*Journal of Nano- and Electronic Physics*
Vol. 8 No 1, 01028(7pp) (2016)

*Журнал нано- та електронної фізики*
Том 8 № 1, 01028(7cc) (2016)

# Atomistic Simulation of Properties of an Ultrathin Layer of Liquid Argon Compressed Between Diamond Surfaces

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(Received 04 January 2016; revised manuscript received 11 March 2016; published online 15 March 2016)

Using the method of classical molecular dynamics we investigate the properties of an ultrathin film of liquid argon, which consists of one or two layers of molecules and is confined by two atomically smooth crystalline diamond surfaces. The aim of the research is validating the use of rigid surfaces and one of the available models of the argon molecule. We study the behavior of the equilibrium and dynamic characteristics of the system. It is shown that with increasing external load the transition of the film in the solid-like state occurs, which is indicated by the behavior of the velocity autocorrelation function of argon molecules, reduction of the magnitude of the diffusion coefficient and the shear viscosity increase. The organization of molecules in layers and the presence of their in-plane ordering are revealed. The dependences of the kinetic friction force on time and load are obtained. The results are compared with experimental data.

Keywords: Molecular dynamics, Tribology, Boundary friction, Ultrathin argon film, Computer experiment, Interatomic interaction potential.

DOI: 10.21272/jnep.8(1).01028 PACS numbers: 46.55. + d, 61.20.Ja, 61.20.Ne, 61.30.Hn, 68.35.Af, 81.40.Pq

## 1. INTRODUCTION

Friction plays a key role in various systems and phenomena, which, at first sight, may seem unrelated with each other, but which at closer examination reveal the general features inherent in all tribological processes in the fields of technology, geology, and biology [1-18]. The development of wear-resistant surfaces with low friction and thin lubricant films has become an important factor in the miniaturization of moving parts in many technical devices. The latter includes micro-electromechanical systems, memory devices, miniature motors, etc. Simple empirical friction laws do not always work in such systems. This is because they are characterized by a high surface-to-volume ratio; and chemistry of surfaces, adhesion and structure (or roughness) of the latter are more important for them. Conventional tribological methods applied for macroscopic objects may turn out to be ineffective on a nanometer scale that requires new approaches to control the study of similar systems. The field of tribological investigation of the biosystems and also lubricant mechanisms in the joints is rapidly developed.

There are two different modes, namely, hydrodynamic (fluid) friction and boundary lubrication (or boundary friction) [2, 3]. In the first case, surfaces are separated by a thick (approximately more than 0.01 mm) liquid lubricant film. In the second case – by a thin lubricant film (of the thickness of several atomic diameters). For the boundary friction, the experimentally measured friction is much higher, for example, $10^2$ or even $10^5$-$10^7$ times, than the hydrodynamic friction. It depends, to a greater extent, not on the lubricant viscosity, but on its chemical composition.

Real and computer experiments indicate the unusual properties of boundary lubricants, which can be briefly summarized as follows [1-6, 8-23]:

– molecules of liquids compressed between two atomically smooth rigid surfaces become more ordered and tend to form layered structures. The density of a liquid in an ultrathin film is non-uniform and has an oscillating profile. The oscillation period is close to the value of the molecular diameter and reflects the forced ordering of the molecules into quasi-discrete layers;

– the mobility of the molecules in ultrathin films is significantly reduced compared with bulk liquids. This is manifested in decreasing the diffusion coefficient and increasing the shear viscosity and molecular relaxation times;

– the boundary lubrication can be divided into two kinds of the reaction to shift: a liquid-like, at which the liquid flows when shear stress is applied, and a solid-like characterized by the presence of yield limits. In a solid-like film, the molecules can be ordered both perpendicularly and parallel to the plates. The behavior of the films in a solid-like state is more reminiscent of two-dimensional nematics or ductile solids, which undergo plastic deformation. In addition, unlike the bulk state of matter in such films, the positional and orientational ordering of the molecules is caused not only by the interaction of the liquid molecules with each other, but also by the strong proximity of two surfaces;

– for velocities and temperatures below some critical values, the stick-slip effect can be observed, for which the movement of the movable surface occurs as a series of alternating stops and slippages. The stick-slip mode is a sign of the presence of a solid-like lubricant state and is considered one of the main causes of wear of the friction surfaces [1-4].

