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Frequency Thresholds in the Energy Transmission Across Kapitza Interfaces

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When heat is conducted from a solid into liquid helium, a thermal boundary resistance occurs across the interface. This effect, known as Kapitza resistance, has been described with limited success by the acoustic impedance theory. An energy exchange mechanism is proposed in this investigation which is distinct from that of the acoustic impedance theory. The resulting model is successful in predicting the existence of frequency thresholds in the energy transmission across Kapitza interfaces. These frequency thresholds were derived from an analysis of the interaction between helium atoms from the coupling fluid and adsorbed particles which are bound to the solid surface by the Van Der Waals potential of the interface. The predicted ratio of the onset frequencies for a solid surface coupled to ^{3}He and ^{4}He gas was found to be approximately equal to the square root of the mass ratio. This result is in good agreement with experimental observations.

1. INTRODUCTION

When heat is conducted from a solid into liquid helium, a finite temperature jump ΔT occurs across the interface.[1] If the amount of heat flow per unit area $\dot{q}$ is sufficiently small, then ΔT should be proportional to it. The ratio,

$$R_K = \frac{\Delta T}{\dot{q}} \tag{1}$$

represents a thermal resistance for the interface. This phenomenon, known as Kapitza resistance, has been a topic of both experimental and theoretical interest for more than half a century. Interest in this subject has remained strong for two main reasons: the inherent difficulties in attaining ever lower temperatures, and the fact that this effect is so poorly understood.

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The most widely accepted theory of Kapitza resistance was developed by Khalatnikov,[2] and is known as the acoustic impedance theory. This theory is based upon the continuum mechanics concepts of classical wave propagation in elastic media. When thermal phonons strike the interface between a solid and liquid helium from the solid side, classical mechanics stipulates that the component of momentum parallel to the interface must be conserved. Since liquid helium and most solids differ in both density and sound velocity by at least an order of magnitude, this requirement can only be satisfied if the majority of the incident phonons are reflected back into the solid. The large mismatch in physical properties of the two media impedes the transmission of phonon energy across the interface. This impedance results in the temperature discontinuity which is observed at the interface when thermal phonons are transmitted across it.

Experimental values of R_K are typically 1 to 2 orders of magnitude less than the predictions of the acoustic impedance theory,[3] except at very low phonon temperatures, where somewhat better agreement has been observed.[4] This discrepancy between theoretical prediction and experimental observation has not yet been resolved, despite several attempts to improve the theory by adding corrective terms which make the model more physically realistic.[5–8] In view of this, it has been proposed[9] that an alternative channel of energy transmission may exist at Kapitza interfaces which is not accounted for by the acoustic impedance theory. If correct, this energy exchange mechanism is responsible for a variety of experimentally observed characteristics including frequency thresholds in the energy transmission, and the dependence of these frequency thresholds upon the mass of the coupling fluid.[10–11]

The purpose of this paper is to describe in detail an energy exchange mechanism that is distinct from that of the acoustic impedance theory. This mechanism is based in part upon the experimental observation[12–13] that foreign atoms and molecules, including those which comprise the coupling fluid, may condense upon the solid surface due to the Van Der Waals potential of the interface. These observations have contributed to the development of several theoretical models[14–18] which attempt to account for the role of both continuous and discontinuous adsorbed films and surface particles in the Kapitza problem. While these theories are each successful in predicting specific characteristics of the Kapitza resistance, none provide a mechanism for energy exchange between the helium and the adsorbed particles on an atomic level. Since the transmission of energy across such an interface actually occurs on an atomic scale, an atomic theory of this process would prove useful.

The construction of such a theory requires as a prerequisite appropriate models for both the liquid helium and the atomic structure of the

solid surface. In a practical sense, this can only be accomplished in certain special cases. For example, a complete theoretical analysis of the interactions which occur between the helium and the adsorbed particles cannot be performed without a detailed statistical knowledge of the dynamic properties of the helium particles. And liquid helium in particular poses a number of difficulties due to its unique properties. Use of the ideal gas model for liquid helium addresses these issues, and may be justified because the thermal properties of liquid helium are similar to those of a high density ideal gas.

