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Interfacial three-phonon processes: Application to pulse experiments and the Kapitza conductance

M. E. Lumpkin* and W. M. Saslow
Department of Physics, Texas A&M University, College Station, Texas 77843

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We have studied interfacial three-phonon processes as a mechanism to explain the enhanced transmission of acoustic energy across the interface between (high phonon velocity) "classical" and (low phonon velocity) "quantum" systems. Because of phase-space considerations, decay of a phonon from the "classical" material to two phonons in the "quantum" material is possible for reasonable values of the interfacial cubic anharmonicity (1–10 eV/Å^2). The transmission rate, for an interaction depending on the cube of the surface strain, varies as ω^6, for an incident phonon frequency ω, so that the process rapidly "turns on" and then must saturate, in agreement with experiment. The mechanism causes most of the transmission to occur outside the critical cone, in agreement with experiment. The mechanism is also consistent with the observed frequency conversion of the incident phonon. When the characteristic wavelength of the transmitted phonons is considered (~16 Å), one can account for experiments showing that a 3-layer (12 Å) ^{4}He film behaves like bulk liquid ^{4}He. The mechanism appears to be consistent with a wide variety of experimental results, including recent pulse work on solid surfaces cleaved *in situ*. It is suggested that experiments be performed to verify the predicted ω^6 onset rate, perphaps by studying the transmitted energy outside the critical cone.

I. INTRODUCTION

It is an embarrassing fact that, for temperatures $T \gtrsim 0.1\,^\circ\mathrm{K}$, current theories of the Kapitza conductance h_k are often smaller than the experimental values by an order of magnitude or more.[1] There are a number of reasons why this might be the case: (i) Even if the dominant mechanisms for interfacial energy transport are known, it is not clear that the usual approach to calculating h_k is correct.[2] (ii) The dominant mechanism, usually taken to be direct transmission of phonons across the interface, may have a larger transmission coefficient than that given by the long-wavelength limit.[3] (iii) There may be significant contributions due to defects and impurities. (iv) Even in the case of a perfectly "clean" system, there may be additional mechanisms which dominate the interfacial energy transport. It is the purpose of the present paper to investigate this last possibility. Specifically, we will consider three-phonon processes mediated by anharmonicity at the interface.

Our motivation for investigating this question has been provided by recent experimental work, which we break up into five classes. First, we note the work of J. T. Folinsbee and A. C. Anderson[4] on measurements of h_k between "classical" and "quantum" systems (by "quantum" we mean liquid or solid ^{3}He, ^{4}He, H_2 or D_2) establishing that it is for such classes of interfaces that the theory is inadequate. Second, we note the work of Maris and coworkers[5,6] (which supports the results of Ref. 4), using a heat-pulse technique to measure phonon reflection coefficients of near-normal incident phonons at the interface between "classical" and "quantum" systems. For $T \rightarrow T$ and $L \rightarrow L$ processes (T and L refer to transverse and longitudinal phonons) they observed much lower reflection coefficients than predicted by the acoustical theory, and for mode-conversion processes ($T \rightarrow L$ and $L \rightarrow T$) they found much larger reflection coefficients than predicted by the acoustical theory. The results of Guo and Maris also show that about three atomic layers of helium on a solid surface are sufficient to make the helium behave like bulk liquid. This indicates that the relevant physical processes must be occurring at the interface, regardless of whether a thin film or bulk liquid is adjacent.

The third class of experiment, by Kinder and Dietsche,[7,8] involved the study of 130- to 870-GHz phonon reflection off an interface between solid surfaces and helium, in which an anomalously small reflectivity was observed, as well as both up-conversion[7] and down-conversion[8] of the pulse frequency. In addition, they too observed that the effect becomes nearly saturated at a thickness of about three atomic layers of helium.

The fourth class of experiment, by E. S. Sabisky and C. H. Anderson,[9] involved an indirect measurement of the frequency dependence of a quantity ϵ, monotonically related to the phonon reflectivity at an SrF_2–liquid-^{4}He interface. They found that ϵ decreases monotonically from a value near unity at 20 GHz, to below 0.5 around 120 GHz, and is independent of frequency between 120 and 315 GHz. These results were essentially independent of temperature

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between 1.4 and 2 °K. In other words, a saturation effect seems to occur. Note that their results do not appear to depend on pulse intensity, which they varied by about a factor of 10.

The fifth class of experiment, and one which we will discuss in detail because of its relevance to our calculations, was performed by Sherlock and Wyatt and their coworkers at Nottingham.[10,11] Acoustics predicts that a critical cone will be found in the material which has the slower phonon group velocities. This means that a phonon incident from any angle in the "fast" material will be transmitted only within the critical cone of the "slow" material. Conversely, only those phonons in the "slow" material which are incident from within the cone will be transmitted into the "fast" material; all others will be totally internally reflected. The Nottingham group, however, found that in the transmission of heat pulses from a solid material ("fast") to liquid helium ("slow"), greater than 85% of the total transmitted energy is transmitted into the helium outside the critical cone. Furthermore, for phonons incident from the helium, the experiments showed that phonons incident outside the cone are indeed being transmitted across the interface, although the transmitted intensity for "slow" to "fast" outside the cone is much smaller than the corresponding intensity for "fast" or "slow". There is, therefore, in addition to the "cone breaking", a fundamental anisotropy involved in energy transport across these interfaces; phonons behave differently depending upon their direction of incidence with respect to the interface. Unfortunately, the incident beam was not collimated, and the temperature dependence of the angular distribution of the transmitted energy (even in relative terms) was not given. As a consequence, a theoretical analysis of their work is rendered difficult. Nevertheless, the qualitative features of their experiments are so striking that we have attempted such an analysis. The results are not completely satisfying, but are sufficiently good that we believe that further work of this nature, both experimental and theoretical, is well worth pursuing. Indeed, pulse experiments of all types probably provide our best hope of understanding the Kapitza conductance problem.

It should be noted, before discussing any detailed calculations, that interfacial three-phonon processes are capable of explaining each of the above experiments, at least in a qualitative sense. Three-phonon processes are compatible with frequency conversion, and cone breaking (with its associated anisotropy). They are also compatible with three-layer ^{4}He films behaving like bulk ^{4}He, once the characteristic phonon wavelengths are considered. They can cause unusually large amounts of mode conversion. Furthermore, as will be seen in the course of the calculations, the effect will be important only for interfaces between "classical" and "quantum" materials. This is not because of an intrinsically quantum-mechanical effect, but rather because the low sound velocity of a quantum system renders the direct transmission process inefficient and, more importantly, causes the phase space for certain interfacial three-phonon processes to become enormously large. It should also be noted that attempts to explain measured h_k values solely in terms of an enhanced transmission coefficient are incapable of explaining a number of the pulse experiments.[7,8,10–12] Finally, we remark that the frequency dependence of the interfacial three-phonon process transmission is so great (ω^6 relative to the direct acoustic transmission) that, once the process becomes important, it rapidly swamps the ordinary process. If this domination is too great, then our perturbation-theory calculation can predict more transmitted energy than incident energy. In this case a more sophisticated, self-consistent, calculation must be done and this should lead to the saturation effect observed in Ref. 9.

For the benefit of the reader, we present a short analysis of the frequency dependence of the interfacial three-phonon transmission. Using Fermi's golden rule we must sum the square of the matrix element over allowable final states, subject to energy conservation. If we consider one phonon of frequency ω in one material splitting into two the unrestricted final-state phase space varies as ω^6. However, conservation of the two components of transverse momentum reduces this to ω^4, and energy conservation further reduces this to ω^3. Since the square of the matrix element goes as ω^3, the fractional energy transmission goes as ω^6. However, only the details of the phase-space analysis provide the prefactor of ω^6, and this prefactor is shown to be large in the case of a "classical"-"quantum" interface.

The remainder of the paper has the following form. In Sec. II we consider the general structure that a phenomenological matrix element for interfacial three-phonon processes can be expected to have. We conclude that the attempts of previous workers to obtain this matrix element, using semimicroscopic approaches, are probably in error, and we trace what we believe to be the source of this error. In Sec. III we consider the available phase space for various three-phonon processes and certain angular distributions are presented. We find that considerable amounts of phase space are available for such processes, which provide transmission of energy outside critical cones. Furthermore, we find that the processes do indeed exhibit anisotropy with respect to direction of incidence, which is observed experimentally. In Sec. IV we consider the effect that a nonconstant matrix element can have on these processes. We show that the general matrix element can be a linear combination of many different terms. We then arbitrarily choose a particular form, and find that the matrix element can considerably modify the shape of the angular distribution of emitted phonons. We also calculate the interaction energy per unit area at the interface which would be required for three-phonon processes to contribute an interfacial heat current equal to the heat current due to ordinary, single-phonon processes. For interfaces between classical and quantum materials the required surface energies compare favorably with the cohesive energy per unit area calculated for real materials, whereas for interfaces between classical materials we find the required surface energies to be unphysically large. In Sec. V we summarize our results and show how interfacial three-phonon processes are consistent with the aforementioned pulse work, as well as the recent pulse work of Weber *et al.*[12] Further discussion of the matrix element and of the interaction strength are also presented there.

