SOME THEORETICAL CONSIDERATIONS CONCERNING IONIZING RADIATION INDUCED ACCOUSTIC CAVITATION

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CHAPTER I

INTRODUCTION

Primarily this paper is proposed as a theoretical study of the possibility of building a bubble chamber or other kind of radiation detection devices working on the ultrasonic cavitation effect. How such an idea has arisen will be explained in the following paragraphs.

One can observe the formation of bubbles in a liquid in various occasions. Further, in some cases, the bubble formation can be greatly amplified by the presence of radiation. Actually the construction of bubble chambers is one of the applications of this effect, - to enhance the bubble formation of a superheated liquid by the radiation to be detected.[1,8,12-20] On the other hand, by putting a liquid under a sonic vibration with a strong enough amplitude and a proper frequency, bubbles will also come into being which has been referred to as a cavitation phenomena. Although it has long been proposed but not observed until the successful experiment due to Lieberman[2], this process can also be enhanced by the introduction of certain radiations. Certain liquids, such as water or propane, when irradiated in beta-ray radiation will reduce their waiting time for sonic cavitation appreciably. Having laid down these two effects in parallel, one can easily arrive at our original proposal.

However, besides those similarities between the above-mentioned effects there are also some radical differences between them. For example, one is able to understand the former effect through a pure thermodynamical consideration and the latter in a more or less dynamical way. Furthermore, the former one is always a fast process as compared with the latter.[2,10,11] Usually, the times required for these two processes yield to a difference of the order of magnitude of 10^7. Such a formidable difference is due, mainly to the fact that the growth of a bubble in a bubble chamber is attributed to the evaporation of a superheated liquid which requires a time of the same order as the collision time of the molecules of the liquid in the chamber.[8] On the other hand, the growth of a bubble under sonic vibration is basically a resonant phenomena which can be detected only after a great number of vibrations.[2,4] We shall discuss the significances of these facts as well as some others as we proceed. Table I will give a rough comparison between the two processes.

TABLE I

A COMPARISON OF BUBBLE CHAMBER AND LIEBERMAN'S EXPERIMENT

	Mechanism of Bubble Formation	Radiation Effects on Bubble Formation	Response Time
Bubble Chamber	A sudden superheating effect is obtained by reducing the pressure of the chamber in a short time. Bubbles will form in this superheated liquid.	Thermal spikes generated by scattered particles serve as the embryos of the observed bubbles.	Few microseconds
Lieberman's Exp.	A small gaseous embryo will grow in the sonic field by means of its amplified vibration of the liquid-gas interface of the bubble, etc.	Increase the number of embryos in the liquid in the same way as stated above.	The waiting time for cavitation is reduced by about half minute as compared to one minute (approx.)

It can be seen from Table I that the amplification of both processes caused by radiation is believed to be of the same character. Furthermore, for reasons we shall explain later, both processes are concerned with a special kind of phase transition which will be called a liquid-to-bubble transition. A phase transition of this kind is only an intermediate transition of a liquid-to-gas transition in a superheated liquid, nevertheless, it is the only phase transition possible, if any, in the case of sonic cavitation.

Ultrasonic cavitation can be detected by various methods. Only two of them are of great importance so far as the present topic is concerned. They are: 1. Detection by the direct observation of the visible bubbles formed; 2. Detection by recording the shock waves caused by collapsing bubbles. If our devices are possible, the first one will give us a continuously operating bubble chamber which enables us to observe the tracts of radiation as long as the sonic field is on and the second one will give us a device similar to an ionization chamber, i.e., the strength of the shock waves as well as their spectrum will inform us the character of the radiation.

Unfortunately, sonic cavitation phenomena have not yet been extensively studied as the bubble chamber characteristics. Consequently, a lot of crude assumptions have been ventured concerned with cavitation phenomena in the following sections whenever it is necessary. An even worse situation will be encountered when we come across the theories of liquid state, which is, actually, a completed unexplored field. Owing to all the difficulties hereby arisen together with some others all the conclusions in the coming sections are by no means critical.

