Absolute momentum transfer in gas-surface scattering

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Two new parameters called the reduced force coefficients are introduced that eliminate the singularity problems associated with the momentum accommodation coefficients and completely describe the macroscopic average momentum transfer to a surface by an incident gas. In addition, the reduced force coefficients can be used to determine the flux-weighted average velocity and translational energy of molecules scattered from a surface, and can be used to approximate the energy accommodation coefficient for many gas-surface interactions. The reduced force coefficients for molecular beams of H_2 and N_2 incident upon SiO_2-coated Kapton and Z-93–coated aluminum were obtained using a specialized torsion balance to measure the magnitudes and directions of the forces exerted on the surfaces by the molecular beams. [S1063-651X(97)50804-X]

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I. INTRODUCTION

Understanding both macroscopic and microscopic properties of momentum and energy transfer between gas and surface molecules is of fundamental and practical importance. Traditionally, gas-surface interactions have been investigated through detailed studies involving measurements of the velocity, angular, and internal state distribution functions of the scattered molecules [1–4]. When molecules scatter from a surface, the exchange of momentum and energy between gas and surface molecules, in general, results in a nonzero force exerted on the surface and a change in surface temperature [1–3]. Because the force on the surface is directly related to the interaction potential between gas and surface molecules [1,2], microscopic details of gas-surface interactions can also be investigated by analyzing the average forces exerted on surfaces by incident gases. In addition, measurements of these forces can be used to obtain macroscopic properties of the scattered molecules, such as the flux-weighted average (hereafter referred to as average) [5] velocity, and average translational energy. This approach bypasses the problems associated with determining macroscopic quantities from measurements of the distribution functions of the scattered molecules [6,7].

In the past, the tangential and normal momentum accommodation coefficients [8] have been used to characterize measurements of the momentum transferred to surfaces by incident gases in rarefied flow regimes. Singularities, however, render the momentum accommodation coefficients nearly unusable for many applications [9]. In this paper a new formalism is presented that eliminates these singularity problems and can be used to completely characterize measurements of the tangential and normal components of the momentum transferred to surfaces by incident gases. The formalism is based upon two new parameters called the reduced force coefficients that can be obtained for a particular gas-surface interaction by measuring the magnitude and direction of the force exerted on the surface by a molecular beam of the gas, the average velocity of the molecular beam, and the number of incident molecules per unit time. These coefficients can be used to approximate the energy accommodation coefficient [1,8] for gas-surface interactions where differences between the average energies associated with the internal state distribution functions of the incident and scattered molecules can be neglected. Also, the reduced force coefficients can be used to calculate the average velocity and average translational energy of the scattered molecules. By analyzing these macroscopic average quantities of the scattered molecules, microscopic details of the gas-surface interaction can be determined.

The reduced force coefficients were determined for H_2, N_2, CO, and CO_2 incident upon a solar cell array material, a polyimide plastic with a trade name of Kapton, SiO_2-coated Kapton, and a coating material used for thermal insulation on space craft surfaces called Z-93. These particular gas-surface interactions are important in the design of the International Space Station. Only the results obtained for H_2 and N_2 incident upon SiO_2-coated Kapton and Z-93 will be presented in this paper. Details determined from these measurements parallel those obtained for the other gas-surface interactions. The forces were measured using a specialized torsion balance with a total uncertainty of less than $\pm 1.5\%$. Molecular beams were incident upon the surfaces at 0°, 25°, 50°, 75°, and 85°. The velocity distribution functions of the molecular beams were measured using standard time-of-flight methods. With the nozzle temperature at 295 K, the average velocities of H_2 and N_2 were 2590 and 1870 m/s, respectively. The seeded beam method [10] was used to accelerate N_2 to this velocity. Uncertainties in the average velocities were estimated to be approximately $\pm 2\%$. The absolute flux densities of the molecular beams were measured using a torsion balance fitted with a beam stop that nullified the force due to the exiting gas molecules and separately using the effusive method. Uncertainties in the flux density measurements were estimated to be approximately $\pm 5\%$. Complete descriptions of the experimental details are given elsewhere [11,12].

