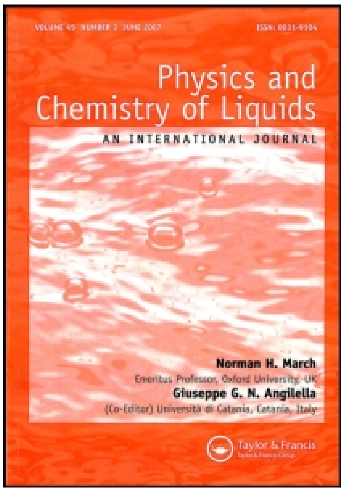

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Bulk Viscosity and the Relation between Transport Coefficients

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BULK VISCOSITY AND THE RELATION BETWEEN TRANSPORT COEFFICIENTS

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The longitudinal and bulk viscosity of the fluid Argon is calculated using its relation with self diffusion coefficient. This relation was derived by developing the relation between coherent and incoherent scattering functions. The results obtained are compared with recent simulation data of bulk viscosity. A good agreement is achieved for a wide range of temperatures at the triple point density. Our results successfully explain the increase in bulk viscosity with decrease in temperature near the triple point. The validity of the relation between diffusion and longitudinal viscosity is also tested for liquid metals. The results obtained for liquid metals of the longitudinal viscosity, at their melting points, are not found to be in agreement with experimental results. A relation between thermal conductivity and self-diffusion coefficient is proposed.

KEY WORDS: Bulk viscosity, diffusion, thermal conductivity, fluid Ar, liquid metals.

1 INTRODUCTION

In the last three decades extensive computer simulation works for hard sphere, Lennard-Jones (LJ) fluids and liquid metals have been done to study the dynamical correlations and transport properties. In these studies dynamical structure factors and transport coefficients: shear viscosity η_l, self-diffusion coefficient D, and thermal conductivity λ, have been evaluated. However, there exist, still, very less work (theoretical as well as experimental) on the longitudinal viscosity η_l ($=4\eta/3 + \eta_v$; η_v is bulk viscosity) or bulk viscosity of the fluids. For liquid metals there exists very little experimental data on bulk-viscosity. The experimental measurements are complicated and can be made only indirectly. No simulation work has been done to calculate the bulk viscosity of the liquid metals. On the other hand, for liquid Argon, recently, some equilibrium[1,2] and non-equilibrium[3,4] molecular dynamics (MD) simulations have been made to study the density and temperature dependence of the bulk-viscosity. In the simulation studies it has been found that the bulk-viscosity increases with decrease in temperature at and near the triple point density which is in contrast to the predictions of the existing kinetic theories. The kinetic theories[2] results are at least 60 percent off from the simulation results. In fact, there exist no kinetic theory which can readily be used to predict the transport propeties of the dense fluids.

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The theoretical approach which is, now, more freqently used for the dense fluids is through the Green Kubo formulae. In this approach the transport coefficients are expressed as the time integrals of the appropriate time correlation functions. These formulae can also be written[5] equivalently as the long wave length and small frequency limits of the dynamical structure factors (self and collective). For example, self diffusion is related to $S_s(k, \omega)$ and η_l and λ are related to $S(k, \omega)$. Since, it is simpler to develop models for $S_s(k, \omega)$ than for $S(k, \omega)$, attempts have been made[6–9] in the past to develop the relations between $S_s(k, \omega)$ and $S(k, \omega)$. These relations ultimately provide a connection between η_l and D. In the present work we use such a relation obtained by Hassan and Lado[10] to calculate the longitudinal viscosity, knowing the diffusion coefficients, of the fluid Argon and of liquid metals. Therefore, the aim of the present work is two fold: (i) to calculate the longitudinal and bulk viscosity of the fluids and (ii) to examine the relation between the transport coefficients.

