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Elastic deformation during dynamic force measurements in viscous fluids

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ABSTRACT

Understanding and harnessing the coupling between lubrication pressure and elasticity provides materials design strategies for applications such as adhesives, coatings, microsensors, and biomaterials. Elastic deformation of compliant solids caused by viscous forces can also occur during dynamic force measurements in instruments such as the surface forces apparatus (SFA) or the atomic force microscope (AFM). We briefly review hydrodynamic interactions in the presence of soft, deformable interfaces in the lubrication limit. More specifically, we consider the scenario of two surfaces approaching each other in a viscous fluid where one or both surfaces is deformable, which is also relevant to many force measurement systems. In this article the basic theoretical background of the elastohydrodynamic problem is detailed, followed by a discussion of experimental validation and considerations, especially for the role of elastic deformation on surface forces measurements. Finally, current challenges to our understanding of soft hydrodynamic interactions, such as the consideration of substrate layering, poroelasticity, viscoelasticity, surface heterogeneity, as well as their implications are discussed.

1. Introduction

Viscous forces caused by the relative movement of two surfaces in a viscous fluid can exert local pressures that can be sufficiently large to cause elastic deformation of the interacting materials (see Fig. 1). This scenario is very common in the tribology of lubricated contacts, where deformation due to viscous forces in oils is a key mechanism to reduce friction and prevent wear (i.e. elastohydrodynamic lubrication or EHL, see Fig. 1a) [1]. Similarly, the presence of an elastic boundary during the drainage or infusion of fluid in a confined gap can lead to elastohydrodynamic deformation (EHD, see Fig. 1b).

Both EHD and EHL play an important role in soft matter where materials such as gels, biological tissues, or elastomers (Young's modulus ~ 10 kPa-100 MPa) can deform during motion in a fluid at relatively low velocities and viscosities [2–5]. For example, elastic deformation will affect the collision and rheological response of soft colloidal particles (Fig. 1c (i) and (ii)), coalescence of bubbles/drops (Fig. 1c (iii)), and has been hypothesized as a mechanism for shear thickening behavior [6]. Studying the dynamic adhesion of cells or capsules attached to a wall is also accompanied by deformation, and the stored elastic energy greatly alters the detachment process (Fig. 1d) [7]. Similarly, insects and several vertebrates are also known to mediate contact between their soft toe pads and surfaces through liquid layers [8], therefore understanding their locomotion and detachment requires understanding how the toe pads and highly confined liquid layers interact. Finally, in joint cartilage, a lubricating layer of fluid and the squeeze out from a polymer network (weeping lubrication) is suspected to be responsible for the low apparent friction across joints [9,10].

These deformations, in turn, can have a profound effect on the hydrodynamic interactions [11], such as those encountered during dynamic force measurements with the atomic force microscope (AFM) [12*,13] or the surface forces apparatus (SFA) (see Fig. 1b) [14,15*]. The SFA and AFM are well-suited to study how hydrodynamic interactions affect soft materials, as demonstrated with droplets or bubbles [16*–21](Fig. 1c(iii)). More recently, these tools have been employed and adapted to study elastohydrodynamic deformation of compliant solids [13–15*]. It is also important to consider the possibility of elastic deformation in the interpretation of surface forces measurements when one (or both) of the interacting surfaces has some degree of compliance (or reversely to know when to confidently ignore it). Neglecting to account for elastic deformation could lead to misinterpretation of the conservative surface forces (such as van der Waals or electrostatic) or slip behavior.

Due to its broad practical significance, a better understanding of the impact of deformation on hydrodynamic interactions and surface forces is necessary. Here we review studies, which in general have considered the classic geometry of a sphere approaching a plate at a constant drive velocity, and where one of the surfaces is supported by a force measuring spring (see Fig. 1b). These measurements are performed in viscous fluids within the lubrication limit, which are the experimental conditions encountered in typical surface forces measurements. This review is organized as follows: first we describe the theoretical framework and present experimental validations, focusing on direct force measurements. We then discuss current scientific challenges in describing or characterizing hydrodynamic interactions in the presence of

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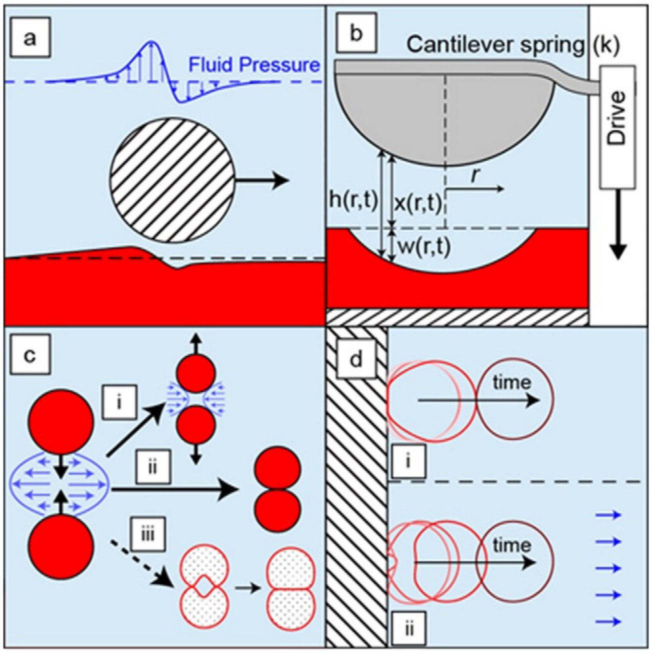

Fig. 1. Hydrodynamic interactions in the presence of deformable materials. (a) Elastohydrodynamic lubrication: lift generated due to the sliding of a rigid cylinder along a soft material. (b) Elastohydrodynamic force measurement in the sphere-wall configuration with labeled variables. (Not to scale) (c) Rheological behavior of particles, bubbles, or drops colliding followed by either (i) rebound or (ii) sticking, or (iii) dimpling and then coalescence for bubbles/drops. (d) Adhesion and detachment of a soft or liquid-filled capsule, or cell, where the morphology and dynamics of detachment changes in the presence of quiescent (i) or external (axisymmetric) flow (ii). (d) is adapted from [22] with permission.

elastic deformation. Finally, we provide broad guidelines to researchers working on surface forces measurements that highlight experimental conditions where elastic deformation might be present in dynamic force measurements with the AFM or SFA. Important and related topics not covered here include bouncing and rebounding of spheres [6](Fig.1c), jamming of deformable spheres [23], elastohydrodynamic lubrication (EHL) [1], and rheological implications [24*].

