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Dynamical Properties of Liquid cesium at 338 K

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ABSTRACT

A theoretical framework is applied to calculate the dynamical structure factors of an alkali metal, liquid cesium (Cs). The study is conducted at 338 K, where 1.825 grams of cesium atoms occupying one cubic centimeter of the liquid. The computations span a wave vector range of κ : 0.3 \AA^{-1} to 2.5 \AA^{-1} , to cover the primary peak region of the static structure factor of liquid cesium at concerned temperature and density conditions. The approach slots in interparticle interactions within the liquid to compute various dynamical quantities, such as the dynamical structure factor and the current-current correlation function. These properties, in turn, provide insights into other key parameters, including collective mode frequencies, sound velocity, and transport properties. This framework reports results for various dynamical properties of interacting liquid of cesium including dynamical structure factors, current-correlation functions, collective mode frequencies, diffusion coefficient, and sound velocity across the entire κ range. A comprehensive method, referred to as the modified microscopic theory, is provided to calculate the full dynamics of liquid potassium at its melting temperature by integrating interparticle interactions and the system's dynamics.

KEYWORDS: Alkali Metals, Inter-particle interaction, Dynamical structure factor, Modified microscopic theory, Current-current correlation function, Collective dynamics, Diffusion coefficient.

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1. INTRODUCTION

Studying the collective dynamics of fluids has always posed a significant challenge, particularly because the degree of correlation among their particles can vary from strong to weak, and the fluids exhibit both individual and collective particle motion. Experimental techniques, such as thermal neutron inelastic scattering and inelastic X-ray scattering, are commonly employed to investigate the collective motion of interacting particle systems, but they have certain limitations. The inelastic neutron scattering (INS) method, for instance, results in a mix of incoherent and coherent scattering cross sections in nearly equal proportions,^{1,2} which complicates the extraction of the coherent dynamical structure component from the data. On the other hand, inelastic X-ray scattering, when performed using high-resolution synchrotron radiation sources, can provide pure coherent scattering data³. Several theories^{4,7} have been developed to explain the complex dynamics of fluids, leading to explanation of these involved dynamical states through certain space-time dependent correlation functions. The dynamical structure factor, is such a function which is obtained from the space-time dependant function when Fourier transformed and can describe how a liquid responds to external disturbances, offers valuable insight into the collective behavior of its atoms.

Cesium, is the heaviest alkali metal with atomic number 55 and atomic mass of 132.7 a.m.u. When compared to other lighter alkali metals, liquid lithium and liquid sodium, it stands out as a much better coherent scatterer of neutrons. The contribution of coherent scattering cross-section, for liquid Cs, in an inelastic scattering spectra is a way higher (~16 times)⁸ than the incoherent cross-section, and hence, it turns out be predominantly coherent spectra. This makes cesium a suitable choice for studying collective modes in simple, highly coupled liquids through INS study. Such a study has been conducted and also has been explained theoretically for an interacting liquid of cesium atoms near its melting temperature, at 308 K. The study revealed that dynamical structures for liquid cesium are endowed with well-formed collective modes even at higher high momentum transfers. These studies, however, remained limited to a certain temperature only and the higher temperatures, which may carry information for dynamical behaviour of the metallic liquid along the phase-curve, remained uninvestigated. Such an effort has been made in the current study to theoretically investigate the dynamical properties and hence, collective dynamics of a liquid of cesium atoms, above its melting point (~ 301 K). The liquid is endowed with 0.824×10^{22} particles occupying one cm^3 of the system and at relative mass density of 0.99 as compared to that at the melting point, is essentially a system of strongly interacting cs atoms. Among some other reported work related to distinct properties of the cesium in its liquid state, no data has been reported at the considered temperature, to best of my knowledge.

In this study, the dynamical structure factors and, consequently, the collective dynamics of liquid cesium has been predicted using an approach that incorporates the liquid's microscopic dynamics. The theory applies appropriate time averages to the solution of the equations of motion for moving particles, examining how an interacting, correlated fluid, exhibiting time-dependent disorder, responds to external radiation (such as X-rays or neutrons)⁷. The familiar Fluctuation-Dissipation Theorem links the Fourier transform in space-time of the imaginary part of the density response function to the dynamical structure factor. The theory incorporates correlations between distinct particles, previously neglected, by introducing a characteristic relaxation time⁹. This relaxation time is influenced by key

properties of the system, such as the static structure factor, density, temperature, mass, interaction potential, and diffusion coefficient.

