International Journal of Scientific Research in Physics and Applied Sciences
Vol.7, Issue.1, pp.56-59, February (2019)
DOI: https://doi.org/10.26438/ijsrpas/v7i1.5659

Research Paper

E-ISSN: 2348-3423

Microscopic Transport Phenomena in a Liquid Alkali Metal: K^{39}

Grima Dhingra

Department of Physics, Maharshi Dayanand University, Rohtak-124001, India

Corresponding Author: grimadhingra@gmail.com

Available online at: www.isroset.org

Received:21/Jan/2019, Accepted: 14/Feb/2019, Online 28/Feb/2019

Abstract— Microscopic equations of an interacting and correlated system of particles has been solved to compute two of the transport properties, namely diffusion coefficient and longitudinal viscosity, of liquid potassium near its melting point, at 343K, for a wave vector range: 0.9 nm^{-1} to 17.0 nm^{-1}. The present theoretical approach uses microscopic theory as a tool to compute the detailed dynamical structure factor, current-current correlation function and hence, the diffusion coefficient as well as the coefficient of longitudinal viscosity. Microscopic theory uses interparticle-interaction present among particles of a liquid to yield density-density response function and hence, its complete dynamics. The diffusion coefficient is evolved as a realistic parameter which has been fit to explain the experimental dynamical structure factors. The coefficient of longitudinal viscosity on the other hand is directly related to static structure factor and diffusion coefficient in the regime where wavevector and frequency approaches zero. It also depends upon velocity of sound which, in the present communication, has been calculated from peak positions of current-current correlation functions in the limit wavevector approaches zero. Computed results for both of the transport coefficients, self diffusion coefficient and longitudinal viscosity are found to agree well with the corresponding experimentally reported values.

Keywords—*transport coefficients, current-current correlation function, microscopic theory, dynamical structure factor, longitudinal viscosity, diffusion coefficient*

I. INTRODUCTION

There are a number of theories [1-3] which explains the transport phenomena and hence, quantitatively determine transport coefficients such as diffusion coefficient, viscosity, thermal conductivity etc. in liquids as well gases. However, these transport coefficients are rarely evaluated in terms of interactions and correlations present among the constituent particles of a given fluid. In the present attempt, modified microscopic theory for liquids has been used to evaluate diffusion coefficient and coefficient of longitudinal viscosity of liquid potassium at 343 K, just near its melting point~336 K, in a wave-vector, K, range 0.9 $nm^{-1} \leq K \leq 17.0$ nm^{-1}. As the melting temperature of liquid potassium is fairly high, its constituent particles interact with quite high energies in this temperature vicinity. The sample under consideration is having 1.26×10^{22} particles in one cubic centimetre volume, which again is closer to 1.276×10^{22} particles/cc at its melting point. Such a fluid is a highly interacting, strongly correlated and is an ordered system of potassium atoms. Use of modified microscopic theory to study the equilibrium dynamics and hence, transport coefficients of such a classical fluid system is well justified.

Study of collective dynamics of fluids at different physical constraints had always been a challenging task for theoreticians as it demands experimental measurement of pure coherent scattering cross section. However, the earlier used inelastic scattering technique(INS) yields blend of both incoherent and coherent scattering cross sections in comparable ratios and extraction of coherent part from such experimental data is not easy. This drawback of INS technique has been overcome by recently developed advanced synchrotron radiation facilities which are capable of yielding high resolution spectral data from inelastic scattering of x-rays. This inelastic x-ray scattering (IXS) produce coherent scattering cross sections which hence, can be conveniently used for theoretical investigation of coherent dynamics of an interacting fluid. Such an experimental IXS coherent spectral line shape data has been recently reported by Monaco et al.[4] for liquid potassium at 343 k for wave vector range: 0.9-17.0 nm^{-1}.

This paper has been organized into five different sections: Introduction, Theory, Mathematical formalism, Results and discussion and Conclusion. In Section I a brief review of literature related to current research problem has been made. Section II describes the basic hypotheses and outline of the theory used in the present communication. In section III, mathematical expression for distinct physical quantities in reference to present microscopic theory has been provided.

