GENERATION OF MICROWAVE FREQUENCY PHONONS IN KCl:Li *

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(Received 22 January 1969)

Phonons have been generated by resonant excitation of the tunneling states of Li^+ in KCl with 24-GHz microwaves. Directional phonon signals of 10^{-3} W have been detected.

We have observed the generation of monochromatic phonons through microwave excitation of paraelectric ions in a crystal host, namely of Li^+ in KCl. The lithium tunneling states have an energy splitting of 10^{-4} eV which can be tuned with an electric field[1] and have a lifetime of about 10^{-9} sec. This very short lifetime allows efficient generation of microsecond pulses of milliwatt acoustic power at microwave frequencies.[2]

24-GHz microwave pulses of about 1-μsec duration from a Klystron tube of 600-mW peak power were transmitted through a wave guide to the sample cavity immersed in helium at 1.3°K. A bolometer made with evaporated superconducting indium film was used to detect the phonons.[3] A gated integrator (boxcar) was often used to improve the signal-to-noise ratio.[4] The power of the phonon signal was typically on the order of 1 mW.

Figure 1 demonstrates that the relative intensity of the longitudinal and transverse ⟨100⟩ phonon signal depends on the angle between the microwave electric field and the phonon propagation direction. These results were obtained by mounting the same sample in different orientations within a low-Q rectangular cavity. Lithium was diffused[5] about 2 mm into a stub at one end of a 1-cm long piece of pure KCl; see Fig. 1. The stub was inserted through a port in the cavity wall so that the lithium ions were in the high electric field region. The first signal was caused by heating of the bolometer resulting from microwave leakage through the cavity port. The two subsequent rapid rises were caused by phonons traveling directly through the crystal at the speed of longitudinal and transverse sound, respectively. The geometry of the crystal, reflections off the walls, and duration of the microwave pulse prevented complete separation of these events, but the arrival times of the longitudinal and transverse modes and their relative intensity (from the slopes of the arriving pulses) could be well resolved. The arrival of the first phonon pulse coincided with the known ⟨100⟩ sound velocity. The second pulse came in 15% sooner than the transverse ⟨100⟩ mode should. We attribute this early arrival to anisotropy of the transverse sound velocity in KCl causing slightly off-axis phonons to travel faster than the ⟨100⟩ phonons.[6]

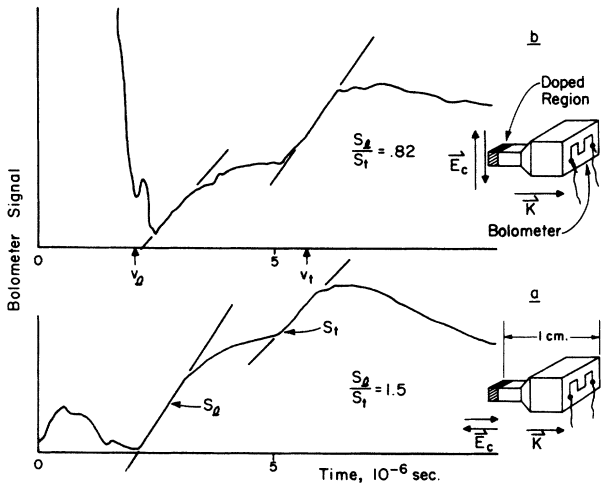

FIG. 1. Bolometer signal (obtained using boxcar integrator) with phonon propagation vector $\vec{k} \| \langle 100 \rangle$ (a) parallel to cavity electric field $\vec{E}_C$ and (b) perpendicular to $\vec{E}_C$. S_l and S_t are longitudinal and transverse slopes and are a qualitative measure of the elastic power. Distortion of field by cavity port prevented complete discrimination between $\vec{E}_C \| \vec{k}$ and $\vec{E}_C \perp \vec{k}$.

With the electric field $\vec{E}_C$ parallel to the propagation direction, Fig. 1(a), the relative intensity of the longitudinal to transverse phonons is 1.5 while in Fig. 1(b), with the field perpendicular, the ratio is 0.8. We take this difference in relative intensity and the ballistic arrival times of the phonons as evidence that the phonons did not come from a thermal process and had not suffered multiple scattering in transit.

