tion for all the $\langle j_0 \rangle$ functions given by Blume, Freeman, and Watson.[4] In each case we can accurately predict where the departure from linearity begins to be comparable to our experimental error. Applying the same analysis to the experimental data on Gd, we find that the linear region of Fig. 1 should extend to $\sin\theta/\lambda = 0.143$, which is inside the first Bragg reflection. We conclude that our local form factor has behavior consistent with that of calculated $4f$ form factors in the questionable region.

Theoretical calculations are not sufficiently advanced to judge whether our diffuse component behaves exactly as conduction electrons should. The form factor shown in the Fig. 1 insert is not similar to that expected for $6s$ or $5d$ electrons in atomic Gd, nor does it correspond to the spin polarization produced in free electrons by the Ruderman-Kittel-Kasuya-Yosida interaction.

Note that for three free electrons in Gd, the $\sin\theta/\lambda$ value corresponding to twice the Fermi wave vector ($2k_F$) is 0.222 $Å^{-1}$. However, our diffuse component is certainly long range and oscillatory, which are properties universally attributed to conduction electrons in the rare-earth metals.

*Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

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Intense Tunable Phonon Fluorescence in Superconductors

V. Narayanamurti and R. C. Dynes
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 21 June 1971

The "phonon fluorescence" spectrum of superconducting Sn and $Pb_{0.5}Tl_{0.5}$ films pumped by a heat pulse is studied through the observation of resonance absorption by Sb donor levels in uniaxially compressed Ge. The spectrum consists primarily of a narrow band of phonons centered about the energy gap 2Δ (1.2 meV for Sn, 1.7 meV for $Pb_{0.5}Tl_{0.5}$). The power generated in this narrow band is at least hundreds of milliwatts. In the case of Sn the phonon energy has been tuned from 1.2 meV down to 0.7 meV by a magnetic field parallel to the plane of the film.

It was first pointed out by Eisenmenger and Dayem[1] that superconducting tunnel junctions could be used for the quantum generation of incoherent phonons with frequencies up to about 10^{12} Hz. In a recent Letter[2] (hereafter referred to as I) we measured the frequency spectrum of phonons emitted by a tunnel junction into a solid and showed that such junctions could be used to generate a much higher power level of effectively monochromatic phonons (of energy 2Δ, the superconducting energy gap) than previously envisioned. This realization made the device of potential use for the study of high-frequency monochromatic phonon transport and interactions in materials. In this Letter we report a new, more intense, simpler method for the generation of these phonons and describe the successful attempt to tune the emitted phonon frequency over a wide range. This tuning of both generator and detector is achieved by the application of a parallel magnetic field ($\lesssim 1$ kG), which adjusts the superconducting energy gap 2Δ controlling the frequency of emitted phonons.

From I it was clear that to generate phonons whose spectrum shows a peak at 2Δ it was not necessary to employ a tunnel junction. It was shown that phonons (generated by the relaxation of high-energy injected quasiparticles) whose energies $\hbar\omega > 2\Delta$ are extremely short lived. They rapidly break Cooper pairs, thus creating quasiparticle excitations above the energy gap. These quasiparticles then relax to the gap edge by emitting phonons and the process repeats until the generated phonons have energies $< 2\Delta$ at which time they become discontinuously long lived. The excitations at the gap edge subsequently recombine, forming Cooper pairs, and emit a phonon of energy 2Δ in the process. (Single-phonon emission is believed to be the dominant mechanism for pair recombination.) By invoking this degeneration of high-frequency excitations, the invariance, as a function of bias, of the measured large peak in the emitted phonon spectrum at 2Δ was explained. The phonons of energy $<2\Delta$, which were also generated, were not detected on account of the selectivity of the detecting tunnel junction.

Assuming the validity of this model, a high-energy phonon ($\hbar\omega > 2\Delta$), incident on a superconductor, will have a high scattering rate for pair breaking and the net result is a *down conversion* of the frequency through this "fluorescence" model. The incident single phonon creates at least two phonons, one necessarily of energy 2Δ. Hence a beam of high-energy phonons will be down converted, with at least an equal number being generated at 2Δ and an additional lower-energy contribution due to the relaxation process.

