Effective Interionic Potentials and Properties of Molten Noble and Transition Metals

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Recent X-ray diffraction data on molten noble and transition metals have been used to calculate effective interionic potentials by means of the Born-Green equation. The results on the noble metals are compared with the generalized pseudopotential calculations of Moriarty. In all cases the potentions were found to be insensitive to temperature and to have long-range oscillations. The results suggest that the effective valence number is close to unity in these molten metals, this giving reasonable values of the electrical resistivity when calculated in the framework of the usual Ziman theory.

The self-diffusion coefficients, viscosities and surface tensions were calculated from the potentials and radial distribution functions using the kinetic theory of fluids. Adequate agreement with experimental data was obtained.

1. Introduction

In the calculation of various properties of molten metals[1], the knowledge of the interaction between the component ions is of primary importance. Effective interionic pair potentials may be derived directly from structural data as obtained by X-ray or neutron diffraction experiments using one of the molecular theories of fluids. In this way, effective interionic potentials of low melting molten metals have been derived using the Born-Green equation[2]. The obtained potentials have been successfully used in estimating various macroscopic properties, e. g. self-diffusion, of these molten metals. Therefore, in this paper a similar approach on molten noble and transition metals is presented.

Using the Born-Green equation, we derive the effective interionic potentials of molten Cu, Ag and Au, and of molten Fe, Co and Ni from the structural data recently reported[3] and compare them with those obtained from the electron theory of metals. In addition, the electrical conductance, self-diffusion, viscosity and surface tension of these molten metals are calculated and compared with experimentally observed values.

2. Method for Deriving Pair Potentials

The numerical calculation using the Born-Green equation was identical with the one described in a previous paper[4]. According to the Born-Green theory[5] the pair potential $\Phi(r)$ is given by the relation

$$\Phi(r) = U(r) + \frac{\pi \varrho_0}{r} \int_0^\infty \frac{\mathrm{d}\Phi(s)}{\mathrm{d}s} g(s)\,\mathrm{d}s \cdot \int_{-s}^{+s} (s^2 - t^2)(t+r)\,[g(|t+r|) - 1]\,\mathrm{d}t\,, \qquad (1)$$

where $U(r) = -k\,T \ln g(r)$. $g(r)$ is the radial distribution function obtained by diffraction experiments. A numerical solution can be obtained by a linearized simultaneous equations method[4]. In this work, the simultaneous equations were solved by a combination of the Sweep out and Gauss-Seidel method. The numerical calculations were performed on the NEAC-2200, Model-700, in the Computer Center, Tohoku University.

3. Effective Interionic Potentials

All the effective interionic potentials obtained show long-range oscillations which are relatively insensitive to temperature. Figure 1 shows the effective interionic potential in molten iron at three temperatures. Slight variations with temperature are only seen in the region of the first minimum and the first maximum. Figures 2 and 3 show the effective interionic potentials of the six molten metals as derived from the structural data near the melting points reported in[3]. With Fig. 1 in view it is supposed that they all are relatively insensitive to temperature within the observed temperature region.

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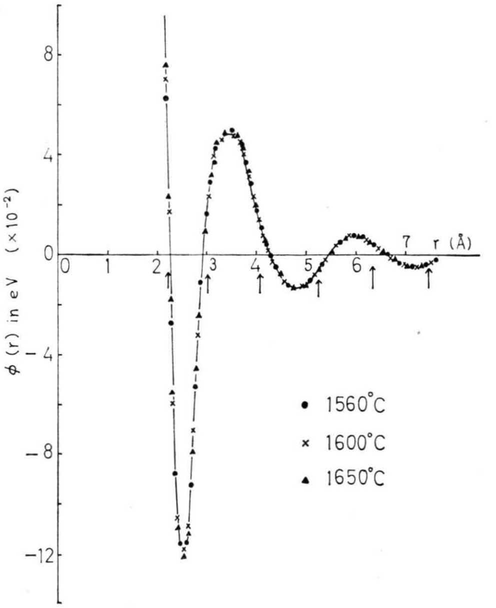

Fig. 1. Temperature dependence of the pair potential in molten iron. The arrows indicate nodes of $g(r)-1$.

