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Research Report

Exact Solution and Simulation Method for Energy Dissipation via Lattice Vibration at a Friction Interface

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ABSTRACT In order to dissipate lattice-vibration energy generated in a molecular dynamics (MD) calculation properly, we developed a numerical method by which an infinitely large lattice system can be connected to an MD calculation cell. MD calculations of kinetic friction models showed that an enormous number of solid atoms is required to simulate energy dissipation due to long-wavelength lattice vibrations. The proposed method derived accurate results without calculating large number of the solid atoms explicitly. This method would be useful in clarifying non-equilibrium phenomena, such as catalysis and fracture dynamics, as well as the kinetic friction mechanism.

KEYWORDS Atomic Scale Simulation, Friction, Lattice Vibration, Energy Dissipation, Heat Exchange, Lattice Heat Bath

1. Introduction

Despite industrial demands related to the growing need for energy conservation, the mechanism involved in friction remains unclear. One difficulty in clarifying the friction mechanism stems from the elementary processes at an atomic scale. For example, if one supposes a system in which two solids are in sliding contact, the surface atoms would repeatedly generate and break chemical bonds at the interface. If the sliding surfaces are lubricated, the lubricant may react with the surface and alter the friction properties. These atomic elementary processes affect friction, but are difficult to observe the friction interface experimentally.

Simulations are useful for accessing this hidden friction interface, and molecular dynamics (MD) and electronic state calculations have been used to investigate tribology phenomena such as lubrication and tribo-chemical reactions.[1,2]

However, these atomic simulations are not free from technical problems. One of the major problems is the insufficient treatment of energy dissipation in kinetic friction. During shear, atoms within the frictional region absorb the kinetic energy of the sliding material, and the lost kinetic energy is transferred to other atomic degrees of freedom, such as lattice vibrations and electron excitations. In the present study, we focus on the energy dissipation via lattice vibrations. In order to simulate a highly non-equilibrium state in kinetic friction, the heat generated in a calculation cell should be removed by means of a thermostat, which exchanges heat with the calculation cell as the rest of the entire system. Usually, the Langevin equation and the Nose-Hoover method have been used to express the thermostat to maintain a specified temperature.[3] Although these methods provide excellent thermostats for liquid and gas systems, they are not useful in a solid system because the Langevin equation is based on a Brownian environment and the Nose-Hoover method cannot retain the ergodic property in harmonic oscillator systems.[4]

We present a method that mimics the heat exchange via lattice vibrations in a solid friction system. Our strategy is simple. As shown in **Fig. 1**(a), in the MD simulation cell without thermostats, the excited lattice vibrations by shear propagate to the surface normal direction and reflect at the boundary. In order to dissipate the excited lattice vibrations, we derive the exact solutions for surface atoms in an infinitely large harmonic-coupled oscillator in Fig. 1(b). Then, as shown in Fig. 1(c), the surface atoms of the infinite harmonic-coupled oscillator are set at a calculation-cell boundary to interact with the MD atoms. Since the derived surface atoms involve infinite degrees of freedom, the excited lattice vibrations propagate toward the infinite lattice system and never return to the surface. Therefore, the proposed method can correctly reproduce the energy dissipation within

the harmonic approximation.

The remainder of the present paper is organized as follows. Section 2 provides theoretical and numerical descriptions for the dynamics of the surface atoms in the infinitely thick lattice. Section 3 presents the simulation results of the proposed method with friction models. Section 4 presents remarks and conclusions of the present study. Although, for simplicity, we discuss a two-dimensional lattice system, this formalism can be readily extended to a three-dimensional lattice system.

