theory by Ambegaokar and co-workers.¹²

The samples, $\frac{1}{16}$ in. thick, were grown with flat parallel faces from 99.99999% pure Hg¹³ and were not subjected to lapping or polishing. Our experiments were carried out using the conventional pulse-echo technique.¹⁴ Attenuation measurements were made in both the normal and superconducting states. The echo pulse was calibrated against a comparison pulse subjected to the same electronic circuitry. At the lower temperatures, magnetoacoustic oscillations were observed in the normal attenuation; however, these were very small compared to the difference in attenuation between the superconducting and normal states. Hence no attempt was made to currect for them; α_n was measured for each temperature at the critical (transverse) field. No amplitude effects¹⁵ were observed in present experiments, although several pulse voltages were used at various frequencies.¹⁶

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¹L. M. Falicov and D. H. Douglass, Jr., <u>Progress in</u> Low Temperature Physics, edited by C. J. Gorter

(North-Holland Publishing Company, Amsterdam, 1964), Vol. 4, p. 97.

²J. K. Hulm, Proc. Roy. Soc. (London) A204, 98

(1950); J. H. P. Watson and G. M. Graham, Can. J. Phys. 41, 1738 (1963).

³P. L. Richards and M. Tinkham, Phys. Rev. 119, 575 (1960).

⁴B. C. Deaton, Phys. Rev. Letters <u>16</u>, 577 (1966).

⁵Our measurements of the critical temperature give $T_c = 4.155 \pm 0.005^{\circ}$ K. This value is in agreement with susceptibility measurements by J. E. Schirber and C. A. Swenson, Phys. Rev. <u>123</u>, 1115 (1961).

⁶B. C. Deaton, Phys. Rev. 140, A2051 (1965).

⁷S. Bermon and D. M. Ginsberg, Phys. Rev. 135, A306 (1964).

⁸L. Mackinnon and A. Myers, Proc. Phys. Soc. (London) 73, 291 (1959).

⁹K. L. Chopra and T. S. Hutchison, Can. J. Phys. 37, 1100 (1959).

 $^{10}\mathrm{R.}$ W. Shaw, private communication.

 $^{11}\text{Deaton's suggestion}$ (Ref. 4) that $(\alpha_S/\alpha_n)\sim\exp(k_S/$

 k_n) appears not to be applicable to Hg. ¹²V. Ambegaokar and L. Tewordt, Phys. Rev. <u>134</u>, A805 (1964); V. Ambegaokar and J. Woo, Phys. Rev.

139, A1818 (1965). ¹³Obtained from United Mineral and Chemical Corpo-

ration, New York, New York.

¹⁴R. W. Morse, Progr. Cryog. <u>1</u>, 219 (1959); G. N. Kamm and H. V. Bohm, Rev. Sci. Instr. 33, 957 (1962). ¹⁵R. E. Love and R. W. Shaw, Rev. Mod. Phys. <u>36</u>, 260 (1964); B. R. Tittmann and H. E. Bommel, Phys.

Rev. Letters <u>14</u>, 296 (1965).

¹⁶R. W. Shaw and co-workers (private communication) have observed a small amplitude effect in their Hg samples.

MICROWAVE ABSORPTION STUDIES OF SUPERCONDUCTORS-MAGNETIC-FIELD-INDUCED ANISOTROPY AND THE EFFECT OF IMPURITIES*

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The effect of a static magnetic field on the electromagnetic absorptivity of superconductors has been studied extensively both experimentally¹⁻⁶ and theoretically.⁶⁻⁸ These studies fall into two groups: those in the frequency region such that the photon energy is very small compared with the energy gap (i.e., $h\nu$ $\ll 2\Delta$)^{2-4, 6-8}; and those where the photon energy is of the same order of magnitude as the energy gap,^{1,5} so that absorption can occur with excitation of a quasiparticle across the gap. In this paper we present experimental results on the effect of electron free path on

the magnetic-field-induced anisotropy of the superconducting energy gap as measured by high-frequency $(h\nu \sim 2\Delta)$ microwave absorption in silver-doped aluminum. These results, along with results for a similar experiment on pure aluminum,¹ will be shown to help substantiate the basic physical postulate used by Maki⁸ in his theory of the magnetic-field dependence of the low-frequency microwave absorption in superconductors.

In the work on high-frequency absorption in pure superconducting aluminum with a static magnetic field parallel to the surface,¹ it was

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seen that the absorption spectrum corresponds to that of an anisotropic energy gap. The anisotropy in the apparent energy gap was shown to be caused by the anistropy in the kinetic energy given to the electrons near the Fermi surface by the Meissner current. The Meissner current, with an associated electron drift velocity \vec{v} , changes the energy of those electrons at the Fermi surface having momentum \vec{p} , by $\vec{\mathbf{p}}\cdot\vec{\mathbf{v}}\equiv pv\cos\theta$. It is known that microwaves are absorbed in a metal with the simultaneous diffuse scattering of the electrons from the sample surface. Thus there is an electron energy change given by $h\nu + vp\cos\theta_i - vp\cos\theta_f$, where θ_i and θ_f are the directions of \vec{p} before and after collision with the surface, respectively. The effect of these considerations was shown¹ to reduce the frequency of the initial onset of absorption at low temperatures, and to cause the rise in absorption to be much more gradual than in the absence of a magnetic field, as can be seen in Fig. 1(b).

