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PHONON DISPERSION CURVES OF LIQUID METALS

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In the present paper, the phonon dispersion curves (PDC) of some alkali metals are reported in second order approach through the equation given by Hubbard and Beeby (HB). The pair correlation function $g(r)$ is directly computed from the interatomic pair potential, which is used in the present computation. Two different forms of local field correction functions proposed by Hartree (H) and Ichimaru-Utsumi (IU) are used in the present study the screening dependence of the phonon frequencies in the metallic elements. Present results of phonon dispersion curves are found to be in qualitative agreement with available experimental results.

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1. Introduction

The problem of an appropriate description of the structure and related properties of liquids arise from their intermediate situation between ideal gases and solids. It is well known that crystalline solids having long range order are completely characterized by their symmetry properties whereas liquids having no such periodicity and can be characterized only by distribution or correlation functions. In liquids the interpretation of neutron inelastic scattering measurements is more complicated than it is in the case of solids, largely because there is no long range order, neither in space nor in time. At low frequencies, the liquids behave as a viscous medium but at higher frequencies its response is elastic, the system behaves like a solid and transverse excitations are supported [1-10]. Recently, Pratap *et al.* [5] and Thakor *et al.* [6] have reported phonon dispersion curves for liquid alkali metals theoretically. While, Jong *et al.* [11] and Pilgrim *et al.* [12] have studied the phonon dispersion curves (PDC) of Li and Na alkali metals experimentally.

Collective excitations in fluids have been studied experimentally, theoretically and by computer simulations for almost several decades. Lot of effort has been put to study the dynamical properties of liquid metals [1-10] both theoretically and experimentally. The investigation of collective modes in liquids has comparatively received less attention. Some few researchers [1-8] have reported the phonon dispersion curves of simple liquid metals. It was found that the maximum deviation takes place in the vicinity of the first spherical Brillouin zone. This region lies nearly at the half distance of the first peak in the structure factor $S(q)$. Thus, the choice of structure factor $S(q)$ also plays a vital role in the study of lattice dynamics of liquid metals. In the present work we have used the pair correlation function $g(r)$ instead of structure factor $S(q)$ calculated using the interatomic pair potential to give the calculation a flavour model potential, which is a beauty of our present paper.

The present article deals with the computation of the phonon dispersion curves (PDC) of some alkali metals with the aim to explore the applications of model potential of Gajjar *et al.* [13] for the first time. The choice of the model potential form factor is certainly an important factor in the study of metallic properties and its actual form is much more sensitive to the choice of the local field correction functions of the electron gas. Hence, the purpose of the present article is not only to study the phonon dispersion curves (PDC), but also to see the influence of the various local field correction functions in the screening. Therefore, we have adopted here two different types of local field correlation functions viz. Hartree (H) [14] and Ichimaru-Utsumi (IU) [15]. Also, we have used here Hubbard and Beeby (HB) [16] approach for studying the phonon dispersion curves (PDC) of liquid alkali metals.

2. Computational methodology

The interatomic pair potential $\Phi(r)$ is calculated from the relation given by [17, 18],

$$\Phi(r) = \left(\frac{Z^2 e^2}{r}\right) + \frac{\Omega_O}{\pi^2} \int F(q) \left[\frac{Sin(qr)}{qr}\right] q^2\, dq\,. \tag{1}$$

Where, Z and Ω_0 are the valence and atomic volume of the metallic elements, respectively. The energy wave number characteristics appearing in the Eqs. (1) is written as [17, 18],

$$F(q) = \frac{-\Omega_O q^2}{16\pi} \left|W_B(q)\right|^2 \frac{\left[\varepsilon_H(q) - 1\right]}{\left\{1 + \left[\varepsilon_H(q) - 1\right]\left[1 - f(q)\right]\right\}}\,. \tag{2}$$

Here $W_B(q), \varepsilon_H(q),\ f(q)$ are the bare ion potential, the Hartree dielectric response function and the local field correction functions to introduce the exchange and correlation effects, respectively.