Within the theoretical framework, the diffusion coefficient is extended to a wave-vector-dependent form, with specific values assigned to each wave vector. The theory is further refined to align with experimentally measured values for fluids with varying physical parameters. This allows for an accurate depiction of the intermediate self-scattering function by making the diffusion coefficient frequency-dependent, rather than relying on numerous arbitrary parameters. Previous work has used this modified microscopic theory to explain the collective dynamics of various correlated liquids¹⁰⁻¹⁹. In this study, the same approach is applied to determine the collective dynamics of liquid cesium.

2. MATHEMATICAL FORMALISM

Microscopic theory examines the microdynamics of a fluid to describe density fluctuations and particle motion. To better understand the complex space-time-dependent correlation functions, they are defined and approximated to derive results for the dynamical structure factor. In earlier versions of the theory, and when calculating the dynamical structure factors, correlations over time were not taken into account. As a result, the resulting dynamical structure factors were overly damped and did not match the spectral lineshapes observed experimentally. To overcome this limitation, Tewari et al.⁹ modified the theory by introducing a characteristic relaxation time to account for the missing correlations. This updated version of the microscopic theory has successfully explained the dynamical structure factors of a number of liquids.

The characteristic time in the theory is given by the following expression:

$$[\tau(\kappa)]^{-1} = \frac{\omega_{\kappa}}{[(\kappa^2 / m\beta S(\kappa)\beta'^2) - 1]^{1/2}} - \beta' \tag{1}$$

In the expression (2) $\beta' = D\kappa^2$, D is the diffusion coefficient that varies with wave-vector, κ , $\beta = (k_B T)^{-1}$, m is the atomic mass and $S(\kappa)$ is the static structure factor can be obtained from the Fourier transform of the $g(r)$ as follows:

$$S(\kappa) = 1 + 4\pi\rho_0 \int_0^{\infty} r^2 (g(r) - 1) \frac{\sin \kappa r}{\kappa r} dr \tag{2}$$

The frequency ω_{κ} occurred in the expression (1) and (2) is given as follows:

$$\omega_{\kappa}^2 = \omega_E^2 \left[1 - \frac{3 \sin \kappa r_0}{\kappa r_0} - \frac{6 \cos \kappa r_0}{(\kappa r_0)^2} + \frac{6 \sin \kappa r_0}{(\kappa r_0)^3} \right] \tag{3}$$

The elastic frequency ω_E and r_0 occurred in the expression (2) are given respectively, as:

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

Here, $V(r)$ is the inter-atomic potential and z is the space coordinate.

And on approximating

$$\frac{\partial^2 V}{\partial z^2} g(r) = A\delta(r - r_0) \tag{5}$$

The peak of the delta function is $r = r_0$.

In this form of the theory, the dynamical structure factor takes the following form:

$$S(k, \omega) = \frac{k^2}{\pi m \beta} \left\{ \left[1 + \omega_k^2 \frac{(\beta'' + \gamma)^2 - \omega^2}{[(\beta'' + \gamma)^2 + \omega^2]^2} \right] \frac{2\beta''}{(\beta''^2 + \omega^2)^2} - \omega_k^2 \frac{(\beta''^2 - \omega^2)}{(\beta''^2 + \omega^2)^2} \frac{2(\beta'' + \gamma)}{[(\beta'' + \gamma)^2 + \omega^2]^2} \right\} \times \left\{ \left[1 + \omega_k^2 \frac{(\beta'' + \gamma)^2 - \omega^2}{[(\beta'' + \gamma)^2 + \omega^2]^2} \right]^2 + \left[\omega_k^2 \frac{2\omega(\beta'' + \gamma)}{[(\beta'' + \gamma)^2 + \omega^2]^2} \right]^2 \right\}^{-1} \quad (6)$$

Here, in the expression (1), $\beta'' = D_{\text{eff}} \kappa^2$, D_{eff} is ω -dependant diffusion coefficient, and, $\Upsilon = \tau(\kappa)^{-1}$, given by expression (1)

The current-current correlation function $C(\kappa, \omega)$ is given by the following expression:

