[25]M. Debe and D. C. Johnson, private communication.
[26]M. B. Webb, T. D. Poppendick, T. C. Ngoc, and T. Tomet, in Proceedings of the Thirty-Sixth Annual Conference on Physical Electronics, Madison, Wisconsin, 7–9 June 1976 (to be published).
[27]See, e.g., J. A. Strozier, Jr., D. W. Jepsen and F. Jona, in *Surface Physics of Materials*, edited by J. M. Blakely (Academic, New York, 1975), Vol. 1; S. Y. Tong, in *Progress in Surface Science*, edited by S. G. Davison (Pergamon, New York, 1975), Vol. 7, Part 2; M. B. Webb and M. G. Lagally, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1973), Vol. 28, p. 301.
[28]J. V. Florio and W. D. Robertson, Surf. Sci. 24, 173 (1971).
[29]H. D. Hagstrum and G. E. Becker, Phys. Rev. B 8, 1580 (1973).
[30]We are gratefull to J. M. Baker for providing the Si(111) wafers used in this study.
[31]D. W. Jepsen and P. M. Marcus, in *Computational Methods in Band Theory*, edited by P. M. Marcus, J. F. Janak, and A. R. Williams (Plenum, New York, 1971), p. 416.
[32]A. Ignatiev, F. Jona, D. W. Jepsen, and P. M. Marcus, Phys. Rev. B 11, 4780 (1975).
[33]K. C. Pandey, T. Sakurai, and H. D. Hagstrum, Phys. Rev. Lett. 35, 1728 (1975).

Macroscopic Calculation of the Kapitza Resistance between Solids and Liquid ^{4}He

David Cheeke
Département de Physique, Université de Sherbrooke, Sherbrooke, Province of Quebec, Canada

and

Henry Ettinger
Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique, 38042 Grenoble, France

(Received 26 July 1976)

We treat the Kapitza thermal boundary resistance R_K between a Debye solid and liquid ^{4}He in the temperature range 0.1 to 1 K, where the phonon transmission coefficient is known to be highly anomalous. Using a continuum model, we consider the consequences of a solid van der Waals layer of helium which is acoustically absorbing. Using values of acoustic wave attenuation in the layer based on experiment, we are able to predict the correct order of magnitude and temperature dependence of R_K for copper.

The anomalously high phonon transmission coefficient between liquid helium and solids has been a well-known problem since its discovery by Kapitza in 1941.[1] Khalatnikov[2] proposed an acoustic mismatch model which in its simplest form failed to predict the observed magnitude of the effect.[3] In this Letter we propose a more general macroscopic model based on acoustic mismatch which successfully predicts the order of magnitude of the experimental results.

The Kapitza effect has been found to exist[4,5] in both liquid and solid ^{3}He and ^{4}He and even solid H_2 and D_2 and it is now generally believed to be characteristic of quantum systems. For simplicity and brevity we restrict our discussion here to the "classic" problem of the steady-state thermal boundary resistance R_K for phonon transport between an isotropic ordinary Debye solid in contact with a bath of liquid ^{4}He in the temperature range 0.1 to 2 K. In this case classic acoustic theory predicts the existence of a small critical cone for total reflection in the helium as a result of the large difference in acoustic properties of the two media. The small transmission coefficient for incidence within this cone explains the high values of R_K predicted by the Khalatnikov theory.

A first attempt to provide a mechanism for an increased phonon transmission coefficient was made by Challis, Dransfeld, and Wilks (CDW),[6] who took into account the formation of a dense layer of helium at the interface due to the van der Waals attraction of helium atoms to the substrate. This layer acts as an acoustic matching unit which leads to an increased transmission coefficient within the critical cone; this is, however, quite insufficient in itself to resolve the discrepancy. More recently, other workers[7,8] have considered the effects of a finite phonon lifetime in the substrate, which led to an effective widening of the critical cone. The two mechanisms have been combined in a single calculation by Opsal and Pollack.[9] However a large body of evidence[10] suggests that substrate lifetime effects cannot provide the sole basis for the correct explanation for $T \gtrsim 1$ K and we do not consider them further here.

The present model uses the existence of a

dense layer as its starting point. For simplicity we consider a model composed of two solid layers each 3-Å thick at pressures of 30 and 260 atm; values of density and longitudinal and transverse sound velocities for each layer were obtained by using experimental results for bulk solid at these pressures.[11] As did previous workers,[6,9,12] we use a continuum model in that the van der Waals layer is chosen as an elastic continuum across which plane waves propagate from one medium to the other,[13] mostly in the limit in which the wavelength is larger than the layer thickness.

