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## Spontaneous decay of TA phonons

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Anharmonic decay of high-energy transverse acoustic (TA) phonons satisfying  $\hbar\omega \gg k_B T$  is studied taking account of the elastic anisotropy of real crystalline solids. Dispersive effects are assumed to be small. We find that the decay rate is very anisotropic. The possibility of observing this anisotropy in a phonon-focusing experiment is discussed.

Anharmonic phonon decay together with mass-defect scattering (impurity or isotope scattering) and phonon-focusing effects are important ingredients governing the dynamics of high-energy phonon transport in nonmetallic solids. Recently, extensive studies have been made both theoretically and experimentally on the isotope scattering<sup>1,2</sup> and the focusing effects<sup>3</sup> of phonons. On the other hand, there are only a few theoretical investigations on the anharmonic interaction of high-frequency phonons in real crystalline solids in which anisotropy is important.<sup>4-6</sup> The isotropic approximation combined with continuum elasticity theory does indeed lead to the correct result, for instance, on the frequency dependence of the spontaneous decay of a LA phonon into two other phonons, i.e., the  $\omega^{-5}$  dependence of the lifetime ( $\omega = 2\pi\nu$  being the angular frequency of a phonon)<sup>7-9</sup> being valid at nondispersive frequencies. This has recently been verified experimentally by Baumgartner, Engelhardt, and Renk.<sup>10</sup>

However, it is also concluded under this approximation that degenerate TA phonons cannot split into two phonons for kinematical reasons.<sup>11</sup> This leads to anomalously long anharmonic lifetimes at superthermal frequencies satisfying  $\omega \gg k_B T/\hbar$  ( $T$  being the ambient temperature), though LA phonons of the same frequency can decay very rapidly into two phonons. Accordingly the high-energy TA phonons are considered to relax into low-energy phonons via

anharmonic processes only after executing mode conversion into LA phonons through mass-defect scattering. There have been several recent works based on this assumption.<sup>12,13</sup>

In this Rapid Communication we consider the effects of the anisotropy of real crystalline solids. By virtue of the presence of elastic anisotropy a TA phonon, even that of the lowest branch, in a crystal can often find a set of phonons with slower phase velocities to decay into. Moreover, we find that for propagation along some directions, the spontaneous decay rates of TA phonons become comparable to those of the LA phonons. In addition, the decay rate of the TA phonons is found to change a large amount as the direction of the wave vector is varied. Owing to this anisotropy it is in principle possible to verify experimentally the predicted decay of the TA phonons by a phonon-focusing experiment.<sup>14-16</sup> We show by numerical calculations for sodium fluoride that the intensity of the phonon flux exhibits indeed some characteristic changes due to anharmonic effects which are observable when the propagation distance or the phonon frequency is varied.

In the continuum approximation the anharmonic decay rate (i.e., the reciprocal of the lifetime  $\tau$ ) of a phonon of mode  $j$  with the wave vector  $\mathbf{q}$  can be written in the frequency regime  $\omega \gg k_B T/\hbar$  as<sup>4</sup>

$$\tau^{-1}(\mathbf{q}, j) = \frac{\pi\hbar}{8\rho^3\omega(\mathbf{q}, j)} \sum_{\mathbf{q}', \mathbf{q}''} \sum_{j', j''} \frac{|M(\mathbf{q}, \mathbf{q}', \mathbf{q}''; j, j', j'')|^2}{\omega(\mathbf{q}', j')\omega(\mathbf{q}'', j'')} \delta_{\mathbf{q}, \mathbf{q}'+\mathbf{q}''} \delta(\omega(\mathbf{q}, j) - \omega(\mathbf{q}', j') - \omega(\mathbf{q}'', j'')) \equiv \alpha(\hat{\mathbf{q}}, j)\omega^5(\mathbf{q}, j), \quad (1)$$

where  $\rho$  is the mass density of the medium,  $\alpha$  a constant depending on the direction  $\hat{\mathbf{q}}$  of  $\mathbf{q}$ , and on the mode polarization  $j$ .  $M$  is the matrix element for the decay  $(\mathbf{q}, j) \rightarrow (\mathbf{q}', j') + (\mathbf{q}'', j'')$  derived for three-phonon processes from the interaction Hamiltonian

$$H' = \int dV \left[ \frac{1}{2!} C_{ijkl} \eta_{ij} \eta_{kl} + \frac{1}{3!} C_{ijklmn} \eta_{ij} \eta_{kl} \eta_{mn} \right]. \quad (2)$$

