



## Investigation of surface tension, viscosity and diffusion coefficients for liquid simple metals

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### ABSTRACT

Surface tension of some liquid simple metals has been investigated employing the hard sphere theories which were derived from the first and second order approximations of the isothermal compressibility. Atomic transport properties such as diffusion and shear viscosity are also studied for the same systems. The premier ingredients involved in the theories are effective HS diameter, the packing fraction associated with it, and the effective pair potential. For effective calculations, these ingredients are evaluated by using a local pseudopotential, variational modified hypernetted chain integral equation theory (VMHNC) and also using the linearized Weeks-Chandler and Andersen thermodynamic perturbation theory. Calculated results, when compared with the available experimental data and other theoretical values, reveal that the first order approximation theory along with VMHNC predicts much better results than those of the other theoretical values for all concerned systems except for Pb and Al. The cause of discrepancy in the results for Al and Pb is also discussed.

### 1. Introduction

Technologists, metallurgists and scientists have been attracted to calculate surface tension [1–7], shear viscosity [8, 9] and diffusion coefficients [10, 11] because of their multiple applications in industrial and academic purposes. Among them, surface tension plays a major role in different phenomena for example, nucleation of gas bubbles, gas absorption, and growth of non metallic inclusions and metal reactions [1]. There are a lot of physical properties related to material processing techniques, for example, soldering, brazing, sintering, dying, wetting, etc. [2–8, 10–13] need the knowledge of surface physics for better understanding. On the other hand, shear viscosity and diffusion coefficients can also play a key role in binary solidification, and controlling rate of chemical reactions [8, 9, 11, 13]. This indicates that, structural and thermodynamical change can play an important role to determine surface tension and hence the atomic transport properties, for example, shear viscosity and diffusion coefficients of liquid metals. Therefore, to investigate the possible interlink among the aforementioned properties is a very interesting problem. In this case, we need a theory which can best describe the liquid structure and thermodynamics of the systems under study.

Surface tension of liquid simple metals can be studied theoretically, using either statistical mechanical theory [12, 14–16], or density functional theory [17, 18], or different semi-empirical models proposed

by Skapski and Schytil [1]. Few popular experimental techniques are sessile drop method [19], levitated drop method [20, 21], oscillation drop technique [22], drop-weight method [19, 23] etc. and computer simulation method in ref. [24] are also used for this purpose. Unfortunately, experiments on the nature and behaviour of surfaces or interfaces between two different phases are generally unsatisfactory. This situation results from the difficulty of precise experimental determinations of metallic liquid's surface tensions or interfacial tensions; the main problem is that it is extremely difficult to remove all impurities from both the surface of the specimen and the atmosphere. Similarly, it is very difficult to measure transport coefficients experimentally due to the reactivity with the containing vessel and atmosphere [13, 15, 25]. Therefore, some attempts have been made to understand surface tension, shear viscosity and diffusion coefficients using different theories [1, 10, 13, 26–28] and computer simulation methods [29–34].

In the meantime, based on the statistical mechanical theory, surface tension of some liquid simple metals were studied by Evans [12] where for interionic interaction, pseudopotential was chosen but the obtained results for the concerned systems underestimated the available experimental data. On the similar ground, Fowler's formula [14] was derived and applied by Korkmaz et al. [4] to the liquid transition metals and the obtained results were consistent with the experimental data but the problem of the Fowler's formula is that, it provides us an approximate

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value since we need to cut the integrand at a certain interionic distance to avoid the divergency if the potential has a long Friedel type oscillating tail. Recently, based on the HS interaction, Egry [35] and Yokoyama [31] derived relations between surface tension and transport coefficients and applied these relations to calculate surface tension of some liquid transition and noble metals where the studied metals received no attention. Similarly, using kinetic theory, Born and Green [28] proposed a simplified theory for transport coefficients and predicted apparently reasonable values for some liquid metals. Following the work of Born and Green, very recently, Sonvane et al. [13] studied the surface and atomic transport properties for the concerned systems using various local field corrections in their pseudopotential. In refs. [13, 28, 31, 35], it is seen that, for calculating surface tension, one needs to know the values of transport coefficients which are not easy to measure experimentally [11, 13, 25]. To overcome those limitations, recently, Mayer's empirical formula [36] has been extended by Ruhul et al. [6] by using second order approximation of isothermal compressibility for evaluating the surface tension of the liquid transition metals where a good agreement with the experimental data was found [for details see ref. [6]]. Similarly, neglecting the vapour pressure, Reiss et al. [37] have also derived a formula for surface tension involving single parameter  $\eta$  (packing fraction) which also can be derived by using first order approximation of isothermal compressibility into Mayer's empirical formula [36]. Theories mentioned in refs. [6, 37] have not yet been applied to calculate surface tension of liquid simple metals.

