

Lifetime of high-frequency longitudinal-acoustic phonons in  $\text{CaF}_2$  at low crystal temperatures

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Using the rigid-ion model, we have calculated the lifetime of longitudinal-acoustic phonons in  $\text{CaF}_2$  with frequency  $\nu$  between 2.5 and 5.5 THz at low crystal temperatures. Neutron scattering data and temperature-dependence data of the second-order elastic constants have been used to determine the parameters of the theory. The results show a marked anisotropy and a small deviation from the long-wavelength approximation. The overall agreement with recent experiments is good but suggests a more careful analysis of the raw experimental data.

## I. INTRODUCTION

In the last few years, several experiments have been performed to study the lifetime of acoustic phonons with frequency  $\nu$  of 1 THz or greater at low crystal temperatures. Some new methods are described in great detail by Eisenmenger<sup>1</sup> and Bron.<sup>2</sup> Transverse-acoustic (TA) phonons have been probed in crystalline quartz,<sup>3</sup> in semiconductors,<sup>4</sup> and in thallium halide crystals,<sup>5</sup> and their lifetime turns out to be very long, of the order of microseconds or greater. This behavior is well understood on the basis of the theoretical works of Slonimskii,<sup>6</sup> Orbach and Vredevoe,<sup>7</sup> and Lax *et al.*<sup>8</sup> The anharmonic multiphonon decay is forbidden, because of energy-momentum conservation, even for TA phonons near the zone boundary so that their lifetime is essentially determined by impurities and boundary scattering.

The case of longitudinal-acoustic (LA) phonons is somewhat controversial. Very recently, Baumgartner *et al.*<sup>9</sup> probed LA phonons with frequencies between 1.5 and 3.0 THz in  $\text{CaF}_2$  and their results undoubtedly indicate that the lifetime is proportional to the inverse fifth power of the frequency. On the other hand, Bron<sup>10</sup> and Bron and Grill<sup>11</sup> studied LA phonons in the same frequency range in  $\text{SrF}_2$  and their results show a lifetime very slowly and quite linearly dependent on the frequency. Obviously, the discrepancy between the two experiments cannot be accounted for by the small differences between the two crystals.<sup>12</sup> In a theoretical approach, the theorem of Lax *et al.*<sup>8</sup> cannot be used to forbid the spontaneous decay of LA phonons and this process becomes the most important. Thus, the LA-phonon lifetime turns out to be an intrinsic property of the crystal, determined by the anharmonic nature of the forces

between ions. So far, any theoretical prediction has been confined only to the long-wavelength approximation for an isotropic model where the TA modes are obviously degenerate. Using the formalism of the elasticity theory for the three-phonon process, Orbach and Vredevoe<sup>7</sup> have obtained an analytical expression for the decay  $\text{LA} \rightarrow \text{LA} + \text{TA}$ , where the lifetime is inversely proportional to the inverse fifth power of the frequency. They also claimed that the process  $\text{LA} \rightarrow \text{TA} + \text{TA}$  is negligible. On the basis of a simplified model involving the Gruneisen parameter, Klemens<sup>13</sup> has obtained expressions for both decays  $\text{LA} \rightarrow \text{LA} + \text{TA}$  and  $\text{LA} \rightarrow \text{TA} + \text{TA}$  with the same frequency dependence of Orbach and Vredevoe's formula, claiming that both processes give comparable contribution. More recently, on the basis of a simple scaling law, one of the authors<sup>14</sup> has shown that the  $\nu^{-5}$  dependence is valid also for anisotropic solids. The results of Baumgartner *et al.*<sup>9</sup> confirm the predictions of Orbach and Vredevoe<sup>7</sup> and Klemens,<sup>13</sup> but there are still some problems to be answered: (a) the formulas of Orbach and Vredevoe<sup>7</sup> and Klemens<sup>13</sup> cannot be used for an accurate and quantitative comparison with experimental data; (b) the relative contribution of the two processes  $\text{LA} \rightarrow \text{LA} + \text{TA}$  and  $\text{LA} \rightarrow \text{TA} + \text{TA}$  to the total lifetime is still unknown; (c) in the frequency region between 1.5 and 3.0 THz the dispersion curves of  $\text{CaF}_2$  show a slight deviation (10–20 %) from the long-wavelength region,<sup>12</sup> and it is not *a priori* clear why this deviation does not affect the total lifetime in the experiment of Baumgartner *et al.*<sup>9</sup>

