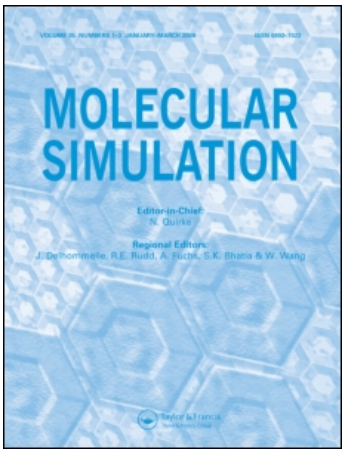

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Multiscale treatment of thin-film lubrication

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Multiscale treatment of thin-film lubrication

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A multiscale technique that combines an atomistic description of the interfacial (near) region with a coarse-grained (continuum) description of the far regions of the solid substrates is proposed. The new hybrid technique, which represents an advance over a previously proposed dynamically-constrained hybrid atomistic-coarse-grained treatment (Wu *et al. J. Chem. Phys.*, **120**, 6744, 2004), is applied to a two-dimensional model tribological system comprising planar substrates sandwiching a monolayer film. Shear–stress profiles (shear stress versus strain) computed by the new hybrid technique are in excellent agreement with "exact" profiles (i.e. those computed treating the whole system at the atomic scale).

Keywords: Film; Interface; Lubrication; Multiscale; Coarse-grained

1. Introduction

Fluid–solid interfaces play a prominent role in boundary lubrication, which involves the sliding of one solid substrate over another separated from it by a thin film of lubricant. Although the surfaces of the substrates appear smooth on the macroscopic scale, they actually make molecular contact at relatively few microscopic asperities [1]. Throughout the vast expanses (far regions) of the substrates well removed from the interface the solid atoms merely oscillate with small amplitude about their equilibrium positions. The impact of such regular motion can be adequately accounted for by continuum mechanics. In contrast, the lubricant molecules diffuse and, under sufficiently large loads, the asperities may deform plastically. It is therefore necessary to describe the details of atomic motions in the near region. The question is how to combine the atomistic description required in the near region with the continuum description that suffices for the far regions.

During the past decade several multiscale schemes for treating processes (e.g. crack propagation) in defective crystals have been devised [2,3]. In particular, Tadmor *et al.* [3] proposed the quasicontinuum (QC) technique, in which the crystal lattice is coarse-grained by covering it with a finite-element mesh and the configurational energy is minimized (as a function of the configuration of mesh nodes) at zero temperature. In order to handle processes in solids at nonzero temperature, we proposed a dynamically constrained extension of the QC method [4]. The (non-nodal) atoms underlying the finite elements are assumed to move in concert with the nodes. The thermophysical properties are computed as ensemble averages over nodal configurations by the Monte Carlo (MC) method. Application of this dynamically constrained MC QC technique to a two- dimensional (2D) Lennard-Jones (LJ) crystal shows that the coarse-grained isotropic stress differs from its "exact" counterpart (computed by fully atomistic MC) by an amount linear in the absolute temperature (T).

Combining an atomistic description of the near region with a coarse- grained description of the far regions, we extended the dynamically constrained MC QC method to handle fluid–solid interfaces [5,6]. The resulting hybrid atomistic-coarse-grained (HACG) treatment was applied to the reversible shearing of 2D LJ substrates separated by an LJ film. The (dynamically constrained) HACG shear–stress profiles (plots of shear stress versus shear strain) are in very good agreement with the exact profiles. However, the HACG profile of mean separation between the substrates differs markedly from its exact counterpart. This discrepancy reflects the neglect of thermal motion of underlying atoms, which is a consequence of the dynamical constraint.

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To eliminate the dynamical constraint, we recently proposed an alternative extension of the original QC method [7]. By integrating the canonical Boltzmann factor over the degrees of freedom of the underlying atoms, we obtain an effective potential energy for the nodal motion which consists of the dynamically constrained potential energy plus a free- energy correction (i.e. the Helmholtz energy of the underlying subsystem). This effective potential replaces the dynamically constrained potential in the MC treatment. Applying this free-energy QC MC technique to the 2D LJ crystal yields the isotropic stress in excellent accord with the exact isotropic stress. The purpose of the present study is to introduce the free-energy correction within the HACG framework and ascertain its impact on the *shear* stress.

