

A direct two-dimensional pressure formulation in molecular dynamics

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ABSTRACT

Two-dimensional (2D) pressure field estimation in molecular dynamics (MD) simulations has been done using three-dimensional (3D) pressure field calculations followed by averaging, which is computationally expensive due to 3D convolutions. In this work, we develop a direct 2D pressure field estimation method which is much faster than 3D methods without losing accuracy. The method is validated with MD simulations on two systems: a liquid film and a cylindrical drop of argon suspended in surrounding vapor.

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Multiscale coupling of atomistic and continuum simulations is of significant importance in the areas of heat transfer, fracture mechanics and bioengineering [1]. These computations typically map properties determined from atomistic simulations onto grid points in continuum simulations [2]. In the atomistic system, a very important property is estimation of pressure or stress from which interfacial energies, surface tension, pressure gradients can be computed. Further, many simulated systems have inhomogeneity only in two dimensions (2D) and thus only require 2D pressure distribution; examples include defect nucleation in bulk and two-dimensional crystals, bio molecular assemblies such as lipid bilayers and membrane proteins, as well as thin film evaporation and heat transfer. However, local pressure estimation is currently based on 3D [6] or 1D [7] pressure estimation. The 2D pressure distribution in a system is obtained by averaging over the 3D pressure data, and is extremely computationally expensive [4] as it involves a 3D convolution. A generalized method for 3D stress calculations which included temporal averaging weight functions was derived by Yang [3]. Recently, Vanegas [4] and Sanchez et al. [5] applied the modified Hardy versions of IK stress to lipid bilayers, coiled coil protein and graphene sheet to determine continuum level properties from atomistic simulations. Further, there exist a few Irving-Kirkwood versions [8] of 1D pressure calculations for 1D inhomogeneous system. However, to the best of our knowl-

edge, no methods are present for a direct 2D pressure estimation. This work presents a 2D pressure estimation algorithm based on Hardy's stress method, and is validated by comparing the results from molecular simulations of suspended liquid film and cylindrical drop with experimental data and classical Young-Laplace equation, respectively.

Historically, the atomic level virial stresses from statistical analysis were first derived by Irving and Kirkwood [9], now generally referred to as IK method. The need for large ensemble averaging due to the delta function in IK method was circumvented by Hardy in his classical paper [8,10] by introducing a spreading function and a bond function. The virial stress has two components, a kinetic component and a force component. There existed an ambiguity among researchers about the equivalence of virial stress with Cauchy stress. The ambiguity is thoroughly discussed in Zhou's paper [11] which claimed that Cauchy stress is not equivalent to virial stress, as the continuum level Cauchy stress is equivalent only to the force component of virial stress. Based on this finding, researchers [12] performed a number of molecular studies. Zimmerman [12a] showed that, for crystals, Hardy's stress formulation gave more accurate results than simple local virial averages. A comparative study of different versions of local virial stress was studied by Murdoch [13]. In contrast to Zhou's work [11], Subramaniyan [14] found that virial stress is indeed the Cauchy stress using specific examples. There were other works [15] which tried to develop the appropriate relation of virial stress and continuum level stresses. Our work also supports the fact that both kinetic com-

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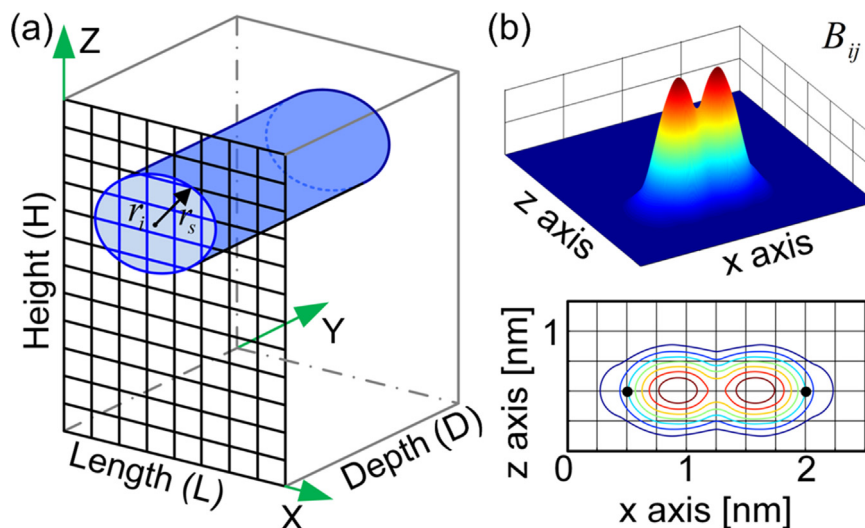


Fig. 1. Weight and bond function developed for two-dimensional pressure formulation. (a) Cylindrical volume of influence associated with an atom located at r_i , where r_s is the spread radius, L, D, H are length, depth and height respectively. (b) Visualization of bond function for two atoms separated at a distance of 1.5 nm. The gradient image (lower) shows the surface plot of the same.

ponent and force component of virial stress should be considered when converting virial stress to a continuum level property.

