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MODULAR SURFACE LIGHT SCATTERING
INSTRUMENT WITH AUTOTRACKING OPTICS
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A preview of a modular surface light scattering instrument with autotracking optics

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ABSTRACT

NASA's Advanced Technology Development (ATD) program is sponsoring the development of a new generation of surface light scattering hardware. This instrument is designed to non-invasively measure the surface response function of liquids over a wide range of operating conditions while automatically compensating for a sloshing surface. The surface response function can be used to compute surface tension, properties of monolayers present, viscosity, surface tension gradient and surface temperature. The instrument uses optical and electronic building blocks developed for the laser light scattering program at NASA Lewis along with several unique surface light scattering components. The emphasis of this paper is the compensation for bulk surface motion (slosh). Some data processing background information is also included.

INTRODUCTION

Surface light scattering provides a way to non-invasively measure surface tension and viscosity. It can also be used for non-contact temperature and surface tension gradient measurements at the surface of a fluid interface. The fluid interface can be either liquid-liquid or liquid-vapor and may include a monolayer. We will begin with a brief description of this technique and its limitations, then discuss the individual optical and electronic components developed to build this instrument. We will discuss the overall instrument design options and our work with unique autotracking optics which compensate for a sloshing surface, a persistent difficulty in surface light scattering research. Conventional optical tables are sufficient for traditional surface light scattering work only if the environment is quiet. The effects of building vibration and sound noise from the environment can degrade the signal to noise ratio

substantially unless the cell is carefully isolated. Even with cell isolation, low frequency driven waves, which we call slosh, are hard to suppress. Our autotracking optics techniques are still evolving, and we will report the results we have obtained up to this point.

DESCRIPTION OF HOW SURFACE LIGHT SCATTERING WORKS

When laser light is reflected from or transmitted through a fluid surface, it is interacting with a weak phase grating created by thermal fluctuations of the interface. These surface waves can be separated into a Fourier sum of many different wavelengths since the amplitude to wavelength ratio is small enough that a linear response is obtained. The optical instrument studies a narrow range of wavelengths as the small surface waves move across the surface, damp out and are born anew due to the thermal fluctuations which occur on any surface (since all real surfaces have a temperature above absolute zero). Consider one of these wavevectors (k-values) on the surface to illustrate how the surface light scattering technique works. (When taking data from a real physical system, instrument broadening requires a generalization of this discussion from a single wavelength ripple to a narrow band of wavevectors.) This illustrative monochromatic, capillary ripple travels along the surface and damps out in time if the fluid is viscous and the interface is not being driven by an external force. If forced, the ripples will damp in space. During its existence, this traveling wave acts as a phase grating which diffracts light beams so that they appear at the detector as weak diffuse rings of light around the reflected or transmitted laser beam. A two-dimensional slice of this intensity profile looks like the diffraction from a phase grating (a series of periodically spaced lines produced by variations in the index of refraction with no absorption). Its intensity profile at a Fraunhofer distance from the surface looks something like Figure 1. The diffraction pattern is given by a number of displaced spots. The strength of each spot is given by the Bessel function evaluated at the modulation depth of the grating (sinusoidal). A slice through the center of this distribution has a bright central spot with dimmer spots placed symmetrically on either side. The diameter of each spot is inversely proportional to the diameter of the beam that illuminates the grating. For a weak grating the zero-order dominates; +1 and -1 may be visible. Higher orders are negligible.

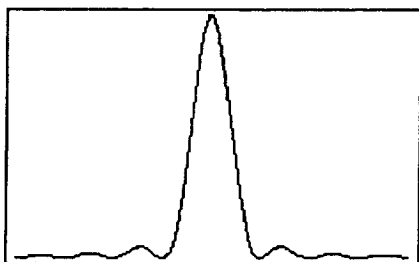


Figure 1

The diffraction pattern from a monochromatic capillary ripple recorded at a Fraunhofer distance from the liquid surface