The calculation of η_l and of η_v from the relation between η_l and D requires the knowledge of interaction potential and the radial distribution function, $g(r)$. We have used the LJ potential for fluid Argon and the theoretically generated $g(r)$. The results obtained for η_l and η_v are compared with recent simulation results of Borgelt *et al.*[2]. A good agreement is achieved at the triple point density for a wide range of temperatures. On the other hand, for liquid metals we have simplified the calculations and avoided the use of the interaction potential and $g(r)$. The final approximate expression obtained for η_l, valid only at the melting point, for the liquid metal requires only the compressibility data as input. We have used experimental results[11] for the compressibility to calculate η_l for liquid Na, K and Al at their melting point. The results obtained have been compared with the experimental data on bulk viscosity[12] and with earlier theoretical predictions[13,14]. We find that our results are of the same order as that of the other theoretical predictions. The values of η_l obtained are not reliable as these are of the order of shear viscosity. Therefore, it may be concluded that the relation, used here, between η_l and D for calculating η_l, valid for fluid Argon, is not true for liquid metals. This may be due to the fact that the dynamics in fluid Argon and in liquid metals are quite different, especially in the small k limits.

In the present work, we also suggest a relation between thermal conductivity and self diffusion coefficient. The relation is obtained by relating the self motion to the collective motion dynamics. We find that thermal conductivity is related to the self diffusion through specific heats and the compressibility. The result obtained for thermal conductivity from this relation, for fluid Argon at the triple point, is remarkably in good agreement with experimental result. We have also applied this relation to liquid Na and compared the result with earlier prediction of ionic contribution to the thermal conductivity. We find that result is also in good agreement. However, it may be pointed out that the relation obtained still requires further investigations.

The layout of the paper is as follows. In Section 2, we have given the available relations between $S_s(k, \omega)$ and $S(k, \omega)$. The relation between η_l and D is also given there. In Section 3 we have carried out the calculations of the bulk viscosity of the fluid Argon and liquid metals. The relation between thermal conductivity and self diffusion constant is derived in Section 4. The concluding remarks are given in Section 5.

2 GENERALITIES

The relation between transport coefficients and dynamical structure factors[5], through the Kubo relations, are given by

$$D = \tfrac{1}{2} \lim_{\omega \to 0} \lim_{k \to 0} \frac{\omega^2}{k^2} S_s(k, \omega), \tag{1}$$

$$\eta_l = \tfrac{4}{3}\eta + \eta_v = \frac{m^2}{2k_B T V} \lim_{\omega \to 0} \lim_{k \to 0} \frac{\omega^4}{k^4} S(k, \omega), \tag{2}$$

$$\frac{n^2 k_B T^2 V \alpha^2}{\lambda} = \tfrac{1}{2} \lim_{k \to 0} \lim_{\omega \to 0} k^2 S(k, \omega), \tag{3}$$

where n, k_B, α and V are the number density, absolute temperature, Boltzmann constant, thermal expansion coefficient and volume of the system, respectively. Noting the orders of the limits in Eqs. (2) and (3), it is seen that thermal conductivity is related to the elastic scattering law i.e.,

$$\frac{n^2 k_B T^2 V \alpha^2}{\lambda} = \tfrac{1}{2} \lim_{k \to 0} k^2 S(k, 0) \tag{4}$$

whereas η_l is related to the inelastic scattering law. However, from Eqs. (2) and (3) one intuitively expects that there may exist a well defined relation between thermal conductivity and the longitudinal viscosity. However, in our knowledge, there exist no work which relates thermal conductivity to the longitudinal viscosity through the relations (2) and (3). On the other hand, relation between viscosity and self-diffusion coefficients, through Eqs. (1) and (2), and between thermal conductivity and diffusion constant, through Eqs. (1) and (3), can be obtained by developing the relation between $S_s(k, \omega)$ and $S(k, \omega)$. Experimentally, these two functions can be measured separately by means of incoherent and coherent inelastic neutron scattering. Since, theoretically, the models now exist that can describe $S_s(k, \omega)$ more accurately than the $S(k, \omega)$, the relation between these self and collective motion functions have been developed. The simplest connection between $S_s(k, \omega)$ and $S(k, \omega)$, known as convolution approximation, is given by

$$S(k, \omega) = S(k) S_s(k, \omega). \tag{5}$$

where $S(k)$ is the static structure factor. Eq. (5) preserve only the zeroth sum rule of the $S(k, \omega)$. The systematic derivation of such relations has been done using

