Surface Light Scattering Close to the Critical Damping of Surface Fluctuations and in the Presence of Molecular Orientation Effects

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Surface light scattering (SLS) represents a contactless method for the absolute determination of viscosity and surface or interfacial tension of fluids in macroscopic thermodynamic equilibrium. The technique probes the dynamics of surface fluctuations which can be accessed by the analysis of the light scattered from the phase boundary. Significant progress has been made in recent decades in the application of SLS for thermophysical property research, but challenges remain.

One of these challenges is related to SLS experiments performed close to the critical damping of surface fluctuations where their dynamics change from an overdamped to an oscillatory behavior. Here, the evaluation of SLS signals is complicated by influences from the rotational flow in the bulk of the fluid close to the phase boundary. The present contribution shows a new evaluation approach which considers the rotational flow in a physically meaningful way. Its application to SLS signals recorded for several vapor-liquid systems near the critical damping demonstrates the success of this evaluation strategy for an accurate determination of viscosity and surface tension.

Another challenge is associated with the presence of surfactant monolayers at vapor-liquid interfaces, which induces a viscoelastic behavior affecting the dynamics of surface fluctuations. By combining SLS experiments with conventional viscometry and tensiometry as well as molecular dynamics simulations, it is revealed that already weakly asymmetric molecules can induce viscoelastic behavior. This is shown as an example for the liquid organic hydrogen carrier system based on diphenylmethane, in which the corresponding reaction intermediate cyclohexylphenylmethane can act as a surfactant. Its influence on the dynamics of surface fluctuations varies with increasing temperature, which reflects the change of the molecular orientation with respect to the interface from a preferentially perpendicular to a parallel alignment. Here, the combination of SLS and conventional measurement techniques allows us to deduce the viscoelastic properties of the surfactant monolayer.