently, there has been growing interest in the physics of nanoclusters. In particular, it is important to investigate the depositions of nanoclusters on solid surfaces for the construction of high-quality films used in nanoscale electronic devices and photonic devices.

The ionized cluster beam (ICB) technique was developed by Yamada et al. The ICB technique is used to produce atomic clusters by employing adiabatic expansion of condensed vapour through a nozzle into a high-vacuum region. In the ICB technique, clusters are ionized by electron impact and then accelerated toward a substrate. Because the ICB technique controls the translational kinetic energy of the cluster, there have been many experimental and theoretical studies aimed at understanding the effect of the incident velocities of the cluster on the outcome of impact.

The outcome of such a cluster impact is largely affected by the incident velocity, as seen from the phase diagrams in Refs. 12) and 13) If the translational kinetic energy per atom becomes too large, the cluster can damage the substrate, and the cluster can break into pieces after the impact. However, if the translational kinetic energy per atom is less than 100 eV, the cluster is adsorbed on the surface or reflected by the surface. Awasthi et al. carried out molecular dynamics simulations for collisions of Lennard-Jones clusters with weakly attractive surfaces. They discovered that the cluster rebounds when the translational kinetic energy of the cluster is larger than the adhesion energy. Moreover, they clarified that a transition

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from adhesion to rebound occurs at the critical translational kinetic energy. Järvi et al. carried out molecular dynamics simulations of low-energy deposition of individual metal clusters on a (100) surface. They revealed that the heat generated by the impact partially or completely melts the deposited cluster. As a consequence, the atoms in the cluster are rearranged and adjusted to the atomic structure of the substrate. They found the maximum size at which single clusters align epitaxially on the substrate.

Recently, Kuninaka and Hayakawa have carried out molecular dynamics simulations of two identical colliding clusters and investigated impact phenomena of nanoclusters subject to thermal fluctuations. They found super-rebound events in which the restitution coefficient is larger than 1. They confirmed the validity of the macroscopic quasi-static theory of cohesive collisions. This suggests that the research on nanoclusters is relevant even for the study for fine powders whose diameters range from 100 nm to 1 μm. They also revealed the mechanism responsible for the super-rebound process, the normal rebound, and the merging.

Although early numerical studies assumed that the clusters are highly crystallised, we also need to know the properties of amorphous clusters. Indeed, it is easy to form metastable amorphous clusters in terms of the quench process from high-temperature liquids. The main purpose of our paper is to understand the behavior of the deposited amorphous Lennard-Jones clusters on the crystalline surface at zero temperature. Here, we report on our molecular dynamics simulation of the depositions with the small incident energies per atom, which lie in the so-called soft-landing regime (0 − 2 eV). In addition, we report on the wettability between different Lennard-Jones atoms.

From the analysis of the final configurations of the deposited clusters, we find the existence of a morphological phase transition from the hemi-spherical droplet to the monolayer film at the critical incident speed. The multilayered adsorption state is described on the basis of the energy conservation law. Furthermore, we find that there are some unstable modes of the boundary shape of the deposited cluster.

This paper is organized as follows. Section 2 consists of two subsections. In §2.1, we explain the model of our numerical simulation. We explain our setup of cluster depositions in §2.2. Section 3 consists of five subsections. In §3.1, we show some time evolutions of impact processes. In §3.2, we explain how the cluster size and cluster adsorption parameter depend on the incident speed after the impact. In §3.3, we discuss the transition from partial wetting to perfect wetting of deposited clusters. In §3.4, we explain the morphological change of the final configuration of adsorbed atoms in clusters. In §3.5, we discuss the wettability between different Lennard-Jones atoms. In §4, we discuss our numerical results and summarize the conclusion.
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§2. Molecular dynamics simulation

2.1. Model

To investigate the nanocluster depositions on a substrate, we perform a molecular dynamics simulation. Figure 1 displays a snapshot of our numerical simulation. Because we are interested in neutral nanoclusters and substrates, the electrostatic interaction between atoms is not considered. We assume that the potential energy of the interaction between two atoms can be described using the Lennard-Jones (LJ) potential:

$$U(r_{ij}) = 4\epsilon_{\alpha\beta} \left\{ \left( \frac{\sigma_{\alpha\beta}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r_{ij}} \right)^{6} \right\},$$

