PHONON GENERATION IN KCl:Li^+ AND KCl:OH^-
WITH PULSED ELECTRIC FIELDS

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PHONON GENERATION IN $\text{KCl}:\text{Li}^+$ AND $\text{KCl}:\text{OH}^-$
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ABSTRACT

A pulse electric field techniques has been developed to explore the possibility of using electrically polarizable tunneling defects in crystals as generators of short pulses of phonons of discrete, tuneable frequencies, and has been applied to $\text{KCl}:\text{Li}^+$ and $\text{KCl}:\text{OH}^-$. Phonons generated by the application of high voltage pulses to one end of an impurity doped sample are studied using bolometric detection. By measuring defect-lattice relaxation times it is concluded that in both $\text{KCl}:\text{Li}^+$ and $\text{KCl}:\text{OH}^-$ at least half of the detected phonon energy is a result of generation at discrete frequencies. The remainder of the signal may be a result of panchromatic generation produced by other relaxation processes faster than could be resolved ($\lesssim 1$ ns). Low field relaxation times of 8 ns and 6.5 ns were obtained for $\text{KCl}:\text{Li}^+$ and $\text{KCl}:\text{OH}^-$ respectively, in reasonable agreement with previous estimates obtained with other techniques.^{1,2} These times exhibit electric field and temperature dependences consistent with a single-phonon relaxation model. Scattering by the Li^+ and OH^- impurities resulted in considerable temporal broadening of the phonon pulses.

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I. INTRODUCTION

Certain impurities in alkali halides exhibit quasi-rotational or tunneling motion which gives rise to a splitting of the vibrational ground state of the defect.³ For impurities having an electric dipole moment this level structure may be altered through the application of an external electric field as has been verified in numerous experiments including measurement of specific heat,⁴ dielectric constant,⁵ para-electric resonance (PER),⁶⁻⁸ and para-electric cooling.^{1,9} The purpose of the present work is to explore the possibility of using this property of polarizable defects for the generation of short pulses of phonons of discrete tuneable frequencies.

Phonons are generated as a result of the defect-host interaction whenever the dipoles orient to an external electric field. It will be shown in Section II that if an electric field is applied with a risetime, τ_E , less than the defect lattice relaxation time, τ_{DL} , the possibility of monochromatic phonon generation exists. To investigate this, a pulsed electric field was applied to one end of an impurity doped crystal while the generated phonons were detected with a bolometer at the opposite end after their ballistic transit of the sample. Information on the frequency spectrum of the generated phonons was deduced from measurements of τ_{DL} obtained from the dependence of the emitted phonon energy on the length of the applied electric field pulses. The method employed in the determination of τ_{DL} is very direct and is a general means of measuring extremely short relaxation times, the shortest time being limited by τ_E , which is on the order of 0.5 ns in the present experiment.

The time resolution available with the pulse technique enables longitudinal and transverse phonons to be resolved. Also it provides information on the amount of scattering undergone by the generated phonons allowing an estimate of the phonon mean free path to be obtained. The temporal resolution

of bolometric detection compliments the spectral resolution achieved by monochromatic detection as used in the PER work on KCl:Li⁺ b. Larson and Silsbee.⁸

KCl:Li⁺ and KCl:OH⁻ were chosen for the experiment since these systems possess large dipole moments and have large polarizabilities near 1°K.⁹ Furthermore, their relaxation times have been estimated by several techniques to be on the order of a few nanoseconds.^{1,2,6} This ensures that the condition $\tau_E \ll \tau_{DL}$ can be satisfied while at the same time τ_{DL} is short enough that high time resolution is, in principle at least, possible.

II. THEORY

The generation of phonons in dipolar impurity systems upon application of an electric field arises through a change in the thermal occupation of the impurity states caused by an alteration of the energy level spacings. The mechanism can be understood by considering a two level model for the impurity ground state (Fig. 1a). This is an oversimplification of the multi-level structures actually encountered in crystals but does not affect the underlying principle.¹⁰ The ground state multiplets of Li⁺ and OH⁻ in KCl can be found in references 4 and 11.

