SUPPRESSION OF TURBULENT DIFFUSION ON THE WATER SURFACE BY VISCOELASTIC NANO LAYER.

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Abstract We study effects of thin (10 nm) layers of adsorbed proteins on the water surface hydrodynamics. We show that extremely small concentrations of protein (less than 1 ppm) form strong viscoelastic layer at the water-air interface. This layer greatly reduces single particle dispersion on the surface perturbed by Faraday waves and turns disordered surface waves into a square stationary oscillating crystal. The viscoelastic film is destroyed by minute addition of surfactant which leads to the recovery of the horizontal mobility of fluid particles and the restoration of the Faraday wave driven turbulence.

It was recently found that parametrically excited Faraday waves in vertically vibrated containers inject vorticity into the horizontal flow (vertical vorticity) [1]. These horizontal vortices interact and spread energy over a broad range of scales eventually leading to the development of two-dimensional (2D) turbulence [2, 3, 4]. Single particle dispersion in the Faraday wave driven turbulence is found to be Brownian at long times [4, 5]. This coincides with the observation of disordered wave filed. However when microscopic concentrations of solvable protein, *bovine serum albumin* (BSA) are added to water, the wave fields turn into perfect oscillating crystals [6]. Fig. 1 shows horizontal *(x-y)* trajectories of tracer particles at the fluid surface filmed for over 30 periods of the Faraday waves on the surface of water with added non-ionic surfactants (a) and water with added 5 ppm (in mass) of BSA (b). A very thin layer of the adsorbed BSA stops the fluid particle motion.

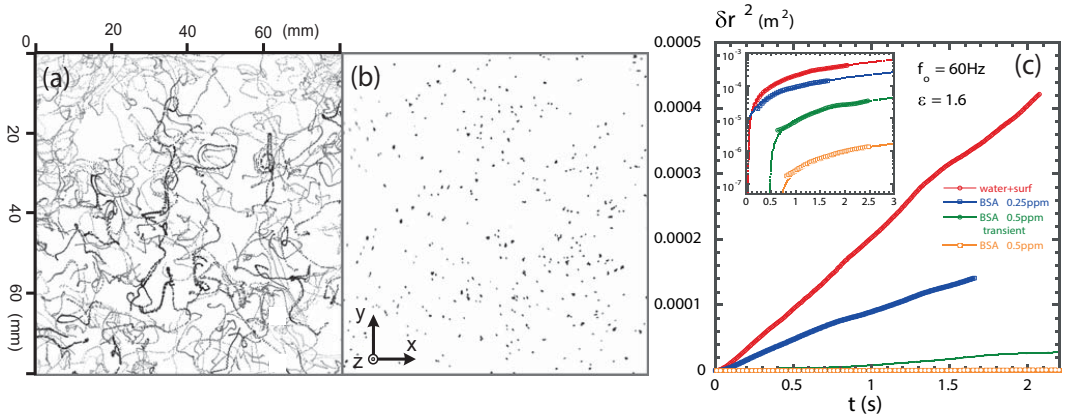

Figure 1. Trajectories of the fluid particles on the water surface perturbed by Faraday waves in case of (a) water with added surfactant, and (b) water with added 5 ppm of BSA. Particles (50 μm diameter polyamide particles) are filmed for 1 s (30 Faraday wave periods). (c) Mean squared displacement as a function time t at different concentrations of BSA in water. Inset: same data in the log-lin scale. The solid lines show linear fitting.

Now we consider single particle dispersion $\langle \delta r^2 \rangle = \langle \, |\vec{r}(t) - \vec{r}(0)|^2 \rangle$ at the Faraday wave surface, where $\vec{r}(t)$ is the horizontal position vector of a fluid particle at time t. If Faraday waves are formed in water with added surfactant, surface particles move chaotically showing Brownian-type diffusion at long times, $\langle \delta r^2 \rangle = 2Dt$ [7]. Figure 1(c) shows the effect of BSA on $\langle \delta r^2 \rangle$ for various concentrations c_{BSA}. The vertical acceleration a in these experiments is fixed such that the super criticality is $\epsilon = 1.6$. Reduction of the particle mobility is observed at the BSA concentrations as low as 0.25 ppm. The diffusion coefficient D at this concentration is reduced by almost a factor of 3 in comparison with water-surfactant case, Fig. 1(c). At 0.5 ppm and higher concentrations, D drops dramatically, by several orders of magnitude.

