

Finite Linewidths and "Forbidden" Three-Phonon Interactions

A. J. LEGGETT* AND D. TER HAAR

Clarendon Laboratory, Oxford, England

(Received 27 October 1964, revised manuscript received 10 March 1965)

We have used thermodynamic perturbation theory to study the effect of the finite energy spread of phonon excitations upon the possibility of three-phonon interactions which could not otherwise conserve energy. It is shown that a proper treatment of this effect is equivalent to a consideration of higher order processes in which the interacting phonons occur in virtual intermediate states, but that such processes cannot in general be adequately taken into account simply by assigning a lifetime to one or more of the interacting phonons. It is also shown that the higher order terms of conventional perturbation theory should be modified, whether or not the three-phonon process is energetically allowed; if it is, these modified terms should be substituted for the three-phonon contribution rather than added to it. We give a qualitative discussion of applications to liquid helium II and to dielectric solids.

1. INTRODUCTION

THE problem of the lifetime of a long-wavelength phonon due to its interaction with other phonons is of great importance in the theory of insulating solids and also in the phenomenological theory of liquid helium II. Provided the medium is sufficiently pure, this lifetime will have a dominant effect upon sound absorption, on neutron-scattering linewidths—which, indeed, measure it directly—and on transport properties. Normally, the lifetime will be primarily due to processes involving three phonons, that is, spontaneous decay processes, $A \rightarrow B+C$, or "collision" processes, $A+B \rightarrow C$.

It has been noted before¹ that since the probability for spontaneous-decay processes varies as the inverse fourth power of the wavelength, collision processes dominate the lifetime in the long-wavelength limit at finite temperatures. However, if there is only a single dispersion curve, and if it is convex, that is, if the phonon velocity decreases with increasing wavenumber—as is the case for helium II—it is impossible to conserve both energy and momentum in collision processes. This is also true for the case of longitudinal phonons in an isotropic solid with a convex dispersion curve. Such processes have, therefore, usually been neglected and it has been assumed that in solids the dominant processes are interactions with phonons from special regions of phase space where two-phonon branches nearly coincide² and, in liquid helium II, "indirect" four-phonon interactions proceeding via a virtual intermediate state.^{3,4}

However, a number of authors⁵⁻⁷ have recently

proposed, in connection with the absorption of sound in helium II, that if the energy uncertainty of the phonons B and C is of the same order as the "energy deficit" for the process $A+B \rightarrow C$, then this process can take place and will dominate the lifetime of A.⁸ The most detailed attempt to justify this hypothesis has been made by Simons.⁹ Using ordinary second-order time-dependent perturbation theory, Simons argues that the expression for the probability that the process $A+B \rightarrow C$ takes place after time t , which is normally taken to be proportional to $\sin\omega t/\omega$ where ω is the energy deficit,¹⁰ should be averaged over t from 0 to ∞ after having been weighted with a factor $\exp(-t/\tau_2)$ where τ_2 is the lifetime of B, since this factor gives the probability that B has been present for a time t . Integrating over ω from some threshold value $\omega_{20}(k_2)$ to infinity, Simons then gets for the lifetime τ of A

$$\tau^{-1} = \text{const} \times \omega_1 \int_0^\infty k_2^4 \frac{\partial n_2}{\partial \omega_2} \text{arccot}(\omega_{20}\tau_2) dk_2, \quad (1.1)$$

where ω_1 is the energy of A, k_2 the wave number of B, and n_2 the number of phonons B. We note that in the normal treatment $\text{arccot}\omega_{20}\tau_2$ would be replaced by $\pi\theta(\omega_{20})$ where $\theta(x)$ is the step function [$\theta(x)=1$, $x>0$; $\theta(x)=0$, $x<0$]. The conclusion is that the process $A+B \rightarrow C$ is, indeed, allowed with a "weight factor" $\text{arccot}\omega_{20}\tau_2$.

Although suggestive, this approach seems to be inadequate for several reasons: (i) the intuitive idea of the "time for which B has been present" seems to be hard to interpret on the quantum level, since B is one of a set of indistinguishable phonons, whose number does not change in time because the system is supposed to be in thermal equilibrium; (ii) intuitively, one feels that the "linewidth" effect must really be equivalent to some higher order effects involving B, but how this

* Present address: Department of Physics, University of Illinois, Urbana, Illinois.

¹ R. E. Peierls, *Quantum Theory of Solids* (Oxford University Press, New York, 1955).

² C. Herring, *Phys. Rev.* **95**, 954 (1954).

³ L. D. Landau and I. M. Khalatnikov, *Zh. Experm. i Teor. Fiz.* **19**, 637 (1949) [English transl.: *Collected Papers of L. D. Landau* (Gordon and Breach Publishers Inc., New York, 1965), p. 494].

⁴ I. M. Khalatnikov, *Zh. Experm. i Teor. Fiz.* **44**, 769 (1963) [English transl.: *Soviet Phys.—JETP* **17**, 519 (1963)].

⁵ K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **26**, 793, 795 (1961); K. Kawasaki and H. Mori, *ibid.* **28**, 784 (1962).

⁶ T. O. Woodruff, *Phys. Rev.* **127**, 682 (1962).

⁷ K. Dransfeld, *Phys. Rev.* **127**, 17 (1962).

⁸ On close examination, the mechanisms proposed by Dransfeld (Ref. 7) and Woodruff (Ref. 6) appear to be equivalent to ordinary phonon interaction theory for three-phonon processes (Dransfeld explicitly assumes the absence of dispersion).

⁹ S. Simons, *Proc. Phys. Soc. (London)* **82**, 401 (1963).

¹⁰ We use throughout units in which $\hbar=1$.

equivalence comes about is not clear and it is, for instance, difficult to see whether the "linewidth" effect replaces four- or five-phonon effects, is added to them, or is connected in an even more complicated way; (iii) it is not clear whether the assumption of an exponential decay of B—which is inexact at very short times—is adequate for this problem; and (iv) it is clearly inconsistent to take account of the finite lifetime of B, but not of the collision product C.

In the present paper we develop a method of taking account of the finite spread in energy of the colliding phonons which avoids most of the above difficulties. Basically, our method consists of using thermodynamic perturbation theory to derive the contribution to the self-energy of A from three-phonon processes. However, by using, instead of free-phonon propagators for B and C, the (unknown) exact propagators, some higher order phonon interactions are taken into account. By a method of successive approximations we elucidate the relation of our approach and the conventional perturbation-theory approach^{3,11} to higher order processes. It turns out that Simons' formula is, generally speaking, incorrect, since the assumption of an exponential decay—or, equivalently, of a Lorentzian line-shape—is unjustified. Under certain circumstances, the generalization of Simons' result obtained by taking account of the lifetime of C as well as that of B gives the correct answer.

As a by-product we obtain in a natural way a correction, previously obtained by Carruthers,¹² to higher order terms of the conventional perturbation series; this correction eliminates the unphysical divergence of these terms as the intermediate energy denominator tends to zero. We show that if we consider these terms at all, they should be substituted for the mediating three-phonon processes rather than added to it, and that usually, if the mediating three-phonon process is energetically allowed, the correction obtained in this way is negligible.

In the next section we discuss the phonon spectral function, and in Sec. 3 we derive our basic expression for the inverse lifetime of a phonon in terms of the exact spectral functions—or, equivalently, the exact propagators—of the other phonons involved in the three-phonon processes. In Sec. 4 we discuss various approximations for the exact propagators and show the relation of our method to conventional perturbation theory. In Sec. 5 we discuss to what extent Simons' simplified approach⁹ is justified. Applications are discussed in Sec. 6, while in Appendix A we give a justification of the Hamiltonian used in the present paper.

