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TRANSFERABLE PAIR POTENTIALS FOR LIQUID IRON, COBALT AND NICKEL

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Effective inter-ionic pair potentials for liquid iron, cobalt and nickel are derived from second order pseudopotential perturbation theory with the transferable electron-ion potential of Fiolhais and co-workers which was originally developed for the solid state. The liquid structure is obtained by the thermodynamically self consistent liquid state theory, the variational modified hypernetted chain (VMHNC) approximation. It has been found that the calculated structural properties at the investigated thermodynamic states just above the melting point agree well with experiment. In this work we have determined the parameters of liquids Fe, Co and Ni for the universal choice of the evanescent core pseudopotentials of Fiolhais'. We have shown that this pseudopotential is transferable to the liquid state if used this parameterisation.

Keywords: Static structure factor; Pair distribution function; Pseudopotentials; Pair potentials

1. INTRODUCTION

The modelling of inter-ionic interactions for liquid transition metals is a long-standing problem and it remains a problem of interest. It is well known that the thermodynamics and structure of the simple metals

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in their liquid state are well described using the effective inter-ionic interaction which is derived from the pseudopotential theory [1,2]. The difficulties arise when this method applied to transition metals because of the hybridisation of d states. The pseudopotential theory was reformulated and generalised by Harrison to include d -band metals [3]. Generalised pseudopotential method had been extended and applied by Moriarty [4] to calculate the dynamic properties of noble and alkaline earth metals, possessing either completely filled or empty d -bands. Most of the theoretical works on liquid transition metals are based on a study of Wills and Harrison (WH) [5] who used a separate treatment of s - p and d states and, in addition, take account of the effect of s - d hybridisation. WH model potential is sufficiently simple to be used in the liquid state calculations and applied by most number of workers [6–9]. One of the main result of WH calculations is that the applicability of WH model for the elements at the beginning and at the end of the $3d$ series is ascertain. Recently, the structure of the $3d$ transition metals series in the liquid state have been calculated by using a local pseudopotential proposed by Brettonet and Silbert (BS) [10]. This model combines the sp band contribution with the contribution of s - d mixing. The empty core model pseudopotential is used to describe the nearly free electron band and a d -band contribution deduced via an inverse scattering approach. It has been proved to be successful for liquid transition metals [10–13]. It has been noted that, in some cases the local model pseudopotentials describe some liquid state properties even better than those of norm conserving non-local ones.

This work is concerned with a recently proposed local model pseudopotential of Folhais *et al.* [14], whose parameters are fitted on the thermodynamic properties of the solid state. Two versions of this potential have been considered with individual and universal choices for parameters. It was originally proposed by Perdew and co-workers who has checked its transferability to finite systems for dimmers and clusters [15,16]. We have shown in our previous work [17] that the individual version of their model potential is transferable for liquid alkaline and alkaline earth metals using very accurate integral equation of liquids, the variational modified hyper-netted chain (VMHNC) approximation [18,19]. Boulahbak *et al.* and

Tammer *et al.* [20] have also shown the universal version of the local pseudopotentials of Fiolhais *et al.* using the soft-core mean spherical approximation (SMSA) is transferable for simple liquid alkaline metals, Li, Na, K, Rb, Cs but not polyvalent ones. Recently we have shown the calculated structure factor $S(q)$ for liquid polyvalent simple metals using the individual choice of Fiolhais' potential with VMHNC integral equation theory disagrees [21] with experiment [22]. According to our knowledge, this model potential has not been employed to study of structural properties of liquid transition metals. One of the aims of this paper is to investigate checking the degree of transferability of these pseudopotentials to the transition metals in their liquid state.

For this purpose, we took interest liquid Fe, Co and Ni for which some authors have the successful structural and thermodynamic calculations using the BS [12–13] potentials. In this work we have taken the universal choice of the local Fiolhais' potential in our calculations. The pair potential contains three parameters: the pseudopotential parameters R and α , the effective s -electron valance number Z_s , although there are some restrictions to the range of the values that R and α can take up [16]. Our second aim is to demonstrate that this pseudopotential is transferable for liquid transition metals if used this parameterisation. We show that the universal choice of Fiolhais' potential predicts more reasonable structural properties for liquid Fe, Co and Ni than others [12–13].

The layout of the paper is as follows: in Section 2, we outline the theoretical basis of our work. First, the effective pair potential with the local pseudopotential of Fiolhais' and BS potential are described. The main features of this liquid state theory used in our calculations are given in the same section. The results of structural calculations for liquid Fe, Co and Ni are presented, together with brief discussion of our results in Section 3. Finally, in Section 4, we sum up and briefly comment on the results of our work.