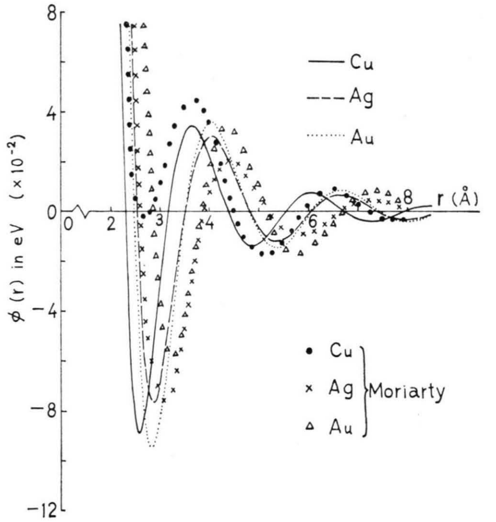

Fig. 2. Pair potentials in molten noble metals.

As reported in[2], the potentials of simple molten metals as calculated with the Born-Green equation agree fairly well with those calculated with the nearly free electron model. A similar comparison is therefore made in this work. Recently Moriarty[6] reported the effective interionic potentials of the solid noble metals calculated from the electron theory of metals in the framework of the generalized pseudopotential approximation suggested by Harrison[7]. His results are plotted in Fig. 2. The calculation of Moriarty includes assumptions for the determination of parameters such as those connected with the overlap of s − d bands and does not correctly predict the structure, i. e. according to his calculation all three metals should show the h.c.p. structure rather than the observed f.c.c. structure. If the parameters were well chosen, the potential minimum of Morialty for copper would probably be deeper, approaching that for silver and gold. Therefore it seems that the potential calculated by

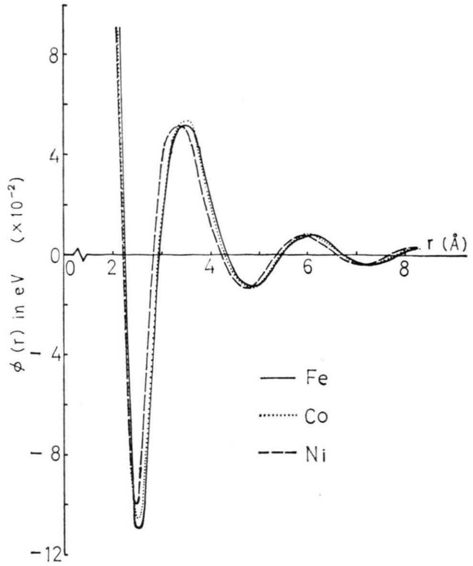

Fig. 3. Pair potentials in molten transition metals.

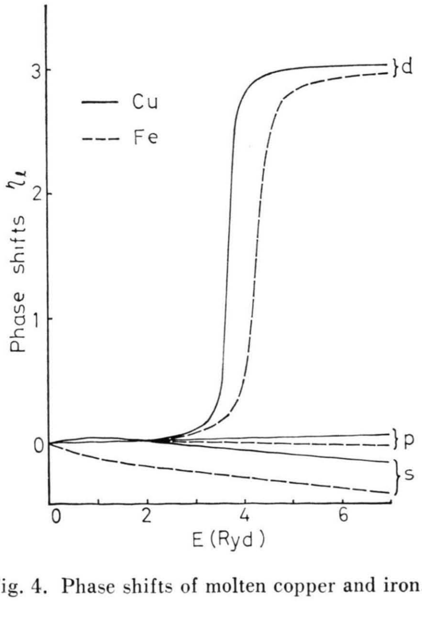

Fig. 4. Phase shifts of molten copper and iron.