Figure 2 shows the influence of Stark splitting of the tunneling states on the phonon generation. The KCl crystal was uniformly doped with about 10^{17}-cm^{-3} lithium ions and mounted in the configuration of Fig. 1(a). Electrodes for applying a static ⟨111⟩ field were silver painted onto the crystal surfaces outside the cavity. The upper trace shows the signal with no applied field. The lack of model resolution and the early arrival shows that the phonons were created substantially throughout the sample, apparently by micro-

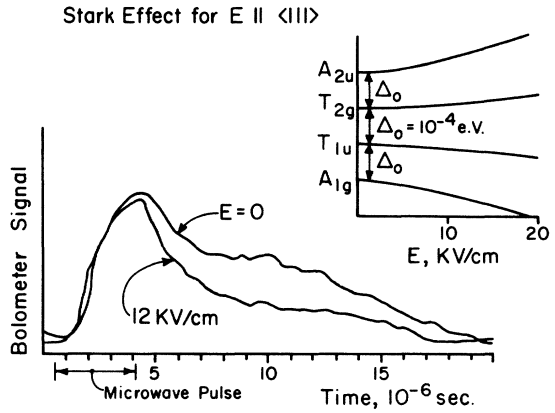

FIG. 2. Bolometer signal (from oscilloscope photographs) obtained with and without applying a static ⟨111⟩ electric field to the doped crystal. The magnitude of the effect was influenced by fringing of the field at the edges of the electrodes and by attenuation of the microwaves by the lithium ions.

waves transmitted out of the port and confined by the "wave guide" formed by the field electrodes. The rapid decay of the phonon pulse shows that there was negligible scattering of the phonons by the lithium in this low concentration. On application of a 12-kV/cm field the tunneling energy levels spread apart and moved out of resonance with the microwaves, which resulted in the decreased phonon generation shown in the lower trace.[7] This demonstrates that the phonons we observed were indeed created through a resonance excitation of the tunneling states by the microwaves.

We now consider the phonon generation mechanism. 24-GHz phonons can couple any two neighboring tunneling states; see the insert in Fig. 2. Thermal-conductivity experiments indicate that elastic waves can couple any two of the tunneling states and give no evidence for anharmonic processes.[8,9] From this we conclude that simultaneous emission of two phonons is unlikely; therefore, the phonons generated should have the energy Δ_0, $2\Delta_0$, or $3\Delta_0$. Note that the generation of phonons of energy $2\Delta_0$ or $3\Delta_0$ depends on the thermal population of the excited tunneling states. At the temperature of our experiment, $3\Delta_0$ phonons should be negligible, and below 1°K, even $2\Delta_0$ phonons should not be generated any longer. In the context of this Letter, the important result is that the emitted phonons are not "thermal" ones, but belong to relatively narrow frequency bands (or even to only one band).

Emission of phonons with energy Δ_0 can be explained as arising from a three-dimensional dipole oscillator which is driven by an ac electric field and which generates stress waves in a surrounding elastic medium, setting up longitudinal waves when the dipole axis is parallel to the propagation direction and shear waves when perpendicular. This explains why the relative intensity of longitudinal and transverse phonons depends on the angle between the microwave electric field and the phonon propagation direction. In terms of the tunneling states it implies that only the particular states of the triplets T_{1u} and T_{2g} that are excited by the microwaves generate the phonons, i.e., the time for the coupling between the different states of each triplet is longer than the time for phonon emission.

Our measurements do not give direct information on the spectral line shape of the emitted phonons. They can be emitted incoherently with the exciting microwaves. In that case their spectral width would be of the order of the linewidth of the tunneling states, which is about 10^9 $\sec^{-1}$, resulting from the lifetime of the states and of random internal stress. They can also be emitted coherently. In that case of resonance fluorescence,[10] the linewidth would be given by the linewidth of the exciting microwaves, i.e., 10^{-6} $\sec^{-1}$. At temperatures where $kT \ll \Delta_0$ we expect this process to dominate, as long as interactions between the individual lithium ions can be ignored. These questions go beyond the scope of this Letter and will be discussed separately.

We plan to use the generation of phonons by microwave excitation of paraelectric ions to study directly the interaction between defect states and phonons. We further hope to develop this technique into a tunable[1] source of directed monochromatic phonons at microwave frequencies usable for phonon spectroscopy.

We gratefully acknowledge stimulating discussions with C. H. Anderson, A. T. Fiory, W. M. Goubau, J. A. Krumhansl, R. J. Rollefson, and E. S. Sabisky. We thank S. Othmer for his help and advice regarding the microwave technology.

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*Work supported by the U. S. Atomic Energy Commission under Contract No. (AT30-1)-2391 (NYO-2391-90). Additional support was received through the Advanced Research Projects Agency by use of the space and facilities of the Cornell Materials Science Center.

