Transmission of high-frequency ballistic phonons in superconducting In, Sn, and Pb films

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Phonon transmission through superconducting In, Sn, and Pb films (with thicknesses of 4 and 8 μ m) is studied with a heat-pulse technique using a semiconductor Si:P detachable bolometer as the phonon detector. Below the critical temperature of the films, ballistic-phonon propagation is observed. From the dependence of the phonon transmission on the incident power and film thickness, we calculate the mean free path of phonons at the gap frequency. The phonon transmission amount changes when a magnetic field is applied and is also different if the field is parallel or perpendicular to the film. By a simple model based on the Landau laminar model for intermediate states, the critical field for the films can be calculated. Good agreement (for In and Sn) with the theoretical values is obtained.

It is well known that electron-phonon interaction plays an important role in the transport properties of metals, in normal as well as in superconducting states. In this latter case, high-energy phonons, having a frequency close to the superconducting gap, are directly involved in several phenomena, such as tunneling and nonequilibrium superconductivity. Unfortunately, the widely used ultrasound technique^{1,2} is limited to the 10^7-10^9 Hz range, far from the energy gap for the most common superconducting materials.

The heat-pulse technique extends the available frequencies to the GHz or Thz range,^{3,4} but in the past it has been rarely used to study superconducting materials,⁵⁻⁷ expecially in the presence of magnetic fields: in fact, the detectors generally used (superconducting tunnel junctions or superconducting bolometers) cannot work under these conditions. However, it should be interesting to study the propagation of high-energy phonons in superconducting materials in the presence of a magnetic field; for instance, in order to clarify the interaction mechanism between fluxons and phonons in II-type materials.⁸

In this paper we report on the preliminary study of phonon transmission through superconducting films (In, Sn, and Pb) by a heat pulse technique. To do that, we use a semiconductor Si:P bolometer able to work on magnetic fields of up to 6 T.⁹

The experimental setup is shown in Fig. 1. The source of ballistic phonons is a thin gold film (heater) of $100 \times 500 \ \mu\text{m}^2$ area, thermally evaporated on a silicon crystal disk (50 mm diam, 10 mm thickness, floating zone *n* type, with a resistivity of $1000 \ \Omega$ cm). The silicon disk is used to separate in time the longitudinal and the transverse phonons, as they have different phase velocities. The phonon pulses are detected by a detachable Si:P bolometer previously described. ^{10,11} The sensitive area is about $0.5 \times 1 \ \text{mm}^2$. On the back side of the bolometer a superconducting film of In, Sn, or Pb with thickness 4 or 8 μ m is realized, starting from materials with a purity of 99.99%. To test the critical temperature of the films, their resistance and magnetic susceptibility are measured. $T_c = 3.25 \pm 0.15 \ \text{K}$ for In, $T_c = 3.5 \pm 0.15 \ \text{K}$ for Sn, and $T_c = 7.3 \pm 0.15 \ \text{K}$ for Pb are obtained.

The bolometer is mechanically pressed onto the surface of the silicon disk opposite to the heater. The heater and the bolometer are aligned along the $\langle 100 \rangle$ direction of the Si disk. The sample is inserted in a cryogenic cryostat (working temperature about 1.5 K) and immersed in the liquid helium bath. An external coil around the dewar is used to generate a magnetic field **H** up to 1000 G, perpendicular or parallel to the film. The magnetic field at the sample position is measured by a Hall probe (Lake Shore Cryotronics Inc.).

In the following, we present first the experimental results for In and Sn films, and discuss the Pb case separately.

In Fig. 2 we show, as an example, a typical time-offlight (TOF) spectrum recorded for an In film at a temperature of 1.5 K and at zero magnetic field. The spectrum presents at 1 μ sec an inductive signal, generated by the electric pulse applied to the heater. The rise of this signal is the zero of the time-of-flight scale. The time scale is affected by an arbitrary delay of $\approx 1 \mu$ sec given by the trigger. The two peaks labeled L and T correspond to the signal contribution of longitudinal and transverse phonons, respectively. As a comparison, the horizontal bars show the expected time of flight (pulse duration 300 nsec) calculated using the phase velocity of phonons along the $\langle 100 \rangle$ direction of Si (isotropic model). Both the time of flight and the shape of the spectrum indicate





0163-1829/94/49(5)/3600(4)/\$06.00



FIG. 2. Time-of-flight spectrum of the phonons transmitted through a 4- μ m-thick In film at T=1.5 K and zero magnetic field (voltage pulse: 6 V amplitude, 300 nsec width). The horizontal bars represent the theoretical time of flight.

the ballistic behavior of the phonon transmission. Similar TOF spectra are obtained using Sn films as coupling media. TOF spectra are measured for different values of the power dissipated on the heater W_h and for two film thicknesses.