II. THE MOMENTUM ACCOMMODATION COEFFICIENTS

The tangential σ and normal σ' momentum accommodation coefficients can be expressed as [8]

$$\sigma = \frac{\overline{v_i}\sin\theta_i - \overline{v_s}\sin\theta_s}{\overline{v_i}\sin\theta_i}, \tag{1}$$

$$\sigma' = \frac{\overline{v_i}\cos\theta_i - \overline{v_s}\cos\theta_s}{\overline{v_i}\cos\theta_i - \sqrt{\pi kT/2m}}, \tag{2}$$

where $\overline{v}$ is the magnitude of the average velocity, θ is the angle between the average velocity and the surface normal, the subscripts i and s represent the incident and scattered molecules respectively, k is Boltzmann's constant, T is the temperature of the scattering surface, and m is the mass of the individual gas molecules. The quantity $\sqrt{\pi kT/2m}$ is the average velocity of the scattered molecules assuming diffuse scattering from the surface with complete thermal accommodation [8].

For many applications the tangential momentum accommodation coefficient has a removable singularity at an angle of incidence θ_i normal to the surface, and is well behaved. However, for applications where θ_s is not equal to zero at normal angles of incidence, the tangential coefficient would diverge. Problems with the singularity in the normal momentum accommodation coefficient result in divergent behavior for essentially every application involving a directed gas flow. In addition, the angle of incidence at which the singularity occurs is a function of the average velocity of the incident molecules and the temperature of the scattering surface. For the gas-surface interactions considered in this paper, the angle of incidence at which the singularity occurred ranged from 55° to 85°. The divergent behavior caused by this singularity made it nearly impossible to use the normal coefficient.

To overcome this singularity problem, Knuth [9] defined a normal momentum transfer coefficient that eliminates the functional dependence of the singularity upon the average velocity of the incident molecules and the surface temperature. However, the coefficient diverges at large angles of incidence for all nonspecular scattering processes. Therefore, a new formalism was developed that completely eliminated the problems caused by singularities.

III. THE REDUCED FORCE COEFFICIENTS

The definitions of the reduced force coefficients were arrived at by considering the force exerted on a surface by an incident molecular beam. The singularity problems were eliminated by defining the coefficients with denominators that are independent of the incident flow direction. The tangential f_t and normal f_n reduced force coefficients are defined as

$$f_t = \frac{\overline{p_{it}} - \overline{p_{st}}}{\overline{p_i}}, \tag{3}$$

$$f_n = \frac{\overline{p_{in}} + \overline{p_{sn}}}{\overline{p_i}}, \tag{4}$$

where $\overline{p}$ is the magnitude of the average momentum, and the subscripts t and n are the tangential and normal components, respectively. The reduced force coefficients can also be written as $f_t = F_t/N_i m\overline{v_i}$ and $f_n = F_n/N_i m\overline{v_i}$, where F is the magnitude of the force exerted on the surface by the incident gas, and N_i is the total incident flux. Thus, the coefficients can be determined from measurements of F_t, F_n, N_i and $\overline{v_i}$. Equations (3) and (4) can be used to express the magnitude and direction of the average velocity of the scattered molecules as

$$\overline{v_s} = \overline{v_i}\sqrt{(f_t - \sin\theta_i)^2 + (f_n - \cos\theta_i)^2}, \tag{5}$$

$$\theta_s = \tan^{-1}\left(\frac{\sin\theta_i - f_t}{f_n - \cos\theta_i}\right). \tag{6}$$

It is assumed that the average velocity of the scattered molecules is in the plane formed by the incident gas flow direction and the surface normal.

