## EXPERIMENTAL EVIDENCE FOR QUANTIZED FLUX IN SUPERCONDUCTING CYLINDERS\*

Bascom S. Deaver, Jr., and William M. Fairbank Department of Physics, Stanford University, Stanford, California (Received June 16, 1961)

We have observed experimentally quantized values of magnetic flux trapped in hollow superconducting cylinders. That such an effect might occur was originally suggested by London<sup>1</sup> and Onsager,<sup>2</sup> the predicted unit being hc/e. The quantized unit we find experimentally is not hc/e, but hc/2e within experimental error.<sup>3</sup>

Although the unit of quantized flux is small  $(hc/2e = 2.07 \times 10^{-7} \text{ gauss cm}^2)$ , it can be produced by a magnetic field easily measured and controlled in the laboratory if the area to which it is confined is sufficiently small. For our samples, one flux unit corresponds to a magnetic field of the order of 0.1 gauss. Measurements were made on two hollow tin cylinders. Cylinder No. 1 was 0.8 cm long,  $2.33 \times 10^{-3}$  cm outside diameter and  $1.33 \times 10^{-3}$  cm inside diameter. Cylinder No. 2 was 0.9 cm long,  $1.64 \times 10^{-3}$  cm outside diameter and  $1.35 \times 10^{-3}$  cm inside diameter. These were fabricated by electroplating tin on a one-centimeter portion of a No. 56 copper wire. The sample, plus protruding wire, was jacketed for protection and strength with electroplated copper to an approximate outside diameter of  $8 \times 10^{-3}$  cm.

A field-free region ( $H = 0 \pm 0.001$  gauss) is prepared using three orthogonal Helmholtz coils. The tin cylinder is placed in this region and cooled through the superconducting transition in the presence of a known applied axial magnetic field. The net flux in the cylinder is measured both with the field on and after the field is turned off. The measurement is made by moving the tin cylinder up and down one hundred times per second with an amplitude of one millimeter and observing the electrical pickup in two small coils, each of ten thousand turns, surrounding the ends of the cylinder. The instrument is similar in concept to that described by Foner.<sup>4</sup> The induced emf measures the difference in the flux contained within the area of the cylinder and that which would have been in the same area if the cylinder were absent (or in the normal state). The system is calibrated by cooling the sample below the superconducting transition in zero field and observing the signal from the completely diamagnetic cylinder when a known magnetic field is applied. From the value of the field and the measured cross-section area of the cylinder, the absolute value of the flux for a given signal is calculated.

The diameter of each cylinder was measured with a microscope equipped with a micrometer eyepiece. X-ray photographs verified the dimensions of the tin cylinder after the application of the copper jacket. For the purpose of calculating flux, the measured radii of the cylinders are reduced by 0.6 micron due to an expected loss of superconducting properties on the surface of the tin in contact with the copper.<sup>5</sup> That this correction was approximately valid is indicated by a 0.2°K decrease in the value of the transition temperature for the sample No. 2 whose cylindrical walls were 1.5 microns thick, leaving, we believe, only 0.3 micron of superconducting material in the center after allowance for the effect of the center copper wire and the outside copper jacket. This is in agreement with the experimental results on electroplated tin.<sup>5</sup>

With this adjustment, the area used for the diamagnetic calibration of cylinder No. 1 is  $3.84 \times 10^{-6}$  cm<sup>2</sup>, and the area of the hole is  $1.65 \times 10^{-6}$  cm<sup>2</sup>. For cylinder No. 2 the corresponding quantities are  $1.81 \times 10^{-6}$  cm<sup>2</sup> and  $1.70 \times 10^{-6}$  cm<sup>2</sup>.

Data on sample No. 1 are shown in Fig. 1, and on sample No. 2 in Fig. 2. The diagonal line through the origin represents the calibration. It is the signal corresponding to zero flux in the cylinder and hole in the presence of the applied field as described above. The experimental points shown on the graph represent two types of data for each value of the applied field. The points on the bottom half of each graph represent the signal in the presence of the applied field after cooling through the transition in that field. The points in the upper half represent the trapped flux after the field is subsequently reduced to zero. For each point in the lower curve there is a corresponding point in the upper curve. The solid lines represent calculated integral values of hc/2e.

