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## Temperature dependence of phonon lifetimes in dielectric crystals

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We present calculations of the temperature dependence of the lifetime of phonons due to anharmonic interactions. The contributions to the lifetime arising from phonon decay and phonon-phonon collisions are considered separately and, in addition, the relative strength of normal and umklapp processes is determined. The calculations are performed for an fcc lattice with nearest-neighbor central forces.

### I. INTRODUCTION

Phonon scattering in crystalline dielectrics can arise from both extrinsic and intrinsic mechanisms.<sup>1-3</sup> The extrinsic processes include scattering of phonons at dislocations, point defects, grain boundaries, and the exterior surfaces of the sample. In addition, mass-defect scattering occurs in most crystals because of the variation of mass between the different stable isotopes of the elements involved. In the absence of these extrinsic processes phonon lifetimes are determined by the rate at which anharmonic processes occur. In the lowest order of perturbation theory these are three-phonon processes which can be divided into collision (two phonons collide to produce a third) and decay (one phonon decays into two) processes. The events can be further subdivided into normal processes ( $N$ ) in which crystal momentum is conserved and umklapp processes ( $U$ ) in which the momentum changes by a reciprocal-lattice vector.

These processes have traditionally been studied through thermal conductivity measurements. The kinetic formula

$$\kappa = \frac{1}{3} C v^2 \tau \quad (1)$$

relates the thermal conductivity  $\kappa$  to the specific heat per unit volume  $C$ , the average phonon group velocity  $v$ , and the average phonon scattering time  $\tau$ . Thus, assuming an appropriate value of  $v$  one can estimate  $\tau$  from the measured value of  $\kappa$ .<sup>4</sup> The  $\tau$  determined in this way includes only the effect of "resistive processes," i.e., those that do not conserve momentum. More recently a variety of new techniques have been invented to investigate phonon scattering.<sup>5</sup> However, many of these techniques (e.g., tunnel junction studies) are only applicable to low-frequency phonons ( $\omega \ll \Omega_D$ ,  $\Omega_D$  is the Debye frequency), or to low temperatures, and therefore quantitative results for phonon interaction rates as a function of temperature, phonon wave vector  $\mathbf{k}$  and polarization  $j$  are not available.

In this paper we present calculations of the rates of phonon-phonon interactions via anharmonic processes for a simple model of a dielectric crystal. The aim is to

obtain quantitative results that can be used to provide at least a rough estimate of phonon lifetimes in real crystals. In addition, we are able to compare the relative strengths of normal and umklapp processes, and to study how the lifetime varies with  $\mathbf{k}$  and  $j$ .

### II. MODEL

As a simple model we consider an fcc lattice of atoms, each of mass  $M$ , which interact via nearest-neighbor central forces. We have previously used this model to calculate the rate of spontaneous decay of phonons at  $T=0$  K,<sup>6</sup> and other aspects of anharmonicity within this model have been studied by Maradudin and co-workers.<sup>7,8</sup> In addition, Maradudin, Fein, and Vineyard<sup>9</sup> have used this model to estimate the lifetimes of longitudinal and transverse phonons propagating in the [100] direction of lead at one particular temperature (425 K). As far as the three-phonon process is concerned the model is completely specified by the values of the atomic mass, the lattice parameter  $a_0$ , and the second and third derivatives  $\phi''$  and  $\phi'''$  of the interatomic potential evaluated at the nearest-neighbor distance. These parameters can be related to the bulk modulus  $B$ , the volume thermal-expansion coefficient  $\alpha$ , and the Grüneisen parameter  $\gamma$  by<sup>6</sup>

$$\phi'' = \frac{3a_0 B}{4}, \quad (2)$$

$$\phi''' = -\frac{3B^2 a_0^3 \alpha}{4\sqrt{2}k_B} = -\frac{3Ba_0^3 \gamma C}{4\sqrt{2}k_B}, \quad (3)$$

where we have used the relation  $\gamma = \alpha B / C$ . The lattice parameter can be related to the density  $\rho$  by  $a_0 = (4M/\rho)^{1/3}$ . The phonon-dispersion curves based on this model are shown in Fig. 1. The maximum phonon frequency  $\omega_{\max}$  is  $(8\phi''/M)^{1/2}$ , and the low-temperature limiting value of the Debye temperature is  $\Theta_D = 2.965(\hbar/k_B)(\phi''/M)^{1/2}$ .<sup>10</sup> For any material one can use Eqs. (2) and (3) to choose the values of  $\phi''$  and  $\phi'''$  to correspond to the experimental values of  $B$  and  $\alpha$ . For crystals with more than one atom in the unit cell the