We have performed a careful numerical calculation of the lifetime of LA phonons in  $\text{CaF}_2$  with frequencies between 2.5 and 5.5 THz *without* using the long-wavelength approximation. The details of

our approach are described in Secs. II and III. The results, which are reported in Sec. IV, show an anisotropy much larger than one would expect from the dispersion curves. Contrary to the assumption of Orbach and Vredevoe,<sup>7</sup> the process LA → TA + TA is dominant. Besides, each decay mode shows small deviations from the  $\nu^{-5}$  law which, curiously, cancels out in the final result. Our results are in agreement with the data of Baumgartner *et al.*,<sup>9</sup> but indicate that a better theory is needed in extrapolating the real lifetime averaged over all directions from the effective one which is observed experimentally. Besides, in Sec. IV we suggest a new interpretation of the raw data of Bron and Grill's experiment<sup>11</sup> which is not in contradiction with our results or the data of Baumgartner *et al.*<sup>9</sup>

## II. GENERAL FORMALISM

The lifetime of LA phonons with energy  $h\nu \gg k_B T$  is essentially determined by the three-phonon interaction, i.e.,

$$M(\vec{k}, \lambda; \vec{k}', \lambda'; \vec{k}'', \lambda'') = v \left[ \frac{\hbar}{4\pi} \right]^{3/2} (\nu_{\vec{k}, \lambda} \nu_{\vec{k}', \lambda'} \nu_{\vec{k}'', \lambda''})^{-1/2} \sum_{\mu, \mu'} m_{\mu}^{-1} m_{\mu'}^{-1/2} e^{i(\vec{k} + \vec{k}' + \vec{k}'') \cdot \vec{S}(\mu)}$$

$$\times \sum_{\alpha\beta\gamma} [\delta_{\mu, \mu'} \xi(\vec{k}, \lambda; \mu)_{\alpha} \xi(\vec{k}', \lambda'; \mu)_{\beta} \xi(\vec{k}'', \lambda''; \mu)_{\gamma} x(\mu)_{\alpha\beta\gamma}$$

$$+ \xi(\vec{k}, \lambda; \mu')_{\alpha} \xi(\vec{k}', \lambda'; \mu)_{\beta} \xi(\vec{k}'', \lambda''; \mu)_{\gamma} x(\vec{k}; \mu', \mu)_{\alpha\beta\gamma}$$

$$+ \xi(\vec{k}, \lambda; \mu)_{\alpha} \xi(\vec{k}', \lambda'; \mu')_{\beta} \xi(\vec{k}'', \lambda''; \mu)_{\gamma} x(\vec{k}'; \mu', \mu)_{\alpha\beta\gamma}$$

$$+ \xi(\vec{k}, \lambda; \mu)_{\alpha} \xi(\vec{k}', \lambda'; \mu)_{\beta} \xi(\vec{k}'', \lambda''; \mu')_{\gamma} x(\vec{k}''; \mu', \mu)_{\alpha\beta\gamma}], \quad (2)$$

and

$$x(\vec{k}; \mu', \mu)_{\alpha\beta\gamma} = \frac{1}{v} \sum_j' \frac{\partial^3 \phi}{\partial R_{(\mu')_{\alpha}}^{(j)} \partial R_{(\mu)_{\beta}}^{(j)} \partial R_{(\mu)_{\gamma}}^{(j)}} \exp\{-i\vec{k} \cdot [\vec{R}^{(j)} - \vec{R}_{(\mu')}^{(0)}]\}, \quad (3a)$$

$$x(\mu)_{\alpha\beta\gamma} = \lim_{\vec{k} \rightarrow 0} \sum_{\mu'} x(\vec{k}; \mu, \mu')_{\alpha\beta\gamma}, \quad (3b)$$

where we have used the following notation:  $v$  is the volume of the unit cell,  $\vec{R}^{(j)}$  is a lattice vector [ $\vec{R}^{(0)} = 0$ ],  $\vec{S}(\mu)$  is a basis vector [ $\vec{S}^{(0)} = 0$ ], and  $\vec{R}_{(\mu)}^{(j)} = \vec{R}^{(j)} + \vec{S}(\mu)$ ,  $m_{\mu}$  is the mass of the ion of type  $\mu$ ,  $\nu_{\vec{k}, \lambda}$  is the eigenfrequency in the quasiharmonic approximation and  $\xi(\vec{k}, \lambda; \mu)$  is the polarization vector relative to phonons  $(\vec{k}, \lambda)$  and ions of type  $\mu$ , and  $\phi$  is the crystal potential. Furthermore, we assume that the short-range part of the potential between two ions of type  $\mu$  and  $\mu'$  at a