In Eq. (2)  $\eta$  is the deformation tensor defined in terms of the displacement vector  $\mathbf{u}$  by

$$\eta_{ij} = \frac{1}{2} \left[ \partial_j u_i + \partial_i u_j + \sum_{k=1}^3 (\partial_i u_k)(\partial_j u_k) \right], \quad (3)$$

and  $C_{ijkl}$  and  $C_{ijklmn}$  are elastic stiffness constants of second

and third orders, respectively. The explicit expression for  $M$  is too lengthy to be given here but will appear elsewhere.<sup>17</sup>

To give a qualitative discussion of the dependence of  $\tau^{-1}$  on  $\hat{\mathbf{q}}$ , we consider first what happens when the factor  $|M|^2/\omega'\omega''$  is treated as a constant which can be removed from the sum in Eq. (1). Then  $\tau^{-1}$  is proportional to the two-phonon density of states  $D_2$  defined by

$$D_2(\mathbf{q}, j) \equiv \sum_{\mathbf{q}', \mathbf{q}''} \sum_{j', j''} \delta(\omega(\mathbf{q}, j) - \Omega(\mathbf{q}, \mathbf{q}'; j, j', j'')) \\ = \frac{V}{(2\pi)^3} \sum_{j', j''} \int \frac{dS_{\Omega}(\mathbf{q}', j')}{|\nabla_{\mathbf{q}'} \Omega(\mathbf{q}, \mathbf{q}'; j, j', j'')|}, \quad (4)$$

where  $V$  is the volume of the medium;  $dS_{\Omega}$  denotes a surface element on the constant-frequency surface determined

by  $\Omega = \omega(\mathbf{q}, j)$  in  $\mathbf{q}'$  space and

$$\Omega(\mathbf{q}, \mathbf{q}'; j, j', j'') \equiv \omega(\mathbf{q}', j') + \omega(\mathbf{q} - \mathbf{q}', j'') \quad (5)$$

Note that

$$\nabla_{\mathbf{q}'} \Omega(\mathbf{q}, \mathbf{q}'; j, j', j'') = \mathbf{v}(\mathbf{q}', j') - \mathbf{v}(\mathbf{q} - \mathbf{q}', j'') \quad (6)$$

where  $\mathbf{v}$  is the group velocity of a phonon. Therefore, provided that two phonons produced as the decay products of an initial phonon ( $\mathbf{q}, j$ ) happen to have identical group-velocity vectors for some  $\mathbf{q}'$ ,  $D_2$  should exhibit some singularity as in the case of the one-phonon density of states.<sup>18</sup> The equation  $\nabla_{\mathbf{q}'} \Omega = 0$  is trivially satisfied by the collinear process, but is not necessarily exactly satisfied by other decay processes. However, we can readily confirm that it is approximately satisfied for some directions of the wave vector  $\mathbf{q}$  of the initial phonons. This may be qualitatively understood by referring to the constant-frequency surfaces ( $\omega$  surfaces) and the corresponding group-velocity surfaces ( $\nu$  surfaces) of phonons.

In Fig. 1 we have plotted the sections in the  $(1\bar{1}0)$  plane of these surfaces for the TA phonons in NaF.<sup>19</sup> ( $T1$  and  $T2$  stand for the lower and upper TA branches, respectively.) NaF is chosen because it consists of isotopically pure elements. It should be noted that in most crystals consisting of isotopically impure elements the scattering of phonons by isotopes is much stronger than the scattering by anharmonicity in the frequency range  $\nu \leq 1$  THz and  $\hbar\omega \gg k_B T$ ,<sup>9</sup> making the observability of anharmonic decay somewhat difficult.