In thermal equilibrium with no applied electric field the levels are occupied according to Boltzmann statistics with

$$N_1(\Delta_0) = N[1 + \exp(-\Delta_0/k_B T)]^{-1} \quad (1)$$

defects in level 1 and

$$N_2(\Delta_0) = N - N_1(\Delta_0) \quad (2)$$

defects in level 2, where N is the total number of polarizable impurities and Δ_0 is the zero-field splitting. When an electric field is applied, the

system seeks a new distribution appropriate for the splitting Δ_E . Relaxation is in general achieved through multi-phonon decay processes. However, at low temperature ($T < 5^\circ\text{K}$) dielectric constant measurements in NaBr:F by Rollefson⁵ as well as relaxation time measurements in OH⁻ doped alkali halides by Lütty¹² indicate that single phonon decay processes dominate. When defect relaxation is by one-phonon processes the total number of phonons emitted upon application of a field E is

$$\hat{N}(\Delta_E) \equiv N_2(\Delta_0) - N_2(\Delta_E) \quad (3)$$

while the energy spectrum of these phonons is governed by the rate at which the electric field is applied compared with τ_{DL} . For $\tau_E \gg \tau_{DL}$ (Fig. 1b) the defects are able to establish a quasi-equilibrium as the levels are being pulled apart. As a result the phonon spectrum will be continuous with energies between Δ_0 and Δ_E . This is the limit which in electrocaloric work results in equal amounts of heating and cooling upon polarization and depolarization of a sample. If, however, $\tau_E \ll \tau_{DL}$ (Fig. 1c), there is no opportunity for phonon emission before the field has reached its maximum value. Thus, when relaxation occurs, all of the phonons $\hat{N}(\Delta_E)$ will have the same energy Δ_E . Since Δ_E is controlled by the electric field, the system acts as a source of tunable monochromatic phonons. The frequency bandwidth of these phonons is determined either by the natural linewidth $(2\rho\pi_{DL})^{-1}$ or by strain broadening and random electric fields.^{6,11}

For $\tau_E \ll \tau_{DL}$ the total phonon energy is

$$U_{\text{tot}}^e = \Delta_E \hat{N}(\Delta_E) \quad (4)$$

This energy is released through the exponential relaxation of the excess population of level 2 to its Boltzmann equilibrium value, the time dependence of the emitted energy being given by

$$U^e(t_p) = U_{\text{tot}}^e [1 - \exp(-t_p/\tau_{DL})] \quad (5)$$

where t_p is the time after application of the field. Experimentally, τ_{DL} may be determined by applying square ($\tau_E \ll t_{DL}$) voltage pulses of variable length t_p to a crystal and monitoring with a bolometer the exponential dependence of $U^e(t_p)$ on pulse length. The amplitude of a bolometer signal $\Delta T(t_p, t)$ is a measure of the power per unit area incident on the detector at the time t and its time integral

$$A(t_p) \equiv \int_0^\infty \Delta T(t_p, t') dt' \quad (6)$$

is proportional to $U^e(t_p)$.¹³ The relaxation time is determined from the expression

$$\tilde{A}(t_p) \equiv A(t_p = \infty) - A(t_p) = A(\infty) \exp(-t_p/\tau_{DL}) \quad (7)$$

Here it should be emphasized that (7) represents the expected behavior for a two level system characterized by a single relaxation time τ_{DL} . In multi-level systems such as KCl:LI^+ and KCl:OH^- one should expect to find several distinct relaxation times associated with the numerous allowed transitions, in which case the observed signal would be of the form

$$A(t_p) = \sum_i A_i(\infty) [1 - \exp(-t_p/\tau_{DL}^i)] \quad (8)$$

Somewhat surprisingly, no multiple relaxation processes have thus far been reported for tunneling defects.

A potential complication associated with the use of voltage pulses is that phonon emission after application of the electric field is followed by phonon absorption when the field is removed, for after the levels return to their zero-field splitting the occupation number of level 2 is less than its thermal equilibrium value. In establishing equilibrium as many phonons must be absorbed as were emitted when the field was applied. Now, however, the

energy per phonon is Δ_0 rather than Δ_E and the absorbed energy is

$$U^a(t_p) = -\Delta_0[U^e(t_p)/\Delta_E] \quad (9)$$

As a result of this 'self-absorption' the total energy appearing at the detector is given by the sum of the energies of the two processes. That is

$$A(t_p) \propto U^e(t_p) + U^a(t_p) = \gamma U^e(t_p) \quad (10)$$

where

$$\gamma \equiv (\Delta_E - \Delta_0)/\Delta_E$$

The temporal separation between emission and absorption is the length of a voltage pulse and is, in this experiment, always at least an order of magnitude shorter than the resolution time of the detector (which is limited by geometry to about 2 μ s). Since the two processes cannot be resolved, not only the total energy, but also the instantaneous observed phonon power is reduced by the factor γ as a result of self-absorption.¹⁴ γ is expected to be in the range 0.4 to 0.8 in both KCl:Li⁺ and KCl:OH⁻ for the fields applied in this experiment.

