Monochromatic Phonon Propagation in Ge:Sb Using Superconducting Tunnel Junctions

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The frequency spectrum of the phonons generated by a superconducting SnO-Sn tunnel junction has been determined through the observation of resonance absorption by Sb donor levels in Ge. These measurements show that one can use such junctions to generate and detect milliwatts of phonon power at energies of 2Δ (1.2 meV).

The use of superconducting tunnel junctions to generate and detect phonons[1] is of considerable potential interest for the study of thermal conductivity, phonon propagation, and electron-phonon interactions in solids and liquids. The question of the frequency spectrum of phonons emitted from a junction into a solid has not yet been answered and consequently the use of such a device as a spectroscopic tool has been limited. In this Letter we report the results of an experiment which shows that, over a wide range of bias conditions, the distribution has a large density of phonons at an energy $\leq 2\Delta$, the superconducting energy gap, and very few phonons of higher energy.

To demonstrate this we use as a phonon spectrometer the properties of the shallow n-type donor Sb in Ge. The hydrogenic-like ground state in the effective-mass approximation[2] is fourfold degenerate because of the four equivalent conduction-band minima. If valley-orbit coupling is considered,[3] this degeneracy is partially lifted into a singlet ground state $A_1(S)$ and an excited triplet state which are separated[4] by the "chemical shift" of 0.32 meV for Sb in Ge. In addition, when the crystal is subjected to uniaxial stress, the site symmetry becomes lower than cubic and the degeneracy of the triplet is generally destroyed. By deformation-potential theory, the shifts of the various valleys can be determined with good accuracy.[3] Assuming the deformation potential $E_u = 16$ eV, the calculated variation of the levels with compressive (111) stress is shown in the insert of Fig. 1. Here the triplet splits into a singlet $A_1(T)$ and a doublet $E(T)$ and all levels rapidly approach their asymptotic limits for modest stress. In fact, it is easy to "tune" the level splitting through energies equivalent to 2Δ of Sn (1.2 meV) or Pb (2.8 meV), and up to several meV.

It is this tunable level splitting that is used as the phonon spectrometer in this work. If allowed by symmetry, phonons propagating through the crystal with energies equal to this splitting will be more readily absorbed than others. By measuring the absorption as a function of stress, the spectrum of generated phonons can be determined.

In the experiment we used a Bell Telephone Laboratories zone-leveled Ge crystal[5] in the form of a rectangular bar $1.2\times0.42\times0.377$ cm^3, containing 1.8×10^{15} cm^{-3} Sb impurities and a surface dislocation density $<10^3/\text{cm}^2$. The stress was applied along the long axis of the bar which corresponded to the [111] direction. The phonons were propagated along the short axis of the bar which coincided with the $[1\bar{1}0]$ direction. Thin-film ($\approx$5000 Å thick) Sn-oxide–Sn tunnel

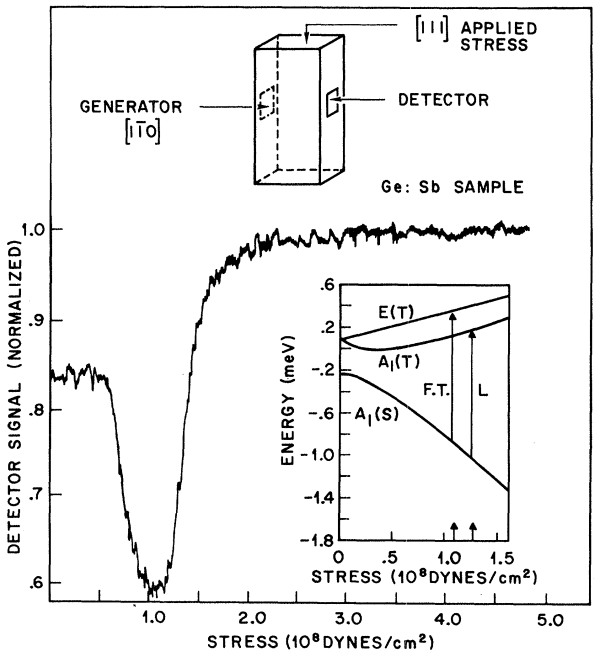

FIG. 1. Tracing of detector signal as a function of stress in the dc experiment. Phonons are propagating along $[1\bar{1}0]$ direction. Insert shows the Sb-donor level splittings as a function of [111] compressive stress. Arrows at the baseline reflect the positions of the measured absorption peaks. The FT and L arrows indicate theoretically expected positions. See text.