2. Model tribological system

The idealized contact consists of identical 2D LJ substrates separated by an LJ film, as illustrated in figure 1. Outside layers of substrate atoms ("walls", indicated by filled squares in figure 1) are rigid and serve as handles by which the substrates are slid quasistatically relative to each other. A portion (the far region) of each substrate is coarse-grained with a finite-element mesh, which consists of congruent equilateral triangles when the atoms of the substrates are in their equilibrium positions (as shown in figure 1). This coarse-graining partitions the original substrate atoms into two subsets: N_n nodal atoms and N_q non-nodal atoms. If one integrates the Boltzmann factor over the $2N_q$ degrees of freedom of the non-nodal atoms, one obtains an effective potential energy governing the motion of just the nodal atoms

$$V_{\text{eff}}(\mathrm{R}^{N_n}) = \sum_{e=1}^{N_e} (N_a^e \tilde{u}_e + N_q^e f_e), \tag{1}$$

where R^{N_n} stands for the nodal configuration, N_e for the number of elements, N_a^e the (total) number of atoms underlying element e, N_q^e the (total) number of non-nodal atoms underlying element e, $\tilde{u}_e$ for the configurational energy per atom (with the underlying atoms fixed in the equilibrium configuration in the field of the nodal atoms), and f_e for the Helmholtz energy per atom. Since the elements are assumed to be "local" [3], we can express $\tilde{u}_e$ as

$$\tilde{u}_e = \frac{1}{2} \sum_{j \neq i} u_{ss}(r_{ij}), \tag{2}$$

where u_{ss} denotes the LJ(12,6) interatomic potential between substrate atoms, and i refers to the centroid atom (i.e. the atom nearest the centroid of e) and j to atoms that lie within the circle of cutoff radius r_c that is the centered on i. Using the local harmonic approximation [8], we estimate f_e as

$$f_e = 2k_B T \ln [h(\det \mathbf{D})^{1/4} / 2\pi k_B T], \tag{3}$$

where k_B is Boltzmann's constant, h is Planck's constant, and $\mathbf{D}$ is the 2D "local" dynamical matrix associated with the centroid atom.

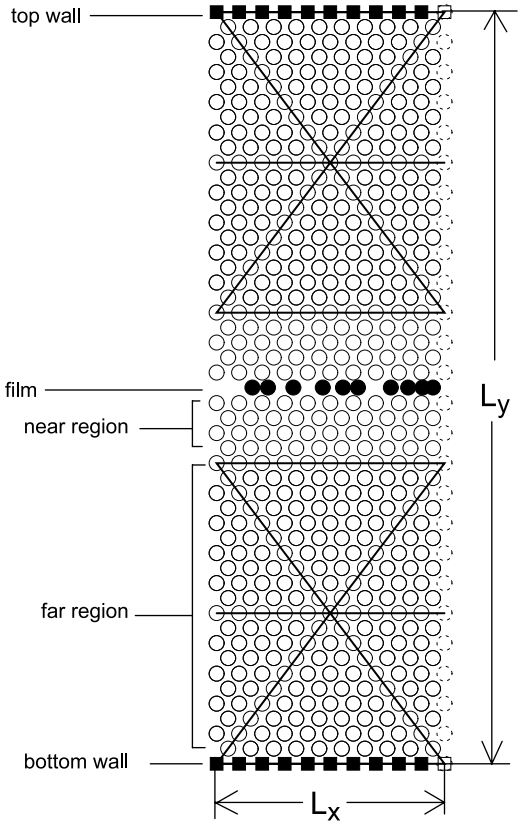

Figure 1. Schematic of two-dimensional tribological system (where substrates shown in perfect alignment $\alpha = 0$ and atoms in their equilibrium positions at zero temperature). Periodic boundary conditions imposed in x-direction. Film atoms denoted by filled circles, substrate atoms by open circles and wall atoms by filled squares. System depicted is the one actually simulated. Total number of atoms $N = 510$; number of wall atoms $N_w = 20$; number of film atoms $N_f = 10$; number of near-region substrate atoms $N_s' = 80$; number of far-region (coarse-grained) atoms $N_s'' = 400$; number of nodal atoms $N_n = 4$; number of non-nodal atoms $N_q = 396$. Cell constant $a = 1.075$; $L_x = 10a$; LJ potential parameters: $\epsilon_{ss} = 1.0$, $\epsilon_{ff} = 1/9$, $\epsilon_{fs} = 1/9, 1/6, 1/3$; $\sigma_{ss} = \sigma_{ff} = \sigma_{fs} = 1$; cutoff radius $r_c = 2.5$. Fixed normal load (stress) $T_{yy} = -0.1$; temperature $T = 0.1$.