CHAPTER II

THEORY

1. The Formation of Embryo Bubbles in a Liquid by Radiation

When a fast moving particle carrying enough energy dissipates all its energy in a small region in a liquid, a thermal spike is formed. Since it can be shown that the heat conduction in this case is a subsonic process,[1,8] it is reasonably safe to say that all the dissipated energy will reappear in a completely randomized form (i.e., heat energy). It should be noted that not all particles carrying sufficient energy can produce thermal spikes in liquid. The criteria is whether this energy can be randomized in a small region or not. In other words, it depends largely on the collisions involved and consequently the path taken by the energetic particle. Further, it is believed that the most effective particles that will generate thermal spikes are secondary particles, i.e., the particles scattered by the primary component of the radiation.

Consider a scattered particle with kinetic energy E in a liquid of density ρ. Statistically this particle will be completely stopped by its collisions with the surrounding molecules within a distance R. F. Seitz[1] suggested the following formula to express R

$$R = 0.58\ E(\text{in kev})^2 / \quad (z/A) \tag{1}$$

where z denotes the charge of the nuclei of one molecule of the liquid and A the molecular weight of the liquid. Here we have assumed deliberately a pure liquid.

One of the most apparent conclusions from the above is that the energy of the scattered particle will be randomized over a wider and wider range as E, the energy of the scattered particle, increases. This explains why only secondary particles are of the most importance in generating thermal spikes for the primary particles are usually too energetic to randomize their energy within a small enough region.[2]

Now confining our discussion in small thermal spikes, we can assume that they are spherical in shape with a diameter R, then the temperature of the spike, immediately after the randomization of the energy of the particle, will be T' and is given by

$$T' = J\rho^5 (z/A)^6 / \tfrac{4}{3}\pi(0.58)^3 E^5 + T_0 = T + T_0 \qquad (2)$$

where J stands for the Joule constant and T_0 to the liquid temperature. Here, we have imposed another assumption that the temperature is uniform throughout the spike.

Under ordinary conditions such spikes will not last very long, for no matter how slow the conduction process is the spike will cool off sooner or later.[1,2,5] Now, suppose that the liquid is already superheated or is subjected to a sonic disturbance, the situation will be quite different. A phase transition breakdown or a shortening of cavitation waiting time will then be observed. Macroscopically, such effects can be explained as follows.

Suppose that the temperature of the thermal spike does not change appreciably during a certain duration (few periods of the sonic vibration, say) and the energy distribution within the spike can be dexcribed in a cannonically way; that is to say the spike is large enough to be further subdivided into even smaller regions with definable temperatures. Then, the probability of a phase transition which converts the spike into a gaseous bubble of the same dimension can be expressed as

$$W' = Ke^{-E'/kT} \tag{3}$$

where E; is the Gibbs function of the gaseous bubble at the same temperature T; of the spike and k of the Boltzmann constant. One of the shortcomings of the above expression which is common to all the thermodynamic expressions is that the constant K cannot be determined here. Fisher,[6,7] after a statistical consideration, suggested an expression for K as

$$K = NkT/h \tag{4}$$

where N is the number of molecules of the spike and h the Plank constant. However, the validity of this suggestion has not yet been confirmed experimentally. In order to get rid of this constant, we consider the more specific problem as follows. Using the same argument as above we have for the transition probability in the region outside of the spike for the formation of gaseous embryo of the same size as specified above as

$$W = ke^{-E/kT} \tag{5}$$

Then, it is also meanful to consider the relative probability of phase transition of the spike against its background. Define C the relative probability as

$$C = W/W' \doteq e^{-(1/kTT')(E\Delta T)} \tag{6}$$

if E' does differ from E appreciably.

The above-defined C not only gives us an expression free from all the undefined terms but also gives us a qualitative measurement of the contrast of the bubbles along the track of the radiation against the background in a bubble chamber or a possible "cavitation chamber."

Since there is a great deal of experimental data in bubble chamber measurements this also suggests an experimental check of the above-mentioned discussion.