2. Methods

2. 1 Semi-infinite Dynamic Green's Function Method

We consider a two-dimensional square lattice in which N and N' atomic layers are aligned in the surface normal direction, z, and lateral direction, x, respectively, as shown in **Fig. 2**(a). Here, η and ξ are used to identify atoms. Periodic boundary conditions are applied in the x direction, and the bottom layer connects to a rigid body. The bonds between the atoms are approximated by two-dimensional harmonic springs, where $Kx(')$ and $Kz(')$ are the spring constants in the x and z directions, respectively, for a bond extending in the $z(x)$ direction (see Fig. 2(b)). We assume that only the surface atomic layer is subjected to an external force. Denoting the x and z displacements of an atom by u, the equation of motion for the lattice is given as follows:

$$m\ddot{u}_{\eta,\xi} + K_u(2u_{\eta,\xi} - \sum_{\Delta\xi} u_{\eta,\xi+\Delta\xi} - \delta_{\xi,1}u_{\eta,1}) + K'_u(2u_{\eta,\xi} - \sum_{\Delta\eta} u_{\eta+\Delta\eta,\xi}) = \delta_{\xi,1}F^u_\eta \ , \quad (1)$$

where δ is the Kronecker delta, and Δ refers to the

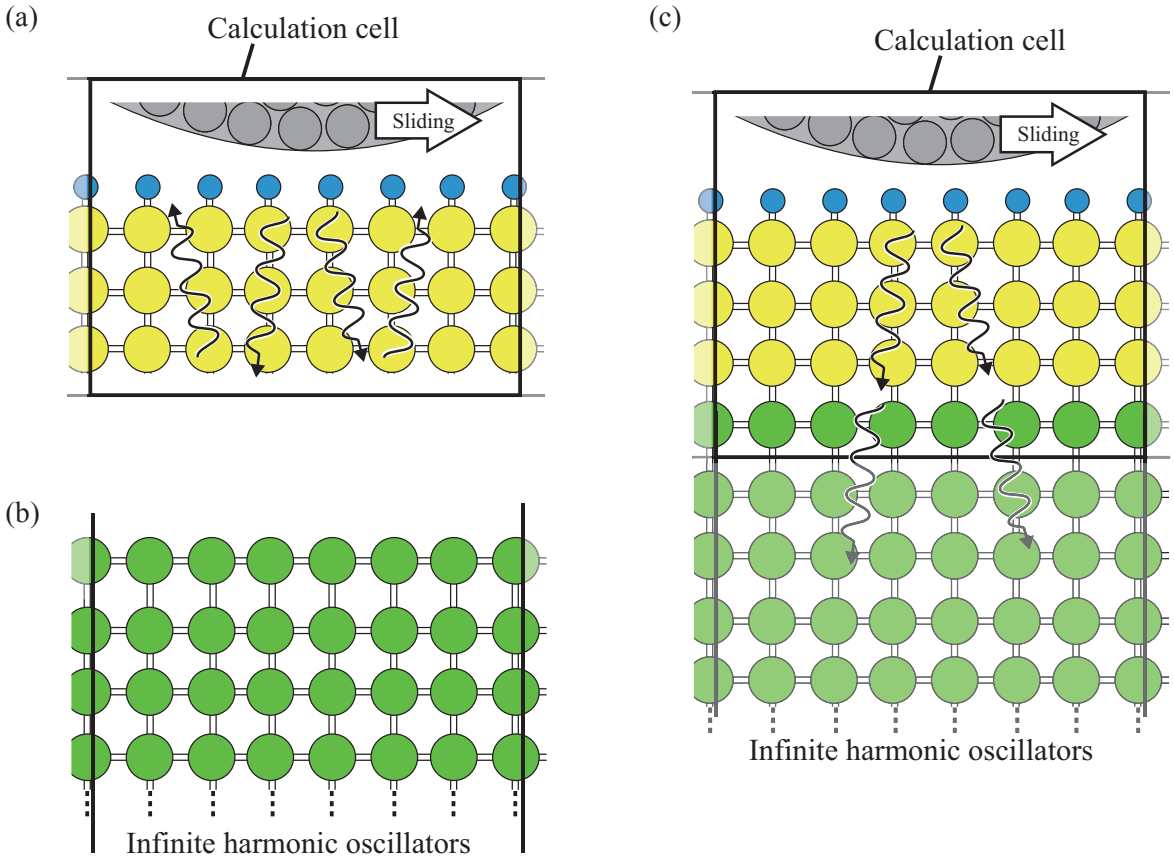

Fig. 1 Conceptual schematic diagram of the method for energy dissipation via lattice vibrations. (a) Calculation cell for a friction system by MD simulation without a thermostat. Periodic boundary conditions are applied along the lateral surface direction. (b) Surface model of a coupled-harmonic oscillator containing an infinite number of atomic layers in the thickness direction. (c) Method for describing the energy dissipation via lattice vibrations. The surface atoms of the infinite coupled harmonic oscillators are included and are connected to the bottom atoms in the MD calculation cell.

nearest-neighbor atoms connected to an atom labeled by η and ξ. The quantities m and F_η are the atomic mass and the external force acting on the surface of atom η, respectively.

Equation (1) can be solved analytically, and we can take the limit of an infinite number of atomic layers N in the solution.[5,6] The solutions for the surface atoms ($\xi = 1$) in the infinite N limit are expressed by discrete Fourier transform as follows:

$$\dot{u}_l(t) = \delta_{l,0}\dot{u}^0 + \int_0^t s_l(t-t')F_l^u(t')dt'/m\,, \qquad (2)$$

