## AMPLITUDE-INDEPENDENT ULTRASONIC ATTENUATION IN SUPERCONDUCTING LEAD\*

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This Letter reports some measurements of the amplitude-independent longitudinal ultrasonic attenuation in superconducting lead. The results are in qualitative agreement with recent calculations<sup>1,2</sup> which show that the phononlimited electron mean free path,  $l_{\rm ph}$ , is different in the normal and superconducting states while the impurity-limited mean free path  $l_0$ is the same in the two states.<sup>2</sup>

Investigations of the amplitude-independent low-temperature ultrasonic attenuation in lead have been plagued by the presence of an amplitude-dependent attenuation.<sup>3,4</sup>. We have examined five well-annealed high-purity singlecrystal lead speciments from three different suppliers, and in every case the amplitude effect was so strong that reliable amplitude-independent data could not be obtained over a sufficiently large frequency range to provide a fair test of the theory. Small amounts of plastic deformation ( $\simeq 0.25\%$ ) as well as alloying decrease the effect; thus, we limited the study to pure deformed specimens and wellannealed alloys where the amplitude independence of the results was experimentally demonstrated. In all cases the amplitude-independent data were taken using transducer voltages at least 4 dB below the amplitude-dependent threshold. Our results show that in pure deformed lead, the superconducting-to-normal electronic attenuation ratio  $\alpha_s/\alpha_n$  decreases with decreasing temperature most rapidly at low frequencies and least rapidly at high frequencies in contradiction to experimental results reported by Deaton<sup>5</sup> for pure well-annealed lead.

Although data were taken between 1.3 and 8°K, reliable estimates of the zero-field normal-state attenuation could be obtained only above about 3.5°K. Below 4.2°K the superconducting data are sensitive to small errors in background-attenuation determination. Therefore, in the following we limit the discussion to temperatures above 4.2°K.

The electron mean free path  $l (l^{-1} = l_{\text{ph}}^{-1} + l_0^{-1})$  was evaluated for the speciments used in the experiments by an extension of the high-field method<sup>6</sup> with an accuracy of ±5%. The normal-state data agreed with the Pippard<sup>7</sup> free-electron attenuation theory to within the accuracy of the l determination. In addition, it was possible to extract the phonon-limited electron mean free path from the data so that one could tell when the mean free path was phonon limited or impurity limited in a given specimen. Experimentally determined ql values at 90 MHz for the pure deformed specimen of the present experiments are given in Table I together with the corresponding phonon-limited quantity  $ql_{\rm ph}$ . The main results of the investigation are as follows:

(1) In pure deformed Pb, where *l* was rigorously phonon limited for  $t = T/T_c > 0.8$  and approximately phonon limited for t > 0.6 [Fig. 1(a)],  $\alpha_s/\alpha_n$  is frequency dependent above 32 MHz and drops fastest at the lowest ultrasonic frequencies used (12.8 and 32 MHz).

(2) In pure deformed Pb and at frequencies (12.8 and 32 MHz) where  $\alpha_n$  is approximately proportional to  $\omega^2$ ,  $\alpha_S/\alpha_n$  is approximately frequency independent and smaller than the BCS prediction based on a zero-temperature energy gap of  $4.30k_BT_c$ .

(3) In pure deformed Pb and above  $5.5^{\circ}$ K,  $\alpha_S$  displays an approximate  $\omega^2$  dependence from 12.8 to 90 MHz, while  $\alpha_n$  displays such a dependence only at the lowest frequencies [Fig. 1(b)]. In Fig. 1(b) the curve labeled "P" is calculated from the Pippard theory<sup>7</sup> using experimentally determined ql values. The straight line labeled "L" has slope 2 and corresponds to a frequency-squared law which is approximately obeyed by the superconducting electronic attenuation  $\alpha_S$ . The straight (dashed) line labeled "S" shows what would be expected if the

Table I. ql estimates in Pb at 90 MHz (q = 2570 cm<sup>-1</sup>,  $\bar{q} \parallel [001]$ ).

Т (°К)	t	ql	$ql_{\mathrm{ph}n}$	$ql_{\mathrm{ph}s}/ql_{\mathrm{ph}n}$
8.0	1.11	1.30	1.36	
7.2	1.00	1.95	2.08	1.00
7.0	0.97	2.17	2.33	0.68
6.5	0.90	2.85	3.14	0.49
5.5	0.76	5.11	6.14	0.28
5.2	0.72	6.13	7.69	0.24
4.7	0.65	8.36	11.6	0.19



FIG. 1. (a) Temperature dependence of  $\alpha_s/\alpha_n$  at various ultrasonic frequencies. The sound was propagating parallel to the [001] axis. (b) Frequency dependence of the normal-state electronic attenuation (circles) compared with the frequency dependence of the superconducting electronic attenuation (squares). See text for further details.

normal-state electronic attenuation obeyed a frequency square law and coincides with the Pippard "P" curve at low frequencies where the normal-state data show such a frequency dependence. At 90 MHz the normal-state data deviate from the frequency-squared dependence by about 100%.

(4) In Pb-Sn alloys ( $\simeq 0.1$  at.% Sn), where the electron mean free path was largely impurity dominated near  $T_c$  and lower than in the phonon-limited case,  $\alpha_s/\alpha_n$  was frequency independent from 12 to 127 MHz (at  $T_c$ , ql ranged from 0.1 to 1.0). In this case the superconducting and normal-state attenuations showed the same frequency dependence, which was in good agreement with Pippard theory.<sup>7</sup>

The above experimental results suggest that for Pb a change in the phonon-limited mean free path in the superconducting transition may be the major cause of the frequency dependence of  $\alpha_s/\alpha_n$  when *l* is phonon limited. In the phonon-limited case, changes in the phonon-limited free path produce changes of comparable magnitude in the total free path. When *l* is impurity limited, changes in the phonon-limited free path do not cause much of a change in *l*. On this basis one expects that  $\alpha_s/\alpha_n$  will be frequency dependent in the phonon-limited case [see item (1) above] and frequency independent in the impurity-limited case [see item (4)].