2. THEORY

The effective inter-atomic pair potential in simple metals $\phi(r)$ is based on the use of pseudopotentials to describe the interaction

between an ion and second ion and its screening cloud of electrons from second order pseudopotential perturbation theory, obtained by,

$$\phi(r) = \frac{Z_S^2}{r} + \phi_{\text{ind}}(r), \quad (1)$$

where Z_S for the effective number of valance electrons per atom and here Fourier transform (FT) of the indirect part is given by,

$$\tilde{\phi}_{\text{ind}}(q) = \chi(q)|\tilde{\omega}(q)|^2, \quad (2)$$

where $\tilde{\omega}(q)$ is the pseudopotential local form factor and $\chi(q)$ is the response of the electron gas. The effects of exchange and correlation between the electrons are accounted for by the introduction of a local field factor $G(q)$ in the response function as

$$\chi(q) = \frac{\chi^0(q)}{1 - (4\pi/q^2)[1 - G(q)]\chi^0(q)} \quad (3)$$

where $\chi^0(q)$ is the response function of a non-interacting electron gas. A wide variety of expression for the local field function are available [23–25]. In this paper we employ the well known local field corrections of the Ichimaru-Utsumi (IU) [23] and the local density approximation (LDA) version of the local-field function $G(q)$ with the correlation energy of Vosko-Wilk and Nussair (VWN) [25]. In the present work, we use the recent local pseudopotential proposed by Fiolhais *et al.* This model pseudopotential has the core repulsion represented by an exponential factor, so called evanescent core (EC) pseudopotential. An analytic expression for the form factor of this pseudopotential in Fourier space:

$$\tilde{\omega}(q) = 4\pi Z_S R^2 \left[-\frac{1}{(qR)^2} + \frac{1}{(qR)^2 + \alpha^2} + \frac{2\alpha\beta}{[(qR)^2 + \alpha^2]^2} + \frac{2A}{[(qR)^2 + 1]^2} \right] \quad (5)$$

where R is the core decay length, β and A parameters are given in terms of α namely

$$\beta = \frac{\alpha^3 - 2\alpha}{4(\alpha^2 - 1)}, \quad (6)$$

$$A = \frac{\alpha^4}{4(\alpha^2 - 1)} \quad (7)$$

Krosko and Gurskii have independently proposed a similar model [26] before. The values α and R can be obtained from the equilibrium condition of the solid state using the LDA [14]. For this reason this model potential has the advantage to be free of adjustable parameters. Fiolhais and co-workers have given the individual and universal pseudopotential parameters, EC(I) and EC(U) respectively, for sixteen simple metals in Ref. [14]. But the parameters R and α of the EC(U) pseudopotential for several values of the average valence electron density r_S and valence Z are given in a table in Ref. [16]. The resulting effective pair potential for liquid transition metals contains three parameters R , α , Z_S whose choice is discussed in the next section.

In the present work we shall use another local pseudopotential suitable for liquid transition metals proposed by Brettonet and Silbert (BS). This local pseudopotential is constructed by the superposition of the sp and d band contributions, written as,

$$w(r) = \begin{cases} \sum_{n=1}^2 B_n \exp(-r/na) & r < R_C \\ -Z_S/r & r > R_C \end{cases} \quad (8)$$

where a , R_C , Z_S are softness parameter, the core radius, effective s electron occupancy number, respectively. The effective inter-ionic potential is obtained from Eq. (1) in usual way and different screening functions, for comparison.

With the effective pair potential known, integral equations are able to provide us the liquid structure for metals. In our structural calculations, one of the integral equation theory which has shown to be very reliable theory of liquids is VMHNC has been carried out [18,19]. The starting point of most of the integral equation theories of liquids is the Ornstein-Zernike (OZ) equation, which for a homogenous, an isotropic system can be written as

$$h(r) = c(r) + \rho \int d\vec{r}' h(|\vec{r} - \vec{r}'|) c(\vec{r}') \quad (9)$$

which defines the direct correlation function $c(r)$, in terms of the total correlation function, $h(r) = g(r) - 1$ where $g(r)$ is the pair distribution function and ρ is the number density. This equation is supplemented by exact closure relation involving the pair potential $\phi(r)$,

$$g(r) = \exp[h(r) - c(r) - \beta\phi(r) - B(r)] \quad (10)$$

where $\beta = (k_B T)^{-1}$, and $B(r)$ the bridge function. In this work $B(r)$ is approximately by the use of the analysis solution of the Percus-Yevick (PY) equation for hard sphere (HS), namely $B(r) = B_{PY}(r, \eta)$ [18]. The packing fraction $\eta = \eta(\beta, \rho)$ is variationally determined by minimizing the VMHNC configurational free energy at a temperature T , as

$$\frac{\partial f^{\text{VMHNC}}(\beta, \rho, \eta)}{\partial \eta} = 0 \quad (11)$$

