

of time constants from an activated process, in this case the *diffusion* of vacancies, and a $1/f$ spectrum. The temperature dependence of the magnitude of the noise, on the other hand, could arise from the creation of vacancies, the magnitude of the noise being proportional to the *number* of diffusing vacancies. In bulk metals, the activation energy E_v/R_B for creation of a vacancy or interstitial is roughly 12 000 K. Over the range of temperatures in Fig. 3, the noise also appears activated with $E_g/k_B \cong 2000$ K for Ag. But vacancies are created preferentially at surfaces and deformations (e.g., at grain boundaries) and so a decrease in the bulk activation energy by roughly a factor of 6 is not unrealistic for films. In addition, E_g becomes somewhat larger as the sample is annealed, consistent with our expectations that a thicker annealed film should be somewhat closer to bulk. The speculation, then, is that creation of vacancies could explain the temperature dependence of the noise (at low temperatures), while diffusion of vacancies characterized by a distribution of activation energies could account for the $1/f$ nature of the frequency spectrum. Unfortunately, this analysis does not relate directly to perhaps the most interesting feature of the data, the peak at high temperatures.

In conclusion, the rapid temperature dependence measured in both Ag and Cu films provides a stringent test for models of $1/f$ noise. A model based on the two-step process of creation and diffusion of vacancies can provide rapid T depen-

dence, but remains quite speculative. The temperature-fluctuation model of VC, on the other hand, has various appealing features, but in its present form, at least, fails in its prediction for the temperature dependence of the noise. Experiments in progress on the temperature dependence of $1/f$ noise in other metals hopefully will help clarify the situation.

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¹A. W. Hull and N. H. Williams, *Phys. Rev.* **25**, 173 (1925).

²F. N. Hooge, *Physica (Utrecht)* **38B**, 14 (1976).

³P. Dutta, J. W. Eberhard, and P. M. Horn, *Solid State Commun.* **21**, 679 (1977).

⁴J. Clarke and T. Y. Hsiang, *Phys. Rev. Lett.* **34**, 1217 (1975).

⁵F. N. Hooge and A. M. H. Hoppenbrouwers, *Physica (Utrecht)* **45**, 386 (1969).

⁶R. F. Voss and J. Clarke, *Phys. Rev. B* **13**, 556 (1976).

⁷A. L. McWhorter, in *Semiconductor Surface Physics*, edited by R. H. Kingston (Univ. of Pennsylvania, Philadelphia, 1957), p. 207.

Ballistic Propagation of Near-Gap Phonons in Bulk Superconducting Tin

B. Pannetier, D. Huet, J. Buechner, and J. P. Maneval

Groupe de Physique des Solides, Ecole Normale Supérieure et Université Paris VII, 75231 Paris 5, France

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Phonon transport in β -Sn crystals was studied by heat-pulse techniques using superconducting tunnel-junction detectors. The temperature-dependent attenuation by the quasiparticles was measured at fixed frequency (≈ 160 GHz), while, at lower temperature (0.6 K), the ballistic transmission was monitored up to the onset of pair-breaking frequencies. A new method of determining the energy gap follows, which is shown to apply to nonequilibrium states of the bulk.

The main source of phonon scattering in metals drops out below the superconducting transition temperature (T_c) as the electrons condense into the pair states. Since this fact was first discovered in ultrasonic attenuation,¹ much information about the electron-phonon interactions was gained

by extending the measurements into the gigahertz range and, in particular, the pair-breaking mechanisms could be directly observed² in the vicinity of T_c . However, the phonon energies $h\nu$ (h is the Planck constant; ν the frequency) fitting the ground-state parameter 2Δ of the BCS theory³ fall

into the thermal range, beyond the ultrasonic frequencies presently available. This Letter reports pulse transmission measurements in bulk tin ($T_c = 3.72$ K) of incoherent thermal phonons⁴ frequency-resolved by superconducting-tunnel-junction (STJ) detectors.⁵ The main objective here is to investigate the properties of the "free-propagation window" ($0 < h\nu < 2\Delta$), namely (i) the quasiparticle-limited phonon lifetimes and (ii) the pair-breaking cutoff at 2Δ . In addition, this window turns out to be a convenient way of characterizing the transient disturbance of a superconducting domain.