III. EXPERIMENTAL ARRANGEMENT

A schematic of the cryogenic arrangement is shown in Fig. 2. Samples were prepared so as to be impurity doped only in a 4 mm long region between the high voltage electrodes (generating region). This was done to reduce scattering of the generated phonons before their detection at the bolometer. These crystals were obtained from <100> cleaving of specially grown "half-doped" boules obtained by seed pulling about two centimeters of pure KCl from a melt, adding the appropriate amount of dopant to the remaining melt and continuing growth. The location of the boundary between pure and doped regions was determined by chemical analysis in the case of KCl:Li⁺ and by

ultraviolet absorption in the case of KCl:OH^- and could be resolved to within ± 1 mm. Overall sample dimensions after cleaving were approximately 15 mm x 6 mm x 4mm. The thickness of the generating region was reduced from 4 mm to 2.2 mm by grinding, enabling fields in excess of 20 kv/cm to be applied. The samples were annealed at 550°C for two hours.

The detector end of the samples was ground with 3 micron garnet powder and chemically polished.^{15,16} Zig-zag indium film bolometers with 50 Ω room temperature resistance were evaporated onto the surfaces as described by Channin.¹⁶ These had resistance ratios between room temperature and 4°K of about 25 and typical sensitivities at 1.2°K of 5 Ω /°K.

The cryostat consisted of a top plate supporting a rectangular loop of 50 Ω stainless steel (s.s.) co-axial conductor terminated at one end with 50 Ω and an RG-174/U cable for the transmission of the bolometer signal. Samples were mounted in a copper box at the bottom of the s.s. loop with the detector end protruding through a hole. The box was effective in shielding the bolometer from electromagnetic radiation produced by the rapid-risetime voltage pulses being applied to the opposite end of the sample. The pulses were tapped from the s.s. conductor, access to the center lead being gained by means of a port cut in the outer sheath. Samples were immersed directly in a bath of He^4 pumped to 1.2°K. The magnetic field for biasing the bolometer was provided by a superconducting solenoid.

Because of the high voltage of the input pulses (about 4.5 kv) and the very small signal voltages (tens of microvolts), the elimination of ground loops in the electronics was extremely important and caused considerable difficulties. It was found essential that the two ends of the stainless steel rectangular loop and the bolometer coax be electrically isolated from each other at the top plate of the cryostat.

The high voltage pulses were generated by discharging an RG-142/U coaxial cable through a mercury wetted reed switch capsule (type HG from C. P. Clare Co., Chicago). Using the switch as a spark gap rather than a vibrating contact, output pulses of up to 4.5 kv could be achieved. A Tektronix 519 oscilloscope showed the pulse risetimes to be about 0.5 ns independent of voltage. By varying the cable length, pulse lengths between 2 ns and 100 ns were obtained.

The bolometers were biased with a current between 40 ma and 70 ma. Voltage signals produced by the phonons were amplified with a Tektronix 1121 amplifier followed by a Hewlett-Packard 465A amplifier. Signals were monitored on an oscilloscope and extracted from the noise with a PAR model 160 boxcar integrator. A PIC C-88 pulse transformer was employed to match the impedance of the bolometer to the Tektronics amplifier. This provided a factor of six in essentially noise free gain. The total gain exclusive of the boxcar was about 3×10^3 and the largest signal to noise ratio under optimum conditions was approximately four to one prior to integration.

