

Normal-state tunnel junction as a tunable quasimonochromatic phonon source

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The frequency distribution of the acoustic phonons emitted from a biased metal-insulator-metal normal-state tunnel junction has been studied. Electrons tunneling across the insulating layer form an excited population with a range of energies up to $\sim eV_B$ above the Fermi level and then relax, producing a phonon spectrum with a frequency cutoff at the same characteristic value. Normal-state tunnel junctions made from aluminum have been produced and phonon spectra determined by using the stress-tuned splitting of boron acceptors in silicon as a spectrometer. It has been shown that, in the range $\sim 1\text{--}4$ meV ($\sim 250\text{--}1000$ GHz), the second differential of the signal with respect to junction bias ($\partial^2 P / \partial V_B^2$) is quasimonochromatic, having a spectrum of an approximately Gaussian form with the peak occurring at an energy of eV_B and a half width of $\sim 6kT_e$.

I. INTRODUCTION

Since the superconducting tunnel junction (SCTJ) was first demonstrated as a continuously tunable source of monochromatic phonons,¹ it has been used extensively in the phonon spectroscopy of a wide variety of systems. It has many advantages over other techniques, such as a large phonon frequency range (~ 0.3 meV–15 meV),² narrow bandwidth (~ 10 μ eV), and relative ease of production. However, its one great disadvantage is that it cannot be used for measurements in a magnetic field above the critical field (B_C) of the superconductor, thus excluding many potentially interesting experiments. At present, the only method of phonon spectroscopy in a magnetic field is by the use of the temperature dependence of the spectra of heat pulses,³ which has the problems of not being a monochromatic source and only offering a limited degree of “tunability.”

We demonstrate here a phonon source arising from the tunneling between the nonsuperconducting (normal state) metal films of a tunnel junction. This process leads to an excited population of electrons with energies up to eV_B , where V_B is the voltage bias applied across the junction. These electrons may then relax, giving rise to a phonon spectrum with a high energy tail reaching approximately eV_B , the effect of which may be accentuated by modulating the bias voltage and measuring the first and second differentials of the signal with respect to the modulation, in order to give an increasingly monochromaticlike distribution.

This normal-state tunnel junction (NSTJ), despite being similar in principle to the SCTJ and also having been previously suggested as a possible phonon source,⁴ has not before been either studied or used in any measurements. This is because there are serious practical problems, mostly due to the nonsuperconducting nature of the materials, in particular, the effects of thermal emission

from the metal films, the problems of precise determination of the voltage drop across the film, and the possibilities of alternative channels of energy relaxation of the excited electrons, for example, via electron-electron scattering.

In the next section, a simple model of the NSTJ is described and an expression is determined for the phonon spectra. In Sec. III the techniques used to produce and measure the spectra from the NSTJ's are detailed and it is also discussed how the problems associated with the finite film resistance were overcome. In Sec. IV the spectra obtained from first and second differential measurements are presented and compared with the model outlined in Sec. II.

II. THEORY

A relatively simple model based upon the rate equation for the phonon occupation numbers can be used. As shown in Fig. 1, when the potential of one metal is raised relative to the others, electrons from the side at higher bias may tunnel through the barrier into the empty states, thus producing an excited population in both films. There are two possibilities for this excited population to redistribute its energy: by the emission of high energy phonons or via the electron-electron interaction, which will lead merely to a raised average electron temperature in the metal producing a thermal phonon spectrum. Thus if the electron-electron interaction dominates, no high energy phonon emission will occur. However, the results of Sec. IV will demonstrate that this is not the case and so this effect is neglected in this analysis.

