Resonance in Phonon-Phonon Scattering*

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The scattering process, two phonons $\rightarrow$ two phonons, is examined in perturbation theory. One of the contributions of second order in the cubic anharmonic potential gives rise to an energy denominator that can vanish. This difficulty is removed by noting that the intermediate phonon has a complex self-energy associated with its instability in the presence of anharmonic forces. Approximating the irreducible self-energy by its lowest-order contribution, one finds a Breit-Wigner form of width equal to the decay rate, one phonon $\rightarrow$ 2 phonons, calculated to lowest order. An experiment is proposed to observe directly the predicted resonance. This experiment appears to be somewhat beyond the capabilities of present apparatus. The effect of the resonance on heat conduction, phonon drag, and the analogous resonance expected in spin-wave scattering, are not discussed in this paper.

DURING the course of a theoretical study of the higher-order effects of anharmonic forces and impurities[1] on phonon interactions in dielectric crystals, we have found a resonance in the scattering process, 2 phonons $\rightarrow$ 2 phonons. In the past this process has been ignored in comparison with the "elementary" process, one phonon $\leftrightarrow$ 2 phonons [Figs. 1(a), 1(b)], so important in the theory of thermal resistance of crystals.[2] The reason for this situation is that the scattering process either involves extra powers of the cubic anharmonic interaction V_3, as in Figs. 1(c) and 1(d), or the presumably small quartic terms V_4 (Figs. 1(e) and 1(f).[3] However, higher order effects in a perturbation series cannot be ignored if the energy denominators become small, or vanish. We find that it is easy to conserve both energy and momentum in the process of Fig. 1(c), so that in the conventional perturbation theory the energy denominator vanishes. Thus it is necessary to evaluate the propagator for the virtual phonon with more care. Since the phonon is unstable in the presence of anharmonicities, the self-energy is complex.[4] Thus we find a resonance denomination of Breit-Wigner form. The other scattering process of second order in V_3, in which the incident phonons are annihilated with one of a virtual triplet of phonons (Fig. 1(d)], is well behaved since the corresponding denominator never vanishes. In Fig. 1(e) the scattering due to the lowest order of V_4 is shown. The denominator corresponding to the process of Fig. 1(f) can vanish, but upon using the correct phonon propagator this term will be small compared to the graph 1(c). Hence we concentrate on the latter. A point worth noting is that the resonance under consideration is an example of what is called a "kinematical" resonance in the literature of elementary particle physics. Such resonances occur whenever the two colliding particles are coupled to an unstable intermediate state by an energy-momentum conserving process.

Now consider the process 1(c) in which phonons of wave vector $\mathbf{p}$ and $\mathbf{q}$ coalesce into a phonon $\mathbf{k}=\mathbf{p}+\mathbf{q}$, which in turn disintegrates into the final phonons $\mathbf{p}'$ and $\mathbf{q}'$. Setting $\hbar=1$ and ignoring umklapp processes, the energy denominator is $\omega(\mathbf{p})+\omega(\mathbf{q})-\omega(\mathbf{p}+\mathbf{q})$. According to well-known selection rules as explained by Ziman,[5] for example, this quantity vanishes only if the intermediate phonon belongs to the longitudinal branch (l) and the initial phonons are longitudinal and transverse (t) or both transverse. The final phonons $\mathbf{p}'$ and $\mathbf{q}'$ must likewise occur in the combination $l+t$ or $t+t$. For illustration we consider the fictitious isotropic dispersionless solid with constant longitudinal

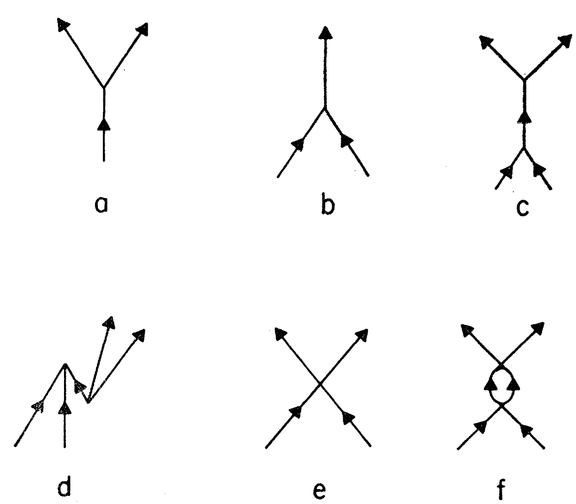

FIG. 1. (a) and (b) show the lowest order decay and absorption processes caused by the cubic anharmonic interaction V_3. (c) and (d) are the lowest order graphs for phonon-phonon scattering induced by V_3. The quartic interaction V_4 gives rise to scattering via processes like (e) and (f). The non-dangerous process of scattering by phonon exchange has been omitted from this figure.