(2.1)

where subscripts $\alpha$ and $\beta$ specify the species of LJ atoms and $r_{ij}$ is the distance between two atoms labeled by $i$ and $j$. Here, $\epsilon_{\alpha\beta}$ and $\sigma_{\alpha\beta}$ are, respectively, the strength of the interaction and diameter of the repulsive core between $\alpha$ and $\beta$ atoms. If $\alpha \neq \beta$, we adopt the cross parameters of the LJ potential using the Lorentz-Berthelot rule as

$$\sigma_{\alpha\beta} = \frac{(\sigma_{\alpha} + \sigma_{\beta})}{2}, \quad \epsilon_{\alpha\beta} = \sqrt{\epsilon_{\alpha} \epsilon_{\beta}}.$$  

(2.2)

We mainly investigate the case that the cluster and substrate are constructed using the same atoms, A. Here, we borrow the values of LJ parameters and mass of a typical inert gas, argon. Therefore, $\epsilon_{AA}$, $\sigma_{AA}$, and the mass of an A atom $m_A$ are $1.65 \times 10^{-21}$ J, $3.405$ Å, and $6.63 \times 10^{-26}$ kg, respectively.\(^{34)} \text{–}^{36)} On the other hand, to study the effect of the interaction energy between different atoms of the cluster and substrate, we introduce a C atom as the constituent of the substrate in §3.5. We use the values of LJ parameters and mass of carbon to the C atom, in which $\epsilon_{CC}$, $\sigma_{CC}$, and the mass of C atom $m_C$ are $3.86 \times 10^{-22}$ J, $3.354$ Å, and $1.99 \times 10^{-26}$ kg, respectively.\(^{34)} \text{–}^{36)} We should note that the interaction energy between the cluster and substrate, $\epsilon_{AC}$, is several times weaker than $\epsilon_{AA}$. In the following, we omit the subscripts of the LJ parameters of the interaction between the A atoms and we adopt

Fig. 1. A snapshot of our simulation of a nanocluster deposition. The incident cluster contains 300 atoms that are bounded by the Lennard-Jones potential. The substrate consists of a single layer (120 × 120) of atoms on a triangular lattice.
simplified notations $\epsilon, \sigma$. We also adopt $m$ as the mass of an A atom. We use $\epsilon, \sigma,$ and $m$ as the units of energy, length, and mass, respectively. Thus, the unit time is given by $\tau = \sqrt{m \sigma^2 / \epsilon}$.

We use a single-layer surface, which involves $120 \times 120$ atoms on a triangular lattice as the substrate with the periodic boundary condition.$^{37,38}$ We set the lattice constant to $2^{1/6} \sigma_{\alpha \alpha} (\alpha = A, C)$ as the equilibrium distance between atoms. To avoid the destruction of the substrate, each atom of the substrate is also tethered to its equilibrium position by an elastic spring. In actual impacts of nanoclusters on substrates, the energy induced by an impact is relaxed to the internal motion of the atoms of the bulk region of the substrate. To represent such energy relaxation, we simply introduce the viscous force proportional to its velocity. The introduction of the viscous force has another advantage to reduce the unrealistic boundary effects. Indeed, if we do not introduce the viscous force, the acoustic wave would be transmitted across the boundary. The atom of the substrate at $r_i$ satisfies the equation of motion

$$m_\alpha \frac{d^2 r_i}{dt^2} = - \sum_j \frac{d}{dr_i} U(r_{ij}) - k (r_i - r_{eq}^i) - \lambda \frac{dr_i}{dt}, \quad (2.3)$$

where $\sum_j$ is a summation over the interacting pairs $i$ and $j$, and $r_{eq}^i$ is its equilibrium position, and $m_\alpha (\alpha = A, C)$ is the mass of an atom of the substrate. Because we consider the substrate as a rigid surface, we use the spring constant $k = 1.0 \times 10^3 \epsilon / \sigma^2$. We simply adopt the value $\lambda = 1.0 \sqrt{m \epsilon} / \sigma$ for the coefficient of viscosity.

