New Kapitza heat-transfer model for liquid ^{4}He

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(Received 4 December 1979)

The Kapitza resistance between a solid and liquid ^{4}He is explained by two parallel processes. One is the well-known Khalatnikov-acoustic-mismatch channel that has a frequency-independent transmission coefficient. The other we propose herewith is due to the difference in quantum effects between the bulk liquid and the compressed surface layers. The crucial point is the presence of an "optical branch," that is a dispersion curve with $\omega_0 \neq 0$ for $k \to 0$. We illustrate this type of coupling with a mechanical equivalent model. The excitations in the surface are stimulated by the transverse part of the sound waves in the solid. The coupling is frequency dependent with two regions of enhancement: one near zero frequency and another near ω_0. In the spin-lattice model of helium the quantum effect is described by a nonzero angle for the spin orientation, and we show that the deviation from the interlayer coupling parameters from the bulk value gives rise to a change in the spin orientation which leads to an optical branch in the dispersion.

I. INTRODUCTION

In 1941 Kapitza[1] observed a temperature jump between liquid ^{4}He and the solid with which it was in contact. He also observed that this thermal boundary resistance R_k increased upon lowering of the temperature. He found a relation $R_k \sim T^{-3}$ in the region between 1.6 K and T_λ. Subsequently, it was realized that the "natural" explanation, assuming that the energy was transported by phonons and that the resistance stems from the reflection of these phonons at the surface, led to a resistance much higher than was observed. The discrepancy between this theory, due to Khalatnikov,[2] and the experiment, has led to an enormous amount of experimental work and several theories. For the older work we refer to the review article of Pollack.[3] For a more recent review, see Snyder.[4]

In order to delineate our work, let us define the Kapitza resistance as the thermal boundary resistance between liquid helium and any given solid. Actually, it does not matter in our model whether the helium is liquid or a solid at relatively low pressures. Furthermore, we will exclude the ^{3}He wetting a surface of a magnetic material, since this leads to dipolar coupling mechanism and an extra conductance in the region below 0.1 K as was explained by Leggett and Vuorio[5] and refined by Maki *et al.*[6] If we assume that the magnetic conductance and the nonmagnetic conductance are additive, which may not be the case, then our ideas will refer to the nonmagnetic part only.

The Khalatnikov theory gives a result for R_k proportional to T^{-3}, but the proportionality constant is two orders of magnitude larger than the experimental value. Since heat flows more easily than the acoustical-mismatch theory indicates, it is reasonable to assume that there must be a second channel through which energy can be transmitted from the solid to the liquid or vice versa. The goal of this paper is to establish another mechanism which, we think, is strongly suggested by some recent experimental results.

Before we go into this we would like to mention a few modifications of the acoustic mismatch theory that have been developed. It is well known that due to the Van der Waals forces at the solid surface, a layer of helium atoms is formed whose properties are different from those of bulk helium. Taking this into consideration, the Khalatnikov theory has been extended by considering this layer as a classical continuum which produces intermediate acoustic matching.[7,8] Since the thickness of this layer is very small compared to the phonon wavelength the effect is very small.

Another, and very difficult, aspect of the Kapitza-resistance problem is its dependence upon the condition of the solid surface.[9] When sound waves travel from a solid, bounded by a perfect surface, to a liquid (a less dense medium), only those waves restricted to the critical cone are allowed to pass through the surface.[10] Accordingly, this implies that about 99% of the phonons have zero probability of transmission.[11] If the surface contains defects, as from machining or mechanical polishing, this critical cone is widened,

and thus more energy is allowed to pass by diffuse scattering. Another view is that the rough surface actually provides a larger surface area through which energy may flow. Incorporating these ideas into the classical scheme improves the heat transport, i.e., decreases the discrepancy between the Khalatnikov theory and experiment. Yet despite this, the largest resistance observed (which we assume, was also due to the roughest surface) is still less than that of the Khalatnikov theory.

In view of the failure of the acoustic-mismatch theory to explain the Kapitza resistance, Sheard *et al.*[12,13] have proposed a theory which takes into account the superfluid character of liquid ^{4}He. By the use of their transfer Hamiltonian, expressions have been derived for the energy transfer by one-phonon, one-roton, and second-order processes. They have found that in the long-wavelength limit, the expression of the energy transfer reduces to that of the acoustic-mismatch theory. The value of this expression is greater than that of the Khalatnikov theory by only a factor of 4. The roton contribution is even less; the ratio of energy transfer by rotons to that by phonons being 0.042 at 2 K. Neither of these two contributions bridge the gap.

Shortly after the publication of Sheard *et al.*,[12] three sets of new experimental results were published. The first set[14–18] involves internal reflection at the boundary using a crystal sample, one side of which is exposed to an evacuated chamber. As helium vapor is introduced into the chamber, several layers of ^{4}He atoms are deposited upon the surface of the sample. The purpose is to measure the reflection coefficient of the boundary and its dependence upon the amount of helium gas introduced into the chamber. Three interesting results emerge from these experiments: (i) A strong echo loss is the result of the adsorption of three atomic layers. At this thickness the loss is almost the same as the loss when bulk liquid is present. (ii) The transverse echo shows no structure when the roton minimum was passed. (iii) The echoes most influenced by the adsorbed layers are the echoes due to transverse waves.

The second set of experiments[11,19–21] studies the angular dependence of phonon transmission through the boundary. The typical angular distribution of the radiated phonons reveals a pronounced narrow peak centered at $\alpha = 0°$, corresponding to the channel for transmission which conserves the wave vector which is parallel to the surface. That is, at $\alpha \simeq 0°$, transmission is governed by classical acoustics. But there is also a channel which does not conserve the parallel wave vector and gives rise to a broad background. After integration over 2π sr, it is revealed that this background gives a greater contribution than does the central peak. Particularly noteworthy is the evidence which shows that it is easier for phonons to travel from the crystal to the helium than vice versa (as judged by comparison of the heights of the two background curves).

The third set of experiments,[22] which is similar to the first, involves measuring reflections of phonons at various interfaces when the liquid ^{4}He is replaced by the solid ^{4}He, solid H_2, solid D_2, and solid Ne. These different materials may be characterized by the de Boer quantum parameter which is a convenient measure of the quantum effects in the system. This parameter ranges from 2.68 for liquid ^{4}He to 0.59 for solid Ne; the larger the parameter, the greater the quantum effects. There is a large discrepancy between theory and experiment, particularly for transverse waves, when liquid ^{4}He, solid ^{4}He, solid H_2, and solid D_2 are used; the largest discrepancy being with the liquid-^{4}He data. The data involving solid Ne reveals little discrepancy, indicating that solid Ne behaves much like a classical system. Hence classical theories adequately describe the heat transmission for this case.

