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Understanding gas-surface interactions from direct force measurements using a specialized torsion balance

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The first comprehensive measurements of the magnitude and direction of the forces exerted on surfaces by molecular beams are discussed and used to obtain information about the microscopic properties of the gas-surface interactions. This unique approach is not based on microscopic measurements of the scattered molecules. The reduced force coefficients are introduced as a new set of parameters that completely describe the macroscopic average momentum transfer to a surface by an incident molecular beam. By using a specialized torsion balance and molecular beams of N_2 , CO , CO_2 , and H_2 , the reduced force coefficients are determined from direct measurements of the force components exerted on surfaces of a solar panel array material, Kapton, SiO_2 -coated Kapton, and Z-93 as a function of the angle of incidence ranging from 0° to 85° . The absolute flux densities of the molecular beams were measured using a different torsion balance with a beam-stop that nullified the force of the scattered molecules. Standard time-of-flight techniques were used to determine the flux-weighted average velocities of the various molecular beams ranging from 1600 m/s to 4600 m/s. The reduced force coefficients can be used to directly obtain macroscopic average properties of the scattered molecules, such as the flux-weighted average velocity and translational energy, that can then be used to determine microscopic details concerning gas-surface interactions without the complications associated with averaging microscopic measurements.

I. INTRODUCTION

Traditionally, experimental investigations of gas-surface interactions have been based upon microscopic measurements of the velocity, angular, and internal-state distribution functions of the scattered molecules¹⁻⁴. Alternatively, properties of gas-surface interactions can be determined from measurements of the forces exerted on surfaces by incident molecular beams⁵. When molecules scatter from a surface, the exchange of momentum and energy between gas and surface molecules, in general, results in a non-zero force exerted on the surface and a change in surface temperature. This force is directly determined by the gas-surface interaction potential. Therefore, a knowledge of the force can be used to study gas-surface interactions. In addition, this alternative approach can be used to determine the macroscopic average momentum and translational energy of the scattered molecules, thus, bypassing the problems associated with determining these quantities from measurements of the microscopic distribution functions of the scattered molecules^{6,7}.

The experiments described in this article were motivated by the need to develop accurate empirical models of the gas-surface interactions encountered in rarefied gas regimes, such as in low earth orbit. The particular interactions investigated are those between Space Shuttle reaction control jet plume gases and surfaces to be utilized in the construction of the International Space Station Alpha. The experimental apparatus was designed to have the capability of simulating the actual gas-surface interactions.

II. EXPERIMENTAL DETAILS

Measurements were made of both the magnitude and direction of the forces exerted on Space Station surfaces by molecular beams of Shuttle plume gases using a specialized torsion balance^{8,9}. The surfaces include: the solar panel array material to be used on Alpha; a polyimide plastic with the trade name Kapton; SiO_2 coated Kapton; and a material called Z-93 used as a thermal insulation coating on surfaces. Measurements were made with molecular beams of the following gases: N_2 , CO , CO_2 and H_2 .

A supersonic nozzle source was used to produce the molecular beams. The source is fabricated from nickel with an orifice diameter of 130 μm and is resistively heated by passing currents up to 900 A through the nozzle tip at rms voltages between 3 V and 7 V. A nearly identical source to the one utilized in this experiment is described in detail elsewhere¹⁰. The seeded beam technique¹¹ was used to produce molecular beams of N_2 , CO , and CO_2 that had translational energies in the range of 1 eV to 2 eV. The flux-weighted average velocities¹² of the molecular beams varied between 1700 m/s and 4600 m/s. The absolute flux-densities of the molecular beams were measured using both

the effusive method and a unique torsion balance. A complete analysis of these methods, the uncertainties associated with each method, the time-of-flight method used to determine the velocity distribution functions of the molecular beams, and the absolute flux-density of each molecular beam used in these experiments are described elsewhere⁹.

