

Supersonic Phenomena

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INTRODUCTION

IN THE past ten years considerable interest has developed among physicists and chemists in several aspects of the phenomena accompanying the production and propagation of ultra-audible sound waves. The purpose of this article is to give, insofar as they are at present understood, the explanations for these phenomena. An attempt has been made throughout to make the exposition as simple as is compatible with clarity, and to refer to experiment at every possible point. It is hoped that this may provide a convenient starting point for the understanding of experimental investigations into the subject. Copious references to a fairly complete bibliography have been given, and these may perhaps be useful to those who are already familiar with what has here been written.

The review consists of three parts. In the first of these, expressions for the propagation of plane waves of sound as a function of amplitude, pressure, density, temperature and frequency are derived. The second part gives a brief review of the various methods of measuring the velocity and absorption of sound, and the interpretation of the measurements which have been reported

by different investigators. The third section reviews the effects of very intense sound waves.

Perhaps it should be stated at the outset that the terms *supersonic*, *ultrasonic*, and the ungraceful *ultra-acoustic* are here considered synonymous, and refer to sound of frequency too great to be audible. An attempt was made by some of the workers at the Loomis Laboratory to give two of them separate meanings, but since this has not become general practice there is little to be gained by laboring the point further.

THE THEORY OF THE PROPAGATION OF PLANE SOUND WAVES

In developing expressions for the propagation of sound in media of various properties it has been considered expedient to return to first principles. This renders unnecessary references to other publications, and makes explicit any approximations later to be used as the foundation for general conclusions.

We shall consider first the velocity of sound in an isotropic homogeneous medium and retain the first order of small quantities throughout. (A numerical example will then be given to show that this is sufficiently accurate to describe

contemporary experiment.) The passage of a plane sound wave through this fluid in the x direction will cause it to undergo periodic local displacements resulting in periodic variations from its equilibrium pressure P and density ρ . In particular, a slice of the medium of unit cross section and finite thickness τ will have its boundaries displaced by the amounts ξ and $\xi + (\partial\xi/\partial x)\tau$, respectively. The total mass in the undisplaced slice is $\rho_0\tau$, which is equal to the mass in the displaced slice $\rho\tau[1 + \partial\xi/\partial x]$, and hence, the density at A' and B' (see Fig. 1) is given by

$$\rho_{A'} = \frac{\rho_0}{1 + \partial\xi/\partial x}$$

and
$$\rho_{B'} = \frac{\rho_0}{1 + \partial\xi/\partial x + (\partial^2\xi/\partial x^2)\tau} \quad (1)$$

It will now be convenient to assume, subject to later proof, that the sound wave is transmitted adiabatically, hence

$$P/\rho^\gamma = P_0/\rho_0^\gamma. \quad (2)$$

If this is true the pressures on the two faces of the displaced slice will be

$$P_{A'} = (P_0/\rho_0^\gamma)[\rho_0(1 + \partial\xi/\partial x)^{-1}]^\gamma$$

and
$$P_{B'} = (P_0/\rho_0^\gamma)\left[\rho_0\left(1 + \frac{\partial\xi}{\partial x} + \frac{\partial^2\xi}{\partial x^2}\tau\right)^{-1}\right]^\gamma.$$

In expanding $P_{B'}$ it is necessary to retain only

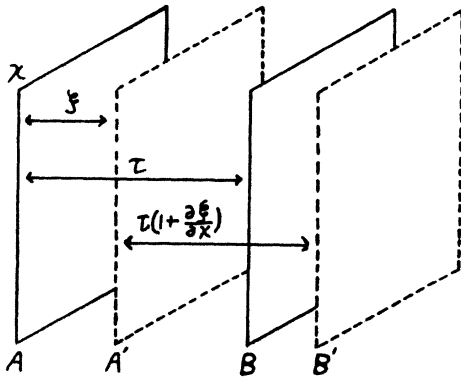


FIG. 1. Distances of the boundaries of an undisplaced slice of fluid (solid lines), and the same slice after a time t displaced by a plane sound wave moving in the x direction (dotted lines). In the text the slice is taken of unit cross section.

terms in the first power of τ , since this quantity may be taken arbitrarily small. This gives

$$P_{B'} = (P_0/\rho_0^\gamma)\left(\frac{\rho_0}{1 + \partial\xi/\partial x}\right)^\gamma \times \left(1 - \frac{\partial^2\xi/\partial x^2}{1 + \partial\xi/\partial x} \cdot \gamma\tau\right). \quad (3)$$

The difference in pressure $P_{A'} - P_{B'}$ multiplied by unit cross section is the displacing force, and is equal to the invariant mass $\rho_0\tau$ times the acceleration $\partial^2\xi/\partial t^2$. This is, to the first order of small quantities,

$$\tau\gamma P_0\partial^2\xi/\partial x^2 = \tau\rho_0\frac{\partial^2\xi}{\partial t^2}(1 + \partial\xi/\partial x)^{\gamma+1},$$

or
$$\frac{\partial^2\xi}{\partial t^2} / \frac{\partial^2\xi}{\partial x^2} = \frac{P_0\gamma}{\rho_0}\left(1 + \frac{\partial\xi}{\partial x}\right)^{-(\gamma+1)} \quad (4)$$

and gives rigorously the equation of motion. It is perhaps worth mention in passing that several more compact derivations in current texts are not correct (cf. Lamb,¹⁹ page 177). For very small amplitudes τ may be neglected, and the solution

$$\xi = Ae^{i\omega(t-x/V)}, \quad (5)$$

where A is the amplitude of ξ and ω is the frequency of a wave traveling in the x direction with a phase velocity V gives

$$V_0^2 = P_0\gamma/\rho_0 = (\partial P/\partial\rho)_{\text{adiab}}. \quad (6)$$

The coefficient $(\partial P/\partial\rho)_{\text{adiab}}$ is, of course, evaluated for the case $\rho = \rho_0$.

When the amplitude is finite the left-hand side of (4) does not represent a phase velocity because in this approximation $\partial\xi/\partial x$ is a periodic function of t and x . It is negative at one time and positive at another. The meaning of (4) is that the denser portions of the wave, in which $\partial\xi/\partial x$ is negative from (1), are traveling more rapidly than the rarified portions. This is without effect on the phase velocity to the first approximation. Fay²³ has shown, in a much more detailed analysis, that although waves of finite amplitude may depart strikingly from the sinusoidal form, becoming "saw-toothed," the wave will not obliterate itself from this cause. Viscosity limits the steepness of the wave front which may be attained because of

the tendency of the crests of the wave to overtake the troughs. A rapid attenuation of waves of finite amplitude does, however, result; this attenuation may be more than twenty times as great as that resulting from the viscous damping of a sinusoidal disturbance. An alteration of frequency is also entailed.²⁹ Audible sound, for example, which comes from a considerable distance, may have lost much of its low frequency components partly by attenuation, and partly by the passing of energy into higher frequencies. Fay's analysis shows that the effect of this on the velocity of propagation is negligible unless the intensity is sufficiently great to alter the mean density of the fluid.

It will be convenient to examine at this point the amplitudes of the waves currently used in the measurement of the velocity of sound and justify by a numerical example the use of (6) for the interpretation of experiments made with small but finite amplitudes. The ratio of the velocities of sound at zero amplitude, V_0 , and at amplitude A , V_A , may be obtained by an expansion of (4)

$$V_0/V_A = 1 + \frac{\gamma(\gamma+1)}{4} (\partial\xi/\partial x)^2.$$

The second term of this expansion is omitted because it is obviously zero for a sinusoidal wave and appears also to be zero, from Fay's data, for a "saw-toothed" wave. Pilemeyer¹⁷⁵ gives 130 decibels as a reasonable maximum for the intensity, and supports this figure by observations made by Andrade on particle motion at resonance in a Kundt's tube. This corresponds to a value for $\partial\xi/\partial x$ of 0.004, and, for a fluid for which $\gamma=1.4$, affects the velocity of sound only about 0.001 percent. This figure is free from the troublesome objection that the amplitude at resonance cannot be obtained from the power output. It is perhaps worth mentioning, however, that 130 decibels corresponds to an acoustical output of 0.1 watt per cm². In view of the inefficiency of the conversion of electrical energy into sound because of reflection phenomena at the surface of an oscillator, and the fact that most ultrasonic generators are driven by only a few watts, this intensity appears generous for gases even at resonance. On the basis of this paragraph, (6) will be used as the fundamental

acoustical equation, and effects due to amplitude will be neglected when the velocity of sound is discussed.

The assumption that the sound is transmitted adiabatically, which was made in writing (2), may now be justified by an appeal to experiment. Actually Stokes¹⁹⁴ and Kirchhoff¹⁵⁴ have shown that viscous loss and loss due to heat conduction will be negligible except near the walls of the containing vessel. Since their arguments are lengthy and of little contemporary interest it has been considered preferable to refer to measurements, which are given in Table I. The agreement with the adiabatic hypothesis for both gases and liquids is excellent, and such disharmony as remains is probably due to the difficulty in obtaining accurate measurements of the isothermal compressibility.