In section IV, detailed computational results as obtained for fluid under consideration has been provided and been thoroughly discussed. Section V yields the conclusions drawn from the computational results.

II. THEORY

Modified microscopic theory of simple fluids has proven to be a self-consistent theory which has successfully explained the collective dynamics of a number of fluids with different extent of correlations and different degrees of disorder[5-10]. This theoretical approach evolves the solution of microscopic equation of motion for an interacting and correlated system of particles. By solving microscopic equation of motion, one can have displacements of correlated particles which are further related to the density-density response function which actually expresses the response of the system to any external weak probe. Density-density response function is a wave vector and frequency dependant quantity whose Fourier transform is related to the dynamical structure factor, $S(\kappa,\omega)$, where, κ is wave-vector and ω is angular frequency, quantity from which complete collective dynamics of a given fluid can be calculated. While solving the microscopic equations of motion one may need to solve involved space-time dependant correlation functions[11] which are not possible to solve exactly. This microscopic theory is, therefore, modified to incorporate the distinct particle correlation[5-7] by introducing relaxation time[8] in the expression for dynamical structure factor, $S(\kappa,\omega)$, such that it satisfies zeroth sum rule which was ignored in the earlier theory. The relaxation time so introduced carries only one fitting parameter, the self diffusion coefficient. Further to make the intermediate correlation functions to be valid for entire time range and not only at time t=0, the self diffusion coefficient which occurs as the only physical parameter in the theory is made frequency dependant. This modified microscopic theory, hence, uses the inter-particle interaction potential as the only input through relaxation time and generates three peak structures for dynamical structure factors for different values of wave-vector, κ. Collective excitations exhibited for finite values of ω, are then analyzed for their variation with wave-vector, κ to yield velocity of sound, $c(\kappa)$. Hence, the theory is capable of predicting dispersion relation and the entire dynamics of given fluid.

In the present theoretical workout, modified microscopic theory has been used as a tool to compute the detailed dynamical structure factor and hence, the diffusion coefficient as well as the coefficient of longitudinal viscosity. The diffusion coefficient is evolved as a realistic parameter which has been fit to explain the experimental (IXS) data for dynamical structure factor and turns to be a frequency dependant physical parameter. The coefficient of longitudinal viscosity, on the other hand, is directly related to static structure factor, diffusion coefficient and velocity of sound in the limit $\omega \to 0$ & $\kappa \to 0$ through a mathematical expression.

III. MATHEMATICAL FORMALISM

This Expression for dynamical structure factor in the present theory is turned to be:

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

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

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

where, ω_E, is maximum elastic frequency which is related to inter atomic interaction potential *V(r)* and static pair correlation function *g(r)*[10,12].

And

$$\gamma(\kappa)=$$

$$\left[\tau(\kappa)\right]^{-1}=\frac{\omega_\kappa}{\left[\left(\kappa^2/m\beta S(\kappa)\beta'^2\right)-1\right]^{1/2}}-\beta' \quad (3)$$

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

Longitudinal current-current correlation function $J_l(\kappa,\omega)$ is given by expression:

$$J_l(\kappa,\omega)=\frac{\omega^2}{\kappa^2}S(\kappa,\omega) \quad (4)$$

Expression for coefficient of longitudinal viscosity is as follows:

$$\eta_l(\kappa)=2m\rho_0 D(\kappa)\left[1+\left\{c(\kappa)\left[m\beta S(0)\right]^{1/2}-1\right\}\right] \quad (5)$$

here, $c(\kappa)$ is the κ-dependent velocity of sound determined from the dispersion relation executed from the peak positions of current-current correlation functions.