$$C(\kappa, \omega) = \frac{\omega^2}{\kappa^2} S(\kappa, \omega) \quad (7)$$

3. RESULTS AND DISCUSSION

As outlined in the mathematical section, the evolution of equilibrium dynamics in any liquid using the modified microscopic theory requires knowledge of the inter-particle interaction potential. Therefore, it is essential to determine a suitable interaction potential for the liquid under the given physical conditions. To calculate the dynamical structure factors, it is more appropriate to use an interaction potential that is derived from or connected to the static structure factor, rather than relying on other algebraic mathematical formulations. Such an inter-particle potential has been worked in the literature²⁰ for liquid cesium to produce the static structure factor in agreement with the experimental X-ray diffraction data for the static structure factor $S(\kappa)$ reported by Hujiben et al.²¹. Though the potential form described in the study seems consistent in pattern and also with the physical quantity of interest to be produced, $S(\kappa)$, the temperature for system of Cs atoms is rather lower, 303 K. Hence, another form of interaction potential as described by Hujiben et al.²¹ while reporting the X-ray diffraction data, has been implied in the present work. The proposed potential is deduced using hyper-netted chain approximation and has successfully reproduced the experimental results for the static structure factor of liquid Cs at a temperature of 338 K. Thus, it has proven to be a more appropriate choice for this investigation. The suggested interaction potential exhibits a variation with interatomic distance which is peculiar for metallic liquids consists of both a hard core and a long-range oscillatory component generated through the screening of electrons in the presence of cesium ions.

Substituting the interaction potential into expression (4) results in the maximum Einstein frequency ω_E and from expression (5) results in r_0 , turn out to be $4.98 \times 10^{12} \text{ s}^{-1}$ and 4.92 \AA , respectively. These calculations are based on the pair distribution function $g(r)$, obtained from the Fourier transform of the experimentally measured static structure factor and derived using the interaction potential for liquid cesium²⁰. These values are substituted in expression (3) for the evaluation of κ -dependant frequency, ω_κ and further these results are then inserted into expression (1) to determine the single characteristic relaxation time. The characteristic relaxation time, $\tau(\kappa)$, is a key component of the modified microscopic theory, accounting for the correlations between distinct particles at different wave-vector values, κ . It represents the evolution of system properties, such as particle positions or densities, over time, considering wave-vector-dependent correlations. In essence, it describes the timescale over which the system returns to equilibrium after a disturbance, with this timescale varying based on the wave-vector. The diffusion coefficient, D , in expression (1), is introduced fitting parameter to ensure that the zeroth sum rule is satisfied for a given wave-vector, κ . As

a result, the modified microscopic theory complies with all sum rules for liquids. With the relaxation time and diffusion coefficient in place, the detailed dynamical structure factors, $S(\kappa, \omega)$, are then computed. These structure factors characterize the system's fluctuations in both space and time, offering crucial insights into its dynamic behaviour and have been derived using expression (6), which connects $S(\kappa, \omega)$ to $\tau(\kappa)$, ω_{κ} , m & temperature and also to the diffusion coefficient.