The next step in generalizing (6) is the introduction of the temperature. This may be done by writing $P=f(\rho, T)$ and considering T as a function of S and ρ . We then have

$$(\partial P/\partial \rho)_S = (\partial P/\partial \rho)_T + (\partial P/\partial T)_\rho (\partial T/\partial \rho)_S,$$

where S is the entropy and T the temperature. Similarly if the internal energy $E=f(\rho, T)$, then

$$\left(\frac{\partial T}{\partial \rho}\right)_S = \frac{(\partial E/\partial \rho)_S - (\partial E/\partial \rho)_T}{(\partial E/\partial T)_\rho}.$$

Since $(\partial E/\partial \rho)_S = MP/\rho^2$ and $(\partial E/\partial T)_\rho \equiv C$, the heat capacity for one gram molecular weight M at constant volume,

$$\left(\frac{\partial P}{\partial \rho}\right)_S = \left(\frac{\partial P}{\partial \rho}\right)_T + \left(\frac{\partial P}{\partial T}\right)_\rho \cdot \frac{M}{\rho^2} \cdot \frac{P + (\partial E/\partial v)_T}{C}.$$

Finally, the so-called "thermodynamic equation of state" gives for the sum of external and internal pressures

$$P + (\partial E/\partial v)_T = T(\partial P/\partial T)_v,$$

which, on substitution in the previous equation, gives the velocity of sound in any homogeneous isotropic substance as a function of P , T , and ρ

$$V^2 = (\partial P/\partial \rho)_T + (\partial P/\partial T)_\rho^2 \cdot (MT/C\rho^2). \quad (7)$$

It is convenient next to consider absorption, and its effect on the velocity. This is usually effected by an analytical device borrowed from

optics, which treats V in (5) as a complex number. For example it may be written that $\xi = A e^{-\beta x} e^{i\omega(t-x/c)}$ or

$$V_c = c/(1 - i\beta c/\omega), \quad (8)$$

in which c has the dimensions of a velocity but is not quite equal to the phase velocity, and β is the absorption coefficient for the amplitude expressed as a reciprocal length. This is exactly equivalent to writing (4) with a damped velocity term, but has the advantage of being considerably more easy to treat analytically. Since β and c are not functions of x and t it follows as before that

$$V_c^2 = (\partial^2 \xi / \partial t^2) / (\partial^2 \xi / \partial x^2)$$

and the effect of the absorption on the velocity is seen to be

$$\text{real part } (V_c^2)^{\frac{1}{2}} = V = c \left(\frac{1 - \beta^2 c^2 / \omega^2}{(1 + \beta^2 c^2 / \omega^2)^2} \right)^{\frac{1}{2}}$$

The absorption coefficient could, of course, be obtained from the imaginary part of V_c^2 , but it happens to be more convenient to use the imaginary part of $1/V_c^2$. The reason for this will be appreciated if the reciprocal of (8) is squared. The result is $-2i\beta/\omega c = \text{imaginary part of } (1/V_c^2)$. It has been briefly mentioned above that absorption may occur because of viscosity, heat conduction, and finite amplitude of the waves; no further attention will be given to these causes. Absorption caused by failure of part of the heat capacity of a polyatomic gas to participate in the adiabatic acoustical cycle at high frequencies is of greater contemporary interest, and an ex-

pression for this, together with corresponding expressions for the velocity, will now be developed.

A glance at the coefficients on the right-hand side of (7) shows that $(\partial P / \partial \rho)_T$ is simply related to the isothermal compressibility, and that $(\partial P / \partial T)_\rho$ is the ratio of the thermal coefficient of expansion and the compressibility. Both these involve adjustments of translational energy, and of the attractive and repulsive forces between molecules, but neither is concerned with the conversion of translational energy into rotational energy or vibrational energy within the molecule. From what we know of the nature of the van der Waals forces we should expect them to follow the changes in the sound wave except at very high acoustic frequencies. For the treatment of gases it is sufficient, therefore, to consider that the heat capacity is the only coefficient which fails to follow the sound wave in the range of frequencies at present experimentally available.

If the change of temperature of the translational energy of a gas due to the passage of a sound wave of frequency ω be momentarily ΔT an amount of heat will be absorbed which may be called $C_\omega \Delta T$. Thus C_ω is the heat capacity which is effective at the frequency ω , and is made up of several parts. Specifically,

$$C_\omega \Delta T = (3/2) R \Delta T + E_A \sum_l \Delta n_l + \sum_m m \Delta n_m + \dots$$

Here the first term on the right refers to changes of translational energy, and the subsequent terms express energy changes in a series of rotational and vibrational states corresponding to the optical frequencies $\nu_A = E_A / h N_a$ etc.; h is Planck's constant and N_a is Avogadro's number. The symbols n_l , n_m , etc. refer therefore to the fractions of gram molecular weights in the l th, m th, etc. states, and are subject to the restrictions $\sum m_l = 1$, etc. Experiment shows, however, that the rotational states are excited by changes in translational energy at a rate sufficient to maintain their equilibrium distribution at all frequencies which have been studied. A simplified expression for $C_\omega \Delta T$ may therefore be written. The simplest case of all is that in which the difference between the heat capacity at very low frequencies C_0 , and that at very high frequencies C_∞ , is due only to a single $0 \rightleftharpoons 1$ transition; the

TABLE I. *Experimental verification of the adiabatic hypothesis.*

SUBSTANCE	T^*	γ^{**}	V_{obs}^\dagger	$V_e^{\dagger\dagger}$	$V_T^{\dagger\dagger}$
Argon	303.1	1.667	324.0	324.2	251.2
Nitrogen	273.1	1.400	337.3	336.7	284.5
Benzol	293.1	1.45	1324	1319	1095
Toluol	293.1	1.34	1328	1317	1138
Carbon tetrachloride	293.1	1.46	935	931	774

* T is in $^\circ$ Kelvin.

** γ has been obtained for the gases from theoretical considerations, and from the ratio of the isothermal to the adiabatic compressibility β_T / β_s (given in *International Critical Tables* 3, 39) for the liquids.

† V_{obs} is the measured velocity of sound in $m \text{ sec.}^{-1}$ and has been reported in reference 179, p. 121 for argon, by several observers in *International Critical Tables* for nitrogen, and in reference 220 for the three liquids.

†† $V_e = (\gamma / \rho \beta_T)^{\frac{1}{2}}$ and $V_T = (1 / \rho \beta_T)^{\frac{1}{2}}$ were calculated from data in the *International Critical Tables*.

higher states of the vibration are not excited appreciably. In this case $h\nu \gg kT$, $n_0 \gg n_1$, $n_2 \cong 0$, and

$$C_0 = C_\infty + E(\partial n_1 / \partial T).$$

Since at equilibrium $\bar{n}_1 = \bar{n}_0 e^{-x}$ where $x = h\nu/kT$, and since at all times $n_0 + n_1 = 1$, it follows that

$$E(\partial n_1 / \partial T) = Rx^2 e^{-x} / (1 + e^{-x})^2 = C_0 - C_\infty. \quad (9a)$$

But, since $\bar{n}_2 = \bar{n}_0 e^{-2x} = 0$, this result is not physically consequent as it contains terms in e^{-2x} . By expanding, and discarding these we obtain the correct first approximation

$$C_0 = C_\infty + Rx^2 e^{-x}. \quad (9b)$$

To find the dependence of $C_0 - C_\infty$ on frequency it is necessary to express the rates of the transitions $0 \rightarrow 1$ and $1 \rightarrow 0$. This is most simply done by writing a reaction equation such as

$$dn_1/dt = f_{01}n_0 - f_{10}n_1, \quad (10)$$

where f_{01} and f_{10} are the probabilities of the $0 \rightarrow 1$ and $1 \rightarrow 0$ transitions per molecule per second, due not necessarily only to collisions, but to any cause whatever. Thus f_{10} is the number of times per second that a molecule in the state 1 leaves the state 1 for the state 0. At equilibrium $dn_1/dt = 0$ and $f_{01} = e^{-x}f_{10}$. It is not necessary, if the reaction equation is stated in this way, to include a term to account for pressure fluctuations of the gas, since n_0 and n_1 are not functions of the pressure.

Since we wish to find the dependence on frequency of this part of the heat capacity, we must examine a case where equilibrium has not been attained, and dn_1/dt is therefore not equal to zero. This means that the variables n_1 , f_{10} , and T will be displaced from their equilibrium values by small finite amounts Δn_1 , Δf_{10} , and ΔT . For example $n_1 = \bar{n}_1 + \Delta n_1$, etc. The displaced equation, in which second-order terms are disregarded, is

$$d\Delta n_1/dt = f_{10}(\bar{n}_0 \Delta e^{-x} + e^{-x} \Delta n_0 - \Delta n_1). \quad (11)$$

The time derivative may be evaluated from the statement $\Delta n_1 = \text{const } e^{i\omega t}$ which expresses the harmonic character of the sound wave. Furthermore $\Delta e^{-x} = e^{-x} \Delta T/T$, since x is a function of temperature only. Incorporating these changes, and remembering that $\Delta n_0 = -\Delta n_1$, we obtain

$$\Delta n_1 = \frac{n_0 f_{10} x e^{-x} \Delta T}{T[i\omega + f_{10}(1 + e^{-x})]}.$$

The energy change is of course $E\Delta n_1$ where $E = h\nu N_a = RTx$. Hence

$$E\Delta n_1 = \frac{n_0 f_{10} R x^2 e^{-x} \Delta T}{i\omega + f_{10}(1 + e^{-x})}.$$

If we substitute from (9a), we obtain

$$E\Delta n_1 = \frac{(f_{10} + f_{01})(C_0 - C_\infty)}{i\omega + f_{10} + f_{01}}$$

which is the expression printed by Kneser¹⁵⁶ and several others. It has been pointed out by Rose¹⁸⁷ and others that this is not the consequent approximation, since it contains terms in e^{-2x} . In order to eliminate these f_{01} must be neglected, and the result is

$$C_\omega = C_\infty + \frac{C_0 - C_\infty}{1 + i\omega\theta}. \quad (12)$$

Here the notation $\theta = 1/f_{10}$ has been introduced since it will serve as a useful abbreviation later; θ is often referred to as the "relaxation time" or better the "mean lifetime" of the state 1. When (12) is substituted into (7) we obtain

$$V_c^2 = \left(\frac{\partial P}{\partial \rho} \right)_T + \left(\frac{\partial P}{\partial T} \right)^2 \frac{MT(1 + i\omega\theta)}{C_0 + C_\infty i\omega\theta}. \quad (13)$$

Equation (13) is the fundamental statement on which is based the interpretation of acoustical dispersion in polyatomic gases. The phase velocity is obtained from the real part of the square root of (13), together with some equation of state. Far from the critical point the equation of state of an ideal gas

$$MP/\rho = RT$$

may be used, and the velocity is in this case

$$V = \left\{ \frac{RT}{M} \left(1 + \frac{R(C_0 + C_\infty \omega^2 \theta^2)}{C_0^2 + C_\infty^2 \omega^2 \theta^2} \right) \right\}^{\frac{1}{2}},$$

whereas the absorption coefficient for the intensity, μ , expressed as a reciprocal number of wave-lengths is (cf. reference 179, page 739).

$$\mu = 2\beta\lambda = \frac{2\pi C_\infty \omega}{C_0 f_{10}} \left\{ \frac{V_\infty^2/V_0^2 - 1}{(V_\infty^2/V_0^2)(C_\infty \omega/C_0 f_{10})^2 + 1} \right\}.$$

We must now pause briefly to comment on some aspects of the reasoning process by which (13) has been obtained, and to trace its history. Since Bourgin¹³⁴ has recently subjected this expression to a thorough critical examination, only the more superficial implications and inaccuracies of the derivation will be considered.