Furthermore, the atomic structure of an interface cannot in most cases be determined exactly. But theoretical attempts to describe the power flux through an arbitrary interface requires the assumption of a specific surface structure. This aspect of the modeling process provides the most difficult challenge because real experimental surfaces vary greatly in their levels of atomic roughness, damage and contamination, and residual stress. With the exception of the special cases where the interface is laser annealed[19] or cleaved in situ,[20] most experimental surfaces deviate to some degree from that of an ideal surface. For the purposes of this investigation, it is assumed that the levels of damage and residual stress at the surface do not result in significant scattering of the incident phonons.

Scattering is also a concern when the wavelength of the incident phonons becomes comparable to the characteristic length scale of the surface roughness. For most Kapitza interfaces enhanced transmission is initiated at predominant phonon frequencies between 10 GHz and 100 GHz. For a typical solid such as sapphire, which has a longitudinal wave speed of 10.9×10^3 m/s, the predominant phonon wavelengths lie between 10^3 Å and 10^4 Å. If the length scale of the surface roughness is much less than 10^3 Å, then the surface should appear relatively flat to the incident phonons. In most cases, the experimental surfaces are atomically rough. If the characteristic length scale of the surface roughness is much less than the predominant phonon wavelength, then to a good approximation these solid surfaces may be modeled as if they were atomically flat.

The model which is developed in this investigation involves a simplified Kapitza interface where an ideal gas is coupled to an atomically flat surface onto which foreign particles are adsorbed. The objective is to determine how these adsorbed particles affect the transmission of energy across the interface. Classical collision theory is employed to construct an idealized 3-dimensional model of the interactions which occur between the adsorbed particles and the ideal gas. As phonons strike the interface from the solid side, the bound surface particles are set in motion. The incident phonons therefore impart both energy and momentum to the bound particles. As helium atoms from the ideal gas strike the interface, energy

may be exchanged between the helium and the bound particles through momentum transfer. The details of this model are presented in the following section.

2. CLASSICAL THEORY OF A SOLID-IDEAL GAS INTERFACE

Consider Fig. 1 which schematically illustrates a spherical particle of mass m_1 and radius r_1 bound to a point on the surface of a linearly elastic, solid half-space. A monatomic ideal gas at temperature T and pressure $\mathscr{P}$ occupies the half-space above the solid, and the spherical atoms which comprise the gas each have mass m_2 and radius r_2. The bound particle and the ideal gas are initially in a state of thermal equilibrium. The origin of a Cartesian coordinate system is set at the bound particle's equilibrium point when the surface is at rest, with the z-axis perpendicular to the solid surface, and the positive y-axis into the page. These axes are fixed in the rest frame of the laboratory, so that this coordinate system represents an

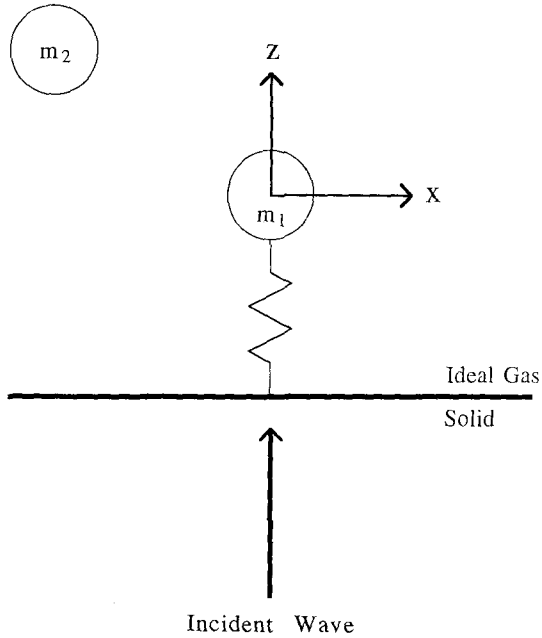

Fig. 1. A particle of mass m_1 is bound to a point on the solid surface by a simple harmonic potential function. Atoms of mass m_2 from the ideal gas strike the bound particle from above, while an incident longitudinal wave from the solid provides a driving force for the oscillator.

inertial frame of reference. A monochromatic longitudinal wave propagates through the solid and strikes the interface at normal incidence.

