

REVIEW

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Atomic transport and surface properties of some simple liquid metal using one component plasma system

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Abstract

In the present paper, we have calculated diffusion coefficient, viscosity coefficient, and surface tension of liquid metals near melting point (Li, Na, K, Rb, Cs, Mg, Al, Pb, and Bi). We have applied our newly constructed model potential to describe electron ion interaction with one component plasma reference system. We have also investigated the effect of different correction functions such as those of Hartree, Hubbard and Sham, Vashista and Singwi, Taylor, Ichimaru and Utsumi, Farid et al., and Sarkar et al. on the above-said properties. It is observed that the present results are found to be in good agreement with those of experimental data as well as with other theoretical results.

Keywords: Diffusion coefficient, Viscosity coefficient, Surface tension, Liquid metal

Review

Introduction

Transport coefficients such as diffusion and viscosity coefficients of liquids are important quantities both from the scientific and technological points of view. For example, knowledge of liquid-state diffusion coefficient is required to understand important material processing techniques. A binary solidification is controlled by the diffusion of reactant species. Experimental measurement of the self-diffusion coefficient of liquid is a difficult task because of the high sensitivity of the phenomenon to external perturbations such as convection effects. The knowledge of viscosity is required in determining the critical cooling rates for glass formation. On the other hand, it is not fully understood how they depend on the structure and the thermodynamics of liquids as well. Finding a relationship between the transport coefficient and structural properties remains one of the most challenging tasks in the field of condensed matter [1-7]. Some attempts have been made since the late 1970s to find relationships between transport and structural properties. Initially, it has been realized for quite different

interaction potentials that the reduced transport coefficient is linked to the reduced entropy by simple expressions using macroscopic reduction parameters such as temperature and density. A similar scaling law which assumes the proportionality between the reduced diffusion coefficient, viscosity coefficient, and excess entropy has been obtained [8,9] using microscopic reduction parameters. The surface properties of liquids have attracted much more interest from both physicists and metallurgists from the long past to the present day because of either scientific or technological importance [10-28]. During soldering, brazing, sintering, and dying, the knowledge of surface properties of the liquid state is required. Although there are a large number of methods to determine the surface properties of liquids such as surface tension, those suitable for liquid metals are limited by their chemical reactivity and high melting points. On the other hand, it is not fully understood how the surface properties depend on the structure and the thermodynamics of the liquids [10].

The surface properties have been studied using different theories, such as the statistical mechanical theory [11,12], density functional theory [13] and computer simulation [15]. The statistical mechanical theory involving surface tension which goes back to Fowler was analyzed and improved by different

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Table 1 Input parameter used in the present study

Metal	Z	T (K) [51]	Density n (gm/cm ³) [51]	r_i (Å)	r_a (Å)	Packing fraction ζ [51]	Plasma parameter Γ
Li	1	453	0.504	0.68	1.52	0.46	210.62
Na	1	378	0.928	0.95	1.86	0.46	206.45
K	1	343	0.826	1.33	2.27	0.46	183.55
Rb	1	313	1.47	1.48	2.47	0.43	187.86
Cs	1	303	1.84	1.69	2.65	0.43	180.21
Mg	2	953	1.545	0.65	1.60	0.46	120.00
Al	3	943	2.37	0.50	1.43	0.45	100.00
Pb	4	613	10.66	1.20	1.75	0.46	85.00
Bi	3	300	10.03	0.96	1.54	0.40	100

authors [12-14]. Very recently, Gosh et al. [16] have checked the reliability of the analytical surface entropy formula derived for liquid alkali metals. Here, we have used the pseudopotential approach to describe interionic interaction and determined pair distribution function $g(r)$ from one component plasma (OCP) [29-31] structure factor $S(q)$.

An important application of pseudopotentials is the calculation of transport properties of disordered materials such as liquid, amorphous materials and their alloys. The problem with model pseudopotentials is that of their transferability because there still is no accurate method to obtain the form factor by which all the properties of liquid metals may be successfully investigated. It is found that a particular pseudopotential may be suitable for some properties of some metals and unsuitable for other properties of other metals. The usefulness of any model potential depends on how many number of parameters it involves. Generally, the potential involving less number of parameters is considered to be, comparatively,

a better one than that involving more number of parameters because it avoids more complexities in the calculation. It has been observed that a unique method of determination of the potential parameter has not been pointed out so far. A number of investigators have used a fitting procedure in which the potential parameters are fitted in such a way that a good agreement with experimental findings be obtained. Such a procedure will generally give good results for a certain property, but the same set of parameters will not give good results for other properties. Hence, we thought it worthwhile to construct a parameter free model potential which, by employing, explains the physical as well as chemical properties of condensed matter.