Our tin samples were mechanically polished slices taken from 99.999%-pure single-crystal ingots (Metals Research, Ltd.). The phonons were generated from an evaporated copper film (350 Å) heated by short electrical pulses. The fabrication of the STJ quantum detectors has been described elsewhere,⁶ except for the granular-aluminum case: The Al metal was here evaporated from an alumina boat and left for five minutes in a reduced pressure (10^{-1} Torr) of pure oxygen to form the tunnel barrier. The films thus made are characterized by an elevated T_c (≈ 2.3 K) and a critical field of several kilo-oersteds. Both the generator and the detector were insulated from the tin crystals by ~ 5000 -Å-thick SiO layers. Most of the experiments were performed at 0.6 K in the evacuated pot of a ³He refrigerator equipped for applying magnetic fields parallel to the faces of the samples.

At low temperatures, the phonons with $h\nu < 2\Delta$ propagate ballistically over several millimeters of bulk tin. Plots versus time of the detected signals corresponding to the [110] direction of the tetragonal β -Sn system (inset of Fig. 1) show pulse amplitude and resolution similar to those obtained with dielectrics or lightly doped semiconductors.⁶ In the present case, the granular-Al STJ imposes a threshold of ≈ 160 GHz for phonon detection. The three acoustic modes, identified as L (longitudinal), T_1 (displacement vector along [001]), and T_2 (displacement along [1 $\bar{1}$ 0]), are found to travel at the respective maximum velocities of 3.66, 1.94, and 1.30 km/sec, close to the corresponding velocities of coherent hyper-sonic waves, and it is noticed that no diffusive trailing accompanies the attenuation (unexpectedly stronger on the transverse modes) taking place above 1 K.

From the rapid fall of the ballistic transmission with increasing T (temperature), we wish to measure the coupling of the phonons to the quasipar-

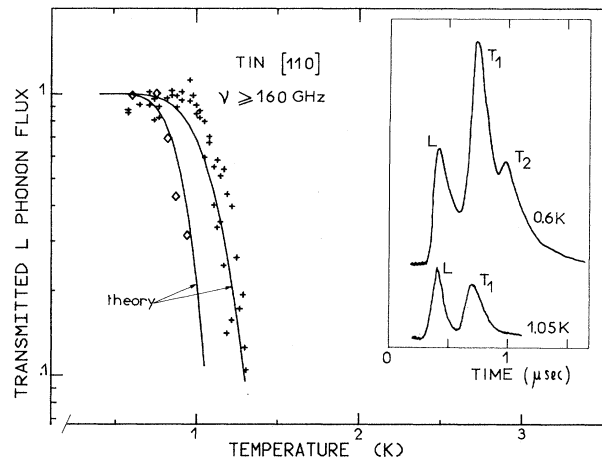


FIG. 1. Plot vs T of normalized ratios $\Delta I_{Sn}/\Delta I_{Ge}$ of L signals transmitted in tin over calibration signals in Ge (see text) for two sample lengths: 1.0 mm (crosses) and 4.0 mm (lozenges). Continuous curves are theoretical fits. Detector is a granular-Al STJ. The inset shows typical phonon pulses transmitted in tin (input flux, 1 W/cm^2 ; duration, 7×10^{-8} sec).

ticle system. The major problem encountered is the T dependence of the STJ's tunnel conductance and of their resultant sensitivity. To obviate this, we adopted the following procedure: The short-circuit detector signals⁷ ΔI_{Sn} were compared, at each temperature, to reference signals ΔI_{Ge} that had been transmitted through a dielectric slab (pure Ge). Two detectors were admitted as suitable for such comparison when their tunnel conductances varied in exact proportion (typically $\sim 10\%$) over the whole T range of interest. The experimental $\Delta I_{Sn}/\Delta I_{Ge}$ ratios thus determined for the L polarization are reported in Fig. 1, with normalization at unity performed on the low-temperature "plateau," which indeed defines the domain of existence of the free-propagation window. According to the Bardeen-Cooper-Schrieffer-Bobetic theory,⁸ the ratio of the normal [$\Lambda_N(\nu, T)$] to the superconducting [$\Lambda_S(\nu, T)$] mean free path is given (for $h\nu < 2\Delta$) by

$$\frac{\Lambda_N}{\Lambda_S} = \frac{2}{h\nu} \int_{\Delta}^{\infty} \left(1 - \frac{\Delta^2}{EE'}\right) [f(E) - f(E')] \rho(E') dE \quad (1)$$

in which formula $E' = E + h\nu$, $f(E)$ is the Fermi-Dirac distribution function, and $\rho(E)$, the relative density of states, is $E/(E^2 - \Delta^2)^{1/2}$. We have then tempted to fit the transmission function $\exp(-d/\Lambda_S)$, where d is the sample thickness, to the experimental data, assuming the emitted phonon flux is a constant vs T . For simplicity, we also

