Fig. 6. The nodal surface consists of eight hexagons, arranged symmetrically in the corners of the cube. The intersection of the nodal surface with each wall of the cavity forms a square, turned  $45^{\circ}$  with respect to the edges, and having a diagonal length equal to  $\lambda/2$ . T' changes sign when the nodal surface is crossed. In this mode, the temperature oscillation adjacent to the heater wall, for 1/8 of the area, is  $\pi$  out of phase with the temperature oscillation adjacent to the remainder of the heater. For n=4, 6, 8,  $\cdots$ , the temperature distributions opposite the heater are also divided into areas having opposite signs, and therefore mode A was not excited with either heater configuration we used.

We observed modes B and C superimposed. They peaked at very nearly the same frequency, as seen for  $\lambda = a$  in Fig. 5 (the difference in frequency is due to

unequal cavity dimensions). The phase relations at maximum amplitude are given in Table V. We also show in Fig. 5 an apparent doublet, actually composed of single resonances at  $\sqrt{2}\lambda = a$  (n=m=2; p=0) and  $3\lambda/2=a$ .

In a separate experiment, we decided to see if modes B and C could be separated by increasing the distance between walls in the y direction by 1%. While assembling the cavity, we inadvertently also increased the distance in the z direction by 0.3%. The resulting resonances for  $\lambda = a$  are shown in Fig. 7. Each mode resonated twice, at the frequency for the x direction, and also at the frequency for the y or z direction. Unlike the doublets in the square cavity, the phase relations are the same at each resonant frequency in Fig. 7 (they are as listed in Table V).

PHYSICAL REVIEW

VOLUME 136, NUMBER 4A

16 NOVEMBER 1964

## Magnetic-Field Penetration and Breakdown of Surface Superconductivity I\*

Myron Strongin, Donald G. Schweitzer, Arthur Paskin, and Paul P. Craig

Brookhaven National Laboratory, Upton, New York

(Received 15 June 1964; revised manuscript received 31 July 1964)

Low-frequency low-field ac susceptibility measurements on type II and certain type I superconductors have shown complete diamagnetism, until the transition at some surface upper critical field  $H_{e3}$ . In this paper we describe measurements in a swept dc field which show that at sufficiently large sweep rates the ac field can be made to penetrate the superconductor, and the real part of the ac susceptibility will be dM/dH along the magnetization curve, with no observable transition at  $H_{e3}$ . At intermediate sweep rates, the value of the susceptibility becomes more diamagnetic and a partial transition is observed at  $H_{e3}$ . For the slowest sweeps and highest frequencies, essentially the full diamagnetic-susceptibility characteristic of the point-by-point measurements is obtained, along with a transition at  $H_{e3}$ . These results are of particular significance since they indicate that an ac field of small amplitude can be made to penetrate a type II superconductor in the mixed state, and can therefore be used to investigate the bulk properties. Two models which we have considered to explain our results are briefly discussed.

#### INTRODUCTION

In this paper we report low-frequency low-field ac susceptibility measurements which have been made in "swept dc fields." In a previous paper it was shown that in fixed dc fields the ac susceptibility showed complete sample diamagnetism (i.e.,  $-\frac{1}{4}\pi$ ) until the upper critical field  $H_{c3}$ . In contrast to the measurements in fixed dc fields, the susceptibility measurements in swept dc fields can yield dM/dH of the magnetization curve up to  $H_{c2}$  and then the normal metal susceptibility above  $H_{c2}$ . These results are of particular significance since they indicate that the small ac field has penetrated the superconductor in the mixed state, and can thus be used to investigate the bulk properties of type II superconductors. The detailed results, which

depend on the sweep rate, ac amplitude, and the ac frequency, are discussed.

Magnetization and ac measurements were made on a series of cylindrical alloy samples¹ subjected to various surface treatments. Both the ac and dc fields were parallel to the axes of the samples.

Sensitive ac susceptibility measurements were made in which the external dc field was increased and decreased at constant rates. Both the real component of the ac susceptibility  $(\chi')$ , and the imaginary component  $(\chi'')$ , were studied as functions of dc sweep speeds.<sup>3</sup> By a proper choice of ac frequency, ac amplitude and dc sweep rate,  $\chi'$  can be made less than completely diamagnetic in the region below  $H_{c3}$  and above the region of bulk diamagnetism, thereby indicating penetration of the ac field into the bulk of the

RESULTS

<sup>\*</sup>This work was performed under the auspices of the U. S. Atomic Energy Commission.

<sup>&</sup>lt;sup>1</sup> M. Strongin, A. Paskin, D. G. Schweitzer, O. F. Kammerer, and P. P. Craig, Phys. Rev. Letters 10, 442 (1964).

<sup>&</sup>lt;sup>2</sup> Saint-James and P. G. de Gennes, Phys. Letters 7, 306 (1963).

<sup>&</sup>lt;sup>3</sup> Under conditions of changing dc fields we define  $\chi'$  and  $\chi''$  to be proportional to the real and imaginary parts of the mutual-inductance-bridge imbalance voltage.

sample. In Fig. 1 we show copies of our x-y recorder traces of  $\chi'$  on a copper-plated 2% Bi-in-Pb sample. (Copper plating reduced  $H_{c3}$  to a convenient field.)