We adopt the velocity Verlet method for numerical integration of the equation of motion for each atom with the time step $dt = 1.0 \times 10^{-3} \sqrt{m \sigma^2 / \epsilon}$. To reduce computational costs, we introduce the cutoff length $\sigma_{\text{cut}} = 3.0 \sigma$ to the LJ potential, and we adopt the periodic boundary conditions in the horizontal $xy$ directions and the free boundary condition in the vertical $z$ direction. It should be noted that the viscous force is evaluated as the value at the previous time step for the numerical integration of Eq. (2.3).

### 2.2. Setup

We make an LJ cluster by the temperature quench$^{39}$ into the metastable phase of LJ fluid.$^{40}$ We prepare $32, 108, 255, 300, 500,$ and $862$ atoms in a periodic box and equilibrate at the temperature $T = 1.0 \epsilon$ with the number density $0.05 \sigma^{-3}$ in the gas state (Fig. 2(a)). It should be noted that the unit of the temperature becomes $\epsilon$, because we set the Boltzmann constant to be unity. To equilibrate the gas at a specific temperature, we perform the velocity scaling until $\tau = 2000 \sigma \sqrt{m / \epsilon}$ for the relaxation to a local equilibrium state. We have confirmed the equilibration of the total energy in the initial relaxation process, and we quench the gas to $T = 0.5 \epsilon$. After an equilibration, a weakly bounded liquid cluster is formed (Fig. 2(b)) and is quenched to $T = 0.01 \epsilon$ to make it rigid. This two-step quenching is adopted to form one cluster from an initial gas state. Indeed, if we quench the system into $T = 0.01 \epsilon$ directly, many small clusters appear. After this equilibration, we obtain an amorphous cluster (Fig. 2(c)). We place the amorphous cluster at $10 \sigma$ above the substrate and give the cluster the translational velocity $V_{\text{imp}}$ to make it collide
against the substrate. It should be noted that the amorphous cluster is metastable to maintain its shape within our observation time. The incident angle of the cluster to the substrate normal is zero. The incident speed of the cluster ranges from $V_{\text{imp}} = 0.1$ to $5.0\sqrt{\epsilon/m}$.

§3. Results

3.1. Time evolution of impacts

Figures 3 and 4 display the time evolutions of the impact of the LJ cluster of 300 atoms on the crystalline surface. Figures 3(a)–(d) represent the case of $V_{\text{imp}} =$
2.0\sqrt{\epsilon/m}$, and Figs. 4(a)–(d) are the case of $V_{\text{imp}} = 4.0\sqrt{\epsilon/m}$.

The incident cluster moves toward the substrate with its translational speed $V_{\text{imp}}$ (Figs. 3(a) and 4(a)), and hits the substrate (Figs. 3(b) and 4(b)). After the hit, the cluster is only deformed to be a hemi-sphere (Fig. 3(c)) for the small incident speed. If the incident speed is, however, larger than a critical value, the deposited cluster is split into many pieces (Fig. 4(c)). After the impact, the deposited cluster is adsorbed on the substrate and settles into the final configuration (Figs. 3(d) and 4(d)).

We observe that the impact process and final configuration depend strongly on the incident speed $V_{\text{imp}}$. In the case of $V_{\text{imp}} < 1.7\sqrt{\epsilon/m}$, no atoms can escape from the cluster during the impact. By contrast, some atoms evaporate during the impact process for $V_{\text{imp}} \geq 1.7\sqrt{\epsilon/m}$. If the incident speed is relatively low, the final configuration is a hemi-sphere on the substrate, as in the case of a partial wetting of a liquid droplet on a dry surface. The deformation becomes larger as the incident speed increases. Above $V_{\text{imp}} = 3.3\sqrt{\epsilon/m}$, the deposited cluster is completely split into fragments, and the absorbed atoms on the substrate form a monolayer coverage. Above $V_{\text{imp}} = 4.5\sqrt{\epsilon/m}$, the deposited cluster bursts into fragments, and the absorbed coverage is no longer characterized by one cluster.