The aim of this work is the study of the properties of ultrathin argon films compressed between atomically-smooth diamond surfaces and the comparison with the behavior of an ultrathin water film [12].

## 2. BASIC PRINCIPLES OF MOLECULAR DYNAMICS

### 2.1 General concepts

Theoretical models and modeling, which are called a computer experiment, help in the interpretation of experimental data and provide prediction of events, which can be eventually confirmed or refuted by experiments. Among these methods, one can emphasize analytical models and large-scale modeling by the molecular dynamics (MD) method [24-30].

 

Modeling by the MD method is a compromise between the analytical models and experimental conditions. Thus, MD modeling is based on approximate interatomic forces and classical dynamics that is close to approximations of analytical models. In addition, computer experiments can discover unexpected phenomena, which require further explanation. This resembles the situation that occurs in real experiments. Moreover, a wrong choice of modeling conditions can lead to a senseless result, which also takes place in experimental studies.

Atomistic computer modeling at first glance seems rather simple, namely, having a set of initial conditions and a way of describing the interatomic forces, it is simply necessary to integrate the Newtonian classical equations of motion with one of several standard methods. Modeling gives new relative atomic locations, velocities and forces, and it is possible to monitor the behavior of atoms in real time using images-animations.

However, the efficient use of MD modeling to study friction on the atomic scale requires understanding of many details, which are evident in this simple analysis. Let us consider some of them.

### 2.2 Interatomic interaction potentials

As it was stated, MD modeling is computer programs, in which Newtonian equations of motion are integrated to trace the motion of atoms in time in response to applied forces,

$$F = ma, \tag{2.1a}$$

$$-\nabla E = m(\partial^2 r/\partial t^2), \tag{2.1b}$$

where $F$ is the force acting on each atom, $m$ is the mass of the atom, $a$ is the acceleration of the atom, $E$ is the potential energy of the atom, $r$ is the atomic location, $t$ is the time. At the beginning of the modeling, the forces acting on each atom are calculated. After that, the atoms move for a short period of time $\Delta t$ (the time step) in response to the action of applied forces. This is accompanied by a change in atomic coordinates, velocities, and accelerations. Then, the process is repeated for a certain number of time steps chosen by the user.

The advantage of this method is that it gives the possibility to monitor the individual motion of all atoms in the considered system in real time. The disadvantage is that time scales are very limited (from pico- to nanoseconds). Moreover, the system sizes, which can be considered at the moment, are restricted by $10^8$-$10^{10}$ atoms, which, although are impressive, but still far from real systems containing $10^{23}$ atoms or more. Thus, although the MD modeling is used to study friction on the atomic scale and provide understanding of this process; however it is still limited by spatial and time scales, which are significantly less than the experimental values.

For atomistic modeling of a material, the mathematical expressions of its potential energy ($E$ in (2.1b)) are necessary. There are currently two approaches to choosing the type of potential. Within the first approach, it is assumed that the potential energy of atoms can be presented as a function of only their relative atomic positions [28]. These functions, which are the so-called empirical potentials, are usually based on simplified interpretations of the general principles of quantum mechanics and, as a rule, are composed of a certain number of free parameters. The latter are chosen in such a way that a set of physical properties of the system is reproduced as best as possible. However, there are many difficulties associated with the search for suitable functions of potential energy. For example, the parameters used to adjust the potential energy are determined using a restricted set of known properties of the system. For metals, the properties, under which a function of potential energy is adjusted, may include a lattice constant, cohesive energy, elastic constants, and vacancy formation energy. The consequence is that other properties, including also those, which can be decisive for determining the results of this modeling, are defined by the only foreseeable functional form. The stipulated properties for the metal can include the surface reconstructions, defect energetics and responses (elastic and plastic) to the applied load. Thus, the potential form is key, if the ability to adequately reflect the system physics is required from modeling.

Another approach, which became more general with the availability of powerful computers, is the calculation of interatomic forces directly from the first principles or semi-empirical evaluations implicitly including electrons [24, 25]. The advantage of this approach is that in the general case the number of unknown parameters is less than those of empirical potentials.