This experiment suggests that the anomalously small Kapitza resistances are characteristics of systems in which quantum effects are important. The similarity of the results for liquid and solid ^{4}He, solid H_2, solid D_2, indicates that the mechanism responsible for this anomalous transfer of energy is perhaps the same for all these systems.

In Sec. II we will describe briefly the results obtained by the acoustic theory. In Sec. III we give the "mechanical equivalent" model of a system that shows a second channel of energy transfer. In Sec. IV we describe the quantum-mechanical model, based on a lattice model of the liquid. Section VI sums up our conclusions.

II. IMPORTANCE OF THE OPTICAL MODE

Since more energy flows from the solid to the liquid than can be explained by either the classical or the quantum-mechanical acoustic-mismatch theory, we expect the existence of a second channel, parallel to the acoustic channel. As indicated in the introduction, there exists strong evidence that a two-dimensional array of helium atoms deposited on the surface of the solid plays a key role in the enhancement of heat transfer. It was also observed that the transmission probability is dependent upon the frequency of the phonons,[11] whereas according to the acoustic-mismatch theory, the transmission probability should be independent of frequency. We suggest that a near resonance process is of importance in this problem. When the surface layer is considered as a classical continuum, the dispersion curve of the phonons in the bulk solid does not intersect that of the phonons in the layer, except at infinitely long wavelengths and zero frequency, see Fig. 1. It is clear that these phonons carry very little energy. But

if there were an optic-like branch present in the spectrum, the situation would be quite different. Then the intersection would take place at $\omega \neq 0$, and the solid phonons would couple with energetic excitations in the surface layer. The mechanical model described in Sec. III is an attempt to simulate the surface structure which produces an optic-like branch in the dispersion relations.

The problem of the energy flux perpendicular to the surface is treated very nicely by Sommerfeld[23] and by Khalatnikov.[2] Note that the conventions used for the amplitudes are different in these two publications: the amplitudes A, B, and C in the Sommerfeld book have to be multiplied by ik in order to obtain the amplitudes A, B, and C in the Khalatnikov book.

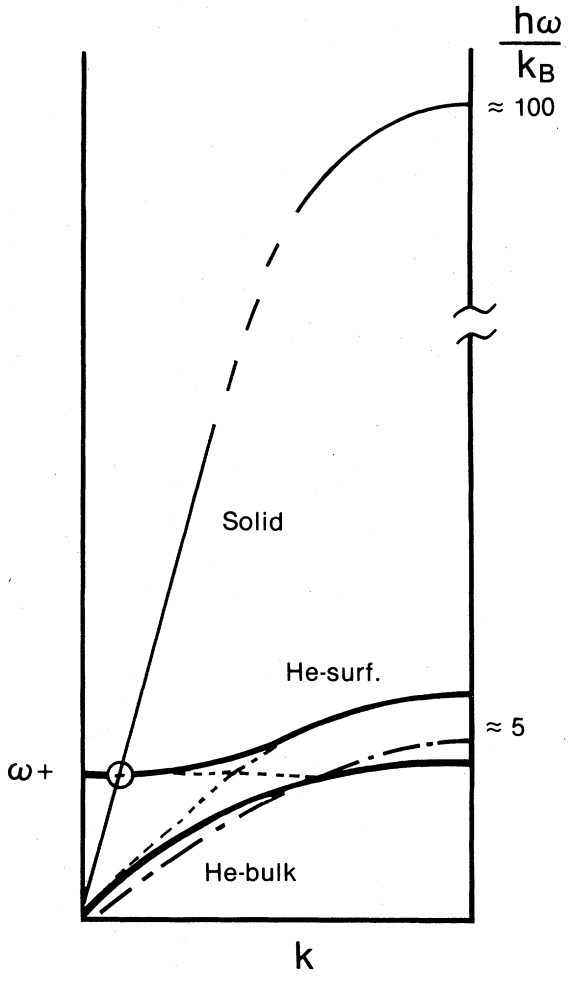

FIG. 1. Dispersion curves of phonons in: the solid, the bulk liquid (— · — ·), the surface layer helium branches (——) resulting from the ordinary dispersion curve (liquid helium at the equivalent higher pressure) and the excitation branch. Finely dotted line is the crossover in the absence of coupling.

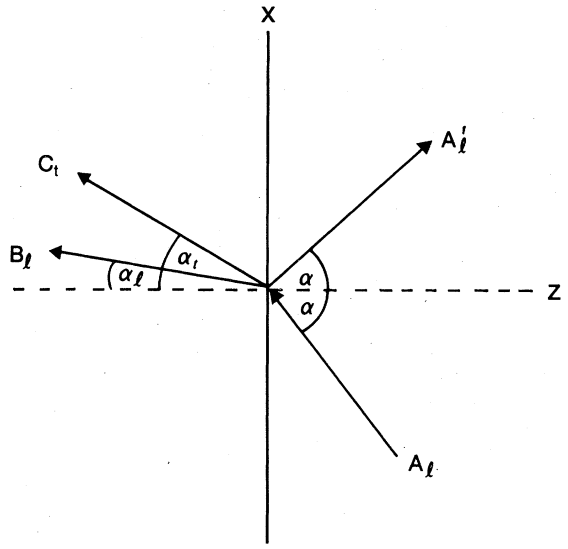

FIG. 2. Reflection and transmission of longitudinal wave from the liquid towards the interface with the solid.

This is because Khalatnikov describes potentials and Sommerfeld describes displacements. Here k is the wave number. We will use the Khalatnikov convention.

If a longitudinal wave with amplitude A_l from the liquid strikes the solid (Fig. 2), it will create a longitudinal wave in the liquid (A_l'), a longitudinal wave in the solid (B_l), and a transverse wave in the solid (C_t). At the boundary, two conditions must be valid: the continuity of normal displacement and the continuity of stress. This leads to two conditions on the amplitude and the fact that the spatial phases must be equal at the boundary (Snell's law) which gives the relation between the direction angles α, α_l, and α_t. Solving the amplitude relations leads to the relative values of A_l', B_l, and C_t.

