would consist of measuring the thermal conductivity over a sufficiently wide temperature range that regions both above and below the break are covered. If a reasonable estimate of the elastic properties of the colloids can be made, and if the total amount of material incorporated in them is known, the fit of a calculated curve to the data is very sensitive to the mean size. In fact, at temperatures below the break such that Rayleigh scattering is dominant, the cross section varies as the radius to the sixth power.

V. SUMMARY

The purpose of this investigation has been to study the scattering of phonons by an elastic sphere through its effect on the thermal conductivity. In particular, the effect of a transition from a Rayleigh to a geometrical scattering law was required. In general, the following effects have been observed:

(i) Geometrical scattering is never truly operative.

(ii) Although the resonances in the transition region can never be observed through their effect on the conductivity, there is a broad maximum which is important.

(iii) The fit of the experimental data is extremely sensitive to the assumed mean particle radius. It is less sensitive to the assumed properties of the particle. Thus, if it is possible to make a reasonable estimate of the elastic properties of inclusion in crystals, it is possible to use the thermal conductivity as a means of measuring their mean size.

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Scattering of Phonons by a Square-Well Potential and the Effect of Colloids on the Thermal Conductivity. II. Theoretical*

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The scattering cross section of an elastic sphere as a function of frequency has been obtained for a number of representative combinations of the properties of the sphere and the matrix. These are obtained from a partial-wave analysis and are intended to be used primarily in the computation of thermal conductivity. To simplify the problem, it has been assumed that both the sphere and the matrix are isotropic and that they obey the Cauchy relations. From the exact results, suitable procedures for obtaining analytic approximations are discussed.

I. INTRODUCTION

It is the purpose of this paper to investigate the effect of the elastic properties of an elastic sphere on its scattering cross section for an incident longitudinal acoustic wave. These results are intended to be used primarily in understanding the effects of a macroscopic scattering center on the thermal conductivity.

Although it is possible to calculate exact numerical values of scattering cross sections using a partial-wave analysis, this is a cumbersome procedure, and it is desirable to find sufficiently general analytic approximations. Our objective will be to search for those approximations which would be most useful in calculating the thermal conductivity. In this respect our task is simplified because an integral over the phonon spectrum is involved.

An obvious approximation would be to assume a Rayleigh law, calculated using Born approximation for long wavelengths. For short wavelengths a constant cross section would be used. The transition from one to the other would occur when the incident wave vector equalled the radius of the sphere. The transport cross section as defined below is of interest here, so the high-frequency limiting cross section will be the geometrical cross section of the sphere.

Our objective has been to compare the complete numerical solutions with the above approximation, and to investigate the possibility of broadening it to include those cases where it proves inadequate. It is obvious at the outset that it will be impossible to adequately represent the fluctuations in the cross section which usually occur in the transition region from a Rayleigh to a constant scattering law. However, the thermal conductivity involves an average over the phonon spectrum, and it is insensitive to this fine structure. In this case, where these results should be most applicable, we may neglect the fine structure.

The problem of scattering of a plane longitudinal wave by an isotropic elastic sphere in an isotropic solid

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has been treated by many authors.[1–4] The scattering of a transverse wave has also been considered.[3] For the longitudinal case some numerical computations are available[4]; however, they refer to the total cross section without the correction factor which yields the transport cross section. Our results will also be for the scattering of a longitudinal wave alone. For the computer we have programmed Pao and Mow's[1] statement of the problem. This is derived from, and is essentially identical to, Ying and Truell's[2] original formulation. We have obtained results over a wide range of elastic properties for a fictitious solid which is both isotropic and obeys the Cauchy relations.

