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STATISTICAL ASPECTS OF ACOUSTIC RETURNS FROM A WIND-DRIVEN WATER SURFACE COVERED WITH ORGANIC FILMS

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The statistical properties of ultrasonic (10 MHz) signals scattered at a wind-driven water surface covered with different petroleum derivative films of well-defined and oceanographically relevant elastic surface properties were examined under laboratory and open-sea conditions. Evolution of the shape, skewness and kurtosis parameters of the signal distribution as a function of wind speed reflects a principal role played by the film elasticity. The elastic properties of composite sea surfaces likely to be present in nature and consisting of oil spills filled with a surface active material, floating solid particles, bubbles or drops of a third fluid with their important implications in remote sensing techniques are also discussed. A two-spatial scale irregularities distribution of the sea surface (capillary ripples present on tilted faces of long gravity waves) expresses itself in the corresponding signatures of at-sea experiment data.

1. Introduction

Ideally, a remote sensing system should be able to routinely detect unknown oil discharges and so act in a policing and alterting role, and also be able to accurately determine the location, extent, thickness and type of oil pollutant. No single sensor system is presently capable of performing all these tasks and so it seems that a multi-sensor approach is the optimum solution.

The hypothesis presented in a few papers by the author [1–3] could lead to a practical acoustic surveillance system for polluted sea areas. It has been demonstrated in open-sea experiments with artificial crude oil films spread over the Baltic Sea surface and a buoy-like high-frequency (10 MHz) acoustic system based on the specular forward scattering geometry that the viscoelastic film parameters can be recovered from the signal modulation spectra by means of the Marangoni damping theory and the 3-parameter best-fit procedure to the acoustic data.

Water wave attenuation by viscoelastic films is attributed to the Marangoni effect [4] which causes a strong resonance-type wave damping in the short gravity-capillary wave region. It is evident that the Marangoni damping depends on the physicochemical nature and concentration of the film-forming substance [5]. Short gravity and capillary waves are characterized by a particular shape i.e., they have large stepness and rather small amplitudes [6]. A directional ultrasonic transducer of narrow transmitting characteristics which irradiates only a small area of the studied surface seems to form the most suitable acoustic tool to investigate scattering from such surfaces [7]. In the light of the high-frequency scattering theory the modulation spectrum of the acoustic scatter is linearly related to a wind-driven surface wave slope spectrum [8]. Although, the relationship between the ocean wave spectra and the (laser, radar, acoustic) image spectra (i.e., the transfer function) is still not satisfactory determined [9]. The same principle is fulfilled in sun glitter and laser surface probing measurements [10], where the wavelength of light is smaller than all other wavelengths on the rough surface in question [11].

Thus, the scattered signal signatures should be related (via wind waves damping) to the viscoelastic properties of the film.

So far, the smoothing effect of an oil film on a ruffled surface is expressed in the corresponding changes of statistical properties of the ultrasonic signal scattered at a wavy surface as has been already shown in laboratory and at-sea experiments [12–14]. Simultaneous analyses of all the statistical distribution parameters could be a starting point for determining the fraction weight of the given substance, its layer thickness, and finally a form of the oil pollutant (monolayer, thick layer or individual dispersed spots) [15].

This paper deals with further detailed statistical analyses of the amplitude fluctuations of the ultrasonic signal scattered from a wind-generated surface of clean water and water covered with a monolayer of well-defined oceanographically relevant elastic properties performed in a small wind-wave tank and in open-sea conditions with artificial oil slicks.

Several film-forming substances compirising a group of commercially available crude oil products which from a physicochemical point of view are characteristic of natural crude oil slicks were used in these studies. The decreases in surface tension of water i.e., surface pressure of the film caused by these material and by sea slicks were of the same order of magnitude. The same holds for the surface elasticity of the films formed by both types of materials [5]. To characterize the elastic properties of the film supplementary Langmuir through measurements were performed using the surface tension-time relationship by means the procedure and set-up proposed by Loglio *et al.* [16, 17], and a novel sampler-elastometer was used to determine "in situ" artificial sea slicks [18].

In nature sea surfaces are often non-uniform. Such composite surfaces consisting of thick oil spills with "holes" filled with a surface-active material can also contain floating solid particles, bubbles or droplets of a third fluid: The composite dilational modulus of such a surface depends on the structure and wetting properties of the components in a complex way [19, 20], that should be borne in mind while interpreting any remote sensing data based upon the smoothing effect of the sea surface due to the surface film. It is intended that the review presented in this paper may serve as a general overview and provide a guide to what is currently possible and what is being developed for future use.

2. Ultrasound scattering from a wind-driven water surface and its statistical features

The scattering of light, radio and acoustic waves by the sea surface depends on the slope distribution, either directly, as in the case of "specular point" scattering [21], or indirectly via tilting and hydrodynamic effects, as in the case of large-angle Bragg diffraction scatter [22]. Certainly, laser backscatter from the ocean surface is caused not by Bragg resonant reflection but by local specular reflection from surfaces (facets) on the small-wave structures that are oriented near normal to the incident laser propagation vector [11]. The same applies to high-frequency acoustic scattering [23, 24].

The scattering coefficient may be defined as the ratio of the received intensity when the acoustic wave is reflected by the surface under study to the received intensity when the wave is reflected in the specular direction by a plane surface [25]. Since the scattered waves are essentially planar over the receiver aperture their intensity is determined from acoustic pressure measurements, and assuming a linear dependence between the output voltage U of the hydrophone and the acoustic pressure the limiting scattering coefficient for a very rough surface (so-called high-frequency scattering) is given by Eq (15) in [2], and is related to the insonified surface area A, angle of incidence δ, and the mean-square slope of the wind-generated surface which can be derived from the omnidirecitional wave height spectrum $S(f)$ [26].