The central peak, which is the undiffracted laser beam, is the zero-order spot, and the first set of side lobes are the first-order diffraction spots, etc. The first-order diffraction spots intensity fluctuates in time as the wave travels across the surface. The first-order diffraction spots for a particular wavelength on the surface are usually very weak (10^{-15} Watts)¹ and the detector and electronics would have to respond in the visible light range at around 5×10^{14} Hz if we did not beat the signal from the first-order spot with a local oscillator at the detector. For this and other reasons we cannot directly detect the frequency in this frequency range. By using acousto-optic modulators², we can drop the detected signal into the kilohertz range or shift it into the megahertz range to avoid laser and other

noise. This can be accomplished with or without an external grating (a grating outside the liquid-fluid surface). A stable reference spot can be attained by placing a holographic phase grating before or after the surface, thereby reflecting or transmitting its zero- and first-order spots, which have a known k-value (wavevector), onto the detector. The reflected first-order spot of the external grating is normally what we use to select a known k-value at the detector. The zero-order spot (incident laser beam) is scattered by the surface at many different k-values, each of which scatter at their appropriate angle according to the momentum balance at the surface which results in $k = k_0 |\sin \theta_m - \sin \theta_{sc}|$. By using the visible first-order spot generated by a grating, we can pick a position with our detector of known k-values being scattered from the surface. This also defines the direction of the capillary waves that we are looking at. These waves are only from 0.3nm to 1nm in amplitude; and the light scattered by the zero-order spot (the incident laser beam) is detected at the position of the first-order spot which acts as a reference beam. The

detector is a photodiode which is a square law detector and which provides the beat frequency signal. We use holographic phase gratings of 240 cm^{-1} to 1500 cm^{-1} . Our 240 cm^{-1} gratings have an efficiency range from 70:1 to 2650:1. This amounts to a measure of the intensity of the zero-order spot to the first-order spot. Earnshaw¹ has pointed out that lower efficiencies are needed for accurate measurements with gratings of low k -values (e.g. 240 cm^{-1}). Additionally, if the first-order spot is too bright (if we have an efficiency higher than 100:1, e.g. 30:1), then the DC offset at the detector is so large compared to the amplitude of the fluctuations in the signal that the detector is swamped, and the superposition of the first-order diffraction spot with the scattered light no longer has a discernible beat signal.

We use band pass filtering to block the DC and low frequency noise before the signal is autocorrelated. While Earnshaw¹ has shown that photon counting techniques can be used, the small modulation depth of the signal makes his scheme awkward. While photon counting has proved superior for volume scattering, the advantage does not carry over to surface scattering. For this reason it is convenient to have a correlator with an analogue input to AC couple to the detector signal. Brookhaven Instruments has modified the BI-9000 correlator which they developed for the laser light scattering program at NASA Lewis. Their BI-8500 correlator has analogue inputs and a zero channel which allows the signal to noise ratio of an analogue signal to be measured directly. Unlike volume scattering, the optimization of the spectrometer is easy if the zero channel is calculated as part of the time series manipulation built into the correlator. Because the correlation function of volume scattering is exponential, there is little interest in knowing the signal to noise ratio accurately. We have chosen to use a correlator instead of a power spectrum analyzer because the correlogram is computed from the photocurrent time series and is directly done by the circuits in the correlator. (We fit to the correlation function and use the transform to the spectrum as quality control in the sense of minimizing unwanted contributions from electronic noise.) The fluctuations of photocurrent forming the time series in current can be viewed as the result of looking at an ensemble of traveling waves moving across the surface. These waves are generated by point-like pressure fluctuations at the interface.