A schematic diagram of the torsion balance used to measure the forces is shown in Fig. 1. Notches cut into the sample holder allowed the angle of incidence to be adjusted in 5° increments from 0° to 85°. The angle of incidence is defined to be the angle between the surface normal and the molecular beam. Measurements were made at angles of incidence of 0°, 25°, 50°, 75°, and 85°. The component of the net force tangential to the surface was assumed to be in the plane formed by the surface normal and the incident beam. With this assumption, the magnitude and direction of the net force was determined by measuring the torque exerted on the torsion balance by the molecular beam with the lever arm in two different configurations, as shown in Fig. 2. The component of the net force parallel to the molecular beam $F_{\parallel}$ is measured with the lever perpendicular to the beam as shown in Fig. 2a, while the component perpendicular to the molecular beam $F_{\perp}$ is measured with the lever parallel to the beam as shown in Fig. 2b. Once the measurements have been made in the two configurations, the force components tangential F_t and perpendicular F_n to the scattering surface can be obtained using

$$F_t = F_{\parallel} \cos \theta + F_{\perp} \sin \theta, \quad (1)$$

and

$$F_n = F_{\parallel} \sin \theta - F_{\perp} \cos \theta, \quad (2)$$

where θ is the angle of incidence.

III. THE REDUCED FORCE COEFFICIENTS

Historically, the classically defined tangential and normal momentum accommodation coefficients¹³ have been used to understand momentum transfer in rarefied flow regimes. The singularity problems associated with these coefficients⁸, however, made it impossible to analyze the force data. Therefore, a new set of parameters, called the reduced force coefficients, were defined that eliminate the singularity problems associated with the momentum accommodation coefficients and can also be used to obtain the macroscopic average quantities of the scattered molecules. The definitions of the new coefficients were arrived at by considering the force exerted on a surface by an incident molecular beam.

The tangential f_t and normal f_n reduced force coefficients are respectively defined as

$$f_t = \frac{\bar{p}_{it} - \bar{p}_{st}}{\sqrt{\bar{p}_{it}^2 + \bar{p}_{in}^2}}, \quad (3)$$

$$f_n = \frac{\bar{p}_{in} + \bar{p}_{sn}}{\sqrt{\bar{p}_{it}^2 + \bar{p}_{in}^2}}, \quad (4)$$

where $\bar{p}_{it}$ and $\bar{p}_{in}$ are respectively the tangential and normal components of the flux-weighted average momentum of the incident gas molecules, $\bar{p}_{sn}$ and $\bar{p}_{st}$ are respectively the tangential and normal components of the flux-weighted average momentum of the scattered molecules, and the bars indicate flux-weighted averages. Eqs. (3) and (4) can also be expressed in terms of the force exerted on a scattering surface by a molecular beam as

$$f_t = \frac{F_t}{A\Phi m\bar{v}_s}, \quad (5)$$

$$f_n = \frac{F_n}{A\Phi m\bar{v}_s}, \quad (6)$$

where A and Φ are respectively the cross sectional area and absolute flux-density of the molecular beam, and m is the mass of the individual molecules in the beam. Using Eqs.(3) and (4), the magnitude $\bar{v}_s$ and direction θ_s of the flux-weighted average velocity of the scattered molecules can be expressed as

$$\bar{v}_s = \bar{v}_i \sqrt{(f_t - \sin \theta)^2 + (f_n - \cos \theta)^2}, \quad (7)$$

and

$$\theta_s = \tan^{-1} \left(\frac{\sin \theta - f_t}{f_n - \cos \theta} \right). \quad (8)$$

It is assumed that the flux-weighted average velocity of the scattered molecules is in the plane formed by the molecular beam and the surface normal. The scattering angle θ_s is the angle between the surface normal and the flux-weighted average velocity of the scattered molecules.

Energy accommodation is also of interest in the understanding of gas-surface interactions. To determine the energy accommodation coefficient¹³ for a particular interaction it is necessary to have a knowledge of the flux-weighted average translational energies and the average energies associated with the internal-state distribution functions for both the incident and scattered molecules. Because the energy associated with the internal-state distribution function of the scattered molecules cannot be obtained from only measurements of the force exerted on a surface by a molecular beam, additional experiments must be made to determine energy accommodation coefficients. In addition, errors as large as 200% can be obtained by simply equating $\bar{v}_s^2$ with the flux-weighted average of the velocity squared of the scattered molecules $\bar{v}_s'^2$. However, for many applications the force measurements can be used to provide an accurate approximation for the flux-weighted average translational energy of the scattered molecules. The approximation method makes use of the types of energy accommodation coefficients. The first coefficient is based upon the flux-weighted average of the velocity squared of the incident $\bar{v}_i^2$ and scattered molecules, and is given by