Figure 1(a) shows the point-by-point measurements, which demonstrate complete diamagnetism up to  $H_{c3}$ . Figure 1(b) demonstrates a case of partial penetration of the ac signal in a swept dc field. The three critical fields  $H_{c1}$ ,  $H_{c2}$ , and  $H_{c3}$  are evident. (H<sub>c3</sub> is well below the value given by Saint-James and de Gennes<sup>2</sup> since the sample is copper plated.) The values of  $H_{c1}$  and  $H_{c2}$  are in agreement with the values obtained from the magnetization curves [Fig. 2(a)]. Figure 1(c) gives the ac susceptibility for forward and reverse dc sweeps and demonstrates complete penetration. The ac frequency is 18 cps and the dc sweep speed is 17 Oe/sec. It should be noted that there is no observable surface superconductivity above  $H_{c2}$  under these conditions. We have compared the ac susceptibility to dM/dH obtained from the magnetization curves. In Fig. 2(b),  $\chi'$  of Fig. 1(c) is compared to dM/dH obtained from Fig. 2(a). The agreement both in shape and magnitude verifies the agreement of the 18-cps ac measurement and the dc magnetization measurements. No arbitrary normalization has been used to obtain these curves.

The general behavior of the ac susceptibility as a function of the frequency and magnitude of the ac field and the dc sweep rate has been investigated. When the value of the susceptibility is neither completely diamagnetic nor paramagnetic (i.e., for partial penetration of the ac field), increasing the frequency from 18 to 250 cps or decreasing the sweep rate makes  $\chi'$  more diamagnetic. (The dc field sweep rates were 1.7–17 Oe/sec and the ac field amplitudes were 0.03 and 0.2 Oe.)

Type I superconductors having surface layers also show similar behavior in the region below  $H_{c3}$  and

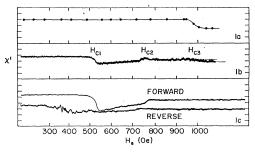
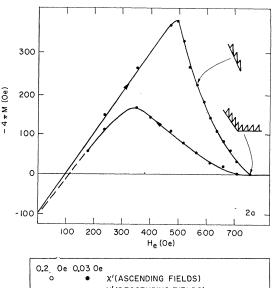


Fig. 1. (a) Point by point susceptibility measurements, at  $4.2^{\circ}$  K, for  $\chi'$  on a copper-plated 2% Bi-in-Pb sample. (b)  $\chi'$  for the same sample with dc sweep rate=1.7 Oe/sec, ac amplitude=0.03 Oe, and ac frequency=18 cps. (c)  $\chi'$  in ascending and descending external fields: dc sweep rate=17 Oe/sec, ac amplitude=0.03 Oe, and ac frequency=18 cps.

detect either  $\chi'$  or  $\chi''$  was fed into an x-y recorder.

<sup>5</sup> In the  $H_{o2}$  to  $H_{o3}$  region, complete diamagnetism suggests that the surface-layer thickness is greater than the penetration depth.



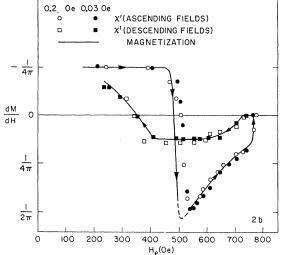


Fig. 2. (a) Magnetization curve for sample in Fig. 1, at  $4.2^{\circ}$ K (insets show enlarged illustrations of possible structure of the magnetization curve; the field-increment size is not necessarily the same in both regions). (b) dM/dH from Fig. 2a and  $\chi'$  from Fig. 1c in ascending and descending fields. Data are also shown for a similar experiment to Fig. 1c in which the ac amplitude was increased to 0.2 Oe. Probable error in obtaining dM/dH from Fig. 2a is about 5%.

above the field at which bulk penetration begins. In type I materials in which there is no  $H_{c3}$ , point-by-point measurements show the paramagnetic susceptibility characteristic of the intermediate state.<sup>1,6</sup>

#### DISCUSSION

We have considered the above results on the basis of two models. In one model we assume that small field increments are required in order to break down the surface [see 2(a) insert] and permit field penetration. Between field penetrations shielding currents are induced in the surface layer thereby making the slope of the magnetization curve diamagnetic. To ex-

<sup>&</sup>lt;sup>4</sup> The signal from our mutual-inductance bridge was detected by a low-level-tuned amplifier which was followed by a Princeton applied research lock-in detector, the output of which, tuned to detect either x' or x'' was fed into an x-y recorder.

