

## Theory of thermal boundary resistance between small particles and liquid helium: Size effect on phonon conduction

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The size effect for the heat transfer between small particles and liquid helium is theoretically investigated at low temperatures. The general expression is presented for the thermal boundary resistance due to phonon conduction between a small particle and liquid helium. The resistances are calculated under some experimental conditions, which exhibit a  $T^{-3}$  variation above some temperature. Above this temperature the absolute magnitude of the calculated resistance also coincides with that of the acoustic mismatch theory for a bulk solid. Below its temperature the resistance increases exponentially with decreasing temperature. This temperature depends on both the size and the elastic properties of a small particle.

### I. INTRODUCTION

It is known that a finite-temperature discontinuity develops between a solid and liquid helium when a heat flux is maintained across the interface. For small temperature differences, the ratio of the temperature jump to the heat flux is defined as the thermal boundary resistance  $R_K$ . The discontinuity was first discovered by Kapitza,<sup>1</sup> and  $R_K$  is now referred to as the Kapitza resistance. The theory of the Kapitza resistance has been given by Khalatnikov,<sup>2</sup> which (the so-called acoustic mismatch theory) predicts that the conductance  $h_K = R_K^{-1}$  exhibits a  $T^3$  variation. The physical ground of this temperature dependence arises from Debye spectrum of the phonons in a solid and therefore one would expect a close  $T^3$  dependence of the conductance  $h_K$  for most *bulk* solids. The experimental data are generally in qualitative agreement with the acoustic mismatch theory<sup>2</sup> below about 0.1 K, although the actual magnitude of  $R_K$  is often one order of magnitude smaller than the calculated one.<sup>3</sup>

In the experiments on the thermal resistance below about 100 mK, solids have been often used in the form of small particles with micron size in order to make the surface-to-volume ratio large.<sup>4-9</sup> There are several cases<sup>7-9</sup> using metallic particles which contain magnetic impurities in low concentration. Mills and Béal-Monod<sup>10,11</sup> discussed in detail the contribution of magnetic coupling for these systems. At low temperatures the finite size of small particles may play some important roles for heat exchange. Harrison and McColl<sup>12</sup> and Harrison<sup>13</sup> have recently made remarks on the importance of the size effect for phonon conduction to the heat transfer between small particles and liquid helium at very low temperatures. However, no theoretical attempts have yet been

made in order to clarify the size effect on the heat transfer.

This paper presents a theory for the thermal boundary resistance due to phonon conduction between small particles and liquid helium. In Sec. II, we describe, at first, the expression of the phonon emission from a small particle to liquid helium. From this expression the explicit form of the thermal boundary resistance  $R_K$  is written down in such a form as it depends on the surface displacement due to thermally excited phonons in a particle. Particles used in experiments take various form such as flakes or powders with micron size.<sup>12,13</sup> In this paper, an elastic sphere is employed as a model of small particles. It will be argued later that the shape of particles does not seriously give rise to the correction against the main result of the present work. In Sec. III, the eigenmodes of elastic waves in a spherical particle are discussed in detail. We show that only the spheroidal modes contribute to the heat transfer. The discreteness of the eigenfrequencies and the presence of the finite lowest eigenfrequency will become clear in this section. In Sec. IV, the displacement field due to the spheroidal waves is quantized. In Sec. V, the thermal resistances  $R_K$  are calculated by illustrating copper and silver particles with various radii. It is shown that the  $R_K$  increases exponentially with decreasing temperature below the temperature corresponding to the finite lowest eigenfrequency. Above this temperature, the  $R_K$  shows the close  $T^{-3}$  dependence as well as those between most *bulk* solids and liquid helium. In addition, it will be shown that the present theory recovers the acoustic mismatch theory<sup>2</sup> at large particle size. In Sec. VI, we summarize and discuss our results. Some remarks are made about the experimental data on the thermal boundary resistance  $R_K$  between metallic particles and liquid helium.

## II. GENERAL EXPRESSION FOR THERMAL BOUNDARY RESISTANCE $R_K$ BETWEEN A SMALL PARTICLE AND LIQUID HELIUM

### A. Emission of sound waves from a small particle

Let us consider an elastic body with finite size oscillating in liquid helium, which causes a periodic compression and rarefaction of the density of liquid helium near it. These propagate in the form of sound waves. The mean rate of emitted energy from the small particle is given in terms of the square of the fluid velocity  $\vec{v}(\vec{r})$ ,<sup>14</sup>

$$\dot{\epsilon} = \rho_L c_L \int \int |\vec{v}(\vec{r})|^2 dS, \quad (2.1)$$

where  $\rho_L$  and  $c_L$  are the mass density and the sound velocity of liquid helium, respectively. The integral is performed over a closed surface surrounding the small particle. The energy carried away by sounds is supplied from the kinetic energy of the surface motion of the small particle. We take the velocity potential in liquid helium as the scalar function  $\phi(\vec{r}, t)$  defined by  $\vec{v}(\vec{r}, t) = \text{grad} \phi(\vec{r}, t)$ . The equation of motion for the velocity potential is derived from the Euler's fluid equation. The general solution can be written as the superposition of the velocity potential  $\phi_q(\vec{r}) \exp(-i\omega_q t)$  belonging to the eigenfrequency  $\omega_q = c_L q$ . At sufficiently far from the spherical particle, having its center at the origin, the solution may be written simply in the form

$$\phi_q(\vec{r}) = f_q(\theta, \phi) \exp[iq(r-R)]/r, \quad (2.2)$$

where  $R$  is the radius of the particle. The function  $f_q(\theta, \phi)$  is determined from the boundary conditions on the surface of the particle. By taking the gradient of Eq. (2.2), we obtain the expression for the square of velocity at  $\vec{r}$  in terms of the surface velocity  $\vec{v}(R, \theta, \phi)$  of the particle as

$$[\vec{v}^q(\vec{r})]^2 = \frac{R^4}{2r^4} \left[ \frac{1+(qr)^2}{1+(qR)^2} |v_r^q(R, \theta, \phi)|^2 + [ |v_\theta^q(R, \theta, \phi)|^2 + |v_\phi^q(R, \theta, \phi)|^2 ] \right]. \quad (2.3)$$