A pseudopotential method employing a simple model of a solid composed of atomic cores and valence electrons can predict the existence and properties of new solids and their properties [32]. By implementing this idea, we construct a new model potential which is split into three regions [31,33-37]:

Table 2 Calculated viscosity (η) (mPa/s) for simple liquid metals

Metal	Calculated viscosity (η) (mPa/s)								
	H [39]	VS [40]	HS [41,42]	S [43]	IU [44]	T [45]	F [46]	Others [22]	Experiments [48,51]
Li	0.407	0.379	0.393	0.382	0.373	0.378	0.393	0.51	0.602
Na	0.761	0.694	0.728	0.702	0.684	0.697	0.728	0.56, 0.256	0.70
K	0.539	0.495	0.518	0.501	0.488	0.499	0.518	0.46, 0.192	0.54
Rb	0.769	0.721	0.747	0.727	0.709	0.727	0.747	0.56	0.674
Cs	0.531	0.486	0.511	0.492	0.473	0.486	0.511	0.59	0.686
Mg	1.166	1.28	1.231	1.251	1.281	1.275	1.231	1.26	1.25
Al	1.403	1.382	1.392	1.383	1.378	1.378	1.392	1.62	1.2
Pb	2.615	2.734	2.213	2.772	2.33	2.487	2.213	2.51	2.61
Bi	1.663	1.55	1.605	1.556	1.498	1.495	1.605	2.34	1.63

Table 3 Calculated surface tension (γ) (N/m) for simple liquid metals

Metal	Calculated surface tension (γ) (N/m)							Experiment [48]
	H [39]	VS [40]	HS [41,42]	S [43]	IU [44]	T [45]	F [46]	
Li	0.281	0.261	0.264	0.258	0.262	0.272	0.258	0.398
Na	0.264	0.242	0.243	0.237	0.241	0.253	0.237	0.191
K	0.136	0.126	0.127	0.123	0.125	0.131	0.124	0.115
Rb	0.126	0.119	0.119	0.116	0.118	0.122	0.116	0.085
Cs	0.068	0.063	0.064	0.061	0.063	0.066	0.061	0.070
Mg	0.624	0.683	0.680	0.686	0.685	0.659	0.686	0.590
Al	0.709	0.697	0.699	0.696	0.699	0.703	0.696	0.917
Pb	0.385	0.366	0.408	0.343	0.402	0.325	0.341	0.458
Bi	0.170	0.153	0.159	0.153	0.159	0.164	0.153	0.378

$$\begin{aligned}
 W_B(r) &= 0; & r < r_i, \\
 &= -\left(\frac{Ze^2}{r_a}\right) \left\{ 2 - \exp\left(\frac{r}{r_a} - 1\right) \right\}; & r_i \leq r \leq r_a \\
 &= -\left(\frac{Ze^2}{r}\right); & r > r_a
 \end{aligned} \quad (1)$$

Basically, this form of the pseudopotential is the modified version of Ashcroft's empty core model [38]. Here, we have considered the actual core of an ion as an empty core rather than pseudizing it. The whole effective region is split into three parts. The effective weak potential felt by an electron is sandwiched between the empty core and the long-range coulomb potential which is continuous at $r = r_a$ and is considered between the ionic radius (r_i) and atomic radius (r_a). It may be noted that most of the pseudopotentials are represented always by twofold splitting in their influence. In the present potential, we have included both the features of the pseudopotential - cancellation as well as the remaining weak interaction around the ion. Another important part of the model potential is that it is parameter free. Looking at the success of the present approach, we believe that it should be interesting to try out all the local forms of the pseudopotential on the line of the present approach.