It should first be noted that (13) has been obtained without any assumption concerning the mechanism by which the $0 \leftrightarrow 1$ transition is brought about. It is not, therefore, a collision theory inherently, although most writers obtain an identical expression by means of explicit assumptions concerning the transition probability and the pressure. Again, the statement $\theta f_{10} = 1$ is true only if the number of molecules in the state 2 is less than the number which experiment can detect. With many gases, such as carbon dioxide, this is not the case, and the significance of θ must be revised in a way which will shortly be indicated. Further, the notation

$$C_\infty = 3R/2 + E_A \sum_i l \Delta n_i + \text{etc.}$$

is, as Bourgin remarks, convenient but physically fictional, since even states of very short lifetime lag slightly at any frequency. A numerical computation shows, however, that the dispersion due to any group of states with a common lifetime may be detected only over about two orders of magnitude of frequency if the accuracy of the velocity measurement is 0.1 percent, as in the best of contemporary work. The influence on absorption measurements is about the same. Since experiment shows that the lifetimes of the rotational states of gases are far shorter than those of most, if not all, vibrational states, the inclusion of the rotational heat capacity in C_∞ is in most cases sound from the standpoint of measurement.

One further consideration underlying (13), which in no way affects the accuracy of the expression, but is often discussed by writers on dispersion, should be mentioned for the sake of completeness. This is the phase lag between the pressure and the density in a dispersive region. This comes about because in the dispersive region

the lifetime of the energy-absorbing process is of the same order of magnitude as the reciprocal of the frequency, whereas the lifetime of a collision must necessarily be much less than the reciprocal of the frequency for the wave to be transmitted at all. Thus adjustments in internal energy lag behind adjustments in translational energy, the pressure impulse leads the density, and part of the energy of the wave is absorbed. This is much simpler to state analytically than it is to say in words. We write for the pressure impulse

$$\Delta P = A' \exp [i\varphi + i\omega(t - x/V_c)]$$

and allow it to lead the density impulse

$$\Delta \rho = A'' \exp [i\omega(t - x/V_c)]$$

by an angle φ . Then we equate real and imaginary parts in the ratio $\Delta P/\Delta \rho$ and obtain

$$\frac{\omega^2}{c^2} - \beta^2 = \frac{A''}{A'} \omega^2 \cos \varphi; \quad \frac{2\beta\omega}{c} = \frac{A''}{A'} \omega^2 \sin \varphi,$$

which lead to a convenient expression for the absorption coefficient

$$\mu = 2\beta\lambda = 2\pi \tan \varphi.$$

The type of expression which (13) represents was apparently first foreseen by Jeans.¹⁵³ It was put in articulate form by Einstein in 1920¹⁵⁸ in connection with the rate of dissociation of polymolecular vapors. In the case of a dissociating gas, of course, the heat capacity at very low frequencies contains the heat absorbed by the dissociation reaction. If the heat capacity is independent of frequency, the only cause for dispersion will be the lag of the dissociation reaction, and this permits the treatment of an ideal dissociating gas to be carried through very simply. Since the resulting expressions have been widely discussed in this country^{169, 179} it does not appear necessary to reprint them. Herzfeld and Rice¹⁴⁸ gave an explanation for anomalous velocities and absorptions in nondissociating gases which was similar in intent to that of Jeans. These treatments are essentially continuous fluid theories. Immediately following this Bourgin¹³⁴ wrote a very complete theory for the propagation of sound in gases using the statistical discipline; he considered viscosity, quantization of the collision process, radiation from excited mole-

cules, and dispersive gaseous mixtures. In 1931 Kneser renewed interest in the subject by reporting a series of measurements on carbon dioxide, and writing a theory for them based on Einstein's treatment. Kneser's exposition¹⁵⁶ was made more rigorous by Rutgers,¹⁸⁹ who followed Einstein even more closely. Meanwhile Bourgin wrote the theory of the three-state gas (a gas with the translational energy and two sets of internal energy states with separate characteristic lifetimes). Other writers have given descriptions of absorption in nondissociating gases,^{144, 147, 158} absorption in a real dissociating¹⁶⁹ gas, and the velocity in an ideal dissociating gas in which the heat capacity and the dissociation reaction have separate characteristic lifetimes.¹⁷⁹ Treatments of real nondissociating gases have been given in which van der Waals' equation,¹⁸¹ Sutherland's equation,¹⁴⁰ and Onnes's second virial coefficient¹⁵² are used, and several versions of the three-state gas have appeared.^{134, 166, 180, 187} All of these papers are based either on the thermodynamic theory developed by Einstein or the quantized kinetic theory given by Bourgin. In his last paper Bourgin has shown that the results of the two are equivalent, but that the statistical theory is the more comprehensive and logical. I agree with this, and regret that, in spite of the expenditure of considerable effort, I was unable to develop a presentation of his work sufficiently simple and compact to be included in this review.

The overwhelming majority of writers prefer to consider that the $0 \rightarrow 1$, etc. transitions are brought solely by effective collisions. Thus the transition probabilities of the $f_{l, l+1}$ type are set equal to the product of a molecular velocity, a target area involving the cross section of the molecules, and one or more efficiency factors. An example of such an expression is

$$f_{10} \sim N\sigma^2 \left(\frac{\pi RT}{M} \right)^{\frac{1}{2}} \zeta_{10} \exp \left[-3 \left(\frac{(\pi a v)^2 M}{2kT} \right)^{\frac{1}{2}} \right], \quad (14)$$

where N is the numerical density, σ the cross section, ζ_{10} a steric efficiency factor, and the exponential factor one suggested by Landau and Teller¹⁶⁶ involving the molecular weight M and the atomic radius a . This formulation of the transition probabilities is open to criticism on several grounds, which are at present best tested

by experiment, and will therefore be discussed in the experimental section.

Landau and Teller have recently generalized (13) to show that all of the energy states associated with a certain mode of vibration will have the same lifetimes, and that consequently they will all fail to follow the acoustical cycle in the same frequency range. Since this argument is based on a detailed consideration of the collision process, and treats only double collisions, a new notation indicating this will be introduced. In a gas of numerical density N there will be l quantum states, and the number of molecules per cc in each state is N_l . In this case, of course,

$$\sum_l N_l = N. \quad (15a)$$

The number of vibrational quanta per cc is designated by Z , and is

$$Z = \sum_l l N_l. \quad (15b)$$

Instead of writing transition probabilities we shall now employ reaction rate constants of the type $k_{l, l+1}$. If the transitions between states are due solely to double collisions

$$f_{l, l+1} = k_{l, l+1} N K, \quad (16)$$

where K is the number of collisions a molecule suffers in unit time at a numerical density of one molecule per cc. Then the general reaction equation becomes

$$dZ/dt = KN \left\{ \sum_l k_{l, l+1} N_l - \sum_l k_{l, l-1} N_l \right\}, \quad (17)$$

since obviously $k_{l, l-1} N_l = k_{l+1, l} N_{l+1}$, provided the oscillator is harmonic. Eq. (17) is practically Bourgin's kinetic equation in a somewhat less general form. Landau and Teller now examine the relation between the various k terms with the aid of four assumptions, all of which are extremely reasonable provided l is not very large. The first of these, that the molecular vibration should be harmonic, has already been invoked. The second is that only one mode of vibration shall be concerned, and that this shall not be degenerate; this is merely to simplify the exposition, for it will soon be seen that the theory is readily generalized. The third supposes that the excitation by collisions may be described by a

first-order perturbation calculation; since in cases where dispersion is observed the probability of excitation is small, this cannot be far from the truth. The fourth supposes that the interchange energy which causes the excitation of the vibration shall be a linear function of the normal coordinates of the vibration; this is essentially the case if the amplitude of the vibration is small compared to the separation of the atoms. According to these assumptions the matrix elements of the perturbation energy are proportional to the matrix elements of the coordinates of an harmonic oscillator, and all elements vanish which have a difference of more than one quantum for the beginning and end states. Thus $k_{l+m} = 0$ if $l-m \neq 1$, and

$$k_{01} : k_{12} : k_{23} : \dots \\ = k_{10} : k_{21} : k_{32} \dots = 1 : 2 : 3 : \dots$$

By use of this simplification, the general reaction Eq. (17) becomes

$$\begin{aligned} dZ/dt &= KN \left\{ k_{01} \sum_l (l+1) N_l - k_{10} \sum_l l N_l \right\} \\ &= KN (k_{01} N - (k_{10} - k_{01}) Z). \end{aligned} \quad (18)$$

By taking partial derivatives as before, neglecting the temperature dependence of Kk_{10} , and discarding terms of the second order it is found that, for substitution in (13),

$$\theta = 1/[KN(k_{10} - k_{01})], \quad (19)$$

which of course reduces to the same solution as the simpler case when $h\nu \gg kT$, but differs importantly from it when $N_0 \cong N_1$.