Since the second and third term in Eq. (2.3) are negligible compared with the first term at large distance  $r \gg R$ , we have

$$[\vec{v}^q(\vec{r})]^2 = \frac{1}{2r^2} \frac{q^2 R^4}{1+q^2 R^2} |v_r^q(R, \theta, \phi)|^2. \quad (2.4)$$

Substituting Eq. (2.4) into Eq. (2.1) and taking the

closed surface of the integration to be a sphere of radius  $r$ , the mean energy emitted per unit time and unit area is found to be

$$\dot{\epsilon}_q = \frac{\rho_L c_L}{8\pi} \frac{q^2 R^2}{1+q^2 R^2} \int_0^{2\pi} \int_0^\pi |v_r^q(R, \theta, \phi)|^2 d\Omega, \quad (2.5)$$

where  $d\Omega = \sin\theta d\theta d\phi$ . It should be noted that in the limit  $R \rightarrow \infty$  Eq. (2.5) reduces to the rate of emission of sound waves per unit area from plane surface. On the surface of the small particle, the fluid velocity  $\vec{v}(R, \theta, \phi)$  must be equal to that of the surface motion of the small particle. Provided that the surface displacement of the small particle is defined as  $\bar{u}(R, \theta, \phi, t)$ , its time derivative is simply the velocity of the surface motion, i.e.,  $\vec{v}(R, \theta, \phi, t) = \partial \bar{u}(R, \theta, \phi, t) / \partial t$ . Thus, we can estimate the rate of emitted energy  $\dot{\epsilon}_q$  from the small particle by the surface displacement  $\bar{u}(R, \theta, \phi, t)$ .

### B. Expression of the thermal boundary resistance $R_K$

Equation (2.5) is the classical formula for the rate of emission of sound waves due to surface oscillation of a particle. In order to obtain the expression of the heat transfer it is convenient to write down the formula (2.5) in terms of the quantized displacement. The displacement vector  $\bar{u}(\vec{r}, t)$  at an arbitrary position  $\vec{r}$  in a spherical body is expressed by the sum of the eigenmodes. If the eigenmode belonging to the eigenfrequency  $\omega_j$  is defined by  $\bar{u}_j(\vec{r})$ , the elastic waves are quantized by replacing the amplitude of the expansion by the boson operators  $a_j$  and  $a_j^\dagger$ .  $J$  stands for a set of possible quantum numbers specifying the eigenmodes in the spherical particle. The explicit form of quantum number  $J$  will be given in the Sec. IV. The displacement-field operator is written in the general form

$$\begin{aligned} \bar{u}_{\text{op}}(\vec{r}, t) &= \sum_J \bar{u}_{\text{op}}^J(\vec{r}, t) \\ &= \sum_J \left( \frac{\hbar}{2\rho_P \omega_j V} \right)^{1/2} [a_J \bar{u}_J(\vec{r}) e^{-i\omega_j t} + \text{H.c.}], \end{aligned} \quad (2.6)$$

where  $V$  and  $\rho_P$  are the volume and the mass density of a spherical particle, respectively. Putting  $v_r(R, \theta, \phi) = \dot{u}_r(R, \theta, \phi)$  into Eq. (2.5) and taking the thermal average of the square of velocity at a finite temperature, the heat flux density  $\dot{Q}_P(T)$  into liquid helium is given by summing up over all of the eigenmodes  $\{J\}$

$$\dot{Q}_P(T) = \frac{\rho_L c_L}{4\pi} \sum_J \frac{\omega_j^2 R^2}{c_L^2 + \omega_j^2 R^2} \int_0^{2\pi} \int_0^\pi T_r [e^{-\beta H} \dot{u}_{\text{op}, r}^J(R) \dot{u}_{\text{op}, r}^J(R)] d\Omega, \quad (2.7)$$

where  $H = \sum_j \hbar \omega_j a_j^\dagger a_j$  and  $\beta = 1/k_B T$ . We have used the relation that the eigenfrequency  $\omega_j$  in the particle is the same with that of the compression wave in liquid, i.e.,  $\omega_j = q c_L$ , originating from the energy conservation law.

Let us consider the heat flow from a small particle at temperature  $T_1$  into liquid helium at  $T_2$ . The net heat flux across the interface is given by the difference of the heat flux per unit area,

$$\Delta \dot{Q}_P = \dot{Q}_P(T_1) - \dot{Q}_L(T_2) , \quad (2.8)$$

where  $\dot{Q}_L(T_2)$  is the heat flux density from liquid helium into the particle. Since the net heat flow has to vanish at equal temperature ( $T_1 = T_2$ ), we have the relation  $\dot{Q}_L(T_1) = \dot{Q}_P(T_1)$ . If the difference in temperature ( $\Delta T = T_1 - T_2$ ) is small ( $T_2 \gg \Delta T$ ), Eq. (2.8) becomes

$$\Delta \dot{Q}_P(T) = h_K \Delta T , \quad (2.9)$$

where the heat conductance is defined as

$$h_K = \frac{\partial \dot{Q}_P(T)}{\partial T} . \quad (2.10)$$

The thermal resistance is obtained by the inverse of the conductance  $R_K = h_K^{-1}$ . The rest to be done is to determine the eigenmodes of elastic waves in a particle and to quantize them.