The total configurational energy of the partially coarse-grained system is then given by

$$\begin{aligned} U_{\text{cf}}(\mathbf{r}^{N_f}, \mathbf{r}^{N_s'}, \mathrm{R}^{N_n}) = {} & \frac{1}{2} \sum_{i=1}^{N_f} \sum_{j \neq i}^{N_f} u_{ff}(r_{ij}) \\ & + \sum_{i=1}^{N_f} \sum_{j=1}^{N_s'} u_{fs}(r_{ij}) \\ & + \frac{1}{2} \sum_{i=1}^{N_s'} \sum_{j \neq i}^{N_s'} u_{ss}(r_{ij}) \\ & + \frac{1}{2} \sum_{i=1}^{N_s'} \sum_{j=1}^{N_s''} u_{ss}(r_{ij}) \\ & + \sum_{e=1}^{N_e} N_a^e \left[\frac{1}{2} \sum_{j \neq i} u_{ss}(r_{ij}) \right] \\ & + \sum_{e=1}^{N_e} N_q^e 2k_B T \ln [h(\det \mathbf{D})^{1/4} / 2\pi k_B T)], \end{aligned} \tag{4}$$

where $\mathbf{r}^{N_f}$ and $\mathbf{r}^{N_s'}$, respectively, denote the configurations of the N_f film atoms and the N_s' substrate atoms in the near

region. N_s'' labels atoms in the coarse-grained far regions. The quantity of central interest, namely the shear stress, is given in the framework of the free-energy corrected HACG treatment by

$$\begin{aligned}T_{yx,cf}=&\frac{1}{2L_x}\sum_{i=1}^{N_f}\sum_{j\neq i}^{N_f}\langle u_{ff}'(r_{ij})x_{ij}y_{ij}/(r_{ij}L_y)\rangle\\&+\frac{1}{L_x}\sum_{i=1}^{N_f}\sum_{j=1}^{N_s'}\langle u_{fs}'(r_{ij})x_{ij}y_{ij}/(r_{ij}L_y)\rangle\\&+\frac{1}{2L_x}\sum_{i=1}^{N_s'}\sum_{j\neq i}^{N_s'}\langle u_{ss}'(r_{ij})x_{ij}y_{ij}/(r_{ij}L_y)\rangle\\&+\frac{1}{2L_x}\sum_{i=1}^{N_s'}\sum_{j=1}^{N_s''}\langle u_{ss}'(r_{ij})x_{ij}y_{ij}/(r_{ij}L_y)\rangle\\&+\frac{1}{L_x}\sum_{e=1}^{N_e}N_a^e\left[\frac{1}{2}\sum_{j\neq i}\langle u_{ss}'(r_{ij})x_{ij}y_{ij}/(r_{ij}L_y)\rangle\right]\\&+\frac{1}{L_x}\sum_{e=1}^{N_e}N_q^e\left[\frac{k_BT}{2}\left\langle\frac{1}{|\mathbf{D}|}\frac{\partial|\mathbf{D}|}{\partial\alpha a}\right\rangle\right],\end{aligned}\tag{5}$$

where

$$\begin{aligned}\frac{\partial|\mathbf{D}|}{\partial\alpha a}&=\frac{\partial D_{xx}}{\partial\alpha a}D_{yy}+D_{xx}\frac{\partial D_{yy}}{\partial\alpha a}-2D_{xy}\frac{\partial D_{xy}}{\partial\alpha a}\\\frac{\partial D_{xx}}{\partial\alpha a}&=m^{-1}\sum_{j\neq i}\Big\{\{u_{ss}'''x_{ij}^3y_{ij}/r_{ij}^3L_y+3u_{ss}''x_{ij}y_{ij}^3/r_{ij}^4L_y\\&\qquad-3u_{ss}'x_{ij}y_{ij}^3/r_{ij}^5L_y\Big\}\\\frac{\partial D_{yy}}{\partial\alpha a}&=m^{-1}\sum_{j\neq i}\Big\{u_{ss}'''x_{ij}y_{ij}^3/r_{ij}^3L_y\\&\qquad+u_{ss}''x_{ij}y_{ij}(x_{ij}^2-2y_{ij}^2)/r_{ij}^4L_y\\&\qquad-u_{ss}'x_{ij}y_{ij}(x_{ij}^2-2y_{ij}^2)/r_{ij}^5L_y\Big\}\\\frac{\partial D_{xy}}{\partial\alpha a}&=m^{-1}\sum_{j\neq i}\Big\{u_{ss}'''x_{ij}^2y_{ij}^2/r_{ij}^3L_y-u_{ss}''y_{ij}^2(2x_{ij}^2-y_{ij}^2)/r_{ij}^4L_y\\&\qquad+u_{ss}'y_{ij}^2(2x_{ij}^2-y_{ij}^2)/r_{ij}^5L_y\Big\}\end{aligned}\tag{6}$$

and the prime denotes the derivative with respect to the argument, α, the register, is the fraction of the lattice constant a by which the substrates are relatively displaced in the lateral (x) direction, m is the mass of the substrate atom and the angular brackets signify the ensemble average.