Since the calculations of the force are based on quantum principles, the type of interatomic interactions can be quite tolerable between different atomic environments. However, this does not necessarily guarantee that the forces from calculations of the semi-empirical electronic structure are precise. Inadequately selected parameters and functional forms can also give wrong results. The disadvantage of this approach is that the used potentials require significantly higher computational efforts than those utilized in the empirical potential functions. A lot of time on modeling causes the smaller available sizes and time scales of the studied systems than when using empirical potentials. For example, the largest systems, which can currently be studied by using the first principles and semi-empirical MD modeling, are composed of several hundreds and several thousands of atoms, respectively. Thus, these methods have not yet become widely used in large-scale MD modeling.

The simplest approach to construct a continuous potential energy function is the assumption that the binding energy $E_b$ can be written as the sum of all pairs of atoms [28, 30]

$$E_b = \sum_i \sum_{j>i} V_{pair}(r_{ij}), \tag{2.2}$$

where indexes $i$ and $j$ are the marks of atoms, $r_{ij}$ is the scalar distance between them, $V_{pair}(r_{ij})$ is the suspected functional form for the energy. Some traditional forms for the potential are given by the expression:

$$V_{pair}(r_{ij}) = D \cdot X(X-1), \tag{2.3}$$

where parameter $D$ determines the energy minimum for pairs of atoms. Two general forms of this expression are presented by the Morse potential ($X = e_{ij}^{-\beta r}$) and Lennard-Jones (LJ) «12-6» $X = (\sigma/r_{ij})^6$. Here, $\beta$ and $\sigma$ are the arbitrary parameters used to adjust the potential to the observed properties. The short-range exponential form of the Morse function provides a reasonable description of the repulsion forces between atomic nuclei, while the term $1/r^6$ in the LD potential describes the main term of the long-range dispersion forces.

### 2.3 Thermostats

Usual MD differs from most experimental studies by the fact that energy $E$ and volume $V$ are constant in the modeling unlike temperature $T$ and the pressure $P$. In terms of statistical mechanics, usual MD gives average with respect to the microcanonical ensemble $NVE$ ($N$ is the number of molecules), while experiments with constant temperature correspond to the canonical ensemble $NVT$ [28, 30].

To perform simulations close to the experiments, it is necessary to use the canonical ensemble to maintain a constant temperature. Since external forces do work on the system, then the constant temperature maintenance means the way how to extract excess heat or connect the system and thermostat. In real bodies, heat extraction occurs due to many mechanisms, for example, phonon excitation or electron-hole pair generation, whose energy is converted into heat [1]. In modeling, it is possible both to explicitly introduce methods of heat extraction and do not do this. For example, for the system studied here, the electronic thermal conductivity cannot be taken into account due to the excellent dielectric properties of the diamond, and for metals, contribution of the electronic thermal conductivity cannot be neglected.

Constant temperature is maintained in the canonical ensemble by using one of a large number of thermostats, some of which are described below. As a rule, in modeling of indentation and friction, the thermostat is applied to the area of modeling cell, which is rather far from the boundary, where friction and indentation occur. In this case, although there is a local heating of the surfaces as a result of the work on the system, however, the excess heat is effectively dissipated for the system as a whole. One can assume that such modeling is carried out in the mixed $NVE$/$NVT$ conditions, which, strictly speaking, do not correspond to any real thermodynamic ensemble, but are found to be useful and widely used.

The simplest approach to control the system temperature is simply to periodically re-scale atomic velocities to achieve the desired temperature [26]. This method is widely used in early MD modeling and often found to be efficient to maintain this temperature during modeling. However, it has several disadvantages, which led to the development of more sophisticated methods. For example, there are little theoretical justifications for re-scaling velocities. For typical system sizes in MD modeling, the average quantities, such as pressure, do not correspond to the quantities obtained from any thermodynamic ensemble. Moreover, the resulting dynamics is irreversible in time that contradicts to classical mechanics. Finally, the velocity and energy dissipation mode are not determined by the system properties, but rather depend on how often atomic velocities are re-scaled. This can influence the system dynamics.

The Langevin dynamics is a more sophisticated method to maintain the system temperature [1, 27-29]. At first, this technique was used to describe the Brownian motion and was widespread in MD modeling. Within this method, the terms corresponding to friction and random force are added to the equation of motion. The equation of motion for the atoms subordinated to the Langevin thermostat has the following form:

$$ma = F - m\xi v + R(t), \qquad (2.4)$$

where $F$ are the forces acting due to the interatomic potential, $m$ and $v$ are the particle mass and velocity, respectively, $\xi$ is the friction coefficient, $R(t)$ represents a random force acting as a white noise.

