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### Structural, Thermodynamic and Transport Properties of Liquid Noble and Transition Metals

G. M. Bhuiyan<sup>a</sup>; A. Rahman<sup>a</sup>; M. A. Khaleque<sup>a</sup>; R. I. M. A. Rashid<sup>a</sup>; S. M. Mujibur Rahman<sup>b</sup>

<sup>a</sup> Physics Department, University of Dhaka, Dhaka, Bangladesh <sup>b</sup> Physics Department, College of Science, Sultan Qaboos University, Muscat, Sultanate of Oman

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# STRUCTURAL, THERMODYNAMIC AND TRANSPORT PROPERTIES OF LIQUID NOBLE AND TRANSITION METALS

G. M. BHUIYAN<sup>a</sup>, A. RAHMAN<sup>a</sup>, M. A. KHALEQUE<sup>a</sup>,  
R. I. M. A. RASHID<sup>a</sup> and S. M. MUJIBUR RAHMAN<sup>b</sup>

<sup>a</sup> *Physics Department, University of Dhaka, Dhaka:1000, Bangladesh;*

<sup>b</sup> *Physics Department, College of Science, Sultan Qaboos University,  
P.O.Box 36, Post Code 123, Al Khod, Muscat, Sultanate of Oman*

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We have investigated a number of structural, thermodynamic and atomic transport properties of various liquid noble and transition metals. The underlying theory combines a simple form of the N-body potential and the thermodynamically self-consistent variational modified hypernetted chain (VMHNC) theory of liquid. The static structure factors calculated by using the VMHNC resemble, as expected, the hard sphere (HS) values. Consequently the HS model is used to calculate thermodynamic properties, viz. the specific heat, entropy, isothermal compressibility and the shear viscosity of liquid Ni, Cu, Ag, and Au. The results are in reasonable accord with the experimental values.

**Keywords:** N-body potentials; noble and transition metals

## 1. INTRODUCTION

The transition and noble metals play a very significant role in materials science. Consequently there have been numerous works aiming to understand the various physical and chemical properties of these metals and their alloys [1]. In this paper we have presented results of calculations for structural, thermodynamic and transport properties of liquid Ni, Cu, Ag, and Au using N-body potentials. Recently the applications of the N-body potentials [2], sometimes referred to as the embedding-atom-model (EAM) for the transition and noble metals and their liquids, have been receiving increasing attention. The

obvious reason is that the EAM potentials have successfully overcome some of the limitations of pairwise models. One prominent example is the Cauchy relation  $C_{11}/C_{44}$ , which is correctly reproduced by the EAM against a value of 1 as incorrectly reproduced by the pairwise models. The two popular EAMs, one by Finnis and Sinclair (FS) [3] and the other by Daw and Baskes [4] are proposed independently from two philosophical points of views; but both of them have included the many-body term described by an embedding energy function and a repulsive pair potential term in the effective interaction.

It is relevant to mention here that even though the N-body formalism is based on a specific physical background, there is still a degree of flexibility to choose suitable forms for the embedding function and for the pairwise repulsive potentials [5, 6]. Taking this into account the FS model has been further developed by various authors [5, 7–10] to extract more realistic potentials for the noble and transition metals. Among these the potentials derived by Ackland *et al.* [5] are simple in form and therefore easier to handle numerically. In addition, these potentials have also been proved to be successful in describing some physical properties of a variety of solid and liquid metals using both molecular dynamics (MD) simulation [11] and integral equation theory [12]. As far as we know no one has applied these potentials [5] to the study of static structure factors  $S(q)$  for liquid Ni, Cu, Ag and Au using the sophisticated thermodynamically self-consistent variational modified hypernetted chain (VMHNC) theory of liquids. Likewise the combination of these potentials and the VMHNC theory has not yet been employed to investigate thermodynamic properties; namely the specific heat, entropy and isothermal compressibilities of these liquid metals.