Moriarty is not necessarily the best one for the noble metals. Anyhow, the potentials of Moriarty also show the long-range oscillations, and the agreement in the general is rather impressive. The effective interionic potentials obtained in this work may give a net information because they are directly derived from precise experimental data [3].As for the transition metals, so far no effective interionic potentials calculated from the electron theory of metals seem to be available.

The characteristics of the effective interionic potentials of molten noble and transition metals obtained in this work shall now be examined. The long-range form of the effective interionic potential in metals is well known to be given by

$$\Phi(r) \cong \cos(2\, k_f \cdot r)/r^3\,, \qquad (2)$$

where k_f is the Fermi wave vector. Therefore the wavelength of the oscillations in $\Phi(r)$ is expected to be $\lambda_f = \pi/k_f$. Eq. (2) is a good approximation for simple molten metals such as the alkali metals [2], but the situation for noble and transition metals is more complicated. The complicated electronic structure due to the overlap of the d-band with the s-band prevents a light-hearted application of a simple electron theory of metals. However, the potentials of Moriarty and those of this work oscillate about zero and this seems to suggest that also these potentials are influenced by a sharp cutoff of the electron distribution function at the Fermi wave vector. On the other hand, Ziman [8] suggested that the screening in these metals would be predominantly caused by fast-moving s-electrons rather than by slowly moving d-electrons which would be strongly localized around the ions and correlated among themselves. For these reasons, at first, the value of k_f was chosen under the following conditions; a) The effective valence (z) is 1.0 for noble metals, whereas it is 2.0 for transition metals in the molten state; b) $k_f = (3\pi^2 z/\Omega_0)^{1/3}$, where Ω_0 is the atomic volume. These values are shown in Table 1 together with the observed wavelength and the position of the first maximum in the radial distribution function. In Table 1, λ_{obs} agrees quite well with λ_f for the molten noble metals, but for the molten transition metals no such agreement is found. Besides, λ_{obs} differs distinctly from the interatomic distances (r_{RDF}) in both molten noble and transition metals. To illustrate this, the nodes of $g(r)-1$ are shown in Fig. 1 by the arrows.

We still have no unique way of determining the number of valence electrons for noble and transition metals in the molten state because of the overlap of the incomplete d-band with the s-band. Therefore, an empirical estimation of the effective valence number (z_{obs}) was made using the observed wavelengths of the oscillations which are given in Table 1 together with the observed values of k_f^{obs}. Recently Miller [9] has found by optical measurements that the effective density of the conduction electrons in noble metals increases slightly on melting. We find the same tendency in our results as shown in Table 1, namely some transfer of d-electrons into s-like conduction-free electrons in the case of noble metals. On the other hand, the transfer of s-electrons into the incomplete d-level can be found in the case of molten transition metals. Although these problems can only be discussed after performing some further experiments such as measurements of Hall coefficients, optical properties and so on, the effective interionic potentials obtained in this work suggest that the effective valence number is nearly equal to unity in both molten noble and transition metals.

Recently the softness of the repulsive core in the pair potential for metals was frequently discussed [10–12]. In our previous paper [2] it was found that

Table 1. Characteristics of the pair potentials obtained in this work. λ_{obs}: observed wavelength of the oscillations of $\Phi(r)$, λ_f: π/k_f, where k_f is the Fermi wave vector, r_{RDF}: the nearest neighbour distance, k_f^{obs} and z^{obs}: the Fermi wave vector and the effective valence number estimated from the observed wavelength of the oscillations in $\Phi(r)$, respectively, σ: hard sphere diameter, ξ: $\Phi_{max}-\Phi_{min}$, δ: parameter of steepness of repulsive core in $\Phi(r)$.