Mori-memory function formalism[7,8] and the perturbation technique[10]. This provides a hierarchy of approximations relating $S(k, \omega)$ to $S_s(k, \omega)$. The first member of the hierarchy is Eq. (5) and the next two approximations are given as

$$S(k, \omega) = \frac{S(k)}{\pi} \frac{\delta_1 \delta_1^s G_s'(k, \omega)}{\left[\frac{\delta_1}{n} - nC(k)\delta_1^s \omega G_s''(k, \omega)\right]^2 + [nC(k)\delta_1^s \omega G_s'(k, \omega)]^2} \tag{6}$$

$$S(k, \omega) = \frac{S(k)}{\pi} \frac{\delta_1 \delta_1^s \delta_2 \delta_2^s G_s'(k, \omega)}{\left[\frac{A(k, \omega)}{n} - \omega B(k, \omega) G_s''(k, \omega)\right]^2 + [B(k, \omega)\omega G_s'(k, \omega)]^2} \tag{7}$$

where

$$\delta^s = \frac{k^2 k_B T}{m}$$

$$\delta_1 = \frac{k^2 k_B T}{mS(k)}$$

$$\delta_2^s = \frac{2k^2 k_B T}{m} + \omega_E^2$$

$$\delta_2 = \omega_l^2(k) - \frac{k^2 k_B T}{mS(k)}$$

$$A(k, \omega) = \delta_1 \delta_2^s + (\delta_2 - \delta_2^s)\omega^2$$

$$B(k, \omega) = \delta_1^s \delta_2 - A(k, \omega)$$

with

$$\omega_E^2 = \frac{n}{M} \int d\mathbf{r} g(r) U_{xx}$$

and

$$\omega_l^2 = 3k^2 k_B T/mS(k) + (n/m) \int d\mathbf{r} g(r)(1 - \cos(\mathbf{k}.\mathbf{r})(\hat{k}.\nabla)^2 U(r).$$

In the above equations $U_{xx} = d^2U(r)/dr_x^2$ i.e., second derivative of the pair potential $U(r)$, with respect to the x Cartesian component. The $G_s'(k, \omega)$ and $G_s''(k, \omega)$ are the real and imaginary parts of the space-time transform of the Von-Hove self correlation function. The real part $G_s'(k, \omega)$ is of course $\pi S_s(k, \omega)/n$. The last approximation (7) has been called the liquid extended phonon model. This model has been tested against the simulation results of dense gas and a good agreement[15] is achieved. The approximations (5) and (6) do not provide a relation between η_l and D, through Eqs. (1) and (2), whereas Eq. (7) gives[10]

$$\eta_l = \frac{k_B T}{a_0 D}, \tag{8}$$

where

$$a_0 = \frac{I_0}{I_1} \tag{9}$$

with

$$I_0 = \frac{m}{k_B T n}\, \omega_E^2 \tag{10}$$

and

$$\begin{aligned} I_1 &= \lim_{k \to 0} \frac{m}{k^2 k_B T}\left[\omega_l^2 - \frac{k^2 k_B T}{m S(k)}\right] \\ &= 3 - \frac{1}{S(0)} + \frac{n}{2 k_B T}\int d\mathbf{r}\, g(r) x^2 U_{xx}. \end{aligned} \tag{11}$$

The relation (8) differs from the prediction of many theories which relate the self diffusion constant to the shear viscosity alone[16]. However relation (8) is supported by some theories like that of Hubbard and Beeby[17] and by the recent approximate result of Zwanzig[18] which relates the diffusion constant to the shear viscosity through the longitudinal viscosity.

In the next section we test the validity of the relation (8) and also use it to predict the bulk viscosity of the fluid Argon and of the liquid metals at their melting points.

3 CALCULATION AND RESULTS

The calculations of the longitudinal viscosity from Eqs. (8) require the knowledge of the self diffusion constant and of the integrals I_0 and I_1. The integrals I_0 and I_1 can be obtained knowing the interatomic potential and radial distribution function. The detailed calculations for fluid Argon and liquid metals are separately given below.

