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THE UNIVERSITY OF MISSISSIPPI
PHYSICAL ACOUSTICS RESEARCH GROUP
DEPARTMENT OF PHYSICS AND ASTRONOMY

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OFFICE OF NAVAL RESEARCH

Contract N0014-79-C-0404

Technical Report No. 802

AIR BUBBLE GROWTH BY RECTIFIED DIFFUSION

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER: 802
2. GOVT ACCESSION NO.
3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle): AIR BUBBLE GROWTH BY RECTIFIED DIFFUSION
5. TYPE OF REPORT & PERIOD COVERED: TECHNICAL Prepts
6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s): LAWRENCE A. CRUM
8. CONTRACT OR GRANT NUMBER(s): N0014-79-C-0404

9. PERFORMING ORGANIZATION NAME AND ADDRESS:
Department of Physics
University of Mississippi

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS

11. CONTROLLING OFFICE NAME AND ADDRESS:
Office of Naval Research
800 N. Quincy St. Arlington, VA. 22217

12. REPORT DATE: 15 Apr 80
13. NUMBER OF PAGES: 41

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)

15. SECURITY CLASS. (of this report): U

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for Public release: Distribution Unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

Approved for Public Release: Distribution Unlimited

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

RECTIFIED DIFFUSION
CAVITATION
AIR BUBBLE

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Measurements are reported of the growth of air bubbles in water by rectified diffusion at 22.1 kHz. Values of the threshold acoustic pressure amplitude were obtained as a function of bubble radius, liquid surface tension and gas concentration. Measurements of the rate of growth of bubbles by rectified diffusion were also obtained as a function of acoustic pressure amplitude for a range of different values of the liquid-vapor surface tension. It was determined that although →

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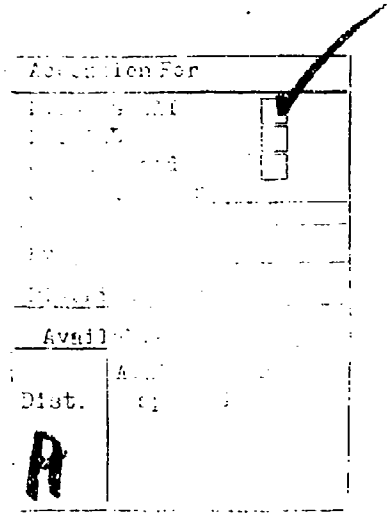
both the threshold and the growth rate were in agreement with theory for normal values of the surface tension of water, the addition of a surfactant caused the observed thresholds and growth rates to deviate from the predicted values. Surface wave activity that could increase the diffusion rate by acoustic streaming was not detected at low radii and is not thought to be the principal mechanism for the increased diffusion. Some possible explanations are given for the effect.



INTRODUCTION

This paper concerns the phenomenon of rectified diffusion of gas into air bubbles that are caused to pulsate in a liquid by the action of an acoustic field. Free bubbles that would normally dissolve in water may be made to grow due to the unequal mass transfer across the air-water interface during bubble oscillation. The phenomenon seems to have originated with Harvey,[1] and was first discussed in some detail by Blake,[2] who formulated an inadequate theory for predicting the threshold. Attempts by Pode[3] and Rosenberg[4] were also found inadequate to explain some of the early measurements by Strasberg.[5] Hsieh and Plesset,[6] by adding a convection term in the diffusion equation, considerably improved upon Blake's approximate results. Their theory was shown by Strasberg[7] to be in good agreement with his measured values. Eller and Flynn[8] extended the theory to include nonlinear or large amplitude effects and Safar[9] showed that the Hsieh-Plesset and Eller-Flynn treatments were essentially equivalent when inertial effects were not neglected in the Hsieh-Plesset approach. Eller[10,11] made several measurements of both the threshold and the growth rate and found that his theory was adequate in predicting thresholds but was unable to account for some very large growth rates that he observed. He suggested that the rapid rates of growth may be accounted for by acoustic microstreaming. Gould[12] has observed bubbles growing by rectified diffusion and has found agreement with Eller's theory in the absence of streaming. By observing the bubble directly in a microscope he was able to detect the onset of surface oscillations of the bubble that in turn introduced significant acoustic microstreaming. This streaming greatly enhanced bubble growth

and was likely the explanation for the large growth rates seen by Eller. Attempts by Davidson[13] and by Kapustina and Statnikov[14] to account for enhanced growth rates by microstreaming appear to be inadequate.[12]

In this paper measurements will be presented of the growth of air bubbles by rectified diffusion, for various values of the surface tension of the host liquid, the acoustic pressure amplitude, the dissolved air content and the bubble radius. Comparisons with theoretical predictions indicate good agreement with theory for both the threshold and the growth rate for normal values of the surface tension of water, provided surface wave activity of the bubble is avoided. However, the addition of a surfactant to the water results in a slight reduction in the experimental threshold, compared with the calculated values, and in a substantial increase in the rate of growth of the bubble. Although the threshold for surface wave activity is lowered for reduced values of the liquid surface tension, an explanation for the increased diffusion in terms of bubble surface oscillations appears to be inconsistent with the observations.

I. EXPERIMENTAL APPARATUS AND PROCEDURE

The various data concerning the growth of bubbles by rectified diffusion were obtained by acoustically levitating the air bubbles near the antinode of an acoustic stationary wave. This technique[5,10,11,12,15,16], although quite common for many applications, has been considerably refined for these measurements. A diagram of the experimental apparatus is shown in Figure 1. The stationary wave system was constructed by cementing a hollow glass cylinder between two matched hollow cylindrical transducers, fitted with a flexible pressure release diaphragm on one end and open at the other. The composite system was approximately 7.5 cm in diameter by 10 cm in height, the width of the glass in the middle about 2.5 cm. This system was driven at its (r,0,z) = (2,0,2) mode at a frequency of 22.1 kHz.

In order to obtain accurate values of the acoustic pressure amplitude required to acoustically levitate a bubble at a specific position, it is necessary to know the spatial variation of the stationary acoustic wave along the axis of the cylinder. This variation was measured with the use of a small calibrated probe hydrophone that was mounted to a micromanipulator. It has been determined previously that stationary wave systems such as the one used here often have distorted acoustic pressure profiles and an accurate measurement of this profile is necessary for accurate acoustic pressure amplitude measurements.[17] In order to reduce effects of the hydrophone itself, the actual acoustic pressure amplitude measurements were made by a small pill transducer mounted externally as shown in Figure 1. The acoustic pressure amplitude at the bubble's

position was determined from the voltage output of the pill transducer and the known spatial variation of the sound field. The average radius of the bubble was determined by first measuring its terminal rise velocity through the host liquid and then by iterating the following equation:[17]

$$R_o^2 = \frac{9\nu u}{2g}\left[1 + 0.20\, R_e^{0.63} + 2.6 \times 10^{-4}\, R_e^{1.4}\right] \quad (1)$$

In Eq. 1, ν is the kinematic viscosity of the liquid, u is the terminal velocity, g is the gravitational acceleration and $R_e = 2R_o u/\nu$ is the Reynolds number. Two refinements were made to increase the accuracy of the radius measurements. First, several fiducial lines were introduced into the cathetometer so that rise-times could be taken over a variety of distances and thus would not be excessively short or long. Second, the rise-time measurements for an individual bubble was introduced directly into a computer program that corrected for the dissolution of the bubble when the sound field was off and the terminal velocity measurement was being made. For small bubbles, say 20 μm or less, an interval of a few seconds used to measure the bubble's rise velocity could result in errors of 10% or more in the bubble radius due to bubble dissolution.

The surface tension of the host liquid was varied by addition to distilled water of small amounts of a commercially available surfactant, ethoxylated octaphenol, marketed under the brand name of Photoflo, and commonly used in photography. The surface tension was measured with a Du Nouy ring tensiometer[18].

It was discovered that small variations in the temperature of the host liquid could result in substantial changes in the rectified diffusion

threshold and growth rate of the bubble. As will be shown in Section III, a small change in temperature could result in a significant effect due to the strong variation in the dissolved gas concentration with temperature. Accordingly, it was necessary to surround the test cell with a constant temperature bath, see Figure 1, to insure temperature stability, and equilibrium gas concentration. The temperature of the host liquid was constantly monitored to insure no change in temperature during the measurements, or just as importantly, no change from the temperature at which the liquid was brought to equilibrium gas concentration.

In general it is estimated that the absolute accuracy of the acoustic pressure amplitude measurements was good to within 5%, and consistent to within 2%. The radius measurements were slightly better except for small radii, < 25um. Surface tension measurements tended to be subjected to contamination during an experiment, but were consistent to within 3% and of similar absolute accuracy. Dissolved gas concentration was not measured directly, but considerable care was made to bring the liquid to equilibrium by vigorous agitation of the host liquid, for several minutes, and then maintaining the temperature to within 0.2°C.