II. PHENOMENOLOGICAL MATRIX ELEMENT

Although a large amount of theoretical work has been done on bulk multiphonon processes, there are very few studies of interfacial multiphonon processes. Two papers, however, are of particular significance. In his original paper on heat exchange between solids and helium, Khalatnikov[13] considered interfacial three-phonon processes using a matrix element derived from semiclassical considerations. He arrived at the conclusion that these processes contribute a negligible interfacial heat current compared with the direct transmission processes involved in the acoustic mismatch theory. In 1973, Sheard *et al.*[14] attempted a completely quantum-mechanical calculation of the Kapitza resistance for the solid–liquid-^{4}He interface. Processes involving phonons and rotons in the liquid helium were treated in a unified manner by using a realistic model for bulk helium. The coupling of the phonons in the solid to the excitations in the helium was derived in the form of a transfer Hamiltonian but the interaction Hamiltonian was derived from the interatomic potential between a helium atom and the solid. In the long-wavelength limit this theory reproduces the classical acoustic mismatch theory results for direct transmission processes. The coupling Hamiltonian also includes roton emission and higher-order processes involving two or more excitations in the helium. In their discussion of second-order processes, Sheard *et al.*, find that the ratio of the heat current due to three-phonon processes to the heat current due to direct transmission processes is 0.033 at 2 °K and becomes negligible at lower temperatures. Both Sheard *et al.* and Khalatnikov considered three-phonon processes employing a three-phonon matrix element M which varies as

$$M \propto (pq/k)^{1/2} , \tag{2.1}$$

where $\vec{k}$ ($\vec{p}$ and $\vec{q}$) are phonon wave vectors in the solid (liquid). For the process defined by $\vec{k}+\vec{p}\rightarrow\vec{q}$, Eq. (2.1) predicts that as the energy (and wave vector) of the phonon in the solid (described by wave vector $\vec{k}$) becomes very small, the matrix element for this transition becomes infinite. We believe that this result is incorrect and we can trace the $k^{-1/2}$ dependence to an oversimplified treatment of the lattice displacement of the solid. The matrix element we calculate, on the other hand, is proportional to $(kqp)^{1/2}$ (for the process $\vec{k}\rightarrow\vec{q}+\vec{p}$) and is obtained as the continuum limit of the cubic interfacial interaction for discrete systems.

For a discrete system one may write the cubic interfacial interaction for a three-phonon process as

$$C_{\alpha\beta\gamma}(\Delta u)_{k\alpha}(\Delta u)_{p\beta}(\Delta u)_{q\gamma} ,$$

where $C_{\alpha\beta\gamma}$ is the strength of the interaction and $(\Delta u)_{k\alpha}$ is the difference in the α-direction displacements from equilibrium (due to the $\vec{k}$ phonon) of the interfacial atoms. For small displacements in the z direction, this interaction leads to a matrix element of the form

$$M \propto \left[\frac{\partial u_z}{\partial z}\right]_k \left[\frac{\partial u_z}{\partial z}\right]_p \left[\frac{\partial u_z}{\partial z}\right]_q a^3 , \tag{2.2}$$

where a is the lattice spacing. Using the expression for the displacement operator $\vec{u}(\vec{r})$ (given in a later section), Eq. (2.2) leads to

$$M \propto (kqp)^{1/2} . \tag{2.3}$$

The matrix element given by Eq. (2.1) does not allow the solid to adjust to the liquids; i.e., the matrix element of Eq. (2.1) is based on displacements of the solid rather than differences in the displacements. On the other hand, the phenomenological interaction given by Eq. (2.2) explicitly depends on differences in displacements. Furthermore, being phenomenological, it permits all three-phonon processes (including one phonon in the liquid and two phonons in the solid, processes which do not appear in the theories of Refs. 13 and 14). As we will show in Sec. III, some processes not considered in Refs. 13 and 14 can be quite important. In addition, the phenomenological interaction does not depend on whether the materials are solid or liquid. (Recall the experimental evidence[4–6] indicating that the "excess" Kapitza conductance values which are measured with helium on one side of the interface do not depend on whether the helium is solid or liquid.) Finally, note that the coefficient of the matrix element is to be determined from experiment. We thus avoid the difficult problem of calculating the surface anharmonicity for systems whose surface harmonic terms are not well understood. In practice, it is the decision to leave this coefficient determined by experiment that enables us to study interfacial three-phonon processes. Previous theories specified the form *and* the coefficient of the matrix element.[13,14]

It is important to note that the argument for a matrix element of the form (2.2), as opposed to the forms given in Refs. 13 and 14, can be strengthened considerably in light of experiments which show that, on most solids, the first overlayer of ^{4}He is itself solid. In addition, this permits an estimate of the coupling strength C. These points will be discussed further in Sec. V, where we will show that the theoretical and experimental estimates of C compare favorably.

Note that it is not clear that Eq. (2.2) is correct in all contexts (particularly at high frequencies and large wave vectors), so it is safest at this time to take the attitude that the mechanism may perhaps be mediated by some unknown cubic anharmonic interaction at the interface. This attitude is especially prudent if we recall that h_k values are very sensitive to surface preparation and, therefore, to what is actually on the surface.[1,12,15] To separate matrix element from phase-space effects we have broken our calculations into two parts. In Sec. III we consider phase-space calculations only, appropriate if the matrix element is constant. In Sec. IV we include the effect of a matrix element consistent with Eq. (2.2). We believe that the existing experiments are, in the absence of detailed analysis, qualitatively consistent with the momentum dependence given by Eq. (2.3). Nevertheless, interfacial three-phonon processes provide so attractive a mechanism for explaining the Kapitza conductance problem that, if detailed experimental analysis casts doubt on our use of a matrix element whose momentum-dependence is given by Eq. (2.3), we would rather abandon that momentum dependence than the interfacial three-phonon process itself.

III. THREE-PHONON PROCESS PHASE-SPACE CALCULATIONS

A. Channel I

Consider the process $\vec{k} \rightarrow \vec{q} + \vec{p}$ (Channel I) in which a phonon in material $\alpha (z < 0)$ decays into two phonons in material $\beta (z > 0)$. To be specific we take material α to be a solid and material β to be a liquid, although in principle the phonons in each material may be either L (longitudinal) or T (transverse). For a clean surface, the matrix element M conserves momentum parallel to the interface. We temporarily assume that M is independent of the directions and energies of the phonons.

To calculate the rate of energy transmission observed by a detector subtending a solid angle $d\Omega_q$, we calculate the transition rate w for this process using Fermi's golden rule. Taking $\vec{q}$ in the direction of the detector, summing over all allowed $\vec{p}$ and $|\vec{q}|$, and weighting by the energy ϵ_q and by $|M|^2$, we find

$$I(\theta_q, \phi_q; \theta_k) = 2 \sum_{|\vec{q}|\vec{p}} \frac{2\pi}{\hbar} |M|^2 \epsilon_q \delta^{(2)}_{\vec{k}_{\|}, \vec{q}_{\|} + \vec{p}_{\|}} \delta(\epsilon_k - \epsilon_p - \epsilon_q) , \tag{3.1}$$

where $I(\theta_q, \phi_q; \theta_k)$ is the energy intensity observed at θ_q and ϕ_q in material β, due to $\vec{k}$. The angles are illustrated in Fig. 1. The factor of 2 appears outside the summation because the $\vec{q}$ and $\vec{p}$ phonons may be interchanged.

Summing over $\vec{p}_{\|}$ (thus eliminating the Kronecker δ), summing over p_z (thus eliminating the Dirac δ), employing the Debye approximation for all phonons, and converting the sum over $\vec{q}$ to an integral, we find

$$I(\theta_q, \phi_z; \theta_k) = \frac{A^2 |M|^2}{4\pi^3 c_L^5 \hbar^6} \int \frac{\epsilon_q^3 (\epsilon_k - \epsilon_q) \, d\epsilon_q}{\{[(\epsilon_k - \epsilon_q)/\hbar c_L]^2 - \vec{p}_{\|}^{\,2}\}^{1/2}} , \tag{3.2}$$

where A is the area of the interface and c_L is the phonon group velocity in the liquid. (The dependence on A^2 is only apparent, being due to our oversimplified matrix element.) Introducing $x = \epsilon_q / \epsilon_k$, Eq. (3.2) becomes

$$I(\theta_q, \phi_q; \theta_k) = \frac{A^2 |M|^2 \epsilon_k^4}{4\pi^3 c_L^4 \hbar^5} Z_1(\theta_q, \phi_q; \theta_k) , \tag{3.3}$$

where the dimensionless function $Z_1(\theta_q, \phi_q; \theta_k)$ is given by

$$Z_1(\theta_q, \phi_q; \theta_k) = \int_{x_l}^{x_u} \frac{x^3 (1-x) \, dx}{(f_1 x^2 + f_2 x + f_3)^{1/2}} ; \tag{3.4}$$

$$f_1 = \cos^2\theta_q , \tag{3.5}$$

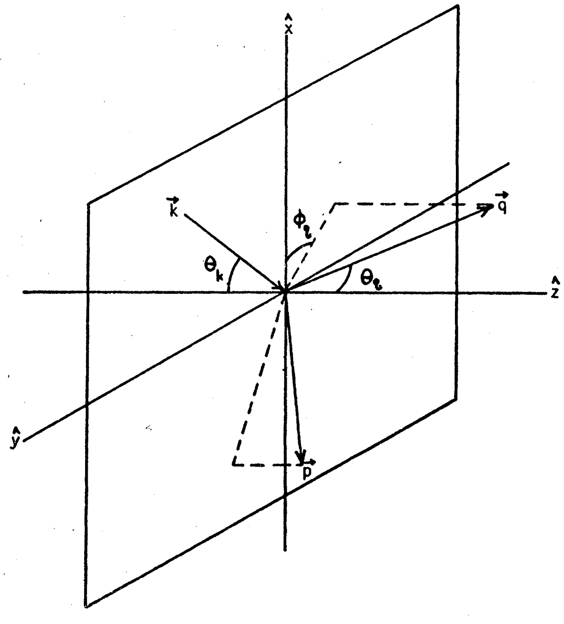

FIG. 1. Interfacial three-phonon process. The interface is the $z = 0$ plane.

