# NONEQUILIBRIUM PHONONS

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## 1. INTRODUCTION

Nonequilibrium or nonthermal phonon distributions in solids in general result from any kind of energy supply to the system. Experimentally thin superconducting films are especially well suited for studies of nonequilibrium excitation distributions. Quasiparticle and phonon distributions depend on the primary excitation processes, e.g. electron-injection by tunneling, phonon, microwave or optical radiation as well on the properties of the system as quasiparticle and phonon lifetimes, mean free path values and phonon escape conditions from the superconductor. Nonequilibrium in superconductors gives rise to an ample field of different phenomena. This chapter reviews work on nonequilibrium phonons resulting from tunnel injection, and phonon generation and detection in Section 2, experimental probing of nonequilibrium phonon emission in Section 3, applications to phonon absorption spectrocopy in Section 4, quantitative phonon intensity measurements in Section 5, applications to phonon emission spectroscopy in Section 6, and aspects of future work in Section 7.

Work in this field started around 1965. Earlier reviews (Eisenmenger 1969, Dayem 1972, Renk 1972, Kinder 1973, Kinder 1975, Eisenmenger 1976a and Bron 1980) discuss fundamental experiments and theory, the application to phonon spectroscopy and compare also with other techniques, as e.g. heat pulse experiments or optical generation and detection of acoustical phonons.

Elastic single electron tunneling between two superconducting films separated by a thin oxide barrier results in a nonequilibrium energy distribution of quasiparticles. The quasiparticles decay predominantly under phonon emission. This results in a nonequilibrium phonon-energy distribution. The phonon frequencies typically extend from 50 GHz to 1.5 THz. Since the phonon mean free path is comparable to or exceeds the film-thickness (100 Å to 1000 Å) a significant number of phonons can escape into the insulator single crystal substrate without reabsorption or thermalization in the superconductor. The phonon spectrum can be analyzed with a frequency selective superconducting tunneling detector, thus giving information on the nonequilibrium distribution. By the characteristic discontinuities of this spectrum, applications for the spectroscopy with acoustic phonons are possible, especially with respect to resonant phonon absorption in the volume of substrate crystals and also phonon reflection and absorption in transmission across boundaries. In addition, phonon absorption and reabsorption processes in superconductors can be studied. Finally, information on phonon spectra can also be obtained by directly analyzing the quasiparticle distribution in superconducting films resulting from phonon absorption by pairbreaking.

## 2. QUASIPARTICLE AND PHONON TRANSITIONS, TUNNELING, PHONON DETECTION AND GENERATION

### 2.1 Quasiparticle and Phonon Transitions

Single quasiparticles in superconductors are characterized by the BCS (Bardeen et al., 1957) energy-wavevector relation $E(k)$, cf. Fig. 1. The dispersion relation has a minimum with $E = \Delta$, the energy gap at the Fermi-wavevector $k_F$. Electron and holelike excitation branches are symmetrical to $k_F$ for small deviations from $k_F$. Excited quasiparticles decay under spontaneous or stimulated

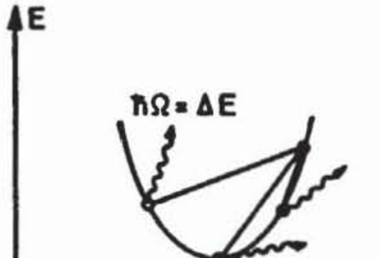


Fig. 1. Phonon emission by quasiparticle relaxation.

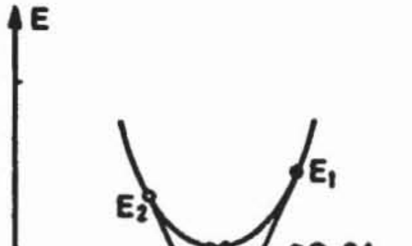


Fig. 2. Phonon emission by quasiparticle recombination.

phonon emission into states of lower energy either within the same branch or changing over to the other branch (Tinkham, 1972). The phonon energies emitted in this relaxation transition (also named phonon "Bremsstrahlung" (Kinder, 1972a)) range from $\hbar\Omega = 0$ to $\hbar\Omega = E - \Delta$ with E the quasiparticle excitation energy. The total spontaneous transition probability $\tau^{-1}$ depends on the quasiparticle energy approximately as $\tau^{-1} \alpha (E-\Delta)^{3,5}$ (Tewordt, 1962), indicating that relaxation of quasiparticles close at the gap edge is vanishing. This can be understood from the decreasing number of phonon states available for the transition and by destructive interference in the BCS transition probability (Bardeen et al., 1957).

The reverse process of relaxation is phonon reabsorption by excited quasiparticles. This process becomes important at high quasiparticle densities; i.e. at temperatures not small compared to $T_c$ or under conditions of high quasiparticle population by injection, resulting in an inelastic decay of nonequilibrium phonons.

The second process of quasiparticle decay is the recombination of two quasiparticles to a Cooper-pair in the superconducting groundstate under the emission of one recombination phonon of the energy $\hbar\Omega = E_1 + E_2$; $E_1$ and $E_2$ are the energies of the recombining quasiparticles. The minimum energy in this process is $\hbar\Omega = 2\Delta$ for quasiparticles at the gap edge. The recombination rate $\tau_R^{-1}$ for quasiparticles at the gap edge depends on their number density as $\tau_R^{-1} \alpha N(\Delta)$. The reverse process, i.e. phonon absorption by pairbreaking, requires the minimum phonon energy $\hbar\Omega = 2\Delta$. This process is used for phonon detection, and is further important in measurements of the quasiparticle recombination lifetime. Phonons emitted in recombination are reabsorbed via Cooper-pairbreaking with high probability (the phonon mean free path for pairbreaking is of the order of 100 Å to 1000 Å, thus leading to an enhanced effective lifetime $\tau_{eff} \equiv \tau_R^* > \tau_R$ by $2\Delta$ phonon trapping (Rothwarf and Taylor, 1967). Depending on the phonon surface escape and volume decay either by quasiparticle excitation or anharmonic phonon interactions, $\tau_{eff}$ in superconducting films is up to two orders of magnitude larger than $\tau_R$ (Eisenmenger et al., 1977). The absolute values for $\tau$ and $\tau_R$ and their dependence on quasiparticle energy and population have been studied theoretically in detail by Kaplan et al. (1976).

Nonequilibrium distributions of quasiparticles and phonons are determined by the processes discussed above. Calculations based on the BCS transition rates make use of coupled kinetic equations (rate equations) of the quasiparticle and phonon system. Important parameters entering such calculations are: The energy distribution of the primary quasiparticle excitation rates as e.g. by tunneling injection, phonon-, microwave- or photon irradiation, the quasiparticle - phonon and the phonon - quasiparticle transition rates, the phonon lifetimes for surface-boundary escape and volume decay, and also elastic scattering rates for quasiparticles and phonons in cases of inhomogeneous distributions; i.e. diffusion. The latter contribution is of importance with respect to high energy phonon escape from superconducting films and the possible occurrence of nonequilibrium instabilities (cf. Langenberg, 1975 and Chapt. 10).

Since the complete numerical treatment of the kinetic equations is complicated, different approximations have been used. With respect to nonequilibrium phonons and quasiparticles at the gap edge a first treatment by Rothwarf and Taylor (1967) showed the important result that the experimental quasiparticle recombination lifetime $\tau_{eff}$ is significantly increased as compared to the intrinsic recombination lifetime $\tau_R$. More general models, trying to simplify the nonequilibrium description by introducing quasithermodynamic quantities as an effective chemical potential $\mu^*$, and a "hot quasiparticle" temperature $T^*$ have been proposed by Owen and Scalapino (1972) and Parker (1975). Nonequilibrium phonon emission of tunneling junctions under weak injection has been treated by taking account of only first step relaxation transitions without recombination (Kinder et al., 1970) and including recombination (Welte 1973, cf. Forkel et al., 1973). For the "weak injection (no gap reduction)" situation the detailed balance of high energy quasiparticles and phonons including limited phonon escape has been treated by Dayem and Wiegand (1972). Detailed phonon spectra by relaxation and recombination at finite temperatures and finite tunnel injection voltages taking account of successive relaxation steps have been calculated by Welte (1976). A simple approximation of high energy phonon emission was used by Forkel (1977). Finally strong tunnel injection without phonon reabsorption (Welte 1974, Welte 1976, cf. Welte and Eisenmenger 1980) and with phonon reabsorption and gap reduction under different phonon escape conditions has been treated (Chang and Scalapino, 1978, Chang 1980). In the "weak" tunneling injection case the energy-gap reduction by a stationary increase of the number density of quasiparticles at the gap edge remains below one percent. This condition is typical for the phonon generation with tunneling junctions; i.e. bath temperatures $T < 0.5\ T_c$ and high phonon escape by the condition $d \leq \Lambda$ ($d$ = filmthickness, $\Lambda$ = phonon mean free path). Although the negligible energy-gap reduction in the weak injection case may appear as characteristic for a minute deviation from thermal equilibrium, the high energy phonon emission spectra in contrast indicate a strong nonequilibrium situation.

Phonon emission spectra of superconducting films, therefore, are very sensitive to deviations from thermal equilibrium. Thus, experimental investigations of phonon emission complement the direct measurements of nonequilibrium quasiparticle distributions by three-layer tunneling experiments discussed in this volume by Gray (1980).

## 2.2 Tunneling Junctions for Phonon Experiments

In using superconducting film tunneling junctions for phonon generation and detection experiments, elastic single particle tunneling (cf. Gray, 1980) is the most important process to be discussed. The junctions are usually prepared by vacuum deposition (Giaever and Megerle, 1961; Giaever, 1964; Solymar, 1972; Eisenmenger, 1976a) of Al, Sn, or Pb layers with thickness of the order of 1000 Å or less separated by an approximately 10 Å thick oxide barrier which is formed by exposing the first layer to air, oxygen or by glow discharge with oxygen background. In order to obtain a sharp gap structure and an enhanced energy gap, also granular Al-films (Al-evaporation with oxygen background $10^{-5}$ Torr) or Pb:Bi alloy films are used. The tunneling overlap area is typically of the order of 1 mm². As evaporation substrates, especially for phonon experiments, optically polished single crystals; e.g. $Al_2O_3$, Si, etc.: with long phonon mean free path, typically exceeding 1 cm, are used. In these experiments glass substrates or other materials with high phonon scattering are not well suited. Details for different possible procedures in junction preparation can be found in the literature (cf. Solymar, 1972; Eisenmenger, 1976a).

For most phonon experiments, junctions composed of two identical superconductors are used. The I-V (current - voltage) characteristic (see Fig. 3) can be most conveniently described in terms of the density of states in the semiconductor picture (Giaever and Megerle, 1961), cf. Figs. 4 and 5.