$$s_l(t) = \frac{4}{\pi}\int_0^{\pi/2}\cos^2\theta\cos(\alpha t\sqrt{\sin^2\theta+\beta_l^2})d\theta\,,$$

$$F_l^u = \frac{1}{N'}\sum_{\eta=1}^{N'} e^{i2\pi\eta l/N'}F_\eta^u\,,$$

$$\dot{u}_{\eta,1} = \sum_l e^{-i2\pi\eta l/N'}\dot{u}_l\,,$$

where l is the wave number index, $\alpha = 2\sqrt{K_u/m}$, and $\beta_l = \sqrt{K'_u/K_u}\sin(\pi l/N')$. The term $\dot{u}^0$ will be discussed later herein. The memory kernel function s_l includes the effect of all of the atoms in the infinite lattice. Hereinafter, we refer to the function s_l as the semi-infinite dynamic Green's function (SI-DLGF). Note that, despite calculating only the surface atoms explicitly described in the Eq. (2), the surface atoms can move as if they are connected to the infinite lattice, thanks to the SI-DLGF.

The function $\dot{u}^0$ is a general solution of Eq. (1) under the initial conditions for the uniform distortion of the lattice. The explicit form is $\dot{u}^0 = -\bar{F}^u\alpha/2K_u$, where $\bar{F}^u$ is the force associated with the uniform distortion. This solution is important when applying uniform forces through the lattice; namely, $\dot{z}^0$ expresses the normal force applied to the lattice, and $\dot{x}^0$ is used to balance the shear force in order to achieve a steady state in a friction system. Therefore, the surface atoms described by the SI-DLGF method act as a heat exchanger but also a role to apply normal pressure and shear stress.

The second term on the right-hand side of Eq. (2) is troublesome because the computational costs for the convolution integral are too high. Using a conventional integral method, the calculation over N_t time steps requires a large memory space $O(N_t^2)$ in order to maintain the force history and $O(N_t)$ operations. Fortunately, this difficulty can be overcome by a fast convolution algorithm that uses the inverse Laplace transformation to reduce the computational cost significantly, so that the memory requirement is $O(\log N_t)$ and the operation requirement is $O(N_t \log N_t)$. Details concerning the algorithm can be found in Ref. (6). For the time integral in Eq. (2), a typical method such as the Runge-Kutta method and varlet algorithms can be used.