A new parameter μ, called the scalar momentum accommodation coefficient, is introduced and defined as

$$\mu = \frac{\overline{p_i} - \overline{p_s}}{\overline{p_i} - \overline{p_d}}. \tag{7}$$

The quantity $\overline{p_d}$ is the magnitude of the average momentum of the scattered molecules assuming diffuse scattering and is given by $\sqrt{\pi kTm/2}$. The scalar momentum accommodation coefficient ranges between zero for specular scattering and one for diffuse scattering with complete thermal accommodation and is well defined for all angles of incidence.

For applications where differences between the average energies associated with the internal state distribution functions of the incident and scattered molecules can be neglected, the energy accommodation coefficient ϵ can be written as [1,8]

$$\epsilon = \frac{\overline{v_i^2} - \overline{v_s^2}}{\overline{v_i^2} - 4kT/m}, \tag{8}$$

where $\overline{v^2}$ is the average of the velocity squared, and $4kT/m$ is the average of the velocity squared of the scattered molecules assuming diffuse scattering. A knowledge of the reduced force coefficients is not sufficient to determine the energy accommodation coefficient using Eq. (8). Additional information is required to determine $\overline{v_s^2}$. The simple assumption of equating $\overline{v_s^2}$ with v_s^2 can result in errors as large as 60%. A more accurate approximation for $\overline{v_s^2}$ can be made with an energy accommodation coefficient ϵ' based on the average velocities of the incident and scattered molecules. The coefficient is defined as

$$\epsilon' = \frac{\overline{v_i^2} - \overline{v_s^2}}{\overline{v_i^2} - \pi kT/2m}. \tag{9}$$

From a knowledge of the reduced force coefficients and the velocity distribution function of the molecular beam, the coefficient ϵ' can be calculated. The uncertainty associated with the approximation of equating ϵ' with ϵ has been shown to be less than $\pm 1\%$ for gas-surface interactions where the average energy of the incident molecules is large compared to kT [11]. With this approximation, $\overline{v_s^2}$ can be written as

$$\overline{v_s^2} = \overline{v_i^2} - \epsilon'(\overline{v_i^2} - 4kT/m). \tag{10}$$

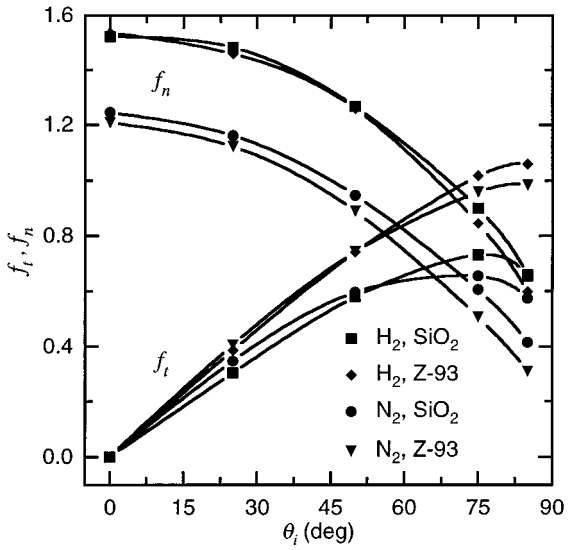

FIG. 1. The reduced force coefficients for molecular beams of H_2 and N_2 with respective average velocities of 2590 and 1870 m/s incident upon SiO_2-coated Kapton and Z-93.

