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Correlated motion of particles in liquid sodium metal

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ABSTRACT

The correlated dynamical modes of liquid alkali metal, liquid sodium, has been theoretically calculated at temperature 388.4 K for six different wave-vector values: *2.4 nm^{-1}, 6.96 nm^{-1}, 22.4 nm^{-1}, 44.8 nm^{-1}, 68.8 nm^{-1}, 88.0 nm^{-1}*. At this temperature, which is just near to its melting point, the fluid under consideration is essentially a classical system of interacting particles and hence, the modified microscopic theory of fluids is applied for the present theoretical investigation. The microscopic theory carries expressions for spectral line shape of dynamical structure factor, $S(\kappa,\omega)$, where $\hbar\kappa$ and $\hbar\omega$ are momentum and energy transfers respectively, which is further related to various physical quantities like velocity of sound and dispersion relation and hence yields the complete dynamics of a given fluid. Detailed dynamical structure factors, $S(\kappa,\omega)$, have been computed in the entire κ -range and are compared with the scale reduced experimental values obtained from IXS(inelastic x-ray scattering) spectra of liquid lithium. The two are found to be in good agreement with each other. Variations of collective mode frequencies and velocity of sound with wave-vector have also been reported.

KEY WORDS: microscopic theory, dynamical structure factor, diffusion coefficient, correlation functions, static structure factor

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1. INTRODUCTION

Fluids comprise a disordered system of correlated particles and the degree of correlations depends upon the various physical parameters. Dynamics of such disordered systems is quite involved and is hard to study. However, the collective excitations of fluids can be observed experimentally when they are perturbed by some external probe like neutron or photons. Data obtained from inelastic scattering of neutrons, however contains a complicated blend of coherent and incoherent cross-sections whereas inelastic x-ray scattering predominantly yields coherent scattering cross-sections. From so obtained double differential scattering cross-sections, the detailed coherent dynamical structure factors, $S(\kappa,\omega)$, can be extracted which are able to explain the complete collective dynamics of any fluid.(Here, κ and ω refers to wave vector and frequency respectively). In the present communication, an attempt has been made to explain the coherent dynamics of a liquid alkali metal, liquid sodium, near its melting point at 388.4 K(melting point of Na is 370 K). At this temperature, liquid Na constitute a dense system of 0.0237 atoms per $Å^3$ whose inter-particle separation is much larger than its thermal de-broglie wavelength, λ_{th} i.e. $2r_s \gg \lambda_{th}$ (r_s, being the radius of a spherical Na atom). Thus, liquid sodium at 388.4 K and density 0.0237 atoms/ $Å^3$ behaves as a classical liquid and its dynamics can be explained using modified microscopic theory of liquids.

Microscopic theory of liquids evaluates the dynamical disorders generated due to some external perturbation by solving classical equations of motion of constituent particles then taking averages of their trajectories over full time span. Solution of equations of motion evolves density-density response function which is further related to the imaginary part of dynamical structure factor, through fluctuation-dissipation theorem. Dynamical structure factor, $S(\kappa,\omega)$, is Fourier transform of space-time dependant correlation function $G(r,t)$ which at $t \to 0$ is generalized to the static pair correlation function $g(r)$. The dynamical structure factor, thus, provides the complete information about the dynamics of a fluid.

The dynamical structure factor so obtained exhibit sharp peaked structures with finite widths. This is unlike the real feasible picture for fluids where these structures are more damped. This gap has been filled by including a characteristic relaxation time, a parameter which is related to inter-particle interaction, mass, temperature, density and diffusion co-efficient. This form of the microscopic theory has been used to explain the dynamics of various fluids[1,2,3,4]. The theory, however, has been further modified to incorporate all time behaviour of realistic intermediate scattering functions. This modified theory has recently been used to successfully explain the coherent dynamics of a wide variety of liquids[5,6,7] and now has been used in the present investigation.

