Parametric studies of the thermal and momentum accommodation of monoatomic and diatomic gases on solid surfaces

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ABSTRACT

The thermal accommodation coefficient (TAC) and the momentum accommodation coefficient (MAC) are the two fundamental parameters quantifying the solid–gas energy and momentum exchange efficiencies. We use molecular dynamics (MD) simulations to study the effect of individual interfacial parameters including, (i) solid–gas interaction strength, (ii) gas–solid atomic mass ratio, (iii) solid elastic stiffness, and (iv) temperature, on TAC and MAC at solid surfaces in contact with monoatomic and diatomic gases. In addition to offering a fundamental understanding on how these individual parameters affect the nature of gas–solid collisions, we provide an extensive database for the TAC and MAC. We also study the effect of surface functionalization with molecular monolayers on the energy and momentum transfer at the interface. These results are useful in developing interfaces with enhanced heat transfer under various operation conditions.

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1. Introduction

Convective heat transfer in rarefied gases is of great importance to the thermal management of microelectronic devices [1–3], microelectromechanical systems (MEMS) based devices [4], and aircraft flying at high altitudes [5,6]. The degree of rarefaction of a gas can be described by the Knudsen number $Kn = \lambda/L$, where λ is the mean free path of gas molecules and L is a characteristic dimension. A gas is regarded as rarefied if $Kn > 0.01$ [6], which means λ is not negligible compared to L. This happens in two ways, either because of a large λ, or due to a very small L. The first case occurs when gas pressure is very low, for example air at very high altitudes. The second case occurs when the dimensions of objects are very small, for example micro/nano-channels in cooling element of microelectronic devices or MEMS based devices. In both cases, the number of collisions between the gas molecules and the wall is larger than the number of collisions between the molecules in vicinity of the solid wall. Accordingly, the energy and momentum transfer at solid–gas interfaces could strongly affect the overall heat transfer efficiency.

In order to investigate the heat transfer to a body in a rarefied gas, the well-established continuum theories must be modified to account for the velocity slip and temperature jump at the

solid wall [4–6]. According to the gas kinetic theory [7,8], the velocity-slip and temperature-jump boundary conditions can be incorporated by introducing two parameters called momentum accommodation coefficient (MAC), defined as the fraction of gas molecules reflected diffusely from the solid surface, and thermal accommodation coefficient (TAC), defined as the fraction of gas molecules incident on the surface scattered in thermal equilibrium with the surface.

Despite the fact that the subjects of the TAC and MAC have over a century-long history of investigation, presently no quantitative theory exists and there are only semi-quantitative or empirical formulas available to fit experimental data [9]. These formulas have limited applicability to a wide range of gas–solid interfaces and temperatures. Moreover, the role of individual parameters is often difficult to determine in experiments [6]. To understand and quantify the role of individual interfacial parameters and to determine how these parameters affect the nature of gas–solid collisions and the value of the TAC and MAC, we resort to molecular dynamics (MD) simulations and a suite of molecular-level thermal characterization techniques.

Molecular simulations are uniquely positioned to explore the mechanism behind the energy and momentum transport at solid–gas interfaces. In the past decade, the MD simulation has been utilized to study the TAC and MAC of noble gases on various solid surfaces [1,2,10–12]. However, each of these work only investigated one or two factors that influence TAC or MAC.

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Moreover, a systematic study of effects of various parameters on the TAC and MAC of diatomic or polyatomic gases is not available. In this work, we carry out a systematic study of the effects of various parameters of importance, including the solid–gas interaction strength, the gas–solid mass ratio, and the temperature and elastic modulus of solid on the TAC and MAC. In addition to study the accommodation coefficients of monoatomic gases on solid surfaces, we also investigate the TAC and MAC of diatomic gases and the effect of surface modification such as the functionalization of the solid with self-assembled monolayers (SAMs) on thermal and momentum accommodation of various gases. Through the extensive MD simulations, we will generate a database for TAC and MAC and provide a fundamental understanding of the nature of thermal and momentum accommodation for monoatomic and diatomic molecules.

2. Determination of TAC and MAC using MD simulations

2.1. Theory

In the case of monoatomic gases TAC is defined by [9]

$$\alpha_T = (T_r - T_i)/(T_s - T_i) \tag{1}$$

where T_i and T_r are the temperatures of incident and reflected gas atoms, respectively, and T_s is the solid surface temperature. $\alpha_T = 1$ means there is a complete thermal equilibration of the incident gas stream with the solid. In the case of diatomic gases, the TAC can be also calculated by Eq. (1), if the diatomic molecule can be approximated as a classical rigid rotor. For diatomic gases, we can further calculate the translational and rotational temperatures of gas molecules, thus obtain the TACs for translational and rotational molecular motions.

In the temperature jump regime ($0.01 < Kn < 0.1$), TAC relates to the solid–gas interfacial thermal conductance G_K by [8]

$$G_K = f k_B N \alpha_T/(2 - \alpha_T) \tag{2}$$

where $f = 4$ for a monoatomic gas and $f = 6$ for a diatomic gas, k_B is the Boltzmann constant, and N is collision rate per unit area. The collision rate is a function of pressure, P, temperature, T, and the atomic mass m, and it is given by [8]

$$N = P/\sqrt{2\pi m k_B T} \tag{3}$$

The interfacial thermal conductance can be readily obtained from non-equilibrium MD (NEMD) simulations using

$$G_K = q/\Delta T \tag{4}$$

where q is the heat flux across the solid–gas interface, and ΔT is a temperature jump at the solid–gas interface. Hence, the relation between G_K and α_T given by Eq. (2) can be used to verify the consistency of the TAC calculated directly by Eq. (1) and via Eq. (2).