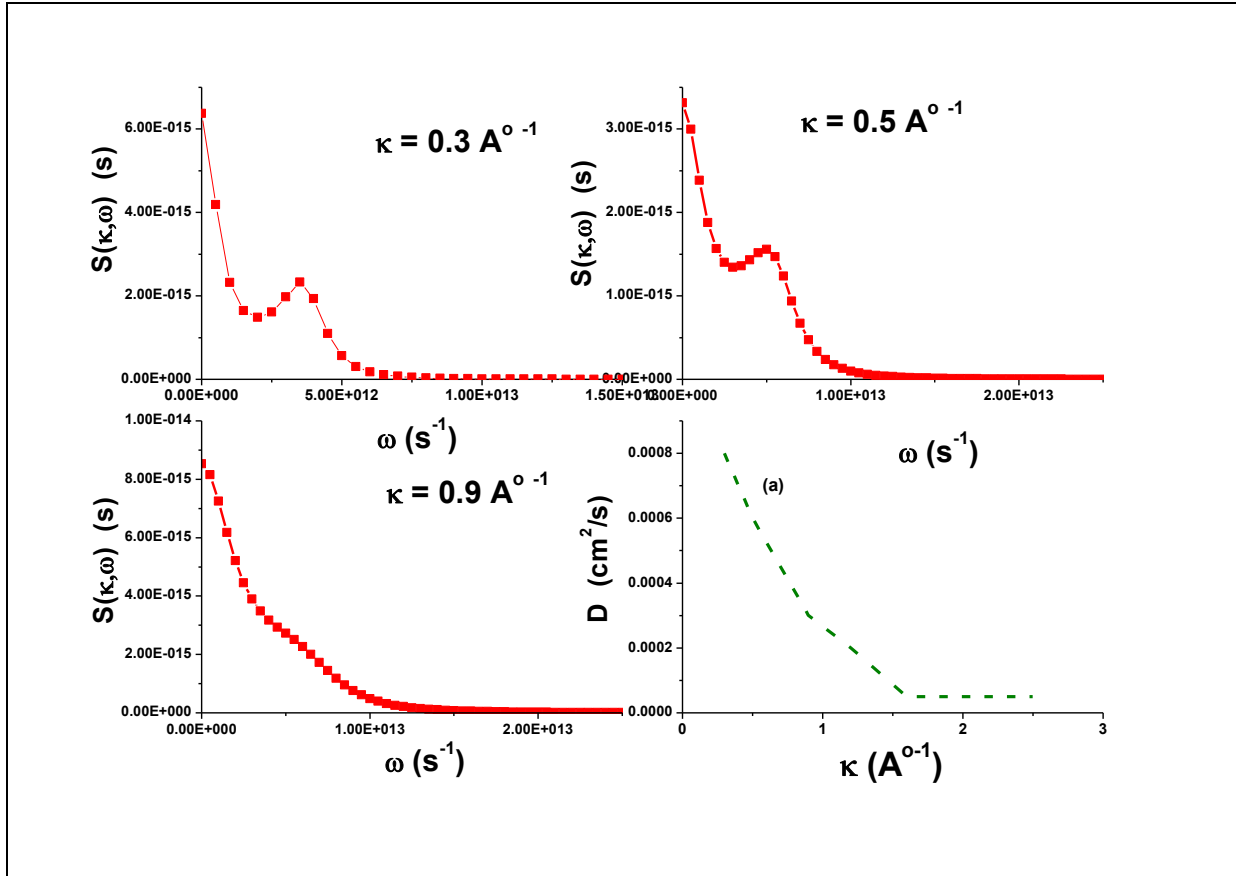


Figure 1: Evaluated results for the dynamical structure factor, $S(\kappa, \omega)$ of liquid cesium at 338 K versus frequency, ω : (—■—) present theory at $\kappa=0.3 \text{ \AA}^{-1}$, $\kappa=0.5 \text{ \AA}^{-1}$ and $\kappa=0.9 \text{ \AA}^{-1}$.

Figure 1(a): Variation of the diffusion co-efficient with wave-vector κ : with (- - -).

The dynamical structure factors were computed using expression (6) for various wave-vector values ranging from 0.3 \AA^{-1} to 2.5 \AA^{-1} . The results, which show the variation of the dynamical structure factors with frequency, ω , are presented in Figure 1, for wave-vectors of 0.3 \AA^{-1} , 0.5 \AA^{-1} , 0.9 \AA^{-1} , represented by a solid square curve (—■—) and in Figure 2 for wave-vectors of 1.2 \AA^{-1} , 1.6 \AA^{-1} , 2.5 \AA^{-1} , shown solid-square curve (—■—). In these calculations, the diffusion coefficient, $D_{\text{eff}} = D$. As shown in the figures, for smaller wave-vector values, up to $\kappa < 0.9 \text{ \AA}^{-1}$, the dynamical structure factors exhibit a three-peak form, with the presence of Brillouin peaks. At $\kappa = 0.9$ a minute side hump can be noticed in the structural function. However, as the wave-vector increases, $\kappa > 0.9 \text{ \AA}^{-1}$, the dynamical structure factors turned more damped, and the Brillouin peaks are no longer observed. The pattern of variation is,

hence, is typical. To provide a comparison, the calculated results are contrasted with data from an inelastic neutron scattering (INS) study of liquid cesium⁸, which was conducted at a lower temperature of 308 K. The comparison has been shown at four different wave vector values, $\kappa = 0.3 \text{ \AA}^{-1}$, $\kappa = 0.9 \text{ \AA}^{-1}$, $\kappa = 1.6 \text{ \AA}^{-1}$ and $\kappa = 2.5 \text{ \AA}^{-1}$. The self-diffusion coefficients, which act as fitting parameters in the theory and thus exhibit wave-vector dependence, were determined during the calculation of the dynamical structure factors. The earlier microscopic theory⁷ did not satisfy the significant zeroth sum rule in its generated dynamical structure factors.