Now, the normal vectors of the  $\omega$  surfaces at points located between  $A$  and  $E_1$  are mapped in the narrow region  $A'$  to  $E'_1$  on the  $\nu$  surfaces which are folded at  $C'$  and  $D'_+$ . ( $C$  and  $D$  are parabolic points of the  $\omega$  surfaces with vanishing curvature.) Thus the  $\nu$  surfaces of the TA phonons form multivalued surfaces, and for a direction between  $A'$  and  $E'_1$  in real space there exists more than one group-velocity vector being very close in magnitude. Accordingly, if the wave vectors  $\mathbf{q}'$ , and  $\mathbf{q} - \mathbf{q}'$  of final TA phonons are oriented between  $\theta_A$  and  $\theta_E$  in the  $(1\bar{1}0)$  section of the wave vector space,  $\nabla_{\mathbf{q}'} \Omega$  becomes very small.<sup>20</sup> This occurs for  $\mathbf{q}$  pointing also in directions between  $\theta_A$  and  $\theta_E$  in the same  $(1\bar{1}0)$  plane. Therefore, the decay rates of the phonons with  $\mathbf{q}$  pointing in such directions may exhibit some characteristic differences from those of phonons with  $\mathbf{q}$  being out of this range. Of course, the equation  $\omega = \omega' + \omega''$  must be satisfied. Hence, this cannot occur for the decay  $LA \rightarrow TA + TA$  because the  $\omega$  surface of the LA mode is definitely separated from those of the TA modes, but may occur for the decays of the TA phonons. As a result we can expect that  $D_2$  and then  $\tau^{-1}$  for TA phonons do not vary smoothly with the direction of  $\mathbf{q}$ . Thus, the mean free path of the TA phonons changes rather rapidly in this range of directions.

Figure 2 shows the calculated spontaneous decay rates in the  $(1\bar{1}0)$  plane (in the wave-vector space) of NaF. The frequency of the phonons is taken to be 1 THz. We have used the third-order elastic constants of Ref. 19 to calculate the matrix element  $M$ . It is assumed in the calculation that the effect of phonon dispersion on the phonon frequencies and on the matrix element  $M$  is very small.

We see that the decay rates of TA modes depend quite strongly on the propagation direction, and show certain structures as we have expected, while the decay rate of the

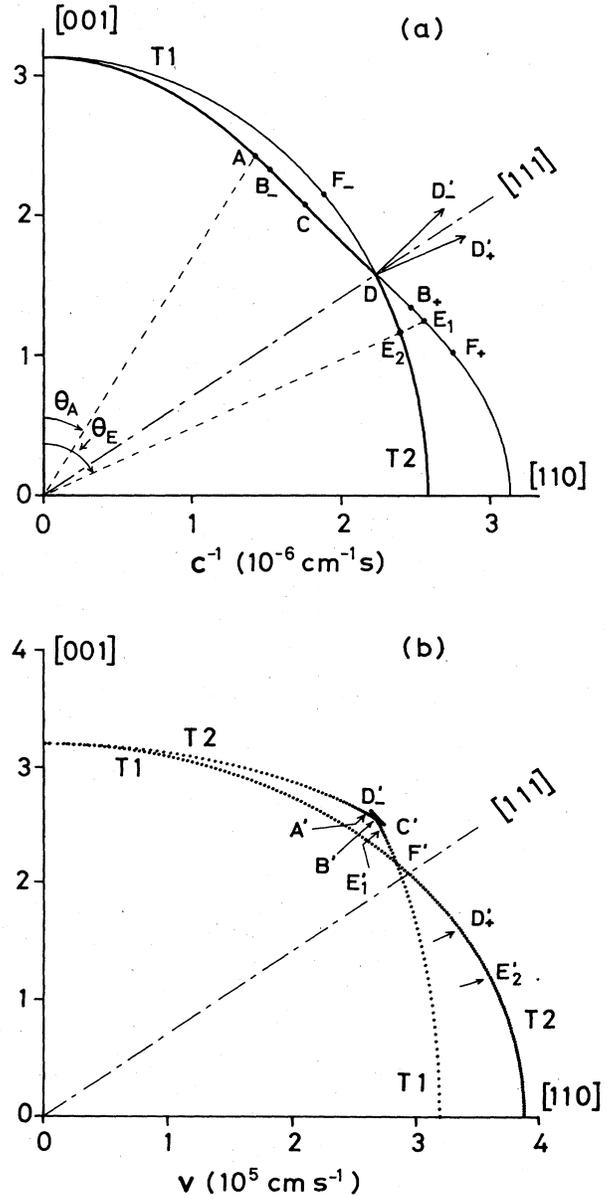


FIG. 1. Sections in  $(1\bar{1}0)$  plane of the first quadrants of  $\omega$  surfaces (a), and  $\nu$  surfaces (b), for TA phonons in NaF. Points  $A$  to  $F_{\pm}$  on the  $\omega$  surfaces are mapped onto  $A'$  to  $F'$  on the  $\nu$  surfaces, respectively. The  $\nu$  surfaces are generated by plotting the group-velocity vectors corresponding to the wave vectors distributed uniformly on the  $(1\bar{1}0)$  plane of the  $\omega$  surfaces. Transitions from  $T1$  to  $T2$  and vice versa happen at  $D'_{\pm}$  on the  $\nu$  surfaces.

