Hydrodynamics in nanoscale confinement: SFA and colloid probe AFM liquid drainage experiments

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Abstract. Flow and drainage of very thin liquid films play an important role in mineral recovery, drop coalescence and emulsion stability, as well as lubrication of micromechanical devices. Studies of liquid flow under strong confinement (i.e., film thickness below a few hundred of nanometers and down to a few nanometers) can reveal the limits of applicability of a classical hydrodynamics description, but are very challenging. The Surface Force Apparatus (SFA) technique has enabled studies of drainage at nanoscale separation between atomically smooth mica sheets. The development of the colloid probe Atomic Force Microscope (AFM) as an alternative technique has allowed a significantly wider variety of confining solid surfaces to be studied. Both the SFA and the colloid probe AFM have been adapted to permit the surfaces confining the film to be soft, e.g., the surface of a drop or bubble, and therefore deformable. We present a succinct review of the experimental and theoretical modeling challenges for such studies and critically discuss the outcomes of recent experiments.

Dedicated to Prof. François Feuillebois on the occasion of his 65th anniversary

1. Introduction
Flow and drainage of very thin liquid films play an important role in, among others, mineral recovery by flotation, drop coalescence and emulsion stability, as well as lubrication of micromechanical devices. The rate of drainage of a Newtonian liquid in situations where the confinement is strong, i.e., when the surfaces bounding and squeezing out the film are separated by distances below a few hundred of nanometers and down to few nanometers, is also of interest for understanding the limits of applicability of classical hydrodynamics. Moreover, such studies can eventually provide a way to assess the validity of the hydrodynamic boundary conditions, for example the no-slip condition usually assumed for liquid flow over a smooth solid surface. A violation of the no-slip boundary condition can be quantified in terms of the so-called hydrodynamic slip length $\lambda$ (see, e.g., Ref. [1]). Slip can have an observable effect on the drainage rate only when the lengthscale of confinement becomes comparable with $\lambda$. For smooth surfaces (roughness smaller than few nm), it has been argued that the slip length, if any, should range from no more than a few nm for lyophilic surfaces to a maximum of few tens of nm for lyophobic ones [1, 2]. Sophisticated techniques and very well controlled experimental conditions are therefore necessary to ensure the required strong confinement and to enable a reliable discrimination of a slip length.

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Various methods, such as bubble-plate collision [3–6], the Thin Film Balance (TFB) balance technique (also known as the Sheludko cell) [7,8], the Surface Force Apparatus (SFA) [9–13], and the colloid probe Atomic Force Microscope (AFM) [14–25] have been employed to study hydrodynamic behaviour in confined systems. In this paper we will focus on investigations with Newtonian liquids involving the latter two methods.  

1.1. Thin film drainage in SFA and colloid probe AFM

A large number of studies used the SFA to study drainage of various liquids between atomically smooth mica sheets (see, e.g., Refs. [9, 13, 26, 27]. Almost two decades ago the colloid probe AFM technique was developed [14]. This permitted the extension of thin film drainage studies to a much larger variety of solids (see, e.g., Refs. [17, 18, 20, 21, 28, 29] and references therein), including the so-called asymmetric systems, i.e., the two bounding solid surfaces being made out of different materials, e.g., mica and silica, silica and titania, or silica and polystyrene. A consistent theoretical framework for the interpretation of these thin film drainage experiments was developed in Ref. [30]. At about the same time, modifications of the SFA have been made to allow one of the surfaces to be soft and thus deformable, e.g., a drop or a bubble [31, 32]. Since then, a number of studies involving such set-ups have been carried out (see, e.g., Ref. [33] and references therein). In the last decade the colloid probe AFM has been also extended to permit studies in which one or even both of the surfaces confining the film are soft (see, e.g., Refs [22–25, 34–36] and references therein). Building upon the theoretical background developed in Refs. [31, 37–39] to describe the static surface forces in the case when one of the two interfaces is soft, in the last decade a very elaborate theoretical framework aiming at a consistent interpretation of the AFM measurements of film drainage between deformable surfaces was developed (see Refs. [40–42] for an excellent, detailed presentation).