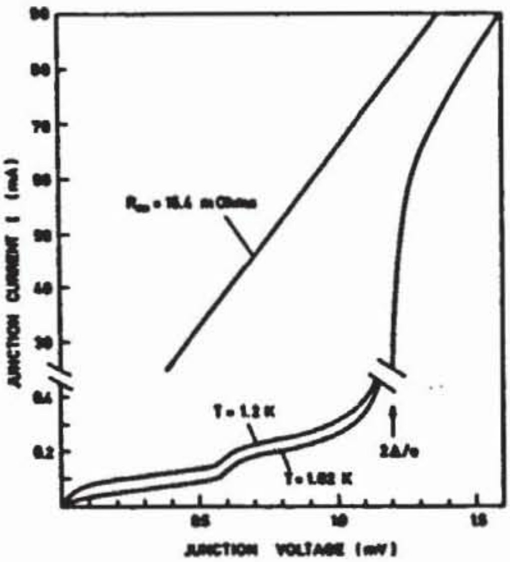


Fig. 3. Current-voltage characteristic of a Sn-I-Sn tunneling junction of 1 mm² area, for phonon generation and detection.

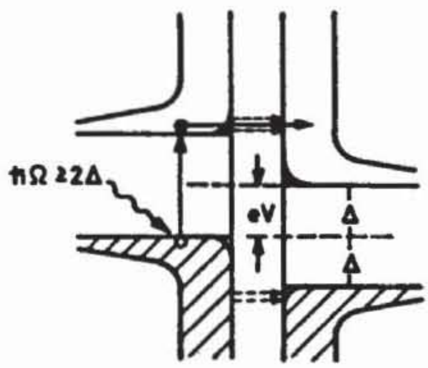


Fig. 4. Phonon detection in the range $0 < eV < 2\Delta$.

For battery voltages $|V| < 2\Delta/e$, with $\Delta$ the superconductor energy gap and e the electron charge, only a small temperature dependent current results from thermally excited quasiparticles. Irradiation with phonons of energy $\hbar\Omega > 2\Delta$ leads to additional quasiparticle excitations via Cooper-pairbreaking and a corresponding increase of the tunneling current. Thus, phonons of sufficient energy can easily be detected with a tunneling junction operated in the voltage range $|V| < 2\Delta/e$, as indicated in Figs. 3 and 4.

## 2.3 Phonon Detection

A quantitative model for phonon detection can be based on parameters which are determined by experiment (cf. Eisenmenger, 1976). Experimentally low ohmic junctions with asymptotic tunneling resistance $R_\infty$ between $10^{-2}$ Ohms and $10^{-3}$ Ohms (1 $mm^2$) are preferable since the phonon detection sensitivity increases with tunneling probability.

In order to suppress the DC-Josephson current, a magnetic field of about 20 to 50 Oe parallel to the junction plane (Helmholtz coils) is required. Shielding the earth-magnetic field can increase the electronic sensitivity, since flux trapping in cooling down below $T_c$ reduces the dynamic resistance and $\tau_{eff}$ in the biasing point. Instead of magnetic shielding an orientation of the junction plane parallel to the earth-magnetic field is mostly sufficient. The detector is current-biased usually at a voltage $V \approx 1.4\ \Delta/e$, just above the current step at $V = \Delta/e$, caused by two particle tunneling.

Biasing below $V = \Delta/e$ is often difficult because small discontinuities (Fiske steps) and Josephson subharmonic structures limit the range of linearity.

Electronically the detector has the properties of a current generator. Phonons absorbed by pairbreaking increase the quasiparticle population at the gap edge. This results in an almost parallel shift of the I(V) characteristic in the thermal tunneling regime $|V| < 2\Delta/e$ to higher currents corresponding to a differential detector current signal $I_d$ at constant biasing voltage. With constant biasing current instead the resulting open circuit detector voltage signal is $v_d = I_d \cdot R_{dyn}$, with $R_{dyn}$ the dynamic resistance at the biasing point. For good detector junctions $R_{dyn}$ ranges from 0.1 to 1 ohm. An optimal signal-to-noise ration, therefore, requires the use of low-ohmic strip lines and pulse transformation for matching to the typical 50 $\Omega$ input impedance of a low-noise-pulse preamplifier. Alternatively a number of small area tunnel detectors in series can be used for matching (cf. Eisenmenger, 1976a).

Detector calibration in terms of the number of phonons absorbed by pairbreaking under stationary conditions is most easily performed using the relation (cf. Eisenmenger, 1976)

$$I_d = 2\dot{N}_\omega \cdot \frac{\tau_{eff}(T)}{\Omega} \cdot \frac{I(T)}{N(T)}$$

with $\dot{N}_\omega$ the number of $2\Delta$ phonons per unit time absorbed by pair-breaking; $\Omega$ = the volume of the tunneling junction; $\tau_{eff}$ the experimentally observed effective quasiparticle recombination time; $N(T)$ = the equilibrium quasiparticle number density at T and $I(T)$ the tunneling current by thermally excited quasiparticles. $N(T)$ can be calculated according to the approximation valid within 5 % for $T < 0.5\ T_c$:

$$N(T) = 4N(0)\Delta(T)\sqrt{\frac{\pi}{2}\frac{kT}{\Delta(T)}} \cdot e^{-\Delta(T)/kT}$$

with $N(0)$ the one spin density of states at the Fermi level, as known from electronic specific heat data. $I(T)$ is experimentally obtained from the temperature dependence of the tunneling current at constant voltage bias. Plotting this current as function of $\sqrt{T}\cdot e^{-\Delta/kT}$ and extrapolating to $T = 0$ gives the almost temperature independent leakage and two-particle-current contributions which have to be subtracted from the measured current, thus obtaining $I(T)$. $\tau_{eff}(T)$ can be easily obtained in a phonon pulse experiment from the exponential decay of the detector signal depending on temperature in proportion to $T^{-1/2}\cdot e^{\Delta/kT}$. $I_d$ is obtained from the measured electronic signal voltage taking account of the total amplification and the ohmic load of the stripline and pulse-transformer system in parallel to $R_{dyn}$. Simple calibration of the total electronic system is accomplished by simulating a current pulse signal (introduced from a high ohmic source) at the tunneling junction and measurement of the amplified voltage signal. The detector relation $I_d \propto \dot{N}_\omega$ holds for the linear detection range; i.e. $I_d \ll I(T)$ with increase of the number of quasiparticles by phonon absorption small compared to the number-density of thermally excited quasiparticles. Experimentally, the fulfillment of this condition is easily checked. A further limitation of linearity results from the curvature of the $I(V)$ curve (variation of $R_{dyn}$) at the detector bias voltage. Mostly a voltage excursion of a few percent of $2\Delta$ can be admitted. The attainable phonon power resolution depends on the noise figure of the preamplifier; minimal phonon powers of $10^{-13}$ Watts with one second integration time can be detected.

The detector equation, so far, describes the sensitivity with respect to $2\Delta$-phonons. Phonons with lower energy cannot directly contribute to pairbreaking, but may be weakly absorbed by thermally excited quasiparticles. The energy supplied by this process is partly upconverted to $2\Delta$-phonons (heating influence). For $T = 0.5\ T_c$ the corresponding detector-signal contribution is, however, at least three orders of magnitude below the sensitivity for $2\Delta$-phonons. For phonon energies between $2\Delta$ and $4\Delta$ the sensitivity with respect to the number of absorbed phonons remains constant. Increasing the phonon energy above $4\Delta$ results in an almost linear energy dependence, since quasiparticles are excited via pairbreaking also to energies above $3\Delta$ and succeeding relaxation contributes to secondary $2\Delta$-phonons within the detector. An additional frequency dependence in principle results from the energy dependence of the phonon mean free path. In Sn- and Pb-junctions of at least 2000 Å thickness the $2\Delta$-phonon mean free path is equal or shorter than this length and all incident $2\Delta$-phonons are absorbed by pairbreaking. In pure Al-junctions the energy dependence of the phonon mean free path must be taken account of. For further details see (Eisenmenger, 1976a; Wyatt 1976).

## 2.4 Phonon Generation with Superconducting Tunneling Junctions

At voltages $V \geq 2\Delta/e$ the tunneling current increases discontinuously (Fig. 3) and approaches with higher voltages the linear normal conductor limit. In this regime quasiparticles are excited via pairbreaking by the battery energy leading to nonequilibrium injection into the films. In addition, also tunneling by thermal or injection-excited quasiparticles contributes to the injection distribution which is shown qualitatively in the upper part of Fig. 5. The sharp maxima at the lowest and highest quasiparticle energies result from the singularities in the BCS-densities of state, c.f. (Gray, 1980; McMillan and Rowell, 1969). A detailed account of the stationary quasiparticle occupation by tunnel-injection, considering especially branch imbalance and the corresponding relaxation and recombination rates in Al-films, has been given by Kirtley et al. (1980). So far, the possible influence of branch imbalance on phonon experiments has not been studied, and will not be further discussed.

The quasiparticle decay takes place by relaxation and recombination transitions predominantly under phonon emission. Since the available densities of state for photon emission is by several orders of magnitude below the density of phonon states, photon emission can be neglected. The phenomenological similarity between relaxation phonon emission and continuous x-rays has led to the name "phonon Bremsstrahlung" (Kinder, 1972a).

Again as consequence of the singularities in the density of states the quasiparticle relaxation and recombination decay results in characteristic structures (Kinder et al., 1970; Kinder, 1972a; Dayem and Wiegand, 1972; Welte et al., 1972; Welte, 1973; Forkel et al., 1973; Forkel, 1974; Welte, 1976; Forkel, 1977; Chang and Scalapino, 1978) of the phonon spectrum, as shown in the lower part of Fig. 5. The most significant structures are:

a) $\hbar\Omega = 2\Delta$, minimum energy of the recombination spectrum.