2. 2 Friction Decomposition with Lattice Vibration Modes

By virtue of the analytically exact solutions, the SI-DLGF formalism provides information about the degree to which a lattice-vibration mode dissipates the kinetic energy of a sliding material. Combining Eq. (2) with the thermodynamic relationship between the friction force and the energy-loss rate, we can derive the equation for the kinetic-friction force $\bar{F}^x$ in the

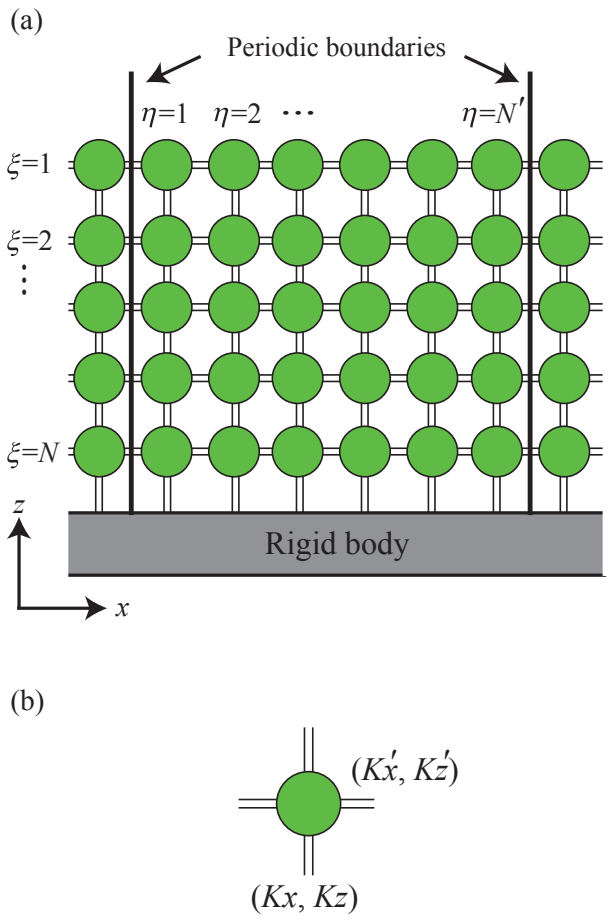

Fig. 2 Schematic diagram of (a) infinite coupled harmonic oscillators and (b) spring constants for the atomic bonds.

stationary state, as follows: [6,7]

$$\overline{F}^x = \sum_{\substack{u=x,z \\ 1\le n\le\infty;\, l}} \frac{2}{mVN'} \left| F_l^u(\omega_{nN'-l}) \right|^2 \mathrm{Re}\, s_l(\omega_{nN'-l}) , \quad (3)$$

$$\mathrm{Re}\, s_l(\omega) = \frac{2\omega}{\alpha^2} \sqrt{\frac{1}{(\omega/\alpha)^2 - \beta_l^2} - 1} ,$$

where ω is the frequency. The power spectrum $|F_l^u(\omega_{nN'-l})|^2$ can be obtained by the history of the surface forces in the simulations. The analytical form of the real part of the SI-DLGF is given and represents the coefficient of the energy absorption by a surface vibration mode. Using Eq. (3), we can determine the contribution of the lattice vibrational modes to the kinetic friction by the analysis of a SI-DLGF simulation. Equation (3) states that the kinetic-friction force can be decomposed by the contributions of the mode indices u, n, and l. Furthermore, the contributions of the bulk and surface effects to the kinetic friction are independent of each other because the friction is the product of the surface term and the bulk term in Eq. (3), rather than their sum.

3. Results

3. 1 Simple Demonstration: Friction Dependence on the Number of Atomic Layers

A friction system, in which the upper and lower bodies have the same lattice constant σ and an atomically flat surface, as shown in **Fig. 3**, are used to compare the MD and SI-DLGF results, i.e., simulations with finite and infinite atomic layers systems are compared. The upper layer slides at V = 10 m/s, and the bonds between the atoms are frozen. The lower layer is a coupled harmonic oscillator with the spring constant of diamond and the atomic mass of a carbon atom. The number of the inner atomic layer is N. The interaction potential between the upper and lower atoms is a repulsive soft core potential. For the calculation of the finite N system, we introduced a velocity-proportional damping term that mimics the energy dissipation. More details of the simulation can be found in Ref. (6).

