

ACOUSTIC SCATTERING AFFECTED BY MONOMOLECULAR FILMS SPREAD OVER A WIND-DRIVEN WATER SURFACE

S. J. POGORZELSKI

Environmental Acoustics Laboratory University of Gdańsk
(80-952 Gdańsk, Wita Stwosza 57)

A spectrum of amplitude fluctuations of the ultrasonic signal scattered from a wind-created water surface covered with an oil substance monolayer of well-defined viscoelastic properties was examined under laboratory conditions. The presence of an oil film causes a shift of the peak frequency towards higher frequencies and increases the fall-off on the high-frequency band of the spectrum if compared to clean surface scattering. The effect turned out to be unequivocally related to rheological parameters of the given monolayer film.

1. Introduction

It is well-known that the organic films on the sea surface both natural and artificial are very predominant in coastal zones. The existence of mineral oil spills at sea is becoming a problem of growing concern to our society because of its adverse effects on marine life in coastal and inland waters. Therefore there is a great need for aerial surveillance systems capable of detecting oil-derivative contaminations under all weather conditions and independent of the time of the day. These slicks strongly affect the surface waves field and thereby influence most air-sea interaction processes, for instance, the momentum and energy transfer from the wind to the wave field, and the gas exchange at the air-sea interface [5], [7], [26].

In modern oceanography studies of this variability are closely connected with the problem of remote sensing techniques of oceanic processes by their manifestation on the wavy surface. For example, the electromagnetic emission in the microwave [2], [3], and visible [9] spectral bands as well as the scattering of electromagnetic waves are influenced by monomolecular surface films [10], [11]. The theoretical background for an acoustic remote sensing method is derived, potentially applicable

for determining physicochemical characteristics of surface films. The presented theory makes use of the bond existing between rheological surface film properties, damping, and signatures of wind-excited waves spectra as turned out strictly related to ultrasonic surface scattering spectra of amplitude fluctuations.

The Marangoni effect has been recently identified as being responsible for resonance like type damping of short gravity waves when the wavy surface is covered with viscoelastic films [3], [13]. Short gravity and capillary waves which play a principal role in Marangoni damping are characterized by a particular shape i.e. they have a large slope and a rather small amplitude [24]. The most suitable acoustic system for investigation of scattering on this kind of wavy surfaces consists of a directional transducer irradiating a small area of the studied surface [8], and creating conditions of scattering on so-called high-frequencies (i.e. at large values of Rayleigh parameter). In light of the scattering theory [25], if the wavelength and beam geometry are known, the specularly scattered sound intensity (relative to the plane surface situation) is solely determined by the mean-square surface slope for "rough surfaces" [6, 17].

All systematic changes of the rough surface undulation, for example due to presence of a layer of oil substance, express themselves in corresponding changes in the scattered signal statistics as already shown in laboratory and at-sea experiments by the author [19–22].

In this paper the author reports about the analysis of the amplitude fluctuations spectrum of the ultrasonic signal scattered from a wind-created surface of clean water and its surface covered with a monolayer of well-defined rheological properties. The substances under study stand for a group of commercially available crude oil derivatives having very different physical properties, and which can be found out in natural waters as contaminations. Surface pressures of the spread films were on the order of a few dynes/cm, similarly the natural sea slick took on the same value [4], [5], [9], [14]. The spectra were analysed to characterize the surface pressure, dilational elasticity modulus and structural diffusion parameter values which completely identify the spread film [1], [7]. Another motivation for this experiment was to predict the sensitivity of the amplitude fluctuations spectra to monomolecular surface films. The wind wave tunnel results indicate that the method based on high-frequency surface scattering appears to be a feasible approach in the field of sea surface contamination monitoring. It is pointed out that as a precursor to the at-sea application additional experiments with well-defined artificial films of different chemical structure ought to be performed.