TABLE I. Values of the coefficient of Z_1 in Eq. (3.7) for various three-phonon channels. The subscripts α and β refer to solid NaF and liquid helium at 20 atm pressure. L and T represent longitudinal and transverse phonons, respectively. z is the ratio of the coefficient of Z_1 for each channel to the coefficient for Channel I. The phonon group velocities are $c_{\alpha L}=6.22\times10^5$ cm/sec, $c_{\alpha T}=3.22\times10^5$ cm/sec, and $c_{\beta L}=3.47\times10^4$ cm/sec. The factor $\tilde{a}$ is defined by $\tilde{a}=(A^2M^2\epsilon_k^4)/4\pi^3\hbar^5$. Note that $c_{\beta T}$ processes have been neglected, since liquid helium does not support propagating transverse waves.

Channel	Three-phonon process	Coefficient of Z_1	z
I	$L_\alpha \rightarrow L_\beta + L_\beta$ or $T_\alpha \rightarrow L_\beta + L_\beta$	$\tilde{a}/c_{\beta L}^4$	1.00
II	$L_\beta \rightarrow L_\beta + T_\alpha$	$\tilde{a}/2c_{\beta L}c_{\alpha T}^3$	6.26×10^{-4}
III	$T_\alpha \rightarrow T_\alpha + L_\beta$	$\tilde{a}/2c_{\alpha T}c_{\beta L}^3$	5.39×10^{-2}
IV	$L_\alpha \rightarrow T_\alpha + L_\beta$	$\tilde{a}/2c_{\alpha T}c_{\beta L}^3$	5.39×10^{-2}
V	$L_\alpha \rightarrow L_\alpha + L_\beta$	$\tilde{a}/2c_{\alpha L}c_{\beta L}^3$	2.79×10^{-2}
VI	$T_\alpha \rightarrow L_\alpha + L_\beta$	$\tilde{a}/2c_{\alpha L}c_{\beta L}^3$	2.79×10^{-2}
VII	$L_\beta \rightarrow T_\alpha + T_\alpha$	$\tilde{a}/c_{\alpha T}^4$	1.35×10^{-4}
$VIII$	$L_\beta \rightarrow L_\beta + L_\alpha$	$\tilde{a}/2c_{\beta L}c_{\alpha L}^3$	8.68×10^{-5}
IX	$L_\beta \rightarrow L_\alpha + T_\alpha$	$\tilde{a}/2c_{\alpha L}c_{\alpha T}^3$	3.49×10^{-5}
X	$L_\beta \rightarrow L_\alpha + L_\alpha$	$\tilde{a}/c_{\alpha L}^4$	9.69×10^{-6}

$$f_2=-2\left[\frac{c_L}{c_S}\sin\theta_k\sin\theta_q\cos\phi_q+1\right], \tag{3.6}$$

and

$$f_3=1-\left[\frac{c_L}{c_S}\sin\theta_k\right]^2. \tag{3.7}$$

(c_S is the phonon group velocity in the liquid.) The $\cos\phi_q$ dependence enters because

$$\vec{p}_{\parallel}^{\,2}=\left[\frac{\epsilon_k}{\hbar c_S}\sin\theta_k\right]^2+\frac{2\epsilon_k\epsilon_q}{\hbar^2c_Sc_L}\sin\theta_k\sin\theta_q\cos\phi_q+\left[\frac{\epsilon_q}{\hbar c_L}\sin\theta_q\right]^2. \tag{3.8}$$

The integral in Eq. (3.4) is evaluated in the Appendix. In calculating numbers, one must carefully consider the upper and lower limits, x_u and x_l, to ensure that the argument of the square root is always positive. Physically, this corresponds to requiring that p_z be real.

Taking c_L/c_S to be 0.1 (to correspond approximately to a solid-liquid helium interface), we computed $Z_1(\theta_q,\phi_q;\theta_k)$ vs θ_q, as shown in Fig. 2. There is little dependence upon θ_k or ϕ_q, but the dependence on θ_q is significant. A critical cone occurs in the liquid, at $\theta_q^c=\sin^{-1}(c_L/c_S)\approx5.7°$. Figure 2 shows that most of the energy transmitted by this three-phonon channel is transmitted outside the critical cone. In fact, for $\theta_k=0°$ and $\phi_q=180°$, 81% of the total energy transmission from this process occurs for $\theta_q>6°$. Thus, Channel I is certainly capable of transmitting large amounts of energy outside the critical cone. There are four other channels for this solid-liquid system in which the incident phonon is in the "fast" material but for these channels (Channel $III-VI$ of Table I) the intensities are smaller than

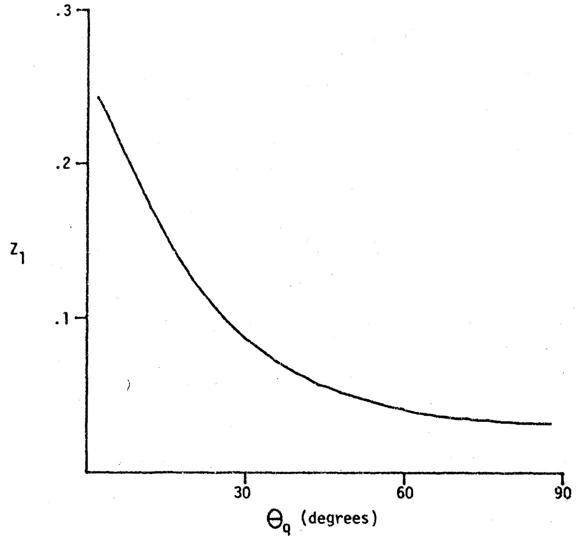

FIG. 2. Dimensionless integral Z_1 in Eq. (3.6) vs θ_q for Channel I. The values of θ_k and ϕ_q are 0° and 180°, respectively.

those of Channel *I* by at least an order of magnitude. This is primarily because, for these other channels, the constant which multiplies the dimensionless integral in Eq. (3.7) is much smaller than the constant for Channel *I*. Note that Channels *IV* and *VI* are responsible for mode conversion.[5]

B. Channel *II*

For a phonon $\vec{k}$ incident from the liquid, or low velocity side, there are also five channels, labeled Channel *II* and *VII*–*X* in Table I. In this case, the dominant channel (Channel *II*) is that for which $\vec{k}$ decays into a phonon $\vec{p}$ in the liquid, and a transverse phonon $\vec{q}$ in the solid. This is because the greatest amount of phase space is available to $\vec{q}$ when $\vec{p}$ emerges in the slow material. The intensity at the detector, which is now in the solid, is as in Eq. (3.3) except that no factor of 2 appears outside the summation, since now only one phonon propagates in the detection material. The intensity is given by

$$I(\theta_q,\phi_q;\theta_k)=\frac{A^2|M|^2\epsilon_k^4}{(2\pi)^3\hbar^5 c_L c_S^3}\,Z_1(\theta_q,\phi_q;\theta_k)\ , \quad (3.9)$$

where the functions f_1, f_2, and f_3 in Z_1 are redefined as

$$f_1=1-\left[\frac{c_L}{c_S}\sin\theta_q\right]^2 , \quad (3.10)$$

$$f_2=-2\left[1+\frac{c_L}{c_S}\sin\theta_k\sin\theta_q\cos\phi_q\right] , \quad (3.11)$$

and

$$f_3=\cos^2\theta_k\ . \quad (3.12)$$

The resulting intensities in Channel *II*, given in Fig. 3 for $c_L/c_S=0.1$, are quite different from those in Channel *I*. The dependence on θ_q and ϕ_q is weak, but its dependence on θ_k is strong.

Figure 3 shows that Channel *II* also permits the transmission of energy even though the incident phonon is outside the critical cone in the liquid. Therefore, both Channels *I* and *II* allow transmission of phonons from regions which are forbidden by classical acoustic theory. This agrees with the experimental observations of Wyatt *et al.*[11] Furthermore, from Table I the ratio of the rate of energy transmission by Channel *I* to the rate of energy transmission by Channel *II* is very large. Thus much more energy is transmitted by these three-phonon channels in going from the solid to the liquid than going from the liquid to the solid. The experimental results of Wyatt *et al.*,[11] show this behavior also, but in a somewhat less striking fashion than our table would indicate. Therefore, although we have not yet considered the details of the matrix element for these processes, it is apparent that three-phonon processes are an attractive candidate for explaining both the transmission of energy outside critical cones and the anisotropy of the transmission of energy which is observed experimentally. They can also explain mode conversion. We consider the matrix element in more detail in Sec. IV.