It is well known that thermodynamic quantities are the average properties of metallic systems and consequently a good degree of consistency with experiments may be expected even with a less accurate form of interacting potential; this happens due to a mode of cancellation of errors at different stages of calculations. On the contrary, since transport properties are not global in nature, only a correct theoretical interpretation of their values would allow us to draw a more accurate conclusion about the interionic interaction in liquid metals. There has been rarely any attempt to study atomic

transport properties, and in particular the shear viscosity of liquid Ni, Cu, Ag, and Au using the N-body potentials. It is needless to mention here that the study of viscosity is important for various metallurgical and industrial purposes. For example, the rates of many industrially viable chemical reactions are controlled by the diffusion of the reaction species. Following these aspects and prospects, it has become tempting to calculate the shear viscosity of these liquid metals.

A careful analysis of the experimental values of liquid structure for transition and noble metals shows that they behave as hard sphere (HS) liquids [13]. In addition, there is also evidence that the HS model describes certain thermodynamic properties of liquid noble and transition metals very well [14–16]. Inspired by the above success of the HS model, we have used it as a first approximation in the calculation of thermodynamic and transport properties. The additional advantage with the HS system is that analytical expressions for shear viscosity and the relevant thermodynamic quantities have long been established using statistical mechanics [15,17]. We have compared the HS results for  $S(q)$  with that of the more sophisticated VMHNC theory and we have found good agreement; this in turn reflects the accuracy of the HS calculation.

The layout of the paper is as follows: In Section 2, the N-body interatomic interaction, in particular the Ackland *et al.* version of the FS potential, is described briefly. The VMHNC integral equation theory and the linearized Weeks–Chandler–Andersen (LWCA) theory used in this paper are also outlined briefly in this section. The theory of shear viscosity is briefly presented in Section 3. Section 4 is devoted to the presentation and discussion of results. Some concluding remarks are presented in Section 5.

## 2. FORMALISM

In this section we briefly describe the underlying theory concerning the effective interactions, the variational modified hypernetted chain (VMHNC) theory of liquids and the linearized Weeks–Chandler–Andersen (LWCA) theory [18].

### A. Effective Interionic Potentials

Following the N-body formalism, the total energy of an atom in a solid may be written as [3]

$$U_{\text{tot}} = \frac{1}{2} \sum'_{i,j} V_{ij} - \sum_i f(\varrho_i)$$

where  $V$  is the core-core repulsive pair interaction and  $f(\varrho)$  an embedding function describing the many-body interaction. In the spirit of the second moment calculation of density of states  $f(\varrho)$  may be written as:

$$F(\varrho) = \sqrt{\varrho} \quad (2)$$

with

$$\varrho_i = \sum'_j \phi(r_{ij}) \quad (3)$$

Now if a pair potential function, namely

$$U(r) = V(r) - 2f(\varrho)\phi(r) \quad (4)$$

is assumed such that

$$U_{\text{tot}} = \frac{1}{2} \sum'_{i,j} u(r_{ij}) \quad (5)$$

then it can be shown that Eqs. (1) and (5) give exactly the same expression for the bulk and shear moduli when the second and higher order derivatives of  $f(\varrho)$  are neglected. Equation (4) thus may be approximated as an effective pair potential for the noble and transition metals. Now one must specify the functional forms of  $V(r)$  and  $\phi(r)$  for practical calculation. Within the N-body formalism there are various forms of  $V(r)$  and  $\phi(r)$  functions. The simple and computationally convenient forms of these functions are [5]

$$V(r) = \sum_K a_k (r_k - r)^3 H(r_k - r) \quad (6)$$

$$\phi(r) = \sum_K A_k (R_k - r)^3 H(R_k - r) \quad (7)$$

where

$$\begin{aligned} H(x) &= 0 & \text{for } x < 0 \\ &= 1 & \text{for } x > 0 \end{aligned}$$

In Eqs. (6) and (7),  $r_k$  and  $R_k$  are chosen knot points of cubical spline such that  $r_1 > r_2 > r_3 > r_4 > r_5 > r_6$  and  $R_1 > R_2$ . The values of  $r_1$  and  $R_1$  represent the cut-off radii of  $V$  and  $\phi$  fitting exactly to the equilibrium lattice parameter, cohesive energy, elastic constants, vacancy formation energy and stacking fault energy. Only  $a_6$  has been adjusted so as to fit the pressure-volume relation [5]. Fitted values, in these calculations, are also taken from this reference.