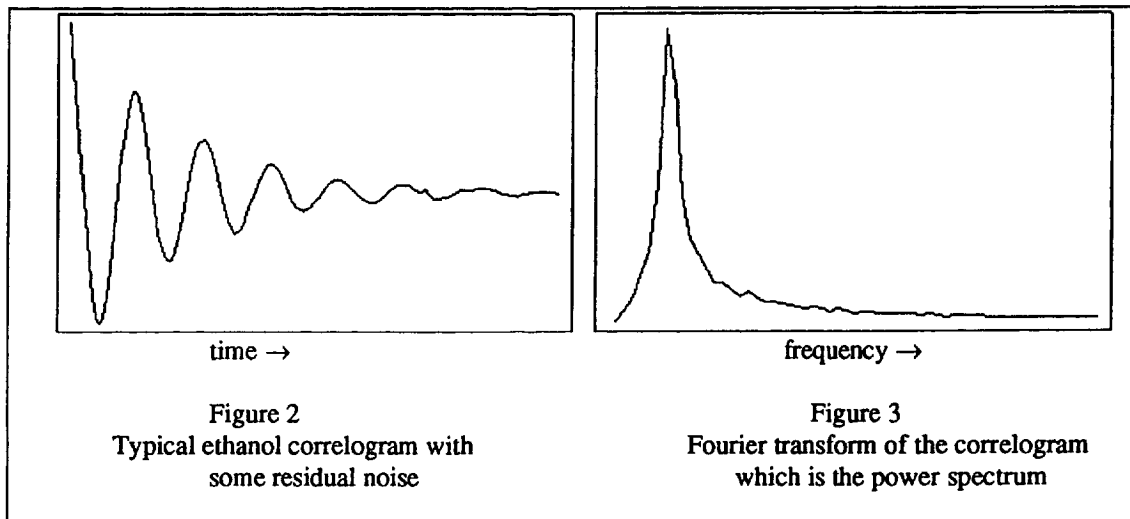


Figure 2 is a typical correlogram with some residual noise. This correlogram was obtained for ethanol with a BI-2030 correlator when transmitting the laser beam once through the surface. This should look like an exponentially damped cosine. The spacing between the peaks is an indication of the surface tension and the damping is an indication of the viscosity. The Fourier transform of the correlogram which is the power spectrum appears next to it in Figure 3, where the power spectrum is the power per unit frequency.

The correlation determined in the experiment is that of the photocurrent i and is $R_i(\tau) = \langle i(0) i(\tau) \rangle$, where $\langle \dots \rangle$ implies averaging over an equilibrium ensemble. Actually, the correlator estimates the time-average,

but it is assumed that the stochastic process is ergodic so that the experimentally determined time average, $\overline{i(o)i(\tau)}$, is equivalent to $\langle i(o)i(\tau) \rangle$. This is equivalent to computing $R_q(\tau) = \langle \zeta_q^*(o) \zeta_q(\tau) \rangle$ where $\zeta_q(\tau)$ is the complex amplitude of the mode with wave number q for this heterodyne experiment. However, there is an additional complexity in that there is an instrument function that is "convolved" with $R_q(\tau)$ to give $R_i(\tau)$:

$$R_i(k_e, \tau) = \frac{1}{2} \iint \langle \zeta_q^*(o) \zeta_q(\tau) \rangle F^2(k_e - q_x, q_y) dq_x dq_y \quad (1)$$

Where the instrument function, F , is

$$F(k_e - q_x, q_y) = \exp \left[-\frac{(k_e \tau)^2}{2\epsilon^2} \left(1 - \frac{q_x}{k_e} \right)^2 \right] \exp \left[-\frac{\sigma^2}{2} q_y^2 \right] \quad (2)$$

and σ is the Gaussian beam width at the surface, $\epsilon^2 = \cos^2 \theta$ where θ is the angle of incidence measured from the normal, k_e the grating wave number as seen at the surface and the ripple wave number is $q = (q_x, q_y)$ with $q = |q|$. The physical idea is that if the beam diameter is too small, R_i will be badly distorted; R_i will decay in time much faster than $R_q(\tau)$. When $k_e \sigma$ is sufficiently large, $k_e \sigma > 100$, F behaves as $\delta(k_e - q_x, q_y)$ and $R_q(\tau)$ is well represented by $R_i(k_e, \tau)$ with $q = k_e$. This integration can be approximated numerically using a simple quadrature formula based on Laguerre polynomials. See Abramowitz and Stegun³ formula 25.4.45 (25.4.46 can work as well). Notice that if the Fourier transform is done on τ , the spectrum function can be calculated with the same formula.

Models for $R_q(\tau)$ have been constructed and the algorithm we use is given in papers by Mann and Edwards when the capillary number is of a magnitude that implies that the ripples are propagating. Of use in reading this paper is the dimensionless group $Y_1 = (\rho + \rho') \omega^2 / \gamma k^3$, where $\rho + \rho'$ is the sum of the phase density, γ the surface tension and $\omega = 2\pi f$ the center frequency of the spectrum. In practice Y_1 varies from about 0.9 to 1.1 where $Y_1 = 1$ is the case for ideal fluid behavior. Y_1 is computed from the full dispersion equations. Thus the center frequency tracks the surface tension reasonably well even for real fluids. Viscous dissipation must be taken into account and so must the properties of any monolayer adsorbed or spread at the surface. A second dimensionless group that must be kept in mind when interpreting either the autocorrelation function or the spectrum is a capillary number, N_{CP} , that emerges from non-dimensionalizing the surface equations of motion:

$$N_{CP} = \frac{\Gamma_0}{2\omega_0} \quad (3)$$

where $\Gamma_0 = 2 \frac{\eta' + \eta''}{\rho' + \rho''} k^2$ is the time damping of capillary ripples moving on a slightly viscous phase and

$\omega_0^2 = \frac{\gamma k^3}{\rho' + \rho''}$. The numbers η' , η'' are the viscosities of the two phases, ρ' , ρ'' are the densities and γ is

the surface tension. For extreme capillary numbers, either large or small, the spectrum of fluctuation is Lorentzian to a good approximation; split from $\omega = 0$ for $N_{CP} \ll 1$, overdamped for $N_{CP} \gg 1$. Dispersion equations have been derived for computing the center frequency and width of the spectrum. See Reference 4 for the most general dispersion equation.