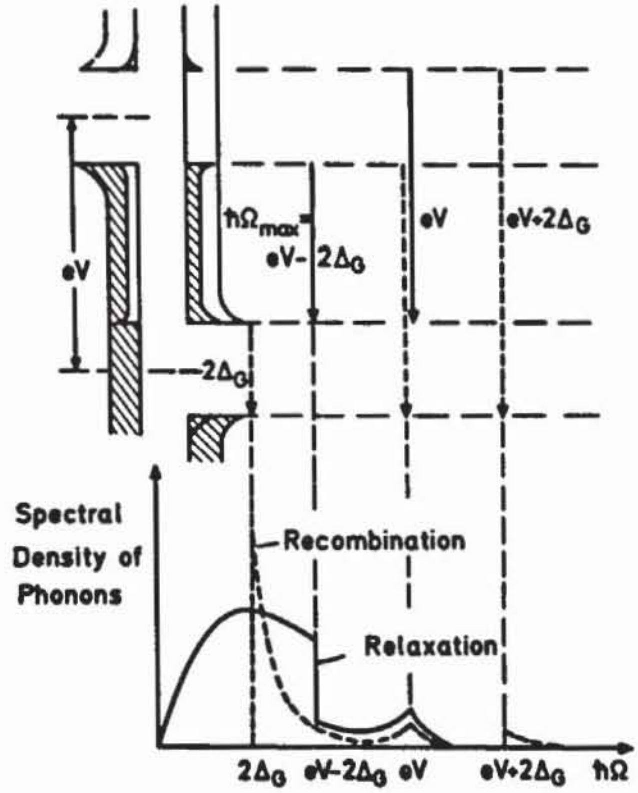


Fig. 5. Phonon emission spectra for relaxation and recombination at finite temperature. Transitions corresponding to density of state- or quasiparticle population singularities give rise to characteristic spectral structures, as discussed in the text.

b) $\hbar\Omega$ = eV, direct recombination of injected quasiparticles with thermal quasiparticles.
c) $\hbar\Omega$ = eV + 2Δ, minimum phonon energy for the direct recombination of tunnel-injected thermal quasiparticles with thermal excitations.
d) $\hbar\Omega$ = eV - 2Δ, upper edge of the relaxation spectrum; the shape and voltage dependence of this structure provides the possibility of voltage tunable phonon spectroscopy.
e) $\hbar\Omega$ = eV, relaxation of tunnel-injected thermal quasiparticles.

In numerical calculations of these spectra, different approximations have been used, cf. Sect. 2.1. In the simplest form which for low injection and low temperatures already reveals the most significant spectral contributions, only first step processes are considered. An example for the calculated first step relaxation spectrum at T > 0 and eV > $2\Delta_G$ is given in Fig. 6 (Welte 1973), indicating the upper discontinuity in the relaxation phonon spectrum at $\hbar\Omega = eV_G - 2\Delta_G$ and the presence of the thermal peak structure at $\hbar\Omega = eV_G$. The height of the relaxation edge decreases with in-

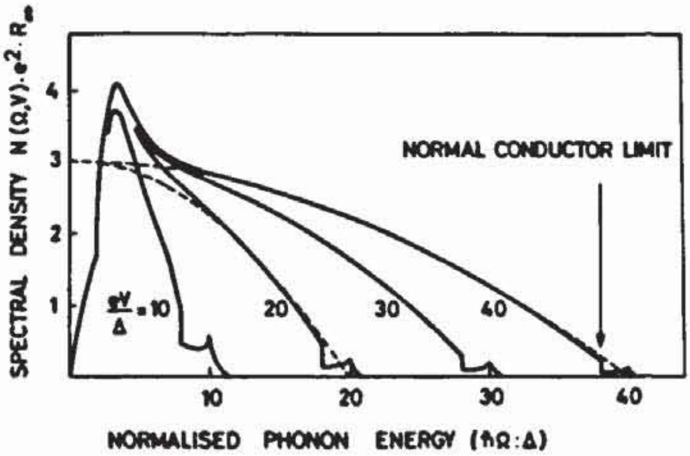


Fig. 6. Calculated relaxation phonon spectra for high generator voltages at finite temperature and weak injection; first step decay; no reabsorption; dotted line: normal conductor limit for T = 0; (Welte, 1973; Forkel et al., 1973).

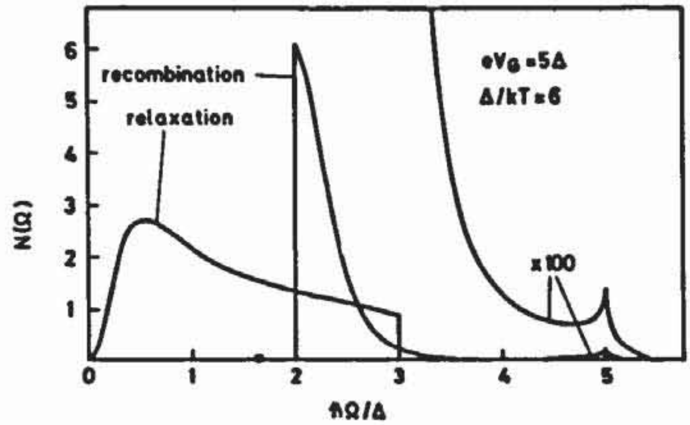


Fig. 7. Calculated relaxation and recombination phonon spectrum at finite temperature and weak injection, no reabsorption, successive quasiparticle decay included (Welte, 1976).

creasing $V_G$ and temperature. For comparison also the phonon spectrum for the normal conducting limit at T = 0 is introduced.

Since quasiparticles decay into all available unoccupied energy levels, a higher degree of approximation must take account of secondary and higher order decay steps. Fig. 7 shows an example for weak injection without phonon reabsorption (Welte, 1976) concerning the relaxation and recombination spectrum at higher temperatures. As seen from this figure, the inclusion of secondary and higher order quasiparticle decay steps does not change the qualitative spectral shape. Recombination between injected quasiparticles becomes important under strong injection as well as reabsorption of phonons by pairbreaking and quasiparticle excitation. The influence of di-

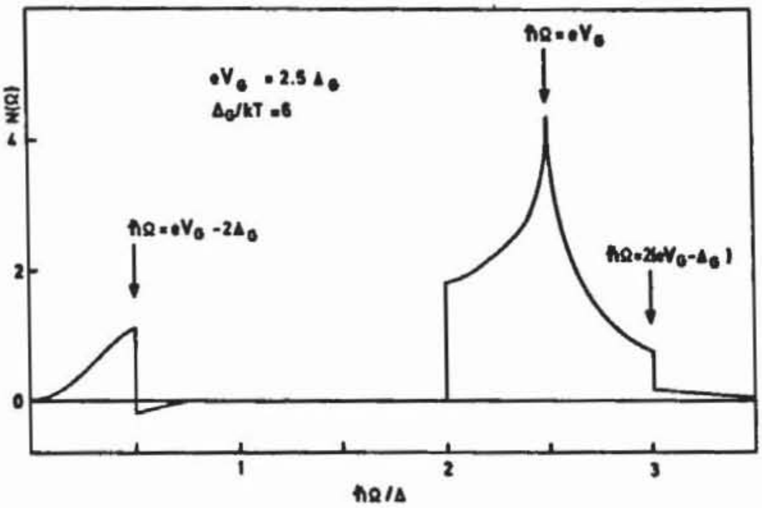


Fig. 8. Calculated relaxation and recombination phonon spectrum at finite temperature and finite tunnel injection, indicating the contribution of the direct recombination of high energy quasiparticles. (Welte, 1976)

rect recombination without reabsorption in the case of strong injection at low temperatures is shown in Fig. 8 (Welte, 1976) by the occurence of an upper spectral edge in the recombination spectrum at $\hbar\Omega = 2(eV-\Delta_G)$, a strong increase of the peak structure at $\hbar\Omega = eV_G$ and by the high population of quasiparticles at the gap edge. Even without taking account of the direct recombination of injected quasiparticles, high injection results in an increased bandwidth of the recombination phonons as consequence of the population of quasiparticle states at and above the gap edge. In this situation a proper account of phonon escape at the film boundaries and the anharmonic phonon decay at the surface or within the volume of the films is necessary. High injection nonequilibrium under limited phonon escape will be discussed in this volume, Chapter 9, by Chang (1980).

## 3. EXPERIMENTAL EVIDENCE FOR NONEQUILIBRIUM PHONON EMISSION

In the first observation of nonequilibrium phonon emission (Eisenmenger and Dayem, 1967) Sn-I-Sn-junctions as phonon generator and detector on $Al_2O_3$ as substrate crystal have been used. In this experiment, a nearly linear increase of the detected phonon signal amplitude, as function of the generator current up to the generator voltage of $V_G = 4\Delta_G/e$ is found, cf. Fig. 9. With higher generator current again a linear signal increase is observed but with a slope nearly three times larger than before; i.e. the dependence of the detector signal on the generator current develops a sharp bend at $V_G = 4\Delta_G/e$. This can be easily understood in terms of the onset of the contribution of relaxation phonons with maximum energy $\hbar\Omega = eV_G - 2\Delta_G$ at the voltage $V_G = 4\Delta_G/e$. Below this generator voltage only recombination phonons can contribute to the detector signal.

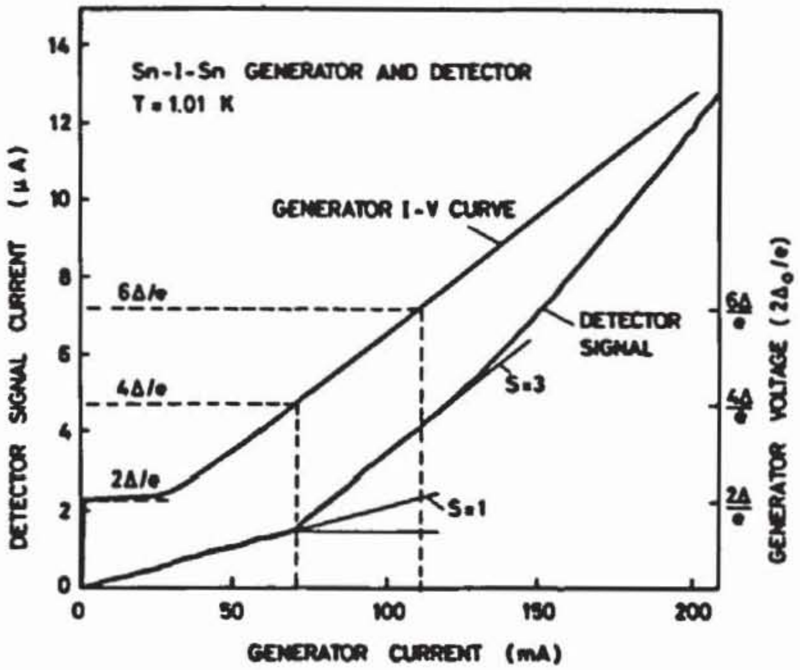


Fig. 9. Detector signal as function of the generator current (voltage: right scale) for Sn-I-Sn generator and detector, T = 1.01 K. For comparison with the characteristic generator voltages 4Δ/e and 6Δ/e, the generator I-V curve is introduced (Eisenmenger and Dayem 1967; Kinder et al., 1970).

