JOURNAL OF SPACECRAFT AND ROCKETS
Vol. 34, No. 3, May–June 1997

Nocilla Model Parameters Obtained from Forces Exerted on Surfaces by Molecular Beams

Steven R. Cook* and Mark A. Hoffbauer†
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

The Nocilla gas–surface interaction model was used to approximate the velocity and angular distribution functions of H_2 and N_2 molecules scattered from surfaces of SiO_2-coated Kapton® and Z-93-coated Al. A new formalism was developed to analyze the momentum transferred to surfaces by incident gases that is founded upon two parameters called the reduced force coefficients. The reduced force coefficients were experimentally determined from measurements of the forces exerted on the surfaces by molecular beams of the gases and used to calculate the Nocilla model parameters.

Nomenclature

A = cross-sectional area of the molecular beam
$d\Omega$ = differential solid angle
F = magnitude of the net force exerted on the scattering surface by the molecular beam
f_n = normal reduced force coefficient
f_t = tangential reduced force coefficient
I = angular distribution function of the scattered molecules
k = Boltzmann's constant
m = mass of individual molecules in the molecular beam
N = velocity distribution function of the scattered molecules
n = number density of the scattered molecules
$\bar{p}$ = magnitude of the flux-weighted average momentum
S = molecular speed ratio of the scattered molecules
T = temperature
$\boldsymbol{U}$ = most probable velocity of the scattered molecules
$\bar{v}$ = magnitude of the flux-weighted average velocity
$\overline{v^2}$ = flux-weighted average of the velocity squared
$\boldsymbol{v}$ = velocity
Δ = quantity defined by Eq. (27)
ϵ = energy accommodation coefficient defined using the incident and scattered flux-weighted averages of the velocity squared
ϵ' = energy accommodation coefficient defined using the squares of the incident and scattered flux-weighted average velocities
Θ = quantity defined by Eq. (29)
θ = spherical coordinate polar angle
θ_i = angle between the surface normal and the flux-weighted average velocity of the incident molecules
θ_s = angle between the surface normal and the flux-weighted average velocity of the scattered molecules
θ_u = angle between the surface normal and the most probable velocity of the scattered molecules
Λ = defined by Eq. (21)
μ = scalar momentum accommodation coefficient
ρ = quantity defined by Eq. (30)
Σ = quantity defined by Eq. (22)
Σ' = quantity defined by Eq. (26)
Φ = absolute flux density
ϕ = spherical coordinate azimuthal angle

Subscripts

d = diffuse scattering with complete thermal accommodation
i = incident molecules
n = normal component
s = scattered molecules
t = tangential component
w = scattering surface

Received July 2, 1996; revision received Feb. 28, 1997; accepted for publication March 3, 1997.

*Postdoctoral Fellow, Chemical Science and Technology Division-1, MS J565. Member AIAA.
†Technical Staff Member, Chemical Science and Technology Division-1, MS J565.

Introduction

WHEN a gas scatters from a surface, the incident molecules experience forces that depend on the microscopic details of the gas–surface interaction potential. Since these forces change the average momentum of the incident molecules, a nonzero force must be exerted on the surface by the gas. In general, the collisions are inelastic and result in a transfer of energy between the incident gas and the surface that usually leads to a change in surface temperature. These interactions can be further complicated by changes in energy associated with the internal degrees of freedom of the gas and by adsorption or reactions of the gas with the surface.

In rarefied flow regimes, where the density is low enough that the gas does not behave as a continuum, forces exerted on and heat transferred to surfaces by incident gases can have a number of important consequences in several disciplines. Accurate models of forces exerted on and heat transferred to surfaces by molecules in the upper atmosphere of a planet during re-entry could be used to simplify spacecraft design.[1–3] These models could also be used to predict the orbital decay of satellites.[4,5] To achieve sufficient accuracy to be used as a pressure calibration standard, rotating-disk pressure gauges require precise knowledge of the tangential component of the force exerted on the rotating disk by different gases as a function of pressure.[6–8] In magnetic disk storage devices, the gap between the disk and the head is now approaching the mean free path for air at atmospheric pressure.[9] The air in this gap is responsible for the force that maintains the position of the head while the disk rotates. The gas–surface interactions between the air and the materials used to construct the disk and the head are poorly understood under realistic conditions, making accurate models of the forces exerted on the disk and the head by the air difficult to construct.