It is important to know that, as well as in the case of any thermostat, atomic velocities vary in the process of temperature control by using the Langevin thermostat. This can have the perturbation effect on the dynamical property being studied. One of the approaches, which is efficient for minimization of this problem, is to add the Langevin forces only to atoms located at some distance from the area, in which the processes take place. Then, the simplified approximations can be used for the friction coefficient without excessive influence on the dynamics produced by interatomic forces.

A random force $R$ in (2.4) is usually specified by the Gaussian distribution, in which the choice of the width takes into account the satisfaction of the fluctuation-dissipation theorem. It is determined by the equation:

$$\langle R(0) \times R(t) \rangle = 2mk_B T\xi\delta(t), \qquad (2.5)$$

where $m$ is the particle mass, $T$ is the necessary temperature, $k_B$ is the Boltzmann constant, $t$ is the time, and $\xi$ is the friction coefficient. We note that random forces do not depend on the forces in the previous steps (that is noted by the delta function), and the width of the Gaussian distribution, from which the random force results, is changed with temperature. The Langevin approach presented above does not require any feedback from the current system temperature. Random forces are determined solely from the expression (2.5).

In the work, the non-equilibrium equations of motion are used to control the temperature or, as they say, the mechanical restrictions (or boundary conditions) are imposed [9, 12].

## 3. MODEL

### 3.1 General scheme of the computer experiment

To find a model of the tribological system, which would explain the properties of an ultrathin argon film, the computer experiments described in this paper are conducted. Let us consider their general scheme.

The modeling was performed for a planar Couette geometry, which resembles experimental systems studied using the surface forces apparatus [2, 10]. An ultrathin argon film limited by two solid walls with periodic boundary conditions in the plane of the plates applied to both the liquid and surfaces was considered. The atomically-smooth plates were studied. In the first case, each wall contains 1152 carbon atoms, which form two crystalline surfaces with the diamond lattice. To simplify the task, the plates are considered to be absolutely rigid, and the model does not include the surface elasticities [12]. But, given that diamond is one of the most solid materials, it is decided in the paper to check this approximation.

The working program contains the code executing the calculation and the interface allowing to directly observe the 3D image animation of the system in real time and is written in the Microsoft VisualStudio 2008 environment using the Microsoft Visual C++ 8.0 compiler.

The films were studied at the initial moment with the content of two and one molecular layers, 196 molecules

in each layer. The initial configurations of the system are shown in Fig. 3.1.

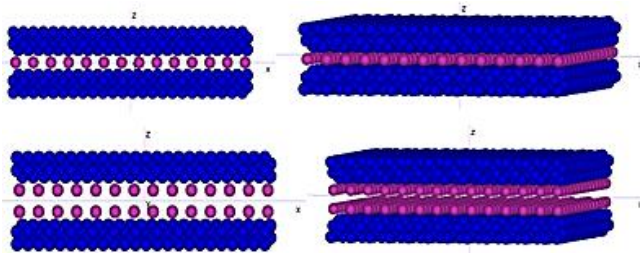

Fig. 3.1 – Configurations of the studied system for an argon film composed of one and two molecular layers at the beginning of the modeling compressed between atomically-smooth surfaces. The carbon and argon atoms are depicted, respectively, by blue and violet balls

To visualize an argon molecule, the calculated value of its radius, which is equal to 71 pm, was used, and the diamond lattice constant was 356.68 pm [30, 31]. Argon molecules at the beginning of the modeling were placed at the vertices of the cubic lattice with the lattice constant corresponding to the value of 942 kg/m$^3$ of the liquid argon density [31]. At that, velocity of each molecule has temperature-dependent fixed values and randomly chosen direction. The initial distance (gap) between the surfaces for one layer is equal to 0.3104 nm and for two layers – 0.6207 nm. The horizontal sizes of the modeling cell along the *x* and *y* axes are the same and make up 42.806 Å. The temperature is 120 K, and the load is modeled by applying a constant force *L* to each atom of the planes along the *z*-axis in the direction of the argon molecules. The shift was also simulated by applying a constant horizontal force $F_s$ to each atom of the upper surface along the *x*-axis [12], and the algorithm of the constant shear force [21] was used in this work.