3. Results of MC simulations

We performed isobaric-isothermal MC simulations on the model tribological system using the prescription previously detailed [5,6]. The configurational energy is given by equation (4) and the shear stress by equation (5). The various parameters of the system are listed in the caption of figure 1. Numerical values are expressed in dimensionless units based on the LJ potential parameters for the substrate-substrate (ss) interaction.

We studied the behavior of the system under *reversible* shearing for several film-substrate coupling strengths ϵ_{fs} (i.e. the depth of the attractive well of the LJ potential between a film atom and a substrate atom) at fixed load $T_{yy}=-0.1$ (i.e. the film is under compression) and temperature $T=0.1$. Figure 2(a) displays shear–stress profiles (plot of shear stress versus register) for the case $\epsilon_{fs}=1/9$ obtained from the dynamically-constrained HACG [5,6], the current free-energy corrected HACG and the exact (fully atomistic) treatments. Each profile consists of two plots corresponding to shearing in the "forward" ($\alpha=0\rightarrow\alpha=1.2$) and "reverse" ($\alpha=1.2\rightarrow\alpha=0$) directions. Since we regard shearing as a quasistatic (reversible) process, each point (α, T_{yx}) corresponds to an equilibrium state of the system. The general characteristics of the shear–stress profile have been discussed in detail [5,6]. Figure 2(a) shows the free-energy corrected HACG profile to be in very good agreement with both the "exact" (fully atomistic) and dynamically-constrained HACG profiles. This suggests that the HACG technique, with or without the free-energy correction, can reliably account for the elastic response of the far regions. In contrast, however, we note from figure 2(b) that the dynamically-constrained HACG method fails to yield the correct mean-separation profile (i.e. plot of mean distance between substrates $\langle L_y\rangle$ versus register), whereas the corrected HACG mean-separation profile is in excellent agreement with the exact one. The error in the dynamically-constrained HACG profile is due to the suppression of random (thermal) motions of underlying

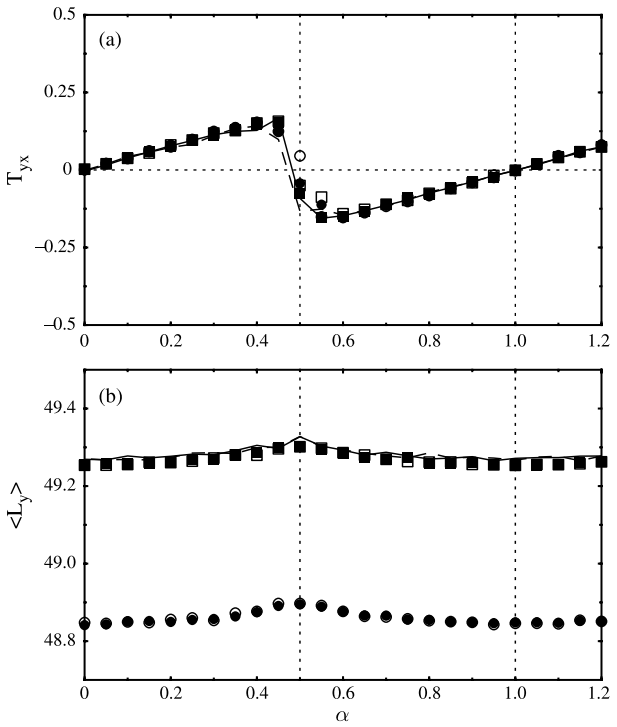

Figure 2. (a) Shear stress profile and (b) mean-separation profile for $\epsilon_{fs}=1/9$. "Exact" (fully atomistic) profile (forward, solid line; reverse, dashed line); dynamically-constrained HACG profile (forward, filled circles; reverse, open circles); free-energy corrected HACG profile [see equation (5)] (forward, filled squares; reverse, open squares). α is fraction of cell constant a by which the upper wall is displaced in x-direction relative to bottom wall (see figure 1).

non-nodal atoms, which prevents the far regions of the substrates from undergoing thermal expansion.

