Thermodynamics and Structure of Liquid Metals from a New Consistent Optimized Random Phase Approximation

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Z. Naturforsch. **56 a,** 605–612 (2001); received March 1, 2001

We study thermodynamics and structural properties of several liquid metals to assess the validity of the generalized non-local model potential (GNMP) of Li et al. [J. Phys. **F16**, 309 (1986)].

By using a new thermodynamically consistent version of the optimized random phase approximation (ORPA), especially adapted to continuous reference potentials, we improve our previous results obtained within the variational approach based on the Gibbs - Bogoliubov inequality.

Hinging on the unified and very accurate evaluation of structure factors and thermodynamic quantities provided by the ORPA, we find that the GNMP yields satisfactory results for the alkali metals. Those for the polyvalent metals, however, point to a substantial inadequacy of the GNMP for high valence systems. – Pacs: 61.25.Mv, 61.20.-p, 61.20.Gy

Key words: Liquid Metals; Liquid Structure; ORPA; Pseudopotentials; Thermodynamics.

1. Introduction

During the last two decades, the structure and thermodynamics of liquid metals have been extensively studied with increasing sophistication in the modeling of the inter-ionic forces and in the classical statistical mechanics treatment of ionic correlations.

The calculations of the atomic interactions in s-p bonded metals have mainly been based on density dependent pairwise potentials derived from electron-ion pseudo-potentials within the linear response theory and second order perturbation theory [1, 2]. The statistical mechanics side of the problem has been studied with perturbation theory [3, 4] integral equations [5 - 7] and computer simulations [8 - 10].

Although a lot of work has been done on both, some questions still await a definite answer. The limitations of linear-response-theory based interactions are well known. But it is also possible to consider the interactions based on second order perturbation theory as effective pair-wise potentials [11, 12]. However, in such case one would expect that a combined analysis of the structural *and* thermodynamic consequences of the interaction could elucidate in a quantitative way the relevance of such effective model interactions for the description of the physical properties of liquid metals.

We believe that only a combined analysis of structural and thermodynamic data should be able to assess the quality of any model for the inter-ionic forces. However, only a few studies have been made addressing simultaneously these two problems. Moreover, many existing studies have been limited to local model potentials with empirically determined parameters [13 - 15].

In the last two decades, some ab-initio pseudo-potentials suitable for perturbation theory of the ionic interactions were generated. One of these, the so called generalized non-local model potential (GNMP) of Li et al. [16] has been shown to be reliable in reproducing a number of metallic properties ([17] and cited refs.).

On the side of the treatment of the atomic correlations, computer simulation has a precision limited only by numerical errors. However, when focusing on trends of thermodynamic quantities like

0932–0784 / 01 / 0900–0605 $ 06.00 © Verlag der Zeitschrift für Naturforschung, Tübingen · www.znaturforsch.com

the excess entropy, approximate but reliable theories aimed at modeling the free energy are still very useful tools.

The simplest of such methods is the variational Gibbs Bogoliubov (GB) approach [18]. However, the quality of the results for the Helmholtz free energy depends on how accurately the reference liquid models the correlations in the system of interest. Moreover, within the GB approximation, the structure factor and the excess entropy are those of the reference system and depend on the model interaction only indirectly through the determination of the optimal reference system.

In a previous paper [3] we used the GB approximation to study the thermodynamics of the GNMP for simple mono and polyvalent metals by using charged hard spheres (CHS) as reference system. Such a system allowed us to explore the smooth interpolation between the two well known limit cases of neutral hard spheres (NHS) and one component plasma (OCP). The main result was that, while for the alkali metals, in agreement with previous investigations, the OCP is a very satisfactory reference system, in the case of the polyvalent metals, the OCP and NHS are essentially equivalent. Only the excess entropy and the structure factor S(k) evaluated from the OCP indicate the tendency of such a reference system to give a too high degree of short range order. Thus, it is natural to ask whether a statistical mechanics approach, less trivial than the GB approximation, would be able to provide accurate results for both structure and thermodynamics by using the OCP reference system.

