

<sup>1</sup>S. Ichimaru, preceding paper, Phys. Rev. **174**, 289 (1968), referred to as Paper I.

<sup>2</sup>Equations in Ref. 1 are cited here by affixing I before the equation numbers; for example, (I.4.23) means the Eq. (4.23) in Paper I.

<sup>3</sup>S. Ichimaru and T. Nakano, Phys. Letters **25A**, 163 (1967); Phys. Rev. **165**, 231 (1968).

<sup>4</sup>L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon Press, Ltd., London, 1958), Sec. 74. To make the comparison clearer, we note that in thermodynamic equilibrium one can write  $G(12) = F(1)F(2)\omega(\vec{r}_1 - \vec{r}_2)$ , where  $\omega$  has the same meaning as that defined by Eq. (74.22) in the Landau-Lifshitz book. Equation (2.2) then becomes

$$f_3(123) = F(1)F(2)F(3) \\ \times [1 + \omega(\vec{r}_1 - \vec{r}_2)] [1 + \omega(\vec{r}_2 - \vec{r}_3)] [1 + \omega(\vec{r}_3 - \vec{r}_1)].$$

This equation corresponds to that appearing at the top of p. 235 in the above-mentioned book.

<sup>5</sup>This point will be further elucidated in Sec. V after a self-consistent equation for  $\mathcal{G}_{\vec{k}}$  is obtained.

<sup>6</sup>The definition of  $\mathcal{G}_{\vec{k}}$  in Eq. (4.8) is different from that in (I.4.30) by a factor  $\exp(2\tilde{\gamma}_{\vec{k}}t)$ .

<sup>7</sup>D. Pines and J. R. Schrieffer, Phys. Rev. **125**, 804 (1962).

<sup>8</sup>S. Ichimaru, in *Fluctuation Phenomena in Solids*, edited by R. E. Burgess (Academic Press Inc., New York, 1965), p. 113.

<sup>9</sup>For simplicity in presentation, we restrict the range of our consideration in the remainder of this section to a half  $\vec{k}$  space such that  $\tilde{\omega}_{\vec{k}}$  and  $\tilde{\epsilon}'(\vec{k}, \tilde{\omega}_{\vec{k}})$  are positive; for the other half, similar discussion also applies. See Sec. VI for a general argument.

<sup>10</sup>Reference 8; further references may also be found here.

<sup>11</sup>See also a discussion in the introduction of a review paper by V. N. Tsytovich, Usp. Fiz. Nauk. **90**, 435 (1966) [English transl.: Soviet Phys. - Usp. **9**, 805 (1967)].

<sup>12</sup>The latter work of Ref. 3 contains a discussion and further references on this superposition principle.

<sup>13</sup>Reference 8, p. 120.

## Ultrasonic Attenuation In Liquid Metals

K. C. Sharma\*

*Department of Physics, Theoretical Physics Institute,  
University of Alberta, Edmonton, Canada  
(Received 12 April 1968)*

Using the thermodynamic theory of relaxation processes, and incorporating the disorder model of melting mechanism in the two-state theory, the ultrasonic attenuation in simple liquid metals has been investigated. Agreement of the results with the experimental bulk viscosity indicates that this might be a genuine approach to explain the excess absorption of ultrasonic waves in monatomic fluids. The possibility of extending the theory to other associated liquids with complex structures is also discussed.

### 1. INTRODUCTION

It is well established that the ultrasonic attenuation<sup>1</sup> in liquid metals is greater than that expected due to classical causes,<sup>2,3</sup> viz., the shear viscosity and the thermal conductivity. The excess attenuation<sup>4</sup> per cm can be conveniently described in terms of a frequency-independent bulk viscosity  $b$  given by

$$b/\eta = \frac{4}{3}(A_{\text{exp}} - A_{\text{cl}})/A_{\eta} = \frac{4}{3}A'/A_{\eta} \quad (1)$$

where  $\eta$  is the shear viscosity,  $A_{\text{exp}}$  is the observed attenuation of sound waves,  $A_{\eta}$  is the absorption due to shear viscosity and  $A_{\text{cl}}$  is the sum of  $A_{\eta}$  and the absorption due to thermal conductivity.  $A_{\text{cl}}$  is usually referred to as the attenuation due to classical causes.

