# Computer simulation of vapor-liquid phase separation in two- and three-dimensional fluids: Growth law of domain size

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The late-time growth law of the vapor-liquid phase separation in two- and three-dimensional one-component fluids has been investigated by molecular dynamics simulations in which instantaneous quenching of fluid composed of 50000 (2D) or 78732 (3D) Lennard-Jones particles into the corresponding two phase (spinodal) region has been performed. The power law growth of the characteristic length scale, i.e.,  $l(t) \sim t^a$  is observed in both the 2D and 3D systems in the late stage. All molecular dynamics simulations which we have carried out, except for the cases in which the scaling regime has not been reached in simulation time, confirm that the asymptotic growth exponent is  $\frac{1}{2}$  in both two and three dimensions. It is also found that thermal noise has no effect on the growth exponent but gives rise to a substantial delay of the transition time to the asymptotic regime. This work provides temporal domain patterns associated with the phase separation processes suggesting that the domain structures are sensitive to the temperature of the system.

## I. INTRODUCTION

The dynamical aspects of phase separations are not very well understood. Many experimental, theoretical, and numerical approaches have been applied to this problem.<sup>1</sup> In experimental studies, metal alloys and binary fluid mixtures in the vicinity of the critical point have been extensively investigated by x-ray and neutron small-angle scattering experiments; the time scales of the phase separations are comparable to the experimental time scales in such systems. These studies related to the spinodal decomposition have been reviewed in Refs. 1 and 2.

Lifshitz and Slyozov have theoretically analyzed the dynamics of aggregation of solutes from supersaturated dilute solutions using a scaling concept.<sup>3</sup> They found that the late-time behavior of the growth law of the solute domain scales as  $l(t) \sim t^{\frac{1}{3}}$ , where l(t) is the characteristic length of the domain size. This relation is known as the Lifshitz-Slyozov rule and agrees well with x-ray scattering experiments on spinodal decomposition.

Numerical studies have also been done for phase separation problems by Monte Carlo (MC) simulations for kinetic Ising models. Two types of kinetic Ising models are studied. One is the Glauber model in which the order parameter is not conserved, and the other is the Kawasaki model in which the order parameter is conserved. The former model is considered to be a good model for magnets, and the latter is good for metal alloys. The MC studies for these models are summarized in Refs. 1 and 4. It has been found that both the Glauber and the Kawasaki models show power law growth of domain size as  $l(t) \sim t^a$ , and the growth exponent a is found to be  $\frac{1}{2}$  for the Glauber model and  $\frac{1}{3}$  for the Kawasaki model. The growth law for the Kawasaki model agrees with the Lifshitz-Slyozov rule.

A different type of numerical studies has been applied to phase separation problems based on either of the following two phenomenological equations, i.e., the timedependent Ginzburg-Landau (TDGL) equation which is the case for nonconserved order parameter,

$$\frac{\partial \psi(\mathbf{r},t)}{\partial t} = -L \frac{\delta H[\psi(\mathbf{r},t)]}{\delta \psi(\mathbf{r},t)},\tag{1}$$

or the Cahn-Hilliard-Cook (CHC) equation which is the case for conserved order parameter,

$$\frac{\partial \psi(\mathbf{r},t)}{\partial t} = L \nabla^2 \frac{\delta H[\psi(\mathbf{r},t)]}{\delta \psi(\mathbf{r},t)},\tag{2}$$

where  $\psi(\mathbf{r}, t)$  is the order parameter of the system at point  $\mathbf{r}$  and at time t, L is a phenomenological parameter, and  $H[\psi(\mathbf{r}, t)]$  is the coarse-grained free-energy functional given by

$$H[\psi(\mathbf{r},t)] = \int d\mathbf{r} \left[ \frac{1}{2} (\nabla \psi)^2 - \frac{\tau}{2} \psi^2 + \frac{g}{4} \psi^4 \right], \qquad (3)$$

with temperature-dependent phenomenological parameters  $\tau$  and g which are positive. The late-time growth behavior of phase separations has been discussed by solving these equations numerically. Oono and Puri have proposed an efficient computational method to solve TDGL and CHC equations using discrete space and time.<sup>6</sup> This numerical method is called the cell dynamical system (CDS). Using the CDS, it is found that the average domain size l(t) shows a power law growth as  $l(t) \sim t^a$ 

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in the late stage, and the exponent a is found to be  $\frac{1}{2}$  for the TDGL equation and  $\frac{1}{3}$  for the CHC equation, respectively. These results coincide with the Monte Carlo results for the Glauber and the Kawasaki models. Thus, it is believable that the growth exponent is  $\frac{1}{2}$  for the case of nonconserved order parameter and  $\frac{1}{3}$  for the case of conserved order parameter.