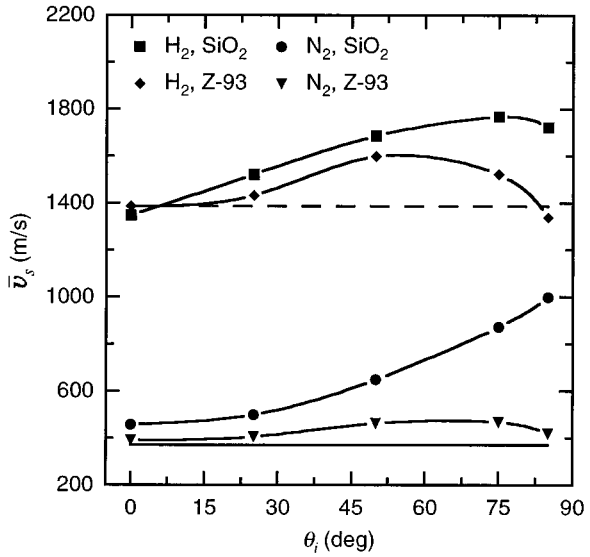

FIG. 2. The average velocities of the scattered molecules as a function of the angle of incidence for molecular beams of H_2 and N_2 with respective average velocities of 2590 and 1870 m/s incident upon SiO_2-coated Kapton and Z-93. The dashed and solid lines represent diffuse scattering with complete thermal accommodation for H_2 and N_2, respectively.

The quantity $\overline{v_i^2}$ can be determined from a knowledge of the velocity distribution function of the incident molecules. The average translational energy of the scattered molecules can be obtained using Eq. (10).

IV. EXPERIMENTAL RESULTS

Figure 1 shows the angular dependence of the reduced force coefficients for H_2 and N_2 incident upon surfaces of SiO_2-coated Kapton and Z-93. The normal reduced force coefficient has a strong dependence upon the gas and a weak dependence upon the surface, with the absolute values for H_2 approximately 25% larger than for N_2. The tangential reduced force coefficient has a weak dependence upon the gas and a strong dependence upon the surface for larger angles of incidence. The tangential coefficient would be close to zero for specular-type scattering and have a $\sin\theta_i$ dependence for diffuse-type scattering with complete thermal accommodation. Thus, the tangential coefficient data indicate that the scattering becomes more specular as the angle of incidence increases for SiO_2-coated Kapton and is almost entirely diffuse with complete thermal accommodation at all angles of incidence for Z-93. Scanning electron microscope photographs of the surfaces revealed that Z-93 was rough on a 100 μm scale, while SiO_2-coated Kapton was smooth on a 1 μm scale. This contrast in surface roughness should account for much of the scattering differences between the two surfaces.

The magnitudes and directions of the average velocities of the scattered molecules are shown in Figs. 2 and 3. For H_2 and N_2 incident upon SiO_2-coated Kapton both $\overline{v_s}$ and θ_s generally increase with the angle of incidence. This behavior is consistent with the scattering becoming more specular for increasing angles of incidence. These results are in contrast to those observed for Z-93, where the variation in $\overline{v_s}$ is small and θ_s is close to zero for all angles of incidence, implying that the scattering is nearly diffuse with complete thermal accommodation.

Figure 3 shows that for SiO_2-coated Kapton at large angles of incidence, the scattering angles for N_2 are larger than for H_2. This effect can be explained by noting that the mass of H_2 is significantly smaller than the mass of N_2 and a given force perpendicular to the direction of motion would deflect H_2 more than N_2 and result in a smaller scattering angle. Thus, for large angles of incidence where the scattering becomes more specular, the large repulsive force exerted on the molecules by the surface will deflect H_2 more than N_2.

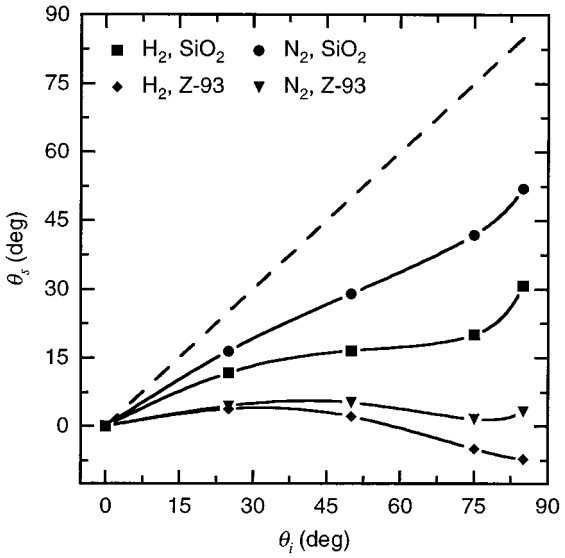

FIG. 3. The scattering angle θ_s as a function of the angle of incidence for molecular beams of H_2 and N_2 with respective flux-weighted average velocities of 2590 and 1870 m/s incident upon SiO_2-coated Kapton and Z-93. The dashed line represents specular scattering.

