Gas flow near a smooth plate

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We examine gas flow adjacent to a molecularly smooth, solid, muscovite mica. The fluctuations in force acting on a glass sphere as a function of proximity to a mica plate were measured in air and were used to obtain the damping. The damping was interpreted as a lubrication force. The measured damping as a function of separation in the slip-flow regime corresponds to a slip length of $480 \pm 70$ nm, which is equivalent to highly specular gas molecule collisions. A slip-flow model fits the data for separations as small as one mean free path.

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I. INTRODUCTION

Recent research on gas flow in confined spaces confirms that boundary slip occurs at the solid-gas interface and that the magnitude of slip depends on the nature of the interface [1–6]. There are reports of particularly large slip lengths adjacent to carbon nanotubes [5,6]. For example, slip lengths up to 70 $\mu$m have been reported for water in a nanotube [5]. The primary objective of this work is to improve the understanding of gas flow in confined spaces adjacent to a molecularly smooth planar wall. Specifically, we examine gas molecule interactions with muscovite mica by measuring the lubrication force on a small, spherical particle approaching a flat plate. Secondary goals include testing the Vinogradova solution to the Navier-Stokes equations for squeeze flow near dissimilar surfaces [7] and developing a basis for more informed design of hard drive air bearings, semipermeable membranes, and microfluidics or microelectrical mechanical systems (MEMS) [8–10] where confined gas flows occur. Comparison to nanotube research can shed light on the importance of the degree of confinement on gas flows. In the work presented here, the gas is confined between two essentially flat parallel plates, whereas in the nanotube experiments, the gas is confined within a cylinder.

The flow of gases is usually classified into different regimes using the Knudsen number, which is the ratio between the mean free path, $\lambda$, and the characteristic length of the system, $L$. For the slip-flow regime, $1000\lambda > L > 10\lambda$, the slip length $b$ is defined by the equation [11]

$$v_y(0) = b \frac{\partial v_y}{\partial z}.$$  (1)

In Eq. (1), $v_y$ is the fluid velocity tangential to the interface, $z$ is a direction normal to an interface, and $v_y(0)$ is the fluid velocity at the surface, i.e., at $z=0$ (see Fig. 1). The slip length is sometimes used as a fitting parameter for experiments not in the slip-flow regime, e.g., for $L \sim \lambda/100$ in nanotube experiments [5,6] and even in liquids, $L > 1000\lambda$. [11–14]. Variation in slip length arises from the fact that, during a collision with a solid surface, a gas molecule will transfer some of its tangential momentum to the solid.

The gas-solid interaction is characterized by the tangential momentum accommodation coefficient, $\sigma$, where $\sigma = 0$ represents specular reflection (gas molecule tangential momentum not conserved, minimum slip length) and $\sigma = 1$ represents diffuse reflection (gas molecule tangential momentum transformed into damping). The damping was interpreted as a lubrication force. The measured damping as a function of separation in the slip-flow regime (i.e., where the concept of slip is defined) and at smaller length scales approaching the free molecular regime.

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II. THEORY

A. Damped oscillator theory

The method of Honig et al. [1] is described briefly here. When a cantilever-sphere probe is immersed in a gas, the cantilever undergoes one-dimensional deflection fluctuations (see Fig. 1) with an average energy, $\frac{1}{2}k_BT$, which are mediated by collisions with the gas. We use the following equation for this motion [1]:

$$m \frac{d^2h}{dt^2} + D(h) \frac{dh}{dt} + kd = F_{\text{noise}} + F_{\text{surf}}(h). \quad (3)$$

Here, $m$ is the cantilever effective mass, $t$ is time, $D$ is the damping coefficient, $k$ is the spring constant, $d$ is the deflection of the cantilever, and $h$ is defined in Fig. 1. $F_{\text{noise}}$ is the force associated with the gas-mediated fluctuations. The influence of the plate is captured in two terms: $F_{\text{surf}}(h)$ which is a quasistatic surface force between the sphere and plate and $D(h)$, which we consider to be a combination of far-field damping $D(\infty)$ and the damping due to the proximity of the plate $D_{\text{hub}}$. We assume that $m$, $D(h)$, $F_{\text{surf}}$, and $k$ are frequency independent. This is usually a good approximation when the damping and surface forces are small. Equation (3) can be rearranged to identify the energy spectrum density (ESD) near resonance as [1]

$$B \left[ 1 - \left( \frac{f}{f_0} \right)^2 \right]^2 + \left( \frac{2\pi f D_{\text{hub}}}{\kappa m \nu} \right)^2 + C, \quad (4)$$

where $f$ is the frequency, $f_0$ is the resonant frequency, and $k_{\text{eff}}$ is the effective spring constant under the influence of external forces. In Eq. (4), $B$, $C$, $f_0$, and $D(h)$ are adjustable parameters, which have distinct effects on the shape of the ESD. $D(h)$ controls the width of the spectrum. $D_{\text{hub}} = D(h) - D(\infty)$ is the contribution of the plate to the damping, which is then compared to a theoretical value to obtain $b$ or $\sigma$.