Recent progress of computers makes it possible to study phase separation phenomena by molecular dynamics (MD) simulation. The advantage of MD simulation is that both static and dynamic correlations and hydrodynamic effects are all taken into consideration. The hydrodynamic effects are known to be important in the late stages of the phase separation dynamics of fluid systems such as fluid binary mixtures and polymer melts.<sup>7–12</sup> Velasco and Toxvaerd<sup>14</sup> have carried out MD simulations for two-dimensional binary fluid mixtures and found that the growth exponent is  $\frac{1}{2}$  in the early stage, and that a crossover to a higher exponent takes place in the late stage. Ma et al.<sup>13</sup> have also performed MD simulations for three-dimensional binary fluid mixtures and found that the growth exponent is 0.55. Both MD results have shown that the growth exponent is clearly larger than  $\frac{1}{3}$ , which is predicted by MC studies or the CHC equation, which ignore the hydrodynamic effects.

In the case of a one-component fluid, the characteristic time scale relevant to the vapor-liquid phase separation is too short to observe experimentally. Koch *et al.*<sup>15</sup> have studied the dynamics of vapor-liquid phase separation in a two-dimensional one-component fluid by MD simulations using a 5041-particle system. They have found that the characteristic length scale of the system grows in proportion to  $t^{\frac{1}{2}}$  for the isothermal simulation and in proportion to  $t^{\frac{1}{3}}$  for the adiabatic simulation. They have analyzed their simulation results by a similar way to Lifshitz and Slyozov and concluded that the growth exponent of  $\frac{1}{2}$  obtained by their isothermal simulations is independent of the system dimensionality.<sup>15,16</sup>

In this study, we investigate more accurately the dynamics of vapor-liquid phase separation in two- and three-dimensional fluids via MD simulations with a 50 000-particle system for two dimensions and with a 78732-particle system for three dimensions. The main purpose of the present work is to examine the analysis of Koch et al. on the isothermal phase separation, which stated that the growth exponent is  $\frac{1}{2}$  and independent of the system dimensionality. First, we check their isothermal simulation results on a two-dimensional fluid by adopting a much larger system size. Then we study the growth law dynamics in a three-dimensional fluid both by MD simulation. If we can show that the growth exponents are  $\frac{1}{2}$  in two and three dimensions, it strongly suggests that their analysis is correct. In the adiabatic condition (the exponent is  $\frac{1}{3}$  in Koch's simulation), the separation dynamics is supposed to be more complicated than the isothermal case. Since the system temperature increases with time because of the latent heat, the free-energy functional has a time dependency; it corresponds to the case in Eq. (3) that the parameters  $\tau$  and q are not constant during the separation process. In this paper, we treat only the isothermal case.

## **II. SIMULATION METHODOLOGY**

In order to investigate the late-time growth behavior of phase separations by computer simulations, we have to use a quite large system because the characteristic length scale, i.e., the average size of the domains, easily becomes comparable to the system size as the separation grows. In this work, we use a 50 000-particle system for a two-dimensional fluid and a 78 732-particle system for a three-dimensional fluid. Because of the finite system size, simulations are stopped when the characteristic length scale l(t) breaks the condition of  $l(t) < \frac{L_c}{2}$  for the threedimensional system and of  $l(t) < \frac{L_c}{2}$  for the threedimensional system, where  $L_c$  is the length of the simulation cell. For both two- and three-dimensional systems, each particle interacts through the Lennard-Jones (L-J) potential within the cutoff radius  $r_{\rm cut} = 2.7\sigma$ ,

$$\phi(r) = 4\epsilon \left[ \left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^6 \right].$$
(4)

The periodic boundary condition is used as usual. We have used the leapfrog algorithm to integrate Newton's equations of motion with a time step of  $0.01\tau$ , where  $\tau \ (= \sqrt{\frac{m\sigma^2}{4\epsilon}})$  is the unit time of the L-J fluid, which is equal to 1.067 ps for an argonlike fluid. The cell index method is employed together with the list vector to save the computation time on a vector processor (Cray Y-MP2E). Since the numerical method for molecular dynamics simulation is available in the literature,<sup>19</sup> we do not repeat it.