In most of the fluids,  $A_{\text{exp}}$  exceeds  $A_{\text{cl}}$  significantly. In polyatomic fluids the energy is distributed between the translational (external) and vibrational and rotational (internal) degrees of freedom. The sound wave disturbs the equilibrium distribution of energy between the external

and internal degrees of freedom. The restoration of equilibrium at a finite rate gives rise to the extra absorption which is known to be due to the thermal relaxation processes. In monatomic fluids the temperature and pressure changes associated with the sound wave will perturb the local spatial arrangements of the molecules in equilibrium. Since the equilibrium is restored at a finite rate, one may expect the occurrence of a relaxation process which is responsible for the excess absorption. If such structural changes involve a change in volume, both the temperature and pressure fluctuations can induce the lack of equilibrium.

There have been a few attempts on the liquids having complex structures, but no theoretical estimates of bulk viscosity  $b$  have yet been made for simple monatomic liquids. Though the structural relaxation processes have already been investigated in hydrogen-bonded<sup>5,6</sup> and other associated liquids,<sup>7,8</sup> a clearer understanding of the mechanisms in liquids with simpler and more defined structure would indeed be of much importance. The monatomic liquids are the only

obvious choice as the associated liquids which are easily distinguishable from the normal ones.

Recently, Eckstein<sup>8</sup> proposed the disorder model for liquid metals, applicable only to those shrinking on freezing. He obtained the values of several thermodynamic properties (such as specific heat, thermal-expansion coefficient, etc.) in fair agreement with the experiments. The model assumes that the changes in physical properties arise from the spontaneous production of thermodynamic defects (or vacancies) in the medium during transition at melting point, where the defect is characterized by its activation energy and the activation free volume rather than the discrete atomic arrangement in the liquid state.

The object of this paper is to develop the theory for excess attenuation of sound waves in liquid metals assuming a model of the liquid having different phases corresponding to the different free energies, and then to apply the theory to simple liquid metals. Finally, implications of the theory for real systems is discussed and, in a sense, compared with the Hall picture<sup>5</sup> of molecular compression in liquids.

## 2. THEORY

### A. Basic Concept of the Two-state Model

In order to describe the physical processes in monatomic fluids, the simplest two-state model is assumed. The states of the molecules in a fluid in thermodynamic equilibrium can be characterized by the considerations of difference in the structural arrangements which might correspond to different free energies. At a particular temperature and pressure a certain fraction of the number of molecules would be in state 1, and the remaining fraction would be in another state. When a sound wave passes through the medium, the equilibrium distribution of the number of molecules in two states is perturbed. The energy required by a molecule to jump from one state to another is known as the activation energy  $E$ . Equilibrium would be established at a finite rate which would lead to an absorption of sound waves.

### B. Free Energy and Defect Concentration

Limiting ourselves to the liquids which shrink on freezing, we make the following simplifying assumptions:

(1) According to the two-state model of liquid metals, each molecule would be assumed to stay in state 1 characterized by smaller volume and — contrary to the Hall's view — lower energy, or in state 2 characterized by larger volume and higher energy.

The smaller volume corresponds to the structure of the solid state and the larger one is assumed to correspond to one containing an equal number of atoms and defects (vacancies), i. e. the volume of structure in state 2 can be written as

$$V_2 = V_1 + Nv_V \quad (1)$$

where  $v_V$  is the volume of one defect and  $N$  is the total number of molecules on the lattice sites. Because the discrete nature of defects seems to lose its physical significance in the liquid state<sup>9</sup>, an approximate quasicontinuous distribution is assumed.