$$H' = \frac{1}{3! \sqrt{N}} \sum_{\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{G}} \sum_{\lambda_1, \lambda_2, \lambda_3} M(\vec{k}_1, \lambda_1; \vec{k}_2, \lambda_2; \vec{k}_3, \lambda_3)$$

$$\times A_{\vec{k}_1, \lambda_1} A_{\vec{k}_2, \lambda_2} A_{\vec{k}_3, \lambda_3}$$

$$\times \delta_{\vec{k}_1 + \vec{k}_2 + \vec{k}_3 + \vec{G} = 0}, \quad (1)$$

where  $A_{\vec{k}, \lambda} = a_{\vec{k}, \lambda} + a_{-\vec{k}, \lambda}^{\dagger}$ ,  $a_{\vec{k}, \lambda}$  ( $a_{\vec{k}, \lambda}^{\dagger}$ ) is the destruction (creation) operator of phonons with wave vector  $\vec{k}$  and mode  $\lambda$ ,  $N$  is the number of unit cells, and the coefficient  $M$  is obtained from the anharmonic lattice dynamics. Since we are interested in acoustic modes of ionic crystals, it is sufficient to consider the first-order effect of the screening due to point dipoles which consists in a renormalization of the actual ionic charges.<sup>15,16</sup> Consequently, we can use the rigid-ion model where the "effective" charges take into account precisely this effect of the atomic polarizability. Within this approximation, it is straightforward to show that

distance  $d$  has the Born-Mayer form<sup>17</sup>

$$U^s(\mu, \mu'; d) = \alpha_{\mu, \mu'} \exp\left[-\frac{d}{\beta_{\mu, \mu'}}\right]$$

$$+ \frac{\delta_{\mu, \mu'}}{d^6} + \frac{\epsilon_{\mu, \mu'}}{d^8}. \quad (4)$$

The first term represents the repulsive interaction between electronic shells, and the second and third terms take into account the dipole-dipole and the

dipole-quadrupole interactions, respectively. In Sec. III we will determine the values of the effective charges and of the parameters of Eq. (4) using the relevant available experimental data. This, in turn, will allow us to obtain an explicit expression for the matrix elements in Eq. (2).

### III. LATTICE DYNAMICS OF CaF<sub>2</sub>

The numerical evaluation of the lattice dynamics of CaF<sub>2</sub> can be divided into two distinct steps. At first, we consider the crystal in the quasiharmonic approximation at room temperature and fit the rigid-ion model to the dispersion curves obtained by neutron scattering experiments<sup>18</sup> and to the second-order elastic constants (SOEC's) measured in the first-sound region.<sup>19</sup> The discrepancy between the two sets of data is smaller than the experimental error, typically 2–3 %, indicating that the energy shift between zero and first sound is negligible for the present purpose. In constructing the short-range part of the crystal potential we have selected only the bonds shown in Table I, because the introduction of additional bonds, and hence of additional parameters, does not improve the fitting to the dispersion curves. In this first step we evaluate the “effective” ionic charges and the first and second derivatives of the short-range potentials at room temperature. The value of the “effective” charge of Ca<sup>2+</sup> ions turns out to be 1.51e while the potential derivatives are shown in Table II. We notice that our results are somewhat different from those obtained by Elcombe and Pryor, even if the overall agreement with the dispersion curves is the same (see Fig. 1). In particular, the nearest-neighbor Ca<sup>2+</sup>-F<sup>-</sup> short-range interaction turns out to be repulsive in our calculations, while Elcombe and Pryor obtain the opposite result which is, obviously, unphysical.

For the bonds Ca<sup>2+</sup>-Ca<sup>2+</sup> and F<sub>1</sub><sup>-</sup>-F<sub>1</sub><sup>-</sup> (or F<sub>2</sub><sup>-</sup>-F<sub>2</sub><sup>-</sup>) with distance  $a/\sqrt{2}$  ( $a$  is the lattice constant) it is plausible to neglect the shell-shell repulsive term in the short-range potentials of Eq.

