Abstract

In my thesis, I study the dynamics of a liquid interface in contact with a solid substrate via large-scale molecular dynamics (MD) simulations.

The first part is devoted to the validation of the simulation tools through equilibrium and dynamic wetting studies. MD simulations of liquid drops spreading on a solid substrate are carried out for a very wide range of solid-liquid interactions and equilibrium contact angles. The results for these systems are shown to be consistent with published results. They show that the molecular-kinetic theory (MKT) for dynamic wetting, which emphasizes the role of contact-line friction as the principal channel of energy dissipation, works well for our simple system and therefore has some underlying validity at a fundamental level. Several predictions of this theory are confirmed by our simulations. These include a quantitative link between the dynamics of wetting and the work of adhesion and the existence of an optimum equilibrium contact angle that maximizes the speed of wetting. A feature of the new work is that key microscopic parameters (κ^0 and λ, the jump frequency and the distance of jump, respectively), normally accessible only by fitting the MKT to dynamic contact angle data, are also obtained directly from the simulations, with good agreement between the two sources. This validates the MKT at some fundamental level. Further verification is provided by interfacial relaxation studies, which also lend support to the interfacial tensions relaxation process invoked in Shikhmurzaev's model for dynamic wetting.

We then use our simulation tools to study shear in a Couette flow geometry, allowing a large-scale study of slip between the same liquid and solid as in the wetting case. This leads to a direct measure of the slip length L_S which is a crucial characteristic in slip phenomena. These results can be explained and quantitatively predicted by the generalization of the MKT model that we have developed in collaboration with T. Blake, involving the same microscopic characteristics as in the wetting case. Additional equilibrium simulations of diffusion rates at the solid-liquid interface have shown the relevance of the solid-liquid friction coefficient.

The last part is devoted to the study of dewetting dynamics of a thin liquid film. MD simulations are carried out to investigate the effects of wettability, film thickness and dewetting front geometry. As observed experimentally and in previous simulations, the films recede at an initially constant speed, creating a growing rim of liquid with a constant receding dynamic contact angle. Consistent with the current understanding of wetting dynamics, film recession is faster on the more poorly wetted surface. These results can be explained and quantitatively predicted by incorporating the contact-line friction coefficient of the MKT model into the classical dewetting dynamics model which does not account for this energy dissipation channel at the dewetting front. It is observed that, for the thinnest films, the rates of recession increase with decreasing film thickness. A possible interpretation is that the effective viscosity of the film is decreasing with thickness.

This work contributes to a more general and unified understanding of the properties of a liquid interface on a solid surface.