Currently, in rarefied flow regimes, poor understanding of gas–surface interactions makes it difficult to accurately model forces exerted on surfaces by incident gases. Modeling is difficult because the velocity distribution functions of gas molecules scattered from surfaces are generally not known.[10] In fact, theoretical models used to predict the velocity distribution functions cannot reproduce the molecular-beam data that currently exist. In the absence of a comprehensive theoretical model, it is commonly assumed that some fraction of the incident molecules scatters diffusely from the surface with complete thermal accommodation while the remaining fraction scatters specularly. However, for most real surfaces and in most applications, the behavior of the scattered molecules cannot be accurately described using this assumption.[10] This problem can be overcome by determining the adjustable parameters in an empirical model from measured quantities for each particular gas–surface interaction of interest.

Nocilla proposed an empirical model that assumes the velocity distribution function of the scattered molecules can be represented by a drifting Maxwellian.[11] It has been shown that the adjustable parameters in the Nocilla gas–surface interaction model can be determined from measurements of the energy and momentum accommodation coefficients.[1,2,12,13] Complete reviews of accommodation coefficient measurements and a summary of work related to the material presented in this article can be found elsewhere.[5,14–20] However, as defined, the momentum accommodation coefficients have singularities[21] that render the equations essentially unusable for many applications.

To alleviate the singularity problems associated with the momentum accommodation coefficients, a new formalism was developed to analyze the momentum transferred to a surface by an incident gas. The foundation of this formalism is two parameters called the reduced force coefficients. The formalism can be used to determine the flux-weighted average[22] velocity and translational energy of the scattered molecules as a function of the velocity of the incident molecules. These macroscopic average quantities for the scattered molecules can be used to determine the Nocilla model parameters.

The experiments described in this article were motivated by the need to accurately model the forces exerted on the large solar panel arrays to be used on the International Space Station by the plume gases emitted from the reaction control jets on the Space Shuttle. These gas–surface interactions were simulated by directing molecular beams of the plume gas species onto the surfaces of interest at varying angles of incidence. A supersonic nozzle source and the seeded beam technique were used to accelerate the molecules in the beams to velocities similar to those of the gases in the reaction control jet plumes.[22] The forces exerted on the surfaces by the molecular beams were measured using a specialized torsion balance. The force measurements were used to determine the reduced force coefficients for the gas–surface interactions, the flux-weighted average (hereafter referred to as average) velocities and translational energies of the scattered molecules, and the Nocilla model parameters.

Reduced Force Coefficients

The definitions of the reduced force coefficients are motivated from a consideration of the force exerted on a surface by a molecular beam and are given by

$$f_t = \frac{\bar{p}_{it} - \bar{p}_{st}}{\sqrt{\bar{p}_{it}^2 + \bar{p}_{in}^2}} \tag{1}$$

$$f_n = \frac{\bar{p}_{in} + \bar{p}_{sn}}{\sqrt{\bar{p}_{it}^2 + \bar{p}_{in}^2}} \tag{2}$$

The coefficients can also be written in terms of the components of the net force exerted on the surface by the incident molecules as

$$f_t = \frac{F_t}{A\Phi_i m \bar{v}_i} \tag{3}$$

$$f_n = \frac{F_n}{A\Phi_i m \bar{v}_i} \tag{4}$$

In the limiting case of specular reflection, the reduced force coefficients are

$$f_t = 0 \tag{5}$$

$$f_n = 2\cos\theta_i \tag{6}$$

whereas for diffuse scattering with complete thermal accommodation, the coefficients are

$$f_t = \sin\theta_i \tag{7}$$

$$f_n = \cos\theta_i + \sqrt{\frac{\pi k T_w}{2m\bar{v}_i^2}} \tag{8}$$

Using Eqs. (1) and (2), the magnitude and direction of the average velocity of the scattered molecules can be written in terms of the reduced force coefficients as

$$\bar{v}_s = \bar{v}_i\sqrt{(f_t - \sin\theta_i)^2 + (f_n - \cos\theta_i)^2} \tag{9}$$

$$\theta_s = \tan^{-1}\left(\frac{\sin\theta_i - f_t}{f_n - \cos\theta_i}\right) \tag{10}$$

Spherical polar coordinates are used to define the directions of the average velocities of the incident and scattered molecules. The polar angle is measured from the surface normal, the azimuthal angle is 180 deg for the average velocity of the incident molecules and is assumed to be 0 deg for the average velocity of the scattered molecules, and the scattering surface is in the plane defined by the polar angle of 90 deg. This coordinate system is used because the velocity and angular distribution functions of the scattered molecules are three-dimensional.