junctions (area ~1 mm^2) were used to generate and detect the phonons. The junctions typically showed resistance ratios between the superconducting and normal states at voltages $<2\Delta$ of ~30 and a normal-state resistance ≈ 10 mΩ. Great care must be taken to apply uniform stress over the whole crystal as line broadening and false structure result otherwise. One of the junctions (the generator) is dc biased (I_{dc}) with a small ac modulation (I_{ac}) superposed, while the other junction (detector) is biased below 2Δ in the usual way and the voltage signal is phase-sensitively detected. The dectector signal as a function of generator current showed characteristics of similar or better quality than previously published data.[1] The stress was monitored by means of a calibrated Kistler charge transducer and an electrometer. The output of the electrometer was used to drive the X axis of an X-Y recorder. The experiments were performed at 1.3°K with a magnetic field ≈ 30 G applied parallel to the [111] axis to suppress undesirable Josephson effects.

In Fig. 1 we plot detector voltage signal as a function of stress. I_{dc} was approximately one half the current rise at 2Δ (80 mA) while I_{ac} was ~10 mA peak to peak. In this figure we see a well-defined resonance line at 1.09×10^8 dyn/cm^2; from the insert this value corresponds to absorption of energy 2Δ from $A_1(S)\rightarrow E(T)$. According to the selection rules,[6] in the $[1\bar{1}0]$ direction this is only allowed for the fast transverse (FT) mode. There is no obvious peak at 1.25×10^8 dyn/cm^2 corresponding to the expected longitudinal mode (L) absorption. It will be shown later that this is simply due to the fact that the large majority of the phonons propagating in $[1\bar{1}0]$ are FT in nature. Selection rules forbid coupling of the slow transverse (ST) mode in the $[1\bar{1}0]$ direction. In addition to the absorption peak, there is a baseline shift at the resonance. This shift we interpret in terms of inelastic scattering and will discuss later. It should also be pointed out here that these results say nothing about the emitted spectrum for energies $<2\Delta$ as we are employing a detector sensitive only to energies $\geq 2\Delta$. The line shape of Fig. 1 is somewhat broadened from that expected on theoretical grounds. The intrinsic phonon shower width can be due to any superconducting energy-gap anisotropy (~5-10%) or pair recombination lifetime broadening[7] (expected to be small). The spectrometer resolution is limited by level broadening due to lifetime effects[8] (~10%), inhomogeneous stress (~10%), and impurity wave-function overlap. Further experimentation is required to determine accurately the intrinsic width of the 2Δ phonons.

By increasing I_{dc} above that equivalent to $V=4\Delta$ we are now in a position to determine the spectrum of emitted phonons due to the injection of hot electrons into the superconductor. It has been suggested[9,10] that this spectrum would reflect the BCS distribution of injected quasiparticles and/or the phonon distribution $F(\omega)$ in Sn. In our plots of absorption versus stress, both of these effects would show up as structure which would depend on I_{dc}. However, for dc biases up to $V=10\Delta$ we see no measurable line-shape change and no additional structure at higher stress which would correspond to phonons $>2\Delta$ generated by quasiparticle relaxation to the top of the energy gap. The absorption strength remains constant with the same baseline shift and no broadening. This result suggests that the phonon spectrum still consists only of phonons of energy 2Δ or less. This is to be contrasted with a heat-pulse measurement where we have observed a broad band of phonons with a frequency maximum which shifts with power.