2. MATHEMATICAL FORMALISM

General expression for dynamical structure factor in the present theory is turned to be:

$$S(\vec{k},\omega)=\frac{k^2}{\pi m\beta}\left\{\left[\begin{array}{c}\left[1+\omega_k^2\dfrac{(\beta''+\gamma)^2-\omega^2}{\left[(\beta''+\gamma)^2+\omega^2\right]^2}\right]\dfrac{2\beta''}{(\beta''^2+\omega^2)^2}-\\[2mm]\omega_k^2\dfrac{(\beta''^2-\omega^2)}{(\beta''^2+\omega^2)^2}\dfrac{2(\beta''+\gamma)}{\left[(\beta''+\gamma)^2+\omega^2\right]^2}\end{array}\right]\times\left\{\left[1+\omega_k^2\dfrac{(\beta''+\gamma)^2-\omega^2}{\left[(\beta''+\gamma)^2+\omega^2\right]^2}\right]^2+\left[\omega_k^2\dfrac{2\omega(\beta''+\gamma)}{\left[(\beta''+\gamma)^2+\omega^2\right]^2}\right]^2\right\}^{-1}\right\}$$ (1)

where $\beta''=D_{eff}k^2=D(\omega)k^2$, $D(\omega)$ being the ω-dependent self diffusion coefficient and 'm' is atomic mass.

Characteristic frequency, ω_k is given as :

$$\omega_k^2=\omega_E^2\left[1-\frac{3\sin kr_0}{kr_0}-\frac{6\cos kr_0}{(kr_0)^2}+\frac{6\sin kr_0}{(kr_0)^3}\right]$$ (2)

Here, ω_E, is maximum elastic frequency which is related to inter atomic interaction potential *V(r)* and static pair correlation function *g(r)* as follows:

$$\omega_E^2=\frac{4\pi\rho_0}{3m}\int\left(\frac{\partial^2 V}{\partial r^2}+\frac{2}{r}\frac{\partial V}{\partial r}\right)g(r)r^2 dr$$ (3)

$$\gamma(\kappa)=[\tau(\kappa)]^{-1}=\frac{\omega_\kappa}{\left[(\kappa^2/m\beta\,S(\kappa)\beta'^2)-1\right]^{1/2}}-\beta'$$ (4)

where $\beta'=D\kappa^2$, D being the κ-dependent self diffusion coefficient and $\beta=(k_BT)^{-1}$.

longitudinal current-current correlation function , $J_l(\kappa,\omega)$, is related to dynamical structure factor by following expression:

$$J_l(\kappa,\omega)=\frac{\omega^2}{\kappa^2}S(\kappa,\omega)$$ (5)

3. RESULTS AND DISCUSSION

As is discussed in prior sections, liquid sodium is a classical system of interacting correlated particles. To work out the dynamics of this fluid system, the quantitative form of an interaction potential is required. In the following computations, interaction potential, *v(r)*, for liquid sodium as given by Paskin and Rahman[8], which explains the experimental static structure factor[9], has been used. The potential for liquid Na is shown in Figure1 with solid curve(————) and exhibit the peculiar behaviour for metallic potentials, consists a soft core part associated with a repulsive oscillatory part. Inter-atomic potentials for two other alkali metals, liquid caesium and liquid

