RECENT INVESTIGATIONS ON SONIC AND ULTRASONIC CAVITATION IN GÖTTINGEN

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INTRODUCTION

The investigations to be discussed here are divided into the following subjects:

1. The noise spectrum of cavitation with steady sinusoidal excitation and its relation to the oscillation of the bubbles.

2. The cavitation caused by a single underpressure pulse,

   a. excited with a barium titanate calotte

   b. excited by the sudden retardation of a moving water column.

3. The relations between sound pressure and luminescence.

4. The measurement of the surface tension and surface viscosity from the excitation of capillary waves at frequencies up to 1.5 Mc/s.

It is generally known that the onset of streaming or ultrasonic cavitation is accompanied by noise; the noise has a very broad frequency spectrum and in many cases the audible part of it is so noticeable that it is often used as a criterion for the onset of cavitation. It was, for example, during the war very easy to determine the upper limit for the creeping speed of a submarine as function of the diving depth by simply listening to the onset of cavitation or the onset of formation of air bubbles caused by a sufficient number of revolutions of the screw with a kind of a stethoscope attached to the screw shaft; already with the first experiment the increase of the critical number of revolutions of the screw with the square root of the diving depth (static pressure) was stated.

As was shown in an older paper by Th. Lange (1) also for ultrasonic cavitation the onset of noise is so sharp that it is preferred to the formation of visible bubbles as a criterion for cavitation. It is to a far extent without significance which part of the noise spectrum is used for detecting the onset of cavitation. From the work of Th. Lange only one figure (Fig. 1) will be given to confirm the above statements. Vibration cavitation was excited with a quartz crystal at 575 kc/s. The driving voltage of the quartz is plotted on the abscissa against the noise level in the frequency interval from 520 to 540 kc/s on the ordinate. Here the onset of cavitation is much easier recognized than by using e.g. the nonlinearity of the sound pressure increase of the exciting frequency (575 kc/s) as a criterion.

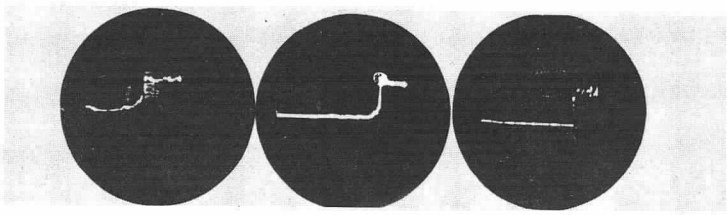

Fig. 1 - Noise voltage (520 to 540 kc/s) vs transmitter voltage (575 kc/s) for different stages of degassing

NOISE SPECTRUM AND SOUND PRESSURE OSCILLOGRAM FOR THE ONSET OF CAVITATION AND THEIR RELATION TO THE OSCILLATIONS OF THE BUBBLES

In a thorough investigation R. Esche (2) has studied the spectrum of sonic or ultrasonic cavitation for a great number of frequencies ranging from 100 c/s up to 3 Mc/s. He found out, that there is always a line and a continuous spectrum simultaneously. In the line spectrum the exciting frequency appears, of course, with high intensity and also the harmonics are observed. This seems to be obvious. But Esche could, perhaps for the first time, ascertain that also a number of subharmonic frequencies occur. In his investigation he came to the conclusion that the occurrence of a continuous spectrum might be a proper method to distinguish true cavitation (formation of bubbles containing only water vapor) from that type of cavitation where air bubbles are formed.

The occurrence of subharmonic frequencies (Fig. 2) is of special interest, as subharmonics one-half, one-third, and one-fourth the exciting frequency are observed. Also the overtones of the subharmonics are present. In a more recent investigation on the detailed structure of the spectra L. Bohn (3) found not only subharmonics within the range of the fundamental frequency but also - mostly as overtones of half-order subharmonics - up to the 30th or 40th order, their level sometimes being 10 db under the level of the integer harmonics.

The occurrence of subharmonics is not difficult to understand. In a high-intensity sound field the oscillating air bubbles change their volume to a great extent during one period. That means that the compliance of air and the mass of water taking part in the oscillation are functions of time. In this case we have a rheolinear oscillation, the differential equation of which admits also subharmonics as solutions. W. Güth (4) starts from the principle that the oscillation is nonlinear, since the compliance of the air cushion is a function of the oscillation amplitude. The compliance increases with increasing amplitude. The resonance curve of such a system is, as is well known, not symmetrical. The maximum of the curve moves towards lower frequencies with increasing amplitude. At the same time the occurrence of subharmonics is possible. It is, however, surprising that especially one-half the exciting frequency and the corresponding overtones are so clearly observable. This means that in the oscillation of the bubble a periodic amplitude or phase modulation with one-half the exciting frequency is very prominent.

Varying the conditions for ultrasonic cavitation such as air content or exciting amplitude, more astonishing results with respect to the sound pressure oscillogram or to the sound spectrum may be obtained. Such investigations were carried out by

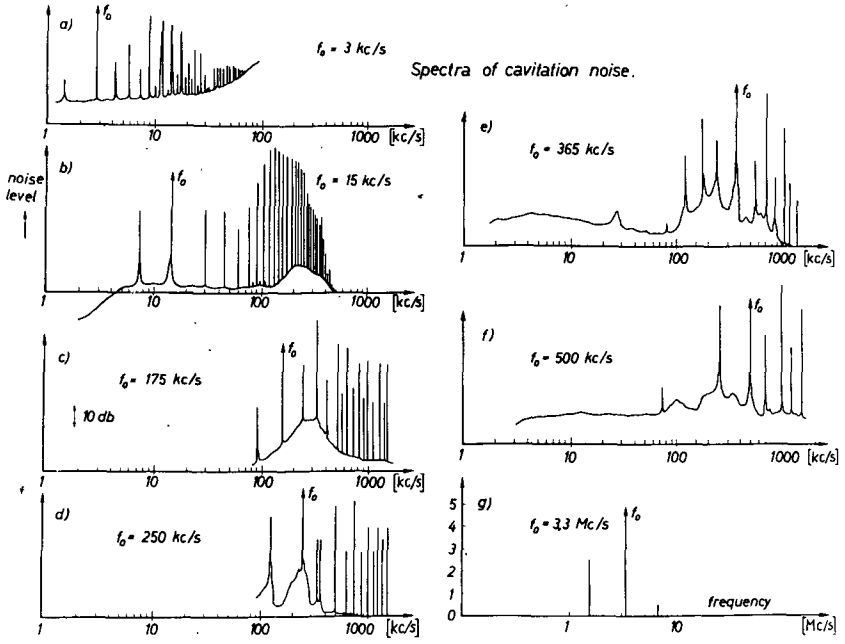

Fig. 2 - Spectra of cavitation noise

L. Bohn (3). His apparatus was very simple in principle (Fig. 3). Twenty-one magnetostrictive systems with a resonance frequency of 14.6 kc/s connected in parallel were arranged in such a way that the sound was focused on a small area. In the focus a microphone was placed, consisting of a thin nickel wire of about 0.5-mm diameter (5). Only the tip of the nickel wire was sensitive to sound since the rest of the wire immersed in the water was covered with a thin plastic tube. The part of the wire outside the water had considerable length and its end was covered with a wedge-shaped layer of wax so that only progressive waves are excited at the tip of the wire. These extensional waves generate an ac voltage in a solenoid around the nickel wire. The microphone responded to frequencies up to 1 Mc/s and it is of minor importance whether the frequency response curve rises with frequency, as corresponds to the principle of the magnetostrictive transducer, or is leveled out by an electrical network.