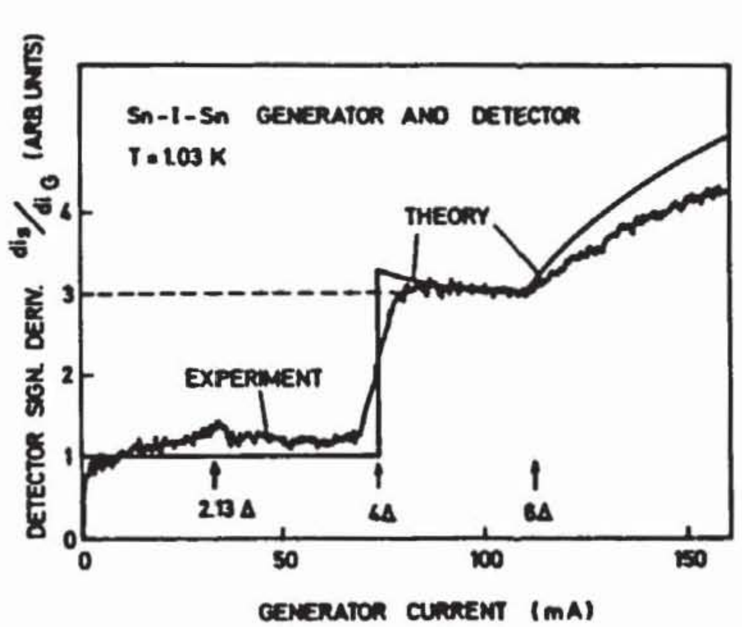


Fig. 10. Derivative of the detector signal with respect to the generator current. Sn-I-Sn generator and detector. T = 1.03 K; cf. Fig. 9 (Eisenmenger and Dayem 1967; Kinder et al., 1970).

This result clearly indicates nonequilibrium phonon emission under weak injection conditions; i.e. strongly impeded phonon thermalization in thin superconducting films. By superimposing small current pulses on the D.C.-generator current it is observed that the differential detector signal is roughly constant with a step at $V_G = 4\Delta_G/e$, and with the onset of a linear increase at $V_G = 6\Delta_G/e$, cf. Fig. 10. This indicates further contributions to the number of detected 2Δ-phonons by repeated phonon reabsorption and reemission in the generator or detector (Kinder et al., 1970); i.e. a splitting or primary relaxation phonons with the energy $\hbar\Omega = 4\Delta$ into two phonons with the energy $\hbar\Omega = 2\Delta$. The theoretical change of slope at $eV_G = 4\Delta_G$ in Fig. 9 and the corresponding step in Fig. 10 for $T = 0$ in the weak injection limit amounts to 3.1 (Kinder et al.,1970; Long and Adkins, 1973). The experiment in general exhibits a somewhat smaller value, especially for low ohmic generator junctions. In this case the high quasiparticle population at the gap edge leads to an enhancement of the direct recombination of high energy quasiparticles and the corresponding reduction of relaxation transitions. The equivalent situation holds for elevated temperatures.

Further experimental evidence for the emission of 2Δ nonequilibrium phonons from Sn-I-Sn-generators was provided by Dynes et al. (1971), using the mechanical stress tunable phonon transition in Sb-doped Ge-substrates as phonon spectrometer.

The first demonstration of the feasibility of nonequilibrium phonon emission from superconducting Sn-I-Sn tunneling junctions for tunable phonon absorption spectroscopy, cf. Sect. 5, was given by Kinder (1972a), who used the voltage dependence of the upper edge of the relaxation phonon spectrum at $\hbar\Omega = eV_G - 2\Delta_G$ for investigating the phonon absorption of $V^{3+}$ in $Al_2O_3$. In this case relaxation phonons with energy $\hbar\Omega = eV - 2\Delta_G < 2\Delta_G$ at $T < 0.5\ T_c$ are not reabsorbed in the superconducting film and can be radiated into the substrate essentially without further interaction. The upper frequency limit of this method results from the fact that relaxation phonons with the energy $\hbar\Omega \geq 2\Delta_G$ are strongly reabsorbed already in the Sn-films of the generator. This holds also for strong coupling superconductors as Pb. In contrast for thin films of the weak coupling superconductor Al a finite escape of relaxation phonons with $\hbar\Omega > 2\Delta_G$ is to be expected, see Fig. 11, since the 2Δ-phonon mean free path is of the order of 1000 Å (cf. Kaplan et al., 1977).

For the escape of relaxation phonons from the generator junctions in addition to the phonon mean free path the following influences have to be taken into account (Welte et al., 1972; cf. Eisenmenger, 1976a), as depicted in Fig. 11.

i) quasiparticles injected by tunneling have a sharp angle distribution normal (maximum momentum) to the barrier. By momentum conser-

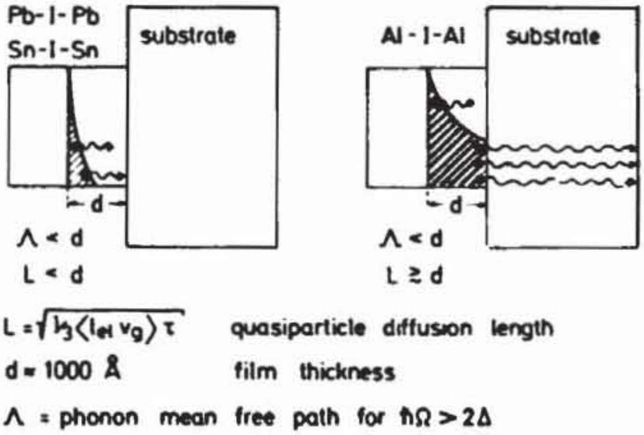


Fig. 11. Phonon escape conditions for $\hbar\Omega > 2\Delta$. The large quasiparticle diffusion length and phonon mean free path $\Lambda$ in Al-films increase the phonon escape probability, as compared to Pb and Sn-films.

vation, phonon emission in the primary relaxation decay is perpendicular to the quasiparticle momentum and therefore confined to directions in the plane of the junction with little escape probability into the substrate, even for phonon mean free path values exceeding the film thickness. The above-mentioned momentum conditions become less stringent for real metal Fermi surfaces.

ii) This situation changes by elastic scattering of quasiparticles before relaxation, corresponding to quasiparticle diffusion in the generator film. Also boundary scattering at the film surfaces result in a wide angle distribution of quasiparticle momenta. Thus, relaxation phonons are generated omnidirectionally and more close to the substrate (cf. Fig. 11) with increased escape probability. This process becomes important if the characteristic decay time for relaxation is long compared to the elastic scattering time of quasiparticles; i.e. again in the case of weak coupling superconductors.

iii) A simple model (Welte et al., 1972; cf. Eisenmenger, 1976a) taking account of these influences and using literature data for the relaxation time, elastic quasiparticle scattering time and the phonon mean free path, indicates a high phonon escape probability of primary relaxation phonons with $\hbar\Omega > 2\Delta_G$ into the substrate for thin Al-films. Thus, Al-junctions can be used preferably for studies of the nonequilibrium phonon emission in a wide frequency range with little phonon reabsorption and also a weak "feedback" coupling of the phonon system to the quasiparticle system. This makes Al-films also well-suited for direct studies of primary nonequilibrium quasiparticle distributions (cf. Kirtley et al., 1980).

With respect to the phonon spectrum, as depicted in Fig. 5, and the calculated spectra, see Figs. 6, 7 or 8, an experimental analysis is possible by using tunneling detectors with different energy gaps, such as Al-junctions evaporated with oxygen background (Welte, 1974, 1976). Using instead a Sn-I-Sn junction as detector, the voltage-dependent contributions of the Al-generator spectrum with $\hbar\Omega \geq 2\Delta_{Sn}$ can be investigated (Welte et al., 1972; Forkel, 1974; Forkel, 1977) applying modulation techniques.

Since the detector is only sensitive to phonons with energies exceeding the detector gap $2\Delta_D$, the detector signal corresponds to the integral of the emitted generator phonon spectrum with a lower cut-off energy at $2\Delta_D$. Modulation of the generator voltage results in a detector signal modulation arising mainly from the changing generator spectral contribution at the energy $\hbar\Omega = 2\Delta_D$. Thus, modulation is equivalent to differentiating the integral generator spectrum at the lower cut-off energy $\hbar\Omega = 2\Delta_D$. The detector modulation signal, therefore, corresponds to the spectral intensity at $2\Delta_D$. By changing the generator bias voltage it is, therefore, possible to scan the voltage dependent structures of the generator spectrum. The experimental arrangement is shown in Fig. 12. Generator, Al-I-Al and detector, Sn-I-Sn are prepared on the opposing flat surfaces of a high quality Si single crystal cylinder (Forkel, 1974 and 1977). The observed detector modulation signal, as function of the generator bias voltage, is shown in Fig. 13, clearly revealing the characteristic spectral structures depicted in Fig. 5 (with inverted frequency scale). This demonstrates the strong deviation of the phonon-, and indirectly also of the quasiparticle population from thermal equilibrium, even under weak injection conditions. The experiment further gives evidence that phonons with $\hbar\Omega > 2\Delta_G$ escape from Al-I-Al generators into the substrate only with little reabsorption by pairbreaking. Corresponding experiments with a Sn-generator, using as reference the phonon absorption line of $^{16}O$ in Si

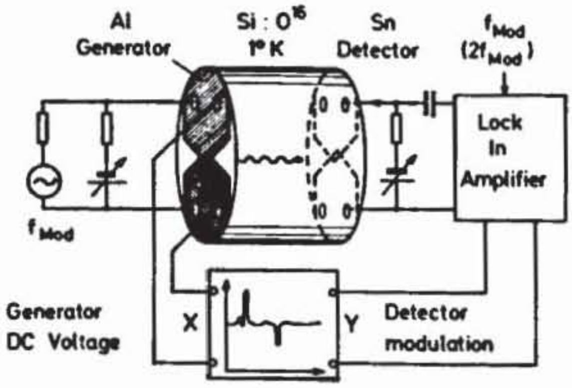


Fig. 12. Experimental arrangement for the analysis of high energy phonon spectra emitted by Al-tunneling junctions as phonon generator. The same measuring technique is used for high energy phonon spectroscopy (Forkel, 1974).