In the following, the scattered waves in the matrix are represented by two potential functions in a well-known expansion,

$$\Phi=\sum_{n=0}^{\infty} A_n h_n(\alpha_1 r)P_n(\cos\theta)\,,$$
$$\Psi=\sum_{n=0}^{\infty} B_n h_n(\beta_1 r)P_n(\cos\theta)\,, \tag{1}$$

where α_1 is a longitudinal-wave vector, β_1 is a transverse-wave vector, h_n is the spherical Hankel function of the first kind, A_n and B_n are constants, and P_n is the Legendre polynomial. Similar expressions apply for the functions inside the elastic sphere. These, of course, are required to solve the problem.

The expression for the total cross section obtained by Ying and Truell[2] is then

$$2\pi\int_0^{\pi} d\sigma\,\sin\theta d\theta=4\pi\sum_n \frac{1}{2n+1}\times\left[\frac{1}{\alpha_1^2}|A_n|^2+\frac{n(n+1)}{\alpha_1\beta_1}|B_n|^2\right], \tag{2}$$

where $d\sigma$ is the differential cross section, and the integration over ϕ is unnecessary because of the symmetry of the problem. We observe, parenthetically, that this is no longer true for an incident transverse wave; here the scattered wave will depend on ϕ oven though the scattering potential is spherically symmetric. This, of course, is simply a consequence of the vectorial nature of the displacement field.

For our problem we require the transport cross section defined as

$$\sigma_T=2\pi\int d\sigma(1-\cos\theta)\,\sin\theta d\theta\,, \tag{3}$$

where $d\sigma$ is the differential cross section. The differential cross section is obtained from the energy flux, which in turn depends on the stresses and displacements. The procedure used is outlined by Ying and Truell. The integration involved in Eq. (3) may be performed using the properties of the Legendre polynomials and the result is that

$$\begin{aligned}\sigma_T=2\pi\sum_n \Bigg\{&\frac{2}{(2n+1)\alpha_1^2}\\ &\times\left[|A_n|^2-\frac{n+1}{2n+3}\left(\frac{A_nA_{n+1}^*+A_n^*A_{n+1}}{2}\right)-\frac{n}{2n-1}\left(\frac{A_nA_{n-1}^*+A_n^*A_{n-1}}{2}\right)\right]+\frac{2n(n+1)}{\alpha_1\beta_1(2n+1)}\\ &\times\left[|B_n|^2-\frac{n+2}{2n+3}\left(\frac{B_nB_{n+1}^*+B_n^*B_{n+1}}{2}\right)-\frac{n-1}{2n-1}\left(\frac{B_nB_{n-1}^*+B_n^*B_{n-1}}{2}\right)\right]\Bigg\}\,.\end{aligned} \tag{4}$$

II. COMPUTER PROGRAM

The cross-section calculations were programmed in FORTRAN for the ORNL 1604 computer. Equation (4) expresses the cross section as a sum of terms which are functions of A_n and B_n, the scattered-wave amplitudes. With the boundary conditions for an elastic inclusion, A_n and B_n may be found by solving a set of four simultaneous equations. The coefficients of these equations are complex numbers. The equations and their coefficients are given by Pao and Mow.[1]

The program solves these equations by elimination and back solution in complex arithmetic form. The cross sections were summed until the terms became negligible as determined by the following conditions: (a) the term index, n, exceeded by at least 5 the largest argument of the corresponding spherical Hankel or Bessel functions in the coefficient formulas; (b) the magnitude of the cutoff term was <0.0001 times the sum of all preceding terms; (c) the magnitude of the cutoff term was less than that of the preceding term.

A subroutine[5] written by Hagin was used to compute spherical Bessel functions from which in turn the Hankel functions were computed. An extensive check of this subroutine was made and, after a slight modification necessary for the zero-order functions, a comparison with the British Association Tables showed the routine to be accurate to at least six decimals for arguments <100.0, and for orders 0–100 the cross sections always reached cutoff far in advance of the accuracy-check limits.

[1] Y. H. Pao and C. C. Mow, J. Appl. Phys. **34**, 493 (1963).
[2] C. F. Ying and R. Truell, J. Appl. Phys. **27**, 1086 (1956).
[3] N. G. Einspruch *et al.*, J. Appl. Phys. **31**, 806 (1960).
[4] G. Johnson and R. Truell, J. Appl. Phys. **36**, 3466 (1965).

