Simultaneous Analysis of Equilibrium Fluctuations at the Surface and in the Bulk of a Binary Liquid Mixture by Dynamic Light Scattering

Thomas M. Koller and Tobias Klein

Institute of Advanced Optical Technologies - Thermophysical Properties (AOT-TP), Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Erlangen, Bavaria, Germany

> Jiaqi Chen and Ahmad Kalantar Shell Global Solutions International B.V., Amsterdam, Netherlands

> > Michael H. Rausch

Institute of Advanced Optical Technologies - Thermophysical Properties (AOT-TP), Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Erlangen, Bavaria, Germany

Alfred Leipertz

Institute of Engineering Thermodynamics (LTT), Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Erlangen, Bavaria, Germany

Andreas P. Fröba^{C, S}

Institute of Advanced Optical Technologies - Thermophysical Properties (AOT-TP), Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Erlangen, Bavaria, Germany andreas.p.froeba@fau.de

Dynamic light scattering (DLS) analyzes microscopic statistical fluctuations present in fluid systems in macroscopic thermodynamic equilibrium to determine corresponding macroscopic thermophysical properties. For light scattering from bulk of fluids, the relaxation of temperature and concentration fluctuations can be studied to access thermal diffusivity and mutual diffusivity. By applying DLS to fluid interfaces, also called surface light scattering (SLS), the relaxation of surface fluctuations can be probed to measure viscosity and surface or interfacial tension. Using two individual setups specifically designed for either light scattering experiment, expanded uncertainties in the various properties in the low percentage level are achievable. Until now, it has not yet been investigated whether surface and bulk fluctuations can be studied simultaneously by DLS within a single experimental setup or even within a single experimental run in a quantitative way. In the present contribution, we demonstrate that it is possible to simultaneously analyze microscopic fluctuations at the surface and in the bulk of a binary liquid mixture by DLS in macroscopic thermodynamic equilibrium. By analysis of light scattered in the region of the vapor-liquid interface of a model system containing n-octacosane and ethanol, three individual light scattering signals could be found on distinctly different time ranges. One oscillatory signal from surface fluctuations at the vapor-liquid interface in the short-time range and two exponential Rayleigh signals from fluctuations in temperature and concentration in the bulk of fluid in the long-time range could be associated with hydrodynamic modes. This microscopic information allows for a simultaneous determination of the macroscopic properties interfacial tension, kinematic viscosity, thermal diffusivity, and mutual diffusivity within a single experimental run probing the very same sample at a welldefined thermodynamic state. Thus, the presented approach represents a worthwhile strategy for an effective multiproperty determination of fluid systems.