Figure 4 shows the friction coefficients obtained by the MD and SI-DLGF simulations. In the case of N = 1, the friction coefficients are approximately zero. The friction coefficients increase with N, and the values converge to those for infinite N, which is obtained by the SI-DLGF method. Over a thousand atoms are needed to obtain the converged friction coefficients.

In order to analyze the friction dependence on the number of atomic layers more closely, the friction decomposition in Eq. (3) was used. Owing to the translation symmetry of the friction system, the wave number l = 0 is only allowed to be excited. **Figure 5** shows the decomposition of the friction with the frequency mode, and the lowest frequency component n = 1 shares a large amount of the total friction coefficient. The lowest frequency is $2\pi V/\sigma$ and is regulated by the sliding velocity V, which is generally in the range of 0 to 10 m/s. In the case of N = 1, the eigenfrequency for the single harmonic oscillator is on the order of the sound velocity in a solid, which is typically several thousand meters per second. Therefore, since the eigenfrequency is too high to resonate with the sliding velocity, the single harmonic oscillator cannot dissipate the kinetic energy of the sliding counter object. As N increases, the lattice can be excited at longer wavelengths and lower frequencies, and energy dissipation eventually occurs when the lattice vibrations become so slow as to resonate at $2\pi V/\sigma$. These result reveal the importance of using an extremely large number of solid atoms when reproducing the energy dissipation via lattice vibrations.

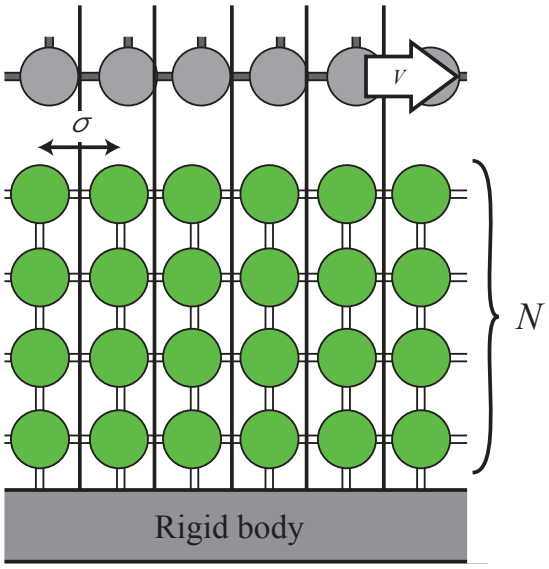

Fig. 3 Schematic diagram of a friction system between flat surfaces. The lower solid is a coupled-oscillator surface model that consists of N inner atomic layers. Vertical lines indicate periodic boundaries.

3. 2 Single Asperity System

The potential effectiveness of the SI-DLGF was examined using the simple model described in the previous subsection. Here, we use a more complex but realistic system involving friction at a single asperity. The friction system is illustrated in **Fig. 6**. In the lower solid, there is a coupled-harmonic oscillator simulated by MD calculation, and the number of surface atoms N' and 10 of the inner lattice layers are in alignment. The atomic mass is the same as that of a silicon atom, and the spring constant is that of bulk silicon. Atoms simulated by the SI-DLGF with the same bulk parameters as the MD lattice are deployed at the bottom of the MD lattice with spring connections. In this manner, the MD lattice can make a conjunction and exchange heat fluxes with the infinitely large lattice. In the upper solid, the surface atoms align in the x direction to form an atomically flat asperity of length L_c. The upper layer slides at $V = 10$ m/s, and the bonds between the atoms are frozen. The upper and lower lattices have the same lattice constant σ, and the interaction potential is a repulsive Lennard-Jones potential. Owing to the periodic boundary conditions, the lower solid has an infinitely large surface area, whereas the upper solids form asperities at intervals of $N'\sigma$.