$$\alpha_{KE} = \frac{\bar{v}_i^2 - \bar{v}_s^2}{\bar{v}_i^2 - 4kT/m}, \quad (9)$$

where k is Boltzmann's constant, T is the temperature of the scattering surface, and $4kT/m$ is the flux-weighted average of the velocity squared of the scattered molecules if the molecules scatter diffusely from the surface with complete thermal accommodation. The quantity $\bar{v}_i^2$ can be determined from the velocity distribution function of the molecular beam. For cases where changes in the internal-state distribution functions can be neglected, α_{KE} is the actual energy accommodation coefficient. The second coefficient is given by

$$\alpha'_{KE} = \frac{\bar{v}_i^2 - \bar{v}_s^2}{\bar{v}_i^2 - \pi kT/2m}, \quad (10)$$

where $\pi kT/2m$ is the square of the flux-weighted average velocity of the scattered molecules if the molecules scatter from the surface diffusely with complete thermal accommodation. From a knowledge of the reduced force coefficients and the velocity distribution function of the molecular beam, the coefficient α'_{KE} can be determined. With the approximation that α'_{KE} and α_{KE} are equal, $\bar{v}_s^2$ can be calculated from

$$\bar{v}_s^2 = \bar{v}_i^2 - \alpha'_{KE} (\bar{v}_i^2 - 4kT/m), \quad (11)$$

The uncertainty associated with this approximation has been shown⁸ to be less than 1% for many gas-surface interactions.

IV. EXPERIMENTAL RESULTS

Force measurements were made for each gas with the nozzle temperature at 300 K and 775 K. With the nozzle at 300 K the flux-weighted average velocities of the molecular beams of H₂, N₂, CO, and CO₂ were 2590 m/s, 1870 m/s, 1870 m/s, and 1670 m/s, respectively. With the nozzle at 775 K the flux-weighted average velocities of the molecular beams of H₂, N₂, CO, and CO₂ were 4610 m/s, 3180 m/s, 3190 m/s, and 2840 m/s, respectively. A linear interpolation method was used to calculate the reduced force coefficients for each gas incident upon each surface with a flux-weighted average velocity of 3000 m/s using the measurements at the two velocities. The details of this interpolation method are given elsewhere⁸.

The flux-weighted average velocity of the scattered molecules is shown in Fig. 3 and the scattering angle is shown in Fig. 4. Excluding the Z-93 surface, it can be seen that $\bar{v}_s$ and θ_s become larger as the angle of incidence increases for N₂, CO, and CO₂. This behavior is consistent with the scattering becoming more specular as the angle of incidence increases. The N₂ and CO data are almost identical for all four surfaces. This result is not surprising since the two gases have almost identical masses, they impinge upon the surfaces with nearly the same momentum, and their

potential energy well depths and the anisotropies of the gas-surface interactions are similar¹⁴. Scanning-electron-microscope images of the four surfaces revealed that Z-93 was rough on a 100 μm scale, while the other three surfaces were smooth on a 1 μm scale. These observations explain why scattering from Z-93 was different from the other three surfaces, and why the scattering from Z-93 was diffuse like with near complete thermal accommodation.

From Fig. 4 it can be seen that the scattering angles for N_2 and CO are nearly identical, and for large angles of incidence, θ_s is smallest for H_2 and largest for CO_2 . A qualitative argument for this effect can be given by noting that H_2 has the smallest mass of the four gases, and that CO_2 has the largest. Therefore, a given force perpendicular to the direction of propagation would deflect a beam of H_2 molecules more than a beam of CO_2 molecules. Thus, for large grazing angles of incidence, where the scattering is more specular, it is reasonable that θ_s is smallest for H_2 and largest for CO_2 . For small angles of incidence, where the scattering is more diffuse, this effect is partially obscured.