It should be noted that upon the application of the preceding theory the change of phase should take place promptly as soon as the scattering energy becomes completely randomized (this corresponds to a high thermal spike temperature) for otherwise the observed C will be much smaller than the computed one. Of course it is quite possible that at the first few collisions a change of phase may have already taken place and all the later collisions (which are less energetic) will only enhance the evaporation of the surrounding molecules to make a bigger gaseous bubble. What really happens in this process can never be revealed in our present consideration since only energy relations are used here and which are irrevelant to the mechanistic details. Nevertheless, if a thermal spike in a liquid bubble chamber becomes a gaseous embryo it will continue to grow and give us a visible bubble. A comparison between the experimental countings of bubbles along the path of the primary particles of the radiation and the number of the theoretically evaluated possible spikes gives a very large probability of transition. As we have mentioned above, this can either be explained by a large value of C or can be considered as an indication of the fact that transition occurs during the first few collisions. Under the present circumstances both are highly possible. The difficulties we encountered here can be understood if we try to study this problem microscopically. In the first few collisions the scattered molecules usually are too energetic to be in a liquid state. However, since they are so few in number it is just meaningless to speak of their state. As the scattering process goes on and on enough number of scattered molecules will be accumulated, however, the energy of the scattered molecules are less energetic and the phase transition will be only a probable one. A detailed study of the mechanistic relation of a fast moving particle with the surrounding molecules in a liquid and the collection of a number of fast moving molecules with the velocity distribution resulted from a scattered charged particle seems necessary to solve this problem, nevertheless, they are out of the scope of this paper.

2. The Phase Transition

As we have pointed out in Section 1, the Formation of Embryo Bubbles in a Liquid, the problem of detection of a thermal spike in a liquid depends largely on whether or not a phase transition will take place at the spike. In this section this problem will be considered in more detail.

First of all let us consider a general phase transition from a certain parent phase to its daughter phase. Denote by ΔF_i the difference in free energy (Gibbs function) of an embryo in the daughter phase from that of the parent phase where the parent phase is the predominant phase. As long as external field can be neglected ΔF_i can be expressed as a linear combination of three terms:[6]

$$F_i = Ai^{2/3} + Bi + Ci \qquad (8)$$

where i is the number of molecules in this embryo. They are correspondingly the interfacial free energy, the bulk free energy and the strain energy. This expression is true provided that the embryo is both spherical and homogeneous.

The total free energy for n_i embryos of i molecules in the parent phase will be

$$F = n_i \Delta F_i - T \Delta S_i \tag{9}$$

where ΔS_i is the entropy of the embryo. By minimizing this expression one gets the probability of the existence of n_i embryos in the parent pH phase as

$$n_i / n = e^{-\Delta F_i / kT} \tag{10}$$

where n is the total number of molecules in the whole system.

It is clear that the transition will not occur unless ΔF_i decreases towards negative infinity as i increases. The problem of phase transition here reduces to the problem of the dependence of the coefficients A, B, and C upon the thermodynamic conditions. In order to make it easy to visualize, let us take the daughter phase as a gaseous bubble, then A will be a function of the surface tension of the liquid which is temperature dependent and B and C will be functions which are highly pressure and temperature dependent.

The behavior of the ΔF_i function for large i will determine which one of the two phases are more stable; whether the phase transition will actually take place or not, depends largely upon the behavior of the ΔF_i function for small i. For large i the property of this function is predominantly determined by (B + C). Suppose that the daughter phase is more stable, then (B + C) will be a negative quantity. However, since A is usually a positive quantity (for example, surface tension coefficient is usually positive) we will find a hump in the $\Delta F_i / kT$ - i plot and consequently any embryo of the daughter phase has to overcome this barrier (hump) in order to jump to a stable phase. This is shown in Figure 1.