The total power emitted by the tunnel junction is given by

$$P^{\text{tot}} = \int_0^\infty d\omega \hbar\omega \dot{N}(\omega, eV_B), \quad (1)$$

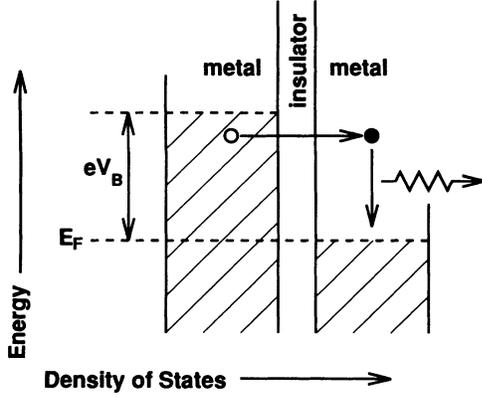


FIG. 1. Energy diagram of a NSTJ. Application of bias raises the electron energies in the metal on the left-hand side, allowing electrons to tunnel across into the empty states on the right-hand side. The excited populations in both films then relax and emit phonons in the process.

where ω is the frequency of the phonons and $\dot{N}(\omega, eV_B)$ their spectral density. This power is equal to the rate of phonon emission due to electron-phonon interaction in the tunnel junction:

$$\int_0^\infty d\omega \hbar\omega \dot{N}(\omega, eV_B) = \left. \frac{\partial E}{\partial t} \right|_{ep}. \quad (2)$$

Starting from the standard Bloch-Boltzmann-Peierls equation for the energy emission rate due to electron-phonon interaction,⁵ we can derive an analytic expression for $\dot{N}(\omega, eV_B)$,

$$\left. \frac{\partial E}{\partial t} \right|_{ep} = 4\pi \sum_{kk'} |M_{kk'}|^2 \hbar\omega_q \delta(\epsilon_k - \epsilon_{k'} - \hbar\omega_q) \times \{(1 + N_q)f_k(1 - f_{k'})\}. \quad (3)$$

Here $M_{kk'}$ is the electron-phonon interaction matrix element ($k' = k \pm q$) and f_k and $f_{k'}$ are the distribution functions for the initial and final electron states during electron-phonon scattering. In the case of our tunnel junction, f_k represents the nonequilibrium distribution function of the electrons after tunneling, which therefore will be denoted by f_k^T in the following, whereas $f_{k'}$ represents the Fermi distribution in the part of the tunnel junction into which the electrons tunnel. Introducing the Eliashberg function $\alpha^2(\omega)F(\omega)$ and the electronic density of states at the Fermi energy $N(E_F)$ (Ref. 6) into Eq. (3), after some algebra we arrive at

$$\left. \frac{\partial E}{\partial t} \right|_{ep} = 2\pi \hbar N(E_F) \int_0^\infty d\omega \hbar\omega \alpha^2(\omega)F(\omega) \times [1 + N(\omega, eV_B)] \int_0^\infty d\epsilon f_{\epsilon+\hbar\omega eV_B}^T (1 - f_\epsilon). \quad (4)$$

Comparing (4) with (2) we get a general expression for the number of emitted phonons per frequency ω of a normal conduction tunnel junction:

$$\dot{N}(\omega, eV_B) = 2\pi \hbar N(E_F) \alpha^2(\omega)F(\omega)[1 + N(\omega, eV_B)] \times \int_0^\infty d\epsilon f_{\epsilon+\hbar\omega eV_B}^T (1 - f_\epsilon). \quad (5)$$

For the determination of $\dot{N}(\omega, eV_B)$, in principle the nonequilibrium distribution function of the tunneled electrons $f_{\epsilon+\hbar\omega eV_B}^T$ has to be known, which in the general case is a difficult many-body problem.^{7,8} For our experimental situation, however, where $eV_B \ll E_F$, the tunneling probability can be assumed to be independent of energy and the following nonequilibrium tunnel distribution is considered to be a good approximation:⁹

$$f_{\epsilon+\hbar\omega eV_B}^T = T \left\{ \left[\exp\left(\frac{\epsilon + \hbar\omega - eV_B - \epsilon_F}{kT_e}\right) + 1 \right]^{-1} - \left[\exp\left(\frac{\epsilon - \epsilon_F}{kT_e}\right) + 1 \right]^{-1} \right\}, \quad (6)$$

where T is the tunneling probability. Furthermore it is appropriate in our case to take the low frequency limit of the Eliashberg function, which is¹⁰

$$\alpha^2(\omega)F(\omega) = b (\hbar\omega)^2. \quad (7)$$

Inserting (6) and (7) into (5) and solving the integral we arrive at the following analytic expression for $\dot{N}(\omega, eV_B)$:

$$\dot{N}(\omega, eV_B) = 2\pi \hbar N(E_F) T b [1 + N(\omega, eV_B)] (\hbar\omega)^2 \times \frac{(\hbar\omega - eV_B)}{e^{-\frac{\hbar\omega - eV_B}{kT_e}} - 1}. \quad (8)$$