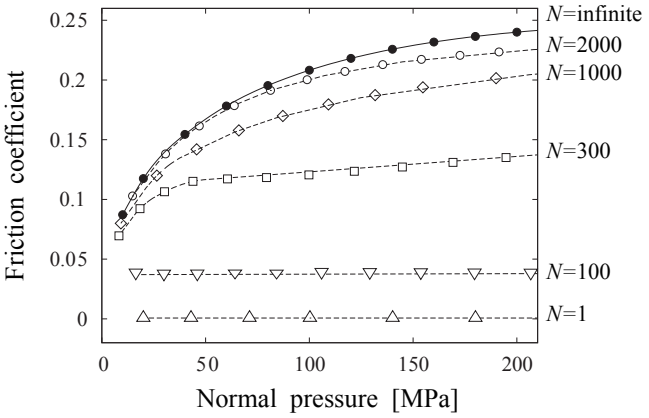

Fig. 4 Dependence of kinetic friction coefficient on the number of atomic layers N. Symbols △, ▽, □, ◇, ○ and ● indicate plots for N=1, 100, 300, 1000, 2000 and infinite, respectively. The infinite data were obtained by the SI-DLGF method.

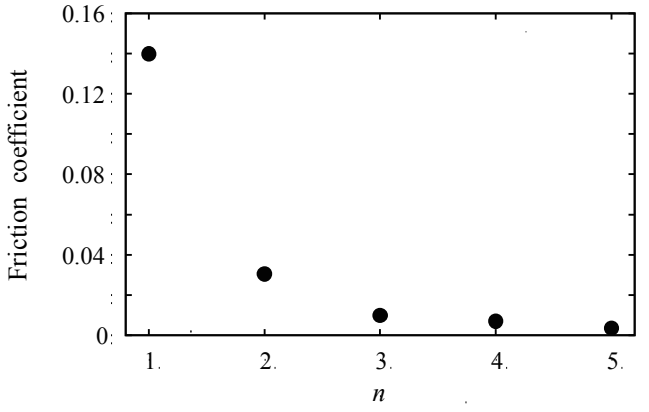

Fig. 5 Decomposition of kinetic friction coefficient with frequency index n. Normal pressure is 100 MPa.

Figure 7 shows the dependence of the kinetic friction on the number of the surface atoms N'. As N' increases, the friction coefficient first decreases for $N' < 1000$. For an N' of approximately 1000, the friction coefficient increases and finally converges to a finite value in the large N' limit. This strong dependence indicates that a large number of surface atoms, and not only internal atoms, is needed for the energy-dissipative surface lattice vibrations.

In order to clarify the origin of the N' dependence, Eq. (3) is modified. The surface force term of $|F_l^u(\omega_{nN'-l})|^2$ is approximated to separate the space