In the extreme cases for a very rough surface, there is only slight variation in scattered signal amplitudes with angle of incidence as the scattered field is totally *diffuse and closely isotropic [23]. Recent measurements of high-frequency spectra of* *nd waves using a wave-following laser surface slope meter have been reported by* ANT [10]. These results show that such spectra follow an $1/f$ law to a good proximation. It should be noted that the modulation spectra of the ultrasonic nal (10 MHz) specularly scattered from a wind-driven surface studied in ind-wave tank by the author [1] follow the same frequency dependence.

To sum up, the modulation spectrum of the specular scattering coefficient (see [2]) merely linearly related to, and can be transformed into, a sea wave slope spectrum $k^2S(f)$, [8]). The relationship between the ocean wave spectra and the (radar or ustic) image spectra (i.e., the transfer function) is still not satisfactory determined

The modulation spectra of the differential scatter examined in the previous hor's papers [2, 3] are derived from the spectra of low-frequency (1–40 Hz) plitude voltage fluctuations of the ultrasonic signal (10 MHz, acoustic wavelength .15 mm) specularly scattered from a wind-created surface of clean water $U_0(f)$ and ered with an oil substance film $U_c(f)$:

$$S_0(f)/S_c(f)=[U_c(f)/U_0(f)]^2. \qquad (2.1)$$

surface due to the surface film. It is intended that the review presented in this paper may serve as a general overview and provide a guide to what is currently possible and what is being developed for future use.

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To sum up, the modulation spectrum of the specular scattering coefficient (see [2]) is merely linearly related to, and can be transformed into, a sea wave slope spectrum ($=k^2S(f)$, [8]). The relationship between the ocean wave spectra and the (radar or acoustic) image spectra (i.e., the transfer function) is still not satisfactory determined [9]. The modulation spectra of the differential scatter examined in the previous author's papers [2, 3] are derived from the spectra of low-frequency (1–40 Hz) amplitude voltage fluctuations of the ultrasonic signal (10 MHz, acoustic wavelength = 0.15 mm) specularly scattered from a wind-created surface of clean water $U_0(f)$ and covered with an oil substance film $U_c(f)$:

$$S_0(f)/S_c(f)=[U_c(f)/U_0(f)]^2. \qquad (2.1)$$

The mean differential specular scatter $\langle DSC \rangle$ ($=20 \log [\langle U_c(f)\rangle / \langle U_0(f)\rangle]$ can be expressed in decibels in terms of the signal recorded at the receiver $\langle U \rangle$ (averaged over each run).

The authors' main concern being the estimation of the relative levels of the acoustic forward specular scatter in the film-covered water surface region, none of the transmission losses inherent to ocean sound propagation, namely geometric spreading and absorption, have been taken into account. Refraction and reflection at the oil-water interface are also neglected since the film thickness is small compared to the incident wavelength.

The principal characteristics of the scattered acoustic signal fluctuations i.e. the autocorrelation functions, the magnitude of signal amplitude variability and the form of the probability density function (p.d.f.) of signal distribution, depend on the value of the Rayleigh parameter R_a defined for specular scattering [6]:

$$R_a = 2k_a h \cos\delta, \tag{2.2}$$

where $k_a = 2\pi/\lambda$ — the wave number of an acoustic wave of length λ, h — the rms surface wave height.

The value of R_a also determines the statistics of the scattered signal. At small R_a ($R_a \ll 1$) the statistical distribution of amplitudes is Gaussian, at large ($R_a \gg 1$) and intermediate values of R_a, the generalized Rayleigh–Rice distribution, gamma, three-parameter lognormal distributions, etc. were postulated [27, 28]. The variety of p.d.f.s observed in many laboratory and field experiments results from the different character of the water wave motion in each case, as suggested by CLAY *et al.*, [29].

A good approximation of the experimental distribution of acoustic returns is obtained by expanding the Gaussian function $p_n(U)$ into a Gram–Charlier series taking into account statistical moments up to the fourth one. The p.d.f. polynomial function has the form [30]:

$$p(U) = p_n(U)\left[1 + \frac{A_1}{6} H_3(z) + \frac{A_2}{24} H_4(z) + \ldots\right], \tag{2.3}$$

where $p_n(U)$ — the Gaussian distribution, $\langle U \rangle$, U — the mean and temporal signal amplitudes, σ — the standard deviation, $z = [U - \langle U \rangle]/\sigma$ — the normalized random variable, $H_3(z) = z^3 - 3z$ and $H_4(z) = z^4 - 6z^2 + 3$ — Hermite polynomials, $A_1 = \mu_3/\sigma$ — the asymmetry coefficient (skewness), $A_2 = (\mu_4/\sigma) - 3$ — the flattening coefficient (kurtosis), μ_3, μ_4 — the third and fourth central statistical moments.

Parameters A_1 and A_2 describe in a regular way the deviations of the experimental distribution from the normal one, in which values of $\langle U \rangle$ and σ are introduced from the experimental data. The fluctuation coefficient η is a measure of signal amplitude variability and is defined as follows [6]:

$$\eta = \sigma / \langle U \rangle. \tag{2.4}$$

The statistical parameters $\langle U^2 \rangle$, η, A_1, A_2 were chosen for further considerations.

3. Acoustic system and surface film characterization

Laboratory acoustic scattering experiments were performed in a small wind-wave flume described in detail elsewhere [1, 31]. In this paper we are largely concerned with very low stream speeds ($V=1-4$ m/s), for which the flow is aerodynamically smooth, i.e. the thickness of the viscous sublayer [26] exceeds the characteristic height of the surface waves for V below 4 m/s [32].

The scattering measurements were carried out for tank surfaces covered with films of commercially available crude oil products: Diesel oil, light engine oils (Extra 15, Selectol and Hipol 15), a heavy gear oil Lux.

Surface pressures of the monolayers under study were ranging from 3.4 to 4.7 mN/m, that was measured using a Wilhelmy plate method [33] directly through the window in a roof of the flume.