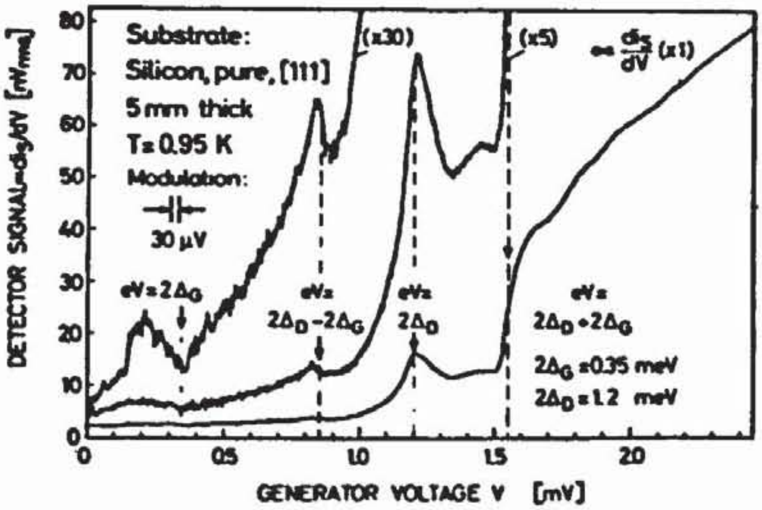


Fig. 13. High energy phonon spectrum emitted by an Al-generator, Sn-I-Sn detector, showing the different spectral structure discussed in Fig. 5 (voltage scale inverted). (Forkel, 1975, see Eisenmenger 1976b).

at 878 GHz, cf. Sect. 4, in contrast demonstrated strong reabsorption (Forkel, 1977). It should be noted that in these experiments with weak injection the strong deviation from equilibrium result from the excitation of quasiparticles by the battery-voltage to energies by far exceeding kT; i.e. the injection of "hot" electrons.

Experiments have been also performed with respect to nonequilibrium phonon emission under strong injection (Welte, 1974, 1976, 1980). These demonstrate the increasing width of the recombination phonon spectrum, the decrease of the lower threshold of 2Δ-recombination spectrum by the reduction of the energy gap, the influence of direct recombination of injected quasiparticles by the enhanced quasiparticle population at the energy gap, thus reducing the relaxation contribution which is also observed at increased temperatures, and finally the direct recombination of injected quasiparticles.

No experimental results on nonequilibrium phonon emission under very high injection with gap reductions up to 50 % are presently known. Such experiments would be valuable with respect to the degree of realization of the $\mu^*$ and $T^*$ quasiparticle distributions in comparison to the results of Willemsen and Gray, (1978) and Kirtley et al., (1980).

## 4. VOLTAGE-TUNABLE MONOCHROMATIC PHONON SPECTROSCOPY

The upper edge of the phonon spectrum emitted in relaxation of quasiparticles at the energy $\hbar\Omega = eV - 2\Delta_G$ can be used for phonon spectroscopy by differentiation (Kinder, 1972a); i.e. modulation techniques. Applying a small sinusoidal or pulse voltage current, in addition to the constant battery voltage $V_G$ to the generator or operating the generator with amplitude modulated pulses, results in a modulation of the emitted spectrum mainly at the energy $\hbar\Omega = eV - 2\Delta_G$ (Fig. 14 b). The detector modulation signal in this case is mainly caused by nearly monochromatic phonons at $\hbar\Omega = eV - 2\Delta_G$ with a linewidth determined by the modulation amplitude. For a substrate crystal containing defects or impurities with resonant phonon scattering simply a variation of the generator voltage allows tunable monochromatic phonon absorption spectroscopy. This technique meanwhile has found a wide area of applications (cf. Proceedings of Phonon Scattering Conferences 1972, 1976, 1980, and reviews: Kinder, 1973; Kinder, 1975; Eisenmenger, 1976a, and Bron, 1979), and is especially useful in studying phonon selection rules, radiationless transitions, phonon resonance backscattering, phonon propagation in insulators, metals, superconductors, semiconductors, amorphous materials, and phonon transmission and reflection at interfaces as e.g. in the Kapitza problem.

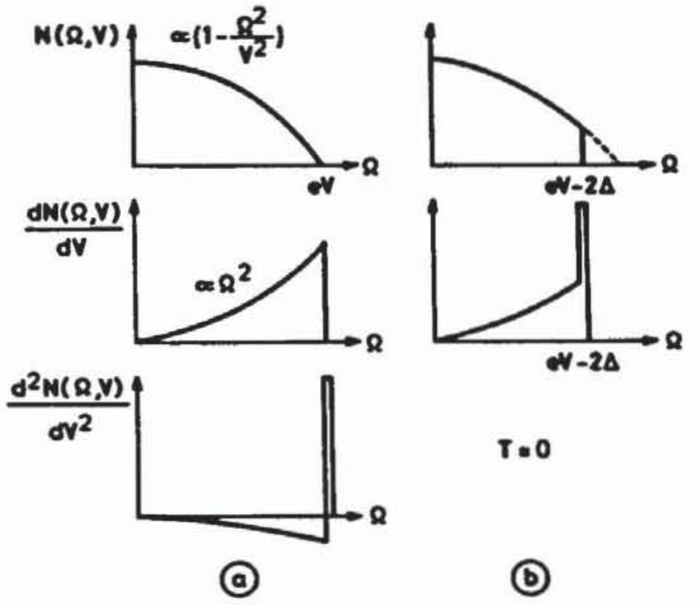


Fig. 14. Modulation of the relaxation spectrum and resulting effective phonon bands: a) approximation for normal conductors or superconductors at energies very large compared to the energy gap, a monochromatic phonon band corresponds to the second derivative (2 ω modulation signal). b) Superconductor relaxation spectrum and monochromatic phonon band of the first derivative (ω modulation signal). (Forkel, 1974)

In phonon spectroscopy two slightly different experimental techniques are used. Avoiding reabsorption in the generator, Kinder (1972a, 1973b) used a Sn-junction as generator for tunable relaxation (Bremsstrahlung) phonons and an Al-junction as detector. In his famous experiment, he demonstrated for the first time monochromatic spectroscopy with nonequilibrium phonons. As a resonant phonon scatterer the Jahn-Teller system $Al_2O_3:V^{3+}$, known from far-infrared and heatconductivity measurements, could be investigated with respect to the selection rules depending on the phonon polarization, see Fig. 15. Also resonant phonon fluorescence in backscattering was demonstrated for the first time. The high phonon-intensity, available in strong coupling superconducting phonon generators, makes this system especially useful for the time of flight separation of longitudinal and transverse phonon contributions.

The upper limiting frequency of 280 GHz for tin junctions can be extended to 870 GHz by using Pb:Bi-junctions as phonon generator (Kinder and Dietsche, 1975). Spectroscopy at higher frequencies may be possible with Nb-junctions.

An alternative way for phonon spectroscopy in the 1 THz-frequency range is the use of Al-generators (Welte et al., 1972, Forkel et al., 1973), since phonon reabsorption for energies $\hbar\Omega > 2\Delta_G$

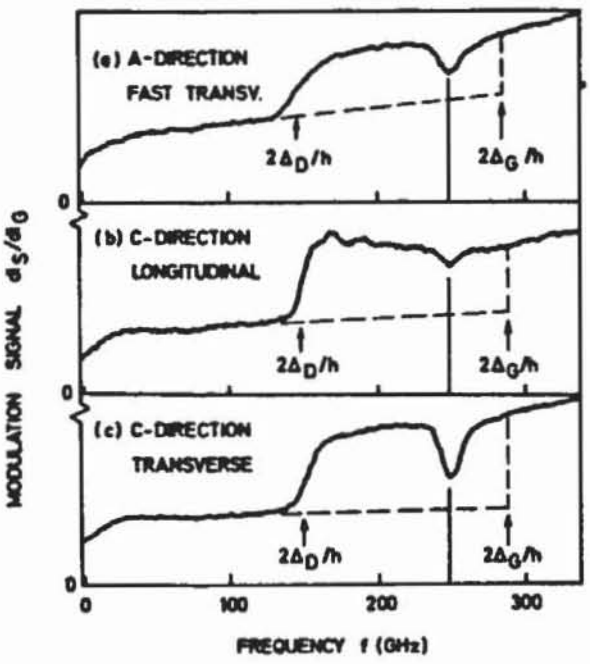


Fig. 15. Phonon-resonant absorption (scattering) in $Al_2O_3:V^{3+}$.
a) fast transverse mode absorption in a-direction.
b) longitudinal mode absorption in c-direction.
c) transverse mode absorption in c-direction.
(Kinder, 1972a).

is comparatively weak (Welte et al., 1972), cf. Sect. 3. The upper frequency limits in Al, resulting from the maxima of phonon dispersion at the Brillouin zone, are 9 THz for longitudinal phonons and 5 THz for transverse phonons. An independent limit results by the momentum conservation between phonons and electrons with $k_{ph\,max} = 2q_F$ at the Fermi surface. Since at higher voltages the relaxation spectrum (Fig. 6) approaches the normal conductor spectrum with a reduction of the amplitude step at $\hbar\Omega = eV - 2\Delta_G$, a second derivative technique (see Fig. 14a) using the second harmonic of the detector signal modulation must be applied. An increase of the available signal power is possible by using very thin Al-films of 100 Å to 200 Å thickness with enhanced energy gap. The high sensitivity and frequency resolution of this technique in the 1 THz range was demonstrated by the resonant phonon absorption (878 GHz) of interstitial oxygen in silicon (Forkel et al., 1973; Forkel, 1977). In the measurement Fig. 16 the resonance absorption of the $^{18}O$ isotope with the natural concentration of $10^{15}$ $cm^{-3}$

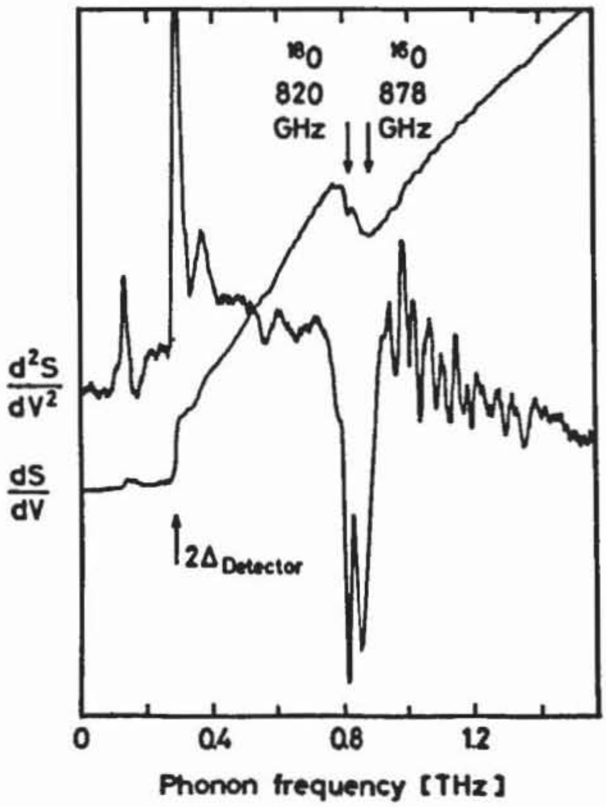


Fig. 16. High frequency phonon absorption of $O_i$ in Si. (Forkel, 1977)

could be detected. The sensitivity is by three orders of magnitude higher than in corresponding FIR measurements and the attainable frequency resolution amounts to 6 GHz. In Fig. 16 also a number of satellite resonances extending to 1.25 THz are observed. These are possibly related to oxygen neighbors with a statistical concentration of $10^{14}$ cm$^{-3}$. The main resonance of $^{16}O$ (concentration $10^{18}$ cm$^{-3}$) is "instrumentally" broadened. At lower concentrations the $^{16}O$ absorption line can be used to study the influence of the generator film-thickness and temperature on the intensity of high frequency phonon emission. Also film-thickness resonances are observable from which the sound velocity and information on phonon reflection at the oxide barrier are obtained. Further spectroscopic examples in the THz-range concern the resonance absorption of the Jahn-Teller systems $Al_2O_3:Ti^{3+}$ at 1.2 THz and $Al_2O_3:V^{4+}$ (Forkel, 1975; Kinder and Dietsche, 1976; Forkel, 1977).