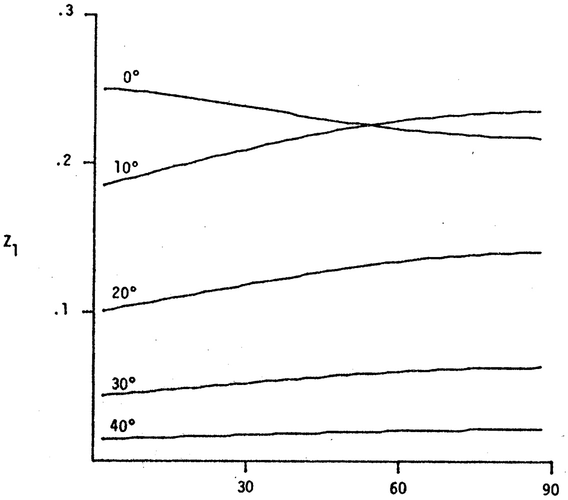

FIG. 3. Dimensionless integral Z_1 in Eq. (3.11) vs θ_q for Channel *II*. The value of ϕ_q is 180° and the family of curves represents various values of the incident angle θ_k.

We close this section with some remarks about two different classes of interfacial three-phonon process; namely, those involving only phonons on one side of the interface, and those involving surface phonons. For all such processes, momentum is not conserved perpendicular to the interface. Therefore, three-phonon decay that from phase-space considerations would be forbidden in the "fast" bulk solid (such as $T\rightarrow L+L$ or $T\rightarrow L+T$) now becomes possible. However, the available phase space for this process is much lower than for the related process discussed in this section (Channel *I*). On the other hand, $L\rightarrow L+L$ processes involving phonons only in the "slow" liquid have a significant amount of phase space relative to Channel *II*. This will be taken into consideration later, when transmission from liquid to solid is discussed. In the case involving surface (S) phonons and two solid phonons (e.g., $L\rightarrow L+S$) there again is phase space available for such processes. However, due to the high velocity of surface phonons there is much more phase space available for processes with the surface phonon replaced by a phonon in the liquid. Nevertheless, one should not offhand dismiss the possibility that such processes occur with a measurable probability. We have not pursued this question in any greater detail.

IV. CALCULATIONS INCLUDING A THREE-PHONON PROCESS MATRIX ELEMENT

Although the constant matrix element analysis is a useful initial approach, it is nevertheless an incomplete one. The matrix element should depend on ϵ_k, θ_k, θ_q, and ϕ_q. In order to obtain such a dependence, we will model each of the two materials by an isotropic elastic continuum and then apply classical elasticity theory to obtain the matrix element. This matrix element may then be incorporated into the phase-space integrations of Sec. III. Only Channel I will be considered.

A. Form of the interaction

Kittel[16] and Ziman[17] give the lattice displacement operator for bulk plane-wave phonons in an isotropic elastic material,

$$\vec{u}(\vec{r}) = \frac{1}{V^{1/2}} \sum_{\vec{k}\mu} \left(\frac{\hbar}{2\rho\omega_{k\mu}} \right)^{1/2} (a_{\vec{k}\mu} e^{i\vec{k}\cdot\vec{r}} + a^{\dagger}_{\vec{k}\mu} e^{-i\vec{k}\cdot\vec{r}}) \hat{e}_{\vec{k}\mu} , \tag{4.1}$$

where $\vec{u}(\vec{r})$ is the displacement of the atom at position $\vec{r}$ from its equilibrium position, V is the volume, ρ is the density, $\hbar\omega_{k\mu}$ is the phonon energy, $a_{\vec{k}\mu}$ and $a^{\dagger}_{\vec{k}\mu}$ are (dimensionless) phonon destruction and creation operators, μ is the polarization index, and $\hat{e}_{\vec{k}\mu}$ is a unit vector in the direction of the polarization of the phonon. Since we are dealing with interfacial decay processes, we must generalize Eq. (4.1) to

$$\vec{u}(\vec{r}) = \frac{1}{V^{1/2}} \sum_{\vec{k}\mu} \left(\frac{\hbar}{2\rho\omega_{k\mu}} \right)^{1/2} (a_{k\mu}\Psi_k + \text{H.c.}) \hat{e}_{k\mu} , \tag{4.2}$$

where Ψ_k is a scattering state containing the incident phonon and its reflected and transmitted components, and H.c. stands for "Hermitean conjugate". (V and ρ are now the volume and density of the material containing the incident phonon; this ensures that the displacements of the two materials at the interface will be equal.) For Channel I, the incident phonon is an incoming $\Psi_k^{(+)}$ state and the outgoing phonons are the states $\Psi_q^{(-)}$ and $\Psi_p^{(-)}$.

Having chosen a form for the displacement operator, it is now necessary to choose a form for the cubic interaction which allows a phonon to decay at the interface into two new phonons. We assume that there is rotational symmetry about the z axis (which is perpendicular to the interface). The following five operators have this symmetry:

$$\partial_z u_z , \quad \partial_x u_x + \partial_y u_y , \quad \partial_x u_y - \partial_y u_x ,$$

$$(\partial_z u_x)^2 + (\partial_z u_y)^2 , \quad (\partial_x u_z)^2 + (\partial_y u_z)^2 .$$

They may be combined to form 16 possible cubic interactions. In the absence of rotational symmetry about the z axis, even more cubic interactions become possible.

Treating each material as an isotropic elastic continuum, thus permitting both longitudinal and transverse phonons, we find 24 possible three-phonon channels, such as

$$L_\alpha \rightarrow L_\alpha + L_\beta , \quad T_\beta \rightarrow T_\alpha = T_\alpha , \quad L_\beta \rightarrow L_\alpha + T_\alpha .$$

Clearly, with 24 possible channels and 16 possible interactions for each channel, we must limit the scope of the investigation; we choose to consider only Channel I in detail, in order to compare with the data of Ref. 11.

A simple choice for the cubic interaction for Channel I is $H' = \int \mathcal{H}' \, d\vec{r}$, where

$$\mathcal{H}' = C(\partial_z u_z)_k (\partial_z u_z)_p (\partial_z u_z)_q \delta(z) , \tag{4.3}$$

C is a phenomenological coupling strength of dimension erg/cm^2, and the δ function ensures that this process occurs only at the interface. Note that the angular dependence of this interaction is not consistent with the results of Ref. 12, which will be discussed in Sec. V. However, the results of Ref. 12 became available only after our calculations had been completed.

Each of the displacement operators is to be evaluated on one side of the interface; we will evaluate them in the "slow" material β, which we now treat as an isotropic solid, rather than a liquid. Consider the $\Psi_k^{(+)}$ state in which a plane-wave phonon of wave vector $\vec{k}_0$ in the xz plane is incident from the "fast" material α. There will be two reflected waves of longitudinal and p-transverse polarization with wave vectors $\vec{k}_{\alpha l}$ and $\vec{k}_{\alpha t}$. (A "p-transverse" phonon is polarized in the plane of incidence; here, in the x-z plane. An "s-transverse" phonon has its polarization parallel to the interface; here, perpendicular to the plane of the incidence.) There will also be two transmitted waves, one longitudinal and one p transverse, described by wave vectors $\vec{k}_{\beta l}$ and $\vec{k}_{\beta t}$. All these wave vectors are in the x-z plane. The wave vectors $\vec{k}_0$, $\vec{k}_{\alpha l}$, and $\vec{k}_{\alpha t}$ are at θ_{ko}, $\theta_{k\alpha l}$, and $\theta_{k\alpha t}$, these angles being measured from the $-z$ axis. The wave vectors $\vec{k}_{\beta l}$ and $\vec{k}_{\beta t}$ are at $\theta_{k\beta l}$ and $\theta_{k\beta t}$, measured from the $+z$ axis. These angles are related by the acoustic analog of Snell's law,

$$\frac{\sin\theta}{\sin\theta'} = \frac{c}{c'} , \tag{4.4}$$

where c and c' are the phonon group velocities.