The 2D pressure estimation method presented in this work is developed by reformulating the 3D weight function, and thus the 3D method is described first. In the 3D method, the modified version of IK stress developed by Hardy [8c] has a kinetic component and a virial component. The 3D pressure estimation methodology lies in smearing the kinetic and the virial component into a 3D grid using a weight function and a bond function respectively. The components are smeared into a spherical volume around the particle location. A typical weight function, as used by researchers [3,15a] for 3D grid, is given as:

$$w(r) = C_1 \left[1 - 3r^2/r_s^2 + 2r^3/r_s^3 \right] \quad (1)$$

Specifically, a 3D pressure method requires $N^2 \times N_X \times N_Y \times N_Z \times N_B$ operations, which can be decreased to $N^2 \times N_X \times N_Z \times N_B$ using the outlined 2D pressure method (N is the number of atoms; N_X , N_Y and N_Z are the number of grid cells along x, y and z-directions respectively; N_B is the number of discrete points for bond function).

The application of pressure estimation theory to a 2D grid involves changing the spherical volume to a cylindrical volume as shown in Fig. 1a. The resulting 2D weight function w is (please refer to supporting information for detailed derivation):

$$w(r) = 10 \left[1 - 3r^2/r_s^2 + 2r^3/r_s^3 \right] / (3D\pi r_s^2) \quad (2)$$

where D is the depth of the system along Y (direction of homogeneity) as shown in Fig. 1a, r_s is the ‘spread’ radius or the ‘smearing’ radius. Thus, based on the new weight function, the Bond function (B_{ij}), pressure tensor (P), and density of the system (ρ) are defined as:

$$B_{ij}(r_p) = \int_0^1 w(\lambda r_{ij} + r_i - r_p) d\lambda \quad (3)$$

$$P(r_p) = \sum_{i=1}^N m_i v_i \otimes v_i w(r_i - r_p) + \sum_{i=1}^{N-1} \sum_{j=i+1}^N r_{ij} \otimes F_{ij} B_{ij}(r_p) \quad (4)$$

$$\rho(r_p) = \sum_{i=1}^N m_i w(r_i - r_p) \quad (5)$$

where, r_i is the position of i^{th} atom, r_p is the position vector of p^{th} grid point, F_{ij} is the force between two atoms, $r = r_i - r_p$, m_i is the mass of i^{th} atom, v is the velocity and $r_{ij} = r_i - r_j$. Fig. 1b shows the variation of bond function for a pair of atoms kept at 1.5 nm apart. The isometric view shows the variation of magnitude of bond function for a spread radius of 0.5 nm. For grid dependent and finite support weight functions like B-splines, a rectangular prism volume should be used instead of cylindrical volume. The computational cost gain from 2D pressure estimation over 3D pressure calculation is also qualitatively discussed in the supporting information. Additional formulated 2D forms for some selected functions, along with their 3D functions are included in supporting information. For completeness, we have also derived the 1D variation of pressure and density which is very suitable for 1D inhomogeneous systems like pressure in thin films, lipid bilayers etc. (details can be found in supporting information and is consistent with the derivation of Hardy stress [8c]).

In order to demonstrate and validate the new 2D pressure formulation, we apply it to study the pressure, surface tension and density variations of argon liquid films suspended in argon vapor using molecular dynamics (MD) simulations. In our chosen example (argon liquid film in vapor), the inhomogeneity is in two dimensions (say, X and Z axes) and there is no significant density variation along the third dimension (Y axis). The MD domain is shown in Fig. 2a. A self-written C++ molecular dynamics code is used for all simulations. The argon liquid film is 10 nm thick with 7.5 nm thick argon vapor on either side along the z-direction. The X–Y cross section size is 5 nm \times 5 nm. Periodic boundary conditions are applied in all directions. The modified Stoddard-Ford LJ potential [16] for argon interactions is used with argon – argon LJ parameters as $\sigma_{Ar-Ar} = 0.34$ nm and $\epsilon_{Ar-Ar} = 1.005841$ kJ/mol. The time step of velocity verlet integration was 5 fs and the thermostat was velocity scaling. The vapor and liquid domains in this molecular system are first equilibrated separately [8b] for 1000 ps in order to get a stable suspended film. The simulation is run for another 1000 ps on which statistical analysis is performed. Using the developed 2D formulation, the temporally averaged 2D contours of density and pressure at 90 K are estimated and shown in Figs. 2b and c. Further, a number of simulations were performed to study the effect of varying cutoff radius, spread radius and system temperatures.

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