

Physicochemical effects of the marine microlayer on air-sea gas transport

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1 Introduction

Observations that films of surfactant material influence wave propagation date back at least two millennia (Plinius Secundus 77 AD). One of the first mathematical treatments of wave damping was that by Levich (1962), who studied the propagation of capillary waves for the limiting cases of a free interface and an interface with an infinite dilational elastic modulus. Levich analytically solved these two limiting cases and found that the time-damping coefficient of short waves was increased by a factor of approximately 1.5 for waves propagating on liquids with infinite elastic films. Levich did not investigate the case of finite surface dilational viscoelasticity, but his work launched a number of theoretical and experimental investigations.

Much of the early interest in the propagation characteristics of short waves stemmed from the increased feasibility of utilizing remotely-sensed microwave backscatter from radars to observe wide areas of the ocean surface. The primary scattering elements for these instruments are ocean surface waves having wavelengths similar to the incident electromagnetic radiation (Bragg scattering), making it necessary to understand the statistical distribution of short waves under different environmental conditions. Remote sensing data products were initially interpreted to yield fundamental

¹In memoriam.

geophysical estimates, including sea state and wind speed (for global weather prediction models), ship wake incidences (for ship detection and tracking), internal wave propagation (for wave interaction models), and later, in assessing mass-transfer rates on global scales (for balancing global budgets for CO₂ and other environmentally important gases).

It became apparent that the presence of surfactant films could affect all of these estimates, but at the time when the first quantitative studies were being performed on capillary ripple propagation, many aspects of the remote sensing field were still in stages of development. For instance, supercomputers had not yet developed to the point where dense maps of wind speed could be assimilated in real time to predict global weather. Similarly, synthetic aperture radar had not yet been developed to produce the scale resolution needed for ship wake tracking or internal wave field interpretation. It was also evident that environmental issues of global warming and greenhouse gases had not yet achieved prominent recognition. These reasons accounted for the ebb in research efforts between the time of the first formulation by Levich in 1962 and the revitalization that occurred two decades ago. The renewed interest that occurred in the 1980s was spurred by the maturation of several independent technologies, including computing power, remote-sensing capabilities, and the development of robust techniques for making short wave measurements *in situ*. This combination enabled studies to assess the amount, extent, and physical properties of the microlayer that forms at the air–sea interface from both natural and anthropogenic sources.

2 Theoretical and experimental studies

Following Levich, subsequent wave propagation studies sought to determine the extent to which surface films modify the air–water interface and its dynamics for intermediate cases of surface dilational viscoelasticity (Mann and Hansen 1963, Hansen and Mann 1964, van den Temple and van de Riet 1965, Lucassen-Reynders and Lucassen 1969, Cini and Lombardini 1978, Hühnerfuss et al. 1982, Bock and Mann 1989). Theory predicted, and laboratory studies validated, that damping enhancements could exceed two orders of magnitude under some circumstances. The bulk of the experimental data was obtained from measurements of the distance-damping characteristics of short ripples. The wavelengths studied ranged from roughly 0.01 cm to 10 cm using waves induced by mechanical, electro-capillary, active thermal, and passive thermal techniques (Sohl et al. 1978, Bock 1987, Stenvot and Langevin 1988).

Other experimental efforts focused on the collection of ocean surface samples in order to investigate properties of the marine microlayer in the laboratory. Samples were retrieved using either inert screens (Garrett 1965) or drum-type samplers (Carlson et al. 1988, Frew and Nelson 1992). Because these samplers physically disturb the natural state of the marine microlayer, *in vitro* results obtained from collected samples could not be proven to represent films in their natural state. Despite this limitation, these studies resulted in characterization of quasi-static film elasticities and led to parameterisations that adequately define bounds within which the marine microlayer typically ranges. By setting such bounds, these works provided constraints for laboratory experiments (e.g., Hirska et al. 1995, McKenna 1997, Saylor 1997). At the same time, techniques for making short wave measurements *in situ* were developed by independent researchers (Hwang 1989, Bock and Hara 1995), and others have followed (Zhang 1995, Suoja 2000).

