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Spontaneous phonon decay selection rule: *N* and *U* processes

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We show that the spontaneous decay of a phonon by anharmonic processes of any order into a set of phonons of higher phase velocity is impossible for *both* N and U processes. The result is valid for crystals of arbitrary symmetry and anisotropy and in the presence of frequency and angular dispersion. The implications for the propagation of large-wave vector phonons is discussed.

The role of anharmonic phonon processes in determining the lattice thermal conductivity was first enunciated by Peierls.[1] He showed that N processes in which the "momentum" of the interacting phonons is conserved do not exhaust all possible processes and that umklapp processes involving a "flip over" into approximately the opposite direction by the addition of a vector of the reciprocal lattice can take place. Such U processes become important in thermal-conductivity-type experiments when the temperature is sufficiently high so that the dominant phonons which carry the heat correspond to a region roughly halfway into the Brillouin zone. Several recent high-frequency-phonon experiments have, however, revealed anomalously long phonon lifetimes at low temperatures.[2,3] In the case of phonons generated during *e-h* recombination[4] in semiconductors macroscopic mean free paths ($\sim$mm) have been observed. In this case the wave vector of the propagating phonons is believed to be close to the Brillouin-zone edge of the crystal. The selection rules for *spontaneous U*-process decay, not normally discussed in the thermal-conductivity or high-frequency-phonon-transport literature, could play a fundamental role in limiting the phonon lifetime of large-wave-vector phonons.

The theoretical work of Herpin,[5] of Klemens,[6] and of Orbach[7] established that for an isotropic solid transverse phonons cannot decay into two other phonons because of energy-momentum considerations. The purpose of this note is to point out that the selection rule against anharmonic transverse-phonon decay can be generalized to the theorem: A phonon cannot decay by anharmonic processes of any order into a set of phonons of higher phase velocity. This result is applicable in the presence of *frequency* and *angular dispersion* in an *anisotropic* crystal of *arbitrary symmetry*. It is applicable to *U processes* as well as *N processes*.

Proof. Conservation of crystal momentum requires that

$$\vec{k}+\vec{G}=\vec{k}_1+\vec{k}_2+\cdots+\vec{k}_n\ , \tag{1}$$

where $\vec{G}=0$ for N processes and $\vec{G}$ is a reciprocal-lattice vector for U processes. (Here $\vec{k}$ is the initial phonon; $\vec{k}_1, \vec{k}_2, \ldots, \vec{k}_n$ are the resulting phonons.)

By a generalization of the triangle inequality, a side of any polygon must be less than the sum of the remaining sides. Thus if $\vec{k}'=\vec{k}+\vec{G}$

$$k' \leq k_1+k_2+\cdots+k_n \tag{2}$$

(see Fig. 1). We will now establish lemma I

$$k \leq k_1+k_2+\cdots+k_n\ . \tag{3}$$

For N processes, $\vec{G}=0$ and the result is trivial. When $\vec{G}\neq 0$, $\vec{k}'$ is outside the first Brillouin zone. But a Brillouin zone is a proximity cell in reciprocal space, that is, *it contains all points closer to its center than to any other (reciprocal) lattice point.*[8] Since $\vec{k}'$ is in a cell with center at O' (see Fig. 1) the vector $\vec{k}''$ from O' to $\vec{P}'$ obeys

$$k'' < k' \tag{4}$$

since k' is the distance from P' to another lattice point at O. But $O'P'$ is parallel to OP and identical in

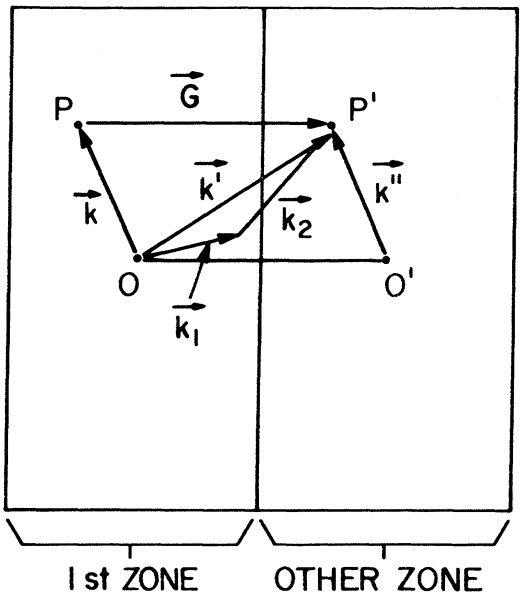

FIG. 1. Possible umklapp process for a phonon of wave vector $\vec{k}$ with the origin of the first zone being O and of the other zone O'. $\vec{G}$ is the reciprocal-lattice vector and $\vec{k}_1$ and $\vec{k}_2$ are the decay products (see text).

length:

$$\vec{k}'' = \vec{k};\ k'' = k \ . \tag{5}$$

Equations (2), (4), and (5) yield the desired lemma, Eq. (3).