Each monolayer of a film-forming substance has certain viscoelastic rheological surface properties characterized by surface pressure P (a drop of water surface tension due to the film's presence, dilational elasticity modulus $E\ (=-A_0(dP/dA_0)$, where A_0 is the film area), and a structural diffusion parameter ω_d [4]. The magnitude of E depends only on the chemical nature and concentration of the substance composing the film, whereas ω_d is governed by the kinetics and mechanism of the rearrangement process which takes place within an insoluble monolayer during compression/expansion cycles [4].

These parameters were determined in the supplementary Langmuir through measurements by means of the surface pressure-time relationship applying the procedure and experimental set-up proposed by Loglio *et al.* [16, 17], for methodology details see [2, 3]. The surface parameters of oil films studied in this experiment are already collected in Table 1 of Refs. [1, 31].

The acoustic system applied in laboratory studies is based on high-frequency (10 MHz) and forward specular geometry scattering as described in detail elsewhere [1, 31]. The fluctuations of the scattered signal voltage amplitude peak value U are statistically analyzed using a computer IBM-aided system by means of a Statgraphics (ver. 5.0) program by Statistical Graphics Corporation. The p.d.f of the distribution was determined on the basis of 10.240 counts sorted out into 41 amplitude channels.

Studies of the effect of an artificial oil slick on the statistics of ultrasonic signals scattered from a rough sea surface were carried out in October 1989 on the Baltic Sea from aboard an oceanographic platform about 200 m off Oksywie under calm sea conditions ($V_{10}=1.7-2.3$ m/s).

The acoustic system — a free drifting, lightweight buoy — has already been applied in remote sensing and monitoring of polluted sea waters [14, 15], and has the same technical features (driving frequency, scattering geometry, data sampling and processing) as used in the laboratory studies.

It should be pointed out that the occurrence of floats can affect conditions of surface wave generation, result in non-uniform film coverage, and produce an additional turbulent flow over the surface screened by the floats as discussed in [2].

Open-sea scattering measurements were performed on a clean water surface and sea surface covered with films of different petroleum derivatives. Three crude oil products: gear oil Selectol, Gasoline 94, and Ethyline spread from hexane solutions were selected as artificial model film-forming materials in open-sea studies. The viscoelastic parameters characterizing artificial sea slicks were simultaneously measured during each acoustic scattering run using a novel film sampler-surface elastometer [18], to provide "ground truth" for the proper interpretation of the results recovered from remote acoustic surface probing.

4. Results and discussion

Figure 1 presents examples of the p.d.f. plots as a function of normalized random variable z, at different air stream speeds measured for a clean water surface in a wind-wave tunnel. The distributions are of very particular form different from that predicted by the theoretical Gaussian or Rayleigh functions. Although, it has been already found that the experimental points in the case of all both the film-coated and clean water surfaces are much closer to the Rayleigh curve than to the Gaussian one, in which values of $\langle U \rangle$ and σ emerge from the experimental data, as confirmed by the X^2 goodness of fit test by the author [14]. It is in agreement with the results of numerous authors [27, 29].

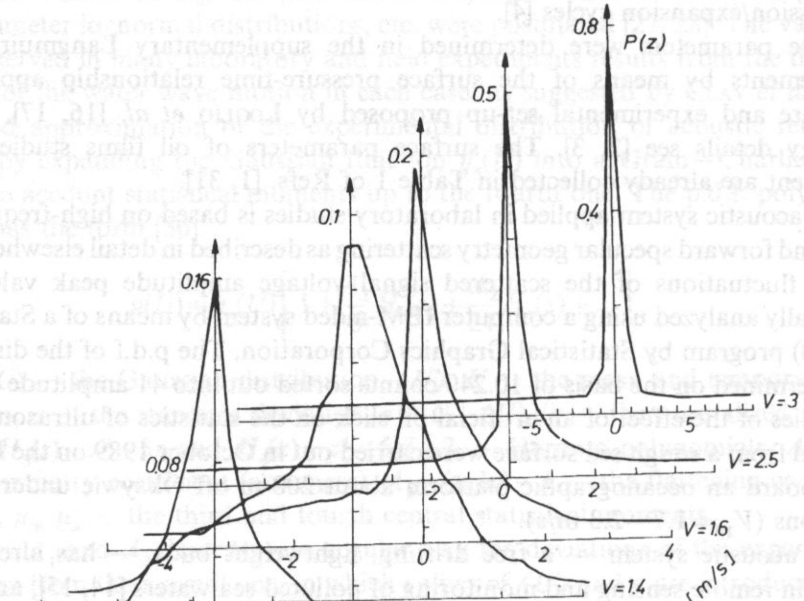

Fig. 1. Evolution of the p.d.f. plots, for a clean water surface as a function of normalized random variable z at different air stream speeds. Laboratory data.

The dependences have sole sharp maxima with wide wings and possess a large left-hand asymmetry. As V increases, a width of the distribution decreases and at the same time a height of the peak grows as well as a left-hand asymmetry is intensified (negative and increasing values of A_1). The effect is apparently related to a particular shape change of water waves being capillary ripples in a small water tank.

It is interesting to note that recent studies on statistics of very large amplitude echoes known as radar sea spikes performed using a ultra-wideband (4 GHz carrier frequency) radar in a wave tank have demonstrated very similar features (see Fig. 7 in [34]). The probability distributions of radar echo power are very peaked (the values of η are of the order of $3-6$) with a longer tail at the high value end. The statistics of open ocean radar sea spikes have been documented empirically as a function of wind speed for both fully-developed and random sea being associated with steep and breaking wave crests [35]. This similarity between the sea spikes and ultrasonic scatter distributions offers an additional argument for a strong dependence of the registered echo on curvature of particular small-scale water waves, and also suggests specular scatter as a dominant mechanism in these experiments in question [34].