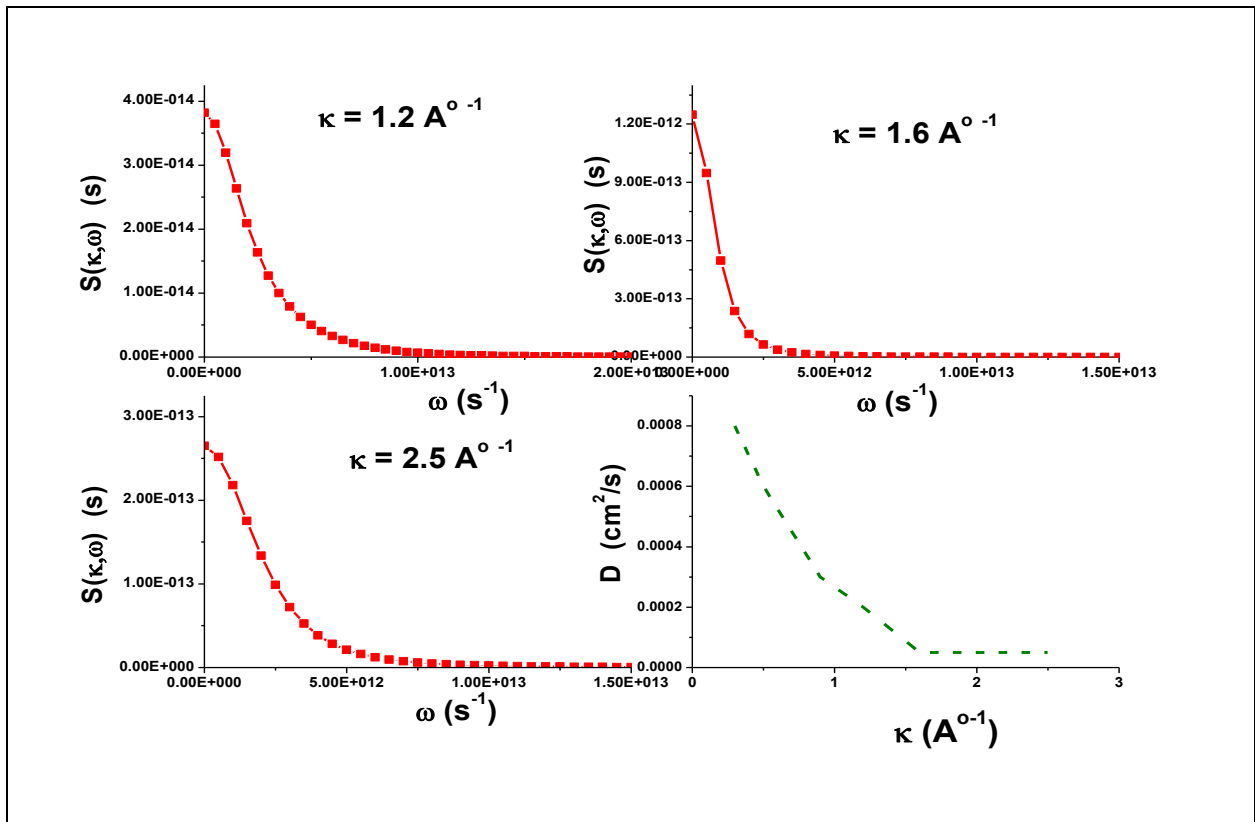


Figure 2: Evaluated results for the dynamical structure factor, $S(\kappa, \omega)$ of liquid cesium at 338 K versus frequency, ω : (—■—) present theory at $\kappa = 1.2 \text{ \AA}^{-1}$, $\kappa = 1.6 \text{ \AA}^{-1}$ and $\kappa = 2.5 \text{ \AA}^{-1}$.
 Figure 2(a): Variation of the diffusion co-efficient with wave-vector κ : with (- - -).

Moreover, the diffusion coefficient is related to the determination of the characteristic time. In Figure 1(a) and 2(a), variation of the diffusion coefficient with wave vector has been plotted with dashed curve (- - -). This can be observed from the figure that the overall variation in the diffusion coefficient is about more than an order where it varies for half an order upto $\kappa = 0.9 \text{ \AA}^{-1}$ and for $0.9 \text{ \AA}^{-1} < \kappa \leq 2.5 \text{ \AA}^{-1}$, it again varies for more than half an order.