2. Physical description

While there are many configurations where elastohydrodynamic deformation might be important (see Fig. 1), the most common alignment encountered surface force measurements is the sphere-plane geometry. This configuration facilitates experimentation and analysis due to axisymmetry and by ensuring point contact (Fig. 1b). A geometrically equivalent configuration would be that of two nearby cross-cylinders which is used in the SFA [25]. The description of the elastohydrodynamic problem needs to address the coupling between fluid dynamics and surface deformations. The lubrication approximation with the no-slip boundary condition (Eq. 1) describes drainage and infusion of liquid from a thin gap in the limit where the surface separation is much smaller than the radial length scale:

$$\frac{\partial h}{\partial t} = \frac{1}{12\mu r}\frac{\partial}{\partial r}\left(rh^3\frac{\partial p}{\partial r}\right), \tag{1}$$

where h is surface separation, t is time, μ is the viscosity for a Newtonian fluid and p is fluid pressure [26,27]. In the absence of elastic deformation the fluid pressure distribution is readily obtained from Eq. 1 for a given initial condition, $h(r,0)$. The pressure can then be integrated radially to determine the total hydrodynamic force.

Surface deformation alters the fluid film thickness profile and shape of the interacting surfaces. This change in local fluid film thickness modifies the fluid pressure which, in turn, determines the deformation profile. The deformation profile, $w(r,t)$, can be obtained from linear elasticity for a spherical half-space, and is given by Eq. 2:

$$w(r,t) = 4\theta\int_0^\infty f(y,t)\frac{y}{y+r}K\left[\frac{4ry}{(r+y)^2}\right]dy, \tag{2}$$

where $\theta = \frac{1-\nu^2}{\pi E}$, ν is Poisson's ratio, E is Young's modulus, f is surface stress distribution, y is a dummy variable used in integration, and K is the complete elliptic integral of the first kind. In Eq. 2 the surface stress distribution comes from the fluid pressure, and due to the solid–fluid coupling, the lubrication equation needs to be solved simultaneously with the deformation profile. Usually a numerical solution is necessary, but analytical solutions can be obtained by making some approximations. For instance, Davis et al. [28*] demonstrated that in the limit of small deformation ($w/x < 0.05$ in Fig. 1b) a closed-form solution can be obtained by using the undeformed surface separation in the lubrication equation. This solution has served as a useful means of examining whether notable deformation is present in drainage flow [25]. Alternatively, a simple algorithm proposed for modeling half-space in the case of significant deformation is to treat the deformation as a non-adhesive contact (Hertz theory [29]), and to replace the contact pressure distribution with the fluid pressure obtained for a given gap thickness by the lubrication equation [30].

A more general consideration than assuming a half-space is necessary to treat the case of hard surfaces with compliant coatings or more generally for layered materials. In this case, the deformation can be calculated by applying a sticky boundary condition on the rigid substrate, using fluid pressure as a surface condition, and then solving for linear elasticity theory [31]. This approach allows for the role of the coating thickness to be investigated directly. For instance, Leroy and Charlaix used this approach to characterize regimes where fluid viscosity or the elasticity of the soft materials dominates the force response when confined by an oscillating rigid indenter (see Fig. 2a) [3*]. Using this method they demonstrated that the absolute modulus of soft coatings could be measured out of contact, and without measuring the contributions of the underlying substrate.

During surface forces measurements one surface is typically mounted on a spring (or cantilever). In this case, the surface separation is different from the displacement of the driving motor due to the deflection of the spring. The different contributions to the total force, F_T, from the hydrodynamic and conservative forces (F_s, e.g. van der Waals force, double layer force) can be considered independently [16*] and this force balance for the sphere-wall geometry is given by Eq. 3:

$$F_T = kS(t) = \int_0^R 2\pi r p(r)dr + F_s, \tag{3}$$

where R is the radius of spherical particle, and $S(t)$ is the deflection of the cantilever with a spring constant k as a function of time.

For rigid surfaces in simple geometries (e.g. sphere-plane, crossed cylinders) the Derjaguin approximation can be used to extract the surface potential $E(h)$ from surface force $F_s = 2\pi RE(h)$. In addition, the hydrodynamic contribution to the total force can be further simplified to the Taylor equation [25]. However, for deformable surfaces, the surface geometry as well as the absolute surface separation, can vary due to fluid pressure. Thus, the knowledge of the surface profile as a function of time is required to decouple the hydrodynamic and surface forces. To do so, one needs to estimate how much deformation is present in the separation range where surface forces are measured. As an example, we present here deformation maps (see Fig. 2b) by solving Eqs. 1-3 for a continuous approach in the absence of surface forces ($Fs = 0$). In Fig. 2b the relative deformation at the center point (w/h) is plotted for a range of Young's modulus (half-space) for experimental conditions that are typical of AFM ($R = 5$ µm, $k = 1$ N/m) or SFA ($R =$