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Thermal resistance network model for thermal conductivity of normal liquid helium-4 and helium-3

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Abstract

Liquid heliums are intriguing substance. Superfluid states below certain critical temperatures, notably liquid helium-4 and helium-3 exhibit ultra-high thermal conductivity (TC) in the superfluid phase. However, the microscopic origin of the TC of liquid heliums in the normal phase remains unclear. In this work, we employ the thermal resistance network model to calculate the thermal conductivities of normal liquid helium-4 (He I) and helium-3. Predicted values are not only in good agreement with the measurements but also reproduce the experimental trend of TC increasing with temperature and pressure.

Keywords: liquid helium, random walk, thermal conductivity

(Some figures may appear in colour only in the online journal)

1. Introduction

Quantum liquid helium-4 (⁴He) and its isotope liquid helium-3 (³He) exhibit unique behaviors different from normal liquids. Liquid heliums without a triple point are not solid even at absolute zero temperature unless pressurized. When the

temperature $T < T_\lambda$, where T_λ is the superfluid transition temperature, liquid heliums transit from a normal fluid to a superfluid. Helium-4 is a superfluid (termed as 'He II') at temperatures below the T_λ of 2.17 K, otherwise it is a normal liquid helium (termed as 'He I'). The T_λ of ³He is 2.6 mK, which is three orders of magnitude lower than ⁴He. And the properties of normal liquid ³He and liquid ⁴He differ significantly as the temperature decreases. Below 0.1 K, liquid ³He is a Fermi liquid and the first sound velocity is independent of temperature [1]. The Pauli Exclusion Principle causes thermal conductivity (TC) to be proportional to T^{-2} as the case of electron thermal conductivities in metals. When the temperature $0.1 \text{ K} < T < 1 \text{ K}$, the TC firstly decreases to its minimum and then increases with increasing temperature [2]. Above 1 K,

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the degeneracy effect of Fermi fluids no longer works, and interestingly, the TC of liquid ^3He becomes similar to the TC of He I. Thus, the aim of this paper is to establish a theoretical model of TCs valid for both He I and liquid ^3He above 1 K.

The TC for simple liquids decreases with increasing temperature except for a few liquids such as He I, ^3He above 1 K, and water near room temperature [3]. In addition, it has also been established that the heat capacity of He I and liquid ^3He above 1 K linearly increases with increasing temperature. Andreev [4, 5] successfully explained the linear growth trend of heat capacity and the TC by introducing tunneling states similar to amorphous materials [6, 7] considering the quantum effects related to zero-point oscillations. As shown by Andreev, semi-quantum liquids are characterized by a universal temperature dependence of the thermodynamic quantities. The main contribution to their thermodynamics is made by a mechanism similar to that proposed by Anderson *et al* [6] and by Phillips [7] to explain the low-temperature properties of glasses. Therefore, in a semi-quantum liquid, the quantum information present in TC is equivalent to that found in heat capacity. Kerrisk and McLaughlin [8] calculated the TC of normal liquid helium based on the classical cell theory of the liquid state of Lennard-Jones and Devonshire [9], assuming that a molecule of the liquid quasi-lattice is confined to its cell composed of adjacent molecules, and the results were all lower than the experimental values. Classical TC models for liquids, such as the Bridgman formula in 1923 [10, 11] and the Eyring model in 1938 [12], deviate substantially from experimental measurements of He I. In 1951, Grenier [13] observed that the TC trend of He I depicts a more gas-like behavior than a normal liquid because of the quantum effects of the uncertainty principle separating atoms from each other. Thus, the kinetic theory of monoatomic gas (KTMG, $\kappa = 2.5\eta C_v$) was employed to describe the TC of He I [13], where η is the viscosity and C_v is the heat capacity at constant volume. Although the predicted values of this theory are comparable to the measurements, it still fails to capture the positive temperature dependence of the TC (i.e. the TC increases with increasing temperature). In 1980s, Tam and Ahlers [14–16] used the group theory to analyze the TC of He I. However, due to the large number of fitting parameters and the complexity of the calculation, this theory is hardly applied in calculating the dependence of TC on temperature, pressures, etc. Numerical simulation is also an essential tool for TC prediction [17, 18]. Imaoka and Kinugawa [19], in 2017, calculated the TC of He I by using the molecule-based simulation of centroid molecular dynamics (CMD), which is a semi-classical method for estimating the collective transport properties of quantum liquids. The predicted values also could not reproduce the experimental trend of TC increasing with temperature.