The top panel of Fig. 2 shows the phonon spectra $\dot{N}(\omega, eV_B)$ calculated for three different bias voltages (1, 2, and 3 mV) and with $T_e = 0.5$ K. The spectra are broad and extend up to frequencies of $\sim eV_B/\hbar$. The middle and bottom panels show the first and second differentials $\partial\dot{N}(\omega, eV_B)/\partial V_B$ and $\partial^2\dot{N}(\omega, eV_B)/\partial V_B^2$, respectively. The first differentials are still continuous spectra with accentuated cutoffs, while the second differentials exhibit quasimonochromatic peaks centered at the frequencies near the respective corresponding bias voltages. The broadening of the peaks corresponds in all three cases to a full width at half maximum of $\sim 6kT_e$, when numerically determined from the second derivative of Eq. (8) with respect to V_B . It can be shown that the broadening of the quasimonochromatic lines scales linearly with temperature, so that the monochromaticity of the lines improves at lower temperatures. For T approaching 0 K the broadening is minimal and only determined by the uncertainty width of the tunneled quasiparticles interacting with the phonons.

In concluding this section we note that this theoretical treatment covers only the phonon emission by the tunneled electrons. Phonon absorption becomes important if $eV_B \sim kT_e$. These processes are, however, not relevant for phonon spectroscopy. Including these processes would also remedy the seeming violation of the energy conservation in Eqs. (5) and (8) at $V_B = 0$.

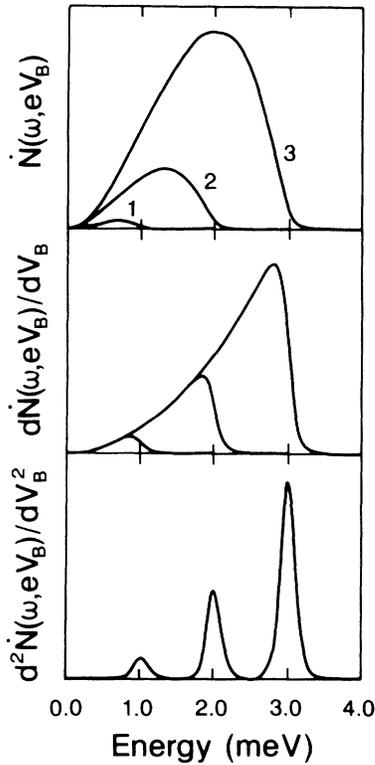


FIG. 2. Spectra of phonon emission from a NSTJ calculated using the analytical forms given in Eq. (8). The top graph shows the form of $\dot{N}(\omega, eV_B)$ calculated for three values of $V_B = 1, 2, 3$ mV. The electron temperature was set at 0.5 K. The center graph shows the first differential with respect to junction bias $d\dot{N}(\omega, eV_B)/dV_B$, for the same three values of V_B , showing that the curves are identical at low phonon energies. In the bottom graph, the three lines are the corresponding forms of $d^2\dot{N}(\omega, eV_B)/dV_B^2$, calculated again from Eq. (8). The width of the quasimonochromatic peaks is $\sim 6kT_e$.

III. EXPERIMENT

The techniques for producing SCTJ's are well established. They consist of depositing a thin film of a suitable superconducting material (e.g., aluminum, tin, lead, etc.), exposing the film to oxygen so as to coat it with a layer of insulating oxide, and then growing a second film. Masks are used during the film growth to produce a completed device which consists of two contact pads (one to each film) and the junction region where the films overlap each other but are separated by the oxide layer on the lower film. Because of the superconductivity of the films, the contact resistances and the voltage drop across the films are zero and the emitted phonons come entirely from electrons tunneling across the oxide and then relaxing. In the preparation of NSTJ's, it becomes important to consider the several effects the finite film resistance.