2. Experimental arrangement and method

The wind-wave flume was open-ended, being 0.24 m wide by 0.10 m high and 0.60 m long. It contained tap water to a depth 0.3 m (see Fig. 1). A stream of air is generated using two fans (1, 2) which allows forming the characteristics of the stream,

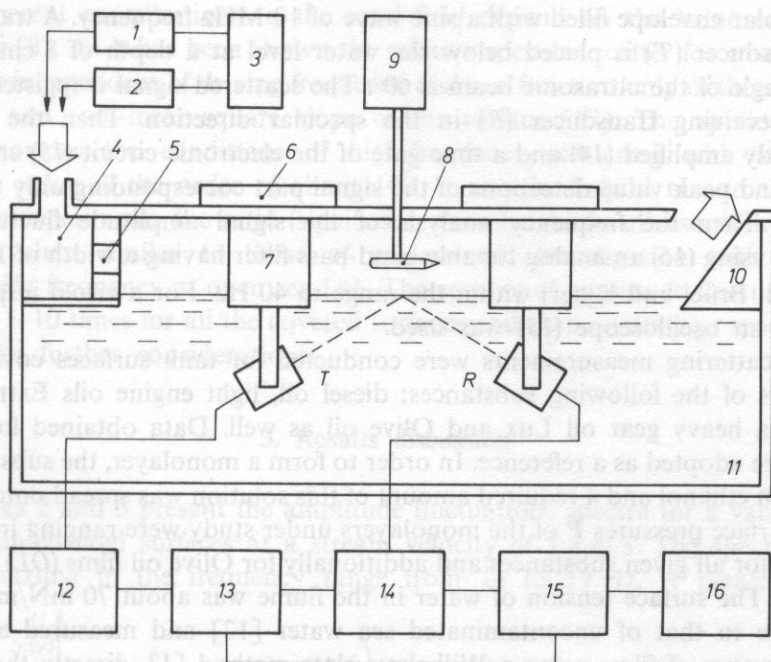


FIG. 1. Block diagram of experimental set-up. For details see text

and introduced it to the tunnel through a nozzle (4) equipped with several guide vanes (5). A supply system allows smooth adjustment of an air stream velocity up to 15 m/s. The tunnel centreline wind speed V was measured by means of a Prandtl tube (8, 9) at a fetch of 0.3 m and situated directly above the point of ultrasound beam scattering. In this paper we are largely concerned with very low winds for which the flow is aerodynamically smooth, the thickness of the viscous sublayer exceeds the characteristic height of the surface waves for stream velocities below 4 m/s [23]. The opposite bank (10) of the channel is inclined at an angle of 40° with respect to the water surface, which significantly reduces the influence of reflected waves. An air stream causes a shear stress at the wavy surface, which results in Stokes drift and unfavourable effect of water accumulation at the end of the linear tunnels of conventional construction [4, 13, 14]. A top-side view of author's tunnel design which consists of special plexiglass shields situated on both sides of the tunnel along its entire length was presented in [19]. Shields screen a part of the surface from action of the air stream and provide the recirculating path between the end and front of the tank. Application of shields also leads to obtain a uniform surface pressure of the spread monolayer over the entire length of the tank surface. Hence, conditions created for generation of wind waves and spreading of oil films are close to natural.

An acoustic part of the experimental set-up consists of a pulse operating ultrasonic transmitter (12), which forms series of pulses lasting several microseconds

of rectangular envelope filled with a sine wave of 10 MHz frequency. A transmitting quartz transducer (T) is placed below the water level at a depth of 8 cm, and the incident angle of the ultrasonic beam is 60° . The scattered signal is registered by an identical receiving transducer (R) in the specular direction. Then the signal is subsequently amplified (14) and a time gate of the electronic circuit (15) enables the envelope and peak value detections of the signal part corresponding only to surface scattering. Then the frequency analysis of the signal amplitude fluctuations is performed using (16) an analog tunable band-pass filter having a width of $1/3$ octave (Type 1621, Brüel and Kjaer) within the range to 40 Hz. For a visual inspection of the signal an oscilloscope (13) was used.