Field measurements of the microlayer and its effects on air-sea processes have advanced with the use of specialized sampling and measurement platforms. Carlson et al. (1988) developed a small catamaran platform (SCUMS, Self-Contained Underway Microlayer Sampler) for making measurements of the extent of chemical enrichment (using an in-line fluorometer and UV-absorbance detector), near-surface microstructure (using high-resolution, fast-response temperature sensors), and surface wave statistics (using a simple laser slope gauge). This platform was refined to incorporate a fixed-point slope gauge capable of measuring temporal wave spectra and was deployed during several field programs in the Atlantic and Pacific Oceans. A scanning laser slope gauge (Bock and Hara 1995) was integrated later to allow full three-dimensional wave spectral measurements of capillary and capillary-gravity waves (wavelengths between 7.9×10^{-3} and 3.1×10^{-1} m could be measured), as well as an array of capacitive wire wave gauges to measure longer gravity waves (wavelengths greater than 10 m). A recent configuration of this platform (shown in Fig. 1) was used in the Coastal Ocean Processes field experiments conducted off Southern California and in the Northwestern Atlantic Ocean in 1995 and 1997. This research catamaran has the capability of simultaneously measuring short and long wave spectra, surface chemical enrichment, wind stress, and near-surface turbulence.



Fig. 1. Research catamaran during 1997 Coastal Ocean Processes experiment.

The catamaran is shown here after deployment from R/V Oceanus in the Northwestern Atlantic Ocean. The platform is manoeuvred autonomously and is controlled remotely from the ship. During this experiment, the catamaran was equipped to measure long and short wave spectra, sub-surface turbulence, chemical enrichment, and various meteorological parameters. A system to measure vertical profiles of dimethylsulfide above the ocean surface was also incorporated into the catamaran measurement suite

One of the most important findings of these studies and parallel laboratory efforts was that surface wave roughness (a parameter that can be remotely-sensed) was found to correlate well with gas-transfer rates across the air–sea interface (Frew et al. 1995, Hara et al. 1995). Results obtained in both laboratory and field experiments indicate that the range of wavelengths pertinent to gas transport may be restricted to wavelengths of millimetres to centimetres. While the correlation between surface roughness and gas transport is robust under differing environmental conditions, the specific mechanisms of interfacial transport have yet to be adequately elucidated and are the topic of recent research (e.g., McKenna 2004).

3 Mechanisms of air–water gas transport

The enhancement of air–sea gas transport (for waterside rate-controlled gases), over that expected for pure molecular diffusion, is affected by sev-

eral interrelated mechanisms that can depend on the wind and wave conditions. Thus, many complex physical processes influence gas transfer, including: the interaction between the wind-driven surface shear layer and the wave field, near-surface turbulence, the modification of interfacial hydrodynamics by surface films, and breaking waves that lead to bubble formation and increased mixing. Consequently, an experiment or model that completely describes gas transfer does not exist. Estimates of gas-transfer coefficients based on wind speed have been favoured for years (e.g., Liss and Merlivat 1986, Wanninkhof 1992). Each of these formulations predicts a monotonic increase in the gas-transfer rate with increasing wind speed. The largest difference between the Liss and Merlivat model and subsequent models is the mechanistic interpretation of distinct regimes occurring under different wind conditions. Liss and Merlivat suggest a low critical wind speed below which the gas-transfer rate increases slowly with increasing wind speed owing to the lack of near-surface mixing associated with a smooth surface without waves. They also suggest a second critical wind speed that occurs when the wind over a wavy surface exceeds the threshold necessary for initiating wave breaking. Above this point, it is presumed that wave breaking, bubble entrapment, and near-surface mixing by rising bubbles contribute significantly to gas transfer. In contrast to this tri-linear model, the Wanninkhof (1992) relationship postulates a smoother enhancement of gas transfer as wind speed increases.