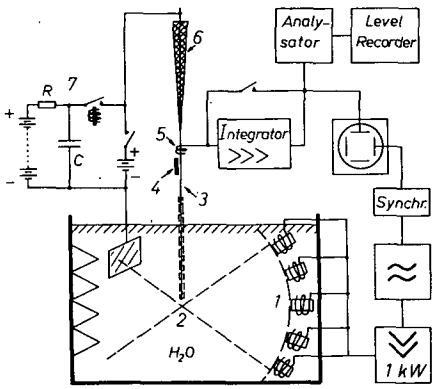

Fig. 3 - Measuring arrangement. 1. Magnetostrictive emitter system tuned at 14.6 kc/s, 2. probe microphone, 3. nickel wire, 4. permanent magnet, 5. pickup coil, 6. absorbing material, 7. electrolysis circuit.

The cavitation bubbles have to be formed at the tip of the microphone to excite directly the microphone with their vibrations. In order to really accomplish

the onset of cavitation at the very tip of the microphone, especially with degassed water, a current pulse (shortened condenser discharge) was led through the wire into the water; so by either increasing the temperature of the water over a very small area or by a weak, transient electrolysis gas bubble nuclei were injected at the tip of the wire.

Of special interest is the cavitation noise for weak excitation in saturated or moderately undersaturated water. Then the cavitation bubbles are excited (by pulses) in their natural pulsation oscillation and reverberate with this frequency. The bubbles are preferably stabilized at certain sizes. Sometimes clouds of very tiny bubbles are then intermittently pushed away from an oscillating bubble. This is probably caused by strong oscillations of the bubble surface excited by the high acceleration related to the pulsation. It therefore suggests itself to assume that in this case the natural pulsation coincides with a natural surface oscillation of the bubble. For weak excitation the surface vibrates on a subharmonic overtone based upon one-half the natural frequency of the pulsation oscillation. For strong excitation a higher order of the surface oscillation coincides directly with the natural frequency of the pulsation oscillation.

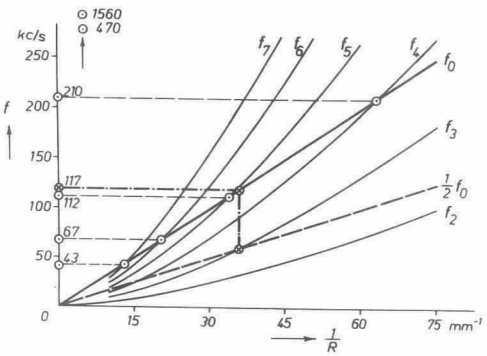

Fig. 4 - Frequencies of the natural radial oscillations and of some modes of surface oscillations for air bubbles in water as a function of the reciprocal radius

In Fig. 4 the eigenfrequency f_0 of the pulsation oscillation and, the half-order subharmonic $f_{0/2}$ as well as the different overtones (f_1 to f_6) from the second to the seventh order are plotted against the reciprocal bubble radius according to the formula given by Minnaert. It is evident from the graphs that for 43, 67, 112, and 210 kc/s the eigenfrequency of the pulsation oscillation for the corresponding bubble size coincides with the seventh, sixth, fifth, and fourth order respectively of the surface oscillation. The third order can also coincide with one-half the fundamental frequency of the pulsation oscillation, resulting in a "stable" oscillation at 117 kc/s.

A quantitative confirmation of statements is given in Figs. 5 and 6.

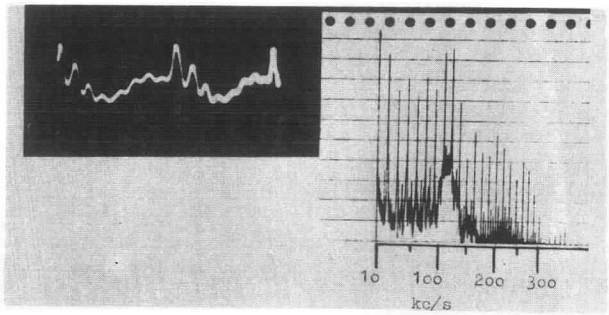

Fig. 5 - Bubble oscillation during cavitation process. Exciting frequency: 14.6 kc/s. Amplitude ca. 1 atm. Natural frequency of bubble: 110 - 120 kc/s. Saturation of water: 70 - 80%.

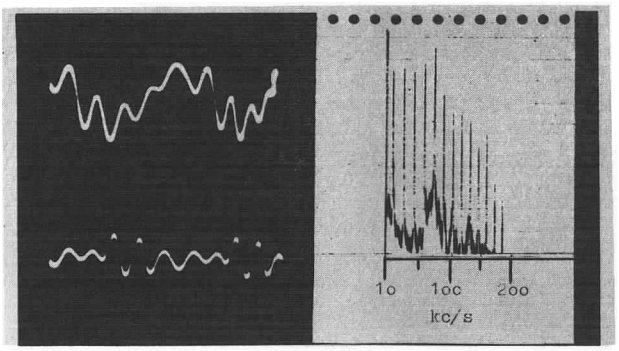

Fig. 6 - Bubble oscillations during cavitation process. Exciting frequency: 14.6 kc/s. Amplitude ca. 1 atm. Natural frequency of bubble: 65 - 70 kc/s. Saturation of water: 80 - 90%.

Here we have weak cavitation; the sound pressure is 0.7 atm and the saturation with air about 70 to 80 percent (Fig. 5). The corresponding values for Fig. 6 are 1 atm and 80 to 90 percent saturation. In the oscillogram as well as in the spectrum the preference of a certain frequency range - using a term from physiological and musical acoustics one might say "formant frequency" - is obvious. In Fig. 5 this is 110 to 120 kc/s, in Fig. 6 it is 65 to 70 kc/s. Also lower "formant" frequencies might occur, as for example the frequencies 15.8 and 16.6 kc/s respectively and 22 kc/s in Fig. 7 for a sound pressure of 1 atm and air-saturated water. On the other hand also higher formant ranges are possible (see Fig. 8); the saturation corresponding to Fig. 8 is 60 to 70 percent and the sound pressure is relatively high (2 atm).

For very strong sound excitation things become rather complex. There occur reverberating oscillations of the bubbles at certain frequencies. Also stabilization in the above described sense is observed. As is expected, the energy of the spectrum in all cases increases with frequency. There are sharp maxima in the oscillograms with a pulse width of only some microseconds. The width of these pulses can be calculated from the minima in the respective frequency ranges of the spectrum. In Fig. 9 a spectrum of this type is given. There is a marked minimum at about 200 kc/s.

Another example illustrating how the noise spectrum extends to higher frequencies when the exciting sound pressure is raised from 1 to 4 atm is given in Fig. 10.

On the other hand in saturated or, better, oversaturated water larger bubbles are formed which are in resonance with the exciting frequency of 14.6 kc/s. Then there are no sharp sound pulses and the oscillogram resembles more or less a sine curve.

Very sharp pulses are naturally observed in extremely undersaturated water at higher exciting sound pressure amplitudes. The spectrum is then exclusively given by the

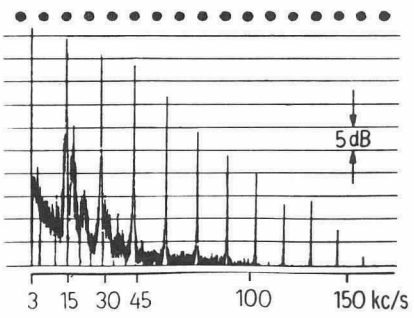

Fig. 7 - Noise spectrum of oscillating bubbles in air-saturated water. Sound pressure about 1 atm.