Very recently, a thermal resistance network (TRN) model was proposed by several co-authors in this paper to calculate the TC of amorphous polymers whose structures can be described by the random coil model [20]. After that, we extended the TRN model to disordered condensed systems by following Einstein's idea of thermal random walk. A unified

formula suitable for various liquids and amorphous solids at high temperatures has been developed in agreement with the experimental results [21]. This unified formula portrays the arrangement characteristics of atoms inside the disordered system, i.e. short-range order and long-range disorder. This physical picture should provide a new insight into depicting the TC behavior of He I and liquid ^3He above 1 K.

In this paper, we extend our unified formula by focus on the temperature and pressure dependence of TC of He I and liquid ^3He for $T > 1$ K. The calculated values are not only in excellent agreement with the experimental data without fitting parameters, but also reproduce the experimental trend of TC increasing with temperature and pressure.

2. Methods

In 1911, Einstein [22] employed the random walk between an atom and its surrounding neighbors for thermal conductivities in liquids. While, our TRN model in this paper is more general and analogous to the nodes-links-blobs (NLB) model [23–25]. The NLB model is based on the concept that an infinite cluster contains a backbone network with a characteristic length scale. This backbone network consists of links, nodes (crossing points of links), and blobs (condensed parts of links). Thermal transfer within the condensed parts of blobs is significantly faster compared to the parts of links. In our TRN model, the backbone network consists of thermal transport channels (links), atoms (nodes), and clusters of atoms (blobs). The clusters of atoms are composed of atoms exhibiting short-range order, such as molecules in polymers or atomic groups in amorphous solids. Thermal transfer within the atoms and clusters of atoms is much faster compared to the parts of the thermal transport channels. By introducing a network structure that characterizes disordered systems, we can derive a unified formula for TC of liquids and amorphous solids as follows [21]:

$$\kappa = \left(\frac{Z}{6}\tilde{n}\right)^{1/3} h, \quad (1)$$

where Z is the average number of adjacent nodes, which is the average coordination number, and \tilde{n} is the number density of nodes. Then, in a three-dimensional isotropic system, the number density of the connecting paths is $Z\tilde{n}/6$. h is the thermal conductance between nodes representing the average energy flow across links per unit time per temperature drop. If only inter-nodal thermal transport is considered, h can be given by

$$h = \frac{C_{\text{per}}v_s}{\delta}, \quad (2)$$

where v_s is the speed of sound, δ is the distance between nodes, and C_{per} is the heat capacity of the particle. Compared to lattice TC caused by phonon diffusion, the TRN model focuses more on thermal transport channels and the atomic/molecular