[5] Available from CDC 1604 user organization as COOP L3 UCSD BFFGH, Z. G. Hagin, Texas Instruments, Inc. The subroutine is based on the method described by Corbato and Uretsky, J. Assoc. Comput. Machines **6**, 366 (1959).

III. RESULTS

The results presented here are for a fictitious solid which is both isotropic and obeys the Cauchy relations. The Cauchy relations lead to a value for the longitudinal velocity of sound which is $\sqrt{3}$ times the transverse. We do not feel that the assumption of the Cauchy relations affects the conclusions we will draw. The results are displayed as plots of $\log\sigma_T$ against $\log x$ where x is the product of the wave number and the radius of the particle. Since we are concerned with a continuum approximation the actual values are unimportant. In our discussion it is convenient to consider first the situation where the properties of the inclusion are very different from those of the matrix or the case where the

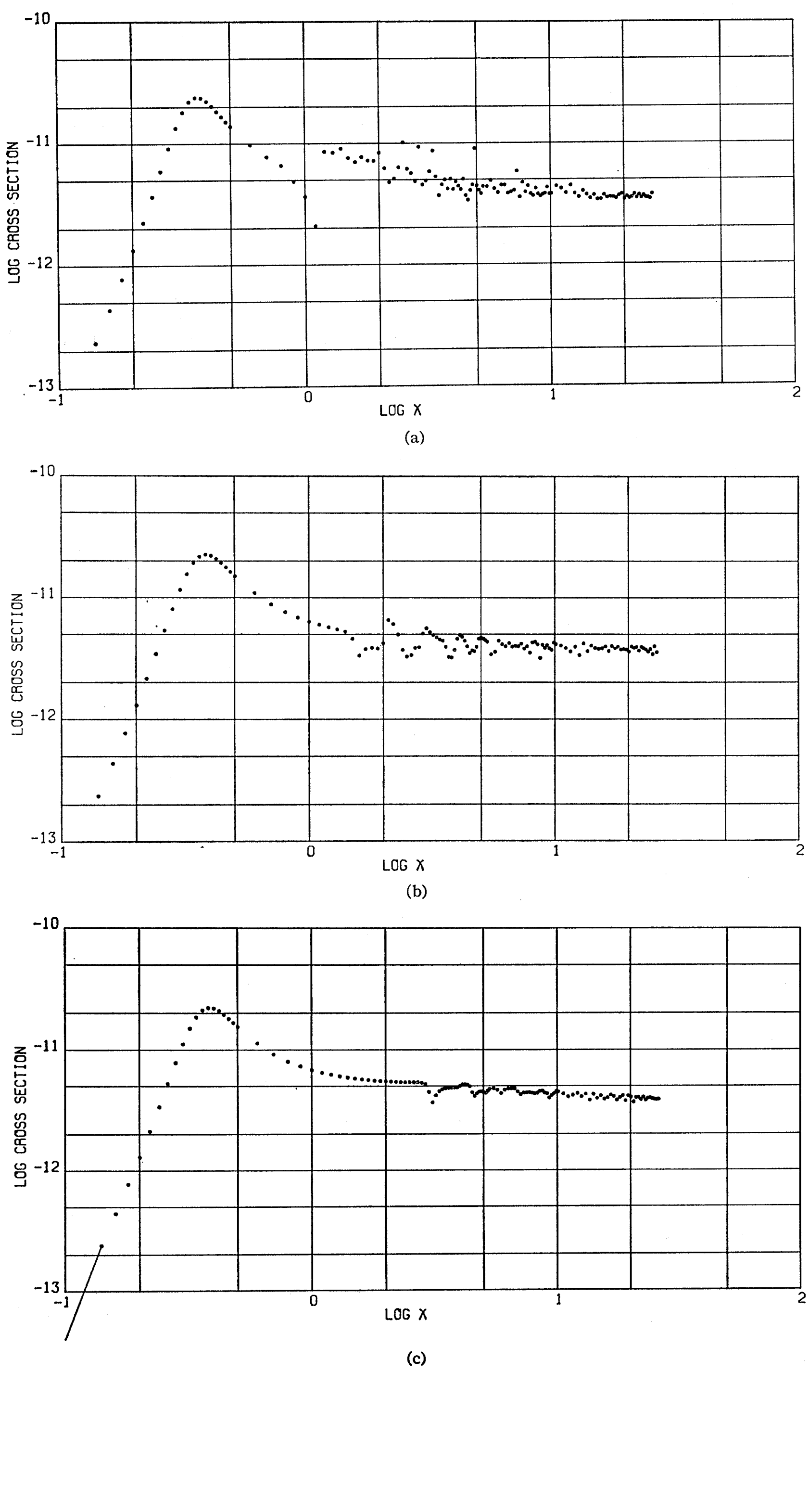

FIG. 1. The scattering cross section as a function of $x=\alpha_1 R_0$ for $\rho_{\text{particle}}/\rho_{\text{matrix}}=10$ and (a) $V_{\text{particle}}/V_{\text{matrix}}=2$; (b) $V_{\text{particle}}/V_{\text{matrix}}=1$; (c) $V_{\text{particle}}/V_{\text{matrix}}=\frac{1}{2}$.

scattering is strong, and then to consider the weak-scattering case where the inclusion and matrix have similar properties.

A. Strong Scattering

Figures 1 and 2 show a series of log-log plots of the scattering cross section against x. In each series the ratio of the density of the matrix to that of the particle is held constant. Figure 1 is a series where the density of the particle is ten times that of the matrix. In Figure 1(a) the ratio of the velocity of sound in the particle to that of the matrix is 2, in 1(b) it is 1, and in 1(c) it is $\frac{1}{2}$. Figure 2 presents a similar series where the density of the particle is $\frac{1}{10}$ that of the matrix.