## B. The VMHNC Theory of Liquid

The variational modified hepernetted chain (VMHNC) theory, originally proposed by Rosenfeld [19], belongs to a new generation of fairly accurate integral equation theory of liquid. The applicability and accuracy of this theory in the calculation of liquid metals are discussed and tested else where. Like most other integral equation theories [12, 20, 21] the VMHNC solves the OrnsteinZernike (OZ) equation with a closure relation.

$$g(r) = \exp[h(r) - c(r) - \beta u(r) - B(r)] \quad (8)$$

The bridge function,  $B(r)$ , is approximated by the use of the analytic solution of the Percus–Yevick (PY) equation for hard sphere, namely  $B(r) = B_{PY}^{HS}(r; y)$ , where the packing fraction  $y$  is the variational parameter;  $y$  is determined by minimizing the VMHNC configurational Helmholtz free energy  $f^{\text{VMHNC}}(T, n; y)$  [22] at a temperature  $T$  and number density  $n$ . Once  $y$  is known for a particular thermodynamic state,  $g(r)$  and the corresponding Fourier transform  $S(q)$ , the static structure factor, are evaluated.

## C. The LWCA Theory

The starting point of the linearized Weeks–Chandler–Andersen theory (LWCA) proposed by Meyer *et al.* [23] is the Weeks–Chandler–Andersen (WCA) thermodynamic perturbation theory. In

the WCA the blip function is defined as:

$$B(r) = Y_\sigma(r) \{ \exp[-\beta u(r)] - \exp[-\beta u_\sigma(r)] \} \quad (9)$$

where  $u(r)$  and  $U_\sigma(r)$  are the soft sphere and hard sphere potentials respectively,  $\beta = (k_B T)^{-1}$ , and  $Y_\sigma(r)$  is the cavity function associated with the hard sphere distribution function. The function  $r^2 B(r)$ , if plotted as a function of  $r$ , gives two sharp tooth-shaped features [23]. In the LWCA approximation this has been approximated by right angle triangles. The Fourier transform of  $B(r)$  is then expanded in terms of Bessel's functions. Thus the thermodynamic condition, that for an approximate hard sphere diameter  $B(q)$  should vanish, leads to a transcendental Eq. [23]

$$\beta u(\sigma) = \ln \{ (-2\beta\sigma u'(\sigma) + Y + 2) / (-\beta\sigma u'(\sigma) + Y + 2) \} \quad (10)$$

The solution of Eq. (10) gives the correct value of the hard sphere diameter; this is determined from a graphical solution.

### 3. THEORY OF SHEAR VISCOSITY

If the liquid metals are approximated as hard sphere liquids, the shear viscosity may be expressed in terms of the effective pair potential,  $u(r)$ , the pair distribution function,  $g_{HS}(r)$  and the soft sphere friction coefficient,  $\xi_s$ , in a very simple form [24]

$$\eta = \eta_h + \eta_2 + \eta_k \quad (11)$$

The contribution to the shear viscosity due to the transfer of momentum occurring during the hard core collision is

$$\eta_h = \eta_h^{(1)} + \eta_h^{(2)}$$

where

$$\eta_h^{(1)} = \frac{k_B T y}{2g_{HS}(y)} \left[ 1 + \frac{8y}{5} g_{HS}(y) \right] D_y \quad (13)$$

and

$$\eta_h^{(2)} = \frac{96y^2 g_{HS}(y) k_B T}{5\pi \Omega} \quad (14)$$

with

$$D_y = \left[ \Omega + \frac{5\xi_s}{8nMg_{HS}} \right]^{-1} \left[ 1 + \frac{4\Omega}{4\Omega + 5\xi_s/(nMg_{HS})} \right] \quad (15)$$

$M$  being the atomic mass.