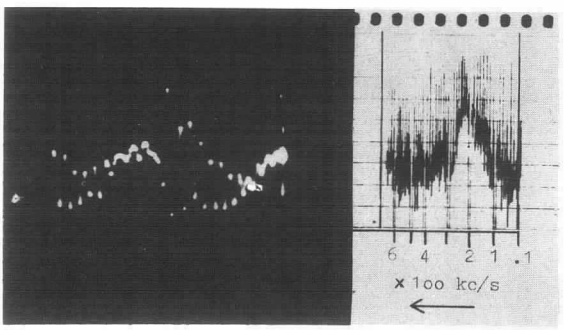

Fig. 8 - Bubble oscillation during cavitation process. Exciting frequency: 14.6 kc/s. Amplitude ca. 2 atm. Saturation of water: 60 - 70%.

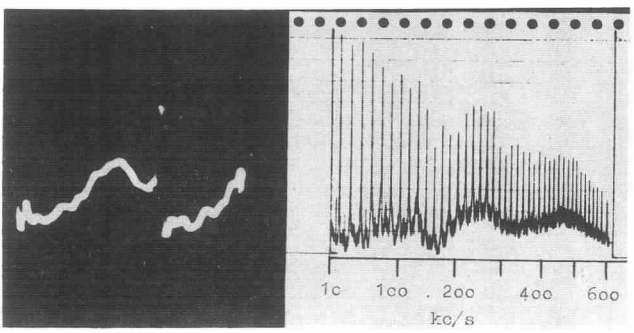

Fig. 9 - Pressure shock caused by bubble collapse. Exciting frequency: 14.6 kc/s. Amplitude about 4 atm. Saturation of water: 70%.

form of the pulses (Fig. 11). The envelope of the spectrum can easily be explained with the assumption of a cosine pulse of 6 to 7 μsec duration.

The occurrence of a continuous spectrum has often been the subject of detailed discussion. For some time it was believed that the continuous spectrum was a criterion for real cavitation, that is, cavitation of vapor bubbles. A more simple reason for the existence of a continuous spectrum is that the amplitude or the phase of the gas bubble oscillation is subjected to statistical fluctuations, which consequently render a continuous spectrum. The vapor bubble cavitation, if it at all has to be distinguished from the formation of gas bubbles, is supposed to be the stronger the higher the frequencies in the spectrum are.

Simultaneously with the above mentioned investigations E. Mundry and W. Güth (6) filmed the gas bubble oscillation itself. In these experiments the oscillating cavitation bubble is generated at the tip of a magnetostrictive nickel rod (2.5 kc/s). The bubble is lighted up with an intermittent electric spark and then recorded on a stationary film with the help of a revolving mirror; the maximal number of exposures is about 105,000 frames/sec. As soon as the bubble has been registered the sound is

cut off at once and the size of the bubble at rest is measured. The film is then evaluated by planimetering the bubble. Thus an "average" bubble radius is found. This radius is plotted as a function of time for a number of bubbles of different sizes in Fig. 12. The average radius of the bubbles in this figure ranges from 1.42 mm down to 0.3 mm. These sizes are indicated with the dash-dot lines in the detailed pictures in Fig. 12. The dashed lines give the corresponding decay curves according to the theory of Rayleigh. Two conclusions may be drawn from the results. Firstly, the time interval during which the bubble is larger than in the equilibrium position (under-pressure phase) is longer than the time interval during which the bubble is smaller. This means that if gas diffusion occurs the direction of the diffusion from the liquid into the bubble is preferred to the opposite direction, so that the bubble is growing. Even more interesting than this phenomenon is the reverberation of the bubble excited by the implosion. Especially for the smaller bubbles this is clearly visible. The reverberation frequency is practically given by the natural frequency of the pulsation oscillation calculated according to the linear theory of the bubble oscillation. In Fig. 13 a number of examples of such measurements are collected. Plotted against the measured length of the period of the natural oscillation of the bubble is the measured radius of the bubble in equilibrium position. The line corresponds to the calculations of Minnaert. The results are obviously in good agreement with the above mentioned investigations on the cavitation spectrum.

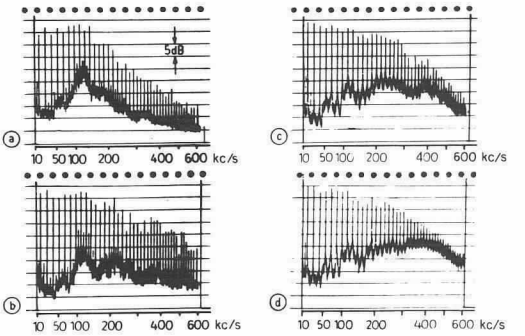

Fig. 10 - Noise spectra for increasing exciting sound pressure

	Exciting sound pressure (atm)	Air saturation (%)
a.	1.0	80
b.	2.0	80
c.	3 to 4	60 to 70
d.	3 to 4	60 to 70

It is also possible to make the shock waves, caused by the implosion of the bubble in the water, visible with a schlieren method (Fig. 14). Thereby a number of shock waves are observed in temporal succession, all of them having their center on the surface of the transmitter but at different points. This is no wonder. It only indicates

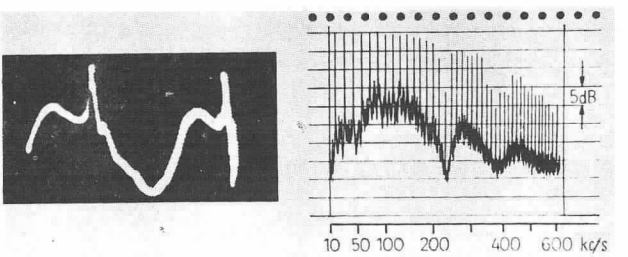

Fig. 11 - Sound pressure and noise spectrum for cavitation in undersaturated water. Air saturation: 60%. Sound pressure 3 to 4 atm.