2. THE SPECTRAL FUNCTION

We shall discuss systems described by the following model Hamiltonian:

$$H = H_0 + V, \quad (2.1)$$

¹¹ I. Pomeranchuk, J. Phys. USSR 4, 259 (1941); 6, 237 (1942); Phys. Rev. 60, 820 (1942).

¹² P. Carruthers, Phys. Rev. 125, 123 (1962); 126, 1448 (1962).

where H_0 is the diagonalized second-quantized operator

$$H_0 = \sum_{\mathbf{k}} \omega(\mathbf{k}) a_{\mathbf{k}}^\dagger a_{\mathbf{k}}, \quad (2.2)$$

while V is given by the equation

$$V = (1/\mathcal{V})(1/3!) \sum_{\mathbf{k}, \mathbf{k}', \mathbf{k}''} V_{\mathbf{k}\mathbf{k}'\mathbf{k}''} (a_{\mathbf{k}} + a_{-\mathbf{k}}^\dagger) \times (a_{\mathbf{k}'} + a_{-\mathbf{k}'}^\dagger) (a_{\mathbf{k}''} + a_{-\mathbf{k}''}^\dagger). \quad (2.3)$$

We discuss the use of this Hamiltonian both for crystals and for liquid helium in Appendix A; \mathcal{V} is the volume occupied by the system, the $a_{\mathbf{k}}^\dagger$ and $a_{\mathbf{k}}$ are, respectively, creation and annihilation operators for bosons (phonons) with momentum \mathbf{k} , the curve $\omega(\mathbf{k})$ (for notational convenience we shall often write $\omega(\mathbf{k}) \equiv \epsilon_{\mathbf{k}}$) is assumed to be weakly convex, and we assume that the $V_{\mathbf{k}\mathbf{k}'\mathbf{k}''}$ satisfy the relation (See Appendix A)

$$V_{\mathbf{k}\mathbf{k}'\mathbf{k}''} = (kk'k'')^{1/2} \delta_{\mathbf{k}+\mathbf{k}'+\mathbf{k}''} \times \text{const.} \quad (2.4)$$

The quantity which plays an important part in our considerations is the so-called spectral function, which is essentially the Fourier transform of the expectation value of the commutator $[a_{\mathbf{q}}(t), a_{\mathbf{q}}^\dagger(0)]_-$, that is,¹³

$$A_{\mathbf{q}}(\omega) = i[G_{\mathbf{q}}^>(\omega) - G_{\mathbf{q}}^<(\omega)], \quad (2.5)$$

with

$$G_{\mathbf{q}}^>(\omega) = -i \int_{-\infty}^{+\infty} dt \langle a_{\mathbf{q}}(t) a_{\mathbf{q}}^\dagger(0) \rangle e^{i\omega t}, \quad (2.6a)$$

$$G_{\mathbf{q}}^<(\omega) = -i \int_{-\infty}^{+\infty} dt \langle a_{\mathbf{q}}^\dagger(0) a_{\mathbf{q}}(t) \rangle e^{i\omega t}, \quad (2.6b)$$

where the angle brackets $\langle \dots \rangle$ indicate an average over a canonical ensemble,

$$\langle \dots \rangle = (\text{Tr} \dots e^{-\beta H}) / (\text{Tr} e^{-\beta H}), \quad \beta = 1/k_B T \quad (2.7)$$

with k_B Boltzmann's constant and T the absolute temperature.

We note for future use the following relations¹³

$$\int_{-\infty}^{+\infty} A_{\mathbf{q}}(\omega) d\omega = 2\pi, \quad (2.8)$$

$$G_{\mathbf{q}}^>(\omega) = e^{\beta\omega} G_{\mathbf{q}}^<(\omega), \quad (2.9)$$

$$G_{\mathbf{q}}^>(\omega) = -i[n(\omega) + 1]A_{\mathbf{q}}(\omega), \quad (2.10)$$

where

$$n(\omega) = [e^{\beta\omega} - 1]^{-1}. \quad (2.11)$$

The probability that in the system described by the Hamiltonian (2.1) a phonon of wave vector \mathbf{q} is created at time $t=0$ and absorbed at time t is proportional to

$$|\langle a_{\mathbf{q}}(t) a_{\mathbf{q}}^\dagger(0) \rangle|^2 = \left| \int \frac{d\omega}{2\pi} [n(\omega) + 1] A_{\mathbf{q}}(\omega) e^{-i\omega t} \right|^2, \quad (2.12)$$

and we thus see that $A_{\mathbf{q}}(\omega)$ is related to the probability of decay of a phonon of wave vector \mathbf{q} which is added

¹³ L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962); our notation differs slightly from theirs (see, for instance, Ref. 14).

to the system in thermal equilibrium. Moreover, if $A_q(\omega)$ is of such a shape (*vide infra*) that it is non-vanishing only over a range of ω values which is small compared to $k_B T$, we can put the slowly varying factor $n(\omega)+1$ equal to its value at the peak of $A_q(\omega)$; the required probability is then simply proportional to the square of the Fourier transform of $A_q(\omega)$.

It is convenient to express $A_q(\omega)$ in terms of two real functions of ω , $\Delta_q(\omega)$ and $\Gamma_q(\omega)$ with $\Gamma_q(\omega)$ positive, as follows

$$A_q(\omega) = \frac{2\Gamma_q(\omega)}{[\omega - \epsilon_q - \Delta_q(\omega)]^2 + [\Gamma_q(\omega)]^2}, \quad (2.13)$$

where ϵ_q is the harmonic-approximation phonon energy. The quantities $\Delta_q(\omega)$ and $\Gamma_q(\omega)$ are related through the dispersion relation (P indicates a principal-value integral)¹³

$$\Delta_q(\omega) = -\frac{1}{\pi} P \int \frac{\Gamma_q(\omega') d\omega'}{\omega - \omega'}. \quad (2.14)$$

We shall in the next section discuss how $\Gamma_q(\omega)$ and $\Delta_q(\omega)$ can be evaluated.

The special case of noninteracting phonons is reached by letting $\Gamma_q(\omega)$ tend to zero for all values of ω , and we then get from (2.13)

$$A_q(\omega) = 2\pi\delta(\omega - \epsilon_q). \quad (2.15)$$

On the other hand, sometimes $\Gamma_q(\omega)$ may tend to zero in the region where $\omega - \epsilon_q - \Delta_q(\omega)$ is small, even though it remains finite in some other region, say, above a “threshold” ω_0 . From (2.15) it then follows that $\Delta_q(\omega)$ remains finite and varies with ω . In the region of the “peak” where $\omega - \epsilon_q - \Delta_q(\omega) \sim 0$, $A_q(\omega)$ will be of the form

$$A_q(\omega) = \frac{2\pi}{1 - (\partial\Delta_q/\partial\omega)} \delta(\omega - \tilde{\epsilon}_q), \quad (2.16a)$$

where $\tilde{\epsilon}_q$ is defined by

$$\tilde{\epsilon}_q - \epsilon_q - \Delta_q(\tilde{\epsilon}_q) = 0;$$

since in this case $\partial\Delta_q/\partial\omega$ is negative [cf. (2.14)] we find

$$A_q(\omega) = \frac{2\pi}{1 + |\partial\Delta_q/\partial\omega|} \delta(\omega - \tilde{\epsilon}_q); \quad (2.16b)$$

that is, we find a reduction of the “strength” of the singularity—necessary in order that the sum rule (2.8) be satisfied. In Sec. 4 we shall discuss a situation which closely resembles this case.