### III. EIGENMODES OF ELASTIC WAVES IN A SPHERICAL PARTICLE

The small particle considered here is assumed to be spherical and isotropic as mentioned in Sec. II. The elastic wave equation for the displacement vector  $\vec{u}(\vec{r}, t)$  can be written down as

$$\rho_P \frac{\partial^2 \vec{u}(\vec{r}, t)}{\partial t^2} = (\lambda + 2\mu) \vec{\nabla} [\vec{\nabla} \cdot \vec{u}(\vec{r}, t)] - \mu \vec{\nabla} \times [\vec{\nabla} \times \vec{u}(\vec{r}, t)] , \quad (3.1)$$

where  $\lambda$  and  $\mu$  are Lamé coefficients. The elastic motion may include three independent orthogonal modes. The displacement vector  $\vec{u}(\vec{r}, t)$  is expressed through a scalar potential  $\psi_1$  and vector potentials  $\vec{\psi}_2$  and  $\vec{\psi}_3$ , using the equation

$$\vec{u}(\vec{r}, t) = \vec{\nabla} \psi_1 + \vec{\nabla} \times \vec{\psi}_2 + \vec{\nabla} \times (\vec{\nabla} \times \vec{\psi}_3) . \quad (3.2)$$

The first term is the longitudinal mode with dilatation. The others indicate the two transverse modes without dilatation. Without loss of generality, we can take the vector potentials as

$$\vec{\psi}_i = (r, 0, 0) \psi_i \quad (i = 2, 3) . \quad (3.3)$$

As will be seen later, the above definition of the vector potentials gives us a good perspective for deter-

mining the eigenmodes in the spherical body.

Now we consider small particles in contact with liquid helium. Since liquid helium has a small mass density ( $\rho_L \sim 0.142 \text{ g cm}^{-3}$  for  $^4\text{He}$ ,  $\rho_L \sim 0.0815 \text{ g cm}^{-3}$  for  $^3\text{He}$ ) compared with those of small particles, the appropriate boundary conditions determining the eigenmodes may be taken as those for a stress-free surface. The boundary conditions are written in terms of the potential functions

$$[(\lambda \vec{\nabla} \cdot \vec{\nabla} + 2\mu \partial_r^2) \psi_1 + 2\mu \partial_r (\Lambda \psi_3/r)]_{r=R} = 0 , \quad (3.4)$$

$$\begin{aligned} & [\partial_\theta [2\partial_r(\psi_1/r) + 2\partial_r(1/r)\partial_r(r\psi_3) - 2\nabla^2\psi_3] \\ & + r\partial_\phi \partial_r(\psi_2/r)/\sin\theta]_{r=R} = 0 , \quad (3.5) \end{aligned}$$

and

$$\begin{aligned} & [\partial_\phi [2\partial_r(\psi_1/r) + 2\partial_r(1/r)\partial_r(r\psi_3) - 2\nabla^2\psi_3]/\sin\theta \\ & - r\partial_\theta \partial_r(\psi_2/r)]_{r=R} = 0 , \quad (3.6) \end{aligned}$$

where the angular momentum operator  $\Lambda$  in Eq. (3.4) is defined as

$$\Lambda = - (1/\sin\theta) \partial_\theta (\sin\theta \partial_\theta) - \partial_\phi^2 / \sin^2\theta . \quad (3.7)$$

Now let us specify the explicit form of the potential functions from the boundary conditions Eqs. (3.4)–(3.6). According to Eqs. (3.1)–(3.3), the potential functions may satisfy the wave equations

$$\ddot{\psi}_j - c_j^2 \nabla^2 \psi_j = 0 \quad (j = 1, 2, 3) . \quad (3.8)$$

The velocities  $c_j$  are expressed in terms of the Lamé coefficients  $\lambda$  and  $\mu$  and the density  $\rho_P$  of the solid as

$$c_j^2 = [(\lambda + \mu)\delta_{j,1} + \mu]/\rho_P , \quad (3.9)$$

where  $\delta_{j,1}$  are Kronecker's  $\delta$ . The general solutions of Eq. (3.8) are expanded by the associated Legendre polynomial  $P_l^{|m|}(\cos\theta)$  and the spherical Bessel function  $j_l(x)$  as

$$\begin{aligned} \psi_j(\vec{r}, t) = \sum_{l,m,\omega_{k_j}} A_j^{lm} j_l(k_j r) P_l^{|m|}(\cos\theta) \\ \times \exp[-i(m\phi + \omega_{k_j} t)] \quad (3.10) \\ (|m| \leq l, \quad j = 1, 2, 3) . \end{aligned}$$