Usually, the GB approximation is introduced as an exact upper bound of the Helmholtz free energy without any simple way to improve it. However, the optimized random phase approximation (ORPA) of the theory of liquids, firstly introduced as perturbation theory in the 1970's by Weeks, Chandler, and Andersen [19 - 21], can be recast [22] in the form of a refinement of the GB approximation.

In [22], we introduced two different ORPA generating functionals from which thermodynamics and correlation functions can be obtained. In particular, one of them (therein referred as GB-ORPA) can be seen as a straightforward improvement of the GB free energy functional by adding the so-called *ring* term to it and treating the resulting approximation as a generalized variational problem. The corresponding approximation for the correlation functions embodies a partial thermodynamic consistency and, at variance with the conventional ORPA, can be used even without a hard core reference system.

In the present paper, we use such GB-ORPA approximation, in connection with the OCP reference system, to extend our previous investigations [3] on the GNMP for mono and polyvalent liquid metals.

The paper is organized as follows. In Sect. 2, we summarize the relevant formulae for GB-ORPA with continuous potentials and we describe the numerical procedure. In Sect. 3, we illustrate our numerical results for the alkali metals at different temperatures and for polyvalent metals (Mg, Cd, Al, Tl and Pb) near freezing. The possibility of a reliable and consistent evaluation of structural and thermodynamical quantities allows us to draw some conclusions about the overall quality of the GNMP model for the investigated systems.

2. Theory and Numerical Method

The high quality of the numerical results of ORPA for simple liquids, in all the existing variants, is well documented [23, 24, 14, 15, 13]. In recent years, modified integral equations like modified hyper-netted chain (MHNC) [25, 26] or HMSA [27] have been used more frequently. There are two main reasons for this preference. The standard numerical methods to solve ORPA equations were extremely inefficient and not very accurate. We have recently shown how to overcome this problem [22] by resorting to numerical methods closer to those used for integral equations.

The second possible reason for a decrease of interest in ORPA is probably due to the feeling that it is mainly a perturbative approach, thus intrinsically less accurate than integral equations. Although ORPA was originally developed in the framework of perturbation theories for the structure of liquids, we consider it as a reference system integral equation. Indeed, we have verified that the algorithm can actually manage "perturbations" of the direct correlation functions that are a few orders of magnitude larger than the reference system values. For all the cases studied in the present paper we have performed MHNC calculations of the correlation functions using the Verlet-Weiss parameterization of the hard spheres bridge functions. Even in the case of a tetravalent metal like Pb, the ORPA and the MHNC results for the structure factors of liquids close to the melting point are extremely close and indistinguishable on the scale of the figures. We have observed significant differences only for non-simple structure factors like that of liquid germanium, not included among the systems studied with the GNMP. Recently, an accurate investigation of the square-well and square-shoulder fluids [28] has shown that in the latter case too ORPA becomes a rather poor approximation. However, we stress that these cases correspond to strong deviations from the typical behavior of the experimental correlation functions of the liquid metals studied in the present paper.

For the systems studied presently, we regard the ORPA as accurate as MHNC for the description of the liquid structure, and therefore of a quality comparable to computer simulation results.

The GB-ORPA is based on the functional [22]

$$F_{\mathrm{ORPA}} = F_0 + \frac{1}{2}\rho \int g_0 \Delta\phi + F_{\mathrm{RING}}[g_0, \Delta\phi], \quad (1)$$

where F_0 and g_0 are the free energy and the pair correlation function of the reference systems, $\Delta\phi$ is the difference between the actual and the reference pair potential and F_{RING} is the RPA functional defined as

$$F_{\mathrm{RING}}[g_0, \Delta\phi, \chi(r)] = \quad (2)$$

$$\frac{k_{\mathrm{B}}T}{[2\pi]^3} \int \mathrm{d}^3\boldsymbol{k}[S_0(k)\hat{P}(k) - \log(1 - \rho S_0(k))\hat{P}(k)].$$