The physical interpretation of Eq. (2.16) is the following one: The “bare-particle” excitation states $a_q^\dagger\Phi_0$, where Φ_0 is any eigenstate of the Hamiltonian, are themselves not eigenstates of the Hamiltonian, while the “dressed-particle” excitation states are eigenstates. If we express the “bare-particle” states in terms of the “dressed-particle” states, they will contain an admixture of states with arbitrary energies, including

those states for which ω is larger than the threshold energy ω_0 . This is indicated by the finite values of $\Gamma_q(\omega)$ and hence of $A_q(\omega)$ for $\omega > \omega_0$. The threshold represents the minimum energy for which spontaneous decay is energetically possible (*vide infra*). At finite temperatures Γ also rises *below* the peak, as there exists a *maximum* energy below which three-phonon collision processes are possible.

We must emphasize that Eq. (2.13) does not necessarily mean that $A_q(\omega)$ has a Lorentzian form. This is only the case if $\Delta_q(\omega) = \text{const} \equiv \Delta$ and $\Gamma_q(\omega) = \text{const} \equiv \Gamma$. In general, this will not be true. Sometimes, however, $\Delta_q(\omega)$ and $\Gamma_q(\omega)$ may be varying slowly in the immediate neighborhood of the peak. In that case we find from (2.12) that—at least for sufficiently large t —

$$|\langle a_q(t) a_q^\dagger(0) \rangle|^2 \approx c e^{-t/\tau}, \quad (2.17)$$

with

$$1/\tau = 2\Gamma_q(\tilde{\epsilon}_q), \quad (2.18)$$

where $\tilde{\epsilon}_q$ is the position of the peak of $A_q(\omega)$.

We must note that the constant c in (2.17) is not necessarily unity. This can be interpreted as follows: the “bare-particle” state $a_q^\dagger\Phi_0$ contains, apart from a single “dressed-particle” state (the amplitude of which is less than unity), states involving more than one “dressed particle.” The single “dressed-particle” state will decay, corresponding to Eq. (2.17). We see thus that (2.17) will hold for values of t larger than the time necessary for the states with more than one “dressed particle” to be “drained off.”

Conversely, we see that for any process (such as the absorption of sound) characterized by a “lifetime,” we need know only $\Gamma_q(\tilde{\epsilon}_q)$ and it is reasonable to approximate $A_q(\omega)$ by a Lorentzian curve. Moreover, although, strictly speaking, a departure from the Lorentzian form should show up in neutron-scattering experiments, which measure $A_q(\omega)$ directly, the accuracy of the experiment is unlikely to be adequate for this purpose for some time to come (see also Sec. 6) so that for a comparison between theory and experiment it is usually the “half-width” of the peak, given by $\Gamma_q(\tilde{\epsilon}_q)$, which is of interest.¹⁴

If we need to know the behavior of $A_q(\omega)$ far from the peak (compare Sec. 4), it may be a very bad approximation to replace $\Gamma_q(\omega)$ by its value at the peak, independent of whether we use a calculated value or one deduced from sound-absorption or neutron-scattering experiments.

3. CALCULATION OF $\Gamma_q(\omega)$

It has been shown in the literature^{13,15} that $\Gamma_q(\omega)$ and $\Delta_q(\omega)$ can be expressed in terms of the so-called irreduc-

¹⁴ Note that in contradistinction to many authors (e.g., Ref. 13) we use $\Gamma_q(\tilde{\epsilon}_q)$ for the half-width and not for the full width.

¹⁵ A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Method of Quantum Field Theory in Statistical Physics*, translated by R. A. Silverman (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963).

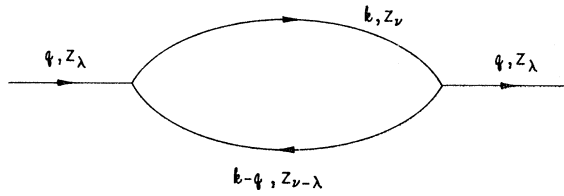


FIG. 1. Simple bubble diagram.

ible self-energy Σ_q , as follows

$$\Gamma_q = \frac{1}{2}i[\Sigma_q(\omega + i\epsilon) - \Sigma_q(\omega - i\epsilon)] = -\text{Im}\Sigma_q(\omega + i\epsilon),$$

$$\Delta_q = \text{Re}\Sigma_q(\omega + i\epsilon), \quad \epsilon \rightarrow +0. \quad (3.1)$$

The rules for the evaluation of $\Sigma_q(z)$ by diagram techniques are given in the monograph by Abrikosov, Gorkov, and Dzyaloshinski¹⁵ to which we refer for details. In the finite-temperature diagram technique we need to find the single-particle propagators (2.6a,b) for a discrete set of points z_n on the imaginary axis, $z_n = 2n\pi i/\beta$ (n : integer). We note that the free-phonon single-particle propagator is given by

$$G_q^{(0)}(z_n) = (z_n - \epsilon_q)^{-1}, \quad (3.2)$$

while the exact phonon propagator satisfies the equation

$$G_q(z_n) = (z_n - \epsilon_q - \Sigma_q(z_n))^{-1}. \quad (3.3)$$

Let us first consider the contribution to the self-energy of the simple bubble diagram of Fig. 1. It is given by the equation

$$\Sigma_q^{(0)}(z_\lambda) = -(\beta)^{-1} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \sum_\nu G_k^{(0)}(z_\nu) \times G_{k-q}^{(0)}(z_{\nu-\lambda}) |V_{k,-q,q-k}|^2. \quad (3.4)$$

This expression can be evaluated by contour integration (*vide infra*) and gives for the corresponding contribution to Γ_q

$$\Gamma_q^{(0)}(\omega) = \pi \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V_{k,-q,q-k}|^2 (n_{k-q} - n_k) \times \delta(\omega - \epsilon_k + \epsilon_{k-q}). \quad (3.5)$$

This result could, of course, have been obtained much more simply by ordinary time-dependent perturbation theory. We notice that the contribution to $\Gamma_q^{(0)}(\epsilon_q)$ from interactions between phonons of the same branch vanishes, if the dispersion curve is convex, since in that case the argument of the δ function cannot be made to vanish.

If we wish to take into account the finite spread in energy of the phonons \mathbf{k} and $\mathbf{k}-\mathbf{q}$, it is natural to replace the free-phonon propagators in (3.4) by the exact propagators $G_k(z_\nu)$ and $G_{k-q}(z_{\nu-\lambda})$. This is equivalent to summing the contributions to $\Sigma_q(z_\lambda)$ of a certain infinite set of diagrams, of the type shown in Fig. 2.; that is, all single-bubble diagrams in which

each internal line may have an arbitrary number of self-energy insertions (represented by the shaded circles in Fig. 2), but in which there are no crosslinks, which means that we neglect vertex renormalization.

The sum over ν in Eq. (3.4) where we now have G in place of $G^{(0)}$, is most easily carried out by substituting for G its spectral representation (cf., e.g., Ref. 13)

$$G_k(z_\nu) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \frac{d\omega A_k(\omega)}{z_\nu - \omega},$$

where A_k is the spectral function defined by (2.5)–(2.6). The summation over ν can then be turned in the well-known way into a contour integration

$$\frac{1}{\beta} \sum_\nu f(z_\nu) = \frac{1}{2\pi i} \oint_C n(z) f(z) dz,$$

where $n(z)$ is given by (2.11) and the contour encircles the poles of $f(z)$ in a clockwise sense. We obtain in this way the following expression for $\Sigma_q(z_\lambda)$:

$$\Sigma_q(z_\lambda) = \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V_{k,-q,q-k}|^2 \int d\omega' \int d\omega'' \times \frac{[f(\omega'' + z_\lambda) - f(\omega')] A_k(\omega') A_{k-q}(\omega'')}{z_\lambda + \omega'' - \omega'}.$$