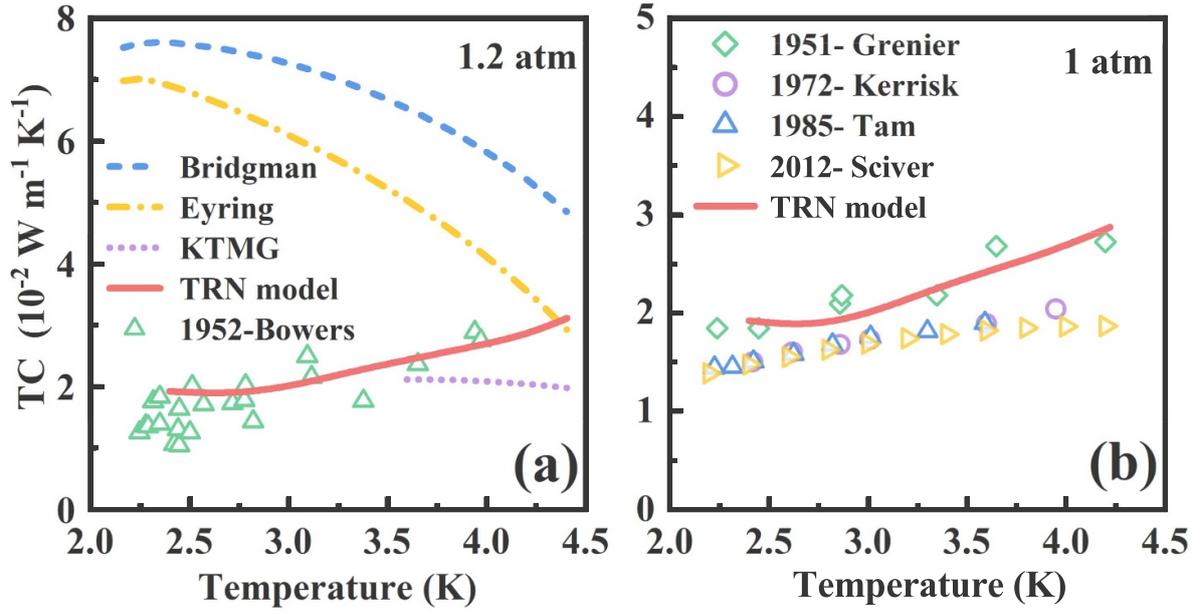


Figure 1. Calculated and observed TC of He I as a function of temperature when (a) $P = 1.2$ atm and (b) $P = 1$ atm. The TCs predicted by the Bridgman formula [10], from the Eyring model [12], and the KTMG, ($\kappa = 2.5\eta C_v$) [27] are plotted for analysis. The experimental data are taken from the measurements by Bowers [29], Grenier [13], Kerrisk and Mclaughlin [8], Tam and Ahlers [15] and Sciver [28].

structure within nodes/blobs. This approach reveals the similarity of heat conduction in liquids and amorphous solids at the microscopic level. In both long-range disordered and short-range ordered systems, thermal transport essentially becomes a thermal random walk within a specific network structure. $(Z\tilde{n}/6)^{1/3}$ in the unified formula corresponds to the number of transport channels along the temperature gradient per unit volume. And h represents the thermal conductance of each channel. Following Einstein’s concept, we assume that each atom transfers energy to its neighboring atoms within half a period. As such, we estimate the thermal conductance of each channel by dividing the heat capacity of the nodes/blobs by the number of half periods. The time taken for one-half period can be estimated using the speed of sound and the length of the links.

In the high-temperatures limit, the heat capacity is $(3/2 + D_v/2)k_B$ with three corresponding to the translational degree of freedom and D_v is the average vibrational degree of freedom of nodes/blobs. The value of D_v should be from 0 to 3. However, at low temperatures, the quantum effect is embedded in the heat capacity, which serves as an input parameter for the TRN model, directly affecting the magnitude of TC.

The cases of He I and liquid ^3He are straightforward to formulate because each atom can be regarded as a node. At a constant pressure, $C_{\text{per}} = C_{\text{per},p}$, where $C_{\text{per},p}$ is the average heat capacity of each particle. $\tilde{n} = n_{\text{mole}} = \rho/m_{\text{mole}}$, where ρ is density and m_{mole} is molecular mass. Thus, the unified formula can be expressed as,

$$\kappa = \left(\frac{Z}{6} \frac{\rho}{m_{\text{mole}}} \right)^{1/3} \frac{C_{(\text{per},p)} v_s}{\delta}. \quad (3)$$

It is obvious that m_{mole} is 4 and 3 atomic mass unit for ^4He and ^3He , respectively. $C_{\text{per},p} = C_p/N_A$, where C_p is the molar heat capacity at constant pressure and N_A is the Avogadro constant. δ can be estimated as $(\rho/m_{\text{mole}})^{-1/3}$, and ρ and v_s can be adopted from experimental measurements. Therefore, Z is the only variable to be determined in our model for different temperatures and pressures. We use the Monte Carlo method to calculate Z by considering ^3He and ^4He atoms as randomly packed spheres. The details of the Monte Carlo method have been presented in our previous work [21].

3. Results and discussions

We now use equation (3) to calculate the TC of He I at different pressures from 2.4 K to 5.1 K. This range is chosen because the gas–liquid transition temperature of helium-4 is 5.2 K, and the superfluid transition temperature is 2.17 K. The mass density, the sound velocity, and molar heat capacity as a function of temperature and pressure are taken from [26]. Figures 1(a) and (b) show the calculated and observed TC of He I as a function of temperature when $P = 1.2$ atm and 1 atm, respectively. The values calculated by the Bridgman formula [10], the Eyring model [12], and the KTMG [27] ($\kappa = 2.5\eta C_v$) are also plotted in figure 1(a) for comparison. The calculated TC from the Bridgman formula and the Eyring model deviate substantially from measurements, as shown in figure 1(a). We further show that the KTMG ($\kappa = 2.5\eta C_v$) cannot lead to the increase of TC with increasing temperature. The values predicted by our model not only agree with the experimental data, but also reproduce the experimental tendency of TC increasing with temperature without using any fitting parameters. In figure 1(b), our calculated results are in agreement with the