In contrast, let us consider exemplary p.d.f.s obtained in at-sea experiments at low wind speeds ($V_{10}=1.7-2.3$ m/s) and depicted in Fig. 2. All the dependences exhibit two distinctly separated local maxima that means they consist of two overlapping each other distributions. This feature seems to support a model of a wavy sea surface of two-spatial scale structure irregularities assuming capillary ripples present on tilted faces of long gravity waves [26]. The film's presence leads to the sharpening of the distribution if compared to the clean reference case but a general shape remains unchanged. It can be noticed that the right-hand maximum for the light oil product (Ethyline) is higher whereas for the havier one (Selectol) the opposite situation is observed. A more detailed interpretation of the observed phenomena awaits precise and direct information on the wavy water surface shape.

Figure 3 presents the mean squared signal voltage amplitude $\langle U^2 \rangle$ as a function of wind speed, for wavy clean water and film-coated water surfaces obtained in laboratory conditions. The scattered power decreases with the wind speed, corresponding to the specular point scattering [36]. In contrast, at large incident angles the backscattered power increases with the wind speed as predicted by the Bragg scattering model [22], that is illustrated in [37], for instance. Higher values of $\langle U^2 \rangle$ for film-coated surfaces if compared to the clean surface reference case, exhibit the smoothing effect of wind waves by surface films attributed to the Marangoni phenomenon. Its strength depends on the ratio (dilational elasticity/surface pressure of the film i.e., E/P) as already discussed in [1]. This ratio is ranging from 2.1 (Hipol) to 4.1 (Selectol).

The mean differential scattering coefficient $\langle$DSC$\rangle$ as a function of wind speed, for several oil substance-coated water surfaces again studied in laboratory conditions is presented in Fig. 4. The $\langle$DSC$\rangle$ dependence increases continuously as wind speed increases in the range $1.3-3.5$ m/s. It suggests that the most convenient wind speed range for further at-sea applications of such a system would lie at rather higher V_{10} where values of $\langle$DSC$\rangle$ are of the order of $10-15$ dB but under the limiting V_{10} ($\cong 7$ m/s, [26]) indicating breaking of wind waves and consequently film disruption.

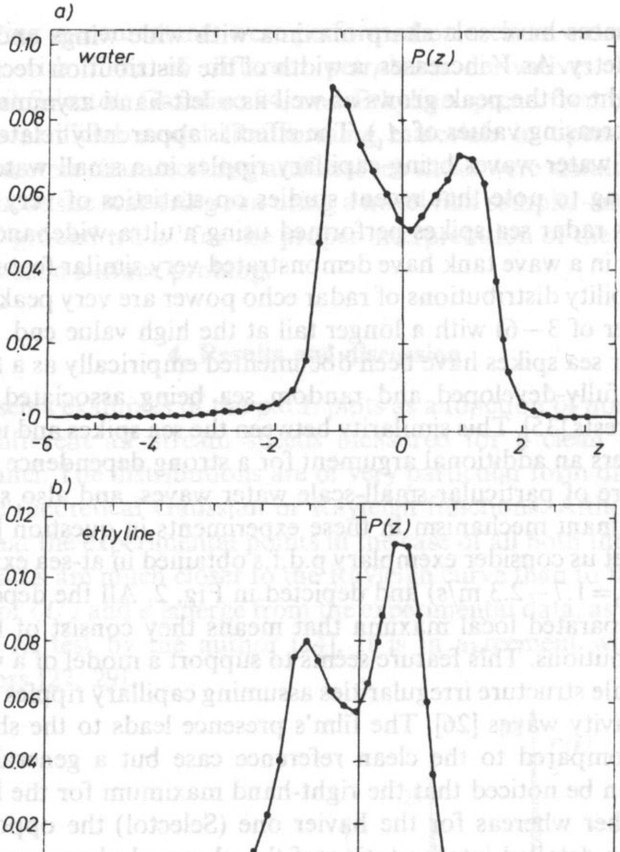

Fig. 2. Exemplary p.d.f.s. for clean (*A*), Ethyline (B), and Selectol (C) covered water surfaces. At-sea experiment data.

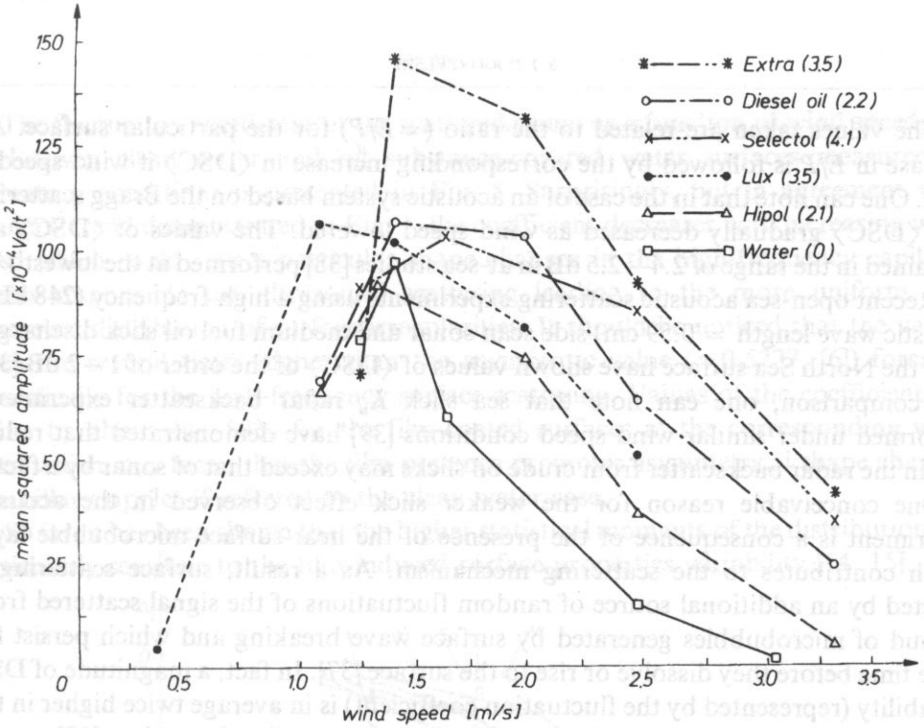

Fig. 3. Mean squared amplitude $\langle U^2 \rangle$ of the ultrasonic signal scattered at a wind-driven surface of clean water and covered with oil substance films versus air stream speed. The ratio E/P of each film is given in brackets. Laboratory data.