In Fig. 1(a) are shown results of a similar experiment on an aluminum sample doped with 0.2% silver, reducing the electron free path (and hence the coherence length) to 0.4 of the



FIG. 1. Surface resistance (absorptivity) ratio versus frequency for (a) silver-doped aluminum, and (b) pure aluminum. In both cases, H is parallel to $H_{\rm rf}$. Crosses are for H=0; circles, for $H=0.8H_C$. These data were taken at $T=0.34T_C$.

superconducting penetration depth, λ . There are two important features to note: (1) There is an anomalous absorption peak in the case $H = 0.8H_C$, which appears just above the initial onset of absorption; and (2) the anisotropy of the energy gap is much less than in the case of pure aluminum, as can be seen from a comparison of the ranges in frequency over which the absorption rises from zero.

We do not know the origin of the absorption peak. Both the amplitude and the frequency of this peak change with field, although the shift in frequency is only about 0.5 Gc/sec as the field is changed from $0.5H_c$ to $0.9H_c$.

The change in absorptivity caused by the magnetic-field-induced anisotropy for both pure and silver-doped aluminum may be seen in Fig. 2. The ordinate is the change in surface-resistance ratio between zero field and $0.8H_C$.

Although the doping brings about several changes in the aluminum (including reducing T_c from 1.175 to 1.121°K, and changing the microwave absorption from the nonlocal nature of the anomalous skin effect almost to the local behavior characteristic of the classical skin effect), it seems that the changes in anisotropy are most likely to be understood in terms of the model of dirty superconductors proposed by Anderson.⁹ According to this model, the electron states in the impure case will not be described in terms of a single momentum state but by an average over several such states, reached by large-angle scattering from an initial state. Thus the kinetic energy term is $\langle \vec{\mathbf{v}} \cdot \vec{\mathbf{p}} \rangle = \vec{\mathbf{v}} \cdot \langle \vec{\mathbf{p}} \rangle$ which in the limit of very short



FIG. 2. Frequency dependence of the change in surface resistance ratio caused by a static magnetic field of $H = 0.8H_c$ - comparison of pure and impure (0.2% silver) aluminum.

free path becomes $\frac{1}{2}mv^2$. This is very much smaller than $vp \cos\theta$, and does not show anisotropy. The electronic free path in the sample used in the present experiment is not so short that all anisotropy is washed out, but is an intermediate case. Nevertheless, in the absence of data on an aluminum sample with shorter electron free paths, the present data confirm the model proposed as well as expected.

Richards and Tinkham⁵ have also made some observations on the effect of magnetic field on the energy gap determined by the electromagnetic absorptivity, concluding that there is no effect. Tinkham,¹⁰ basing his argument on the observation above, suggested that conservation of momentum required that the $\mathbf{\hat{p}} \cdot \mathbf{\hat{v}}$ term be conserved in the absorption of a photon and therefore could not effect the observed energy gap. Our results imply that Tinkham's suggestion¹⁰ is incorrect. In terms of our model we can only understand the Richards and Tinkham⁵ observation if we assume that their samples have short electron free paths.

Maki⁸ in his theory of the magnetic field dependence of the low-frequency surface impedance of superconductors changed the energies of the quasiparticles by just the quantity $\vec{v} \cdot \vec{p}$. In the low-frequency case, this energy shift affects only the absorption by the thermally excited quasiparticles. Although Maki⁸ has worked out detailed formulas for the surface impedance in various temperature domains, it still seems worthwhile to discuss the effect of the $\vec{v} \cdot \vec{p}$ term in a more qualitative way that may help in understanding the physical origin of the "anomalous" behavior of the low-frequency microwave absorption in a static magnetic field. We limit the discussion to a BCS superconductor¹¹ in the extreme anomalous limit. Then the surface impedance ratio for H = 0as given by Mattis and Bardeen¹² is

$$Z/Z_{n} = (\sigma_{1}/\sigma_{n} - i\sigma_{2}/\sigma_{n})^{-1/3}, \qquad (1)$$

where σ_1 and σ_2 are the real and imaginary parts of the conductivity in the superconducting state, σ_n is the conductivity in the normal state, and

$$\frac{\sigma_{\perp}}{\sigma_{n}} \equiv \frac{2}{h\nu} \int_{\Delta}^{\infty} \left[1 + \frac{\Delta^{2}}{E(E + h\nu)} \right] \\ \times \left[\rho(E) \rho(E + h\nu) \right] \left[f(E) - f(E + h\nu) \right] dE, \quad (2)$$

$$\frac{\sigma_2}{\sigma_n} = \frac{i}{h\nu} \int_{\Delta - h\nu}^{\Delta} \left[1 + \frac{\Delta^2}{E(E + h\nu)} \right] \\ \times \left[\rho(E) \rho(E + h\nu) \right] \left[1 - f(E + h\nu) \right] dE.$$
(3)

In these integrals the first bracket in the integrand encloses the matrix element connecting initial and final states. The second encloses the product of initial and final density of states where $\rho(E) = E/(E^2 - \Delta^2)^{1/2}$. The final bracket contains the Fermi functions, where $f(E) = [1 + e^{E/kT}] - 1$.