Making the unique correct analytic continuation of this expression, and using (3.1), we find

$$\Gamma_q(\omega) = \frac{1}{2} \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V_{k,-q,q-k}|^2 \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} n(\omega') \times [A_{k-q}(\omega') A_k(\omega + \omega') - A_{k-q}(\omega' - \omega) A_k(\omega')]. \quad (3.6)$$

This is our fundamental result for that part of the “linewidth” of a phonon of wave vector \mathbf{q} , which is due to its collisions with other phonons to which one can assign a finite linewidth. To see its physical significance we shall make a slight approximation. The first term of the integrand is large only when $\omega \sim \tilde{\epsilon}_{k-q}$ or $\omega' \sim \tilde{\epsilon}_k - \omega$. In cases of physical interest the inequality $\Omega \equiv \omega - \tilde{\epsilon}_{k-q} + \tilde{\epsilon}_k \ll k_B T$ is likely to be satisfied, as $k_B T$ is of the order of magnitude of the energies $\tilde{\epsilon}_k$ and $\tilde{\epsilon}_{k-q}$ themselves and ω will lie in the region of $\tilde{\epsilon}_q$ so that Ω is the energy “deficit” due to the dispersion which is assumed to be weak. For present purposes we shall also assume that the widths of the spectral functions are small compared to $k_B T$. We can thus approximate $n(\omega')$ in the region where the integrand is important by $n(\tilde{\epsilon}_{k-q})$ and take it out from under the second integral sign.



FIG. 2. Diagrams summed by using exact propagators.

Approximating the second term in a similar way, we have

$$\Gamma_q(\omega) \approx \pi \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V_{\mathbf{k},-\mathbf{q},\mathbf{q}-\mathbf{k}}|^2 [n(\tilde{\epsilon}_{\mathbf{k}-\mathbf{q}}) - n(\tilde{\epsilon}_{\mathbf{k}})] \times \int \frac{d\omega'}{4\pi^2} A_{\mathbf{k}}(\omega') A_{\mathbf{k}-\mathbf{q}}(\omega' - \omega). \quad (3.7)$$

Comparing (3.7) with (3.5) we see that the effect of the “broadening” of the other phonons is to replace the δ function by a convolution of spectral functions. This result can be interpreted physically as follows: $A_q(\omega')/2\pi$ can be regarded as the probability that the phonon of wavevector \mathbf{k} has an energy ω' . In fact, in the special case where $A_{\mathbf{k}-\mathbf{q}}(\omega' - \omega) = 2\pi\delta(\epsilon_{\mathbf{k}-\mathbf{q}} - \omega' + \omega)$, we find from (3.7) that the contribution to $\Gamma_q(\epsilon_q)$ from the decay $\mathbf{q} + (\mathbf{k} - \mathbf{q}) \rightarrow \mathbf{k}$ is simply proportional to $A_{\mathbf{k}}(\epsilon_q + \epsilon_{\mathbf{k}-\mathbf{q}})$, that is, to the probability that the phonon which can conserve wavevector with \mathbf{q} and $\mathbf{k} - \mathbf{q}$ can also conserve energy with them. One easily verifies that in the limit when both interacting phonons can be regarded as “free,” so that we can use (2.15), (3.7) reduces to (3.5) as it should.

We now have to add to expression (3.7) the contribution from the spontaneous decay process (see Fig. 3). The calculations proceed as before and to (3.7) we must add the contribution

$$\Gamma_q(\omega) = \pi \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V_{-\mathbf{k},\mathbf{q},\mathbf{k}-\mathbf{q}}|^2 [n(\tilde{\epsilon}_{\mathbf{k}}) + n(\tilde{\epsilon}_{\mathbf{q}-\mathbf{k}}) + 1] \times \int \frac{d\omega'}{4\pi^2} A_{\mathbf{k}}(\omega') A_{\mathbf{q}-\mathbf{k}}(\omega - \omega'). \quad (3.8)$$

Expression (3.8) also reduces to that obtained from ordinary second-order perturbation theory if we can use (2.15). Expressions (3.7) and (3.8) together give us, in the approximation used, the complete expression for $\Gamma_q(\omega)$. We have neglected (i) vertex renormalization, (ii) three-phonon annihilation processes, and (iii) scattering by impurities or at the boundaries.

4. APPROXIMATIONS FOR THE SPECTRAL FUNCTIONS

Equations (3.7) and (3.8) still involve the unknown spectral functions $A_{\mathbf{k}}$, $A_{\mathbf{k}-\mathbf{q}}$, and $A_{\mathbf{q}-\mathbf{k}}$. If we want to use these expressions or want to compare them with expressions obtained by conventional perturbation theory, we must approximate these spectral functions in some way. In this section we shall examine two cases in which we may make plausible approximations to them, in order to illuminate the relation between our approach and the results of ordinary perturbation theory. The first case is that of a single, uniformly convex dispersion curve; in this case, ordinary perturbation theory gives a zero result for the linewidth in

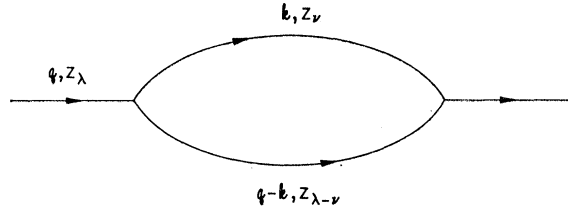


FIG. 3. Contribution from spontaneous decay.

lowest order [cf. Eq. (3.5)] while the next-order result diverges in the limit of vanishing dispersion. We shall see that, if reasonable approximations are made for one or more of the $A_{\mathbf{k}}$, our method gives a result which agrees with the ordinary higher order formula for strong dispersion, but remains finite as the dispersion vanishes. Our second case is a situation where the “three-phonon” process is energetically allowed, so that the ordinary lowest order result for the linewidth is finite; in this case we find that our method gives a result essentially identical to the normal one.

Let us then consider Eq. (3.7); we shall assume, unless otherwise stated, that we are interested in the value of $\Gamma_q(\omega)$ for $\omega = \tilde{\epsilon}_q$. Moreover, we shall generally neglect the difference between $n(\tilde{\epsilon}_{\mathbf{k}})$ and $n(\epsilon_{\mathbf{k}})$.

The lowest order approximation to $\Gamma_q(\omega)$ is obviously the use of (2.15) for the spectral functions. We saw in the preceding section that we then get the usual second-order perturbation-theory result; in particular $\Gamma_q = 0$, if the dispersion is normal. A better approximation is obviously obtained by giving $A_{\mathbf{k}}(\omega') A_{\mathbf{k}-\mathbf{q}}(\omega' - \omega)$ the value calculated by second-order perturbation theory. The resulting expression can easily be written down; it involves four “five-phonon” terms, each of which can be seen to correspond (in a sense to be discussed below) to a possible pair of processes responsible for the lifetime of \mathbf{k} and of $\mathbf{k} - \mathbf{q}$. The expressions are rather cumbersome. As we want to compare our results with those of ordinary perturbation theory which usually involve four-phonon rather than five-phonon processes, we shall not give the results explicitly, but shall make a further approximation, as follows: Let us consider for illustration the case where the dispersion curve is convex ($\Omega \equiv \omega + \tilde{\epsilon}_{\mathbf{k}-\mathbf{q}} - \tilde{\epsilon}_{\mathbf{k}} > 0$). In this case, in second-order perturbation theory $A_{\mathbf{k}}(\omega')$ has a pole at $\tilde{\epsilon}_{\mathbf{k}}$ ($\Gamma_{\mathbf{k}}(\omega') \rightarrow 0$ there); above $\tilde{\epsilon}_{\mathbf{k}}$, $\Gamma_{\mathbf{k}}(\omega')$ rises sharply due to spontaneous decay processes until $\omega' = ck'$, after which it stays virtually constant. At finite temperatures, $\Gamma_{\mathbf{k}}$ also rises below $\tilde{\epsilon}_{\mathbf{k}}$ due to collision processes. Considering the convolution of the spectral functions $\int (d\omega'/4\pi^2) A_{\mathbf{k}}(\omega') A_{\mathbf{k}-\mathbf{q}}(\omega' - \omega)$ and setting $\omega = \tilde{\epsilon}_q$, we see, since by hypothesis $\tilde{\epsilon}_q + \tilde{\epsilon}_{\mathbf{k}-\mathbf{q}} > \tilde{\epsilon}_{\mathbf{k}}$, that at the value of $\omega' - \omega$ (viz., $\tilde{\epsilon}_{\mathbf{k}-\mathbf{q}}$) for which $A_{\mathbf{k}-\mathbf{q}}$ has a pole, $A_{\mathbf{k}}$ is in its “spontaneous-decay range,” while at the value where $A_{\mathbf{k}}$ has a pole ($\tilde{\epsilon}_{\mathbf{k}}$) $A_{\mathbf{k}-\mathbf{q}}$ is in its “collision range.” If we assume fairly weak interactions, by far the greatest contribution to the convolution integral will