At the moment of impact, the temperature of the deposited cluster increases because the initial kinetic energy is transformed into internal motion. Then, the temperature decreases owing to the heat conduction into the bulk region of the material through the contact area.\cite{23,41} The configuration of the deposited cluster is changed into an energetically favorable position during cooling. Furthermore, the atomic structure of the deposited cluster is adjusted to the substrate. Finally, the
configuration is frozen because of the loss of heat.

3.2. Incident speed dependences of the scaled cluster size and cluster adsorption parameter

In our simulation, the main cluster is detected using the clustering algorithm. Following Allen and Tildesley, we adopt the critical atom separation \( r_C = 1.6 \sigma \). After the cluster settles into the final configuration, we represent \( N_{\text{cls}} \) as the number of atoms in the cluster. With the number of atoms in the cluster before the impact \( N \), we introduce the scaled cluster size:

\[
\xi \equiv \frac{N_{\text{cls}}}{N}.
\] (3.1)

If \( \xi = 1 \), no atoms can escape from the cluster after the impact. On the other hand, if \( \xi < 1 \), some atoms evaporate during the cluster impact.

Let us define an absorbed atom in the cluster if an atom at \( \mathbf{r} \) in the cluster satisfies the relation \( |\mathbf{r} - \mathbf{r}_s| < r_C \), where \( \mathbf{r}_s \) is the position of its nearest neighbor constituent of the substrate. Using the number of these adsorbed atoms \( N_{\text{adh}} \), we can introduce the cluster adsorption parameter:

\[
\eta \equiv \frac{N_{\text{adh}}}{N_{\text{cls}}}.\] (3.2)

If \( \eta < 1 \), the cluster is regarded as a multilayered adsorption. However, if \( \eta = 1 \), the deposited cluster is perfectly spread on the substrate, and it is a monolayered adsorption.

Figures 5(a) and (b) plot the incident speed dependences of \( \xi \) and \( \eta \) for the cluster of 300 atoms. We find that \( \xi \) equals 1 below \( V_{\text{imp}} = 1.7 \sqrt{\epsilon/m} \), but it decreases above \( V_{\text{imp}} = 1.7 \sqrt{\epsilon/m} \). On the other hand, \( \eta \) increases with the incident velocity below \( V_{\text{imp}} = 3.3 \sqrt{\epsilon/m} \), but it is satisfied to \( \eta \simeq 1 \) above \( V_{\text{imp}} = 3.3 \sqrt{\epsilon/m} \). Figure 6(a) plots several results on \( \xi \) for \( N = 255, 300, 500, \) and 862, while Fig. 6(b) is \( \eta - \eta_0 \) for \( N = 32, 108, 255, 300, 500, \) and 862, where \( \eta_0 \) is \( \eta \) at \( V_{\text{imp}} = 0 \). It seems that \( \eta - \eta_0 \) is independent of the size of clusters, while \( \xi \) exhibits weak size dependence.

![Fig. 5](https://academic.oup.com/ptp/article-abstract/122/5/1081/1532028)

Fig. 5. (a) A plot of the scaled cluster size \( \xi \) and (b) a plot of the cluster adsorption parameter \( \eta \) as functions of the incident speed for the cluster of 300 atoms.
How can we understand the behaviors in Figs. 5 and 6? During the impact, the temperature in the cluster increases because the kinetic energy is transformed into internal motion.\cite{23} We assume that the energy flux to the substrate $\Phi_{\text{bulk}}$ during the impact is small, and the temperature becomes maximum $T_{\text{max}}$ when the speed of the center of mass of the cluster becomes zero. Thus, the energy conservation law can be written as

$$
\frac{1}{2} m N V_{\text{imp}}^2 + \frac{3}{2} N T_0 \simeq \frac{3}{2} N T_{\text{max}} + \Delta S, \quad (3.3)
$$

where $T_0$ is the temperature of the cluster before the impact, and $\Delta S$ is the change in the surface energy. With the introduction of the surface tension $\gamma$, the height of the deposited cluster $h$, the contact radius of the deposited cluster $R$, and the ratio $\phi = h/R$, $\Delta S$ is given by

$$
\Delta S = \gamma (2 \pi \phi R^2 - 4 \pi R^2_0), \quad (3.4)
$$

where $R_0$ is the mean radius of the cluster before the impact. By introducing the mean area fraction of the contact area $\rho_{\text{adh}} = N_{\text{adh}} / \pi R^2$, $\Delta S$ can be rewritten as

$$
\Delta S = \gamma \left( \frac{2 N_{\text{adh}}}{\rho_{\text{adh}}} - 4 \pi R^2_0 \right). \quad (3.5)
$$