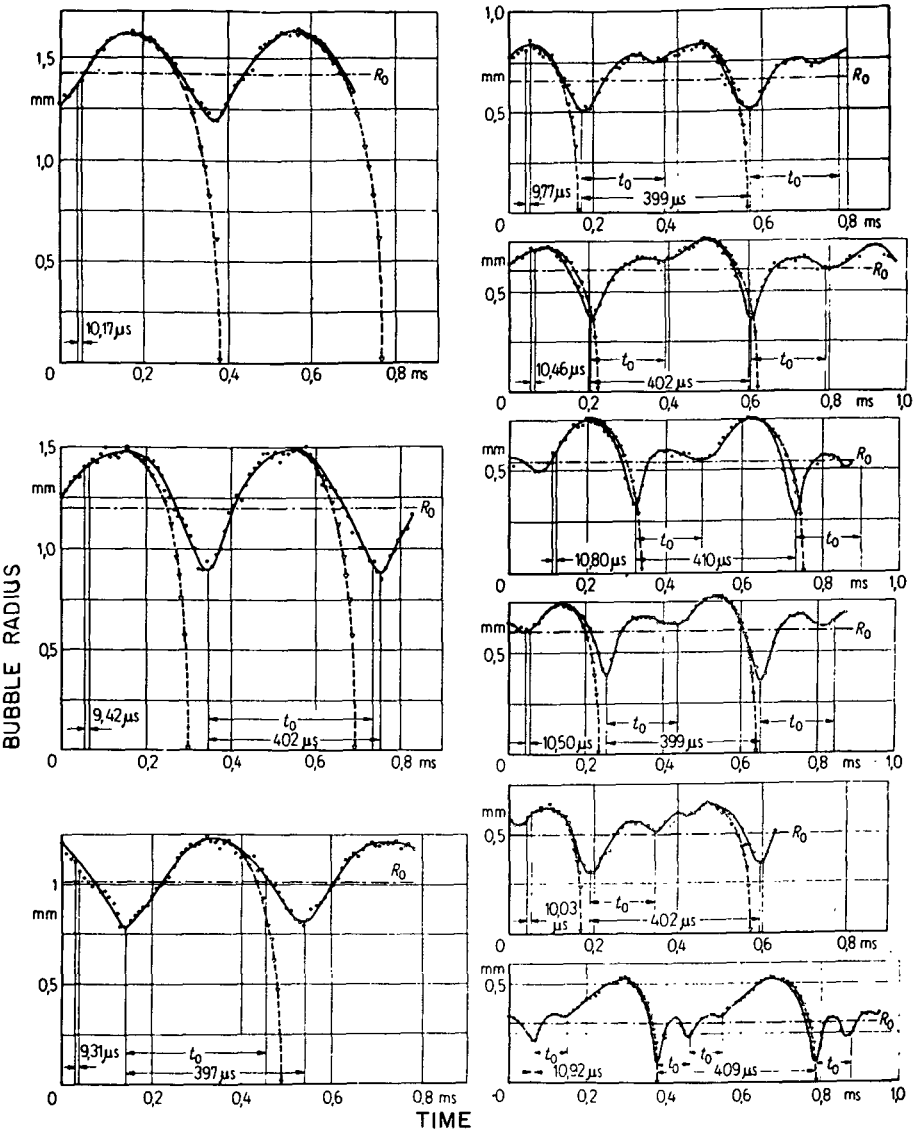

Fig. 12 - Radius of oscillating gas bubbles as a function of time for bubbles with different diameters

that there are different centers of implosion. That again may be taken as an optical argument or proof for the occurrence of a continuous spectrum in the cavitation noise; the shock waves illustrated in Fig. 14 for example are generated within a time interval of at least 80 μsec.

A more detailed study of the shock waves shows a slow pressure increase at the front of the shock waves whereas on the backside there is a steep decrease. According

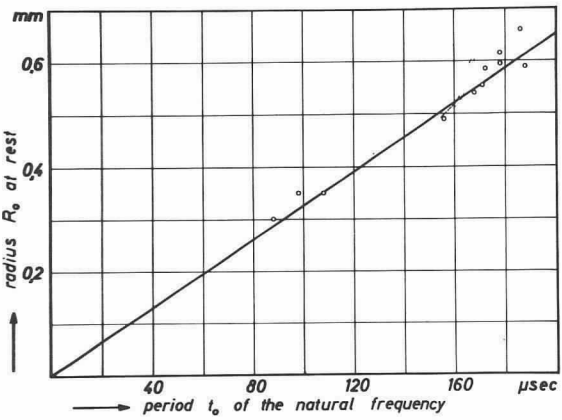

Fig. 13 - Periods of the (linear) natural oscillations of gas bubbles as a function of their radius at rest.

to W. Güth (7) this can be explained by looking at the acceleration of the water as a function of time; we have an increase of the acceleration to the center and then a sudden retardation.

At this point some short remarks will be put in on cleaning of surfaces with ultrasonics, which in the past years became an often-used practice. J. Olaf has worked on this problem in our institute. By making measurements in a larger frequency range he has found out that for the speedy cleaning of a dirty surface (in his case glass plates with polishing rouge (Fe_2O_3) contamination) cavitation is absolutely necessary. If - without changing the sound pressure - the cavitation is prevented by a high static overpressure the cleaning effect is practically gone. The differences in the cleaning effect at different frequencies are of secondary nature. They are for example caused by the directivity of the transmitter a.o. It has also to be kept in mind that at higher frequencies cavitation is generated at higher expense.

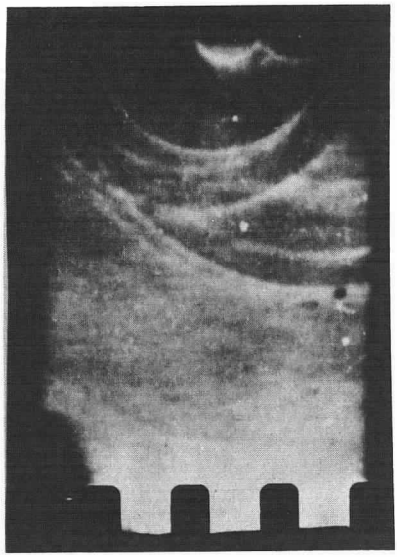

Fig. 14 - Schlieren optical pictures of shock waves

CAVITATION BY A SINGLE UNDERPRESSURE PULSE

For the elucidation of the formation and growth of cavitation bubbles it is of advantage not to use periodic underpressure intervals but to apply only one single underpressure pulse. This problem was tackled by G. Kurtze (9). His experimental setup is given schematically in Fig. 15. A barium titanate calotte of 10-cm diameter and 6-mm thickness is terminated at its backside by a long

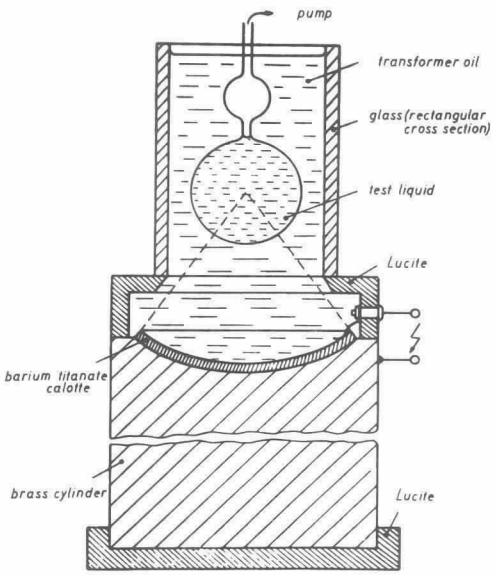

Fig. 15 - Experimental setup for the excitation of cavitation by a single underpressure pulse

brass cylinder. The front surface is adjoining to a liquid. The sound is focused into this liquid. The liquid under test is contained in a thin-walled glass sphere in the focal point. The static pressure inside this glass sphere can be reduced to the vapor pressure of the test liquid. The sound is generated by slowly charging the barium titanate calotte to 12 kv and discharging it with a time constant of 2.5 μsec. The time constant is slightly higher than half the period of the natural vibration of the barium titanate calotte. Thus during the reverberation of the barium titanate the underpressure amplitude is dominating. The brass cylinder on the backside of the barium titanate calotte is long enough to delay the sound reflected at its other end until the process in the test liquid is finished. The oscillogram of the sound pressure in the focal point as a function of time was measured with the help of a tiny barium titanate cube of 1-mm^3 volume. There is practically only an underpressure pulse (Fig. 16).

With this apparatus it is possible to measure the sound pressure necessary to initiate cavitation and to observe the growth of the cavitation bubble. Of course, for that purpose it is indispensable to make a large visible bubble. This could only be achieved by subjecting the test liquid to a static underpressure. On the other hand the growth of the bubble could be calculated under certain assumptions as for example a disappearing hydrostatic pressure and an underpressure pulse of 1-μsec duration and 20-atm amplitude. The results are plotted as a dotted line in Fig. 17. The two experimental curves show that the assumptions made for the calculation can be realized to a certain extent.

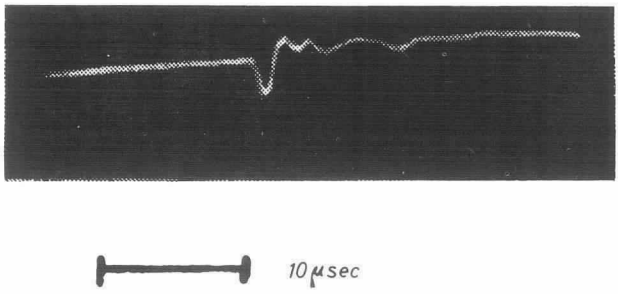

Fig. 16 - Pressure in the focal point of the barium titanate calotte as a function of time. Sound pressure: 10 atm.