Here the suffix  $j$  in Eq. (3.10) means the longitudinal ( $j=1$ ) and the transverse ( $j=2, 3$ ) modes, respectively.  $\{A_j^{lm}\}$  are the expansion coefficients. The wave number  $k_j$  is defined by the relation  $\omega_{k_j} = k_j c_j$ . Substituting Eq. (3.10) into Eqs. (3.4)–(3.6) we have the boundary conditions as follows:

$$\begin{aligned} k_1^2 [-(2\mu + \lambda) + 2\mu l(l-1)/\xi^2 + 4\mu T_1^+/\xi] \psi_1 \\ - 2\mu l(l+1) k_3^2 [(1-l)/\eta^2 + T_1^+/\eta] \psi_3 = 0 , \quad (3.11) \end{aligned}$$

$$\begin{aligned} (2k_1^2/\xi) [(l-1)/\xi - T_1^+] \partial_\theta \psi_1 \\ + k_3^2 [2(l^2-1)/\eta^2 - 1 + 2T_1^+/\eta] \partial_\theta \psi_3 \\ + \partial_\phi (\partial_r - 1/r) \psi_2 / \sin\theta = 0 , \quad (3.12) \end{aligned}$$

and

$$[2k_1^2/(\xi \sin\theta)][(l-1)/\xi - T_l^+] \partial_\phi \psi_1 \\ + (k_3^2/\sin\theta)[2(l^2-1)/\eta^2 - 1 + 2T_l^+/\eta] \partial_\phi \psi_3 \\ + \partial_\theta(1/r - \partial_r) \psi_2 = 0 \quad , \quad (3.13)$$

where  $\xi = k_1R$ ,  $\eta = k_2R = k_3R$ .  $T_l^+$  is the raising operator of the spherical Bessel function defined as  $T_l^+ j_l = j_{l+1}$ . Equations (3.12) and (3.13) bear two independent equations, one of which gives the relation between the potentials  $\psi_1$  and  $\psi_3$ , the other is about  $\psi_2$ . The independent condition for the potential  $\psi_2$

becomes

$$(\partial_r \psi_2 - \psi_2/r)_{r=R} = 0 \quad . \quad (3.14)$$

The modes obtained from the above condition (3.14) have no radial component of oscillation as understood from the definition of displacement  $\bar{u}(\bar{r}, t) = (0, \partial\psi_2/\sin\theta\partial\phi, -\partial\psi_2/\partial\theta)$ . These modes are called the toroidal waves<sup>15</sup> by seismologists. Then these waves do not contribute to the heat transfer between liquid helium and a small particle. The modes determined from the other condition on potentials  $\psi_1$  and  $\psi_3$  and Eq. (3.11) are called the spheroidal waves. These modes have the radial component of surface oscillation and contribute to the heat transfer. By using the relation  $(k_3/k_1) = (\eta/\xi)$  and Eq. (3.10) we have the two equations

$$(\xi/\eta)^2[-(2\mu + \lambda) + 2\mu l(l-1)/\xi^2 + 4(\mu/\xi)T_l^+] A_1^{lm} j_l(\xi) - 2\mu l(l+1)[(1-l)/\eta^2 + T_l^+/\eta] A_3^{lm} j_l(\eta) = 0 \quad (3.15)$$

and

$$2(\xi/\eta^2)[(l-1)/\xi - T_l^+] A_1^{lm} j_l(\xi) + [2(l^2-1)/\eta^2 - 1 + 2T_l^+/\eta] A_3^{lm} j_l(\eta) = 0 \quad . \quad (3.16)$$

By coupling the conditions (3.15) and (3.16), the eigenvalue equation is obtained as for  $l \geq 0$

$$2j_{l+1}(\xi) \frac{\xi}{\eta^2} \left[ 1 + \frac{(l-1)(l+2)}{\eta} \left( \frac{j_{l+1}(\eta)}{j_l(\eta)} - \frac{l+1}{\eta} \right) \right] \\ + j_l(\xi) \left[ -\frac{1}{2} + \frac{(l-1)(2l+1)}{\eta^2} + \frac{1}{\eta} \left[ 1 - \frac{2l(l-1)(l+2)}{\eta^2} \right] \frac{j_{l+1}(\eta)}{j_l(\eta)} \right] = 0 \quad . \quad (3.17)$$

The above equation will be solved numerically with respect to the single variable  $\eta$  by the use of the relation  $\eta^2/\xi^2 = k_3^2/k_1^2 = (2\mu + \lambda)/\mu$ . It should be noted that the eigenvalues  $\eta$  obtained from Eq. (3.17) depend only on the integer  $l$ .

#### IV. QUANTIZATION OF THE SPHEROIDAL WAVES: PHONONS IN A SPHERICAL ELASTIC BODY

The displacement vector  $\bar{u}(\bar{r}, t)$  in a spherical body is expanded by the sum of the toroidal modes and the spheroidal modes as mentioned in Sec. III. Since the toroidal modes do not contribute to the heat transfer, we need only the quantized displacement of the spheroidal modes for describing the heat transfer. The quantization of the spheroidal modes is the subject of this section. The displacement field due to the spheroidal modes is expressed by the sum of the first and third term of Eq. (3.2). The potential

functions  $\psi_1(\bar{r}, t)$  and  $\psi_3(\bar{r}, t)$  take the general form

$$\psi_1(\bar{r}, t) = \sum_{l,m,\omega_{k_1}^l} A_1^{lm} j_l(k_1^l r) P_l^{|m|}(\cos\theta) \\ \times e^{-im\phi} \exp(-i\omega_{k_1}^l t) \quad , \quad (4.1)$$

$$\psi_3(\bar{r}, t) = \sum_{l,m,\omega_{k_3}^l} A_3^{lm} j_l(k_3^l r) P_l^{|m|}(\cos\theta) \\ \times e^{-im\phi} \exp(-i\omega_{k_3}^l t) \quad . \quad (4.2)$$