The energy flow perpendicular to the boundary must be conserved. By using the above mentioned amplitudes, it is easy to verify that

$$\left|\frac{A_l'}{A_l}\right|^2 + \frac{D}{\rho}\frac{c_l \cos\alpha_l}{c\cos\alpha}\left|\frac{B_l}{A_l}\right|^2 + \frac{D}{\rho}\frac{c_t \cos\alpha_t}{c\cos\alpha}\left|\frac{C_t}{A_l}\right|^2 = 1 \quad . \quad (1)$$

where c, c_l, and c_t are, respectively, the velocity of sound in the liquid, the longitudinal velocity in the solid, and the transverse velocity in the solid. Here ρ is the density of the liquid, D the density of the solid. Similar expressions can be obtained for a longitudinal wave coming from the solid[24,25] and a transverse wave coming from the solid.[23,24] These equations simply confirm the energy balance and could have been written down without obtaining the solution.

III. MODEL OF A DISCRETE STRUCTURE COUPLED TO A CONTINUUM SOLID

At very low temperatures, the wavelength of sound is large compared to the lattice spacing in the solid. Therefore, the discrete structure of the solid does not play a major role in the following problem. Our model consists of a continuum solid, covered by a layer of atoms (surface helium). These atoms have two states, represented by two sets of masses, and they are in turn coupled to the bulk helium by a damping constant. This damping constant is associated with one set of masses, namely that which represents the following situation: energy is carried by transverse waves, incident from the solid, which impinge upon the surface. If the frequency of the incoming waves is nearly equal to one of the natural frequencies of the surface layer, then the energy is absorbed resonantly by the layer. Energy "leaks" into the liquid via the dampers attached to one set of masses. We propose that some of the surface atoms must be excited to higher-energy states before giving up the energy to the liquid.

This model is shown in Fig. 3 where the surface atoms are represented by spheres of different masses M and m; and the coupling consists of a bending force constant γ, representing flexible rods, a longitudinal spring constant β_s between the masses and a damping constant κ.

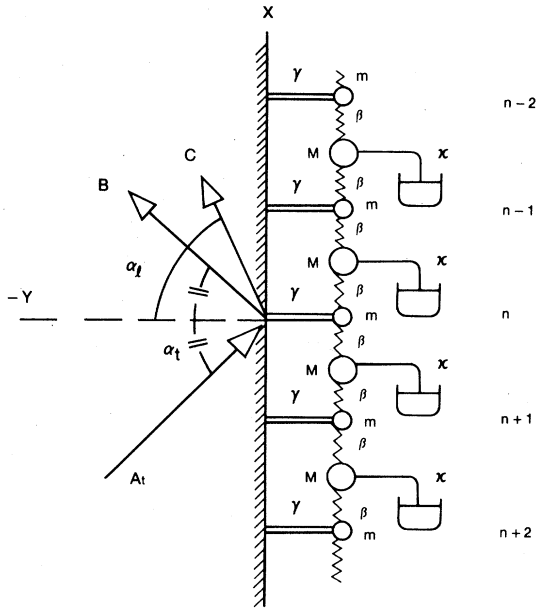

FIG. 3. Discrete surface structure that contains optical branch attached to continuum. The parameters γ, β_s, and κ are the bending, spring, and damping constants; m and M are different masses. Incident transverse wave with amplitude A_t, creates reflected transverse wave B and reflected longitudinal wave C. The dampers represent the coupling of the surface layer to the bulk liquid.

The transmission coefficient **T** for small γ is given in Eq. (22) of the Appendix. This coefficient is a function of ω and has the following characteristics: it is proportional to the damping in the liquid and it shows broadened resonances one near $\omega \approx 0$ (for small k) and one near $\omega \approx \omega_0$, the "optical" frequency. Both resonances are broadened by the damping: $\Delta\omega = \kappa/2m$.

The heat flux due to the phonons is given by

$$W_H = \frac{\hbar}{(2\pi c_t)^2} \int_0^\infty d\omega \int_0^1 dx\, n_\omega \mathbf{T} \omega^3 x \ ,$$

where $x = \cos\alpha_t$ and n_ω the Bose-Einstein function for the phonons. The integral over x gives a factor 0.1 and the integral over ω is approximated by replacing $g(\omega)$ as in Fig. 4(a) by simple rectangles: see Fig. 4(b); the height for $\omega \approx \omega_0$ is $2/\kappa^2\omega_0^3$.

Thus we have

$$W_H = \frac{0.1\hbar\gamma^2}{\mu K_t (\pi c_t)^2 m} n_{\omega_0}(T) \ .$$

Although the temperature dependence in this equation arises soley from the Planck function at $\omega \simeq \omega_0$, there may be also a temperature dependence hidden in the frequency ω_0. Note that the result is independent of κ, provided $\kappa \neq 0$, as it should be.

We find that this process leads to a heat conductance given by

$$\sigma_K = \frac{1}{A}\left(\frac{\partial W_H}{\partial T}\right) \simeq \left(\frac{dn}{dT}\right)_{\omega_0} \ ,$$

where A is the area. This quantity is constant for

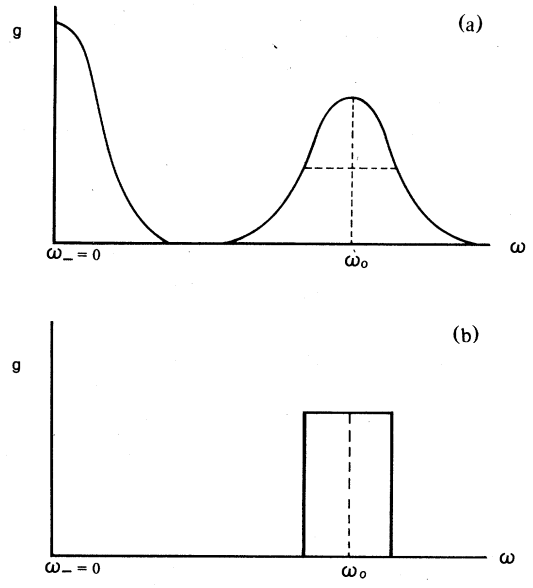

FIG. 4. Transmission function $g(\omega)$; (a) complete form and (b) approximation used.

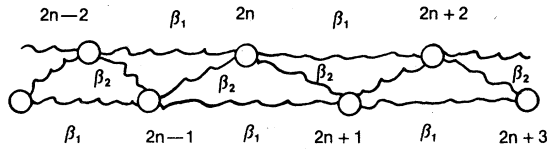

FIG. 5. Equal-mass structure that gives rise to optical branch in dispersion curve.

$kT > \hbar\omega_0$ and goes to zero for $kT < \hbar\omega_0$.