IV. RESULTS AND DISCUSSION

Atoms of potassium are subject to a temperature just above its melting point (T/T_m=1.025), constitute a system of strongly interacting particles (thermal de Broglie wavelength~ 1.26×10^{-9}m). The interaction potential of such a system of K-atoms is worked out by Brettonet-Jakse[12] to yield static structure factor well in agreement with experimental result and the same potential is used in the present article as an input to evaluate collective dynamics. This potential is shown in Figure1. as a function of inter-particle separation with solid curve. For the sake of comparison interaction potential for other alkali metals is also shown in Figure1.: Na(dash-dotted curve), Rb(dotted curve), Cs(dashed curve) and Li(dash-double dotted curve).

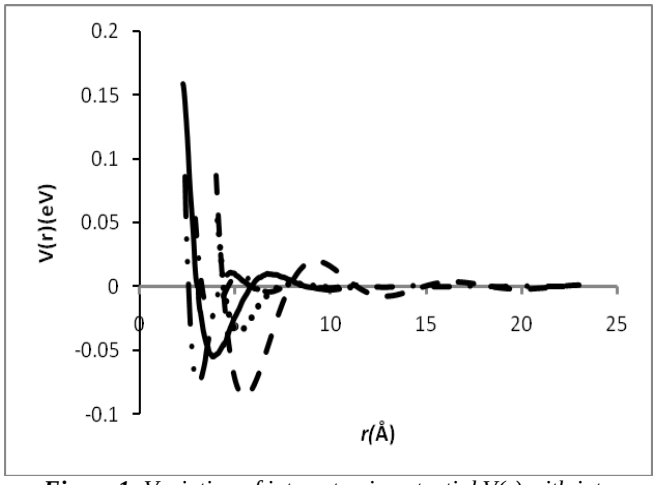

Figure 1: *Variation of inter-atomic potential V(r) with inter particle separation* ***r,*** *for liq. K at 345 K(—);liq. Na at 373 K (– · –);liq. Cs at 308 K(---);liq. Rb at 319 K(·····);liq. Li at 475 K(– ·· –).*

Using this form of potential *V(r)* and static pair correlation function *g(r)* of liquid potassium, elastic frequency , ω_E , turns to be 1.0712×10^{13} s^{-1}. From ω_E and r_0=4.2Å in expression(2) dispersion relation is executed which yields sound velocity $c(\kappa)$ in hydrodynamic limit to be ~1350 m/s which is quite closer to isothermal value obtained from ultrasonic measurements. These results when expressed in equation (1) yields detailed coherent dynamical structure factors, $S(\kappa,\omega)$, for four different values of wave-vector κ [13], which are fairly in agreement with experimental results of Monaco et al.[4].The corresponding current-current correlation function $J_l(\kappa,\omega)$ is plotted in Figure2 for different values, κ .

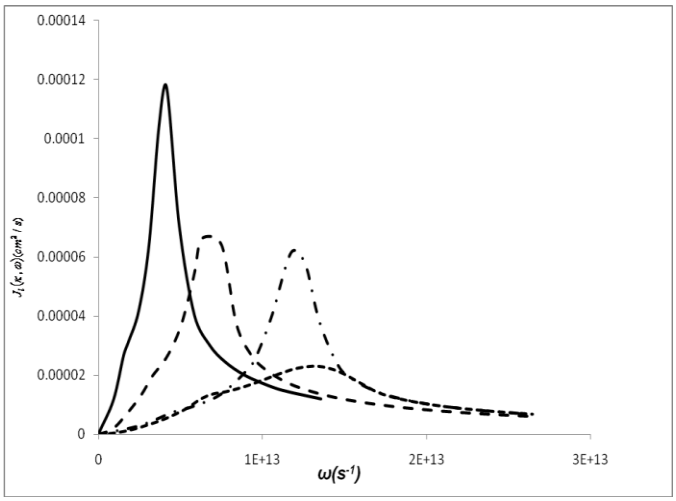

Figure 2: *Variation of current-current correlation function with angular frequency,* ω *,* κ *=1.75 nm^{-1} (——);* κ *=3.0 nm^{-1}(---);* κ *=6.0 nm^{-1}(– · –);* κ *=10.0 nm^{-1} (·····).*

As has already been discussed in theory section, diffusion coefficient $D(\omega)$ appears as a realistic parameter while fitting the expression (1) to achieve agreement with IXS spectra values such that all time behavior of intermediate scattering function is being slotted in.