Here  $\omega_{k_j}^l$  means the eigenfrequency of the spheroidal mode specified by a set of integers  $(l, m)$ . We allow the potentials to be complex valued. From Eq. (3.16), we find the ratio of the amplitude  $\alpha(l) = A_1^{lm}/A_3^{lm}$  to be

$$\alpha(l) = \frac{2(l-1)j_l(\xi) - 2\xi j_{l+1}(\xi)}{[\eta^2 - 2(l^2-1)]j_l(\eta) - 2\eta j_{l+1}(\eta)} \quad . \quad (4.3)$$

By using the above relation, the displacement vector due to the spheroidal modes is expressed by

$$\bar{u}_S^{l,m}(\bar{r}, t) = A \{^l m \{ \bar{\nabla} [j_l(k_1^l r) P_l^{|m|}(\cos\theta) e^{-im\phi}] + \alpha(l) \bar{\nabla} \times \bar{\nabla} [(r, 0, 0) j_l(k_3^l r) P_l^{|m|}(\cos\theta) e^{-im\phi}] \} e^{-i\omega_S^l t} \}, \quad (4.4)$$

where the lower suffix of the displacement  $S$  means the spheroidal mode. The wave numbers  $k_1^l$  and  $k_3^l$  are related through the dispersion relation  $\omega_S^l = c_1 k_1^l = c_3 k_3^l$ . It is clear that the spheroidal modes expressed by Eq. (4.4) are orthogonal to each other. The normalization condition is expressed by the following integral:

$$\int_0^R \int_0^{2\pi} \int_0^\pi [\bar{u}_S^{l,m}(\bar{r}, t)]^* \cdot [\bar{u}_S^{l,m}(\bar{r}, t)] r^2 dr d\Omega = 1. \quad (4.5)$$

Substituting Eq. (4.4) into Eq. (4.5), we have

$$|A \{^l m \}^{-2} = \int_0^R \int_0^{2\pi} \int_0^\pi \{ |\bar{\nabla} [j_l(k_1^l r) P_l^{|m|}(\cos\theta) e^{-im\phi}]|^2 + \alpha^2(l) |\bar{\nabla} \times \bar{\nabla} [(r, 0, 0) j_l(k_3^l r) P_l^{|m|}(\cos\theta) e^{-im\phi}]|^2 \} r^2 dr d\Omega. \quad (4.6)$$

The first term on the right-hand side of Eq. (4.6) becomes the simple relation using the integral formula of the associated Legendre polynomial

$$2\pi I(l, m) J_1(l) / k_1^l, \quad (4.7)$$

where  $I(l, m)$  is defined as

$$I(l, m) = \frac{2(l + |m|)!}{(l - |m|)! (2l + 1)}, \quad (4.8)$$

and

$$J_1(l) = \int_0^{k_1^l R} [\partial_\xi j_l(\xi)]^2 \xi^2 d\xi + l(l+1) \int_0^{k_1^l R} j_l^2(\xi) d\xi. \quad (4.9)$$

From the similar procedure the integral of the second term of Eq. (4.6) can be written

$$2\pi \alpha^2(l) I(l, m) J_3(l) l(l+1) / k_3^l, \quad (4.10)$$

where

$$J_3(l) = \int_0^{k_3^l R} l(l+1) j_l^2(\xi) d\xi + \int_0^{k_3^l R} [(\partial_\xi + 1/\xi) j_l(\xi)]^2 \xi^2 d\xi. \quad (4.11)$$

From Eqs. (4.7) and (4.10), Eq. (4.6) becomes

$$A \{^l m \} = [J_1(l) / k_1^l + \alpha^2(l) I(l, m) J_3(l) / k_3^l]^{-1/2} \times [2I(l, m) \pi]^{-1/2}. \quad (4.12)$$

Thus we have obtained the normalized spheroidal mode  $\bar{u}_S^{l,m}$  of Eq. (4.4). The quantized displacement-field  $\bar{u}_{op,S}(\bar{r}, t)$  can be expanded in terms of  $\bar{u}_S^{l,m}(\bar{r}, t)$  as follows by taking its being real into account

$$\bar{u}_{op,S}(\bar{r}, t) = \sum_{l,m,\omega_S^l} \left[ \frac{\hbar}{2\rho_p \omega_S^l V} \right]^{1/2} \times [a_{l,m,\omega_S^l} \bar{u}_S^{l,m}(\bar{r}) e^{-i\omega_S^l t} + \text{H.c.}] \quad (4.13)$$

It is clear now that the quantum number  $J$  used in Eq. (2.6) is specified by a set of  $(l, m, \omega_S^l)$ . In Eq. (4.13)  $a_{l,m,\omega_S^l}$  and  $a_{l,m,\omega_S^l}^\dagger$  are the annihilation and creation operator of  $(l, m, \omega_S^l)$ -mode phonons satisfying the commutation relations for boson operators.