In Fig. 1 we show schematically the typical experimental setup of the AFM or SFA thin film drainage studies. We note that the relative velocities of the two bodies in the typical AFM or SFA experiment are rather small (usually not greater than few tens of micrometers per second). The corresponding Reynolds number $Re = \frac{\rho_f U h_0}{\mu_f}$ (where $\rho_f$ and $\mu_f$ are the density and viscosity of the liquid, respectively, $U$ is the average flow velocity, and $h_0 := h(0, t)$ is the separation) is small; for example, in the case of a water film draining with an average flow velocity $U' \approx 1 \text{ mm/s}$ (corresponding to surfaces approaching at speeds of the order of micrometers/s) at separation $h_0 = 10 \text{ nm}$, the Reynolds number is $Re \approx 10^{-5}$. The hydrodynamics of a Newtonian liquid is then well-described by the Stokes equations [17, 20, 42, 43]. (We note here that the Newtonian character of the liquid under such strong confinement is actually a model assumption which should be validated independently.) Moreover, in the range of film thicknesses of interest here, i.e., $h \lesssim 100 \text{ nm}$ the lateral extent of the interaction zone, which is defined as the part of the system where the pressure differs significantly from that of the bulk liquid [42], is of the order of microns or more (see Fig. 1), i.e., much larger than the film thickness. This allows one to describe the film drainage within the approximations of the Reynolds lubrication theory.

The corresponding thin-film hydrodynamics equation, basically employed for the data analysis by all the AFM and SFA studies that we discuss here, is written in cylindrical coordinates $\{r, \phi, z\}$ (see Fig. 1) as [30, 41, 42]

$$\mu_f \frac{\partial^2 u}{\partial z^2} = \frac{\partial p}{\partial r},$$

where $p := p(r, t)$ denotes the pressure field and $u := u(r, z, t)$ is the flow field. This is to be

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2 The use of SFA and colloid probe AFM techniques in studies of hydrodynamics under nanoscale confinement was one of the points of “Challenges in microhydrodynamics” session of the workshop “Microparticles in Stokes flows”, August 21-24 2011, Warsaw (Poland).
Figure 1. Schematic drawing of a typical AFM or SFA liquid drainage experiment. The two bodies of viscosities $\mu_{1,2}$ are moved one towards another through a fluid of viscosity $\mu_f$ by displacing either the lower stage (as shown here), or the top one, with an externally prescribed velocity $V(t)$. Either or both of the two bodies can be spheres, part of spheres, or flats (infinite radius of curvature). In the modified SFA discussed in Refs. [31,33] the lower stage is replaced by a rigidly fixed capillary filled with the fluid 2, which is kept at a constant pressure that is sufficient for creating the spherical protrusion, as shown in the figure. The case of a solid body is recovered by taking the limit $\mu_i \to \infty$ ($i = 1$ or 2), while that of fluid corresponds to finite values of the viscosity. The values $h(r,t)$ shown in the figure indicate the range of interest for the present work. The top stage or cantilever is connected to a device that measures the force $F(t)$, either directly (SFA) or by converting the measured cantilever deflection (AFM), experienced by the body 1 during the motion. The experimental setup involves a careful alignment such that the collision is “head-on” and the system has rotational symmetry around the axis indicated in the figure. The distance $r$ is the polar radius measured from this axis. (For clarity, the coordinates system is shown shifted to the left of the symmetry axis, rather than with the $z$-axis coinciding with it.)

solved within a domain $0 \leq r \leq r_{max}$ subject to appropriate boundary conditions on the surfaces $z_{1,2}(r,t)$ of the two bodies, which define the film thickness as $h(r,t) := z_1(r,t) - z_2(r,t)$.

If the two surfaces are rigid, the lateral extent $r_{max}$ is determined from matching the solution for the flow inside the thin film with the macroscopic hydrodynamic flow around the two bodies [30]. (The case of small elastic deformations of the two rigid bodies has also been discussed in Refs. [19,20].) If one or both surfaces are soft, an additional condition, that of mechanical equilibrium for the shape of the fixed volume deformable body (the so called “augmented Young-Laplace equation” [41,42]), is necessary in order to determine the value of $r_{max}$. The analysis, in particular in the cases of deformable bodies, is quite involved. For the purpose of our paper the details of these derivations are not needed; therefore we will not review them here, but we encourage the interested reader to consult, e.g., the Refs. [19,20,30,41,42].