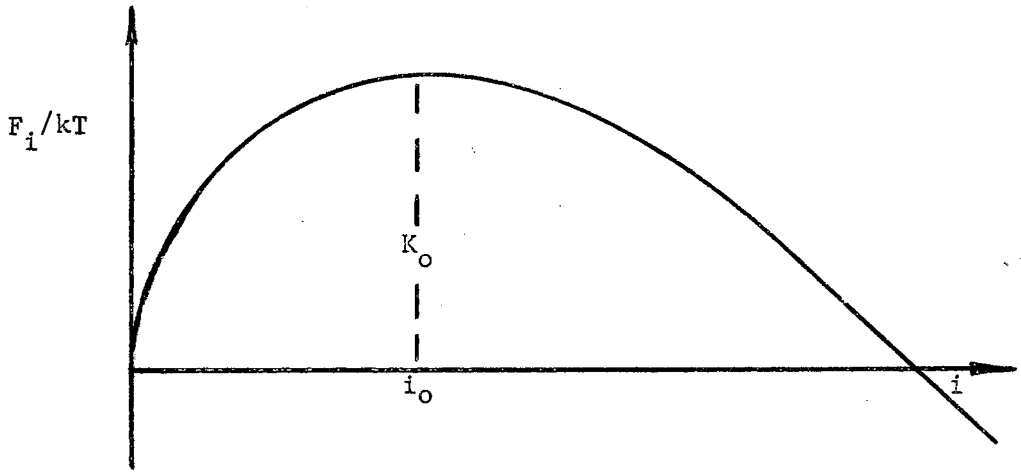

Figure 1. Representation of $\Delta F_i/kt$ vs. i for Stable Daughter Phase

By a simple mathematical manipulation we can see that the height increases as the critical value of i at the hump goes down. However, as i_o, the critical value of i approaches one all the above arguments lose their meaning for we can never consider the phase of just one molecule. Furthermore, in all the above computations i has been used as a continuous variable which becomes a very bad approximation for small i's. the dependence of the $\Delta F_i/kT$ - i line on i_o is represented in Figure 2.

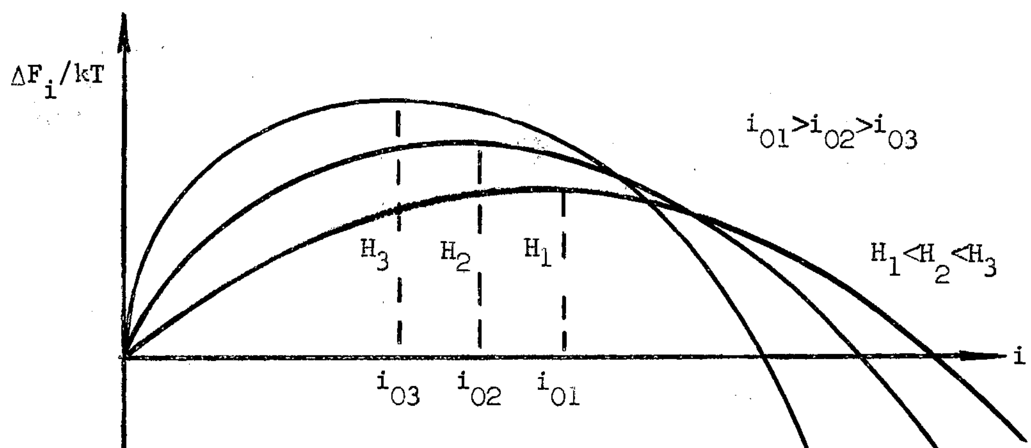

Figure 2. The $\Delta F_i/kT$ - i line for different i's.

If, on the other hand, the parent phase is more stable, $\Delta F_i/kT$ will be a monotonic increasing function with respect to i since all the coefficients A and (B + C) are positive and as i increases this function will be almost linear. This case is illustrated in the following plot.

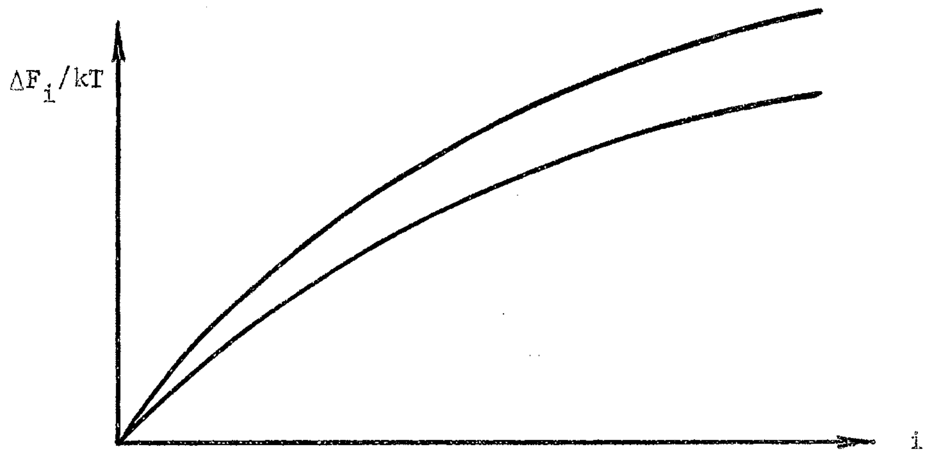

Figure 3. F_i/kt - i Line for Stable Parent Phase

Of course there is an intermediate case where the function (B + C) becomes zero. In this case, the fluctuation of the environment will also be taken into account for its effect may be considerable in a statistical consideration.