If the stresses and displacements are continuous at the boundary, then classical wave theory (and the acoustic impedence theory) correctly predict that a transmitted longitudinal wave will propagate in the ideal gas, and a reflected longitudinal wave in the solid. If the solid surface is periodically displaced along the z axis with an amplitude A and frequency ω, then this oscillation provides a driving force for the adsorbed particle. Provided the displacements of the adsorbed particle from its equilibrium position are sufficiently small, the potential field which binds it to the solid surface may be approximated by a simple harmonic potential function. The potential function used to model the interactions between the bound particle and the ideal gas particles is the hard sphere interaction potential V_I which is given by

$$V_I = \begin{cases} 0, & |\overrightarrow{R_2} - \overrightarrow{R_1}| > r_1 + r_2 \\ \infty, & |\overrightarrow{R_2} - \overrightarrow{R_1}| \leqslant r_1 + r_2 \end{cases} \tag{2}$$

where $\overrightarrow{R_1} = (x_1, y_1, z_1)$ and $\overrightarrow{R_2} = (x_2, y_2, z_2)$ are the position vectors of the bound particle and an ideal gas particle, respectively.

When a collision occurs between these particles, the separation between the particle centers is equal to the sum of their respective radii, and the strength of the interaction potential in (2) is much larger than the binding potential. Under these circumstances the binding potential may effectively be ignored during the collision. This impulse approximation treats the bound particle as if it were a free particle at the time of collision, and reduces the complexity of a 3-body problem to that of a 2-body problem which can be solved easily. The final momentum of each particle may be determined by applying the laws of conservation of linear momentum, and conservation of energy. These expressions for the final momenta may then be used to calculate the change in kinetic energy of each particle due to the collision. If the energy exchanged per collision is multiplied by the number of collisions per unit area per unit time, and the result is averaged over all possible collisions, then the average power flux through the oscillator will be determined. In this manner it may be determined how the adsorbed foreign particle produces a channel of energy transmission in addition to that of the acoustic mismatch theory.

If the collision between the bound particle and the ideal gas particle is completely elastic, then there is no dissipation of energy due to frictional forces. Applying the impulse approximation, conservation of linear momentum requires

$$\overrightarrow{P_1} + \overrightarrow{P_2} = \overrightarrow{P_1'} + \overrightarrow{P_2'} \tag{3}$$

while conservation of energy requires

$$\frac{|\overrightarrow{P_1}|^2}{2m_1}+\frac{|\overrightarrow{P_2}|^2}{2m_2}=\frac{|\overrightarrow{P_1'}|^2}{2m_1}+\frac{|\overrightarrow{P_2'}|^2}{2m_2} \tag{4}$$

Given the assumptions made in this model, the change in each particle's momentum due to the collision will occur along the direction vector which joints their centers at the time of impact. Solving (3) and (4) simultaneously, the final momentum of each particle is given by $\overrightarrow{P_1'}$ and $\overrightarrow{P_2'}$ where

$$\overrightarrow{P_1'}=\overrightarrow{P_1}+\frac{2m_2}{m_1+m_2}\left[\left(\frac{m_1}{m_2}\overrightarrow{P_2}-\overrightarrow{P_1}\right)\circ\vec{e}\right]\vec{e} \tag{5}$$

$$\overrightarrow{P_2'}=\overrightarrow{P_2}-\frac{2m_2}{m_1+m_2}\left[\left(\frac{m_1}{m_2}\overrightarrow{P_2}-\overrightarrow{P_1}\right)\circ\vec{e}\right]\vec{e} \tag{6}$$

and the unit vector $\vec{e}$ is given by

$$\vec{e}=\frac{1}{r_2+r_1}[\overrightarrow{R_2}-\overrightarrow{R_1}] \tag{7}$$

The change in kinetic energy of the ideal gas particle due to the collision with the bound particle is therefore

$$\Delta KE_2=\frac{1}{2m_2}[|\overrightarrow{P_2'}|^2-|\overrightarrow{P_2}|^2] \tag{8}$$