In the above equations

$$\Omega = \left( \frac{4\pi k_B T}{M} \right)^{1/2} \left( \frac{6y}{\pi n} \right)^{2/3} \quad (16)$$

and  $y$  denotes the packing fraction,  $y = (\pi/6)n\sigma^3$ . The contribution of the soft part of the potential to the viscosity is

$$\eta_s = \frac{4\pi Mn^2}{30\xi_s} \int_0^\alpha r^4 \left( \frac{d^2u}{dr^2} + \frac{4}{r} \frac{du}{dr} \right) g_{HS}(r) dr \quad (17)$$

For the soft core friction coefficient, we have used the well known small step diffusion formula [17]

$$\xi_s^2 = \frac{4\pi Mn}{3} \int_{r > (6y/\pi n)^{1/3}}^\alpha r^2 \left( \frac{d^2u}{dr^2} + \frac{2}{r} \frac{du}{dr} \right) g_{HS}(r) dr \quad (18)$$

The kinetic energy contribution to the shear viscosity reads

$$\eta_K = \frac{5k_B T [1 + 8yg_{HS}(y)/5]}{8g_{HS}(y)[\Omega + 5\xi_s/4nMg_{HS}(y)]} \quad (19)$$

#### 4. RESULTS AND DISCUSSION

Results of calculations for static structure factors,  $S(q)$ , of liquid Ni, Cu, Ag, and Au near the respective melting temperatures are presented in Figure 1.  $S(q)$ s are obtained by using both the VMHNC theory of liquid and the LWCA. The VMHNC calculations are carried out using Gillan's algorithm [25]. In all cases we used a 1024 point grid with step size  $\Delta r = 0.06 \text{ \AA}$  from the figure it appears that the VMHNC results overestimate the values of the principal peaks when compared with the experimental data. But the positions of all principal peaks are



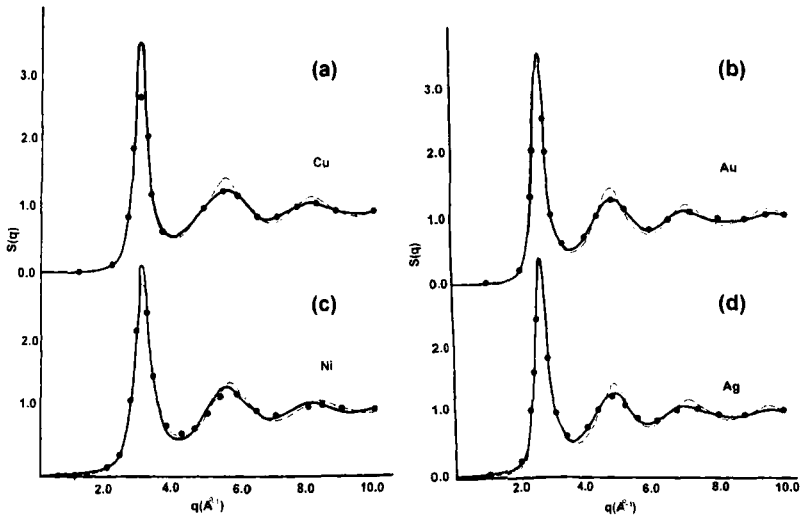


FIGURE 1 The structure factors  $S(q)$  for liquid (a) Cu, (b) Au, (c) Ni and (d) Ag. The solid line represents the VMHNC calculations, the broken line the HS-LWCA calculations and the circles the experimental values. The melting point densities are obtained from Brandes [27].

predicted correctly for all the systems. Except for the higher values at principal peaks, the general agreement with experiment is good both in terms of positions and heights of the other peaks of oscillations. The overestimated results at the first peak may arise from the potentials or from the liquid state theory used. The accuracy of the VMHNC calculations for liquid transition and noble metals has been tested elsewhere [19, 20] and was found to be comparable with molecular dynamics simulation data. It is well known that the position and height of the principal peak result from a delicate balance between the repulsive and attractive parts of the interatomic interactions (see Fig. 2). Thus it appears that the relevant potentials are responsible for the discrepancies. It is worth noting that similar discrepancies for liquid transition and noble metals have been observed for other EAM potentials [6, 20].