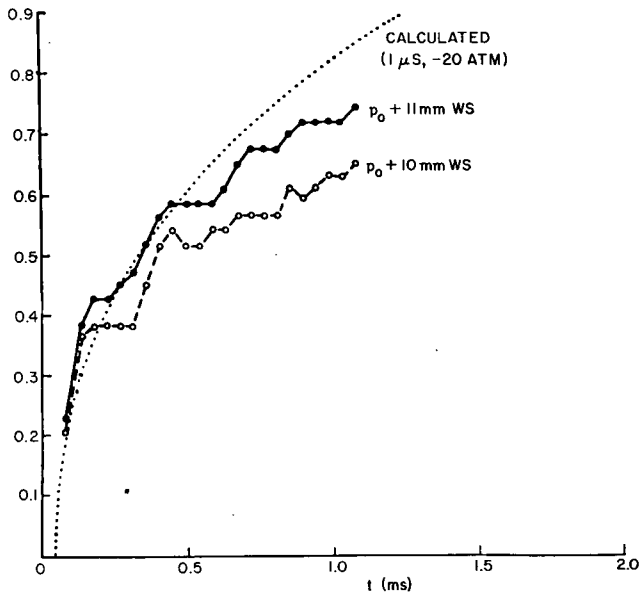

Fig. 17 - Bubble radius as a function of time with a single pulse of 2-μsec pulse width and 20-atm pressure amplitude ($P_o \sim 0$)

In the apparatus described here the cavitation threshold reaches 20 atm for all liquids under test such as water, transformer oil, carbon tetrachloride, and others after exposing them for a longer or shorter time to air and is independent of the dissolved air content.

Also another method to obtain a single cavitation process was studied in Göttingen (J. Schmid (10)). A water column contained in a glass tube is given a high translatory velocity which is then suddenly retarded. Thus high retardation forces are set free and the water is torn apart. To let this happen at a given point a bubble nucleus is injected by a short-time electrolysis. The process is filmed with a high-speed camera. Two examples of such registration are given in Figs. 18a and 18b. The maximal diameter of the bubbles is 1.5 and 1.2 cm respectively. The camera operates at the rate of 5000 and 63,000 frames/sec. The growth of the bubble takes about 2.5 ms, the decay about 0.9 ms. Naturally also in this case an oscillation is observed during the decay of the bubble. The growth of the bubble during the second period is diffuse and irregular in relation to space. This is generally known and was already observed by W. Güth (7) for the collapse of hot water vapor bubbles. It is worth mentioning that the implosion at the first as well as at the second collapse of the bubble is so violent that the radiated shock waves can directly be photographed without any schlieren optics (Fig. 18c). Figure 18c shows that there is a number of implosion centers which are located close together but act in temporal succession.

RELATION BETWEEN PRESSURE AND LUMINESCENCE FOR VIBRATION CAVITATION

Closely related to sonic or ultrasonic cavitation is sonoluminescence, this being the phenomenon that small oscillating gas bubbles contained in the liquid emit light.

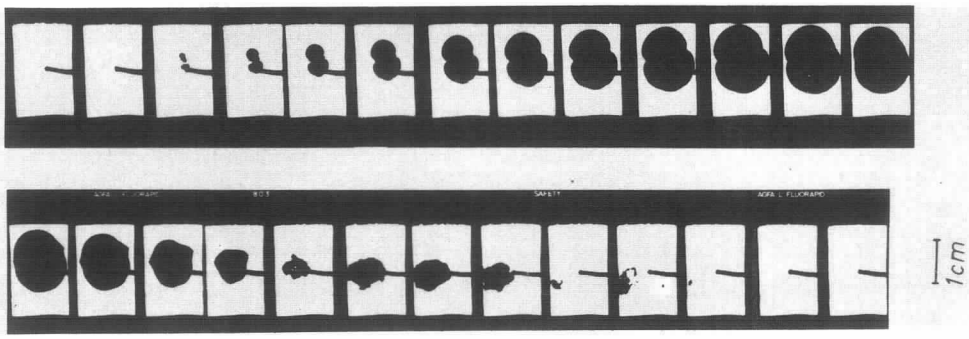

Fig. 18a - Cavitation excited by the sudden retardation of a fast moving water column. Repetition frequency, 4800 frames/sec. Maximum diameter of the bubble about 1.5 cm. Time of growth about 2.5 ms. Time of decay about 0.9 ms (without rebound).

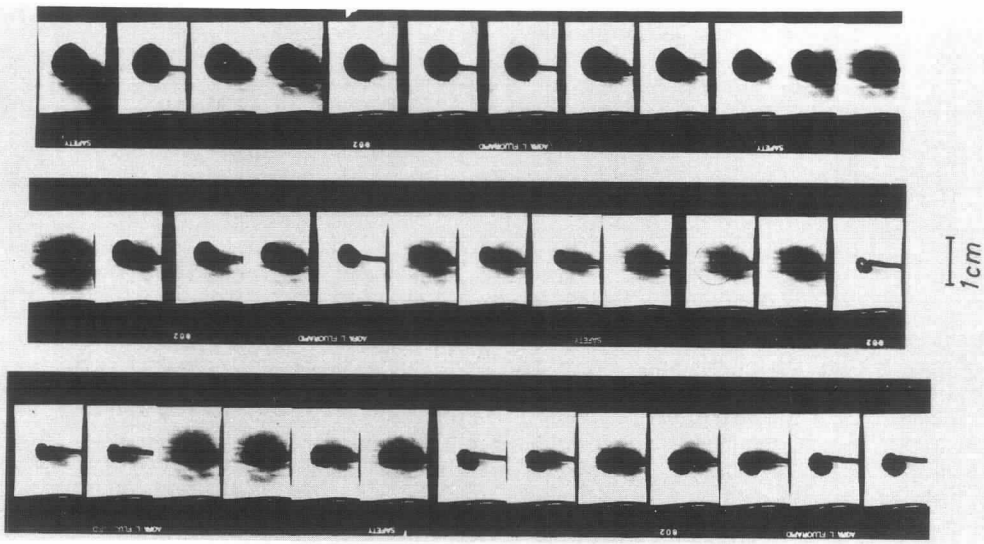

Fig. 18b - Decay and rebound of a bubble. Repetition frequency, 63,000 frames/sec. Maximum diameter of the bubble about 1.2 cm.

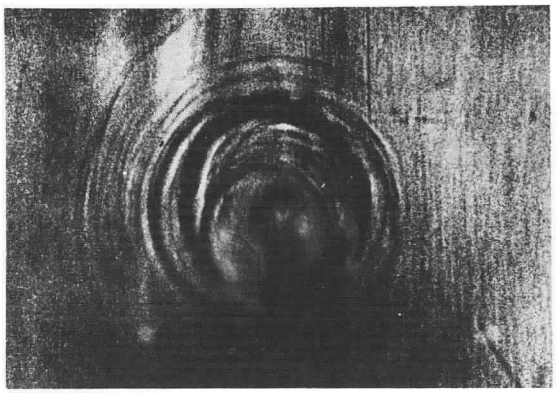

Fig. 18c - Shock waves radiated by collapsing bubbles without Schlieren optics

The sonoluminescence has often been the subject of investigations, and there are a number of theories trying to explain it. One of them is the "hot-spot theory." According to this theory the gas in a collapsing bubble is exposed to an extremely high compression and is consequently heated up, thus emitting light. Another theory starts with the separation of electric charges at the surface of a bubble when this is torn apart; the gas discharge inside the bubble is the decisive effect. Also the chemoluminescence and the triboluminescence are discussed in this connection. Now very recently W. U. Wagner has in our institute completed a series of experiments.