Bubbles were introduced into the test cell by increasing the gain on the amplifier until the threshold for gaseous cavitation was achieved. By suitable manipulation of the acoustic pressure amplitude, bubbles of the desired size could be acquired within a few minutes.

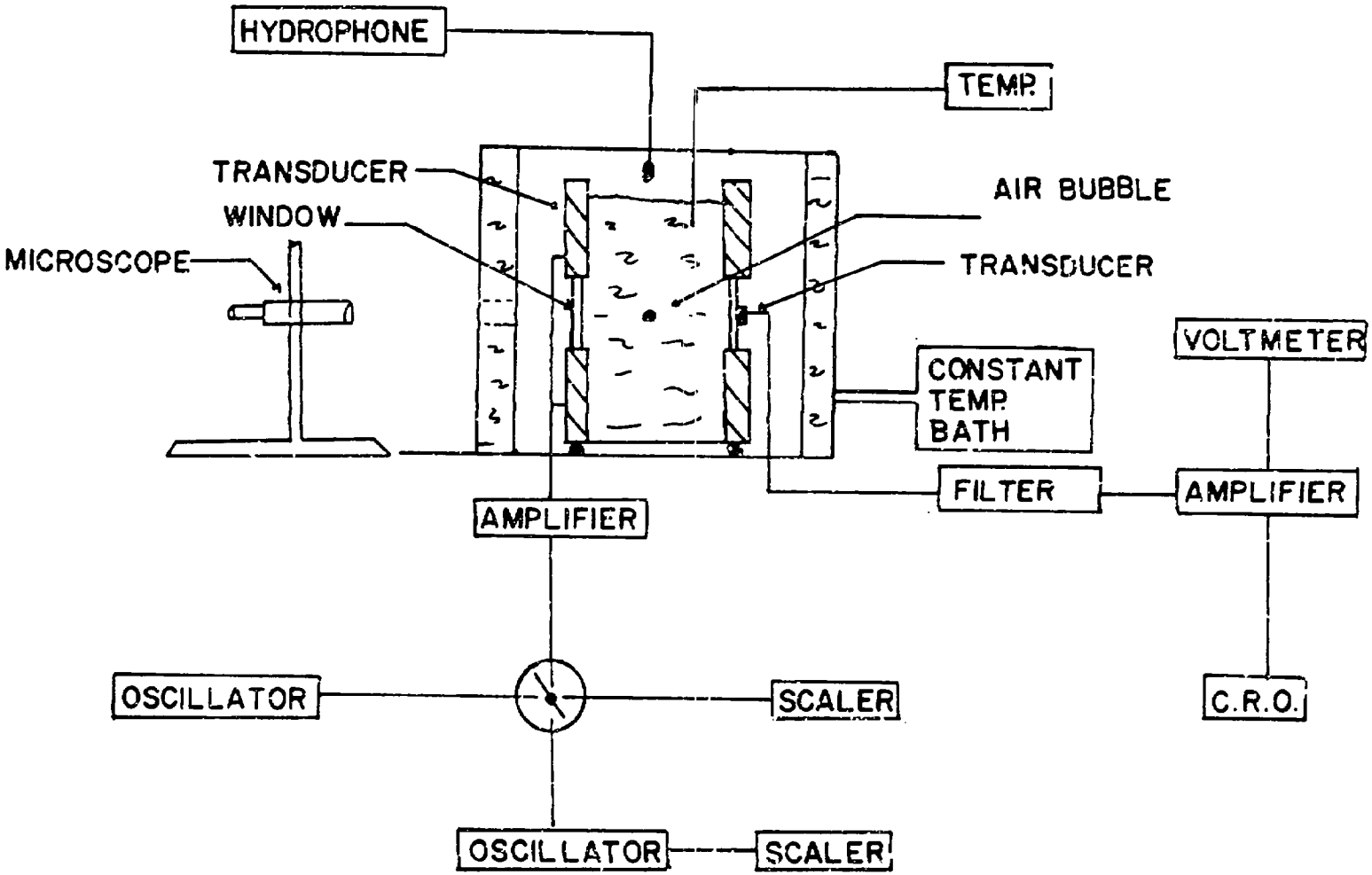


FIG 1. Block diagram and sketch of experimental arrangement

II. EQUATIONS FOR BUBBLE GROWTH

In this section we obtain the equations that describe the change in size of an air bubble that is present in a liquid and experiencing a periodic fluctuation in pressure described by $P_A \cos \omega t$, where P_A is the acoustic pressure amplitude and ω is the angular frequency of the oscillation. The bubble will either grow or dissolve depending upon a variety of conditions; we examine the conditions necessary for size stabilization (threshold for rectified diffusion) and for change in size of the bubble as a function of time (growth rate).

Previously, expressions have been obtained for the threshold and the growth rate by Hsieh and Plesset[6], Strasberg[7], Safar[9] and Eller[10] for isothermal pulsations of the bubble, and by Eller[11] for adiabatic pulsations. Our experimental measurements at 22.1 kHz are for bubbles that span the gap between purely isothermal pulsations ($R_o < 30\mu m$) and those that approximate adiabatic pulsations ($R_o > 70\mu m$). Accordingly, we have attempted to obtain expressions for the growth rate and the threshold acoustic pressure amplitude that apply to the entire range of bubble sizes.

Eller[11] has simplified the more general theory and has obtained an equation for the rate of change in the number of moles of gas in a bubble to be

$$\frac{dn}{dt} = 4\pi D R_o \langle R/R_o \rangle C_o H \qquad (2)$$

where a time-dependent term has been neglected, D is the diffusion constant, R, R_o are the instantaneous and equilibrium values of the bubble radius, C_o is the "equilibrium" or "saturation" concentration of the gas in moles/unit volume,

the pointed brackets imply time-average, and H is defined by

$$H = \frac{C_i}{C_o} - \frac{\langle (R/R_o)^4 \, (P_g/P_o) \rangle}{\langle (R/R_o)^4 \rangle} \quad , \tag{3}$$

where C_i is the concentration of dissolved gas in the liquid far from the bubble, P_o is the absolute pressure and P_g, the instantaneous pressure of the gas within the bubble is given by

$$\frac{P_g}{P_o} = (1 + \frac{2\sigma}{R_o P_o}) \, (\frac{R_o}{R})^{3\eta} \quad , \tag{4}$$

where σ is the surface tension of the host fluid and η is the polytropic exponent. It is further assumed that the bubble radius obeys the equation of motion

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 + \rho^{-1}\left[\frac{2\sigma}{R} - (P_o + \frac{2\sigma}{R_o})\,(\frac{R_o}{R})^{3\eta} + P_o + P_A \cos \omega t\right] = 0 \tag{5}$$

where ρ is the density of the liquid and the dot above the symbol implies differentiation with respect to time. Following Eller[11] we seek an approximate solution of Eq. 5 by an expansion in (P_A/P_o), namely,

$$R/R_o = 1 - \alpha(\frac{P_A}{P_o}) \cos \omega t + \alpha^2 K \, (P_A/P_o)^2 + \ldots \tag{6}$$

where we have determined

$$\alpha = [3\eta \, (1 + 4\sigma/3P_o R_o - \beta^2)]^{-1} \quad , \tag{7}$$

$$K = \frac{(3\eta + 1 - \beta^2)/4 + 4\sigma/3P_o R_o}{1 + \frac{4\sigma}{3P_o R_o}} \quad , \tag{8}$$

and $$\beta^2 = \frac{\rho\omega^2 R_o^2}{3\eta P_o} \qquad (9)$$

The time averages $\langle R/R_o \rangle$, $\langle (R/R_o)^4 \rangle$ and $\langle (R/R_o)^4 (P_g/P_o) \rangle$ will be required later and are given by

$$\langle R/R_o \rangle = 1 + K\alpha^2 (P_A/P_o)^2 \quad , \qquad (10)$$

$$\langle (R/R_o)^4 \rangle = 1 + (3 + 4K)\alpha^2 (P_A/P_o)^2 \quad , \qquad (11a)$$

and $$\langle (R/R_c)^4 (P_g/P_o) \rangle = [1+(4-3\eta)K\alpha^2 (P_A/P_o)^2](1+2\sigma/R_o P_o) \quad . \qquad (11b)$$

We can relate the equilibrium value of the radius to the number of moles of gas by the equation

$$P_o + \frac{2\sigma}{R_o} = 3nR_g T/4\pi R_o^3 \quad , \qquad (12)$$

where R_g is the universal gas constant and T is the absolute temperature. Combining Eq. 12 with Eq. 2 and Eq. 3, we obtain an expression for the rate of change of the bubble radius with time

$$\frac{dR_o}{dt} = \frac{Dd}{R_o} \{\langle R/R_o \rangle (1+4\sigma/3P_o R_o)^{-1} [\frac{C_i}{C_o} - \frac{\langle (R/R_o)^4 (P_g/P_o) \rangle}{\langle (R/R_o)^4 \rangle}]\} \qquad (13)$$

where $d = R_g T C_o/P_o$.