3a *Fluid Argon*

The integrals appearing in Eqs. (10) and (11) are evaluated using the LJ potential and the theoretically generated[19] $g(r)$. This $g(r)$ has been found to be in good agreement with the simulation data[20]. For $S(0)$ we have used the recent theoretical results of Lomba[21]. The results obtained for I_0, I_1 and a_0 are given in Table 1. From the table it can be seen that a_0 remains almost constant with increase in temperature. With a_0 to be constant and if one assumes that η_v/η does not vary with temperature, we obtain a relation of Stokes-Einstein form, i.e.,

$$\eta = C\frac{k_b T}{D\sigma}$$

where C is a numerical constant. However, in practice η_v/η is not a constant and in the present case, it varies from 0.44 to 0.30 for the temperatures ranging from 85 K to 273 K for fluid Argon at the triple point density. Therefore, C will be a variable depending on the ratio η_v/η, as has, recently, been investigated by Zwanzig[18].

The results obtained for η_l from Eq. (8) using the simulation values of the self diffusion constant and the values of a_0 from Table 1, are also given in Table 1. The values of the self diffusion coefficients were smoothened before use, as also done by Borgelt *et al*[2]. The simulation results of $\eta_l (= 4/3\eta + \eta_v)$ of Borgelt *et al.* are also given in Table 1 for comparison. From Table 1 it can be seen that a good agreement is achieved for the whole temperature range investigated here. Since the values of the bulk viscosity are sensitive to the knowledge of shear viscosity, the values of the shear viscosity were also smoothened before subtracting it from η_l for calculating η_v. The results obtained are compared with simulation results for the bulk viscosity of Borgelt *et al.* in Table 1. From Table 1, we find that Eq. (8) predicts the η_l and η_v in agreement

Table 1 Value of I_0, I_1, a_0 and viscosities (Pa. sec) for fluid Argon at a density of 1.414 g.cm^{-3}. The MD results are from Ref. 2.

T (°K)	I_0 (10^{-8} cm)	I_1	a_0 (10^{-8} cm)	η_l (calc)	η_l (MD)	η_v (calc)	η_v (MD)
91.2	1435.6	88.7	16.18	4.34	4.44	0.95	1.06
98.2	1389.0	85.7	16.20	4.14	3.9	0.83	0.81
100.5	1374.0	84.7	16.22	4.15	4.39	0.91	0.81
102.1	1364.6	84.1	16.23	4.12	4.02	0.90	0.84
119.4	1272.4	78.1	16.29	3.88	4.02	0.77	0.77
122.5	1258.0	77.3	16.27	3.84	3.94	0.77	0.81
140.4	1185.0	72.6	16.32	3.68	3.53	0.74	0.56
156.4	1130.7	69.2	16.34	3.69	3.63	0.78	0.75
178.7	1067.1	65.4	16.32	3.48	3.67	0.68	0.66
203.3	1009.9	61.8	16.34	3.39	3.41	0.63	0.65
224.7	967.8	59.3	16.32	3.35	3.28	0.64	0.58
246.1	931.23	57.2	16.28	3.32	3.27	0.67	0.62
265.7	901.9	55.5	16.25	3.29	3.47	0.69	0.71
287.3	872.8	53.9	16.19	3.10	3.13	0.55	0.58

with the simulation results. We also find that calculated η_v decreases with increase in temperature, as expected for dense gas or liquid, in agreement with the simulation results. However, for dilute gases one expects that η and η_v should increase with increase in temperature. Therefore, it is of interest, here, to see the dilute gas limit of Eq. (8). In $n \to 0$ limit, one finds that

$$a_0 = \text{const.}$$

and

$$D \to \sqrt{T}$$

Substituting these in Eq. (8), we find that

$$\eta_l \to \sqrt{T}$$

as predicted by the kinetic theory. From the above results and the good agreement with the simulation results at the triple point density, we expect that the relation (8) is valid for the fluids of the Lennard Jones type for a wide range of densities and temperatures. In order to see the validity of the Eq. (8) for liquid metals in which the details of self and collective motions are quite different from that of the fluid Argon, we have made calculations for liquid Na and K for which experimental results of bulk viscosity are available. The results are presented below.

