Study of the Collective Dynamics of Liquid Ni Using Pseudo-Potential Theory

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Abstract

The Phonon dispersion curves of liquid Ni near its melting temperature have been investigated using two different models: Takeno-Goda and Hubbard-Beeby. Bretonnet-Silbert (BS) pseudo-potential has been used for the interatomic potential calculation. The pair distribution function g(r) obtained by Variational Modified Hypernatted Chain (VMHNC) method. The phonon dispersion data thus obtained is in good agreement with those of Gopala and his colleagues Also the first minimum in $\boldsymbol{\omega}$(k) for longitudinal phonon modes and the first maximum in the static factor S(k) occur nearly at the same value of k according to the framework of Ziman's Formalism. In addition, stiffness constants (C_{11} and C_{44}) and compressibility have been calculated and compared with the available experimental data.

Keywords: BS Pseudopotential theory; Variational Modified Hypernatted Chain (VMHNC) method.

1. Introduction

1.1 Bretonnet and Silbert (Bs) Model of Pseudo Pot1ntial

An approximation for the simplified description of complex system effective potential is used. Pseudo Potential offers a calculation tool that increases the range of problems in solid that can be brought within the computational reach.

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There are many pseudo potential model for liquid transition metals. As we have used Bretonnet-Silbert (BS) model in our calculation, we discuss it. Bretonnet and Silbert (BS) have proposed a model potential [1] to describe s and d bands of a liquid transition metal. The bare potential is written as

$$W_i(r) = \begin{cases} \displaystyle\sum_{m=1}^{2} B_m^{(i)} \exp\left(\dfrac{-r}{ma_i}\right) & if\ r < R_{ci} \\[3mm] \dfrac{-z_i e^2}{r} & if\ r > R_{ci} \end{cases} \tag{1}$$

Where, R_c is the core radius, a is the softness parameter, Z is the effective s-electron occupancy number and B_1 and B_2 are the Dirichlet coefficients.

1.2 Vmhnc Theory

For brief review of the main features of VMHNC, the initial equation of all integral equation theories of liquid can written as [5,6,7]

$$h(r) = c(r) + \rho \int dr' h(|r - r'|) c(r) \tag{2}$$

Where, c(r) is direct correlation function, $h(r) = g(r) - 1$, where h(r) is total correlation function(r) is pair distribution function.

This relation can be written as

$$c(r) = h(r) - \log\{g(r) e^{\beta\varphi(r) + \beta(r)}\} \tag{3}$$

Where, ϕ is interatomic potential, β is $(kT)^{-1}$ the inverse of temperature times the Boltzmann constant $\beta(r)$ bridge function.

So the configurational part of the VMHNC free energy functional is,

$$f^{VMHNC}(\beta, \rho, \eta) = f^{VMHNC}(\beta, \rho, \eta) - \Delta^{(0)}(\eta) \tag{4}$$

taking,

$$\Delta^{(0)}(\eta) = f_{PY}^{VMHNC}(\eta) - f_{pyv}(\eta) - \partial_\emptyset(\eta)$$

$\partial_\emptyset(\eta)$ is a fittimg function where $\partial_\emptyset(\eta) = f_{cs}(\eta) - f_{pyv}(\eta)$

Where, $f_{pyv}(\eta)$ and $f_{cs}(\eta)$ the py virial and free energy.

1.3 The Takeno-Goda (TG) Model

The effective potential and pair distribution function *g(r)* are then used to calculate the longitudinal and

transverse phonon frequencies. The product of the static pair-correlation function '$g(r)$' and the second derivative of the interatomic potential $V(r)$ is peaked at 'σ', which is the hard sphere diameter. The longitudinal phonon frequency $\omega_L(k)$ and transverse phonon frequency $\omega_T(k)$ based on harmonic approximation according to the Takeno-Goda (TG) model [1,2] are given by,

$$\omega_L^2(k) = \left(\frac{4\pi\rho}{M}\right)\int_{r=0}^{\infty} dr\, g(r)\left[rv'(r)\left(1 - \frac{sin(kr)}{kr}\right) + \{r^2v''(r) - rv'(r)\}\right.$$
$$\left. \times\left\{\frac{1}{3} - \frac{sin(kr)}{kr} - \frac{2cos(kr)}{(kr)^2} + \frac{2sin(kr)}{(kr)^3}\right\}\right] \tag{5}$$

$$\omega_T^2(k) = \left(\frac{4\pi\rho}{M}\right)\int_{r=0}^{\infty} dr\, g(r)\left[rv'(r)\left(1 - \frac{sin(kr)}{kr}\right) + \{r^2v''(r) - rv'(r)\}\right.$$
$$\left. \times\left\{\frac{1}{3} + \frac{2cos(kr)}{(kr)^2} - \frac{sin(kr)}{(kr)^3}\right\}\right] \tag{6}$$