We point out at this moment, for later use, that the two-mass model is a special case of a two-sublattice system. Similar results, i.e., two branches, are obtained by two-spring models. In particular, we would like to mention a model that comes close to the picture we will propose for helium. This is the model in which one set of atoms, say the even sites, are coupled to another set, say the odd sites, by spring constants β_2. At the same time both the even and the odd atoms are coupled amongst themselves by spring constants β_1. The arrangement is shown in Fig. 5. It is easy to show that this gives an excitation spectrum with frequencies

$$m\omega_{1,2}^2 = 4\beta_1^2 \sin^2(ka) + 2\beta_2 \pm \beta_2[4\sin^2(\tfrac{1}{2}ka) - 2] \ .$$

For small k this leads to $m\omega_2^2 = O(k^2)$ and $m\omega_1^2 = 4\beta_2 + O(k^2)$, the optical branch. The eigenvectors are given by the sum and difference of the basis vectors, very similar to the case of almost equal masses. If $\beta_2 = \beta_1$, the optical branch disappears, since the periodicity of the system is now half the original periodicity.[26]

We use this model to illustrate that a longitudinal wave along the surface will have the usual phonon behavior if all interaction constants are the same, but that an optical branch will appear as soon as the interaction constants are the same, but that an optical branch will appear as soon as the interaction constants between the layers are different. This we know is the case in the first few layers of helium. These layers are so strongly adhered to the solid that the atoms are more closely packed than in the liquid. This means that both the force constants as well as the kinetic-energy factors are modified. The kinetic energy is modified since, by virtue of the uncertainty relations, the velocity is modified when the interatomic distance is changed.

IV. QUANTUM-MECHANICAL MODEL

It is of course inadequate to explain the Kapitza resistance by a classical model since experimental evidence[22] clearly indicates that the discrepancy between the acoustical model and the observation is proportional to the de Boer parameter. The previous considerations were to emphasize the need for an optical mode.

In order to introduce the quantum effects we express the property that the helium atoms are not localized due to the uncertainty principle. They jump from site to site even in the ground state at $T = 0$. Assuming that the liquid, as well as the surface layer, can be adequately described by a lattice model, this zero-point motion of the atoms is described by a term that annihilates the particle at a given site and recreates it at a neighboring site.[27] Proper evaluation of the operator wave function leads, in this way, to a kinetic-energy term with a factor $K = \hbar/md^2$, where d is the size of the lattice periodicity. We take this distance about equal to the interatomic distance of the atoms in the liquid. The Hamiltonian contains a potential energy term, representing the attractive interaction (the well of the Lennard-Jones potential) between the atoms. The repulsive interaction is taken into account by requiring that not more than one atom can stay (instantaneously) at the same lattice site. In order to be able to use the grand canonical ensemble, we add a chemical potential term. This Hamiltonian is equivalent to a spin Hamiltonian representing an anisotropic Heisenberg coupling in an external field. If the coupling is stronger in the xy direction than in the z direction, the latter being the direction of the external field, then the excitation spectrum is linear in k (phonon-like). This result is found via a rotation-transformation in spin space.[28]

Despite its schematic nature, we adopt this model for liquid helium, for the bulk or for one layer at a time. This is justified through the fact that the model expresses, from the uncertainty principle, the nonlocalized nature of the atoms by means of a tilted spin. If the spin were exactly along the z direction, the particles are either absent or present, but the kinetic effect (the xy coupling) leads to the necessity for a set of rotated axes to obtain the ground state.

We further hypothesize that the increased density of the first two layers of helium on the interface of the solid leads to a decrease of this quantum effect, that is, the coupling *between* the layers is different from the coupling *in* the layers. We assume the latter is about the same as in the bulk liquid. In this manner we arrive at a picture similar to a two sublattice model: two systems each with internal coupling constants (for computational reasons we take these coupling constants to be the same inside each subsystem) coupled to each other by constants that are different. If we follow the scheme of Matsubara and Matsuda,[27] eliminating the terms linear in the operators, we obtain one condition for two unknowns. It was pointed out by Matsuda and Tsuneto[29] that the variation principle applied to the ground state leads to two conditions. This means we can in principle determine the two angles of quantization θ and ϕ. It

can easily be seen, upon elimination of the field terms, that in the case where the intercoupling parameters are different from the intracoupling parameters, the two angles of quantization are *unequal*. Note that, as is the case in similar eigenproblems, it is not correct to associate an angle with a subsystem. This inequality of angles leads, after considerable mathematical labor, to a second branch in the excitation sprectrum: the optical branch. The mathematics is only partly displayed here since the results are identical to the calculations of Liu and Fisher[30] for two subsystems in a three-dimensional lattice. Since the approximations are of molecular-field nature, the dimensionality plays only a trivial role. We repeat the crucial point: due to the fact that the interlayer coupling is different from the intralayer coupling, the system has two different "angles of quantization" and consequently has an optical branch in its excitation spectrum. These "optical" excitations are coupled to the bulk phonons by means of a secondary energy exchange. If this picture is correct, the two surface layers should have their own temperature T_s ($T_{\text{solid}} < T_s < T_{\text{liquid}}$). The temperature dependence remains T^3 at least as long at $T > \hbar\omega_0/k_B$ because the number of bulk phonons is proportional to T^3. It is also trivial that there should be no "cone" associated with this form of energy transport.

We would like to report a very similar development by Nakayama.[31] His theory is based on the same observations as we started out with in Sec. I: the importance of the first few layers, the preference for quantum systems, the absence of Snell's law and the effectiveness of transverse phonons. However, his proposed mechanism, the tunneling of helium atoms, is quite different from what we propose.

Although we also assume that the helium atoms are nonlocalized it is essential in our model that we deal with two different coupling constants and it does not matter whether the hopping terms are different, which it is somewhat similar to tunneling, or whether the interaction term is different.