If more than one mode of vibration must be considered either all the vibrations will leave the wave at a common frequency, as appears very often to be the case judging from experiment, or the rate of excitation of each mode of vibration will be independent of the others. The treatment of the former case is identical with the treatment of a single vibration, and in the latter case separate equations of the type of (19) may be written for each vibration. Thus the heat capacity at any frequency will be

$$C_\omega = C_\infty + C_A \left(\frac{1}{1+i\omega\theta_A} \right) + C_B \left(\frac{1}{1+i\omega\theta_B} \right) + \dots, \quad (20)$$

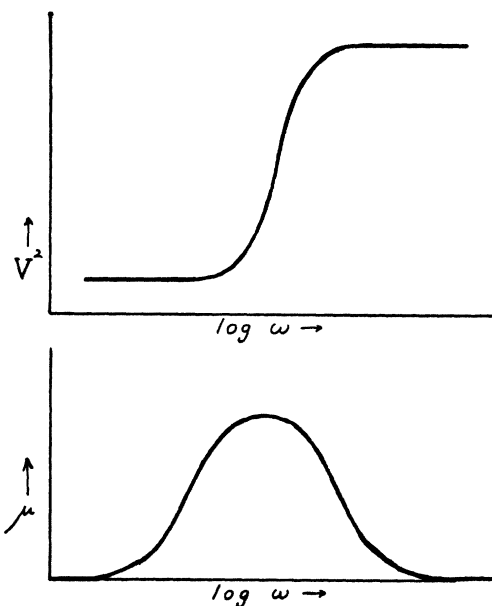


FIG. 2. The velocity and absorption in an ideal gas in the dispersive region according to Eq. (13). The point of inflection of the velocity curve is at $\omega = C_0/\theta C_\infty$, and the maximum of the absorption at $\omega = V_0 C_0/\theta V_\infty C_\infty$. If double collisions are solely responsible for the transitions between states, $\log \omega/N$ may be used as abscissae.

where A, B , etc. designate the various vibrations. C_A is then the equilibrium heat capacity at constant volume of one vibration, θ_A its lifetime, and so on. This value of C_ω is substituted in (13) and the real and imaginary parts of the result taken, as before, to represent the velocity and the absorption. If the band-spectral analysis of the molecule has been made C_A may be calculated from Planck-Einstein oscillators and the van der Waals constants; if, conversely, the vibrational frequencies are unknown they may be estimated from C_A , etc. which are obtained by velocity or absorption measurements.

From what has been said above it should be clear that a believer in the collision theory can find expressions sufficient to describe any measurements he may obtain, provided his experimental technique is equal to the occasion. The view expressed by Richardson and his co-workers which they have called the "resonance theory" appears to be the only rival to the collision theory. It is based on a series of measurements of the velocity of sound which do not show the relationship between velocity and frequency (or pressure) indicated in Fig. 2, but show rather an

initial drop in velocity as the frequency is increased, followed by a sharp rise to a maximum, and by a subsequent fall. Since several other investigations have been made under comparable conditions without confirming these irregularities, and since it is difficult to conceive of anything which would resonate in a polyatomic molecule at 10 kc, this theory will not be discussed in this review.

Turning to liquids, it should be remarked that the appearance of dispersion because of a lag of heat capacity states of long lifetimes is not to be expected in the acoustical frequency range at present available to experimenters. There are at least two reasons for this statement. In the first place the rate of attainment of translational equilibrium in liquids is considerably greater than in gases because of the greater numerical density. This amounts to an increase in the number of "collisions," if such a term may be used, and correspondingly lowers the lifetime of the internal energy states. Secondly, the heat capacities of most liquids are large, and the absence of a small amount of vibrational energy at high frequencies would be difficult to detect. It may seem somewhat surprising that γ in most liquids except water is much greater than unity (indeed many writers appear to share the opinion of Lamb¹⁹ page 166, that "in liquids the difference between the isothermal and the adiabatic elasticities may be neglected"). This is due, of course, to the work which must be done against the cohesion of the liquid to produce thermal expansion. This work is $(\partial E/\partial v)_T(\partial v/\partial T)_P$ per degree centigrade, and in liquids is usually of the same order of magnitude as C_v , whereas for an ideal gas it is zero.

It is not dangerous to predict, however, that the investigation of dispersion in liquids will eventually become a fertile field for measurement when sufficiently high frequencies have been attained, and the theory of the liquid state is in more satisfactory condition. Besides dispersion caused by heat capacity lag, dispersion caused by the lag of a dissociation reaction with a high heat of dissociation could be detected. Indeed it appears that such a case has been reported during the preparation of this review.²⁴⁵ The theory for this may easily be written with (13) as a start. Again, the phenomena in very viscous liquids

should provide an interesting study. In this case the density, compressibility, and thermal expansion are all functions of time, and a radically different attack is necessary.²²¹

There is little to be said concerning solids. Two papers have appeared dealing with the theory of the absorption of sound at high temperatures in solids,^{231, 232} but most of the attention devoted to solids has been confined to transverse vibrations, with which we are not here concerned. In some crystals, when the free rotation of certain groups sets in below the melting temperature, measurements of the dispersion of sound should provide information concerning the rate at which energy is supplied to these groups by the vibrating molecules of the lattice.

EXPERIMENTAL METHODS AND RESULTS FOR THE VELOCITY AND ABSORPTION OF SOUND

In this section an attempt will be made to serve a double purpose. The text will be made complete enough so that the casual reader need not turn to the end of the paper to consult the large number of references which must perforce be included, and the references will be given in a way which it is hoped will facilitate the bibliographical labor of a closer student of the subject.

The most popular instrument for the measurement of the velocity and absorption of sound is the sonic interferometer originally devised by Pierce.¹¹⁵ This consists of an oscillator driven by a valve tube circuit and a movable reflector attached to a micrometer screw. As the reflector is approached to or withdrawn from the surface of the oscillator the plate current of the driving circuit undergoes periodic changes which are easily measurable with a microammeter. If the oscillator were giving off plane waves, the distance between two points of maximum reaction would be one-half the wave-length; very often this is practically the case. If the change of plate current were a linear function of the intensity of sound at a point of maximum reaction, the absorption coefficient could be obtained directly from the decrement of the electrical reaction; this is seldom if ever the case. Thus the interpretation of interferometric data is simpler for the velocity^{96, 102, 105, 118} than for the absorption.^{79, 82, 95, 117} For the determination of

the fundamental constants of dispersive fluids there is, however, little to choose, since the information is obtained from the velocity as the difference between two large numbers, whereas the absorption is concerned with the position and magnitude of the maximum of a function which virtually vanishes on either side (cf. Fig. 2). A large number of modifications of the original interferometer have been described. Double⁸⁷ and even triple¹²⁹ oscillators have been employed in sending-receiving systems. Instruments suitable for wide temperature^{61, 116} and pressure¹²⁵ ranges have been devised. In addition several papers dealing with the theory of interferometry have appeared.^{62, 82, 90, 95}

The sources of sound in interferometers are usually either piezoelectric or magnetostriction oscillators, which change periodically in dimensions with the frequency of an applied electromagnetic field. Single quartz crystals, cut in a variety of ways, are usually employed as piezoelectric sources because of their mechanical strength and lack of chemical reactivity; they are suitable for frequencies between 50 and 50,000 kc. The general characteristics of the piezoelectric effect^{80, 103, 104} have been investigated, the modes of vibration of quartz plates studied,⁷⁸ and the characteristics of the wave field they generate described.^{100, 119} In addition their acoustical energy yield,⁶⁵ their suitability as frequency standards,^{75, 115} and the alteration of their frequencies with changing environment have been studied.^{81, 112} Magnetostriction oscillators have been successfully employed between 1 and 500 kc,^{114, 179} and probably would be practical to 1000 kc. A general study of the magnetostrictive effect has been made,⁷² a revised piece of apparatus for the study of the velocity recently described,¹⁰⁹ and some of the effects of the waves reported.¹¹⁰

Three difficulties have caused confusion in the interpretation of interferometric measurements. In the first place, the wave given off by the oscillator is usually by no means plane.^{82, 119} The face of the oscillator usually does not vibrate back and forth as a whole like a piston, but various portions of the surface vibrate in different ways.⁷⁸ Thus the point of maximum reaction is a compromise between the points of maximum reaction of the various components of the wave

and, particularly near the surface of the oscillator, irregular spacing which may differ from the half-wave-length by several percent is usually found. This has been discussed by Grossmann⁸² and by Grabau¹⁴⁸ from different points of view; Eucken and Becker have devised a useful procedure for compensating for these irregularities.¹⁴⁰ In addition, when measurements are made in a confined space reflections of the various components of the wave from the walls may occur and cause very complicated patterns indeed. The second difficulty arises from larger variations in the frequency of the oscillator due to changes of temperature and pressure than are to be expected from dimensional changes. Thus, to cite an extreme example from my own experience, the change from 250 to 500 mm pressure of acetic acid vapor caused a change in frequency of 3 percent in a massive nickel tube, and this change could be reproduced. Cases are on record where relatively small changes of pressure have caused crystals to stop oscillating.¹⁹⁶ The explanation which suggests itself for this is that chemical adsorption takes place on the crystal and its mount, adding its effect to any direct damping of the crystal which the change of pressure may entail. The third difficulty in the interpretation of interferometric measurements occurs in evaluating the absorption coefficient from the decrement of the plate current as the reflector is withdrawn from the oscillator. This situation has been subjected to detailed analysis, especially by Hubbard,⁹⁵ and is now sufficiently understood to yield reliable absorption coefficients. Much of the early work on the subject, however, treats the electrical and acoustical decrements as if they were identical, and therefore requires revision.*

Two other considerations enter when the velocity or the absorption is measured in small tubes. The Helmholtz-Kirchhoff tube correction^{88, 89, 101} has been tested and found good over a wide range of conditions, and may be employed with plane waves with some confidence.^{77, 98, 108} When the length of the tube is great compared to its diameter, however, care must be taken to

* *Note added in proof.* A series of studies by S. K. Kulkarni Jatkar has recently appeared in *J. Indian Inst. Sci.* **21A**, 245, 455, 477 (1938) and **22A**, 1 (1939). These deal comprehensively with the aberrations of sonic interferometers, and should be helpful to anyone who intends to undertake interferometric measurements.

avoid the phenomenon of radial resonance which was discovered by Boyle and Field,²¹⁵ has been treated theoretically by Field,²¹⁸ and has been the subject of considerable experiment.^{97, 219} This occurs because the walls of the tube are not infinitely rigid, and is far greater in effect with liquids than with gases.