Figures 3 and 4 exhibit profiles for greater film-substrate coupling strengths. Again, we see that both dynamically-constrained and free-energy corrected HACG shear–stress profiles accord well with the exact ones, but that the dynamically-constrained HACG mean-separation profile disagrees markedly with both its free-energy corrected HACG and fully atomistic counterparts. We also note that in the case $\epsilon_{fs} = 1/3$ the effective elastic constant (i.e. the slope of the linear segment of the shear stress profile) for the corrected HACG profile agrees somewhat better with the exact effective elastic constant than does that for the dynamically-constrained HACG profile.

4. Conclusion

These comparisons demonstrate that the free-energy corrected HACG technique eliminates the spurious effects introduced by the dynamical constraints implicit in the original HACG treatment.

Figures 2–4 show that the dynamically constrained HACG method yields mean-separation profiles ($\langle L_y \rangle$ vs α) in disagreement with both the exact profiles and the free-energy corrected ones. This discrepancy is of course due to the neglect of thermal motion of the (dynamically constrained) non-nodal atoms in the vast coarse-grained regions of the substrates. In other words, thermal expansion of the substrates is suppressed. The impact of this suppression on the shear–stress profiles at *constant normal load* is essentially negligible. In sharp contrast, however, shear–stress profiles at *constant wall separation* (i.e. constant density) would be expected to suffer a huge impact. The implication of this observation for the multiscale simulation of a practical device, such as the atomic force microscope, is significant. In *constant-force* mode the neglect of thermal expansion of the substrate would have little effect on the shear–stress profile. On the other hand, such neglect would be expected to distort grossly the profiles measured in *constant-height* mode.

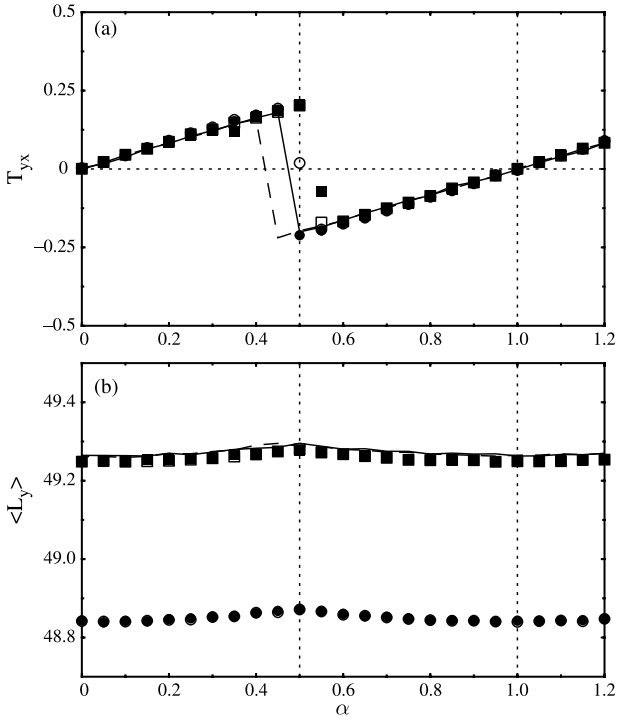

Figure 3. Same as figure 2, except $\epsilon_{fs} = 1/6$.

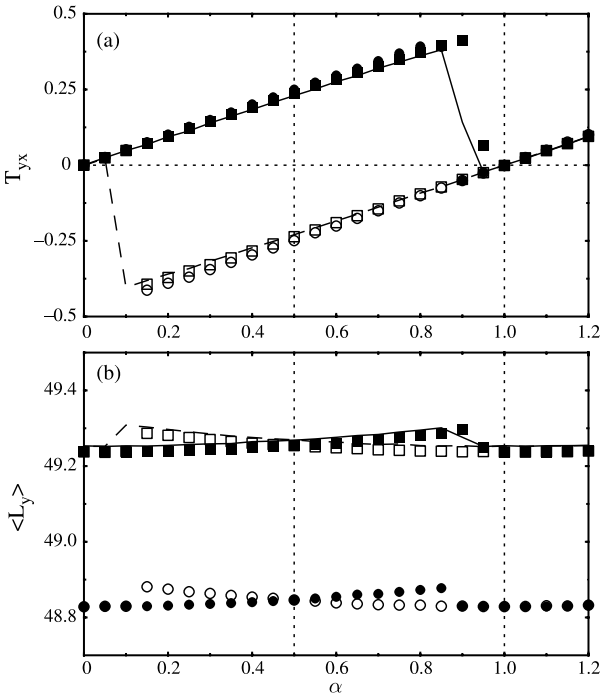

Figure 4. Same as figure 2, except $\epsilon_{fs} = 1/3$.

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