Due to Ohmic heating, there will be emission of thermal phonons from the metal films as well as from the junction. Because of the effects of phonon focusing (whereby a detector which is placed directly opposite the emitter will be strongly acoustically coupled by the fo-

cusing action of the crystal), only emission from the films in the area of the oxide need be considered. The resistance of this area is given as R_F , which for the junctions used will be approximately equal to the film resistance per unit area R_\square . The ratio R_F/R_T (where R_T is the total tunneling resistance of the junction) will give a measure of the relative magnitudes of the signal and background and will be considered further in the following sections.

The film resistance will also affect the determination of V_B since in order to measure the potential drop due solely to tunneling across the junction it is necessary to utilize a four point geometry, and this is shown in Fig. 3 (top). It may also be expected that there is some nonuniformity of the bias voltage V_B over the area of the junction; however, its influence will be negligible since the film resistances of the top and bottom are the same, leading to a virtually uniform tunneling current over the entire junction area.

In principle, any metal may be used for the films, but in these initial measurements aluminum was chosen since it can first be used as a superconducting tunnel junction to test the quality of the metal-insulator interface and then may be set into the normal state by raising the bath temperature slightly above T_c , which is ~ 1.3 K for the films used. The films were grown by evaporating Al from tantalum boats at pressures of $\sim 10^{-6}$ mbar and to a thickness of typically ~ 250 Å since it has been shown² that, in aluminum SCTJ's, thicker films cause high energy phonons to be reabsorbed before they are emitted into the substrate. The thickness of the insulating oxide

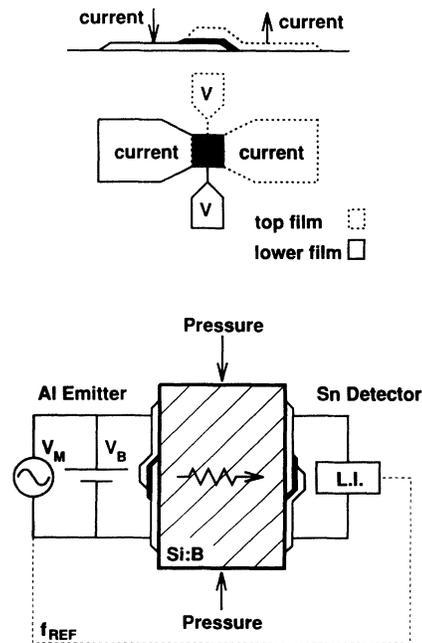


FIG. 3. Top: typical tunnel junction. The first film is deposited, partially oxidized, followed by the second film. Masks are used to produce distinct contact and overlap areas. Center: four-point geometry used in the NSTJ to minimize the effects of finite film resistance on the measured value of V_B . Bottom: schematic of the pressure experiment.

was varied by changing the time that the first film was exposed to air before being returned to vacuum for the growth of the second film.

The phonon spectrum was measured using the stress dependent splitting of impurity atoms in semiconductors (e.g., the boron acceptor in silicon). The stress-tuned splitting (which leads to the resonant scattering of phonons at the splitting frequency) acts as a band-stop filter and may be used in conjunction with a broadband detector in order to obtain spectroscopic information about the phonon source.¹¹

The experimental setup is shown in the bottom section of Fig. 3. The junction was grown on one face of a rectangular cross sectional area (3 mm × 4 mm × 15 mm) silicon crystal which was lightly doped with boron (10^{13} m^{-3}), chosen so as to give the optimum balance between resolution and absorption of the phonon scattering. A tin SCTJ was positioned directly opposite the aluminum junction to be used as a broadband detector (although in actual fact it detects only phonons with energies above $\sim 1.2 \text{ meV}$). Uniaxial stress was applied perpendicularly to the sample with a compressed air system and the pressure on the sample was monitored directly with a quartz crystal.

The present setup allowed a reasonably accurate calibration by using the aluminum junction in the superconducting state as a detector and biasing the tin junction at an energy below that of the gap, so that it was emitting phonons at an energy determined by the Josephson frequency.¹² These Josephson phonons have an extremely narrow bandwidth ($\lesssim 1 \mu\text{eV}$) and so they can also be used to determine the resolution of the system. In the present experiment, due to inhomogeneities in the stress application, the maximum resolution was found to be $\sim 0.1 \text{ meV}$.