Under the assumption of incompressibility, the thickness $h := h(r,t)$ of the film obeys the
equation \[30,41,42\]
\[
\frac{\partial h}{\partial t} = -\frac{1}{r} \frac{\partial}{\partial r} \left( r \int_{0}^{h(r,t)} u \, dz \right).
\]

For two rigid bodies, this couples directly with the externally imposed motion of the stage or that of the cantilever and with the cantilever deflection \(D\) as \(\frac{\partial h}{\partial t} = -V(t) + \frac{dD}{dt}\) (positive deflection of cantilever is chosen to mean above its equilibrium position). For the case in which at least one of the bodies is deformable, the external motion and the cantilever deflection enter the description via a coupling condition for the thickness \(h(r_{\text{max}})\) at the edge of the interaction zone \[41,42\]. The total force acting on the body 1 and measured by the force measuring device is determined as \[42\]
\[
F(t) = 2\pi \int_{0}^{\infty} \left[ p(\xi, t) + \Pi(\xi, t) \right] \xi \, d\xi,
\]
where \(\Pi(\xi,t) := \Pi(h(\xi,t))\) denotes the so-called disjoining pressure (the negative of the derivative, in respect to the separation, of the excess free-energy of two flat interfaces being in close proximity to one another and the polar radius \(\xi\) is measured from the symmetry axis of the system. Note that Eq. (3) implicitly uses the proximity approximation in that the total contribution of the disjoining pressure is written as a linear superposition of contributions arising from the parts of film at each position \(\xi\) with the corresponding thicknesses \(h(\xi,t)\). Therefore once (i) the viscosity \(\mu_f\) of the intervening liquid, (ii) the hydrodynamic boundary conditions on the surfaces 1 and 2, and (iii) the expression of the disjoining pressure \(\Pi(h, t)\) are given, the force \(F(t)\) and the film's thickness \(h(r, t)\) in the interaction zone can be computed. The experimentally accessible quantities are \(F(t)\) and, in the SFA measurements, \(h(r, t)\), as functions of time or, as typically done for the AFM experiments, of the stage displacement \(\Delta X(t) = \int_{0}^{t} dt' V(t')\).

The experimental studies mentioned in the beginning of this section, together with the development of a theoretical background for the interpretation of the results, have significantly contributed to a better understanding of thin film drainage under nanoscale confinement. However, a number of aspects are yet to be clarified. In Sec. 2 we briefly review the theoretical modeling and experimental challenges faced by the thin film drainage studies with colloid probe AFM or SFA. We then critically discuss in Sec. 3 the outcomes of the recent experiments, including the issue of an unexpected no-slip boundary condition at a fluid-fluid interface which seems to be needed for a quantitative description when soft, deformable interfaces are involved. Finally, our conclusions and a brief outlook at future developments are presented in Sec. 4.

2. Intrinsic theoretical and experimental challenges.

When a comparison of the theoretical predictions for the time dependence of the force \(F(t)\) or of the shape \(h(r, t)\) of the film with the experimental data is attempted, a number of challenging issues for both the theory and the experiment occur. We discuss below those that we consider most relevant.

2.1. Theoretical challenges

In Eq. (1) the viscosity \(\mu_f\) of the liquid film influences the time scale of the film thinning (drainage rate). Once the film thickness is in the nanometer range, i.e., \(h(r, t) \lesssim 100\) nm, it is no longer a priori clear that the viscosity is the same as that of the bulk liquid, nor even that the assumption of a Newtonian liquid behavior remains valid. For molecularly thin films a stick-slip behavior and shear-thinning or shear-thickening transitions are well documented in
the literature (see, e.g., Refs. [9,13,26,27]). The analysis of the AFM and SFA thin films experiments thus must be complemented by independent measurements of the viscosity of the liquid in thin films, if the aim is to probe the validity of Stokes equations under nanoscale confinement or specific hydrodynamic boundary conditions.