<sup>&</sup>lt;sup>6</sup> D. Schoenberg, Proc. Cambridge Phil. Soc. 33, 559 (1937).

plain the experiments the field increments necessary for penetration must be of the order of magnitude of one tenth of an oersted.7 The number of field penetrations per unit time is then dependent upon the dc sweep speed. On the basis of this model it can be shown<sup>8</sup> that the ac field penetration (and hence  $\chi'$ ) depends on the number of times the surface is broken down by the dc field during an ac cycle. If there are many breakdowns the average internal ac field follows the external field and  $\chi'$  will equal the slope of the magnetization curve. For many ac cycles per breakdown,  $\chi'$  will appear diamagnetic since the ac fields are excluded most of the time. The above discussion assumes the ac amplitude to be comparable to or less than the field increments necessary for penetration. Under these conditions almost all break-ins are caused by the dc sweep field.

The results of Park<sup>9</sup> and Le Blanc<sup>10</sup> on "semireversi-

ble" superconductors provide an alternate explanation. They find that for small changes in the external field a minor hysteresis loop of negligible area, with slope  $-\frac{1}{4}\pi$ , is traversed, when the direction-of-field scan along the magnetization curve is reversed. Thus at fixed dc fields, the ac field reverses the scan direction and travels along the diamagnetic hysteresis loop. In a sweeping dc field when  $\omega H_{ac} < dH_{dc}/dt$ , the external field always scans in one direction and will not trace out the diamagnetic minor-hysteresis loop. Under these conditions<sup>11</sup> the susceptibility  $\chi'$  measures dM/dH of the magnetization curve. While this model appears reasonable below  $H_{c2}$ , it is not clear how it can explain the diamagnetism in the  $H_{c2}$  to  $H_{c3}$  region where the mechanism which would cause hysteresis is not obvious.

Sensitive magnetization measurements with sufficient field resolution may be able to determine whether the proposed structure in the inset of Fig. 2(a) exists, and thereby distinguish between the two models.

It is worth noting that our experiments imply that resonance experiments on bulk superconductors may now be possible by choosing dc sweep rates and ac frequencies which allow the ac field to penetrate the sample.

PHYSICAL REVIEW

VOLUME 136, NUMBER 4A

16 NOVEMBER 1964

# Magnetic Properties of Superconducting Films

R. C. CASELLA AND P. B. MILLER IBM Watson Research Center, Yorktown Heights, New York (Received 11 May 1964; revised manuscript received 14 July 1964)

The general Gor'kov equations are solved for a superconducting film in a parallel magnetic field. The method determines the best pairing in the superconducting state without the need for ad hoc assumptions about pairing such as are used in other theories. The critical field  $H_e(T)$  and energy gap  $\Delta(H, T)$  are determined for temperatures near the transition temperature at zero field  $T_c$ . The energy gap in the quasiparticle excitation spectrum is shown to be approximately equal to the spatial average of the order parameter. For films whose thickness d is less than the coherence length  $\xi_T$  the Gor'kov equations are nonlocal and differ from the Ginzburg-Landau (GL) equations. In this range we find  $H_c \propto d^{-3/2}$  in agreement with experiment. For films with  $d > \xi_T$  the solution of the Gor'kov equations are the same as the GL results, as expected, since this is a local regime. We find that for all  $\hat{d}$  (excepting ultrathin films) and in the temperature range  $(1-T/T_c)$  <<1 the field dependence of the energy gap is the same as that given by the GL equations, i.e,  $\Delta(H)/\Delta(0) = \{1 - (H/H_c)^2\}^{1/2}$ . Thus, nonlocal effects do not change the field dependence of the gap. Most of the experimental data are in accord with this equation. However, some recent results for aluminum films show deviations which we interpret as probably being due to the important role played by energy-level quantization of single-particle states in ultrathin films. The extension of the method to lower temperatures and higher fields is also discussed.

### I. INTRODUCTION

E obtain nonlocal solutions of the Gor'kov<sup>1</sup> integral equations for the critical temperature  $T_c(H)$ and order parameter  $\Delta(H,T)$  for  $T < T_c(H)$  of a type-I

superconducting film with equal magnetic fields H applied parallel to both film surfaces. Our derivation is restricted to the temperature range,  $1-t \ll 1$ , where  $t = T/T_c(0)$  is the reduced temperature, and to sufficiently pure samples for which the film thickness d is considerably less than the mean free path l for scattering

<sup>&</sup>lt;sup>7</sup>The increment size is expected to depend upon the sample, sample preparation, and the external field. In some samples we and under the initial and under the region between the initial and upper-bulk critical fields. This noise can be observed in a swept dc field by looking at the signal coming out of the secondary coil with no ac field on the sample. These results would imply that in these samples the field increment necessary for breakdown is very irregular. Experiments to investigate the details of the increment size are underway.

8 A. Paskin, P. P. Craig, D. G. Schweitzer, and M. Strongin

<sup>(</sup>unpublished).

J. G. Park, Rev. Mod. Phys. 36, 87 (1964).
 M. A. R. Le Blanc, Phys. Letters 9, 9 (1964).

<sup>&</sup>lt;sup>11</sup> In these measurements, dM/dH does not change appreciably over one ac cycle. If the sweep rate is large enough so that dM/dH changes in this interval, correction terms must be considered which depend on the sweep rate and  $d^2M/dH^2$ .

<sup>&</sup>lt;sup>1</sup>L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **36**, 1918 (1959) [English transl.: Soviet Phys.—JETP **9**, 1364 (1959)].