IV. RESULTS

Junctions of varying oxide thickness (and so tunneling resistance R_T) were prepared and spectra measured using the stress-tuned spectrometer as detailed in the preceding section. In all cases the junction bias V_B was fixed at a value in the range 1–4 mV and a small modulation voltage V_M was superimposed on top of it. In all measurements here, V_M was set to 1 mV since this is approximately the expected linewidth due to the thermal broadening at 1.5 K (the temperature of the helium bath) and so gives the maximum signal with minimum loss of resolution. The modulation frequency f_M was set at $\sim 1500 \text{ Hz}$ and the voltage on the detector was phase detected with the phonon output from the emitter at a frequency of either f_M or $2f_M$ in order to give the first and second differential spectra in the usual way. The spectrum from the junction (determined by V_B) was kept constant while the stress applied to the crystal (and so the splitting of the acceptors) was swept and the pressure monitored by the quartz transducer.

Figure 4 shows the first differential of the phonon spectrum with respect to bias voltage ($\partial P/\partial V_B$). As can be seen, the spectrum is dominated by a large thermal background which we attribute to the Ohmic heating of the

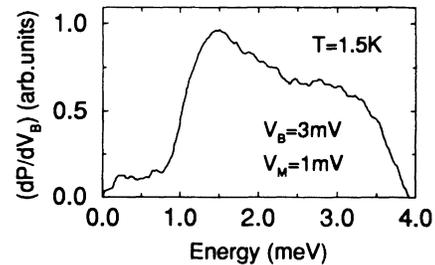


FIG. 4. Experimentally measured spectra of $\partial P/\partial V_B$. The main peak is the thermal emission from the resistively heated films. The steep onset at 1.2 meV is caused by the detector threshold. The signal from the tunneling electrons is seen as a side peak positioned at 3 meV and is equivalent to the curves shown in the middle graph of Fig. 2.

metal films. The sharp onset seen at an energy of $\sim 1 \text{ meV}$ is due to the threshold of the detector. Most importantly, the emission from the tunneling process is also clearly visible as a side peak at an energy of $eV_B = 3 \text{ meV}$, which corresponds to the steep rise close to the cutoff frequency as shown in the central graph of Fig. 2.

In Fig. 5, spectra measured at double the modulation frequency are shown for a series of values of V_B as indicated. These curves are equivalent to the second differential spectra $\partial^2 P/\partial V_B^2$ and the spectra show the quasi-monochromatic form predicted. As can be seen, other than the case of $V_B = 1 \text{ mV}$ (which is below the onset energy of the detector), for each spectrum the peak coincides with the junction bias indicating a good degree of tunability. The linewidth of the peaks is of the order of 1.5 meV and is as expected from the combined effects of the bias modulation and the thermal broadening. Comparing these curves with that in Fig. 4 clearly shows that the thermal background is now much reduced in relation to the monochromatic part. The fact that the thermal signal is larger than the tunneling signal in

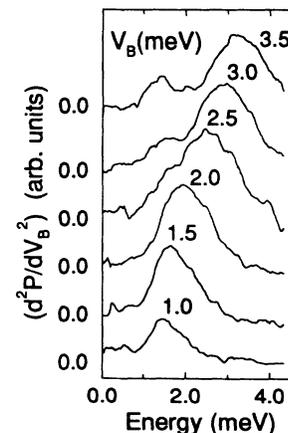


FIG. 5. Experimentally measured spectra of $\partial^2 P/\partial V_B^2$ for a series of values of V_B as indicated, where the quasi-monochromatic nature of the signal and its tunability are clearly demonstrated. The thermal background is seen as a small bump only at the highest bias. In this device, both R_J and R_F are $\sim 0.5 \Omega$.