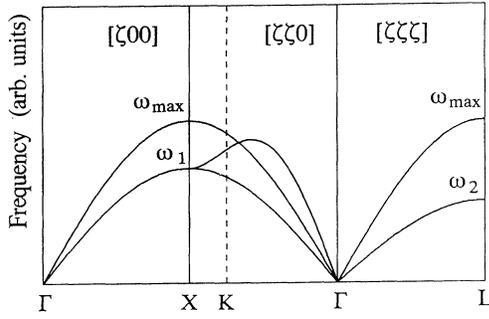


FIG. 1. Frequency as a function of wave vector for phonons propagating in the principal crystallographic directions of the fcc lattice. The maximum phonon frequency  $\omega_{\max}$  is  $(8\phi''/M)^{1/2}$ . The frequencies  $\omega_1$  and  $\omega_2$  have the values  $\omega_{\max}/\sqrt{2}$  and  $\omega_{\max}/2$ , respectively.

mass  $M$  can be chosen as the total mass per unit cell, but of course the model completely fails to include the optical models for such crystals.

### III. CALCULATION OF INTERACTION RATES

General expressions for the interaction rate of a phonon of wave vector  $\mathbf{k}$  and polarization  $j$  as determined by the three-phonon process have been given by Maradudin and Fein.<sup>11</sup> The total rate can be divided into two parts

as follows:

$$\Gamma_{\text{decay}} = \frac{\pi\hbar}{8N\omega} \sum_{1,2} |\Phi(\mathbf{k}j; -\mathbf{k}_1j_1; -\mathbf{k}_2j_2)|^2 \times \frac{(n_1 + n_2 + 1)}{\omega_1\omega_2} \Delta(\mathbf{k} - \mathbf{k}_1 - \mathbf{k}_2) \times \delta(\omega - \omega_1 - \omega_2), \quad (4)$$

$$\Gamma_{\text{coll}} = \frac{\pi\hbar}{4N\omega} \sum_{1,2} |\Phi(\mathbf{k}j; \mathbf{k}_1j_1 - \mathbf{k}_2j_2)|^2 \times \frac{(n_1 - n_2)}{\omega_1\omega_2} \Delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2) \times \delta(\omega + \omega_1 - \omega_2). \quad (5)$$

$\Gamma_{\text{decay}}$  is the rate at which the phonon  $\mathbf{k}j$  decays into two lower energy phonons and  $\Gamma_{\text{coll}}$  is the rate at which the phonon collides with other phonons to produce a third phonon. In these equations  $\omega$  is the frequency of the phonon  $\mathbf{k}j$ , the summations are over the wave vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$  and polarizations  $j_1$  and  $j_2$  of phonons 1 and 2, and  $\omega_1$ ,  $\omega_2$ , and  $n_1$ ,  $n_2$  are the frequencies and occupation numbers for phonons 1 and 2, respectively. The function  $\Delta(\mathbf{K})$  is unity if  $\mathbf{K}$  is a reciprocal-lattice vector or zero, and is zero otherwise. The  $\Phi$  parameters can be regarded as Fourier transforms of the cubic anharmonic part of the interatomic potential. For the fcc lattice that we are considering the explicit form of  $\Phi$  is

$$\Phi(\mathbf{k}j; \mathbf{k}_1j_1; \mathbf{k}_2j_2) = (2i)^3 \frac{\phi'''}{2(2M)^{3/2}} \sum_{\mathbf{n}} \exp\left[\frac{ia_0}{4} \mathbf{n} \cdot (\mathbf{k} + \mathbf{k}_1 + \mathbf{k}_2)\right] [\mathbf{n} \cdot \mathbf{e}(\mathbf{k}j)] \times [\mathbf{n} \cdot \mathbf{e}(\mathbf{k}_1j_1)] [\mathbf{n} \cdot \mathbf{e}(\mathbf{k}_2j_2)] \sin\left[\frac{a_0 \mathbf{n} \cdot \mathbf{k}}{4}\right] \sin\left[\frac{a_0 \mathbf{n} \cdot \mathbf{k}_1}{4}\right] \sin\left[\frac{a_0 \mathbf{n} \cdot \mathbf{k}_2}{4}\right], \quad (6)$$

where  $\mathbf{n}$  is a vector with integer components whose sum is even, and the sum over  $\mathbf{n}$  is restricted to those vectors such that  $\frac{1}{2}a_0\mathbf{n}$  is one of the vectors going from an atom in the fcc lattice to its 12 nearest neighbors.