In the reciprocal space, the corresponding bare-ion form factor of the present model potential is given by the following [31,33-37]:

$$W_B(q) = \frac{-4\pi Ze^2}{q^3 r_a \Omega (1 + q^2 r_a^2)^2} \left[\begin{aligned} &2(1 + 2q^2 r_a^2) \sin(qr_a) - qr_a(1 + 3q^2 r_a^2) \cos(qr_a) + \\ &2(1 + q^2 r_a^2)^2 \{qr_i \cos(qr_i) - \sin(qr_i)\} - \\ &\exp\left(\frac{r_i}{r_a} - 1\right) \left\{ \begin{aligned} &(-2q^3 r_a^3 + q^3 r_a^2 r_i + q^5 r_a^4 r_i) \cos(qr_i) - \\ &(-q^2 r_a^2 + q^4 r_a^4 + q^4 r_a^3 r_i + q^2 r_a r_i) \sin(qr_i) \end{aligned} \right\} \end{aligned} \right] \quad (2)$$

Here, Z , e , Ω , q , r_i , and r_a are the valence, electronic charge, atomic volume, wave vector, ionic radius, and atomic radius, respectively.

The choice of the pseudopotential form factor is certainly an important factor in the study of metallic properties, and its actual form is much more sensitive to the choice of dielectric function of the electron gas. Hence, the purpose of the present paper is not only to calculate surface tension (γ), diffusion coefficient (D), and viscosity coefficient (η) of liquid metals near melting point (Li, Na, K, Rb, Cs, Mg, Al, Pb and Bi), but also to see the influence of various local-field correction functions in the screening along with the newly constructed model potential [31,33-37]. Thus, we incorporated seven different forms of the exchange and correlation functions, *viz.* Hartree (H) [39], Vashista and Singwi (VS) [40], Hubbard and Sham (HS), [41,42], Sarkar et al. (SS) [43], Ichimaru and Utsumi(IU) [44], Taylor (T) [45], and Farid et al. (F) [46]. To the best of our knowledge, only few attempts have been made on γ , D , and η from transferable pseudopotential. These properties are important quantities from the scientific as well as technological points of view. However, the accurate measurement of these quantities is exceedingly difficult for high-temperature melts such as liquid metals. Furthermore, theoretical calculations of the surface and atomic transport properties are also difficult to perform convincingly. For surface tension, it is necessary to know the ionic and electronic structures in the inhomogeneous transition zone between liquid metal and gaseous phases, the studies of which are still in their infancy in comparison with those in bulk liquids [22]. As for viscosity coefficient, Born and Green [47] have proposed a simplified expression using their kinetic theory. In spite of oversimplified assumptions involved in the derivation, this Born-Green equation has been shown to predict apparently reasonable values for the viscosity coefficient of liquid metals with the help of pair potentials extracted from experimental structural data. However, it is remarkable that these results are by no means quantitatively reliable since pair potentials deduced from the inversion method have large uncertainties [10-28].

Theory

There is a correlation between η and γ . Egly et al. examined this correlation [17,18] and derived the formula for the surface tension of a fluid, approximating the smooth density profile by a step function and replacing the gas by vacuum:

$$\gamma = \frac{\pi \rho^2}{8} \int_0^\infty r^4 \frac{du(r)}{dr} g(r) dr \quad (3)$$

where $g(r)$ is the pair distribution function and $u(r)$ is the effective pair potential. These pair potential is commonly evaluated by treating the electron-ion coupling through the use of the pseudopotential concept and of linear screening theory. The usual form for the effective ion-ion interaction potential $u(r)$ in pure liquid metal is given by the following:

$$u(r) = \left(\frac{z^2 e^2}{r} \right) + \frac{\Omega}{\pi^2} \int F(q) \left[\frac{\sin(qr)}{qr} \right] q^2 dq \quad (4)$$

where $F(q)$ is the energy wave number characteristic in Equation 4 which is written as follows:

$$F(q) = \frac{-\Omega q^2}{16 \pi} |W(q)|^2 \frac{\epsilon_H(q) - 1}{1 + [\epsilon_H(q) - 1][1 - f(q)]} \quad (5)$$

Here, $W(q)$ is the bare ion potential, $\epsilon_H(q)$ is the Hartree dielectric, and $f(q)$ is the local-field correction function to introduce the exchange and correlation effects. Born and Green [47] derived an expression for the viscosity of a fluid using the kinetic theory:

$$\eta = \frac{2\pi\rho^2}{15} \left(\frac{m}{k_B T} \right)^{\frac{1}{2}} \int_0^\infty r^4 \frac{du}{dr} g(r) dr \quad (6)$$