Substituting (6) into (8),

$$\Delta KE_2=\frac{2m_2\Gamma^2}{(m_1+m_2)^2}-\frac{2\Gamma}{m_1+m_2}(\overrightarrow{P_2}\circ\vec{e}) \tag{9}$$

where Γ is given by

$$\Gamma=\left(\frac{m_1}{m_2}\overrightarrow{P_2}-\overrightarrow{P_1}\right)\circ\vec{e} \tag{10}$$

The number of collisions $d\dot{N}$ between the ideal gas particles and the oscillator per unit area of the oscillator surface per unit time is given by,[21]

$$d\dot{N}=-\frac{\rho_2\Gamma}{m_1m_2}\left(\frac{1}{2\pi m_2kT}\right)^{3/2}\exp\left[\frac{|\overrightarrow{P_2}|^2}{-2m_2kT}\right]d^3P_2;\ P_{2z}\leqslant O \tag{11}$$

where the term d^3P_2 is the differential element defined by

$$d^3P_2 = dP_{2x}\, dP_{2y}\, dP_{2z} \tag{12}$$

and ρ_2 is the density of the ideal gas. The power flux $d\dot{q}$ through the oscillator is the product of ΔKE_2 and $d\dot{N}$. It is given by

$$d\dot{q} = -\frac{\rho_2}{m_1 m_2}\left(\frac{1}{2\pi m_2 kT}\right)^{3/2}\left\{\frac{2m_2\Gamma^3}{(m_1+m_2)^2} - \frac{2\Gamma^2}{m_1+m_2}(\overrightarrow{P_2}\circ\vec{e})\right\}$$
$$\times \exp\left[\frac{|\overrightarrow{P_2}|^2}{-2m_2kT}\right] d^3P_2 \tag{13}$$

By definition the bound particle is localized in space, with an amplitude of oscillation which is sufficiently small such that its motion may be described as simple harmonic. The bound particle is therefore close to the equilibrium point during the course of its oscillations. The spatial dependence of $d\dot{q}$, which is contained in the term $\vec{e}$, is a function of the relative position vectors of the two colliding particles. Imposing the approximation that the position vector of the bound particle is the origin of the coordinate system (i.e., $\overrightarrow{R_1} = (0, 0, 0)$) at the time of collision should simplify the calculation of the average power flux without compromising the accuracy of the final result. Applying this approximation to (13), averaging over all possible position coordinates of the ideal gas particles at the time of impact, and integrating over the momentum components of the ideal gas,

$$\dot{q}_{avg} = -\frac{\rho_2}{4m_1m_2(m_1+m_2)^2}$$
$$\times\left\{\frac{5m_1^2}{2\pi^2 m_2}(2\pi m_2 kT)^{3/2} + \frac{1}{2}m_2kT\left[\frac{8m_1m_2 - 4m_1^2}{m_2}P_{1z}\right]\right.$$
$$+\sqrt{2\pi m_2 kT}\left[\frac{1}{2\pi}(m_2 - 2m_1)(P_{1x}^2 + P_{1y}^2 + 2P_{1z}^2)\right]$$
$$\left.-\frac{3}{2}m_2P_{1z}\left(P_{1x}^2 + P_{1y}^2 + \frac{2}{3}P_{1z}^2\right)\right\} \tag{14}$$

Consider the expressions which appear on the right hand side of (14) which involve the terms P_{1z}, $P_{1z}P_{1x}^2$, $P_{1z}P_{1y}^2$, and P_{1z}^3. These terms average out to zero over an integer number of cycles of the incident wave provided the oscillator is in steady state during its interaction with the ideal gas particles. This can be most clearly understood by noting that each of these

terms is an odd function of time in steady state. If the brackets $\langle\ \rangle$ denote the average value of a quantity over an integer number of cycles of the incident wave, then the time-averaged power flux through the oscillator becomes,

$$\langle \dot{q}_{avg} \rangle = -\frac{1}{2\pi} \frac{\rho_2 \sqrt{2\pi m_2 kT}}{4m_1 m_2 (m_1 + m_2)^2} \times [10 m_1^2 kT - (2m_1 - m_2)\langle P_{1x}^2 + P_{1y}^2 + 2P_{1z}^2 \rangle] \tag{15}$$