His experimental setup is in principle arranged as in Fig. 19. A quartz crystal with a resonance frequency of 260 kc/s generates standing ultrasonic waves in front of a $\lambda/4$ metal reflector. With a very small barium titanate microphone (M) with a volume of 1 mm^3, carefully isolated acoustically from the leads and its mounting, the alternating sound pressure is measured. The microphone can be moved in the field of standing waves. The tiny gas bubbles oscillating probably below their resonance frequency are collected in the pressure maxima of the standing wave. This effect is, of course, generally known and it can again clearly be seen in Fig. 20 showing the emission of light. One might, in this case, speak of a radiating grating made up of a great number of separate light sources, namely, the light-emitting bubbles. Because of the alternating phase only every second row of radiating bubbles in the standing wave is focused (L) on a photomultiplier (PM) by means of a suitable grating (G). The microphone, sensitive to sound pressure, is situated in one of those pressure maxima, which are used "optically." Pressure and underpressure indication of the microphone have been carefully determined with several calibration methods. On the screen of an oscilloscope the alternating sound pressure as well as the emission of light are registered simultaneously.

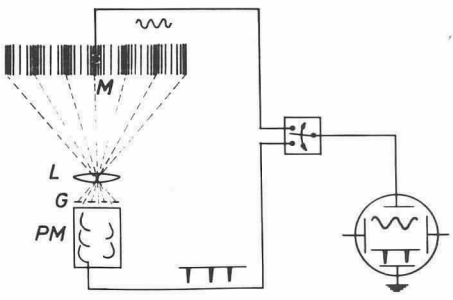

Fig. 19 - Schematic of the experimental setup for the investigation of sonoluminescence: M, microphone; G, grating; PM, photomultiplier; L, lens

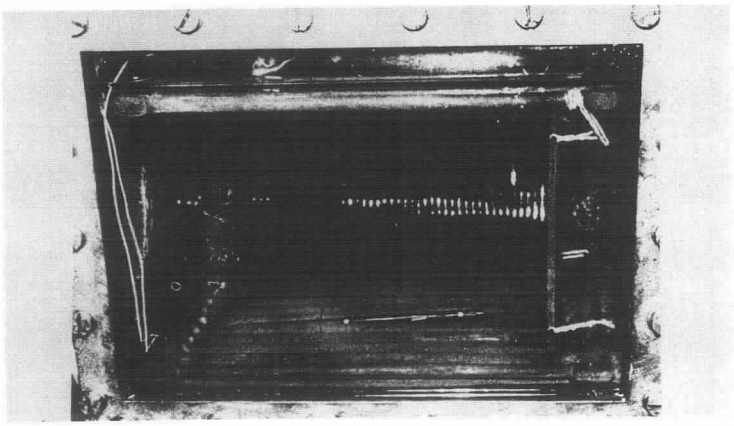

Fig. 20 - Sonoluminescence in a standing wave

Wagner found out that the emission of light is certainly not constant with time, as is the case when, for example, Luminol is added to the water, but that it is emitted in pulses. The most important result, however, is that the emission of light coincides rather accurately with respect to time with the underpressure phase of the alternating sound pressure (Fig. 21). These measurements were carried out in water saturated with air or with krypton. In the case of krypton the luminescence is considerably stronger. In Fig. 21 the upper oscillogram gives the course of the sound pressure and the lower one the corresponding output voltage of the photomultiplier. In the sound-pressure diagram, overpressure is always plotted upwards.

In Fig. 21 the temporal duration of the emission of light is relatively long, which is probably due to statistical fluctuations of the cavitation as well as the emission of light. The sound pressure curves are often distorted because of the cavitation, and without cavitation there is no luminescence. But also in the case of a more sinusoidal course of the sound pressure light pulses are observed. With proper exciting conditions the light pulses can be short and high.

It must be mentioned that the measuring curvette allows for variation of static pressure and temperature of the liquid under test. Most of the figures given here were made with an overpressure of 0.5 atm. At very high overpressures cavitation is prevented and also luminescence is not to be observed anymore.

This is just a short summary of the results of the investigation. They show that the hot-spot theory probably does not hold true in this case, since the luminescence

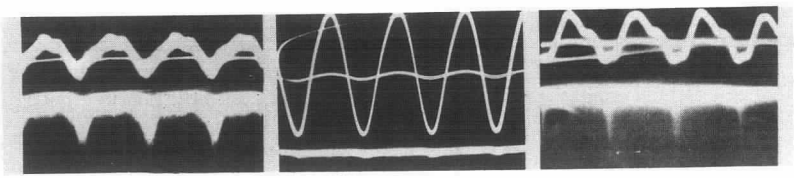

Fig. 21 - Sound pressure and luminescence. Upper oscillogram: sound pressure (overpressure upwards). Lower oscillogram: output voltage of the photomultiplier.

does not coincide with the overpressure phase. But there is the preassumption that the bubbles are smaller than the resonance size, i.e., that they are vibrating in phase with the sound pressure, which is not known with certainty.*

SURFACE TENSION AND SURFACE VISCOSITY

Surface tension and an eventual surface viscosity may play an essential part for the nucleation in the liquid, and I will go into the particulars of a new work by W. Eisenmenger (12) dealing with the dynamic surface tension and a potential surface viscosity in liquids. During the recent years we have made several approaches to this problem at Göttingen. In the beginning the problem was studied with gas bubbles in liquids, especially very small bubbles, where the surface tension has a decisive influence on the resonance frequency. The logarithmic decrement of the natural vibration of the bubble was measured. Unfortunately these measurements, which were made with all sizes of bubbles down to very small ones with a resonance frequency of about 300 kc/s, show that the high thermal damping in the gas contained in the bubble makes any statement about the surface viscosity impossible. One thing was learned from these experiments: one has to make a really free surface and to use capillary waves at the boundary surface between liquid and air. This has been tried formerly, but without much success. In our case the trick is to excite rheolinear oscillations of the surface with the help of a parametric excitation: a small volume of water is set vibrating vertically and the exciting amplitude is increased until at a critical amplitude capillary waves occur at the surface. The frequency of these capillary waves is one half of the exciting frequency.

The plane capillary wave is described by

$$\eta = a\eta^*(t) \cos kx$$

where a is the amplitude. The onset of the capillary waves is given by the solution of a Mathieu differential equation in the first ranges of instability:

$$\frac{\partial^2\eta^*}{\partial t^2} + 2\beta_0 \frac{\partial\eta^*}{\partial t} + \eta^*(\omega_K^2 + kh\,\omega_A^2 \cos \omega_A t) = 0 .$$

Here $\omega_k = \sqrt{Tk^3/\rho}$ is the capillary wave frequency, in which T is the surface tension and ρ is the density, $h \cos \omega_A t$ is the exciting oscillation of the water volume, and β_0 is the viscosity damping. For small capillary wave amplitudes, $\beta_0 = 2\mu k^2/\rho$ where μ is the viscosity and k is the wave number. The solution of the above equation renders for the onset amplitude of the rheolinear oscillation the expression

$$h_0 = 2\frac{\mu}{\rho}\sqrt[3]{\frac{\rho}{T\omega_k}} .$$

This onset amplitude h_0 of the exciting frequency is given by the surface tension T and the viscosity μ.

*Since the preparation of this manuscript some other observations have been made. For very low frequencies (2 kc/s) a relatively strong luminescence has been found too, in water, in glycol, and in glycerine. In this case the phase of luminescence did not coincide with the underpressure phase. Therefore, it was necessary to coordinate the phase of the bubble motion and the phase of luminescence. The result is that the luminescence pulses coincide with the collapsing bubbles, more precisely with the last collapsing bubbles out of a larger number of bubbles (E. Meyer and H. Kuttruff (11)).