The following procedure is recommended for measuring the velocity in dispersive gases. The apparatus should permit complete evacuation and, if possible, degassing in order to eliminate traces of adsorbed impurities. Moisture is especially troublesome if it is not thoroughly pumped off; not only does it make the composition of the gas an undetermined function of time and temperature, but, because of the phenomenon of "trace catalysis" which will be discussed below, it may, even if present in minute quantity, shift the dispersive region to higher frequencies. The apparatus should be calibrated by measurements on some standard gas or mixture of gases which are known not to be dispersive over the entire range of pressure, temperature, and frequency to be employed. The calibration will give information concerning the reproducibility of the wave pattern, any irregularities which it may contain, and the comparability of the various measurements. The frequency and the length are easily measurable to 0.1 percent or better, but experience shows that the accuracy of the velocity measurements may be far inferior to this. If irregularities in the wave pattern appear which are reproducible and not greater than 1 percent it is reasonably safe to treat them as apparatus corrections, and compensate for them in the same way that a tube correction is applied. This is permissible, however, only if the velocity in the dispersive gas is much the same as that in the gas used for calibration (i.e., if the acoustical geometry remains the same). The frequency of the oscillator should be measured at the beginning and end of each determination, and the result discarded if any variation is found. Finally, the degree of purity of the gas studied should be accurately known and frequently verified. When these precautions are taken it makes little difference if the values resulting from measurement are as much as 1 percent in error, for (7) and (13) combine to give, for an ideal gas,

$$\theta = \frac{C_0}{C_\infty \omega} \left\{ \frac{V_\omega^2 - V_0^2}{V_\infty^2 - V_\omega^2} \right\}^{\frac{1}{2}}. \quad (21)$$

Systematic errors in the measured quantities V_0 , V_ω , and V_∞ , almost eliminate each other, and thus leave the determination of the fundamental quantities θ and C_∞ unaffected.

A modification of the interferometric technique has been devised by Richardson, who explores the standing-wave field with a hot wire.¹⁷⁷ This appears to be a promising approach to the determination of the absorption coefficients of gases.¹²⁷ A thermopile may be used in much the same way.^{84, 85}

The reverberation technique developed by Knudsen¹⁶² has proved very effective for the determination of the absorption coefficients of gases. This method utilizes a source of sound, such as a magnetostriction oscillator, a large gas-tight chamber, a crystal microphone or some other device for measuring the intensity of sound, and suitable amplification and recording devices. A burst of sound is introduced into the chamber, and its decay measured from the decrease in amplitude with time of the microphone. This apparatus has proved especially valuable in the audible range, where the physical dimensions of an interferometer become prohibitively large, but it may also be used at higher frequencies.¹⁶³

The most important rival of the interferometric method of determining the velocity is the recently perfected optical technique. Before this is discussed, however, it will be well to mention a few miscellaneous matters pertinent to the subject. For example, both pneumatic jets⁸⁶ and condenser microphones¹²⁰ have been suggested as sources of sound, although neither appears to have been used in velocity determinations. Velocity determinations have been made in gases by the distribution of coagulated smoke,^{111, 113} and by the introduction of a film of liquid on the wall of the vessel which, with suitable illumination, gives bright lines at the antinodes.⁷⁴ In liquids, velocity determinations have been made from the distribution of charcoal powder in a standing-wave system,^{123, 262} and from the interference of capillary waves on the surface of the liquid.⁷⁶ Modifications of the Rayleigh disk have been devised to measure the absorption in

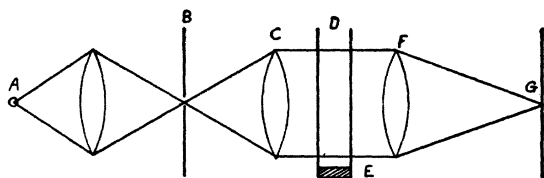


FIG. 3. Apparatus for demonstrating the diffraction of light by ultrasonic waves according to Debye-Sears-Lucas-Biquard. *A* is a lamp and lens, *B* a slit normal to the paper, *C* a lens to make the light parallel, *D* a trough containing liquid, *E* a piezoelectric oscillator, and *F* another lens which focuses the light on a screen at *G*, where the diffraction lines appear.

gases,^{131, 171} and in liquids,^{73, 205} and it has been pointed out that the interpretation of the results with this instrument requires correction.⁹⁹ Finally, two modifications of methods for determining the absorption coefficient which do not fall readily into any of the classifications here adopted have been described.^{70, 71}

The optical methods which have recently been developed for measuring velocities and absorptions, particularly in liquids, are outgrowths of a discovery announced independently and almost simultaneously by Debye and Sears⁴² and by Lucas and Biquard.⁵⁰ These writers, realizing that the periodic condensation and rarification in a sound wave progressing through a liquid would act as an optical diffraction grating, suggested the illumination of the liquid normal to the direction of propagation of the sound wave with parallel light from a slit source, as illustrated in Fig. 3. The emergent light, when focused on a screen by another lens, consists of one or more sets of diffraction lines evenly spaced about the primary image. The spacing of the lines is related to the grating constant as in the case of an optical transmission grating, and hence is a simple function of the wave-length of sound. The elementary theory of the effect was written by Debye,⁴² and has been greatly extended by Brouillin,³⁹ Lucas,⁴⁹ and by Raman and Nath^{52, 53, 55, 56, 58} as well as by a host of others.^{35, 37, 40, 41, 44, 45, 47, 57, 60} Such matters as the change in frequency of the diffracted light,^{30, 43, 48, 230} the shifting in intensity of various orders of diffraction by changing acoustical intensity,^{38, 46} and several others have been investigated both theoretically and experimentally. Since it would almost double the length of this review to include a description of these

developments, the reader is requested to consult a special section of the bibliography where they are listed.

The Debye-Sears-Lucas-Biquard method has stimulated a number of experimenters to produce variations,^{37, 107, 200, 206} some of which have led to important and even dramatic results. For example, Töpler's "*Schlieren-Methode*," which is obviously closely related, permits the direct visualization and photographing of the waves.^{34, 126, 229} Again, in place of the second lens and screen in Fig. 4, Hiedemann and Hoesch use simply a microscope and comparator, and thus observe standing waves as a series of bright lines and dark bands, whose separation may be observed with great accuracy.^{92, 227, 229} As Hiedemann points out, the standing-wave system is comparable to a series of glass rods mounted parallel to each other and separated by their diameters; thus the breadth of the lines is finite, practically all of the light falling within 1/15 of the spacing of the lines. Hiedemann and others have also done away with the necessity for standing waves by the use of stroboscopic light sources, such as a Kerr cell or another acoustic grating, which illuminates the system with the same frequency as the sound wave itself.^{64, 229} This permits direct observation of any irregularities in the wave field, and facilitates the observation of the diffraction, reflection, refraction, and interference of sound waves.¹²² It also yields values for the velocity of sound in liquids which are accurate to 0.02 percent if suitable precautions to avoid heating the liquid by power loss near the crystal are taken.³³ It is possible also to distinguish the transverse from the longitudinal vibrations in transparent solids, and to secure accurate measurements of their elastic constants.^{63, 91, 227} It has been known for some time that the distribution of energy in the sound field produced by a vibrating quartz plate is very uneven; there are "rays" of great intensity, and regions where almost no sound is perceptible. The optical methods make these visible.^{93, 225}

Turning now from methods to results, it will be convenient first to consider the so-called "zero-frequency" velocities and absorptions. It should be mentioned in passing that the zero-frequency properties may, as in the noble gases, persist to very high frequencies indeed, or, as in

oxygen and carbon dioxide, be replaced by other properties well within the audible range.

Gases and vapors give confirmation of the band-spectral analysis in the low frequency range if the expression

$$C = C' + \int_{\infty}^v T(\partial^2 P / \partial T^2) dv,$$

which corrects for the van der Waals forces, is used in computing the velocity. Here C' is, of course, the heat capacity at infinite dilution which is provided by the spectroscopic data. It is often more profitable to find C' by extrapolation, and compute the second virial coefficient from the velocity of sound.^{150, 165} The accuracy of acoustical measurements is sufficient to compete successfully with other methods for the determination of the heat capacity, but one serious difficulty arises. With some gases a part of the heat capacity disappears at very low frequencies, and the observer can never be sure that the value which he obtains at ultrasonic frequencies does not lack such a term. Acoustical measurements are also about as accurate as any for the determination of P , v , T data. The measurement of the low frequency absorption coefficient of gases has yielded little of interest.

Liquids present quite a different picture. Since the heat capacity of most polyatomic liquids can not be evaluated theoretically, the velocity of sound yields, in conjunction with the density and the isothermal compressibility, an empirical constant γ which awaits interpretation on a molecular-kinetic basis. Measurements on a large number of organic liquids and liquid mixtures have been reported;^{220, 236, 239, 243, 246, 249, 251} of special interest are the results of Parthasarathy, which he has correlated with the chemical constitution of the liquid.²³⁹ It is not necessary to use large volumes of liquid; Bär, indeed, was able to determine the velocity in heavy water by using only 1 cc. Measurements on liquid oxygen^{116, 233} and hydrogen have been reported. Several investigations dealing with the velocity^{201, 217, 237, 247} and absorption²³⁸ in electrolyte solutions have shown that the concentration-compressibility function depends on the valence of the dissolved ions. Theoretical^{223, 224} and experimental^{241, 242} investigations into the properties of colloidal systems show that, to a first

approximation, the velocity is independent of the presence of a disperse phase, and that to a second approximation the velocity is independent of the degree of dispersion of the disperse phase.