$\chi(r)$ is the optimizing potential , $S_0(k)$ the reference system structure factor and $\hat{P}(k)$ the Fourier transform of the function $P(r)$ defined as

$$P(r) = \begin{cases} \chi(r) & r < r_{\mathrm{c}} \\ \beta\Delta\phi(r) & r > r_{\mathrm{c}} \end{cases} \quad (3)$$

r_{c} being the a free parameter of the theory measuring the extent of the region where the reference system $g(r)$ is unaffected by the ORPA modification. Notice that in our treatment of the ORPA with soft reference systems there is no hard core. If the reference system $g_0(r)$ is a continuous function, the ORPA correction introduces only a discontinuity on the first derivative of $g(r)$ at r_{c}.

Thus, the practical implementation of the GB-ORPA [29] requires to choose a reference system interaction and to determine its parameters, the unknown function $\chi(r)$ as well as the parameter r_{c}, by making the generating functional extremum.

In practice, after discretizing all the functions on radial numerical grids in r and k space, we have to solve the set of equations

$$\frac{\delta F_{\mathrm{ORPA}}}{\delta\chi(r_i)} = 0 \text{ for } i \text{ such that } r_i < r_{\mathrm{c}}, \quad (4)$$

$$\frac{\partial F_{\mathrm{ORPA}}}{\partial r_{\mathrm{c}}} = 0 \quad (5)$$

and

$$\frac{\partial F_{\mathrm{ORPA}}}{\partial a_i} = 0, \quad (6)$$

where a_n, $n = 1, \ldots.., N$ is the set of the reference system parameters.

A byproduct of such a choice is to make the ORPA functional a free energy functional, thus exactly satisfying the virial-energy thermodynamic consistency [29]. Such consistency is not exact in the standard ORPA calculations where the reference system is chosen through a non variational procedure [19].

Notice that, at variance with the GB functional, F_{ORPA} is not an upper bound of the exact free energy.

By using the equality between the value at the extremum of (1) and the Helmholtz free energy per particle, thermodynamics gives the deviation from the reference system entropy per particle s_{ex} and the static structure factors as

$$s_{\mathrm{ex}} = s - s_0 = -\frac{1}{T}F_{\mathrm{RING}}, \quad (7)$$

$$S_{\mathrm{ORPA}}(k) = \frac{S_0(k)}{[1 + \hat{P}(k)S_0(k)]}. \quad (8)$$

High numerical accuracy in the evaluation of (4) can be achieved by using the algorithm described in [29]. This scheme allows one to obtain a $\Delta g(r) = 0$ within machine precision inside r_{c} without too much numerical effort. This should be compared with typical values of $\Delta g(r) \approx 0.1$ which can be obtained using the older methods [13 - 15, 30].

In the present calculations, in order to start with the overall best case found in our previous investigation [3], we have chosen the OCP as our reference system for all the metals. In any case, since it is reasonable that the dependence of the results from the reference system becomes weaker as the accuracy of the approximation increases, we do not expect an important dependence of our conclusions from such a choice.

Table 1. Optimal OCP reference system coupling parameter Γ and the corresponding Helmholtz free energy for Gibbs-Bogoliubov [3] (GB) and ORPA at temperatures above freezing and atomic volume $\Omega_o(au^3)$ [32].