When the capillary number is close to one, the spectrum deviates from the Lorentzian line shape. A full response function theory is available⁵. This theory has not been used often; Earnshaw advocates its use routinely. In principle, a non-linear least squares procedure fitting the experimental autocorrelation function (or spectrum) with the full response function formula "convolved" with the instrument function will allow evaluation of more than the two parameters of the Lorentzian fitting function (center frequency and width). However, when N_{CP} is less than about 1/20 and greater than 20, the fit is effectively limited to two parameters by a sensitivity analysis.⁶ We leave details for data analysis to the references, e.g. - 5, 6,

7, 15 and 16. Reference 16 gives a complete formulation of the convolution problem to correct for instrument broadening; however we now work with the integral representation directly rather than the approximation scheme discussed in Edwards, et al.; modern personal computers handle the integration quickly enough for practical non-linear least squares fitting. Various researchers are using the dispersion equation published by Kramer.⁸ There is an error of interpretation of his equation which is due to an inappropriate choice of the form of the stress tensor; what he calls the compression modulus K is in fact a sum of the surface shear and dilation coefficients. Compare material in references 4, 8, 9 and 12.

Light scattering spectroscopy can be used to measure the temperature of a liquid interface. This non-contact method was outlined in reference 10 and is based on the observation that the surface ripple spectrum depends on temperature. The center frequency depends on the surface tension through γ_1 , and the surface tension depends on temperature. It is straight forward to calibrate the frequency shift with temperature. Such a frequency shift is observed for the foot-print of the laser beam on the surface; a local temperature is reported. The spatial limitation of resolution is that $k\sigma$ should be larger than about ten to twenty. Only the center frequency is required so that $k\sigma$ may be smaller than what is required to deconvolve the instrument function. This method has possibilities for several space applications including the determination of the surface temperature of levitated drops. The true surface tension is determined, not that of the microlayer.

The theory and practice of surface light scattering are also discussed in a number of papers, for example, 11, 12, 13, 14, 15, 16, 17 and in a recent book by Langevin (Ref. 5).

INDIVIDUAL HARDWARE COMPONENTS

To take data to determine the surface amplitude correlation function, we use a laser diode which emits at 775 nm (near infrared) and is capable of 100 mWatts CW output power. This diode laser is the Spectra Diode SDL-5412-H1, where H1 indicates that the laser is mounted on a temperature controlling Peltier cooler. This is a single mode laser diode; but as Jørgensen has pointed out, this is unnecessary. A multi-longitudinal mode laser diode is satisfactory so long as it supports a sufficient number of modes.¹⁸ Lading's group has shown that an investment in battery powered laser diodes, detectors, and preamplifiers helps to reduce noise when taking data. Noise of the laser (not photon noise) sets the limit to how strong a reference beam can be applied.

A pin diode detector makes a fine analogue detector. It is inexpensive and provides better signal to noise than either a photomultiplier or an APD when the signal is not weak¹⁹. United detector makes a hybrid photodetector/preamplifier combination (e.g. - UDT455) which is less than \$50 (U.S.), and this can be coupled to an inexpensive homemade instrumental amplifier circuit or a low noise amplifier available from United Detector (e.g. - UDT1000A) which costs less than \$1K. We use a low noise, battery powered EG&G 9551 preamplifier which contains the necessary low-pass and high-pass filters to control the band pass and costs about \$1500 (U.S.). Care must be taken when selecting a detector to ensure that it does not contain low frequency noise (or a high-pass filter) which will not allow you to detect signals in the frequency range between about 100Hz and 100kHz.

VIBRATION AND AUTOTRACKING OPTICS

Surface light scattering instruments are sensitive to vibrations for a number of reasons. Since we are using a heterodyning technique to detect the signal from the ripples, any movement of the local oscillator signal with respect to the scattered light will cause spurious beats at the detector. In practice this is usually not a problem when a grating is used. An even more delicate problem is the sloshing of the surface due to vibrations. When a large light pipe (around 3 mm in diameter) is used in a vibration free room, the first-order spot can be maintained on the entrance to the light pipe, while still rejecting the zero-order spot. As the spot moves around at larger angles on the entrance to the light pipe, a larger selection of k -values is seen at the detector and more often than not the zero-order spot falls onto the light

pipe and swamps the detector (which causes additional worry for the researcher using a PMT). When the surface sashes, the first-order diffraction spot from a grating will dance around so much that it is nearly impossible to track it to get a signal to correlate. When the sloshing on the surface can be accounted for or compensated for, we suddenly have an extremely robust instrument which is vibration insensitive.