	λ_{obs} (Å)	λ_f (Å)	r_{RDF} (Å)	k_f^{obs} (Å^{-1})	z^{obs}	σ (Å)	ξ (K)	δ
Cu	2.38	2.33	2.57 (1150 °C)	1.32	1.04	2.25	1773	14.6
Ag	2.58	2.64	2.87 (1000 °C)	1.22	1.20	2.55	1334	18.3
Au	2.60	2.62	2.86 (1150 °C)	1.21	1.15	2.54	1692	16.2
Fe	2.32	1.91	2.58 (1560 °C)	1.35	1.11	2.25	2044	15.2
Co	2.35	1.88	2.54 (1550 °C)	1.34	1.03	2.22	1925	14.0
Ni	2.28	1.88	2.53 (1500 °C)	1.38	1.14	2.21	1820	13.6

the repulsive core in the pair potential of molten metals is definitely softer than with that of the rare-gas elements. The potetnials of molten noble and transition metals shall be checked here in the same way. We introduce a non-dimensional parameter δ which is a measure of the steepness of the repulsive core at the first node r_0 in $\Phi(r)$:

$$\delta = \frac{\sigma}{\xi}\left(\frac{\Delta\Phi(r)}{\Delta r}\right)_{r=r_0} \tag{3}$$

σ is the hard sphere diameter calculated from $\pi\,\varrho_0\,\sigma^3/6 = 0.45$ with ϱ_0 the average number density of atoms, $\xi\,(=\Phi_{\max}-\Phi_{\min})$ is the difference in energy between the first maximum and the first minimum of $\Phi(r)$. The average value of δ for molten simple metals is 15.1, whereas that for rare-gas elements is 27.8 (see [2]). As shown in Table 1, the values of δ for molten noble and transition metals are about half those for the rare-gas elements and very close to those for molten simple metals. If the repulsive core part of $\Phi(r)$ is approximated by a function $(1/r)^n$, it is found that $n \approx 6$ for molten noble and transition metals and $n \approx 12$ for rare-gas elements. This is consistent with the conclusion of the previous works [10–12] and should be connected with the overlap of the s and d bands in these metals.

To summarize the results in this section, the effective interionic potentials for molten noble and transition metals are characterized by a fairly soft repulsive core, a subsequent minimum at the position corresponding to the nearest neighbour distance, and long-range oscillations.

4. Electrical Resistivity

It is well known that reasonable numerical agreement with experimental data is oftained [1] if the electrical resistivity of simple molten metals is described by the usual Ziman theorie [13]. Recently Evans, Greenwood, and Lloyd [14] extended this approach to molten transition metals by means of the t-matrix of a muffin-tin potential. Their attempt is of considerable interest, but in their calculation the choice of the Fermi energy is obviously somewhat arbitrary and they adopted the muffin-tin potential of an expanded solid with the lattice spacing determined by the density of the liquid. For these reasons we re-calculated the electrical resistivity for molten noble and transition metals near the melting point by means of the concept of a simple approach suggested by Evans, Greenwood, and Lloyd [14]. In our calculation, the muffin-tin potential in the molten state was constructed from the radial distribution function directly observed by X-ray diffraction experiments [3], and the Fermi energy was estimated from the observed value of $k_{\mathrm{f}}^{\mathrm{obs}}$ given in the previous section.

According to Ziman [13], the electrical resistivity (ϱ_{el}) is given by the equation

$$\varrho_{\mathrm{el}} = \frac{3\,\pi\,\Omega_0}{e^2\,\hbar\,v_{\mathrm{f}}^2}\int_0^1 4\left(\frac{q}{2\,k_{\mathrm{f}}}\right)^3 a(q)\left|t\left(\frac{q}{2\,k_{\mathrm{f}}}\right)\right|^2 \mathrm{d}\left(\frac{q}{2\,k_{\mathrm{f}}}\right), \tag{4}$$

$$t(k,k') = \frac{-2\,\pi\,\hbar^3}{m\,(2\,m\,E)^{1/2}}\left(\frac{1}{\Omega_0}\right)\Sigma\,(2\,l+1)\sin\eta_l(E) \cdot \exp\{i\,\eta_l(E)\}\,P_l(\cos\theta)\,. \tag{5}$$

The notation is identical to the one in [14].

