HIGH-FREQUENCY EFFECTS IN THE ACOUSTIC ATTENTUATION OF SUPERCONDUCTING ALUMINUM*

E. A. Fagen and M. P. Garfunkel University of Pittsburgh, Pittsburgh, Pennsylvania (Received 10 April 1967)

All published experimental results of acoustic attenuation in superconductors have employed frequencies so low that the phonon energy $h\nu$ is small compared to the superconducting energy gap at absolute zero, $2\Delta(0)$.^{1,2} In this limit, absorption of phonons can only occur by scattering of thermally excited quasiparticles.³ At energies greater than $2\Delta(T)$, however, phonon absorption can also occur by the creation of a pair of quasiparticles from the ground state (gap jumping). Calculations of the resultant acoustic attenuation at all phonon energies and temperatures have been made by Privorotskii⁴ and by Bobetic⁵ on the basis of the microscopic theory of superconductivity of Bardeen, Cooper, and Schrieffer.³ In the above theories the total attenuation is expressed as the sum of two terms: a scattering term similar to but everywhere smaller than the low-frequency result which gives the ratio of the attenuation coefficients in the superconducting and normal states as α_S/α_n $=2f(\Delta)$, where f is the Fermi function]; and a gap-jumping term which has a discontinuous threshold at the temperature where $h\nu = 2\Delta(T)$. On the high-temperature side of this discontinuity the total attenuation exceeds the normalstate attenuation.

In an attempt to verify these predictions we have measured the attenuation of longitudinal acoustic waves in superconducting aluminum at 9.3 GHz. This frequency, corresponding to a reduced phonon energy $h\nu/\Delta(0) \approx 0.24$, is much higher than in earlier experiments,^{1,2} and high enough to make observations of the discontinuity due to the onset of the gap-jumping process. Our results exhibit a discontinuity in the attenuation slightly below the transition temperature, the magnitude and location of which are in qualitative agreement with the theoretical predictions^{4,5} at this frequency. Less prominent features of the theory, such as the rise in attenuation between the transition temperature and the discontinuity, are so small that they would be obscured by our experimental error.

The acoustic absorption measurements⁶ can

be described with the help of Fig. 1, which gives a schematic diagram of the apparatus. A pulse of 9.3-GHz electromagnetic radiation of about 0.3 μ sec duration generates a similar pulse of longitudinal acoustic waves in the quartz rod. This pulse travels through the sample and is transduced back into an electromagnetic signal in the receiving cavity about 5 μ sec after the primary electromagnetic pulse. The amplitude of this received signal is then used to measure the attenuation in the sample as a function of the temperature. The quartz rods and the sample are cooled to temperatures in the range 0.35 to 1.3°K by a He³ refrigerator. The earth's magnetic field is cancelled to less than 0.05 G.

By extrapolating the attenuation coefficient α_S to 0 at absolute zero, and normalizing with respect to the normal-state attenuation, $\alpha_n = 37.8 \pm 1.0$ dB, we obtain the data of Fig. 2, which gives α_S/α_n vs T/T_c for a single crys-



FIG. 1. Diagram of the acoustic part of the apparatus including components for generation, transmission, absorption, and detection of pulses of 9.3-GHz acoustic waves. See the text for a description of the functions.



FIG. 2. Acoustic attenuation ratio, α_s / α_n , as a function of reduced temperature, T/T_c . The dashed line, indicating the discontinuity, was too steep to obtain individual data points. See the text for a discussion of how the discontinuity was measured. The point above T_c (carefully measured by long-time averaging of the received pulses with a multichannel pulse-height analyzer) was used to normalize all the data.

tal of pure aluminum, 0.011 cm long, and oriented so that the direction of sound propagation is about 13° from the (100) direction. The shape of the curve from just below the discontinuity to the lowest measured temperature, T = 0.36 °K, can be fitted by a curve obtained from the Bobetic⁵ calculation with the gap $2\Delta(0)$ adjusted to $(3.4 \pm 0.3)kT_c$. [Within the required accuracy at this frequency, we have found it satisfactory to represent the Bobetic result below the discontinuity as a fixed fraction β of the lowfrequency result; thus $\alpha_s / \alpha_n = \beta \times 2f(\Delta)$.] The discontinuity is so steep that individual data points could not be taken. Instead, continuous observation of the transmitted pulse amplitude was made as the temperature was allowed to vary slowly through the discontinuity. From these observations it was established that the discontinuity is $7.5 \pm 1.5 \text{ dB} [(0.20 \pm 0.04)\alpha_n]$ occuring in a temperature interval $\lesssim 5 \times 10^{-4}$ °K at $T = 1.170 \pm 0.003$ °K without perceptible hysteresis. This is about 0.005°K below the transition temperature of pure aluminum (T_c) =1.175°K) as determined in other experiments at this laboratory.⁷

In the absence of a magnetic field we know of three possible sources of a discontinuity in the temperature dependence of acoustic attenuation of longitudinal waves in superconductors at these frequencies:

(1) There is a possibility that near T_c , where the attenuation rises most rapidly with temperature, an increase in absorption further increases the temperature of the sample, making a continuous variation appear as a discontinuity. This can ordinarily be checked by determining the amplitude dependence of the absorption, but this was not possible in our thick sample since the normal-state signal was barely retrievable from the noise at our maximum sound-power input (peak pulse power < 30 mW). However, an estimate of the magnitude of the effect shows that it can cause a temperature rise of only 0.0002° K.