In order to establish conclusively the phonon nature of the signals and the mode coupling to the impurity levels we performed a time-of-flight experiment. In this experiment ~0.3-μsec wide current pulses from a Hewlett-Packard 214A pulse generator were applied to the generating diode. A rise-time decreaser and current transformer were used to minimize ringing in the circuitry. The detector was dc biased and the voltage pulses from the free terminals of the junction were amplified and fed into a boxcar integrator. The dectector signal as a function of time for a generator current of 1.5 A and for three different stresses is shown in Fig. 2. Three distinct pulses are observed whose arrival times correspond to sound velocities in good agreement (within 5%) with those expected from ultrasonic work for rectilinear propagation[11] of the three different modes. The relative intensities of the different modes, far away from resonance, are in agreement with those expected from phonon focusing effects[12] due to the elastic anisotropy of Ge. Variations in the mode intensities due to differences in the electron-phonon coupling in Sn are expected to be small since U processes dominate in the recombination. It is clear from Fig. 2 that the majority of the signal in the dc experiment must have come from the FT mode, and consequently no doublet was observed. In addition these results show that, as

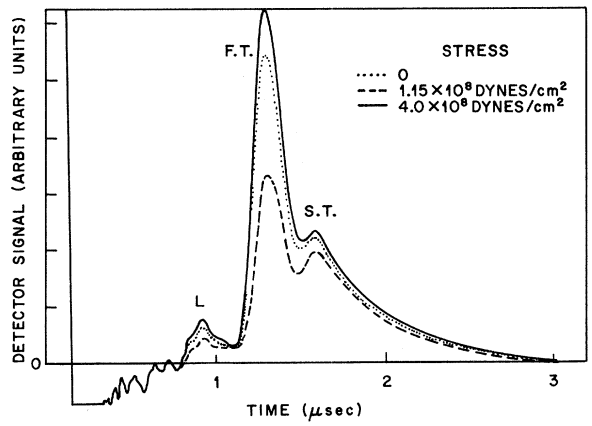

FIG. 2. Tracing of boxcar output as a function of time for three values of stress. Pulsed experiment. Some ringing in the circuit before the L mode is evident.

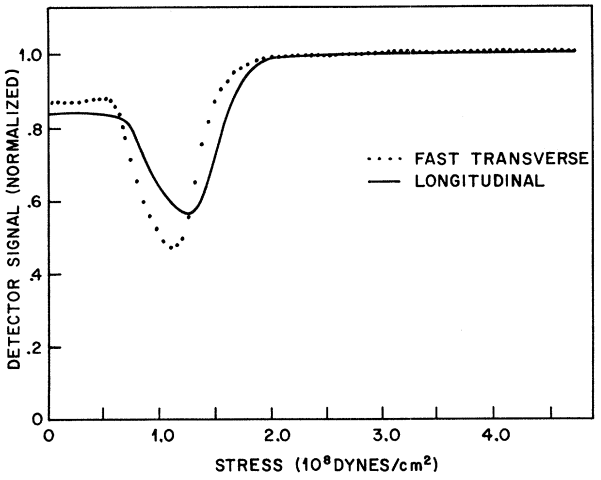

FIG. 3. Tracing of peak intensities of longitudinal and fast-transverse modes at a function of stress.

expected from matrix-element considerations,[6] the ST mode propagating along $[1\bar{1}0]$ does not couple to the Sb levels. The small differences in the intensity of the ST mode observed in Fig. 2 are believed to be due to the changes with stress of the overlapping tail of the much stronger FT mode.

To resolve the coupling explicitly, we sit on the peak of the L and FT modes and apply stress. The results of these two measurements are shown in Fig. 3, where clearly we see absorption peaks occurring at different values of stress. The peak positions are in excellent agreement with that expected from the insert of Fig. 1. The agreement with calculations is within 3% using $E_u = 16$ eV, a value close to that obtained from infrared measurements.[4] The line shapes for these absorptions are qualitatively similar to those in the dc experiment although the absorption is stronger as the dc experiment is an integral over all three modes. As before, decreasing the power by a factor $\simeq 2500$ resulted in no visible effect on the line shape, suggesting that the spectrum of phonons emitted for any generator power consists of phonons of energy $\leq 2\Delta$ with only the 2Δ ones being detected. This invariance of the spectrum with power indicates that the higher-energy excitations (phonons and/or injected electrons) rapidly degenerate[13] into superconducting paired electrons and multiple phonons of energy $\leq 2\Delta$. The most probable degeneration process is that of high-energy phonons created by the initial decay of the injected particle rapidly creating excitations at or above the gap edge by pair breaking. These excitations then recombine into the superconducting condensate creating the 2Δ phonons observed. Only when the phonons are of energy 2Δ or less are they sufficiently long lived to escape into the Ge.