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[1] In this paper we consider only "perfect" crystals in which there are anharmonic forces but no impurities.
[2] R. E. Peierls, *Quantum Theory of Solids* (Oxford University Press, New York, 1960), Chapter 2.
[3] The nth order anharmonic interaction term V_n involves n products of the phonon displacement field.
[4] P. Carruthers, Revs. Modern Phys. 33, 92 (1961). The graphical representation of phonon interactions is explained in Appendix D of this reference.
[5] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960), Chapter 3.

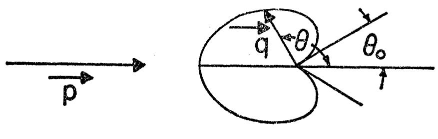

FIG. 2. The graphical solution of Eq. (2) is indicated for the longitudinal phonon l and the transverse phonon t. The minimum scattering angle θ_0 is given by $\cos\theta_0 = v_t/v_l$.

and transverse sound velocities v_l and v_t, $v_t < v_l$ with $\omega = vq$. For the transition $(\mathbf{p},l)+(\mathbf{q},t) \rightarrow (\mathbf{k},l)$ the energy conservation condition is

$$(p+\eta q)^2 = (\mathbf{p}+\mathbf{q})^2, \quad \eta \equiv v_t/v_l < 1, \tag{1}$$

which has the solution

$$q = 2p(\eta - \cos\theta)/(1-\eta^2). \tag{2}$$

In Eq. (2), θ is the angle between $\mathbf{p}$ and $\mathbf{q}$, given by $\cos\theta = \mathbf{p}\cdot\mathbf{q}/pq$. Since q must be positive, we find the restriction $\cos\theta \leq \eta$. A similar calculation for the process $t+t \rightarrow l$ leads to the more restrictive condition $\cos\theta \leq -1+\eta^2 < 0$. The minimum angle for which the denominator can vanish is thus given by

$$\cos\theta_0 = \eta = v_t/v_l, \quad l+t \rightarrow l; \tag{3}$$

$$\cos\theta_0 = -1+\eta^2 = -(1-v_t^2/v_l^2), \quad t+t \rightarrow l. \tag{4}$$

Figure 2 shows the graphical representation[6] of Eq. (2), which gives q in terms of p and $\cos\theta$. In actual crystals the effect of anisotropy is to make the conditions even less restrictive than the nondispersive case treated here, as shown by Herring.[7]

In order to study the analytic and physical properties of the complete phonon propagator it is essential to eliminate the so-called reducible self-energy parts. (If a graph falls into two disjoint pieces when one phonon line is cut, then that graph is reducible.) In our problem the effect of summing the reducible graphs is to replace $[E_0-\omega(\mathbf{k})]^{-1}$ by $[E_0-\omega(\mathbf{k})-\Sigma(\mathbf{k})]^{-1}$, where $E_0=\omega(\mathbf{p})+\omega(\mathbf{q})$ is the energy of the initial state and $\Sigma(\mathbf{k})$ is the sum of all irreducible contributions to the phonon self-energy. It is of course impossible to evaluate $\Sigma(\mathbf{k})$ exactly. However, the most important contribution to $\Sigma(\mathbf{k})$ is most likely just the second-order term. In Figs. 3(a) and 3(b) we show the contribution in second order. The contribution of Fig. 3(b) has no imaginary part, since the denominator in the intermediate state never vanishes. Hence its only effect is to renormalize the frequency of the bare phonon. Thus we do not consider this term further. The "bubble" of Fig. 3(a) has a contribution which we call $\Sigma_2(\mathbf{k})$. This was calculated in reference 4 to be