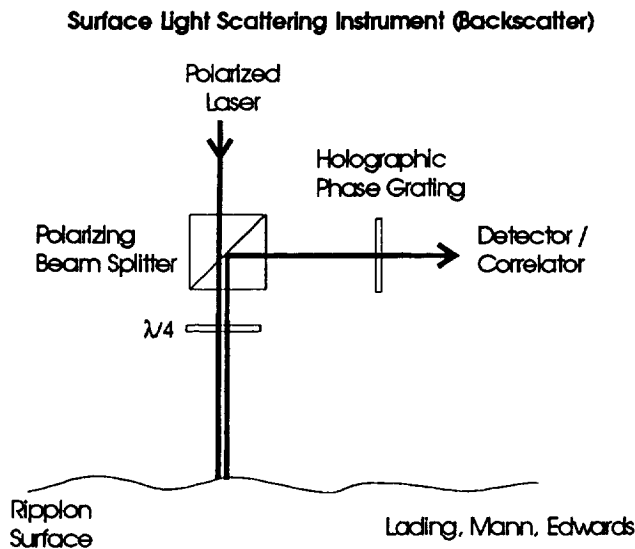


Figure 4

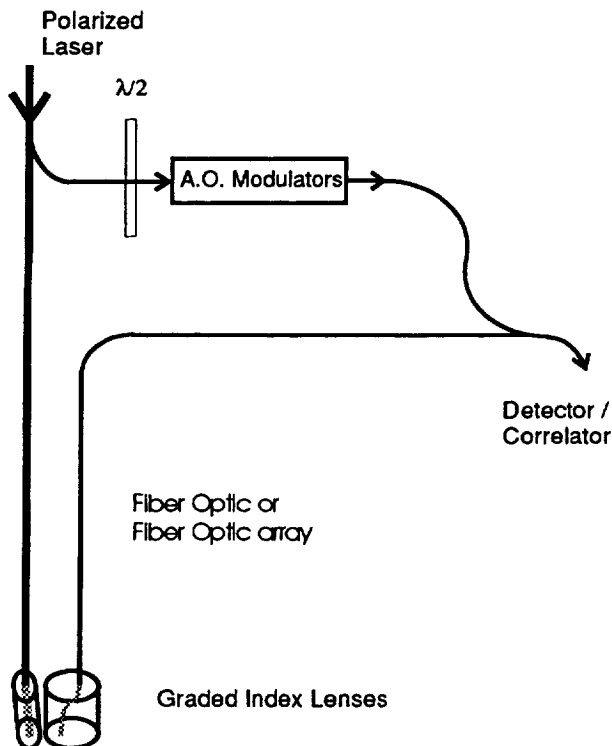


Figure 5

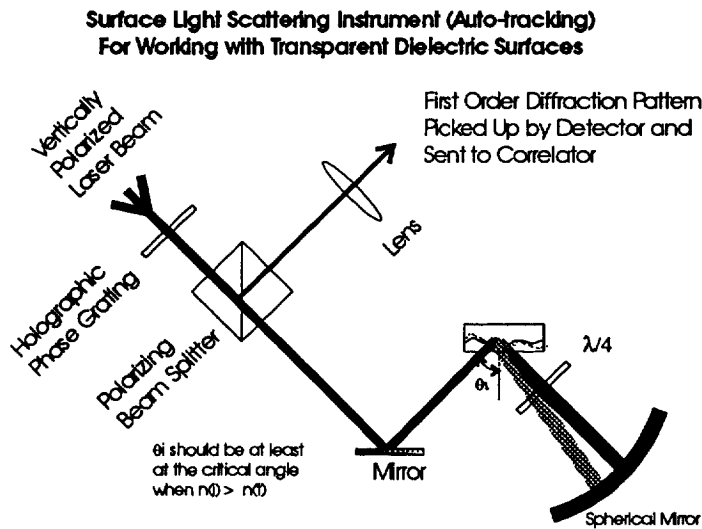
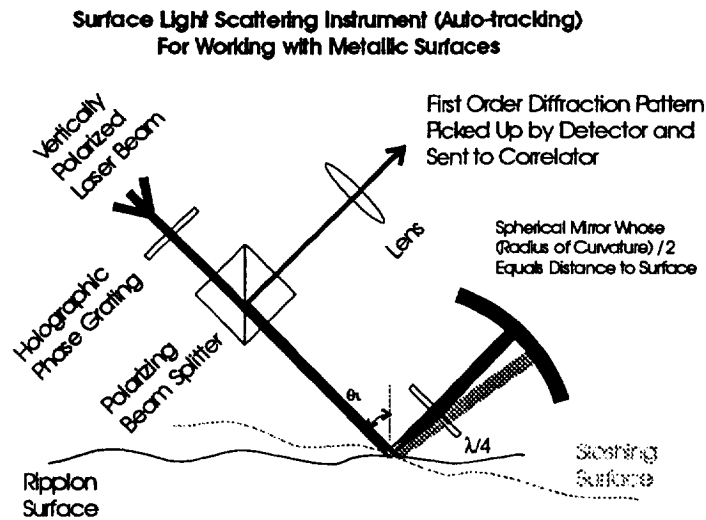
A fiber optics implementation using AO modulators in series to shift the center frequency of the reference beam

