Modeling Bearing and Shear Forces in Molecularly Thin Lubricants

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Abstract Under the effects of high shear rate and confinement between solid surfaces, the behavior of a thin lubricant film deviates from that of the bulk, resulting in significant increases of lubricant viscosity and interfacial slip. A semi-empirical model accounting for the breakdown of continuum theory at the nanoscale is proposed—based on film morphology and chemistry from available experimental and molecular dynamics simulation data—to describe lubricant behavior under shear. Viscosity stiffening and interfacial slip models are introduced into the formulations of the normal (bearing) and shear forces acting on a sphere that moves within a thin lubricant film parallel to a rigid plane. The experimentally measured ‘apparent’ viscosity confounding the effects of both stiffening and slip is used to predict the hydrodynamic forces acting on a sphere that moves within a thin lubricant film parallel to a rigid plane. The proposed sphere-on-flat model forms the basis of a future, dynamic contact with friction model that will account for lubricant contact in the context of molecularly thin lubricated rough surface contact.

Keywords Nanotribology · Magnetic data storage · Sub-boundary lubrication · Non-Newtonian hydrodynamic effects · Viscoelasticity · Rheology

List of Symbols

\( A \) Oscillation amplitude

\( a, b \) Viscosity-gap model coefficients

\( a', b' \) Viscosity-shear rate model coefficients

\( C \) Molecular coverage of surface area

\( c \) Damping coefficient (for fluid ‘contact’)

\( d_0 \) Liquid gap

\( f \) Driving frequency of oscillation

\( f^\# \) Dimensionless slip factor

\( G' \) Storage modulus

\( G'' \) Loss modulus

\( G^* \) Complex modulus

\( H \) Heaviside function

\( h \) Film thickness

\( h_0 \) Average molecular height of bonded layer

\( h_0 \) Solid-solid gap (minimum film thickness)

\( L_s \) Slip length

\( p \) Pressure

\( P_{\text{fluid}} \) Normal (bearing) hydrodynamic force

\( P_{\text{solid}} \) Normal solid force

\( P_{\text{trans}} \) Normal transitional force

\( Q_{\text{fluid}} \) Shear hydrodynamic force

\( Q_{\text{solid}} \) Solid friction force

\( Q_{\text{trans}} \) Shear transitional force

\( R \) Sphere (probe or asperity) radius

\( S \) Spherical cap area

\( t \) Total lubricant layer thickness

\( U \) Shearing velocity

\( u \) Fluid velocity along \( x \)-direction

\( u^* \) Dimensionless form of \( u \)

\( u_s \) Slip velocity

\( u_s^* \) Dimensionless form of \( u_s \)

\( u_w \) Wall velocity (as boundary condition)

\( v \) Fluid velocity in \( y \)-direction

\( v^* \) Dimensionless form of \( v \)

\( \dot{c'} \) ‘Apparent’ shear rate

\( \dot{c} \) ‘True’ shear rate (accounting for slip)
1 Introduction

Molecularly thin lubricant layers are used in nanotechnology to lubricate and protect contacting surfaces. However, modeling their behavior is not trivial. Consider, for example, the head–disk interface in magnetic storage, where a slider flies at a prescribed clearance of a few nanometers above a rapidly rotating disk. To protect against accidental contact, the disk and slider are coated with 2-nm-thick diamond-like carbon (DLC) films, while the disk is also coated with a polymeric lubricant layer of atomic thickness that also protects against corrosion [1–3].

The presence of this lubricant layer alters the surface energy of the disk and, in turn, the adhesive forces acting on the slider, thereby affecting its dynamic response. Furthermore, as the flying clearance between the slider and disk is sought to be minimized to increase storage density, possible operation within the lubricant layer—denoted herein as lubricant contact—cannot be simplified by assuming that the lubricant is readily displaced upon contact, and that it offers no resistance to shearing and compression. While the effect of molecularly thin lubricant layers on adhesion has been studied previously, e.g., in [4], there is a lack of models predicting normal and shear forces during contact with such layers.

Polymeric lubricants confined in nanometer-scale gaps can have different rheological properties from the bulk [5, 6]. For example, when lubricant molecules are confined between two solid surfaces that slide past each other with high velocities, the lubricant viscosity starts to increase at gaps smaller than a few hundred nanometers; similarly, elasticity appears suddenly when the gap becomes smaller than a few nanometers and both viscosity and elasticity increase monotonically with decreasing gap. Then, the viscoelastic properties of the film start to differ from those of the bulk and exhibit enhanced shear viscosities or prolonged relaxation times. This is considered to be caused by a confinement-induced glasslike transition of the polymer chains at very high shear rates [7], which in magnetic storage can be of the order of $10^{10}$ s$^{-1}$ [8]. Sliding velocity is a critical parameter such that viscous forces dominate at the interface [9].

Interfacial slip is expected to occur at the nanoscale as well. Several authors have performed experiments [10–12] and molecular dynamics simulations [13–16] to investigate interfacial slip in sheared films. At low shear rates, the slip length depends strongly on velocity [12]; however, at high shear rates such as those encountered in magnetic storage, the slip length approaches a constant value that depends on the material properties of the solid [13]. Viscosity stiffening and interfacial slip are expected to be confounded in experimental measurements of the shear response. It is possible to decouple their effects by making certain assumptions about the state of ‘friction’ at the interface where slip occurs and applying these to the viscosity and slip models, as shown in the Appendix.

An experimental technique, referred to as fiber wobbling method (FWM), has been developed to measure the dynamic shear force at high shear rates with precise gap control [5], and was based on a modified surface force apparatus (SFA) [17, 18]. Unlike the SFA, which used atomically smooth mica surfaces as substrates, the FWM utilizes a vibrating spherical probe to shear a liquid parallel to a rough, rigid substrate such as a magnetic storage disk. Since liquid confined in nanometer-scale gaps is expected to be non-Newtonian, an effective (dynamic) viscosity represented by a complex function of the real (in phase) and imaginary (out of phase) components is measured in dynamic shearing experiments [7]. The real and imaginary components are termed viscosity $\eta'$ and elasticity $\eta''$, respectively [17]. While elasticity and local intermolecular interactions [11] dominate in atomically smooth surfaces (with the SFA) [6, 9, 17], this picture is reversed when roughness is present (in the FWM): the effect of roughness seems to be that it loosens the confinement of molecules such that the viscosity dominates the elasticity [7], while meniscus forces do not appear to play a role in the response [12].