$$\Sigma_2(\mathbf{k}) = \delta\omega(\mathbf{k}) - \tfrac{1}{2}i\Gamma(\mathbf{k}), \tag{5}$$

where the real part $\delta\omega(\mathbf{k})$ serves to renormalize $\omega(\mathbf{k})$ and the positive quantity $\Gamma(\mathbf{k})$ is the rate at which the phonon $\mathbf{k}$ decays into phonons $\mathbf{p}$ and $\mathbf{k}-\mathbf{p}$ according to the process of Fig. 1(a):

$$\Gamma(\mathbf{k}) = 2\pi \sum_{\mathbf{p}} |\langle \mathbf{k}|V_3|\mathbf{p}\,\mathbf{k}-\mathbf{p}\rangle|^2 \times \delta[\omega(\mathbf{k})-\omega(-\mathbf{p})-\omega(\mathbf{k}-\mathbf{p})]. \tag{6}$$

Keeping only Fig. 3(a) in $\Sigma(\mathbf{k})$ corresponds to replacing the bare phonon propagator by the sum of the bubble graphs in Fig. 3(c).

Thus we obtain the following expression for the matrix element for phonon-phonon scattering:

$$M(\mathbf{p}+\mathbf{q}\rightarrow \mathbf{p}'+\mathbf{q}') = \sum_{\lambda}\left\{\frac{\langle \mathbf{p}'\mathbf{q}'|V_3|\mathbf{k}\lambda\rangle\langle \mathbf{k}\lambda|V_3|\mathbf{p}\mathbf{q}\rangle}{\omega(\mathbf{p})+\omega(\mathbf{q})-\omega(\mathbf{k},\lambda)+\frac{1}{2}i\Gamma(\mathbf{k},\lambda)}\right\}+M'. \tag{7}$$

In (7) the sum goes over the three polarization modes λ. For resonance scattering only the longitudinal mode is important. M' represents contributions from the processes like 1(d) and 1(c). These will be unimportant near resonance. The phonon frequencies appearing in (7) are renormalized. The width Γ is, not surprisingly, equal to the inverse of the lifetime of the phonon $\mathbf{k}$, according to (6).[8]

Equation (7) will be valid only in relatively pure crystals in which the decay rate (6) exceeds that due to impurities. For isotope and strain scattering the forward scattering amplitude vanishes in Born approximation[4] so that the lowest order contribution to $\Sigma(\mathbf{k})$ changes (7) only by adding to Γ the rate of decay due to these impurities calculated in lowest order. Thus the resonance is broadened and lowered by these impurities. The width in (7) is rather narrow, except possibly for phonons of frequency of the order of the Debye frequency, since $\Gamma(\mathbf{k})$ increases rapidly with k.

The successful detection of the proposed resonance would provide a direct measurement of the decay rate $\Gamma(\mathbf{k})$. The ideal experiment would involve two monochromatic beams of phonons at appropriate angles and polarizations. Varying the frequency of one beam through the resonance region then gives rise to a maximum in the absorption of the other beam. In order to minimize the background and absorption of beam phonons due to the presence of the "host" phonons in the lattice, very low temperatures are required (less than liquid helium temperatures). The decay of beam phonons by the process of Fig. 1(a) should not be a serious problem. In a perfect lattice the selection rules say that transverse phonons will not decay. If the longitudinal beam is detected, then requiring the detected phonon to have an energy equal to the initial

[6] This construction is due to Professor Wayne Bowers.

[7] C. Herring, Phys. Rev. 95, 954 (1954).

[8] *Note added in proof:* A few remarks are in order concerning the renormalization of $\omega(\mathbf{p})$ and $\omega(\mathbf{q})$. If one prepares a harmonic phonon at time 0, then it takes a time of order $1/\delta\omega$ for the frequency shift to occur. The imaginary part is not admixed until a time of order $\hbar/\Gamma$ has passed. Thus the imaginary part of the frequency change of the external phonons may be ignored if $i/\Gamma\tau \gg \hbar v$, where τ is the collision time for the process of Eq. (7).