Figure 3: *Variation of Diffusion coefficient with angular frequency,* ω *,* κ *=1.75 nm^{-1} (——);* κ *=3.0 nm^{-1}(------);* κ *=6.0 nm^{-1}((– · –);* κ *=10.0 nm^{-1} (·····). Experimentally reported value of diffusion coefficient (●)*

This approach leads to a frequency dependant form of diffusion coefficient whose variation with angular frequency, ω , is shown in Figure3 for four different values of wave-vector, κ ; for κ =1.75 nm^{-1}(solid curve); κ =3.0 nm^{-1}(dashed curve); κ =6.0 nm^{-1}(dash-dotted curve) and κ =10.0 nm^{-1}(dotted curve).Experimentally measured values for self diffusion coefficient[1] of liquid potassium at 337 K and at 400 K are also shown in the graph with solid circles. As is evident from the figure, variation of diffusion coefficient with angular frequency never exceeds more than

two orders and remain within one order vicinity of experimental value for entire ω range.

Peaks of current-current correlation functions, given by expression (4), when plotted against wave vector, κ , indicates dispersion relation which yields value for velocity of sound, $c(\kappa)$. This result for sound velocity along with evaluated self-diffusion coefficients and value of static structure factor when substituted into equation(4) produces coefficient of longitudinal viscosity as a function of wave vector, κ .The calculated variation of $\eta(\kappa)$ is plotted in Figure 4.

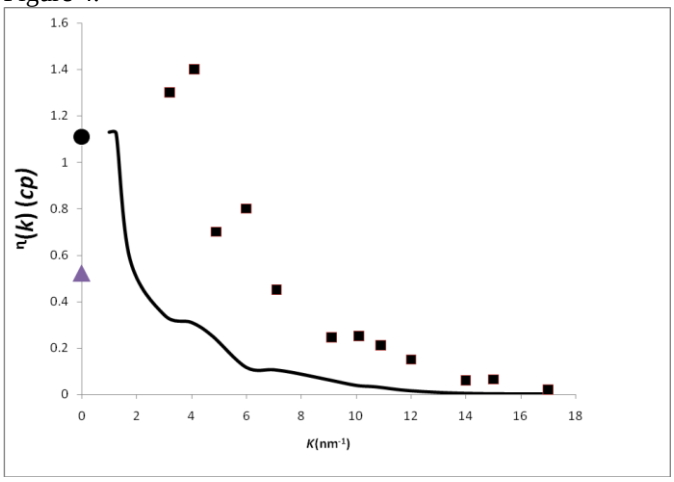

Figure 4: *Variation of coefficient of longitudinal viscosity η_l , with wave vector κ , present theory(—); memory function approach(■■■); hydrodynamic value(●); experimental value at m. pt., 337 K (▲).*

In Figure4., values of $\eta(\kappa)$ as calculated from present theory are shown with solid curve whereas those evaluated from memory function approach are shown with solid squares. Also, are shown the experimental hydrodynamic result [14] with solid circle and experimental measured value [1] for liquid potassium at 337K (m. pt.) with solid triangle. This is clearly indicated by the figure that variation of $\eta(\kappa)$ as computed from modified microscopic theory remain in well agreement to experimental value at melting point of potassium for entire κ range and approaches experimental result[4] in the limit $\kappa \to 0$. However, results obtained from another alternative approach [4] seems to be slightly off.

V. CONCLUSION

It may be concluded from the study that the modified microscopic theory is capable of generating not just the collective dynamics of interacting fluids but can also explain the transport phenomena in these correlated fluids. This is evident since transport coefficients of liquid potassium , diffusion coefficients and coefficient of viscosity, evaluated using this above mention theory are found to be in good agreement with the corresponding experimental results for a range,0.9 $nm^{-1} \leq \kappa \leq 17.0$ nm^{-1}. This can also be noted that theory acquires a single input for whole framework, i.e. inter-atomic interaction potential.