Energy conservation yields

$$\omega(\vec{k}+\vec{G}) \equiv \omega(\vec{k}) = \omega(\vec{k}_1) + \omega(\vec{k}_2) + \cdots + \omega(\vec{k}_n) \tag{6}$$

or with $v_i = \omega(\vec{k}_i)/k_i$,

$$vk = v_1 k_1 + v_2 k_2 + \cdots + v_n k_n \ . \tag{7}$$

By hypothesis

$$v < v_i \ \text{ for all } i \tag{8}$$

so that

$$k > k_1 + k_2 + \cdots + k_n \ , \tag{9}$$

which contradicts Eq. (3). Thus energy and "momentum" conservation will not allow a low-velocity phonon to decay into a set of phonons of higher phase velocity.

In most materials the lowest velocity branch is a transverse branch with normal dispersion. Thus barring extreme anisotropy, this theorem will mean that the lowest transverse-acoustic branch will be unable to decay by anharmonic processes of any order, even with the help of U processes. Although the higher transverse-acoustic mode can, in principle, decay with the help of the lower acoustic branch the phase space will be so restricted by energy and momentum conservation that the decay rate will be small. Thus, in general, the selection rule for anharmonic decay will be of the type $l \rightleftarrows t + t$, or $l \rightleftarrows t + l$, where l stands for a longitudinal phonon and t for a transverse phonon. Even in highly anharmonic materials the lowest branch will be fundamentally stable at low temperatures and will not down-convert in frequency. Such behavior would then be analogous to the extremely short-wavelength-phonon-propagation predicted[9] and observed[10] in liquid helium where, of course, U processes are irrelevant. The propagation of such large-wave-vector phonons in crystalline materials will be limited only by the presence of scattering due to impurities or naturally occurring isotopes. We shall show elsewhere that in diatomic crystals some zone-boundary modes have zero motion for one of the two particles. If the nonvibrating atom has isotopes, but the other does not, isotope scattering will be forbidden. More generally, the motion will be shared between the two particles and isotope scattering will be reduced from the formula appropriate to the monatomic case. Such behavior may be expected, e.g., in fluorides, arsenides, and phosphides. In such materials (besides isotopically pure materials like solid He and Bi) a "window" for ballistic propagation of near-zone-edge phonons may occur. Even in other cases as long as the density of states for the transverse phonons is much larger than that of the longitudinal phonons, the elastic scattering by naturally occurring isotopes may result in diffusive propagation of these high-energy phonons but without frequency down-conversion for times which may often lie in the μs range for materials with "soft" TA branches. Such long-lived high-energy phonons and their propagation properties will play an important role in the understanding of nonradiative energy transport in semiconductors and insulators. Furthermore, because of their zone-edge nature such phonons will have a considerable spread in frequency and velocity. Thus their propagation, focusing, and scattering will be affected by a combination of angular and frequency dispersion. The correct numerically obtained vibrational spectrum of the solid must be used and care is needed in both the experiment and comparison with the theory.

Orbach[11] many years ago proposed that optical techniques may be used to generate high-frequency phonons. Our generalization of the decay processes to include U processes and crystals of arbitrary symmetry and anisotropy suggests that extremely short-wavelength acoustic phonons of the lowest branch can be extremely stable at low temperatures. The propagation of such phonons is *fundamentally inaccessible* to thermal-conductivity-type measurements which involve kT phonons but may be explored by any narrow-band high-frequency excitation technique at low temperatures.

Note added in proof. Maris [Phys. Lett. 17, 228 (1965)] has pointed out the long lifetime of slow

transverse phonons at low temperatures and has shown by numerical calculation that the selection rule of this paper is valid over a large portion of the Brillouin zone for an fcc crystal with nearest-neighbor central forces.

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