It has been shown previously that the frequency spectrum of phonons emitted into a solid[3] from a thin-film Constantan heater can best be represented by a blackbody distribution[4] with a "characteristic temperature" T_H above ambient, determined by the power dissipated in the voltage pulse. Incident on a superconductor, all those phonons with $\hbar\omega > 2\Delta$ have a short lifetime and will experience the degeneration process described above, the resultant emitted spectrum being represented schematically in the upper right-hand corner of Fig. 1. The phonons represented by the cross-hatched region are converted into the dotted area. It should be emphasized that on account of the inherent nonlinearity in the process the dotted area will be greater than the original area above 2Δ. We expect, as illustrated, a large peak in the resultant spectrum at 2Δ.

To test this hypothesis, we use the experimental arrangement illustrated in Fig. 1, which is similar to the system reported previously except for the fact that the generator consists of a conventional thin-film Constantan heater ($R \approx 50\ \Omega$) screened by a superconducting film. A layer of silicon oxide ($d \approx 3000$ Å) was used to insulate the heater from the superconductor electrically. To determine the emitted spectrum, we employ as a spectrometer a Sn-I-Sn tunnel junction and the large variation in the valley-orbit splitting of Sb in Ge with applied [111] uniaxial stress. This stress dependence, shown in Fig. 1, has been discussed in detail in I. For phonons propagating

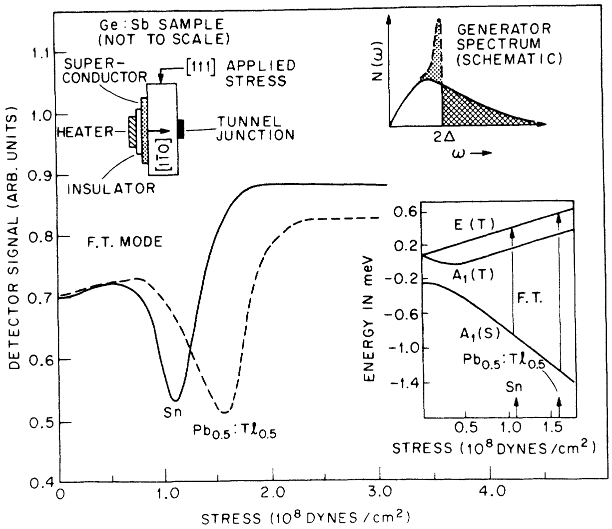

FIG. 1. Tracing of peak intensity of FT mode, propagating along [1$\bar{1}$0] as a function of [111] compressive stress for Sn and $Pb_{0.5}Tl_{0.5}$ generators pumped by a heat pulse (pulse power, 2 W; generator area, 9 mm^2; pulse width, 5×10^{-8} sec). Insert shows the Sb-donor level splittings as a function of stress. Arrows at the baseline reflect the positions of the measured absorption peaks. The FT arrows indicate theoretically expected positions for the two generators.

in the [1$\bar{1}$0] direction, symmetry allows coupling of the fast transverse (FT) phonon mode from the ground state $A_1(S)$ to the excited $E(T)$ state. The resonance absorption of phonons by the Sb donors "burns a hole" in the emitted spectrum at the stress splitting energy thus affecting the detector signal for phonon energies $\geqslant 2\Delta_{\text{detector}}$. As before, we emphasize that great care must be taken to apply uniform stress as spurious structure is produced otherwise.

In the experiment a Ge crystal of dimensions $0.43\times0.36\times1.3$ cm^3 containing 1.8×10^{15} Sb impurities was used. The superconducting films were $\approx 600-700$ Å thick in both the generator and detector in order to effect the tunability which will be discussed below. Typical voltage pulses to the heater (3×3 mm^2) were of ~10 V amplitude and ~0.05 μsec width and the detector (size 1×1.5 mm^2) was biased in the usual manner. The sample was in a He^4 bath at 1.3°K and the heat pulses had a "temperature" characteristically $\sim5\pm1$°K. Care should be taken when quoting this number as it is used here simply to characterize the heat-pulse phonon frequency distribution; it does not represent a thermodynamic equilibrium. Characteristic ballistic longitudinal- and transverse-phonon pulses, of quality similar to or better than those reported in I, were observed at the Sn tunnel detector. In this paper, we present only data on the FT mode since it was the strongest in intensity.

