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10th Anniversary Highlights



Ultralow interfacial tension driven demixing dynamics

D G A L Aarts¹, R P A Dullens¹ and H N W Lekkerkerker²

¹ Physical and Theoretical Chemistry Laboratory, University of Oxford, UK² Van't Hoff Laboratory for Physical and Colloid Chemistry, University of Utrecht, The Netherlands

'Interfacial dynamics in demixing systems with ultralow interfacial tension' D G A L Aarts et al 2005 New Journal of Physics 7 40



LSCM images (1400 x 1400 μ m²) of a phase separating colloid–polymer mixture. Gravity points downwards (in all images). Directly after homogenization a spinodal structure is observed, which immediately coarsens in time. It is evident that coarsening occurs through pinch-off instead of coalescence and one can follow such events in time; the insets (109 x 109 μ m²) zoom in on the pinch-off event (from left to right, *t* = 29, 33, 37 s). The spinodal structure is collapsing under its own weight leading to strong directional flow. After several minutes the macroscopic interface is being formed.

Whenever one tries to mix water and oil, one observes that the system quickly phase separates. One of the driving terms of this demixing is the interfacial tension, which leads to considerable velocities in the phase separation process. In order to study this process in more detail we resort to colloid–polymer mixtures.

Mixtures of colloids and polymers display rich phase behaviour, involving colloidal gas (rich in polymer, poor in colloid) and colloidal liquid (poor in polymer, rich in colloid) phases. Due to the colloidal lengthscale the interfacial tension is much lower than in the molecular case (nN m^{-1} instead of mN m^{-1}). Consequently, the demixing can be followed in great detail.

Using laser scanning confocal microscopy clear and well-defined images are obtained, which makes quantitative comparisons with theory possible, and is highly instructive. Simple scaling arguments can be given as to why in a single experiment three steps of the phase separation can be observed: an interfacial-tension-driven coarsening, gravity-driven flow and finally the interface formation. The first stage can be quantitatively described by viscous hydrodynamics. Coarsening occurs through pinch-off events. The second stage is reminiscent of the Rayleigh–Taylor instability. The liquid phase breaks up and becomes discontinuous. There is strong directional flow in the system, but the Reynolds number remains much smaller than unity. Finally, the macroscopic interface is formed, growing upwards, with a velocity comparable to the coarsening velocity in the initial stage. Recent developments in both confocal microscopy and colloidal model systems now make it possible to study these demixing processes even at a particle level.

Biomolecular dynamics in model biological membranes

M Smits 1 , A Ghosh 1,2 , J Bredenbeck 1 , S Yamamoto 3 , M Müller 4 and M Bonn 1,2

- ¹ FOM Institute for Atomic and Molecular Physics, Amsterdam, The Netherlands
- ² Leiden Institute of Chemistry, Leiden University, The Netherlands

³ Stanford Synchrotron Radiation Laboratory, CA, USA

 $^{\rm 4}$ Swammerdam Institute for Life Sciences, University of Amsterdam, The Netherlands

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Cell membranes both act as passive barriers that separate the inside and outside of the cell, and play an active role in physiological functions such as intercellular signaling and the transport of small molecules to and from the environment. Molecularly, the membrane is primarily composed of lipids, proteins and their surrounding water. Membrane function thus involves the interplay of these components. Molecular dynamics simulations have shown that this interplay evolves over sub-nanosecond timescales but experimental insights into the dynamics of these interactions have lagged. This lag is the result of the considerable challenge of investigating the single layer of molecules that compose the membrane with the required time resolution. In our article we overcome this challenge by using femtosecond laser spectroscopy to follow the evolution of a biomolecular system in real-time after a laser-pulse induced change (e.g. a temperature jump or vibrational excitation). The strength of our approach lies in probing the sample using an even-order nonlinear optical process that has inherent interfacial sensitivity but maintains the time resolution of simpler bulk techniques. Our study is the first surface-specific study of vibrational relaxation and energy transfer in lipid monolayers prepared over a water subphase. This model membrane allows us to easily control parameters such as surface pressure and lipid phase. Generally we find remarkably fast dynamics: heat transfer across the monolayer, for instance, occurs on picosecond timescales. This study demonstrates the potential of using ultrafast surface-specific spectroscopies to elucidate biomolecular dynamics in membranes on the timescale over which they occur.



Schematic of the experiment: a self-assembled monolayer of lipid molecules (with polar headgroups shown in yellow and apolar alkyl chains shown in grey) on water. Laser pulses at different wavelengths are incident on the lipid monolayer. One pulse triggers a temperature jump or vibrational excitation in the monolayer, and a delayed pair of pulses interrogates the transient changes in the monolayer, in a process where the sum-frequency of the incident pair (shown here as red and green) is generated (blue beam). This technique provides surface-specific information on model membrane relaxation processes.