A double-ended tuning fork (DETF) resonator [19] was recently added to the FWM apparatus to simultaneously measure the vertical force. The dynamic viscosity obtained from FWM measurements was used with classical continuum theory to predict the normal (bearing) force acting on the shearing probe [20]. Agreement with the experimentally
measured force indicates that continuum theory can adequately predict the lubricant behavior, under certain considerations. Specifically, experiments that utilize bulk fluid—even if that is a micron-size droplet—correspond well to continuum theory because the bulk dominates the shear response. In contrast, the same approach has limitations in the case of molecularly thin films where breakdown of continuum occurs. This is investigated in this work by interpreting available experimental and molecular dynamics simulations results, and proposing a model of the lubricant morphology at the interface that captures the behavior of lubricant molecules under shear. Experiments with molecularly thin layers, rather than bulk droplets, would help to illustrate the potential validity of this model when continuum breaks down. Finally, a semi-empirical, partially submerged sphere-on-flat model is proposed that can predict the hydrodynamic normal (bearing) and shear forces in the presence of scale effects, such as the breakdown of continuum and interfacial slip, as well as confinement and high shear rates in a molecularly thin lubricant layer.

2 Proposed Physical Model

Before any consideration of lubricant contact, it is important to characterize the lubricant morphology as accurately as possible. The main challenge is the expected breakdown of continuum theory at the nanoscale: are continuum quantities such as ‘shear rate’ valid with few lubricant molecules present at the interface? We address this fundamental question by examining experimental and molecular dynamics simulation data to propose a model of lubricant morphology that captures the limits of continuum theory. Using this model allows for the prediction of hydrodynamic forces up to the point where continuum breaks down. A qualitative description of lubricant behavior is offered beyond that point and up to the inception of solid contact.

Molecularly thin lubricant films, such as those used in magnetic storage, should have good retention and replenishment characteristics: both characteristics are achieved by using a mixture of bonded and mobile molecules. While at low speeds replenishment of the lubricant seems to be the critical factor in avoiding wear, at high speeds, surface mobility needs to be augmented by a strongly adhered bonded lubricant layer that helps resist asperity penetration (solid–solid contact). A strongly bonded layer coupled with a mobile layer of high viscosity is important at high speeds.

Perfluoropolyether (PFPE) lubricants such as Fomblin Zdol2000 and Zdol4000 have polar (hydroxyl) end groups that cause some molecules to adsorb onto the magnetic disk surface and become immobile. Surface-induced vitrification and van der Waals interactions are thought to contribute to the formation of a ‘glassy’ layer, approximately one radius of gyration in thickness, in the liquid near a solid surface [17]. In addition, chemical treatments of the lubricant with ultraviolet rays or annealing as well as the adsorptive nature of the substrate can increase the fraction of molecules that are bonded to the substrate in this ‘glassy’ state where the polymer segments at a certain distance from the solid surface appear to be effectively broken down. A strongly bonded layer coupled with a restricted geometry [17]. In a monolayer, both bonded and mobile molecules exist within the same plane and the mobile molecules have to spread in the spaces not covered by the bonded molecules, which conform to the solid surface texture [22].

The effect of the adsorbed layer is to move the plane of shear into the liquid, localized at the mid-plane of the film [10, 23]. Beyond a limiting value of the normal load where the film has transitioned into a ‘glassy’ state, the film appears to be nearly rigid and shear is confined to the wall–film interface in what is frequently called ‘plug-like flow’ [23]. With reduced shear rate, PFPE lubricants show viscous behavior since they have more time to conform to the external flow. However, very fast sliding at room temperature may not lead to lubricant depletion because the shear rate may be too fast for the molecules at the boundaries to catch up to external perturbations [8].

The transition to solid-like behavior in thin polymeric lubricant films under high shear rates has also been observed in molecular dynamics simulations [8]. While ‘shear thinning’ behavior is observed with increasing shear rate where functional end groups slow down the dynamics, confinement plays a more dominant role in lubricant rheology [8]. Simulations have shown that chain molecules, rather than crystallizing, change their configuration and orientation markedly as they approach the glass transition; this structure seems to become insensitive to chain length when the film thickness is much smaller than the radius of gyration in a bulk melt [23]. Confinement also retards the lubricant film relaxation process and increases its viscosity; this effect is more pronounced for lubricants with functional ends [8].

The average molecular height of the bonded layer $h_B$ depends on the coverage of the surface area and can be measured experimentally through ellipsometry. The molecular height for a coverage ratio less than one ($C < 1$ a.u.) is less than the monolayer height, which for Zdol4000 is equal to 2.3 nm; in such a case the layer is termed a submonolayer [22]. For example, if the coverage ratio is 0.8 a.u. then the molecular height is $h_B \approx 1.5$ nm, and is
less than the monolayer height resulting in the formation of a submonolayer. Note that $C = 0.2$ a.u. is the saturated value of the coverage ratio such that the molecular height of the bonded layer without any treatment, i.e., the smallest possible bonded layer thickness, is equal to $h_{B_{\text{min}}} = h_B$ ($C = 0.2$ a.u.) $\approx 1$ nm [22].

It is reasonable to assume that some of the mobile lubricant molecules will bond to the shearing surface when lubricant contact is established, creating a second submonolayer, which, in the absence of treatment, will have a thickness of $h_{B_{\text{min}}}$. Fukuzawa et al. [20] used non-polar Fomblin Z03 PFPE lubricant (molecular weight = 4,000 - a.m.u.) in their viscosity measurements. Since the non-polar lubricant has polymer chains with non-functional ends, only the smallest possible number of molecules will bond to the substrate corresponding to the saturated coverage ratio of $C = 0.2$ [22]. Then, the molecular height will be $h_0 = h_{B_{\text{min}}} = 1$ nm. In other cases where the coating material is not DLC, the minimum molecular height will depend on the chemistry between the lubricant molecules and the shearing surface’s material. As the gap between the solid surfaces decreases, mobile molecules will be expelled from the interface until, in the limiting case, the two bonded layers will come into contact and will shear past each other. The bonded molecules in the bottom layer will be blocked in their motion because they cannot cut across the molecules tethered to the shearing surface [17]. The minimum solid–solid gap corresponding to this scenario will be equal to the sum of the two layers’ thicknesses, i.e., $h_{0_{\text{min}}} = h_B + h_{B_{\text{min}}} = 2$ nm. Beyond this point, where arguably the definition of shear rate would stop being valid, experimental results seem to show that the interface ‘breaks down’ and one transitions from lubricant-solid to solid–solid contact through the rupture of the lubricant film. Rupture refers to the destruction of bonded layers through chain scission and breaking of bonds, a process through which previously bonded molecules are expelled from the interface allowing for the inception of solid contact.

