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Theory of non-equilibrium force measurements involving deformable drops and bubbles

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ABSTRACT

Over the past decade, direct force measurements using the Atomic Force Microscope (AFM) have been extended to study non-equilibrium interactions. Perhaps the more scientifically interesting and technically challenging of such studies involved deformable drops and bubbles in relative motion. The scientific interest stems from the rich complexity that arises from the combination of separation dependent surface forces such as Van der Waals, electrical double layer and steric interactions with velocity dependent forces from hydrodynamic interactions. Moreover the effects of these forces also depend on the deformations of the surfaces of the drops and bubbles that alter local conditions on the nanometer scale, with deformations that can extend over micrometers. Because of incompressibility, effects of such deformations are strongly influenced by small changes of the sizes of the drops and bubbles that may be in the millimeter range. Our focus is on interactions between emulsion drops and bubbles at around 100 μm size range. At the typical velocities in dynamic force measurements with the AFM which span the range of Brownian velocities of such emulsions, the ratio of hydrodynamic force to surface tension force, as characterized by the capillary number, is ~10^{-6} or smaller, which poses challenges to modeling using direct numerical simulations. However, the qualitative and quantitative features of the dynamic forces between interacting drops and bubbles are sensitive to the detailed space and time-dependent deformations. It is this dynamic coupling between forces and deformations that requires a detailed quantitative theoretical framework to help interpret experimental measurements. Theories that do not treat forces and deformations in a consistent way simply will not have much predictive power. The technical challenges of undertaking force measurements are substantial. These range from generating drop and bubble of the appropriate size range to controlling the physicochemical environment to finding the optimal and quantifiable way to place and secure the drops and bubbles in the AFM to make reproducible measurements. It is perhaps no surprise that it is only recently that direct measurements of non-equilibrium forces between two drops or two bubbles colliding in a controlled manner have been possible. This review covers the development of a consistent theory to describe non-equilibrium force measurements involving deformable drops and bubbles. Predictions of this model are also tested on dynamic film drainage experiments involving deformable drops and bubbles that use very different techniques to the AFM to demonstrate that it is capable of providing accurate quantitative predictions of both dynamic forces and dynamic deformations. In the low capillary number regime of interest, we observe that the dynamic behavior of all experimental results reviewed here are consistent with the tangentially immobile hydrodynamic boundary condition at liquid–liquid or liquid–gas interfaces. The most likely explanation for this observation is the presence of trace amounts of surface-active species that are responsible for arresting interfacial flow.

Contents

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1. Introduction

1.1. Background and motivations

Studies of non-equilibrium interactions involving deformable drops and bubbles predated the formulation of the Derjaguin–Landau–Verwey–Overbeek theory of colloidal stability [1,2] with the studies of Derjaguin and Kussakov [3] on time-dependent behavior of a rising bubble towards a flat plate under buoyancy force. Subsequent non-equilibrium studies concentrated on the drainage phenomena of the liquid film between deformable menisci [4].

Early direct measurements of non-equilibrium forces were based on the Surface Forces Apparatus to measure the time-dependent approach between two cross-cylinders of mica down to nanometer separations in aqueous [5] and non-aqueous liquids [6]. Forces under conditions of steady state oscillations of the mica surfaces were also studied in the context of examining the possible variations in fluid viscosities of nanometer thick confined liquid films [7] and the lubricating properties of adsorbed polymers [8].

With the advent of the atomic force microscope, interest continued in the hydrodynamic interaction involving solid spheres in the tens of micrometer size range. Although much interest was generated by reports of hydrodynamic boundary slip at the solid–liquid interface [9], particularly in the context of microfluidic applications [10], recent repeated measurements suggest that instrumental artifacts are likely to be responsible for such observations at smooth well defined surfaces [11–13].

In the first applications of the atomic force microscope to measure equilibrium forces involving deformable bubbles, the deformational response of the bubble was treated as a Hookean spring [14,15]. In subsequent equilibrium studies involving drops, the Young–Laplace equation was used to account for the drop deformational behavior [16,17].

In considering non-equilibrium interactions, the time-dependent force between, for instance, two approaching deformable drops at any instant, does not only depend on the instantaneous shapes and separation between the drops, but also on initial conditions that determine the drop shape and interfacial velocities. In addition, flow of the continuous fluid phase also contributes to the hydrodynamic interaction. Therefore appropriate experimental data needs to be recorded to provide initial and boundary conditions for theoretical analysis.