Several laboratory and field studies, from the early work of Broecker et al. (1978) to the more recent studies of Frew (1995), have shown that a unique relationship between the gas-transfer rate and wind speed is not likely to exist for natural waters where surfactants are influential. Fundamentally, the exchange of gases between water and air occurs through very thin boundary layers at the air–water interface where turbulent motions are suppressed giving way to molecular diffusion processes. The details of these layers — their thicknesses and the gradients across them — are determined by the aero- and hydrodynamics near the surface. For sparingly soluble gases (e.g., CO₂, O₂, N₂) the airside effect is less critical, and the transport is determined by the waterside boundary layer. The fluid flow in this boundary layer is highly sensitive to the presence of a surface film and can be modified considerably, which is the reason gas transfer exhibits a similar sensitivity. Therefore, studies of the relevant physical mechanisms of interfacial gas transport and the role of surface films are crucial to improving our understanding of the exchange process.

Conceptual models aimed at explaining the dominant processes associated with gas transfer can be generalized as being either turbulent eddy diffusivity or surface renewal models; others are based on similarity considerations with reference to experimental data. Irrespective of the model

type, it may be assumed that the vertical velocity fluctuations near the surface, $w'(z)$, which bring fresh fluid from the bulk to the interface for diffusion, contribute to gas-transfer enhancement. Assuming incompressibility, these velocity fluctuations can be expressed in terms of the surface divergence as $w'(z) \approx -(\nabla_h \cdot \mathbf{v}')_o z$ to first order, where the subscript 'o' indicates evaluation at the interface, $z = 0$. Expressing $w'(z)$ in such manner suggests a strong connection between the surface divergence, $(\nabla_h \cdot \mathbf{v}')_o$, and gas transport. Some models (e.g., Lamont and Scott 1970, Brumley and Jirka 1988) attempt to predict gas-transfer rates by considering surface renewal by turbulent eddies. Such a process is sketched conceptually in Fig. 2, which shows how upwelling eddies bring about a horizontal surface divergence that renews the near-surface region with bulk fluid from below. This figure also makes clear how the presence of a surfactant can influence surface renewal and divergence. A common difficulty with many gas-transfer models has been relating the model inputs (velocity, length, and/or time scales) to relevant, measurable hydrodynamic parameters. Therefore, a direct experimental measurement of the surface divergence could prove very useful. In addition, such a measurement could be used to explore the model results of Ledwell (1984) and Csanady (1990), which provide direct relationships between surface divergence and the gas-transfer rate.

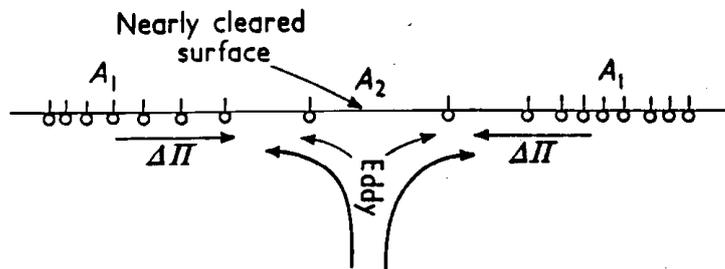


Fig. 2. Conceptualisation of surface renewal by a turbulent eddy in the presence of a surfactant. The eddy upwells fresh fluid to the surface, while the differential spreading pressure, $\Delta\Pi$, opposes the transport of new fluid to the interface. Adapted from Davies and Rideal (1963)

In order to gain insight into the impact of an interfacial film on surface divergence, the case of linear, monochromatic capillary-gravity waves was studied. Although it is generally believed that the mechanical straining of the surface due to orbital wave motions is not sufficiently large enough to bring about significant gas-transfer enhancement, this simple case provides a useful and illustrative result.