The dependence of the total force on the disjoining pressure $\Pi(h)$, Eq. (3), requires that the various contributions, e.g., the electrostatic double layer and the van der Waals classic DLVO interactions, are known precisely in order for a quantitative comparison between theory and experiment to be possible. A quantitative calculation of the classic DLVO interactions remains a challenging point because: (i) the exact phenomenology of the charging of the interface, which determines the boundary conditions for the Poisson-Boltzmann equation, is generally unknown and at best accounted for by a so-called “charge regulation” boundary condition dependent on parameters to be determined by fitting experimental data (see Ref. [44] and references therein). For soft interfaces, for example that between a bubble and a salt solution, reliable values of these parameters are notoriously difficult to find in the literature (see, e.g., the discussion in Ref. [25]); (ii) the van der Waals interactions require precise knowledge, and are very sensitive to the details, of the dielectric permittivity functions of the two bodies and of the liquid. For water, for example, precise information is not yet available and various constructions of the dielectric permittivity function, giving rise to significantly different interactions, are a priori plausible (see Ref. [45]). Even for the most well-studied systems, those involving mica surfaces, the agreement between theoretical predictions and the experimental measurements of van der Waals interactions is seldom better than within 30% [46]; (iii) in the case when the liquid is a salt solution, which is the case in many of the experimental studies, it has been argued that a screening effect can occur even for the van der Waals interactions [52]. The exact form of the screening is presently under investigation, while only a few experimental studies of such effects have been reported, with so far rather inconclusive results [45,46,53]; (iv) a theoretical description of dynamic surface interactions, i.e., the time dependent equivalent of the DLVO theory, is yet to be developed. The validity of the quasi-static approximation, i.e., using for the disjoining pressure the equilibrium expression at the instantaneous film thickness $h(r,t)$, which is invoked in order to close the Eqs. (1)-(3), can be assessed only a posteriori. This is even more stringent in cases when the liquid separating the two bodies is actually a salt solution, because the convective transport of the ions in films of small thickness may disrupt the double layer charge distributions (which relax diffusively).

The hydrodynamic flow field $u(r,t)$ in Eq. (1) is determined subject to boundary conditions on the two surfaces. For a liquid-solid interface, the condition on the tangential velocity is postulated: either a no-slip boundary condition, i.e., $u \equiv 0$ at the surface of the solid, which is the generally accepted one for macroscopic hydrodynamics, or a Navier slip boundary condition, i.e., $u = \lambda du/dn$ at the surface of the solid, where $n$ denotes the direction normal to the surface of solid and oriented into the liquid, while $\lambda$ is the slip length (note that in the limit $\lambda \to 0$ the no-slip boundary condition is recovered). There exists no general prescription of how to calculate the slip length $\lambda$ for a given liquid-solid pair [54], and therefore $\lambda$ remains a fitting parameter for all the models of film drainage in AFM and SFA experiments. For a fluid-fluid interface, the hydrodynamic boundary conditions, also known as “mobile interface” boundary conditions, are the continuity of the tangential velocity and of the tangential shear stress (as well as continuity

Salt solutions are extensively used in experimental studies because of the possibility to control the range of the electrostatic interactions via the salt concentration. Unfortunately, they are also subject to potentially significant complications such as, e.g., the fact that the multivalent and the asymmetric electrolytes are poorly described by the mean-field Poisson-Boltzmann theory (see, e.g., Ref. [47] and the references therein). Moreover, the ion-specificity, i.e., the so-called “Hofmeister effect” (see, e.g., Ref. [48] and references therein) has been a puzzle for almost a century, and it is only recently that a consistent theoretical description for it seems to emerge [49-51].
of the normal velocity and the Laplace jump in pressure due to surface tension and curvature of the interface; they are determined (rather than being postulated) by the requirement that such interfaces cannot sustain any tangential stress. If a third species (e.g., surfactants) is present at a fluid-fluid interface, the interface can become “partially” or “completely” immobilized, i.e., the boundary conditions may change into one of partial or no slip [42]. Even in the absence of such surface active contamination an effective no-slip boundary condition may also apparently hold if the ratio of the viscosities of the two fluids satisfies certain conditions [42]. Therefore the choice of the hydrodynamic boundary conditions remains in most cases a parameter of the theoretical modeling.

2.2. Experimental challenges

The experimental challenges occurring when employing the colloid probe AFM technique in drainage studies have been thoroughly reviewed in the recent Ref. [17, 28], as well as in Refs. [29, 55]. Here we discuss only few of the experimental issues occurring in a typical thin-film drainage experiment.

Whenever solid surfaces are employed in the SFA or AFM studies, and in particular the spherical probes in the AFM, the issue of the surface roughness typically cannot be avoided. Except for the case of the mica sheets, which are atomically smooth (and also sufficiently elastic to be glued to SFA quartz lenses), for most of the other materials used as planar substrates or as colloid probes the surface is rough: (root-mean-squared) roughness of at least 0.5 nm and peak-to-valley values of at least a couple of nanometers. This has several implications, all of which make very difficult (if at all meaningful) a quantitative comparison between the experimental results and the theoretical predictions obtained under the assumption of smooth, flat surfaces [18, 56].