From these results, the ratio between the intensity of peak l in the 4 μ m film and in the 8 μ m film as function of W_h is obtained and is reported in Fig. 3. The ratio is about constant for $W_h > 20$ W/mm². For lower W_h values, a part of the power dissipated on the heater prefers to escape in the surrounding liquid helium, as observed by Kappus and Weis.¹²

Two sets of data are obtained with the same bolometer at the same experimental conditions. The ratio is found (for $W_h > 20$ W/mm²) to be 2.00 ± 0.05 for In and 1.75 ± 0.05 for Sn. A similar analysis on peak T yields the same results. In this case, it is necessary to take into account the fact that peaks L and T overlap. Then the shape of the peak L is fitted with a Gaussian curve and subtracted from the spectrum. These values are used to determine the phonon mean free path in our films.

The peak intensity of the bolometer signal when the

film is in the superconducting state, is given by the following proportionality:

$$S \propto \int_0^{\nu_{\max}} p(\nu) \exp[-(\alpha_s + \alpha')d] d\nu , \qquad (1)$$

where d is the thickness of the film, α_s is the absorption coefficient in superconducting state, α' is the absorption coefficient for scattering on defects, dislocations, grain boundaries, etc., p(v) is the incident phonon power density, and v_{max} is the maximum frequency of the incident phonon spectrum determined by isotope scattering in the Si disk. Strictly speaking, the attenuation coefficients α_s and α' are frequency dependent and it is necessary to analyze them in detail before applying Eq. (1).

In order to evaluate α_s , we consider that for highfrequency phonons $(h\nu\approx 2\Delta)$ two different processes involving electronic excitations contribute to the phonon attenuation in superconducting state.¹³ The first is the phonon absorption by thermally excited quasiparticles, which is effective at all temperatures and only falls to zero at T=0 K. The second one is the phonon absorption by direct breaking of Cooper pairs which is effective for phonons with energy $h\nu \ge 2\Delta(T)$. The ratio γ between attenuation coefficient for phonons in superconducting (α_s) and normal (α_n) state is given by

$$\gamma(\nu, T) = \frac{\alpha_s}{\alpha_N} = \frac{\alpha_{\rm QP} + \alpha_{\rm PB}}{\alpha_N} , \qquad (2)$$

where α_{QP} and α_{PB} are the attenuation coefficients for quasiparticles and pair-breaking mechanism, respectively.

The detailed expressions for $\alpha_{\rm QP}$ and $\alpha_{\rm PB}$ are derived in Ref. 13; from these, γ can be computed via numerical integration. γ is strongly dependent on temperature but for $t = T/T_c < 0.5$, as in our experimental conditions, it is almost a step function (Fig. 4).

The spectral power density of the phonons emitted from the heater into the crystal has a Gaussian-like shape.¹² The frequency corresponding to the maximum v^* depends, among other parameters, from the electrical power dissipated on the heater. In our experimental conditions v^* is always higher than $2\Delta(T)/h$.



FIG. 3. Ratio of the peak heights of the longitudinal phonons for two In films of thickness 4 and 8 μ m, respectively, as function of the power W_h .



FIG. 4. Ratio of phonon attenuation in superconducting and normal state as function of frequency for In and Sn at T=1.6 K. The solid line represents the "step function" approximation used.