From these results it is straightforward to show that the interaction rates  $\Gamma_{\text{decay}}$  and  $\Gamma_{\text{coll}}$  can be written in the forms

$$\Gamma_{\text{decay}}(\mathbf{k}j) = \Gamma_0 G_{\text{decay}}(\mathbf{k}j, T/\Theta_D), \quad (7)$$

and

$$\Gamma_{\text{coll}}(\mathbf{k}j) = \Gamma_0 G_{\text{coll}}(\mathbf{k}j, T/\Theta_D), \quad (8)$$

where

$$\Gamma_0 = \frac{\hbar\gamma^2\rho^{2/3}}{M^{5/3}}, \quad (9)$$

and  $G_{\text{decay}}$  and  $G_{\text{coll}}$  are dimensionless functions of the wave vector (expressed in terms of reciprocal-lattice vec-

tors, for example), the polarization, and temperature. Values of the coefficient  $\Gamma_0$  calculated for several crystals are listed in Table I.<sup>6</sup>

In the calculation of the interaction rates from Eqs. (4)

TABLE I. The values of the decay rate parameter  $\Gamma_0$  for several crystals as calculated from Eq. (9).

Crystal	$\Gamma_0$ $10^{12} \text{ sec}^{-1}$
Ne	0.29
Ar	0.10
Kr	0.044
Xe	0.024
LiF	0.097
NaF	0.040
Si	0.0032
Ge	0.0018

and (5) the sum over the wave vector of either phonon 1 or 2 can be performed trivially because of the presence of the  $\Delta(\dots)$  factor. This factor fixes the value of  $\mathbf{k}_2$  for a given  $\mathbf{k}_1$ . The result for the sum then has the general form

$$\sum_1 f(\mathbf{k}_1 j_1) \delta(\omega - \Omega_1), \quad (10)$$

where  $\Omega_1 \equiv \pm\omega_1 + \omega_2$ . The function  $f$  varies smoothly as  $\mathbf{k}_1$  is varied. Equation (10) has the same general form as the expression for the one-phonon density of states. An accurate and rapid method for the evaluation of this type of sum has been found by Gilat and Raubenheimer.<sup>12</sup> With some modification we have applied their sophisticated method of Brillouin-zone integration to the determination of  $\Gamma_{\text{decay}}$  and  $\Gamma_{\text{coll}}$ . Briefly, the approach is the following. The first Brillouin zone is divided into small cubic cells with sides of length  $\pi/na_0$  where  $n$  is an integer. One then finds those cubic cells within which there is a section of the surface on which the quantity  $\omega - \Omega_1$  vanishes. The area of this section, assumed to be a plane, is calculated using the dispersion relation for the fcc model that we are using.  $f(\mathbf{k}_1 j_1)$  is taken to be a constant throughout the cell. In the present calculation we have chosen values of  $n$  between 30 and 40, and have checked that the results are essentially unchanged if the cell size is made smaller. It was possible to make some reduction in the required computation time through the use of symmetry. Thus, for example, for  $\mathbf{k}$  along the [100] direction it was only necessary to sum  $\mathbf{k}_1$  or  $\mathbf{k}_2$  over one-eighth of the Brillouin zone. When the magnitude of  $\mathbf{k}$  was small it was necessary to use a finer mesh of points to obtain accurate results. The time to calculate the interaction rates for a single wave vector  $\mathbf{k}$  and polarization  $j$  and for a given choice of  $j_1$  and  $j_2$  was typically at 15 sec on a HITAC S-820 supercomputer.

#### IV. NUMERICAL RESULTS

For definiteness we assign "polarizations" to the three phonon modes with a given wave vector according to their frequency. In order of increasing frequency we label the modes slow transverse (ST), fast transverse (FT), and longitudinal ( $L$ ). For low frequencies,  $L$  phonons have polarization vectors approximately parallel to the wave vector, and FT and ST are approximately transversely polarized. However, for high-frequency phonons in some parts of the Brillouin zone the polarization vectors are unrelated to the label that is assigned in this way so, for example, the mode  $L$  does not necessarily have a polarization vector even approximately parallel to the wave vector.

##### A. Longitudinal phonons

In Fig. 2 we show the total interaction rates  $G_{\text{total}}$  for  $L$  phonons propagating in the three principal directions. For phonons propagating in the [100] direction the rate is shown divided into contributions from decay and collision processes in Fig. 3. We note the following features of these results.

##### 1. Decays

(1) At  $T=0$  the decay rate increases rapidly with increasing wave number. For very small  $k$  the decay rate of  $L$  phonons is known to vary as  $k^5$ . This strong dependence on  $k$  arises because both the matrix element and the region of phase space into which decay can occur increase.