The new and crucial point is that in the present work we consider *explicitly* that the van der Waals layer is an acoustically absorbing medium. We can obtain an estimate of the attenuation constant for 1-K thermal phonons in such a layer from the work of Anderson and Sabisky[14,12]; for slightly thicker films this gives a mean free path $l \sim 100$ Å at a frequency of about 60 GHz varying approximately linearly with wavelength. The attenuation is expected to be even greater for the thin inhomogeneous layer considered here even though it is solid. In any case, we treat the absorption constant ν as a free parameter in the calculation, of the order of the above value ($\nu \sim 0.2$ for an average wave in our film).[15]

The actual calculation (both of the acoustic properties of the multilayered system and of the heat flow) follows very closely the now standard formalism for such problems which has been presented in great detail by Opsal and Pollack.[9] The boundary conditions lead to a secular expression which is solved by computer to determine the wave amplitudes in the incident and substrate media for a given incident wave in the liquid helium at angle θ. The corresponding energy fluxes are determined in the usual way, and for the simple and known case of no absorption in the film we verify conservation of energy at angle θ, as well as verifying an enhanced transmission with respect to Khalatnikov of at most 3 or 4, as obtained by previous workers.[6,9] We now include absorption in the film by expressing the phonon wave vector as $k = k' + ik''$ in the van der Waals layer, with $k'' = \nu k'$. Wave amplitudes are again calculated by computer and, following the astuce of Peterson and Anderson[7] in a similar problem, we obtain by subtraction the energy flux in the layer for incidence at angle θ. Integration over θ and ω then gives the total heat flux absorbed in the layer; and the crucial question now is to determine what fraction β of this heat flux is subsequently transmitted into the substrate. As the calculation is macroscopic we are unable to treat questions related to the frequency spectrum of the re-emitted phonons in the layer (e.g., the possibility of frequency conversion, etc.).

The determination of β is based entirely upon acoustic mismatch.[16] At each interface we determine an average transmission and reflection coefficient deduced from the calculated R_K for an interface between the corresponding semi-infinite media.[3,7] We then consider that there are incoherent multiple reflections in the film, a hypothesis that has been used successfully in similar problems of transmission of thermal phonons across thin foils or sandwiches.[17,7] This argument leads to the expression

$$\beta = \tfrac{1}{2} t_2 (1 + r_1)/1 - r_1 r_2),$$

where r_1 and t_1 are average reflection and transmission coefficients at the layer-to-liquid interface and r_2 and t_2 the corresponding entities at the layer-to-substrate interface. β is typically of the order of 10^{-3} in the cases of interest here,[18] but this is quite sufficient to obtain agreement with experiment as most of the energy flux from the liquid comes from outside the critical cone.

By adding the above heat flux $\dot{Q}_2$ to that $\dot{Q}_1$ transmitted directly inside the cone we thus determine $R_K T^3$ for each value of T. The results are presented in the Fig. 1 for the case of copper and compared with a representative selection of available experimental values.[19,20] It is clear that the basic Khalatnikov model as well as that with a dense layer with $\nu = 0$ are quite incapable of explaining the results, while the present model with ν in the range 0.1 to 0.5 reproduces the correct general magnitude and temperature dependence. For $\nu = 0.5$ and $T = 1$ K, we have $\dot{Q}_2/\dot{Q}_1 \sim 30$ which illustrates the predominance of the present mechanism. For materials other than copper, a representative comparison is made in Table I where we see that aluminum[17] and quartz[6] can be satisfactorily explained while the discrepancy is rather bigger and perhaps significant for silicon.[21] Disagreement by a factor of 2 to 4 seems rather typical for such high-θ_D materials (e.g. Ni and Al_2O_3). Whether or not this disagreement is significant will depend upon the outcome of a more elaborate treatment of our model, and upon detailed calculations on possible competing parallel mechanisms.

It is easy to see the physical significance of the additional heat flow calculated in the present