made the working approximations that (i) Λ_N has no T dependence and (ii) a single frequency component, namely the detector-threshold frequency, is important for the evaluation of Λ_S . A reasonable agreement can be obtained (see Fig. 1), for two sample lengths, with a unique choice of the two parameters $\Delta = 1.94k_B T_c$, where k_B is the Boltzmann constant, and

$$\Lambda_N = 1.2 \pm 0.3 \mu\text{m} \quad (2)$$

around 1 K for 160-GHz longitudinal phonons pointing along [110]. If one accepts the usual linear ν dependence of Λ_N at high frequencies, one finds a mean free path of 0.5 μm between pair-breaking events for ($h\nu \geq 2\Delta$) phonons.

The temperature-dependent attenuation of the T_1 and T_2 modes necessarily involves a "deformation-potential" type of interaction⁹ since pure electromagnetic coupling is negligible in the limit $ql \gg 1$ (q , the phonon wave vector, is about $5 \times 10^6 \text{ cm}^{-1}$; l , the electronic mean free path, was measured to be 0.15 mm at 4.2 K). It is therefore justified to derive Λ_N as for the L case. One thus finds $\Lambda_N \approx 0.7 \mu\text{m}$ for T_1 and $\Lambda_N \lesssim 0.4 \mu\text{m}$ for T_2 , 160-GHz phonons.

The direct absorption mechanisms involving pair breaking are the dominant ones at phonon energies exceeding the bulk pair-condensation energy $2\Delta_{\text{bulk}}$. This range was investigated in its turn by using a tin STJ detector whose threshold frequency ν_{det} was slightly displaced with a parallel magnetic field smaller than the bulk critical field H_c ($H_c \approx 260 \text{ Oe}$ at 0.6 K in our geometry). Actually, in pure metals ($ql \gg 1$), each phonon mode samples a few definite points on the Fermi surface and senses orientation-dependent values of $2\Delta_{\text{bulk}}$ (the detector is assumed not to possess this property of "anisotropy"). For that reason, a variety of situations are possible according to the crystalline orientation chosen. With the longitudinal modes propagating along [001] (direction of small gap¹⁰), the following observation is made [Fig. 2(a) and inset]: As ν_{det} is tuned down by the field, the L signal remains essentially zero until a sharp kink is reached; thereafter a considerable *net increase* of the signal occurs (note that the STJ is working at such low bias that, within experimental uncertainty, its differential conductance is constant throughout, i.e., voltage signals are proportional to phonon fluxes⁷). Such an increase of the signal with the magnetic field, contrasting with the smooth decrease that one observes if using a nontunable (granular-Al) STJ, proves that a modulation of ν_{det} , rather than an

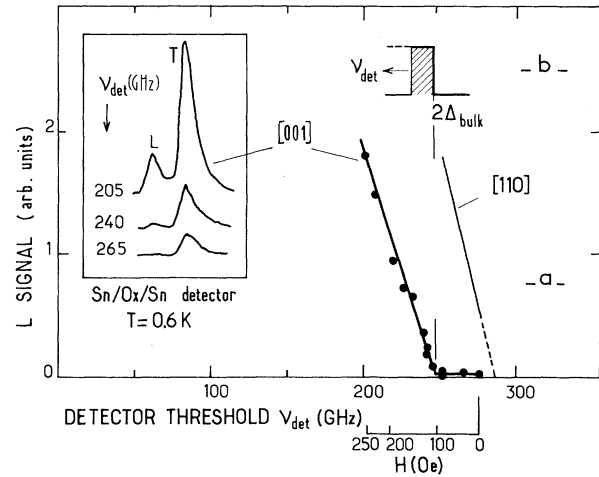


FIG. 2. Inset: quasimonochromatic phonon propagation along [001]. $T = 0.6 \text{ K}$. The threshold frequency ν_{det} of the tin STJ detector is tuned by a magnetic field H . (a) Plot of L peak amplitudes vs effective ν_{det} (solid circles); L amplitudes along [110] are also shown for comparison. (b) Frequency window defined by Eq. (3).