The phase diagrams of the two- and three-dimensional L-J fluids are presented in Refs. 17 and 18. Table I summarizes the critical point and the triple point constants for these model fluids. Our MD simulations are carried out by the instantaneous quenching of homogeneous systems, which are initially equilibrated at the supercritical (one phase) region, into an unstable (two phase) region. We have performed four experiments (A–D) for the two-dimensional (2D) fluid and four experiments (E–H)

TABLE I. Critical and triple point constants for two- and three- dimensional Lennard-Jones fluids.

Dimension	$T_{c}^{*}$	$\rho_{c}^{*}$	$T_t^*$	Source	Method
2	0.56	0.325	0.41	Ref. 17(a)	Theory <sup>a</sup>
	0.533	0.335	0.415	Ref. 17(a)	MC
			0.40	Ref. 17(b)	MC
3	1.36	0.36	0.68	Ref. 18(a)	MC
	1.35	0.35		Ref. 18(b)	MD
	1.313	0.310	0.69	Ref. 18(c)	MD, MC

<sup>a</sup>Perturbation theory and cell theory.

for the three-dimensional fluid under the following conditions, so that we can investigate the effects of the system dimensionality and the thermal noise on the phase separation dynamics.

# A. 2D fluid $(T_c^* \approx 0.56)$

Exp. A. The system is initially equilibrated at the density  $\rho^* = 0.325$  and the temperature  $T^* = 0.8$ , corresponding to the one phase region in the phase diagram of the two-dimensional L-J fluid. Then, the system is quenched into  $T^* = 0.41$  (the triple point temperature) by the instantaneous velocity scaling of all particles, while the density remains unchanged.

*Exp.* B. The same as Exp. A, but the system is quenched into  $T^* = 0.45$ .

*Exp.* C. The same as Exp. A, but the system is quenched into  $T^* = 0.48$ .

Exp. D. The same as Exp. A, but the system is quenched into  $T^* = 0.50$ .

## B. 3D fluid $(T_c^* \approx 1.35)$

*Exp.* E. The system is initially equilibriated at the density  $\rho^* = 0.35$  and the temperature  $T^* = 1.8$ , corresponding to the one phase region in the phase diagram of the three-dimensional L-J fluid. Then, the system is quenched into  $T^* = 0.7$  (just above the triple point temperature), while the density remains unchanged.

*Exp.* F. The same as Exp. E, but the system is quenched into  $T^* = 1.0$ .

*Exp.* G. The same as Exp. E, but the system is quenched into  $T^* = 1.05$ .

*Exp. H.* The same as Exp. E, but the system is quenched into  $T^* = 1.1$ .

In all the above simulations, the temperature is kept constant during each simulation run by the constraint isotherm method,<sup>19,20</sup> which generates a trajectory on the constant-NVT (canonical) ensemble. In Ref. 14 one can find a discussion on a comparison of some isotherm methods for the phase separation dynamics of binary liquid mixture. It seems that the growth dynamics is not sensitive to the choice of the isotherm method. The phase separation begins immediately after the quenching by the mechanism of spinodal decomposition. Since we are interested only in the late-time growth behavior of the vapor-liquid phase separation, we did not analyze the data in the early stage of the separation process. The time-dependent (temporal) structure factor of the system is calculated throughout the simulation runs by

$$S(k = |\mathbf{k}|, t) = \frac{1}{N} \left[ \left( \sum_{i=1}^{N} \cos \mathbf{k} \cdot \mathbf{r}_{i} \right)^{2} + \left( \sum_{i=1}^{N} \sin \mathbf{k} \cdot \mathbf{r}_{i} \right)^{2} \right], \quad (5)$$

where N is the total number of particles,  $\mathbf{r}_i$  is the position vector of the *i*th particle, and  $\mathbf{k}$  is a wave vector: (2D)

$$\frac{L_c}{2\pi}\mathbf{k} = (1,0), (0,1), (1,1), (1,-1), (0,2), \dots$$

(3D)