These vibrations at the surface are sufficiently removed in frequency from those of interest that our present signal inversion algorithms suffice for data analysis. Our algorithms do not assume a flat surface, but the situation is considerably more complicated if the curvature is changing in time.

To solve the problem of sloshing we began by placing a plano-convex lens with a mirrored backing after the quarter-wave plate of the backscatter surface light scattering instrument (an idea contributed by Lars Lading). This instrument builds on that sketched in Figure 4, as described by Lading, et al. (Our adaptation of this idea is shown in Figures 6 and 7.)

This surface light scattering instrument works by launching polarized light through a polarizing beam splitter cube. It then goes through a quarter-wave plate to become circularly polarized light. Upon reflecting or scattering from the surface it passes through the quarter-waver plate again and becomes linearly polarized with its polarization orthogonal to that of the incoming beam; so it is now reflected rather than transmitted through the polarizing beam-splitting cube. This configuration has advantages and other arrangements (see Lading, et al., Ref. 17, for a discussion).

The ability to track the scattered light near one particular k-value will also allow us to switch to a completely fiber optic system and hence reduce the alignment problems and the size of the optical portion of the instrument. The fiber optic system will be about the size of a pencil and a small collection of lenses and mirrors which are needed to implement this scheme. An autotracking instrument will also allow the use of miniature arrays of fiber optics placed behind GrIn lenses. Scattered light collected in this way can be fiber optically coupled to the laser output for heterodyning. This allows us to adjust the ratio of the scattered to local oscillator light intensities;



Figures 6a and 6b

typically expect to get about one percent reflection every time we encounter an interface. If we strike the surface once, encounter a mirror and strike the surface a second time the original signal is diminished by a factor of 10^{-4} . So although this idea can be modified to work with reflective surfaces (e.g. metals) by placing the mirrored lens at a slight angle to ensure that the light is always doubly reflected, we need to find an alternative scheme for surfaces of small reflectivity such as critical fluids. To work with dielectrics we can either come in from below the surface of the fluid at an angle which is near the Brewster angle, as outlined in Figure 6b, or transmit through it.

By reflecting from a mirror along a path which causes the light ray to retrace its path at the speed of light, we find that no matter how the surface sloshes the ray will return to its origin. This is illustrated by projecting a grating onto the surface and reflecting it from a phase conjugate mirror. The three visible spots from the grating which are visible near the detector are immobile when the surface sloshes or the laser table is pounded upon (within reason). However, we have found that Figures 6 and 7 hide a subtle problem. When the surface first diffracts the light, the rays for a particular k-value (angle) will end up at

we change the amount of light that is attenuated in one of the coupling fibers. This also suggests a fiber optics realization of Mazur's ²⁰ idea of placing a set of acousto-optic modulators in series to shift the center frequency of the reference beam (see Figure 5). Brimrose Corporation has supplied us with a fiber optically coupled set of two acousto-optic modulator cells which are cascaded in series. They have a common driver for stability which allows us to select a frequency difference between them of 5, 10 or 20 kHz which is added to the light passing through them. By shifting the center frequency, the Rayleigh-Brillouin triplet is resolved and nonequilibrium effects (gradients) become evident because the Brillouin bands caused by inelastic scattering are separated on either side of the Rayleigh band centered on the shift frequency. Without this technique the two Brillouin bands are superimposed as one band and the difference in their heights is lost. This configuration is difficult to align and to keep aligned when using a classical optical arrangement. Fiber optics obviates this concern (see Figure 5). But to use small diameter fiber optics for this purpose it will be necessary to counter the effects of a sloshing surface by judiciously placing the appropriate mirror as shown in Figures 6 through 8.