Results for the static structure factor,  $S(q)$ , calculated using the LWCA theory are also presented in Figure 1. In this case we have used the well established hard sphere reference system. From the figure it appears that the principal peak values for liquid Ni, Cu, and Ag are

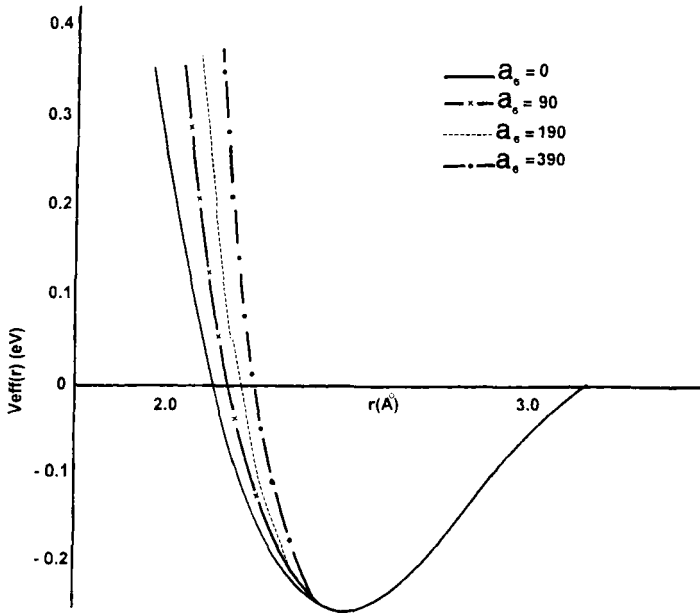


FIGURE 2 The effective pair potential  $V_{\text{eff}}(r)$  (Eq. 6) calculated at various values of  $a_0$ . Fitted values have been taken from Ackland *et al.* [5].

slightly lower than the corresponding VMHNC results implying a better agreement with experiment. For Au, the VMHNC and LWCA predict approximately the same peak value for  $S(q)$ . Beyond the principal peak *i.e.*, at the level of the second and third peaks, LWCA values are somewhat overestimated for all systems, but the positions of the peak remain unchanged. However, the overall agreement with corresponding experimental data is reasonably good. From Figure 1 it is also seen that the discrepancy between the LWCA and the VMHNC results is not significant. This indicates that the accuracy of the LWCA theory is comparable with that of the VMHNC.

The thermodynamic results of calculations for specific heat at constant pressure,  $C_p$ , entropy per atom,  $S/Nk_B$ , isothermal compressibility,  $\chi_T$ , are presented in Table I. The expressions for the thermodynamic properties are taken from Tamaki and Waseda [15]. From the table it appears that the values of  $C_p$  near the melting temperatures are in good agreement with the corresponding

TABLE I Calculated and experimental values of specific heat at constant pressure,  $C_p$  (Cal/mol.K), entropy per atom,  $S/Nk$  (Cal/K) and isothermal compressibility,  $\chi_T$  ( $10^{12}$  dyne $^{-1}$  cm $^2$ )

System	$T(K)$	$C_p$		$S/Nk$		$\chi_T$	
		Theory	Expt.	Theory	Expt.	Theory	Expt.
Ni	1773	8.34	10.30	9.80	11.72	1.0	1.03
Cu	1422	8.98	7.50	8.92	10.26	0.99	1.49
Ag	1273	9.08	8.00	9.81	10.94	1.55	2.11
Au	1423	9.29	7.40	10.63	12.00	1.26	----

experimental data. Results for Cu, Ag and Au are somewhat higher. For Ni,  $C_p$  is found lower and the discrepancy between the theory and experimental data is quite noticeable. A comparison of the calculated values of entropy with the experimental data also shows a good agreement. In this case the calculated values are somewhat lower than the experimental data for all systems. We have found that the Helmholtz free energy calculated using the Carnahan–Starling formula [17] gives 1.19, 0.86, 0.86 and 1.07 eV for Ni, Cu, Ag and Au respectively. These values are almost one fourth of the corresponding experimental data [26]. It is well known that the attractive part of the interionic interaction provides the major contribution to the cohesive energy and consequently to the free energy. Since we have not taken account of the attractive part of the potential and neglected the contribution of electronic entropy to the free energy, calculated values are somewhat low.