System	Temp. (K)	Ω_o	Γ_{GB}	F_{GB}	Γ_{ORPA}	F_{ORPA}
Li	463	152.410	160	−0.2660	171	−0.2664
Na	373	277.942	153	−0.2357	163	−0.2362
	473	285.264	109	−0.2382	125	−0.2387
	573	292.388	82	−0.2409	92	−0.2415
K	336	528.500	145	−0.2011	152	−0.2015
	378	534.921	123	−0.2023	138	−0.2026
	473	549.686	92	−0.2050	97	−0.2055
Rb	312	654.600	138	−0.1929	152	−0.1932
	373	661.366	109	−0.1948	126	−0.1952
	473	685.510	80	−0.1982	89	−0.1986
Cs	302	810.800	145	−0.1810	154	−0.1813
	373	829.178	109	−0.1835	125	−0.1844
	473	856.356	80	−0.1872	89	−0.1876
Mg	953	176.298	160	−0.8969	184	−0.8987
Cd	623	158.333	170	−0.9697	168	−0.9707
Al	943	127.337	170	−2.0759	224	−2.0788
Al(IU)			160	−2.0632	196	−2.0797
Tl	588	203.182	160	−2.0868	172	−2.0873
Pb	613	217.732	160	−3.5916	232	−3.5916
Pb(IU)			160	−3.6245	194	−3.6410

Table 2. ORPA and experimental long-wavelength limit of the structure factor and comparison of the GB, ORPA and experimental excess entropy s. The experimental data for $S(0)$ are taken from [33] (alkali metals) and from [34] for the other metals. The experimental data for the excess entropy at the melting point are taken from [35]. Data for the alkali metals at high temperatures are from [36].

System	Temp. (K)	$S_{ORPA}(0)$	$S_{Expt.}(0)$	$-s_{GB}/k_B$	$-s_{ORPA}/k_B$	$-s_{Expt.}/k_B$
Li	463	0.0219	0.026	4.09	3.74	3.70/3.54
Na	373	0.0220	0.023	3.86	3.58	3.45
	473	0.0293	0.032	3.19	3.22	2.90
	573	0.0379		3.04	2.84	2.70
K	336	0.0212	0.024	3.86	3.51	3.45
	378	0.0231	0.028	3.48	3.57	3.31
	473	0.0313		3.04	2.84	2.70
Rb	312	0.0210	0.022	3.74	3.49	3.63
	373	0.0258		3.34	3.10	3.16
	473	0.0342		2.87	2.67	2.59
Cs	302	0.0179	0.024	3.74	3.60	3.56
	373	0.0235	0.035	3.34	3.12	3.06
	473	0.0312		2.87	2.68	2.48
Mg	953	0.0258	0.025	4.09	3.69	3.45/3.70
Cd	623	0.0200	0.011	3.86	3.56	4.15/3.68
Al	943	0.0255	0.017	4.41	3.35	3.60/3.61
Al(IU)		0.0182		4.20	3.91	
Tl	588	0.0132	0.010	3.98	3.30	4.05/4.31
Pb	613	0.0106	0.009	4.41	4.42	4.1/3.69
Pb(IU)		0.0087		3.98	5.13	

Since for a given density and temperature the OCP is characterized by the valence Z of the charged particles, and recalling that the typical coupling parameter for such a system is the dimensionless quantity $\Gamma = \beta e^2 Z^2/a$ (where $a = (3/4\pi\rho)^{1/3}$ is the ion sphere radius), we use Γ as reference system variational parameter in (6). The reference system data used in the present work are exactly the same as in [3]: the OCP structure and thermodynamics were evaluated only once for different values of Γ in the MHNC approximation [31]. The resulting structural and thermodynamic data agree excellently with the simulation data. They have been stored in a file and used in the ORPA code through fast table interpolation. We have verified that the results are not affected by this numerical procedure.

The numerical solution of (4 - 6) is obtained in a sequential way: we start with the value of Γ obtained from our previous GB calculations, and we choose an r_c close to the size of the exclusion hole of the corresponding OCP, as initial guess. Then, we solve the ORPA equations at fixed Γ for different values of r_c until condition (5) is satisfied with an absolute accuracy of 10^{-3}. Thus, we repeat the calculations, varying Γ by steps of 5, until we have evidence that we bracket the minimum. At this point a parabolic interpolation gives a very good estimate of the minimum. We have found that it is not necessary to go on with additional refinements of the minimum.