(2) The volume of the defects produced in melts is quite comparable to that  $V_1$  of the solid having no vacancies at all. This can be assumed to be a constant fraction  $F$  such that  $V_V = FV_1$ .

$$\begin{aligned} \text{Then } V'/V &= (V_2 - V_1)/V = Nv_V/(V_1n_1 + V_2n_2) \\ &= F/(1 + n_2F) \end{aligned} \quad (2)$$

with  $n_2$  the relative concentration of structure in state 2 present in the liquid.<sup>10</sup> This expression is independent of any volume but assumes that at melting point the total volume of defects is a constant fraction of that before the melting point. The factor  $F$  should in principle be different for different liquids.

Mathematically, the Gibbs free energy of the fluid can be written as

$$G = G_0(P, T) + n_2NE - kT \ln W. \quad (3)$$

The function  $G_0(P, T)$  is the energy of the solid in the absence of defects as a function of pressure and temperature;  $n_2$  is the fraction of the number of defects present in the liquid;  $E$  is the activation energy needed to create one defect, and the third term represents the contribution of entropy. The quantity  $W$  is the number of ways in which  $n$  defects can be distributed over  $N$  lattice sites, which equals  $N!/[N-n]n!$ . For equilibrium

$$\begin{aligned} (dG/dn)_{P, T} &= 0 = E - kT \ln[(N-n)/n] \text{ or} \\ n_2 &= n/N = (1 + e^{E/kT})^{-1}. \end{aligned} \quad (4)$$

The second derivative of  $G$  with respect to  $n$  can be written as

$$\begin{aligned} (d^2G/dn^2)_{P, T} &= kTN/n(N-n) \\ &= RT(n_1 + n_2)/n_1n_2 \end{aligned} \quad (5)$$

where  $n_1 = 1 - n_2$ , and will be utilized later in this section.

### C. Excess Absorption of Ultrasonic Wave

Following the notation of Bhatia,<sup>4</sup> the excess absorption per cm at low frequencies can be written as

$$A' = 2\pi^2 f^2 \tau_S \tau_{SP} / V_0, \quad (6)$$

where  $f$  and  $V_0$  are the frequency and the velocity of the sound wave respectively. The quantity  $\tau_S$  denotes the relaxation strength, which is neither purely temperature-induced nor purely pressure-induced, and is given by

$$r_S = (1/k_S)(V/\Phi)(c_P/c_P^\infty)(V'/V - H'\beta/c_P)^2 \quad (7)$$

with  $\Phi = (d^2G/dn^2)_{P,T} = RT(n_1+n_2)/n_1n_2$

$$\text{and } \tau_{S,P} = (c_P^\infty/c_P)\tau_{T,P} = (c_P^\infty/c_P)n_2(V/RT)\eta.$$

The quantity  $k_S$  is the adiabatic compressibility,  $c_P$  and  $c_P^\infty$  are the static and instantaneous heat capacities at constant pressure respectively,  $R$  is the gas constant, and  $T$  is the temperature in absolute units. The quantities  $\tau_{S,P}$  and  $\tau_{T,P}$  are the relaxation times at constant entropy and pressure, and at constant temperature and pressure respectively. In the above expressions, a single ordering parameter, the characteristic of the two-state model, has been used. However, in reality several mechanisms of relaxation may involve more than one ordering parameter. The quantities  $V'$  and  $H'/T$  represent the change in volume and entropy of the system for a unit change in the ordering parameter at constant temperature and pressure, i. e.

$$V' = (\partial V/\partial n)_{P,T}, H'/T = (\partial S/\partial n)_{P,T}. \quad (8)$$