The question of the absorption and dispersion of sound in liquids offers much greater interest. Several observers starting apparently with Biquard,²¹² have found that the absorption coefficients of many polyatomic liquids are more than a hundred-fold as great as is to be expected from considerations of viscosity and the like;^{205, 216, 238} Bär confirms this, and adds the welcome proof that the effect is not due to excessive amplitude or, indeed, to any apparatus effect, by finding a normal coefficient for mercury.²⁰⁴ Lucas has made an extensive theoretical investigation into possible causes for this phenomenon, and has shown that fluctuations in density are not enough to account for it. He is inclined to the belief that, as is so often assumed in modern physics and physical chemistry, the liquids made up of polyatomic molecules have a sub-microscopic structure.²³⁴ If this were true it would account for the dispersion recently found by Hiedemann and his associates²²⁸ in several liquids. Other observers have failed to detect dispersion,^{214, 240, 248} or have reported a decrease in velocity with increasing frequency;^{244, 245} this is doubtless due to the inferiority of their methods of determining the velocity compared to that of Hiedemann. It seems to me that the case for cybotactic groups is strong but not conclusive; doubtless the same investigators who have carried it so far will decide it within the next two or three years. The anomalous temperature coefficients of the absorption in many liquids^{207, 244} should contribute valuable evidence when the case is complete. It is not necessary to go so far afield to account for the dispersion²⁴⁵ and absorption²⁰⁸ which have been recently reported in acetic acid; a reaction with a heat of dissociation of some 14 Calories per mole is involved, and dispersion may occur from exactly the cause foreseen by Einstein.¹³⁸

Several methods for the determination of the velocity in solid plates,^{69, 102, 202, 209, 213, 320} rods,¹²⁴ and sheet materials²⁵⁰ have been described. Although many measurements have been reported, apparently the only important general-

ization which has emerged is that deduced by Boyle and his students for the reflection coefficient, R , between two contiguous media in which the velocity of sound is, respectively, V_1 and V_2 , and the thickness of the second is τ

$$R = \frac{(\rho_1 V_1 / \rho_2 V_2 - \rho_2 V_2 / \rho_1 V_1)^2}{4 \cot^2 g^2 (2\kappa\tau/\lambda_2) + (\rho_1 V_1 / \rho_2 V_2 + \rho_2 V_2 / \rho_1 V_1)^2}$$

This has been extensively confirmed by experiment,²¹³ and, although theoretically true for media of infinite extent only, has proved correct for relatively small plates.

The reviewer is faced with a disheartening task in discussing the measurements which have been reported concerning the dispersion of sound in gases. Although the evidence is overwhelmingly in favor of the collision theory, the details of the picture vary from laboratory to laboratory. This is in part due to difficulties in interpreting interferometric data, but in far greater part to the failure of many investigators to provide adequate means for securing information concerning the chemical composition of the gas they are studying. It is only by the assumption that unreported traces of impurities have displaced the dispersive region that one can reconcile the statements that the properties of nitrous oxide are identical with carbon dioxide^{157, 177} and that there is no resemblance in their behavior.¹⁹⁹ For this reason no attempt will here be made to fix the numerical value of the relaxation time for any gas. A list of gases will be given which have been studied, either alone or as the principal constituent of a mixture, with references to the various papers in which the measurements may be found. The words "dispersive" or "not dispersive" will be affixed if the evidence seems to point clearly in either direction. A general discussion, in which numerical values are sparingly used, will follow.

Acetic acid, inconclusive, 195.
 Aerosols, dispersive, 135, 164.
 Air, dispersive, 108, 114, 143, 151, 162, 171, 178, 185, 197.
 Ammonia, dispersive, 193.
 Carbon dioxide, dispersive, 90, 95, 114, 131, 132, 137, 140, 142, 144, 149, 150, 156, 157, 170, 174, 176, 177, 178, 181, 183, 186, 198, 199.
 Carbon disulfide, dispersive, 181.
 Carbon monoxide, dispersive, 152, 191.
 Chlorine, dispersive, 140.
 Ethylene, dispersive, 181, 182.

Ethylene dichloride, 133.
 Helium, not dispersive, 149, 152.
 Hydrogen, not dispersive, 131, 136, 146, 149, 152, 160, 163, 181, 188, 197, 198.
 Nitrogen, not dispersive, 149, 150, 162, 172.
 Nitrogen tetroxide, dispersive, 145, 155, 161, 179, 196.
 Nitrous oxide, dispersive, 131, 141, 142, 157, 173, 177, 199.
 Oxygen, dispersive, 139, 147, 152, 159, 163, 165, 172, 174, 192.
 Sulfur dioxide, dispersive, 144, 173, 177, 181, 199.

The general conclusions which result from a study of these papers are as follows:

(1) Dispersion and absorption of sound are to be expected in any polyatomic gas in which an appreciable amount of vibrational energy is excited, at pressures, temperatures, and frequencies which are at present experimentally available. The reason for this is primarily the inevitable inefficiency of the conversion of translational energy into a quantum of vibrational energy by collision. Very elementary and qualitative consideration of the process of collision shows that most of the force of the impact will be consumed by the repulsive forces of the colliding molecules, and that only a small fraction of it is available for the alteration of the vibrational energy.¹⁸⁷ A more detailed analysis by quantum mechanical methods confirms this.¹⁶⁶ A steric factor is also to be expected, since the structure of most molecules is such that the direction of the impact will be important in effecting excitation by collision. Experiment has found relaxation times for excited states, at room temperature and atmospheric pressure, ranging from $\theta = 10^{-8}$ sec. in sulfur dioxide to $\theta = 10^{-2}$ sec. in oxygen.

(2) The effect of pressure variations is in accord with the kinetic theory of heat; an increase of pressure increases the number of collisions, and displaces the dispersive region to higher frequencies. Very thorough measurements on carbon dioxide by several investigators^{140, 149, 150, 181} concur with but one dissenting voice¹⁷⁷ that the excitation of the transverse vibration of this gas is effected by double collisions in the sense demanded by (14) above. Double collisions appear to be the usual mechanism by which vibrational energy is excited, although in a few cases, such as the excitation of oxygen by water vapor,¹⁵⁹ triple collisions appear to be a possibility. The collision theory

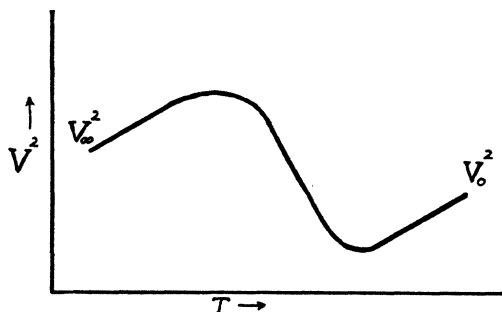


FIG. 4. The dependence of the velocity on temperature at constant pressure and frequency as found experimentally in several gases. The falling off of the velocity from V_∞ occurs because the transitions between states are more frequent and the collision lifetimes diminish as the temperature rises. At high temperatures the dispersive region has been traversed, and the velocity becomes V_0 .

has received impressive support from the study of the dependence of the dispersion on pressure. Indeed, no case has yet been reported where the effect of radiation has been detected, although with molecules having a large dipole moment and vibrational states of long relaxation times this is to be expected.

Incidentally, much confusion may be avoided in reporting the results of pressure studies if the old-fashioned custom of "reducing the velocity to 0°C " is abandoned. In the first place, velocities in the dispersive region are dependent on rates which are unspecified functions of temperature, and treating the velocity as if it were a function only of the square root of the absolute temperature neglects this completely. In the second place, the equation of state of an ideal gas is a poor approximation for a real gas, even over relatively small pressure intervals. For example, in carbon dioxide the zero frequency velocity at infinite dilution is, at 30°C , $271.2 \text{ m}\cdot\text{sec}^{-1}$ from band spectra. At 1700 mm pressure the apparent increase in molecular weight caused by the van der Waals forces has reduced V to $269.8 \text{ m}\cdot\text{sec}^{-1}$. A glance at (13) shows that a corresponding reduction in velocity will appear at all frequencies if the pressure is high. Hence an isotherm should never be plotted with V^2 and $\log \omega/P$ as its coordinates, but either V^2 and $\log \omega/N$ should be used, or V^2 should be idealized by correcting for van der Waals forces, and plotted¹⁸¹ against $\log \omega/P$.

(3) The effect of increasing the temperature is to increase the transition probability, which

produces an isobar of the type illustrated by Fig. 4. Since increasing the temperature at constant pressure decreases the number of collisions in proportion to $T^{3/2}$, according to the collision theory a function of this type necessarily indicates a decrease of the collision lifetime of the state causing the dispersive region. Such behavior was first noted in carbon dioxide, carbon disulfide, and ethylene,¹⁸¹ and has since been reported for carbon dioxide and several other gases.^{140, 142, 152, 199} The measurements which Penman¹⁷³ published, and which he considered to refute the collision theory, have been repeated by Warner,¹⁹⁹ and shown to be without sufficient accuracy to warrant any conclusions.

Although they do not agree in detail, measurements indicate that the increase in the $0 \rightarrow 1$ transition probability is exponential with temperature, and that the exponent involved is larger than that dictated by the Boltzmann factor corresponding to the particular state involved. Landau and Teller¹⁶⁶ point out that only molecules of high translational energy will be able to effect its conversion into vibrational energy, and that the process, when it occurs, will take place classically; they suggest the exponential factor which terminates (14). This would increase the transition probability in carbon dioxide at 100°C by a factor of about 2 in a 30° temperature interval. A rough analysis of Warner's results, which, although he has preferred to publish them without interpretation, appear to be the most complete available, shows that the probability is about doubled by raising the temperature 15° . In view of the difficulty of reading with accuracy the graphs which Warner has published, this is not considered a serious disagreement.