It would be possible to improve the efficiency of our strategy by solving (5) simultaneously with (4) and using a more efficient algorithm for solving (6) or even solving simultaneously all the equations (4-6). However, we did not exploit this possibility in the present calculations.

The thermodynamical parameters of the states we have investigated are given in Table 1.

3. Results and Discussions

In Tables 1 and 2 we show the system parameters and the calculated results for the thermodynamics of the liquid metals near their freezing points. Unless otherwise indicated, we have used the Singwi et al. (SSTL) [37] dielectric function to screen the bare ionic pseudo-potential. Only in the case of Al and Pb we have used the Ichimaru-Utsumi [38] dielectric

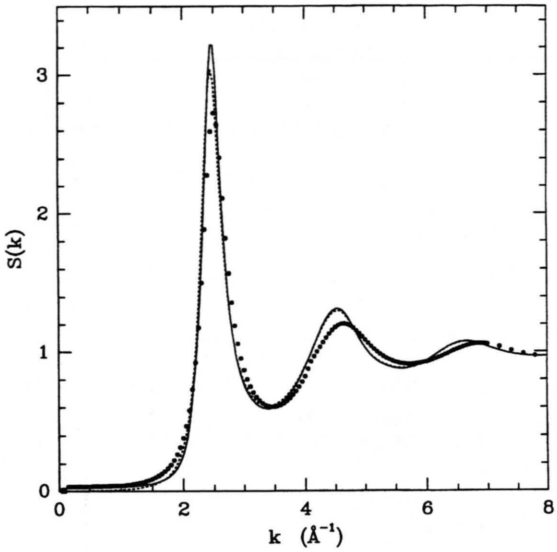

Fig. 1. Structure factor of liquid lithium close to its melting point. Full circles: experimental data (see text), dotted line: GB approximation, full line: ORPA.

function, since for those systems the change in the dielectric function results in the largest change of the atomic interaction. More refined choices for the dielectric functions are now possible [39], however we have used the same screening as in our previous GB results [3] for the purpose of direct comparison, and also because the original GNMP potential has been developed and extensively tested for liquid metals with such dielectric functions. Comparison is shown, where appropriate, with our previous GB results with OCP as reference system [3].

3.1. Alkali Metals

For the alkalis, the ORPA level optimization of the reference system yields values of the reference system coupling parameter, $\Gamma = \beta e^2/a$, generally higher than the optimal values obtained from the GB level, although the difference is not large.

The corresponding ORPA values of the free and internal energies are by a few per mill smaller than the GB values, thus confirming the weakness of the perturbation potential in the case of the alkalis described via a OCP reference system. Notice, however, that such a small change of the Helmholtz free energy corresponds to a more noticeable difference in the excess entropy and also allows us to calculate the long wavelength isothermal compressibilities. These two

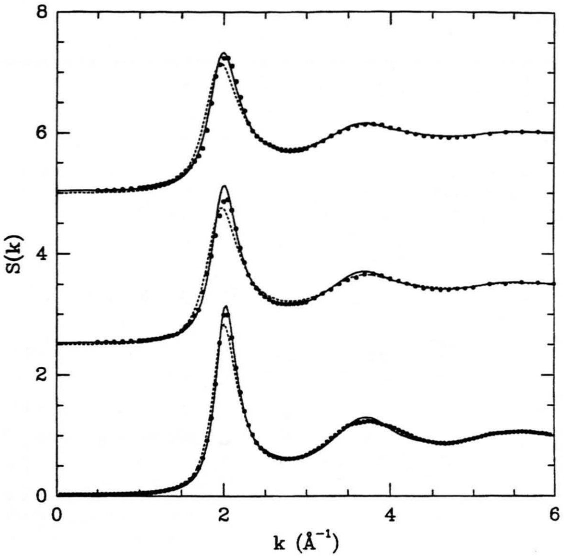

Fig. 2. Structure factor of liquid sodium at three temperatures above its melting point. The three sets of data have been shifted vertically. From the bottom: 373, 473, and 573 K data. Symbols as in Figure 1.

quantities at the GB level were given by the corresponding quantities for the OCP reference system, thus yielding a trivial $S(0) = S_0(0) = 0$ in the case of the long-wavelength isothermal compressibility. In the present calculations, the specific corrections to the excess entropy s_{ex} and to $S(k = 0)$, explicitly depending on the perturbation, give a significant improvement of the values of the excess entropy and a realistic estimation of the long wavelength compressibilities.