One of the potentialities of acoustic phonon spectroscopy lies in the direct investigation of radiationless transitions, unobservable in FIR-spectroscopy. One of the first examples was the observation of sharp phonon resonance lines in undoped Verneuil $Al_2O_3$ (Kinder, 1972c; Welte et al., 1972; Kinder and Dietsche, 1975) which are still unexplained. A second example is the phonon absorption in Si:In at 4.2 meV. Ultrasonic absorption measurements (Schad and Lassmann, 1976; Lassmann and Zeile, 1980) indicated that Si:In represents a Jahn-Teller system with a ground-level splitting of 4.2 meV. This could directly be verified by phonon spectroscopy (Schenk et al., 1978). The observed resonance energy and the large linewidth (see Fig. 17) could be explained theoretically (Sigmund and Lassmann, 1980). Corresponding results of ultrasonic absorption and phonon spectroscopy were obtained for the Jahn-Teller system Si:B (cf. Lassmann and Zeile, 1980).

Resonant phonon scattering in solids has been also investigated in paraelectric systems with respect to selection rules under level splitting by mechanical stress (cf. Windheim and Kinder, 1975a,b) in ferrimagnetic systems giving evidence for, so far, unobservable sharp resonant magnon - phonon interactions (cf. Mattes et al., 1978); in $MgO:Cr^{2}$ and $MgO:Mn^{3+}$ (Hasan et al., 1978); in $Al_2O_3:Cr^{3+}$ indicating the possibility of virtual phonon exchange coupling between neighbors (Hasan et al., 1979a); in highly doped $Al_2O_3:V^{3+}$ also possibly indicating neighbor interactions (Hasan et al., 1978b); and in $Al_2O_3:Fe^{2+}$ with the direct determination of the groundstate zero field splitting (Smith et al., 1978). Especially the observation of neighbor-interactions which is often inhibited by the low concentration of neighbor pairs demonstrates the high sensitivity of tunable phonon spectroscopy.

Further, this technique provided significant evidence for the anomalous phonon dispersion in liquid $^4He$ in the range from 0 to 300 GHz (cf. Dynes and Narayanamurti, 1974, and Narayanamurti and

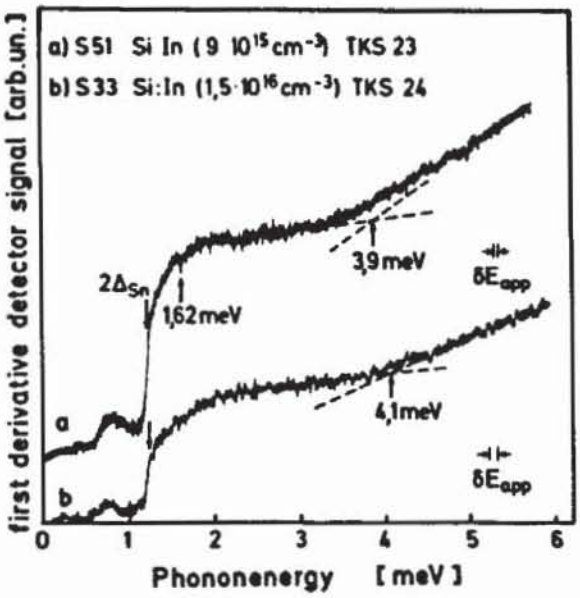


Fig. 17. Phonon absorption in In-doped Si by Jahn-Teller splitting of the In-ground state (Schenk et al., 1978).

Dynes, 1976) and also for inelastic phonon scattering (phonon energy decay) in the anomalous high phonon transmission (Kapitza-anomaly) at the boundary between solids and liquid $^4$He (Dietsche and Kinder, 1976), (Wyatt and Crisp, 1978). The present state of knowledge on the Kapitza-anomaly which has been significantly improved by phonon experiments with superconducting tunneling junctions is discussed by Kinder et al. (1980) and Wyatt (1980). Using tunnel junction detectors also the inelastic scattering of two phonon beams in liquid $^4$He resulting in frequency up-conversion was observed (Korczynskyj and Wyatt, 1978).

Phonon and quasiparticle propagation in bulk Pb single crystals was studied by Hu et al. (1977), and Narayanamurti et al., (1978), using tunnel junctions and bolometers as detector. Depending on temperature, ballistic phonon propagation and combined diffusion of phonons and quasiparticles could be observed. In similar experiments with bulk Sn single crystals and with tunnel junction phonon detection, Pannetier et al. (1977) were able to determine the temperature-dependent attenuation of ballistic phonons by quasiparticles; also the onset of strong absorption by pairbreaking could be observed.

The first dispersion reduced ballistic phonon propagation in solids was reported by Huet et al. (1972) for InSb in 100 direction. In these experiments superconducting tunneling detectors were used for frequency selection. For degenerately doped n-InSb with different carrier concentrations Huet and Maneval (1974) observed the reduction of longitudinal phonon absorption for $k_{ph} > 2q_F$; i.e. the condition of the Kohn anomaly, using a Sn-I-Sn detector for frequency selection. In addition, the detector was magnetically tuned, (cf. Narayanamurti and Dynes, 1971). Recently also resonant phonon absorption by a superlattice of GaAs/AlGaAs could be demonstrated by Narayanamurti et al. (1979), using the tunnel junction technique. Also other applications in the study of phonon emission and propagation in semiconductors are promising (cf. Reupert et al., 1976; Narayanamurti, 1980).

In addition to phonon propagation in single crystals with dimensions in the mm and cm range also the transverse or longitudinal phonon velocities and scattering mean free path values in thin films have been determined. This was possible by observing standing wave film-thickness resonances (Kinder, 1972) in granular Al-films indicating that the sound velocities do not deviate from the bulk values but that the phonon mean free path is reduced. Additional information by measuring the effective recombination lifetime of quasiparticles in these films indicates an increased electron - phonon interaction enhancement. Other film resonance studies (Forkel, 1977) also indicate an unchanged bulk phonon velocity up to 800 GHz in thin Al-films. Phonon mean free path determinations by direct attenuation in thin Al- and Cu-films have been reported by Long (1973). A very recent measurement of phonon resonances in amorphous Ga-films (Dietsche et al., 1980) showed a reduction of the transverse sound velocity by a factor of 2.7, as compared to the crystalline phase, indicating the breakdown of shear stiffness in amorphous Ga. Applications to inelastic phonon scattering in other amorphous systems; e.g. glasses; will be discussed in Sect. 6.

Further literature on the application of tunable spectroscopy with nonequilibrium phonons can be found by the references given in the introduction of Sect. 4.

## 5. QUANTITATIVE PHONON INTENSITY MEASUREMENTS

For the analysis of nonequilibrium phonon distributions and also for phonon spectroscopy a quantitative comparison of experiment and theory is desirable. With respect to the phonon emission and detection by tunneling junctions this has been investigated mainly with respect to phonon emission by recombination (cf. Trumpp and Eisenmenger, 1977). For tunneling junctions with equal energy gap as generator and detector the calculation of detector signal amplitudes is straightforward on the basis of a simple ballistic model (cf. Eisenmenger, 1976a).

Within the phonon energy range $2\Delta_D < \hbar\Omega = 4\Delta_D$ the absolute number of phonons absorbed in the detector is obtained by the calibration procedure described in Sect. 2.3. The number of $2\Delta_G$-phonons

$\dot{N}_{\omega G}$ created per unit time in the generator at voltages $2\Delta_G \leq eV_G < 4\Delta_G$ is given by

$$\dot{N}_{\omega G} = \frac{I_G}{e}$$

with $I_G$ the single particle tunneling current, since each tunnel electron corresponds to the breaking up of one Cooper-pair which in recombination delivers one phonon of $2\Delta_G$.

From this generation rate the number of phonons reaching the detector and absorbed by pairbreaking are given (cf. Trumpp and Eisenmenger, 1977) by

$$\dot{N}_{\omega d} = \frac{I_G}{e} \cdot A_d \cdot \frac{1}{\pi r^2_{GD}} \cdot \overline{(f \cdot \Gamma \cdot p)}^{\sigma,\phi_D}_{\sigma,\phi}$$

with $A_d$ = detector area; $r_{GD}$ = distance generator - detector; f = phonon focussing factor; $\Gamma$ = phonon transmission probability at the substrate - detector interface; p = pairbreaking absorption probability of phonons in the detector, $\sigma$ = phonon mode, $\phi$ = angles of ballistic propagation directions between generator and detector areas.

This equation applies to the weighted average of all contributing phonon modes and directions. For time of flight pulse measurements corresponding equations hold separately for each mode where also the different phonon mode contributions in the escape from the generator into the substrate have to be regarded. It is assumed that all phonons created in the generator escape into the substrate (vacuum condition at the free generator surface). Phonon decay by anharmonic processes in the generator or by scattering of excited quasiparticles is neglected. Experimental support for the validity of this assumption is given by the measurement of $\tau_{eff}$ in tin junctions on Si as function of film-thickness and temperature (Eisenmenger et al., 1977). In these measurements $\tau_{eff}$ agrees within 15 % with the calculation on the basis of acoustic phonon escape into the substrate.The experimental result, see Fig. 18, shows a linear increase of $\tau_{eff}$ with film-thickness in accord with the calculated slope, instead bulk processes would lead to a saturation of $\tau_{eff}$ with increasing film-thickness.