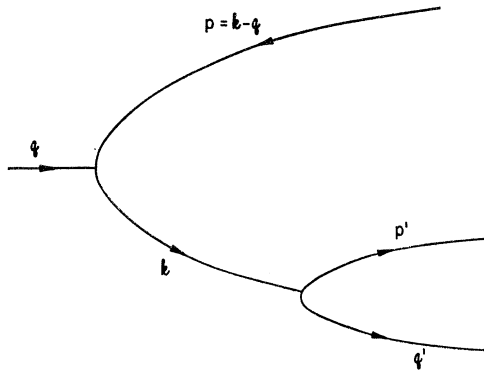


FIG. 4. Diagram corresponding to the process of Eq. (4.5).

come from these two points.¹⁶ Thus, in this case, provided ω is equal or nearly equal to $\tilde{\epsilon}_q$, it is a good approximation to replace the integral over ω' in (3.7) by

$$(1/2\pi)[A_k(\omega + \tilde{\epsilon}_{k-q}) + A_{k-q}(\tilde{\epsilon}_k - \omega)].$$

We introduce a certain error by neglecting the reduction of the pole strengths; this is, however, unimportant for the purposes of comparison with the usual treatment. Let us first look at the term containing $A_k(\omega + \tilde{\epsilon}_{k-q})$ which we shall denote by $\Gamma_q^{(a)}$; it is given by the equation

$$\Gamma_q^{(a)}(\omega) = \pi \int \frac{d^3k}{(2\pi)^3} |V_{k,-q,q-k}|^2 [n(\tilde{\epsilon}_{k-q}) - n(\tilde{\epsilon}_k)] \times \frac{2\Gamma_k(\omega + \tilde{\epsilon}_{k-q})}{[\omega + \tilde{\epsilon}_{k-q} - \epsilon_k - \Delta_k(\omega + \tilde{\epsilon}_{k-q})]^2 + \Gamma_k(\omega + \tilde{\epsilon}_{k-q})^2}, \quad (4.1)$$

where we have used (2.13). We shall generally assume that Δ_k is slowly varying so that we can replace $\epsilon_k + \Delta_k(\omega + \tilde{\epsilon}_{k-q})$ by $\tilde{\epsilon}_k$.

If Γ_k is due to several kinds of process, we can write $\Gamma_q^{(a)}(\omega)$ as a sum of terms corresponding to each of these, but of course we must keep the full Γ_k in the denominator. In the case of a single convex dispersion curve there is only one contributing process, spontaneous decay, which ‘goes’ at energy $\omega + \tilde{\epsilon}_{k-q}$. Substituting, therefore, the second-order perturbation theory expression for the contribution to Γ_k from spontaneous decay, we obtain

$$\Gamma_q^{(a)}(\omega) = \int \frac{d^3k}{(2\pi)^3} \int \frac{d^3l}{(2\pi)^3} \times C_{q,k,l} \frac{\delta(\omega + \tilde{\epsilon}_{k-q} - \epsilon_l - \epsilon_{k-l})}{[\omega + \tilde{\epsilon}_{k-q} - \tilde{\epsilon}_k]^2 + \Gamma_k^2(\omega + \tilde{\epsilon}_{k-q})}, \quad (4.2)$$

¹⁶ We note that we are dealing here with a convolution of two functions which have a sharp peak surrounded by a region where the functions are practically equal to zero, while the functions have again non-vanishing values in regions somewhat removed from the sharp peak. In the more usual case of a convolution of two functions which each have a more or less sharply pronounced maximum, the convolution itself will be a function with one maximum [compare, for instance, Eq. (4.7) below].

where

$$C_{q,k,l} = |V_{k,-q,q-k}|^2 |V_{-k,l,k-l}|^2 \times \{n(\tilde{\epsilon}_{k-q}) - n(\tilde{\epsilon}_k)\} \{n(\epsilon_l) + n(\epsilon_{k-l}) + 1\}. \quad (4.3)$$

Using the relation

$$\frac{n_A + 1}{n_A} = \frac{n_B + 1}{n_B} \times \frac{n_C + 1}{n_C}, \quad \text{if } \epsilon_A = \epsilon_B + \epsilon_C, \quad (4.4)$$

and writing $\mathbf{p} = \mathbf{k} - \mathbf{q}$, we can rewrite $C_{q,k,l}$ in a form not involving ϵ_k ; hence

$$\Gamma_q^{(a)}(\omega) = \pi \int \frac{d^3p}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} \int d^3q' \delta(\mathbf{p} + \mathbf{q} - \mathbf{p}' - \mathbf{q}') \times |V_{-p,q,-p-q}|^2 |V_{-p',q',-p-q}|^2 \times [n_p(n_{p'} + 1)(n_{q'} + 1) - (n_{p+1})n_{p'}n_{q'}] \times \frac{\delta(\omega + \tilde{\epsilon}_p - \epsilon_{p'} - \epsilon_{q'})}{[\omega + \tilde{\epsilon}_p - \tilde{\epsilon}_{p+q}]^2 + \Gamma_{p+q}^2(\omega + \tilde{\epsilon}_p)}. \quad (4.5)$$

This expression obviously corresponds in some sense to the process of Fig. 4. The usual perturbation-theoretic expression for the contribution to the line width of this process is³

$$\Gamma_q^{(a)}(\omega) = \pi \int \frac{d^3p}{(2\pi)^3} \int \frac{d^3p'}{(2\pi)^3} \int d^3q' \times C(\mathbf{p}\mathbf{p}'\mathbf{q}\mathbf{q}') \frac{\delta(\omega + \epsilon_p - \epsilon_{p'} - \epsilon_{q'})}{(\omega + \epsilon_p - \epsilon_{p+q})^2} \quad (4.6)$$

where $C(\mathbf{p}\mathbf{p}'\mathbf{q}\mathbf{q}')$ is the expression appearing in (4.5). It is obvious that (4.6) cannot be exact as it diverges in the limit of zero dispersion; in fact, Carruthers¹² has pointed out that when the energy denominator can vanish, it must be replaced by a Breit-Wigner type expression. We shall return to this point in a moment, but here want to point out that (4.5) does not depend on whether or not the three-phonon process is energetically allowed, provided only that the approximation we have made for the convolution integral is a good one.