Figure 1 is a schematic representation of the lubricant polymer chains being sheared between two solid surfaces with the top surface moving at a constant velocity $U$. The mean shear rate in the presence of interfacial slip is customarily determined by fitting a straight line to the average velocity profile in the central part of the channel [13, 15]. This is analogous to using a ‘hydrodynamic film thickness’ where the effective thickness of the immobilized layers is subtracted from the solid–solid distance [24]. We propose that for shear rate to exist as such within the film and in accordance with existing models [13], the presence of at least two monolayers or submonolayers is necessary. In that case, the shear rate within the fluid film will be equal to the difference in average velocity of the top and bottom layers over the minimum liquid gap. Defining the average layer velocity at the center of each layer’s thickness requires the use of the liquid gap $d_0$, which can be calculated from the solid–solid gap (minimum film thickness) $h_0$ as:

$$d_0 = h_0 - \kappa$$

The minimum liquid gap $\kappa$ depends on the lubricant morphology and chemistry and is defined relative to the minimum solid–solid gap as:

$$\kappa = \frac{1}{2} (h_B + h_{B_{\text{min}}})$$

When deriving the hydrodynamic forces, the liquid gap, given in Eq. 1, is used in place of the solid–solid gap and is valid for the range of $\kappa \leq d_0 \leq t - \kappa$, where $t$ is the total lubricant thickness. For example, in a lubricant of total thickness of $3$ nm and a minimum liquid gap of $1$ nm, the hydrodynamic forces exist for $1$ nm $\leq d_0 < 2$ nm. In terms of the solid–solid gap, this is equivalent to the range of $2$ nm $\leq h_0 \leq 3$ nm, while transition to solid contact occurs for $0 \leq h_0 \leq 2$ nm.

Figure 2 shows a schematic representation of the different regimes of contact. Itoh et al. [7] reported data of the probe amplitude and phase from fiber wobbling experiments with profiles similar to those depicted in Fig. 2 and postulated that intermittent solid contact starts at the inflection point in the plot of the amplitude gradient $(dA/dh_0)$—moving from right to left. They then suggest that steady solid contact occurs at a distance of $3 \times \sigma$, where $\sigma$ is the combined root-mean-square (RMS) roughness of the solid–solid interface for a Gaussian distribution of asperity heights—to the left of the inflection point. However, the reported combined RMS roughness is small ($3 \times \sigma = 1.89$ nm) and does not cover the entire range of transition. It is not entirely clear what happens in this transitional region. What seems to be clear from the experimental results is that there are two regimes to the
right and left of the transition that correspond to steady lubricant and steady solid contact, respectively, as shown in Fig. 2. Therefore, we propose that the transition region represents the approaching and shearing of the two bonded layers as well as the transition from intermittent to steady solid contact.

Specifically, the transition region contains the following two ‘events’ that may or may not be happening concurrently:

1. Rupture of the adsorbed lubricant layers through the destruction/expulsion of bonded molecules from the interface, as explained previously; continuum behavior breaks down in this region.

2. Transition from intermittent to steady solid contact.

Hence, with decreasing probe-substrate gap (from right to left) the probe comes into steady lubricant contact and reaches the maximum of the hydrodynamic forces just before it transitions through intermittentasperity contact and lubricant film rupture into steady solid contact. For \( h_{0,\text{min}} = 2\kappa = 2 \text{ nm} \), the width of the transition region is equal to \( 2\kappa + 3\sigma \approx 4 \text{ nm} \), which agrees with the experimental data reported in [7]. As described previously, roughness plays a role in the shear response of the FWM experiments in that the shear response is no longer quantized with respect to separation (as is the case for experiments utilizing atomically smooth surfaces), while the viscosity becomes larger than elasticity for rough surfaces. Since the FWM data is ‘capped’ up to the breakdown of continuum, the effect of roughness is diminished given that \( 2\kappa > 3\sigma \), and a smooth sphere-on-flat model can be used to analyze the experimental results.

Given that the viscosity reaches its maximum value when the minimum film thickness is \( 2\kappa \), the hydrodynamic forces, which also reach a maximum at this point, become equal to the solid forces over the transition gap. It is not the purpose of the current work to describe exactly the behavior of the lubricant in the transition gap, since the goal is to predict the hydrodynamic forces present up to the breakdown of continuum, i.e., for \( h_0 > 2\kappa \). It is possible that compressive stiffness increases until the bonded molecules are confined to a thickness comparable to their radius of gyration [17]. On the other hand, assuming simply that resistance to shear and compression ceases in the transition gap, the transitional forces can be modeled through linear equations:

\[
P_{\text{trans}} = \frac{P_{\text{fluid,max}} - P_{\text{solid,min}}}{2\kappa} h_0 \tag{3}
\]

\[
Q_{\text{trans}} = \frac{Q_{\text{fluid,max}} - Q_{\text{solid,min}}}{2\kappa} h_0 \tag{4}
\]

\( P_{\text{fluid,max}} \) and \( Q_{\text{fluid,max}} \) are the hydrodynamic forces at the limiting maximum viscosity and shear rate. \( P_{\text{solid,min}} \) and \( Q_{\text{solid,min}} \) are the normal and frictional forces at the inception of solid contact. The range of solid–solid gaps where these equations hold is \( 0 < h_0 \leq 2\kappa \), whereas hydrodynamic forces are valid for the range \( 2\kappa \leq h_0 \leq t \). At and beyond the inception of solid contact, i.e., for negative solid–solid gap \( h_0 \), we describe the forces as functions of interference \( \omega \) (note that solid forces are zero for \( \omega \geq 0 \)).

Using one of the available interfacial models for a sphere-on-flat geometry [25], the normal and frictional forces can be calculated at the inception of solid contact based on material parameters. Let the hardness, Young’s modulus, and Poisson’s ratio of DLC be 18 GPa, 180 GPa, and 0.24, respectively [26]. Since both the sphere and the substrate are coated with DLC, the reduced modulus can be calculated to be 95.5 GPa. Then, \( P_{\text{solid,min}} = 1.5 \mu \text{N} \) and \( Q_{\text{solid,min}} = 120 \mu \text{N} \) for \( \omega = 1 \text{ Å} \) (the same forces are 40 and 1,120 \( \mu \text{N} \) for \( \omega = 1 \text{ nm} \)). It will be shown later that the hydrodynamic forces can be of the same order of magnitude as the solid forces at the inception of solid contact. In the future the authors plan to incorporate these transitional forces in a generic rough surface model, expanding on previous work [1–3, 27].

The ‘true’ average shear rate in the lubricant film is [13]:

\[
\dot{\gamma}_{\text{true}} = \frac{U - u_s}{d_0} \tag{5}
\]

The slip velocity \( u_s \) is defined later. However, in the absence of knowledge of slip behavior, the ‘apparent’ shear rate measurable in experiments is based on the displacement of the shearing surface and is defined as:

\[
\dot{\gamma} = \frac{U}{d_0} \tag{6}
\]

The maximum shear rate occurs when the minimum solid–solid gap is equal to \( 2\kappa \) corresponding to a minimum liquid gap \( d_{0,\text{min}} = h_{0,\text{min}} - \kappa = \kappa \).
3 Modeling Viscosity and Interfacial Slip

3.1 Apparent Lubricant Viscosity Model

Several experimental measurements of dynamic viscosity as a function of gap or shear rate have been published for polymeric melts. Early studies used the SFA with the fluid being sheared between atomically smooth mica surfaces [6, 17, 28]. The FWM [5, 7, 20], based on the same principle as the SFA, uses high shearing frequencies of 100 Hz to few kHz to accurately measure the shear force with a vibrating, spherical-shaped optical fiber probe that is smaller than the one used in a typical SFA [7]. Earlier experiments utilized a lubricant droplet with thickness of 1 μm and a 102 μm radius probe; in such an apparatus, the spherical probe is only partially submerged in the lubricant film. In later experiments, the lubricant droplet was sufficiently thick for the probe to be fully submerged in the lubricant [7, 20].