For each of the elements the neutral atom charge density has been calculated using the wave function of Herman and Skillman [15]. The Coulomb part of the atomic potential was obtained as a solution of Poisson's equation. The muffin-tin radius is taken to be half the distance between the origin and the position of the first peak in the radial distribution function. Using this muffin-tin potential, the phase shift $\eta_l(E)$ as a function of the energy E is calculated by integrating the Schrödinger equation numerically inside the muffin-tin. The method of numerical calculation was essentially identical to the one reported by Mukhopadhyay, Jain, and Ratti [16]. For example, Fig. 4 shows the energy dependence of the phase shifts for molten copper and iron near the melting point. As shown in this figure the d-phase shifts of the metals change strongly in contrast to the s- and p-phase shifts.

To calculate the electrical resistivity in terms of the t-matrix of Eq. (5), we need the phase shifts at the Fermi energy E_{f} in the molten state. This energy was estimated using the expression suggested by Dreirach [17].

$$E_{\mathrm{f}} = E_{\mathrm{b}} + \frac{\hbar^2}{2\,m^*}\,k_{\mathrm{f}}^2\,, \tag{6}$$

where E_{b} is the energy of the bottom of the band measured from the muffin-tin zero, and m^* is an effective mass. Here we adopted the values of $k_{\mathrm{f}}^{\mathrm{obs}}$ obtained in this work as k_{f} in Equation (6). The estimated values of the phase shifts and some other

Table 2. Comparison of calculated electrical resistivity with experimental data. E_b: energy of the bottom of the band measured from the muffin-tin zero, E_f: Fermi energy, m^*: effective mass, η_l: phase shifts at the Fermi energy. Source of experimental data: J. R. Willson, Met. Rev. **10**, 381 [1965]; R. A. Howe and J. E. Enderby, J. Phys. F. Metal Phys. 3, L 12 [1973].

	Temp. (°C)	E_b (Ryd)	E_f (Ryd)	m^*	η_0	η_1	η_2	ϱ_{el} ($\mu\Omega$ cm) cal.	 exp.
Cu	1150	−0.023	0.612	0.77	−0.136	0.060	3.014	29.2	21.1
Ag	1000	−0.010	0.563	0.73	−0.238	0.004	3.038	37.5	17.2
Au	1150	0.028	0.607	0.71	−0.425	−0.093	2.983	36.2	31.2
Fe	1560	0.032	0.627	0.86	−0.395	−0.031	2.902	108.5	139
Co	1550	0.028	0.635	0.83	−0.312	0.001	2.954	56.8	100
Ni	1500	0.011	0.640	0.85	−0.250	0.024	2.961	43.8	85

relevant data are listed in Table 2. The numerical calculation of Eq. (4) was made using the structure factor $a(q)$ near the melting point as measured by the authors[3]. The results are shown in Table 2 together with the experimental values[18, 19]. Although some information for accurate calculations, as for example the density of states in these molten metals, is missing, the agreement between calculation and experiment is satisfactory. From this calculation we may suggest that a simple muffin-tin potential model coupled with the effective interionic potentials obtained in this work gives a good description of the electrical conductance of molten noble and transition metals.

5. Application to Self-Diffusion, Viscosity and Surface Tension

In our previous paper[2] it has been shown that the kinetic theory of fluids combined with the knowledge of the pair potential, the radial distribution function and the number density of atoms allows to fairly estimate the self-diffusion coefficient and the viscosity of most simple molten metals. This approach does not need any intutive model, contrary to e. g. the hole theory[20], the free volume theory[21], and the fluctiation theory[22]. In this section the pair potentials obtained in this work are applied to calculate the self-diffusion coefficient, the viscosity and the surface tension of the six metals studied.