3. RESULTS

We have calculated the effective inter-ionic pair potentials for liquids Fe, Co and Ni using the transferable electron-ion potentials of Fiolhais' using the following parameterisation. The liquid structure of Fe, Co and Ni has been obtained at the thermodynamic states just above the melting point. The input temperature and corresponding number densities ρ are taken from Waseda [22] given in Table I. In our structural calculations for the numerical solution of OZ Eq. (9) and closure relation (10) we have used Zerah's algorithm [27]. For the parameterisation of the universal evanescent core pseudo-potential of Fiolhais', firstly we choose the Z_S values for liquids Fe, Co and Ni. It is well known that the effect valence Z_S takes non-integer values in transition metals to account for s - d hybridisation. Self consistent calculations of Moriarty indicate that Z_S takes the values in a narrow range $1.1 < Z_S < 1.7$ for the three series of the transition metals. For the choice of Z_S values in this work, we have searched which values of Z_S , within the above range, predict the best result for $g(r)$ comparing with experiment. For this purpose we have changed Z_S from 1.5 to 1.4. Thus we have obtained the parameters from the data table given in Ref. [54] of Ref. [16]. The values of R and α used in this work are reported in Table I. We shall use a couple

TABLE I Input data: temperature T , number density ρ , potential parameters R and α

<i>Metal</i>	T (K)	ρ (atoms/Å ³)	Z_S	R (a.u.)	α (a.u.)
Fe	1833	0.0756	1.5	0.276	4.320
			1.4	0.275	4.415
Co	1823	0.0787	1.5	0.271	4.365
			1.4	0.267	4.460
Ni	1773	0.0792	1.5	0.270	4.375
			1.4	0.266	4.470

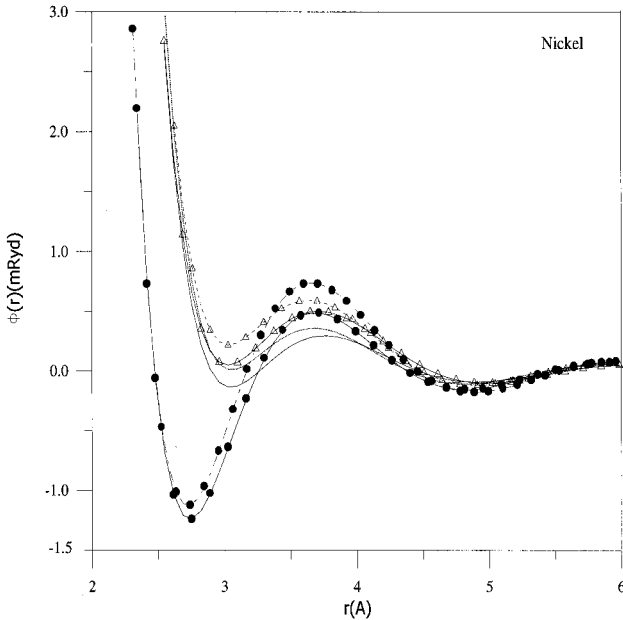


FIGURE 1 Effective pair potentials for liquid Ni. Solid lines and solid lines with empty triangles show Fiolhais' potentials for $Z_S=1.4$ with LDA and IU, respectively. Dashed lines and dashed lines with empty triangles denote Fiolhais' potentials for $Z_S=1.5$ using LDA and IU screening functions, respectively. Solid lines with full circles and long-dashed lines with full circles present BS potential using LDA and IU screening functions for the parameters given in Ref. [13].

sets of a , Z_S , R_C values for BS model potential which are taken from Ref. [12,13], for comparison.

In Fig. 1, we compare the calculated effective pair potentials of nickel, with Fiolhais' and BS model potentials using LDA and IU screening functions where we have changed Z_S from 1.5 to 1.4.