For gaps smaller than 500 nm, PFPE lubricant viscosity increases monotonically to a maximum value about 30 times larger than the bulk-state viscosity in FWM experiments [7]. An increase in elasticity (not synchronized with the increase in viscosity) is observed when opposed adsorbed layers touch each other. However, elasticity also includes the compliance of the apparatus in dynamic shearing experiments and does not represent solely the elastic compression/shearing of the adsorbed polymer layers [24]. In ideally smooth and flat geometries, shear response is linear below a certain lubricant film deformation amplitude. Overall, the transition from linear to non-linear response is associated with significant changes in the structure of the confined molecules and its mechanism is slippage along an interface [9]. Linearity can be assured in dynamic shearing experiments by using small oscillation amplitudes and by limiting the interpretation of experimental results up to the breakdown of continuum theory, beyond which the shear response becomes non-linear. In the FWM, the sphere displacement is measured directly with optical methods and the dynamic viscosity is extracted by solving the resulting linear ordinary differential equation for the damping term [5]. During lubricant contact, this is equal to the shear force acting on the sphere:

\[ \eta_{\text{fluid}} = c\dot{x} = \eta \Omega U \]  

(7)

The average linear velocity of the probe is \( U = 2fA \), where \( f \) is the driving frequency, \( A \) is the vibration amplitude of the probe, \( \eta \) is the dynamic viscosity, and the damping coefficient is \( c = \eta \Omega \). The geometric term \( \Omega \) used by Fukuzawa et al. was derived from the work of Goldman et al. [29] and is valid for a fully submerged sphere. The fully submerged formulation is not applicable in magnetic storage applications where slider features are larger than the lubricant thickness; hence, a new geometric term for the case of a partially submerged sphere is introduced later in the present work for that purpose.

Dynamic viscosity \( \eta^* \) is a complex quantity that describes the system response in terms of the out-of-phase viscous term \( \eta' \) and the in-phase elastic term \( \eta'' \):

\[ \eta^* = \eta' - i\eta'' \]

(8)

It is related to the commonly used dynamic modulus \( G^* \) [6, 17, 28] through the vibration frequency such that \( G^* = 2\pi f \eta^* \); the storage modulus \( G' = 2\pi f \eta' \) (in-phase) and the loss modulus \( G'' = 2\pi f \eta'' \) (out-of-phase) are the components of \( G^* \) [30].

The maximum shear rate that occurs prior to the inception of solid contact is found from Eq. 6 to be \( 2 \times 10^5 \text{ s}^{-1} \). Note that in order to estimate the ‘true’ shear rate, i.e., the shear rate within the fluid, it is necessary to know the slip behavior as well.

The viscosity-liquid gap data adapted from Fukuzawa et al. [20], shown in Fig. 3a, can also be expressed as the viscosity-shear rate data shown in Fig. 3b where the shear rate is calculated from Eq. 6. Using the dynamic viscosity data up to the gap where it reaches its bulk value (≈500 nm) would enable the comprehensive description of lubricant viscoelasticity, for example by using Prony series representation of the viscosity-shear rate data [30]. Based on the above-mentioned experimental measurements [20], the limiting viscosity-gap behavior of the lubricant can be described as follows:

1. As the gap \( d_0 \) approaches the limiting value of \( \kappa = 1 \text{ nm} \), or equivalently, as the shear rate becomes very large, the dynamic viscosity (both \( \eta' \) and \( \eta'' \)) shows a significant increasing trend. When the liquid gap is \( d_0 = \kappa^+ \) just prior to the inception of solid contact the viscosity reaches a maximum limiting value \( \eta_0 \). This value can be extracted from experimental data; for example, from the measurements of Fukuzawa et al., \( \eta_0 = 0.31 \text{ Pa} \text{ s} \). Beyond the maximum limiting shear rate of \( 2 \times 10^5 \text{ s}^{-1} \) where this occurs, the viscosity remains constant at \( \eta_0 \).

2. At the other end of the spectrum, as the liquid gap becomes very large corresponding to very low shear rates, the viscosity approaches its bulk value \( \eta_\infty = 0.06 \text{ Pa s} \) [20].

Therefore, the dynamic (‘apparent’) viscosity is bounded between the bulk and maximum values. The shear rate of interest in magnetic storage is of the order \( 1 \times 10^{10} \text{ s}^{-1} \) as reported in the literature [8]. Hence, the viscosity during hard disk drive operation can be assumed to be constant and equal to the limiting viscosity \( \eta_0 \).
We propose that the viscosity-shear rate behavior can be adequately modeled from experimental data through a second-order polynomial such that:

\[ \eta = \eta(d_0) = ad_0^2 + bd_0 + \eta_0 \quad (9) \]

To express the viscosity as a function of shear rate rather than the gap, the above equation can be reformulated as:

\[ \eta = \eta(\dot{\gamma}) = a'(\dot{\gamma})^{-2} + b'(\dot{\gamma})^{-1} + \eta_0 \quad (10) \]

\[ a' = aU \quad (11) \]

\[ b' = bU \quad (12) \]

For the experimental measurements of Fukuzawa et al. [20] discussed above, the modified coefficients are: \( a' = 7.014 \times 10^6 \) Pa/s and \( b' = -2.090 \times 10^3 \) Pa. Only the viscous component of the dynamic viscosity is used in the hydrodynamic force formulations; therefore, from this point on and for the sake of simplicity the dash is dropped from the notation such that \( \eta = \eta' \). Furthermore, it should be stressed again that \( \eta \) is the ‘apparent’ viscosity that includes the coupled effects of both stiffening and slip.

### 3.2 Lubricant Interfacial Slip Model

To account for deviation from classical hydrodynamics, a dimensionless factor can be introduced to append the classical equations [10–12, 31]. The extent of the deviation from the classical prediction increases with increased velocity under constant film thickness [12]. The degree of slip increases both with the viscosity and the shear rate, while it also depends on the cohesive force at the interface: for example, a hydrophobic surface is expected to give rise to partial slip [10]. In fact, the contact angle drastically affects the boundary condition [15]. It has been shown experimentally that, while in a completely wetting system in the absence of polymer the degree of slip increases as the surface roughness increases, the opposite is true in the presence of polymer [10]. In experiments with equivalent surfaces having variable roughness, it was observed that the critical shear rate and stress for the initiation of partial slip increased nearly exponentially with increasing roughness [11].