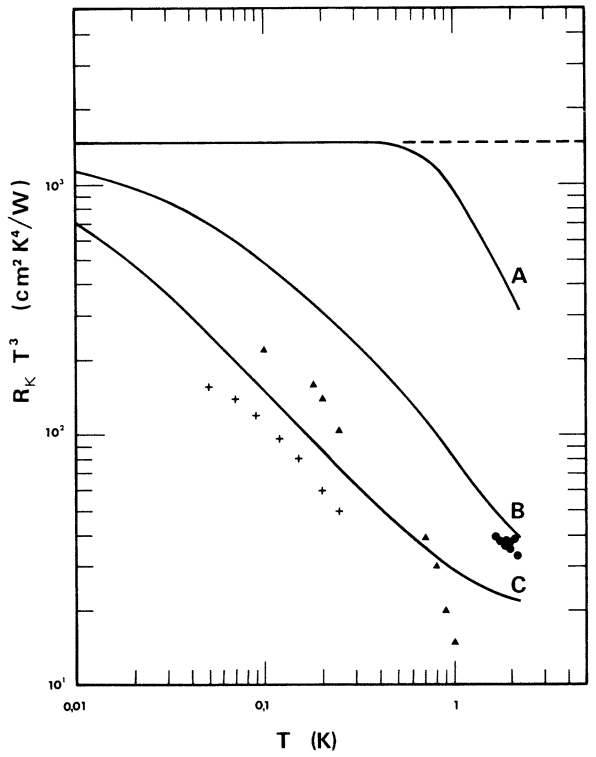

FIG. 1. Calculated values of $R_K T^3$ vs T compared with experimental results for copper: ---, Khalatnikov (term F_1) from Ref. 2; curve A, dense layer with $\nu=0$; curve B, dense layer with $\nu=0.1$; curve C, dense layer with $\nu=0.5$; ●, experiment (Ref. 20); ▲, experiment (Ref. 19); +, experiment (Ref. 19).

model. The presence of absorption (imaginary k vector) in the layer relaxes Snell's law, permitting absorption of the incident energy flux that enters the layer by direct transmission. A consequence is that, for $\nu>0.1$, we calculate a transmission coefficient approximately independent of angle. This agrees with an empirical suggestion by Challis[3] concerning the dependence of the transmission coefficient on the relative densities and sound velocities. Physically it corresponds to an "opening out" of the critical cone to most of the incident half space.

In conclusion, the simple macroscopic model presented here seems capable of predicting the rough temperature dependence and the order of magnitude for R_K of Cu–liquid ^{4}He. This model is consistent with the idea of an anomalous transmission for quantum systems, for which the high compressibility leads to the formation of a dense layer; the high attenuation in the layer may also have a characteristic quantum origin, but this is beyond the scope of the present analysis. The model does not explain *per se* the Kapitza anomaly, but for the first time it does indicate the lines along which a successful microscopic calculation could be carried out.

TABLE I. Values of $R_K T^3$ calculated for aluminum, quartz, and silicon at $T=1.5$ K compared to experiment in watt-centimeter units.

	Al	SiO_2	Si
Khalatnikov (F_1) [a]	1060	1900	3880
Solid layer with [b]			
$\nu=0$	460	788	1490
Our theory with			
$\nu=0.1$	42.3	72	129
$\nu=0.5$	18.2	31	58
Experiment	~20 [c]	~20 [d]	~24 [e]

[a] Ref. 2, and L. J. Challis and J. D. N. Cheeke, Proc. Roy. Soc. London, Ser. A 304, 479 (1968).
[b] We include both longitudinal and transverse waves in the layer.
[c] Ref. 17.
[d] Ref. 6
[e] Ref. 21.

We thank V. Bortolani, L.-G. Caron, H. J. Maris, and L. P. Kadanoff for comments and discussions, as well as A. C. Anderson for communication of unpublished results. This work was partially supported by the GRSD (Groupe de Recherches sur les semiconducteurs et diélectriques).

[1] P. L. Kapitza, J. Phys. (Moscow) 4, 181 (1941).
[2] I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. 22, 687 (1952).
[3] L. J. Challis, J. Phys. C 7, 481 (1974).
[4] J. S. Buechner and H. J. Maris, Phys. Rev. Lett. 34, 316 (1975).
[5] C. L. Reynolds and A. C. Anderson, Phys. Rev. B 14, 4114 (1976).
[6] L. J. Challis, K. Dransfeld, and J. Wilks, Proc. Roy. Soc. London, Ser. A 260, 31 (1961).
[7] R. E. Peterson and A. C. Anderson, J. Low Temp. Phys. 11, 639 (1973).
[8] H. Haug and K. Weiss, Phys. Lett. A40, 19 (1972).
[9] J. L. Opsal and G. L. Pollack, Phys. Rev. A 9, 2227 (1974).
[10] To fit the sapphire–liquid ^{4}He data with a bulk solid attenuation model requires an attenuation constant ν ~0.5. The same value used for an indium-sapphire interface would predict an R_K about 12 times less than that observed, this latter value being in good agreement with the simple acoustic-mismatch model [J. D. N. Cheeke, B. Hébral, and C. Martinon, J. Phys. (Paris) 34, 257 (1973), and D. A. Neeper and J. R. Dillinger, Phys. Rev. 135, A1028 (1964)]. This suggests that at thermal frequencies higher than 1 K,

attenuation, roughness, etc., in the solid surface make, at most, secondary contributions. The experimental situation is similar for metals [J. D. N. Cheeke, B. Hébral, and J. Richard, J. Low Temp. Phys. 12, 359 (1973)].

