# ON T<sub>c</sub> ANISOTROPY OF THE SUPERCONDUCTING TRANSITION

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Results of a precision study of longitudinal ultrasonic attentuation in pure tin single crystals having different orientations are presented. The pulse technique was used for measurements at temperatures between 4.2° and 1.3°K, at frequencies from 130 to 215 MHz, while varying the experimental conditions. It was found that, with  $\pm 0.0002^{\circ}$ K accuracy, the onset of the decrease in the coefficients of electronic ultrasonic attentuation (for  $\hbar\omega \ll 2\Delta$ ) is independent of the crystallographic orientation at the transition of the metal from the normal to the superconducting state.

### INTRODUCTION

IN connection with studies in the neighborhoods of second order phase transition points it is interesting to obtain precision measurements of ultrasonic attenuation coefficients at the transitions of metals from the normal to the superconducting state. Interest has been increased by the fact that in a study of longitudinal ultrasonic attenuation at 70 MHz<sup>[1]</sup> in a tin single crystal singular behavior of the temperature dependence of the attenuation coefficient was observed close to T<sub>c</sub>. The investigators interpreted their results as "... anisotropy of the transition temperature  $T_c$ , which as a sample underwent a transition from the normal to the superconducting state, was 0.004°K higher in all measurements for the  $C_2$  direction than for the  $C_4$  direction" (p. 11 of<sup>[1]</sup>). Although this interpretation of the measurements conflicts with the theory of phase transitions and with the microscopic theory of anisotropic superconductors,<sup>[2]</sup> the nature of the anomalous behavior exhibited by the ultrasonic attenuation coefficient remained obscure. As the same authors correctly stated, the effect can be attributed neither to differences in the heat transfer from the sample to the helium bath, because ultrasonic attenuation was measured in both crystallographic directions simultaneously, nor to the influence of the earth's magnetic field, because the effect did not disappear when the field was compensated.

We note also that neither the presence of thermodynamic fluctuations in the metal near T<sub>c</sub>, nor the possibility of phonon-phonon interactions in the sample can account for the described anomaly of the attenuation coefficient. Indeed, the fluctuation correction in the pure metal is commensurable with ordinary attenuation only for  $\Delta T_c \sim 10^{-13}$  K,<sup>[3]</sup> while nonlinear acoustics shows<sup>[4]</sup> that phonon-phonon interactions do not occur in the case of two mutually perpendicular longitudinal waves. Also, impurities (or their nonuniform distribution) in a sample cannot be the cause of the observed anomaly. It has recently been shown<sup>[5]</sup> that even a precision study of the temperature dependence exhibited by the ultrasonic attentuation at 50-250MHz in pure tin single crystals with 0.01-0.02% impurities does not reveal additional anomalies associated with quantum processes of phonon decay to a pair of

quasiparticles in a superconductor. In addition, an impurity-induced lowering of  $T_{\rm C}$  by  $0.004^{\circ} {\rm K}^{[6]}$  occurs at impurity concentrations >0.01%; the electronic ultrasonic attenuation coefficient  $a_n^{\rm m}$  in the normal state is then reduced by an order of magnitude.<sup>[7]</sup> However, the values of  $a_n^{\rm m}$  given in<sup>[1]</sup> showed that the sample was pure. (The values of  $a_n^{\rm m}$  for pure tin will be elucidated below.)

#### STATEMENT OF THE PROBLEM AND CHARACTER-ISTICS OF THE MEASUREMENT TECHNIQUE

Since none of the theoretically possible causes can account for the experimental results reported in<sup>[1]</sup>, we are left with the hypothesis that the given effect is of instrumental origin. This hypothesis is suggested by the anomalous behavior of the coefficient of electronic ultrasonic attenuation in an actual anisotropic superconductor. For  $kl \gg 1$  (where k is the acoustic wave vector and l is the electron mean free path) both the magnitude of the electronic ultrasonic attenuation coefficient  $a_n^{(8,9]}$  and the temperature dependence of the corresponding coefficient  $a_S^*$  in the superconducting state (specifically, the steepness near  $T_C$ ) are functions of the orientation of k with respect to the crystallographic axes (see<sup>[10,11]</sup>, for example).