$$\frac{L_c}{2\pi}\mathbf{k} = (1,0,0), (0,1,0), (0,0,1), (1,1,0), \dots$$

The first moment of the wave number  $k_1(t)$  was evaluated by

$$k_1(t) = \frac{\int_0^{k_{\text{cut}}} k\tilde{S}(k,t)dk}{\int_0^{k_{\text{cut}}} \tilde{S}(k,t)dk},\tag{6}$$

which measures a characteristic wave number of the system. Here  $k_{\rm cut}$  is an appropriate cutoff wave number, and  $k_{\rm cut} = \pi$  is adopted in this work.  $\tilde{S}(k,t)$  is the "macroscopic" structure factor defined by

$$\tilde{S}(k,t) = S(k,t) - S^{eq}(k), \qquad (7)$$

where  $S^{eq}(k)$  denotes the equilibrium structure factor for a fully segregated macroscopic two phase system. The equilibrium structure factor  $S^{eq}(k)$  was obtained by quenching to the point across the coexistence curve and waiting for the system to reach an equilibrium. The first moment of the wave number  $k_1(t)$  is regarded as a length proportional to the inverse of the characteristic length scale, i.e.,  $l(t) = \frac{2\pi}{k_1(t)}$ .

#### **III. RESULTS AND DISCUSSION**

#### A. Two-dimensional fluid

Temporal sequences of atomic configurations after the quench for the experiments A, B, C, and D are shown







FIG. 1. Snapshots of the simulation experiment A  $(T^* = 0.41)$ . The labels refer to simulation time in units of  $\tau$ .





100



300

FIG. 2. Snapshots of the simulation experiment B  $(T^* = 0.45)$ . The labels refer to simulation time in units of  $\tau$ .

t = 30





FIG. 3. Snapshots of the simulation experiment C  $(T^* = 0.48)$ . The labels refer to simulation time in units of  $\tau$ .

### t = 30







500

FIG. 4. Snapshots of the simulation experiment D  $(T^*=0.50)$ . The labels refer to simulation time in units of  $\tau$ .

TABLE II. Summary of simulation runs for the phase separation experiments for a two-dimensional fluid.

		Density	Temperature	Time	
Exp.	N	$ ho^*$	$T^*$	au	$\mathbf{Exponent}^{\mathtt{a}}$
Α	50000	0.325	0.41	300	$0.499 \pm 0.008$
в	50000	0.325	0.45	300	$0.467 \pm 0.030$
$\mathbf{C}$	50000	0.325	0.48	600	ь
D	50000	0.325	0.50	500	ь

<sup>a</sup>Evaluated by least square fitting using data for  $k_1 < 0.28$ .  $\pm$  on the listed exponent values shows the accuracy of the data fitting. It does not mean the accuracy of present the simulations.

<sup>b</sup>Asymptotic regime has not yet been reached.

in Figs. 1, 2, 3, and 4, respectively. One can see that the phase separation begins immediately after the quench and the domain size increase with simulation time in all the experiments. However, there is a notable difference between domain structures. We observe that the roughness of the vapor-liquid interface increases with increasing annealing temperature of the system. This is attributed to the following effects: the surface tension decreases, and the thermal noise increases with increasing temperature.

The time evolution of the structure factor for the experiment A is shown in Fig. 5 as a typical example. One can observe that the peak position shifts to a smaller k value and its height increases with simulation time. This shift of the peak position indicates the growth of the domain size according to  $l(t) \sim \frac{1}{k(t)}$ . In Fig. 6, we have plotted the first moment of the wave number  $k_1(t)$  for



FIG. 6. First moment of the wave number  $k_1(t)$  vs simulation time for the experiments A-D. The growth exponents converge to  $-\frac{1}{2}$  at late stages  $(k_1 < 0.28)$  in experiments A and B. The solid lines show the least square fitting results using data for  $k_1 < 0.28$ . The dashed lines show the slopes of  $-\frac{1}{2}$  and  $-\frac{1}{3}$  for comparison.

each experiment against the simulation time in log-log scale. The growth exponent is given by the negative of the slope. We confirm that power law growth clearly exists in the late-time region  $(k_1 < 0.28 \text{ in Fig. 6})$ , and the asymptotic growth exponent is found to converge to  $\frac{1}{2}$  for both the experiments A  $(T^* = 0.41)$  and B  $(T^* = 0.45)$ .