From a comparison of (4.5) and (4.6) we draw the following important, if somewhat obvious conclusion: Taking the finite lifetimes of the other phonons properly into account is simply equivalent to an improved calculation of the effect of higher order processes mediated by them. In particular, there must be available some ‘final state’—in the case under consideration $(\mathbf{p}') + (\mathbf{q}') - (\mathbf{p})$ —with which it is possible for the initial state (\mathbf{q}) to conserve energy. If the mediating three-phonon process itself is energetically possible, we must slightly reformulate our results (see next section). We have chosen a particular form of the spectral functions in order to compare our results explicitly with the normal expression for ‘indirect’

four-phonon processes; however, it is clear that the general conclusion stated in this paragraph is independent of this choice through the comparison with, for instance, “five-phonon” expressions will be much more cumbersome.

We now consider our second case. A possible advantage of the procedure outlined in Sec. 3 is that it is applicable whether or not the mediating three-phonon process is energetically allowed—even though hitherto we have assumed that it is not. In our derivations we nowhere made any assumptions about the value of $\Omega (\equiv \omega - \tilde{\epsilon}_k + \tilde{\epsilon}_{k-q})$ so that our equations are valid also in the case when Ω can vanish. Let us, moreover, consider for illustration the case in which one of the phonons involved, say $(\mathbf{k} - \mathbf{q})$, is well defined in energy compared to \mathbf{k} , that is, $\Gamma_{\mathbf{k}-\mathbf{q}} \ll \Gamma_{\mathbf{k}}$ everywhere in the region of their respective peaks. In this case, it is again valid to replace the convolution integral by

$$(1/2\pi)[A_k(\omega + \tilde{\epsilon}_{k-q}) + A_{k-q}(\tilde{\epsilon}_k - \omega)],$$

where, in fact, the second term will be small compared to the first; note that in the limit $\Gamma_{\mathbf{k}-\mathbf{q}} \ll \Gamma_{\mathbf{k}}$ this expression agrees with (5.1) below. Thus, the expansion in terms of higher order processes remains valid and (4.5) holds, in agreement with Carruther’s result.¹²

One might intuitively think that the four-phonon process of Fig. 4 is the one corresponding to equation (4.5). This is, however, misleading in that (4.5) is not an *additional* contribution, to be added to the usual three-phonon process, but part of a better approximation for the latter. In the limit as $T \rightarrow 0$ it should constitute the whole of this approximation, as then the probability of interactions absorbing two phonons besides \mathbf{q} is vanishingly small compared to the probability of those absorbing one; in that case we can, for a good approximation, substitute (4.1) for (4.5). Indeed, we see that if $\Gamma_{\mathbf{k}}$ and the matrix elements vary slowly as functions of k ,¹⁷ (4.1) gives exactly the same result as the ordinary expression for decay due to three-phonon collision processes. Thus, calculating the decay probability due to four-phonon processes mediated by an “allowed” three-phonon process is simply an improved way of calculating that due to the three-phonon process itself, and in general is unlikely to give very different results from the simple, “energy-conserving,” treatment of the latter.

5. COMPARISON WITH SIMONS’ APPROACH

In this section we shall investigate the conditions under which the approach proposed by Simons⁹ is essentially correct. For this purpose we consider once more the case in which a three-phonon process which would otherwise be an important decay mode of a long-

wavelength phonon is energetically forbidden. However, we now relax the very restrictive assumption, made in the first half of Sec. 4, that a single convex dispersion curve represents the *only* low-energy excitations of the systems. We shall show that under certain alternative assumptions, a slight modification of Eq. (1.1) gives the correct result.

It is evident from the first case considered in Sec. 4 that, if the three-phonon process is forbidden, it is *in general* by no means sufficient to characterize the structure of the intermediate spectral function by a single parameter Γ , set equal to the half-width at the peak. Indeed, the very fact that the three-phonon process is forbidden, while the four-phonon process is energetically possible, must imply that $\Gamma_{\mathbf{k}}(\tilde{\epsilon}_k)$ is zero while $\Gamma_{\mathbf{k}}(\omega + \tilde{\epsilon}_{k-q})$ is finite, so that it is undoubtedly incorrect to replace one by the other. In fact, if we were really dealing with a uniformly convex dispersion curve in the absence of other branches, impurities, etc., this prescription would never lead to a finite lifetime for any phonon at all—to any finite order in the interaction strength: to get a finite value of $\Gamma_{\mathbf{q}}(\tilde{\epsilon}_q)$ by this prescription we should need a finite value of at least one of $\Gamma_{\mathbf{k}}(\tilde{\epsilon}_k)$, $\Gamma_{\mathbf{k}-\mathbf{q}}(\tilde{\epsilon}_{k-q})$, and so we would be no further forward. In this case, the use of a single parameter would give too small a result; as we shall see in Sec. 6, there are also dispersion curves for which it would give too large a result, as there is nothing to prevent $\Gamma_{\mathbf{k}}(\tilde{\epsilon}_k)$ from being *larger* than $\Gamma_{\mathbf{k}}(\omega + \tilde{\epsilon}_{k-q})$.

In practice, of course, we never have to deal with perfect liquids or solids with uniformly convex dispersion curves. In particular, it is almost certain that the hydrodynamic model for helium II is inadequate, even in the long-wavelength region, in the sense that there may be present other low-energy excitations besides phonons. Perhaps we may argue as follows: Consider our (energetically forbidden) three-phonon process, $(\mathbf{q}) + (\mathbf{p}) \rightarrow (\mathbf{k})$, in which an acoustic phonon collides with a thermal phonon of much higher momentum, producing another thermal phonon. The experimentally observed linewidths of the thermal phonons may well be due, partly or even mainly, to some unknown type of process, which we denote by $(\mathbf{k}) \rightarrow ?$. If this is so, then it is very unlikely that $\Gamma_{\mathbf{k}}(\omega)$ is sharply varying in the regions of interest, so that it seems legitimate in this case to approximate the spectral functions of (\mathbf{k}) and (\mathbf{p}) by a Lorentzian shape and to substitute this into (3.6). This would be equivalent to considering the processes $(\mathbf{q}) + (\mathbf{p}) \rightarrow (\mathbf{k}) \rightarrow ?$, which processes might well be the most important decay modes of an acoustic phonon (\mathbf{q}) , even though they go via a virtual intermediate state (\mathbf{k}) . For this approach to be valid it is necessary that (a) all *and only* those types of processes which “go” at $\tilde{\epsilon}_k$ also “go” at $\tilde{\epsilon}_k + \Omega$, where Ω is again equal to $\omega - \tilde{\epsilon}_k + \tilde{\epsilon}_{k-q}$, and that (b) there are no direct processes which can interfere with the indirect ones. The first condition is equivalent to the statement that $\Gamma_{\mathbf{k}}(\omega)$ varies slowly over a range comparable with Ω .

¹⁷ For longitudinal phonons in Carruthers’ fictitious isotropic solid (Ref. 12) this is generally the case, since the process where one longitudinal phonon decays into two transverse ones is always energetically possible.