Using the BCS<sup>8</sup> weak-coupling model, Tsu-

neto<sup>9</sup> has shown that the electronic attenuation in the superconducting state is

$$\alpha_{s} = \alpha_{0} F(ql) [2f(\beta \Delta)], \qquad (1)$$

where  $\alpha_0$  is independent of l and proportional to frequency, q is the wave number of the sound,  $\beta = (k_{\rm B}T)^{-1}$ , and  $\Delta(T)$  is the temperature-dependent energy-gap parameter of the BCS theory. In Eq. (1),  $f(x) = [1 + \exp(x)]^{-1}$  and

$$F(x) = \frac{6}{\pi x} \left[ \frac{1}{3} \frac{x^2 \tan^{-1}(x)}{x - \tan^{-1}(x)} - 1 \right].$$

In the normal state  $\Delta = 0$  and Eq. (1) reduces to the Pippard formula<sup>7</sup> for the normal-state attenuation. Although Eq. (1) was derived only for the case of impurity scattering, it has been usual to assume its validity even when phonon scattering is important, and this assumption will be made here. If the mean free path is different in the normal and superconducting states, it is natural to assume that

$$\alpha_{s} = \alpha_{0} F(ql_{s})[2f(\beta\Delta)],$$
  
$$\alpha_{n} = \alpha_{0} F(ql_{n}),$$
 (2)

where  $l_s$ ,  $l_n$  are average (energy-independent) total electron mean free paths in the superconducting and normal states, respectively. If  $l_s$  is less than  $l_n$  in accord with the calculations of Ambegaokar and Woo,<sup>10</sup> it follows from Eq. (2) that for the phonon-limited case  $\alpha_s$  can display an  $\omega^2$  dependence even at temperatures where  $\alpha_n$  does not show such a dependence [see item (3)]. From Eq. (2),

$$\alpha_s / \alpha_n = [F(ql_s) / F(ql_n)] 2 f(\beta \Delta).$$
(3)

In the low-ql limit  $F(ql) \propto ql$ , so that from Eq. (3),  $\alpha_s / \alpha_n = (l_s / l_n) 2 f(\beta \Delta)$ , which is frequency independent and less than the BCS result  $2f(\beta\Delta)$  see item (2). The result of analyzing data on pure deformed Pb using Eq. (3) is shown in Fig. 2. It was assumed that the zero-temperature gap in Pb is  $2\Delta(0) = 4.30k_{\rm B}T_c$  as given by tunneling measurements<sup>11</sup> and that the temperature dependence of the gap is correctly given by the BCS theory. The superconducting mean free path  $l_s$  was obtained using Eq. (3) from the experimental  $\alpha_s/\alpha_n$  data at 51 MHz. The superconducting-to-normal ratio of the phonon-limited mean free path  $l_{\text{ph s}}/l_{\text{ph n}}$  so derived is given at various temperatures in Table I. The mean free path ratio as given



FIG. 2. Temperature dependence of  $\alpha_s/\alpha_n$  at 32 MHz compared with BCS theory and Eq. (3) of the text.

by the present measurements decreases more rapidly with decreasing temperature than that calculated by Ambegaokar and Woo.<sup>10</sup> While the ratio calculated in Ref. 10 is energy dependent, the one given here is not and represents an average over all electron energies. Using the values of  $l_s$  derived from the 51-MHz data together with the normal-state mean-freepath values derived from high-field data, Eq. (3) is compared with data at 32 MHz in Fig. 2. In Fig. 2 the curve labeled "BCS" shows the BCS prediction based on a zero-temperature gap of  $4.30k_{\rm B}T_c$ , while the curve labeled "T" shows  $\alpha_S/\alpha_n$  calculated from Eq. (3). Similar results were obtained at other frequencies. At 90 MHz the fit was not as good as shown in Fig. 2 and at 12.8 MHz it was better. In all cases Eq. (3) reproduced the fast drop in the experimental  $\alpha_S/\alpha_n$  data at high reduced temperatures. No attempt was made to improve the fit by varying the zero-temperature gap.

In summary we find that  $\alpha_S/\alpha_n$  shows qualitatively different behavior in the phonon- and impurity-limited cases, and that the frequency dependence of  $\alpha_S/\alpha_n$  in the phonon-limited case is consistent with a change in the phononlimited electron mean free path in the superconducting transition.

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## ENERGY OF MOTION OF VACANCIES IN TUNGSTEN<sup>†</sup>

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Various indirect measurements of recovery phenomena in metals have been undertaken to establish the activation energies associated with the formation and motion of specific defects in a wide variety of metals.<sup>1</sup> In spite of the large amount of research in this area many specific defects remain unidentified primarily because of the indirect nature of the measurement (for instance, resistivity,<sup>2</sup> yield stress,<sup>3</sup> and stored energy<sup>4</sup>). It is known, though, that a direct measurement of excess defect configurations can be obtained through use of the field-ion microscope.<sup>5,6</sup> The capabilities of this tool have been used in the present study to observe directly the isothermal removal of single vacancies in neutron-irradiated tungsten and, thereby, to obtain an estimate of the energy of motion of single vacancies in tungsten. This estimate precludes the possibility that single vacancies move extensively in stage III of the recovery spectrum of tungsten.

Briefly, the procedure used in this study is as follows: Commercial-purity tungsten

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