## V. THERMAL BOUNDARY RESISTANCE BETWEEN LIQUID HELIUM AND A SMALL PARTICLE

We have given the general formula for the heat conductance  $h_K$  for an arbitrary size of a particle by Eqs. (2.7) and (2.10). The eigenvalue equation of the spheroidal modes which contribute to the heat transfer are obtained by Eq. (3.17). In this section, we give a numerical estimation of the thermal boundary resistance  $R_K$  due to phonon conduction between liquid helium and copper or silver particles. As seen from Eq. (2.7), we need the thermal average of the radial component of the velocity at the surface. The radial component of the quantized displacement of the spheroidal waves is expressed by using Eqs. (4.13) and (4.4) as

$$u_{op,S,r}(\bar{r}, t) = \sum_{l,m,\omega_S^l} \left[ \frac{\hbar}{2\rho_p \omega_S^l V} \right]^{1/2} \times [a_{l,m,\omega_S^l} u_{S,r}^{l,m}(\bar{r}) e^{-i\omega_S^l t} + \text{H.c.}] \quad (5.1)$$

where

$$u_{S,r}^{l,m}(\bar{r}) = A \{^l m \} [\partial_r j_l(k_1^l r) + \alpha(l) l(l+1) j_l(k_3^l r) / r] \times P_l^{|m|}(\cos\theta) e^{-im\phi}. \quad (5.2)$$

Taking the time derivative of Eq. (5.1) and putting  $r = R$ , we have for Eq. (2.7)

$$\int_0^\pi \int_0^{2\pi} T_r [\exp(-\beta H) \dot{u}_{\text{op},s,r}^J(R)^\dagger \dot{u}_{\text{op},s,r}^J(R)] d\Omega = \frac{\hbar(\omega_s^l)^2}{2\rho_s v_s(l)} \left[ |\partial_r j_l(k_3^l r)|_{r=R}^2 + \alpha^2(l) l^2 (l+1)^2 \frac{j_l^2(k_3^l R)}{R^2} \right] 2n(\omega_s^l, T) \quad (5.3)$$

where  $n(\omega_s^l, T)$  is the Bose-Einstein distribution function. We used in Eq. (5.3) the following definition from Eq. (4.12)

$$2\pi I(l, m) |A_l^m|^2 = \omega_s^l v_s(l) \quad (5.4)$$

Here  $v_s(l)$  has the dimension of the velocity, which is defined as

$$v_s(l) = c_1 J_1(l) + c_3 \alpha^2(l) l(l+1) J_3(l) \quad (5.5)$$

The heat conductance  $h_K$  is obtained by differentiating Eq. (5.3) with respect to temperature  $T$  as defined by Eq. (2.10). Thus the explicit form for the heat conductance becomes

$$h_K = \frac{\hbar \rho_L c_L}{4\pi \rho_p} \sum_{l, \omega_s^l} \frac{(2l+1)(\omega_s^l)^4 R^2}{v_s(l) [c_L^2 + (\omega_s^l R)^2]} \{ |\partial_r j_l(k_3^l r)|_{r=R}^2 + \alpha^2(l) l^2 (l+1)^2 j_l^2(k_3^l R)/R^2 \} \partial_T n(\omega_s^l, T) \quad (5.6)$$

The prefactor  $(2l+1)$  in Eq. (5.6) means the degeneracy. The integer  $m$  is eliminated through the expression (5.4).

The eigenvalue equation (3.17) for the spheroidal modes has been solved numerically on the variable  $\eta = k_3 R$ . We have used the following set of parameters  $c_1 = 5.01 \times 10^5 \text{ cm sec}^{-1}$  and  $c_3 = 2.27 \times 10^5 \text{ cm sec}^{-1}$  for copper particles, and  $c_1 = 3.65 \times 10^5 \text{ cm sec}^{-1}$  and  $c_3 = 1.66 \times 10^5 \text{ cm sec}^{-1}$  for silver particles, respectively. The mass densities are taken from the corresponding bulk values to be  $\rho_p = 8.96 \text{ g cm}^{-3}$  for copper and  $\rho_p = 10.5 \text{ g cm}^{-3}$  for silver particles. The eigenvalues up to  $l = 10$  for copper particle are given in Table I in Appendix A. The other eigenvalues used in the calculation need too much space to be included here and are available elsewhere.<sup>16</sup> It will be seen from Table I that the number of states per unit interval of  $\eta$  increases with approaching to the larger part of the eigenvalues. The reason of this tendency is clear from the fact that the wavelength corresponding to the large eigenvalue is small compared with the size of a particle. Then the number of states in the region of the large eigenvalues becomes denser. The eigenvalues for silver particle are not given here.

Figure 1 shows the calculated results of the thermal boundary resistances  $R_K$  between liquid  $^4\text{He}$  and copper particles with radii  $R = 500$  and  $5000 \text{ \AA}$  for the temperature range from 10 mK up to 1 K. We have taken the eigenvalues up to  $k_3 R = 100$  in order to calculate the full curves in Fig. 1. This figure shows that the resistances  $R_K$  is almost proportional to  $T^{-3}$  above 200 mK for the particle with radius  $R = 500 \text{ \AA}$ , and above 20 mK for  $R = 5000 \text{ \AA}$ . Below these temperatures, the resistance  $R_K$  begin to increase exponentially. This is because these temperatures perceive the presence of the lowest eigenfrequency. Figure 2 shows the thermal boundary resistances  $R_K$  between liquid  $^4\text{He}$  and silver particles with radii  $R = 500$  and  $5000 \text{ \AA}$ . The eigenvalues are also taken into account up to  $k_3 R = 100$ . Since Lamé

coefficients and the mass density of silver particles are smaller than those of copper particles, the smaller resistances are obtained compared with those of copper particles. The temperatures, above which the exponential behavior appears, are also found in the case of silver particles. These temperatures are about

