Thin Film Instability and Nanostructure Formation: a Molecular Dynamics Study

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ABSTRACT

One of the recent studies showed that the structures can be self-constructed by controlling the instability of thin fluid film of a nano-scale [S.Y. Chou and L. Zhuang, , *J. Vac. Sci. and Technol. B*, **17**, 3197-3202 (1999)]. The Molecular dynamics simulation is used to probe the phenomena. And two sources of instability are investigated: the normal temperature gradient and the long-range attractive potential by the wall. In the first case, the wall temperature is controlled so that the film maintains its temperature gradient. It is observed that the growth rate decreases in a monotonic fashion. The effect of the surface tension gradient, rather than evaporation, may drive the growth. Another source of instability is considered that is the interaction between the upper wall and the fluid film through a potential originating from the interaction between ion and non-polar molecule. The solid potential induces the formation of one or more vertical structures. This may result from the distortion of the pressure distribution.

Keywords: molecular-dynamics simulation, thin film, nanostructure

1 INTRODUCTION

Recently it is the subject of intense researches to build small structures of a scale less than 100 nm. Manufacturing the nanostructures with efficiency and low cost has great significance in the application of the emerging small-scale technologies; the good examples are the electronic circuits integrated in an increasingly smaller dimension or the nano-electro-mechanical systems. One of the recent studies showed that the structures can be self-constructed by controlling the instability of thin fluid film of a nano-scale[1,2]. The processes involve several physical elements, which may determine the characteristics like the structure arrangements and formation time. Previous studies on the subject rely on the instability theory of continuous media based on the long-wave approximation [3]. In this study, the Molecular dynamics simulation is used to probe the thin fluid film on a substrate and its dynamical behavior during destabilization and structure formation. Two sources of instability are investigated: one is the temperature gradient applied in the normal direction and the other is an attractive potential by the wall in the distance.

2 RESULTS

2.1 Simulation Methods

Non-equilibrium molecular dynamics simulation is conducted [4]. The simulation system consists of the argon atoms confined by two insulated platinum walls. The argon atoms are equilibrated to form a thin fluid film on a platinum wall. The size of the computational domain is W*2.4*29.4 nm^3, where that of width, W, is varied from 2.4 to about 136 nm. The depth is set to be a smallest size possible, 7.06 nm, to minimize the state variation in the direction as in the Hele-Shaw Cell. The argon atom interacts with other argon and platinum atoms through 12-6 Lennerds-Jones potential.

$$u(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \quad (1)$$

u is the interaction energy between two molecules that are separated by the distance of r when they have the energy parameter ε and intermolecular diameter σ. And the interaction is cut off in the distance of 1.02 nm. The walls are of a f.c.c. lattice in which the platinum atom interacts with others with a harmonic potential. The temperature of the system is controlled by the particles embedded in the farthest rows in the walls from the inside. They observe the Langevin Equation whose parameters are given according to a known phonon characteristics [5].

2.2 Instability by temperature gradient

The temperature of the wall the thin film is on is controlled to induce the instability. And the film maintains its temperature gradient of about 150.4 degrees per nanometer in a relatively constant manner except for an initial time (~0.5 ps), when it increases up to about 225.2 degrees per nanometer. These are about the maximum possible amount in the given system. A more amount of gradient induces the evaporation at the solid-liquid interface. The growth rate of the fluctuations in the early stages (0.3<

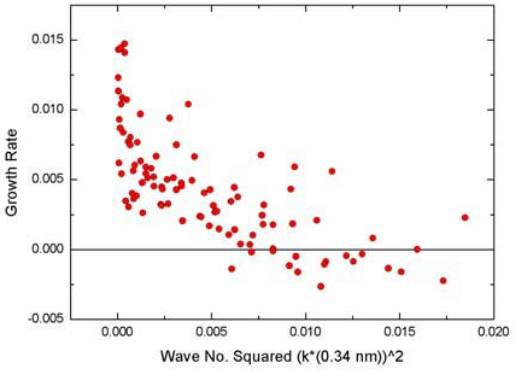

Figure 1. Growth rate of fluctuations vs. wave no. squared

t <0.75 ps) is observed. According to the continuum theory based on the long-wave approximation, the growth rate has a parabolic behavior with a peak with respect to the wave number squared. In the measured results in Fig. 1, it decreases in a monotonic fashion. Two thing may contribute to this discrepancy: one is from the fact that the major group of waves in the figure are not appropriate for the long-wave approximation. The other is that the amplitude of capillary waves in the nano-scale may increase during the initial stage. And the growth of the lower wave numbers continue in the later while that of the higher quickly decays. This instability may mainly come from the surface tension gradient, due to the temperature, with respect to the interfacial position. The vapor-coil effect from the evaporation may not contribute any noticeable effect. Finally, even with the large temperature gradient applied, any fluctuation does not grow up to an enough height to build a structure with the wall in the distance.