## 3.2 Model of the argon molecule

We have used the dimensionless MD measurement units convenient for solving the task. We determined the measurement units of the distance $\sigma$ and energy $\varepsilon$. The mass of an argon atom equal to $m = 3.995 \cdot 1.6747 \cdot 10^{-23}$ g is chosen as the mass unit. The physical and dimensionless measurement units are related as follows: $\sigma = 3.4$ Å, $\varepsilon = 120 \cdot 1.987 \cdot 10^{-3}$ kcal/mole; the time unit *t* is equal to $t = \sqrt{m\sigma^2/\varepsilon} \approx 2.161 \cdot 10^{-12}$ s. Since the energy unit corresponds to $\varepsilon/k_B = 120$ K, the typical temperature of 120 K is equal to 1 in the dimensionless form. The time step of $\Delta t = 0.0005$ was used, and in the dimensional units it is approximately $10^{-15}$s.

The value of the force *L* per each atom varied from 20 (in the SI system it is 68.38 pN) to 300 (1.026 nN) that corresponds to the values of pressure on each surface, respectively, from 4.3 GPa to 64.506 GPa. Taking into account that the plasticity threshold of diamonds is of the order of 100 GPa [1], the approximation of absolute rigidity of walls accepted in the model can be assumed reasonable for the considered values of pressure. The horizontal shear force $F_s$ per each atom of the upper plate was changed from 0.5 (1.71 pN) to 100 (0.342 nN) or the total horizontal force on the surface varied from 1.97 nN to 394 nN.

Argon molecules were modeled by elastic balls, the interaction for which has the LD form:

$$u_{ij} = \begin{cases} 4\varepsilon_{Ar}\left[\left(\frac{\sigma_{Ar}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{Ar}}{r_{ij}}\right)^{6}\right], \\ 0, \end{cases} \qquad (3.1)$$

where the first equality holds at $r_{ij} < r_c$, and the second one – at $r_{ij} \geq r_c$; $r_c = 2.38$. Parameters in (3.1) have the following values: $\varepsilon_{Ar} = 1.534$ and $\sigma_{Ar} = 1.078$ [31].

## 3.3 Equations of motion

In this modeling, the classical equations of motion were used. Due to the absolute rigidity of the surfaces, the motion of their center of mass was considered. For the upper wall, the equations are written as

$$\begin{aligned} M\dot{X} &= F_x + \mathrm{F_s} \cdot N_p, \\ M\dot{Y} &= F_y, \\ M\dot{Z} &= F_z + L \cdot N_p, \end{aligned} \qquad (3.2)$$

where *X*, *Y*, *Z* are the coordinates of the center of mass of the upper plate, $N_p$ is the number of atoms of the wall, $M = N_p \cdot m_c$ is the surface mass, $m_c = 0.3$ is the mass of a carbon atom, $F_x$, $F_y$, $F_z$ are the components of the resulting force acting on the surface from argon. They are usually determined as a sum of derivatives (with minus sign) with respect to the corresponding coordinate of the potential (3.1) for all molecules. For the lower wall, equations of motion have the form similar to (3.2), but in the first equation there is no second term in the right-hand side representing the shear force available in (3.2). In the absence of external horizontal forces, the law of conservation of momentum along the *x* and *y* axes should be maintained; the center of mass of the plates should not move in these directions. However, in the modeling under the specified condition, there were observed small displacements of the center of mass (of the order of the diamond lattice constant). They could be caused by the numerical rounding and, probably, effects induced by the way of maintaining temperature constancy.

The translational equations (without taking into account the term responsible for maintaining constant temperature) for the center of mass of the *i*-th argon molecule have the following form:

$$\ddot{r}_i = \sum_j F_{ij} + 48 \sum_{j(\neq i)} \left( {r_{ij}}^{-14} - \frac{1}{2} {r_{ij}}^{-8} \right) r_{ij}, \qquad (3.3)$$

where $\sum_j F_{ij}$ is the force acting on the part of all carbon atoms located from the *i*-th molecule at the distance less than $r_c$ and determined similarly to the force for the surfaces; $\sum_{j(\neq i)}( {r_{ij}}^{-14} - \frac{1}{2} {r_{ij}}^{-8}) r_{ij}$ is the force acting on the *i*-th molecule on the part of other argon molecules, which are located from the *i*-th one not far than $r_c$. The equations of motion for the coordinates of the centers of mass of the plates and molecules were integrated using the predictor corrector algorithm of the fourth order of accuracy.