High phonon trapping within the generator establishes a detailed balance between longitudinal and transverse phonon densities according to the phonon density of states. The ratio of longitudinal and transverse phonons emitted into the substrate is determined by the phonon occupation in the film and the mode-dependent acoustic boundary transmission (Weis, 1969). The strong anisotropy of the ballistic energy flux by phonon focussing (Taylor et al., 1969;

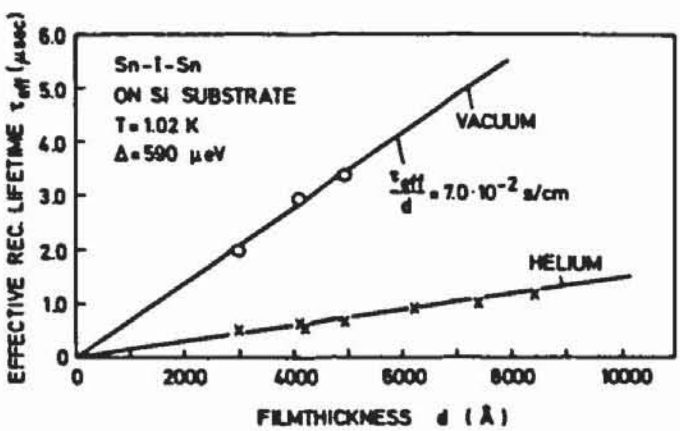


Fig. 18. Effective quasiparticle recombination time $\tau_{eff}$ in Sn-I-Sn tunneling junctions of different thickness. The theoretical film thickness dependence (vacuum) results in $\tau_{eff}/d = 6.8 \cdot 10^{-2}$ sec (T = 1.02° K). Eisenmenger et al., 1977)

Rösch and Weis, 1976) requires careful integration over all energy propagation angles between generator and detector in evaluating the average phonon focussing factor f. With respect to the transmission factors Γ this is less critical, these factors being close to unity for normal incidence at the Si - Sn boundary. The same holds for the ratio of absorbed and incident phonons. For the Sn-detector filmthickness of at least 2000 Å, taking account of the reflection at the vacuum boundary, the phonon mean free path for 2Δ-phonons of the order of 1000 Å for longitudinal and transverse phonons is short compared to the total propagation path in the film of 4000 Å. The detector has been calibrated according to the procedure outlined in Sect. 2.3. It is important to note that the density of electronic states N(0) at the Fermi-level entering the detector sensitivity must be directly taken from experimental electronic specific heat data. This has been verified by independent measurements of $\tau_{eff}$ under pulsed and stationary conditions (cf. Epperlein et al., 1978, and Epperlein and Eisenmenger, 1979). Comparing with the experiment, the observed signal amplitude ranges from 10 % to 15 % of the calculated value for Sn-junctions and generators on Si-substrates. Similar small values are obtained for Sn-junctions on $Al_2O_3$ and for various propagation directions in Si, see Fig. 19 (Trumpp and Eisenmenger, 1977).

Possible reasons for this discrepancy are: limited phonon emission into the substrate by inelastic volume loss in the Sn-film, boundary decay at the Sn-Si-interface, phonon decay in the bulk of the substrate, or anomalous phonon reflection and phonon decay at the detector - substrate boundary. Since only limited information for distinguishing between these possibilities is available from ballistic propagation, other independent experiments are needed.

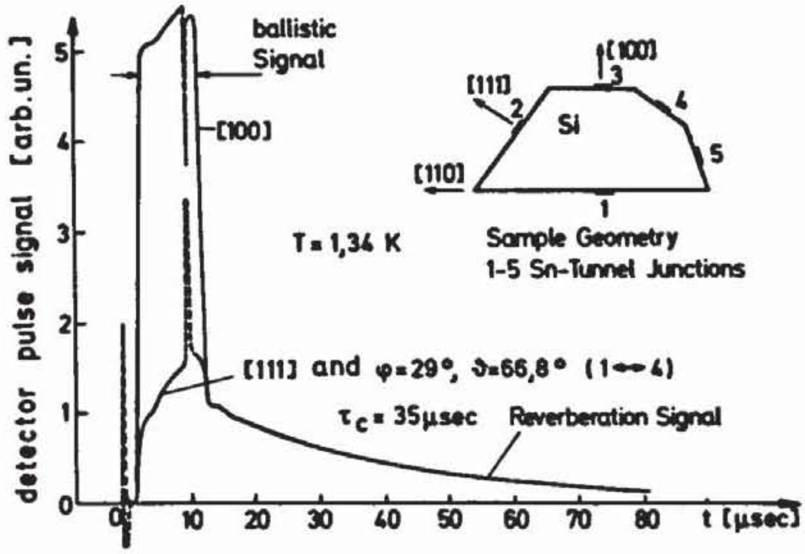


Fig. 19. Detector signal amplitudes for different propagation directions of $2\Delta_{Sn}$ phonons in Si (Sn-I-Sn generator and detector). The reverberation signal amplitude does not depend on the propagation direction in contrast to the ballistic signal. (Trumpp and Eisenmenger, 1977).

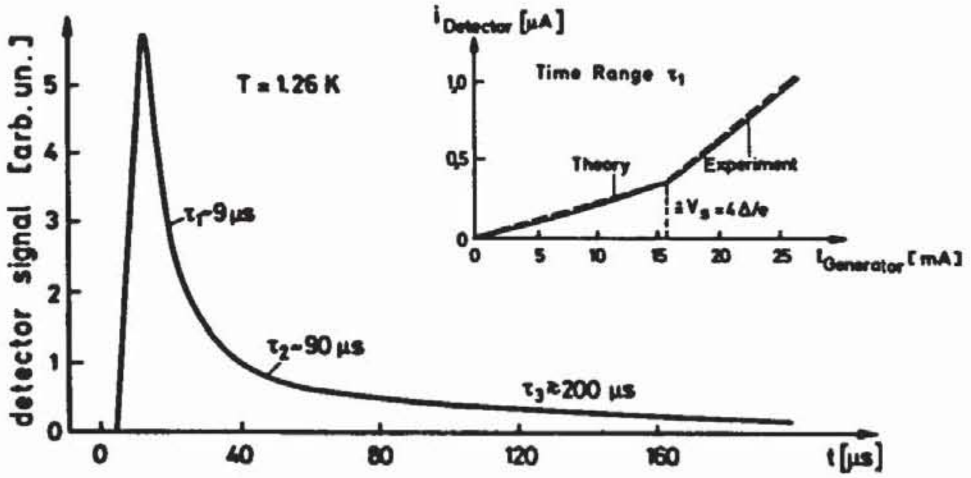


Fig. 20. Frequency check of the reverberation signal by measurement of the detector signal - generator current dependence in the range $\tau_1$. The characteristic bend at eV = $4\Delta$ (insert) is significant for $2\Delta$ phonon reverberation.

One of these is the study of 2Δ-phonon lifetime or reverberation lifetime in the substrate crystal. Fig. 19 shows a slow exponential decay of the detector signal following the ballistic response. Time constant and amplitude agree for different generator positions in contrast to the ballistic signal amplitudes which are strongly influenced by angle-dependent phonon focussing. Measurements of the signal amplitude, as function of the generator voltage, see Fig. 20, show the change of slope at $eV_G = 4\Delta$, indicating that the signal is caused by 2Δ-phonons "reverberating" in the crystal and not by "heat". Experimentally the reciprocal reverberation time depends linearly on the ratio of the crystal surface area covered by the tin film and the crystal volume, see Fig. 21. A simple analysis of the slope of this dependence shows that the phonon absorption coefficient of the Sn-covered area amounts to 87 %. Neglecting boundary reflection, this can be attributed to losses in phonon transmission into the film and after phonon trapping in Sn, losses in backradiation into the substrate; i.e. only 13 % of 2Δ-phonons incident on the film-covered area are backradiated into the Si-crystal after twice traversing the Si - Sn interface. Assuming the absence of bulk losses in the Sn-film as consistent with the $\tau_{eff}$ measurements, this can be interpreted as phonon loss or decay in the crystal-superconductor boundary. Since in ballistic experiments phonons also traverse this boundary twice, the results of both measurements are consistent. A similar finding for Al- and In-films on $Al_2O_3$; i.e. "surface detuning" of phonons; has been reported by Day (1972). As

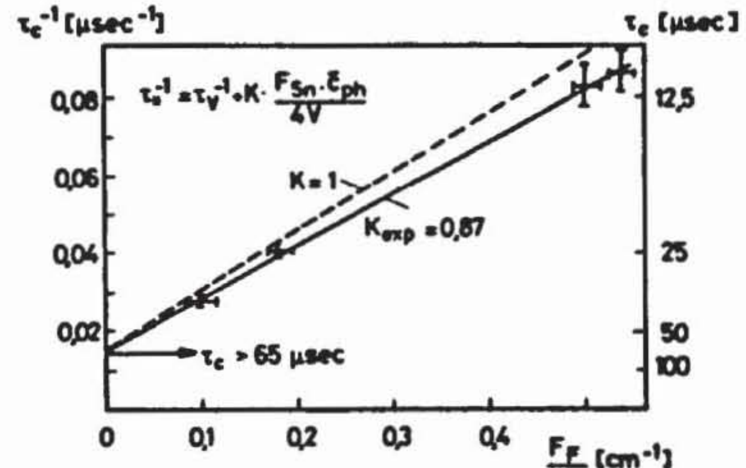


Fig. 21. Dependence of the 2Δ phonon reverberation decay rate $\tau^{-1}$ on the Sn coverage surface to volume ratio. The slope K = 1 corresponds to total phonon absorption. (Trumpp and Eisenmenger, 1977).

an additional result of the reverberation measurement of Fig. 21, the extrapolation to vanishing coverages with Sn indicate a phonon lifetime of 65 μs corresponding to a mean free path (dominant transverse phonons) of 25 cm. This indicates that inelastic substrate losses in ballistic experiments and also phonon decay (inelastic scattering) at the uncovered surface are negligible. Since weak residual inelastic scattering at the free crystal surface and also mode conversion to longitudinal phonons with high spontaneous decay rate may contribute to the observed lifetime of 65 μs, the free propagation transverse phonon lifetime may even be longer.

The nature of the interface phonon decay processes is presently unknown but may be related to the nonideal structure of the film-substrate-boundary caused by impurity atoms or other interface defects. These depend on the methods of surface and tunneling junction preparation. In principle, a comparison with the Kapitza anomaly; i.e. the enhanced phonon escape rate at the boundary between solid and liquid $^4$He is possible. In the latter case, presumably anharmonic processes by the imperfect interface result in enhanced phonon transmission, whereas at the solid-superconductor boundary anharmonic processes result in a decay of high energy phonons.