Using the Stokes-Einstein relation [48-50], the following is obtained:

$$\eta = \frac{k_B T}{2\pi\sigma D} \quad (7)$$

Results and discussion

The input parameter and constants used for the present study for simple liquid metals are tabulated in Table 1. The viscosity (η) of liquid metals near melting point (Li, Na, K, Rb, Cs, Mg, Al, Pb and Bi) is shown in Table 2. From Table 2, it is noticeable that the different forms of exchange and correlation functions do not have much sensible effects on the numerical values of η . The present results for η are far better than the others available in the theoretical data [22], except for liquids Li and Cs. An excellent agreement between the present results and experimental data [48,51] has been achieved due to the Hartree [39] local-field correction function for K, Pb, and Bi. The local-field correction function for Na and Mg is due to Sarkar et al. [43], whereas for Rb and Al, it is due to Ichimaru and Utsumi [44].

Table 3 shows that the present results are quite in good agreement with those of experimental data [22] for simple liquid metals. It could also be noticed that the local-field correction function does not affect the numerical values of γ . An excellent agreement has been found between the present results and those of the experimental data [48], which have been achieved due to Hartree [39]; for K, Cs, Mg, and Bi, they have been achieved due to local-field correction functions of Sarkar et al. [43], Hartree [39], Hartree [39], and Hubbard [41] and Sham [42], respectively, while for Na and Al, they have been achieved due to local-field correction functions of Sarkar et al. [43] and Hartree [39], which are reasonably in good agreement with those of experimental data [48]. There are results that do not agree well with those of experimental data [41], and they have been found for liquids Li, Rb, and Bi. Table 4 shows the calculated self-diffusion coefficient (D) of simple liquid

Table 4 Calculated self-diffusion coefficient (D) (m^2/s) for simple liquid metals

Metal	Calculated D ($m^2/s \times 10^{-9}$)								Experiments [48,51]
	H [39]	VS [40]	HS [41,42]	S [43]	IU [44]	T [45]	F [46]	Others [22]	
Li	9.00	9.69	9.59	9.81	9.66	9.32	9.81	7.33	6.80
Na	3.30	3.61	3.58	3.67	3.62	3.45	3.67	6.80, 6.40	4.23, 4.30, 4.19
K	3.41	3.68	3.67	3.77	3.71	3.55	3.76	6.60, 6.10	3.76, 3.70, 4.01
Rb	2.08	2.20	2.20	2.26	2.22	2.14	2.25	2.79	2.68
Cs	2.72	2.97	2.93	3.05	2.97	2.82	3.05	2.37	2.31
Mg	6.32	5.78	5.80	5.75	5.76	5.99	5.75	5.87	5.63
Al	5.83	5.94	5.91	5.94	5.92	5.88	5.94	5.00	4.87
Pb	1.69	1.78	1.59	1.90	1.62	2.00	1.91	1.72	1.68
Bi	1.33	1.48	1.42	1.48	1.43	1.38	1.48	1.72	-

metals. From Table 4, it is clear that the different forms of exchange and correlation functions do not have much sensible effects on the numerical values of D . The present results are quite in good agreement with other available theoretical data results [22], except for liquid Li. A good agreement has been found between the present results and those of experimental data [22,48,49] for Na, K, and Mg due to the local-field correction function of Farid et al. [46], while for liquid Pb, it is due to Hartree [39]. The present results are in poor agreement with those of liquids Li and Al.

In general, it is found in Tables 2, 3, and 4 that the overall present results match very well with the results of other theoretical data [22] as well as other experimental data [48,51] of simple liquid metals, except for Li and Rb. It could also be noticed that the local-field correction functions do not greatly affect the numerical values of atomic transport and surface properties. However, significantly better results could be obtained by the use of other local-field correction functions.

Conclusions

Good qualitative and quantitative data of atomic transport and surface properties of some liquid metals near melting point have been obtained; they would be beneficial to other theoreticians as well as experimentalists working in the same field. The present obtained results using our newly constructed parameter free model potential [31,33-37] seem to yield better results than those results obtained by other theoreticians [22]. The structure factor $S(q)$ due to the OCP reference system [29-31] is key in the explanation of atomic transport and surface properties of simple liquid metals near melting point. This shows that sensitivity is required in choosing the reference system and local-field correction function to calculate γ , D , and η of liquid metals. Thus, the application of our newly constructed model potential [31,33-37] in the present study definitely adds a new contribution to understand the atomic transport and surface properties of simple metals in liquid state near melting point.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

The three authors, YAS, PBT, and ARJ, contributed equally in all steps of the present paper. All authors read and approved the final manuscript.

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