The $\Psi_k^{(+)}$ state in the β material is given by

$$\Psi_{k\beta}^{(+)} = \hat{k}_{\beta l} t_{kl} e^{i\vec{k}_{\beta l}\cdot\vec{r}} + (\hat{k}_{\beta t} \times \hat{y}) t_{kt} e^{i\vec{k}_{\beta t}\cdot\vec{r}} , \tag{4.5}$$

where t_{kl} and t_{kt} are the longitudinal and transverse transmission coefficients. The $\Psi_q^{(-)}$ state has two "transmitted" waves in the α material joined with two "reflected" waves in the β material to give a phonon $\vec{q}_0$ in the β material; in the β material, it may be written (for $\phi_{q_0}=0$),

$$\Psi_{q\beta}^{(-)}=\hat{q}_0 e^{i\vec{q}_0\cdot\vec{r}}+\hat{q}_{\beta l} r_{ql}^{(-)} e^{i\vec{q}_{\beta l}\cdot\vec{r}}+(\hat{q}_{\beta t}\times\hat{y}) r_{qt}^{(-)} e^{i\vec{q}_{\beta t}\cdot\vec{r}}, \tag{4.6}$$

where $r_{ql}^{(-)}$ and $r_{qt}^{(-)}$ are the reflection coefficients for the $\Psi_q^{(-)}$ state. The $\Psi_p^{(-)}$ is treated in the same way. For $\phi_{qo}\neq 0$, $\Psi_{q\beta}^{(-)}$ is obtained from Eq. (4.6) by a rotation of ϕ_{q0}.

Now define

$$M=\langle n_k-1,1,1|H'|n_k,0,0\rangle\ , \tag{4.7}$$

where the indices correspond to the occupation numbers for phonons of wave vectors $\vec{k}$, $\vec{q}$, and $\vec{p}$, respectively. Using Eqs. (4.2)–(4.6) in Eq. (4.7) we find

$$M=\frac{i\hbar CA}{V_\beta c_{\beta l}^3}\left[\frac{\hbar\omega_k\omega_q\omega_p n_k}{8\rho_\alpha\rho_\beta^2 V_\alpha}\right]^{1/2} B_k B_p B_q \delta^{(2)}_{\vec{k}_{\|},\vec{p}_{\|}+\vec{q}_{\|}}\ , \tag{4.8}$$

where

$$B_k=t_{kl}\cos^2\theta_{k\beta l}-\frac{c_{\beta l}}{c_{\beta t}}t_{kt}\sin\theta_{k\beta t}\cos\theta_{k\beta t}\ , \tag{4.9}$$

$$B_q=\cos^2\theta_{q_0}+r_{ql}\cos^2\theta_{q_0}-\frac{c_{\beta l}}{c_{\beta t}}r_{qt}\sin\theta_{q\beta t}\cos\theta_{q\beta t}\ , \tag{4.10}$$

$$B_p=\cos^2\theta_{p_0}+r_{pl}\cos^2\theta_{p_0}-\frac{c_{\beta l}}{c_{\beta t}}r_{pt}\sin\theta_{p\beta t}\cos\theta_{p\beta t}\ . \tag{4.11}$$

Note that although B_k and B_q contain no energy dependence, B_p depends upon ϵ_q/ϵ_k because the angular position of the wave vector $\vec{p}$ depends on the angular position of $\vec{q}$ and the length of $\vec{q}$.

B. Intensity

Using Eq. (4.8) in Eq. (3.1), and setting $V_\alpha=V_\beta=V$, we find

$$I(\theta_{q_0},\phi_{q_0},\theta_{k_0})=\frac{C^2 n_k A\epsilon_k^7}{32\rho_\alpha\rho_\beta^2\pi^3 V\hbar^5 c_{\beta l}^{10}}\times Z_2(\theta_{k_0},\theta_{q_0},\phi_{q_0})\ , \tag{4.12}$$

where the dimensionless function Z_2 is given by

$$Z_2(\theta_{k_0},\theta_{q_0},\phi_{q_0})=\int\frac{x^4(1-x)^2\,dx}{(f_1x^2+f_2x+f_3)^{1/2}}\times|B_kB_qB_p(x)|^2\ , \tag{4.13}$$

f_1, f_2, and f_3 are given by Eqs. (3.5), (3.6), and (3.7), and $x=\epsilon_q/\epsilon_k$.

We now find the dependence of B_p on x. Equation (4.4) allows us to express B_p in terms of θ_{p_0}. With $\epsilon_p=\epsilon_k-\epsilon_q$ and $\vec{k}_{\|}=\vec{p}_{\|}+\vec{q}_{\|}$, we find

$$\phi_{p_0}=\tan^{-1}\left[\frac{x\sin\theta_{q_0}\sin\phi_{q_0}}{x\sin\theta_{q_0}\cos\phi_{q_0}+(c_{\beta l}/c_{\alpha_l})\sin\theta_{k_0}}\right], \tag{4.14}$$

and

$$\theta_{p_0}=\sin^{-1}\left[\left(\frac{x}{x-1}\right)\frac{\sin\theta_{q_0}\sin\phi_{q_0}}{\sin\phi_{p_0}}\right]. \tag{4.15}$$

The argument of $\sin^{-1}$ in Eq. (4.15) will not exceed unity or diverge because the range of integration is only over those values of x for which $0\leq\theta_{p_0}\leq 90°$, corresponding to real values of p_{0z}.

Before evaluating the integral in Eq. (4.13), we need the reflection and transmission coefficients which appear in Eqs. (4.9), (4.10), and (4.11). To obtain them, one must solve four equations (two involving stresses and two involving displacements), in

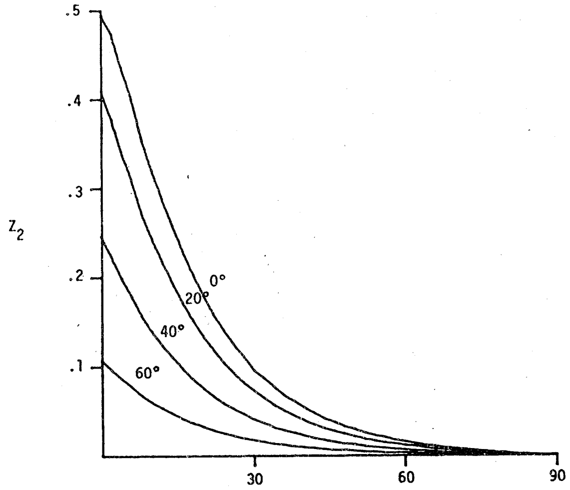

FIG. 4. Dimensionless integral Z_2 in Eq. (4.13) vs the detector zenithal angle θ_{q_0}. The value of ϕ_{q_0} is 0° and the family of curves represents various values of the incident angle θ_k.

four unknowns[18] (the reflection and transmission coefficients), for the three phonons $\vec{k}$, $\vec{p}$, and $\vec{q}$, given the three input parameters θ_{q_0}, ϕ_{q_0}, and θ_{k_0}.

Equation (4.13) was evaluated numerically by using Simpson's rule, with suitable modifications when a singularity appeared in the region $0 \leqslant x \leqslant 1$. A typical plot of $Z_2(\theta_{k_0}, \theta_{q_0}, \phi_{q_0})$ vs θ_{q_0} for $\phi_{q_0}=0$ and various values of θ_{k_0} is shown in Fig. 4. The material dependent parameters used are

$$c_{\alpha_l}/c_{\beta l} = c_{\alpha t}/c_{\beta t} = \rho_\alpha/\rho_\beta = 10$$

and $c_{\alpha l}/c_{\alpha t} = c_{\beta l}/c_{\beta t} = 3$. These ratios are approximately the values appropriate for a NaF—solid-^{4}He interface. Although the basic shape of these intensity curves are unchanged by the inclusion of the matrix element, the matrix element causes a much stronger dependence upon θ_{k_0}. As before, most of the energy is transmitted outside the critical cone. Note that here we have treated the He as a solid, rather than a liquid; this should not affect the results qualitatively, and should have only a minor effect on the quantitative results.

We now estimate the magnitude of the surface energy C, which is the only adjustable parameter in this phenomenological theory. Since the transmitted intensities for the matrix element given by Eq. (4.3) are weakly dependent on θ_{k_0}, we will assume normal incidence ($\theta_{k_0}=0$). The incident energy flux is given by

$$I_{\text{in}} = \frac{1}{V} n_k \epsilon_k c_{\alpha l} \ . \tag{4.16}$$

Therefore, the rate of energy transmission by three-phonon processes (normalized to the incident rate) is

$$\begin{aligned} U &= \int_0^{\pi/2} \int_0^{2\pi} \left(\frac{I}{I_{\text{in}}} \right) \sin\theta_q \, d\theta_q \, d\phi_q \\ &= \frac{C^2 \epsilon_k^6}{32 \rho_\alpha \rho_\beta^2 \pi^3 \hbar^5 c_{\beta l}^{10} c_{\alpha l}} \int_0^{\pi/2} \int_0^{2\pi} Z_2(\theta_{k_0}=0, \theta_{q_0}, \phi_{q_0}) \sin\theta_{q_0} \, d\theta_{q_0} \, d\phi_{q_0} \ . \end{aligned} \tag{4.17}$$