Numerous boundary conditions were proposed in phenomenological models that allow for finite slip at the liquid–solid interface, but these fail to provide a universal picture of the momentum transport there. Slip will depend, among others, on the strength of the liquid–solid coupling (wall friction, \( \eta_w \)), the thermal roughness of the interface, and the commensurability of the wall and liquid densities. At high shear rates, the slip length increases rapidly with the shear rate and appears to diverge as the shear rate approaches a critical value. For flows in the vicinity of the critical shear rate, small changes in the surface properties can lead to large fluctuations in the apparent boundary condition; hence, the same liquid molecules sheared against different substrates will experience varying amounts of slip and vice versa [16]. Beyond the critical shear rate, the wall can no longer impart additional momentum to the liquid layer, reaching an asymptotic value of the slip length, which increases monotonically with increasing chain length [14]. Scaling the slip length by its asymptotic limiting value and the shear rate by its critical value collapses the data onto a master curve; therefore, the Navier model describes only the low-shear-rate limit of slip behavior [16]. This behavior was shown to be more generally applicable to polymeric systems [14].

Molecular dynamics simulations that predict unbounded slip length at high shear rates use rigid walls where the wall atoms have no thermal motion; if, instead, the wall atoms are allowed to oscillate about their positions in response to collisions with liquid molecules, momentum transfer is accounted for and the slip length is bounded [13]. This simulation result is verified by dynamical modeling and continuum fluid dynamics: specifically, the slip length is nearly constant and close to zero at low shear rates and increases sharply to reach a constant value that depends on...
the value of the wall friction \( \eta_w \). If the wall friction is zero then the slip length becomes unbounded at high shear rates \[13\].

It should be noted that, in keeping with the model presented herein, slip will not occur at the fluid–solid interfaces per se, but rather at the interface between the bonded and mobile layers as those were described previously. Since the top and bottom interfaces are similar, the slip behavior at each can be assumed to be the same. Hence, the net slip behavior can be modeled, through the slip velocity as a function of gap or shear rate, as occurring only at the top submonolayer–mobile layer interface: we shall refer to as a function of gap or shear rate, as occurring only at the top submonolayer–mobile layer interface: we shall refer to this interface as the ‘top wall.’ The classical definition of the slip velocity is that it is equal to the slip length multiplied by the local shear rate \( U/d_0 \) \[13\]. Here we incorporate the aforementioned friction constant at the top wall \( \eta_w \) that describes the strength of the solid–liquid interface and the slip length as a function of the liquid gap to propose the following form for the slip velocity:

\[
u_s = \frac{U \nu_s}{\eta_w d_0}
\]  

(13)

It is more convenient to use the dimensionless slip velocity defined as:

\[
u_s^* = \frac{u_s}{U} = \frac{L_s}{\eta_w d_0}
\]  

(14)

When the friction at the wall \( \eta_w \) is infinite, then the slip velocity is zero as is expected with the no-slip condition. If the friction is zero then the slip velocity becomes unbounded (infinite). Furthermore, when the slip velocity becomes larger than the shearing velocity, the slip practically becomes unbounded. Let us then propose the following boundary condition at the top wall (wall velocity):

\[
u_w = H[1 - \nu_s^*](1 - \nu_s^*)U
\]  

(15)

The Heaviside step function is used such that:

\[H[1 - \nu_{s0}] = \begin{cases} 0, & \text{if } \nu_s > U \\ 1, & \text{if } \nu_s \leq U \end{cases}
\]  

(16)

A dimensionless slip factor \( f^* \) can be defined that scales the classical Reynolds equation for the flow and hence the normal force acting on the sphere. This is shown later to be:

\[
f^* = H[1 - \nu_s^*](1 - \nu_s^*) = \frac{\nu_w}{U}
\]  

(17)

The factor \( f^* \) has the following limiting behavior: at the limit of one, the equations revert to the no-slip condition; the non-physical limit of zero corresponds to unbounded slip. Characteristic values of the slip and wall velocities as well as \( f^* \) are given for various cases in Table 1.

The Heaviside function accounts for unbounded slip by ‘turning off’ the velocity at the wall. The predicted normal force that can be exerted on the sphere in the case of unbounded slip is infinite as can be seen from the value of \( f^* \). Obviously, this cannot be realized in practical applications. Therefore, \( f^* \) is valid in the range \( 0 < f^* \leq 1 \). It should be noted that, through algebraic manipulation of Eq. 5, the ‘true’ shear rate accounting for slip can be expressed as:

\[
\dot{\gamma}_{true} = f^* \frac{U}{d_0}
\]  

(18)

Molecular dynamics simulations \[13\] show that for shearing speeds larger than about 8 m/s, the slip length becomes constant and equal to about 3 nm; the no-slip condition appears to be valid for speeds smaller than 2 m/s. Based on the approach of Zhu and Granick \[12\], it would be possible to extract the slip behavior of the lubricant from experimental measurements of the hydrodynamic normal force; however, those authors assumed a constant value for the viscosity whereas the viscosity stiffens as well under the effects of confinement and high shear rate. This is why, after fitting the theoretical prediction of the normal force to the experimental data, Zhu and Granick attributed any deviation from the experiments to interfacial slip alone \[12\]. We propose, instead, that both viscosity stiffening and slip are present at the interface.

Fukuzawa et al. \[20\] measured in their experiments the normal load during shearing and compared it to the theoretical prediction of classical solution without slip: they found that the classical solution is in good agreement with the experimental data. We further propose that any differences between the experimental and theoretical data arise from violation of the boundary conditions used in the formulation of the normal force as will be discussed later. The general agreement between the reported experimental and theoretical data is not surprising since the ‘apparent’ viscosity measured in the experiments and used in the calculation of the normal force includes both the effects of viscosity stiffening and interfacial slip. In order to investigate slip specifically, it is necessary to decouple its effects from those of viscosity stiffening. To this end, simultaneous measurements of both hydrodynamic—normal and shear—forces or molecular dynamics simulations specific

<table>
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to the studied system/lubricant morphology are necessary. A qualitative method to decouple viscosity stiffening and interfacial slip is shown in the Appendix and requires making certain assumptions on the strength of the solid–liquid interface, i.e., the value of the wall friction $\eta_w$.

4 Hydrodynamic Forces

4.1 Reynolds Equation for a Sphere-on-Flat

Accounting for Slip

Only the viscous response, i.e., the viscosity $\eta'$, which is 90° out of phase with the sinusoidal input and in phase with the rate of deformation, is used in the hydrodynamic force calculations [17]. To derive the hydrodynamic forces, a sphere-on-flat formulation is used, as shown in Fig. 4. A rigid smooth sphere of radius $R$ moves with constant velocity $U$ along the $x$-direction parallel to a rigid smooth plane; the assumption of rigidity is used only during fluid contact. This is done in lieu of the classical elastohydrodynamic approach [32, 33] so as to yield easily usable, closed-form solutions of the hydrodynamic forces that can be integrated into existing rough surface dynamic contact models [34]; solid deformations are indeed negligible during hydrodynamic contact (of the order of pm) when the substrate is DLC. It should be noted that the formation of ripples (moguls) under the effect of air stress on the lubricant layer [35, 36] is neglected in the present work and we assume that the lubricant layer is of constant thickness everywhere. Ripples can be modeled within the context of a dynamic contact model [1], for example, by dynamically varying the lubricant thickness under the slider at the head–disk interface.