[11]We use for layer 2, $\rho = 0.258$ g/cm^3, $C_l = 0.96$ km sec^{-1}, and $C_t = 0.48$ km sec^{-1}; for layer 1, $\rho = 0.195$ g/cm^3, $C_l = 0.5$ km sec^{-1}, $C_t = 0.25$ km sec^{-1}. The results are not critically dependent upon the choice of these values, which has been made as realistically as possible; full details of the determination of such constants and of their relative importance is given in Ref. 9. The model used here corresponds to the currently accepted value of *about* 1.6 statistical atomic layers of solid helium.

[12]A. F. G. Wyatt, N. A. Lockerbie, N. G. Mills, and R. A. Sherlock, Solid State Commun. 11, 1089 (1972).

[13]It has been shown quite conclusively [J. G. Dash, in *Critical Reviews in Solid State Sciences* (Chemical Rubber Company, Cleveland, Oh., 1976), p. 209] absorbed helium can be described surprisingly well as an elastic two-dimensional Debye continuum insofar as its low-temperature acoustic and thermal properties are concerned.

[14]C. H. Anderson and E. S. Sabisky, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1971), Vol. 8.

[15]Even supposing the simple k dependence of the absorption term assumed here, ν will generally be a temperature- and thickness-dependent parameter, which could of course be included in more sophisticated versions of the present model.

[16]Previous workers [W. Dietsche and H. Kinder, J. Low Temp. Phys. 23, 27 (1976)] have discussed the possibility of a similar model to explain phonon reflection experiments. However these authors ruled out an explanation based upon the use of acoustic mismatch. In the present work we show that the classic Kapitza problem can be explained by such a model using acoustic mismatch alone.

[17]L. J. Challis and R. A. Sherlock, J. Phys. C 3, 1193 (1970).

[18]For the copper-^{4}He interface, the numerical values and methods stated in the text lead to $r_1 = 0.6$, $t_1 = 0.4$, $r_2 \sim 1$, and $t_2 = 1.5 \times 10^{-3}$.

[19]J. T. Folinsbee and A. C. Anderson, Phys. Rev. Lett. 31, 1580 (1973), and J. Low Temp. Phys. 17, 409 (1974).

[20]N. S. Snyder, J. Low Temp. Phys. 22, 257 (1976). These measurements were made on what were probably the most carefully controlled R_K surfaces ever studied.

[21]R. C. Johnson and W. A. Little, Phys. Rev. 130, 596 (1963).

Stability of Spin-Aligned Hydrogen at Low Temperatures and High Magnetic Fields: New Field-Dependent Scattering Resonances and Predissociations*

William C. Stwalley
Department of Chemistry, University of Iowa, Iowa City, Iowa 52242
(Received 12 October 1976)

Spin-aligned hydrogen (H↑) and deuterium (D↑) are generally predicted to be stable for $\mathcal{H}/T \gtrsim 10^6$ G/K. Magnetic-field-dependent resonances (inverse to field-induced predissociations of high vibrational-rotational levels of HD and D_2) provide exceptions to this general rule. Their existence suggests 50 kG (H↑ + D↑) and 19 kG (D↑ + D↑) should be avoided for stable H↑ and D↑. Induced predissociation observations should yield ultraprecise resonance information and dissociation limits.

Spin-aligned hydrogen (H↑) and deuterium (D↑) are extremely interesting but heretofore hypothetical substances with all electronic spin projections (M_s) parallel. Here only the lowest energy M_s is considered (e.g., $M_s = -\frac{1}{2}$ for H↑). H↑ and D↑ are especially simple, and complete understanding of all phenomena should be possible. For example, H↑ is predicted[1-3] to remain a gas down to absolute zero temperature (for pressures $\lesssim 50$ atm where it should solidify[2]) and to show strong quantum behavior[2] characteristic of a nearly ideal Bose gas (including Bose-Einstein condensation and superfluidity). D↑ is predicted[2] to be on the verge of liquidity at absolute zero temperature and low pressures (compared to a solidification pressure of ~ 8 atm), and unique liquid-gas equilibria of this highly quantal Fermi fluid should occur. T↑ is predicted[2] to be very similar to that of ^{4}He. The solid phases[4,5] and magnetic properties of these ferromagnetic substances should also be of major interest. The energy content per gram of H↑ is higher than any known substance; hence, it is possibly of interest for rocket propulsion[1,6-10] (which is the original motivation[6,7]).

It has been suggested[1,2,6,9] that H↑ and D↑ would be stable under low-temperature ($T \lesssim 1$ K), high-magnetic-field ($\mathcal{H} \gtrsim 50$ kG) conditions. Experiments under roughly these conditions on D/D_2 mixtures[8] and H/H_2 mixtures[9] suggest marginal stability for partial electronic spin alignment. The primary purpose here is to argue that long-