(i) Contact between such surfaces, thus the zero-separation point required for the linear-compliance calibration of an AFM, can be defined only in a statistical sense. (ii) Only an “effective” separation \( h(r, t) \), measured between the average (in respect to roughness) shape of the surfaces can be defined. Because the main interest is in film drainage at (effective) separations in the nanoscale range, e.g. ca 10 nm and below, even 1-2 nm peak to valley roughness can lead to significant (10% - 20%) differences between the separation in the experiment and the “effective” one employed in the theoretical modeling, both in Eq. (2) and in the definition of disjoining pressure. (iii) In the presence of rough surfaces it is not clear where the “effective” surface lies and what are the hydrodynamic boundary conditions which should apply there. The commonly taken approach is to apply a slip boundary condition at the position of the “average” surface [1, 18]; obviously, the slip-length occurring in such a boundary condition remains a parameter which is very difficult to connect with the microscopic details (peak-to-valley values, lateral correlation length of roughness features, etc.).

Moreover, when dissolved gas is present in the fluid (and this is the case of most of the experimental situations), nanoscale bubbles may nucleate in the valleys of the rough topography, giving rise to a heterogeneous surface partially covered by bubbles. In this case, Ref. [65] has shown that the slip length can be either positive, which intuitively means a smaller resistance to liquid drainage than that for a no-slip surface, or become negative, which would mean a larger resistance than that of a no-slip

4 We note here that recently a number of theoretical and numerical studies investigating the “effective” slip-length for model surfaces with well defined distributions of patches with different slip-lengths (usually two-values distributions) have been published. Significant progress has been made in this direction, including: calculations of the effective slip-lengths for flat surfaces with periodically distributed “stripe-like” patterns of given slip-lengths [54, 57], predictions and exact calculations of a tensorial slip boundary condition for anisotropic surfaces (e.g., containing topographical grooves, or stripes of varying slip-lengths) [58], exact bounds for the effective slip lengths for flows in thin-channels with textured walls [59], theoretical calculations and Lattice-Boltzmann simulations results for the effective slip lengths on model superhydrophobic surfaces [60–62] as well as for the drainage rate between a smooth sphere and model anisotropic or superhydrophobic flat (on average) surfaces [63, 64].
A rigorous calculation of the DLVO surface interactions between rough surfaces when the separation between the surfaces is of the order of the peak-to-valley roughness is not available yet.

As mentioned in Sec. 1, in the colloid probe AFM technique only the deflection of the cantilever is measurable, through a deflection of a reflected laser beam, from which the force is calculated based on a calibration of the elastic constant of the cantilever and of the voltage response of the photo-detectors. At small separations between the surfaces the forces become large (and have steep spatial gradients), and the accuracy of the measurement, which depends on the assumption of a linear response of the photo-detector, should be carefully considered. The deflection of the cantilever can be eventually converted into the central separation \( h(0, t) \) between (smooth) rigid surfaces based on the so-called "linear compliance regime". This requires the surfaces to be brought into real mechanical contact, and therefore can be applied only when the two surfaces are rigid. But for rough surfaces this procedure cannot lead to more than just an estimate, rather than an absolute value, for an effective separation; obviously, this is particularly important at very small effective separations (e.g., 10 nm or less). For soft, deformable surfaces, no information about the separation \( h(r, t) \) can be obtained with the present set-up of the colloid probe AFM. In contrast, the SFA apparatus allows for a direct (and independent) measurement of the separation \( h(r, t) \) between the two surfaces even when they are deformable by using fringes of equal chromatic order (FECO) interferometry. However, for solid surfaces which are rough the challenges listed for the AFM techniques are also relevant; additionally, the interpretation of the FECO interferometry spectra is no longer straightforward.

For colloid probe AFM, the motion of the cantilever through the liquid is subject to viscous friction and gives rise to an additional deflection. In general, this cannot be rigorously calculated because the shape and the material properties of the cantilever are too complex. Moreover, the interplay between length-scales: micrometer, for the length and thickness of cantilever and the diameter of the body 1, and nanometer for the film thickness separating the two bodies, makes a numerical calculation of the viscous friction on the cantilever a very difficult task.