The approach of the solid surfaces at the lowest point of the sphere is termed the minimum film thickness or solid–solid gap $h_0$ [32]. Based on the preceding discussion, the minimum film thickness can be replaced by the liquid gap $d_0$ using Eq. 1. In the sphere-on-flat configuration, shown in Fig. 4, the total effective bonded layer, comprising half of each bonded layer, is placed on the substrate to simplify the analysis and the solid–solid and liquid gaps are shown relative to the overall lubricant thickness. The following solutions are valid only when the total fluid film thickness $t$ is much smaller than the sphere radius, i.e., $t \ll R$.

The following list of assumptions is used in the derivation of the Reynolds’ equation describing the pressure distribution in the lubricant:

1. The fluid is isoviscous along its thickness and its viscosity depends only on the degree of confinement, i.e., $\eta = \eta(h_0)$. The latter can be achieved by using a very small oscillation amplitude to ensure the linearity of the response and isolate the effect of confinement (rather than that of the shear rate) [9].
2. The sphere and elastic half-plane are rigid during lubricant contact. This was reported to hold true in the literature [7].
3. The gap between the sphere and the plane surface is much smaller than the sphere radius, i.e., $h_0 \ll R$. Due to the problem definition where we have specified that $t \ll R$ and $h_0 < t$ for fluid contact to occur, this assumption is valid.
4. Because the gap is small compared to the sphere and substrate dimensions, the pressure is assumed to be constant across the film thickness, i.e., $\dot{c}p/\dot{c}z = 0$.
5. The flow is laminar. Indeed, the Reynolds number is very small for the length scale of the problem even at very high shear rates [14].
6. Inertial and surface tension forces are negligible relative to viscous forces. This has been reported to hold [12], not least because the probe displacement in the vertical direction can be precisely controlled.
7. Shear stress and velocity gradients are only significant across the film thickness.
8. The materials of the sphere and substrate are the same or sufficiently similar such that the slip behavior is the same at both solid–fluid interfaces. This is true, for example, in magnetic storage where both the slider (sphere) and disk (substrate) are coated with DLC [1–3].

The velocities in the liquid, denoted $u$ and $v$ in the $x$ and $y$ directions, respectively, can be obtained by integrating the forces acting on an infinitesimally small element of dimensions $dx \times dy \times dz$ with respect to $z$. The necessary boundary conditions are: $u(0) = v(0) = v(h) = 0$, and $u(h) = u_w$, where $h$ is the film thickness with the origin of the coordinate system located under the bottom of the sphere a distance $\kappa$ away from the substrate (as shown in Fig. 4) such that:

$$h = d_0 + \frac{x^2}{2R} + \frac{v^2}{2R}$$  \hspace{1cm} (19)

The viscosity used in the following equations is the ‘true’ viscosity that is assumed to be decoupled from interfacial slip and only includes the effects of viscosity.
stiffening due to confinement and high shear rates. The following velocity distributions are obtained after integrating the force balance equations twice and applying the boundary conditions:

\[
u = \frac{1}{2\eta_{\text{true}} \frac{\partial p}{\partial y}} (z^2 - z h) + \frac{z}{h} H \left[ 1 - u_s \right] \left( 1 - u_s \right) U \quad (20)
\]

The pressure gradient term in \( u \) corresponds to Poiseuille flow and the velocity term corresponds to Couette flow with an adjustment for slip. Using conservation of mass, the Reynolds equation can be obtained and corresponds to the classical solution given in the literature [32] appended for slip:

\[
\frac{\partial}{\partial x} \left( h^2 \frac{\partial p}{\partial x} \right) + \frac{\partial}{\partial y} \left( h^2 \frac{\partial p}{\partial y} \right) = 6H \left[ 1 - u_s \right] (1 - u_s) \eta_{\text{true}} U \frac{\partial h}{\partial x}
\]

\[
= 6\eta U \frac{\partial h}{\partial x}
\] (22)

Comparison with the classical solution leads to the definition of the dimensionless slip factor introduced previously in Eq. 17, i.e., \( f^* = \frac{H}{\eta_{\text{true}}} \left[ 1 - u_s \right] (1 - u_s) \). Furthermore, the ‘apparent’ and ‘true’ viscosities are correlated through the equation:

\[
\eta = \frac{H}{\eta_{\text{true}}} \left[ 1 - u_s \right] (1 - u_s) \eta_{\text{true}} = f^* \eta_{\text{true}}
\] (23)

4.2 Pressure and Velocity Distributions

The classical solution of the Reynolds equation to obtain the normal (bearing) force is based on the work of Kapitza [32]. Including the slip factor yields the following equation for the pressure distribution:

\[
p = -\frac{6f^* U \eta_{\text{true}}}{5} \frac{x}{h^2} = -\frac{24f^* \eta_{\text{true}} U}{5(2h_0 R + x^2 + z^2)^2}
\] (24)

The full forms of the velocity distributions can be obtained by plugging the pressure gradients \( \frac{\partial p}{\partial x} \) and \( \frac{\partial p}{\partial y} \) calculated from the above equation into Eqs. 20 and 21. The normalized velocities can be found as \( u^* = \frac{u}{U} \) and \( v^* = \frac{v}{U} \), while the \( x \) and \( y \) coordinates can be normalized as \( \xi = x(2h_0 R)^{-1/2} \) and \( \zeta = y(2h_0 R)^{-1/2} \), respectively [32].

Using the half-Sommerfeld boundary condition, i.e., disregarding the pressure distribution for positive \( \xi \) and using only the positive pressure (for \( \xi < 0 \)) gives a reasonable estimate of the load-bearing capacity [32]. Even though the half-Sommerfeld conditions violate mass continuity at \( \zeta = 0 \) [32], they can capture the magnitude of the normal force sufficiently accurately and are used in this work. The effect of slip is to decrease the magnitude of the pressure by scaling the distribution.

The normalized velocity in the \( \zeta \) direction immediately under the sphere (\( \zeta = 0 \)) is shown in Fig. 5, as a function of the normalized \( z \)-coordinate \( z = z/h_0 \). Here we can clearly see the two boundary conditions corresponding to no-slip and finite slip: under the effect of interfacial slip, the velocity in the fluid is less than the velocity of the sphere. The thin lines represent the simplified solutions for the velocity distributions where the pressure gradient is neglected; we denote these solutions with the subscript ‘Couette.’