The integral over $d\phi_{q_0}$ gives 2π since normal incidence gives azimuthal symmetry. The integral over $d\theta_{q_0}$ was performed numerically. In order to find a typical value of ϵ_k, the energy of an incident phonon, we define a thermal value of $\langle \epsilon_k^6 \rangle$ by writing

$$\langle \epsilon_k^6 \rangle = \frac{\int n_k \epsilon_k^6 \rho(\epsilon) \, d\epsilon}{\int n_k \rho(\epsilon) \, d\epsilon} \tag{4.18}$$

where $\rho(\omega)$ is the density of phonon states. The result is

$$\langle \epsilon_k^6 \rangle = 1.16 \times 10^{-91} T^6 \ , \tag{4.19}$$

where T is the temperature in °K. Using values appropriate to NaF and solid ^{4}He, and setting $T=1$ °K, Eq. (4.17) gives

$$U = 1.19 \times 10^{-12} C^2 \ .$$

The results of Wyatt *et al.*,[11] suggest that at temperatures around 1 °K the heat current due to three-phonon processes is the same order of magnitude as the direct transmission heat current predicted by Khalatnikov. (Although our calculation is for solid ^{4}He, whereas the experiments are for liquid ^{4}He, our results should be appropriate to the experiments, at least with respect to order-of-magnitude calculations, which is all that is needed.) Thus, in order to solve for C, we now equate U to α, the acoustic transmission coefficient for normal incidence. For NaF against solid ^{4}He, $\alpha=0.029$. Solving for C and changing units, we find $C=9.8$ eV/Å^2. If one multiplies the crystal cohesive energy density for a typical solid (e.g., Cu) by the lattice spacing, one finds the cohesive energy per unit area to be in the order of 1 eV/Å^2. Therefore, realistic values of the coupling strength C will allow the three-phonon contribution to the interfacial heat current to be as large as the direct transmission contribution. (Section V provides a more extended discussion of C.) Because of the limited available phase space and the similarity between the phonon group velocities, we do not expect three-phonon processes to contribute significantly to the heat current across interfaces between classical solids (e.g., NaF against Cu). Indeed, our calculations show that for NaF against Cu, the three-phonon heat current is equal to the direct transmission heat current at 1 °K only if the surface interaction strength is 1.5 GeV/Å^2. (If the surface interaction strength takes on the more reasonable value of 1.5 eV/Å^2, the three-phonon heat current equals the direct transmission heat current for NaF-Cu only at pulse temperatures of 10^3 K.) Furthermore, experimental measurements on interfaces between classical solids show that the Khalatnikov theory is sufficient,[1] which implies that three-phonon contributions to the heat current are negligible in these systems.

Recall that our choice of the interfacial cubic anharmonic interaction was only one of sixteen possibilities, if we can limit ourselves to a symmetrical interface. One might argue that we have been too specific in our choice of this interaction, which will affect the details of the angular distribution of the transmitted energy. The angular dependence of the matrix element in $I(\theta_q, \phi_q; \theta_k)$ is contained in the terms B_k, B_q, and B_p. Since B_k and B_q do not depend on x, and $B_p(x)$ is a slowly varying function of x, we decided to absorb B_x, B_q, and B_p (which contain the angular dependence of the matrix element) into the phenomenological constant C in Eq. (4.12) and call this new constant $\tilde{C}$. The result is

$$I(\theta_{q_0}, \phi_{q_0}, \theta_{k_0}) = \frac{\tilde{C}^2 n_k A \epsilon_k^7}{32 \rho_\alpha \rho_\beta^2 \pi^3 V \hbar^5 C_{\beta l}^{10}} \times Z_3(\theta_{k_0}, \theta_{q_0}, \phi_{q_0}) , \tag{4.20}$$

where

$$Z_3(\theta_{k_0}, \theta_{q_0}, \phi_{q_0}) = \int \frac{x^4 (1-x)^2 \, dx}{(f_1 x^2 + f_2 x + f_3)^{1/2}} , \tag{4.21}$$

and the phase-space angular dependence of Z_3 is contained in the functions f_1, f_2, and f_3. Using Eqs. (4.17) and (4.21) and again setting $U = \alpha$ at 1 °K, we find $\tilde{C} = 0.96$ eV/Å^2. Again, the required coupling strength has a reasonable value.

We have also studied our predicted energy (temperature) dependence. Wyatt *et al.*[11] plot the experimentally measured dependence of E_P/E_T versus temperature for NaF → liquid ^{4}He, where E_P is the energy transmitted within the critical cone and E_T is the total transmitted energy. Our theory gives

$$\frac{E_P}{E_T} = \frac{U_P + U_k}{U + U_K} , \tag{4.22}$$

where U is given by Eq. (4.17), U_P is the same as U except that the integration $d\theta_{q_0}$ is only within the critical cone, and we assume that U_K is the acoustic contribution within the critical cone, given by

$$U_K = 2 \int_0^{\pi/2} \alpha(\theta_k) \sin\theta_k \cos\theta_k \, d\theta_k . \tag{4.23}$$

Here we have

$$\alpha(\theta_k) = \frac{\rho_\beta c_{\beta_l} [1 - (c_{\beta l} \sin\theta_k / c_{\alpha l})^2]^{1/2}}{\rho_\alpha c_{\alpha l} \cos\theta_k} \, | t_{kl}(\theta_k) |^2 \tag{4.24}$$

and $t_{kl}(\theta_k)$ is solved for each value of θ_k numerically. For the NaF–liquid-^{4}He interface, where the ^{4}He is considered now as a liquid in evaluating Eq. (4.23), the value of U_k is 8.74×10^{-3}. Using Z_3 in Eq. (4.17) we find

$$U = 5.57 \times 10^{-11} \tilde{C}^2 T^6 , \tag{4.25}$$

where T is the temperature measured in °K. Equation (4.4) gives the critical angle in the helium for the NaF–liquid-^{4}He interface as 3.2°, and this results in a value for U_P given by

$$U_P = 2.08 \times 10^{-13} \tilde{C}^2 T^6 . \tag{4.26}$$

We now rewrite Eq. (4.22)

$$\frac{E_P}{E_T} = \frac{a \tilde{C}^2 T^6 + 1}{b \tilde{C}^2 T^6 + 1} , \tag{4.27}$$

where a and b are given by

$$2.38 \times 10^{-11} \text{ cm}^4/\text{erg}^2 \, {}^\circ\text{K}^6$$

and

$$6.37 \times 10^{-9} \text{ cm}^4/\text{erg}^2 \, {}^\circ\text{K}^6 ,$$

respectively. We performed a least-squares fit of Eq. (4.27) for the data from 0.6 to 4.4 K given by Wyatt *et al.*,[11] using $\tilde{C}$ as the fitting parameter. The best fit is given by $\tilde{C} = 10.1$ eV/Å^2.

A comparison of theory with experiment is shown in Fig. 5. Clearly, the T^6 dependence in the theory is much too rapid to explain the data. In particular, the fit for 1–2 °K is not good, and $(E_P/E_T)_{\text{exp}}$ may just be saturating near 4 °K, whereas $(E_p/E_T)_{\text{th}}$ saturates very near 1 °K. In addition, at the higher temperatures our calculation has the energy within the peak being dominated by the three-phonon process. This is contrary to experiment,[11] and can be taken to mean that the angular dependence of the matrix element given by Eq. (4.3) is inappropriate because it permits too much energy to enter within the critical cone. Other evidence that Eq. (4.3) is inappropriate[12] will be discussed in Sec. V. Because the transmitted pulse within the critical cone seems to maintain its shape for all incident pulse temperatures, it is likely that E_P is probably not very temperature dependent. It would be of some interest, therefore, to try to fit E_P/E_T to the form $(AT^6 + 1)/(BT^6 + 1)$ for $A = 0$. Without attempting this fit, it is nevertheless clear that once the three-phonon process "turns on", it will rapidly come to dominate, with E_P/E_T approaching zero rapidly as T increases. In fact, the data relevant to this question is not really definitive.[11] Physically, however, the BT^6 term must saturate. Such an effect is observed in the Sabisky-Anderson experiment.[9] We believe it is worthwhile to consider this question in some detail.

From previous considerations it is clear that our simple golden-rule calculation (a type of first Born scattering calculation) gives the result that the ratio of the flux of transmitted energy (via the interfacial

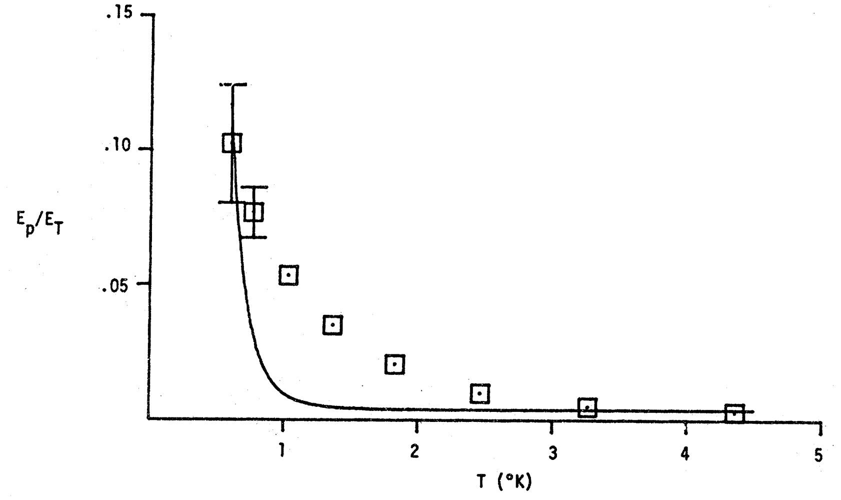

FIG. 5. Dimensionless ratio E_p/E_t in Eq. (4.28) vs temperature T. The solid curve represents the values predicted by the theory. The data points (⊡) are those given by Wyatt *et al.* (Ref. 11), where the error bars are shown when they are larger than the symbol.

three-phonon process) to incident energy has the form $D\omega^6$, where ω is the incident frequency. For large ω, $D\omega^6$ exceeds unity, which is unphysical. A self-consistent theory would replace $D\omega^6$ by a quantity which does not exceed unity. In the context of trying to fit E_P/E_T, we must replace BT^6+1 by a quantity which does not exceed the ratio of the total incident energy to the energy transmitted by direct processes. For example, we may let

$$BT^6+1 \rightarrow BT^6/(1+FT^6)+1 ,$$

which approaches $B/F+1$ for large T. This has the correct physical behavior, but varies too rapidly with temperature, when compared with experiment. Until the question of how the process actually approaches saturation is answered, it will not be possible to fit certain aspects of the data. Note that a related saturation problem (for one-phonon processes via the transfer Hamiltonian method) has been studied by Toombs and Bowley.[19,20] They were able to solve for their T-matrix exactly, because of the simple form of their interaction, involving only two-phonon processes. However, the T-matrix equation for the present problem, involving three-phonon processes, can be expected to be much more difficult to solve, being an intrinsically many-body problem.