LA mode varies rather smoothly with the direction of wave vector. Another important characteristic of the TA phonon decay observed in Fig. 2 is that along some directions around the  $[111]$  axis the decay rates become the same order of magnitude as those of the LA mode. All these results, i.e., the magnitude, large anisotropy, and the presence of cusp structures of the decay rates, are really substantial and play important roles for the observability of the anharmonic effects of TA phonons.

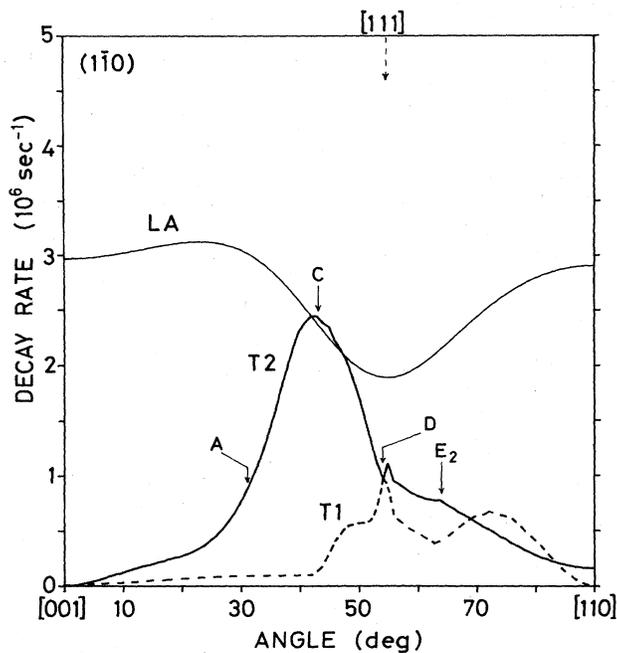


FIG. 2. Spontaneous decay rates of phonons in the  $(1\bar{1}0)$  plane (wave-vector space) of NaF plotted for LA mode (thin-solid line), T2 mode (thick-solid line), and T1 mode (dashed line). Note that polarization vectors of TA phonons interchange in the  $[111]$  direction.

These predictions on the spontaneous decay of TA phonons would best be verified by phonon-imaging experiments<sup>15,16</sup> with which the highly anisotropic spatial dependence of the phonon flux can be observed vividly. It is well established that by virtue of elastic anisotropy of crystalline solids the TA phonon flux emanated from a point source is focused sharply in some directions if their mean free paths are much longer than the propagation distances.<sup>3</sup> The intensities of these focused phonon flux should be reduced when they can undergo anharmonic decay. Therefore, by varying the propagation distances or the phonon frequencies the anharmonic effects should be detected as an anisotropic reduction of phonon intensity.

For comparison's sake we plot, first of all, the ballistic-phonon intensity in the absence of the anharmonic decay. Figure 3 displays the phonon intensity versus propagation direction in the  $(1\bar{1}0)$  plane (in the real space) of NaF. Sharp peaks of phonon flux located at about  $46^\circ$ ,  $61^\circ$ , and  $70^\circ$  can be seen in this plane. Among them those located at  $61^\circ$  (T1 mode) and  $70^\circ$  (T2 mode) originate from the phonons with wave vectors pointing out of the  $(1\bar{1}0)$  plane. It is interesting to recognize that the directions with strong anharmonic effects and strong focusing effects are the same.