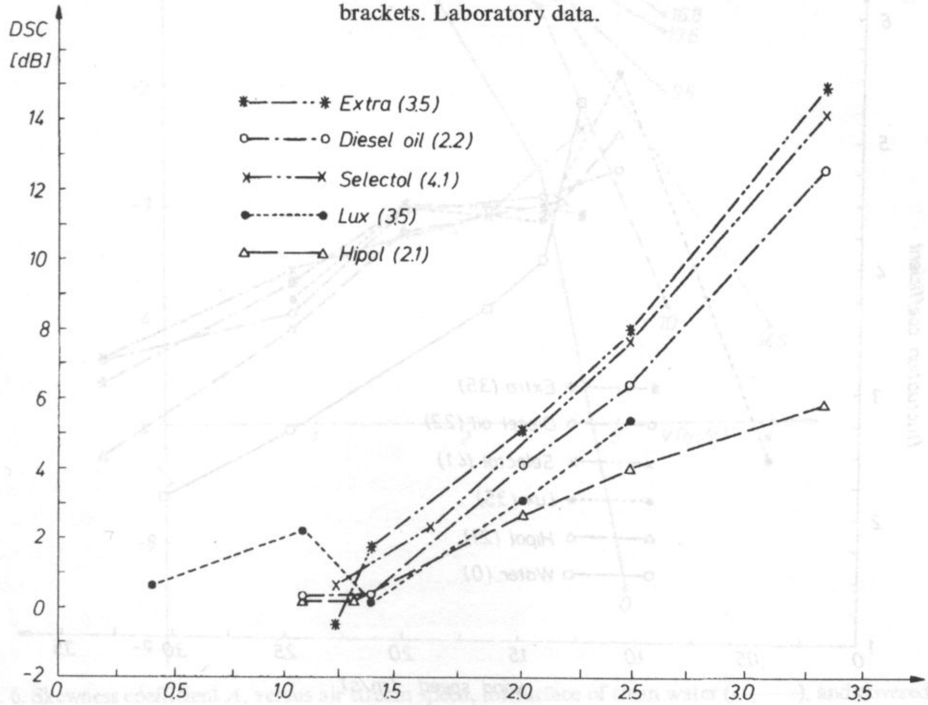

Fig. 4. Mean differential scattering coefficient $\langle DSC \rangle$ as a function of air stream speed. Denotations as in Fig. 3. Laboratory data.

The values taken are related to the ratio ($=E/P$) for the particular surface. An increase in E/P is followed by the corresponding increase in ⟨DSC⟩ if wind speed is fixed. One can note that in the case of an acoustic system based on the Bragg scattering [37], ⟨DSC⟩ gradually decreased as wind speed lowered. The values of ⟨DSC⟩ are contained in the range of 2.4 – 2.5 dB in at-sea studies [38] performed at the lowest V_{10}.

Recent open-sea acoustic scattering experiments using a high-frequency (248 kHz, acoustic wave length = 0.59 cm) side scan sonar and medium fuel oil slick discharged onto the North Sea surface have shown values of ⟨DSC⟩ of the order of 1 – 2 dB [37]. For comparison, one can note that sea slick K_u radar backscatter experiments performed under similar wind speed conditions [39] have demonstrated that reduction in the radar backscatter from crude oil slicks may exceed that of sonar by a factor 5. One conceivable reason for the weaker slick effect observed in the acoustic experiment is a consequence of the presence of the near-surface microbubble layer which contributes to the scattering mechanism. As a result, surface scattering is affected by an additional source of random fluctuations of the signal scattered from a cloud of microbubbles generated by surface wave breaking and which persist for some time before they dissolve or rise to the surface [37]. In fact, a magnitude of DSC variability (represented by the fluctuation coefficient) is in average twice higher in the open-sea experiment than in laboratory studies as shown by the author [38].

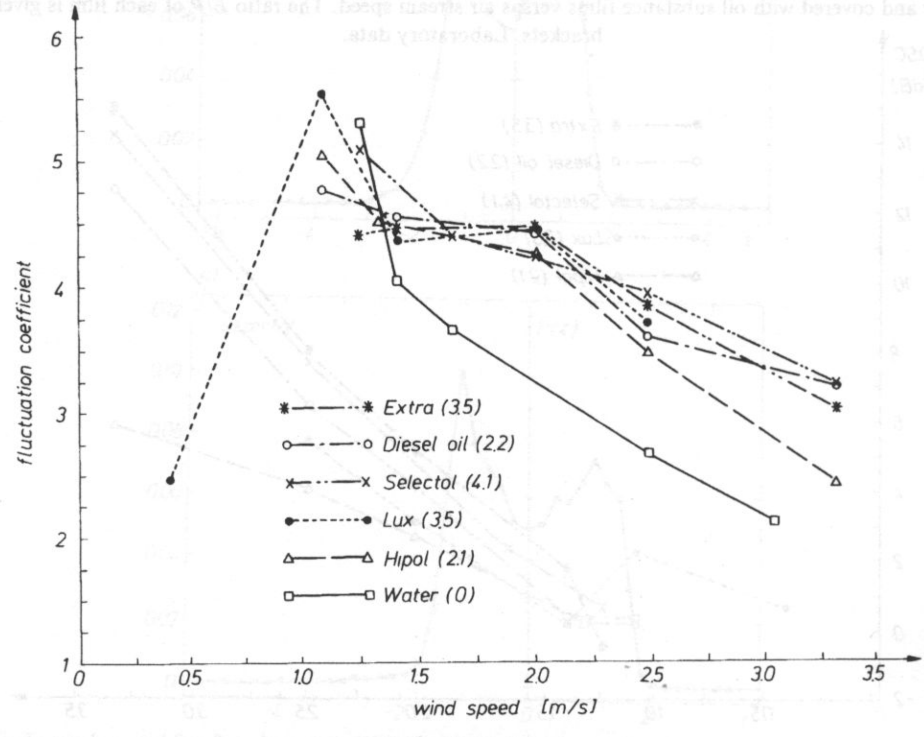

Fig. 5. Fluctuation coefficient η of the scattered signal amplitude variability as a function of air stream speed. Denotations as in Fig. 3. Laboratory data.