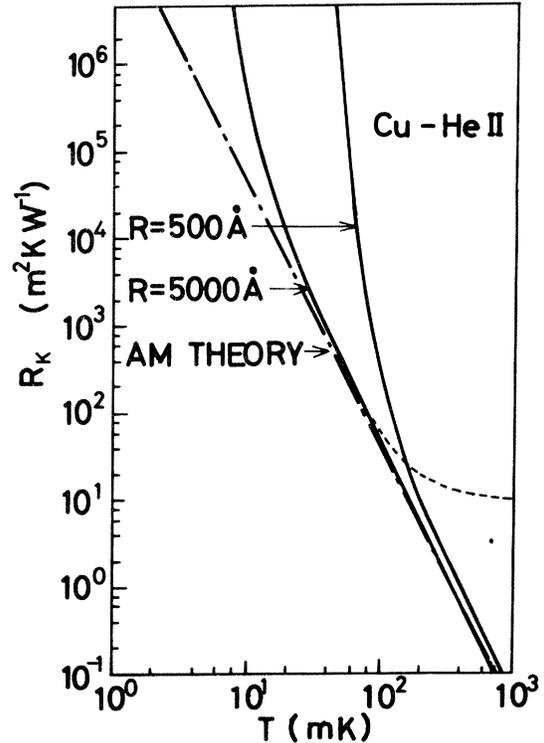


FIG. 1. Thermal boundary resistance between copper particles and liquid  $^4\text{He}$ . The radii of particles are taken to be 500 and  $5000 \text{ \AA}$ , respectively. The dotted curve is the thermal boundary resistance for a particle with radius  $5000 \text{ \AA}$  by taking only the eigenvalues up to  $k_3 R = 20$  into account. The full curves represent the results by taking the eigenvalues up to  $k_3 R = 100$ . The dot-dashed straight line indicates the result for a bulk copper (Ref. 2).

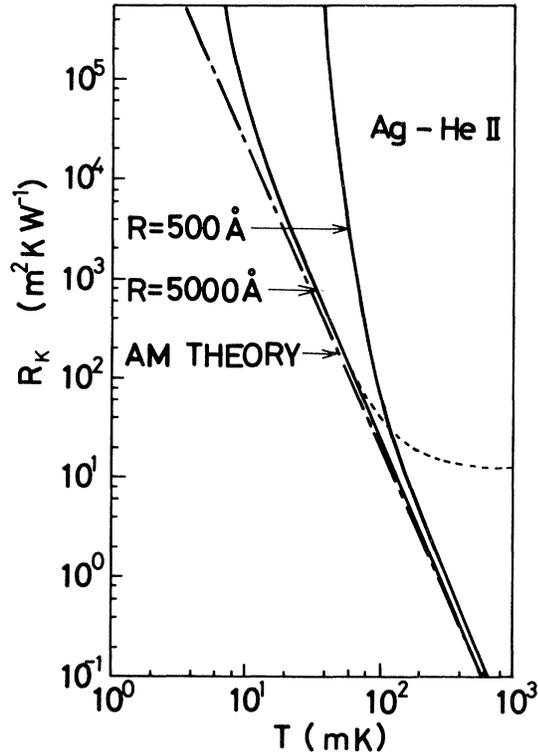


FIG. 2. Thermal boundary resistance between silver particles and liquid  ${}^4\text{He}$ . The radii of particles are taken to be 500 and 5000 Å, respectively. The dotted curve is the thermal boundary resistance for a particle with radius 5000 Å by taking the eigenvalues up to  $k_3R = 20$  into account. The full curves represent the results by taking the eigenvalues up to  $k_3R = 100$ . The dot-dashed line indicates the result for a bulk silver (Ref. 2).

100 mK for a silver particle with  $R = 500$  Å and about 10 mK for  $R = 5000$  Å. The other behavior is similar to the cases of copper particles. The magnitude of the thermal boundary resistances  $R_K$  for bulk copper and silver are given by the dot-dashed straight line in Figs. 1 and 2. These are from the acoustic mismatch theory,<sup>2</sup> which is given by

$$R_K(\text{bulk}) = \frac{15\rho_p(c_3)^3\hbar^3}{2\pi^2\rho_L c_L k_B^4 F_{LS}} T^{-3}. \quad (5.7)$$

The numerical estimation of the factor  $F_{LS}$  for various solids has been performed by Challis and Cheeke,<sup>17</sup> which gives  $F_{LS} = 1.53$  for both copper and silver. It should be noted that the magnitude of the calculated resistance of small particles becomes the same with that of a bulk solid in the high-temperature regime. This indicates that the present theory approaches to the acoustic mismatch limit<sup>2</sup> at large particle size. We have also added in Figs. 1 and 2 the thermal boundary resistances  $R_K$  only taking up

to the eigenvalue  $k_3R = 20$  by the dotted curve. It will be seen that these are inadequate to recover the high-temperature regime.

The exponential behavior in the low-temperature regime is easily understood by taking the few lowest eigenvalues into account. For instance, in the case of copper particle with radius  $R = 5000$  Å the lowest eigenfrequency becomes  $\omega_S^0 = 1.11 \times 10^{10} \text{ sec}^{-1}$  using  $k_3^0 R = 2.45$  from Table I. Taking this lowest eigenfrequency  $\omega_S^0$  into account the resistance  $R_K$  becomes

$$R_K = \frac{16\pi\rho_p v_S(0)k_B c_3^2}{\hbar^2 \rho_L c_L (\omega_S^0)^5} \frac{c_L^2 + (\omega_S^0 R)^2}{(\omega_S^0 R)^2} \times [j_1(k_3 R)]^{-2} \sinh^2\left(\frac{\hbar\omega_S^0}{2k_B T}\right) T^2. \quad (5.8)$$

We see the exponential behavior of the resistance  $R_K$  originates from the lowest eigenfrequency in the low-temperature regime.