It may perhaps be unnecessary to point out, in passing, that if, for some reason not at present clear, a molecule which is in an excited state has an advantage over a molecule which is in the zero state in bringing about the conversion of translational into vibrational energy, an exponential temperature dependence of θ resembling a chemical activation energy will appear. In (14) it is implicitly assumed that a $0-1$ collision and a $0-0$ collision are equally effective in causing the $0 \rightarrow 1$ transition. If the $0-1$ collision is markedly more effective, (14) will be multiplied by $\exp[-h\nu/kT]$, and the tempera-

ture coefficient of the lifetime will be greater. Although this possibility seems remote, it is urgently necessary that more determinations of the effect of temperature be made if the full significance of dispersion measurements is to be understood.

(4) As a result of the researches of Knudsen¹⁶² with the reverberation method, which have been amply confirmed by others,^{172, 192} it has been found that very small traces of foreign gases may have an enormous effect on the efficiency of the collision process, and consequently on the location of the dispersive region. The effect of the presence of these traces is always to shift the dispersive region to higher frequencies; any other result would necessitate a revision of the collision theory. Kneser and Knudsen interpret this result¹⁵⁹ as meaning that a collision of a molecule of the dispersive gas with a molecule of the "trace catalyst" is more effective in exciting the vibrational energy of the dispersive gas than is a collision with one of its own species. Measurements on mixtures by a large number of investigators bear out this interpretation, but there is no general agreement why this is true. One school of thought regards the excitation of vibrational energy as a preliminary to chemical reaction, and seeks correlation between collision efficiency and the probability of a reaction involving the destruction of one or both of the colliding molecules.^{140, 170} Eucken, who has championed this view, even distinguishes between "chemical" and "physical" methods of excitation.¹⁴¹ In order to test the general validity of this interpretation the efficiencies of hydrogen and of deuterium in exciting ethylene were compared,¹⁸² and hydrogen found eight times as effective per collision. In this case the reaction probabilities of the two isotopes are practically identical, and the only decided difference lies in the moments of inertia, and consequent magnitudes of the rotational quanta. On this basis water and ammonia, which have small moments of inertia, should be effective trace catalysts, and appear to be so for a number of gases.^{140, 142, 159, 162, 186} It is clear, however, that the magnitude of the moment of inertia cannot supply an explanation for many cases; for example Knudsen reports that ethyl alcohol is the most effective catalyst for oxygen of the many

he has investigated. If, as is currently believed, there is no free rotation about the carbon-carbon bond in ethanol, the moment of inertia of this molecule must be very large, and its rotational quanta correspondingly ineffective. Nor does the suggestion that the magnitude of the dipole moment influences the transition probability¹⁷⁰ prove especially helpful. It seems necessary, therefore, tentatively to assume that the energy in several degrees of freedom in the ethanol molecule transfers itself simultaneously to the oxygen molecule during a collision. If this were the case, the torsional quanta of ethanol, which must be very large, might participate in the exchange. Processes of this nature are not unknown in chemical reaction kinetics.

To summarize: the efficiency of the conversion of translational energy into vibrational energy by the collision of a molecule of one species with one of another is highly specific, and no one property of the molecules is sufficient to define it. Further study will be necessary before the facts which are at present known may be fully explained.

(5) No information has been obtained concerning the rates of adjustment of the rotational energy or of the van der Waals forces, since both appear to follow the acoustical cycle to the highest frequencies yet employed. The report that rotational dispersion could be found in hydrogen¹⁸¹ has been refuted.^{160, 182, 188, 198}

(6) In many gases several sets of energy states are adjusted with a common frequency, with the result that they show only one dispersive region which is common to all of them. Examples of such behavior are carbon dioxide, carbon disulfide, ethylene,¹⁸¹ and nitrous oxide.¹⁴² In this case (20) combined with (13) gives, for an ideal gas where $\theta_A = \theta_B = \text{etc.}$

$$V = \left\{ \frac{RT}{M} \left(1 + \frac{R(C_\infty + C_A + C_B + \dots + C_\infty \omega^2 \theta^2)}{(C_\infty + C_A + C_B + \dots)^2 + C_\infty^2 \omega^2 \theta^2} \right) \right\}^{\frac{1}{2}},$$

where θ represents the lifetime of the most rapidly adjusted set of states from the standpoint of conversion of translational into vibrational energy. It is unlikely that the rates of adjustment of the several states are identical in these cases; more probably they fall out together because of rapid redistribution of energy within

the molecule. It is remarkable that the symmetrical linear vibration of carbon dioxide should have an independent rate which is some two orders of magnitude less than that of the transverse vibration, since the first excited state of the linear vibration perturbs the second state of the transverse vibration.^{189, 181, 198}

(7) In cases where there is doubt whether the molecule is colinear the value of the high frequency velocity provides conclusive proof. For example Reid's measurements on carbon dioxide prove it to be linear, although in this case the proof was unnecessary.¹⁸¹

(8) Chemical reaction rates may be determined by the acoustical method, but only gases with low heat capacities and high reaction heats are suitable at present.¹⁹⁵ Indeed, nitrogen tetroxide is the only success to date,^{161, 180} and this gas not only has a low heat capacity, but is exceptionally easy to purify.

THE EFFECT OF SOUND WAVES OF LARGE AMPLITUDE

Sound waves of great intensity may be obtained in the audible range from submarine signaling oscillators which give, under the most favorable conditions, acoustical outputs of about 500 watts.²⁷⁴ Sound waves in the audible and ultrasonic ranges may also be obtained by the pneumatic method,⁸⁶ but this is suitable for gases only. Magnetostriction oscillators¹¹⁴ made of nickel or chromium steel have been employed both at audible and ultrasonic frequencies; these are somewhat less efficient than the submarine oscillators. Commonly, however, the source is a piezoelectric crystal, usually quartz, driven in an insulating liquid. The liquid is necessary to prevent shattering of the crystal at large amplitudes, and is usually xylol or transil oil. Under transil oil the shattering point of crystal quartz is reached with an output of the order of 3 watts per cm² if the crystal is 5 cm in diameter; the shattering point may be reached with an electrical input of the order of 200 watts. A safe output is perhaps a fifth of this.^{298, 345} Whatever the source of sound or its frequency, the physical, chemical, and biological effects are usually much the same.

Although most observations with high amplitude waves are concerned with liquids, the

coagulation of smokes and other aerosols is an exception. Hiedemann and his co-workers^{265-270, 297} have shown both theoretically and experimentally that there is an optimum particle size for coagulation at any frequency. Other aspects of the process, including the absorption of sound by aerosols,^{135, 164} have been investigated.²⁵³ The formation of mists by a thin film of liquid in contact with a solid surface, which several observers have reported,^{101, 265, 339} should also be mentioned. Mist formation is evidently due largely to the transverse waves in the solid; in accordance with this a liquid is more easily "atomized" the lower its viscosity.

The presence of intense sound waves in liquids gives rise to several phenomena which all observers report. The surface of the liquid on which the waves impinge becomes disturbed, and at high intensities is actually broken up into drops which may be thrown several feet. It is not entirely clear how much this is due to actual radiation pressure and how much it may be due to secondary electrical and mechanical pumping actions entailed by the generation of sound. Again, the liquid heats slowly, evidently due to viscous absorption of sound. In measuring the heating due to absorption it is essential to eliminate dielectric power loss which, in early investigations, sometimes contributed nine-tenths of the heating effect.³¹⁸ The intensity of the waves may be measured either by modifications of the Rayleigh disk,^{65, 69, 73, 205, 342} or by total absorption in a solid such as pitch.³¹⁹ It has recently been observed that waves of relatively small amplitude are visible in liquids,³¹² and several demonstrations of their existence suitable for large gatherings have been described.^{68, 94, 262}

By far the most important consequence of the passage of intense sound waves through liquids is cavitation,^{261, 263, 298} or the formation of bubbles of gas or vapor. This sets in at intensities of about 0.03 watt per cm² in the ultrasonic range.²⁶⁴ Its importance lies in the fact that almost all of the phenomena described below disappear if easily visible cavitation is suppressed, either by degassing the liquid, or by applying to it a large hydrostatic pressure.^{295, 299, 302, 332, 343} Cavitation has been shown to exist in a degassed liquid very near its boiling temperature,³¹⁵ but only one observer has re-

ported it in pure water at room temperature,³⁴⁰ and he finds not easily visible bubbles, but opalescence. Since the gas bubbles formed by cavitation do not redissolve, but rise jerkily to the surface, the most obvious result is degassing of the liquid. This is a gradual process, however; unless the liquid is radiated many hours the volume of gas liberated per unit time appears independent of time.^{315, 341} The degassing of metal melts has been reported to have valuable technical application.¹

One of the most spectacular concomitants of cavitation is the faint luminescence of liquids which was first discovered in water.^{287, 348} Chambers has found the intensity of light to be proportional to the product of the dipole moment of the liquid and its viscosity.²⁷⁷ Several observers report that the luminescence originates at the liquid-gas interface of the cavitated bubble.^{277, 285, 301} This is a very fundamental observation, since it shows that enough energy is produced locally to give visible light of 6×10^{14} per sec. frequency. Clearly sound waves of frequency less than 10^6 per sec., such as are employed, cannot cause this by any primary process.