1.4 The Hubbard-Beeby Model

The longitudinal phonon frequency $\omega_L(k)$ and transverse phonon frequency $\omega_T(k)$ are given by the expressions due to HB [3],

$$\omega_L^2(k) = \omega_E^2\left[1 - \frac{3sink\sigma}{k\sigma} - \frac{6cosk\sigma}{(k\sigma)^2} + \frac{6sink\sigma}{(k\sigma)^3}\right] \tag{7}$$

$$\omega_T^2(k) = \omega_E^2\left[1 - \frac{3cosk\sigma}{(k\sigma)^2} + \frac{3sink\sigma}{(k\sigma)^3}\right] \tag{8}$$

Where,

$$\omega_E^2 = \frac{4\pi n}{3M}\int_0^{\infty} g(r)r^2V''(r)dr \tag{9}$$

ω_E is the maximum phonon frequency. V'' is the second derivative of the pair potential. M is the atomic mass. Both phonon frequencies, i.e. transverse and longitudinal are proportional to the wave vector (k) and obey the relationships in the long wavelength limit of the frequency spectrum,

$$\omega_L \propto k \quad and \quad \omega_T \propto k,$$

$$or, \omega_L = V_Lk \quad and \quad \omega_T = V_Tk \tag{10}$$

Where V_L is the longitudinal sound velocity and V_T is the transverse sound velocity. Various elastic properties are then determined by the longitudinal and transverse phonon frequencies. We have calculated the stiffness constant C_{11}, C_{44} for *Ni* at melting temperature using the phonon frequency data in long wave length limit, from Schofield's equation. And also compressibility has been calculated, the elastic stiffness constants C_{11} and C_{44}

are related to the angular frequency (ω) and the wave vector (k) for longitudinal wave and transverse wave through the following equations respectively [14]

$$\omega^2 \rho = C_{11} k^2 \tag{11}$$

$$\omega^2 \rho = C_{44} k^2 \tag{12}$$

where, ρ is the mass density.

The elastic stiffness constants can also be calculated from Schofield's equations [15]. These are related to stiffness constants C_{11} and C_{44} through the following equations [16]

$$C_{11} = Q k_B T \left(3 + 2 \frac{I_1}{15} + \frac{I_2}{5} \right) \tag{13}$$

$$C_{44} = Q k_B T \left(1 + 4 \frac{I_1}{15} + \frac{I_2}{15} \right) \tag{14}$$

Thus the integrals I_1 and I_2 are calculated through the use of the RDF and the potential energy derivatives.
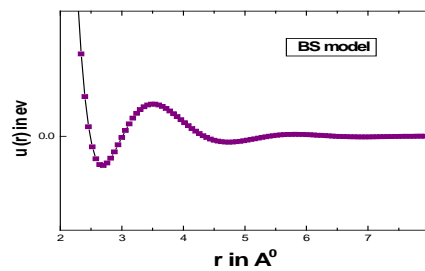
2. Materials and Method

Figure 1: Effective potentials for liquid Ni Obtained from BS model Using the following parameterizations (T=1773).
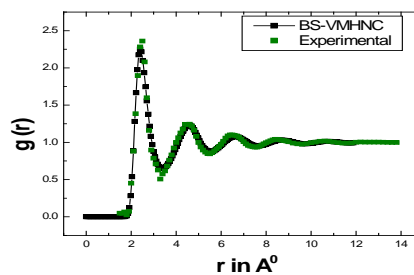

Figure 2: Pair distribution function $g(r)$ for liquid *Ni* at melting temperature (1773 K) calculated by using BS model potentials along with VMHNC.