rubidium have also been plotted in Figure1 with dash dot curve(− · − · −) and dotted curve(⋯⋯⋯⋯) respectively, near their melting points. Though all these alkali metals show oscillatory behaviour of *v(r)*, the cycle of oscillations is smaller in Na which can be attributed to its smaller atomic size.
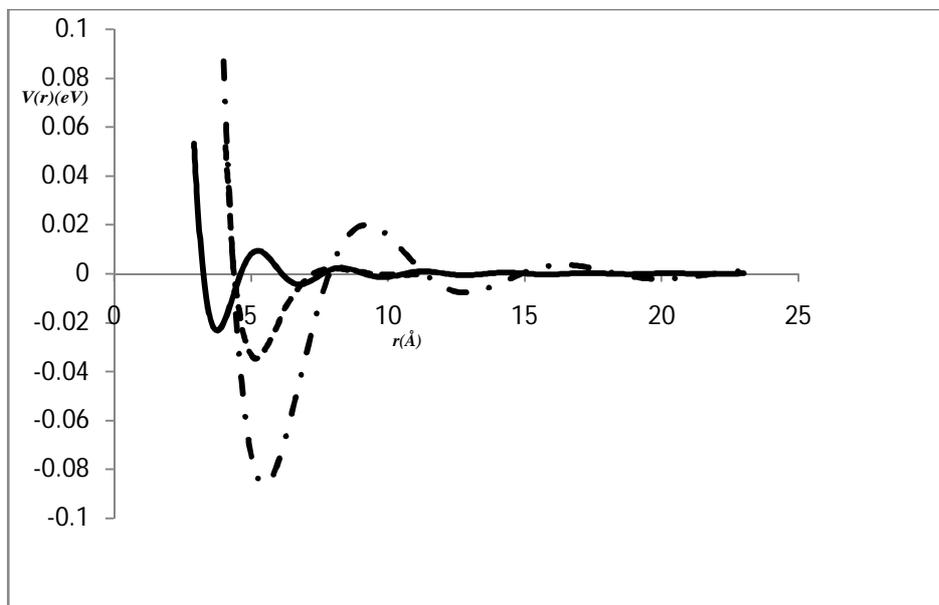
Figure1: Variation of inter-atomic potential V(r) with inter particle separation *r*, for lq. Na(⎯) ; lq. Cs(− · −);lq. Rb (⋯⋯).