The acceleration of chemical reactions by intense sound has also been shown to be due to cavitation.^{285, 331} The great majority of reactions involved are oxidations such as the conversion of iodine ions into iodine molecules,^{255, 302} but effects on hydrolysis³¹⁵ and on the inversion of cane sugar³³⁶ have also been reported. There is some evidence that depolymerization results in certain liquids.³⁴² Several explanations have been advanced for the acceleration of chemical reactions, but none of them can here be accepted with any enthusiasm. The first³¹⁵ attributed the acceleration to a direct temperature effect: since the maximum values of $+\Delta T$ and $-\Delta T$ are practically equal, the net effect of the wave is to accelerate the reaction in proportion to its activation energy; in other words, since the rise and fall of temperature are linear and the dependence of the reaction rate on temperature is exponential an acceleration must result. This explanation was founded on a very exaggerated estimate of the amplitude of the waves, and may be refuted unequivocally by a thermodynamic argument. If we take 3×10^7 ergs per

cm² per sec. as the maximum power output of a quartz crystal, the pressure amplitude obtained from Rayleigh's expression is (reference 18, Vol. II, page 16)

$$I = P^2 / 2\rho V.$$

For water $\rho \cong 1$, $V \cong 1.5 \times 10^5$ cm per sec., and P is in this case therefore 2.6×10^6 dynes per cm². The resulting adiabatic temperature change from the relation first given by Joule

$$(\partial T / \partial P)_S = (T / C_P)(\partial v / \partial T)_P$$

is less than 0.01°C, and therefore too small to influence appreciably any known reaction.

Three secondary results of cavitation might be accelerating agents. One of these is the large pressure which Rayleigh has shown to accompany the collapse of bubbles.²⁵⁸ It might be argued that pressure surges from this cause are sufficiently great to cause the adiabatic temperature changes required. This explanation is improbable for several reasons. In the first place, easily visible cavitation disappears at a fairly sharp point with increasing hydrostatic pressure, and beyond this point no acceleration of reactions occurs. This is an indication that the formation of bubbles which contain gas, and which do not collapse readily, is the cause of the acceleration. Again, many reactions are accelerated only when certain dissolved gases are cavitated, and are unaffected by the cavitation of others. Finally, sonic cavitation is in general almost noiseless, whereas the bubble collapse which produces high pressures is accompanied by a rattling sound. A more attractive explanation has been suggested by Marinenco,³⁰⁶ who has ingeniously measured the surface temperatures of bubbles by determining whether explosives detonate when immersed in liquids which do not wet them. He finds that explosives with detonation temperatures below 230°C detonate, whereas those with higher detonation temperatures do not. The consistency of his results make it appear unlikely that resonance phenomena are present, and indicates that small regions of very high temperature exist in the liquid because of the presence of cavitated bubbles. An alternative suggestion results from the computations of Smith,³³⁴ which show that enormous pressures are built up in the neighbor-

hood of bubbles which are vibrating at their resonance frequencies. This will occur when the frequency of the sound wave is equal to a frequency determined by the diameter, surface tension, and gas pressure of the bubble. A sharp dependence on the acoustic frequency is of course entailed, after the manner of all resonance phenomena, and the explanation fails in the audible range because the dimensions of the resonant bubble become impossibly large. All three of these explanations provide regions in the liquid where the temperature is far higher than the average, and consequently they are perhaps sufficient to account for the acceleration of reactions which are already in progress. None can account, however, for luminescence, nor for the formation of hydrogen peroxide from oxygen and water which appears to be the basis for the acceleration of most oxidation reactions.

It appears necessary, therefore, to postulate a ballo-electrical (*Wasserfallelektricität*) mechanism in order to obtain sufficient energy. Just as the spraying of a liquid in air produces charged drops, the spraying of holes in a liquid, which is cavitation, must produce charged bubbles. In search of this effect, Mr. Hobart and I in the Loomis Laboratory investigated the electrical potentials in a liquid which was forced into strong cavitation by driving it through a constriction in a wide tube with high hydrostatic pressure. We found considerable random variations in potential in the region which was visibly cavitating, and none in other parts of the liquid which were flowing equally rapidly. It appears possible, therefore, that over distances comparable to molecular dimensions electrical potentials of the order of magnitude necessary to produce chemical reaction may be generated by cavitation. Apparently the first to appreciate this was Föttinger. (See reference 258, footnote.) A somewhat similar mechanism was suggested by Beuthe,²⁵⁵ who considered the origin of the charge frictional, and by Levšin and Kževkin in connection with luminescence.³⁰¹

An important member of the group of phenomena produced by cavitation is the formation of colloidal systems, more especially the emulsification of one liquid in another.^{283, 304, 336, 345} This, in distinction to the acceleration of oxidation reactions, is usually independent of the identity

of the cavitating gas. The emulsification of various liquids, including mercury,³⁴⁵ in water has been reported by many observers, and studied in detail. Heavy liquid metals such as mercury and gallium do not appear to require cavitation in order to be emulsified,^{257, 314, 316} but other emulsifications are greatly facilitated if it occurs.^{258, 259, 295, 323} The emulsions produced are often more stable than those produced by colloid mills or similar devices. There appears to be a small group of technical products, ranging from cosmetics to photographic plates, which can be produced in better quality by supersonic emulsification than in any other way.^{279, 282} Colloidal metals have been produced by subjecting the cathode to sonic vibration during electrolysis,²⁸¹ and by analogous procedures.^{280, 326} Many inorganic and organic sols³²⁴ and gels²⁹⁰ are peptized by supersonic waves without the presence of peptizing agents. On the other hand, coagulation will result if the particle size bears a certain relationship to the acoustical wavelength,^{271, 311, 338} the analogy to the coagulation of aerosols naturally presents itself. Naturally flow orientation of unsymmetrical particles occurs at small intensities; this may readily be demonstrated with aged vanadium pentoxide sols between crossed Nicol prisms.²⁷¹ Naturally also thixotropic gels are liquefied by the motion and local heating of the medium.²⁸⁹

Several other physical and physico-chemical effects which have been described³¹⁵ should briefly be mentioned. Explosives are detonated not only in liquids, as mentioned above, but also on vibrating glass surfaces in air. Sound waves will alter sensitive metastable systems such as, for example, the yellow to red enantiotropic change in mercuric iodide at room temperature. Superheated liquids break into almost explosive ebullition on radiation with intense sound.³¹⁵

Undercooled liquids and supersaturated solutions are forced to crystallize by intense sound, but only under restricted conditions.³²¹ Since there have been conflicting reports about this,^{341, 345} the matter will be discussed a little more fully. It has long been known that many liquids, such as benzophenone, remain undercooled indefinitely at room temperature if they have been preheated high above their melting temperature, but recrystallize readily if melted

and preheated only a few degrees. When the preheating has been high no amount of shaking produces crystallization; when it has been low shaking produces the immediate appearance of crystals. The presence of certain adsorbing substances, such as carbon or clean glass, requires that the preheating shall be far higher than if they are absent. Often dust, which has been included by accident and is of unknown composition, serves as the adsorbent; many liquids will not crystallize at any temperature if they have been passed through sufficiently fine filters and sealed in flamed glass tubes. The role of supersonic waves is exactly equivalent to shaking: when the liquid may be caused to crystallize in a sealed glass tube by shaking, supersonic waves will also cause it to crystallize, but if it has been sufficiently preheated neither will have any effect. Another aspect of the influence of supersonic waves on crystallization is the decrease in grain size of metals on acoustical treatment of the crystallizing melt.^{328, 333} This is to be expected with all crystallizing substances, since the sonic agitation shears off small particles of the growing crystals, and these particles act in turn as nuclei.

The sonic irradiation of metals which are in the passive state is sometimes to increase the passivity and sometimes to relieve it; similarly, metals which are cathodically polarized during electrolysis are depolarized by sound if the current density is low.³²⁷ Some polarized elec-

trodes are microphonic.³⁰⁹ The quality of some electrolytic deposits is improved by sonic radiation during electrolysis.³⁴⁷ The fogging of photographic plates in a standing-wave field has been reported by Marinisco,^{305, 307} who attributes it to sonic activation of molecules; the ballo- electrical hypothesis accounts for this. Finally, it has been shown that the electrical conductivity is increased and the electrical breakdown voltage diminished in a standing-wave field.³⁰⁸

Numerous biological observations have been made since the beginning of supersonic history.^{254, 346} Blood is laked,^{272, 295} small animals are killed,^{256, 299} and organisms such as paramoecia, algae, and many bacteria are destroyed.^{273, 292, 293, 296, 329, 344} These effects appear to depend largely on cavitation. The reduction of curd tension in milk has been studied,²⁷⁴ and many dairies use sonic treatment in the preparation of infant's milk. The influence of cavitation on the activity, stability, and degree of dispersion of several proteins,^{275, 284} including some pepsins,²⁷⁸ has been studied. Chambers appeared to have made an especially valuable discovery when he demonstrated the sonic extraction of some labile bacterial constituents;²⁷⁶ he found later, however, that the method was not of as general value as was to be hoped.^{322, 325} Several therapeutic suggestions for the use of supersonic waves have been made,^{286, 288} but none seems to have yielded results of general value.

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The reader is entitled to know how a bibliography has been compiled, and just how comprehensive he may expect it to be. In collecting these papers I have gone through the section marked "Acoustics" in *Science Abstracts A* from January 1934 to April 1938, recording all pertinent work, but omitting references to explosions, signaling, and theoretical treatments of the modes of vibration of rods, disks, and the like. For earlier work I have leaned heavily on Gross-

* Note added on going to press. This bibliography was completed early in June 1938, and is being sent to the printer in January 1939. In the intervening months several important papers, dealing especially with the dispersion of sound in liquids, have appeared both in Germany and in India. I regret that it is not practical to include them here, and assure their authors, several of whom have sent me reprints, that they are not being wilfully ignored.

mann's *Ultraakustik (Handbuch der Experimentalphysik*, Vol. 17 (1934), p. 463), which has been compiled with exceptional scholarship and understanding. I am well aware that the result is incomplete, since I should have missed my own last two papers by following this procedure. Since many of the titles convey little or no information (for example my own "The Chemical Effects of High Frequency Sound Waves. I. A Preliminary Survey"), I have tried to indicate in a few words what seem to me the important points of the paper. The separation into categories is often somewhat arbitrary since many of the papers belong in several categories at once, but it is hoped that the reader may find it useful. The instrumental section is less complete than the others.

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