(4) because the sum of the Ca<sup>2+</sup> and F<sup>-</sup> ionic radii estimated by Pauling<sup>20</sup> and Goldschmidt<sup>21</sup> is smaller than the interionic distance. Thus we obtain

$$\frac{a_0}{e^2} U^s(\text{Ca}^{2+} - \text{Ca}^{2+}; d) \simeq 0.117 \left[ \frac{a_0}{d} \right]^6 - 0.0313 \left[ \frac{a_0}{d} \right]^8$$

$$\frac{a_0}{e^2} U^s(\text{F}_1^- - \text{F}_1^-; d) \simeq -0.0138 \left[ \frac{a_0}{d} \right]^6 + 0.00365 \left[ \frac{a_0}{d} \right]^8$$

for  $d \simeq a_0/\sqrt{2}$ , where  $a_0$  ( $=5.4635$  Å) is the lattice constant at room temperature.<sup>22</sup> We notice that the Ca<sup>2+</sup>-Ca<sup>2+</sup> dipole-dipole interaction turns out to be repulsive, and we may conclude that the presence of the neighbor F<sup>-</sup> ions modifies the interaction between two isolated Ca<sup>2+</sup> ions remarkably. The discrepancy should be investigated further, but it would go far beyond the purpose of the present work. For the other two bonds, i.e., Ca<sup>2+</sup>-F<sub>1</sub><sup>-</sup> (or Ca<sup>2+</sup>-F<sub>2</sub><sup>-</sup>) with distance  $(\sqrt{3}/4)a$  and F<sub>1</sub><sup>-</sup>-F<sub>2</sub><sup>-</sup> with distance  $(1/2)a$ , it is plausible to neglect the dipole-quadrupole interaction in Eq. (4), so that for each bond there are three parameters left. They will be evaluated in the second part of our calculations because additional experimental data are needed.

In the second part of our calculations, we consider the experimental temperature dependence of the lattice constant<sup>22</sup> and of the SOEC's  $c_{11}$  and  $c_{44}$ .<sup>23</sup> The expressions for the SOEC's, as obtained from the long-wavelength approximation of lattice dynamics,<sup>18</sup> refer to the unrenormalized phonon propagator. The SOEC's depend upon the temperature only through the lattice constant, as long as the crystal is free of stress. Therefore, we can fit our model to the above mentioned data with the only assumption being that the “effective” ionic charges are temperature independent. The fit of  $c_{11}$  and  $c_{44}$  is shown in Fig. 2 and the remaining short-range potentials turn out to be

TABLE I. Bonds and relative distances ( $a$  is the lattice constant) used in constructing the short-range part of the crystal potential of CaF<sub>2</sub>.

Bonds	Ca <sup>2+</sup> -F <sub>(i)</sub> <sup>-</sup>	F <sub>1</sub> <sup>-</sup> -F <sub>2</sub> <sup>-</sup>	Ca <sup>2+</sup> -Ca <sup>2+</sup>	F <sub>(i)</sub> <sup>-</sup> -F <sub>(i)</sub> <sup>-</sup>
Distance	$\frac{\sqrt{3}}{4}a$	$\frac{1}{2}a$	$\frac{1}{\sqrt{2}}a$	$\frac{1}{\sqrt{2}}a$

TABLE II. Calculated first and second derivatives of the short-range potentials of  $\text{CaF}_2$  free of stress at room temperature [ $a_0 = 5.4635 \text{ \AA}$  is the lattice constant at room temperature (Ref. 21)].

Bond and distance $d$	$\frac{1}{d} U^s(\mu, \mu'; d)' \left[ \frac{a_0^3}{4e^2} \right]$	$U^s(\mu, \mu'; d)'' \left[ \frac{a_0^3}{4e^2} \right]$
$\text{Ca}^{2+} - \text{F}^-$ , $d = \frac{\sqrt{3}}{4} a_0$	-0.1040	8.746
$\text{F}^- - \text{F}^-$ , $d = \frac{a_0}{2}$	-0.0312	2.759
$\text{Ca}^{2+} - \text{Ca}^{2+}$ , $d = \frac{a_0}{\sqrt{2}}$	0.8063	1.643
$\text{F}^- - \text{F}^-$ , $d = \frac{a_0}{\sqrt{2}}$	0.0985	-0.223

$$\frac{a_0}{e^2} U^s(\text{Ca}^{2+} - \text{F}^-; d) \simeq 9.46 \times 10^4 \exp \left[ -33.06 \frac{d}{a_0} \right] - 8.18 \times 10^{-4} \left[ \frac{a_0}{d} \right]^6 \left[ d \simeq \frac{\sqrt{3}}{4} a \right]$$

and

$$\frac{a_0}{e^2} U^s(\text{F}_1^- - \text{F}_2^-; d) \simeq 1.14 \times 10^8 \exp \left[ -47.2 \frac{d}{a_0} \right] + 3.18 \times 10^{-4} \left[ \frac{a_0}{d} \right]^6 \left[ d \simeq \frac{a}{2} \right].$$

In conclusion, we are now able to numerically evaluate Eqs. (3), and hence the transition matrix elements in Eq. (2). Besides, knowing the temperature dependence of the lattice constant, our method can predict the temperature dependence of the dynamical and transition matrices. The difference between zero and room temperature is of the order of 3–5 %.