The fluctuation coefficient of the scattered signal as a function of wind speed, for wind-driven clean water and oil substance-covered water surfaces measured in laboratory conditions is presented in Fig. 5. Surprisingly, but in agreement with a general trend demonstrated in Fig. 1, the coefficient decreases with increasing wind speed which could reflect particular shape changes in the high-frequency capillary ripples responsible for ultrasound scattering leading to the more uniform and symmetric distribution of surface irregularities. It should be noticed that the values taken are several times higher than the asymptotic value (=0.5227, [6]) foreseen theoretically for the high-frequency surface scattering. Values of the coefficient are higher by about 40–50% for the film-coated surfaces at the corresponding wind speed which may mean that the film presence promotes asymmetrical shape changes in capillary ripples if referred to the clean water case.

So far, it has been shown that the higher statistical moments of the distribution are particularly sensitive to the film-induced surface properties variability [14, 15].

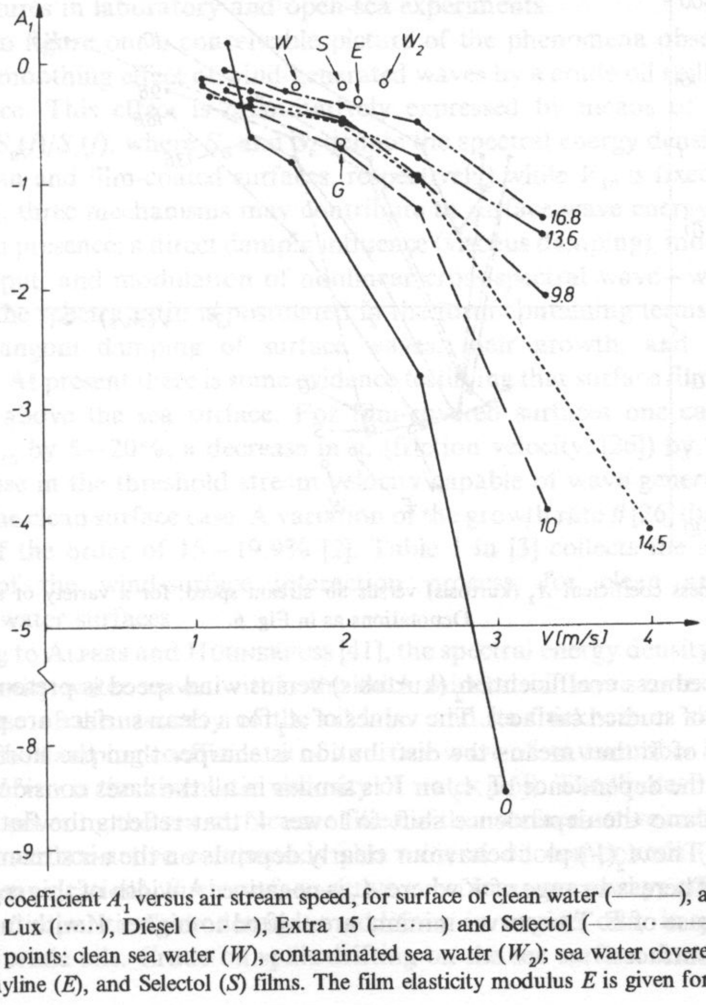

Fig. 6. Skewness coefficient A_1 versus air stream speed, for surface of clean water (———), and covered with Hipol (— — —), Lux (······), Diesel (– · –), Extra 15 (——···—) and Selectol (———···———) oils. Open-sea experiment data points: clean sea water (W), contaminated sea water (W_2); sea water covered with Gasoline 94 (G), Ethyline (E), and Selectol (S) films. The film elasticity modulus E is given for each film.

Figure 6 presents the asymmetry (skewness) coefficient A_1 as a function of V for clean and film-coated water surfaces. For all speeds, the coefficient has negative values that means that the distribution has a strong left-hand asymmetry if compared to the normal reference one, in which values of $\langle U \rangle$ and σ are introduced from the experiment data. An increase in V intensifies a left-hand asymmetry ($| A_1 |$ grows). The presence of an oil film symmetrizes the distribution (the values of A_1 are higher and closer to zero for the covered surfaces, while the air speed is fixed). The difference between the curves for the control and film-coated surfaces evidently depends on the film elasticity modulus E (given in the end of each curve), and increases with increasing E.

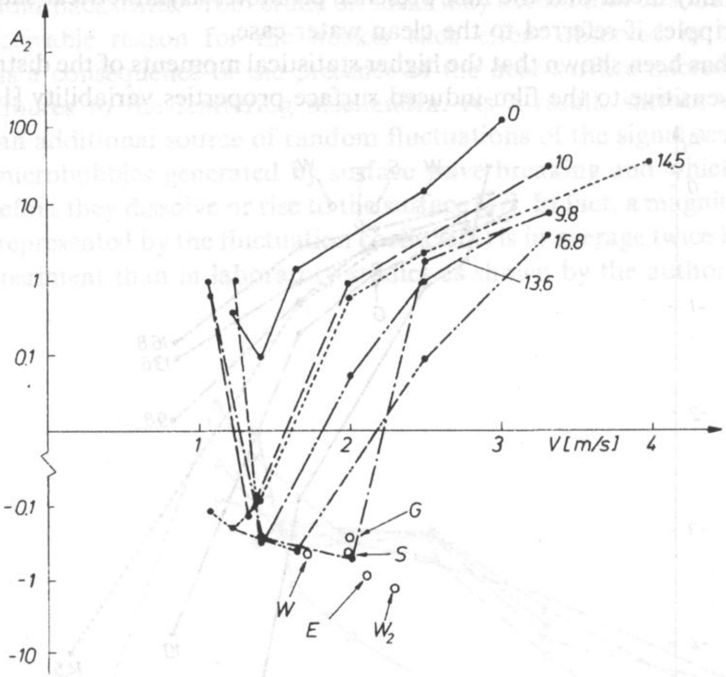

Fig. 7. Peakedness coefficient A_2 (kurtosis) versus air stream speed, for a variety of studied surfaces. Denotations as in Fig. 6.