IV. RESULTS AND DISCUSSION

An oscilloscope trace of a bolometer signal for $\text{KCl}:\text{Li}^+$ ($n_{\text{Li}^+} = 1.8 \times 10^{18} \text{ cm}^{-3}$) is shown in Fig. 3. The absence of a signal in pure KCl and a scaling with concentration indicate that the phonons result entirely from the polarizable defects. Traces for an OH^- doped sample ($n_{\text{OH}^-} = 1.2 \times 10^{18} \text{ cm}^{-3}$) were similar to those of $\text{KCl}:\text{Li}^+$ but about fifty percent larger for a given concentration and electric field. The bolometer signal shows a sharp rise at $2.5 \mu\text{s}$ and a kink at $6 \mu\text{s}$. These are caused by the ballistic arrival of longitudinal and transverse phonons respectively. A broad diffusive peak following the transverse signal as well as the breadth of the ballistic pulses is an indication of

phonon scattering within the sample. This scattering is caused largely by the defects in the generating region and only to a lesser degree by sample boundaries. Broadening resulting from the geometric extent of the generating region and detector is expected to be at most 2 μ s and cannot account of the width of the ballistic signals. Implications as to the frequency spectrum of the observed phonons as a result of scattering have been discussed by Larson and Silsbee.⁸ From the position of the diffusive peak an approximate value for the phonon mean free path λ_R in the generating region can be obtained using Guyer's theory for diffusive heat pulse propagation.¹⁷ This yields a mean free path on the order of 0.5 mm which is consistent with a value of 0.3 mm determined from thermal conductivity.¹⁸ $\lambda_R \approx 0.5$ was also obtained for the OH⁻ doped sample, in reasonable agreement with a thermal conductivity value of 0.2 mm.¹⁹

The energy per unit bolometer area $u_b(t_p)/A_b$, as determined from the area under the trace of Fig. 3 (see footnote 13) is also in reasonable agreement with expectation. An estimated value $u_b^{calc}(t_p)/A_b$ is obtained as described in footnote 13 using $U^e(t_p) \approx U_{tot}^e$ where U_{tot}^e is given by

$$U_{tot}^e \approx C \Delta T_g \quad (11)$$

Here C is the heat capacity of the generating region and ΔT_g is the amount by which the temperature of that region would increase upon application of an electric field if it were thermally isolated from its surroundings. ΔT_g is obtained from electrocaloric data.²⁰ For the experimental conditions of Fig. 3, $u_b^{calc}(t_p)/A_b \approx .06$ erg/cm² as compared with the value $u_b(t_p)/A_b \approx .02$ erg/cm² obtained from the observed signal. For the OH⁻ doped sample $u_b^{calc}(t_p)/u_b(t_p) \approx 0.6$.

The general shape of the trace in Fig. 3 was found to be independent of electric field and pulse length. This scaling enabled the peak amplitude

$S(t_p)$, to be used in place of the area under the trace, $A(t_p)$, as a measure of the energy incident on the detector. By monitoring $S(t_p)$ as a function of pulse length the phonon signal for both KCl:Li^+ and KCl:OH^- was seen to consist of two distinct components. The one decreased in magnitude with decreasing pulse length in accordance with (7) [replacing $A(t_p)$ by $S(t_p)$] while the other was pulse-length independent even for the shortest pulse of 2 ns. These two components are resolved on a semi-log plot of $\tilde{S}(t_p) \equiv S(\infty) - S(t_p)$ vs. t_p (Fig. 4). The pulse-length independent residual signal S_R is determined by extrapolating the straight line of the figure to the intercept at $t_p = 0$. S_R is equal to $S(\infty) - S(0)$ and was found to be $40 \pm 10\%$ of $S(\infty)$ in KCl:Li^+ and $50 \pm 10\%$ in KCl:OH^- . Although the residual signal did not depend on the length of the pulses a dependence on electric field rise-time was observed. Fluctuations in τ_E between 0.5 ns and 1 ns resulting from variations in the position of the spark inside the reed switch capsule produced signal fluctuations of as much as $\pm 10\%$. Such fluctuations were extremely difficult to control over a time span sufficiently long to record data (10 min) and it is fluctuations in S_R which are directly responsible for the large error bars. A discussion of possible mechanisms which could produce the pulse-length independent signal is deferred to a later point.

The relaxation time τ_{DL} for the processes associated with the pulse-length dependent component of the signal is determined from the slope of the $S(t_p)$ vs. t_p plot and is 6 ns in KCl:Li^+ for the electric field and temperature indicated in Fig. 4. The electric field dependence of τ_{DL} is shown in Fig. 5 for KCl:Li^+ . The relaxation time is 8 ns independent of field to about 15 kv/cm but decreases rapidly at higher fields, falling to half its low field value at about 25 kv/cm ($pE/k_B T = 3$). In KCl:OH^- τ_{DL} was found to be 6.5 ns independent of field to 15 kv/cm which was the highest field applied to that sample. If the result $pE/k_B T = 3$ obtained for KCl:Li^+ is used as a criterion, τ_{DL} for

KCl:OH⁻ might be expected to decrease to half its low field value at a field of 40 kv/cm. The values of τ_{DL} measured here are in favorable agreement with those obtained previously by other workers. Kapphan¹ has estimated a relaxation time of about 10 ns for KCl:Li⁺ from paraelectric cooling measurements, while Brugger² has obtained 3 ns for KCl:OH⁻ from ultrasonic attenuation.