Taking the thermodynamic quantities at infinite and zero frequency, one gets the relation between frozen and equilibrium coefficients, as

$$k_T - k_T^\infty = (V'/V\Phi)^2, \quad (9)$$

$$\beta - \beta^\infty = V'H'/VT\Phi, \quad (10)$$

$$\text{and } c_P - c_P^\infty = H'^2/T\Phi. \quad (11)$$

From (10) and (11) we can write

$$H'\beta/c_P = (V'/V) [(1-c_P^\infty/c_P)/(1-\beta^\infty/\beta)], \quad (12)$$

where  $\beta$  is the coefficient of thermal expansion. Using these formulas, Eq. (6) can be written in the form

$$A'/f^2 = 2\pi^2(V_0/\rho)(M/RT)^2\eta n_1n_2^2(V'/V)^2 \times [1 - (1-c_P^\infty/c_P)/(1-\beta^\infty/\beta)]^2 \quad (13)$$

where  $M$  is the molecular weight of the liquid. The Eq. (13) can also be rearranged in the form

$$b/\eta = \frac{4}{3}A'/A_\eta = (MV_0^2/RT)^2 n_1n_2^2(V'/V)^2 \times [1 - (1-c_P^\infty/c_P)/(1-\beta^\infty/\beta)]^2 \quad (14)$$

with  $A_\eta = 4/3\eta(2\pi^2f^2/\rho V_0^3)$ .

### 3. CALCULATION AND RESULTS

For calculating the excess attenuation of ultrasonic waves from Eq. (13), it is obvious that there are four unknowns, viz.  $n_2$ ,  $V'$ , the  $c_P^\infty$ , and  $\beta^\infty$ .

Under assumption (2) in the theory, the total volume change at melting point is attributed to the spontaneous production of vacancies, and  $n_2$  is calculated from

$$\Delta V/V_S = Fn_2, \quad (15)$$

where  $\Delta V$  is the change in volume and  $V_S$  is the volume of the crystalline state at melting point. Then Eq. (4) can be written as

$$E = kT \ln(1/n_2 - 1). \quad (16)$$

The constant-volume fraction  $F$ , corresponding to vacancies produced at melting temperature, is mentioned by Hirai and Eyring to be between 0.16 and 0.20, from which Eq. (2) gives  $V'$ .

Once  $n_2$  is known, the temperature- and pressure-induced part of the relaxation processes given by

$$(V'/V)^2 [1 - (1-c_P^\infty/c_P)/(1-\beta^\infty/\beta)]^2$$

can be calculated from the experimental values of  $b/\eta$ . Since the exact values of  $c_P^\infty$  and  $\beta^\infty$  are not known, we compare the total dilational relaxation contribution with the theoretical value of this term by approximating the instantaneous values as given by those in the solid state at melting point. Comparison has also been made with Fürth's<sup>11</sup> theory in which the actual volume of the vacancy as a function of temperature and surface tension was derived from the statistical treatment. These results for both values of the constant  $F$  are presented in Table I. The agreement is found to be very good within the limits of experimental data available and of the assumptions made in this paper.

### 4. CRITIQUE

In liquid metals the important contribution to the ultrasonic attenuation comes from the thermal-conductivity part which is very sensitive to the presence of impurities and is approximately 75% of the total absorption. This contribution masks the effect of shear viscosity in  $A_\eta$  which is only ~15%. Because of discrepancies in the data used to calculate the thermal-conductivity term, we should expect some disagreement between the theoretical and experimental values of  $b/\eta$  given by Eq. (14). Furthermore, except for a few cases, detailed and reliable measurements on the attenuation of sound waves in liquid metals are lacking. The case of sodium is very strange in this respect, and because of this the value calculated here lies between those given by Lechter and Beyer<sup>12</sup> and by Jarzynski and Litovitz<sup>13</sup>.

It is to be noticed that the change in instantaneous values of  $C_p$  or  $\beta$  even by 3 to 4% may affect  $(V'/V - H'\beta/C_p)$  by as much as 30 to 40%. Therefore the validity of  $C_p^\infty$  and  $\beta^\infty$  taken from those in the solid state at melting point can be questioned but is not arbitrary. For they must satisfy strictly the condition

TABLE I. Comparison of results.  $\Delta V/V_S$  is the fractional change in volume at melting point,  $V_0$  is the velocity of ultrasonic wave,  $\rho$  is the mass density of liquid at melting point,  $b/\eta$  is the ratio of bulk to shear viscosity from experiments (nearer the melting temperature),  $A'$  is the observed excess attenuation per cm proportional to the frequency  $f^2$  of the ultrasonic wave,  $F$  is the ratio between volumes of a vacancy and of the molecule corresponding to  $V_1$ , and  $n_2$  is the concentration of the structure corresponding to  $V_2$ . Finally,  $(V'/V - H'\beta/c_P)$  represents the two terms corresponding to pressure- and temperature-induced relaxation processes respectively.

