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Bilateral International Exchange Program (BIEP, invite) report
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Research Project

Title	Dissipative Structure Formation in Surfactant Films
Duration	2 months

During my stay at Kyoto University I had the opportunity to talk to many scientists. Besides discussing with several group members of the laboratory of Prof. Yoshikawa, I met twice with Prof. Ohta and Prof. Araki. Also the many guests to the laboratory of Prof. Yoshikawa provided valuable input to my research. In most of the discussions the starting point was the question: "What could be the physical origin of the separation observed in my experiments?" To summarize all the discussions, I could classify the ideas into three scenarios:

1.) The separation is driven by the underlying subphase which exhibits a pattern on the same length scale as observed on the transferred monolayer along the contact line.

2.) Upon transfer the conformal space of the polymers is reduced leading to a loss in entropy which drives them towards each other as being solved in water is less preferential.

3.) There exists a difference in wetting towards the surface between the lipopolymers and the lipids. This leads to an instability of the contact line with the observed spatial pattern.

From the hydrodynamics of the experimental geometry there is no need of a flow in the direction along the contact line and has never been considered. Furthermore, experiments with a different polymer (mPEG) did not show the observed long range pattern, rendering the first scenario unlikely.

The second explanation leads to an expected dependency on the size of the polymers. This has not been observed in our experiments and is considered unlikely therefore as well.

The third scenario would modify the contact line spatially. For a higher affinity to the substrate the contact angle is expected to be smaller. From fluorescence microscopy experiments on the three-phase-contact line an upper boundary of several microns for these modulations can be given. To validate this idea, measurements of the contact angle of the pure lipid and the pure lipopolymer have to be carried out. The difference in contact angle can then be directly related to the wetting properties.

Beside these discussions I started modeling the spatial structure using the Cahn-Hilliard equation. The Cahn-Hilliard equation is known to describe many phase-separation processes and leads to periodic structures for certain sets of parameters. To account for the effect of transfer speed v I used the fact that the second derivative of the free energy has dimensions of speed squared (times mass). As the mixture is homogeneous at the air/water interface, the free energy was modeled to increase proportional to the transfer speed squared v^2 starting from zero and reaching the same final value for all transfer speeds. This resulted in a dependency of the spatial structure being proportional to $t^{1/3}$. This matches well with the observation from the experiments.

Further experiments dealt with the variation in subphase viscosity η. The relationship to the diffusion constant of the lipopolymer D considered in the Cahn-Hilliard equation can be thought of being roughly antiproportional $\eta \propto D^{-1}$. Varying the diffusion constant D linearly in the simulations thus led to a linearly increasing spatial structure with decreasing η. The data, both experimentally and numerically can be collapsed onto one curve by plotting against $\eta^3 v$.

The theoretical findings outlined above will be accompanying the experimental observations in a publication I will start to prepare after returning back.