(2) Because the excitation of nominally longitudinal waves is not along a principal direction of the sample and because the Fermi surface in aluminum is not spherical, the acoustic attenuation may exhibit a discontinuity below T_c .⁸ This discontinuity is a consequence of the shorting out of transverse electromagnetic fields by supercurrents. The high symmetry of aluminum would seem to make this a small effect, but a detailed calculation appears forbiddingly difficult. Previous observations of the shorting-out effect for longitudinal waves have been made only in tin,⁸ which has a Fermi surface of considerably greater asymmetry than aluminum.

(3) The calculations of Privorotskii⁴ and Bobetic⁵ show that there is a discontinuity in the attenuation, $\delta(\alpha_S/\alpha_n) = (\pi/2)[1-2f(\Delta)]$, occuring at a temperature given by $h\nu = 2\Delta(T)$. In aluminum the theoretical discontinuity is $\approx 0.15\alpha_n$, occurring at a temperature about 0.005° K below the transition temperature. This is somewhat smaller than the discontinuity of $0.20\alpha_n$ shown in Fig. 2, but in view of (2) above, it is not unreasonable to suppose that the shorting-out effect can account for the remaining $0.05\alpha_n$ in the discontinuity.

In summary, we find that at 9.3 GHz there is a discontinuity in the attenuation of longitudinal waves in a single crystal of pure aluminum just below T_C . The evidence suggests that it results largely from the onset of quasiparticle creation by phonons when $h\nu = 2\Delta(T)$, but possibly in part from shorting out of the transverse electromagnetic field by supercurrents. Below this temperature, we have been able to fit the data by scaling the low-frequency formula with the gap $2\Delta(0)$ adjusted to $(3.4\pm0.3)kT_c$.

We gratefully acknowledge helpful discussions with T. Holstein, J. A. Rayne, and particularly C. K. Jones, whose interest and advice have been especially valuable throughout the course of the experiment. We also wish to thank G. Pike who has helped with the data taking in the last stages of this work.

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²The highest frequency previously reported is that at 9.16 GHz on polycrystalline mercury $[h\nu/\Delta(0) \sim 0.07]$ by R. B. Ferguson, Bull. Am. Phys. Soc. <u>12</u>, 330 (1967). There is also the work of H. E. Bömmel on indium at

≈10 GHz reported informally in the discussion at the Cambridge Colloquium on Superconductivity, June 1959, and variously quoted since. E. A. Lynton [Phys. Today <u>12</u>, No. 11, 26 (Nov. 1959)] incorrectly gives the frequency in Bömmel's experiment as 30 GHz. This error was unfortunately repeated in a theoretical paper by I. A. Privorotskii, Zh. Eksperim. i Teor. Fiz. <u>43</u>, 133 (1962) [translation: Soviet Phys.-JETP <u>16</u>, 945 (1963)], who compared Bömmel's presumed results with the theoretical high-frequency absorption curve. It seems unlikely that this comparison is valid.

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MEISSNER EFFECT FOR SUPERCONDUCTORS WITH MAGNETIC IMPURITIES*

W. R. Decker, D. T. Peterson, and D. K. Finnemore

Institute for Atomic Research and Departments of Physics and Metallurgy, Iowa State University, Ames, Iowa (Received 20 April 1967)

Th-Gd alloys show a full Meissner effect and magnetization curves which are reversible to 0.6% of H_c . Critical field curves agree with the Abrikosov-Gor'kov theory to an accuracy of about 3%.

A special interest has centered around superconductors with magnetic impurities since Abrikosov and Gor'kov (AG)¹ first predicted that they might exhibit gapless superconductivity. Thermodynamic properties,² the thermal conductivity,^{3,4} and the acoustic attenuation⁵ have been calculated for this theory and many aspects have been qualitatively confirmed by criticaltemperature (T_c) ,⁶⁻⁹ electron-tunneling,¹⁰ specific-heat,¹¹ and microwave-absorption¹² experiments. An accurate confirmation of the theory for bulk materials, however, has been difficult because there are serious sample preparation problems. The measurement of critical field curves has heretofore been complicated by severe hysteresis and almost perfect flux trapping.¹¹ For the Th-Gd alloys which are reported here, however, the magnetization curves confirm a full Meissner effect and establish that the bulk properties of superconductors doped with paramagnetic impurities obey the AG theory to an accuracy of approximate-

ly 3%.

Isothermal magnetization measurements were made by the ballistic induction technique which was developed by Finnemore and Mapother.¹³ Above 1.1° K, the T-58 vapor pressure scale¹⁴ was taken as the primary standard, and below 1.1°K, temperatures were determined from the susceptibility of cerium magnesium nitrate. The sixth order Garrett¹⁵ solenoid which was used to produce the magnetic fields was calibrated by the nuclear magnetic resonance of protons in glycerine. To prepare the alloy samples, appropriate quantities of Th and Gd were arc melted four or five times, sealed in Ta containers and annealed at 1200°C for 1 week, pressed into a block, swaged to 0.040-in. diam wire, and annealed at 800°C for 1 h to allow recrystallization and the relief of strain. As a final step, the samples were electropolished in a perchloric acid and methanol solution. Samples prepared in this way have an electrical resistivity at 4.2°K which is proportional