Care must be taken that the above reasoning is rather qualitative, yet it can be served as a guide in the study of specific cases such as the liquid - gas transition.

For this particular topic, using r instead of i as the variable the most comprehensive expression for the energy of a gaseous bubble in a liquid can be set up as follows.[8]

$$E_b = 4/3\pi r^3(P_H - P_v) + 4\pi r^2(\sigma - T_o d\sigma/dT)$$
$$+ 4/3\pi r^3 \int \rho_v(C_v dT + L) + 2\pi\rho_L r^3 r2 = \alpha(T) \qquad (11)$$

The first term of the right-hand-side of the above equation gives the bulk energy of this bubble and the second term the interfacial energy and the rest are respectively the energy required for a pure phase change (i.e. no expansion or change in shape etc.), the kinetic energy of the liquid and the viscous energy as a function of temperature. And,

p_H = the hydrostatic pressure of the liquid

P_v = the pressure in the bubble

σ = the coefficient of surface tension of the liquid in its vapor

T_o = the liquid temperature

L = latent heat of the gas bubble

C_v = specific heat of the vapor of constant volume

ρ_v = density of the vapor

ρ_L = density of the liquid

In the above equation if we drop the second order term in the surface tension energy and if the kinetic energy can be neglected in this case; and we further assume that $(P_H - P_v)$ decreases with increasing r and approaches a lower limit for large r provided that r_o is small, then we can find the same basic characteristics in the E_b vs. r plot as we find in the $\Delta F_i/kT$ vs. i plot.

In a bubble chamber the criteria ofwhether a bubble will grow or not will depend on the initial radius of the bubble; (if r is greater than r_o, it will grow and vice versa). Actually, Seitz' has developed his theory of bubble chamber based on this idea and which is, as far as all present experiments are concerned, a successful theory.

However, in our problem the situation is quite different. For in a non-superheated liquid the critical radius of a stable gaseous embryo bubble will be infinity which, certainly can never be reached by a thermal spike. It seems that the dynamic property of a bubble in a liquid under sonic vibration plays a big role, and this point will be considered in the coming section.

3. Sonic Induced Cavitation

Among all the theories and arguments which have arisen from this problem one point in general agreement has been reached that the cavities generated in a sonic field are due to the pre-existing embryos in the liquid. Actually, this argument comes from the detailed dynamical picture of the growth of embryos.[4,5,22] Following this route a lot of theories have been developed and to certain extent tested. Unfortunately, all the theories so far developed are still more or less qualitative in character which makes it difficult to apply them to our problem. Therefore, by setting up some assumptions, we shall try to arrive at some quantitative ideas.

In the cavitation process one assumes that the gaseous bubbles can exist in liquid for a long time as compared with the period of the sonic vibration used. Further, if the liquid has been properly degassed, no real gaseous state in large can be produced, as one may see from the following thermodynamic considerations. Here, the main differences from ordinary applications of thermodynamics are that we have an external field and the equilibrium is held between a liquid state and a rather particular "bubble state". However, we may assume that the usual thermodynamic method still applies to this situation and consider the external field as a kind of pertubation. To this connection, naturally, one has to consider the mechanical motion of the bubble under sonic vibration.

The most simplified picture of the motion of a gaseous bubble in liquid under sonic aggitation was given by Smith.[21] According to his theory the predominate mood of motion of the bubble is its radial motion -- this is of course only true for small vibrations. Further, the redial motion of the bubble can be described by an equation similar to that of a periodically forced vibration and the resonance will establish when the rest radius (the radius without sonic vibration) of the bubble r_o

$$r_o = r_{sc} = \sqrt{\frac{3\gamma P}{\rho}} \Big/ 2\pi fn \qquad n = 1, 2, 3, \ldots; 1/2, 1/3, \ldots$$

$$\gamma = C_v / C_p \tag{12}$$

where f is the frequency of the sonic vibration.