Our measurements, to be described here, were undertaken for the purpose of testing the given hypothesis.<sup>1)</sup> An enhanced accuracy of the measurements was achieved in two ways. First, in our experiments the ultrasonic frequency was increased by a factor of 2 to 3; since the electronic attenuation is linearly dependent on the frequency<sup>[8,9]</sup> the measured quantities were increased similarly. Secondly, in contrast with the use of precalibrated detectors in<sup>[1]</sup>, at each temperature we used a comparison pulse whose amplitude was matched with that of the investigated signal by means of a continuous attenuator. We were registering relative measurements; also, the comparison pulse and the test signal were being received through the same channel. Therefore any variations in the operation of

<sup>&</sup>lt;sup>1)</sup> T. N. Marchenko, a student at Kharkov State University, assisted us with the measurements.

the channel affected both signals in an identical manner and introduced no errors into the measurements. (A detailed description of the pulse technique apparatus has been published in<sup>[12]</sup>.)

Our samples were tin single crystals (with  $<10^{-4}\%$  impurities) that had been grown by the Obreimov-Shubnikov method. The rf pulses were transformed into ultrasound (and vice versa) by means of thin plates of X-cut monocrystalline quartz in which longitudinal vibrations were induced by the radio pulses. Ultrasound was transmitted from the quartz to the sample (and vice versa) through a thin layer of a vacuumcooked mixture of rubber and vaseline or of organosilicon liquid (GKZh-94).<sup>[13]</sup>

During the course of the measurements the sample was in direct contact with liquid helium, and the earth's magnetic field was carefully compensated. (Since for tin near  $T_{C}$  we have  $dH_{C}/dT\approx$  -150 Oe/deg, the magnitude  $\approx$  0.5 Oe of the earth's magnetic field could induce an appreciable shift of the superconducting transition point). Temperature measurements (on the  $T_{58}$  scale) were based on the pressure P of saturated helium vapor measured with a mercury manometer and a KM-8 cathetometer, and taking into account the temperature change that resulted from the hydrostatic pressure of the liquid helium column above the sample.<sup>[14]</sup> The large value of the derivative, dP/dT $\approx$  510 Torr/deg, and the extreme steepness of the  $\alpha_{\rm S}^*$ curves near the superconducting transition point of tin ensured maximum accuracy of the measurements  $(\pm 0.0002^{\circ} \text{K})$  in this temperature region.

#### RESULTS AND DISCUSSION

Since our investigation was of a basic character we performed three kinds of experiments in the ultrasonic range f = 130-215 MHz at temperatures  $4.2-1.3^{\circ}$ K (as in<sup>[1]</sup>, at the transition of tin from the normal to the superconducting state):

A. Separate measurements of the temperature dependence exhibited by the electronic attenuation coefficients of longitudinal ultrasound, using two ultrasonic frequencies such that the electronic attenuation coefficients  $\alpha_n^*$  in the normal state coincided for the orientations  $\mathbf{k} \parallel \mathbf{C}_2$  and  $\mathbf{k} \parallel \mathbf{C}_4$ . These ultrasonic frequencies for tin are easily determined from the known dependences of  $\alpha_n^*$  on the frequency f.<sup>[8,9]</sup> Figure 1 shows the results obtained near T<sub>c</sub> for one of the tin single crystals (with the dimensions 0.6355 cm along the C<sub>2</sub> axis and 0.8415 cm along the C<sub>4</sub> axis) at f = 174 MHz for  $\mathbf{k} \parallel \mathbf{C}_2$  ( $\alpha_n^* = 51.8$  db/sample) and





f = 215 MHz for  $\mathbf{k} \parallel \mathbf{C_4}$  ( $\alpha_n^*$  = 51.3 db/sample);  $\alpha_s^*$  and  $\alpha_n^*$  are the total values of the electronic attenuation coefficients in the superconducting and normal states of tin. (Details concerning the extrapolation to 0°K of the  $\alpha(T)$  curves for 1.3 < T°K < 4.2 are given in<sup>[10]</sup>.)

B. The temperature dependences of the attenuation coefficients measured separately for  $k \parallel C_2$  and  $k \parallel C_4$  at an identical frequency. The results for  $\alpha_S^*/\alpha_n^*$  near  $T_c$  are similar to those shown in Fig. 1.

C. Simultaneous measurements of the temperature dependences, at an identical frequency, for both orientations of the tin sample. The apparatus described in<sup>[12]</sup> was used for the first two kinds of experiments, but several changes were required for this last kind of measurements. A block diagram of the modified pulse circuit is shown in Fig. 2; the circuit elements are similar to those used previously. With two transmitting and two receiving tracks and an S1-15 oscillograph (with an S1-15/3 preamplifier designed to study two processes independently) we were able to obtain simultaneous measurements at each temperature. The sample was mounted in a specially designed holder that permitted the affixing of quartz plates on four sides of the sample.

Figure 3 shows the data obtained on the simultaneous attenuation near  $T_C$  of longitudinal ultrasound at 133 MHz along both directions of a tin single crystal. The difference in an individual experiment with regard to the onset of reduction in the electronic attenuation coefficients along k ||  $C_2$  and k ||  $C_4$  was at most 0.004°K, which lies within our experimental error limits.