alteration of the bulk, is in effect. Eventually the whole heat-pulse pattern discontinuously vanishes, whatever the detector may be, at the superconducting-to-normal transition of the bulk (260 Oe). In our opinion, the residual L signal present at low fields scales the maximum amount of low-frequency phonons thermally detected by the STJ. On the other hand, the kink at the onset of the L signal rise marks the *pair-breaking cut-off* of the phonon transmissivity. Because of the very short free path associated with these processes [see Eq. (2)], what is selected out of the original heat spectrum is the restricted energy window [Fig. 2(b)]

$$h\nu_{\text{det}} \leq h\nu < 2\Delta_{\text{bulk}}. \quad (3)$$

The above leads to a direct determination of the ground-state gap parameter, as we will now show. For a superconducting tunnel junction in a magnetic field, the existence of an abrupt detection threshold and its construction from the static current-vs-voltage characteristics were specifically justified by independent chromatic-dispersion measurements.¹¹ If identified to $h\nu_{\text{det}}$ (kink value), the bulk pair-condensation energy at 0.6 K is found to be $3.2k_B T_c$ for the L modes along [001], in accordance with the result $(3.2 \pm 0.1)k_B T_c$ derived from the T dependence of the quasiparticle-induced ultrasonic attenuation.¹⁰ For other directions, the gap may be off the de-

tection range of the tin STJ: It is the case, for instance, of L phonons along [110] whose detected flux does not vanish in zero field. Instead, it extrapolates to zero [Fig. 2(a)] at a detector energy corresponding to $3.7k_B T_c$, again in agreement with Morse, Olsen, and Gavenda.¹⁰ More intricate is the behavior of the T modes propagating along [001] since it cannot be accounted for by a single gap parameter. However, the detailed interpretation of such effects has to be postponed to a later publication.

In addition to the linear transport phenomena discussed so far, our method lends itself naturally to the study of far-from-equilibrium situations. Strong irradiation with $h\nu > 2\Delta_{\text{bulk}}$ phonons should result in the formation of a "hot domain" of quasiparticles interacting with the trapped pair-breaking phonons. The local gap ($2\Delta^*$) would then stand as the effective upper limit of the free-propagation window. What is indeed observed¹² is a nonmonotonicity (vs heat-pulse power) of the STJ signals, featuring saturation and eventually complete extinction (at a few W/mm²) as $2\Delta^* = h\nu_{\text{det}}$. Similar experiments performed with different STJ's show the gradual decrease of $2\Delta^*$ with the injected power. Besides, the hot domain thus formed is recognizable on the detector as an independent signal, which bears similarities with the quasiparticle signal recently reported¹³ in lead.

In conclusion, a bulk superconductor is an essentially perfect low-pass acoustic transmission medium. Because they are strongly coupled to the Cooper pairs, phonons at the limit of the pair-breaking energy may serve to probe very small

changes of the gap, arising either from crystal-line anisotropy or from the formation of transient out-of-equilibrium domains.

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¹H. E. Bömmel, Phys. Rev. **96**, 220 (1954).

²M. P. Garfunkel, J. W. Lue, and G. E. Pike, Phys. Rev. Lett. **25**, 1649 (1970).

³J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

⁴The observation of ballistic phonons in bulk lead was already reported by V. Narayanamurti, Bull. Am. Phys. Soc. **15**, 821 (1970).

⁵W. Eisenmenger, in *Physical Acoustics*, edited by W. P. Mason (Academic, New York, 1976), Vol. XII, p. 80.

⁶D. Huet, B. Pannetier, F. R. Ladan, and J. P. Maneval, J. Phys. (Paris) **37**, 521 (1976).

⁷At fixed temperature and constant-current bias of the STJ, the short-circuit signal ΔI is defined as the peak voltage signal multiplied by the slope of the current-vs-voltage tunnel characteristics (see Ref. 5). This ΔI was verified to be independent of the voltage for a wide range of biases.

⁸V. M. Bobetic, Phys. Rev. **136**, A22 (1964).

⁹R. W. Morse, T. Olsen, and J. D. Gavenda, Phys. Rev. Lett. **3**, 15 (1959).

¹⁰D. Huet, B. Pannetier, and J. P. Maneval, in *Phonon Scattering in Solids*, edited by L. J. Challis, V. W. Rampton, and A. F. G. Wyatt (Plenum, New York, 1976), p. 283.

¹¹B. Pannetier, D. Huet, and J. P. Maneval, Bull. Am. Phys. Soc. **22**, 339 (1977).

¹²P. Hu, R. C. Dynes, V. Narayanamurti, H. Smith, and W. F. Brinkman, Phys. Rev. Lett. **38**, 361 (1977).