When the amplitude h of the exciting vibration is increased over the critical point of onset h_0, the capillary wave amplitude a follows this law:

$$\frac{h - h_0}{h_0} = C \frac{a^2}{\lambda^2}$$

where C is a constant. In other words, the damping of the capillary waves has an amplitude- and frequency-dependent additional expression of the form

$$\beta = \beta_0 \left(1 + C \frac{a^2}{\lambda^2}\right)$$

where C is, according to the theory of Klemm, given by

$$C = \frac{\pi^2}{8} \frac{e'}{\mu} k$$

where e' is introduced by Klemm as a surface viscosity. Unfortunately in the expression for $(h - h_0)/h_0$, that is, in the constant C, a further additional expression has to be added, given by the second power of the normal viscosity damping. This term is, however, independent of frequency.

The experimental setup is very simple in principle. For frequencies up to about 200 kc/s hollow barium titanate cylinders with Mason horns attached to them were

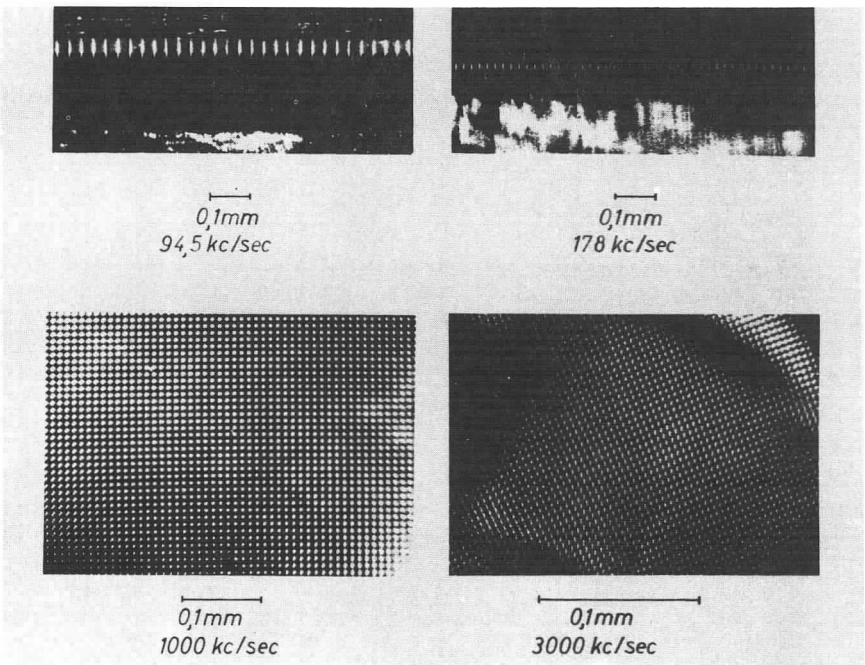

Fig. 22 - Capillary waves generated by parametric excitation

used. There are small grooves at the tops of the Mason horns filled with water. Thus plane capillary waves are forcibly excited. The absolute amplitude (h) of the exciting Mason horn is measured electrostatically with a second Mason horn. The amplitude (a) of the capillary waves is determined optically from the reflection of a light beam at the steepest parts of the sinusoidal capillary wave.

In the frequency range from 400 kc/s to 3 Mc/s concave barium titanate shells filled with water are used. A very thin, platinized copper foil is placed in the focal plane of the shell so that a water droplet on its surface is at the focal point. In the first case plane capillary waves are excited in the grooves; in the second case the capillary waves generated have the form of crossed gratings. Examples for both cases are found in Fig. 22 for exciting frequencies of 94.5 kc/s, 178 kc/s, 1 Mc/s, and 3 Mc/s. Photographs of this kind allow for an easy measurement of the capillary wavelength and for the determination of the surface tension therefrom. In Fig. 23 this was done for a larger frequency range (up to $\nu_A/2 = \nu_k = 1.5$ Mc/s) in pure water for the onset amplitude of the oscillation, i.e., for a capillary wave amplitude $a/\lambda = 0.01$. In another case the same measurements were made with a higher amplitude ($a/\lambda = 0.12$) and there it became obvious, as a first result, that the surface tension is amplitude dependent, but this only at higher frequencies. This effect is obviously caused by the fact that the filling-in of the extended surface with water molecules from inside the liquid needs a certain time.

It is also not without interest that the measured onset amplitude h_0 of the capillary waves, substantially given by the viscosity of the medium, satisfies the equation

$$h_0 = \frac{2\mu}{\rho} \sqrt[3]{\frac{\rho}{T\omega_k}}$$

very well. In Fig. 24 the measured values are compared with the theoretical curves for $T = 75$ dyne-cm^{-1}, $\theta = 20°C$, and $\mu = 10^{-2}$ dyne-cm^{-2}-sec for the given frequency range.

The relation between exciting amplitude h and capillary wave amplitude a for values up to $a = 0.12\lambda$ is treated very thoroughly for 41 kc/s. The curve is displayed in Fig. 25. Given as abscissa is the exciting amplitude h or $(h - h_0)/h_0$. The curve

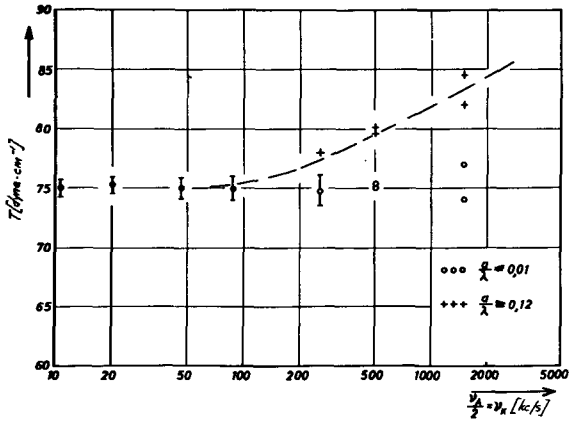

Fig. 23 - Surface tension in pure water as a function of frequency and amplitude

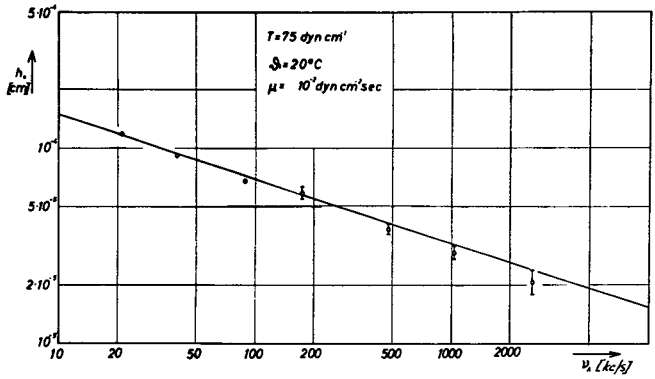

Fig. 24 - Onset amplitude of the exciting oscillation as a function of the exciting frequency for pure water

is in very good agreement with the theoretical relation. For the constant C, introduced above, one finds C = 18.7. The respective values for other liquids with different surface tensions are of the same order of magnitude.

It is only necessary to determine the constant C for water for a number of frequencies in order to find out about a potential surface viscosity. It is thereby stated that for water the frequency dependence is very small and that a potential surface viscosity is certainly smaller than 10^{-5} dyne-cm^{-1}-sec.