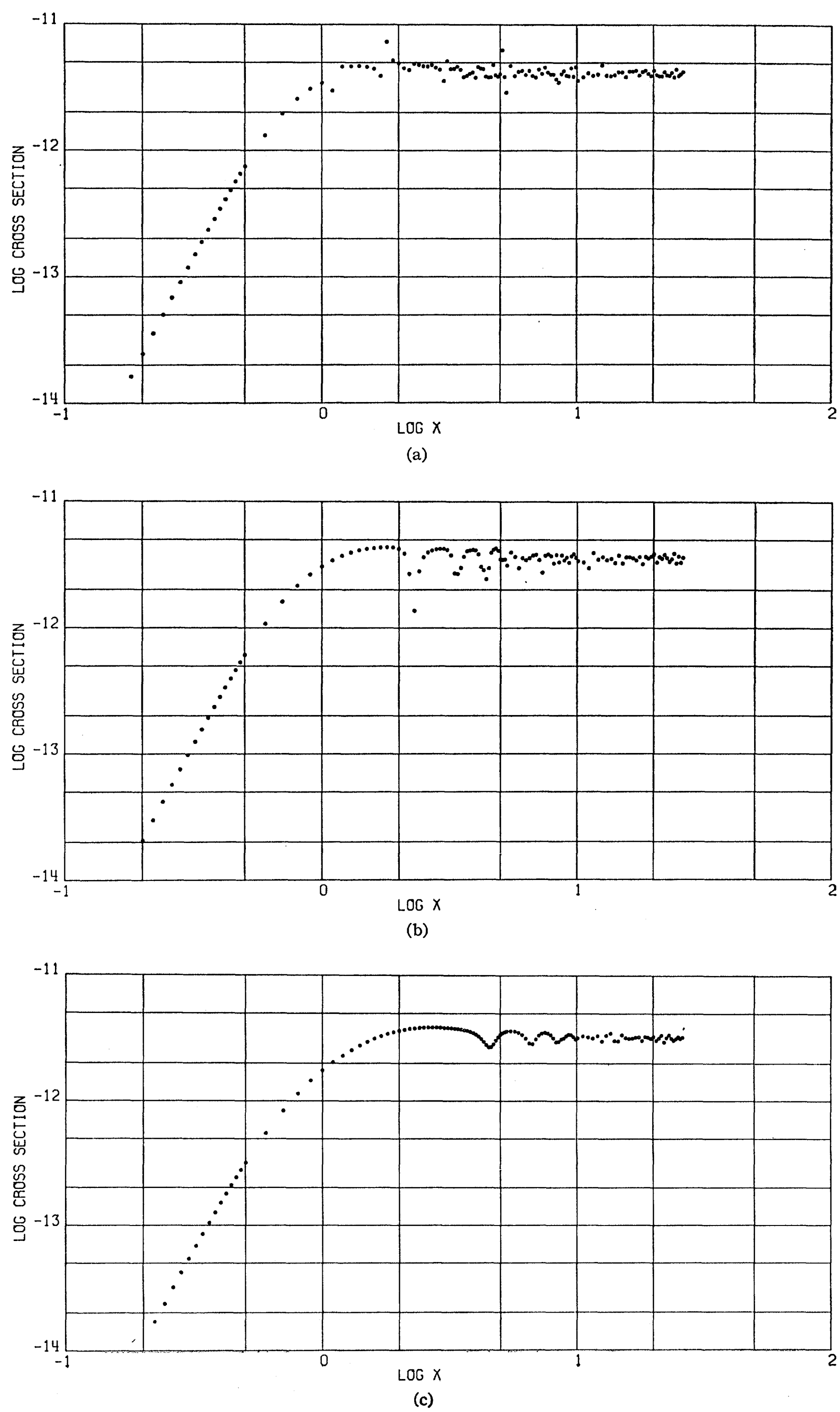

FIG. 2. The scattering cross section as a function of $x=\alpha_1 R_0$ for $\rho_{\text{particle}}/\rho_{\text{matrix}}=\frac{1}{10}$ and (a) $V_{\text{particle}}/V_{\text{matrix}}=2$; (b) $V_{\text{particle}}/V_{\text{matrix}}=1$; (c) $V_{\text{particle}}/V_{\text{matrix}}=\frac{1}{2}$.