Some more insight into the results can be obtained by direct inspection of the ORPA structure factors shown in Figures 1 - 5. The experimental results are taken from [33] for the alkalis at freezing and [32] for higher temperatures.

By comparing the ORPA results (continuous line) with the experimental values (full circles) and those for the best OCP system as obtained from the GB variational approach (dotted line), one can make some interesting observations. We see that the modifications in the $S(k)$ calculated within the ORPA improve the long wavelength limit, shift the main peak towards higher wave-vectors and improve the position of the first minimum. All these changes result in better agreement of the calculated $S(k)$ with the experiment at the melting point, although at higher temperatures, and in particular in the case of potassium at 378 K,

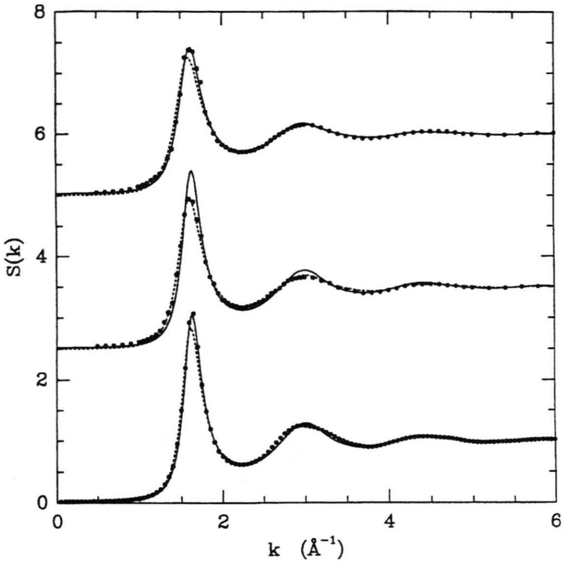

Fig. 3. Structure factor of liquid potassium at three temperatures above its melting point. From the bottom: 336, 378, and 473 K. Symbols as in Figure 1.

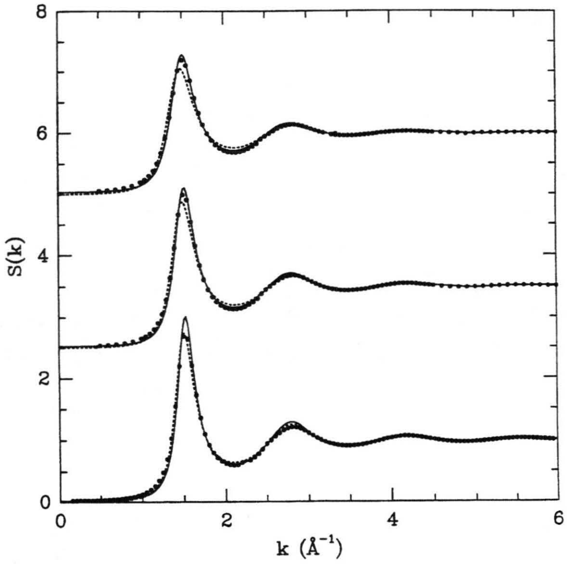

Fig. 4. Structure factor of liquid rubidium at three temperatures above its melting point. From the bottom: 312, 373, and 473 K. Symbols as in Figure 1.

the region of the first peak seems to be described with less accuracy by the ORPA results.