In general, we can expand the time dependent function of the radius r of the bubble in its Fourier expansion in terms of f.

$$r = r_o = \sum^{\infty} r_n e^{i2\pi f_n t} \tag{13}$$

where the rn's are certain functions of the frequency and the amplitude of the sonic vibration.

Without any further information, one thing can be said about the r_n's that they are usually very small quantities unless r_o coincides with the Nth resonante radius of the vibration.

Now, we can consider how such a radial motion affects the energy of a gaseous bubble in liquid. Since the vibration of a bubble in this case is almost a perfect adiabatic process there will be no change in the bulk energy of the bubble. Moreover, for small vibrations of the bubble the effects due to the viscosity of the liquid and the gas can be neglected. Finally, in equation (13) the only term we have to take into account will be the interfacial energy term, in our case, the surface tension term.

$$E_s = 4\pi r^2 \sigma \tag{14}$$

Where we have neglected the second order term. The contribution of this term to the Gibbs energy of the bubble can be written as

$$F_s = 4\pi\sigma(r_o^2 + r_1^2 + r_2^2 + \dots\dots\dots) \tag{15}$$

We can consider the first term in the parenthesis as the contribution to the Gibbs energy without the sonic pertubation and the rest terms the perturbating terms. Since, we have already mentioned that the r_j's are usually neglibible we can, therefore, write,

$$F_s = 4\pi\sigma r_o^2 \tag{16}$$

$$F_s = 4\pi\sigma(r_o^2 + r_j^2) \qquad r_o = r_{sc} = \sqrt{\frac{3\gamma P}{\rho}}\, 2\pi fj \tag{16a}$$

It can be seen that the Gibbs energy of gaseous bubble under sonic vibration does not differ from that of the static case unless its rest radius coincides with one of the resonant radii of the vibration. This will give, in the F/kT vs. r plot, peaks at each resonant radius as follows,