## VI. SUMMARY AND DISCUSSIONS

We have theoretically studied the thermal boundary resistance between liquid helium and small particles. The formula (5.6) for the resistance  $R_K$  has been derived by considering a spherical elastic particle. It has been shown that only the spheroidal modes contribute to the heat transfer. The eigenvalues of the spheroidal modes have been obtained numerically from Eq. (3.17). The resistances  $R_K$  have been calculated by using these eigenvalues for silver and copper particles with radii  $R = 500$  and 5000 Å. The calculated resistances  $R_K$  in Figs. 1 and 2 show the  $T^{-3}$  variation above some temperature corresponding to the finite lowest eigenfrequencies of small particle. This temperature dependence is identical to the prediction of the acoustic mismatch theory<sup>2</sup> for a bulk solid. It should be emphasized that at the high-temperature regime the magnitude of the resistances  $R_K$  coincides with those of bulk solids. The physical meaning of these results is clear because the wavelength of the thermally excited phonons becomes small at high temperatures in comparison with the size of a small particle and the size effect is irrelevant in such a temperature region. In other words, the present theory is an alternative derivation of the acoustic mismatch theory<sup>2</sup> at large particle size. The resistance  $R_K$  increases exponentially with decreasing temperature. The exponential behavior of the resistance  $R_K$  arises from the finite lowest eigenfrequency which depends on the size and the elastic properties of a particle.

The shape of a particle has been assumed to be spherical. The exponential behavior for the temperature dependence would not be altered even if taking

TABLE I. The dimensionless eigenvalues  $\eta = k_3 R$  for a copper particle. The integer  $l$  of the column means the order of Legendre polynomial. The integer  $n$  of the row indicates the  $n$ th modes belonging to the  $l$ th-order oscillation.

$l \setminus n$	1	2	3	4	5	6	7	8	9	10
0	· · ·	2.45	5.96	6.25	9.21	12.41	13.57	15.58	18.74	20.61
1	· · ·	3.66	7.33	9.43	10.76	13.91	16.72	17.17	20.27	23.41
2	2.66	5.20	8.70	11.76	12.68	15.38	18.52	19.88	21.77	24.91
3	3.96	6.80	10.08	13.31	15.13	16.88	19.88	22.64	23.37	26.39
4	5.09	8.41	11.45	14.71	17.40	18.54	21.42	24.50	25.74	27.87
5	6.15	9.95	12.83	16.07	19.61	20.63	22.87	25.97	28.32	29.41
6	7.16	11.41	14.21	17.43	20.61	22.84	24.39	27.40	30.39	31.42
7	8.16	12.82	15.62	18.78	21.99	24.82	26.15	28.82	31.91	33.87
8	9.15	14.16	17.02	20.11	23.35	26.43	28.20	30.27	33.34	36.12
9	10.13	15.43	18.42	21.46	24.69	27.87	30.27	31.82	34.75	37.80
10	11.11	16.68	19.79	22.81	26.02	29.25	32.11	33.60	36.17	39.26

into account the difference in the shape of particles. This is due to the fact that the finite lowest eigenfrequency for any particles with finite size leads to exponential behavior on the temperature dependence. At high temperatures, the dominant phonons for the heat transfer have much shorter wavelength than the size of a particle, then the shape is irrelevant and the resistance  $R_K$  will approach to the acoustic mismatch limit. The  $T^{-3}$  dependence of the resistance  $R_K$  in the high-temperature regime has been observed experimentally between liquid  $^3\text{He}$  and metallic particles.<sup>7-9</sup> The observed resistances,<sup>7-9</sup> however, show the dip around about 10 mK. Metallic particles<sup>7-9</sup> have contained small amounts of various types of magnetic impurities. Mills and Béal-Monod<sup>10,11</sup> discussed in detail the magnetic contribution to the heat transfer in these systems.

Finally, we should note that metallic particles used in experiments<sup>7-9</sup> would be covered with the oxide layer or the  $\text{O}_2$ ,  $\text{H}_2\text{O}$ , and other adsorbed gases with about 100-Å thickness. The oxide layers may include the molecules with paramagnetic moments as suggested by Potter,<sup>18</sup> which will be possible to exchange heat through the magnetic coupling with liquid  $^3\text{He}$ . In addition, it is known that phonons in a metal cou-

ple with electrons and expected<sup>19</sup> that these effects may play some roles for the heat exchange. We hope to consider these problems in a future paper. In conclusion, although we have studied only the acoustic coupling, the present results will permit us to discuss some aspects of the thermal boundary resistance between small particles and liquid helium.

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#### APPENDIX: EIGENVALUES OF SPHEROIDAL MODES

In this Appendix we give the eigenvalues which have been obtained by solving Eq. (3.17) numerically. Table I shows the dimensionless eigenvalues  $\eta = k_3 R$  for a copper particle. We used the following set of parameters  $c_1 = 5.01 \times 10^5 \text{ cm sec}^{-1}$  and  $c_3 = 2.27 \times 10^5 \text{ cm sec}^{-1}$ . The eigenvalues  $k_3 R$  up to ( $l = 10, n = 10$ ) are included in Table I.

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