We also mention the work of Namaizawa,[32] who introduced surfon waves as intermediary between the phonons in the solid and the phonons in the liquid. The quantum properties of helium come in through the structure factor, no exchange of atoms between the surface layer and the bulk atoms takes place. Similar work was done by Gel'fgat and Syrkin.[33]

V. HAMILTONIAN OF THE MODEL

Let us assign the a sites to the helium atoms adhering to the surface and the b sites to the helium atoms that are part of the bulk liquid. Furthermore, a_i and $a_i^\dagger$ are the annihilation and creation operators on the a sites, b_i and $b_i^\dagger$ the operators on the b sites. We introduce the Matsubara-Matsuda Hamiltonian with

$$\tilde{H} = \tilde{H}_{aa} + \tilde{H}_{bb} + \tilde{H}_{ab} \ , \tag{2}$$

where the separate parts are given by

$$\tilde{H}_{aa} = \frac{\hbar^2}{md^2} \sum_{\langle ij \rangle} (a_i^\dagger - a_j^\dagger)(a_i - a_j) - v_0 \sum_{\langle ij \rangle} a_i^\dagger a_i a_j^\dagger a_j + \mu \sum_i a_i^\dagger a_i \ ,$$

$$\tilde{H}_{bb} = \frac{\hbar^2}{md^2} \sum_{\langle ij \rangle} (b_i^\dagger - b_j^\dagger)(b_i - b_j) - v_0 \sum_{\langle ij \rangle} b_i^\dagger b_i b_j^\dagger b_j + \mu \sum_i b_i^\dagger b_i \ , \quad \tilde{H}_{ab} = \frac{\hbar^2}{md_{ab}^2} \sum (a_i^\dagger - b_j^\dagger)(a_i - b_j) - v_0' \sum_{\langle ij \rangle} a_i^\dagger a_i b_j b_j^\dagger \ ,$$

where we included the terms that stem from the use of the grand canonical ensemble. The chemical potential μ is the same for both type of sites. The interaction energy is v_0 between atoms at the same sublattice, and v_0' in the ab case. The lattice constant is d in each lattice and d_{ab} between the lattices, m is the mass of the helium atoms. We have in mind the mass of ^{4}He, but the result should hold for the nonmagnetic part of ^{3}He as well. Since the optical frequency turns out to be inversely proportional to the mass, the ratio of excess conductances of ^{4}He and ^{3}He can be obtained.

It is more convenient to replace the creation operators $a_i^\dagger$ and $b_i^\dagger$ by the spin operators S_i^+ and σ_i^+ (and the annihilation operators a_i and b_i by S_i^- and σ_i^-). The transformation leads to a set of spin Hamiltonians (compare Ref. 28)

$$\begin{aligned} H_{aa} &= -J_{aa} \sum_{\langle ij \rangle} (S_i^+ S_j^- + S_i^- S_j^+) - J_{aa}' \sum S_i^z S_j^z - H \sum_i S_i \ , \\ H_{bb} &= -J_{aa} \sum_{\langle ij \rangle} (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) \\ &\quad - J_{bb}' \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - H \sum_i \sigma_i \ , \\ H_{ab} &= -J_{ab} \sum_{\langle ij \rangle} (S_i^+ \sigma_j^- + \sigma_i^+ \sigma_j^-) - J_{ab}' \sum_{\langle ij \rangle} S_i^z \sigma_j^z \ , \end{aligned} \tag{3}$$

where $J = \hbar^2/md^2$, $J' = v_0$ in each case, and

$$H = \mu - \tfrac{1}{2} z_{ab}(J_{ab} - J_{ab}') - \tfrac{1}{2} z_{aa}(J_{aa} - J_{aa}') \ ,$$

where z_{ab} and z_{aa} are the intersublattice and intrasublattice coordination numbers. We have assumed, and will continue to do so, that the coordination numbers and the coupling constants of each sublattice are equal: $z_{aa}=z_{bb}$, etc. This does not change the essential features of the model and makes the calculation easier.

The first step in the solution is to rotate each spin space so that linear terms are eliminated; this is done by means of two sets of Euler angles: π, θ, and $-\frac{1}{4}\pi$ in the S space and π, θ, and $-\frac{1}{4}\pi$ in the σ space. It is essential that θ, the angle in S space, need not be the same as ϕ, the angle in σ space. After this transformation, the terms of the Hamiltonian can be grouped in three sets: (i) a linear part, that is, all terms containing S^+, S^-, σ^+, or σ^-, either by themselves or multiplied by S^z or σ^z; (ii) a diagonal part, or self-part; and (iii) a coupling part containing all products of two spin-raising operators or two spin-lowering operators. The third part represents the excitation spectrum if there were only one subsystem. However, in our case we deal also with excitations between the subsystems, and this will lead to an additional excitation mode.

After this transformation is inserted we obtain a revised Hamiltonian; for details we refer to Ref. 28. This Hamiltonian contains a self-part and an excitation part. The self-part contains Ising terms and xy terms. The first terms give the ground-state energy. If we consider the angles θ and ϕ as the variables of a variational problem, then we can determine their values by minimizing the ground state with respect to θ and ϕ. This leads to two relations, both containing the magnetic field (i.e., the chemical potential). If we eliminate the field term, we are led to the following condition:

$$(z_{aa}J'_{aa}-z_{aa}J_{aa}-z_{ab}J'_{ab})\sin\theta\sin\phi = z_{ab}J_{ab}(1+\cos\theta\cos\phi) \quad . \tag{4}$$

We will call this the Matsuda-Tsuneto or MT condition. This condition plays an essential role in our description.

The relations obtained by the variational process can be inserted in the linear term, and the result is that this term disappears. The remaining Hamiltonian is used to construct the equations of motion after we take the Fourier transform. The result is

$$\begin{aligned} i\dot{S}_k^\dagger &= -\epsilon_1 S_k^\dagger + i\delta_1 S_{-k} - \epsilon\sigma_k^\dagger + i\delta\sigma_{-k} \quad , \\ i\dot{S}_{-k} &= i\delta_1 S_k^\dagger + \epsilon_1 S_{-k} + i\delta\sigma_k^\dagger + \epsilon\sigma_{-k} \quad , \\ i\dot{\sigma}_k^\dagger &= -\epsilon S_k^\dagger + i\delta S_{-k} - \epsilon_2\sigma_k^\dagger + i\delta_2\sigma_{-k} \quad , \\ i\dot{\sigma}_{-k}^\dagger &= i\delta S_k^\dagger + \epsilon S_{-k} + i\delta_2\sigma_k^\dagger + \epsilon_2\sigma_{-k} \quad , \end{aligned} \tag{5}$$

where $S_k=\sum_i S_i\exp(ikr_i)$ and $\sigma_k=\sum_i \sigma_i\exp(ikr_i)$ and the coefficients are given by

$$\epsilon_1=\Delta J\sin^2\theta\langle s\rangle + z_{ab}J_{ab}\langle\sigma\rangle(\sin\phi/\sin\theta) \quad ,$$

$$\epsilon_2=\Delta J\sin^2\phi\langle\sigma\rangle + z_{ab}J_{ab}\langle s\rangle(\sin\theta/\sin\phi) \quad ,$$