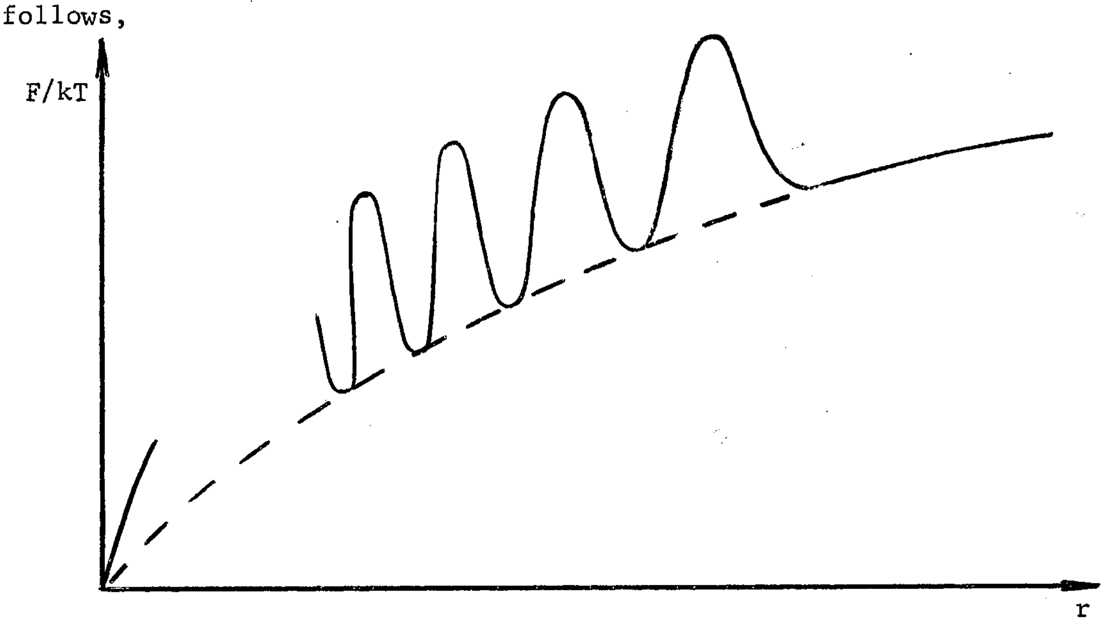

Figure 4. . F/kT Schematic Plot of a Gaseous Bubble in a Sonic Field

From Figure 4, we can see that between each pair of peaks there is a minimum point for the F/kT value. This means, thermodynamically, that bubbles of certain radii can exist in meta-stable states between the peaks of the F/kT vs. r plot. Furthermore, bubbles with radii less than the smallest appreciable radius will be forced to collapse and consequently end up with shock waves. This conclusion checks with the results worked out by Nolting and Nepires in 1949 based upon a pure mechanistic consideration.[3]

Like all the thermodynamic theories this consideration does not give any reason for the inception of such bubbles and the actual mood of motion of the bubbles when their radii happen to be different from the radius of minimum Gibbs energy. The pre-existance of gaseous embryos in the liquid or merely the fluctuation of the free volumes of the liquid can, perhaps partially, answer the former question. However, strictly speaking, the latter can only be answered by a mechanistic study of the bubble vibration such as those developed by Nolting et al., and others or, in a more practical way, by means of experiments. Despite the differences in their bases and other respects, all the studies in this concern have one result in common that the collapse of a bubble takes a time of about one oscillation of the sonic vibration whereas the amplification of a bubble takes many cycles.[3,4,9] This fact greatly reduces the possibility of observing the bubbles produced by radiation promptly and thus reduces the possibility of constructing a "cavitation chamber" to trace the track of radiations. Nevertheless, even if all these expectations are true still it does not rule out completely the possibility of our original proposal. In the next section we shall see how such difficulties can be possibly removed.

4. The Detection of Radiations Through Sonic Cavitation Phenomena

Sonic cavitations have been detected through several different ways, however, the usual criteria utilized in the detections are either based on the shock waves produced by the collapsing bubbles or the direct visualization of the cavitaties (bubbles) in the liquid. If, it is possible for a radiation of a certain kind to enhance the cavitation of a liquid so that this enhancement can be measured by one way or the other, then such a radiation is sonically detectable.

It seems inevitable that radiations will produce thermal spikes in liquids and consequently gaseous embryos of certain radii. If n bubbles in a bubble chamber are observed along the track of a particle from the radiation then, according to Seitz's theory this particle is capable of producing, at least, n gaseous embryos with radius larger than r_c along the track where

$$r_c = 2\sigma/p$$

It has also been pointed out in Section 3 that whether an embryo will give a noticeable effect (i.e. amplified bubble or shock wave) depends on its initial radius. In order that the n embryos in the bubble chamber be sonically detectable in a "cavitation chamber" using the same kind of liquid we must have, by bringing the two theories together,

$$r_c > r_{sc}$$

Such a relation can be illustrated in the following two figures (5 and 6) (remembering that the critical condition is that)