In Eq. 13, $\langle R/R_o \rangle$, $\langle (R/R_o)^4 \rangle$ and $\langle (R/R_o)^4 (P_g/P_o) \rangle$ are given by Eqs. 10 and 11 respectively.

The threshold for rectified diffusio[illegible]tained by setting $dR_o/dt = 0$ in Eq. 13, and is given by

$$P_A = \frac{P_o}{\alpha}\left[\frac{1 + 2\sigma/R_o P_o - C_i/C_o}{(3+4K)C_i/C_o - (1+2\sigma/R_o P_o)(4-3\eta)K}\right]^{1/2} \tag{14}$$

The polytropic exponent η is given by a set of complicated expressions by Eller[19] based on the work of Devin[20], and by Prosperetti[21], using a somewhat different technique. For completeness we list the expression obtained by Eller[19]

$$\eta = \gamma(1+d_t)^{-1}\left[1 + \frac{3(\gamma-1)}{X}\left(\frac{\sinh X - \sin X}{\cosh X - \cos X}\right)\right]^{-1}, \tag{15}$$

where

$$d_t = 3(\gamma-1)\left[\frac{X(\sinh X + \sin X) - 2(\cosh X - \cos X)}{X^2(\cosh X - \cos X) + 3(\gamma-1)X(\sinh X - \sin X)}\right], \tag{16}$$

and $X = R_o(2\omega/D_l)$. (17)

In these equations γ is the ratio of specific heats and $D_l = K_l/\rho_l C_{p_l}$, where K_l is the thermal conductivity of the gas in the bubble, ρ_l is the density of gas and C_{p_l}, is the specific heat at constant pressure for the gas.

If the polytropic exponent η is set equal to 1.0 in Eqs. 7-9, then the expressions for the growth rate, Eq. 13, and for the threshold, Eq. 14, reduce to those derived for the isothermal case by Eller.[10] If η is set equal to γ, then Eqs. 13 and 14 reduce approximately to those obtained by Eller[11] for the adiabatic case. Eller[11] neglected the effect of surface tension and set $3\gamma = 4.0$, approximations not made in this study. Comparisons of the equations obtained for the threshold by various authors, together with some experimental measurements, will be given in section III.

The constants that were used in the above equations and applicable to this experiment are f = 22.1 kHz, $\gamma = 1.4$, $D_l = 0.20$ cm^2/sec, $P_o = 1.01 \times 10^6$ dyn/cm^2, and $\rho = 1.0$ g/cm^3. The constants D and d were corrected for

temperature variations by the following equations: $D = (6.15 \times 10^{-7} T - 15.6 \times 10^{-5})$ cm^2/sec and $d = 1.02 \times 10^{-1} - 2.80 \times 10^{-4} T$, T being the absolute temperature in degrees Kelvin. The data used in the equations for the variation of D and d with temperature were taken from the International Critical Tables[22].

III. EXPERIMENTAL RESULTS

Measurements of the threshold and the rate of growth of bubbles by rectified diffusion were made for a variety of experimental conditions. In this section we examine the results of the measurements and the comparisons with theory.

A. Experiments at normal surface tension

1. Threshold measurements

Figure 2 shows measurements of the threshold for rectified diffusion of air bubbles in water at a surface tension of 68 dyn/cm (distilled water having an initial surface tension of 72 dyn/cm allowed to stand in our laboratory at a temperature of 25°C for a few hours would reach an equilibrium value of 68 dyn/cm no matter what precautions we would take against contamination). The values of the threshold acoustic pressure were obtained by levitating an air bubble and monitoring its radius as a function of time. An acoustic pressure amplitude that resulted in a radius change of less than 1% over a time interval of a few minutes was considered to be a threshold value. Since this value is a position of metastable equilibrium, considerable patience is required in "tracking down" a point. Also shown in Fig. 2 are a set of curves representing the various theories that can be applied to the measurements. Eller[10] and Safar[9] have developed equations that assume isothermal pulsations of the bubble; Eller[11] has also extended his work to include adiabatic pulsations.

It is seen that Eq. 14 more closely approximates the data than the two extreme treatments, and

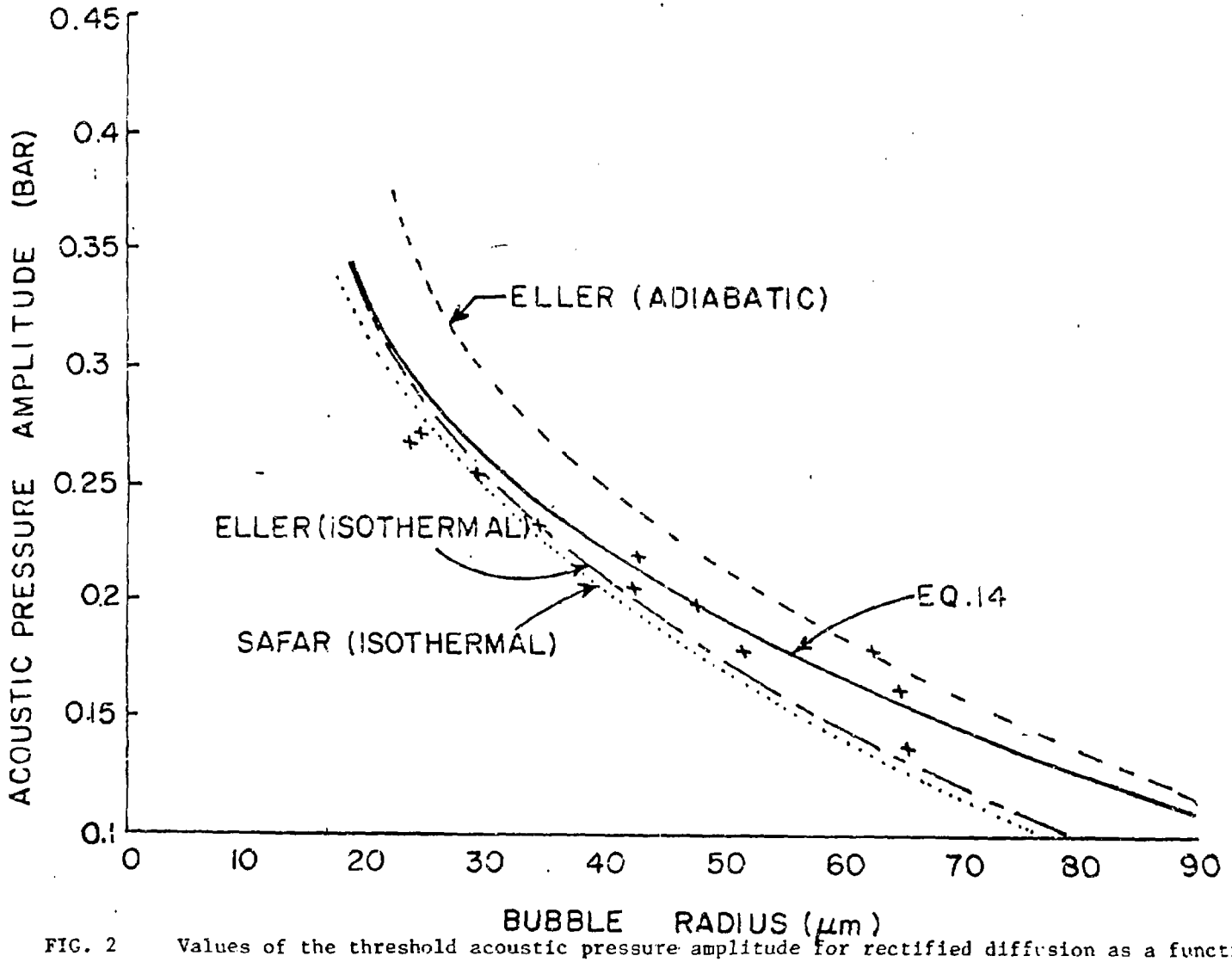


FIG. 2 Values of the threshold acoustic pressure amplitude for rectified diffusion as a function of radius for a surface tension of 68 dyn/cm and a frequence of 22.1 kHz. The dashed curves indicate the calculated thresholds for the limiting cases of isothermal and adiabatic pulsations. The solid curve is calculated from Eq. 14.

that good agreement with theory is obtained for the threshold for rectified diffusion for air bubbles in pure water.