If we choose to work with interfaces between dielectric surfaces, we can

a well defined location with respect to the reflected laser beam. The distance these k-values appear from the reflected laser beam is dependent upon how far back the detector is placed from the surface. This is easily visualized by imaging a cone of light spreading out from the surface where the reflected laser beam acts as its axis of symmetry. When we pass the laser beam through the surface twice, a detector placed at a position to observe one set of k-values will actually see two sets of k-values at one location. It will see one set from the first time the laser beam passes through the surface and scatters light. It will see a second set of k-values from when the laser beam is returned by a phase conjugate mirror beneath the surface and passes through the surface a second time, forward scattering the light. While this corrects for sloshing, it creates a second "virtual" surface which focuses the same k-values from each surface to a different location. Worse yet, two different k-values now focus to one location. This problem also occurs

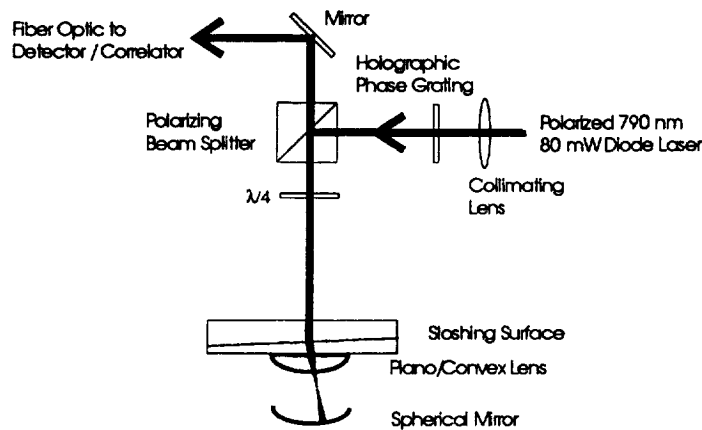


Figure 7
Early version of antislosh optics

when transmitting the laser beam through the surface as shown in Figure 6. We have been able to get reasonable correlograms using this technique when we are careful to slightly misalign the system. However, this is difficult to reproduce and we can do better.

To attain a reasonable correlogram from the surface we need to have a $k\sigma$ value greater than 20 at the surface. In other words the $1/e$ portion (e.g. $-\sigma$) of the laser beam diameter d at the surface times the k value ($2\pi/\lambda$) of the ripplon must be large enough to contribute a reasonable signal, $k\sigma > 20$. So if we focus the laser

beam onto the surface on its first pass through it and then reflect from a mirror or mirror/lens combination which is $r/2$ away, where r is the radius of curvature of the mirror, a parallel ray of light will be returned along the same path from which it came. Figure 8 illustrates this idea. Figure 9 is a picture of the setup which we used to take the correlogram shown in Figure 2 and its accompanying power spectrum in Figure 3. In Figures 2 and 3 we transmitted the laser beam once through the surface.

We are still testing both this prototype system and the ideas presented in this paper, and we believe we can refine the cat's eye geometry discussed in this paper to provide high quality correlograms even in the presence of environmental vibration. Indeed, quite reasonable correlograms have been obtained by us with the arrangements shown in Figures 4, 7 and 9. We were able to compute the surface to better than 1 mN/m for various liquids including water. The instrumental broadening was consistent with that of classical designs and can be corrected using the "deconvolution" formulas that appear in the literature. We prefer the new method mentioned in this paper. The system has the accuracy and precision expected of these spectrometers, but is far less sensitive to surface slosh. This is offered as a preview of potential flight hardware that is being developed in NASA's Advanced Technology Development program, a program which both explores and develops the hardware capabilities in a field and ensures that a microgravity need exists for it. A number of potential investigators have expressed a need for non-invasive surface tension, viscosity and non-contact temperature measurement capabilities, and this paper is being presented to invite additional interest.