Momentum accommodation is difficult to determine using Eqs. (5–8). Therefore, a new parameter called the scalar momentum accommodation coefficient is introduced that eliminates the singularity problems[21] associated with the classical momentum accommodation coefficients and is given by

$$\mu = \frac{\bar{p}_i - \bar{p}_s}{\bar{p}_i - \bar{p}_d} = \frac{\bar{v}_i - \bar{v}_s}{\bar{v}_i - \sqrt{\pi k T_w/2m}} \tag{11}$$

where the term $\sqrt{(\pi k T_w/2m)}$ represents the magnitude of the average velocity of the scattered molecules assuming diffuse scattering from the surface with complete thermal accommodation. Note that μ ranges between zero for specular reflection and one for diffuse scattering with complete thermal accommodation.

It will be shown that the Nocilla model parameters for a particular gas–surface interaction can be determined if $\underline{\Phi_s}$, $\bar{v}_{st}$, $\bar{v}_{sn}$, and $\overline{v_s^2}$ are known. However, an exact determination of $\overline{v_s^2}$ cannot be made from a knowledge of only the reduced force coefficients and the absolute flux-density and velocity distribution function of the incident molecules. With the simple assumption of equating $\overline{v_s^2}$ with $\bar{v}_s^2$, errors as large as 60% can result for diffuse-like scattering. A more accurate approximation can be obtained for $\overline{v_s^2}$ with the introduction of two types of energy accommodation coefficients. Neglecting the average energy associated with the internal degrees of freedom of the gas molecules, a kinetic energy accommodation coefficient can be defined based upon the average translational energies as

$$\epsilon = \frac{\frac{1}{2}m\overline{v_i^2} - \frac{1}{2}m\overline{v_s^2}}{\frac{1}{2}m\overline{v_i^2} - \frac{1}{2}m\overline{v_d^2}} = \frac{\overline{v_i^2} - \overline{v_s^2}}{\overline{v_i^2} - 4kT_w/m} \tag{12}$$

where $4kT_w/m$ is the average of the velocity squared of the scattered molecules assuming diffuse scattering from the surface with complete thermal accommodation. A second coefficient, based on the squares of the average velocities of the incident and scattered molecules, is defined by

$$\epsilon' = \frac{\frac{1}{2}m\bar{v}_i^2 - \frac{1}{2}m\bar{v}_s^2}{\frac{1}{2}m\bar{v}_i^2 - \frac{1}{2}m\bar{v}_d^2} = \frac{\bar{v}_i^2 - \bar{v}_s^2}{\bar{v}_i^2 - \pi k T_w/2m} \tag{13}$$

where the term $\pi k T_w/2m$ is the square of the average velocity of the scattered molecules assuming diffuse scattering from the surface with complete thermal accommodation. Note that both ϵ and ϵ' range between zero for specular reflection and one for diffuse scattering with complete thermal accommodation. It has been shown that the error introduced by equating ϵ with ϵ' is less than $\pm 1\%$ for molecules incident upon surfaces with energies that are large compared to kT (Ref. 23). With the justifiable assumption that ϵ and ϵ' are equal,

$$\overline{v_s^2} = \overline{v_i^2} - \epsilon'\left(\overline{v_i^2} - 4kT_w/m\right) \tag{14}$$

The reduced force coefficients for a particular gas–surface interaction can be determined using Eqs. (3) and (4) from measurements of the components of the force exerted on the surface by a molecular beam of the gas, and the absolute flux-density and velocity distribution function of the molecular beam. The velocity distribution function of the molecular beam is used to determine $\bar{v}_i$ and $\overline{v_i^2}$. Once the reduced force coefficients are known, Eqs. (9) and (10) can be used to determine $\bar{v}_s$ and θ_s. Equation (13) can then be used to determine ϵ', and finally, Eq. (14) can be used to obtain $\overline{v_s^2}$.