Since the specific heat and entropy depend on the derivative of the free energy, and these are found to be in good agreement, one can conclude that the Carnahan–Starling formula predicts, at least qualitatively, the correct features of  $F$  vs  $T$  curve as suggested by experiments. From Table I it is also seen that the calculated results for isothermal compressibility agree reasonably well with available experimental data.

We now turn to the results of calculation for the transport properties, namely, the viscosity. The calculated values for Ni, Cu, Ag and Au are presented in Table II; these values are here compared with the corresponding experimental data [27]. The table shows good agreement with experiment for all the systems except for Ni, for which the agreement is only fair. The possible causes of discrepancy in the case of Ni will be discussed later. From Table II it is noticed that the

TABLE II Calculated and experimental values of shear viscosities  $\eta(cP)$ . Values within the parentheses are obtained from our parametrization of potential for Ni

<i>System</i>	$\eta_h^{(1)}$	$\eta_h^{(2)}$	$\eta_s$	$\eta_k$	$\eta(\text{theo})$	$\eta(\text{expt})$
Ni	0.0543 (0.0499)	1.713 (2.317)	1.571 (2.480)	0.0710 (0.0600)	3.41 (4.9)	4.9
Cu	0.0547	1.884	1.417	0.067	3.421	4.0
Ag	0.0487	1.859	1.592	0.0596	3.560	3.88
Au	0.0749	2.847	2.179	0.0904	5.186	5.0

major contribution to the viscosity arises from the hard core part of the interaction. The contribution from the soft part of the potential is also significant and comparable to contribution to that of the hard core contribution. The kinetic energy contribution to the viscosity is very small relative to the other two. It is worth noting that in the case of simple liquid metals the soft part contribution to the potential was found to be predominant [13, 17, 28]. Itami and Sugimura [15] in their work for liquid Sn, used the Stoke–Einstein (SE) relation to study the viscosity with the slip boundary condition. They found good agreement with viscosity measured in the space laboratory [29]. We have found that the application of the SE formula including the back scattering factor as suggested by IS gives values of 0.44, 0.625, 0.678 and 1.193 ( $cP$ ) of  $\eta$  for Ni, Cu, Ag and Au respectively. These values are much smaller than the experimental values. Note that since there is no simple way to determine the back scattering effect theoretically, we have not included this in the present calculation of viscosity.

In this paper we have also reported on the temperature dependence of the shear viscosity. To do this accurately, within the present formalism, we indeed need a temperature dependent potential. In our case, the potentials are assumed to be temperature independent. So an exact theoretical approach is not possible with the present N-body potentials. Protopapas *et al.* [30] in their original work on transport properties of liquid metals proposed an empirical formula to evaluate the temperature dependence where hard sphere diameter (HSD) at melting has been assumed to be 0.472 for all liquid metals investigated. Itami and Sugimura (IS) [28] in their work proposed another HSD. They showed that the use of their prescription with the Stoke–Einstein relation for viscosity predicts the exact trends of shear viscosity *versus* temperature curve as suggested by the experiment done in the space laboratory. We note that IS, also chose the HSD at

melting temperature and suggested that the packing fraction should be determined theoretically using the WCA type of method. Following the suggestion of IS, we have first determined the effective HSD near the melting temperature using the LWCA method and then followed the prescription of Protopapas *et al.* (PAP) [30]. Therefore we believe that our approach is theoretically more reliable than the PAP and IS approaches independently.

The temperature dependence of the HSD has been illustrated in Figure 3. From the figure it appears that in both cases the HSD decreases linearly with temperature except near the melting temperature. In the PAP the approach the HSD increases non-linearly when one moves from the higher temperature to the melting point. However, beyond the melting temperature, the PAP and IS approaches maintain a constant difference and the IS gives higher values. The variation of density with temperature used in the calculations is determined by the formula  $n = n_0 + (T - T_0)dn/dT$  [27]. The temperature dependence of the