Due to an initial inconsistency in our measurements we examined the effect of slight changes in temperature on the threshold. For the measurements shown in Fig. 3, the following procedure was employed. A sample of water was vigorously agitated, brought to saturation at a particular temperature, and then placed into the test cell within the constant temperature bath, set at the same temperature. After a settling period of an hour or so, the bath was raised or lowered several degrees in temperature, until the host liquid temperature changed a few degrees, and then the bath temperature was manipulated to maintain this new temperature. Measurements of the threshold were then made at this temperature. Using handbook values of the dissolved gas concentration as a function of temperature, and assuming no gas diffusion into or out of the liquid during this period, values of the gas oversaturation or undersaturation of the liquid could be obtained. It is seen in Fig. 3 that a rapid temperature change of 1°C could be easily detected, and temperature changes of 2.3°C, resulting in a gas oversaturation of approximately 4%, could result in a dramatic change in the threshold. Comparisons with theory are good considering the inexactness of the technique. It is seen that if special care is not taken to maintain the gas content of the liquid at saturation, significant errors can be introduced into the measurements by a slight drift in temperature.

2. Growth rate measurements

Measurements were also made of the rate of growth of bubbles by

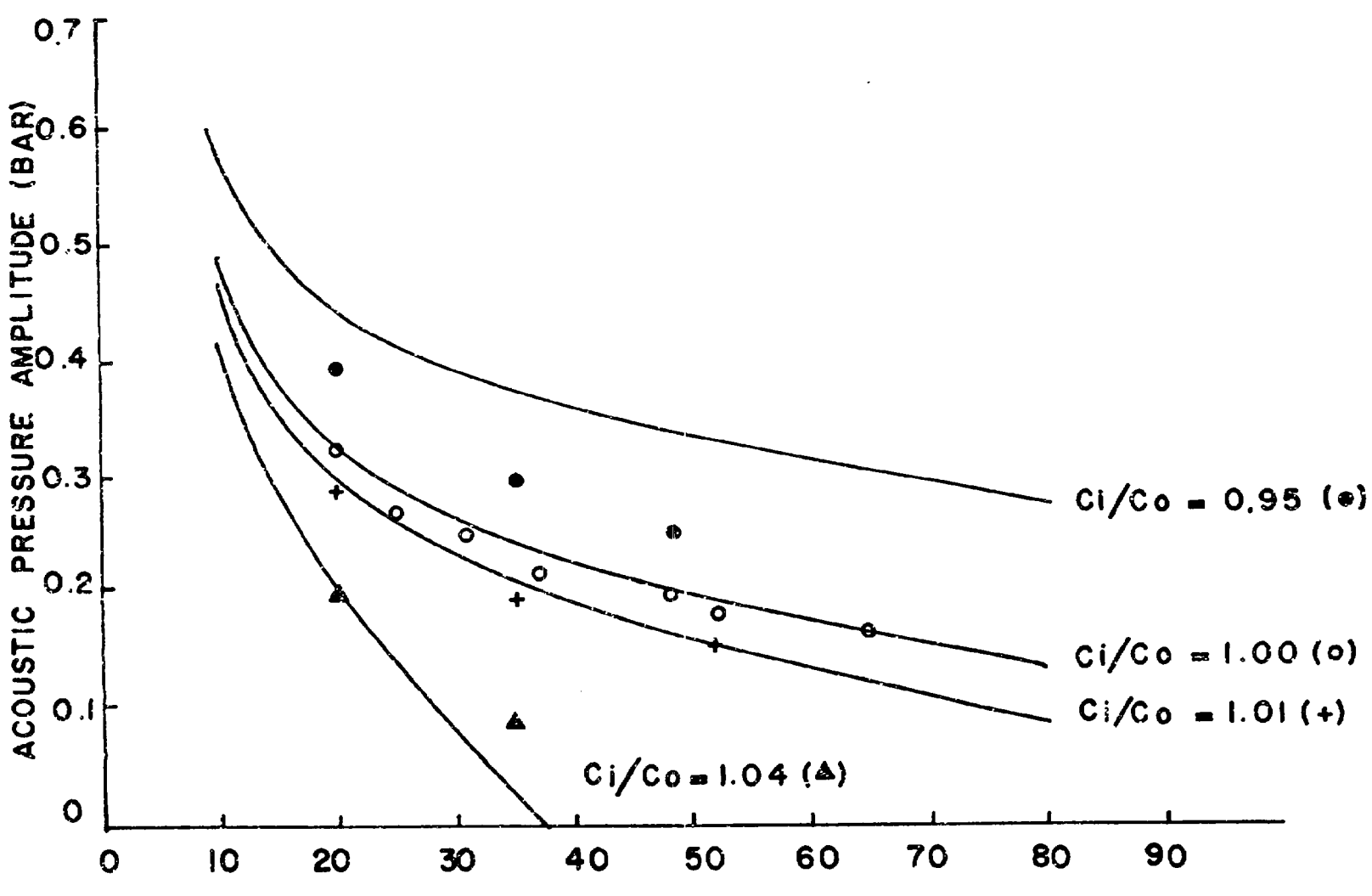


FIG. 3 Variation of the threshold for rectified diffusion with bubble radius for various values of the dissolved gas concentration. The open circles represent no temperature change of the liquid after it was brought to saturation; solid circles, 3°C decrease; plus signs, 1°C increase; triangles, 2.3°C increase. The solid lines are calculated from Eq. 14.

rectified diffusion. Figures 4 and 5 show measurements of the radii of bubbles as a function of time. In Fig. 4, the radius-time curves of four bubbles are shown, each with an initial radius of 35 microns, but for different values of the acoustic pressure amplitude. In Fig. 5, the radius-time curves of three bubbles are shown, each with the same value of the acoustic pressure amplitude, but with different values of the initial radius of the bubble. The concept of a threshold pressure, or a threshold radius is readily seen from the figures. Plotted on the graphs are theoretical curves generated by numerically integrating Eq. 13. It is seen that good agreement is obtained with theory for the growth rates of the bubbles provided surface waves are not obviously present. In Fig. 4, for the bubble with the largest acoustic pressure amplitude, a rapid change in the growth rate was observed near a radius of 60 microns. It was also observed that the scattered light from this bubble changed character, becoming more intense and began fluctuating. The bubble soon began large scale oscillations that have previously been called "dancing motion."[23] We have designated this point on Fig. 4 by an arrow and the letters SW to indicate our belief that the bubble was undergoing surface wave oscillations.

Figure 6 shows the results of a study of the growth rate as a function of acoustic pressure amplitude for a given value of the bubble radius. The experimental points in Fig. 6 were obtained by measuring the slope of the radius-time curves of bubbles at a radius of 45 microns. For example, the top set of data in Fig. 4 show a bubble that has a growth rate of 4.3 microns per 100 seconds at a radius of 45 microns. The data in Fig. 6 also show negative growth rates, that is, for bubbles