The Hartree (H) screening function [14] is purely static, and it does not include the exchange and correlation effects. The expression of it is,

$$f(q) = 0\,. \tag{3}$$

The Ichimaru-Utsumi (IU) local field correction function [15] is a fitting formula for the dielectric screening function of the degenerate electron liquids at metallic and lower densities, which accurately reproduces the Monte-Carlo results as well as it also, satisfies the self consistency condition in the compressibility sum rule and short range correlations. The fitting formula is

$$f(q) = A_{IU}\left(\frac{q}{k_F}\right)^4 + B_{IU}\left(\frac{q}{k_F}\right)^2 + C_{IU} + \left[A_{IU}\left(\frac{q}{k_F}\right)^4 + \left(B_{IU} + \frac{8A_{IU}}{3}\right)\left(\frac{q}{k_F}\right)^2 - C_{IU}\right]\left\{\frac{4 - \left(\frac{q}{k_F}\right)^2}{4\left(\frac{q}{k_F}\right)} \ln\left|\frac{2 + \left(\frac{q}{k_F}\right)}{2 - \left(\frac{q}{k_F}\right)}\right|\right\}. \tag{4}$$

The parameters A_{IU}, B_{IU} and C_{IU} are the atomic volume dependent parameters of IU local field correction functions. The mathematical expressions of these parameters are narrated in the respective papers of the local field correction function [15].

The bare-ion pseudopotential due to Gajjar *et al.* [13] is given by

$$W_B(q) = \frac{-8\pi Z}{\Omega_O q^2}\left(\cos(q r_C) - \frac{(q r_C)^2}{\left(1 + (q r_C)^2\right)}\right). \tag{5}$$

here, Z and r_C are the valence and parameter of the model potential, respectively. The details of the model potential are narrated in the literature [13].

To compute the phonon dispersion relations of liquid metals, the most frequently used approach of Hubbard and Beeby (HB) [16] is adopted. With the physical argument that the product of the static pair correlation function $g(r)$ and the second derivative of the interatomic pair potential $\Phi(r)$ is peaked near the hard sphere diameter σ, Hubbard and Beeby (HB) [14] have derived the expressions for the longitudinal phonon frequencies $\omega_L(q)$ and the transverse phonon frequencies $\omega_T(q)$ as [16],

$$\omega_L^2(q) = \omega_E^2 \left[1 - \frac{\sin(q\sigma)}{q\sigma} - \frac{6\cos(q\sigma)}{(q\sigma)^2} + \frac{6\sin(q\sigma)}{(q\sigma)^3} \right], \tag{6}$$

and

$$\omega_T^2(q) = \omega_E^2 \left[1 - \frac{3\cos(q\sigma)}{(q\sigma)^2} + \frac{3\sin(q\sigma)}{(q\sigma)^3} \right]. \tag{7}$$

with $\omega_E^2 = \left(\frac{4\pi\rho}{3M} \right) \int_0^\infty g(r)\Phi''(r) r^2 dr$ is the maximum frequency.

Where, ρ, M, $g(r)$ and $\Phi(r)$ are the number density, atomic mass, pair correlation function and interatomic pair potential of the element, respectively.

The fundamental ingredient, which goes into the calculation of the phonon dispersion curves (PDC) of liquid metals, is the interatomic pair potential $\Phi(r)$. In the present study, the interatomic pair potential $\Phi(r)$ is computed from Eqs. (1). A quantity which is equally important as the interatomic pair potential Φ(r) while studying a disorder system is the pair correlation function (PCF) $g(r)$. It provides the statistical description of the structure of the system under investigation. The complete information of the precise position and momentum of each particle at each instant of time is contained in this function. The function $g(r)$ can be obtained either experimentally by X-ray diffraction and neutron diffraction technique or computed theoretically from the interatomic pair potential $\Phi(r)$ [19]. Instead of using experimentally available $g(r)$, here the pair correlation function for all disordered systems are generated from presently obtained interatomic pair potential $\Phi(r)$. The function $g(r)$ is presently calculated using the expression [19],

$$g(r) = \exp\left[\frac{-V(r)}{k_B T} \right] - 1. \tag{8}$$

Here k_B is the Boltzmann’s constant and T the room temperature of the system under investigation.