Using a formulation similar to that of Lucassen-Reynders and Lucassen (1969), the near-surface two-dimensional velocity field was numerically solved for both ideal and viscoelastic surfaces. Before considering surface divergence, Fig. 3 shows an example of the near-surface profile of the velocity component in the direction of wave propagation for three different conditions: ideal flow (irrotational and inviscid), viscous flow with zero elastic modulus, and viscous flow with a finite viscoelastic modulus. The results are for a 15 Hz water wave, with the profiles taken under a crest. The wave amplitude was set at 5% of the wavelength to accommodate linear theory, and velocities were forced to be equivalent at depth. The effect of a viscoelastic surface on the surface velocity is marked. The sharp gradient in the velocity within the upper 0.3 mm for the surfactant case is responsible for wave damping. This strong gradient is acted upon by the fluid viscosity, which dissipates the wave motion effectively. The surface divergence for the same group of conditions and for the same wave motion is shown in Fig. 4. The divergence is plotted along the phase of the wave. The considerable reduction ($\sim 50\%$) in divergence due to a viscoelastic surface is apparent. Reduction in surface divergence implies less surface renewal, which leads to reduced gas-transfer enhancement. In this way, the role of surface films in air–water gas exchange is revealed.

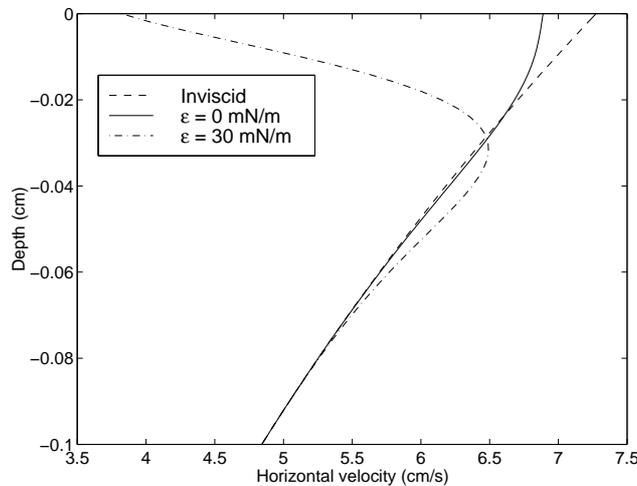


Fig. 3. Theoretical streamwise velocity profiles for a 15 Hz capillary-gravity wave. Curves are shown for an ideal inviscid flow, a real flow with zero viscoelasticity, and a surfactant flow with a finite viscoelasticity. All profiles are taken under a wave crest. The wave amplitude : wavelength ratio is 1 : 20

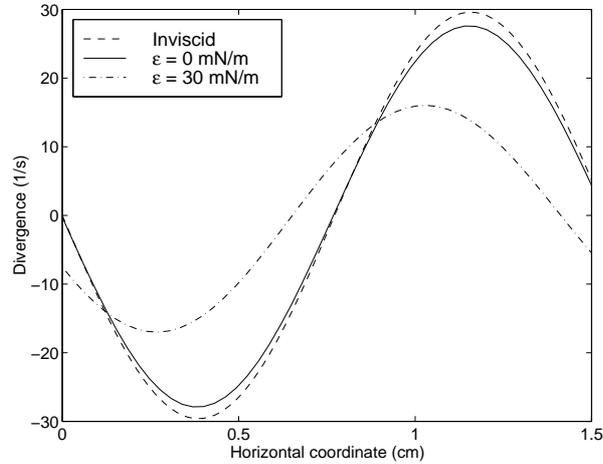


Fig. 4. Theoretical surface divergence for a 15 Hz capillary–gravity wave.

Curves are shown for an ideal inviscid flow, a real flow with zero viscoelasticity, and a surfactant flow with a finite viscoelasticity. Each curve is along the phase of each wave, with the horizontal position at zero corresponding to a wave crest. The wave amplitude : wavelength ratio is 1:20