FIG. 5. Time evolution of the structure factor for the experiment A.



FIG. 7. Plots of the scaled structure factor for the experiment A.



FIG. 8. Plots of the scaled structure factor for the experiment B.

For the experiments C ( $T^* = 0.48$ ) and D ( $T^* = 0.50$ ), the asymptotic growth regime has not yet been reached. As we show later, this is also confirmed by our scaling analysis. Table II summarizes the simulation results on the growth exponents for the two-dimensional fluid.

We have tested the scaling hypothesis for  $\tilde{S}(k,t)$ . The scaling hypothesis has been established by experiments on metal alloys and by simulations for the Ising model in



FIG. 9. Plots of the scaled structure factor for the experiment C.



FIG. 10. Plots of the scaled structure factor for the experiment D.

an asymptotic growth regime as follows:

$$F(x) = (k_1)^d \tilde{S}\left(\frac{k}{k_1}, t\right) , \qquad (8)$$

where d is the dimensionality of the system and F(x) is a time-independent universal structure function. The scaled structure factor  $(k_1)^2 \tilde{S}(\frac{k}{k_1}, t)$  obtained from the experiments A, B, C, and D at different times is shown in Figs. 7, 8, 9, and 10, respectively. For the experiments A and B, the data points can be seen to lie on a smooth master curve. Thus, we conclude that an asymptotic growth (scaling) regime has been reached for these two experiments A and B. However, for the experiments C and D, we see that no asymptotic growth regime has yet been obtained within our simulation time.

TABLE III. Summary of simulation runs for the phase separation experiments for a three-dimensional fluid.

		Density	Temperature	Time	
Exp.	N	$ ho^*$	$T^*$	au	Exponent <sup>a</sup>
Е	78732	0.35	0.70	50	$0.529 \pm 0.002$
$\mathbf{F}$	78732	0.35	1.00	70	$0.502 \pm 0.008$
G	78732	0.35	1.05	100	$0.404 \pm 0.025$
Н	78732	0.35	1.10	100	ь

<sup>a</sup>Evaluated by least square fitting using data for  $k_1 < 0.45$ .  $\pm$  on the listed exponent values shows the accuracy of the data fitting. It does not mean the accuracy of the present simulations.

<sup>b</sup>Asymptotic regime has not yet been reached.



Temporal sequences of atomic configurations after the quench for the experiments E and G are shown in Figs. 11 and 12, respectively. As in the case of the two-dimensional fluid, phase separation begins immediately after the start of the quench. The domain size increases with simulation time in all the experiments, and the roughness of the vapor-liquid interface seems to increase with increasing annealing temperature.

B. Three-dimensional fluid

The time evolution of the structure factor for the experiment E is shown in Fig. 13 as an example. In Fig. 14, we have plotted the first moment of the wave number  $k_1(t)$  for each experiment against the simulation time in log-log scale. As in the two-dimensional fluid, power law growth exists in the late-time region  $(k_1 < 0.45 \text{ in Fig. 14})$ , and the asymptotic growth exponent is found to converge to  $\frac{1}{2}$  for the experiments E  $(T^* = 0.70)$ , F  $(T^* = 1.00)$ , and G  $(T^* = 1.05)$ . For the experiment H  $(T^* = 1.10)$ , the asymptotic growth regime has not yet been reached. Table III summarizes the simulation results on the growth exponent for the three-dimensional fluid.

The test of the scaling hypothesis for  $\tilde{S}(k,t)$  of the three-dimensional fluid has also been made. The scaled structure factors  $(k_1)^3 \tilde{S}(\frac{k}{k_1},t)$  obtained from the experiments E, F, G, and H are shown in Figs. 15, 16, 17, and 18, respectively. The data points seem to deviate from each other for the experiments G and H, but almost lie on a smooth master curve for the experiments E and F. Thus, we can conclude that the scaling regime has been reached at least for these two experiments E and F, and the asymptotic growth exponent is  $\frac{1}{2}$  for the three-dimensional fluid as well.