2.3 Instability by solid potential

Another source of instability is considered in the context of the structure formation. The interaction of a longer range than that of Van der Waals kind between the upper wall and the fluid film may have the significant effect on the film evolution. This contribution is implemented as a solid potential acting on the fluid atoms. The potential

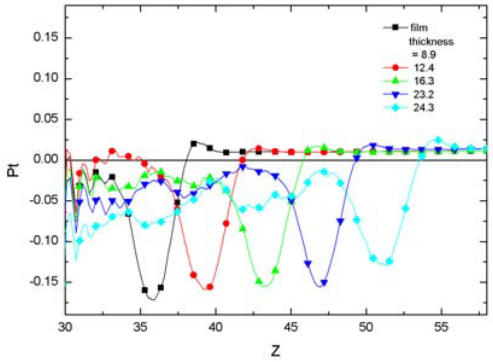

Figure 3. Tangential pressure distribution according to the interfacial postion (ε_{lr} =1; The data are smoothed with 10 points by Savitsky and Golay's method. All scales are reduced in Argon's units)

relevant for the interaction between ion and non-polar molecule is integrated in the solid domain to give the following expression.

$$potential(z_o) = \varepsilon_{lr} \frac{\pi}{z_o} \rho \qquad (2)$$

ε_{lr} is the proportionality that represents the strength of the interaction. z_o is the distance between the argon atom and the solid surface. ρ is the mean number density of the solid. The coulomb interactions among fluid atoms are assumed to of a negligible influence compared to the Van der Waals. This case simulate the system where the upper wall is composed of ionic atoms and induces the dipoles on the non-polar fluid atoms. In the given values of ε_{lr} (=0.5~2), it is observed that the solid potential induces the formation of one or more vertical structures. (Fig. 2) This may be the direct result of the distortion of the pressure distribution. (Fig. 3) As the position of the liquid-vapor interface gets close to the upper wall, the tensional stress (the negative pressure) in the direction tangential to the interface become stronger. This phenomenon is usually

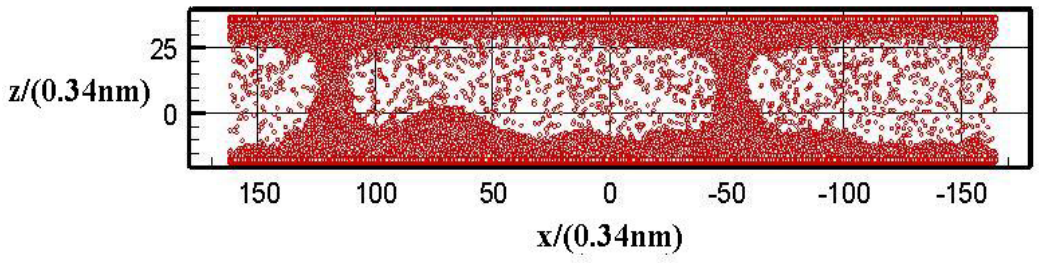

Figure 2. Vertical structure formation by solid wall potential (ε_{lr} =1, t=0.4 ns)

treated as the concept of "disjoining pressure" in the continuum model.

The time that it takes for the fluctuation to reach the upper wall for the first time tends to decrease as the strength of the potential increases. (Fig. 4) The linearly decreasing dependence of the slope in the figure corresponds to the prediction of the continuum model that is based on the long-wave approximation. On the other hand, it can be observed that the spacing of neighboring structures is inversely proportional to the strength. However, more quantitative result for the trend is not pursued because of the limitation in the computational size of the problem.

3 CONCLUSION

In this work, the instability of thin fluid film and the nanostructure formation is studied by the Molecular dynamics simulation. Liquid argon film is located on the platinum substrate and two physical conditions are applied to induce the vertical structures that are observed in the literatures. The primary findings are summarized as follows

1) When a large thermal gradient is applied to the fluid film of a nano size, the instability that is observed in the larger scale is still realizable. And the surface tension gradient due to the temperature, that is, the thermocapillary effect, was the major source in the simulation.

2) It is shown that the interaction energy of the solid whose range is longer that the Van der Waals is very effective in inducing the instability of the thin film and vertical structures formation between the solid walls. The formation time is inversely proportional to the strength of the solid potential.

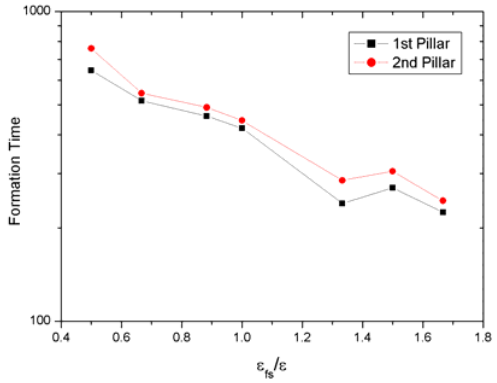

Figure 4. Formation time of the vertical structure vs. the relative strength of the solid potential

4 ACKNOWLEGEMENT

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REFERENCES

[1] S.Y. Chou and L. Zhuang, J. Vac. Sci. Technol. 17, 3197, 1999.

[2] E. Schaffer, T. Thurn-Albrecht, T.P. Russell, and U. Steiner, Nature 403, 874, 2000.

[3] L. Wu, and S.Y. Chou, Appl. Phys. Lett. 82, 3200, 2003.

[4] D.C. Rapaport, "The Art of Molecular Dynamics Simulation," Cambridge, 1995.

[5] M.P. Allen and D.J. Tildesley, "Computer Simulation of Liquids," Oxford, 1987.