The field dependence of τ_{DL} exhibits the qualitative behavior expected from a one phonon relaxation model²¹ which predicts a field-independent τ_{DL} at low fields and a Δ_E^{-1} decrease at high fields. This model has been used quantitatively by Pirc. et al.²² to explain the stress dependence of the orientation time of the O₂⁻ defect in alkali halides observed by Kanzig²³ and also by Kapphan²⁴ to explain the field dependence of τ_{DL} in other doped alkali halides.

The temperature dependence of τ_{DL} measured in KCl:OH⁻ (Fig. 6) is consistent with a T^{-1} behavior also predicted by the one-phonon relaxation model and is evidence that phonon generation is indeed via single-phonon processes. Measurements are restricted to a narrow range of temperature as a result of the rapid decrease in signal with increasing temperature and to difficulties in maintaining a sufficient temperature stability above the λ -point of He⁴. A T^{-1} dependence has been observed at low temperatures ($T < 5^\circ\text{K}$) by Rollefson⁵ and Lüty¹² in dipolar systems with longer relaxation times. It should be noted that theoretical predictions of the temperature dependence of single-phonon processes have been made only for the case $pE \ll \Delta_0$. Experimental data of Lüty, however, shows that the T^{-1} behavior extends even to the regime $pE \lesssim \Delta_0$.¹²

In light of the temperature dependence of τ_{DL} and the fact that $\tau_{DL} \gg \tau_E$, it is concluded that the pulse-length dependent component of the observed phonon signal is a result of generation at discrete frequencies. As for the

large residual signal S_R , the frequency spectrum here depends on the mechanism by which these phonons were produced. Two possible explanations for a pulse-length independent signal are (1) broad-band microwave pumping of the defect levels by high frequency Fourier components in the voltage pulses and (2) the existence of a second relaxation process faster than the shortest pulse of 2 ns. The first of these could imply discrete-frequency generation characterized by the same relaxation time measured for the pulse-length dependent signal, whereas the second would imply a panchromatic spectrum for S_R since the criterion $\tau_E \ll \tau_{DL}$ is not met.

Microwave phonon generation can arise if the electric field pulses contain Fourier components within the strain-broadened linewidth of the defects near the resonant frequency $\Delta_E/2\pi\hbar$. These components would contribute only in the rapid rise and fall of the pulses since $\Delta_E/2\pi\hbar \gg \tau_E^{-1}$. The additional defects pumped to the higher states would relax as discussed in Section II (with the same τ_{DL}). However, as a result of the zero-field splitting (which is comparable with Δ_E) Δ_0 - phonon generation will occur after the end of every pulse, independent of length. An estimate of the magnitude of the signal resulting from microwave excitation depends strongly on the functional form used to characterize the rise and fall of the voltage pulses. An upper limit estimate using a linear rise and fall indicates that a signal of the observed magnitude could conceivably be produced. However, the 10 GHz and higher frequencies required to resonantly excite the OH^- and Li^+ transitions should be severely attenuated in propagating through the co-axial conductor and in coupling to the sample. Attenuation of these frequencies would have had no observable effect on the 0.5 ns risetime pulses. Thus it is felt unlikely that the signal results from microwave generation. However, on the basis of the upper limit estimate the effect cannot a priori be ruled

out.