The peakedness coefficient A_2 (kurtosis) versus wind speed is presented in Fig. 7, for a variety of studied surfaces. The values of A_2 for a clean surface are positive in the whole range of V that means the distribution is sharper than the normal one. The character of the dependence of A_2 on V is similar in all the cases considered. The film presence leads to the dependence shift to lower V that reflects the flattening of the distribution. The $A_2(V)$ plot behaviour clearly depends on the air stream speed range in question. There is a range of V where A_2 is negative. A width of this region expands with an increase of E. The curves minima are shifted to higher V with increasing E of the covered surfaces.

The assumption that the experimental distribution can be approximated by the polynomial function according to Eq. (2.3) seems to be very rough. Since absolute values of both A_1 and A_2 are lower than 1 only in the lowest speed range, thus for stronger winds the statistical moments of higher orders than the fourth one should be included in order to reproduce the distribution in a more representative form.

Figures 6 and 7 also contain data points corresponding to the open-sea experiment. They are located very close to the laboratory dependences which reflects selfconcidence of both scattering experiments and suggests that artificial sea slicks formed films of comparable elasticity moduli ranging from 9.8 to 16.8 mN/m as laboratory-studied surfaces did. In fact, the values of E for artificial sea slicks determined "in situ" were equal to 7.1 mN/m (Ethyline 94), 9.2 mN/m (Gasoline), 15.1 mN/m (Selectol), and 9.4 mN/m (contaminated sea water region). Moreover, it seems that capillary ripples responsible for high-frequency scattering have similar statistical features in laboratory and open-sea experiments.

In order to figure out a conceivable picture of the phenomena observed, let us consider the smoothing effect of wind-generated waves by a crude oil spill floating on the sea surface. This effect is quantitatively expressed by means of the relative spectrum ($=S_0(f)/S_c(f)$, where S_0 and S_c denote the spectral energy densities of wind waves for clean and film-coated surfaces, respectively) while V_{10} is fixed [5].

In general, three mechanisms may contribute to surface wave energy dissipation due to the film presence: a direct dampig influence (viscous damping), modification of wind-wave input, and modulation of nonlinear cross-spectral wave–wave interactions. In [3], the spectra ratio is postulated in the form containing terms responsible for the Marangoni damping of surface waves, their growth, and spatial film homogeneity. At present there is some evidence testifying that surface films transform a wind field above the sea surface. For film-covered surfaces one can notice an increase in V_{10} by 5–20%, a decrease in u_* (friction velocity, [26]) by 7–9%, and a large increase in the threshold stream velocity capable of wave generation [40] in reference to the clean surface case. A variation of the growth rate β [26] due to the film presence is of the order of 15–19.9% [2]. Table 1 in [3] collects the aerodynamic parameters of the wind-surface interaction process, for clean and different film-covered water surfaces.

According to Alpers and Hühnerfuss [41], the spectral energy density of capillary and short gravity water waves within the thick mineral oil layer zones is modified mainly because of the viscosity of the oil layer and its thickness as shown by the author [42]. The damping coefficient α of a surface wave of wavenumber k follows the k^2 law ($\alpha=4k^2\nu$, ν is the kinematic viscosity of water [26]). The "holes" between the thick patches covering the sea surface are filled with a surface-active material and/or a mixture of surface-active compounds plus mineral oil compounds (Hühnerfuss, personal communication). Such surface-active substances are always encountered in crude oil as "impurities" or detergent additives in engine oils, in particular in "weathered" crude oils. Crude oil spills drifting on the sea surface do not consist of pure hydrocarbon fractions but they contain considerable amounts of surfactant-type materials which are being formed by photo-oxidation processes and bacterial decomposition [43]. They tend to spread very easily from the thick oil spill centers over the surrounding sea surface. Only within the sea surface area covered with these monomolecular organic films surface tension gradients and thus the Marangoni wave damping can be induced in a rough surface. The result is the resonance-like behaviour of the damping coefficient α as a function of surface wave frequency f. The $\alpha(f)$ dependence attains a maximum in the frequency range 3 – 20 Hz i.e., for capillary and short gravity waves, depending on the elastic properties of the spread film [5, 41]. The relative spectrum is postulated in terms of the film viscoelasticity (E and ω_d parameters), and the film homogeneity represented by the film filling factor F [3].

It has been found for natural films that F appears to be a linear function, as a first approach, of wind speed V_{10} [44]. It is believed that artificial crude oil films follow the same $F(V)$ pattern as naturally-formed ones do. The modulus E playing a principal role in the intensity of the damping effect is often assumed to be single-valued. In nature, sea surface films experience high surface pressure variations of the order of 12 – 14 mN/m, that leads to the formation of multilayers and phase transitions within the film, and is accompanied by a steep rise in the elasticity modulus E. It would appear that the aforementioned assumption can lead to a wrong result if introduced to the Marangoni damping relation.

As a consequence, two zones exhibiting different mechanisms of wave attenuation can be specified within a mineral oil spill.