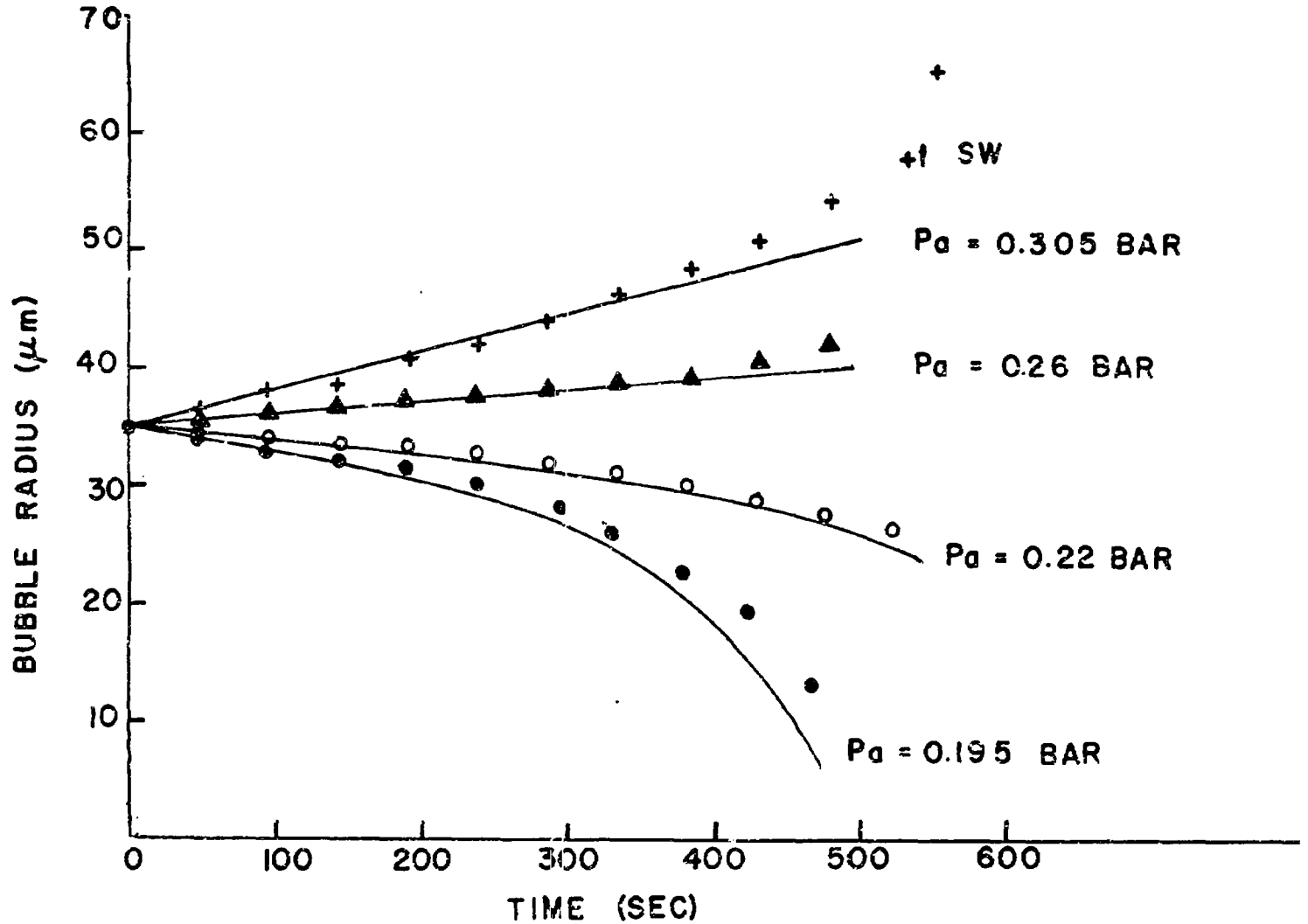


FIG. 4 Variation of the bubble radius with time for a liquid surface tension of 68 dyn/cm, and for various values of the acoustic pressure amplitude. The arrow indicates the detection of obvious surface waves. The liquid was saturated with gas and the solid lines are calculated by numerical integration of Eq. 13

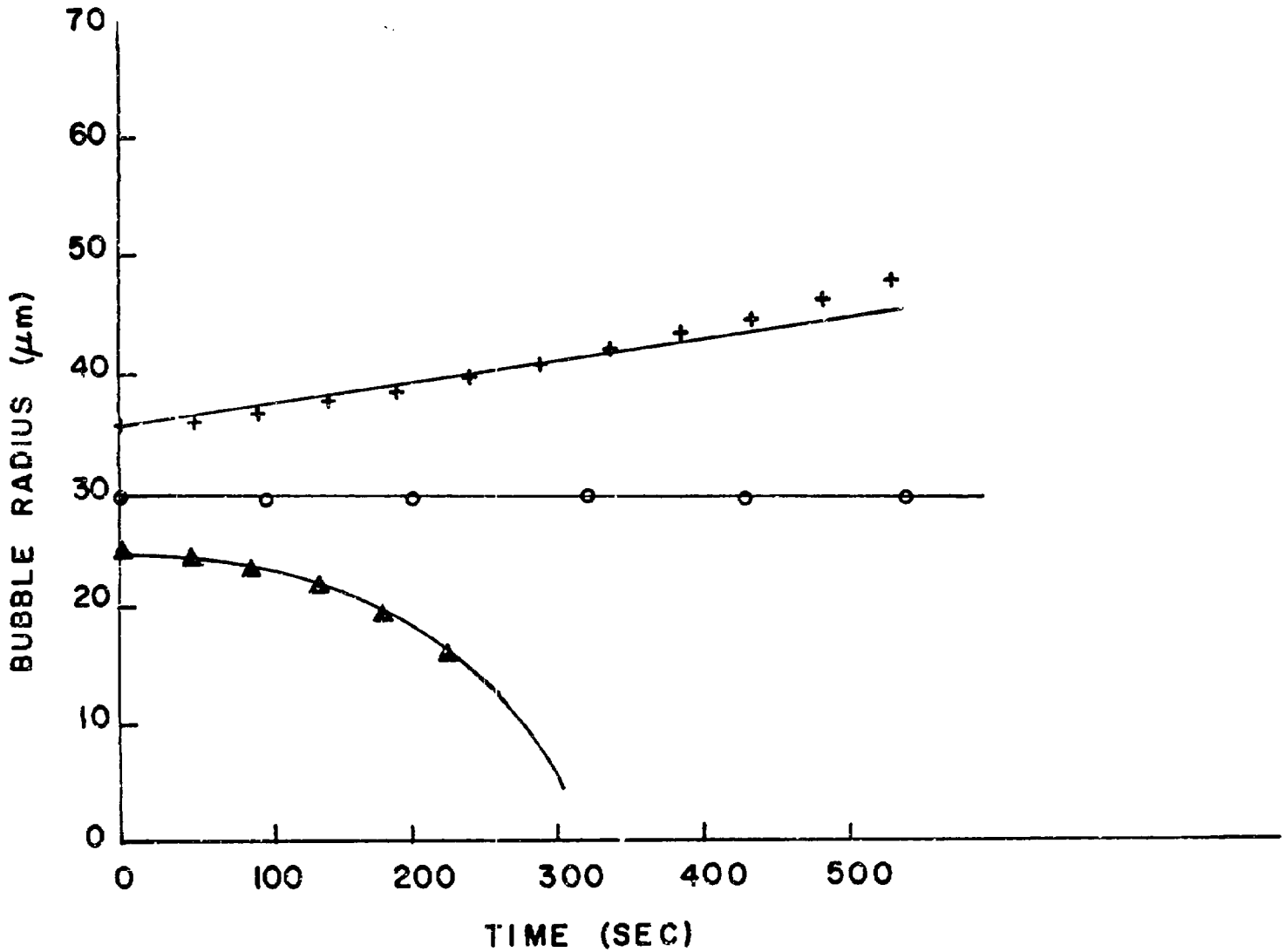


FIG. 5 Variation of the bubble radius with time for a liquid surface tension of 68 dyn/cm and an acoustic pressure amplitude of 0.27 bar. The liquid was saturated with gas. The solid lines are calculated by numerical integration of Eq. 13.

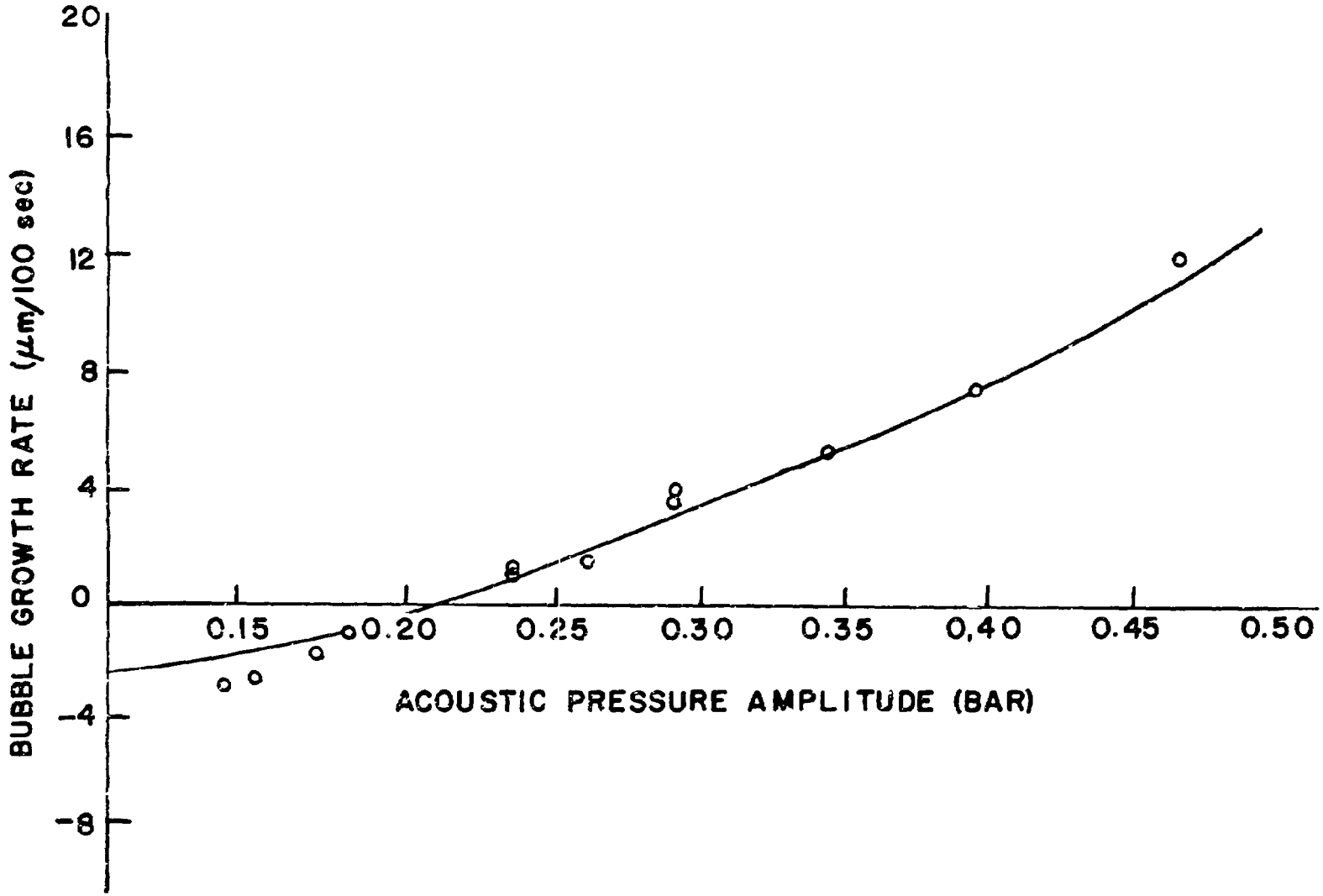


FIG. 6 Variation in the growth rate of air bubbles by rectified diffusion with acoustic pressure amplitude for a surface tension of 68 dyn/cm, a gas saturated liquid, and for a radius of 45 μm. The solid line is calculated from Eq. 13.