Next, we report results of a laboratory investigation focused on the effects of surface films on the flow field near the air/water interface. For repeatability and simplicity, we selected an axisymmetric vortex ring for the source of surface expansion and dilation (divergences) of the near surface. A detailed description of the experimental apparatus can be found in McKenna (1997). In summary, the technique relied on forcing a small slug of fluid from the mouth of a knife-edged stainless steel tube vertically into a visualization tank. Upon exit from the tube orifice, the fluid rolled up into a vortex ring and propagated upward where it then interacted with the free surface. Surfaces with varying surfactant concentration were used to examine the influence of different surfactant composition on ring impact and the ensuing hydrodynamics. Shown in Fig. 5 are flow visualization results taken from McKenna (1997) illustrating the effect of surfactants on the near-surface flow field. Four cases are presented: (a) approach toward a free surface, (b) interaction with a nominally clean free surface, (c) interaction with a surface covered with a microlayer of stearic acid, and (d) interaction with a solid wall boundary.

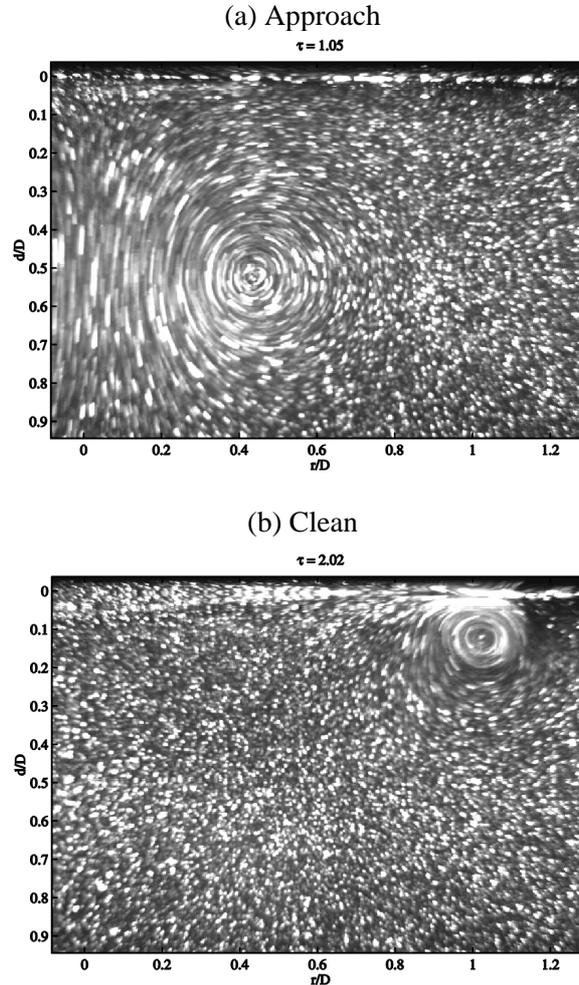


Fig. 5. Flow visualization of vortex ring interaction with various interfaces.

Each panel represents a vertical cross-section of the flow and only one half of the vortex ring is imaged. In each panel, r indicates the radial distance from the vertical axis of the ring, d measures the vertical distance from the interface, D is the major diameter of the ring prior to surface interaction, and τ is a dimensionless time. In this figure, panel (b) represents a surface that has not been contaminated intentionally. For this case, accompanying measurements of the spatial damping parameter (using the capillary wave technique) indicate the damping was, on average, within 7% of the theoretical value for pure water. Panel (c) is that for a stearic acid microlayer having concentration $3.5 \times 10^{-10} \text{ mol cm}^{-2}$. Taken from McKenna (1997)