In this case, we can approximate the function γ as

$$\gamma(\nu, T) = \begin{cases} \gamma_0 & \text{for } h\nu \le 2\Delta \\ 1 & \text{for } h\nu > 2\Delta \end{cases},$$
(3)

where γ_0 is the value of $\gamma(\nu, T)$ for $h\nu$ close to $2\Delta(T)$ (Fig. 4, solid line). At T = 1.6 K, γ_0 is 0.016 for In and 0.011 for Sn. As $\gamma_0 \ll 1$, the superconducting film acts as a "low pass" frequency filter which permits only the transmission of phonons having a frequency lower than the energy gap.⁷

Defining $v_{gap} = 2\Delta(T)/h$ as the phonon frequency corresponding to the gap $\Delta(T)$ and taking into account the temperature dependence of the energy gap, ¹⁴ we find v_{gap} equal to 250 and 270 GHz, respectively, for In and Sn at T = 1.6 K. Also α' can be considered as frequency independent because, as we have shown before, only the phonons close to the energy gap give a significant contribution to the signal.

Inserting Eqs. (3) and (2) into Eq. (1) and defining $\alpha_{\rm eff} = \alpha_N + \alpha' / \gamma_0$ after frequency integration up to $v_{\rm gap}$, we can write the peak intensity as

$$S \propto \exp(-\gamma_0 \alpha_{\text{eff}} d) . \tag{4}$$

From the ratio between the signal intensity measured for two different film thicknesses in the same experimental conditions, we can calculate α_{eff}

$$\alpha_{\rm eff} = \frac{\ln(S_{d_1}/S_{d_2})}{\gamma_0(d_2 - d_1)} , \qquad (5)$$

where d_1 and d_2 are the film thicknesses and S_{d_1} and S_{d_2} are the corresponding signal intensities.

From the observed intensity ratio the value of $\lambda_{\text{eff}} = 1/\alpha_{\text{eff}}$ can be derived for each film. λ_{eff} has to be considered as the mean free path in the normal state for phonons at the energy gap frequency. This value turns out to be 920 A for In and 800 A for Sn. The main error source in this calculation (±10%) is due to the uncertainty in the film thickness.

To our knowledge, there is not very much data available in literature for comparison. For Sn, Pannetier and Maneval⁷ measured the phonon mean free path in single crystals using phonons of frequency 150 GHz. They found that α_N is in the range $0.4-2 \mu m$, depending on the particular crystallographic directions. Rescaling their values with the usual law $(\alpha_N \propto 1/\nu)$ at our frequency 270 GHz, α_N results in the range $0.2-1 \mu m$. Our result can then be considered to be in agreement with those in Ref. 7, taking into account that in our thermally evaporated film the contribution of defects, dislocations, and grain boundaries to the phonon scattering is stronger than in a single crystal.

In order to study the influence of the magnetic field on phonon transmission, for each film of series of TOF spectra is measured by varying the magnetic field **H**.

Figure 5 shows the TOF spectra for 4 μ m In film in a perpendicular magnetic field. The top spectrum (trace a) is recorded at H=0 and the others correspond to H>0. It can be seen that the shape of the TOF spectra does not change; however, the signal intensity decreases by in-



FIG. 5. TOF spectra for phonons passing through 4- μ m-thick In film in various magnetic fields perpendicular to the film $W_h = 52 \text{ W/mm}^2$: (a) H = 0 G; (b) H = 32 G; (c) H = 87 G; (d) H = 178 G.

creasing the applied magnetic field.

With W_h fixed, we measure the intensity of the peak L as function of **H**. The experimental results for the 4 μ m In film are shown in Fig. 6 (circles), where the intensities are normalized to the **H**=0 value. When the magnetic field is perpendicular to the film, the bolometer signal decreases linearly in first approximation, and vanishes at a magnetic field **H**^{*}=175 G for the In film, and at **H**^{*}=170 G for the Sn film. The same behavior is observed for the T peak. We note that **H**^{*} is smaller than the critical field of the bulk material, which, at a temperature of 1.6 K, should be 213 G for In and 241 G for Sn.

To analyze our results, we notice that when the applied magnetic field is perpendicular to the film, it is in the intermediate state. The intermediate state is assumed to consist of a periodic arrangement of straight superconducting and normal domains (Landau nonbranching model¹⁵). The periodicity length L is the sum of the width of the superconducting and normal domains, which we denote by L_s and L_n , respectively.