4.3 Forces Acting on the Sphere

Half-Sommerfeld conditions are used as boundary conditions of the integration to yield a closed form expression for the normal (bearing) force:

\[
P_{\text{fluid}} = \int_{-\infty}^{+\infty} dy \int_{-\infty}^{0} p \, dx
\] (25)

The result of the integration given in classical hydrodynamic theory [32] has now been appended for interfacial slip through the slip factor \( f^* \):

\[
P_{\text{fluid}} = \frac{6\pi}{5} f^* \eta_{\text{true}} U \sqrt{\frac{2R^3}{d_0}}
\] (26)

The shear stress on the surface of the sphere can be calculated as:

\[
\tau = \eta_{\text{true}} \left[ \frac{\partial u}{\partial \zeta} \right]_{\zeta = h} = \frac{4RU \eta_{\text{true}}}{5} f^* \left[ \frac{2d_0 R + 7x^2 + y^2}{(2d_0 R + x^2 + y^2)^2} \right]
\] (27)

The shear force can be calculated by integrating the shear stress over the surface of the sphere submerged into the lubricant layer. The area of the submerged spherical cap is:
S = 2\pi \int_{R-(t-d_0)}^{R} R \, dy = 2\pi R(t - \kappa - d_0) \tag{28}

Hence, the shear force is:

\[ Q_{\text{fluid}} = 2\pi \int_{R-(t-n-d_0)}^{R} \tau \left|_{y=\sqrt{R^2-y^2}} \right. \, R \, dy \tag{29} \]

The full solution is:

\[ Q_{\text{fluid}} = \frac{8\pi U \eta_{\text{true}}}{5 R} \left[ \frac{(t - \kappa - d_0)}{(2d_0 + R)^2} \right] \times \left[ 2d_0^2 - R^2 + 4d_0(R - t + \kappa) - 6R(t - \kappa) + 2(t - \kappa)^2 \right] \tag{30} \]

Neglecting the pressure gradient in Eq. 20 results in a velocity that includes only the Couette term. Then, calculating the velocity gradient at \( z = h \), multiplying with viscosity to find the shear stress, and integrating over the spherical cap surface to find the shear force as was done with Eqs. 27–30, yields the following simplified expression:

\[ Q_{\text{fluid,Couette}} = 4\pi U \eta_{\text{true}} \left[ \frac{R^2 - \kappa - d_0}{2d_0 + R} \right] \tag{31} \]

Hydrodynamic forces can be normalized by dividing through with \( U \eta_0 R \) where \( \eta_0 \) is the maximum limiting viscosity at the minimum liquid gap. Figure 6 is a comparison of the dimensionless shear force with and without the Poiseuille term (i.e., non-zero and zero pressure gradient, respectively). It can be observed that neglecting the pressure gradient and calculating the shear force using a Couette flow approximation [28] underestimates the shear force by two orders of magnitude. While this effect can be expected to diminish away from the bottom of the sphere (i.e., for \( \zeta = \zeta \neq 0 \)) as well as at higher shear rates where the Couette term will dominate the Poiseuille term, it cannot be neglected when interpreting the experimental data. Note that the limit of the shear force when the liquid gap \( d_0 \) tends to \( t - \kappa \) is zero.

Henceforth, the following equations, shown in terms of the measured dynamic ‘apparent’ viscosity \( \eta = f^* \eta_{\text{true}} \), can be used to calculate the normal and shear forces:

\[ P_{\text{fluid}} = \frac{6\pi U}{5} \sqrt{\frac{2R^3}{d_0}} \tag{32} \]

\[ Q_{\text{fluid}} = \frac{8\pi U \eta(t - \kappa - d_0)}{5} \left[ \frac{(2d_0 + R)^2}{2d_0^2 - R^2 + 4d_0(R - t + \kappa) - 6R(t - \kappa) + 2(t - \kappa)^2} \right] \tag{33} \]

4.4 Applicability of Force Formulations to Shearing Experiments

The shear force was given earlier as:

\[ Q_{\text{fluid}} = \eta \Omega U \tag{7} \]

The geometric factor for a partially submerged sphere is obtained from Eq. 33 as:

\[ \Omega_{p-s} = \frac{8\pi(t - \kappa - d_0)}{5} \left[ \frac{(2d_0 + R)^2}{2d_0^2 - R^2 + 4d_0(R - t + \kappa) - 6R(t - \kappa) + 2(t - \kappa)^2} \right] \tag{34} \]

Correspondingly, the geometric factor used by Fukuzawa et al. [20] is based on the work of Goldman et al. [29] for a fully submerged sphere:

\[ \Omega_{f-s} = \frac{16\pi}{5} R \ln \left( \frac{d_0}{R} \right) \tag{35} \]

Figure 7 presents a comparison between the two geometric factors. It can be observed that a fully submerged formulation corresponds to a geometric factor that is more than three orders of magnitude larger than the
partially submerged equivalent. This is expected, since the shear force acting on a fully submerged sphere being dragged through a liquid is expected to be larger than if the sphere were only ‘surfing’ on the liquid surface. Since the partially submerged formulation requires that the film thickness be smaller than the sphere radius, it does not converge to the fully submerged sphere. Therefore, special care should be taken to match the experimental conditions to the appropriate shear force formulation.

In their experiments, Fukuzawa et al. used a droplet of lubricant as the ‘reservoir’ where the probe is vibrated; the droplet thickness was either about 1 μm in earlier work [5] or sufficient to fully submerge the spherical probe in later experiments [7, 20], i.e., of the order of 200 μm. In either case, the formulation used for the hydrodynamic forces should take into account the fact that the lubricant film does not extent sufficiently in the lateral direction for the full- or half-Sommerfeld boundary conditions [32] to be valid. Specifically, even in the case of a fully submerged probe and given that the droplet roughly takes the shape of a hemisphere, the diameter of this hemisphere will only be twice the probe diameter, i.e., of the order of 400 μm. As shown previously, full- or half-Sommerfeld conditions require limits of integration for the pressure in the film that tend to infinity in the lateral directions: the limits of integration for the full-Sommerfeld conditions are \(-\infty\) and \(\infty\), while half-Sommerfeld conditions require integration between \(-\infty\) and zero. Perhaps more importantly, the same consideration holds for the use of the fully submerged shear force formulation, which requires that the lubricant be treated as a semi-infinite fluid [29]. Hence, the methodology used in Fukuzawa et al. [20] is expected to be under-predicting the viscosity. This is because a larger mass of lubricant would provide larger shear resistance than a droplet of the same order of magnitude as the probe.

5 Hydrodynamic Forces at Higher Shear Rates

The presented model can be used to predict the forces acting on a sphere of any radius \(R\) (while \(R \gg t\)) moving in a lubricant of thickness \(t\) with a constant velocity \(U\). As explained previously, expressing the viscosity measurements as functions of shear rate rather than gap allows for the extraction of the ‘apparent’ viscosity at any velocity since we postulate that, for a given lubricant morphology/chemistry, the maximum ‘apparent’ viscosity is bounded in the upper limit by \(\eta_0\). Therefore, even at shear rates larger than the corresponding limiting shear rate, the ‘apparent’ viscosity will assume this constant limiting value.