An analysis of the driven harmonic oscillator shows that[22]

$$\langle P_{1x}^2 \rangle = m_1 kT \tag{16}$$

$$\langle P_{1y}^2 \rangle = m_1 kT \tag{17}$$

$$\langle P_{1z}^2 \rangle = m_1 kT + \frac{1}{2} \frac{m_1 CA^2 \omega_n^2 \omega^2}{(\omega_n^2 - \omega^2)^2 + 4\beta^2 \omega^2} \tag{18}$$

where the oscillator's natural frequency ω_n is defined as

$$\omega_n = \sqrt{\frac{C}{m_1}} \tag{19}$$

The term $m_1 kT$ results from the equilibrium state between the oscillator and the ideal gas prior to the arrival of the incident longitudinal wave. C is the curvature of the binding potential at the equilibrium separation and β represents the damping which the oscillator experiences due to its interaction with the solid surface. Substituting (16), (17), and (18) into (15) and regrouping terms,

$$\langle \dot{q}_{avg} \rangle = -\frac{1}{2\pi} \frac{\rho_2 \sqrt{2\pi m_2 kT}}{(m_1 + m_2)^2} \times \left[\left(1 + \frac{m_1}{2m_2} \right) kT - \left(\frac{m_1}{2m_2} - \frac{1}{4} \right) \frac{CA^2 \omega_n^2 \omega^2}{(\omega_n^2 - \omega^2)^2 + 4\beta^2 \omega^2} \right] \tag{20}$$

3. FREQUENCY THRESHOLDS

A net power flux from the adsorbed particle to the ideal gas will be present when

$$\langle \dot{q}_{avg} \rangle > 0 \tag{21}$$

Examination of (20) reveals that this condition will be satisfied provided

$$\left(1+\frac{m_1}{2m_2}\right)kT<\left(\frac{m_1}{2m_2}-\frac{1}{4}\right)\frac{CA^2\omega_n^2\omega^2}{(\omega_n^2-\omega^2)^2+4\beta^2\omega^2} \tag{22}$$

By solving the inequality in (22) for ω, the general solutions for the threshold frequencies may be determined. Consider the special case where individual atoms from the ideal gas condense upon the solid surface due to the Van Der Waals potential of the interface. If these adsorbed atoms serve as non-interacting oscillators, then the mass of the oscillator equals the mass of an ideal gas particle, i.e., $m_1=m_2$. Under these circumstances (22) reduces to

$$\frac{3}{2}kT<\frac{1}{4}\frac{CA^2\omega_n^2\omega^2}{(\omega_n^2-\omega^2)^2+4\beta^2\omega^2} \tag{23}$$

The term on the left hand side of the inequality in (23) is the mean translational kinetic energy of an ideal gas particle. The term on the right hand side of this inequality represents the time averaged kinetic energy imparted to the oscillator by the incident longitudinal wave. In this special case, the initiation criteria stated in (21) reduces to a simple energy balance. A net power flux will occur from the oscillator to the ideal gas when the time averaged kinetic energy imparted to the oscillator by the incident longitudinal wave exceeds the mean translational kinetic energy of an ideal gas particle. If the oscillator is sufficiently underdamped such that $\beta \ll \omega_n$ then (23) simplifies to

$$\frac{3}{2}kT<\frac{1}{4}\frac{CA^2\omega_n^2\omega^2}{(\omega_n^2-\omega^2)^2}; \qquad \omega\neq\omega_n \tag{24}$$

Solving (24) for ω, a net power flux will occur from the oscillator to the ideal gas when the driving frequency of the incident wave falls between the following values,

$$\omega_L<\omega<\omega_U \tag{25}$$

where

$$\omega_L=\omega_n\left[\sqrt{1+\frac{CA^2}{24kT}}-\sqrt{\frac{CA^2}{24kT}}\right] \tag{26}$$

and

$$\omega_U=\omega_n\left[\sqrt{1+\frac{CA^2}{24kT}}+\sqrt{\frac{CA^2}{24kT}}\right] \tag{27}$$