Nocilla Gas–Surface Interaction Model

The Nocilla gas–surface interaction model assumes that the velocity distribution function of the scattered molecules can be approximated using a drifting Maxwellian distribution function with most probable velocity $\boldsymbol{U}$ defined as[11]

$$N(\boldsymbol{v}) = n\left(\frac{m}{2\pi k T_s}\right)^{\frac{3}{2}} \exp\left[-\frac{m(\boldsymbol{v}-\boldsymbol{U})^2}{2kT_s}\right] \tag{15}$$

The adjustable parameters in the Nocilla model are n, T_s, and $\boldsymbol{U}$. The most probable velocity of the scattered molecules is assumed to lie in the plane formed by the surface normal and the average velocity of the incident molecules. Equations (16–19), obtained by integrating Eq. (15), can be used to determine the four Nocilla model parameters:

$$\Phi_s = (nU/2\pi S)\left(e^{-\Lambda^2} + \sqrt{\pi}\Lambda\Sigma\right) \tag{16}$$

$$\bar{v}_s \sin\theta_s = U \sin\theta_u \tag{17}$$

$$\bar{v}_s \cos\theta_s = \frac{U}{S}\frac{\Lambda e^{-\Lambda^2} + \sqrt{\pi}\left(\frac{1}{2}+\Lambda^2\right)\Sigma}{e^{-\Lambda^2}+\sqrt{\pi}\Lambda\Sigma} \tag{18}$$

$$\overline{v_s^2} = \frac{U^2}{S^2}\frac{(2+S^2)e^{-\Lambda^2} + \sqrt{\pi}\left(\frac{5}{2}+S^2\right)\Lambda\Sigma}{e^{-\Lambda^2}+\sqrt{\pi}\Lambda\Sigma} \tag{19}$$

where

$$S = \frac{U}{\sqrt{2kT_s/m}} \tag{20}$$

$$\Lambda = S\cos\theta_u \tag{21}$$

and

$$\Sigma = 1 + \operatorname{erf}\Lambda \tag{22}$$

Using Eqs. (17–19), it can be shown that

$$\tan\theta_s = \frac{S\sin\theta_u\left(e^{-\Lambda^2}+\sqrt{\pi}\Lambda\Sigma\right)}{\Lambda e^{-\Lambda^2}+\sqrt{\pi}\left(\frac{1}{2}+\Lambda^2\right)\Sigma} \tag{23}$$

$$\frac{(\bar{v}_s\sin\theta_s)^2}{\overline{v_s^2}} = \frac{(S\sin\theta_u)^2\left(e^{-\Lambda^2}+\sqrt{\pi}\Lambda\Sigma\right)}{(2+S^2)e^{-\Lambda^2}+\sqrt{\pi}\left(\frac{5}{2}+S^2\right)\Lambda\Sigma} \tag{24}$$

To determine S and θ_u, Eqs. (23) and (24) can be evaluated iteratively. Then Eq. (17) is used to determine U, and Eq. (20) is used to obtain T_s.

When the scattering angle θ_s is equal to zero, Eq. (17) implies θ_u must equal zero or 180 deg. For this special case, S can be determined from an equation obtained by combining Eqs. (23) and (24), given by

$$\frac{\overline{v_s^2}}{\bar{v}_s^2} = \frac{\Delta\left(e^{-S^2}\pm\sqrt{\pi}S\Sigma'\right)}{\left[\sqrt{\pi}\left(\frac{1}{2}+S^2\right)\Sigma' \pm Se^{-S^2}\right]^2} \tag{25}$$

where

$$\Sigma' = 1 \pm \operatorname{erf} S \tag{26}$$

$$\Delta = (2+S^2)e^{-S^2} \pm \sqrt{\pi}\left(\frac{5}{2}+S^2\right)S\Sigma' \tag{27}$$

The plus sign is used if θ_u equals zero, the minus sign if θ_u equals 180 deg. Once S is known, U can be evaluated iteratively using Eq. (18). For all cases, after θ_u, U, and S have been determined, n can be obtained from Eq. (16) by equating the incident and exit flux densities.