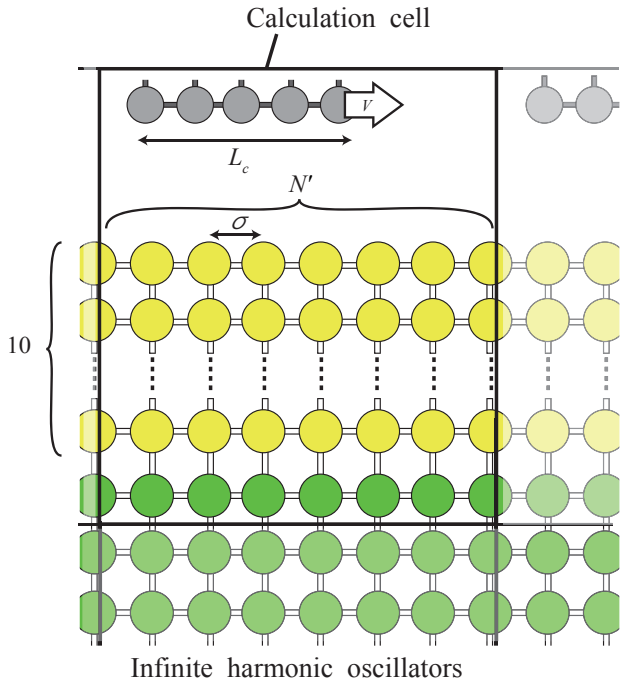

Fig. 6 Schematic diagram of a friction system for a single asperity. The yellow atoms are simulated by MD calculation, whereas the green atoms within the calculation cell are simulated by the SI-DLGF method.

and time variables. (A detailed description is provided in Ref. (7).) The modified equation is as follows:

$$\overline{F}^x = \sum_{\substack{u=x,z \\ 1\le n\le\infty \\ |l|<AnN'}} \frac{2C_u(n)}{mVN'} J_0^2(L_c k_l / 2)\, Res_l(\omega_{nN}), \qquad (4)$$

where J_0 and k_l are a zero-order Bessel function of the first kind and the surface wave number, respectively, and $A = 2V\sqrt{K_u / K_u'}\,/\,\alpha_u\sigma$. The Bessel function term is derived from the shape of the upper contacting surface. The term $C_u(n)$ is a quantity related to the autocorrelation of the temporal fluctuation of the surface force during shear. **Figure 8** shows $C_u(n)$ for this friction simulation with N' = 128 and 4096. As in the previous case of the flat contact system, the contribution of the lowest frequency mode, n = 1, is the largest. The other n components decay exponentially.

Since the n = 1 component is the largest, we can reasonably consider only the n = 1 term in Eq. (4). Parameter A in the maximum restriction for $|l|$ in Eq. (4) refers to the ratio of the sliding velocity to the speed of sound in a solid. The actual value of A in this setup is approximately 1/1000. Thus, the surface wave number l modes that satisfy $|l| < N'/1000$ act as energy-dissipation channels. When N' is less than 1000, only the l = 0 mode is excited in order to dissipate the shear energy. In this case, Eq. (4) yields

$$\overline{F}^x \propto \sum_{u=x,z} \frac{C_u(n=1)}{\alpha_u N'}.$$

As shown in Fig. 8, $C_u(n)$ does not change significantly with N'. Therefore, the above equation assumes the kinetic friction to decrease as $1/N'$. This assumption coincides well with the results shown in Fig. 7. When $N' > 1000$, $|l| \ge 1$ modes begin to contribute to the energy dissipation. The number of energy dissipative l modes increases by 1 with each increment of N' by 1000, whereas the magnitude of each energy-dissipation mode decreases as $1/N'$. Therefore, the value of the friction coefficient converges in the large N' limit.

The N' dependency can be intuitively understood as follows. When N' is small, surface lattice vibration modes other than l = 0 cannot be excited, because the wavelength of $|l| \ge 1$ is too short and the time scale of the frequency is too fast for resonance to occur with the force fluctuation due to the sliding velocity V. As N' increases, the surface wavelength increases and the frequency gradually decreases. Finally, the l modes can resonate to dissipate the kinetic energy of the sliding material.