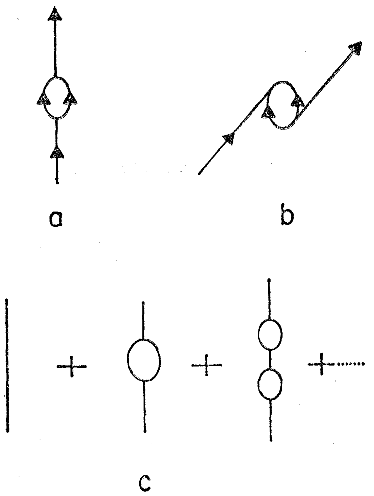

FIG. 3. (a) and (b) show the second-order (in V_3) contributions to the phonon self-energy. The sum of graphs indicated in (c) is the approximate phonon propagator obtained by approximating the irreducible self-energy by Fig. 1(a).

phonon will eliminate the decay events since the undetected phonon has zero energy.

In order to estimate the order of magnitude of the effect, we keep only the (resonant) longitudinal part of the sum in (7). Summing on all final states $\mathbf{p}'$ and $\mathbf{q}'$ gives for the rate at which the initial phonons collide:

$$w=\frac{\Gamma|\langle\mathbf{k}l|V_3|\mathbf{pq}\rangle|^2}{[\omega(\mathbf{p})+\omega(\mathbf{q})-\omega(\mathbf{k})]^2+\frac{1}{4}\Gamma^2}. \quad (8)$$

The total counting rate w_{tot} under the resonant curve obtained by varying $\omega(\mathbf{p})$ or $\omega(\mathbf{q})$ is $\int wdE_0$, $E_0=\omega(\mathbf{p})+\omega(\mathbf{q})$. Making the usual assumption that the matrix element is slowly varying over the width Γ, one obtains

$$w_{\text{tot}}=(2/\pi\hbar^2)|\langle\mathbf{k}l|V_3|\mathbf{pq}\rangle|^2, \quad (9)$$

on reinstating the factors of $\hbar$. Using the matrix elements given in reference 4, Eq. (9) becomes

$$w_{\text{tot}}=\frac{\hbar N(\mathbf{p})N(\mathbf{q})[N(\mathbf{k})+1]|C(\mathbf{pqk})|^2}{4\pi\rho^3\Omega^3\omega(\mathbf{p})\omega(\mathbf{q})\omega(\mathbf{k})}. \quad (10)$$

In Eq. (10), $N(\mathbf{p})$ is the occupation number for the pth mode, assumed to be due to the source alone. For sufficiently low temperatures $N(\mathbf{k})$ will be zero. Ω is the volume of the crystal; ρ is the mass density. Approximating the coupling coefficient $C(\mathbf{pqk})$ by Eq. (2.23) of reference 4 and introducing the intensities of the initial beams by $I(\mathbf{p})=N(\mathbf{p})v(\mathbf{p})/\Omega$, Eq. (10) becomes (for frequencies appreciably less than the Debye frequency)

$$w_{\text{tot}}\approx\hbar\xi^2F(pqka^3)I(\mathbf{p})I(\mathbf{q})\Omega/16\pi vM, \quad (11)$$

where ξ is a number, $\xi\approx40$, a is the lattice constant, F is a complicated angular factor of order unity,[9] v is an average sound velocity, and M is the mass per unit cell. The effect is clearly largest at high frequencies $ka=O(1)$. For frequencies almost within reach of present apparatus, $ka\approx pa\approx qa\approx10^{-2}$ ($\omega\approx10^{11}$/sec) using the constants: $v\approx3\times10^5$ cm/sec, $a\approx3\times10^{-8}$, $M\approx20$ amu, gives (per cm^3) $w_{\text{tot}}/(I(p)I(q))\approx10^{-20}$ cgs. In order that w_{tot} be 10^{16}/sec, we need $[I(p)=I(q)=I]$ a beam intensity of $I\approx10^{18}$ phonons/cm^2 sec. Converting to a power flux, $P=\hbar\omega I$, this is 10 μw/cm^2. Of course w_{tot} refers to *all* directions for the final phonons. If we introduce a factor 10^{-1} for the detector (say of area ≈1 cm^2), the scattered power flux in the peak is of order 10^{-2} μw. For lower frequencies the observable effect decreases rapidly.

In a subsequent paper the role of the proposed resonance and other higher order effects in the theory of thermal conductivity will be assessed.

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[9] However, in certain directions F vanishes.