Metal	$\Delta V/V_S$	$V_0$	$\rho$	$\frac{b}{\eta}$	$\eta$	$A'/f^2$	$F$	$n_2$	$(V'/V - H'\beta/c_P)$			Reference
		( $10^5$ cm/sec)							(g/cc)	( $10^{-2}$ poise)	$\times 10^{-17}$	
Na	0.025	2.526	0.926	2.60	0.73	2.37	0.16	0.156	0.2293	0.1093	0.071	13
							0.20	0.125	0.2815	0.1366		
K	0.0255	1.820	0.830	1.90	0.55	0.81	0.16	0.159	0.0886	0.0544	0.042	13
							0.20	0.127	0.1087	0.0680		
Zn	0.042	2.790	6.92	1.10	3.027	0.42	0.16	0.262	0.0523	0.1064	0.0579	18
							0.20	0.210	0.0632	0.133		
Cd	0.047	2.221	8.02	2.70	1.48	1.37	0.16	0.293	0.0742	0.1311	0.0513	18
							0.20	0.235	0.0891	0.1639		
Pb	0.036	1.790	10.88	0.30	2.70	0.30	0.16	0.225	0.0226	0.0125	0.0055	18
							0.20	0.18	0.0274	0.0156		
Sn	0.026	2.27	6.97	1.50	2.22	1.48	0.16	0.162	0.0756	0.1268	0.0792	18
							0.20	0.130	0.0927	0.1584		

<sup>a</sup>From Eq. (15).

$$(1 - c_P^\infty/c_P)/(1 - \beta^\infty/\beta) \ll 1 \quad (17)$$

if the relaxation processes are primarily pressure-induced. Otherwise the temperature fluctuations will also induce the relaxation significantly. Recently the bulk and shear modulus at infinite frequency have been calculated for liquid argon using the molecular theory by Zwanzig and Mountain.<sup>14</sup> If such calculations are extended to get the instantaneous values of  $c_P$  and  $\beta$ , etc. for monatomic fluids, it would be a convenient and easier task to compare the theory with experiments.

In general, thermodynamically, the liquid in comparison with the solid state can be viewed as a substance with higher entropy and higher internal energy. But the change in volume and in coordination number in particular would have to be decided by the resultant effect of different mechanisms causing transition. The total entropy would be additive. To understand the distribution of energies in different phases of the liquid, the higher energy must be attributed to the larger volume. Here it can be noted that the theory developed in this paper is more suited to monatomic liquids having closest packing in the solid state for which the Hall's theory gives an incorrect perspective.

Though the presence of multistructure<sup>15, 16</sup> in liquids does not necessarily imply the failure of Hall's picture of molecular compression in liquids, this picture, as the only relaxational process,

appears to be somewhat unphysical on the grounds mentioned above. Mathematically, from Eq. (13) or (14) it is very apparent that either the negative or the positive value of  $V'$ , because of its square, would contribute identically to the excess attenuation. The decisive role of the mechanisms would come naturally from the pressure dependence of  $A'$ . Taking a similar view, Carnevale and Litovitz<sup>17</sup> contradicted Hall's picture of the change in volume from one structure to another, on the basis of their experiments on pressure dependence of  $A'$ . Therefore the agreement obtained by Hall seems to be fortuitous.

In order to generalize the approach to other associated liquids, one must take into account the quasidiscontinuous increase of vacancies and the contraction of spatial molecular structure depending upon the openness of the configuration in the crystalline state during transition. Therefore all the monatomic fluids should be treated on the same footing. But at the moment, because of the complexity in structure, molten Hg, Bi, Ga, etc. are excluded until the accuracy of the data used in the analysis for simple metals is achieved with confidence.