Globally, from our data we conclude that the variation of the structure factor with temperature shows a decreasing ability of the GNMP based pair potential

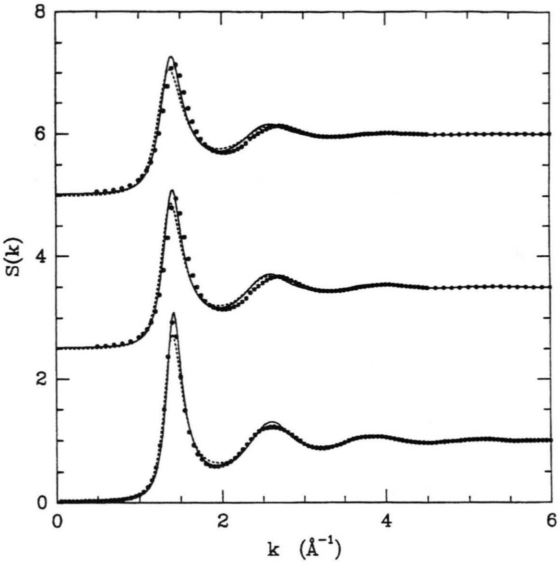

Fig. 5. Structure factor of liquid cesium at three temperatures above its melting point. From the bottom: 302, 373, and 473 K. Symbols as in Figure 1.

to model the pair correlation in real systems. Such discrepancies with respect to T are less severe however, than those observed using the very crude empty core Ashcroft potential [5, 30].

Some residual discrepancies between the rising part of $S(k)$ before the first peak and the second peak widths point towards some genuine, although small, differences between the GNMP and an effective pairwise potentials able to reproduce the experimental diffraction data.

In recent years, Chen and Lai [6] studied liquid alkali metals at freezing using the variational modified hyper-netted chain (VMHNC) closure of Rosenfeld [40, 41]. Their results, based on the same pseudopotential as used in the present study, are close to ours as far as the free energy and structure factors are concerned, although our free energies are slightly lower than theirs. In addition, our calculated excess entropies are much closer to experiment than theirs. Comparison of Chen and Lai's results for the alkali (Fig. 2 in [6]) structure factors from VMHNC and ours shows a remarkable agreement.

3.2. Polyvalent Metals

With di-, tri- or tetravalent metals, we investigate liquid metals whose experimental $S(k)$ depart more significantly from the bare OCP $S(k)$. Increasing the

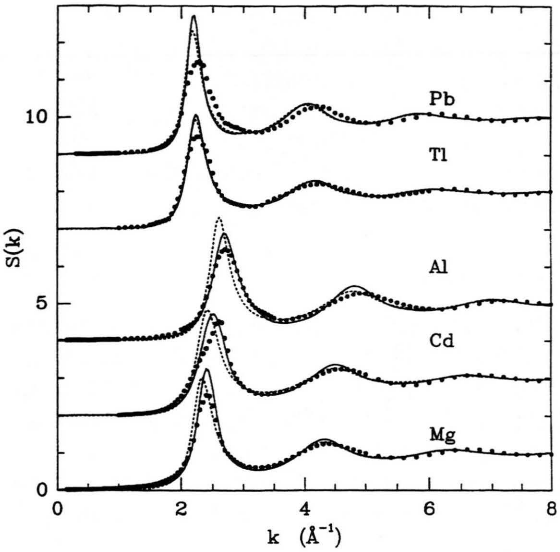

Fig. 6. Structure factor of liquid Mg (953 K), Cd (623 K), Al (943 K), Tl (588 K), and Pb (613 K). Symbols as in Figure 1.