The properties of surface in dilation and compression, as expressed by their dilational moduli E, have so far been studied only for uniform homogeneous surfaces. In real systems, however, surfaces are often non-uniform. Such composite surfaces can contain floating particles, bubbles or droplets of a third phase. Floating solid particles in fluid-fluid interfaces will interact with one another if they have an irregular wetting perimeter which disturbs the smoothness of the interface. Any deformation of this system (liquid surface covered with "rough-edged" solid particles) will be resisted, and this leads to finite values of E even in the total absence of a surfactant [19, 20]. Thus the anticipated dilational modulus is of the order of the surface tension multiplied by the square of the ratio between the roughness amplitude and particle size. For the simplest case of non-interacting floating particles, the smaller the fraction of the interface which is not covered with particles, then the larger is the perceived modulus of the entire interface. For a more complicated system with solid particles, which can cover a variable area, the particle will adopt a position in a fluid-fluid interface which is determined by its contact angle ϑ. While the range of contact angles for natural particles in seawater is not known yet, the advancing contact angles for various other substances have been reported [45], and include Teflon (98 – 112°), polyethylene (88 – 103°), paraffin (110°), TFE-methanol telomer wax (100 – 160°) and human skin (90°). It may be concluded that the presence of partially-wetter ($\vartheta < 180°$) spherical particles can either increase or decrease the apparent modulus of the whole surface [20]. Thus, when the contact angle is close to

90° or when the ratio between the modulus E (surface without particles) and surface tension T of the air-liquid interface is small, there will be an increase.

The effect of contact angle on the composite modulus is illustrated in [20], and on the basis of a number of assumptions (number of particles per $mm^2=50$, particle radius = 50 μm) at $\vartheta=90°$, an increase by 60% of the composite modulus is predicted where $E/T=0.1$.

For oil droplets in air-water interfaces, when easily extensible droplets emerge into the surface, we find for the composite modulus a sudden decrease. This decrease is more pronounced when the area covered with the droplets is larger. It seems likely that capillary interaction plays a large and underestimated role in the various interfacial systems what should be borne in mind while interpreting any remote sensing data based upon the smoothing effect of the sea surface due to the Marangoni damping. Several ideas presented above are in agreement with studies of Ebuchi *et al.*, [46]. They have concluded that fine structures of short wind waves surfaces (a 3-dimensional rhombic structure, a train of capillary waves on the forward face with wavelengths gradually decreasing with distance from the crest, and a streaky structure on backward face in the direction of the wind) are not symmetrical with the wind direction but are the main contributor to high-frequency scattering. In the specular point scattering model the normalized cross section is proportional to the probability density of surface slope normal to the incident direction. Usually, the distribution of the surface slopes is assumed to be the Gaussian one, which may no longer be valid for the mentioned surfaces. So there is still a great need for research to understand signatures of the ultrasound specular scatter at "real" sea water surfaces.

Further conceivable sources of the discrepancy between the acoustic data and theoretical predictions may be related to the simplified model of the air-sea interaction commonly adopted but neglecting the surface drift effect on the wind wave growth, parasitic capillary and instability waves (known as "cat's paws") which increase more rapidly than regular waves and are expected to roll up and to form breakers at very modest steepness [47, 48].

In addition, nonlinear interactions can affect the shallow-water spectrum of wind waves. The offshore frequency spectrum is bimodal with peaks at swell f_1 and sea f_2 frequencies, whereas the observed shallow-water spectrum has additional peaks at frequencies corresponding to harmonics $2f_1$, $2f_2$, $3f_1$... and combination tones f_1+f_2, $2f_1+f_2$, f_1+2f_2... of the deep-water swell and sea [49]. As a result, the relative spectrum may not be a smooth function of frequency.

It may be helpful to add that the applied directional ultrasonic system consisting of two transducers based on a forward specular scattering geometry has two features of significant importance in future applications at sea [1, 2], and can provide far better resolution in studies of small spatial scale air–sea interaction processes taking place at the sea surface than radar and may be more convenient and less costly to use.

5. Conclusions

In general, the scattered ultrasonic signal signatures can be explained in the framework of the "specular point" scattering theory applicable also to laser and microwave surface probing.

The signal distributions are of very particular form that differ significantly from any of the standard functions (Gaussian or Rayleigh). So, for stronger winds the statistical moments of higher orders than the fourth one should be included in the Gram – Charlier series.

As V increases, a width of the distribution decreases and at the same time a height of the maximum grows that is apparently related to a particular shape change of water waves being capillary ripples in a small water tank. Moreover, the fluctuation coefficient decreases with increasing winds that points to a more uniform or symmetric distribution of surface irregularities. Although, the values of η are higher by about 40 – 50% for the film-coated surfaces, while the wind speed is fixed, which may mean that the film presence promotes asymmetrical shape changes in capillary ripples in reference to the clean surface case.

In contrast, all the distributions registered in open-sea experiments exhibit two distinctly separated local maxima that means they consist of two overlapping each other distributions. This feature seems to support a commonly accepted model of a wavy sea surface of two-spatial scale structure irregularities assuming capillary ripples present on tilted faces of long gravity waves.

The smoothing film effect expresses itself in the wind speed dependences of $\langle DSC \rangle$. The particular values taken are related to the ratio E/P (elasticity modulus/surface pressure of the film), and an increase in E/P is followed by the corresponding increase in $\langle DSC \rangle$.

The presence of an oil film symmetrizes the distribution the values of A_1 are higher and closer to zero for the covered surfaces in reference to the clean surface case, and leads to the flattening of the distribution (the values of $|A_2|$ are lower for the coated surfaces) at the fixed V. The difference between the wind speed dependences of A_1 and A_2 for the control and film-coated surfaces depends on the film elasticity E, and increases with increasing E.

The open-sea experiment data points are located very close to the laboratory $A_1(V)$ and $A_2(V)$ dependences which reflects selfcoincidence of both scattering experiments, suggests the same range of E taken by the artificial sea slicks, and points to the similar statistical features exhibited by capillary ripples in the laboratory and open-sea experiments.

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