It can be seen that certain features of the data are common to both samples. (1) Below a certain value of applied field, the total cross section of the cylinder acts as a perfect diamagnet, excluding all the flux, and no flux is trapped when the applied field is turned off. (We believe this



FIG. 1. (Upper) Trapped flux in cylinder No. 1 as a function of magnetic field in which the cylinder was cooled below the superconducting transition temperature. The open circles are individual data points. The solid circles represent the average value of all data points at a particular value of applied field including all the points plotted and additional data which could not be plotted due to severe overlapping of points. Approximately two hundred data points are represented. The lines are drawn at multiples of hc/2e. (Lower) Net flux in cylinder No. 1 before turning off the applied field in which it was cooled as a function of the applied field. Open and solid circles have the same significance as above. The lower line is the diamagnetic calibration to which all runs have been normalized. The other lines are translated vertically by successive steps of hc/2e.

provides a way of obtaining a truly zero-magneticfield region.) (2) When the applied field exceeds a certain value, flux is trapped both with the field on, and after the applied field is turned off. The amount of this trapped flux within the experimental accuracy of the data is hc/2e.

The amount of trapped flux remains constant as the applied field is increased until a value approximately three times that for the initial trapping is reached, at which point the trapped flux increases to about twice the original amount. There appears to be evidence for additional changes at five and seven times the field for the first trapping.

Fluctuations in the data are caused by variations in the zero of the magnetic field, changes in the gain, vibration amplitude, drift, and random noise in the detection system. The approximately two hundred data points for sample No. 1 were taken over a three-week period during which the drift and noise were gradually improved. The fluctuations of the data around the values 0 and hc/2e represent, we believe, expected scatter from drift and noise. This scatter has been greatly improved for sample No. 2. For both samples the data are consistent with values 0 and hc/2e for the trapped flux as described above.

Near the transition to the second and third steps the fluctuations in the data are greater, and in addition points lie between the steps. Some increased scatter is expected since the absolute fluctuations due to changes in gain and vibration amplitude are proportional to the size of the signal. The points between the steps do not necessarily indicate trapping of nonintegral values of flux. Since the observed signal is the sum of the emf's from coils at the two ends of the sample, a flux line passing out of the cylinder at some point other than the end may produce different signals in the two coils. The two ends of the cylinder are not quite identical; so near the transition region it is probable that the two ends might trap a different number of units

of flux, the extra unit being shoved out the side of the cylinder. This is especially probable for sample No. 1 since the x-ray photograph showed a break in the tin coating near the middle of the cylinder. Also, it is known that flux can create a normal region in a superconductor by shrinking in size until the critical field is exceeded. In this experiment we were unable to measure independently the signals from the two coils. However, in future experiments this will be done to remove this ambiguity. It is interesting to note that no intermediate points are found outside the expected scatter of the data near the first step. One point for which no flux was trapped was found near the center of the first step with sample No. 1.

In conclusion, we find:

1. The flux trapped in a superconducting cylinder both in the presence and absence of an applied magnetic field is not continuous but exhibits a step behavior, the first step occurring for  $\Phi = hc/2e$ , within experimental error in the data. Considering all sources of error, we estimate that the value of the trapped flux at the first step is  $hc/2e \pm 20\%$ . If the correction to the size of the cylinder due to the presence of the copper should prove invalid, an additional 11% error could arise for the large cylinder and 17% for the small cylinder.

2. The data seem to indicate additional steps at hc/e, 3hc/2e, and 2hc/e. The points appearing between these levels will be investigated further.

3. The ratio of the fields at which the steps occur are approximately 1, 3, 5, and 7. In the first cylinder (for which the effective cross-sectional area of the cylinder is 2.33 times the area of the hole), the first jump occurs when the flux passing through the total effective cross section of the cylinder in the normal state is approximately hc/2e.