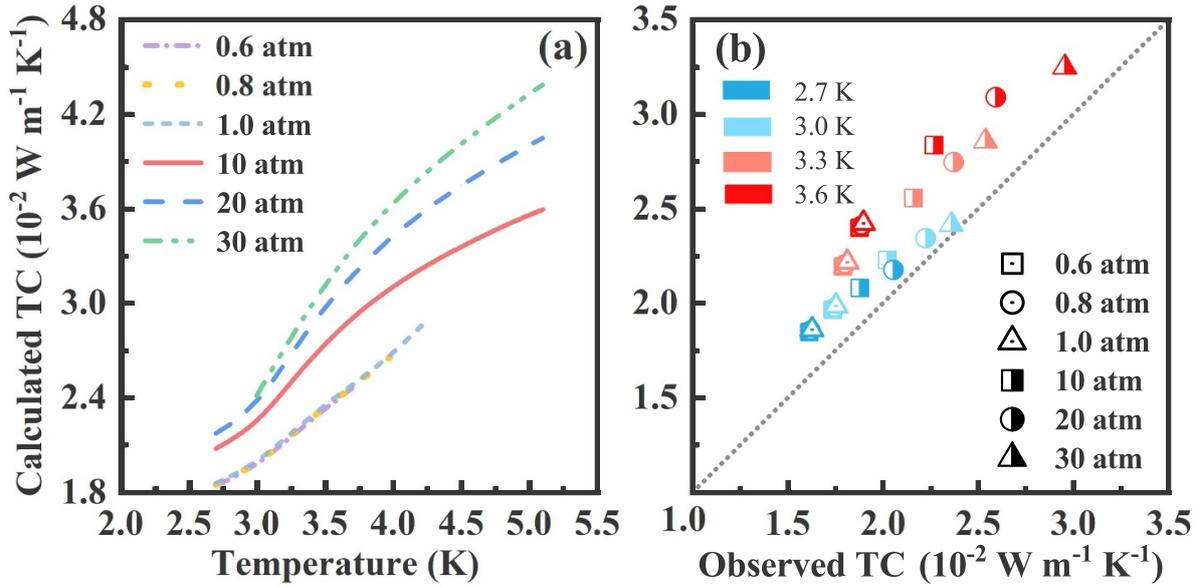


Figure 2. (a) Calculated TC of He I as a function of temperature with different pressures. (b) Calculated TC with different pressures versus the observed values by Tam and Ahlers [15].