The results of present MD simulations for two- and three-dimensional fluids strongly suggest that the asymptotic growth exponent for the vapor-liquid phase separation is  $\frac{1}{2}$  in both two and three dimensions. This coincides with the simulation result of Koch et al. on a two-dimensional fluid<sup>15,16</sup> and also supports their analysis, which stated that the growth exponent is  $\frac{1}{2}$  and independent of system dimensionality. It is also confirmed that thermal noise has no effect on the asymptotic exponent, but gives rise to a substantial delay of the transition time till the asymptotic regime is reached. A similar result has been obtained from numerical simulation with the CHC equation including thermal noise.<sup>5</sup> These results seem to be consistent with the phenomenon called critical slowing down. We suppose that the failure of some simulations at high temperatures to converge to the  $\frac{1}{2}$  exponent within the simulation time is affected by this effect. The analysis of the domain structures is particularly of interest. It will be discussed elsewhere.

## **IV. CONCLUDING REMARKS**

We have studied the asymptotic growth law of the vapor-liquid phase separation for the two- and threedimensional one-component Lennard-Jones fluids by MD simulations, using a 50 000-particle system for twodimensional fluids and a 78 732-particle system for threedimensional fluids. MD simulations have been carried





FIG. 11. Snapshots of the simulation experiment E  $(T^* = 0.70)$ . The labels refer to simulation time in units of  $\tau$ .



FIG. 13. Time evolution of the structure factor for the experiment E.

out by instantaneous quenching of a homogeneous system, which has been equilibriated at supercritical temperature, into the two phase region. The phase separation begins immediately after the quench of the homogeneous system, and the time-dependent (temporal) structure factors S(k,t) has been calculated throughout the simulation runs. The characteristic length scale has been found to grow as  $l(t) \sim t^a$  in the late-time (scaling) regime, which correspond to  $k_1 < 0.28$  for the two-dimensional fluid and  $k_1 < 0.45$  for the three-dimensional



FIG. 14. First moment of the wave number  $k_1(t)$  vs simulation time for the experiments E-H. The growth exponents converge to  $-\frac{1}{2}$  at late stages  $(k_1 < 0.45)$  in experiments E, F, and G. The solid lines show the least square fitting results using data for  $k_1 < 0.45$ . The dashed lines shows the slopes of  $-\frac{1}{2}$  and  $-\frac{1}{3}$  for comparison.



FIG. 15. Plots of the scaled structure factor for the experiment E.

fluid in this work, and the asymptotic growth exponent was found to be  $\frac{1}{2}$  in both two and three dimensions. This strongly suggests that Koch *et al.*'s analysis is correct. Thermal noise was found to have no effect on the asymptotic exponent, but to give rise to a substantial delay of the transition time to the asymptotic regime.

We have observed that the roughness of the vaporliquid interface increases with increasing annealing temperature in both two- and three-dimensional fluids. This is attributed to the following two effects: the surface tension decreases, and thermal noise increases with increasing system temperature. The atomic configurations ob-



FIG. 16. Plots of the scaled structure factor for the experiment F.



FIG. 17. Plots of the scaled structure factor for the experiment G.  $\,$ 

tained by the MD simulations indicate that the domain structure is notably sensitive to the system temperature. In fact, the domain structures of the two-dimensional L-J fluid have been reported to suggest a fractal nature at  $T^* = 0.45$  (Langevin dynamics simulation)<sup>21</sup> or at  $T^* \approx 0.5$  (constant-*NVE* MD simulation).<sup>22</sup> Analysis of the domain structures for the present simulations will be reported in a forthcoming paper.

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FIG. 18. Plots of the scaled structure factor for the experiment H.

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FIG. 1. Snapshots of the simulation experiment A  $(T^*=0.41)$ . The labels refer to simulation time in units of  $\tau$ .



FIG. 11. Snapshots of the simulation experiment E  $(T^* = 0.70)$ . The labels refer to simulation time in units of  $\tau$ .



FIG. 12. Snapshots of the simulation experiment G  $(T^* = 1.05)$ . The labels refer to simulation time in units of  $\tau$ .





FIG. 2. Snapshots of the simulation experiment B  $(T^* = 0.45)$ . The labels refer to simulation time in units of  $\tau$ .









FIG. 3. Snapshots of the simulation experiment C  $(T^* = 0.48)$ . The labels refer to simulation time in units of au.



FIG. 4. Snapshots of the simulation experiment D  $(T^* = 0.50)$ . The labels refer to simulation time in units of  $\tau$ .