3b *Liquid Metals*

It is of interest to calculate the bulk and longitudinal viscosity of the liquid metals from Eq. (8), having found good results for the fluid Argon. For liquid metals we have simplified the calculations and avoided the use of the interatomic potential and radial distribution function. In order to calculate a_0, we essentially require ω_l^2 and ω_E^2. The ω_l^2 can be approximately written in terms of ω_E^2 as

$$\omega_l^2(k) = \omega_E^2\left[1 - 3\,\frac{\sin(k\sigma)}{k\sigma} - 6\,\frac{\cos(k\sigma)}{(k\sigma)^2} + 6\,\frac{\sin(k\sigma)}{(k\sigma)^3}\right], \tag{12}$$

which for $k \to 0$, provides

$$\lim_{k\to 0} \omega_l^2(k) = \frac{3}{10}\,\omega_E^2\sigma^2. \tag{13}$$

Here it may be noted that the approximation (12) has been tested against the exact results for liquid metals and provide good agreement[13,14] for all values of k. Equation (13) has also been tested for present calculation on fluid Argon and results obtained are not found to differ by more than 2 percent. Substituting Eq. (13) in Eq.

(11) and using Eq. (9) and (10), we obtain

$$a_0 = \frac{1}{\frac{nk_BT}{m\omega_E^2}\left(3 - \frac{1}{S(0)}\right) + \frac{3}{10}n\sigma^2}. \tag{14}$$

This expression is valid for all the densities and temperatures. However, at the melting point the Einstein frequency is not very different from the Debye frequency. For example the ω_D is 5.6×10^{13} sec^{-1} and ω_E calculated[14] using Shiff potential for liquid aluminium is 5.16×10^{13}. Therefore, replacing ω_E by ω_D in Eq. (14) and substituting in Eq. (8), we obtain

$$(\eta_l)_m = \frac{k_BT_m}{D_m}\left(\frac{nk_BT_m}{m\omega_D^2}\left(3 - \frac{1}{S_m(0)}\right) + \frac{3}{10}n\sigma_m^2\right) \tag{15}$$

This formula for longitudinal viscosity, at the melting point, appears to be new. The subscript m in Eq. (15) denotes the various quantities at the melting point. The expression (15) is to be compared with that obtained by Brown and March[22] which is given by

$$(\eta_l)_m \approx \frac{9\pi nk_BT_m}{10\omega_D S_m(0)}. \tag{16}$$

Equations (15) and (16) both relates the η_l to S(0) and ω_D. However, in Eq. (16) the $S(0)$ dependence appears as a result of the approximation which relates the zeroth sum rule of the stress auto-correlation function to $S(0)$. On the other hand $S(0)$ dependence in Eq. (15) appears as a result of the relation between $S_s(k, \omega)$ and $S(k, \omega)$.

In order to obtain η_l from Eq. (15) we now require only σ_m, D_m and $S(0)$. For σ_m we have used

$$\sigma_m = \left(\frac{6v_m}{\pi n}\right)^{1/3}, \tag{17}$$

where v_m is the packing fraction taken to be 0.472. For $S(0)$ we use the experimental isothermal compressibility (K_T) data and the results of density fluctuation theory

$$S(0) = nk_BTK_T \tag{18}$$

The longitudinal viscosity calculated from Eqs. (15) and (16) using experimental results of D_m for metals Na, K and Al are given in Table 2. The results obtained for liquid Na and Al of longitudinal viscosity by Bansal[13,14] are also given for comparison. It is found that the theoretical results obtained from Eqs. (15), (16) and by Bansal are of the same order for the metals investigated here. It can be seen from the table that the values of the η_l are not reliable for liquid Na and K as these are of

Table 2 The values of the viscosities (in 10^{-3} Poise) for liquid metals at their melting points.