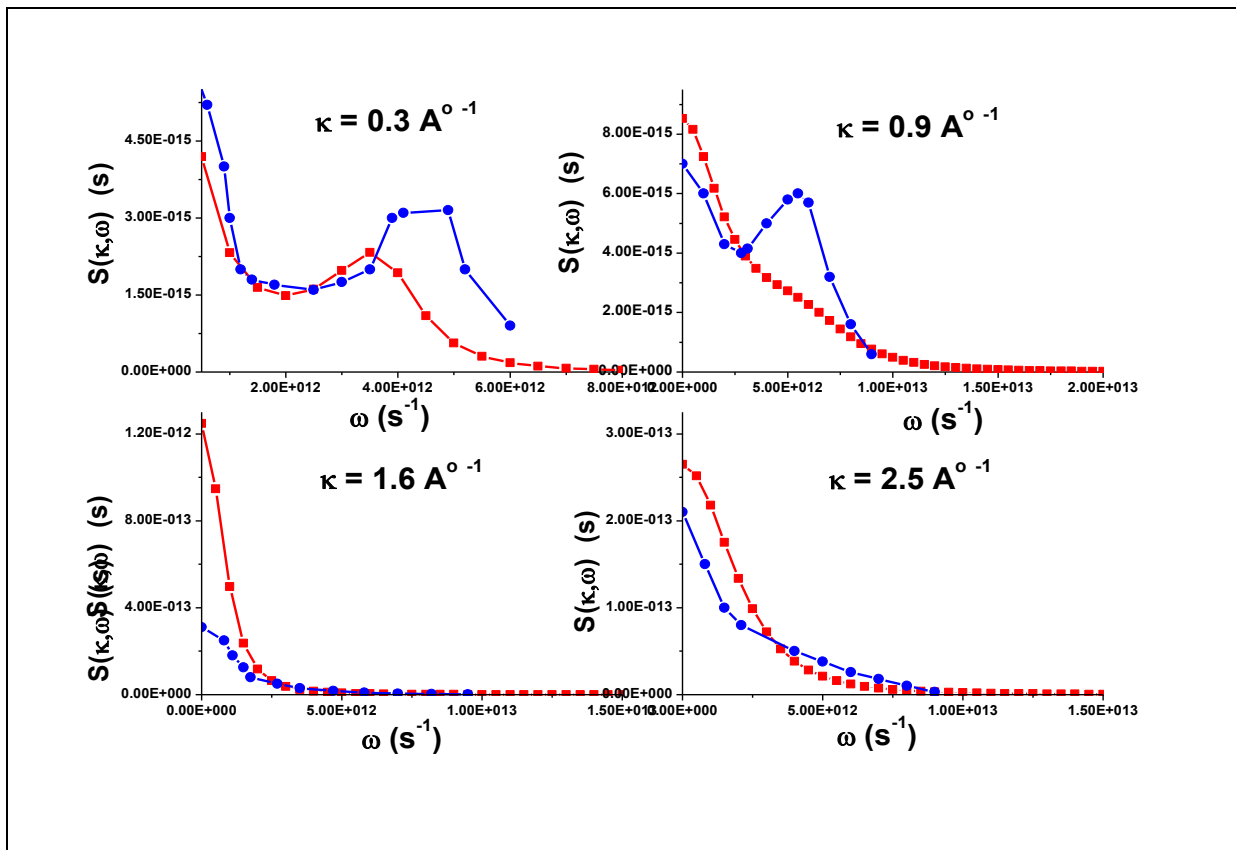


Figure 3: Dynamical structure factor, $S(\kappa, \omega)$ of liquid cesium versus frequency, ω : (—■—) present theory at T=338 K; (—●—) experimental results at T= 308 K.

The experimental data from INS study are shown in Figure 3 as solid-circle curves (—●—), while the theoretical results are represented by solid-square curves (—■—) for all κ values. The comparison indicates that, at the lower temperature, the sharper and more well-defined structures with more prominent Brillouin peaks are present, as for $T=308$ K structures from INS study exhibit a clear defined shoulder peak at $\kappa = 0.9 \text{ \AA}^{-1}$. Additionally, the peak positions shift towards larger frequencies at the lower temperature of 308 K. The observed differences between the dynamical structure factors from the two studies can be attributed to the temperature difference.