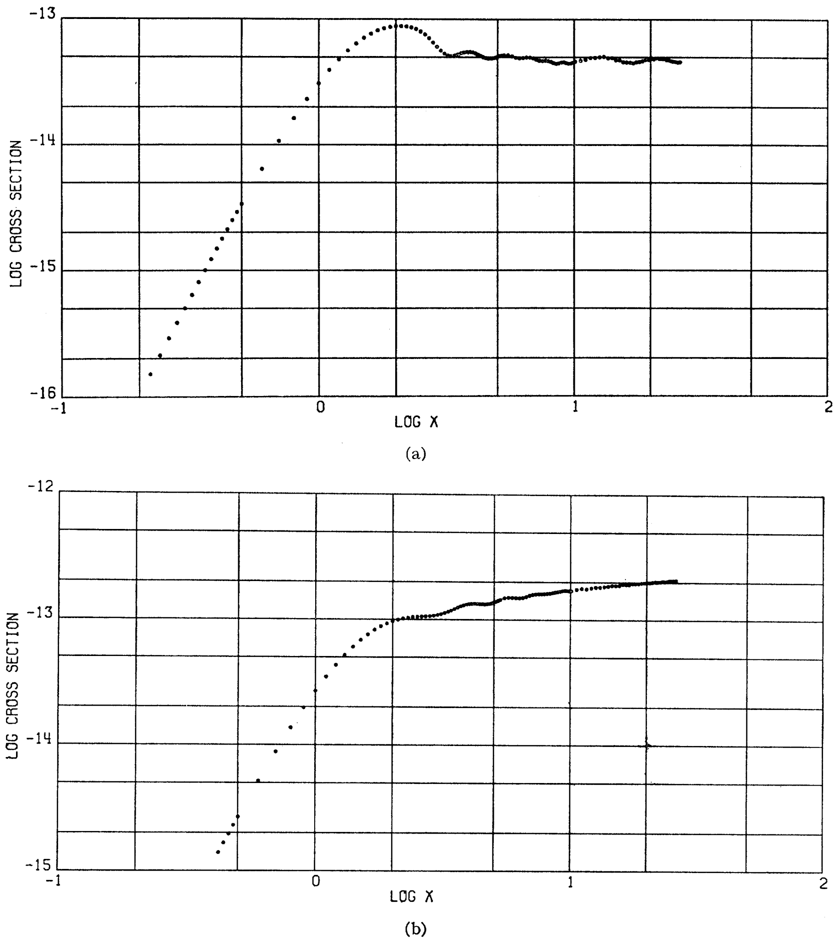

FIG. 3. The scattering cross section as a function of $x=\alpha_1 R_0$ for (a) $\delta\mu/\mu=-0.2$; (b) $\delta\alpha_1/\alpha_1=-0.1$.

B. Weak Scattering

Here we have abandoned the assumption of the Cauchy relations because the meaningful parameters are the fractional changes in the elastic constants. Figure 3(a) shows the results for a 20% change in μ, and Fig. 3(b) refers to a 10% change in the longitudinal velocity of sound. The reason for these two choices will become apparent in the discussion.

IV. DISCUSSION

A. Strong Scattering

1. Density of Scattering Center Greater than That of the Matrix

A comparison of the results shown in Fig. 1 shows a remarkable similarity if the fluctuations in cross section which occur in the transition region are ignored. This similarity clearly indicates that the difference in density between the particle and the matrix is mainly responsible for the scattering. In addition there is a large peak in the cross section at $x=0.36$ which is broad enough that it must be taken into account. Since the peak occurs at a value of x which is less than 1, it is possible to obtain an approximate analytic expression for the cross section which reproduces this maximum. Ludwig[6] has performed this calculation for the case where the density difference is large and has obtained the following expression for the resonant frequency:

$$\omega_R^2=\frac{6}{(\delta\rho/\rho)R_0^2}\left(\frac{2}{C_t^2}+\frac{1}{C_l^2}\right)^{-1} \tag{5}$$

where ρ is the density, R_0 is the radius of the particle, and C_l and C_t are the longitudinal and transverse velocities of sound, respectively.

From this expression we obtain

$$x_R^2=6\left(\frac{2C_l^2}{C_t^2}+1\right)^{-1}\left(\frac{\delta\rho}{\rho}\right)^{-1}. \tag{6}$$

In our case, $C_l=\sqrt{3}C_t$, and

$$x_R^2=\frac{6}{7}\left(\frac{\delta\rho}{\rho}\right)^{-1}. \tag{7}$$

[6] W. Ludwig (to be published).

For $\delta\rho/\rho=9$, $x_R=0.31$. Ludwig's expression for the total cross section is

$$\sigma_{\mathrm{total}}=2V_0\epsilon\gamma\left(\frac{\omega}{\omega_R}\right)^4 q/\omega|1+F_R|^2,$$

where $V_0=\frac{4}{3}\pi R_0^3$, $\epsilon=\delta\rho/\rho$,

$$\gamma=\frac{2}{\epsilon R_0}\left(\frac{2}{C_t^3}+\frac{1}{C_l^3}\right)\left(\frac{2}{C_t^2}+\frac{1}{C_l^2}\right)^{-2},$$

$$F_R=\frac{\omega^2}{\omega_R^2-\omega^2-i2\gamma\omega^3/\omega^2}.$$

For our case, $R_0=10^{-6}$ cm and the cross section at resonance calculated from Ludwig's approximate expression is 3.6×10^{-12} cm^2, in good agreement with the exact value. (For these long wavelengths, of course, there is no difference between the transport cross section and the total cross section.)