Note that, because of the saturation effect, the form of Eq. (4.27) can be expected to hold only near the onset of interfacial three-phonon processes. Unfortunately, this involves the lowest pulse temperatures, for which good data was difficult to obtain. (See Fig. 2 of Ref. 11, where the error bars are largest at the lowest pulse temperatures.)

So far we have only discussed the NaF → ^{4}He data. We now consider the ^{4}He → NaF data, whose most prominent energy-transfering interfacial three-phonon process (Channel *II*) involves a ^{4}He phonon breaking up into a ^{4}He and a NaF phonon. In this case, not all of the incident phonon energy gets across, so the transmission E_T is less, and therefore E_P/E_T is larger. Indeed, it seems to saturate at a value near 0.13, as opposed to a value on the order of 0.005 (or less) in the NaF → ^{4}He case. The reason E_P/E_T is not so small in this case, despite the ω^6 dependence of the interfacial three-phonon process, may be due to the competition of the more dominant interfacial three-phonon process involving phonons from the liquid only ($L \rightarrow L+L$). This process was discussed briefly in the previous section.

We close this section with an estimate of the ratio of the rate for interfacial four-phonon processes to the rate for interfacial three-phonon processes. If a matrix element of the form

$$C^{(4)}(\partial_z u_z)^4\delta(z)$$

is taken, then $C^{(4)}$ has the same units (energy/area) as the C we previously consider for interfacial three-phonon processes. It is probable that $C^{(4)}$ and C are of the same order so that the relative rates of the two processes are in the ratio

$$r=(\partial_z u_z)^2\left[\frac{V}{(2\pi)^3}\left(\tfrac{1}{3}2\pi l^3\right)\right]$$

where the last factor is the maximum phase available to the fourth phonon $\vec{l}$, which can lie in a hemisphere of radius l. We make l as large as possible by considering it to propagate in the liquid, so $l=\omega_l/c$. The characteristic value of ω_l is $\frac{1}{3}\omega$, where ω is the incident phonon frequency (which must split up its energy into three final phonons). Replacing $\partial_z u_z$ by an upper limit, $l|\vec{u}_l|$, where $\vec{u}$ is given by Eq. (4.1), we obtain

$$r \approx \left(l^2 \frac{h}{2\rho V \omega_l} \right) \frac{V}{2\pi)^3} \frac{1}{3} 2\pi l^3$$

$$= \frac{\hbar \omega_l^4}{24\pi^2 \rho c^5} \ .$$

For liquid ^{4}He we may take $\rho \approx 0.15\ \mathrm{cm}^{-3}$ and $c \approx 2.4 \times 10^4$ cm/sec. If we consider $\omega_l = 5 \times 10^{11}\ \mathrm{sec}^{-1}$ so

$$f = (1/2\pi) 3\omega_l \approx 2.5 \times 10^{11}\ \mathrm{Hz} \ ,$$

then we obtain

$$r \approx 2 \times 10^{-4} \ .$$

This implies that four-phonon processes are relatively negligible at the frequencies and temperatures of interest for the Kapitza problem.

V. DISCUSSION

In considering the problem of the anomalous Kapitza conductance, which has eluded solution for so many years, one must take a somewhat different attitude toward theory than in the case of many other problems. Rather than subjecting the theory to rigid quantitative tests in a few selected experiments, it is here more important to see first if the theory is capable of satisfying a broad range of criteria specified by the experimental data. Indeed, since there is no universal quantitative characterization of this phenomenon, it is not clear that any particular quantitative fit would be meaningful. We therefore summarize our qualitative results.

We have seen in the above that the mechanism of interfacial three-phonon processes is capable of explaining the qualitative aspects of Wyatt *et al.*[11] Furthermore, our estimate of the interaction strength is consistent with the mechanism being important only in the case of interfaces between classical and quantum materials. In addition, Channels *IV* and *VI* are consistent with the mode conversion reported in Ref. 5. Detailed analysis of this question is worthy of further investigation.

Another satisfactory aspect of the mechanism is that it becomes important only in the case of high-frequency phonons; this agrees with the measurements of Sabisky and Anderson.[9] Furthermore, the interfacial three-phonon process mechanism increases so rapidly with frequency that it must rapidly saturate. This also is in agreement with the work of Ref. 9.

We should repeat the very important point that, being a process mediated by the interaction at the interface, it should be very sensitive to surface preparation, contamination, etc. This is a very strong argument in favor of a surface-mediated mechanism of any sort, and the present one is certainly in that category.

We now discuss what is perhaps the most puzzling aspect of the pulse experiments. In the work of Guo and Maris,[5] and in that of Kinder and Dietsche,[7,8] the reflectivity as a function of helium film thickness saturates for very thin film thicknesses. Specifically, in the case of Kinder and Dietsche, using 290 GHz pulses and an ambient temperature of 1 °K, saturation is nearly reached for a 12 Å thickness. If the phonon incident from the solid splits into two phonons in the liquid, we may take 145 GHz as a characteristic frequency for the phonons in the final state. This corresponds to a 16 Å wavelength. Given that most phonons will not move normal to the interface, and therefore will see an effective thickness larger than 12 Å, it is not unreasonable that nearly bulk behavior is observed at such thicknesses. It should be remarked that, in terms of the three-phonon process mechanism, the inelastic processes observed by Kinder and Dietsche certainly involve both an initial and a final state phonon in the solid, and probably a third phonon in the liquid. This is not the most efficient of the interfacial three-phonon processes, but there is probably sufficient phase space available to be consistent with the observed intensities (note that no absolute intensity measurements were made).

After our calculations were completed Weber *et al.*[12] published the results of 290 GHz pulse experiments involving LiF and NaF (100) surfaces cleaved in high vacuum at 1 °K (in the absence of a helium leak, presumably better than 10^{-12} Torr). The pulses were only 3° from normal incidence, and their reflectivity was studied, as in the related work of Kinder and Dietsche. The most novel of their results occurred in the case of the fast transverse (FT) pulse in LiF, for which the polarization $\partial_z u_\alpha$ $(\alpha = x,y)$ is completely in the surface plane; there the relative reflectivity against liquid helium as opposed to high vacuum was unity, within an accuracy of at least 1%. In terms of interfacial three-phonon processes, we interpret this to mean that a $\partial_z u_\alpha$ term is absent or nearly absent from the interfacial cubic anharmonic interaction for a clean (100) surface in LiF. Further, it must mean that ordinary ("dirty") surfaces either have a much larger anharmonicity than clean surfaces (which we are inclined to doubt), or that they are sufficiently irregular that a phonon which appears to be polarized in the surface plane is, in fact rather far from being polarized in the surface. It would be very helpful in this regard to study pulses impinging on clean surfaces at non-normal incidence. We anticipate that the FT pulse would retain its relative reflectivity of almost unity, since it would still be polarized in the surface plane, and would thus have the same weak (or absent), interfacial cubic anharmonic interaction. The FT data further means that, since the FT pulse at 3° from normal incidence has a small component of $\partial_x u_y - \partial_y u_x$, this term cannot have a large anharmonic coefficient C.

The slow transverse (ST) pulse in LiF had a 94% reflectivity. This mode is dominantly of $\partial_\alpha u_z$ polarization, with small amounts of $\partial_z u_z$ and $\partial_x u_x + \partial_y u_y$, and very small amounts of $\partial_\alpha u_z$. The non-100% reflectivity must thus be due to a large coefficient C for $\partial_z u_z$ or $\partial_x u_x + \partial_y u_y$,or to a very large coefficient C for $\partial_\alpha u_z$.

The longitudinal (L) pulse in LiF could not be seen. However, for NaF it gave a 96% reflectivity. This mode is dominantly of $\partial_z u_z$ polarization, with small amounts of $\partial_\alpha u_z$ and $\partial_z u_\alpha$, and very small amounts of $\partial_x u_x + \partial_y u_y$. The non-100% reflectivity must thus be due to a moderate coefficient for $\partial_z u_z$, or a large coefficient for $\partial_\alpha u_z$ or $\partial_z u_\alpha$, or a very large coefficient for $\partial_x u_x + \partial_y u_y$.