The angular distribution function I is defined by letting $I(\theta,\phi)\,d\Omega$ represent the fraction of Φ_s that scatters into the differential solid angle $d\Omega$ about the direction defined by θ and ϕ. Using the Nocilla model to approximate the angular distribution function of the scattered molecules gives

$$I(\theta,\phi) = \frac{\Theta}{\pi}\cos\theta\,\frac{e^{-S^2}}{e^{-\Lambda^2}+\sqrt{\pi}\Lambda\Sigma} \tag{28}$$

where

$$\Theta = 1+\rho^2+\sqrt{\pi}\rho\left(\tfrac{3}{2}+\rho^2\right)(1+\operatorname{erf}\rho)e^{\rho^2} \tag{29}$$

and

$$\rho = S(\sin\theta\cos\phi\sin\theta_u + \cos\theta\cos\theta_u) \tag{30}$$

The angular distribution function can also be thought of as representing the probability that a molecule with incident velocity $\boldsymbol{v}_i$ scatters into the solid angle $d\Omega$ about the direction defined by θ and ϕ. Once the Nocilla model parameters have been determined, Eq. (28) can be used to approximate the angular distribution function of the scattered molecules.

Experimental Results and Discussion

The Nocilla model parameters were determined from measurements of the forces exerted on surfaces of SiO_2-coated Kapton®, Kapton, Z-93-coated Al, and the solar array material to be used on the International Space Station by molecular beams of H_2, N_2, CO, and CO_2. The torsion balance apparatus used to measure the forces, along with the techniques used to measure the flux densities and velocity distribution functions of the molecular beams, are described elsewhere.[22,23] The average velocities of the molecular beams ranged from 1670 m/s for CO_2 to 4620 m/s for H_2. The total uncertainty in the reduced force coefficient measurements was estimated to be less than $\pm 10\%$.

Measurements of the reduced force coefficients for N_2, CO, and CO_2 incident upon SiO_2-coated Kapton, Kapton, and the solar array material showed that these gas–surface interactions have many similarities. Therefore, of these gas–surface interactions only the data for N_2 incident upon SiO_2-coated Kapton will be discussed in this article. A complete discussion of all the measurements is given elsewhere.[23] Since differences were observed for H_2 scattering from these surfaces, the data for H_2 incident upon SiO_2-coated Kapton will also be discussed.

Scanning electron microscope measurements revealed that SiO_2-coated Kapton, Kapton, and the solar array material had similar surface roughnesses and were smooth on a scale of less than 1 μm, while the Z-93-coated Al surface was rough on a 100-μm scale. This contrast in surface roughness could account for the significant differences between the way the four gases scattered from Z-93-coated Al and from the other three surfaces.

Figure 1 shows $\bar{v}_s/\bar{v}_i$ for H_2 and N_2 measured as a function of the angle of incidence θ_i for SiO_2-coated Kapton and Z-93-coated Al at the indicated average incident velocities. Figure 2 shows the scattering angle θ_s, and Fig. 3 shows the scalar momentum accommodation coefficient μ for the same gas–surface interactions. N_2 scattering from Z-93-coated Al can be seen to be nearly diffuse, with almost complete thermal accommodation, by noting that θ_s is close to zero and μ is close to one at all angles of incidence. H_2 scattering from Z-93-coated Al can be seen to be nearly diffuse by noting that θ_s is close to zero for all angles of incidence. However, μ varies widely over the range of angles of incidence.

For H_2 and N_2 incident upon SiO_2-coated Kapton, $\bar{v}_s$ and θ_s increase as the angle of incidence increases, consistent with the scattering becoming more specular. For 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 that a given force perpendicular to the direction of motion would deflect H_2 more than N_2. Thus, for large angles of incidence where the scattering becomes more specular, it is reasonable to assume that the large repulsive force exerted on the molecules by the surface will deflect H_2 more than N_2.

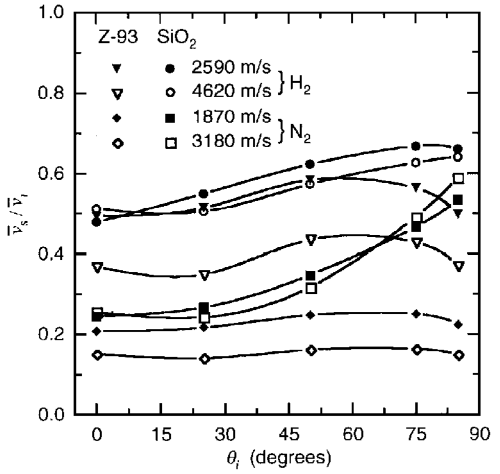

Fig. 1 Ratio $\bar{v}_s/\bar{v}_i$ for H_2 and N_2 incident upon SiO_2-coated Kapton and Z-93-coated Al at the indicated average velocities.