Finally, contamination is an issue for both SFA and AFM experiments. For fluid-fluid interfaces, traces of surface active contaminants are sufficient to change the fully mobile character of the interface into a (partially) rigid-interface one (see Ref. [42] and references therein). For solid-liquid interfaces, insoluble nano-particles (e.g., dust) can adsorb on the solid surface; this gives rise to a spurious smaller value of the "measured" film thickness because the mechanical contact occurs on the contaminant particle rather than on the surface, a feature which by now is very well documented in the literature [17, 28, 66, 67]. The presence of dissolved gases and nucleation of nano-bubbles not only affects the hydrodynamic boundary conditions, but can also give rise to a van der Waals attraction between bubbles on the opposite surfaces [17, 28]. Such an effect is an example of the so-called "long-ranged hydrophobic attraction". This affects the value of the disjoining pressure in Eq. (3), but its occurrence cannot be a priori assessed and the magnitude of the effects cannot be exactly quantified.

3. Investigations of thin film drainage using the SFA and the colloid probe AFM

In view of the various challenges and issues briefly reviewed in the previous section we summarize here the results of the SFA and AFM studies in the last two decades, with a focus on their interpretation as a test for the hydrodynamic description of film drainage.

The early colloid probe AFM experiments employed sucrose solutions, silica probes and muscovite mica surfaces covered with layers of titania and gold, on top of which self-assembled monolayers of certain chemical reagents have been used to tune the "hydrophobicity" of the surfaces [16,68,69]. The strategy followed was to use high velocities of the AFM stage such that the hydrodynamic effects are significant and eventually dominant over the disjoining pressure contributions [see Eq. (3)]. The authors claimed to observe good agreement with the theoretical
predictions reported in Refs. [30] but this is achieved by fitting the experimental data with slip lengths that are dependent on the shear rate (presumed to be constant in an experiment at a given nominal approach speed of the AFM stage). This is unfortunately inconsistent with the theoretical model, a point clearly made in Refs. [15, 21, 29, 42]. Moreover, these experiments have been seriously questioned for a number of possible problems with the experimental set-up and artifacts. Problems include those of too soft cantilevers and contamination of surfaces by nano-particles, as identified and discussed by Refs. [15, 20, 21, 29, 70]; these also report for the same systems, as well as for other liquid and solids, and for similar dynamic conditions (fast motion of the AFM stage) a no-slip, or very small slip lengths of up to few nanometers, boundary conditions. Recently, these pioneering experiments and the experimental set-up have been thoroughly reviewed [17, 28]; additional reports of no-slip boundary conditions at hydrophilic surfaces and of various degrees of slip at hydrophobic or rough surfaces have been made [17, 28, 56]. In spite of the relatively large number of studies, the conclusions reached (slip or no slip) are often conflicting and the reported values for slip-lengths vary wildly between the various papers. Moreover, recently it has been observed that the shape of the cantilever actually influences the outcome of the experiment in terms of the type of boundary condition stemming out of the experimental data [55]; as correctly pointed out by Ref. [42], in the absence of a systematic study to understand the origin, and the exact magnitude, of the effect that the cantilever shape has on the outcome of the force measurements, this leaves doubt as to the reliability of all previous experiments.

The various SFA and colloid probe AFM experiments on thin film drainage between soft, deformable bounding surfaces, as well as experiments involving different experimental techniques, have been generally performed using approach velocities for which both the hydrodynamic pressure and the disjoining pressure contributions are relevant; these have been recently reviewed by Refs. [41, 42], with a view focused on the theoretical modeling. We therefore discuss here only few specific points and encourage the interested reader to consult these reviews and the extensive list of references included there. The most complete test of the theoretical models is provided by the SFA experiments, for both drainage and filling (i.e., bounding surfaces moving towards or away from each other, respectively) with drops and bubbles of Horn et al (see, e.g., Refs. [12, 31–33, 42] and the references therein) because they provide the whole thickness profile $h(r, t)$ as a function of time. According to Ref. [42], the theoretical modeling based on the Eqs. (1) - (3) and the augmented Young-Laplace equation (see Sec. 1) captured very well the qualitative features observed in the experiment, in particular: the occurrence of “dimple” and “wimple” shapes (i.e., regions of curvature with opposite signs) in the interaction zones, as well as the time scales and approach velocities over which these instabilities develop and relax, down to film thicknesses (minimum of $h(r, t)$) in the interaction zone in the order of 50 nm. In view of the complexity of the experimental results, these findings provide a strong argument that the hydrodynamic description reviewed in Refs. [41, 42] seems to capture the essential physics.