#### IV. LIFETIME OF LA PHONONS IN $\text{CaF}_2$

Following Landau and Rumer,<sup>24</sup> the anharmonic low-temperature relaxation of LA phonons in  $\text{CaF}_2$  with frequency  $\nu$  greater than 1 THz can be calculated by the golden rule and the three-phonon Hamiltonian of Eq. (1). This method is justified because the energy shift between the first and zero sound is negligible, as we have mentioned in Sec. III. On the other hand, Niklasson<sup>25</sup> has shown that the ladder diagram approximation contributes only in the first-sound region that we do not consider.

We select LA phonons only in the three principal directions and with frequency  $\nu$  between 2.5 and 5.5 THz. This frequency region is roughly halfway between the center and the boundary of the Brillouin zone and hence the umklapp processes can be safely neglected. For  $T = 5 \text{ K}$ , the relation  $h\nu \gg K_B T$  is satisfied and the boson occupation factors can be neglected. Thus, the LA-phonon lifetime is temperature independent and is given by the relation

$$\frac{1}{\tau_{\vec{k}}} = \frac{1}{2} \sum_{\lambda', \lambda''} W_{\vec{k}}(\lambda', \lambda''),$$

$$W_{\vec{k}}(\lambda', \lambda'') = \frac{\nu}{(2\pi)^3} \int_0^\pi d\theta \sin\theta \int_0^{2\pi} d\varphi \left[ \left| \frac{1}{\hbar} M(\vec{k}, \text{LA}; -\vec{k}', \lambda'; -\vec{k}'', \lambda'') \right|^2 \right. \\ \left. \times \frac{|\vec{k}'|^2}{\frac{\partial}{\partial |\vec{k}'|} (\nu_{\vec{k}', \lambda'} + \nu_{\vec{k}'', \lambda''})} \right]_{\vec{k}' = \vec{k}'_0(\theta, \varphi)},$$

where the summation is over the acoustic branches,  $\vec{k}'' = \vec{k} - \vec{k}'$ ,  $\vec{k}'_0(\theta, \varphi)$  has spherical components  $\theta$  and  $\varphi$  and its magnitude is determined by the equation

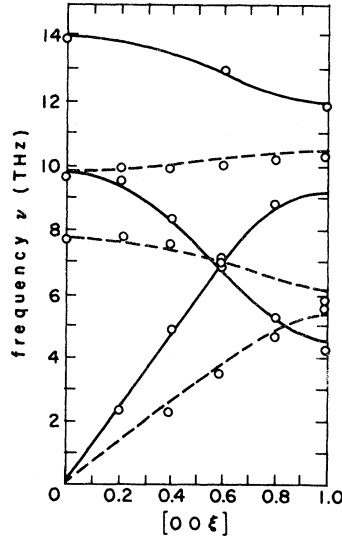


FIG. 1. Phonon dispersion relations of  $\text{CaF}_2$  at room temperature in the [001] direction [white circles: data from Elcombe and Pryor (Ref. 17); continuous and dashed lines: present theoretical prediction for longitudinal and transverse polarization, respectively].

$$v_{\vec{k}, \text{LA}} - v_{\vec{k}_0(\theta, \varphi), \lambda'} - v_{\vec{k}'', \lambda''} = 0.$$

The full dynamical and interaction matrices have been used without the long-wavelength approximation. The integration over the solid angle has been performed with a mesh of 2200 directions. The partial lifetimes are shown in Figs. 3 and 4, where the TA mode of lower energy is labeled with TA1 and the other TA mode with TA2. We notice that the  $\nu^{-5}$  dependence is very well verified, except for the process  $\text{LA} \rightarrow \text{LA} + \text{TA1}$  in the [001] direction. For each decay mode there are small deviations which increase with energy, but they cancel out in the total lifetime, so that the experimental results of Baumgartner *et al.*<sup>9</sup> are justified from a theoretical point of view. We can extrapolate the coefficient of  $\nu^{-5}$  in the total lifetime in the limit  $\nu \rightarrow 0$ , obtaining

$$\tau = \alpha \nu^{-5} \quad (5)$$

with

$$\alpha = \begin{cases} 3.30 \times 10^{-7}, & \text{[001] direction} \\ 1.21 \times 10^{-7}, & \text{[011] direction} \\ 6.22 \times 10^{-8}, & \text{[111] direction} \end{cases}$$

in units of  $\text{sec THz}^5$ . The partial lifetime for the decay  $\text{LA} \rightarrow \text{LA} + \text{TA}$  turns out to be larger than that for the decay  $\text{LA} \rightarrow \text{TA} + \text{TA}$  and hence the