3. Results and discussion

The constants and parameters used in the present computations of the phonon dispersion curves (PDC) of the liquid alkali metals are tabulated in Table 1. The computed phonon dispersion curves of liquid alkali metals are displayed in Figures 1-5. Here, also it may be seen that the dispersion of the longitudinal phonon exhibits oscillatory behaviour extending to the large wave vector transfer region. But in the case of transverse phonon, the oscillatory behaviour seems quite insignificant for high q value. This indicates that the transverse phonon undergoes larger thermal modulation than the longitudinal phonon, which may be connected with the instability of transverse phonons in liquids. The ω→q curves for transverse phonons attain maxima at a high q value than the longitudinal phonon curve. The influence of the exchange and correlated motion of electron through various local field correction functions lowers the phonon modes more than those due to static Hartree (H) effect. The inclusion of local field correction does not affect the position of the maxima, minima and the crossing of ω_L and ω_T modes, very significantly. The position of the first minimum roughly coincides with the first peak in the structure factor of the respective systems. The computer simulations and analytical calculations have demonstrated that this minimum arises from a process analogous to the Umklapp scattering in the crystalline solids. This sharp first maximum in the static structure factor acts like a smeared-out reciprocal lattice vector. The experimental or theoretical data of most of the alkali metals are not available in the literature. But, the behaviour of the present results does not show any abnormality. From the Figures 1-5, it can be noted that when we go from Li→Cs, the peak of the phonon dispersion curves reduces. Also, we have compared our results of phonon dispersion curves of Li and Na alkali metals with available experimental results of Jong *et al.* [11] and Pilgrim *et al.* [12] and found qualitative agreement with them. Also, the present results are found superior than the experimental data. From the Fig. 1-5, it is seen that, the present results obtained from H-local field correction function show higher values in comparison with IU-local field correction function for most of the alkali metals. The experimental or theoretical data for K, Rb and Cs metallic complexes are not available for further comparison.

Table 1. Constants and parameters for alkali metals.

Metal	Z	k_F (au)	$\Omega_O\,(\mathrm{au})^3$	r_C (au)
Li	1	0.5890	144.9	0.7738
Na	1	0.4882	254.5	1.0765
K	1	0.3947	481.4	1.3880
Rb	1	0.3693	587.9	1.4837
Cs	1	0.3412	745.5	1.9108

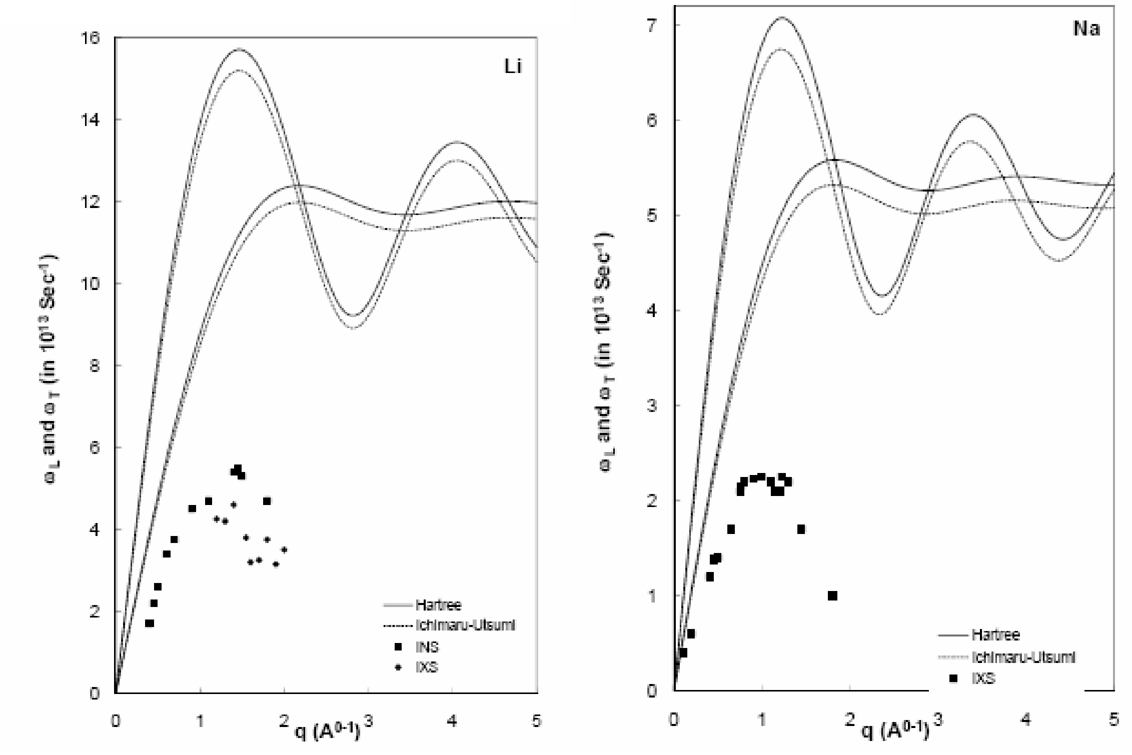

Fig. 1. Phonon dispersion curves of liquid Li.