If these conditions are fulfilled, we can write for the integral in (3.7)

$$D(\Omega) \equiv \frac{1}{4\pi^2} \int_{-\infty}^{+\infty} A_{\mathbf{k}}(\omega') A_{\mathbf{k}-\mathbf{q}}(\omega' - \Omega) d\omega'$$

$$= \frac{\Gamma_{\mathbf{k}} \Gamma_{\mathbf{k}-\mathbf{q}}}{\pi^2} \int_{-\infty}^{+\infty} \frac{d\omega'}{[(\omega' - \bar{\epsilon}_{\mathbf{k}})^2 + \Gamma_{\mathbf{k}}^2][(\omega' - \Omega - \bar{\epsilon}_{\mathbf{k}-\mathbf{q}})^2 + \Gamma_{\mathbf{k}-\mathbf{q}}^2]} = \frac{1}{2\pi} \frac{2(\Gamma_{\mathbf{k}} + \Gamma_{\mathbf{k}-\mathbf{q}})}{\Omega^2 + (\Gamma_{\mathbf{k}} + \Gamma_{\mathbf{k}-\mathbf{q}})^2}. \quad (5.1)$$

In order to compare our results with those of Simons,⁹ we put $\omega = \bar{\epsilon}_{\mathbf{q}}$, and consider the case where $\bar{\epsilon}_{\mathbf{q}} \ll k_B T$. Simons has shown that in that case the integration over angles can be turned into an integration over Ω . Instead of $\text{arccot}(\omega_{20}\tau_2)$ in (1.1) we now have the integral

$$I = \pi \int_{\omega_{20}}^{\infty} D(\Omega) d\Omega, \quad (5.2)$$

where $k_2 = |\mathbf{k} - \mathbf{q}|$ and τ^{-1} is related to $\Gamma_{\mathbf{q}}(\bar{\epsilon}_{\mathbf{q}})$ given by (3.7) through $\tau^{-1} = 2\Gamma_{\mathbf{q}}(\bar{\epsilon}_{\mathbf{q}})$. Performing the integration in (4.8), we find $I = \text{arccot}(\omega_{20}\tau_{2,3})$ where

$$\tau_{2,3} = 1/(\Gamma_2 + \Gamma_3) = 2\tau_2\tau_3/(\tau_2 + \tau_3), \quad (5.3)$$

where Γ_2 and Γ_3 are the Γ 's corresponding to the phonons $(\mathbf{q} - \mathbf{k})$ and (\mathbf{k}) . Since these phonons are supposed to be of the same branch and close in energy, $\tau_{2,3} \sim \tau_2$ and we have formally the same results as (1.1) provided that the energies defining Ω are taken to be the $\bar{\epsilon}_{\mathbf{q}}$ rather than the $\epsilon_{\mathbf{q}}$. In conclusion we emphasize that we obtain (1.1) only when the conditions (a) and (b) stated above are satisfied; we shall discuss in Sec. 6 in how far this is true for the applications considered by Simons.

6. SPECIFIC PROBLEMS

We have seen that by taking into account the finite linewidths of the phonons involved in an interaction is equivalent to considering some of the higher order processes mediated by them. In general, these effects cannot be adequately described by describing the "structure" of the phonon spectral functions by a single parameter, τ , the lifetime. However, such a simplified treatment was seen in Sec. 5 to be justified,

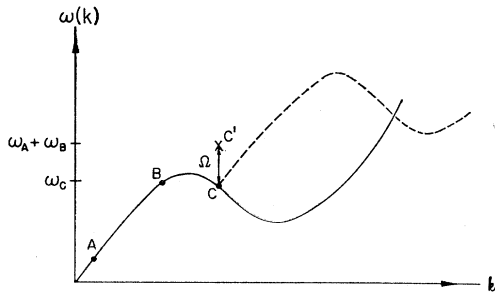


FIG. 5. Dispersion curve for excitations in He II.

if (a) all and only those types of processes "go" at $\bar{\epsilon}_{\mathbf{k}} - \Omega$, which "go" at $\bar{\epsilon}_{\mathbf{k}}$, and (b) there are no direct processes which can interfere with the indirect ones.

Condition (a) is certainly never completely satisfied for the case of a convex dispersion curve; if it were, four-phonon processes such as the ones considered by Landau and Khalatnikov³ would never occur. Whether we can treat it as satisfied in practice depends on the relative importance of the various decay mechanisms for the intermediate phonon \mathbf{k} and on their variation over the energy interval Ω .

A simple argument shows, for instance, that it is by no means certain that condition (a) is satisfied in liquid helium II. Consider, for instance, a process where a long-wave acoustic phonon A collides with a thermal phonon B, producing another thermal phonon C and assume that B and C lie near the first maximum of the dispersion curve (Fig. 5). This process is, of course, energetically forbidden, as the energy deficit Ω is of the order ω_A . Now, quite an appreciable contribution to the linewidth of C (wavenumber \mathbf{k}) will come from collisions with excitations (\mathbf{k}') in the roton area. Such collisions can take place if $\omega(k) + \omega(k') \leq \omega(k + k')$: the minimum value of k' is thus given by the intersection with the original $\omega(k)$ -versus- k curve of a similar one drawn with C as origin (dotted line in Fig. 5). We see thus that $\Gamma_{\mathbf{k}}(\bar{\epsilon}_{\mathbf{k}})$ has contributions from collisions with excitations of momenta greater than some critical value k_C . On the other hand, the corresponding process can contribute to $\Gamma_{\mathbf{k}}(\bar{\epsilon}_{\mathbf{k}} + \Omega)$ only, if k' is greater than the value $k_{C'}$ at which a curve drawn with respect to C' as origin cuts the original curve. In the case of liquid helium $k_{C'}$ may be much greater than k_C and may even not exist, even if k_C exists. The simplified treatment may in this case thus give far too large a value for the width of the acoustic phonon. Strictly speaking, a study of the "wings" of the neutron-scattering peaks should be sufficient to resolve this question; however, it is very doubtful whether present data^{18,19} are sufficiently accurate to allow us to draw definite conclusions.

At very low temperatures the "thermal" phonons are well below the first maximum of the dispersion curve and their decay is in all probability due mainly to the four-phonon processes considered by Landau and Khalatnikov,³ although not necessarily only their

¹⁸ J. L. Yarnell, G. P. Arnold, P. J. Bendt, and E. C. Kerr, Phys. Rev. **113**, 1379 (1959).

¹⁹ D. G. Henshaw and A. D. B. Woods, Phys. Rev. **121**, 1266 (1961).

process I. It is therefore a question whether the decay of very long wave acoustic phonons may not be due primarily to “linewidth” effects, that is, to five-phonon processes, such as the one of Fig. 6a, which are not just modifications, in the sense of the discussion of the preceding section, or possible energy-conserving four-phonon processes. On the other hand, it is a little difficult to see why these processes should be much more important than, say, the process of Fig. 6b, which is a modification of an energy-conserving four-phonon process. Probably, the experimental data at present hardly justify a detailed investigation of this question.

We notice, incidentally, that according to the theory developed in the present paper, Landau and Khalatnikov’s expression³ itself is incorrect in the limit of small dispersion. The effect of replacing their simple energy denominator by a Breit-Wigner type of expression will only be important if $\Omega/\Gamma \lesssim 1$, where Γ is the half-width of the intermediate state. A crude order-of-magnitude estimate using the experimental values of Γ at the longest wavelengths measured¹⁹ and the experimental value of the dispersion indicates that this condition is quite likely to be satisfied; if this is so, the Landau-Khalatnikov expression³ for the phonon-phonon scattering cross section and thus also Khalatnikov’s expression⁴ for the sound absorption coefficient are likely to be too large. The situation is, however, complicated, as five-phonon and higher order processes may partially compensate for this effect.

We must also note that at present the experimental data²⁰ on sound absorption in helium II at low temperatures are not really capable either of discriminating between the T^4 law predicted by Kawasaki⁵ and Simons⁹ on the basis of “linewidth” effects and the T^6 behavior derived by Khalatnikov,⁴ or of demonstrating that Khalatnikov’s expression is too large, as we have suggested here.

In the case of phonons in solids we are on more hopeful ground. The mechanism responsible for the decay of long-wavelength phonons in solids at low temperatures is predominantly collisions with phonons from regions of phase space near “degeneracy lines.”² It is extremely unlikely that the possibility of these processes is critically dependent on ω , so that the simplified approach is probably justified.