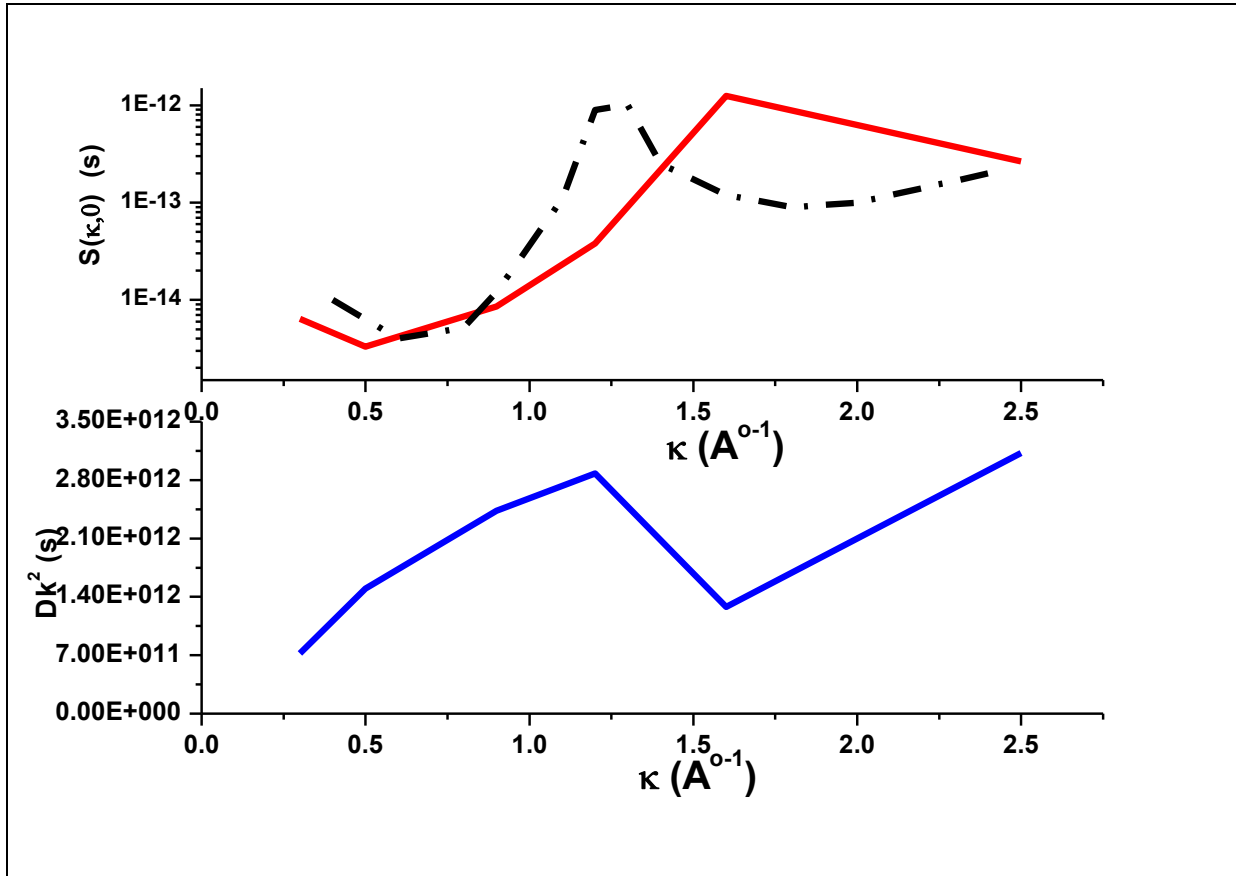


Figure 4(a): Zero dynamical structure factor, of liquid cesium versus frequency, ω : (—) present theory at $T=338$ K; (— · —) experimental results at $T= 308$ K.

Figure 4(b): Variation of diffusive frequency, $D\kappa^2$ with κ : (—).

In Figure 4(a), the zero frequency dynamical structures for liquid cesium obtained from the present computations, have been compared to the earlier INS study performed at $T=308$ K. They are plotted as against wave vector: with solid curve (—) at 338 K and dashed-dotted curve (— · —) INS results at 308 K. The zero dynamical structure factor, $S(\kappa, \omega=0)$, is a significant factor to ensure the enhancement of the modified microscopic theory over the older theory of Hubbard and Beeby [16] which was not efficient enough to generate accurate zero dynamical structure factors. In present form, the microdynamics can be solved to obtain quantitative $S(\kappa, \omega=0)$ at both temperatures, however, the results for $S(\kappa, \omega=0)$ are higher for the higher temperature and further, are shifted towards larger κ -values. The trend for variation in both temperature considerations, however, remains the same. In Figure 4(b), the variation of $D\kappa^2$, corresponding to the inverse of the diffusive time as obtained in the present calculations, is plotted against the wave-vector, κ with a solid curve (—). This can be observed from the figure that the variation of $D\kappa^2$ with κ yields a dispersion relation of diffusive frequencies.

In Figure 5(a), the dispersion relation of collective mode frequencies, ω_p , for liquid Cs at 338 K is shown with solid-squares (■). The values of ω_p represent the peak positions of the