$$r_c = r_{sc}$$

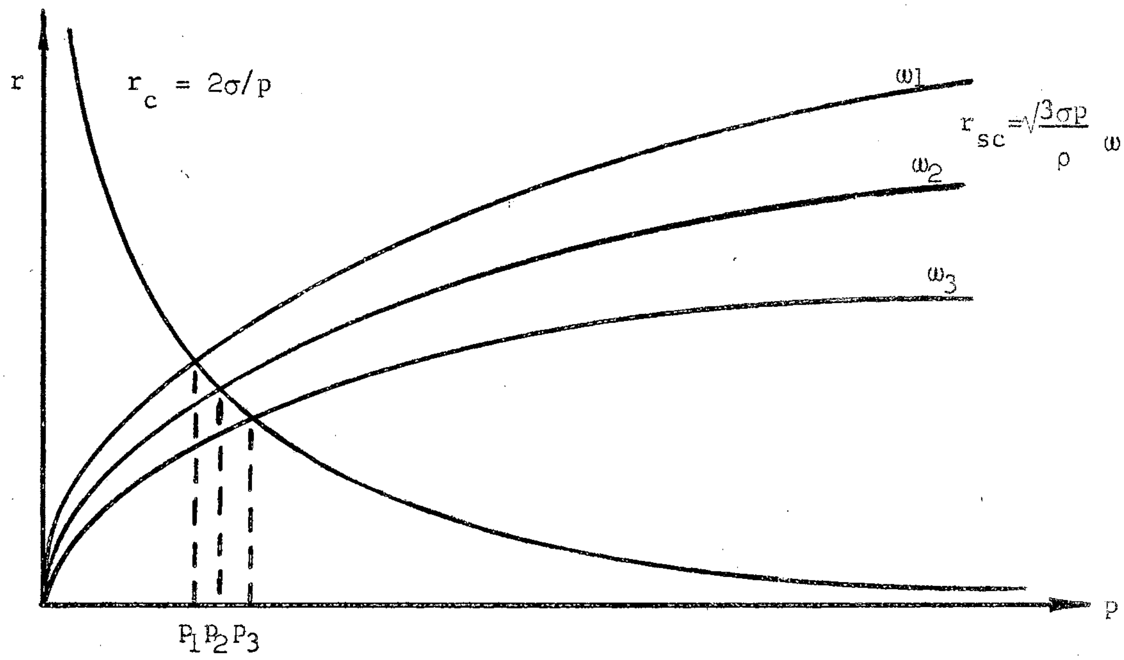

Figure 5. p vs r Plot of Sonically Detectable Embryos

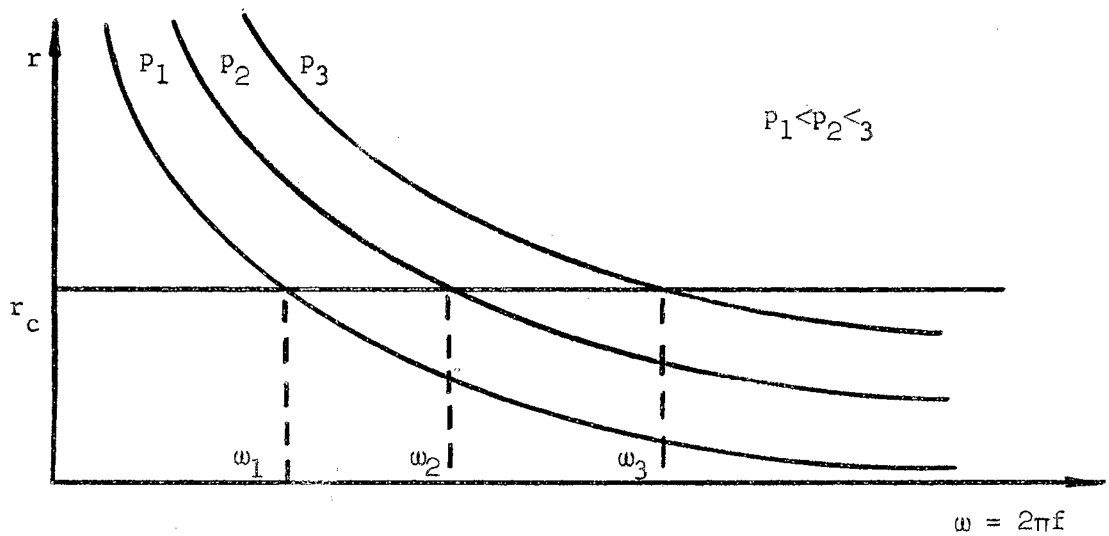

Figure 6. r vs ω Plot of Sonically Detectable Embryos

Once more one has to notice that the sonic cavitation is a comparatively slow process which makes a prompt detection of radiation effect through this method almost impossible.

CHAPTER III

CONCLUSION

The formation of gaseous embryos in liquid as well as their destinies under a sonic aggitation have been studied in the previous sections. Also the effect of radiation upon them has been scrutinized. It reveals at least one fact that the formation of gaseous embryos and consequently, cavities in a liquid can be enhanced by radiation with suitable characteristics. As an instrumentation for the radiation detection the result is rather pessimistic; for owing to the radical properties of the cavitation phenomena a direct tracing or a prompt detection of radiations are very impossible.

Nevertheless, this paper does open a quite unexplored field of study, namely, the relation between radiation and the states or the structure of liquids. So many theoretical possibilities are evident that, as usual, in physics, experimental determination of possibilities must be made.

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