B. Calculation of slip length

Vinogradova derived an equation for the lubrication damping force acting on a sphere driven towards a plate [7], which, via the fluctuation dissipation theorem, is the same as the damping observed in the ESD of the thermal fluctuations:

$$D_{\text{hub}} = \frac{6\pi \eta R^2}{h} f^*. \quad (5)$$

The fluid dynamic viscosity is given as $\eta$ and $R$ is the radius of the sphere. The slip lengths on the two solids, $b_1$ and $b_2$, enter into Eq. (5) via $f^*$:

$$f^* = -\frac{2\alpha h}{\beta \gamma} - \frac{2h}{\gamma - \beta} \left( \frac{\beta + h}{\beta^2} \right) \ln \left( 1 + \frac{\beta}{h} \right) - \frac{(\gamma + h)(\gamma - \alpha)}{\gamma^2} \ln \left( 1 + \frac{\gamma}{h} \right), \quad (6)$$

$$\alpha = b_1 + b_2, \quad \beta = 2b_1(2 + q + \sqrt{1 + q + q^2}), \quad \gamma = 2b_1(2 + q - \sqrt{1 + q + q^2}), \quad q = \frac{b_2}{b_1} - 1. \quad (7)$$

For $b_1 = b_2 = 0$, $f^* = 1$, recovering the result of Brenner [16]. In this work, we consider an asymmetric interaction [between glass coated with trimethylchlorosilane (TMCS) and mica]. It is important to note that for mild asymmetry ($q$ small), $f^*$ is controlled by the sum of the slip lengths, $\alpha = b_1 + b_2$. This effect is shown in Fig. 2 for $b_1 + b_2 = 600$ nm: $D_{\text{hub}}$ is very similar for $b_1 = b_2 = 300$ nm and for $b_1 = 200$ nm, $b_2 = 400$ nm. When the lubrication force is sensitive only to the sum of the slip lengths, the effect of the probe cancels out for a series of measurements on different plates with the same probe.

III. EXPERIMENT

Mica (S&SJ, New York) for the plate was freshly cleaved immediately preceding an experiment. AFM images of these
surfaces over a 5 μm scan size indicate an rms roughness <0.1 nm. No steps were observed in a typical 5 μm scan. The fluctuation experiments used the D lever on an ORC8 chip (Veeco Metrology). Spring constants were determined by the Hutter method at h > 1 mm [17]. The three cantilevers employed here had k = 0.067, 0.070, and 0.062 N/m. A borosilicate glass probe (Duke Scientific) was attached to each cantilever with an epoxy (Epon Resin 1004F; Hexion Specialty Chemicals). Probe roughness was determined by inverse imaging with a TGT01 grating (NT-MDT). Probes with asperities greater than 10 nm above the apex of the sphere were rejected. The radii of the probes were measured using optical microscopy and were R = 10.3, 11.9, and 8.4 μm, respectively. Probes were coated with trimethylchlorosilane (99+% purity, Aldrich Chemical) to reduce the formation of water films on the surface, which make the system more difficult to model. The probes were irradiated for 90 min in a UV/Ozone ProCleaner (BioForce Nanosciences) and then placed in a vapor deposition chamber with 1–2 mL of TMCS overnight. The quality of the TMCS film was assessed by a contact angle study performed on a glass cover slip (FisherScientific) that underwent the same deposition procedure. Deionized water (Elix 3 followed by Synergy UV finishing; Millipore) was placed on the TMCS film in a FTA125 Contact Angle Analyzer (First Ten Angstroms). The contact angles were 95 ± 3° advancing, and 86 ± 3° receding. After removal, the probe again underwent inverse imaging and was rejected if it had been fouled.

Our AFM is an MFP-3D (Asylum Research) operating with closed-loop control on the piezo drive. The probe-plate contact area was exposed to atmospheric conditions (temperature: 21–24°C; humidity: 38–78 %). Experiments consisted of performing a 5–20 μm/s force curve to determine probe-plate separation. After returning (approximately) to its starting position, the probe dwelled for 10 s without controlled change of separation. Data were collected during this dwell at 50 kHz. A second force curve was then performed to identify any separation drift that occurred. This process was repeated in a series of increasing probe-plate separation steps to obtain the damping as a function of separation.