It appears that both model potential using the LDA screening give rise to a deep potential well while IU yields a shallow well. We note the first maximum of the both calculated pair potential is around 3.7 Å. The calculated BS potential with the parameters of Ref. [13] using LDA screening is closer to our results than those obtained by IU screening with the parameters for $Z_S=1.4$. However the first minima in our potentials are shallower than those of BS. The energies of potential wells are about four times smaller than corresponding BS energies. The positions of the wells are displaced towards slightly larger values of r . In order to influence both exchange-correlation effect and effective valence on $S(q)$, in Fig. 2(a) we present a

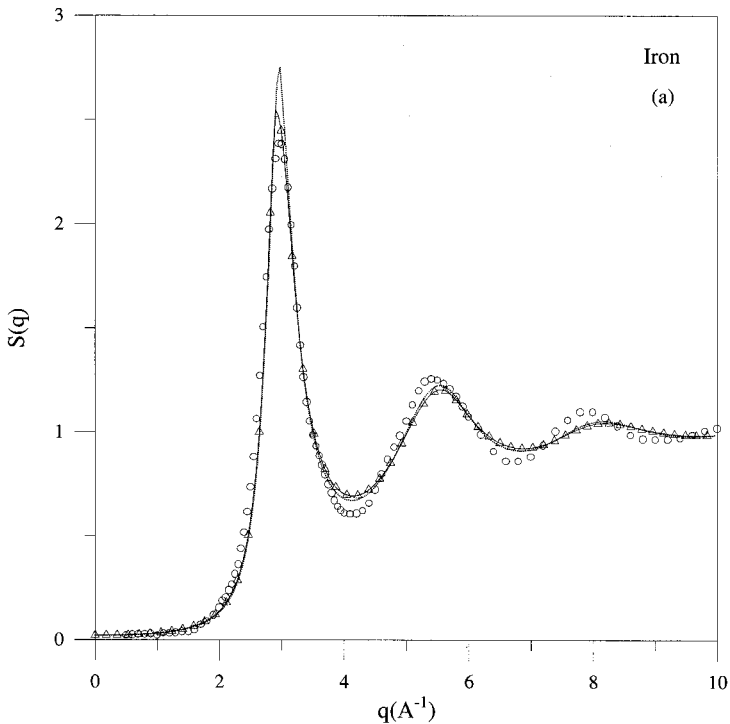


FIGURE 2 (a) Structure factor of liquid Fe. Solid and dashed lines denote the present results using the LDA where $Z_S=1.4$ and $Z_S=1.5$, respectively. Solid lines with open triangles show the results with IU screening for $Z_S=1.4$. Open circles correspond to the experimental data of Waseda [22]. (b) Same as in (a) but effective Fiolhais' potentials and corresponding $g(r)$ s.

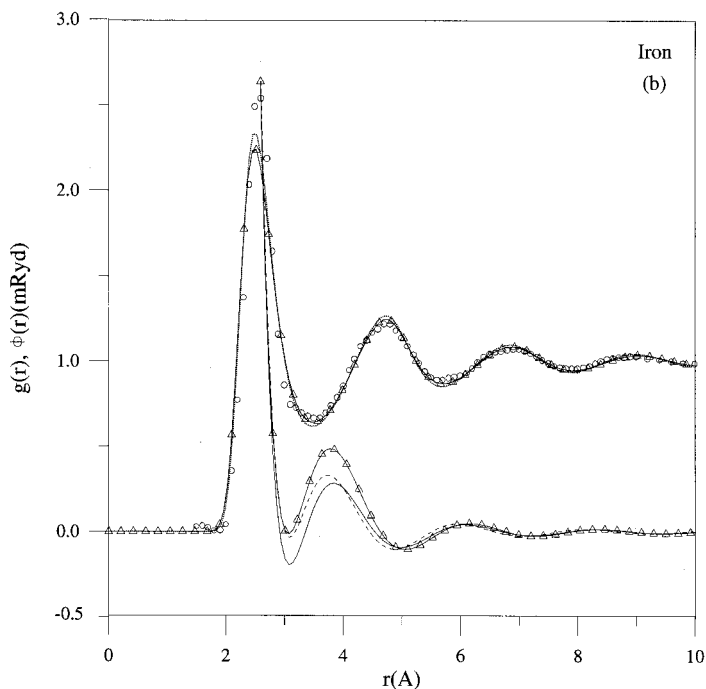


FIGURE 2 Continued.

comparison of our VMHNC result $S(q)$ calculated with the pair potentials given in Fig. 2(b) for liquid iron. These figures have also included the experimental $S(q)$ and $g(r)$ data of Waseda [22] for comparison. Except height of the first peak for $Z_S=1.4$, the VMHNC results $S(q)$ calculated with Fiolhais' potential using LDA and IU screening functions are almost the same however these potentials are so different. The calculated values of $S(0)$ are 0.0215 and 0.0214 using LDA and IU screening functions, respectively. But it is 0.0205 for $Z_S=1.5$ comparing with the experimental value of 0.021 for liquid Fe. The effective valence is affected more on $S(0)$ value than screening. We also find that the oscillations in the calculated $S(q)$ die out more rapidly than in the experimental data. It suggests too soft a repulsive potential.