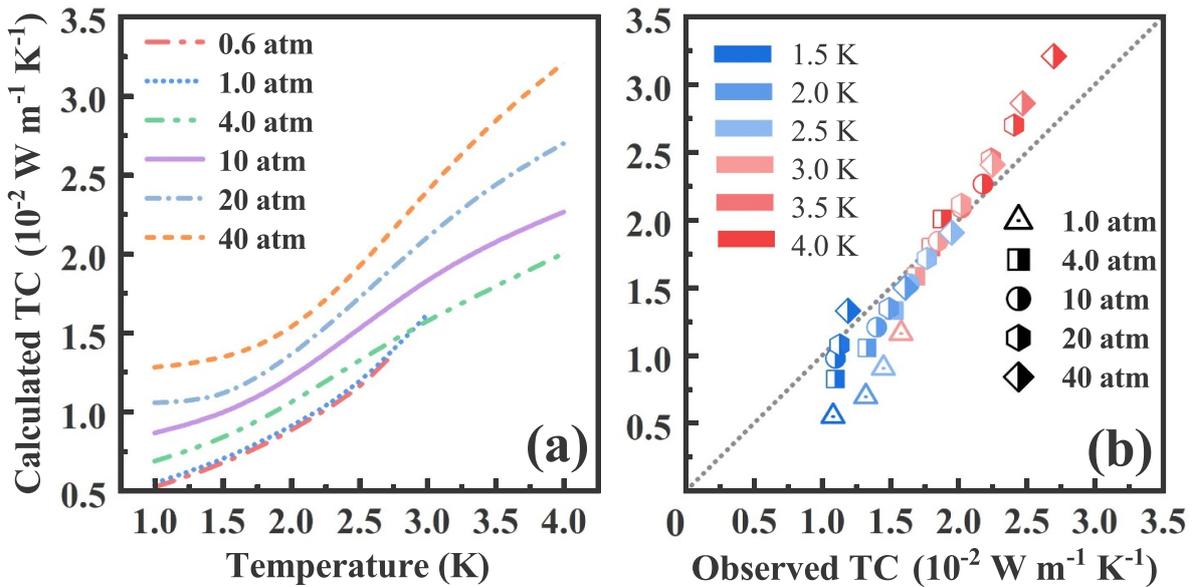


Figure 3. (a) Calculated TC of liquid ^3He as a function of temperature with different pressures. (b) Calculated TC with different pressures versus the observed values. The observed values with 1, 10, 20, and 40 atm are from Betts and Marshall [33] and 4 atm is from Tam and Ahlers [15].

TC data by Grenier [13] and slightly larger than the TC data measured by other groups [8, 13, 15, 28, 29]. This deviation mainly originates from the experimental errors due to the interfacial Kapitza resistance [30, 31]. In Grenier’s work [13], the effect of the Kapitza resistance between plates and He I was eliminated by adjusting the separation distance between the plates.

Figure 2(a) shows the calculated TC of He I as a function of temperature with different pressures. Below 1 atm, the effect of pressure on TC is negligible. When $P \geq 1$ atm, the increase in pressure leads to an increase in TC. When $T > 4$ K, the TC

of He I is found to be strongly affected by pressure. Figure 2(b) shows that TC calculated from our model is slightly larger than the observed values taken from [15]. The main reason could be the underestimation of TC due to the presence of Kapitza resistance in measurements. Our model captures the characteristics of the thermal transport behaviour of He I, which in turn perfectly reproduces the tendency of TC as a function of temperature and pressure.

We further study the TC of liquid ^3He when $T > 1$ K. The mass density, sound velocity, and molar heat capacity as a function of temperature and pressure are taken from

[32]. Although many physical properties of fermionic ^3He and bosonic ^4He are fundamentally different, the thermal transport behavior of liquid ^3He and He I is surprisingly similar. Figure 3(a) shows temperature dependence of TC of liquid ^3He under different pressures. The calculated values of TC always increase with increasing temperature. When $P < 1$ atm, the effect of pressure on TC is negligible. When $P > 1$ atm, the increase in pressure leads to a significant increase in TC. Figure 3(b) compares the calculated results with experimental ones from several groups [15, 33, 34]. Excellent agreement between calculation and experiments is achieved at high pressures. A slight deviation is found at low pressures which may result from the measurement error due to convective thermal transfer [35].

The successful prediction of the TC of normal liquid helium extends the applicability of the high-temperature TRN model to encompass the regime below Debye temperature. The main distinction between the high-temperature and low-temperature models lies in the definition of heat capacity, which incorporates information not only about the system's degrees of freedom but also its state.

4. Conclusion

By redefining the heat capacity, we utilized the TRN model to calculate the thermal conductivities of normal liquid helium-4 and helium-3 at temperatures above 1 K. The calculated values not only exhibit excellent agreement with experimental measurements but also accurately reproduce the observed trend of TC increasing with temperature and pressure. This theoretical explanation elucidates the anomalous temperature dependence of the thermal conductivity of normal liquid helium, which deviates from that of other simple liquids due to the variability of heat capacity, reflecting the quantum characteristics inherent in liquid helium.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Author contributions

Dan Huang: Formal analysis (equal); Writing-original draft (lead); Writing-review & editing (equal); Data curation (lead);

Visualization (equal). Jinxing Zhong: Writing-review & editing (equal); Software (lead); Visualization (equal); Formal analysis (equal). Saqlain Raza: Validation (equal). Tsuneyoshi Nakayama: Writing-review & editing (equal). Ran Niu, Baishan Fu, Dapeng Yu: Validation (equal). Jun Liu: Writing-review & editing (equal); Supervision (equal); Methodology (supporting); Conceptualization (supporting). Jun Zhou: Formal analysis (equal); Methodology (lead); Supervision (equal); Conceptualization (equal); Writing-review & editing (equal).

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