Bretonnet and Silbert (BS) proposed a model which incorporates both  $sp$  and  $d$ -bands separately to describe interionic interactions for liquid transition metals [38]. This model was established within the well-known pseudopotential formalism and it is very simple to handle numerically. It has a form factor which is constructed by the superposition of the  $sp$  and  $d$ -band calculation. Its local form, which appears to be similar to that of the well-known Heine-Aberenkoff model [39], permits one to extend this model to other liquid metals for which the effects of hybridization are significant. The BS pseudopotential model has proven to be successful to describe the structural [38] and electron transport [40] properties of liquid less simple metals. Envisaged elements to be studied in this work lie in group 1A and 2A in the periodic table except liquids Al and Pb. The  $s$  and  $p$  bands of them are completely filled but the effects of  $s-p$  hybridization in Al [41, 42] and  $sp-d$  hybridization in Pb [40] still exist. So this effect is accounted for approximately by changing the relative occupancy of the  $s$ ,  $p$  and  $d$  bands. There is no restriction in doing this as such to use suitable values of the effective  $s$  electron occupancy number,  $Z_s$ , provided the self consistent calculation of charge transfers supports it. It should be mentioned that Sonvane et al. [13] used  $Z_s = 1$  for all elements of 1A group and 2, 3 and 4 for Mg, Al and Pb, respectively. In addition to that, Bhuiyan et al. used  $Z_s = 1.4$  to describe structure for 3d transition metals [43]. In the study of electrical resistivity, Sharmin et al. [40] used different values of  $Z_s$  for liquid less simple metals. Besides those input parameters, this model potential also can be tuned using various local field corrections such as Ichimaru-Utsumi (IU) [44], Vashishta-Singwi (VS) [45], etc. It is worth noting that BS pseudopotential model has proved to be successful for determination of surface properties of liquid less-simple metals [2, 7]. In the present work, we have chosen the BS pseudopotential model as a serious test to see whether the theories mentioned in refs. [6, 37] work for determination of both the surface and atomic transport properties of liquid simple metals.

It is well known that structure and the thermodynamics can affect both surface and atomic transport properties of liquid transition metals [11]. In this regard, approximated liquid state theories such as integral equation and perturbation theory, already proved to be successful [2, 6, 7, 43]. However, it is still difficult to determine which equation is superior. From Linearized Weeks-Chandler-Andersen (LWCA) perturbation theory, it is known that the degree of accuracy is comparable to the variational modified hypernetted chain (VMHNC) integral equation

theory in case of simple liquid metals. It is also true that the presence of  $d$ -bands in the transition metals and in their liquids near melting makes a sharp distinction between the simple and transition metal properties [46]. On the other hand, VMHNC theory can impact the effect of full effective pair potential on different physical properties. So it is natural to expect that it will predict better results.

Finally, it appears that the present study is a serious test to see which theory can best describe surface tension and also the transport coefficients of liquid simple metals namely, Li, Na, K, Rb, Cs, Mg, Ca, Al and Pb.

## 2. Theories

Theories relevant to the present study of surface tension, viscosity coefficient, and diffusion coefficient are briefly described below.

### 2.1. Surface tension

The density of liquid metals is higher than other common liquids. As a result, to explain liquid metals we need sophisticated theories. Several theories have been proposed depending on the idea that the repulsive intermolecular force determine the structure of liquid metals [47–50]. From this idea, we can say that the shape of the molecules determines the intermolecular correlations. Usually a hard sphere potential is purely repulsive. For this reason, if a hard sphere potential is used to represent the interaction of real liquid systems, it should mimic the repulsive part of the potential only [27, 51]. Under this assumption, several expressions had been developed and successfully applied to determine atomic transport [27] and surface properties [1, 6].

Based on the HS interaction, Mayer [36] proposed the following empirical relationship for the compressibility,  $\chi$  and surface tension,  $\gamma$  which is expressed as

$$\chi\gamma = \sigma \left[ \frac{(2 - 3\eta + \eta^3)}{4(1 + 2\eta)^2} \right], \quad (1)$$

where  $\sigma$  and  $\eta$  denote the effective HS diameter and packing fraction, respectively.

From this empirical relation, recently, Ruhul et al. [6] derived a formula of surface tension for liquid transition and noble metals involving single parameter,  $\sigma$  by using an expression of isothermal compressibility obtained from second order approximation of Percus-Yevick [26] solution. The formula reads as,

$$\gamma = \frac{\sigma n k_B T [2\eta(4 - \eta) + (1 - \eta)^4](2 - 3\eta + \eta^3)}{4(1 + 2\eta)^2(1 - \eta)^4}. \quad (2)$$

Here  $n$ ,  $T$  and  $k_B$  denote the number density, temperature and Boltzmann constant, respectively.

Using rational function approximation, which is an alternative to the integral equation approach, the compressibility equation obtained from first order approximation of Percus-Yevick solution can be written as,

$$\chi = \frac{(1 - \eta)^4}{n k_B T (1 + 2\eta)^2}. \quad (3)$$

Substitution of Eq. (3) into Eq. (1) provides the following formula of surface tension

$$\gamma = \frac{\sigma n k_B T (2 + \eta)}{4(1 - \eta)^2}. \quad (4)$$

Surface tension expression in Eq. (4) can also be obtained from a relation proposed by Reiss et al. [37] for real fluids which reads as,

$$\gamma = \frac{k_B T}{4\pi\sigma^2} \left[ \frac{12\eta}{1 - \eta} + \frac{18\eta^2}{(1 - \eta)^2} \right] - \frac{p\sigma}{2} \quad (5)$$

where  $p$  denotes the vapour pressure. To develop the above equation,

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