Frequency thresholds have been observed in phonon reflection studies of Kapitza interfaces. Koblinger *et al.*[10,11] used superconducting tunnel junctions to emit frequency-tunable monochromatic phonons at normal incidence into insulator crystal substrates. A sharp amplitude reduction in the reflected phonon signal was observed at 85 GHz when the tunneling diodes were covered by liquid or gaseous ^{4}He. When the same surface was placed in contact with ^{3}He gas, a shift in the threshold frequency to 100 GHz was observed. These results can be accounted for by the current theory.

Examination of (26) shows that the lower threshold frequency ω_L is a function of several variables: the natural frequency of oscillation (ω_n), the helium temperature (T), the amplitude of the surface displacement (A), and the curvature of the binding potential at the equilibrium separation (C). A helium temperature of 1 K was used in the experiments conducted by Koblinger *et al.* for contact with both ^{3}He and ^{4}He, so the values of T are the same in both cases. Since the phonon signal was generated by the same source under the same set of conditions, the amplitudes A are also approximately equal. ^{3}He and ^{4}He differ only in mass. The "spring constant" of the binding potential is not a function of mass, so to a good approximation the values of C are also equal. If subscripts of 3 and 4 are used to denote properties associated with ^{3}He and ^{4}He, respectively, then the ratio of the lower threshold frequencies for contact with ^{3}He and ^{4}He is given by

$$\frac{\omega_{L3}}{\omega_{L4}} = \frac{\omega_{n3}}{\omega_{n4}} \tag{28}$$

where ω_{n3} and ω_{n4} are the natural frequencies of oscillation for ^{3}He and ^{4}He atoms, respectively. Substituting for ω_{n3} and ω_{n4} from (19) into (28), and noting $C_3 = C_4$,

$$\frac{\omega_{L3}}{\omega_{L4}} = \sqrt{\frac{m_4}{m_3}} \tag{29}$$

The atomic masses for ^{3}He and ^{4}He are[23]

$$^3\text{He mass } m_3 = 5.0095 \times 10^{-27}\ \text{kg} \tag{30}$$

$$^4\text{He mass } m_4 = 6.6482 \times 10^{-27}\ \text{kg} \tag{31}$$

Substituting (30) and (31) into (29),

$$\frac{\omega_{L3}}{\omega_{L4}} = \sqrt{\frac{6.6482 \times 10^{-27}\ \text{kg}}{5.0095 \times 10^{-27}\ \text{kg}}} = 1.15 \tag{32}$$

The ratio of the onset frequencies measured by Koblinger *et al.* is

$$\frac{\omega_{L3}}{\omega_{L4}} = \frac{100\ \text{GHz}}{85\ \text{GHz}} = 1.18 \tag{33}$$

The ratio of the onset frequencies predicted by the current theory differs from the experimentally determined value of 1.18 by less than 3%.

4. CONCLUSIONS

The model which is presented in this paper is successful in predicting the existence of frequency thresholds in the energy transmission across Kapitza interfaces. These frequency thresholds were derived from an analysis of the interaction between helium atoms from the coupling fluid and adsorbed foreign particles which are bound to the solid surface by the Van Der Waals potential of the interface. For the special case where He atoms are adsorbed onto the solid surface, the ratio of the onset frequencies for a solid in contact with ^{3}He and ^{4}He was found to be approximately equal to the square root of the mass ratio. This mass dependence was observed in the investigations of Koblinger *et al.*[10,11] where superconducting tunnel junctions were used to emit frequency-tunable monochromatic phonons into insulator crystal substrates. A sharp reduction in the reflected phonon signal was observed at 85 GHz when the tunneling diodes were covered by liquid or gaseous ^{4}He. When the same surface was placed in contact with ^{3}He gas, a shift in the threshold frequency to 100 GHz was observed. The ratio of the threshold frequencies measured experimentally is 1.18, which differs from the theoretically predicted value of 1.15 by less than 3%.

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