Realizing that a higher surface viscosity, as it was supposed by other authors, was not found in water, the question arises whether the admixture of surface active substances would render a higher frequency dependence of C, thereby also indicating a higher surface viscosity. Therefore different concentrations (10^{-4} and 10^{-1} weight percent) of an emulgator (Renex 688) and two wetting agents (Hostapal and Tween 80) were examined. In this case the whole range of amplitudes of the capillary waves as

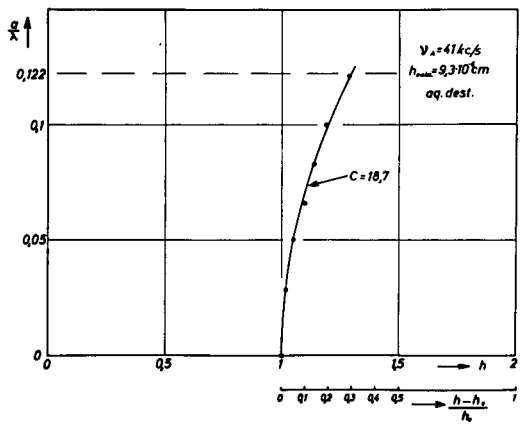

Fig. 25 - Capillary wave amplitude as a function of the exciting amplitude

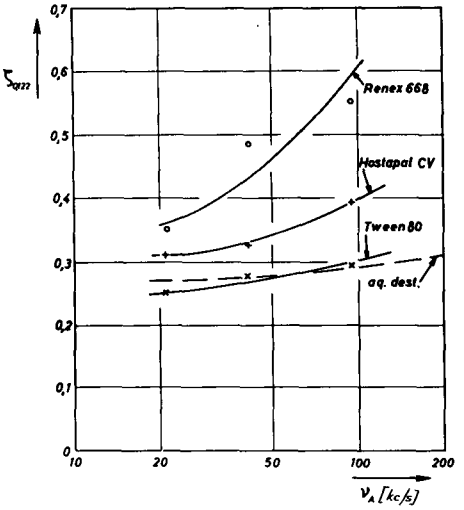

Fig. 26 - Relative difference of the exciting amplitude $\overline{h} - h_0/h_0 = \zeta_{0.122}$ as a function of the frequency for different surface-active substances. Capillary wave amplitude a = 0.122.

a function of the excitation was not measured. Only the onset amplitude h_0 and the exciting amplitude h for a given ratio $a/\lambda = 0.122$ was examined. The corresponding value $(h - h_0)/h_0 = \zeta_{0.122}$ is plotted in Fig. 26 versus frequency. The considerable frequency dependence of Renex 688 is evident. Here we find a surface viscosity of about 1×10^{-4} dyne-cm^{-1}-sec, being much higher than for "pure" water.

The above paper contains a short survey of some investigations specializing on sonic and ultrasonic cavitation carried out in Göttingen. I do hope, that some of the results might be of general interest.

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DISCUSSION

T. Brooke Benjamin (University of Cambridge)

I wish to comment upon the formation of capillary waves on the surface of a mass of liquid which is contained in a vessel undergoing vertical vibrations. This was the subject of a paper by Ursell and myself* and the problem has continued to interest me since it appears to exemplify very nicely a general class of phenomena, many instances of which arise in practical acoustics. For example, Murray Strasberg and I have shown that the waves in question have counterparts in the behavior of small bubbles undergoing radial pulsations in response to a sound field: such bubbles may develop large nonspherical oscillations when the amplitude and frequency of the radial motion take certain values, and the explanation which we suggested is the same, exactly, in principle as that for the waves on a plane free surface.

I think it may be of interest to remark that the mechanism of these waves was the subject of a historic controversy extending well over a century. They were first studied by Michael Faraday about 130 years ago. He observed that a pattern of standing waves sometimes appeared on a layer of liquid covering a vertically vibrating plate, and he noticed that the frequency of the waves was only one-half that of the plate. About 50 years after, the famous German physicist Matthiessen re-investigated the problems and found in his experiments that the waves were synchronous with the

*Proc. Roy. Soc. 225(A):505 (1954).

vibrations of the vessel. The discrepancy between this and Faraday's result led Lord Rayleigh to make a further series of experiments, which supported Faraday's observation. You may recall, incidentally, that Rayleigh gave to the waves the appealing term "crispatious": this term will perhaps be remembered by anyone familiar with Rayleigh's textbook "Theory of Sound."

The theory set out in the paper by Ursell and myself appeared, happily, to reconcile the observations of Faraday and Rayleigh and those of Matthiessen; for it was shown that both half-frequency and synchronous waves can be generated in suitable circumstances. The essential explanation of these waves, and also of the many similar phenomena which I wish to keep in view here, can be summarized without need for much mathematics.

I think that to emphasize the general character of the explanation we should consider any mechanical system in which free vibrations are possible in certain normal modes. Now, suppose that one of the physical parameters of the system is made to vary in simple-harmonic fashion with time through the action of some external agency. (We may consider, to fix our ideas, that the parameter varied is one that would enter the equation for the frequency of the normal modes: e.g., it is a stiffness or inertial coefficient.) Now, with this modification, small displacements in the system will be determined by an equation of motion having at least one periodic coefficient. This equation will be linear, of course, provided the displacements are small enough and, in the absence of dissipation, it will nearly always be reducible to Mathieu's equation, whose standard form may be written

$$\frac{d^2 a_n}{dT^2} + (p - 2q \cos 2T) a_n = 0 .$$

In general, a_n is the amplitude of, say, the nth mode of disturbance, and $T = \omega t$ where ω is the frequency of the applied oscillation. Also, the parameter q is proportional to the amplitude of this oscillation, and $p = (2 \omega_n/\omega)^2$ where ω_n is the frequency of free vibrations in the nth mode.

Now, the general solution of Mathieu's equation is known to be unstable for certain values of p and q, i.e., it becomes unbounded as $T \to \infty$, and this provides the explanation of why the mechanical system in question may spontaneously develop large oscillations. The stability chart is sketched in Fig. D1: the diagram is symmetrical about the p-axis, and the third and fourth quadrants do not concern us here. Note that when $p \approx 1$, i.e., when $\omega = 2 \omega_n$, instability is possible for small values of q.

When $p \approx 4$, instability may again occur for small values of q, and the developed frequency is then ω, i.e., the applied and developed oscillations are synchronous.

In the capillary wave problem, the disturbed motion of the free surface is the same as if the

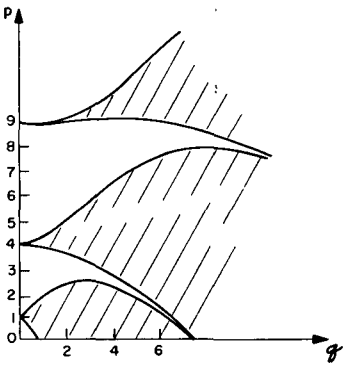

Fig. D1 - Plot of p–q values showing stable and unstable behavior of Mathieu's equation

vessel were at rest, and the gravitational acceleration were given a periodic component in addition to the constant g. We now see that the plane, free surface becomes unstable when the amplitude and frequency of this component have the appropriate values. The waves are not, of course, forced vibrations in the usual sense: they develop quite suddenly as p or q is varied so as to bring the point (p,q) into an unstable region of the stability chart, and they grow in amplitude until eventually they are restrained by non-linear effects, not so far considered, or until the free surface disintegrates.

The effects of friction tend, of course, to inhibit the growth of waves. For instability to occur in practice, the point (p,q) must lie somewhere inside an unstable region of the chart, so that the rate of growth of the frictionless solution exceeds the rate of frictional damping. Thus, as Professor Meyer has pointed out in his paper, an estimate of the damping can be obtained from a careful experimental observation of the onset of instability. In my own experiments on these waves, the frequency of the vessel was about 30 cycles per second, which is considerably less than in the experiments at Göttingen. In mine, most of the damping was evidently due to the boundary layer at the sides of the vessel and was quite small. I was unable to get a good agreement between experimental and theoretical estimates of the rate of damping, and I concluded that the discrepancy was mainly due to extra dissipation in the vicinity of the meniscus at the edge of the free surface. However, I am very pleased to learn that at Göttingen this method has given satisfactory measurements of the damping at high frequencies.

E. Meyer

The point here is that this is not only the damping of this surface, but also the damping given by the normal damping, i.e., by the viscosity of the liquid itself. Then there is an additional term, the damping by the surface, and this is very difficult to separate. There is also a quadratic term for the normal damping and you have to distinguish this from the surface damping. The distinguishing term is the frequency.

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