In order to observe the effects of anharmonic decay an experiment with monochromatic phonons, whose decays should faithfully reflect the angular dependence given by Fig. 2, would be most favorable. However, a simpler experiment would use a heated film as a phonon source. The nonequilibrium phonons radiated into a crystal may be regarded approximately as a Planckian distribution characterized by the temperature  $T_h$  of the heat source. In Fig. 4 we have plotted for several traveling distances the phonon in-

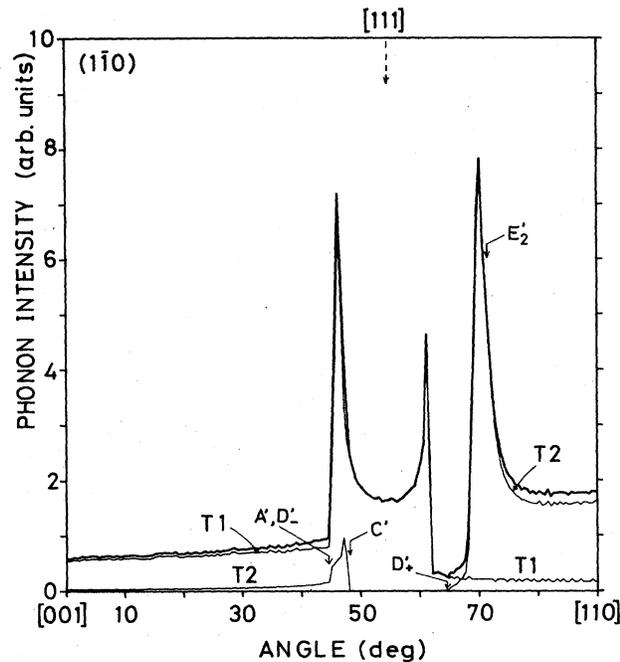


FIG. 3. Angular dependences of TA phonon intensities in the  $(1\bar{1}0)$  plane of NaF. The anharmonic effects are neglected. Results show the energy flux integrated over  $\pm 0.25^\circ$  and  $\pm 0.5^\circ$  within and perpendicular to the  $(1\bar{1}0)$  plane, respectively.

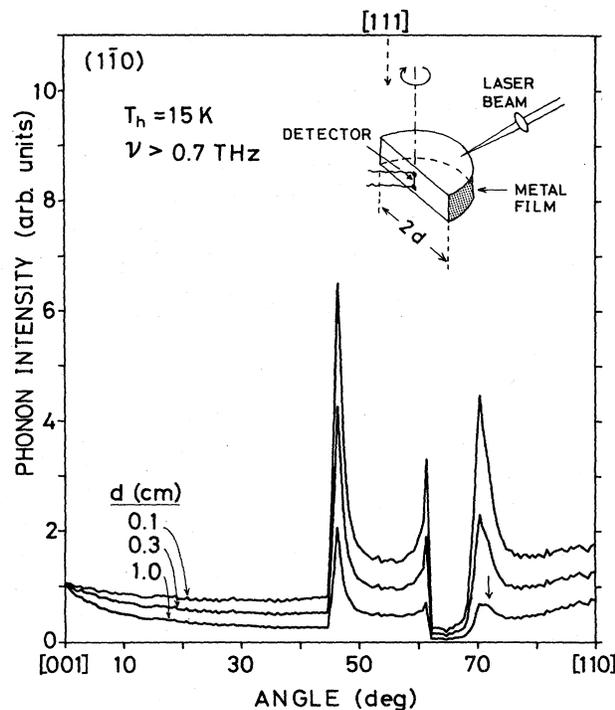


FIG. 4. Angular dependences of phonon intensities for several traveling distances  $d$  in the  $(1\bar{1}0)$  plane of NaF. The anharmonic effects are included, and intensities are obtained by integrating over frequencies  $\nu > 0.7$  THz. The temperature of heater is assumed to be 15 K. Inset shows a possible geometry of heat-pulse experiment (Ref. 14).

tensities versus propagation directions in the same plane calculated under the above assumption with  $T_h = 15$  K. This figure shows intensities for phonons integrated over frequencies higher than 0.7 THz.<sup>21</sup> Accordingly these results correspond to the signals to be detected by a Pb-Ox-Pb tunneling junction detector which responds to the phonons in this frequency range.<sup>22</sup> As can be seen from Fig. 4 the ballistic phonon intensity observed in the (110) plane of NaF changes considerably with the propagation distance. One of the remarkable features observed in Fig. 4 is the fact that the relative magnitude of the ballistic-phonon intensity associated with the caustic of  $T_2$  phonons (located at  $69^\circ$ ) decreases rapidly with increasing the propagation distance  $d$ , and a shoulder (marked by the arrow) appears for a large value of  $d$ . We find that the appearance of this shoulder is really due to a structure of the decay rate being present in Fig. 2. Here, we note that the direction at which the cusp  $E_2$  in Fig. 2 is situated in wave-vector space corresponds to  $E_2'$  in real space (Fig. 3). On the side of  $E_2$  closer to the [110] axis (which corresponds to the directions closer to the [110] axis in the real space) the decay rate of  $T_2$  phonons decreases rather rapidly as the propagation directions are rotated toward the [110] axis, while it decreases more slowly on the other side of  $E_2$ . Therefore, as the frequency or the propagation distance increases, the ballistic phonon intensity of the  $T_2$  mode decreases in the directions closer to the [001] axis of  $E_2'$  more heavily than that in the directions closer to the [110] axis. This explains the remarkable di-