Similar to Eq. (1), the equation to calculate tangential-MAC is [13]

$$\alpha_v = (v_{x,r} - v_{x,i})/(v_{x,s} - v_{x,i}) \tag{5}$$

where $v_{x,i}$ and $v_{x,r}$ are the tangential velocities of incident and reflected gas molecules, respectively, and $v_{x,s}$ is the tangential velocity of solid surface. $\alpha_v = 1$ means all gas molecules are diffusely scattered by the surface. If $v_{x,s} = 0$ which is the case in our MD simulation, Eq. (5) can be simplified to Eq. (6).

$$\alpha_v = 1 - v_{x,r}/v_{x,i} \tag{6}$$

According to Eq. (6) $v_{x,r}$ is linearly proportional to $v_{x,i}$ if the macroscopic velocity of the solid is zero. This relation is utilized in our MD simulations to calculate the tangential-MAC.

2.2. MD model

The model system consists of a solid Au slab in contact with Ar or N_2 gas, as depicted in Fig. 1(a). The Au slab is formed by a nine layers of FCC (1 1 1) planes of solid Au with a cross section area of 5.2 nm by 5.0 nm. The length of the gas region, L, is 102 nm. Periodic boundary conditions are applied in all three directions. The embedded-atom-method (EAM) potential [14] is used for Au–Au interactions. The Lennard-Jones (LJ) 12-6 potential, with parameters $\sigma = 3.41$ Å and $\varepsilon = 10.3$ meV [15], is employed for Ar–Ar interactions. The N_2 molecule with a bond length of 1.10 Å [16] is considered as a rigid rotor in the MD simulation. The LJ potential, with parameters $\sigma = 3.31$ Å and $\varepsilon = 3.21$ meV [17], is employed for N–N interactions between molecules.

In the case of Au surfaces functionalized with SAMs, 120 1-octanethiolate [-S-(CH$_2$)$_7$-CH$_3$] chains are covalently bonded to Au surface and form a two-dimensional triangular lattice with a surface density of $1/(21.6$ Å$^2)$. The interatomic interactions for SAM molecules are described by the Hautman–Klein united atom (UA) model [18], which treats the hydrocarbon groups as single interaction sites. LJ potentials are used for non-bonded interactions in SAM molecules in the UA model. The Koike–Yoneya bond stretching potential [19] is included in the UA potential to take into account the C–C and C–S bond stretching motions. The Au–S interactions are modeled by the Morse potential $V(r) = D(e^{-2a(r-r_e)} - 2e^{-a(r-r_e)})$ with parameters $r_e = 2.65$ Å, $a = 1.47$ Å^{-1} and $D = 0.15$ eV [1,20]. LJ potentials for interactions between Au and other atoms in SAMs are taken from the universal force field (UFF) [21].

The solid–gas interactions are all described by the LJ potential with parameters calculated by the Lorentz–Berthelot mixing rule.
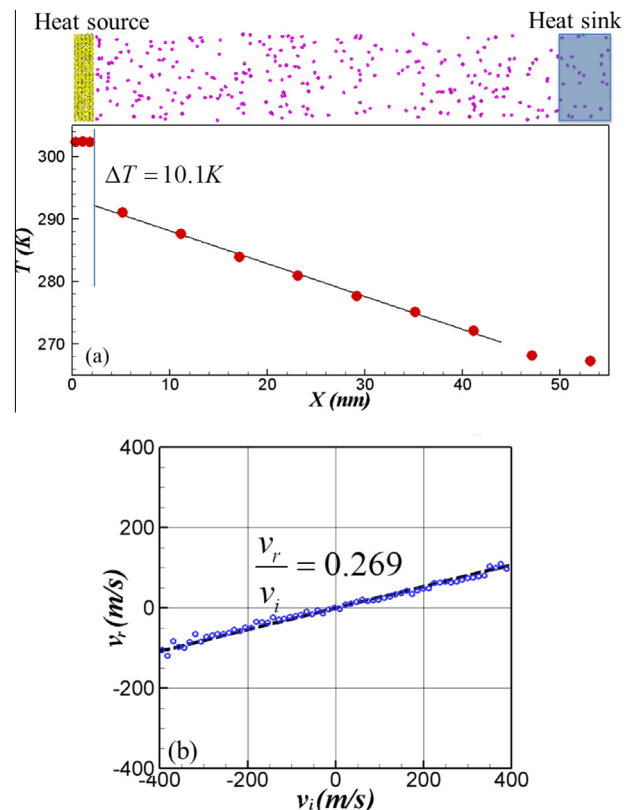

Fig. 1. (a) A snapshot of solid–gas system and the steady state temperature profile. Due to the symmetry of the system, only half of the simulation box is shown. (b) The reflected tangential velocity as a function of the incident tangential velocity. The dashed line is a linear fit to the simulation data.