At high temperatures and low frequency, the integral for σ_1/σ_n has a large contribution from the neighborhood of the infinities in the densities of states; the smaller $h\nu$, the larger the contribution (diverging in the limit $h\nu - 0$). The effect of a static magnetic field in a pure superconductor is to change the energies of the electrons near the Fermi surface by an amount $\vec{p} \cdot \vec{v} = pv \cos \theta$.¹ This affects all the factors in the integrand of Eq. (2) (and, in fact, destroys the validity of this equation). However, in a qualitative way, we can see that at high temperatures and low frequencies the effect on the Fermi functions and matrix element in σ_1/σ_n will be small compared to the smearing out and reduction of the peak of the density-of-states term near the edge of the gap. Thus at high temperatures σ_1/σ_n will decrease with increasing field for low frequencies. As we increase frequency in zero field, the large contribution to σ_1/σ_n of the integrand near the edge of the gap decreases and the effect of introducing anisotropy by a magnetic field is thus reduced. This is just the frequency dependence that has been observed experimentally⁶ in the magnetic field dependence of the absorptivity.

As we reduce temperature, the effect of the anisotropy on the Fermi functions grows, eventually dominating the integrand. This factor always tends to increase σ_1/σ_n , causing a positive change at sufficiently low temperatures, just as observed.⁶ Since impurities wash out the anisotropy induced by the field, the reduction in σ_1/σ_n at high temperatures and the increase in σ_1/σ_n at low temperatures would be expected to be reduced by short free paths. In fact, the negative effect might be expected to be eliminated, leaving only a positive effect for all temperatures for sufficiently short free paths. This is just the effect observed by Richards⁴ in his experiments on impure tin. Now it must be pointed out that, while we have discussed the effect of magnetic-field-induced anisotropy on σ_1/σ_n , this is not the same thing as the effect on R and X, the real and imaginary parts of the surface impedance. In fact, if we consider Eq. (1) for the case $\sigma_1/\sigma_2 \ll 1$ (approximately true for most of the range we are concerned with), then

$$R_s/R_n \approx (\sigma_1/\sigma_n)(\sigma_2/\sigma_n)^{-4/3}$$

and

$$X_{s}/X_{n} \approx 1/\sqrt{3} (\sigma_{2}/\sigma_{n})^{1/3}.$$

These equations do not obviously lead to the observations discussed above unless the changes in σ_2/σ_n cause changes in R_S/R_n that are small, zero, or have the same sign as those caused by σ_1/σ_n . In fact, in the theory of Maki,⁸ the expressions he obtains for X_S/X_n (which is dominated by σ_2/σ_n) do not agree with the results of experiment, suggesting that the origin of the effect of magnetic field on the surface resistance (or on σ_2/σ_n) is not yet completely understood.

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¹W. V. Budzinski and M. P. Garfunkel, Phys. Rev. Letters <u>16</u>, 1100 (1966); W. V. Budzinski and M. P. Garfunkel, in <u>Proceedings of the Ninth International</u> <u>Conference on Low-Temperature Physics</u> (Plenum Press, New York, 1965), Pt. A, p. 391.

²R. Glosser and D. H. Douglass, Jr., in <u>Proceed-ings of the Ninth International Conference on Low-</u><u>Temperature Physics</u> (Plenum Press, New York, 1965), Pt. A, p. 385.

³R. T. Lewis, Phys. Rev. <u>134</u>, A1 (1964).

⁴P. L. Richards, Phys. Rev. <u>126</u>, 912 (1962).

⁵P. L. Richards and M. Tinkham, Phys. Rev. <u>119</u>, 575 (1960).

⁶A. B. Pippard, in <u>Proceedings of the Seventh Inter-</u> national Conference on Low-Temperature Physics, <u>Toronto, 1960</u>, edited by G. M. Graham and A. C. Hollis Hallett (University of Toronto Press, Toronto, Canada, 1961), pp. 320-327; A. B. Pippard, Rev. Mod. Phys. <u>36</u>, 328 (1964).

⁷G. Dresselhaus and M. S. Dresselhaus, Phys. Rev. <u>118</u>, 77 (1960).

⁸K. Maki, Phys. Rev. Letters <u>14</u>, 98 (1965).

⁹P. W. Anderson, J. Phys. Chem. Solids <u>11</u>, 26 (1959).

¹⁰M. Tinkham, IBM J. Res. Develop. <u>6</u>, 49 (1962).
¹¹J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. <u>108</u>, 1175 (1957).
¹²D. C. Mattis and J. Bardeen, Phys. Rev. <u>111</u>, 412

"D. C. Mattis and J. Bardeen, Phys. Rev. <u>111</u>, 412 (1958).