The calculational procedure was identical to the one reported in our previous paper[2]. The self-diffusion coefficient (D) is given by the relation

$$D = k\,T/(\zeta^{\mathrm{H}} + \zeta^{\mathrm{S}} + \zeta^{\mathrm{SH}}) \,, \tag{7}$$

where ζ^{H}, ζ^{S} and ζ^{SH} are friction coefficients due to the repulsive hard-part interaction, the soft-part interaction between neighbouring atoms and the cross-effect between the hard and soft forces in the pair potential, respectively. They were calculated by the equations

$$\zeta^{\mathrm{H}} = \frac{8}{3}\,\varrho_0\,g(\sigma)\,\sigma^2\,(m\,k\,T)^{1/2} \,, \tag{8}$$

$$\zeta^{\mathrm{S}}_{\mathrm{SS}} = \left[\frac{4\,\pi\,m\,\varrho_0}{3}\int_{\sigma}^{\infty}\nabla_r^{\,2}\,\Phi^{\mathrm{S}}(r)\,g(r)\,r^2\,\mathrm{d}r\right]^{1/2} , \tag{9}$$

$$\zeta^{\mathrm{S}}_{\mathrm{LT}} = -\frac{1}{3}\,\frac{\varrho_0}{(2\,\pi)^2}\,(\pi\,m/k\,T)^{1/2}\int_0^{\infty} \cdot\, q^3\,\tilde{\Phi}^{\mathrm{S}}(q)\,\tilde{G}(q)\,\mathrm{d}q \,, \tag{10}$$

$$\zeta^{\mathrm{SH}} = -\frac{1}{3}\,\varrho_0\,g(\sigma)\,(m/\pi\,k\,T)^{1/2} \times \int_0^{\infty}[(q\,\sigma)\cos(q\,\sigma) - \sin(q\,\sigma)]\,\tilde{\Phi}^{\mathrm{S}}(q)\,\mathrm{d}q \,, \tag{11}$$

where ϱ_0 is the number density of atoms, σ the hard sphere diameter, $g(\sigma)$ the value of the radial distribution function at $r = \sigma$ and m the atomic mass. $\tilde{\Phi}^{\mathrm{S}}(q)$ and $\tilde{G}(q)$ are the Fourier transforms of the soft part of the pair potential and of $[g(r) - 1]$, respectively. q is the wave-vector transfer. Equation (9) is based on the small-step-diffusion theory of Rice-Kirkwood[23], (10) is the based on the linear-trajectory theory of Helfand[24] and (11) is due to Davies-Palyvos[25].

Table 3 shows a comparison of calculated and experimental values. The results obey the relation $\zeta^{\mathrm{S}} > \zeta^{\mathrm{H}} + \zeta^{\mathrm{SH}}$, and this relation implies that the soft part in the pair potential plays a dominant role in determining the magnitude of self-diffusion in molten noble and transition metals, as was also found the alkali metals[2]. The calculated values in terms of both the small-step-diffusion theory and the linear-trajectory theory for molten copper and

Table 3. Comparison of calculated self-diffusion coefficients with experimental data. The units of ζ and D are g sec^{-1} $\times 10^{-10}$ and cm^2 sec$^{-1} \times 10^{-5}$ respectively, ζ^{H}: hard-core friction coefficient, $\zeta^{\mathrm{S}}_{\mathrm{LT}}$: soft-part friction coefficient by linear-trajectory theory, $\zeta^{\mathrm{S}}_{\mathrm{SS}}$: soft-part friction coefficient by small-step-diffusion theory, ζ^{SH}: cross-effect friction coefficient, D_{LT}: value calculated by linear-trajectory theory, D_{SS}: value calculated by small-step-diffusion theory, D_{exp}: value observed experimentally. Sources of experimental data. D_{Cu}: J. Henderson and L. Young, Trans. Met. Soc. A.I.M.E. **221**, 72 [1961]. D_{Ag}: V. G. Leak and R. A. Swalin, Trans. Met. Soc. A.I.M.E. **230**, 426 [1964].