For all four surfaces, the flux-weighted average velocity of the scattered H_2 molecules increases as the angle of incidence increases, and then begins to decrease for very large angles of incidence. This result seems to imply that for very large angles of incidence, accommodation increases as the angle of incidence increases. However, excluding Z-93, Fig. 4 clearly indicates that for very large angles of incidence, the scattering angle increases as the angle of incidence increases, and for the solar array material this increase is dramatic. This result implies that the scattering becomes more specular as the angle of incidence increases. To resolve this apparent contradiction it should be noted that the incident gas molecules interact with the potential energy surface over a longer period of time for very large angles of incidence than for smaller angles of incidence. The longer interaction time would result in the force acting on the molecules over a longer period of time, and therefore, could result in more momentum loss. As the molecules leave the surface, the increased loss in momentum would allow the attractive van der Waals forces to act on the molecules over a longer period of time. Since this interaction will tend to be along the surface normal, the receding molecules will lose more of their normal momentum component while retaining their tangential component. As a result, the molecules would scatter closer to the specular direction for very large angles of incidence. The fact that this effect is not observed for the other three gases may be due to the much larger incident momenta of these gases.

Figure 3 shows that $\bar{v}_s$ is largest for H_2 and smallest for CO_2 . From this observation alone it cannot be concluded that H_2 scatters with the least accommodation and that CO_2 scatters with the most. If all gases were incident upon the surface with the same flux-weighted average velocity and were equally accommodated, $\bar{v}_s$ would be largest for H_2 and smallest for CO_2 because H_2 has the smallest mass and CO_2 has the largest. Fig. 5 shows the energy accommodation coefficient for all four gases incident upon all four surfaces. Because the potential energy well depth and anisotropy of the gas-surface interaction¹⁴ is largest for CO_2 and smallest for H_2 , it might be expected that H_2 would scatter with the least momentum accommodation while CO_2 would scatter with the most. However, excluding Z-93, H_2 scatters with the largest accommodation for very large angles of incidence on all four surfaces, and with the largest accommodation for angles of incidence near the surface normal on Kapton and SiO_2 coated Kapton. To explain these results, note that the magnitude of the flux-weighted average velocity of molecules that scatter diffusely with complete thermal accommodation, given by the term $\pi kT/2m$ in Eq. 10, is nearly 50% of the incident velocity for H_2 , about 12% of the incident velocity for N_2 and for CO , and only about 10% of the incident velocity for CO_2 . Thus, the denominator in Eq. 10 is much smaller for H_2 than for the other three gases, implying that α'_{KE} can be largest for H_2 , even if $\bar{v}_s$ is largest for H_2 . Fig. 3 also shows that for all four surfaces, α'_{KE} for N_2 and CO are nearly identical, and they are smaller than the scalar momentum accommodation coefficient for CO_2 . These results can be explained by noting that the potential energy well depths and the anisotropies of the gas-surface interactions are largest for CO_2 , and nearly the same for N_2 and CO ¹⁴, and that the denominator in Eq. 31 is similar for N_2 , CO , and CO_2 .

It has been shown that measurements of both the incident molecular beam velocity and the magnitude and direction of the forces exerted on a surface by incident molecules can be used to obtain reliable and accurate information concerning the scattered molecules flux-weighted average velocity and translational energy. 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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FIG. 1. Schematic diagram of the torsion balance used to measure the tangential F_t and normal F_n force components. The force components can be measured in 5° increments from angles of incidence of 0° to 85° .

FIG. 2. Schematic diagram of the torsion balance configurations used to measure the force components perpendicular and parallel to the molecular beam. Configuration (a) is used to measure the force component parallel to the beam, while configuration (b) is used to measure the component perpendicular to the beam. The view is looking down from above the torsion balance.

FIG. 3. The flux-weighted average velocity of the scattered molecules as a function of the angle of incidence. The results in this figure were obtained for each gas by interpolating between experimentally determined reduced force coefficients to estimate the coefficients for an incident flux-weighted average velocity of 3000 m/s.

FIG. 4. The scattering angle θ_s as a function of the angle of incidence. The results in this figure were obtained for each gas by interpolating between experimentally determined reduced force coefficients to estimate the coefficients for an incident flux-weighted average velocity of 3000 m/s.

FIG. 5. The energy accommodation coefficient α'_{KE} as a function of the angle of incidence. The results in this figure were obtained for each gas by interpolating between experimentally determined reduced force coefficients to estimate the coefficients for an incident flux-weighted average velocity of 3000 m/s.

Fig. 1, S.C.

Fig. 2, S.C.

Fig. 3, S.C.

Fig. 4, S.C.

Fig. 5, S.C.