From Eq. (3.3), $T_{\text{max}}$ satisfies

$$
T_{\text{max}} = T_0 + \frac{m}{3} V_{\text{imp}}^2 - \frac{2 \Delta S}{3 N}. \quad (3.6)
$$

Because the binding energy per atom in the cluster is roughly equal to $\epsilon$, the evaporation takes place at $T_{\text{max}} \simeq \epsilon$. In our simulation, $T_0$ is much smaller than $T_{\text{max}}$, and the last term on the right-hand side of Eq. (3.6) is negligible for large $N$. Thus, the evaporation is considered to take place near $V_{\text{imp}} \simeq \sqrt{3 \epsilon/m}$. In Figs. 5(a) and 6(a), the scaled cluster size becomes $\xi < 1$ above $V_{\text{imp}} = 1.7 \sqrt{\epsilon/m}$, which is consistent with the above estimation. For the clusters with 32 and 108 atoms, $\xi$ decreases faster.
than the other cases. In such cases, we cannot ignore the last term on the right-hand side of Eq. (3.6).

During the impact, an evaporated atom carries away the volume energy \( uV \), which is the potential energy per atom and the kinetic energy \( \frac{3}{2}T_{\text{max}} \) from the cluster. We assume that the internal energy of the deposited cluster decreases because of the energy flux to the bulk of the substrate. Therefore, after the cluster settles into the final configuration, the energy conservation law can be written as

\[
\frac{1}{2}mNV_{\text{imp}}^2 + \frac{3}{2}NT_0 = \Delta S + \Phi_{\text{bulk}} + (1 - \xi)N\bar{E},
\]

(3.7)

where \( \bar{E} = \frac{3}{2}T_{\text{max}} - uV \) is the energy carried away by an evaporated atom. Here, \((1 - \xi)N\) represents the number of evaporated atoms. If the incident kinetic energy is not large, the number of atoms in the cluster is approximately preserved during the impact. Therefore, it is reasonable that the scaled cluster size satisfies \( \xi \approx 1 \). If we assume \( \phi \approx 1 \), the energy conservation law (Eq. (3.7)) can be simplified as

\[
\eta = \frac{m\rho_{\text{adh}}}{4\gamma}V_{\text{imp}}^2 + \frac{\rho_{\text{adh}}}{2\gamma} \left( \frac{4\pi\gamma R_0^2}{N} - \frac{\Phi_{\text{bulk}}}{N} + \frac{3}{2}T_0 \right),
\]

(3.8)

where we have used Eqs. (3.2) and (3.5).

We use \( \rho_{\text{adh}} = 0.91 \), because the adsorbed atoms should match the hexagonal lattice on the substrate. The mean radius \( R_0 \) of a cluster consisting of \( N \) atoms satisfies \( R_0 = r_0N^{1/3} \), where we use \( r_0 = 0.68\sigma \) as a fitting parameter. The solid line in Fig. 5(b) is the theoretical prediction (Eq. (3.8)), where the surface tension \( \gamma \approx 3.5\epsilon/\sigma^2 \) and the energy flux per atom \( \varphi_{\text{bulk}} = \Phi_{\text{bulk}}/N \approx 1.7\epsilon \) are the two other fitting parameters.

The second term on the right-hand side of Eq. (3.8) can be written as

\[
\eta_0(N) = \frac{\rho_{\text{adh}}}{2\gamma} \left( 4\pi r_0^2\gamma N^{-1/3} - \varphi_{\text{bulk}} + \frac{3}{2}T_0 \right).
\]

(3.9)

It is interesting that \( \eta - \eta_0(N) \) is independent of the cluster size. Figure 6(b) plots our numerical results \( \eta - \eta_0(N) \) for \( \eta < 1 \), which support the validity of the theoretical prediction (solid line).