Condition (b) does not present much difficulty in practice, at least in the long-wavelength limit. The reason is that although there are matrix elements connecting the “true” initial and final states, namely those due to the quartic terms in the potential energy, in the long-wavelength limit they vanish compared to the “indirect” matrix elements which connect them via an intermediate state. If we assume that the wavevectors of all phonons involved are of the same order of magnitude $\sim q$, that is, that we are interested in the

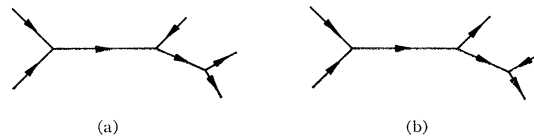


FIG. 6. Five-phonon processes. (a) Typical process that is not a mere modification of a four-phonon process; (b) one that is.

limit $q \rightarrow 0$, $k_B T \rightarrow 0$, $q'/k_B T$ constant, the “direct” elements are of order q^2 and the indirect ones of order q^3/Ω . As, in general,⁹ $\Omega \sim q^3$, the ratio vanishes as $q \rightarrow 0$.

In conclusion we note that there is nothing in our treatment which limits its applicability to a system of interacting phonons. However, there are few many-body systems apart from the phonon system where the lowest order collisions are forbidden; higher order effects are therefore on the whole of less interest.

ACKNOWLEDGMENTS

We should like to acknowledge helpful discussions with many members of the Department of Theoretical Physics, Oxford University, and especially R. V. Lange, W. E. Parry, C. J. Pethick, R. B. Stinchcombe, and M. J. Zuckermann. We also want to thank Dr. J. Wilks for interesting us in the problem of sound absorption in helium II and Dr. P. Carruthers for helpful correspondence. Finally we wish to express our gratitude to the Department of Scientific and Industrial Research and the United Kingdom Atomic Energy Authority for maintenance grants to one of us (A.J.L.).

APPENDIX A

The Basic Hamiltonian

In this Appendix we discuss the use of our basic Hamiltonian (2.1). The derivation is a standard one for the case of solids, if the cubic anharmonic terms are included. We shall therefore consider only the case of helium II. In that case we can, in the hydrodynamical model, write for V the expression

$$V = \frac{1}{3!} \frac{1}{\sqrt{V}} \left(\frac{\hbar \rho_0}{2c} \right)^{3/2} \sum_{\mathbf{k}\mathbf{k}'\mathbf{k}''} \delta_{\mathbf{k}+\mathbf{k}'+\mathbf{k}''} (kk'k'')^{1/2} \times \left[3 \frac{c^2 (\mathbf{k} \cdot \mathbf{k}')}{\rho_0^2 k k'} + \frac{\partial}{\partial \rho} \left(\frac{c^2}{\rho} \right) \right] u_{\mathbf{k}} u_{\mathbf{k}'} u_{\mathbf{k}''}, \quad (A1)$$

where the $u_{\mathbf{k}}$ are the displacement operators, ρ_0 is the equilibrium density, and $c^2 = \partial P / \partial \rho$. The annihilation and creation operators are related to the $u_{\mathbf{k}}$ by $u_{\mathbf{k}} \equiv a_{\mathbf{k}} + a_{-\mathbf{k}}^\dagger$. The effects we are interested in are those when the phonons involved are nearly parallel so that we can replace $(\mathbf{k} \cdot \mathbf{k}')/kk'$ by unity, whence (2.3) and (2.4) follow.

There is a difficulty in the case of helium II, absent in the case of a crystal. In the effects considered in the present paper a crucial role is played by the shape of

²⁰ C. A. Chase and M. A. Herlin, Phys. Rev. 77, 1447 (1955); K. Dransfeld, J. A. Newell, and J. Wilks, Proc. Roy. Soc. (London) A243, 500 (1957).

the $\omega(k)$ curve, and in particular by the sign of the departure of $\omega(k)$ from a linear law. In the case of a crystal this is no problem, as owing to the finite "structure" of the lattice there is a well-defined departure from a linear law, even in the harmonic approximation; in fact, for all known crystals the deviation is negative: normal dispersion.

In the hydrodynamic theory of helium II, however, the harmonic approximation is precisely the approximation of a perfectly elastic continuum, so that exactly $\omega(k) = ck$; the effects of the atomicity of the liquid enter only through the anharmonic terms. To obtain sensible results we must replace the theoretical "harmonic" spectrum by the "experimental" elementary-

excitation curve as deduced, for instance, from neutron scattering. This curve is almost certainly convex at very long wavelengths. The Hamiltonian (2.1) should thus give a reasonable description of the interaction of long-wavelength phonons in helium II.

Strictly speaking, this approach is incorrect as neutron-scattering experiments measure the energies $\tilde{\epsilon}_q$, as renormalized by the phonon-phonon interactions. We have therefore assumed that the $\omega(k)$ curve is really convex in the absence of interaction but use experimental data to determine $\tilde{\epsilon}_q$ rather than $\epsilon_q = \omega(q)$. The theoretical unsatisfactoriness of this approach is an inherent limitation of the hydrodynamic model of helium II.

Magnetic-Field Dependence of the Knight Shift in Superconductors*

PETER FULDE

Department of Physics, University of California, Berkeley, California

AND

KAZUMI MAKI†

The Enrico Fermi Institute for Nuclear Studies, The University of Chicago, Chicago, Illinois

(Received 1 March 1965)

Assuming that spin-orbit interaction is the reason for the appearance of a Knight shift in superconductors at zero temperature, we have calculated the magnetic-field dependence of the shift caused by the depairing effect of the field. Numerical results for the spin susceptibility as a function of the field are presented.

I. INTRODUCTION

THE occurrence of a shift in the frequency of the nuclear magnetic resonance (Knight shift)¹ in a superconductor at zero temperature has been attributed by Ferrell² and Anderson³ to the importance of spin-orbit interaction in small specimens. Because of this interaction, the component of the electron spin with respect to a certain axis will no longer be a good quantum number. In the superconducting state, electrons in mutually time-reversed states will be bound in pairs. Since this will no longer correspond to simple spin-up, spin-down electron pairing, the electrons in a pair will have the ability to respond with their spins to an imposed magnetic field. In this way a finite susceptibility arises even at zero temperature. If the applied field is comparable with the critical field of the sample, the tendency of the field to break the electron pairs will also have to be taken into account. This will affect the

response of the paired electrons to the field. Hence the Knight shift will become field dependent.

The pair-breaking effect of the field is incorporated into the theory by using the Green's-function formulation of the Knight-shift problem as given by Abrikosov and Gorkov.⁴ The treatment by those authors, which is valid for weak fields, will have to be generalized to arbitrary fields. The next section deals with the details of such a generalization. In the last section, we shall discuss the results.

II. CALCULATION OF THE SUSCEPTIBILITY

In the following we shall restrict ourselves to cases in which the critical field of the sample is such that contributions of the Pauli paramagnetism to the free energy are not yet of importance.⁵ Furthermore, we shall assume that the mean free path $v_F\tau$ between electron collisions is much smaller than the coherence distance. Since a Knight shift of the observed magnitude requires a mean free path $v_F\tau_1$ between spin flips of the order of

* Work supported in part by the Office of Naval Research and the National Science Foundation.

† On leave of absence from Research Institute for Mathematical Sciences, Kyoto University, Kyoto, Japan.

¹ F. Reif, *Phys. Rev.* **106**, 208 (1957); G. M. Androes and W. D. Knight, *ibid.* **121**, 779 (1961).

² R. A. Ferrell, *Phys. Rev. Letters* **3**, 262 (1959).

³ P. W. Anderson, *Phys. Rev. Letters* **3**, 325 (1959).

⁴ A. A. Abrikosov and L. P. Gorkov, *Zh. Eksperim. i Teor. Fiz.* **42**, 1088 (1962) [English transl.: *Soviet Phys.—JETP* **15**, 752 (1962)].

⁵ A. M. Clogston, *Phys. Rev. Letters* **9**, 266 (1962); B. S. Chandrasekhar, *Appl. Phys. Letters* **1**, 7 (1962).