The scattering measurements were conducted for tank surfaces covered with monolayers of the following substances: diesel oil, light engine oils Extra 15 and Hipol 15, a heavy gear oil Lux and Olive oil as well. Data obtained for a clean surface were adopted as a reference. In order to form a monolayer, the substance was dissolved in ethanol and a required amount of this solution was spread onto the tank surface. Surface pressures P of the monolayers under study were ranging from 3.4 to 4.7 mN/m for all given substances and additionally for Olive oil films (OL) at 11 and 15 mN/m. The surface tension of water in the flume was about 70 mN/m which is comparable to that of uncontaminated sea water [12] and measured as well as surface pressure of films using a Wilhelmy plate method [1], directly through the window in a roof of the tunnel. Each monolayer of a film-forming oil substance has certain viscoelastic rheological properties characterized by the surface pressure P , which represents a surface tension reduction of a clean water surface due to the film presence, by the dilational elasticity modulus E_0 , and finally structural diffusion parameter w_d values. The magnitude of E_0 , defined as $E_0 = -d\gamma/d(\ln\Gamma)$, [1], where γ — is the surface tension of studied surface and Γ — is the surface concentration of film-forming substance, depends only on a chemical nature and concentration of the material composing the film. The remaining parameter w_d is governed by the kinetics and mechanism of the rearrangement process which takes place within an insoluble oil monolayer during compression/expansion cycles [7]. $1/w_d$ corresponds to a doubled characteristic time of the relaxation phenomenon. In order to determine these parameters additional measurements were carried out in which the dynamical method proposed by LOGLIO et al. [15] was adopted. It has been shown by HÜHNERFUSS et al. [12], [13] that these values of E_0 and w_d obtained under dynamic conditions are much more suitable for describing the wave damping effect of monolayers. These investigations were performed in a Langmuir trough with a Wilhelmy plate connected to a wire balance as a surface pressure sensor at the same temperature and surface pressures as encountered on the tunnel surface.

The scattering coefficient of acoustic waves can be defined as the ratio of the received intensity when the acoustic wave is reflected by surface under study to the received intensity when the wave is reflected in the specular direction by a plane surface according to BECKMANN [6]. Generally, the scattered field registered in a specular direction consists of both coherent and incoherent components. Coherent

and incoherent contributions to the total field depend on the value of Rayleigh parameter [8]. As it has been shown the relative specular scatter for a very "rough surface" is independent of driving frequency and is a function only of the experiment geometry and root-mean-square slope of the surface [17]. The applied acoustic system provides the conditions of high-frequency scattering with a Rayleigh parameter value of the order of about 100. It may be helpful to add that in the laboratory experiments the ratio (significant wave height to ultrasonic wavelength) took on values similar to those of an acoustic investigation with waves of 80–100 kHz frequency at open sea [8]. The spectra of amplitude fluctuation were measured 5–10 times for all the covered surfaces and then an average spectrum was adopted for further considerations.

3. Results discussion

Figures 2 and 3 present the amplitude fluctuations spectra for a variety of the studied film-covered surfaces at a stream velocity of 1.4 m/s. The spectra exhibit distinct maxima in the frequency range from 14 to 19 Hz. A presence of the