## 3.4 Measurement features

Two sets of measurements were conducted during the modeling. Some measurements took place in the absence of the shear force $F_s$, and the diffusion coefficient is mea-

sured there. For another set, the shear force was non-zero, and the kinetic friction force acting on the surface was measured here.

To determine the integrated autocorrelation function of the velocity $\varphi(t)$, the following relation [32-36] is used:

$$\varphi(t) = \langle \sum_{j=1}^{Nm} v_j(t) \cdot v_j(0) \rangle, \qquad (3.4)$$

where $N_m$ is the number of argon molecules and $v_j$ is the velocity of the $j$-th molecule. In (3.4), angular brackets denote averaging over a sufficiently large number of independent specimens of the system.

The diffusion coefficient was calculated by using the Einstein formula, which for a large (compared with the "time of collisions") time $t$ has the form of [32-35],

$$D = \lim_{t\to\infty} \frac{1}{6N_m t} \langle \sum_{j=1}^{N_m} [r_j(t) - r_j(0)]^2 \rangle. \qquad (3.5)$$

In the modeling during the first 2000 time steps, the equilibrium state was achieved, after that or immediately the diffusion coefficient was measured, or the shear force was applied and the friction force was measured. The maximum duration of the computer experiment was equal to 55000 time steps or 144.4 ps.

## 4. RESULTS

In Fig. 4.1-Fig. 4.6 we present the results obtained in the modeling of ultrathin argon films located between atomically smooth diamond surfaces. In Fig. 4.1 we illustrate the calculated time dependences of the autocorrelation functions of the velocities of argon molecules for different load values. It is seen that the amplitude and number of oscillations increase with increasing load. This indicates that velocities of the molecules become more correlated at high loads.

Fig. 4.2 indicates the transition of an ultrathin film of liquid argon to a solid-like state with increasing external load. The feature of the dependences $D(t)$ is that for low loads, the film is in a liquid state as implied by the attainment of saturation by the dependences.

There is a weakly expressed ordering in the plane of the layer. The typical view of configurations of the argon molecules in such films is illustrated in Fig. 4.4.

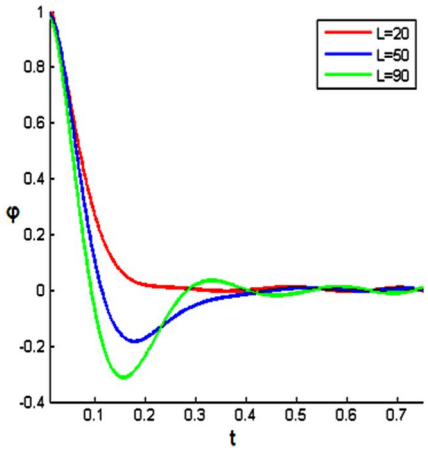


Fig. 4.1 – Time dependences of the autocorrelation function of velocity for an argon film of a thickness of two molecular diameters for different load values

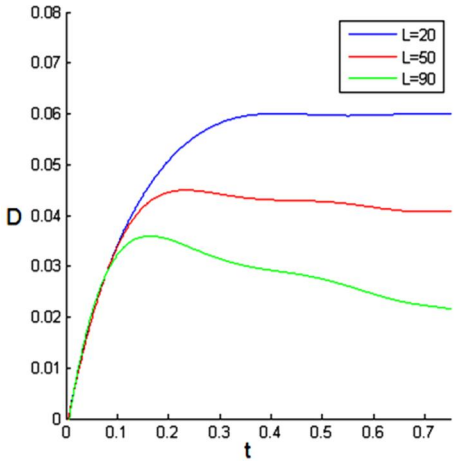


Fig. 4.2 – Time dependences of the diffusion coefficient calculated by the Einstein formula (3.5) for an argon film of two molecular diameter thick for different loads

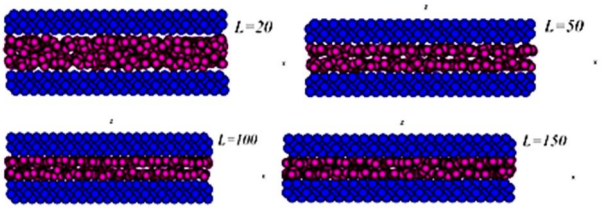


Fig. 4.3 – Formation of quasi-discrete layers with increasing external load for an argon film of two molecular diameter thick at zero shear