If a faster relaxation time existed this would be the first such observation for tunneling impurities. Considering that the ground states of KCl:Li^+ and KCl:OH^- actually consist of eight and six levels respectively and are not the idealized two level model described in Section II, it is not unreasonable to expect that the numerous allowed phonon transitions (the number of which is increased by random internal stresses) are characterized by different relaxation times. That a relaxation time comparable with the 0.5 ns pulse risetime may in fact exist has been shown in recent electron spin resonance experiments on KCl:Li^+ by Osswald and Wolf.²⁵

As pointed out earlier, the observed bolometer signal (Fig. 3) has a broad diffusive appearance resulting from phonon scattering in the generating region. The loss of temporal resolution makes a determination of the relative amplitude of longitudinal to transverse phonon signals impossible. In principle such a measurement could be used to extract information about the matrix elements coupling the defects to these modes and hence about the phonon transition probabilities, as has been discussed by Channin.¹⁶

This broadening also prevents KCl:Li^+ and KCl:OH^- from being useful generators of sharp pulses of discrete-frequency phonons. In order to produce such pulses systems with a weaker defect-host coupling must be found, relaxation times on the order of 100 ns being ideal. Defects longer lived than this lose their attractiveness since time resolution of the emitted phonons is lost when the lifetime becomes comparable with the phonon transit time across the crystal. A 100 ns lifetime would also considerably alleviate experimental difficulties associated with the use of extremely fast voltage pulses. Unfortunately, all tunneling impurities studied thus far have relaxation times either less than 10 ns or greater than 1 μs . If defect systems

with an appropriate relaxation time and energy level structure were found, the experimental technique could prove useful for the generation of pulses of tunable monochromatic phonons.

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REFERENCES

1. S. Kapghan and F. Lüty, Solid State Commun. 6, 907 (1968).
2. Brugger, Fritz, Kleinman, J. Acoust. Soc. Am. 41, 1015 (1967).
3. For a review of tunneling defects in solids see V. Narayanamurti and R. O. Pohl, Reviews of Modern Physics 42, 201 (1970).
4. J. P. Harrison, P. P. Peressini, R. O. Pohl, Phys. Rev. 171, 1037 (1968).
5. R. J. Rollefson, Ph. D. Thesis. Department of Physics, Cornell University, Cornell Materials Science Center Report No. 1382, 1970, and Phys. Rev. B 5, 3235 (1972).
6. R. A. Herendeen, Ph. D. Thesis, Department of Physics, Cornell University, Cornell Materials Science Center Report No. 1323, 1970 (unpublished).
7. D. J. Channin, V. Narayanamurti, and R. O. Pohl, Phys. Rev. Letters 22, 524 (1969).
8. T. R. Larson and R. H. Silsbee, Phys. Rev., this issue.
9. R. O. Pohl, V. L. Taylor, and W. M. Goubau, Phys. Rev. 178, 1431 (1969).
10. For a more extensive treatment see S. Kapghan, Ph. D. Thesis, Univ. of Utah, 1970 (unpublished).
11. W. E. Bron and R. W. Dreyfus, Phys. Rev. 163, 304 (1967).
12. S. Kapghan and F. Lüty, Solid State Commun. 8, 349 (1970).
13. In detail, $\Delta T(t_p, t)$ is the change in temperature of the detector above its ambient value T_a caused by the thermalization of the incident phonons. From black-body theory, for $\Delta T \ll T_a$, $A(t_p)$ is related to the energy per unit bolometer area $u_b(t_p)/A_b$ incident on the detector according to the expression

$$A(t_p) = (1/4\sigma T_a^3) u_b(t_p)/A_b$$

where σ is the phonon equivalent of the Stefan-Boltzmann constant of photon radiation theory and has the value $1.7 \times 10^6 \text{ erg cm}^{-2} \text{ sec}^{-1} \text{ } ^\circ\text{K}^{-4}$

for a perfect acoustic match of the detector to KCl. $u_b(t_p)$ in turn is given by $\Omega U^e(t_p)$ where Ω is the solid angle subtended by the bolometer at the generating region and is in first approximation equal to $A_b/4\pi\ell^2$, ℓ being the distance between generating region and detector.