	η_l *Eqn. (15)*	η_l *Eqn. (16)*	η_l *(Ref. 13, 14)*	η	η_v *(Expt.)*
Na	6.91	6.29	4.01	6.9	16.5
K	4.80	5.87	—	5.4	13.1
Al	24.85	20.96	27.52	11.0	—

Experimental values of η_v are taken from Kim *et al.* (1971).

the order of the shear viscosity. On the other hand, result obtained for liquid Al is reasonable and it provides the ratio $\eta_v/\eta = 0.92$. Overall, we find that the relation (8), valid for fluid argon does not provide acceptable results for the liquid metals. This may be due to the fact that the dynamics in liquid metals are quite different from that in fluid Argon, particularly in small k region.

4 RELATION BETWEEN λ AND D

The relation between thermal conductivity and self diffusion constant can be obtained using Eqs. (4) and (1) and exploiting the relations of $S_s(k, \omega)$ and $S(k, \omega)$ given by Eqs. (5)–(7). We find that Eqs. (5) and (6) provide a relation between λ and D, whereas Eq. (7) does not. The Eq. (6), satisfying the second sum rule of the $S(k, \omega)$ in ω tending to zero limit gives

$$S(k, 0) = (S(k))^2 S_s(k, 0). \tag{19}$$

In the hydrodynamic limit $S_s(k, \omega)$ is governed by the diffusion equation and one finds

$$S_s(k, \omega) = \frac{1}{\pi} \frac{Dk^2}{\omega^2 + (Dk^2)^2}. \tag{20}$$

Substituting Eq. (20) in Eq. (19) and the result in Eq. (4), we obtain

$$\lambda = \frac{2\pi n^2 k_B T^2 \alpha^2 V D}{(S(0))^2}. \tag{21}$$

Using $\alpha^2 = K_T(C_p - C_v)/TV$ and Eq. (18), we obtain

$$\lambda = \frac{2\pi n(C_p - C_v)D}{S(0)}. \tag{22}$$

The above relation, relating thermal conductivity to diffusion constant, appears to be new.

In order to check the validity of Eq. (22), we have calculated thermal conductivity of fluid Argon at its triple point using the value of C_p, C_v, $S(0)$ and D given by Egelstaff[23]. We find that our result for thermal conductivity is 2.85×10^{-4} in comparison to the experimental result 2.9×10^{-4} (in units of cal $\sec^{-1}$ K^{-1}). This agreement is remarkably good. However, for liquid metals it is known that heat conduction takes place due to electron and ion transport. The electronic contribution λ_e to λ has been found to be dominant and deviation from λ is quantitatively very small. However, if we calculate λ_{ion} from Eq. (22) for liquid Na at its melting point we find that it is 0.00026 in comparison to value 0.00021 (in same units as above) calculated theoretically by Kitajima[24] given in the book by Shimoji. From above results we find that Eq. (22) is reasonably a good approximation. However, it still requires further investigations.

5 CONCLUDING REMARKS

In this paper we have used the relation between longitudinal viscosity and the self-diffusion coefficient to predict the bulk viscosity of the fluid Argon and liquid metals. We find that the relation (8) successfully predicts the bulk viscosity of fluid Argon in agreement with the simulation results. However, for liquid metals the relation (8) is found to be invalid. This may be attributed to the difference in the dynamics of the fluid Argon and liquid metals.

We have also suggested the relation between thermal conductivity and self diffusion constant through the relation between $S_s(k, \omega)$ and $S(k, \omega)$. Therman conductivity obtained from the relation (22) for liquid Argon and liquid Na at their melting points are found to be very good.

In conclusion we find that relation (8) and (22) can be used to predict the viscosity and thermal conductivity of the fluids of the LJ types. Recalling that the sixth sum rule of $S(k, \omega)$ was used[25,26] in predicting the collective density excitations in liquid metals, we expect that the next approximation relating dynamical structure factors which satisfy the sixth sum rule may provide good results for the bulk viscosity of the liquid metals. Here it may be mentioned that theory for the bulk viscosity, similar to the other transport coefficients[27], can be developed which does not require the knowledge of the shear viscosity. The work is in progress[28] and will be applied to the liquid Argon and liquid metals.

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