In the solid line of Fig. 1 labeled Sn the superconducting film used as a down converter was tin. We clearly see an absorption line at a stress corresponding to a level splitting of 2Δ for Sn. As the detector is a Sn tunnel junction, the system is effectively monochromatic (phonons $\hbar\omega < 2\Delta$ are not detected), the results indicating a substantial peak in emitted density at 2Δ. The line shape is very similar to that published previously using tunnel junctions, the linewidth being somewhat narrower. This we attribute to improved techniques in applying uniform stress and should not be considered a linewidth measurement. As before, we attribute the background shape to off-resonance scattering. No such peak is observed if a heat pulse (without superconducting converter) is incident directly onto the detector. The fact that we do observe this strong resonance suggests that the mean free path of the phonons for pair breaking is $\lesssim 600-700$ Å. Our free-electron estimates indicate that this is a reasonable order of magnitude value for phonons of energy $> 2\Delta$. A precise theoretical determination of such a number is extremely sensitive to allowed umklapp processes and hence to the fine details of the Fermi surface near the zone boundaries. This sensitivity makes such an estimate very difficult.

In order to determine, at least qualitatively, the relative strength of the emitted spectrum at energies $< 2\Delta$, the experiment was repeated using a film of the alloy[5,6] $Pb_{0.5}Tl_{0.5}$ as generator and a Sn-oxide-Sn tunnel junction as a detector. The gap $2\Delta_{alloy}$ measured independently by tunneling was equal to 1.7 meV. Employing a Sn tunnel junction as a detector we were sensitive to phonons in the range $2\Delta_{Sn} < \hbar\omega < 2\Delta_{alloy}$. By repeating the experiments as a function of stress it was possible to determine the density of phonons between 1.2 and 1.7 meV. The measured detector signal as a function of stress for the FT mode is shown as the dashed line of Fig. 1, where again we see a resonance absorption corresponding to a stress splitting of $2\Delta_{alloy}$ (see inset Fig. 1). There is very little qualitative difference between the solid and dashed line shapes, suggesting that the peak in the spectrum at 2Δ is substantial relative to the lower-energy modes. There is, however, an asymmetry to the dashed line, indicating a contribution to the total signal from the 1.2- to 1.7-meV range of the spectrum. This is not

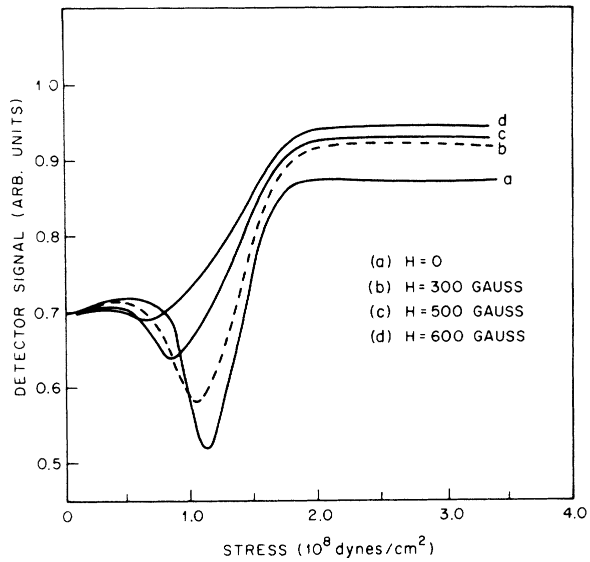

FIG. 2. Intensity of the FT mode for four different values of magnetic field applied parallel to the plane of the Sn generator film.

surprising in view of the fact that the fluorescence model predicts that $\frac{1}{2}$ to $\frac{2}{3}$ of the phonons generated in the decay will have a continuous distribution of energies from 0 to 2Δ and the remainder will be centered in a narrow band about 2Δ. The precise intensity ratio is determined by the width of this band. Our results clearly show that the spectrum is certainly peaked at 2Δ.

We have also displayed the tunability of such a device over a substantial range. In Fig. 2 we show a series of absorption curves, as a function of stress, for various parallel magnetic fields. The low fields used here do not affect the Sb levels but cause significant variation of the gap parameter of the superconductor. The Ginsburg-Landau theory[7] shows that when the film thickness $d \lesssim \lambda$, the London penetration depth, the gap parameter goes continuously to zero as a parallel magnetic field is increased, giving a second-order phase transition at H_c, the critical field. In the limit of very thin films the gap parameter is constant across the film and varies with field as

$$[\Delta(H)/\Delta(0)]^2 = 1 - (H/H_c)^2. \quad (1)$$

As $d \sim \lambda$ in our case we expect behavior of this form for Δ and hence for the energy of the phonon beam. Figure 2 clearly shows a monotonic behavior of the absorption peak position as a function of field. In Fig. 3 we plot the square of the peak position as a function of the square of magnetic field and indeed, for the field range studied, the points do fit on a straight line as expected