REFERENCES

[1] C. Chen, H. Eyring, "*Significant Liquid-Structure Theory of Viscosity and Self-Diffusion of the Alkali Metals*", Proc. Nat. Acad. Sci., USA, Vol. **69**,pp. **1342-1345**,**1972**.

[2] M. Sayem, R. D. Murphy, "*Temperature Dependence of Transport Properties of Liquid Sodium and Potassium*", Phy. Chem. Liq., Vol. **40**, No. **4**, pp. **469-475**, **2002**.

[3] P. A. Egelstaff, "*An Introduction to The Liquid State*", Clarendon Press, **Oxford**,1994.

[4] M. Monaco,T. Scopigno, G. Monaco et al., "*Collective Dynamics in Molten Potassium: An Inelastic Scattering Study*", Journal of chemical physics, Vol.**120**, No. **17**, pp. **8089-8094**, **2004**.

[5] S.P. Tewari, S.P. Tewari, "*Theory of Collective Motion in Liquids*", Journal of Phys. C: Solid State Phys.**,** Vol. **8**, pp. **L569-L572**, **1975**.

[6] S.P. Tewari, S.P. Tewari, "*Zero Frequency Dynamical Structure Factor of Liquids*", Phys. Letters, Vol. **56A**, pp. **99-100**, **1976**.

[7] S.P. Tewari, S.P. Tewari, "*Theory of Long Wavelength Collective Motion in Liquids*", Phys. Letters, Vol. **65A**, pp. **241-243**, **1978**.

[8] S. P. Tewari, S. P. Tewari, , R. Gupta *"Dynamics of a Supercooled Liquid", Solid State Communications*, Vol. **27**, pp. **1385-1387**, **1978.**

[9] S.P. Tewari, G. Dhingra, P. Silotia, J. Sood, " *Microdynamics of a monoatomic liquid metal Journal of Non- Crystalline Solids*", Vol. **355**, pp. **2522**, **2009**.

[10] S. P. Tewari, G. Dhingra, P. Silotia, J. Sood, "Pronounced variation in the collective dynamics of highly correlated lightest liquid alkali metal: Lithium", *Journal of Non-crystalline Solids*, Vol. **357**, pp. **3702-3710**, **2011**.

[11] J. Hubbard, J. L. Beeby, "*Collective Motion In Liquids*", J. Phys. C, Vol. **2**, pp. **556**, **1969**.

[12] J. K. Bretonnet, N. Jakse, "*Use of Integral-Equation Theory in Determining the Structure and Thermodynamics of Liquid Alkali Metals*", Phys. Rev. B, Vol. **50**, pp. **2880**, **1994**.

[13] S.P. Tewari, P. Sioltia, G. Dhingra, J. Sood, "*Coherent Dynamics of Molten Potassium*", AIP Proceedings Vol. **1731**, pp. **110018**, **2016**.

[14] O. Ohse, "*Handbook of thermodynamics and transport properties of alkali Metals*", Blackwell Scientific, **Oxford**, **1985**.

AUTHORS PROFILE

Dr. Grima Dhingra did M.Sc. in Physics from Department of Physics and Astrophysics, University of Delhi, with specialization in Condensed Matter Physics. She completed her Ph.D. in 2009 from University of Delhi. Her areas of research are 'Fluid Dynamics' and 'Plasma Physics' and 'Cryptography'. She has published around twelve research papers in reputed, peer-reviewed and citation indexed international journals. She has presented many of her research papers in international and national conferences. She has been granted and successfully completed UGC minor project in 2012-14. Dr. Grima is a life member of materials research society of India (MRSI). Currently, she is working as Assistant Professor in the Department of Physics, M.D. University, Rohtak, Haryana. She has 3 years of UG and 8 years of PG teaching experience and has 11 years of research experience.