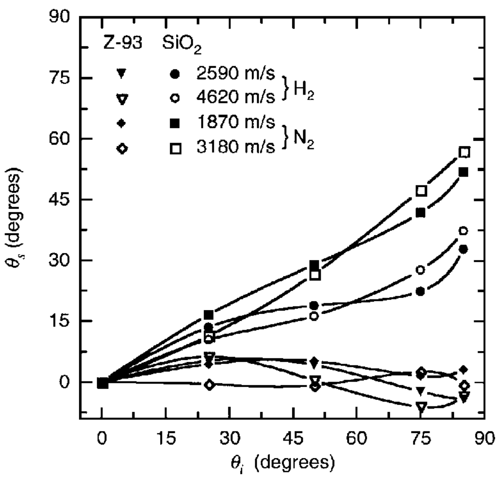

Fig. 2 Scattering angle θ_s for H_2 and N_2 incident upon SiO_2-coated Kapton and Z-93-coated Al at the indicated average velocities.

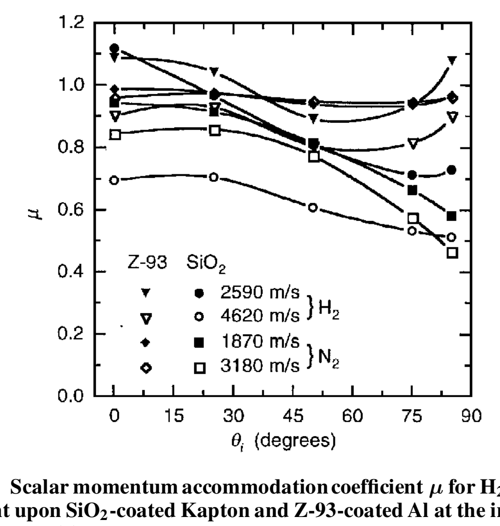

Fig. 3 Scalar momentum accommodation coefficient μ for H_2 and N_2 incident upon SiO_2-coated Kapton and Z-93-coated Al at the indicated average velocities.

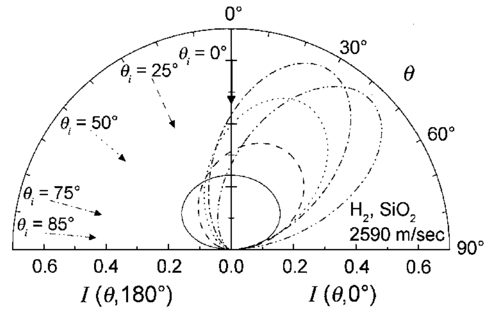

Fig. 4 Angular distribution functions for H_2 incident upon SiO_2-coated Kapton with an average velocity of 2590 m/s.

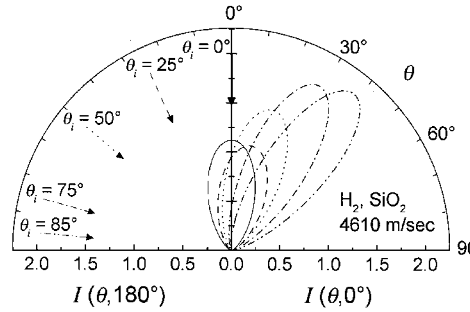

Fig. 5 Angular distribution functions for H_2 incident upon SiO_2-coated Kapton with an average velocity of 4620 m/s.

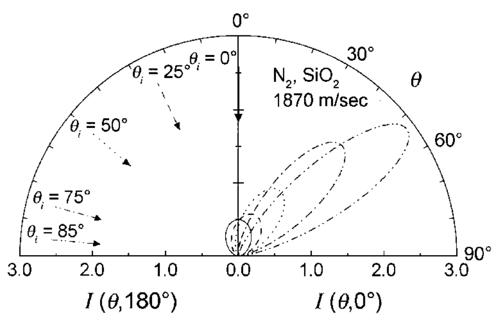

Fig. 6 Angular distribution functions for N_2 incident upon SiO_2-coated Kapton with an average velocity of 1870 m/s.