Fig. 2. *Phonon dispersion curves of liquid Na.*

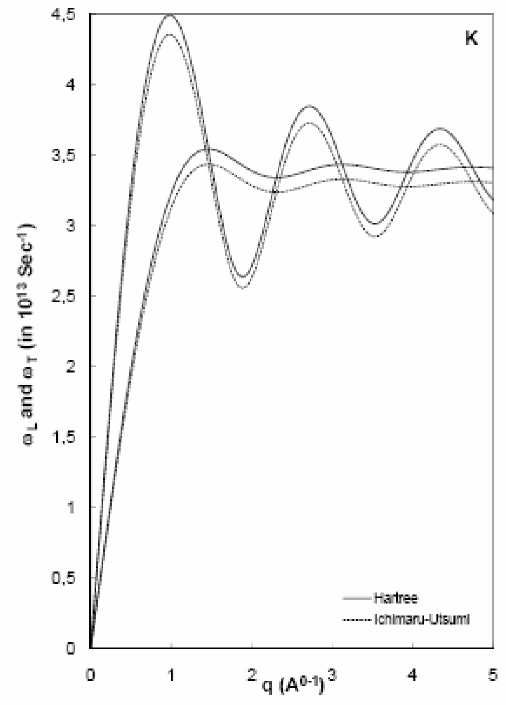

Fig. 3. Phonon dispersion curves of liquid K.

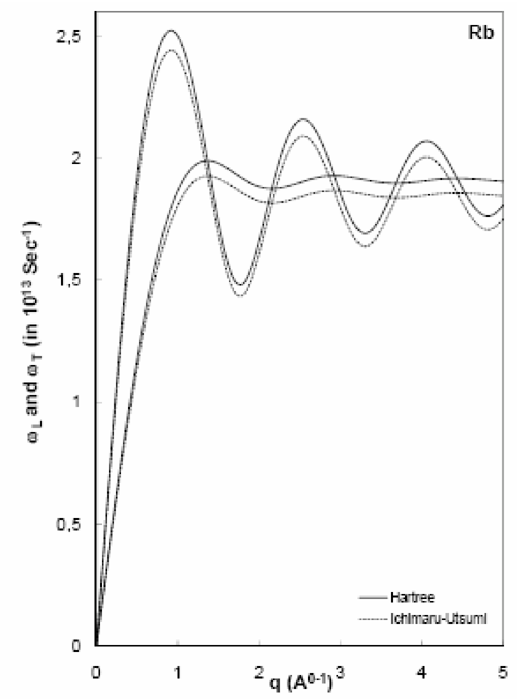

Fig. 4. Phonon dispersion curves of liquid Rb.

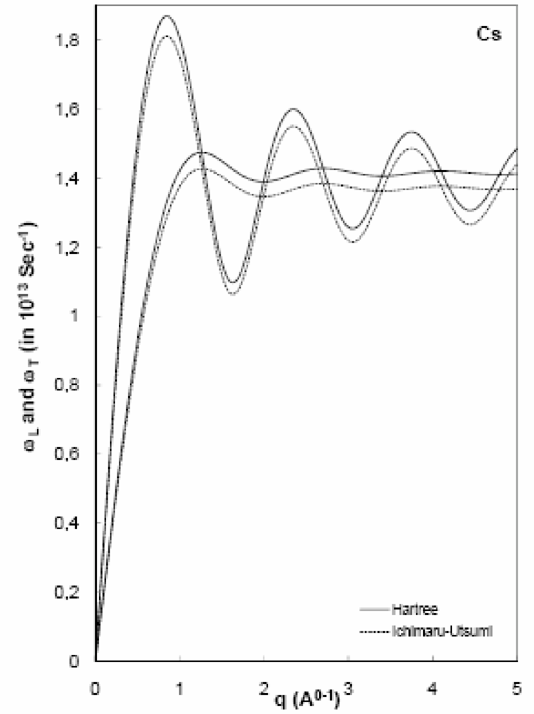

Fig. 5. Phonon dispersion curves of liquid Cs.

4. Conclusions

At the end, we conclude that the presently computed results of the phonon dispersion curves (PDC) of liquid alkali elements are showing consistent nature. The present results of phonon dispersion curves (PDC) for Li and Na are shown qualitative agreement with available experimental data and also found superior with them. The experimental data for another liquid metals are not available for further comparison. Thus, in the absence of experimental results such calculations may be considered as one of the guidelines for further theoretical or experimental investigations. This is very much essential for obtaining concrete conclusions. Also, the model potential along with H and IU local filed correction functions is capable of explaining the phonon dispersion curves (PDC) of liquid alkali metals. From the present experience, we also conclude that it should be interesting to apply other local pseudopotentials for such comprehensive study to judge and confirm the wider applicability of the potential.

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