$$\epsilon=\Delta J\sin\theta\sin\phi\langle s\rangle \quad ,$$

$$\delta_1=\Delta J\sin^2\theta\langle s\rangle \quad ,$$

$$\delta_2=\Delta J\sin^2\phi\langle s\rangle \quad ,$$

$$\delta=(z_{ab}J_{ab}+\Delta J\sin\theta\sin\phi)\langle s\rangle \quad .$$

We took the structure factor $\gamma(k)=1$, the low-k limit, and wrote: $\Delta J=z_{aa}(J_{aa}-J'_{aa})$. If we consider the right-hand side of Eq. (5) written in matrix form, the resulting matrix M has the property[34]

$$M=-\begin{pmatrix}0&1\\1&0\end{pmatrix}M^*\begin{pmatrix}0&1\\1&0\end{pmatrix} \quad ,$$

which implies that $\lambda_1=-\lambda_2$ and $\lambda_3=-\lambda_4$. The first pair corresponds to an acoustic mode: $\lambda_{1,2}\to 0$ for $k\to 0$; the second pair corresponds to an optical mode, i.e.,

$$\lambda^2=\tfrac{1}{2}z_{ab}^2J_{ab}^2\left(\frac{\sin\phi}{\sin\theta}-\frac{\sin\theta}{\sin\phi}\right)^2+O(k^2) \quad . \tag{6}$$

This mode is only present if θ and ϕ are not equal to zero. The spin model for liquid helium is based on the idea that the kinetic energy is enhanced through the quantum effect and consequently these angles are not zero. The relative influence of this effect diminishes when the temperature is increased; i.e., each angle becomes smaller. In a strict molecular-field model, the angle becomes zero at the lambda temperature, but we will assume that in a more realistic model the short-range order will persist above this temperature and that even in this region the angle is not zero. The justification we present here is that the other manifestation of quantum effects in helium, the presence of rotons, does not disappear either above the λ temperature.

Since the equations that determine θ and ϕ are difficult to solve in terms of general coupling parameters and chemical potential, we will make the approximation that the parameters associated with the surface layer have values not too far from the parameters of the bulk liquid. If we make the coupling constants between the layers equal to the coupling constants in the layer: $J_{aa}=J_{ab}$ and $J'_{aa}=J'_{ab}$ we find from the MT relation that

$$\sin\theta\sin\phi+\cos\theta\cos\phi=-1 \quad .$$

Hence $\phi=\pi+\theta$. In this case, the optical mode is not present. If the interlayer coupling is slightly different, we take $\theta=\theta_0+\Delta\theta$ and $\phi=\pi+\theta_0-\Delta\theta$ and the optical mode can be expressed in terms of θ as follows

$$\lambda^2=\tfrac{1}{2}z_{ab}^2J_{ab}^2(4\cot\theta_0\Delta\theta)^2 \quad , \tag{7}$$

while $\Delta\theta$ is determined by the difference in coupling constants. Expanding the MT relation we find

$$(\Delta\theta)^2 = \frac{(J'_{ab} - J'_{aa}) - (J_{ab} - J_{aa})}{2J_{aa}} \sin^2\theta_0 \ . \tag{8}$$

Inserting this in the equation for the optical mode, gives:

$$\lambda^2 = 4J_{ab}(J'_{ab} - J'_{aa} - J_{ab} + J_{aa}) \cos^2\theta_0 \ . \tag{9}$$

It is interesting to notice that for small θ_0 this becomes independent of θ_0, consequently, there will still be an optical mode provided the J parameters are different from the J' parameters.

The result obtained substantiates the mechanical model: There is energy transferred to surface excitations at a *finite* frequency. This mechanism will disappear when $kT << \lambda$ in much the same way as the Debye specific heat will disappear below the characteristic temperature. There is indeed experimental evidence[35,36] that at lower temperatures, somewhere around 0.1 K, the conductivity approaches the Khalatnikov limit.

VI. CONCLUSION

In this paper we argue that the discrepancy between the observed thermal conductivity resulting from the phonon mismatch is due to the presence of an extra channel, through which energy can be transferred from the solid. This channel consists of a two step process: energy transfer from the transverse waves in the solid into the surface layers of helium atoms, and coupling from these helium atoms to the bulk liquid. The essential assumption is that this surface structure has an optical mode, i.e., a resonance at $\omega_0 \neq 0$ for $k \to 0$. We show in a mechanical-equivalence model how such a structure reacts to an impinging transverse acoustical wave.

In the second part of the paper we use the spin model of Matsubara and Matsuda to show that the quantum-mechanical off-diagonal long-range order is different in the surface layer from that in the bulk. This difference is due to the compression of the first few layers, and it leads to an optical branch.

This optical branch will contribute an additional term to the heat transfer coefficient; this term will also have a T^3 dependence, similar to the phonon-mismatch term, except at temperatures $T < \omega_0/k_B$. This drop-off has been observed and its position can be used to fit ω_0 and hence $\Delta J_{ab} - \Delta J'_{ab}$. At much longer lower temperatures, the heat transfer should follow the Khalatnikov value. Another prediction of our model is that the surface layer should have a temperature different from the temperature in the liquid and from the temperature in the solid. We hope that the experimental effort could be directed towards an observation of this prediction.

ACKNOWLEDGMENTS

This research was started with the help of an NSF grant and the final phase was accomplished when one of us (P.M.) held a Fulbright grant at the Ecole de Physique et Chimie in Paris; I would like to thank Professor P. Papon for his hospitality.

APPENDIX

Suppose a transverse wave, polarized in the plane of incidence, impinges upon the $z = 0$ plane, as shown. The transverse wave is reflected, while a longitudinal wave is generated in the continuum by the well-known mode conversion mechanism. In addition, a disturbance will now propagate through the structured, i.e., atomic, layer.