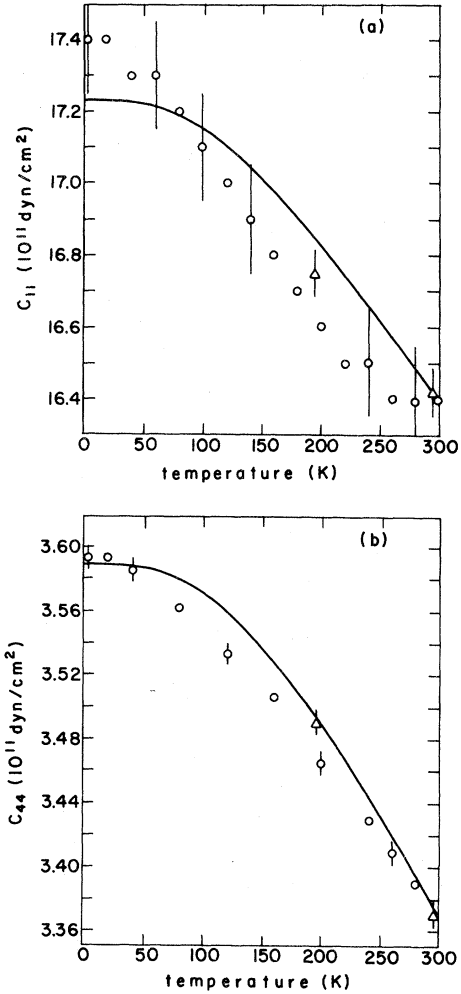


FIG. 2. Temperature dependence of the second-order elastic constants (a)  $c_{11}$  and (b)  $c_{44}$  of  $\text{CaF}_2$  [white circles: data from Huffman and Norwood (Ref. 22); triangles: data from Wong and Schuele (Ref. 18); continuous line: present prediction].

latter process is dominant to the contrary of the assumption of Orbach and Vredevoe.<sup>7</sup> The large deviation for the decay mode  $\text{LA} \rightarrow \text{TA1} + \text{LA}$  in the [001] direction cannot be explained by the dispersion curves and is due only to the fact that the long-wavelength approximation of the interaction matrix elements fails for that decay mode. Equation (5) shows that the lifetime of LA phonons depends upon the direction within a factor of 5, even if the angular dependence of the phonon velocity in the nondispersive region is small ( $\sim 10\%$ ). This feature makes it difficult to compare our prediction with experimental data where the angular distribution of the nonequilibrium pho-