In the case of magnetic storage with velocities of \(\sim 30\) m/s the shear rates in the lubricant will be much greater than those found in the experiments of Fukuzawa et al. A simple calculation of the minimum shear rate for the same lubricant morphology of with total thickness \(t = 3\) nm, and \(d_{0,max} = t - \kappa = 2\) nm, shows this to be \(\sim 1 \times 10^{10}\) s\(^{-1}\), similar to what is reported in the literature [8]. In contrast, the maximum shear rate observed in the shearing experiments for \(d_{0,min} = \kappa = 1\) nm and \(U = 200\) μm/s is much smaller at \(1.4 \times 10^5\) s\(^{-1}\). Therefore, in magnetic storage applications we postulate that the maximum viscosity in the lubricant layer is equal to the limiting value of \(\eta_0 = 0.31\) Pa s.

In magnetic storage, contact at the head–disk interface can be approximated as that between a smooth flat and an equivalent rough surface; the asperities on the latter can be assumed to be spheres with a radius of \(\sim 500\) nm [1–3]. Using the presented sphere-on-flat formulation, the resulting hydrodynamic forces acting on a single asperity will be as shown in Fig. 8.

Lubricant contact is valid only up to the total lubricant film thickness of 3 nm, and, equivalently, up to \(d_{0,max} = 2\) nm. Both forces are increased relative to the reported experimental values [20] by about two orders of magnitude stemming from a five order of magnitude increase in shearing velocity from 200 μm/s to 30 m/s. Even though the forces are linearly dependent on the shearing velocity, the correspondence is not one-to-one due to the effect of the viscosity, which is capped at the value of \(\eta_0\) at higher shear rates. Note that using the bulk value of the viscosity results in significant under-prediction of the hydrodynamic forces as shown in Fig. 8. In fact, the shear force is practically zero (data lies on the x-axis) if the bulk viscosity is used.

For a given set of material parameters for DLC, the minimum solid forces were calculated to be \(P_{solid,min} = 1.5\) μN and \(Q_{solid,min} = 120\) μN for \(\omega = 1\) Å, and 40 and 1,120 μN for \(\omega = 1\) nm as shown earlier. These magnitudes are comparable to the hydrodynamic forces developed for the sliding speeds encountered in hard disk drives. Therefore, previous models where the lubricant was

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**Fig. 8** Hydrodynamic normal (a) and shear (b) forces for a 500 nm radius sphere (relevant to magnetic storage)
assumed to offer no resistance to shearing and was simply displaced during contact [1–3] need to be corrected to account for lubricant contact. More importantly, it seems possible that the bearing capacity of the molecularly thin lubricant could benefit the interface by protecting the DLC coating from light contact.

6 Conclusions

A description of the morphology and behavior of molecularly thin lubricant films has been presented that can explain the experimental results obtained from dynamic shearing experiments based on the available literature. Models have been proposed that account for viscosity stiffening and interfacial slip. The validity of using the dynamic viscosity measured in shearing experiments has been discussed and an improved formulation has been proposed for the shear force acting on a partially submerged sphere shearing a thin lubricant film. ‘Apparent’ viscosity data were used to predict the hydrodynamic forces acting during lubricant contact found in the head–disk interface of hard disk drives and these were compared to the forces developed during solid–solid contact. The proposed sphere-on-flat model can be readily incorporated in existing rough surface contact models with friction to provide a more complete picture of nanoscale, rough, lubricated contacts. Future work utilizing molecular dynamics simulations and experiments with molecularly thin films would help investigate further the breakdown of continuum.

Appendix: Decoupling Viscosity Stiffening and Interfacial Slip

Simultaneous measurement of the normal and shear forces with the dynamic ‘apparent’ viscosity can be used to decouple viscosity stiffening from interfacial slip. However, in the absence of such data, another way to decouple the two effects is to use molecular dynamics simulation data. For example, Martini et al. [13] report that the slip length \( L_s \) for velocities larger than \( \sim 8 \, \text{m/s} \) is constant at \( \sim 3 \, \text{nm} \). Even though simulations pertaining to the exact lubricant morphology/chemistry are necessary for a more accurate estimate of the slip length, we shall assume that \( L_s = 3 \, \text{nm} \) is valid in this work for the purpose of demonstrating the procedure.

The dimensionless slip factor \( f^* \) can be calculated from Eqs. 14 and 17. As a first approximation, the wall friction \( \eta_w \) can be assumed to be unity. Knowing \( f^* \) allows for the extraction of the ‘true’ viscosity from Eq. 23 and the ‘true’ shear rate from Eq. 18. These calculations result in \( f^* \) becoming zero—corresponding to unbounded slip—at \( d_0 \approx 2.5 \, \text{nm} \) and \( \eta_{\text{true}} \) tending to infinity at the same liquid gap value. This is an artifact arising from the arbitrary assumption that the wall friction is unity. The minimum liquid gap of \( d_{0,\text{min}} = 1 \, \text{nm} \) corresponds to a maximum ratio of slip length to liquid gap \( L_s/d_0 = 3 \). Since unbounded slip is unrealistic, we can constrain \( f^* \) such that it is larger than zero at the minimum liquid gap by tuning the wall friction \( \eta_w \) accordingly; taking \( \eta_w = 3 \) results in the slip factor becoming zero. Figure 9a shows plots of the ‘true’ viscosity and slip factor as functions of gap for the given parameter values of \( L_s = 3 \, \text{nm}, \, d_{0,\text{min}} = 1 \, \text{nm}, \) and \( \eta_w = 3 \).

The plots of Fig. 9a represent the lower limit of the wall friction where the ‘true’ viscosity again tends to infinity as \( f^* \) tends to zero. As is shown in Fig. 9b, if the wall friction \( \eta_w \) is increased to a value of 5, the maximum ‘real’ viscosity becomes only 2–3 times larger than the maximum ‘apparent’ viscosity and the slip factor ranges between \( \sim 0.4 \) and unity. In the upper limit of \( \eta_w \rightarrow \infty \) (not shown), the ‘true’ and ‘apparent’ viscosities become the same and \( f^* \) is everywhere 1, resulting in the no-slip boundary condition: only viscosity stiffening happens in this case.

It is obvious that the exact values of the ‘true’ viscosity and interfacial slip as functions of the liquid gap are very sensitive to the interaction between the bonded molecules and the substrate, i.e., to the value of the wall friction \( \eta_w \). Future experiments or molecular dynamics simulations could focus on investigating this interaction. Based on the proposed models and methodology, the following appears to hold for the wall friction: \( L_s/d_{0,\text{min}} \leq \eta_w < \infty \). Therefore, we can say that lubricants confined in molecularly thin gaps and sheared at high speeds either exhibit the confounded effects of both stiffening and slip as the wall friction tends to the lower limit of \( L_s/d_{0,\text{min}} \), or stiffening overrides slip as the wall friction tends to infinity.

![Fig. 9 ‘True’ viscosity and slip factor as functions of the gap for: a \( \eta_w = 3 \) and b \( \eta_w = 5 \)](image-url)
References


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