Let ξ and ζ be the total displacements in the x and z directions, respectively, in the solid, while X_n is the displacement, parallel to the surface, of a mass at position nx ($n = 0, 1, 2, \ldots$) on the surface structure. The total displacement of the continuum is the result of three displacements

$$\xi = \xi_t^i + \xi_t^r + \xi_l^r, \quad \zeta = \zeta_t^i + \zeta_t^r + \zeta_l^r \ , \tag{A1}$$

which comprise a composition of incident and reflected parts, where i refers to incident, r to reflected, and t and l to transverse and longitudinal, respectively. Each displacement can be written

$$\begin{aligned}
\xi_t^i &= \cos\alpha_t A \exp[ik_t(x\sin\alpha_t - z\cos\alpha_t) - i\omega t] \ , \\
\zeta_t^i &= \sin\alpha_t A \exp[ik_t(x\sin\alpha_t - z\cos\alpha_t) - i\omega t] \ , \\
\xi_t^r &= \cos\alpha_t B \exp[ik_t(x\sin\alpha_t + z\cos\alpha_t) - i\omega t] \ , \\
\zeta_t^r &= -\sin\alpha_t B \exp[ik_t(x\sin\alpha_t + z\cos\alpha_t) - i\omega t] \ , \\
\xi_l^r &= \sin\alpha_l C \exp[ik_l(x\sin\alpha_l + z\cos\alpha_l) - i\omega t] \ , \\
\zeta_l^r &= \cos\alpha_l C \exp[ik_l(x\sin\alpha_l + z\cos\alpha_l) - i\omega t] \ .
\end{aligned} \tag{A2}$$

The displacement of the discrete masses is given by:

$$\begin{aligned}
X_{n(\text{even})} &= (\tfrac{1}{2}N)^{-1/2} \sum_{k_s} Y_k e^{ik_s na} \ , \\
X_{n(\text{odd})} &= (\tfrac{1}{2}N)^{-1/2} \sum_{k_s} X_k e^{ik_s na} \ ,
\end{aligned} \tag{A3}$$

where N is the total number of masses on a strip of the surface, and k_s is the wave number in the surface structure. A is an arbitrary constant, B, C, X_k, and Y_k follow from the matching conditions.

The condition that must hold on the boundary is that of continuity of stress. Obviously, the displacements of the masses are not necessarily equal to the displacement of an element of surface on the solid, hence, continuity of displacement does not hold in

this model. The continuity of stress gives,

$$\sigma_{zz}=0 \ , \tag{A4}$$

$$\sigma_{zx}=\gamma(X_n-\xi) \ . \tag{A5}$$

This notation indicates the position of the surface by the first subscript and the direction of the stress by the second subscript (compare Sommerfeld, Ref. 23, p. 38).

The bending constant, γ, has the proper units so as to make the right-hand side of Eq. (A5) and expression of the force per interparticle distance.

We determine the four unknown amplitudes from the two conditions (A4) and (A5) and the equations of motion of the masses on the surface. These equations are given by:

$$m\ddot{X}_n=\beta_s(X_{n+1}+X_{n-1}-2X_n)+\gamma(\xi-X_n) \ , \tag{A6}$$

$$M\ddot{X}_{n+1}=\beta_s(X_{n+2}+X_n-2X_{n+1})+i\omega\kappa X_{n+1} \ , \tag{A7}$$

where n is even. The last term of Eq. (A6) describes the coupling of mass m to the solid. That of Eq. (A7) describes the coupling of mass M to the liquid.

First, we determine the natural frequencies of the structured layer. This is the problem of the classical diatomic linear chain.[26] We take the Fourier transform of Eq. (A6)

$$m(\tfrac{1}{2}N)^{-1/2}\sum_n \ddot{X}_n e^{-ik_s na}=\beta_s(\tfrac{1}{2}N)^{-1/2}\left(\sum_n X_{n+1}e^{-ik_s(n+1)a}e^{ik_s a}+\sum_n X_{n-1}e^{-ik_s(n-1)a}e^{-ik_s a}\right)$$

$$-2\beta_s(\tfrac{1}{2}N)^{-1/2}\sum_n X_n e^{-ik_s na}+\gamma(\tfrac{1}{2}N)^{-1/2}\sum_n(\xi-X_n)e^{-ik_s na} \ . \tag{A8}$$

We evaluate the different terms separately.

$$(\tfrac{1}{2}N)^{-1/2}\sum_n \xi e^{-ik_s na}=(\tfrac{1}{2}N)^{-1/2}\sum_n[(A\cos\alpha_t+3\cos\alpha_t)\exp(ik_t x\sin\alpha_t+C\sin\alpha_t)\exp(ik_l x\sin\alpha_l)]e^{-ik_s na} \text{ for } x=na \ .$$

At this point (using the condition that at $z=0$, the phase should be equal, regardless of the values of x or n) we recover Snell's law

$$\frac{2}{N}\sum_n \exp[i(k_t\sin\alpha_t-k_s)na]=\delta_{k_s,k_t\sin\alpha_t} \ ,$$

$$\frac{2}{N}\sum_n \exp[i(k_l\sin\alpha_l-k_s)na]=\delta_{k_s,k_l\sin\alpha_l} \ ,$$

with

$$k_s=\frac{\omega}{c_s}, \quad k_l=\frac{\omega}{c_l}, \quad k_t=\frac{\omega}{c_t} \ ,$$

with the c's being the respective velocities of sound. That is, we find:

$$k_s=k_t\sin\alpha_t=k_l\sin\alpha_l \ . \tag{A9}$$

Both equalities express the same idea; the components parallel to the surface must be equal. The second equality is the refraction law. Since the velocity of sound in helium is much smaller than that in the solid, on account of the low density, the angle of the cone is rather small. The first equality can only be fulfilled for one value of α_t which is also small; that means the cone is empty. We assume that there is a small spread in the surface velocities, and hence the cone has a certain thickness. The result is that the space angle associated with the second equality is less than but may be comparable with the space angle associated with the first equality.

Hence, we obtain:

$$(\tfrac{1}{2}N)^{-1/2}\sum_n \xi e^{-ik_s na}=\xi_k=(\tfrac{1}{2}N)^{1/2}[(A+B)\cos\alpha_t+C\sin\alpha_l] \ .$$

Noting that the time dependence enters by way of the factor $e^{i\omega t}$, that

$$X_k=(\tfrac{1}{2}N)^{-1/2}\sum_n X_n e^{-ik_s na} \ ,$$

$$Y_k=(\tfrac{1}{2}N)^{-1/2}\sum_n X_{n\pm1}e^{-ik_s(n\pm1)a} \ ,$$

and that the same arguments apply to Eq. (A7), we find for the Fourier transformed equations of motion

$$\begin{aligned} -m\omega^2X_k&=2\beta_s Y_k\cos k_s a-2\beta_s X_k+\gamma(\xi_k-X_k) \ , \\ -M\omega^2Y_k&=2\beta_s X_k\cos k_s a-2\beta_s Y_k+i\omega\kappa Y_k \ . \end{aligned} \tag{A10}$$

The eigenfrequencies are given by

$$\omega_\pm^2=\frac{\beta_s}{Mm}\{M+m\pm[(M-m)^2+\mu Mm\cos^2(k_s a)]^{1/2}\} \ . \tag{A11}$$