3.3. Transition from partial wetting to perfect wetting of the deposited cluster

Let us introduce the radius of the equimolar dividing surface (Gibbs Surface)\(^{42}\)

\[
R^2 = -\frac{1}{\rho_{\text{adh}}} \int_0^\infty \frac{dp(r)}{dr}r^2dr
\]

(3.10)

as the contact radius of a deposited cluster, where \( \rho(r) \) is the area fraction of the contact area with radial distance from the center of mass of the adsorbed atoms in the cluster. We also define the cluster height \( h \) as \( z_{\text{max}} - z_0 \), where \( z_{\text{max}} \) is the maximum vertical position in the atoms in the cluster, and \( z_0 \) is the minimum vertical position. Assuming a meniscus shape to the deposited cluster, we geometrically calculate the contact angle \( \theta \).
Figure 7 displays $h$, $R$, and $\theta$ for the deposited cluster consisting of 300 atoms as functions of the incident speed. We observe that the cluster height $h$ decreases and the contact radius $R$ increases as the incident speed increases. Above $V_{\text{imp}} = 3.3 \sqrt{\epsilon/m}$, the height and contact angle become zero, which implies that the deposited cluster becomes a monolayer film and is perfectly wetting on the substrate. In this regime, the monolayer film is spread further and its boundary is partially chipped. Therefore, the contact radius decreases. The clusters consisting of 108, 255, 500, and 864 atoms are also perfectly wetting on the substrate at critical incident velocities. However, it should be noted that the transition from multilayer film to monolayer film is only the morphological change of the deposited cluster. In the case of the cluster consisting of 32 atoms, the number of adsorbed atoms is too few to define the wetting parameters $h$, $R$, and $\theta$.

3.4. Morphology of the final configuration of the adsorbed atoms in the cluster

The boundary shape of the contact area depends strongly on the incident speed. To investigate the morphology of the boundary shape, we define the radial distance of the boundary $r = f(\psi)$. Here, $r$ and $\psi$ are the usual radial and azimuthal coordinates. We take the origin to the center of mass of the adsorbed atoms in the cluster. Moreover, we define a dimensionless variable $g(\psi)$ for the boundary as

$$g(\psi) = \frac{f(\psi) - R}{R}. \quad (3.11)$$

We also use its Fourier representations $g(\psi) = \sum_n g_n e^{in\psi}$, with the integer $n = 0, \pm 1, \pm 2, \ldots$.

Figures 8(a)–(h) are the variety of the final horizontal configurations of the adsorbed atoms in the cluster consisting of 300 atoms. We find that there are three phases in the boundary shape. Below $V_{\text{imp}} = 1.5 \sqrt{\epsilon/m}$, the boundary shape is grainy (Figs. 8(a), (b)), and $|g_n|^2$ has some peaks at higher modes. This may be caused by the small number of adsorbed atoms. In the case of $1.5 \sqrt{\epsilon/m} < V_{\text{imp}} < 3.0 \sqrt{\epsilon/m}$, the deposited cluster is uniformly spread on the substrate (Figs. 8(c), (d), (e), (f)).
Fig. 8. Configurations of the adsorbed atoms in a deposited cluster of 300 atoms for each incident speed. $V_{\text{imp}}$ equals (a) 0.5, (b) 1.0, (c) 1.5, (d) 2.0, (e) 2.5, (f) 3.0, (g) 3.5, and (h) 4.0 $\sqrt{\epsilon/m}$.

Thus, peaks of $|g_n|^2$ vanish and their boundaries can be fitted by circles. In the case of $3.0 \sqrt{\epsilon/m} < V_{\text{imp}} < 4.0 \sqrt{\epsilon/m}$, the deposited cluster becomes a thin film or a monolayer film. In this regime, the boundary is partially chipped (Figs. 8(g), (h)), where $|g_n|^2$ has intense peaks at some modes in our simulation. Above $V_{\text{imp}} = 4.0 \sqrt{\epsilon/m}$, the deposited cluster bursts into fragments and the number of adsorbed atoms is too few to define $R$. Thus, we cannot define $g(\psi)$ in this regime.