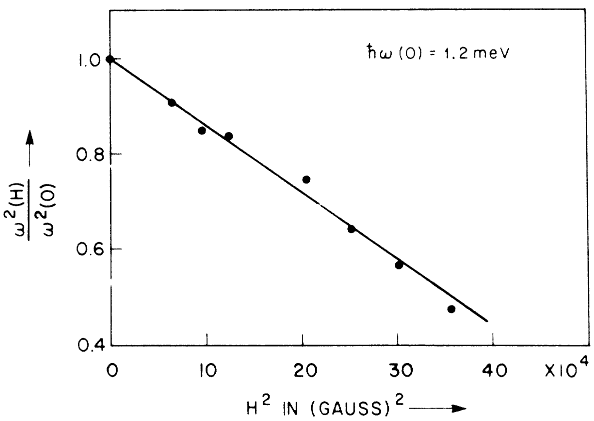

FIG. 3. Plot of the square of resonance frequency $\omega(H)$ as a function of square of magnetic field H. Sn generator.

from (1). This confirms that we are in fact tuning the phonon beam energy by tuning the gap parameter in the superconducting film.

A more careful consideration of this problem of magnetic field dependence of the gap parameter in thin films leads to more profound changes than are considered above. It is well-known[8] that an applied H field, in this limit of $d \sim \lambda$, not only affects the energy gap $\Delta(H)$ but also alters the shape of the density of excitations, the familiar BCS singularity disappearing for finite fields and the gap edge becoming less sharp. $\Delta(H)$ is nevertheless a monotonic function, decreasing continuously to zero, and we are certainly seeing this effect in Figs. 2 and 3. It is possible that the decrease in absorption peak strength with increasing field is due to this change of the density of excitations in the superconductor, broadening of the spectrum of emitted phonons being the result of this distortion. If this is the case, the tunability of such a device is limited simply by magnetic field broadening to a range $\sim\Delta < \hbar\omega < 2\Delta$. On the other hand, part of the decrease in signal with lower ω is almost certainly due to the expected strong frequency dependence of the elastic resonance-fluorescence scattering strength[9] of the Sb levels. Thus, the tunability of the phonon source may be greater than that indicated above.

In conclusion we have, in analogy to an optical fluorescence experiment, succeeded in pumping a superconductor with a blackbody source of phonons and have observed the system emit a discrete spectrum with a large peak at 2Δ, the superconducting energy gap. It appears that one can generate phonon powers of the order of watts by this technique. In addition, we have shown that one can tune the emitted phonon frequency over a substantial range by the application of a parallel magnetic field. This tunability and the high power level of our generator makes it of practical use for phonon spectroscopy in a previously inaccessible frequency region and it has been used by us to observe the ground-state splitting of V^{3+} in Al_2O_3. The details of these latter measurements, together with the pulse-power, pulse-width, and ambient-temperature dependence of the phonon fluorescence effect will be published in a more detailed publication.

It is a pleasure to acknowledge the very skillful technical assistance of M. A. Chin and W. F. Flood in various stages of this work. Also, for continued interest and discussions we thank S. Geschwind and J. M. Rowell.

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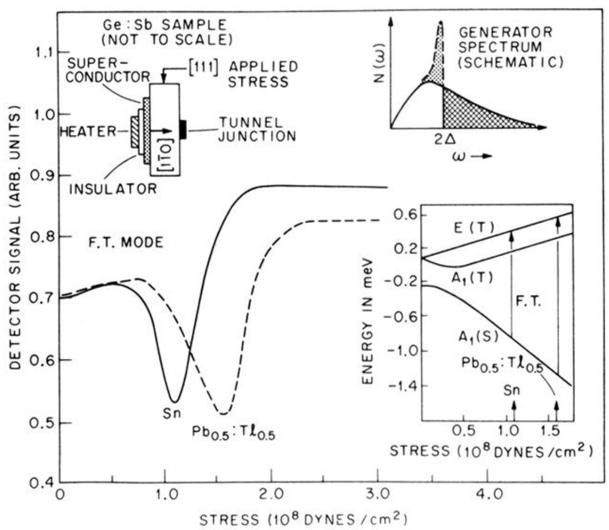

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