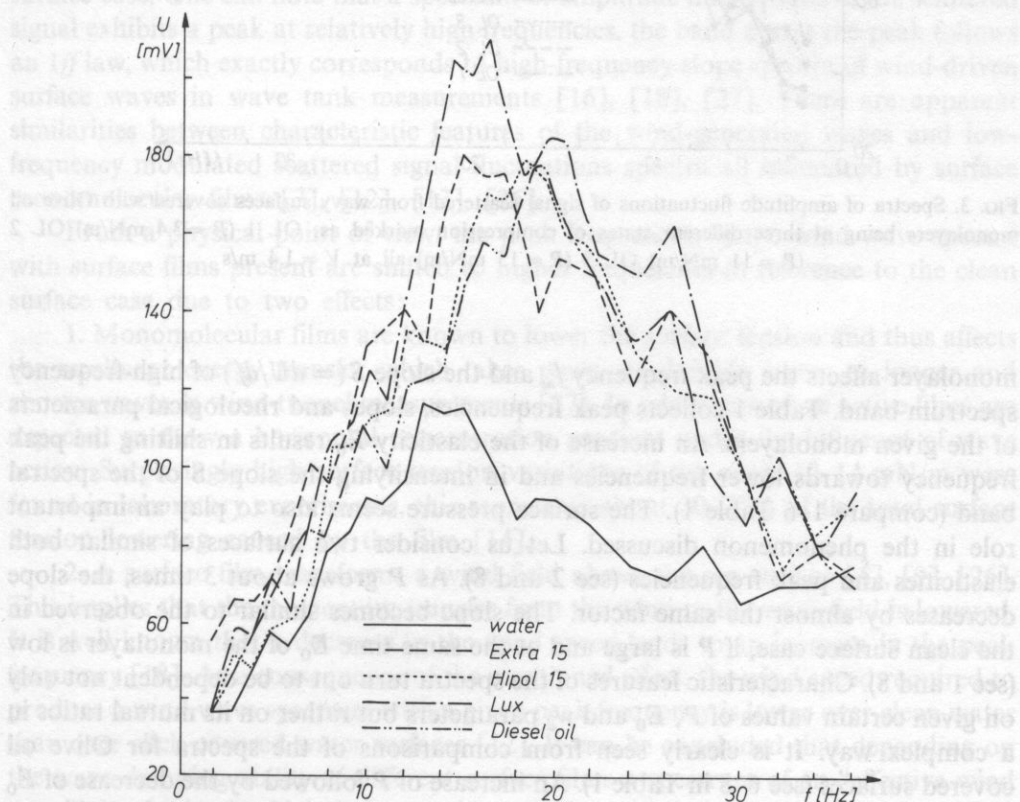


FIG. 2. Spectra of amplitude fluctuations of signal scattered from wavy surfaces covered with monolayers of chosen oil substances at $V = 1.4$ m/s. Clean water surface reference curve —

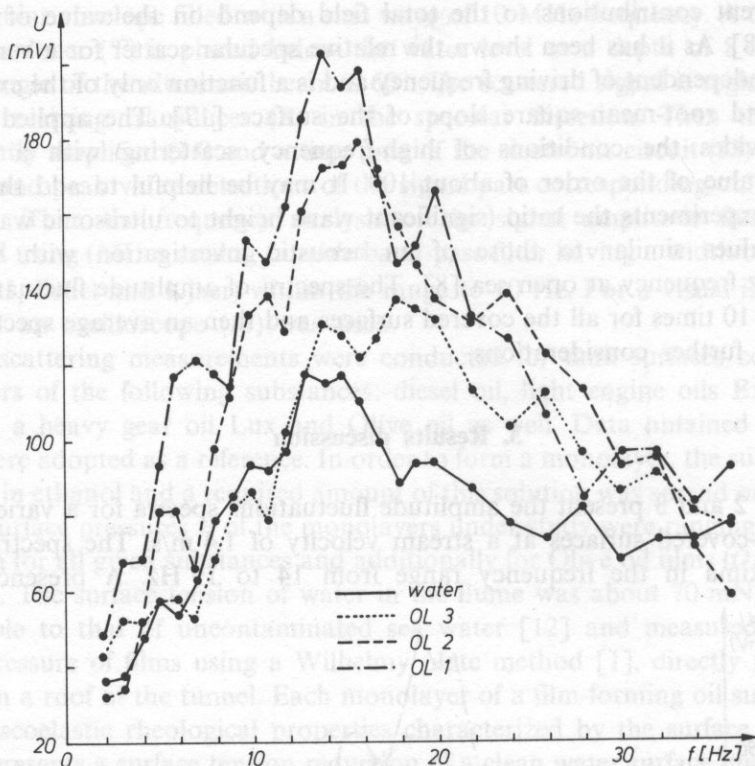


Fig. 3. Spectra of amplitude fluctuations of signal scattered from wavy surfaces covered with Olive oil monolayers being at three different states of compression marked as: OL 1 ($P = 3.4$ mN/m) OL 2 ($P = 11$ mN/m) OL 3 ($P = 15$ mN/m) all at $V = 1.4$ m/s