We have also compared the VMHNC results of $S(q)$ and $g(r)$ for liquid Co with those obtained from BS model potential for the

parameters given in Ref. [12] using LDA screening. These are demonstrated along with the experimental data, in Figs. 3(a) and (b), respectively. The results of potentials for both Fiolhais' and BS model are shown in Fig. 3(b). The VMHNC results of Co are closer to experimental data than those obtained from BS potentials.

The results of VMHNC calculations $S(q)$ and $g(r)$ for liquid nickel along with experimental data are shown in Figs. 4(a) and (b), respectively. In Fig. 4(a) we have also included the results of $S(q)$ with BS model potentials with LDA screening using the parameters given in Ref. [12], for comparison. Comparing the illustrated VMHNC results of $g(r)$ with both parameters given in Ref. [12,13] in Fig. 4(b) shows that they are almost the same. Figures show that, curves agree quite well with the experimental data of Waseda [22] than others [12,13]. However, as a general feature, the height of the first calculated peak of $S(q)$ is overestimated. Except Ni, we have observed that the positions of main peaks in calculated $S(q)$ are similar. However, for Ni, it is shifted slightly toward smaller q values as compared with experimental data. Both Bhuiyan and co-workers [12] and Jakse *et al.* [13] have observed this shift with VMHNC and HMSA integral equations using BS model potentials. With this exception, our results for $S(q)$, in the phase of oscillations are in very good agreement. In particular, height of the main peak and the depth of the first minima of $g(r)$ for Ni are in better agreement than those obtained by others.

As we examine the figures, it can be seen that the low- q region is well reproduced all cases. The calculated $S(0)$ values of Co for $Z=1.5$ and Ni for $Z=1.5$ are 0.0192 and 0.019, compared to the experimental values of 0.0186, 0.0199, respectively. However, the values of $S(0)$ with both BS model parameters are around 0.033, 0.032 and 0.030 for Fe, Co and Ni, respectively.

It is clearly seen in Figs. 2–4 that a larger value of Z_S , increases the height of the peaks of $g(r)$ altering their positions a little since universal choice of Fiolhais' potential parameters R and α depend on the effective valence. It appears that a larger value of α gives rise to a deep potential well and small Friedel oscillations, while the others yield a shallow potential. A larger R results in a softer repulsive potential, although the positions of both the first zero and minimum are affected more less by changes in R .