minishing of the phonon intensity in the vicinity of the  $T_2$  caustic and the production of a shoulder for long propagation distances.

We also note that the relative magnitudes of phonon intensities in the [001] and [110] directions become larger for larger values of  $d$ . This is because the TA phonon decays are zero or very small in these directions, and so the phonon intensities in these directions are either not attenuated at all with distance or are attenuated only slightly. The characteristic behaviors of the phonon intensity in the presence of the TA phonon decay could equally be seen at lower heater temperatures if longer path lengths were used.

To summarize, we have found that in crystalline solids the TA phonons of superthermal frequencies can for some directions decay spontaneously as frequently as the LA phonons, though the decay rates show large spatial anisotropy. This is quite different from the conclusion deduced from the isotropic approximation that the TA phonons cannot decay spontaneously except through the collinear processes. The result we have obtained should yield effects observable in the phonon-focusing experiments. Our study also suggests the potential of anharmonic interaction of TA phonons combined with isotope scattering for resolving the unsettled problems on the transport of large-wave-vector phonons in GaAs.<sup>23-25</sup>

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<sup>11</sup>Even a very small amount of normal dispersion prohibits the decay of transverse acoustic phonons.

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<sup>17</sup>S. Tamura (unpublished).

<sup>18</sup>H. J. Maris, *Phys. Lett.* **22**, 402 (1966).

<sup>19</sup>For NaF we have employed the values  $C_{11} = 1.085$ ,  $C_{12} = 0.229$ , and  $C_{44} = 0.290$  in units of  $10^{12}$  dyne  $\text{cm}^{-2}$ , and  $\rho = 2.85$   $\text{g cm}^{-3}$  from J. T. Lewis, A. Lehoczy, and C. V. Briscoe, *Phys. Rev.* **161**, 877 (1967); and  $C_{111} = -14.8$ ,  $C_{112} = -2.7$ ,  $C_{123} = 2.8$ ,  $C_{144} = 0.46$ ,  $C_{166} = -1.14$ , and  $C_{456} = 0.0$  in the units of  $10^{12}$  dyne  $\text{cm}^{-2}$  from W. A. Bensch, *Phys. Rev. B* **6**, 1504 (1972).

<sup>20</sup>For wave vectors marked by  $B_-$  and  $B_+$ , and  $F_-$  and  $F_+$  the group-velocity vectors coincide at  $B'$  and  $F'$ , respectively. We have confirmed that there exists no phonon which can decay into a pair of phonons at  $F_-$  and  $F_+$ . However, for the decay into a pair of phonons at  $B_-(\mathbf{q}')$  and  $B_+(\mathbf{q}-\mathbf{q}')$  there exists a  $T_1$  phonon (with  $\mathbf{q}$  oriented close to the [111] direction) which satisfies exactly the conservation of the wave vectors and  $|\omega(\mathbf{q}) - \omega(\mathbf{q}') - \omega(\mathbf{q}-\mathbf{q}')|/\omega(\mathbf{q}) < 10^{-4}$ .

<sup>21</sup>For NaF the lowest zone-boundary frequency of the TA phonons is 4.4 THz, and for frequencies lower than 3 THz the dispersive effects are negligible as far as the focusing and anharmonic effects are concerned. See, W. J. L. Buyers, *Phys. Rev.* **153**, 923 (1967).

<sup>22</sup>Pb-Ox-Pb tunneling junction should be more suitable than an Al bolometer as the detector, because the anharmonic effects become critical at high frequencies.

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<sup>25</sup>J. P. Wolfe and G. A. Northrop, in Ref. 13, p. 100.