(2) Regardless of the magnitude of the wave vector, the temperature dependence of the decay rate is similar [see Fig. 3(a)]. At low  $T$ ,  $G_{\text{decay}}$  is independent of  $T$  and at high  $T$  there is a linear temperature dependence. This behavior follows directly from consideration of the temperature dependence of the phonon occupation numbers in Eq. (4). The increase in occupation numbers for the phonon states into which decay can occur increases the probability of decay. The range in which the temperature dependence is linear begins when the temperature is high enough that most of the phonon states into which the phonon can decay begin to be thermally occupied.

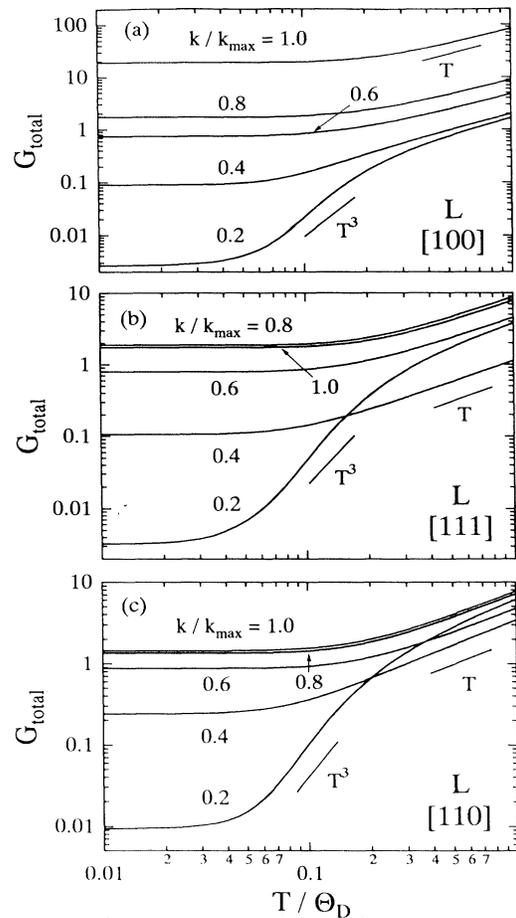


FIG. 2. Total interaction rate  $G_{\text{total}}$  for  $L$  phonons propagating in principal directions as a function of wave number and temperature  $T$ . The curves are labeled by the wave number  $k$  divided by the wave number  $k_{\text{max}}$  at the zone boundary in the same direction.

From Fig. 3(a) it is seen that this transition occurs at around  $k_B T \approx \hbar\omega/5$ .

## 2. Collisions

The behavior of the collision contribution to the interaction rate is considerably more complex.

(3) At high temperatures ( $T \approx \Theta_D$ ) the rate varies linearly with temperature because the phonon occupation numbers in Eq. (5) have this temperature dependence. The collision rate increases with increasing wave number in the range up to about  $0.2k_{\max}$  where  $k_{\max}$  is the wave number at the zone boundary, but decreases rapidly as  $k$  approaches  $k_{\max}$ . The reasons for this are as follows. At high temperatures most of the thermal phonons with which a phonon  $\mathbf{k}$  can collide have high frequency. An  $L$  phonon with  $\mathbf{k}$  near to the zone boundary has a frequency close to the maximum frequency in the lattice, and thus cannot collide with a typical thermal phonon because this would lead to the production of a phonon that would lie outside the range of lattice frequencies. Hence, the collision rate decreases as the wave vector increases, and must vanish for a phonon that has a frequency equal to

$\omega_{\max}$ . At low temperatures the average frequency of the thermal phonons is small and so this suppression does not occur until  $\omega$  is very close to  $\omega_{\max}$ .

(4) For sufficiently small temperature the frequency of the typical thermal phonon ( $\omega_1 \sim k_B T/\hbar$ ) becomes much less than the frequency  $\omega$  of the phonon  $\mathbf{k}$ . In a collision process the conditions of conservation of energy and momentum are

$$\omega + \omega_1 = \omega_2, \quad (11)$$

$$\mathbf{k} + \mathbf{k}_1 = \mathbf{k}_2. \quad (12)$$

Thus, the phonon 2 that is produced must have nearly the same frequency and momentum as the phonon  $\mathbf{k}$ . It follows that since  $\mathbf{k}$  is an  $L$  phonon, the phonon 2 must also be an  $L$  phonon. In addition, one can show from Eqs. (11) and (12) that the collision can take place only if the phase velocity of phonon 1 is less than the group velocity of the phonon  $\mathbf{k}$ . Hence, possible processes are

$$L + ST \rightarrow L, \quad L + FT \rightarrow L. \quad (13)$$

If  $\mathbf{k}$  is not too large the group velocity of the phonon  $\mathbf{k}$  will be greater than the velocity of transverse phonons and these collision processes can occur. This gives a collision rate varying as  $T^3$ . This type of behavior can be seen in Fig. 3(b) for  $k = 0.2k_{\max}$  and  $k = 0.4k_{\max}$ . It follows from the results of Herring<sup>13</sup> that in this range of  $k$  the interaction rate varies approximately as  $k^2$ . For larger  $k$  the increasing effect of phonon dispersion has the consequence that the group velocity of phonon  $\mathbf{k}$  decreases to a value below the transverse velocity and the interaction rate decreases dramatically, implying that decay processes dominate the collisions.