IV. RESULTS AND DISCUSSIONS

An example of the raw data is shown in Fig. 1 and the damping due to proximity to the plate, Dhab, is shown in Fig. 3. Dhab, as mentioned earlier, is obtained from the total damping, D(h) by subtracting the damping at infinite separation [Dhab = D(h) − D(∞)]. D(∞) can be fitted from Eq. (4), evaluated at infinite separation, but in practice was derived from the quality factor, Q, in the Asylum thermal tune software using [1]

$$D(\infty) = \frac{k_\infty}{2\pi f_\infty Q_\infty}. \quad (8)$$

When h becomes large, the D(∞) correction becomes a significant fraction of the Dhab(h) signal. Therefore, a substantial error in D(∞) results in a considerable error in Dhab. To minimize this effect we elect to only use data where $D(\infty) < 1/3D(h)$, which in practice corresponded to h < 100 nm.

Figure 3 shows that at large separation, 1/Dhab is approximately linear, as expected. We fit Equation (5) to the data in the range 500–1000 nm, i.e. in the slip-flow regime (h > 7λ). For this fit we use the slip length of TMCS-coated glass established in literature (b1 = 125 ± 50 nm) [1]. The best fit returns a slip length on the mica of b2 = 443 nm for the data shown in Fig. 3. The average over seven experiments gives b2 = 480 ± 50 nm for the mica plates. After including the error in b1, the slip length on mica is b2 = 480 ± 70 nm.

From Fig. 3 one can see that the fit between the experimental data for separations in the range 500–1000 nm (h > 7λ) and the asymmetric Vinogradova solution is also excellent for separations approaching the order of a mean free path of air (λ = 67 nm) [18]. In Fig. 4, the near contact region is examined for another plate-probe combination. For this 1/Dhab(h) curve the parameters were fit to the data in the range 500–1000 nm with a b2 = 517 nm. This fit is extrapolated to h = 0.1 nm. Excellent agreement is seen until h ~ 100 nm. Here, the gas is rarefied and continuum behavior (Navier-Stokes’ equation) is not assured to adequately describe the confined gas flow, but in fact the theory and experiment agree quite well.

There are significant experimental difficulties in the region h < 100 nm, which are not included in the error bars shown, and prevent us from making a conclusion about the accuracy of the theory for small separations. Three experimental issues are noted here. First, the probe becomes overdamped near probe-plate contact making it difficult to ascertain the location and shape of the resonance peak. Second, the broadening of the resonance increases the range of frequencies examined. If any of the coefficients in Eq. (3) are functions of frequency in this range, then the solution [Eq. (4)] also has diminished accuracy.
The principal result of this work is that the slip length of air on mica is $480 \pm 70$ nm. This corresponds to $\sigma = 0.25 \pm 0.03$, where at each point we fix $b_1$ and plot the minimum sum of squared residuals obtained when $b_2$ is allowed to vary. We see two distinct minima, at about 100 and 565 nm, showing that a single lubrication measurement on an asymmetric system can give slip lengths for the two surfaces simultaneously. These slip lengths are close to the values identified when we determined each individually: $b_1 = 125$ nm from an experiment where both surfaces had the same chemistry and $b_2 = 517$ nm using the asymmetric technique described in this work on this particular inverse damping curve.

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gas.) Similarly, the pressure drop in a capillary is affected by the boundary condition. For a gas in a cylinder (radius = 0.5 μm), solution of the Navier-Stokes equation [20] shows that the pressure drop is about three times smaller for a slip length of 500 nm as compared to that for a slip length of 100 nm.

The slip length scales with the mean free path which is, in turn, inversely proportional to \( P \) [15]:

\[
b \propto \lambda \propto \frac{1}{P}.
\] (9)

Thus, the effect of the slip length extends further from the surface at lower pressures and thereby influences wider channels. For example, at \( P = 0.01 \text{ atm} \), the slip length on mica would be 50 μm.

V. CONCLUSION

The slip length on muscovite mica is large: \( b = 480 \pm 70 \) nm, as expected for a very smooth and stiff material. This slip length is much greater than for a rough material, and thus the properties of a solid surface can have a significant effect on the gas pressure drop across channels that are smaller than 1 μm when the pressure is less than or equal to 1 atm. There is quantitative agreement between the measured damping-separation data and the Vinogradova slip-flow solution. This agreement extends to the region when the gas is confined to a film that has a thickness that is similar to the mean free path of the gas. This ability to extend continuum force and flow models to molecular dimensions is seen elsewhere [10,21] and is useful for modeling. The experiment is sufficiently sensitive to distinguish between damping from asymmetric pairs of surfaces. Thus an experimenter could determine whether the sample and probe have different surfaces, if this were not already known.

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