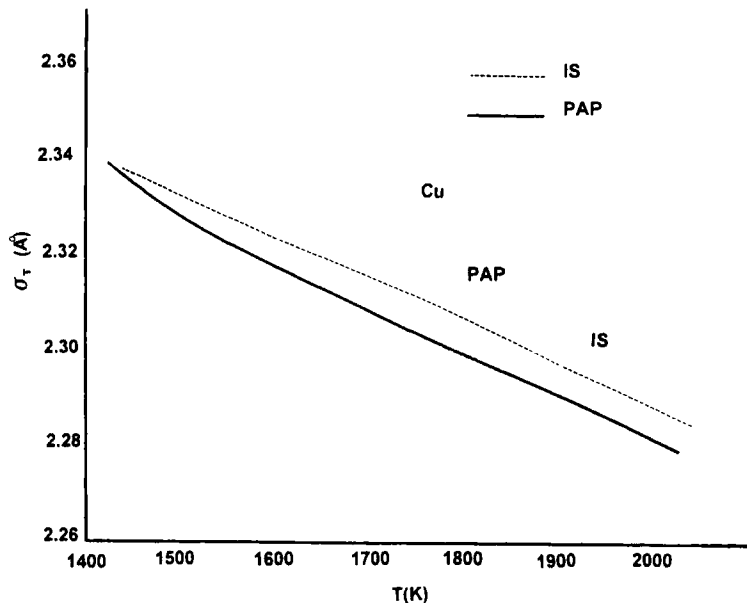


FIGURE 3 The temperature variation of the hard sphere diameter  $\sigma_T$  Å for liquid Cu. The solid line is based on Protopapas-Anderson-Parlee [30] method and the broken line on the Itami-Sugimura [28] prescription.

shear viscosity is illustrated in Figure 4. It is seen that the shear viscosity decreases linearly with temperature. Note that for Cu and Ag values of  $\eta(T)$  are very close to each other. So the IS results have not been shown in the figure. Unfortunately, high temperature experimental data for  $\eta$  are not available to us to compare our results with. Rahman and Bhuiyan [31] have studied the temperature dependence of shear viscosity for liquid Cu using the Gibbs–Bogoliubov variational scheme and potentials derived from the pseudopotential theory. Our results at different temperature are found higher than those predicted by Rahman and Bhuiyan [31]. But our results in the vicinity of the melting temperatures appear to be better with the corresponding available experimental data.

Ackland *et al.* [5] in their paper note that the potential for Ni is not very reliable: this happens presumably due to the incomplete parametrization because of the nonavailability of high pressure-

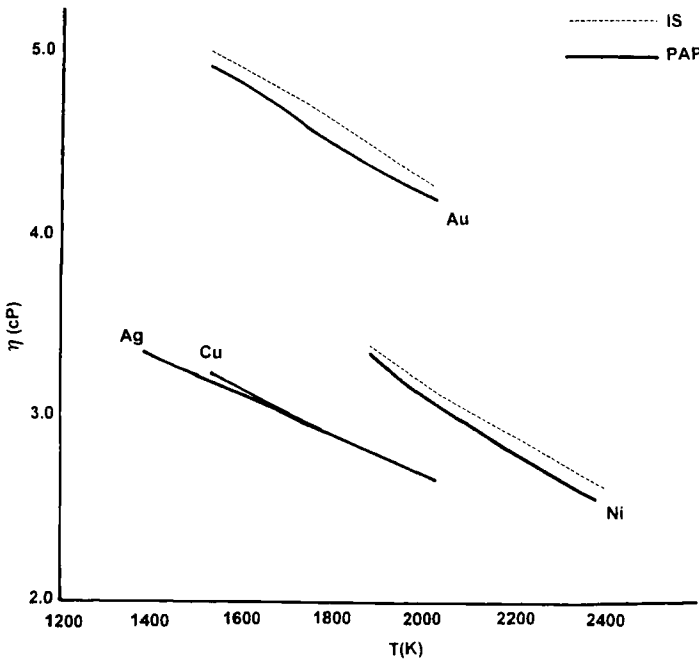


FIGURE 4 The temperature dependence of the shear viscosity  $\eta(cP)$  for liquid Cu, Au, Ag and Ni. The solid line is based on the Protopapas–Andersen–Parlee [30] method and the broken line on the Itami–Sugimura [28] prescriptions.