(5) In an important paper Herring<sup>13</sup> has investigated the collision rate of *low-frequency longitudinal phonons*, and argued that these phonons may play a special role in heat conduction in crystals. He showed that the interaction rate for these  $L$  phonons is dominated by collisions of type

$$L + ST \rightarrow FT, \quad (14)$$

where the  $ST$  and  $FT$  phonons have wave vectors that lie close to points, lines, or surfaces of degeneracy in the Brillouin zone. For the cubic crystal that we have considered here, Herring's results imply that the collision rate should vary as  $\omega^2$ . As Herring emphasizes, his analysis is limited to consideration of what happens for longitudinal phonons of frequency much less than the frequency of the phonons with which they collide ( $\omega \ll \omega_1$ ). In a particular case that he analyzes in detail he finds that the proportionality to  $\omega^2$  should hold fairly well when  $\omega < 0.05\bar{\omega}_1$ , where  $\bar{\omega}_1$  is the average frequency of the  $ST$  thermal phonons, but should deviate rapidly from an  $\omega^2$  law at higher frequencies. Our results confirm these earlier conclusions of Herring. In Fig. 4 we show the total interaction rate of low-energy longitudinal phonons propagating in the  $[100]$  direction as a function of temperature and frequency. Investigation of the contributions to the interaction rate from the different possible processes reveals that the rate is dominated by the  $L + ST \rightarrow FT$

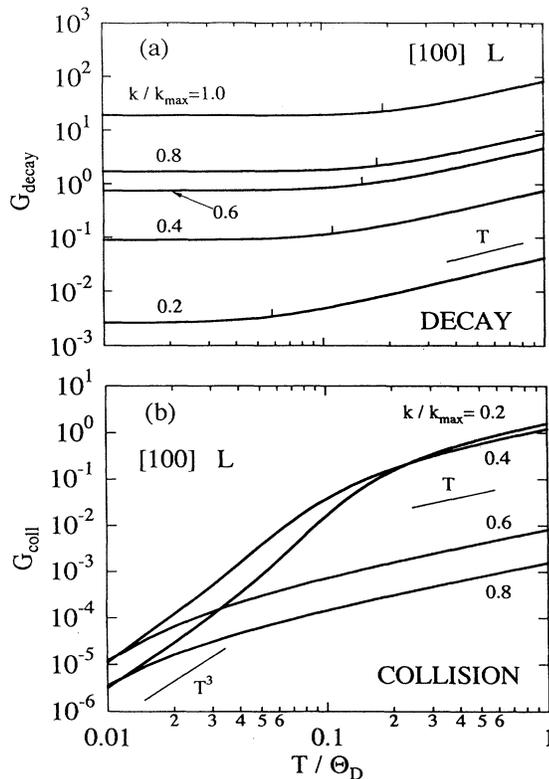


FIG. 3. Interaction rates for  $L$  phonons propagating in the  $[100]$  direction as a function of wave number and temperature  $T$ . The rates are separated into (a) decay  $G_{\text{decay}}$  and (b) collision processes  $G_{\text{coll}}$ . The curves are labeled by the wave number  $k$  divided by the wave number  $k_{\max}$  at the zone boundary in the same direction. Note that  $G_{\text{coll}} = 0$  for  $k/k_{\max} = 1$ . The small vertical lines in part (a) show the temperatures corresponding to  $k_B T = \hbar\omega/5$ .

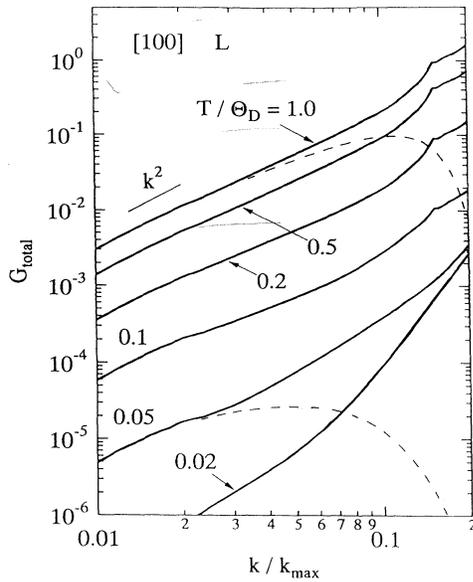


FIG. 4. Total interaction rate  $G_{\text{total}}$  for low-frequency longitudinal phonons propagating in the [100] direction as a function of temperature  $T$  and wave number  $k$  divided by the wave number  $k_{\text{max}}$  at the zone boundary. The dashed curves show the contributions from the process  $L+ST \rightarrow FT$ . The curves are labeled by the temperature  $T$  divided by the Debye temperature  $\Theta_D$ .

