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LIFETIMES OF HIGH FREQUENCY PHONONS

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Abstract - We discuss the inhibition of anharmonic spontaneous multiphonon decay by energy-momentum conservation and the reduction of isotope scattering by the sharing of ionic motion to permit a frequency window for long-lived transverse acoustic phonons.

1. Introduction - Recent high frequency phonon experiments have revealed anomalously long phonon lifetimes at low temperatures. Indeed, for phonons generated by e−h recombination in GaAs mean free paths of a few mm have been observed for near zone boundary acoustic phonons.

Since elementary theories of isotope scattering and of two-phonon decay lead to scattering rates that grow as $\omega^4$ and $\omega^5$ respectively, the experimental results require some mechanism or mechanisms that inhibit such scattering processes at least over some frequency window near the zone boundary.

2. Spontaneous Phonon Decay - In an isotropic solid transverse phonons can not decay into two other phonons because of energy and momentum conservation. Maris recognized that the same conclusion would be likely to apply to an anisotropic crystal, but that conservation of energy and momentum would be model dependent. He demonstrated the result for a face-centered cubic crystal with central forces between nearest neighbor atoms.

Lax, Hu and Narayanamurti proved the following theorem: A phonon can not decay by anharmonic processes of any order into a set of phonons each of whose phase velocities is higher than that of the initial phonon. This result is applicable in the presence of frequency and angular dispersion in an anisotropic crystal of arbitrary symmetry. It is applicable to $U$ (umklapp) as well as $N$ (normal) processes.

The above theorem does not completely preclude the possibility that a transverse acoustic phonon can decay by anharmonic processes, but as found in Maris's example, only modes with small wave-vector $q$ can decay. The reason for this is that the phase velocity of a phonon will normally decrease with increasing $q$ making it more difficult to find phonons of lower phase velocity to decay into.

3. Isotope Scattering - To explain anomalously long-lived phonons, we must also demonstrate that isotope scattering, at least over some frequency "window", is weaker than expected from the usual theoretical treatments of monatomic lattices. We have suggested that this weakness is to be expected in multi-atomic lattices when not all of the constituents possess isotopes. If only one atom (say Ga) has isotopes, there is a reduction factor of the form $1 - \frac{\omega^2(q')}{\omega^2(q)} \cdot \frac{\omega^4(q)}{\omega^4(q')}$ where $\omega^4(q)$ is a (normalized) amplitude for Ga in a mode of type $t$ with propagation vector $q$. Here $t$ and $q$ refer to the initial phonon state and $t'$ and $q'$ refer to the final state.

For the case of GaAs, which has cubic site as well as space group symmetry, the initial and final state amplitudes can be disentangled and the decay rate $1/\tau(q)$ associated with a single scattering center in a lattice of $N$ sites is given by

$$\frac{1}{\tau(q)} = \frac{1}{12} \frac{\omega^2(q)}{N} \left| \omega^4(q) \right|^2 \frac{\left< (\Delta M_{\text{GaAs}})^2 \right>}{(M_{\text{GaAs}})^2} W(f); \text{ where } f = \omega(q)/2\pi$$

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Here all the initial state information is concentrated in \( \omega_f(q) \) and \( u^{Ga}(q) \) and the final state information is concentrated in the density of states weighted by the Ga motion:

\[
W(f) = \sum_{t'} g_t(f) R_{t'}(f) = \sum_{t'} W_{t'}(f) ; \text{ where } g_t(f) = <\delta(f - f_t(q'))> ,
\]

is the ordinary density of states for mode \( t' \) and

\[
g_t(f) R_{t'}(f) = <\delta(f - f_t(q'))> |u^{Ga}(q')|^2 > .
\]

Here \( f_t(q') = \omega_f(q')/2\pi \) is the frequency in Hertz of the scattered phonon, and the amplitudes are normalized in accord with

\[
|u^{Ga}(q)|^2 + |u^{As}(q)|^2 = 1 .
\]

The averages shown in Eqs. (3) and (4) are integrals over the Brillouin zone performed using the Gilat-Raubenheimer algorithm.\(^6\) \( W_{t'}(f) \) is the density of final states of type \( t' \) weighted by the Ga motion, and \( R_t(f) \) is the final state reduction factor at frequency \( f \) caused by the fact that the Ga motion contains only a portion of the kinetic energy. The factor \(|u_t(q)|^2\) is plotted versus \( q \) in the \([qq0]\) direction in Fig. 1a. Plots of \( g_t(f) \) and \( W_{t'}(f) \) for the acoustic modes are shown in Fig. 1b.

These calculations are based on a simplified extension from Ge to GaAs of Weber's adiabatic bond charge model\(^7\) by adding only an ionic charge asymmetry \( \Delta \) between Ga and As.\(^5\) The results for \( |u^{Ga}|^2 \) are sensitive to \( \Delta \) and favor \( \Delta < 0 \) for Ga.

4. Anharmonic Two Phonon Scattering - The extent to which two-phonon spontaneous decay of a phonon of wave-vector \( k \) and frequency \( \omega \) is possible is described by the two-phonon density of states

\[
g^2(\Omega, k) = <\sum_{G} \delta(\Omega - \Omega^+ (k,q))> \text{ where } \Omega^+ (k,q) = \omega_f(q) + \omega_r(k - q - G) ,
\]

embodies the simultaneous requirements of energy and momentum conservation with the sum over \( G \) including umklapp-processes.
Fig. 2. The two phonon density of states for pairs of transverse acoustic (TA) phonons of the slow (S) and fast (F) variety is plotted versus frequency for (a) \( k = [1,0,0] \), and (b) \( k = [0.3,0,0] \) phonons as slow transverse phonons.

For decay of a transverse phonon, the relevant final states are also transverse. We display \( g_{\alpha}(\omega,k) \) for \( t \) and \( t' \) slow (S) or fast (F) transverse (T) phonons, with \( k \) at \([3,0,0]\) and \([1,0,0]=X\) (the zone boundary point) in Figs. 2a and 2b respectively. Since the two phonon density of states exists only above some minimum \( \omega_m \), for any given \( k \), phonons whose \( \omega(k) < \omega_m \) can not decay, such as the T phonons at X. This conclusion is undoubtedly valid in all directions for large enough \( k \) permitting propagation over a substantial portion of \( k \) space.

5. Summary - We have shown that anharmonic decay by a phonon into two (or more) phonons is rigorously prevented if the final phonons have higher phase velocities than the initial phonon, and is strongly inhibited for acoustic transverse phonons near the zone boundary. Moreover, isotope scattering in GaAs is reduced near the zone boundary by two factors involving the squared amplitude of the Ga atoms. Thus a window is available for long-lived high frequency phonons. Since these results depend on the eigenvectors they provide a more sensitive test of vibrational models than measurements of dispersion alone.

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References