	Temp. (°C)	ζ^{H}	$\zeta^{\mathrm{S}}_{\mathrm{LT}}$	$\zeta^{\mathrm{S}}_{\mathrm{SS}}$	ζ^{SH}	D_{LT}	D_{SS}	D_{exp}
Cu	1150	11.5_3	23.5_7	29.2_5	4.26_7	4.99	4.36	4.66
	1300	12.4_1	20.1_9	28.6_4	4.15_2	5.91	4.80	
	1500	13.9_2	17.4_9	28.0_9	4.13_4	6.88	5.30	
Ag	1000	11.5_5	19.5_5	29.6_6	4.47_3	4.94	3.85	3.22
	1150	12.3_3	13.3_4	28.9_3	4.27_3	6.56	4.31	
	1300	13.8_1	13.7_4	28.7_3	4.32_9	6.81	4.63	
Au	1150	15.9_0	29.2_1	40.7_0	6.33_3	3.82	3.12	
	1300	17.9_3	24.5_4	39.7_7	6.46_0	4.44	3.38	
	1500	20.7_9	20.6_8	39.0_0	6.64_6	5.09	3.68	
Fe	1560	11.0_3	19.3_7	23.0_9	2.71_8	7.64	6.87	
	1600	11.7_5	18.0_2	22.5_0	2.85_3	7.92	6.97	
	1650	11.9_6	16.2_3	22.3_2	2.82_2	8.56	7.15	
Co	1550	11.8_6	16.8_3	16.7_5	3.40_5	7.84	7.86	
	1600	12.9_2	15.1_4	15.0_3	3.63_0	8.16	8.19	
	1650	13.5_8	14.8_3	14.2_8	3.68_2	8.27	8.41	
Ni	1500	11.5_7	16.7_6	16.8_3	2.15_4	7.37	7.35	
	1600	12.8_8	14.8_9	15.1_0	2.28_7	8.60	8.54	
	1650	13.4_4	14.0_1	14.4_3	2.26_1	8.93	8.81	

silver are in good agreement with the observed values. A detailed discussion may not be given here, because experimental data for molten gold and the transition metals are not reported.

The viscosity (η) and surface tension (γ) have been estimated using the relations [26, 27]

$$\eta = \frac{2\pi}{15} (m/k\,T)^{1/2} \varrho_0^2 \int_0^\infty g(r) \frac{\mathrm{d}\Phi(r)}{\mathrm{d}r} r^4 \,\mathrm{d}r\,, \tag{12}$$

and

$$\gamma = \frac{\pi}{8} \varrho_0^2 \int_0^\infty g(r) \frac{\mathrm{d}\Phi(r)}{\mathrm{d}r} r^4 \,\mathrm{d}r\,. \tag{13}$$

Table 4 shows the results of these calculations together with the observed values. The calculated values of the viscosity are in good agreement with the observed values, but those of surface tension are not as good. This may be due to the inadequacy of the assumption that the density changes abruptly from the vapour phase in the liquid-vapour transition zone.

Table 4. Comparison of calculated and observed viscosity and surface tension. Source of experimental data: J. R. Willson, Met. Rev. **10**, 381 [1965].

	Temp. (°C)	η (Poise) cal.	η (Poise) exp.	γ (dyne/cm) cal.	γ (dyne/cm) exp.
Cu	1150	0.036	0.038	1450	1300
	1300	0.032		1370	
	1500	0.029		1310	
Ag	1000	0.038	0.040	1120	980
	1150	0.035		1090	
	1300	0.032		1060	
Au	1150	0.049	0.051	1130	1130
	1300	0.046		1110	
	1500	0.043		1090	
Fe	1560	0.046	0.048	2250	1850
	1600	0.041		2030	
	1650	0.039		1960	
Co	1550	0.043	0.039	2040	1890
	1600	0.040		1930	
	1650	0.037		1810	
Ni	1500	0.043	0.046	2020	1800
	1600	0.040		1930	
	1650	0.036		1761	

To summarize the results in this section, the effective interionic potentials obtained in this work are shown to be adequate for the estimation of various macroscopic physical properties, such as self-diffusion and viscosity, in molten noble and transition metals. Moreover, also information regarding their temperature dependence may be obtained as shown in Tables 3 and 4.

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