(c) Microlayer

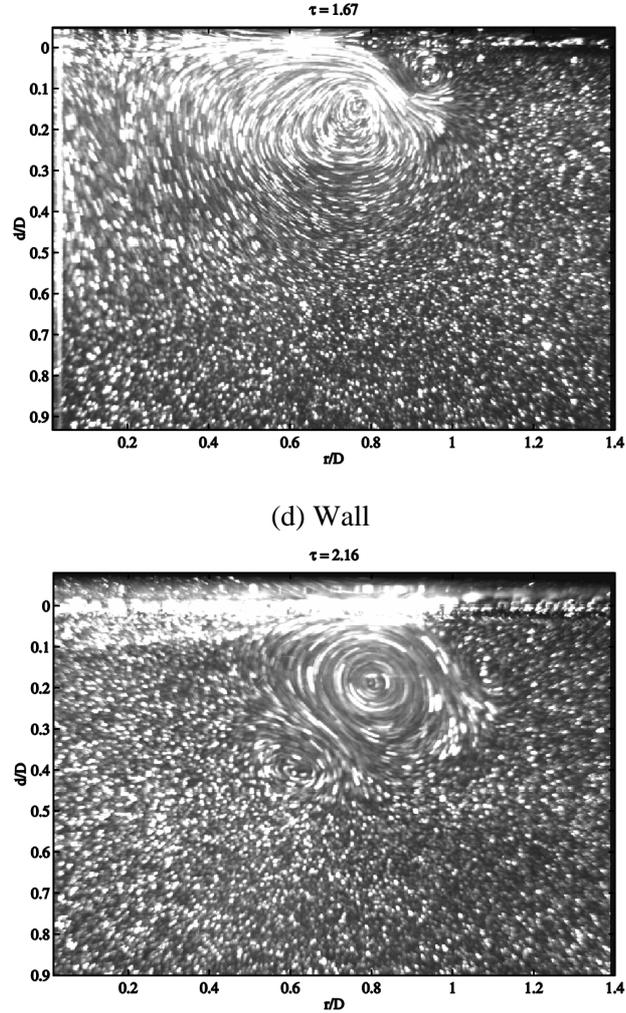


Fig. 5 (cont.) Visualization of vortex ring interaction with various interfaces.
 Panel (c) is that for a stearic acid monolayer having concentration 3.5×10^{-10} mol/cm². Taken from McKenna (1997)

In an ideal fluid with a free surface and potential flow, a vortex ring nearing the surface will tend to dilate. This result can be rationalized by the mathematical construction of an ‘image’ vortex ring reflected about the interface and approaching from the opposite direction. Fig. 5b depicts the experimental observation of a physical case similar to this type of behaviour. On the other hand, when a no-slip or intermediate-slip interface is present, the dilation of the ring major diameter is altered, as is the trajectory subsequent to surface interaction. The solid boundary demonstrates

the behaviour of a no-slip interface (Fig. 5d), and the viscoelastic surface condition, obtained by spreading a surfactant film on the water surface, represents an intermediate-slip interface (Fig. 5c). A surfactant-covered surface constitutes an intermediate-slip condition because of the Marangoni effect. This effect arises from gradients in surfactant concentration that lead to gradients in surface tension, which then give rise to surface stresses and surface flows. Owing to Marangoni stresses, a surfactant surface can support a finite surface shear, whereas a free surface in the absence of any adsorbed film cannot. These Marangoni stresses develop as a result of the vortex-induced surface velocities, which act to redistribute surfactant and compress the film as the ring interacts with the surface. Such compression results in spatial gradients of the surface tension that seek to equilibrate and produce a surface flow field by themselves. These opposing flows in the surface viscous boundary layer can have strong implications for the bulk flow. Vorticity, opposite in sign to that of the primary vortex cell, can be generated in this near-surface layer. In all the cases studied, save for the clean case, coherent secondary and tertiary vortex rings were formed as a result of this newly generated vorticity. In Fig. 5c, a second vortex outside the primary cell can be seen forming very near the interface, and in Fig. 5d, two additional vortex rings are observed developing as a result of the wall interaction. Thus, a surface with an adsorbed surfactant can act intermediate between a free-slip boundary and a no-slip boundary. The ability of a surfactant surface to hydrodynamically resemble a solid boundary is widely evidenced in studies of air/water gas exchange. For mass transfer at a smooth solid boundary, the gas-transfer velocity is found to follow a $-2/3$ exponential Schmidt-number dependence (Deacon 1977). It has been observed (e.g., Jähne et al. 1984) that film-covered free surfaces, which inhibit formation of small-scale waves, tend to follow the same $-2/3$ dependence rather than a $-1/2$ dependence that is expected for an ideal free surface.