monolayer affects the peak frequency f_m and the slope $S (= dU/df)$ of high-frequency spectrum band. Table 1 collects peak frequencies, slopes and rheological parameters of the given monolayers. An increase of the elasticity E_0 results in shifting the peak frequency towards lower frequencies and in intensifying the slope S of the spectral band (compare 1–6 Table 1). The surface pressure seems also to play an important role in the phenomenon discussed. Let us consider two surfaces of similar both elasticities and peak frequencies (see 2 and 8). As P grows about 3 times, the slope decreases by almost the same factor. The slope becomes similar to the observed in the clean surface case, if P is large and at the same time E_0 of the monolayer is low (see 1 and 8). Characteristic features of the spectra turn out to be dependent not only on given certain values of P , E_0 and w_d parameters but rather on its mutual ratios in a complex way. It is clearly seen from comparisons of the spectra for Olive oil covered surfaces (see 6–8 in Table 1). An increase of P followed by the decrease of E_0 results in lowering of the mean spectrum level, in shifting of the peak frequency

Tab. 1 Peak frequencies f_m and high-frequency band slopes S of measured spectra versus viscoelastic properties of the spread oil substance monolayers. SD — standard deviation

SUBSTANCE	P [mN/m]	E_0 [mN/m]	w_d [10^{-3} rad/s]	f_m [Hz]	S [mV/Hz] (SD)
1 water	—	—	—	14.0	—2.10 (0.17)
2 Diesel oil	4.4	9.8	0.97	18.8	—8.0 (0.56)
3 Hipol 15	4.7	10.0	0.59	18.0	—8.18 (0.49)
4 Extra 15	3.9	13.5	1.02	16.0	—6.67 (0.46)
5 Lux	4.1	14.5	1.08	15.0	—10.0 (0.76)
6 OL 1	3.4	16.0	0.43	14.5	—8.0 (0.42)
7 OL 2	11.0	13.0	„	16.4	—5.31 (0.29)
8 OL 3	15.0	8.0	„	18.2	—2.73 (0.22)

towards higher frequencies, and in a drop of S to the value observed for the clean surface case. One can note that a spectrum of amplitude fluctuations of the scattered signal exhibits a peak at relatively high frequencies, the band above the peak follows an $1/f$ law, which exactly corresponds to high-frequency slope spectra of wind-driven surface waves in wave tank measurements [16], [18], [27]. There are apparent similarities between characteristic features of the wind-generated waves and low-frequency modulated scattered signal fluctuations spectra all influenced by surface monomolecular films [7], [12], [13], [26].

From a physical point of view, the peak frequencies of the wind wave spectra with surface films present are shifted to higher frequencies in reference to the clean surface case due to two effects:

1. Monomolecular films are known to lower the surface tension and thus affects the nonlinear energy transfer which takes place via 5–9 Hz waves to longer and shorter waves in wind-tunnel measurements [27]. In addition surface active films are expected to show a horizontal concentration gradient under the influence of wave action. Surprisingly high surface tension variations of the order 12–14 mN/m were found in laboratory experiments, this represents about 30–70% of the total surface tension lowering caused by the film [14].

2. A surface film transforms a wind field above the sea surface [5], [9], [26]. This implies that the momentum transfer from the wind to the wave field is lowered. It is well known that a decrease in the wind speed leads to an increase in the peak frequency [18]. As a consequence of the mentioned effect, the wind speed required to produce a wind wave spectrum with a given peak frequency is lower over clean water than over slick-covered water surfaces [12]. It can be concluded that depending on the wave damping ability of different surface films a spectrum of an “effective wind speed” is obtained which is lower than the actual wind speed [3], [13].

4. Conclusions

The presence of an oil substance monolayer spread onto a wavy water surface affects the peak frequency and slope of the high-frequency band of the scattered signal amplitude fluctuations spectrum. The effect is unequivocally related to the viscoelastic rheological properties of the film.

Taking together reported apparent similarity between the wind wave spectra and amplitude fluctuations spectra of scattered acoustic signals, if influenced by surface oil derivative films, the observed peak frequency shift might be a measure of the covered surface susceptibility to deformation caused by an air stream.

It is of course too early to deduce the physicochemical surface films characteristics by remote sensing acoustic methods based on the presented results, but the method appears to be a feasible approach. However, it has to be stressed that additional experiments with well-defined artificial sea slicks have to be performed.

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