With this form of inter-atomic potential and pair correlation function $g(r)$[9], r_o and Einstein frequency, ω_E, are calculated using expression(3) and turns to be 0.35nm and 13.1 ps^{-1} respectively. These are further used in expression (2) to evaluate ω_k for different values of wave-vector, κ. These computed ω_k values along with other physical quantities like static structure factor, mass, temperature, diffusion coefficient are substituted in expression (1) to evaluate dynamical structure factor, for six different values of wave vector, κ: 2.4 nm^{-1}, 6.96 nm^{-1}; 22.4 nm^{-1}; 44.8 nm^{-1}; 68.8 nm^{-1} and 88.0 nm^{-1} .One may notice that the only parameter involved in the process is diffusion coefficient which is not an arbitrary but is a feasible parameter for a given fluid. The computed results for $S(\kappa,\omega)$ have been plotted in Figure2 for, κ= 2.4 nm^{-1}, 6.96 nm^{-1}; 22.4 nm^{-1} with solid curve(⎯⎯⎯) along with $S(\kappa,\omega)$ calculated using earlier microscopic theory[4], with dashed curve(- - - - -). The computed values are compared with the scale reduced experimental values of IXS spectra of liquid lithium[10], near its melting point, with solid circles(●●●),[keeping in view the unitary description of the dynamical features of liquid alkali metals[11]. Also, the variation of diffusion coefficient, $D(\omega)$, with ω for these three values of κ have been plotted in Figure2, with solid curve(⎯⎯⎯), dashed curve(- - - - -)and dash dotted curve(− · − · −) for κ, 2.4 nm^{-1}, 6.96 nm^{-1}, 22.4 nm^{-1} respectively. In Figure3 similar results have been plotted for κ=44.8 nm^{-1}; 68.8 nm^{-1} and 88.0 nm^{-1}.
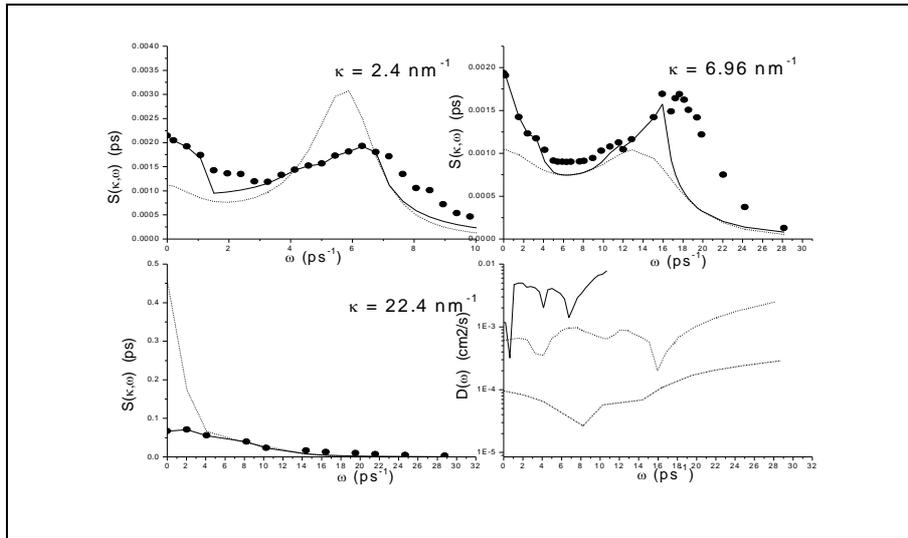
Figure2: Variation of dynamical structure factor, *S(k,ω)*with frequency *ω* for different values of *k:* (——) present calculations using modified microscopic theory, (- - - -) calculation using previous microscopic theory; (●●●) scale reduced experimental data. Variation of *D(ω)* with *ω* for: *k=2.4 nm⁻¹*(——); *k=6.96 nm⁻¹*(- - - -); *k=22.4 nm⁻¹*(– · –).
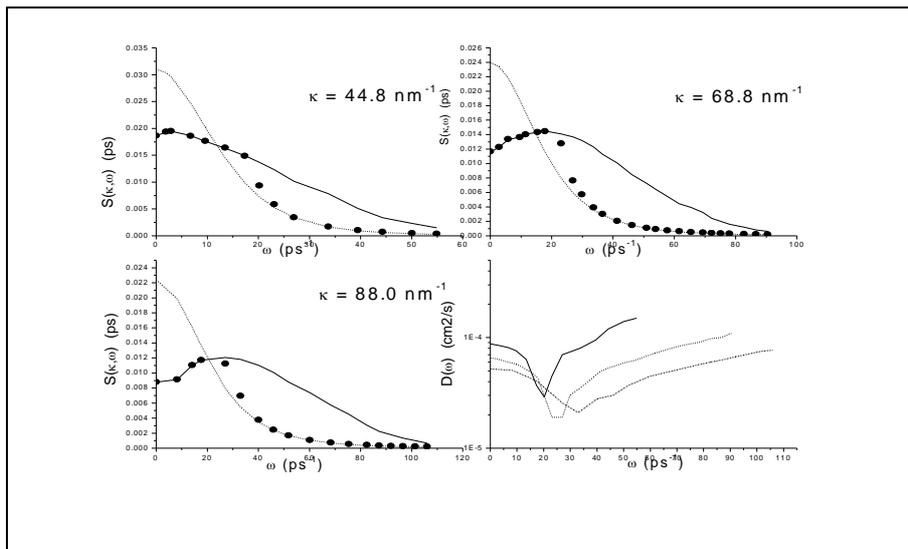
Figure3: Variation of dynamical structure factor, *S(k,ω)*with frequency *ω* for different values of *k:* (——) present calculations using modified microscopic theory, (- - - -) calculation using previous microscopic theory; (●●●) scale reduced experimental data. Variation of *D(ω)* with *ω* for: *k=44.8 nm⁻¹*(——); *k=68.8 nm⁻¹*(- - - -); *k=88.0 nm⁻¹*(– · –).