In order to elucidate the affects of a surfactant on the near-surface flow further, the temporal evolution of the radial component of the near-surface velocity as the vortex ring approaches the surface is plotted in Figs. 6a, b. Velocity measurements were carried out using the non-invasive full-field flow measurement technique known as digital particle image velocimetry (DPIV). In addition to a general attenuation of the tangential velocity, we observe the quasi-no-slip condition occurring outside of the vortex core for the microlayer situation. This is a direct consequence of surfactant-induced Marangoni forces that develop at the interface. These vortex ring experiments illustrate the degree to which surface films can modify near-surface flow. By acting like a solid interface, a surfactant surface can dramatically alter the flow field at the interface. Fluid velocities at the interface are re-

duced, inhibiting eddy motion near the surface. This affects the degree of vertical upwelling at the surface through a reduction in the surface divergence.

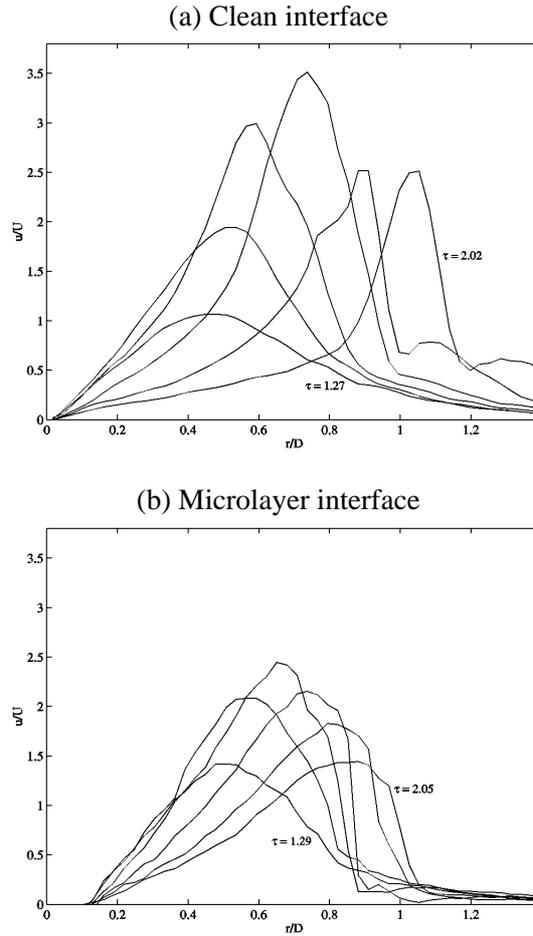


Fig. 6. Variation of the near-surface radial component of velocity. Panel (a) is for a clean interface; panel (b) is for the stearic acid microlayer. All variables are as defined in Fig. 5, u is the radial component of velocity, and U is the vertical free propagation speed of the vortex ring. Velocities are taken along a horizontal section at a depth $d/D = 0.03$ (0.125 cm). The temporal spacing between curves, $\Delta\tau$, is 0.15. Taken from McKenna (1997)

4 Concluding remarks

Much progress has been made in understanding the role of surface films in air–sea gas transfer. Advances in both laboratory and field techniques have provided improved measurements and needed insight into this complex and important process. Current efforts focused toward better understanding the key physical mechanisms involved in interfacial gas transfer and the role of surfactants, along with innovative and interdisciplinary field and laboratory measurements, will help to further our present knowledge.

Acknowledgements. This paper was originally presented in 1998 at EUROMECH 387, “Surface Slicks and Remote Sensing of Air-sea Interactions” in Coventry, UK. It was during the final preparation of the manuscript that Erik John Bock unexpectedly passed away. Erik was an exceptionally talented experimentalist and his passing represents a great loss to the marine sciences community. Erik was a pioneer in sea-surface measurements, most notably three-dimensional frequency-wavenumber spectra of ocean waves, and his contributions have been integral to many innovative air–sea interaction studies. Erik was also a supportive advisor and mentor, as well as a good friend. He will be dearly missed. –SPM

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