Phenomenologically the phonon coupling at the insulator - superconductor boundary can be described by an elastic coupling (spring model) parallel to a dissipative or inelastic (dashpot) element, assuming that the interface width is small compared to the phonon wavelength. The elastic contribution represents the binding potential, whereas inelastic processes are possible by multilevel transitions caused by dangling bonds, weakly bound interfacial atoms or other imperfection states. The elastic and dissipative coupling contributions in principle can be determined by a measurement of the elastic phonon transmission and reflection at the interface. For the determination of the elastic reflection and scattering properties the reverberative method has the advantage of simple evaluation. Since it averages over all propagation directions, phonon focussing does not enter, also a quantitative treatment of the phonon generator and detector properties is not necessary. Finally, taking account of the boundary losses, as obtained in reverberation, also the ballistic signal amplitudes are in better agreement with the calculation. This verifies the validity of the theoretical model for ballistic 2Δ-phonon emission and detection if anomalous inelastic and elastic boundary processes are taken into account. If bulk phonon decay in the generator, as for example by magnetic flux trapping or quasiparticle absorption, reduces the phonon signal amplitude, this influence can be experimentally controlled by the measurement of $\tau_{eff}$ and the linearity of the signal increase with generator current in the voltage range $0 < eV_G < 4\Delta_G$.

It should be further noted that the reverberation method can be also used for the absolute measurement of the total 2Δ-phonon population in the substrate crystal. For evaluation of the detector signal amplitude it can be either assumed that 2Δ-phonons incident from all directions in the crystal are contributing, or that the quasiparticle and phonon distribution in the detector is in detailed balance equilibrium ($\tau_{eff} << \tau_{crystal}$) with the 2Δ-phonon population of the substrate. In the first case the evaluation results in boundary losses of the same magnitude as determined ballistically, and from the reverberation time constant. In the second case somewhat higher boundary losses were found. This indicates that the assumption of a detailed balance between 2Δ-phonons in the crystal and the detector may be wrong, as interface loss processes represent an irreversible 2Δ-phonon drain.

More specific information on anomalous interface properties can be obtained by phonon reflection and transmission measurements (Marx et al., 1978), as have been successful in studies of the Kapitza-anomaly at the solid He-boundary (cf. Kinder et al., 1980; Wyatt, 1980).

The results of such measurements will be useful for improved crystal surface- and metal film preparation techniques.

It is to be noted that in some interface systems, especially metallic heaters evaporated on insulator crystals, no deviations from the acoustic mismatch model have been observed (Weis, 1969; Weis, 1972; Martinon and Weis, 1979). Also $\tau_{eff}$ measurements for Sn-films on silicon show agreement with the acoustic model (Eisenmenger et al., 1977) within 15%. The influence of the observed boundary losses on phonon reflection, therefore, should be comparatively small. In phonon scattering experiments with amorphous layers (Lang et al., 1980) and epitaxial layers (Narayanamurti et al., 1979) no indication of phonon boundary loss was found. This indicates the possibility of avoiding boundary losses by ideal interface conditions.

Experiments with Al-recombination phonons (Trumpp and Eisenmenger, 1977; Welte and Eisenmenger, 1980) showed reduced interface losses as compared to 2Δ-Sn-phonons, indicating a significant but smooth frequency dependence.

Also low energy decay phonons, resulting from interface losses, have been observed (Trumpp and Eisenmenger, 1977; Korczynskji and Wyatt, 1978). With respect to the spectral distribution of phonons resulting from inelastic boundary decay, a high probability with half the original frequency is to be expected from the maximum of available lattice phonon states in a three-phonon interaction. This does not apply to localized boundary states, where a continuous phonon energy distribution or a maximum at low frequencies by multiple decay steps appears possible.

Quantitative evaluation of phonon-transmission experiments have been also reported for relaxation phonons (Welte et al., 1972; Welte, 1976; Forkel, 1977; Welte and Eisenmenger; 1980). The treatment of phonon generation is less simple than in recombination, since primary decay processes and phonon escape are influenced by quasiparticle and phonon scattering in the generator film (see Sect. 3). In addition, it must be expected that this is strongly influenced by the microcrystalline film structure. Model calculations, therefore, remain of approximate nature. The comparison with experimental data resulted in order of magnitude agreement with phonon signal amplitudes within 10 % - 40 % of the calculated values, indicating again phonon interface decay or increased reflection.

An interesting possibility is the observation of changes in the relative amplitude for longitudinal and transverse phonons at the onset of phonon reabsorption by pairbreaking in the generator (Long, 1972; Eisenmenger, 1976a; Welte and Eisenmenger, 1980). Such changes reflect the difference between the relative phonon population by primary relaxation decay in comparison to the equilibrium distribution established by the detailed balance in the case of strong $2\Delta$-phonon trapping. From observing the relative transverse and longitudinal signal amplitude ratios below and above the generator voltage $eV_G = 4\Delta_G$, in principle conclusions about the strength of the longitudinal and transverse electron - phonon interaction in relaxation decay can be drawn. Such changes of the amplitude ratio are reported by Welte and Eisenmenger (1980). In the quantitative interpretations of these measurements mode conversion of relaxation phonons by elastic bulk and boundary scattering processes, as well as the mode dependence of boundary phonon decay, have to be taken into account.

As a general result it is observed, that primary relaxation decay leads to a dominant transverse phonon contribution caused by the nonspherical Fermi-surface of real metals and the higher densities of transverse phonon states.

Although quantitative evaluation of phonon-transmission experiments are roughly in agreement with simplified model calculations, more information is needed especially with respect to nonideal interface and film-structure properties.

## 6. EXPERIMENTAL PROBING OF THE QUASIPARTICLE DISTRIBUTION AND APPLICATION TO PHONON SPECTROSCOPY

Tunneling measurements with junctions composed of superconducting films with different energy gaps can be used for an analysis of the energy distribution of excited quasiparticles (Miller and Dayem, 1967; Chang and Scalapino, 1976). The principle of this method, which has been applied to the study of nonequilibrium distributions (Kaplan et al., 1977; Willemsen and Gray, 1978) in superconducting films, is depicted in Fig. 22, showing a three layer Al-I-Al-I-Pb:Bi sandwich tunneling structure. The first and second films are used for quasiparticle injection producing the nonequilibrium distribution. The quasiparticle distribution in the middle film is probed by the third film with the higher energy gap. In varying the voltage between the middle (second) and the third film the tunneling current is mainly determined by excited quasiparticles tunneling into the unoccupied singularity of the density of states of the third film. In the corresponding experiment, Kaplan et al. (1977) were able to analyze the nonequilibrium quasiparticle distribution by weak tunnel injection in Al and to compare their results with theory. Excellent agreement was found including clear evidence for branch imbalance. Recently, the same method has been successfully used to study the quasiparticle distribution (Willemsen and Gray, 1978) in Al-films under high quasiparticle injection. (See Gray, 1980; this volume, Chapter 5). It has been demonstrated that the approximate description of the nonequilibrium distribution by a $\mu^*$ or $T^*$ model (Owen and Scalapino, 1972; Parker, 1975) depends characteristically on the strength of injection.

As demonstrated by Dietsche (1978), it is also possible to use this method for the analysis of quasiparticle distributions produced by phonon-induced pairbreaking. In this case the first film is not necessary and the tunneling junction, consisting of Al-I-Sn or Al-I-Pb:Bi, is directly used as phonon detector. Phonons absorbed by pairbreaking in the Al-film produce a quasiparticle distribution with an upper energy limit at $E = \hbar\Omega - \Delta_{Al}$ ($\hbar\Omega$ = energy of incident

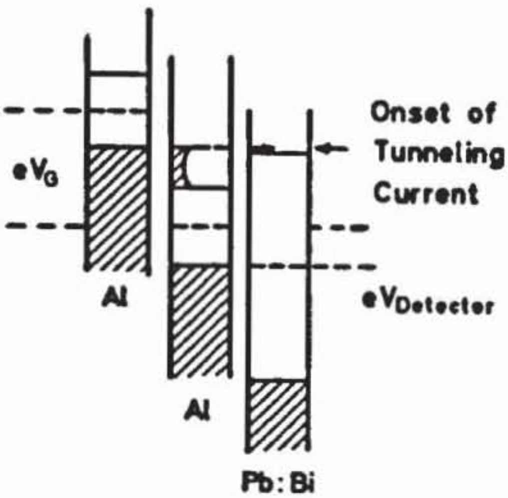


Fig. 22. Direct probing of the quasiparticle distribution in a three layer tunneling sample.

phonons). This can be analyzed in probing the quasiparticle distribution with the higher energy gap film by modulation technique and bias voltage tuning. Using such hetero-junction detectors, it is possible to determine different energies of incident phonons.

This new technique of tunable phonon detection has been successfully applied to the analysis of phonon spectra emitted from Sn-generators (Dietsche, 1978; Lang et al., 1980; Koblinger and Forkel, 1980) and, in addition, to the investigation of inelastic phonon scattering in thin glass films (Dietsche and Kinder, 1979; Lang et al., 1980). By these measurements it was possible to determine the frequency dependence of the phonon mean free path and to give evidence that the dominant scattering process is inelastic.

With respect to nonequilibrium phonons, the generator frequency analysis verified the extreme sharpness of the relaxation phonon edge at $\hbar\Omega = eV_G - 2\Delta_G$, whereas the lower cut-off energy of the recombination spectrum at $\hbar\Omega = 2\Delta_G$ in earlier experiments showed a significant contribution of spectral diffusion extending to energies below $2\Delta$. In later experiments (Koblinger and Forkel, 1980; Lang et al., 1980) less spectral diffusion was observed. A possible origin of spectral diffusion is magnetic flux trapped in the generator junction which leads to gap smearing and in-gap quasiparticle states which participate in phonon reabsorption and emission thus shifting the $2\Delta$-phonon spectrum to lower energies.

Tunable phonon detection, using hetero-tunneling junctions or phonon spectroscopy with stress-tunable absorption lines in the substrate, are very promising for investigations of inelastic phonon scattering in solid systems, as e.g. for phonon decay at solid - solid and solid - liquid boundaries.

## 7. RESULTS AND OPEN QUESTIONS

Nonequilibrium phonon experiments with superconducting film tunneling junctions have demonstrated that phonon spectra produced by nonequilibrium quasiparticle distributions are well in accord with the predictions of theory. This is also demonstrated by the successful application of superconducting tunneling junctions for phonon absorption spectroscopy and recently also for phonon emission spectroscopy.

Open questions are related to the detailed description of the separate contributions of longitudinal and transverse phonon emission and reabsorption (i.e. transverse and longitudinal phonon mean free path data by pairbreaking, quasiparticle scattering and volume decay) in the superconducting films, the influence of branch imbalance, and the properties of film substrate boundaries in the inelastic interface decay of phonons. In addition, more information is needed on quasiparticle and phonon distributions under different - for instance high injection - conditions.

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