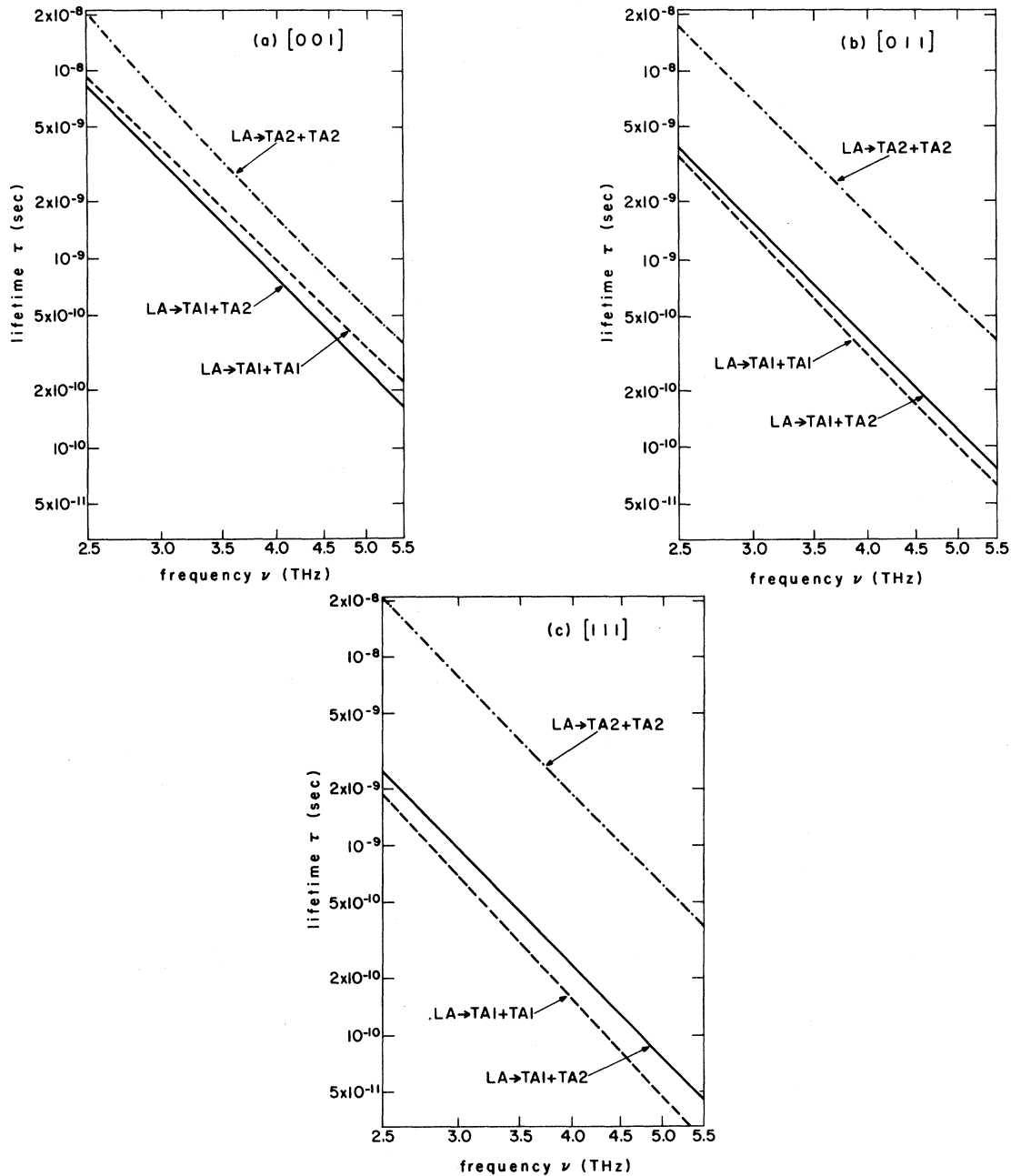


FIG. 3. Partial lifetimes for the process  $LA \rightarrow TA + TA$  of high-frequency longitudinal-acoustic phonons in the three principal directions in  $CaF_2$  at low crystal temperatures.

non population is unknown. A comparison with the experimental results is shown in Table III. We have chosen the phonon frequency of 2 THz because this is the only frequency probed by Akimov *et al.*<sup>26</sup> The agreement is fairly good but indicates that a more rigorous interpretation of the raw data is needed. The mode conversion due to the impurities leads to a quasiequilibrium of the populations of phonons with the same frequency, but with dif-

ferent polarizations. Experimentally, the decay rate of the combined population of LA and TA phonons is observed and the effective lifetime is related to the LA-phonon lifetime by the expression

$$\frac{1}{\tau_{\text{eff}}} = \frac{1}{1 + N_{TA}/N_{LA}} \frac{1}{\tau_{LA}}, \quad (6)$$

where  $N_{TA}$  and  $N_{LA}$  are the quasiequilibrium TA- and LA-phonon populations, respectively. Since