To investigate the reason for such a strong suppression of the particle motion in the presence of the BSA we visualise 3D particle trajectories using a recently developed method which combines the diffusive light imaging method with the 2D particle tracking velocimetry technique [1, 8]. Figs. 2(a, b) show several trajectories corresponding to the case of Fig. 1(a,b). In water with added surfactant, particles move chaotically generating turbulence on the surface. Trajectories represent complex curves such that particles wander erratically in a succession of "traps" and "flights". Trajectories become dramatically different if microscopic quantities of BSA are added. In this case *all* trajectories on the water surface [red in Figs. 2(b,d)] are straight vertical lines. However particles 0.25 mm *below* the surface [green trajectories in Figs. 2(b,d)] show strictly vertical motion near the wave crests, and a pendulum type oscillations near the wave nodes. These sub-surface particles stay confined within the wave period.

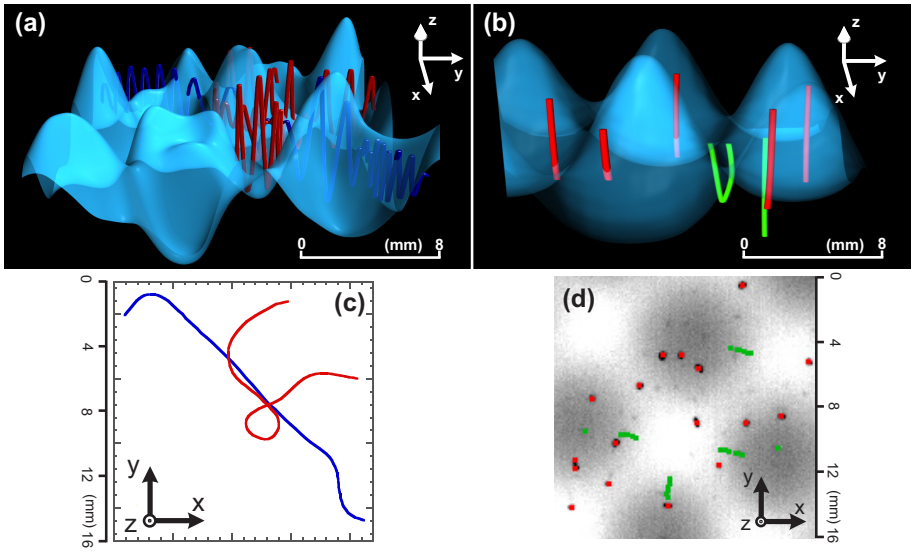

Figure 2. 3D particle trajectories of the fluid particles on the surface of water (a) with added surfactant, and (b) water with 5 ppm of BSA. Red trajectories are for particles moving on the surface, while the green trajectories correspond to the particles 0.25 mm *below* the surface. (c) A top view of the trajectories shown in (a). (d) A top view of the trajectories shown in (b) (some trajectories in (b) are discarded to improve visibility). The black and white blobs in the background correspond to the wave crests and troughs respectively. Red dots represent surface particles while the green dots represent particles 0.25 mm *below* the surface.

In 2D standing waves particles perform the pendulum-type motion near the wave nodes, while near the wave trough/crest particles oscillate vertically. Similar trajectories have been measured in 3D Faraday wave crystal formed in highly viscous Newtonian liquid (glycerol+water solution) [1]. This pendulum-like motion is a signature of the incompressibility of the liquid. In contrast, in the presence of BSA, only the particles *below* the surface show a motion similar to that in viscous Newtonian liquid. All surface particles oscillate strictly vertically regardless of their position Fig. 2(b). Such anomalous trajectories of fluid particles violate the incompressibility assumption and suggest the existence of a very thin gelatinous layer of adsorbed BSA possessing viscoelastic properties.

The above results are consistent with properties of adsorbed BSA as well as with the previously studied rheological properties of protein layers [9]. These proteins adsorb at the water-air interface forming very thin viscoelastic layer, or a grid-like network possessing non-trivial and well studied rheological characteristics. The surface rheology determines different types of viscoelastic properties related to specific deformation of the surface of the liquid, such as a dilatational surface elasticity with respect to dilation deformation (the area of the surface is increased or decreased, but its shape remains the same), or a shear modulus with respect to shearing deformation (the shape of the liquid interface changes, but its area is kept constant) [9, 10, 11].

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