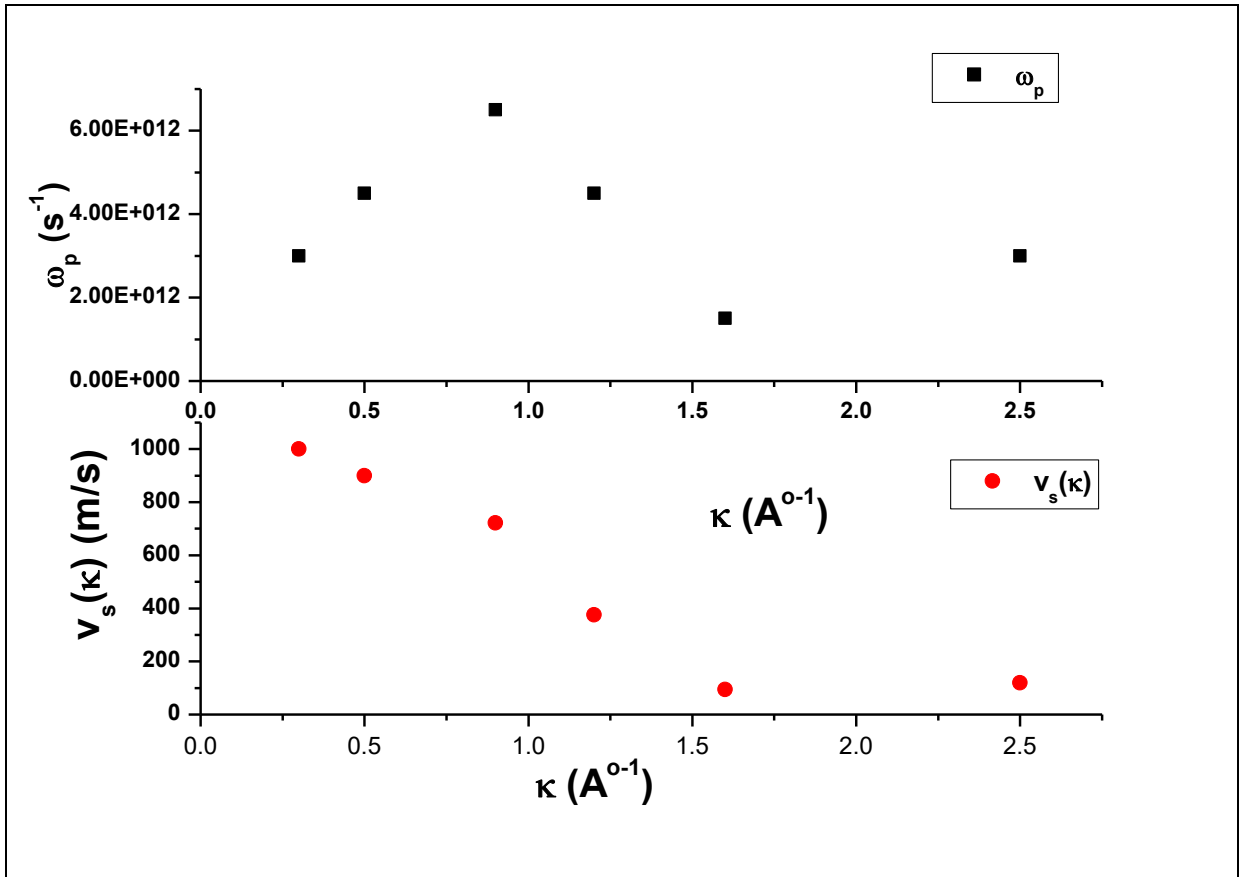


Figure 5(a): Dispersion relations for liquid Cs at 338 K: (■) ω_p deduced from current correlation function.

Figure 5(b): Velocity of sound in liquid Cs at 338 K as depicted from figure 5(a) vs. wave-vector, κ : (●).

computed current-2 correlation functions for different wave-vector values, plotted against the frequency ω . As seen in the figure, the frequencies of the collective modes, ω_p , initially increases with κ to a maximum at $\kappa = 0.9 \text{ \AA}^{-1}$, after this, the frequencies decrease for higher wave-vectors to attain a minimum value at $\kappa = 1.6 \text{ \AA}^{-1}$ and then increases again. Thus, the typical form of the dispersion relation for liquid Cs is obtained across the entire wave-vector range. Figure 5(b) shows the sound velocities, represented by solid-circles (●), deduced from the dispersion relation in Figure 5(a) and plotted as a function of the wave-vector κ . This can be depicted from the figure when extrapolated to $\kappa=0$, the κ -dependant sound velocity turns out to be nearly 995 m/s which is closer to the experimental measurement²² (~990 m/s).

4. CONCLUSION

The study concludes that the modified microscopic theory effectively predicts the collective dynamics of liquid Cs at 338 K, representing a significant improvement over the previous microscopic theory. This approach incorporates particle correlations by defining a relaxation time that accounts for inter-particle interactions. As a result, it allows for the calculation of key properties such as the diffusion coefficient, collective modes, and sound velocity along with the dynamical structure factors. By defining the relaxation time to reflect the specific particle correlations, the theory provides precise results for these properties.

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