process, provided that the frequency is sufficiently low. When  $T = \Theta_D$  one finds that the interaction rate varies as  $\omega^2$  (or  $k^2$ ) for frequencies below about 0.05 of the maximum lattice frequency (or  $k \leq 0.03k_{\text{max}}$ ). At lower temperatures the upper frequency limit of the  $\omega^2$  regime for the  $L+ST \rightarrow FT$  process occurs at a lower frequency. This is to be expected on the basis of Herring's ideas because  $\bar{\omega}_1$  decreases as the temperature is lowered.

### B. Transverse phonons

(6) The total interaction rates (including decay and collision) for FT phonons propagating in the [110] direction are shown in Fig. 5. These results are qualitatively similar to the corresponding results for  $L$  phonons (Fig. 2). However, it is important to note that generally the magnitude of the scattering rate for the FT phonons is significantly less than the rate for  $L$  phonons.

(7) The interaction rates for ST phonons propagating in the [100] direction are shown in Fig. 6.<sup>14</sup> The behavior at low temperatures is quite different from that seen for  $L$  and FT phonons. In the [100] direction ST phonons can decay only if they have a wave number less than a critical value  $k_c$  which is approximately  $0.55k_{\text{max}}$ , where  $k_{\text{max}}$  is the wave number at the zone boundary in this direction. This follows from the conditions of conservation of energy and momentum.<sup>6</sup> Thus, for  $k > k_c$  the total interaction rate vanishes as  $T \rightarrow 0$ , whereas for  $k < k_c$  the rate tends to a constant in this limit. A similar behavior is

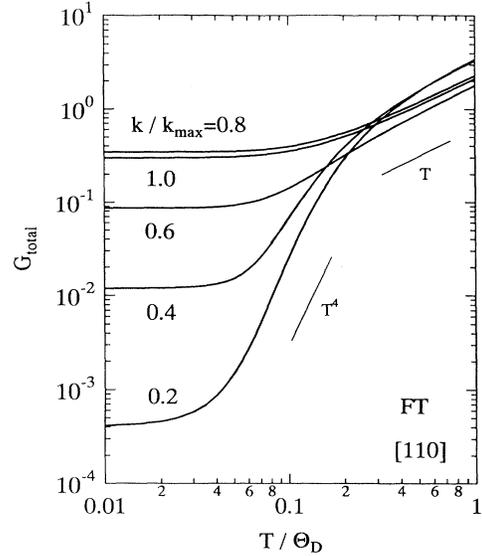


FIG. 5. Total interaction rate  $G_{\text{total}}$  for fast transverse (FT) phonons propagating in the [110] direction as a function of wave number and temperature  $T$ . The different curves are labeled by the wave number  $k$  divided by the wave number  $k_{\text{max}}$  at the zone boundary in the [110] direction.

found in the [111] direction but with a different value of  $k_c$ . In the [110] direction  $k_c$  is zero, i.e., no ST phonons can decay.

### C. Umklapp processes

The collision and decay processes can be divided into normal ( $N$ ) and umklapp ( $U$ ) processes. For the um-

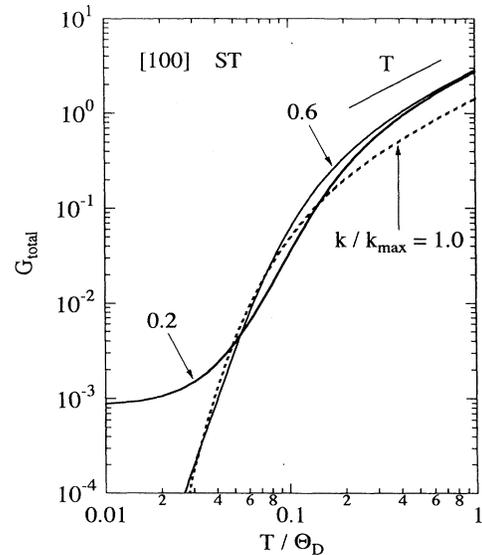


FIG. 6. Total interaction rate  $G_{\text{total}}$  for slow transverse phonons propagating in the [100] direction as a function of wave number and temperature  $T$ . The different curves are labeled by the wave number  $k$  divided by the wave number  $k_{\text{max}}$  at the zone boundary in the [100] direction.