that were below threshold and were dissolving. The smooth curve in Fig. 6 is calculated from Eq. 13 with R_o = 45 microns, and shows good agreement with the experimental values.

It is thus seen from the above measurements that so long as there is no surface waves present on the bubble, both the threshold and the growth rate of bubbles by rectified diffusion can be adequately predicted by theory for the modest range of variables studied here.

B. Experiments at reduced surface tension

Measurements were made of the threshold and the growth rate of bubbles in water for which the surface tension was reduced by the addition of small amounts of a surface active agent or surfactant. These measurements tended to give lack of agreement with theory for both thresholds and growth rates.

1. Threshold measurements

Measurements similiar to those shown in Fig. 2 were made for a variety of liquid surface tensions. The procedure for the measurements was identical to those for pure water, except that small amounts of surfactant was added to the liquid after it had been placed into the test cell and had reached temperature equilibrium. A few drops of surfactant added to the 200 cm^3 volume of water in the levitation transducer was sufficient to reduce the surface tension from 68 dyn/cm to 32 dyn/cm.

Figure 7 shows the results of measurements of the rectified diffusion threshold as a function of liquid surface tension for two values of the bubble radius. Also shown are the theoretical predictions from Eq. 14.

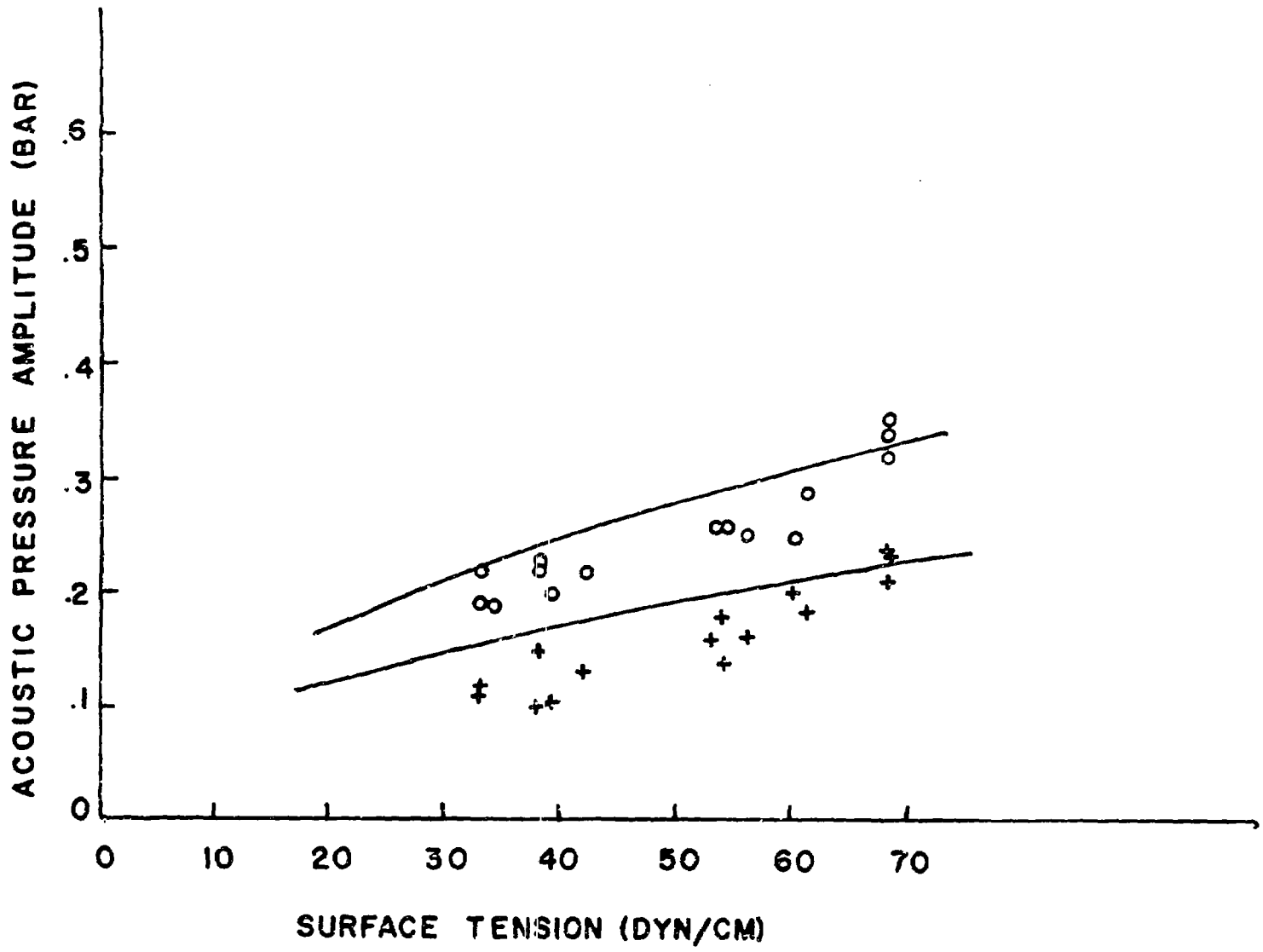


FIG. 7 Variation in the threshold acoustic pressure amplitude for rectified diffusion with surface tension for a gas saturated liquid. The open circles are for a radius of 20 μm; the plus signs are for a radius of 40 μm. The solid lines are calculated from Eq. 14.

It is seen that the data are approximately represented by the theoretical curves but lower values of the surface tension tend to give values of the threshold acoustic pressure amplitude that are consistently less than theory. As an example, the lower 6 data points on the lower curve in Fig. 7 average to a threshold of approximately 0.12 bar, with a standard deviation of 0.02. The theoretical value for the threshold at a surface tension of 37 dyn/cm is 0.165 bar. Thus, the six experimental points average to a value that is slightly more than 3 standard deviations from the predicted value. At best it can be said that there are indications that the threshold is lowered with addition of a surfactant to the host liquid.

2. Growth rate measurements

Measurements were also made of the growth rate of bubbles by rectified diffusion for various values of the liquid surface tension. Figure 8 shows measurements of the radius-time curves of three bubbles, each with an initial radius of 35 microns, but having different values of the acoustic pressure amplitude. The measured surface tension for these data was 32 dyn/cm. Plotted on the graph are the theoretical curves, taken from numerical integration of Eq. 13, that predict the radii of the bubbles as a function of time. Note that the measured growth rates are significantly larger, for those bubbles that are growing, than the predicted values.