14. This statement assumes implicitly that the time characteristic of absorption is not severely limited by the availability of phonons of energy Δ_0 within the strain-broadened line-width of the Li^+ and OH^- defects. Evidence that such a "phonon bottleneck" does not exist is given by Kapphan and Lüty¹² who have obtained identical relaxation times from polarization as well as depolarization of OH^- doped crystals for values of τ_{DL} as short as 10^{-6} sec.
15. A. Taylor, H. R. Albers, and R. O. Pohl, J. Appl. Phys. 36, 2270 (1965).
16. D. J. Channin, Ph. D. Thesis, Department of Physics, Cornell University, Cornell Materials Science Center Report No. 1111, 1970 (unpublished).
17. C. C. Ackerman and R. A. Guyer, Annals of Physics 50, 128 (1928).
18. This estimate is obtained in the dominant phonon approximation using Fig. 22 of Ref. 3. Phonons generated by applying an 18 kv/cm field have a mean free path approximately given by the mean free path of thermal phonons at 1°K. The mean free path was scaled to correct for the different concentration.
19. Estimate based on data of R. L. Rosenbaum, Ph. D. Thesis, Department of Physics, University of Illinois, 1968.
20. Data obtained from Fig. 5. and Fig. 6 of Ref. 7 scaled to the appropriate concentrations and electric fields.
21. B. G. Dick, Phys. Stat. Sol. 29, 587 (1968).
22. R. Pirc, B. Žekš, and P. Gosar, J. Phys. Chem. Solids 27, 1219 (1966).

23. W. Kanzig, J. Phys. Chem. Solids 23, 479 (1962).
24. S. Kapphan, Ph. D. Thesis, University of Utah, 1970 (unpublished).
25. R. Osswald and H. C. Wolf, Phys. Stat. Sol. 50, K93 (1972).

FIGURE CAPTIONS

Figure 1. (A) Energy level splitting for a two level model as a function of electric field. The levels are occupied according to Boltzmann statistics, equilibrium being achieved by phonon emission (indicated by arrow) characterized by the defect-lattice relaxation time τ_{DL} . At low temperatures the phonon frequency spectrum is determined by the rate at which the electric field is applied compared with τ_{DL} . (B) Level splitting as a function of time for $\tau_E \gg \tau_{DL}$. The defects reach a quasi-equilibrium distribution as the levels are pulled apart by the field, resulting in a continuous phonon spectrum with energies between Δ_0 and Δ_E . (C) Level splitting as a function of time for $\tau_E \ll \tau_{DL}$. There is no opportunity for phonon emission before the electric field has reached its maximum value. When relaxation takes place all phonons have the same energy Δ_E .

Figure 2. Schematic of the cryogenic arrangement. The sample is mounted in a copper box (not shown) with the detector end protruding through a hole. Samples are doped only in the generating region between silver paint electrodes. High voltage pulses are tapped from a hole in the stainless steel conductor at the bottom of the loop. Phonons generated by the pulses are detected after ballistic transit of the undoped region of the crystal with an indium bolometer biased in its transition region with a magnetic field. Samples are immersed in liquid He^4 pumped to 1.2°K.

Figure 3. Oscilloscope trace of a phonon signal for $KCl:Li^+$. Ballistic longitudinal phonons cause an abrupt initial rise at 2.5 μs which is followed by a kink at 6 μs caused by ballistic transverse phonons. A diffusive signal peaking at 10 μs appears as a result

of phonon scattering by the Li^+ ions in the generating region. The initial spike at the left is direct electromagnetic pickup from the high voltage pulses. The area under the trace corresponds to an energy per unit bolometer area of 0.02 erg/cm^2 while the amplitude at the peak corresponds to a temperature change of the detector of $0.1 \text{ m}^\circ\text{K}$. Bolometer bias current was 40 ma . $T = 1.2^\circ\text{K}$.

Figure 4. Change in phonon signal vs. pulse length for $\text{KCl}:\text{Li}^+$. The data indicate two independent components in the signal, one decreasing with pulse length, the other pulse-length independent. The pulse length independent residual signal S_R is determined by extrapolating the straight line to $t_p = 0$ and is given by $S(\infty) - S(0)$. Here $S(\infty) = 14$ while $S(0) = 9$ implying that S_R is approximately 40% of $S(\infty)$. Fluctuations in S_R are the major contribution to the error bars. The pulse length dependent signal follows an exponential decay as predicted by (7). τ_{DL} is 6 ns for the particular electric field indicated. $T = 1.2^\circ\text{K}$.

Figure 5. Electric field dependence of τ_{DL} for $\text{KCl}:\text{Li}^+$. The relaxation time is field independent below 15 kv/cm but decreases for higher fields reaching half its low field value at about 25 kv/cm ($pE/k_B T = 3$). This behavior is qualitatively similar to the stress dependence of the O_2^- defect in alkali halides.

Figure 6. Temperature dependence of τ_{DL} in $\text{KCl}:\text{OH}^-$. The data is consistent with the T^{-1} behavior expected for single-phonon decay processes.

Figure 1.

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.