As is evident from figure1 & figure2, computed results for $S(\kappa,\omega)$ are in excellent agreement with the corresponding experimental results(shown with solid circles) particularly at lower ω values, ω < 25 ps⁻¹ for all six values of κ. However, for larger values of ω the calculated results deviates~15% from the corresponding scale reduced experimental values. This can also be noticed from the figures that modified microscopic theory yields results which are in much better agreement with experimental results as compared to that obtained from earlier microscopic theory.

One may calculate the current-current correlation function, whose peak position yield the collective mode frequency, ω_p, using expression (5). Variation of collective mode frequencies, calculated from microscopic theory, with wave vector have been plotted in Figure 4 with hollow circles(○○○) and are compared with values deduced from experimental data (solid circles of fig.2 & fig.3) with solid circles(●●●). Here also, the agreement between two is good. Variation of velocity of sound as computed from dispersion relation, ω_p vs κ , have been plotted in figure4: triangles(ΔΔΔ) present calculation; solid triangles (▲ ▲ ▲) reduced experimental results.
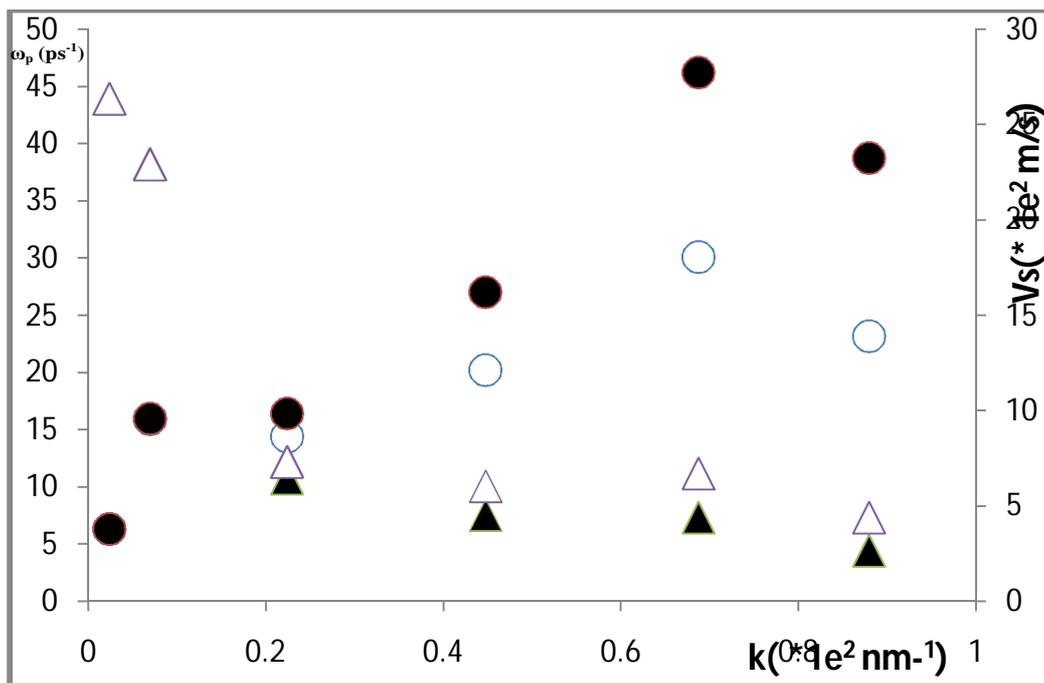
Figure 4:Variation of collective mode frequency, ω_p with wave vector k: (○○○) present calculations using modified microscopic theory; (●●●) scale reduced experimental data.

Variation of v_s with$k$:(ΔΔΔ) present calculations using modified microscopic theory; (▲ ▲ ▲) scale reduced experimental data.

4. CONCLUSION

It may be concluded from present study that the modified microscopic theory of liquids successfully explains the coherent dynamics of the molten sodium metal, which is a light alkali metal, by using the diffusion coefficient as a parameter to obtain quantitative results.

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