In Figure 9 we have collected the growth rate measurements for three different values of the liquid surface tension as a function of acoustic pressure amplitude. Plotted on the graph are the calculated values from Eq. 13. It is seen that substantial increases in the growth rate from

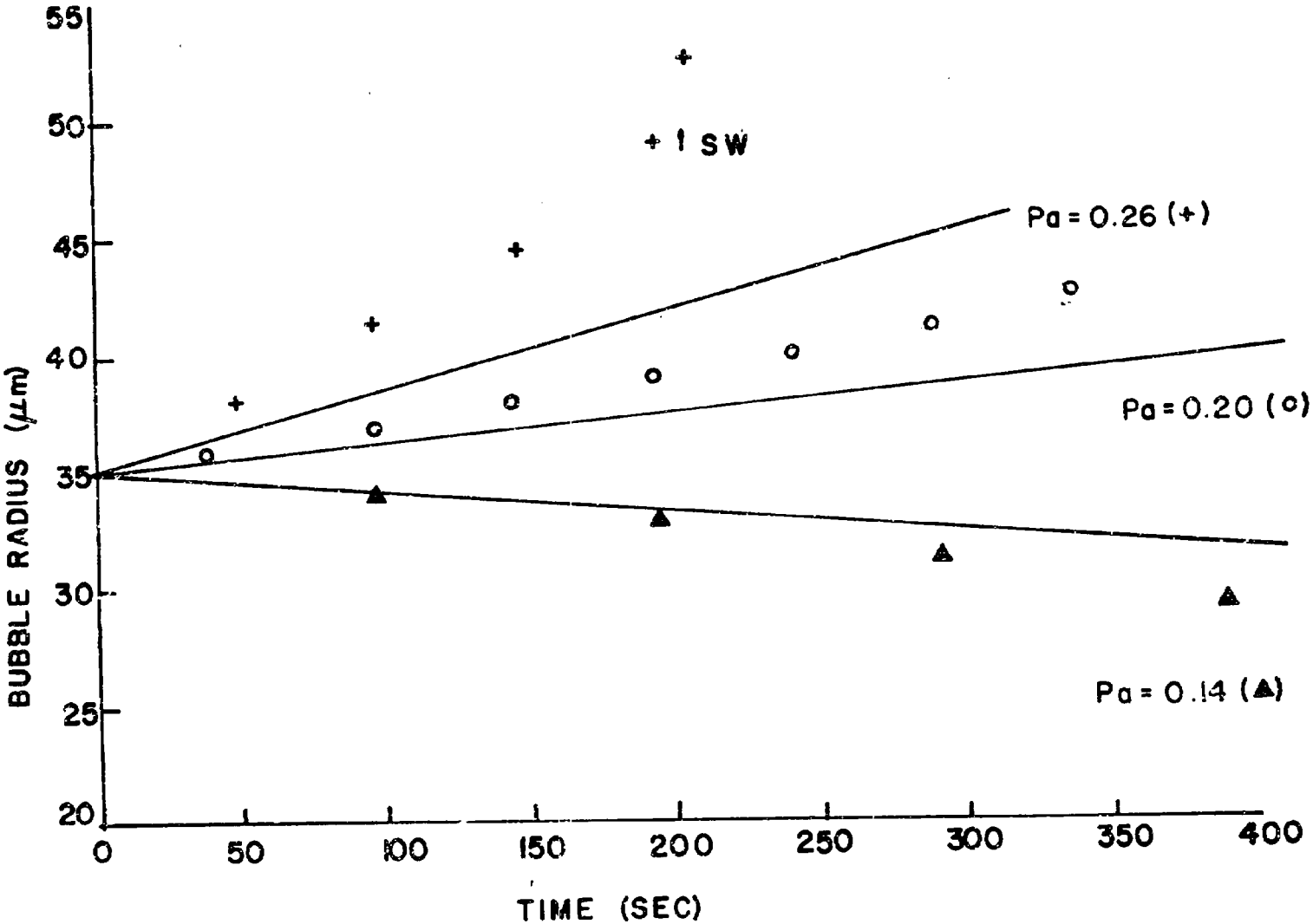


FIG. 8 Variation of the bubble radius with time for a liquid surface tension of 32 dyn/cm, and for various values of the acoustic pressure amplitude. The arrow indicates the detection of obvious surface waves. The liquid was saturated with a gas and the solid lines are calculated by numerical integration of Eq. 13.

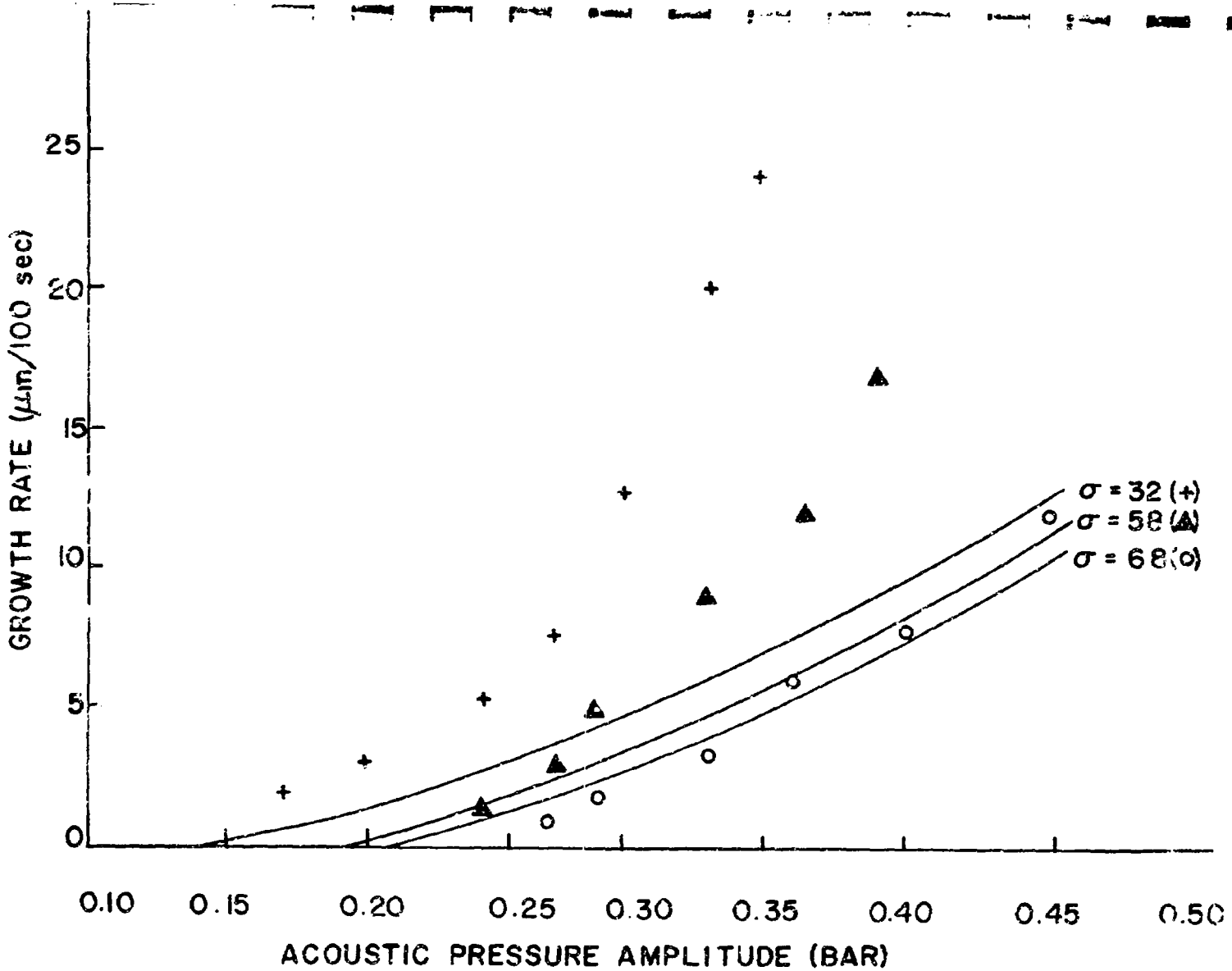


FIG. 9 Variation in the growth rate of air bubbles by rectified diffusion with acoustic pressure amplitude for a radius of 45 μm, a gas saturated liquid, and a range of liquid surface tensions. The solid lines are calculated from Eq. 13.

that predicted by theory are observed when a surfactant is added to the water.

In Figure 10, the measured growth rate for a given acoustic pressure amplitude and for a given radius is shown as a function of surface tension. Substantial disagreement is observed between the experimental points and the predicted values, shown as the curve, taken from Eq. 13.