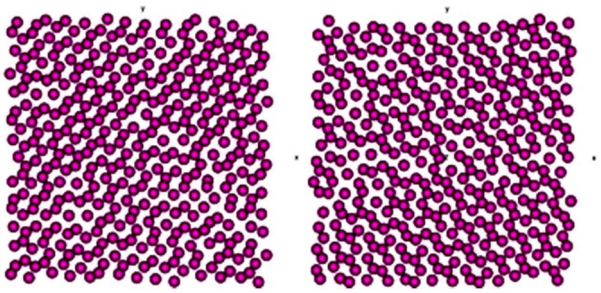

Fig. 4.4 – Typical configurations of argon molecules in the film of two molecular diameter thick in the absence of shear and at loads of 150 (on the left) and 200 (on the right)

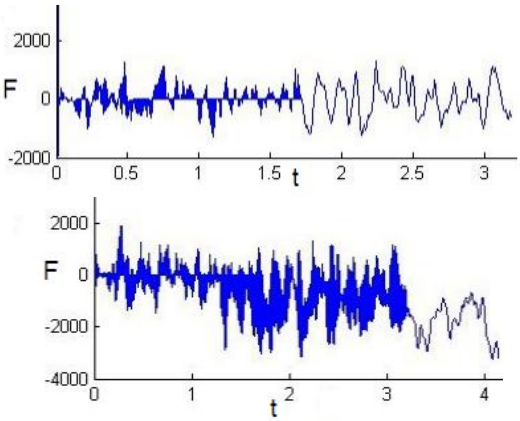


Fig. 4.5 – Time dependences of the friction force for argon films of a thickness of one (upper) and two (lower) atomic diameters at load of 10 and shear of 100

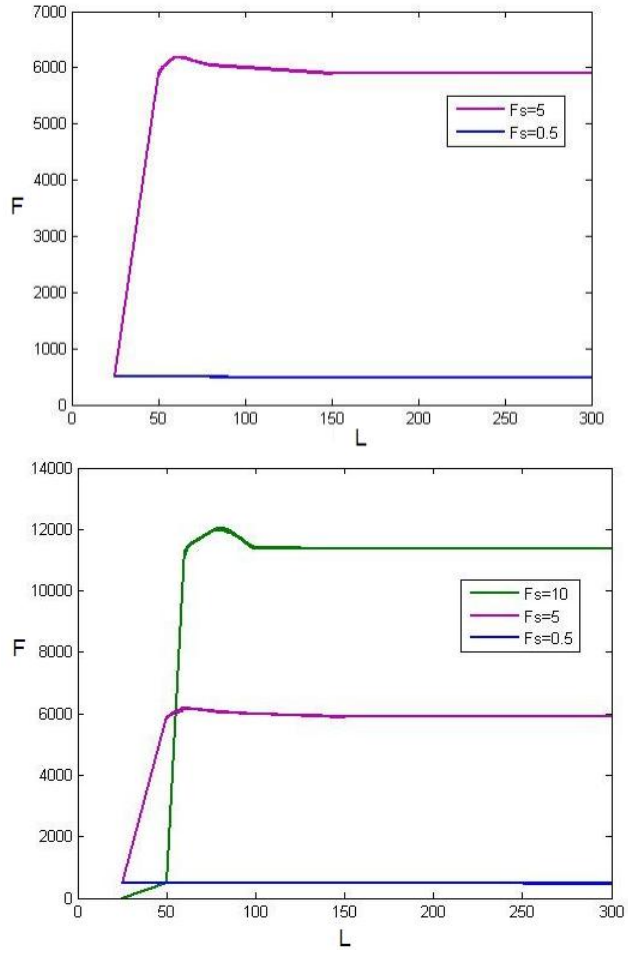


Fig. 4.6 – Dependences of the average value of the friction force on the load for the films of a thickness of one (upper) and two (lower) molecular diameters

The typical time dependences of the friction force for argon are presented in Fig. 4.5. One can see the presence of peaks of the friction force corresponding to the stick-slip mode and a gradual decrease with time of the oscillation frequency. It is also seen that the amplitude and frequency of oscillations in the lower figure are larger that is caused by an increase in the film thickness.