Table 1: The values of the BS model parameter r_{cut}, Z, α , thermodynamic inputs temperature T and ionic number densities n [12, 13] are given in Table:1

System	T(K)	r_{cut}	α (au)	n $(\dot{A}^{-3})$	z
Ni	1773	1.060	0.220	0.0792	1.5

Collective dynamics results specifically the curve of longitudinal and transverse frequencies with respect to wave number k. The dispersion curves were generated using the method of Takeno Goda [2,3], Hubbard-Beeby [4].
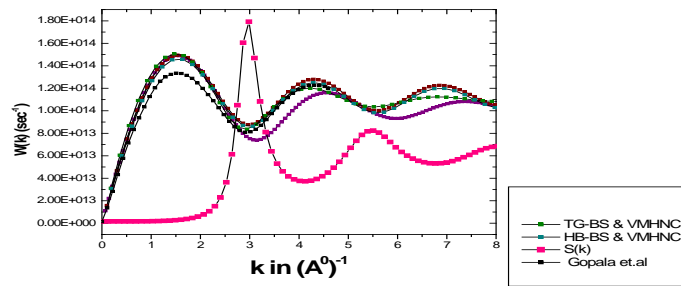

Figure 3: Phonon Dispersion (Longitudinal) curve for liquid *Ni* at melting temperature

Phonon Dispersion (**Longitudinal**) From the phonon dispersion curve of Ni, it appears that the first minimum in $\omega(k)$ for longitudinal phonon modes and the first maximum in the static structure factor S(k) occur nearly at the same value of $k \approx 3\left(A^0\right)^{-1}\left[\approx 2k_F\right]$.This is within the framework of Ziman's formalism [11]. It is seen that $\omega_L^{max}(k)$ occurs midway between zero and the structure factor maximum. That is the position of the maxima of the longitudinal phonon mode for Ni at $k \approx 1.5\left(A^0\right)^{-1}\left[\approx k_F\right]$ position of the first minimum of ω_l is found to be near the position of the first peak of the structure factor data as predicted by Takeno-Goda. Oscillations are presented in $\omega_L(k)$ up to large values of **k** showing the collective nature of these excitons. However, $\omega_L(k)$ does not show any oscillations. It can be noted that the structure factor S (k) maxima corresponds to minima in $\omega_L(k)$. Thus the first maximum of S (k) coincides with the first minimum of $\omega_L(k)$ while the first minimum of S (k) coincides of 2nd maximum of $\omega_L(k)$. Therefore when the correlation in S(k) is strong as exhibited by the peaks, the corresponding frequency is less.
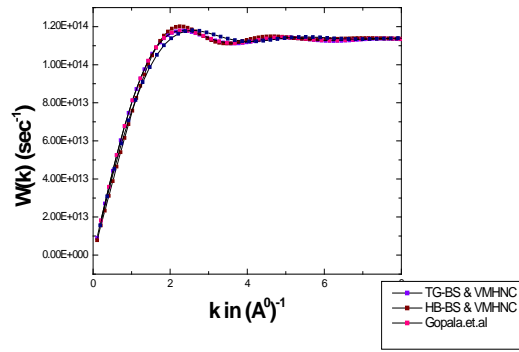
Figure 4: Phonon Dispersion (Transverse) curve for liquid *Ni* at melting temperature

The peak of transverse frequency appears at a higher wave vector than that of the longitudinal frequency and exhibits no significant oscillatory behavior. The principal peak of the transverse phonon mode for *Ni* occurs at

$$k \approx 2.20\left(A^o\right)^{-1}$$

2.1 Elastic Properties: Stiffness Constants And Compressibility

stiffness constant C_{11}, C_{44} for *Ni at* melting temperature are calculated. These constants have been calculated using the phonon frequency data in long wave length limit, from Schofield's equation. a = Ref. [18].

Table 2: Stiffness and compressibility obtained for liquid Ni at melting temperature

Metal	Stiffness constant	From phonon frequency	From Scofield's equation	Gopala and his colleagues	Experimental Value [a]
Ni	C_{11}	.83	.88	.84	1.02
	C_{44}	.38	.32	.29	
	Compressibility				
	From C_{11}	1.20	1.13	1.18	.98

3. Conclusion

In conclusion we would like to summarize the main points of this work. We have presented a comprehensive study of the collective dynamics knows as Phonon of Ni at melting temperature. A better agreement can be obtained if we can do a better parameterization to obtain the pair-potential and use that to get g(r), however data obtained as above agrees well with each other and also with that of Gopala and his colleagues. The Stiffness Constants and Compressibility have been calculated and the values are found to be in good agreement with each other.

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