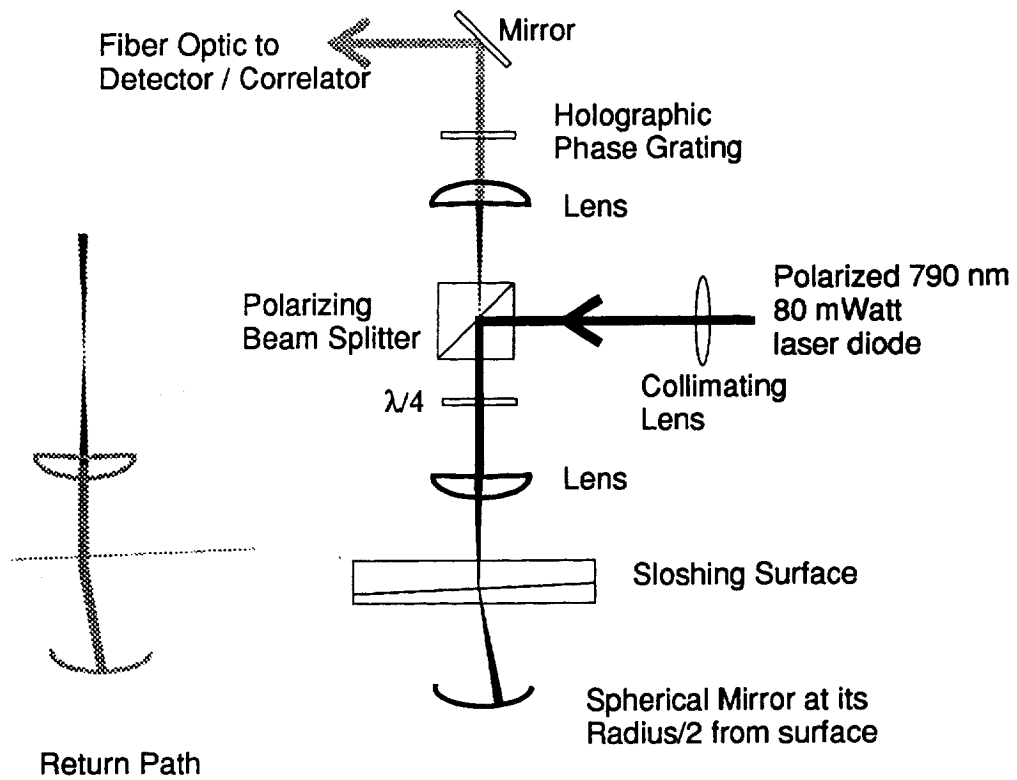


Figure 8

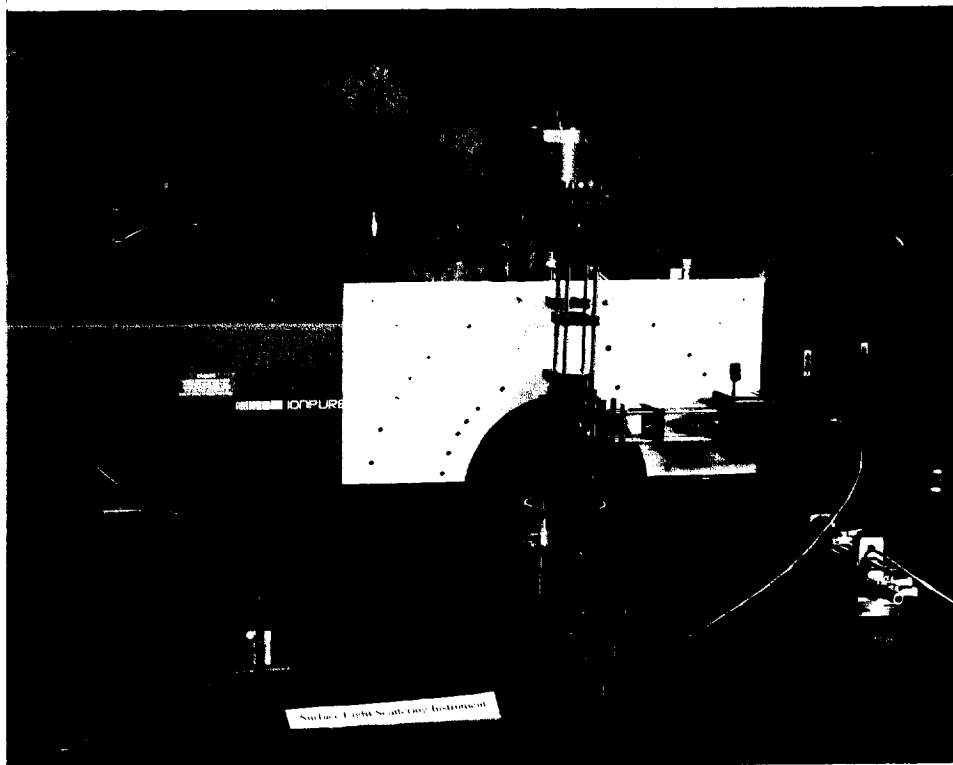


Figure 9

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