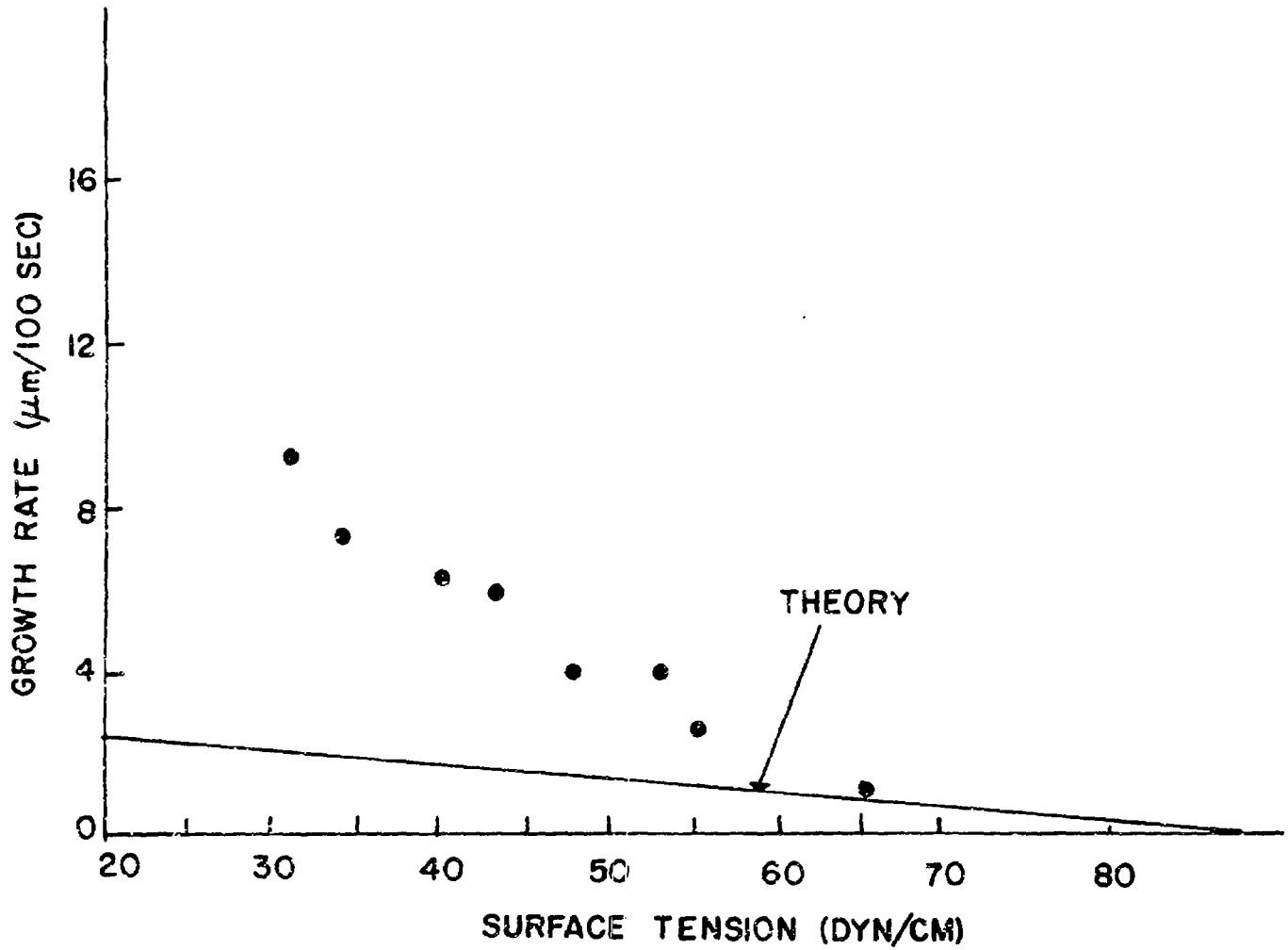


FIG. 10 Variation in the growth rate of air bubbles by rectified diffusion with surface tension for a radius of 50 μm, a gas saturated liquid, and an acoustic pressure amplitude of 0.22 bar. The solid line is calculated from Eq. 13.

IV. DISCUSSION

Eller[10,11] had found that observed growth rates were larger than predicted even though observed and predicted thresholds were about the same. He suggested that a possible explanation for the increased growth rate was due to acoustic microstreaming that greatly increased the availability of gas for diffusion into the bubble. Gould[12] examined the effect of microstreaming on diffusion rates and concluded that the onset of surface wave activity did indeed induce streaming which markedly increased the diffusion rate. Further, he found essential agreement for the predicted and observed growth rates when microstreaming was absent. The results of this experiment corroborate these findings. However, it has been discovered in this experiment that at reduced surface tension, observed and predicted growth rates began to increasingly diverge as the surface tension is reduced. It was first thought that surface wave activity was responsible for the increased growth rates observed. However, in the observations by Gould[12] (see for example his Figs. 7 and 8) these surface oscillations were accompanied by such a rapid increase in the growth rate that a readily observable kink occurred in the radius-time curve. Figures 4 and 8 of this study, which are similar to over 100 other radius-time observations made, show cases of similar kinks in the growth curves, but only after the observed growth rate has deviated significantly from theory for the lower surface tensions.

It is possible to make an estimate of the threshold acoustic pressure amplitude required for generating surface waves. Eller and Crum[23], Hsieh and Plesset[6], and Benjamin[24] have made calculations of the threshold for surface wave production. An approximate expression was given

by Hsieh and Plesset[6] that agrees reasonably well, at a radius of 45 microns, with the more detailed calculations of Eller and Crum[23]. The threshold accustic pressure amplitude required to generate n = 2 oscillations is given by

$$P_A = \frac{14.4 \quad \sigma \; P_o(1 - \beta^2)}{\rho\omega^2 R_o^3} \quad . \tag{18}$$

For a radius of 45μm and a surface tension of 68 dyn/cm, P_A = 0.49 bar; for a surface tension of 58 dyn/cm, P_A = 0.42 bar; and for a surface tension of 32 dyn/cm, P_A = 0.23 bar. It is seen from Fig. 9 that the deviation of the measured growth rate from the predicted value occurs before these surface wave thresholds are reached. It is noted, of course, that the surface wave thresholds listed here are calculated values, and actual thresholds may be somewhat less. Some measured values of the threshold for surface wave generation do exist[12,23], but there is considerable scatter in the results by Gould[12], and no mode identification in the results of Eller and Crum.[23] Precise measurements of the threshold for surface wave generation are required in order to reject or accept this explanation for the increased diffusion at low surface tensions.

A second possible but rather speculative explanation is now proposed to explain the increased diffusion.

It has been known for some time[25,26,27] that surface active agents can reduce the evaporation rate of water. Mansfield[26] has shown that the water loss by evaporation from large lakes may be reduced by as much as 50% by addition of an appropriate monomolecular film. Archer and La Mer[28] found that the rate of evaporation may be reduced by a factor

of 10^4 by the application of a fatty acid monolayer. In an attempt to account for these large reductions in the evaporation rate, Barnes[29] and co-workers calculated the evaporation resistance assuming that water molecules could only evaporate through holes in the monolayer film. With a reasonable estimate of the ratio of hole area to monolayer area they were able to calculate values of the evaporation resistance that approximated the measured values. Mansfield[26] also found that expansion of the film by as little as 10% reduced the evaporation resistance considerably, while compression of the film had little effect on the evaporation resistance.

The existence of surface active materials present on air bubble surfaces have been previously suggested as affecting the dissolution,[30,31] and the resonance frequency[32] of small air bubbles. Elder[33] visually observed the presence of a layer of surface active materials on air bubbles and its effect on acoustic microstreaming. Liebermann[34] examined the effect of surface contamination in reducing the diffusion of a stationary air bubble that was allowed to slowly dissolve in water. However, he states that he was unable to find a significant (by a factor of two) effect on the diffusion rate by adding surface-active agents such as detergents. He did find that air bubbles left a solid particulate residue when dissolved on a glass slide.

It appears that some facts point toward the presence of a surface active monolayer as a possible explanation for the large growth rates observed in rectified diffusion. The observation by Mansfield[26] that expansion of a film allows more evaporation than when the film is compressed is directly applicable to the case of a pulsating air bubble. If the film

allows more penetration of molecules on expansion than on compression, a rectification of diffused air would occur, in agreement with the above observations on rectified diffusion. Further, if, on expansion, the film is broken up, the time required for the film to repair itself is long compared to the period of the acoustic cycle. Lord Rayleigh,[35] in an ingenious experiment performed in 1890, showed that surface active agents needed at least 10 msec for their migration to the surface and their resultant reduction in surface tension. On the other hand, the compression may cause the repair mechanically so that no diffusion of the film from the liquid substrate is required. At any rate, it seems logical to assume that if evaporation rates can be retarded by four orders of magnitude, diffusion rates can be affected enough each cycle to cause a significant increase in the amount of gas contained within the bubble. The observations by Liebermann[34] that diffusion rates may be slightly affected by surface contaminants support this view. With the rapid pulsation, 22.1 kHz, very little rectification is required each cycle to significantly affect the growth rate. Further, if the film has a rectifying effect, it should lower the threshold for rectified diffusion. Although the measurements do not show an obvious and distinct lowering, there are indications of a threshold reduction in these measurements. Finally, the absence of any kinks in the radius-time curves imply no immediate change in boundary conditions, as would occur if a bubble underwent surface wave oscillations.

A third explanation, and perhaps the most plausible one, is that some microstreaming can be induced without surface wave generation. It was observed by Elder[33], for example, in his studies of microstreaming

that when surface active agents were added to water, a thin film would form which tended to present a no-slip boundary condition for the bubble. This boundary layer effect increased the microstreaming until it was broken up by the bubble pulsations. It is possible that the addition of small amounts of surfactant induces microstreaming without surface waves. This streaming would increase the growth rate for all values of the acoustic pressure amplitude and bubble radius, and would show no distinct inception threshold.

V. ACKNOWLEDGEMENT

The author wishes to acknowledge the assistance of Ron Roy in the data acquisition, and of the Office of Naval Research for their financial support.

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