Figures 1 and 3 show that detailed measurements of electronic ultrasonic attenuation along the different crystallographic directions in an extremely pure tin sample can be represented near  $T_C$  by a single curve and reveal no traces of " $T_C$  anisotropy."

Our numerous different experiments thus show un-

FIG. 2. Block diagram of pulse circuit. 1 - sample, 2 quartz receivers, 3 - quartz transmitters, 4 - receivers, 5 - continuous attenuators, 6 - oscillograph, 7 - comparison pulse generator, 8 - synchronizing circuit, 9 - modulator, 10 - rf generator



FIG. 3. Temperature dependence, based on simultaneous measurements, of the ratio between the electronic longitudinal ultrasonic attenuation coefficients at 133 MHz in a tin single crystal near  $T_c$ .



ambiguously that the onset of a reduction in the electronic ultrasonic attenuation coefficient for  $kl \gg 1$ and  $\hbar\omega \ll 2\Delta$  at the transition to the superconducting state (consequently, also, the appearance of energy gaps and the superconducting transition temperature) is independent of crystallographic orientation. (Here  $\omega = 2\pi f$  is the ultrasonic frequency, and  $2\Delta$  is the energy gap in the superconductor spectrum.)

Let us now consider the results obtained in our investigation of longitudinal ultrasonic attenuation in monocrystalline tin between  $4.2^{\circ}$  and  $1.3^{\circ}$  K. Figure 4



FIG. 4. Temperature dependence of the ratio between the coefficients of electronic longitudinal ultrasonic attenuation in superconductive tin for  $kl \ge 1$ .

shows the temperature dependences exhibited by the ratio  $\alpha_s^{\rm s}/\alpha_n^{\rm n}$  of the electronic attenuation coefficients, from simultaneous measurements on pure tin in both principal directions. Figure 4 agrees well with results obtained by other investigators<sup>[1,15]</sup> in the entire temperature range. From an analysis of Figure 4 (similar to the analysis in<sup>[10]</sup>) we obtain the following minimum gaps in the energy spectrum of superconductive tin:  $2\Delta = (3.6 \pm 0.2) \text{kT}_{\text{C}}$  for k || C<sub>2</sub>, and  $2\Delta = (3.2 \pm 0.1) \text{kT}_{\text{C}}$  for k || C<sub>4</sub>, in good agreement with results reported in<sup>[1,15]</sup>. (The gaps are zones on the Fermi surface that are designated by kv = 0, where v is the electron velocity on the Fermi surface.<sup>[16]</sup>)

The absolute values of the electronic attenuation coefficients  $\alpha_{11}^{\pi}$  obtained in our present work for  $k \parallel C_2$  in tin (Fig. 5) are in agreement with<sup>[8,9]</sup>; however, the value  $\alpha_{11}^{\pi} = 47.6$  db/cm given in<sup>[1]</sup> for f = 70 MHz conflicts with the result 31.5 db/cm that



FIG. 5. Frequency dependences of the electronic ultrasonic attenuation coefficients  $\alpha_n^*$  in normal tin:  $\bigcirc, \bullet -$  measurements in [<sup>1</sup>];  $\Box, \blacksquare$ measurements in [<sup>8</sup>];  $\bigcirc -$  measurements in our earlier investigation [<sup>9</sup>];  $\Delta, \blacktriangle$  measurements in [<sup>17</sup>];  $\heartsuit, \blacktriangledown -$  extrapolation in [<sup>17</sup>];  $\heartsuit, \blacklozenge -$  present work.

was obtained in other investigations. For the same orientation of k Perz and Dobbs<sup>[17]</sup> obtained the incorrect result  $lpha_n^*$  = 60 db/cm by extrapolating to f = 100 MHz. Following a correct extrapolation of the measurements performed by the last-mentioned authors (at f = 42 MHz), we find that the frequency dependence of all the values obtained for  $\alpha_n^*$  (in<sup>[8,9,17]</sup> and the present work) are linear in the entire frequency region; this is in accordance with the theory for kl $\gg 1.^{[18]}$  We note that for the other principal orientation of tin,  $\mathbf{k} \parallel \mathbf{C}_4$ , all the known values of  $\alpha_n^*$  (in<sup>[1,8,17]</sup> and the present work) exhibit linear frequency dependence. As we know, the exact values of the electronic ultrasonic attenuation coefficients for different orientations of single crystals are needed when determining details of the electron energy spectrum and of electron-phonon interactions in metals.<sup>[18,9]</sup>

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