At sufficiently low temperatures it should be possible, in parallel magnetic fields, to vary 2Δ over a large range without significant loss in sensitivity. This will provide a tunable phonon source in the far-infrared frequency region.

The scattering of phonons by shallow impurity donors in Ge has been investigated theoretically[8,14] to describe thermal conductivity and acoustic attenuation results. The three separate types of interactions considered are (1) elastic scattering, (2) inelastic scattering, and (3) phonon-assisted absorption. In (1) the interaction can be considered as a resonant absorption process causing virtual excitations from the ground state, elastically scattering the resonant-energy phonons out of the beam. In (2) the interaction can be written $[\text{phonon} + A_1(S)] \rightleftarrows [\text{phonon}' + A_1(T)$ or $E(T)]$. At 0°K this interaction will only go from left to right and from conservation of energy considerations will cut off at a stress equivalent to a splitting of 2Δ. In (3) we consider $[\text{phonon} + \text{phonon}' + A_1(S)] \rightleftarrows A_1(T)$ or $E(T)$. This will not show the same cutoff at 2Δ that the inelastic process will show. In view of these considerations we feel that we are observing the effects of interaction (1), the resonance line, and (2), the background shift at 2Δ.

In conclusion, by using the unique strain-dependent properties of Sb in Ge, we have determined the spectrum of phonons emitted from a superconducting tunnel junction into a solid. It appears that as a result of rapid relaxation and multiple scattering the high-energy excitations

degenerate quickly into phonons $\hbar\omega \leq 2\Delta$ and only then are they long enough lived to be emitted into the Ge. This indicates that we have succeeded in generating milliwatts of phonon power over a very restricted spectrum and have detected only a small portion (the upper cutoff region) of the distribution which makes the system effectively a monochromatic source of phonons. We have observed scattering of these modes by the hydrogenic-like levels of Sb in Ge and the coupling obeys the symmetry restrictions. We believe that we have seen two types of scattering: (1) a resonance absorption indicated by the resonance peak, and (2) inelastic scattering manifested by a step in the background of the absorption curve, both structures occurring at a stress equivalent to a splitting of 2Δ.

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Calculation of Energy-Band Pressure Coefficients from the Dielectric Theory of the Chemical Bond*

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The dielectric theory of the chemical bond has been extended to calculate pressure coefficients of interband energy differences in acceptable agreement with experiment. Some improvements in Van Vechten's prescriptions are suggested by these new, more stringent tests. An approximate empirical rule for pressure coefficients is confirmed, and deviations from it predicted.

Van Vechten has recently proposed[1] a general model for tetrahedrally coordinated A^NB^{8-N} compounds which is based on Phillips's[2] spectroscopic theory of electronegativity differences and appears to yield good agreement between predicted energy-band separations and experimental results. In this Letter we extend this model to calculate the expected pressure dependences of three direct and two indirect energy gaps in nineteen group IV, group III-V, and group II-VI semiconductors. With a few exceptions which will be discussed in detail, we have adopted Van Vechten's prescriptions for these energy gaps and have avoided adjusting his numerical values for parameters, so that our comparison of calculated with measured pressure coefficients may provide a new test for his model.

A brief summary of Van Vechten's prescriptions follows:

(1) A direct interband energy separation E_i in a compound neither of whose elements possesses core states (in practice, d states) close in energy to the valence-band energies is given by

$$E_i = E_{h,i}[1 + (C/E_{h,i})^2]^{1/2}, \quad (1)$$

where $E_{h,i}$, the homopolar gap, is assumed to be a function of nearest-neighbor distance d only and to be given by[3]

$$E_{h,i} \propto d^{s_i}. \quad (2)$$