Furthermore, it has been shown that the same model can lead to predictions in quantitative agreement with the experimental results for: $h(r, t)$ in the SFA setup and the approach and retract force curves $\{\Delta X(t), F(t)\}$ in the AFM set-up. But this is achieved only if a no-slip boundary condition is invoked at the fluid-fluid interface(s). These findings hold for various configurations of the bodies 1 and 2: bubble-plate, drop-plate, drop-drop, or bubble-bubble, as well as for various choices of the intervening liquid (see, e.g., the review in Ref. [42]). This is very surprising. For example, by using as an estimate the criteria expressed in Ref. [42] (p. 5 We note, however, that Ref. [25] mentions that for quantitative agreement to be achieved for a few parameters slightly different values than the experimentally measured ones had to be used. It is unfortunately difficult to assess if this was also the case for other experiments because in many cases – see, e.g., Ref. [23] – no complete information is given about the values of the parameters, for example the spring constant of the cantilever, used in the theoretical modeling and experimentally measured.
for an air bubble - water interface, the viscosities of the two fluids, the radius of the bubble in the experiment, and the film thickness in the range of tens of nanometers would argue in favor of a fully mobile interface. The most frequently invoked argument for such a no-slip boundary condition is that of surface active contaminants (see, e.g., Ref. [23]) which, even in minute amounts, would be sufficient to partially or fully immobilize the interface. However, this is unlikely to be the case in Refs. [12, 25, 31, 33], where explicit reference is made to significant efforts made to ensure the cleanness of the system; for example, for the bubble approaching a titania plate studied in Ref. [25] the presence of contaminants would likely make the titania surface hydrophobic and lead to the film rupture, which is not what is observed experimentally. The fact that the experimental conditions indeed can be controlled to ensure the absence of surface active contaminants is further supported by the reports of mobile boundary conditions for bubbles in water and aqueous electrolytes [6]. Moreover, a mobile boundary condition at a clean air-water interface has also been clearly evidenced independently in Ref. [71], which used a different experimental technique (the motion of optically trapped colloids near the air-water interface). The recent arguments that a no-slip boundary condition is needed to obtain quantitative agreement with the results of experimental bubble-rise studies of micro air-bubbles colliding with a titania plate [42, 72] are also debatable. At closer inspection, some of the experimental data for the time dependence of the separation between the apex of the bubble and the titania plate, determined from the intensity of an interference pattern, show “jumps”, i.e., non-physical discontinuities (see, e.g., the curves (b) and (c) in Fig. 5 of Ref. [72]). It is possible to identify the source of these as an incorrect choice of the corresponding interference order. After correcting the experimental data for these spurious discontinuities (starting from the long time region of the curve, as this corresponds to the minimum separation and thus a clearly defined order of interference), the experimental results (b) and (c) mentioned above would lay significantly below the theoretical predictions shown there, indicative of a faster drainage and thus of a mobile (at least partially) interface, rather than of the claimed no-slip condition.

In the light of these observations, the necessity for a no slip boundary condition at a fluid-fluid interface to quantitatively explain the SFA and AFM film drainage experiments remains questionable and it seems that at this time a consistent explanation for its source is yet to be found.

4. Conclusions and outlook
We have discussed a number of theoretical and experimental challenges in the SFA and AFM studies of thin film drainage and have critically reviewed the outcomes of recent (last two decades) experimental studies. While significant progress has been made, it seems clear that the results so far cannot yet overcome all the issues outlined in Sec. 2. Therefore, we argue that conclusive SFA or AFM tests concerning the validity or otherwise of a hydrodynamic description for the drainage of liquid films with a thickness below tens of nanometers are yet to be provided. Although the theoretical modeling is greatly simplified for rigid surfaces, it is clear that the issue of a nano-rough surface reduces the likelihood of such tests being successful when solid surfaces are involved. Therefore, the most promising direction appears to be that of studying the case of deformable bounding surfaces pursued so far in, e.g., Refs. [22–25, 31, 33] (see also the reviews in Refs. [41, 42]. The continuing development of an interferometry enhanced AFM [73], which would allow a determination of the separation $h(r, t)$ independently of the cantilever deflection (similarly to the situation in the SFA experiments), could significantly contribute in this endeavor.

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