klapp processes the condition of conservation of crystal momentum is

$$\mathbf{k} + \mathbf{k}_1 = \mathbf{k}_2 + \mathbf{g} \quad (15)$$

for collisions, and

$$\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 + \mathbf{g} \quad (16)$$

for decay processes, where  $\mathbf{g}$  must be a nonzero reciprocal-lattice vector. The results shown in Figs. 1–6 are the sum of the rates at which normal and umklapp processes take place, i.e., plots of  $G_{\text{total}} = G_{\text{total}}^{\text{normal}} + G_{\text{total}}^{\text{umklapp}}$ . In Fig. 7 we show for selected phonons the fraction  $f_U$  of the total rate that comes from umklapp processes. The relative strength of umklapp processes depends in a complex way on the phonon wave vector and on the temperature.

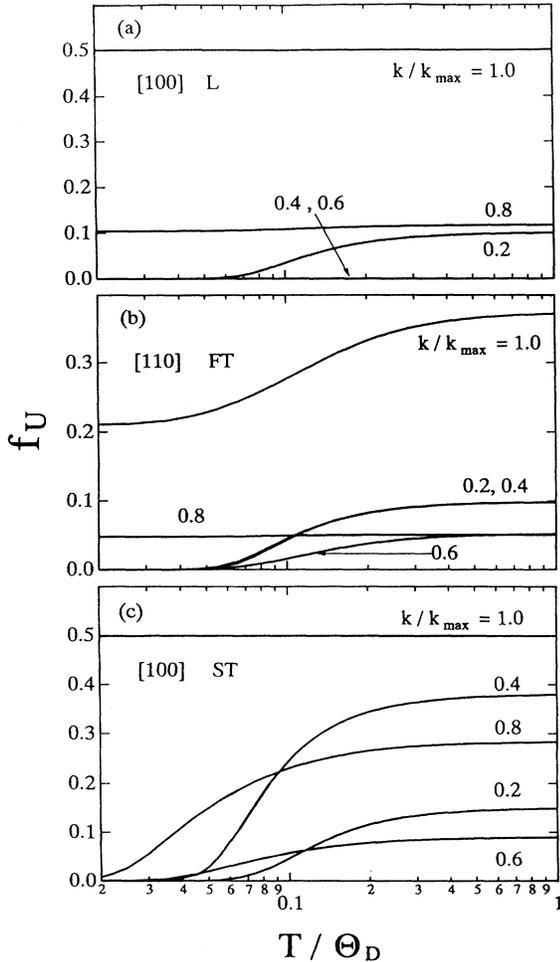


FIG. 7. Fraction  $f_U$  of interaction rate that arises from umklapp processes as a function of the wave number and the temperature  $T$ . (a) Longitudinal phonons propagating in the [100] direction, (b) fast transverse phonons in the [110] directions, and (c) slow transverse phonons in the [100] direction.  $k$  is the phonon wave number, and  $k_{\text{max}}$  is the wave number at the zone boundary in the given direction.

### 1. *L* phonons

In Fig. 7(a) we show  $f_U$  as a function of temperature for *L* phonons propagating along [100]. We find qualitatively the same temperature dependence in the [110] and [111] directions.  $f_U$  has the value  $\frac{1}{2}$  when the wave vector lies on the zone boundary at the *X* point (and also at the *L* point). These phonons have frequency equal to the highest frequency of any phonon in the lattice, and thus cannot collide with another phonon but can only decay into lower-frequency phonons. To understand why  $f_U$  is equal to  $\frac{1}{2}$  for the *L* phonon at the *X* point, consider a particular *N*-process decay mode into phonons with wave vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$  having components  $(k_{1x}, k_{1y}, k_{1z})$  and  $(k_{2x}, k_{2y}, k_{2z})$ . Since at the *X* point the wave vector is  $(2\pi/a_0)(1, 0, 0)$  where  $a_0$  is the lattice parameter, we must have  $k_{1x} + k_{2x} = 2\pi/a_0$ . The *L* phonon at the *X* point can also decay via a *U* process into phonons  $\mathbf{k}'_1$  and  $\mathbf{k}'_2$  with components  $(-k_{1x}, k_{1y}, k_{1z})$  and  $(-k_{2x}, k_{2y}, k_{2z})$ . The frequencies of these two phonons are the same as the frequencies of the phonon produced in the *N*-process decay and the magnitude of the matrix element for the event is also the same. Thus, for every *N*-process decay mode there is a matching *U*-process decay mode which occurs at the same rate. A similar argument can be used to explain why  $f_U$  is  $\frac{1}{2}$  at the *L* point.