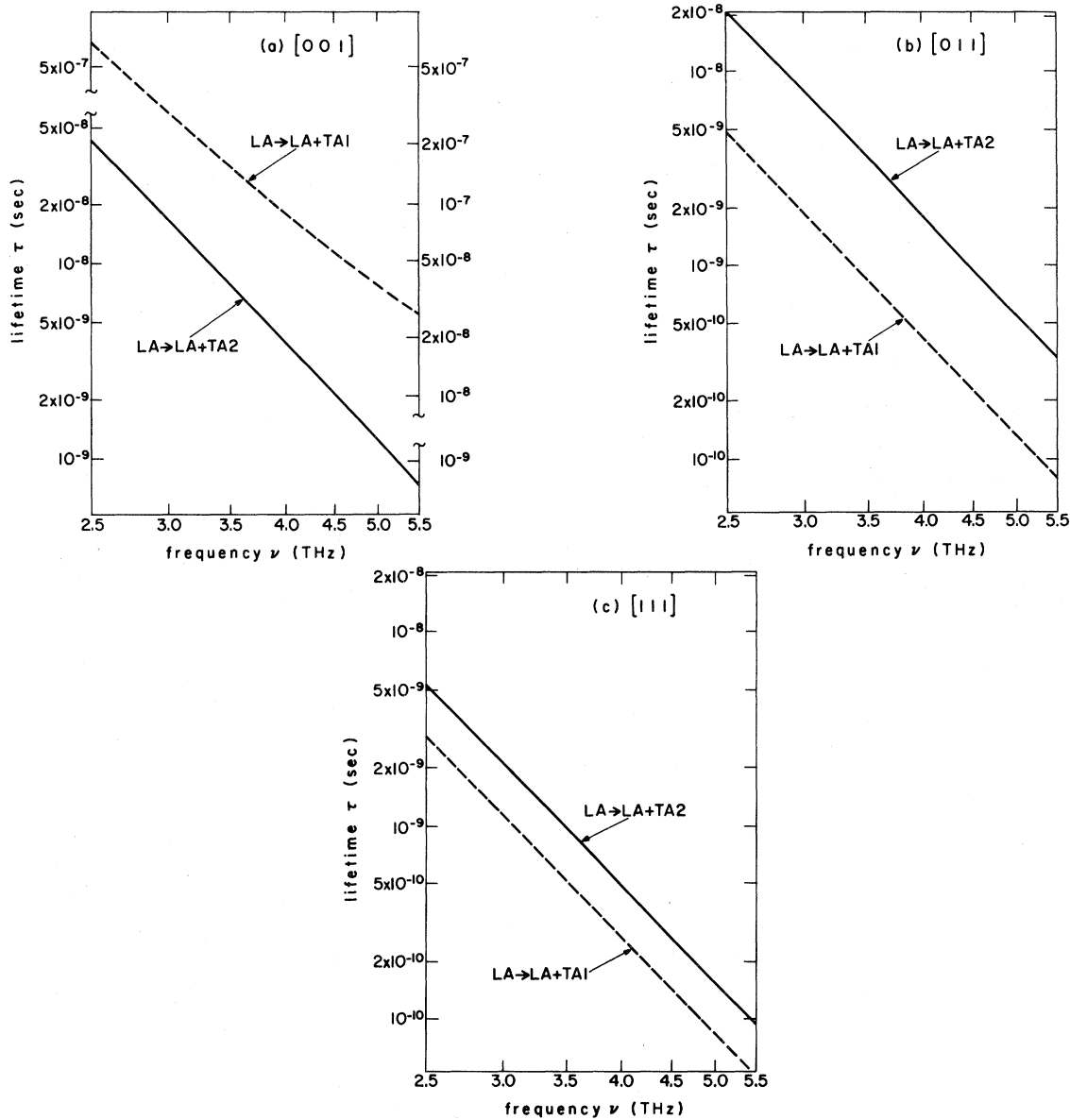


FIG. 4. Partial lifetimes for the process  $LA \rightarrow LA + TA$  of high-frequency longitudinal-acoustic phonons in the three principal directions in  $CaF_2$  at low crystal temperatures.

the process of mode conversion is essentially unknown, it is assumed that the ratio  $N_{TA}/N_{LA}$  is equal to the ratio between the density of states. This is a very sensible assumption but any small deviation from it would imply a variation of  $\tau_{LA}$  of the order of the discrepancy between our predictions and the experimental data. Our calculation indicates that  $N_{TA}/N_{LA}$  is relatively larger than the ratio between the corresponding density of states.

Since the crystals  $CaF_2$  and  $SF_2$  are very similar,<sup>12</sup> the LA-phonon lifetime should have the

same frequency dependence and roughly the same order of magnitude in both crystals. The experiment of Bron<sup>10</sup> shows a LA-phonon lifetime much

TABLE III. Lifetime (nsec) of 2-THz longitudinal-acoustic phonons in  $CaF_2$  at low crystal temperatures.

Experimental		Present prediction		
a	b	[001]	[011]	[111]
38.0	13.0	10.3	3.78	1.94

<sup>a</sup>Baumgartner *et al.* (Ref. 9).

<sup>b</sup>Akimov *et al.* (Ref. 26).

higher and linearly dependent upon the frequency. The discrepancy is striking but may be explained by a better analysis of the heat pulse technique<sup>11</sup> used by Bron. In his experiment, during the first microsecond after the heat pulse the distribution of phonons with  $\nu > 1$  THz is similar to that given by the Bose factor with  $T = 13$  K, while the distribution of phonons with  $\nu < 1$  THz indicates a much higher temperature. Therefore, the repopulation process from below cannot be neglected and the ef-

fective lifetime of the LA phonons with  $\nu \geq 1$  THz becomes a functional of the population density of phonons with lower energy so that Eq. (6) cannot be used.

#### ACKNOWLEDGMENTS

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