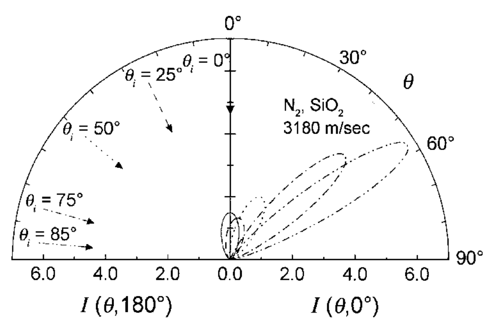

Fig. 7 Angular distribution functions for N_2 incident upon SiO_2-coated Kapton with an average velocity of 3180 m/s.

In Figs. 4–8, polar plots of the angular distribution functions of the scattered molecules are shown for the indicated gas–surface interactions. The Nocilla model parameters were obtained for the gas–surface interactions using the previously outlined procedure. Equation (28) was then used to calculate the angular distribution functions. In these figures, the origin can be thought of as the impingement point of the molecular beam, the vertical axis is perpendicular to the scattering surface, and the horizontal axis is defined by the projection of U onto the plane of the scattering surface. Figures 4–8 show the intersection of the plane formed by the average velocity of the incident molecules and the surface normal with the three-dimensional angular distribution functions. The distance from the origin to a point on a lobe represents the probability that a molecule with the indicated average velocity scatters from the surface into a differential solid angle $d\Omega$ about the direction defined by θ and ϕ. These distances are defined by the tick marks on the horizontal and vertical axes. The portion of the horizontal axis to the right of the vertical axis corresponds to $\phi = 0$, and the portion

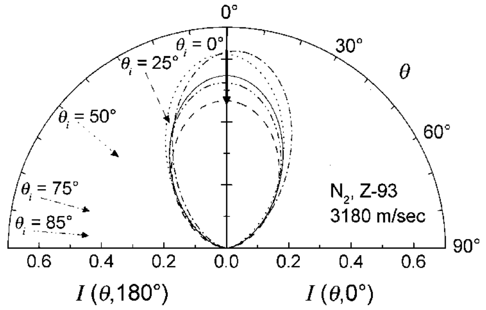

Fig. 8 Angular distribution functions for N_2 incident upon Z-93-coated Al with an average velocity of 3180 m/s.

to the left of the vertical axis corresponds to $\phi = 180$ deg. The polar angle is given along the circular axis of the graphs.

The general trend that the scattering becomes more specular with increasing angles of incidence for gases scattering from SiO_2-coated Kapton can be inferred from Figs. 4–8. As the angle of incidence increases, the lobes become much larger because of the way in which the angular distribution function is normalized according to the equation

$$\int I(\theta, \phi)\, d\Omega = 1 \tag{31}$$

This behavior has fundamental physical significance. As the angle of incidence increases, the size of the lobes also increases, meaning fewer particles scatter out of the plane formed by the average velocity of the incident molecules and the surface normal. This effect is more pronounced for N_2 than for H_2 and for the larger incident velocities.

Figure 8 shows that scattering from Z-93-coated Al is nearly diffuse for all angles of incidence. This result was observed for all gases incident upon Z-93-coated Al and at all incident velocities. Nearly diffuse scattering is most likely due to the extremely rough surface morphology of Z-93-coated Al.

Conclusions

A new formalism has been used to analyze the average momentum transferred to surfaces by incident gases. This new formalism eliminates the singularity problems associated with the momentum accommodation coefficients and can be used to uniquely determine the Nocilla gas–surface interaction model parameters. The formalism eliminates the need to measure energy accommodation coefficients, thereby reducing the number of experimentally determined parameters required to obtain the Nocilla model parameters.

The results discussed in this article show that molecules can scatter from technical surfaces more specularly as the angle of incidence increases and that the scattering has a strong dependence on both the incident gas and velocity. These results show that for some technical surfaces the simple assumption of diffuse scattering with complete thermal accommodation is entirely inadequate.

Acknowledgments

This work was supported by NASA Johnson Space Center (JSC). The authors also acknowledge the technical assistance of Frank Archuleta, David Clark, and Jeff Patterson. Finally, we thank Carl Scott and Michael Jensen at NASA JSC for their continued support of this project.

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I. D. Boyd
Associate Editor