As the wave vector is reduced, the value of  $f_U = G_{\text{total}}^{\text{umklapp}}/G_{\text{total}}$  decreases rapidly and for  $k/k_{\text{max}}$  of 0.4 and 0.6,  $f_U = 0$ . These phonons have momenta too far from the surface of the Brillouin zone for them to be able to decay via a *U* process. In addition, they are also unable to undergo *U*-process collisions. To see this let us consider as a specific case the collisions that can occur when  $k/k_{\text{max}} = 0.4$ . It is clear that the conditions of conservation of energy and momentum make it impossible for the *L* phonon to collide with another *L* phonon. The allowed processes thus involve collisions with ST or FT phonons. The longitudinal phonon with  $k/k_{\text{max}} = 0.4$  has a frequency  $\omega_{\text{max}} \sin(0.4\pi/2) = 0.59\omega_{\text{max}}$ . Thus, the highest frequency transverse phonon with which this phonon can collide has frequency  $\omega_1 = 0.41\omega_{\text{max}}$ . The magnitude of the wave vector of such a transverse phonon depends on the propagation direction (in the [100] direction it is approximately  $0.4k_{\text{max}}$ ), but it is always too small to enable an umklapp process to occur.

When the wave vector is reduced further, *U* processes again become possible [see curve for  $k/k_{\text{max}} = 0.2$  in Fig. 7(a)]. The frequency  $\omega$  is now small enough that  $\omega_1$  approaches the upper limiting frequency for transverse phonons. Thus, the phonon 1 can have a wave vector close to the zone boundary and umklapp processes are again possible. The rate at which this process occurs is proportional to the number of phonons 1 which have sufficiently large wave vector to produce an umklapp process. This number decreases rapidly as the temperature is lowered giving the temperature dependence shown in Fig. 7(a).

### 2. *FT* phonons

Results of calculations for the [110] direction are shown in Fig. 7(b). Note that according to the definition

we have adopted, the FT mode for any wave vector is always the mode of second highest frequency. Thus, in the [110] direction the dispersion relation (Fig. 1) for this mode is complicated, and has a discontinuous change in slope at the wave number  $k'_c = 8k_{\max}/9$ , where  $k_{\max}$  is the zone boundary wave number in this direction. For  $k = k_{\max}$  the umklapp fraction is 0.37 when  $T = \Theta_D$ . At this temperature these phonons have comparable rates for collision and decay. Both collision and decay can give an umklapp process, but the fraction of umklapp processes is larger for collision events than for decay processes. As the temperature is lowered the rate of collisions decreases more rapidly than the number of decays, and hence there is a decrease in the fraction of the total number of interactions which are umklapp. On the other hand for  $k = 0.8k_{\max}$ , which is below  $k'_c$ , the decay rate at all temperatures is greater than the collision rate by at least a factor of 6. Thus, the overlap umklapp fraction is effectively determined by the umklapp fraction for the decay rate, which is nearly independent of temperature. For  $k = 0.2, 0.4$ , and  $0.6k_{\max}$ , the only umklapp processes come from the collision contribution. Thus, the fraction decreases rapidly at low temperatures because there are very few thermal phonons of large enough momentum to combine with the FT phonon to produce a phonon outside the Brillouin zone.

### 3. ST phonons in the [100] direction (Ref. 14)

Results are shown in Fig. 7(c). For these phonons the decay process can only occur for  $k$  less than the critical wave number  $k_c \approx 0.55k_{\max}$ , and all of these decays are  $N$  processes. Thus, for  $k > k_c$  all interactions come from

collisions. When  $k = k_{\max}$   $f_U$  is equal to  $\frac{1}{2}$  regardless of the temperature, and this can be understood by the same type of reasoning as was used above to explain the results for  $L$  phonons with this wave vector. Note that although the fraction of  $U$  processes is constant, the actual rate becomes zero as  $T \rightarrow 0$  K. For smaller wave vectors the umklapp fraction is approximately constant at high temperatures, but then decreases rapidly below a characteristic temperature  $T_c(k)$ . This decrease occurs because of the decrease in the population of thermal phonons with high momentum (see comments in FT phonon section above). The temperature  $T_c(k)$  is smallest for large  $k$  because then a collision with a thermal phonon having even a small wave number is sufficient to produce an umklapp collision.

## V. SUMMARY

In this paper we have presented the first detailed calculations of the rates of phonon collision and decay as a function of wave vector and temperature. The calculations are performed for a simplified model of a monatomic fcc crystal, and include the effects of phonon dispersion and anisotropy. By appropriate scaling one can use these results to make rough estimates of the interaction rates for real crystals.

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