We may conclude, then, that for the case where the density of the particle is much larger than that of the matrix the differences in the elastic constants may be neglected. The analytic approximation which can be used for this case, then, can consist of an approximate calculation of the long-wavelength cross section which includes the first resonance. At short wavelengths a constant cross section equal to the geometrical area of the particle can be used.

2. Density of Scattering Center Less than That of Matrix

In this case, again, the geometrical cross section can be used at short wavelengths, while at long wavelengths a Rayleigh law is operative. However, a calculation in Born approximation yields the wrong numerical value. An approximation derived from the long wavelength limit for the partial-wave analysis (due to Ying and Truell),[2] on the other hand, will work. A comparison of the two shows that Born approximation breaks down because it does not include the energy scattered into a *d* wave.

It is interesting that no evidence is found for a low-frequency resonance (values of $x<1$). Of course none would be expected due to the density difference, but none has been found even when the Lamé constants of the particle exceeded those of the matrix by a factor of 10. Reference to Fig. 1(c) also reveals no apparent evidence of a low-frequency resonance aside from that due to the density difference.

B. Weak Scattering

Here the Born approximation can be expected to work at long wavelengths and this is, in fact, the case. At short wavelengths, however, the cross section falls well below the geometrical cross section. Qualitatively, this can readily be explained by an increasing transparency of the particle to sound. It develops that an approximate expression for the cross section can also be obtained which takes this into account.

It is possible to obtain expressions for A_n and B_n valid in the high-energy limit by replacing the Bessel functions with the approximation valid for large arguments. Assuming that the properties of the matrix and the sphere do not differ greatly, it is found (after some manipulation) that

$$|A_n|\sim\left(\frac{\delta\rho}{\rho}-\frac{\delta\alpha_1}{\alpha_1}\right)|A_n{}^L|+\sin(R_0\delta\alpha_1).$$

In this expression, $A_n{}^L$ is the limiting value of the coefficient for a large difference in properties, $\delta\rho=\rho_2-\rho_1$, and $\delta\alpha_1=\alpha_2-\alpha_1$. Similarly,

$$|B_n|\sim\left(\frac{\delta\rho}{\rho}-\frac{\delta\mu}{\mu}\right)|B_n{}^L|.$$

On substitution in Eq. (4) the terms in $\sin^2(R_0\delta\alpha_1)$ cancel and the high-energy limiting value for the cross section is approximately

$$\sigma\sim\pi a^2\left\{\left|\frac{\delta\rho}{\rho}-\frac{\delta\alpha_1}{\alpha_1}\right|+\left|\frac{\delta\rho}{\rho}-\frac{\delta\mu_1}{\mu_1}\right|\right\}^2. \qquad (8)$$

V. SUMMARY

It is apparent that the approximation mentioned in the Introduction rarely works. On the other hand, it is possible to find the following analytic approximations which are valid:

(a) If the scattering is strong and due to a large difference in density, Born approximation may be used at long wavelengths provided the first resonance is included. The geometrical cross section is applicable at short wavelengths.

(b) If the scattering is strong and due principally to a difference in elastic properties, the long-wavelength approximation obtained from the partial-wave analysis should be used followed by a geometrical cross section at short wavelengths.

(c) If the scattering is weak, Born approximation is applicable at long wavelengths and the geometrical cross section multiplied by the factor in Eq. (8) is applicable at short wavelengths.

Finally, let us consider the effect of colloids on the thermal conductivity. The appropriate scattering cross section will depend most strongly on the radius of the colloid and the elastic properties are of lesser importance. It is possible to use this fact to obtain the mean size of the colloids from thermal-conductivity data. Since the cross section is so much more sensitive to the colloid radius, it should not be necessary to have a precise knowledge of their elastic properties.