For the two surfaces, the magnitude of the average velocity of the scattered H_2 molecules increases with the angle of incidence, and then begins to decrease at large incident angles. This result seems to imply that H_2 becomes more accommodated in this limit. However, the scattering angles for H_2 incident upon SiO_2-coated Kapton seem to indicate that the scattering becomes more specular at increasing angles of incidence. This apparent contradiction can be resolved by considering that the total time during which the incident gas molecules interact with the surface would increase with increasing angles of incidence. As the H_2 molecules approach the surface, the attractive van der Waals forces would act on the gas molecules over a longer period of time, thereby increasing the momentum loss to the surface. Likewise, as the scattered H_2 molecules recede from the surface, their decreased velocities also allow for a longer interaction time. Since the attractive forces will tend to be along the surface normal, the receding H_2 molecules will lose more of their normal momentum component while retaining most of their tangential component [4]. As a result, the molecules scatter closer to the specular direction for very large angles of incidence. This effect may not be observable for N_2 due to its larger incident momentum.

Figure 4 shows the scalar momentum accommodation coefficient for the various gas-surface interactions. These results show that even for technical surfaces, the simple assumption of diffuse scattering with complete thermal accommodation is entirely inadequate. For a particular gas, the accommodation for Z-93 is greater than for SiO_2-coated Kapton. Accommodation generally decreases as the angle of incidence increases for SiO_2-coated Kapton, while N_2 scatters with near complete accommodation from Z-93.

Comparisons of these results with data measured for similar gas-surface interactions are difficult to make due to the limited number of experimental investigations related to momentum transfer in gas-surface interactions. The results obtained from a number of experiments are compared elsewhere [6]. The majority of these studies used beams of N_2^+ and Ar^+ in the energy range of 15–25 eV. The results obtained by various investigators agree qualitatively that the normal momentum accommodation coefficient decreases as the angle of incidence increases. However, the measurements of the tangential momentum accommodation coefficient show no agreement between different investigators.

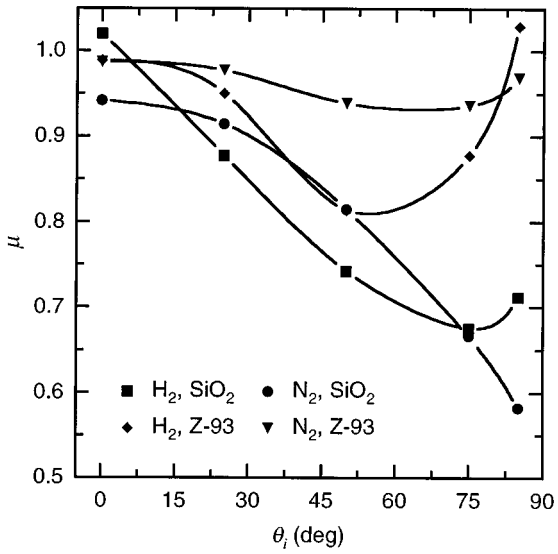

FIG. 4. The scalar momentum accommodation coefficient as a function of the angle of incidence for molecular beams of H_2 and N_2 with respective average velocities of 2590 and 1870 m/s incident upon SiO_2-coated Kapton and Z-93.

It has been shown that measurements of both the incident molecular beam velocity distribution function and the magnitude and direction of the forces exerted on a surface by the incident beam can be used to obtain reliable and accurate information concerning the average velocity and average translational energy of the scattered molecules. These new results and methods of analyzing forces exerted on surfaces by incident gases will be extremely useful in the study of gas-surface interactions.

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