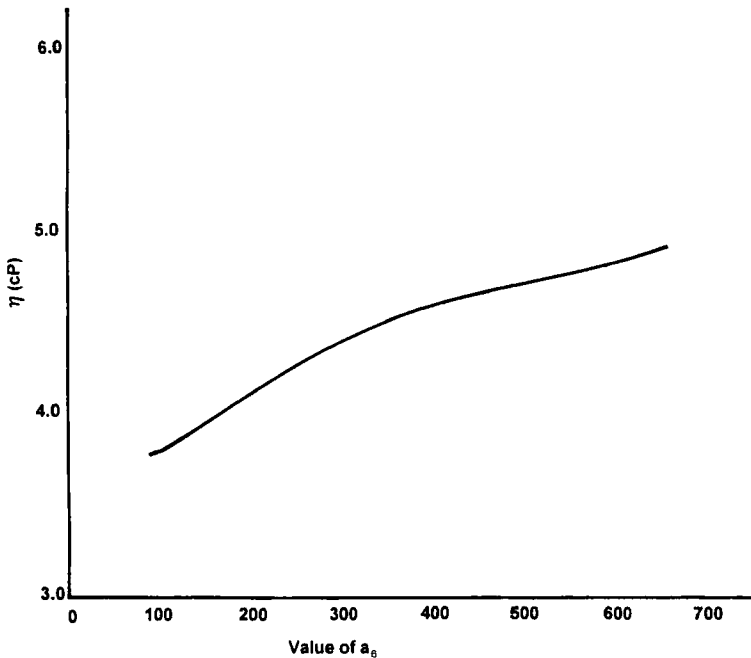


FIGURE 5 The shear viscosity  $\eta(cP)$  as a function of the parameter  $a_6$  for liquid Ni.

volume experimental data. This shortcoming is reflected in our calculation when we look particularly at the results of specific heat and viscosity for Ni. So the N-body potential for Ni should be improved for subsequent work. To this end we have first plotted the shear viscosity as a function of parameter  $a_6$  (see Fig. 5). From the figure it is seen that for  $a_6 = 620.32$  we can predict the experimental value of shear viscosity. With this value of  $a_6$  we also found that the value of  $C_p$  agrees better with experiment. This suggests that the systems for which high pressure-volume dependent data are not available, our approach of fitting  $a_6$  may be followed at least for the calculations of transport properties.

## 5. CONCLUSIONS

In this report we have presented the results of calculations for the static structure factor  $S(q)$ , thermodynamic properties *e.g.*,  $C_p$ ,  $S$ ,  $\chi_T$

and a transport property, namely, the shear viscosity. Reasonable agreement with experimental data has been found for all systems except Ni. The reason for the discrepancy in the case of Ni is attributed to the incomplete parametrization of  $a_6$ . We have prescribed a new approach to determine  $a_6$ , which essentially fits to the shear viscosity of liquid metals near melting. With the fitted values  $a_6$ , we found that the value of  $C_p$  also improves. For further justification of the parametrization one should investigate other transport properties, such as thermal conductivity *etc.* Results for the Helmholtz free energy suggest that the Carnahan–Starling formula is not adequate for quantitative results and consequently one must include the contributions arising from the attractive part of the potential and the electronic entropy for this purpose.

The Stoke–Einstein relation for viscosity is not adequate for the liquid transition and noble metals although it successfully works for liquid simple metals. The temperature dependence of viscosity has been studied using empirical formulas [28, 30]. For a better description, one should do it theoretically using a self-consistent basis. The Daw and Baskes (DB) formalism [4] for N-body potentials includes the electron density in the embedding function. As electron density depends on thermodynamic states the potentials will be somewhat temperature dependent. Therefore, temperature dependence of the HS diameter and consequently the shear viscosity may be studied from the DB model using a purely theoretical approach. This work is in progress and will be reported on completion. Finally the results of calculations for the static structure, thermodynamic and transport properties indicate that the Ackland *et al.* potential [5] has the correct nature to represent real liquid metals.

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