4. Conclusion and Remarks

A method for energy dissipation via lattice vibrations was proposed herein. We successfully derived analytical solutions for surface atoms in an

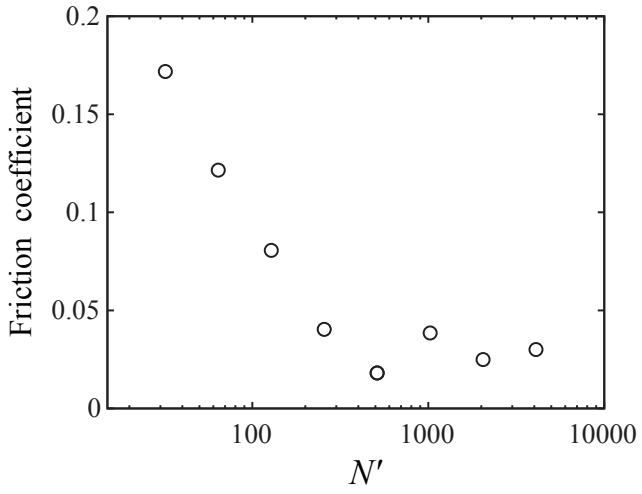

Fig. 7 Dependence of the friction coefficient on the number of the surface atoms N' by SI-DLGF method. The normal force for an upper surface atom is 0.0473 nN. The number of the upper surface atoms is 20.

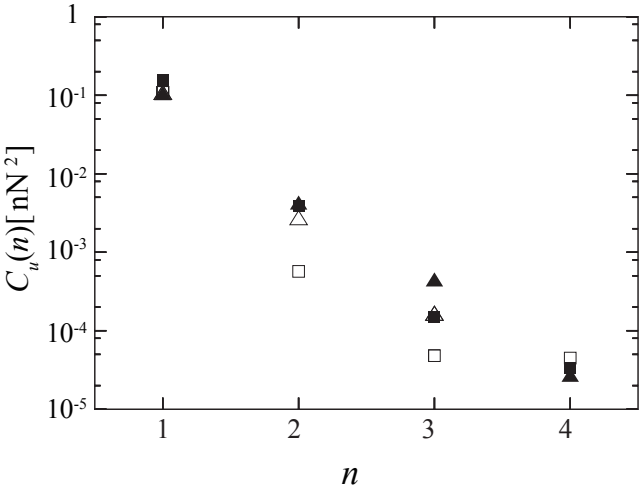

Fig. 8 Results of $C_u(n)$. Symbols □ and △ indicate the results for $u = z$ and $u = x$ for N' = 4096, respectively. Symbols ■ and ▲ indicate the results for $u = z$ and $u = x$ for N' = 128 respectively.

infinite harmonic-coupled oscillator and obtained the SI-DLGF method, which can simulate the dynamics of the surface atoms on which the existence of an infinitely large lattice is projected implicitly. Since the SI-DLGF atoms and MD atoms can coexist in a calculation cell, the SI-DLGF atoms dissipate the vibrational energy generated by a non-equilibrium phenomenon occurring in the MD atoms.

We demonstrated the SI-DLGF method in friction systems, and the strong dependence of friction on the number of solid atoms was found. In order to dissipate the kinetic energy of a sliding counter material, the energy-dissipative lattice vibrations should have a long wavelength and be slow enough to resonate with the sliding velocity. These results have two important implications. First, in terms of simulations, the number of solid atoms is a key parameter because the energy-dissipative lattice vibrations are long coherent movements. Second, in terms of applications, kinetic friction can be changed by not only surface modifications but also by modifications of bulk characteristics, because Eq. (3) implies that the contributions of the bulk and surface effects to the kinetic friction are independent.

The SI-DLGF method is not a solid heat bath completely, because this method can only treat the infinitely large lattice at zero Kelvin, but not at a finite temperature. Introducing a finite temperature to the Si-DLGF method is possible by extension of the formulation in Eq. (2) to the generalized Langevin framework; this extension will be presented elsewhere. Another important shortcoming is that the SI-DLGF method cannot treat the general structure of solids, such as face-centered cubic and body-centered cubic structures. No analytical method by which to solve this problem has yet been established, but numerical methods for obtaining Green's functions for general structures have been proposed.[8] Although there remain problems to overcome, the presented method may be a fruitful step for the goal of the proper solid heat bath, that could contribute to automotive fields, such as tribology, catalysis, and fracture dynamics in atomic simulations.

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