The eigenvectors are

$$X_k'=\sqrt{m}\cos\theta X_k+\sqrt{M}\sin\theta Y_k \ , \tag{A12}$$

$$Y_k'=-\sqrt{m}\sin\theta X_k+\sqrt{M}\cos\theta Y_k \ , \tag{A13}$$

where $tg(2\theta)=2(Mm)^{1/2}\cos(k_s a)/(M-m)$. Applying this transformation to Eqs. (A8) and (A9), we obtain

$$X_k'=\gamma\sqrt{m}\,\cos\theta[(Mm)^{1/2}(\omega^2-\omega_-^2)+i\omega\kappa(m/M)^{1/2}]\xi_k/G\equiv P\xi\ , \quad \text{(A14)}$$

$$Y_k'=\gamma\sqrt{M}\,\sin\theta[(Mm)^{1/2}(\omega^2-\omega_+^2)+i\omega\kappa(m/M)^{1/2}]\xi_k/G\equiv Q\xi\ , \quad \text{(A15)}$$

where

$$G=Mm(\omega^2-\omega_+^2)(\omega^2-\omega_-^2)-\gamma M[\cos^2\theta(\omega^2-\omega^2)+\sin^2\theta(\omega^2-\omega_+^2)]+i\omega\kappa\{m[\cos^2\theta(\omega^2-\omega_+^2)+\sin^2\theta(\omega^2-\omega_-^2)]-\gamma\}\ .$$

Note that G contains resonating terms even when γ and κ are zero. This implies that, X_k' or Y_k' become large with the proper values of the driving frequency, ω, when κ and γ are small.

Let us now return to Eqs. (A4) and (A5). Using the index of refraction, $n=k_l/k_t=(2+\lambda/\rho)^{-1/2}$ where μ and λ are Lame constants, and Snell's law, Eqs. (A4) and (A5) may be rewritten in the form

$$-(A+B)\sin2\alpha_t+(1/n)C\cos2\alpha_l=0\ , \quad \text{(A16)}$$

$$-i\mu k_t(A-B)\cos2\alpha_t+i\mu k_l C\sin2\alpha_l=\gamma(X_k-\xi_k)\ . \quad \text{(A17)}$$

Remember that ξ_k is a linear combination of A, B, and C. Inserting Eqs. (A14) and (A15) in the right-hand side of Eq. (A17) using the inverse transformation of Eqs. (A12) and (A13), and solving Eqs. (A16) and (A17) simultaneously, we find

$$\frac{B}{A}=\frac{\mu k_t B_0-\gamma(R-iS)}{\mu k_t A_0+\gamma(R-iS)}\ ,\qquad \frac{C}{A}=\frac{\mu k_t n\sin4\alpha_t}{\mu k_t A_0+\gamma(R-iS)}\ , \quad \text{(A18)}$$

where

$$A_0=\cos^2 2\alpha_t+n^2\sin2\alpha_l\sin2\alpha_t\ ,\qquad B_0=\cos^2 2\alpha_t-n^2\sin2\alpha_l\sin2\alpha_t\ , \quad \text{(A19)}$$

and

$$R=-(\cos\alpha_t/\sqrt{m})\,\mathrm{Im}(\cos\theta P-\sin\theta Q)\ ,\qquad S=\cos\alpha_t[1-(1/\sqrt{m})\,\mathrm{Re}(\cos\theta P-\sin\theta Q)]\ . \quad \text{(A20)}$$

The transmission coefficient **T** is found from the equations of conservation of energy flow perpendicular to the surface similar to Eq. (A3); i.e.,

$$\left|\frac{B}{A}\right|^2+\frac{\sin2\alpha_l}{\sin2\alpha_t}\left|\frac{C}{A}\right|^2=1-\mathbf{T}\ ,$$

with

$$\mathbf{T}=\frac{4\mu k_t\gamma R\cos^2 2\alpha_t}{(\mu k_t A_0-\gamma R)^2+\gamma^2 S^2}\ . \quad \text{(A21)}$$

Note that **T** vanishes, as it should, under each of the following conditions: $\kappa\to0$, $\gamma\to0$, $\beta_s\to0$.

If the damping constant κ is zero, there is no "sink" for the energy on the right-hand side ($z>0$), and consequently all the incoming energy will be reflected back into the solid. If the bending constant γ is zero, the discrete structure on the surface will be disconnected from the solid, and if the spring constant β_s is zero, there will be no connection with the viscous damping into the liquid.

The rate of energy transport to the right side is given by **T**, the transfer probability. This function is rather complicated. Therefore, in order to understand its behavior, let us make the simplification that $k_s\simeq0$ and $M\simeq m$. We now have $\cos\theta=\sin\theta=1/\sqrt{2}$, and the problem contains only three parameters: $4\beta_s/m=\omega_0^2$ as well as the coupling constant γ and the damping constant κ. We will assume γ to be small and omit all terms of order γ^3. This leads to a simple form of **T** given by

$$\mathbf{T}=(\text{const})\,\gamma^2\kappa g(\omega,\omega_\pm,\kappa)\ ,$$

where

$$(\text{const})=\frac{4\cos^2 2\alpha_t\cos\alpha_t}{\mu k_t A_0^2}$$

and

$$g(\omega,\omega_\pm,\kappa)=\frac{\omega(\omega_+^2-\omega_-^2)}{2m^2(\omega^2-\omega_+^2)^2(\omega^2-\omega_-^2)^2+\frac{1}{2}\kappa^2\omega^2[(\omega^2-\omega_+^2)+(\omega^2-\omega_-^2)]^2}\ . \quad \text{(A22)}$$

In the region of small k_s, $\omega_-\simeq0$ and ω intersects the ω_+ branch at ω_0.

From Eqs. (A14) and (A15) we can see that for small k_s, we obtain two resonance peaks, one at $\omega \simeq 0$ and the other at $\omega \simeq \omega_0$. The first peak does not contribute very much to the energy transport since the integrand contains a factor of $\hbar\omega$. So, despite the fact that there are many phonons at that frequency and that they have a large transfer probability, they carry little energy. The situation at $\omega \simeq \omega_0$ is different; there are fewer phonons present but the transfer probability is large and so is the amount of energy transferred. Moreover, it has been observed that phonons of high frequency are produced in the helium. That is, the spectrum of phonons produced in the helium does not necessarily have the same characteristic temperature as the heater.[21] This indicates that the "tail end" of the blackbody distribution from the heater interacts with the surface to produce high-frequency phonons in bulk helium. Although the model deals with two specific natural frequencies, $\omega_{\pm}$, the surface layer may be characterized by more frequencies, thus allowing a larger number of phonons from the heater to interact with the surface excitations.

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