Since the normal domains carry a magnetic field equal



FIG. 6. Peak height of longitudinal phonons, normalized at the H=0 value, as a function of the magnetic field for 4- μ m-thick In film. Circle: *H* perpendicular to the film. Triangle: *H* parallel to the film. ($W_h = 34$ W/mm².)

to the critical one \mathbf{H}_{c} , flux conservation requires

$$\frac{L_n}{L} = \frac{H}{H_c} = h \quad . \tag{6}$$

Then the region of the film in normal state grows linearly with \mathbf{H} . The bolometer signal in the presence of the magnetic field consists of the phonons propagating through the superconducting domains and the normal ones. It can be written as

$$S(h) \propto \frac{L_s(h)}{L} \exp(-\alpha_s d) + \frac{L_n(h)}{L} \exp(-\alpha_n d) . \quad (7)$$

In the above equation the second term is negligible for films with a thickness of 4 or 8 μ m since the mean free path in the normal state is of the order of 0.1 μ m or less.

The bolometer signal in the absence of the magnetic field is simply proportional to

$$S(0) \propto \exp(-\alpha_s d) \tag{8}$$

and the ratio S(h)/S(0) reads

$$\frac{S(h)}{S(0)} = \frac{L_s(h)}{L} = 1 - h \quad . \tag{9}$$

This relation shows that the bolometer signal should decrease linearly with the field **H** and becomes zero at the critical field. However, as stated before, we find that the signal vanishes at a value H^* which is lower than the critical field H_c in bulk material. The ratio H^*/H_c is 0.82 for In and 0.70 for Sn.

 H^* represents the effective critical magnetic field in our films. Guyon, Caroli, and Martiner¹⁶ calculated the ratio H^*/H_c in the framework of the Landau laminar model of the intermediate state. Applying their results to our case, we find H^*/H_c equal to 0.70 and 0.67 for In and Sn, respectively. On the other hand, Onori and Rogani¹⁷ obtained, with a microwave sensitive technique, 0.75, for Sn, very close to our value. This agreement is indicative that our phonon technique is a sensitive tool and can represent an alternative method to probe the magnetic effects on the superconducting state of films.

When the magnetic field is parallel to the superconducting film (triangles in Fig. 6), the drop of the intensity is more pronounced with respect to the perpendicular case. We observe a similar trend in In, Sn, and Pb films

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of both thicknesses. This behavior is surprising. In fact, the demagnetization factor for H parallel to the film is zero and the field penetration into the film should be zero up to the critical field. The signal should then be constant up to H_c and drop to zero for $H > H_c$. The experimental results, however, show a monothonic decrease of the intensity, also for H close to zero. To explain this behavior, we assume that the phonon transmission is strongly attenuated in the most external region of the film, in which the superconducting current flows to counter the field penetration.

Finally, we briefly discuss the case of Pb films. The experimental results are very similar to those reported before for In and Sn. The ratio between the intensities of the L peak for the 4 and 8 μ m films is 1.80 \pm 0.05, close to the values for the others; moreover, the bolometer signal decreases in perpendicular magnetic field, and at $H^* = 230$ G it vanishes.

However, the same analysis used before yields inconsistent results if applied to the Pb film. For example, λ_{eff} results to be about 15 A, too small a value as compared with the result of Narayanamurti¹⁸ (≈ 100 A for 200 GHz phonons in normal state). This difference is related to the morphology of the Pb film. In fact, the magnetic susceptibility data show that the transition is spread off in temperature with a large dissipative component. This observation indicates a granular composition of the film and in these conditions the simple models used cannot be further applied: in fact, the phonon absorption can be important in intergranular region.

In conclusion, we have reported experimental results on phonon transmission through superconducting films, also in presence of magnetic field. These results can be interpreted by simple models based on the BCS and Landau theory of the intermediate state. The agreement with the proposed models is satisfactory for In and Sn films and the values of phonon mean free path λ_{eff} and critical magnetic field **H**^{*} can be derived by using the heat pulse technique.

The assistance of the Technical Service of CFSBT-CNR has been necessary for the success of the experiment. We are also grateful to Dr. P. Fabbricatore for the susceptibility measurements on our samples.

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