Dependences of the average in time value of the friction force on the load are shown in Fig. 4.6. One can distinguish two main sections, namely, the linear section for small *L* corresponding to the first Amontons law and the horizontal one for higher loads. These dependences are explained by using the "Cobblestone" model, according to which the friction force for the boundary mode is determined by two components [3, 4, 11]. The first one arises due to the presence of internal adhesion forces between the film molecules and the surfaces; the second component is caused by the action of external load. The friction force is defined by the ratio:

$$F = S_c A + CL, \qquad (4.1)$$

where constants $S_c$ and $C$ are, respectively, the critical shear stress and friction coefficient, $A$ is the contact area, $S_c$ depends on adhesion interactions of the film with the surface, and $C$ is associated with the atomic granularity (roughness) of the surfaces and with sizes, shape and configuration of the liquid molecules between the surfaces. In the general case, the smoother surfaces are, the smaller the value of $C$. We should note that the macroscopic friction coefficient $\mu$ for damaged surfaces has different origin and magnitude with respect to $C$.

The obtained dependences can be explained as follows. In the model, the surfaces are absolutely smooth and the argon molecules have a simple shape, so one can assume that only adhesion interactions contribute to the friction force, thus $C \approx 0$, and the friction force is determined by the first component in (4.1). At the beginning, with increasing load, the number and magnitude of adhesion bonds of argon molecules with surfaces sharply increases. Upon reaching a certain value of the load for the given magnitude of the shear force, some stationary values of the number of adhesion bonds and magnitude of adhesion forces are established. Since the model does not include surface strains, then with increasing *L*, the contact area is not changed, the first term in (4.1) and, respectively, the friction force remains constant. One can also talk about the same changes in the shear stresses $S = F/A$, which are often measured in the experiments.

Let us compare the obtained in the modeling dependences *F*(*L*) (or *S*(*L*)) in Fig. 4.6 with the experimental dependences of the shear stress $S = F/A$ on the load for different liquids [2, 4]. In liquids, octamethylcyclotetrasiloxane and cyclohexane molecules are of a simple spherical shape, and molecules of other liquids have the form of chains. As emphasized in [4], for spherical molecules the main contribution is given by the first term in (4.1) and $F \propto A$. At high loads, the shear stresses remain constant for a given number of layers of molecules, since, although the friction force increases, but also there is a proportional increase in the contact area because of the deformation of the surfaces. For complex molecules, the main contribution to the friction force is given by the second term in (4.1) and $F \propto L$. One can note the similarity of the experimental and obtained in the modeling dependences $S(L)$. However, for the first ones, the constancy of *S* with increasing *L* is caused by the proportional changes in *F* and *A* due to the deformation of surfaces, and in computer experiments, the constancy of shear stresses is induced by the constancy of these quantities.

Thus, the model gives results largely corresponding to the behavior of water films and other liquids with the molecules of a spherical shape [2, 4, 12]. It can also be concluded that the linear increase in the friction force with the load is defined not by the surface relief, but by their elasticity that explains a rapid attainment of saturation by the force with increasing load.

## 5. CONCLUSIONS

We have performed the computer modeling by the MD method of an ultrathin layer of liquid argon compressed between absolutely rigid diamond surfaces. In the paper, the model of elastic balls was used for argon. The equilibrium and dynamic characteristics of the system depending on the external load (surface pressure) and shear forces have been studied. Based on the study, we can conclude the following:

1) with increasing load, one can observe a transition of ultrathin films of liquid argon to a solid-like state that is manifested in a decrease of the diffusion coefficient and, for most cases, in the formation of ordered configurations of molecules;

2) time dependences of the friction force reflect, in general, a solid-like film structure. However, a significant difference in time scales occurring for the corresponding measurements prevents an unambiguous comparison of modeling results with experiments for the friction force;

3) change of the average (in time) value of the kinetic friction force with the load for both types of surfaces satisfies the "Cobblestone" model in the approximation of the prevalence of adhesion interactions. The shear stress dependences on the load obtained by the modeling are similar to the experimental ones for simple spherical molecules [2, 4].

## ACKNOWLEDGMENTS

The work has been performed within the Project "Non-equilibrium thermodynamics of metal fragmentation and friction of spatially non-uniform boundary lubricants between the surfaces with nanosized irregularities" under the financial support of the Ministry of Education and Science of Ukraine (Grant No 0115U000692).

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