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## Measurement of the Coherent Neutron Scattering Amplitude of Deuterium, Mercury, and Fluorine by Mirror Reflection\*

W. Bartolini, R. E. Donaldson, and D. J. Groves

*Lawrence Radiation Laboratory, University of California, Livermore, California*

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The bound coherent neutron-scattering amplitudes of deuterium, mercury, and fluorine have been measured by the mirror-reflection method. The measured values are  $a_D = +6.21 \pm 0.04$  F,  $a_{Hg} = +12.67 \pm 0.13$  F, and  $a_F = +5.83 \pm 0.17$  F. The mercury and fluorine values are in agreement with presently accepted values. The deuterium value departs seriously from most previously reported measurements. This value implies  $+6.13 \pm 0.04$  F for the quartet scattering amplitude,  $+0.13 \pm 0.05$  F for the doublet scattering amplitude, and  $3.15 \pm 0.04$  b for the free-atom scattering cross section of deuterium.

### INTRODUCTION

This paper describes measurements of the coherent neutron scattering amplitude of deuterium, fluorine, and mercury done by the mirror-reflection method of the Livermore Pool-Type Reactor. The theory of measurement of coherent neutron scattering amplitudes by reflection from mirror surfaces, as well as the experimental arrangement used in these measurements, has been described in the literature.<sup>1-5</sup>

### METHOD OF MEASUREMENT

Our experimental procedure consists of finding the angles at which the reflected neutron intensities from a standard liquid and from a liquid containing the unknown element are the same. Since equal reflected intensities imply equal reflection coefficients, we have the relation

$$(\phi_1^2)^{-1} \sum_i N_i a_i = (\phi_2^2)^{-1} \sum_j N_j a_j, \quad (1)$$

which does not depend explicitly on the spectral distribution of the incident neutron beam. The subscripts  $i$  and  $j$  in Eq. (1) refer to the constituents of mirrors 1 and 2 respectively;  $N$  and  $a$  stand for the number of scattering centers per unit volume and their coherent neutron scattering amplitudes; and the angles  $\phi_1$  and  $\phi_2$  are those yielding equal intensities from the corresponding mirrors. That is, in order to carry out the measurement, the angles of equal intensity, the chemical composition of both mirrors, and the coherent neutron scattering amplitude of all constituents but the one to be measured must be known.

Equation (1) is not valid, however, unless the reflected intensities are corrected for incoherence and absorption. Some knowledge of the incident neutron spectrum is required before these corrections can be made. The corrections applied to the experimentally determined intensities account for: (1) gain drifts in the detectors and amplifiers, (2) changes in the mirror density with temperature, (3) fast- and thermal-neutron backgrounds, (4) scattering of both incident and reflected beams in the vapor above the liquid surface, (5) variation of the incident beam intensity with angle, (6) finite angular divergence of the beam, and (7) incoherent scattering and absorption in the mirror. These corrections are described in detail in Ref. 3, pp. 535-538. Corrections (1) through (5) are simple and straightforward. Once they have been made, the reflected intensity is given by

$$I_{\text{exp}}(\phi_0) = \int_{\phi_0 - \Delta\phi}^{\phi_0 + \Delta\phi} \left[ \int_0^{k_{\text{Be}}} R(\phi, A, \gamma, k) \times \Phi(k) T(\phi) dk \right] d\phi \quad (2)$$

where

$R$  is the reflection coefficient of the surface,  
 $A = \sum_j N_j a_j$  for the  $j$  constituents of the mirror,  
 $\gamma = \sum \{ [N\sigma(k)] \text{ incoherent} \}_j + \sum \{ [N\sigma(k)] \text{ absorption} \}_j$ ,  
 $\Phi(k)$  is the incident flux distribution (beryllium-filtered spectrum),  
 $T(\phi)$  is the triangular transmission function of the collimator,  
 $\Delta(\phi)$  is the angular divergence of the beam, and  
 $k_{\text{Be}}$  is the beryllium cutoff wave number.