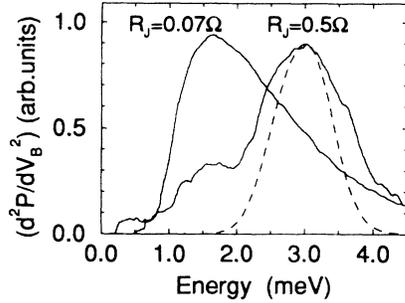


FIG. 6. Experimentally measured spectra of $\partial^2 P/\partial V_B^2$ for two different values of R_J and $R_F=0.5 \Omega$. In each case $V_B=3$ mV and $V_M=1$ mV. The two signals are not to scale, but, as can be seen, the reduction in R_J has led to the complete domination of the spectrum by the thermal background caused by the phonon emission due to film heating. The dashed curve is the spectrum calculated numerically from Eq. (1) for supposedly identical conditions of temperature and modulation and clearly shows that the degree of broadening is worse than expected, indicating that the electron temperature may be above that of the helium bath.

$\partial P/\partial V_B$, and vice versa in $\partial^2 P/\partial V_B^2$, is an indication that a relatively abrupt edge must occur in the first differential signal from the tunneling electrons whereas the thermal signal depends only weakly on V_B .

This device had a film resistance of $R_\square \sim 0.5 \Omega$ and a tunneling resistance of $R_T=0.5 \Omega$ at an overlap area of 0.25 mm^2 . Since the value of V_B across the devices is fixed by the experiment at <4 mV, the only way of increasing the power output from the junction is by changing the junction resistance, with a reducing resistance leading to higher powers.

Another junction, grown with $R_T=0.07 \Omega$ (and so with a larger expected monochromatic phonon power output), was also tested and the results of the measurement of $\partial^2 P/\partial V_B^2$ for both junctions are compared in Fig. 6. In the junction with the smaller value of R_T , the thermal background has become overwhelming, with the tunneling signal not readily visible. It can be easily shown that the ratio of total power from the junction to that from the films is given as $P_T/P_F = R_F/R_J$, which thus explains the differing ratios of background to monochromatic signal. Junctions with much higher values of R_T have also been prepared; however, in these the signal to noise ratio was too poor to make useful measurements of the second differential.

V. DISCUSSION

As outlined in the preceding section, it is possible to produce quasimonochromatic high frequency phonons by modulating a biased NSTJ. One disadvantage of the

present device is that it can be operated successfully only in a narrow range of tunnel resistance for which the monochromatic power output is rather low. In order to improve this, it would require a reduction of the resistance of the aluminum films; however, the resistivity R_\square of the films used is already close to the limiting value set by boundary scattering at a thickness of 250 \AA .

One possibility for the reduction of R_F would be to use Au instead of Al, which has an electron phonon scattering time τ_{ep} about ten times greater¹³ and so allows films to be made accordingly thicker while still avoiding phonon reabsorption, resulting easily in a reduction of R_\square by a factor of about two orders of magnitude. Preliminary measurements have been made using Au-AlO_x-Au tunnel junctions, but showed no evidence of high frequency phonons. We conclude that in gold $\tau_{ee} < \tau_{ep}$, i.e., the electron-electron scattering is the dominant relaxation process.

However, in Al, the observation of high frequency phonon emission demonstrates that $\tau_{ep} < \tau_{ee}$. Estimating τ_{ep} of Al from Ref. 13 gives 40 ps at 2 meV, which sets the lower limit of τ_{ee} for electrons at the same energy. Using Fermi liquid theory¹⁴ one can estimate the thermalization time by electron-electron scattering to be 5 ns, in agreement with our result. Analysis of alternative metals is presently under way, which should give further information about the relative values of τ_{ee} and τ_{ep} as well as possibly providing an even better material for use in tunnel junction fabrication.

We conclude this article by stating that quasi-monochromatic and tunable phonon emission from an NSTJ was observed and the ultimate resolution of the presently available junctions is limited by both the modulation of the bias voltage required to give a detectable signal and also by the thermal broadening of the excited population. The thermal broadening has been found theoretically to be $\sim 6kT_e$, which is ~ 1 meV at 1.5 K (corresponding to $\lesssim 30 \mu\text{eV}$ at a temperature of 50 mK), which is the lowest temperature feasible for this type of experiment. The frequency range should be as broad as that of the SCTJ, which has been shown to exceed 10 meV.²

Most importantly, this device allows spectroscopic measurements to be made in a magnetic field. Preliminary measurements have already been made in fields up to 18 T, which show evidence of the monochromaticity of the source, and these will be published in due course.

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