The data on transverse pulses in NaF, with a 94% reflectivity, seem to provide no new information.

If we combine the information obtained from each of these experiments, we conclude that $\partial_\alpha u_z$ and $\partial_x u_x + \partial_y u_y$ have the largest cubic anharmonic coefficients, that $\partial_z u_z$ and $\partial_x u_y - \partial_y u_x$ have smaller coefficients, and that $\partial_z u_\alpha$ has the smallest cubic anharmonic coefficient. This analysis can only be considered a tentative one, due to the assumptions about the long-wavelength limit and rotational symmetry about the interface, to make no mention of the fact that the experiments of Weber *et al.* were not performed with the interfacial cubic anharmonic interaction in mind.[21]

It should be noted that the angle of incidence (nearly normal to the interface) in the experiments of Weber *et al.* is very atypical of a thermal distribution, unlike the case of the phonon distribution in a typical h_k experiment. It is therfore quite possible, if the reflectivity of typical (non-normally incident) phonons is not changed much when the surface is clean, that the overall value of h_k is not much affected by the clean surface. Indeed, this appears to be the case for the work of R. C. Johnson, who measured h_k on KCl crystals cleaved in liquid ^{4}He along a (100) plane.[22] To within about 20% there was no change in h_k on going to the crystal face cleaved in the liquid.[23]

As a final piece of experimental evidence in favor of interfacial three-phonon processes, we mention the results of Wyatt and Crisp on the frequency of phonons emitted into liquid ^{4}He from NaF.[24] These authors studied the angular distribution of emitted phonons using both a broadband phonon detector (a bolometer) and a high-pass phonon detector (a tunnel junction). They found that the bolometer detected a relatively larger signal for phonons outside the critical cone than did the tunnel junction. In other words, the frequency spectrum for the phonons emitted outside the critical cone is lower than for the phonons emitted within the critical cone. This is just as one would expect if interfacial three-phonon processes are responsible for "cone breaking".

We now present additional support for a matrix element of the form (2.2), and a theoretical estimate of the expected value of the coupling strength C. During the past decade, it has been established that the first overlayer of ^{4}He on a typical solid surface (with the possible exception of some of the rare gases) is in the form of a solid. Therefore, whether the bulk ^{4}He is liquid or solid, the interfacial cubic anharmonic interaction must be that for a solid-solid interface, as in Eq. (2.2). Furthermore, because the first layer is in many ways quasiclassical (as will be discussed shortly), it is possible to estimate the coupling strength C. We find a value in the range of 0.2–2 eV/Å^2.

The evidence that the first overlayer of ^{4}He is solid has been obtained in many ways. Specific heat and vapor pressure data make it clear that there is a sharp change in behavior as the first overlayer forms and the second overlayer begins, with the specific heat of the completed first overlayer well described by a two-dimensional Debye model.[25,26] Furthermore, neutron scattering data show clear Bragg peaks, permitting the deduction that (at least on a Grafoil substrate) the first overlayer forms a triangular lattice.[27] It should also be noted that quantum-mechanical calculations show that the first overlayer of ^{4}He has an rms deviation normal to the interface of less than 0.3 Å.[28] Thus the ^{4}He atoms in the first overlayer are reasonably well-localized relative to the ^{4}He atoms in the second overlayer. Although we do not know how localized the atoms are within the first overlayer, since the Debye temperature of the first overlayer is comparable to the bulk value we may suppose that, for their motion along the surface, atoms in the first layer are about as quantum mechanical as they are in the bulk.

We thus conclude that motion normal to the surface, being relatively well localized, is probably less quantum mechanical than motion along the surface. For purposes of estimating the surface anharmonicity, we will assume that motion normal to the surface is quasiclassical. Corrections to our estimate will be a function of a deBoer parameter for the surface, Λ_s, and will be small if Λ_s is small (corresponding to the classical limit). We will not be able to obtain a reliable estimate of the surface anharmonicity associated with motion along the surface, since the associated harmonic motion of the ^{4}He atoms is so quantum mechanical. Nevertheless, we would find it surprising if it differs much from the value associated with motion normal to the surface.

To obtain the cubic surface anharmonic energy density B_s we argue as follows. This quantity, in bulk, has the form ϵ/σ^3, where ϵ and σ are the Lennard-Jones parameters. The surface anharmonicity should also be of this form, with appropriate values for ϵ and σ. Thus the ratio $(\epsilon/\sigma^3)_s/(\epsilon/\sigma^3)_b$ should give, more or less, the ratio of the surface cubic anharmonicity to the bulk cubic anharmonicity. In equation form, this means that

$$B_s \approx B_b[(\epsilon/\sigma^3)_s/(\epsilon/\sigma^3)_b] \ .$$

Bulk values of B_b run from 10^{12}to10^{13} erg/cm^3.[29] To obtain $(\epsilon/\sigma^3)_s/(\epsilon/\sigma^3)_b$ we consider a Cu-^{4}He interface. The value of σ_s, ususally taken to be the arithmetic mean of the σ's for the two types of interacting atoms,[30] does not differ much from σ_b for bulk Cu. However, the value of ϵ_s, usually taken to be the geometric mean of the ϵ's for the two types of interacting atoms,[30] is probably about $0.1\epsilon_b$ for bulk Cu. This estimate assumes that Cu can be described by an equivalent Lennard-Jones potential, whose value can be obtained via dimensional analysis from

$$\theta_D = \alpha(\epsilon/M\sigma^2)^{1/2} \ ,$$

where θ_D is Debye temperature, α is a dimensionless constant, and M is the atomic mass.[31] By comparing bulk Cu to bulk Ne (both of which form an fcc lattice) we obtain

$$(\epsilon/M\sigma^2\theta_D^2)_{\text{Cu}} = (\epsilon/M\sigma^2\theta_D^2)_{\text{Ne}} \ ,$$

thus permitting an estimate of ϵ_{Cu}, if we take $\sigma_{\text{Cu}}/\sigma_{\text{Ne}}$ to be in the ratio of their lattice constants. Specifically, we find $\sigma_{\text{Cu}} = 2.22 \times 10^{-8}$ cm, $\epsilon_{\text{Cu}} = 2120 \times 10^{-16}$ erg, so that $\sigma_s = 2.38 \times 10^{-8}$ cm, $\epsilon_s = 172 \times 10^{-16}$ erg. This yields $B_s \approx 0.1 B_b$, so that we expect B_s to be on the order of $10^{11}-10^{12}$ erg/cm^3. To convert this to the cubic anharmonic energy per unit area, we multiply by the distance across the interface, about 3×10^{-8} cm, to obtain $C \approx B_s a$ lying in the range $3 \times 10^3 - 3 \times 10^4$ erg/cm^2, or 0.2–2 eV/Å^2. This overlaps with the range of values 1–10 eV/Å^2 needed to provide a semiquantitative fit to the data of Wyatt *et al.*[11]

To summarize, we have shown that interfacial three-phonon processes are consistent with the qualitative aspects of a large number of experiments relating to the Kapitza conductance problem. In addition, the number of interfacial interactions is so great that is is plausible that the model can be made to fit the quantitative aspects of these experiments. Nevertheless, the following should be kept in mind: the calculations we have performed are valid only in the vicinity of onset, whereas the existing experiments have been performed at frequencies away from onset. Therefore no quantitative comparisons can be made until the theory is extended beyond the onset, or until experiments are performed in the vicinity of the onset, where the ω^6 dependence is predicted.

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APPENDIX

Equation (3.4) for Z_1 may be evaluated from expressions given by Gradshteyn and Ryzhik.[32] The result is

$$Z_1(\theta_q,\phi_q;\theta_k) \equiv \int_{x_l}^{x_u} \frac{x^3(1-x)\,dx}{(f_1x^2+f_2x+f_3)^{1/2}}$$

$$= [X(x)]^{1/2}[RS(x) - C(x) + DE(x)]_{x_l}^{x_u} + (RP+DQ)f_1^{1/2}(\ln 2[f_1X(x)]^{1/2} + 2f_1x + f_2)_{x_l}^{x_u} \ , \quad \text{(A1)}$$

where

$$X(x) = f_1x^2 + f_2x + f_3 \ , \quad S(x) = \frac{x^2}{3f_1} - \frac{5f_2x+8f_3}{12f_1^2} + \frac{5f_2^2}{8f_1^3} \ , \quad C(x) = \frac{x^3}{4f_1} \ , \quad E(x) = \frac{x}{2f_1} - \frac{3f_2}{4f_1^2} \ , \quad \text{(A2)}$$

and

$$R = 1 + \frac{7f_2}{8f_1} \ , \quad D = \frac{3f_3}{4f_1} \ , \quad P = \frac{f_2(12f_1f_3 - 5f_2^2)}{16f_1^3} \ , \quad Q = \frac{3f_2^2 - 4f_1f_3}{8f_1^2} \ . \quad \text{(A3)}$$

[*] Present Address: Boeing Commercial Aircraft Company, P.O. Box 3707, Seattle, Wash., 98124.

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