The thermal fluctuation of a circular geometry step is estimated as $\langle |g_n|^2 \rangle = T/2\pi\beta R n^2$ from the equipartition of energy among the $g_n$. Here, $\beta$ is the step edge stiffness. In our simulation, the thermal fluctuation is estimated as $\langle |g_n|^2 \rangle \sim 0.1$, while the $|g_n|^2$ has peaks ranging from 1000 to 3000 above $V_{\text{imp}} = 3.0 \sqrt{\epsilon/m}$. Therefore, these intensive peaks reflect on the growth of some unstable modes of $g_n$ during the spread of the deposited cluster on the substrate.

3.5. Wettability between different Lennard-Jones atoms

To investigate the effect of the interaction energy on the outcome of impact, we also perform the molecular dynamics simulation in which the LJ parameters of A and C are used for the atoms in the cluster and the atoms of the substrate, respectively. Henceforth, we call this situation A-C case. The Lorentz-Berthelot rule (Eq. (2.2)) is adopted to calculate the cross LJ parameters $\epsilon_{AC}$ and $\sigma_{AC}$. Other simulation settings are the same as those described in §2.

Figure 9 displays the cluster adsorption parameter $\eta$ and contact angle $\theta$ as functions of the incident speed for the LJ cluster of 300 atoms. We also plot the data in the case of the LJ parameters of the A-A case. We observe that the cluster adsorption parameter $\eta$ remains low even if the impact speed is high (Fig. 9(a)). We also stress that any A cluster does not become a monolayer film in which $\theta$ becomes zero during the deposition onto a C surface. This is because the wetted state of the argon cluster is unfavorable on the crystalline carbon surface, which results from
Fig. 9. (a) Plots of the cluster adsorption parameter $\eta$ and (b) plots of the contact angle $\theta$ as functions of the incident speed for both A-C case (open square) and A-A case (filled circle).

$\epsilon_{AC} < \epsilon$. Thus, it is clear that not only the incident velocity, but also the choice of composites is important to determine the final configuration of the deposited cluster.

§4. Discussion and conclusions

In this paper, the incident kinetic energy per atom in the cluster is less than 2 eV. In this case, the damage of the substrate due to the impact of a cluster can be ignored and we considered a single-layer substrate. However, the effect of the interaction between the deposited cluster and the bulk of the substrate is important. In general, the adsorption state is strongly affected by the surface temperature,\textsuperscript{45} but the substrate was assumed to be at $T = 0$ before the cluster impact in our simulation. Therefore, the effect of the surface temperature is also important for future study. Moreover, we performed simulation only for one deposition event of the cluster at each incident speed. Thus, we should take an ensemble average of impact processes for future study.

In conclusion, we found that deposited LJ clusters consisting 32, 108, 255, 300, 500, and 862 atoms exhibit a transition from multilayered adsorption to monolayered adsorption at the critical incident speed, $V_{\text{imp}} = 3.3\sqrt{\epsilon/m}$. From our simulation, we clarified that the deposited clusters are perfectly wetting on the substrate above the critical incident speed. Employing the energy conservation law, we estimated the critical value of the incident speed at which the evaporation begins to occur during the impact. The estimated critical value, $V_{\text{imp}} = 1.7\sqrt{\epsilon/m}$, exhibits good agreement with our simulation results of $\xi$. Using the energy conservation law, we also found that the scaled cluster adsorption parameter is independent of the cluster size and is proportional to $V_{\text{imp}}^2$. These results exhibit good agreement with our simulation results. We performed the Fourier analysis of $g(\psi)$ and found that some modes become unstable for $3.0\sqrt{\epsilon/m} < V_{\text{imp}} < 4.0\sqrt{\epsilon/m}$. We also performed the molecular dynamics simulation of the A-C case and we found that the A cluster does not become a monolayer film on the C surface. Thus, we concluded that not only the incident speed, but also the strength of the interaction between the cluster and
the substrate is important to form a monolayer film on a substrate.

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