KELVIN-HELMHOLTZ INSTABILITY OF LANGMUIR MONOLAYERS

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Although Langmuir monolayers exhibit various important class of two-dimensional phenomena, their fluctuation and morphology into third dimension have also attracted great interests both theoretically and experimentally. For instance, Milner *et al.* predicted a buckling of Langmuir monolayers in a fluid phase under a lateral compression which decreases the surface tension to $\gamma = \gamma_0 - \Pi$, where γ_0 is a surface tension of pure water and Π is a two-dimensional surface pressure due to the compression [1]. In a real system, however, buckling has not been observed in fluid monolayers and they collapse to form multilayers even at positive tensions ($\gamma_0 > \Pi$) upon increasing the surface pressure.

In this article, we discuss the possibility of a dynamical instability of Langmuir monolayers. This instability is generally known as "Kelvin-Helmholtz instability" which occurs when two superposed fluids flow one over the other with a relative horizontal velocity. For Langmuir monolayers, above a certain critical value of the velocity of the air relative to the water, the Kelvin-Helmholtz instability will arise before the static buckling transition takes place.

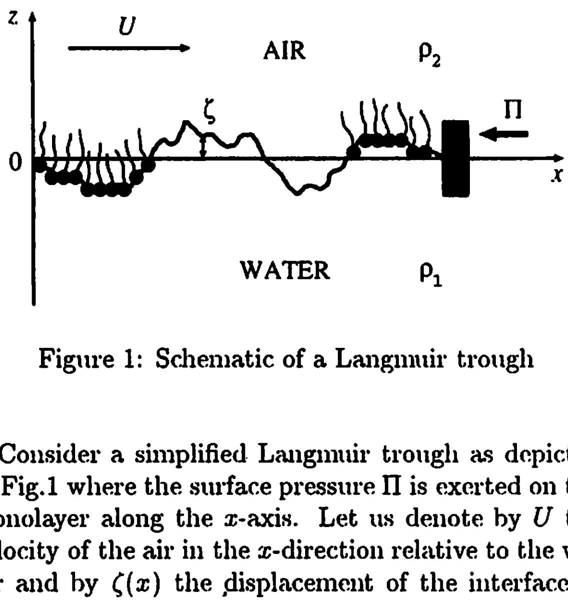

Figure 1: Schematic of a Langmuir trough

Consider a simplified Langmuir trough as depicted in Fig.1 where the surface pressure Π is exerted on the monolayer along the x-axis. Let us denote by U the velocity of the air in the x-direction relative to the water and by $\zeta(x)$ the displacement of the interface in the z-direction. We describe here both the air and the water as incompressible inviscid fluids. For the force balance condition at the interface, we use the deformation energy of the monolayer given by the well-known Helfrich Hamiltonian:

$$F = \gamma \int dA + \frac{\kappa}{2} \int (c_1 + c_2 - c_0)^2 dA. \qquad (1)$$

In the above, dA is the surface element, κ is the bending rigidity, c_1 and c_2 are two principle curvatures, and c_0 is the spontaneous curvature. We look for the displacement ζ in the periodic form $\zeta = a\sin(kx - \omega t)$. After some calculation, we obtain the dispersion relation given by

$$\omega = \frac{k\rho_2 U}{\rho_1 + \rho_2} \pm \sqrt{\frac{kf(k)}{\rho_1 + \rho_2}}, \qquad (2)$$

$$f(k) = \kappa k^4 + \left(\gamma + \frac{\kappa c_0^2}{2}\right) k^2 - \tilde{\rho} U^2 k + \Delta\rho g, \qquad (3)$$

where $\tilde{\rho} = \rho_1\rho_2/(\rho_1 + \rho_2)$ and $\Delta\rho = \rho_1 - \rho_2$. Here ρ_1 and ρ_2 are the density of water and air, respectively, and g is the acceleration of gravity. If ω is a complex number with a positive imaginary part, the motion becomes unstable and this is called as Kevin-Helmholtz instability. Hereafter we shall only discuss the case of vanishing spontaneous curvature ($c_0 = 0$) just for the simplicity.

Results for several special cases can be deduced from Eq. (2). First, when $U = 0$ and if the surface tension satisfies $\gamma < \gamma_c = \gamma_0 - \Pi_c = -2(\kappa\Delta\rho g)^{1/2}$, $f(k)$ can be negative for a certain range of the wave vector k. The most unstable mode is given by $k_c = (\Delta\rho g/\kappa)^{1/4}$ in this case. This situation exactly corresponds to the buckling instability predicted by Milner *et al.* [1]. When there are no amphiphilic molecules, the interfacial property is solely determined by the surface tension of water, γ_0. In this case, $f(k)$ can be negative provided $U > U_{c1} = (4\gamma_0\Delta\rho g/\tilde{\rho}^2)^{1/4}$, and the most unstable mode is $k_{c1} = (\Delta\rho g/\gamma_0)^{1/2}$. Typical values for air over water ($\rho_1 \sim 1$ g/cm^3, $\rho_2 \sim 10^{-3}$ g/cm^3, $\gamma_0 \sim 74$ dyn/cm) provide $U_{c1} \sim 730$ cm/s and $k_{c1} \sim 3.6$ cm^{-1} ($2\pi/k_{c1} \sim 1.7$ cm). As the surface pressure Π is increased towards γ_0 in the presence of adsorbed amphiphilic molecules, it turns that the interface is mainly governed by the bending rigidity κ. Here we consider the case for $\gamma = 0$ which is still larger than γ_c at which the static buckling takes place. In this case, the critical velocity is $U_{c2} = (2/3^{3/8})\kappa^{1/8}\tilde{\rho}^{-1/2}(\Delta\rho g)^{3/8}$, and the most unstable mode is $k_{c2} = (\Delta\rho g/3\kappa)^{1/4} = k_c/3^{1/4}$. With the same orders of magnitude as used before, and $\kappa \sim 10^{-13}$ erg, we find that $U_{c2} \sim 13$ cm/s and $k_{c2} \sim 7.6 \times 10^3$ cm^{-1} ($2\pi/k_{c2} \sim 8 \times 10^4$ Å). Comparing with the pure water surface case, we see $U_{c1} \gg U_{c2}$ and $k_{c1} \ll k_{c2}$ indicating a dramatic effect of adsorbed amphiphilic molecules on the surface property. One can expect a crossover of these critical values by changing the surface pressure Π. Details are given in Ref.[2].

References

[1] S. T. Milner, J.-F. Joanny, and P. Pincus, Europhys. Lett. 9, 495 (1989). [2] S. Komura and T. Iwayama, to be published in J. Phys. II France (1997).