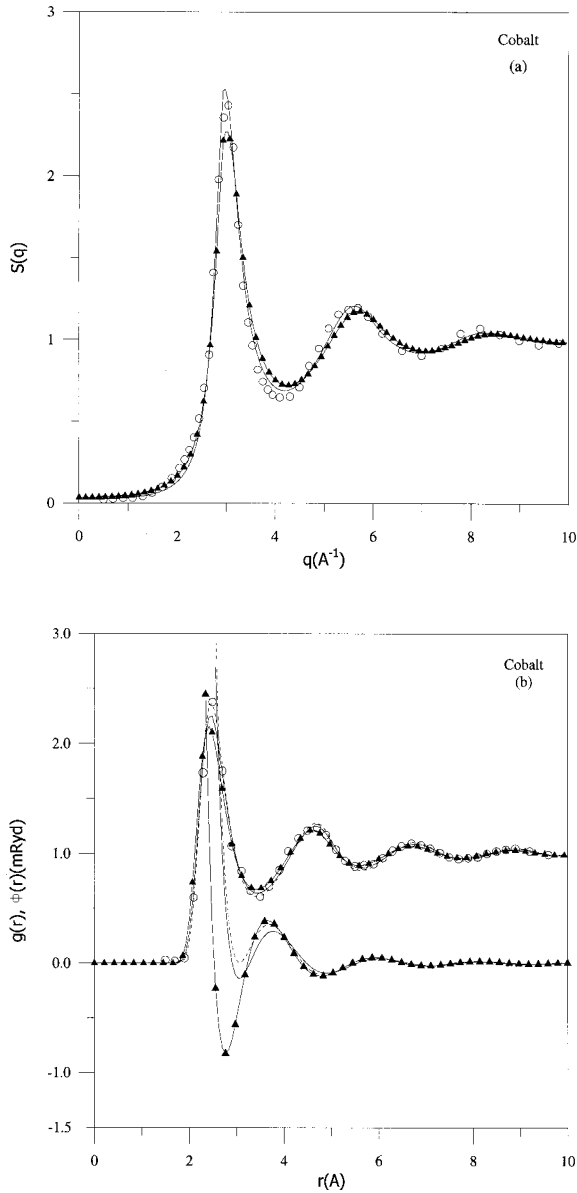


FIGURE 3 (a) Structure factor $S(q)$ for liquid Co. Lines with full triangles show the result of BS-LDA model with the parameters given in Ref. [12]. Other notations are the same as in Fig. 2. (b) Same as in (a) but effective potentials and corresponding $g(r)$ s.

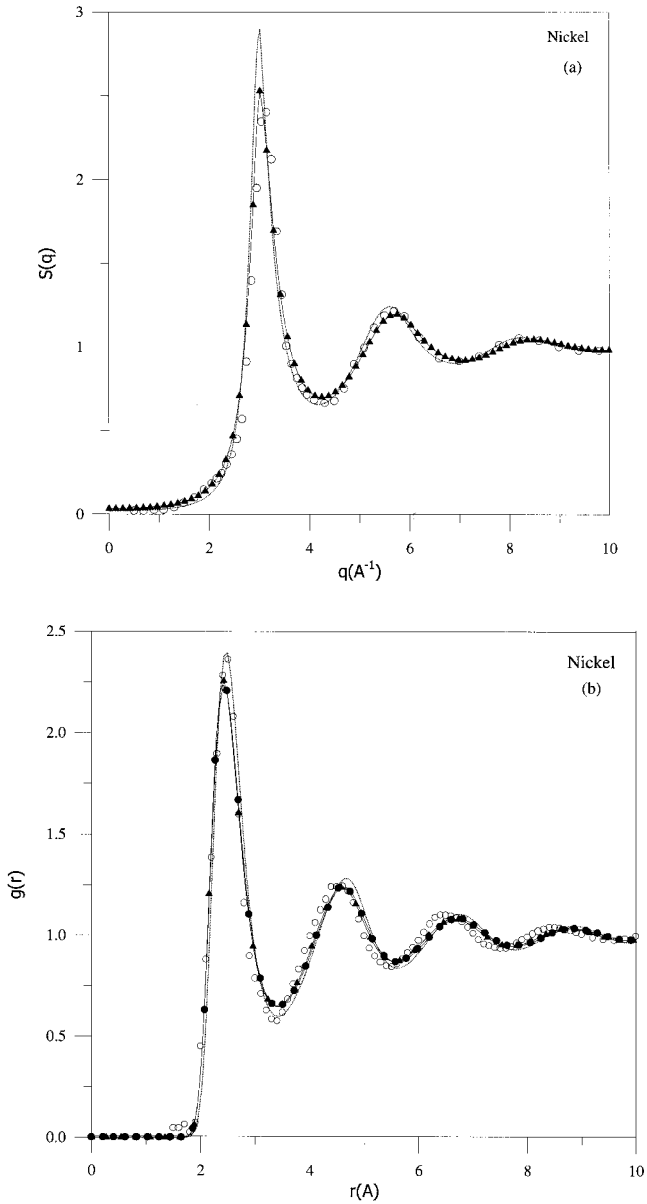


FIGURE 4 (a) The calculated $S(q)$ using the BS potential parameters given in Ref. [12]. All notations are the same as in Figs. 2 and 3; (b) The calculated $g(r)$ using the BS potential parameters given in Ref. [12,13]. All notations are the same as in Figs. 2 and 3.

4. DISCUSSION

In this paper, We have shown that a simple evanescent model potential of Fiolhais' can be successfully applied to the study of liquid Fe, Co and Ni using the presented parameterisation. We have analysed the structure of liquid Fe, Co and Ni using the VMHNC liquid state theory. Our results are comparable to those from one of the BS model potentials. A good agreement is found between our calculated structural properties and experimental data. A number of properties need to be studied in order to have a full picture of the applicability of this potential to liquid transition metals. At present, we are more interested in applying the present method for study of pair structures in liquid noble and less simple metals, and its results will be reported later.

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