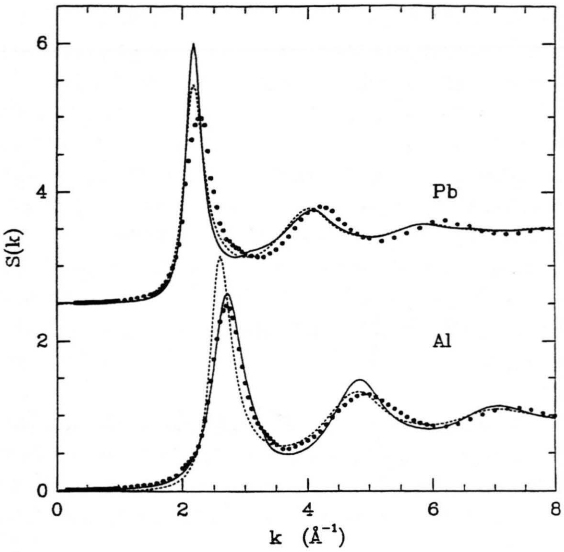

Fig. 7. Structure factor of liquid Al and Pb. GNMP potential derived from the Ichimaru-Utsumi dielectric function. Symbols as in Figure 1.

bare ionic valence, we could expect a systematic decrease of the quality of any linear screening-based effective interaction. Furthermore, the inter-ionic repulsion, as determined from the pair potentials of most of the polyvalent metals [35], is significantly stiffer than in the alkalis.

Our GB-ORPA results for Mg, Cd, Al, Tl, and Pb (Fig. 6), indistinguishable from MHNC results for the same effective potentials, show the difficulty, within the GNMP potential, to reproduce quantitatively the experimental data.

The ORPA theory performs well, since even in the case of lead ($Z = 4$) we obtain qualitatively correct results. Comparison with the GB calculations [3] shows that the modifications of $S(k)$ brought about by the ORPA are in qualitative agreement with the difference between the OCP and the experimental structure factor. The thermodynamics too shows some improvements over the GB.

Then, the observed differences between experimental data and calculations should be ascribed to inaccuracies of the used GNMP to account for the interactions in these systems.

Results obtained with different models for the interactions in polyvalent metals (see for example [42]), seem to indicate a general difficulty to get a uniform quality in the description of the structure and thermodynamics for such systems.

In order to assess the role played by the dielectric function, in the cases of Al and Pb, in addition to the SSTL screened potential, we have used potentials corresponding to the Ichimaru-Utsumi IU [38] local field $G(k)$. The resulting structure factors, shown in Fig. 7, and the corresponding thermodynamics show that some marginal improvement could be obtained.

However, our conclusion is that for such polyvalent systems with a strong pseudo-potential, we are facing the intrinsic limits of an effective potential based on a low order perturbation scheme. In such cases, where the linear response regime is not expected to describe appropriately the effective interactions, a better effective potential, including the effect of higher orders in perturbation theory, in the spirit of those derived in [11, 12] might improve significantly the agreement with the experiments as far as the polyvalent metals are concerned. Work in this direction is planned.

4. Conclusions

We have used a new consistent form of the ORPA to transcend the GB approximation and to evaluate *simultaneously* the structure and thermodynamics of mono and polyvalent metals using the GNMP of Li et al. From the methodological point of view, the quality of GB-ORPA results for the studied systems is comparable with that of the most refined integral equation theories with a relatively simpler implementation.

The overall improvement on the variational GB theory is visible and conveys specific information on the performance of the potential on quantities like the excess entropy and the long-wavelength compressibility, which are only indirectly related to the pseudo-potential at the GB level. The small modifications observed in the case of the alkalis confirm the suitability of the OCP for describing the inter-ionic correlations and the good quality of the GNMP for such systems.

The situation for polyvalent metals is less satisfactory and points to the need of a substantial refinement of the pair potential. We believe that a combined and consistent evaluation of *both* the structural and thermodynamic properties, like in recent work [7] and in the present paper, could be a fundamental tool to develop and assess a refined version of the effective interactions. In this respect, our consistent ORPA theory seems to offer a better control of the thermodynamic consistency due to its explicit origin from a free energy approximation.

Acknowledgements

O. A is grateful to SAREC for funds allowing his participation in the activities of the condensed matter group as a a regular associate of the centre. Z. B wishes to thank the International Atomic Energy Agency and UNESCO for grant allowing her participation in the Condensed Matter activities at the International Centre for Theoretical Physics in Trieste.

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