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[1]In a field $E = 25$ kV/cm applied along a ⟨111⟩ direction the tunnel splitting doubles. See also the insert of Fig. 2. J. P. Harrison, P. P. Peressini, and R. O. Pohl, Phys. Rev. 171, 1037 (1968). This paper also

contains references to earlier work on the tunneling states in KCl:Li.

[2]Previous work in this field has emphasized paramagnetic impurities. Their long spin-lattice relaxation times prevent generation of ballistic pulses of phonons and the low saturation-power levels, and weak microwave absorption compared with paraelectric systems reduces the maximum power available. See C. H. Anderson and E. S. Sabisky, Phys. Rev. Letters 21, 987 (1968), and W. J. Brya and D. E. Wagner, Phys. Rev. 157, 400 (1967), for recent progress and reference to earlier work.

[3]Such bolometers can be used to detect both thermal pulses and monochromatic ultrasonic pulses. See John M. Andrews, Jr., and M. W. P. Strandberg, J. Appl. Phys. 38, 2660 (1967).

[4]W. Gilbert Clark and A. L. Kerlin, Rev. Sci. Instr. 38, 1593 (1967).

[5]R. C. Hanson, Bull. Am. Phys. Soc. 13, 902 (1968).

[6]For phonons traveling under 30° from the ⟨100⟩ axis, this anisotropy in KCl can increase the transverse velocity by 50% while reducing the longitudinal velocity by only 10%.

[7]The original intent of this experiment was to demonstrate the monochromatic nature of the phonons by transmitting them through a scattering medium of resonant lithium ions and observing the signal to increase on application of a field that tunes the scatterers out of resonance with the phonons. Weak scattering and microwave leakage produced the opposite effect, namely a decrease of signal with electric field.

[8]D. Walton, Phys. Rev. Letters 19, 305 (1967).

[9]P. P. Peressini, J. P. Harrison, and R. O. Pohl, to be published.

[10]Optical resonance fluorescence is discussed in detail by W. Heitler, Quantum Theory of Radiation (Oxford University Press, London, 1966), 3rd ed., Sec. 20, case (b).

PHONON SPECTRUM OF SUPERCONDUCTING AMORPHOUS BISMUTH AND GALLIUM BY ELECTRON TUNNELING*

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The phonon spectra for amorphous Bi and Ga are derived by using McMillan's inversion program on our tunneling data. The results indicate an extreme "softening of the phonon spectrum" from what is found for crystalline superconductors. Parameters derived from our phonon spectra are used to test recent theories of the transition temperature of amorphous and strong-coupling superconductors, and also to test the McMillan equation for the maximum possible transition temperature in the limit of extremely strong coupling.

In this paper we present results on the phonon spectrum of superconducting amorphous Bi and Ga derived from tunneling measurements. The results are in the form of plots of $\alpha^2(\omega)F(\omega)$, where $\alpha^2(\omega)$ is the energy-dependent electron-phonon coupling parameter and $F(\omega)$ is the phonon density of states, with associated values of μ^*, the Coulomb pseudopotential. These results were obtained by inverting the strong-coupling gap equation, using the computer program of McMillan[1] which employs the experimental tunneling density of states $N(\omega)$ and the superconducting energy-gap parameter Δ_0 as input data. The resulting $\alpha^2(\omega)F(\omega)$ gives some insight into why the amorphous phases of Bi and Ga are such strong-coupling superconductors.

The tunnel junctions used in this investigation were Al-Al_xO_y-Bi and Al-Al_xO_y-Ga, and were prepared in situ in an evaporator cryostat. The Bi and Ga films were 1000 Å thick and were condensed onto a substrate held at 1.5°K. A brief description of the preparation of the tunnel junctions and of the method of measuring transition temperatures and of obtaining Δ_0 and $N(\omega)$ from the tunneling measurements have been given in an earlier publication.[2] A more detailed description of the experimental method will be included in a later article.

The values of the transition temperature T_C, twice the superconducting energy-gap parameter at zero temperature $2\Delta_0$, and the ratio $2\Delta_0/k_BT_C$ obtained for amorphous Bi and Ga in the present investigation are given in Table I, along with other values of these quantities obtained by some previous investigators. We have found that to obtain reproducible values of T_C and Δ_0 for amorphous Bi and Ga the substrate temperature during the condensation of the film has to be kept at a very low value (e.g., 1.5°K in the present investigation) compared with the temperature at which the amorphous phase converts to the crystalline phase (approximately 20°K for Bi and 15°K