For cylinder No. 2 (in which the effective crosssectional area of the cylinder is 1.1 times the area of the hole), the first jump occurs when the flux passing through the total effective cross section of the cylinder in the normal state is approxi-



of magnetic field in which the cylinder was cooled below the superconducting transition temperature. The circles and triangles indicate points for oppositely directed applied fields. Lines are drawn at multiples of hc/2e. (Lower) Net flux in cylinder No. 2 before turning off the applied field as a function of the applied field. The circles and triangles are points for oppositely directed applied fields. The lower line is the diamagnetic calibration to which all runs have been normalized. The other lines are translated vertically by successive steps of hc/2e.

FIG. 2. (Upper) Trapped flux

in cylinder No. 2 as a function

mately 0.6hc/2e. In a following Letter, Byers and Yang<sup>6</sup> conclude that in a thin ring the first jump should occur at 0.5hc/2e.

4. Since the time constant of our measuring circuit is 25 seconds, this experiment gives only a large upper limit for the time involved in reaching these quantized flux values. Mercereau and Vant-Hull<sup>7</sup> have reported a negative experiment designed to observe quantized flux in a 1-mm ring cooled 6000 times per second through the superconducting transition in a small magnetic field. It is possible that the difference in their results and the results of our experiment are due to a minimum time necessary to establish equilibrium. We are planning to investigate this relaxation time.

We have had the pleasure of discussing the results of this experiment with N. Byers, C. N. Yang, and L. Onsager, whose interpretation of these results appear in the following Letters.<sup>6,8</sup> One of us (WMF) also wishes to acknowledge his indebtedness to F. London and M. J. Buckingham who greatly influenced his concept of the superfluid state. We also wish to thank F. Bloch, L. I. Schiff, and J. D. Bjorken for many stimulating discussions of the experiment. We wish to acknowledge the invaluable assistance of M. B. Goodwin.

\*Work supported in part by grants from the National Science Foundation, the Office of Ordnance Research (U. S. Army), and the Linde Company.

<sup>1</sup>F. London, <u>Superfluids</u> (John Wiley & Sons, New York, 1950), p. 152.

<sup>2</sup>L. Onsager, <u>Proceedings of the International Con-</u> ference on Theoretical Physics, Kyoto and Tokyo, <u>September, 1953</u> (Science Council of Japan, Tokyo, 1954), pp. 935-6.

<sup>3</sup>Such a possibility was mentioned by Lars Onsager to one of us (WMF) at the conference on superconductivity in Cambridge, England, 1959 (unpublished).

<sup>4</sup>S. Foner, Rev. Sci. Instr. <u>30</u>, 548 (1959).

<sup>5</sup>E. Burton, H. Grayson-Smith, and J. Wilhelm, <u>Phenomena at the Temperature of Liquid Helium</u> (Reinhold Publishing Corporation, New York, 1940), p. 120.

<sup>6</sup>N. Byers and C. N. Yang, following Letter [ Phys. Rev. Letters 7, 46 (1961)].

<sup>7</sup>J. E. Mercereau and L. L. Vant-Hull, Bull. Am. Phys. Soc. 6, 121 (1961).

<sup>8</sup>L. Onsager, this issue [Phys. Rev. Letters  $\underline{7}$ , 50 (1961)].

## THEORETICAL CONSIDERATIONS CONCERNING QUANTIZED MAGNETIC FLUX IN SUPERCONDUCTING CYLINDERS\*

## N. Byers and C. N. Yang<sup>†</sup>

Institute of Theoretical Physics, Department of Physics, Stanford University, Stanford, California (Received June 16, 1961)

In a recent experiment,<sup>1</sup> the magnetic flux through a superconducting ring has been found to be quantized in units of ch/2e. Quantization in twice this unit has been briefly discussed by London<sup>2</sup> and by Onsager.<sup>3</sup> Onsager<sup>4</sup> has also considered the possibility of quantization in units ch/2e due to pairs of electrons forming quasi-bosons.

The previous discussions<sup>3</sup> leave unresolved the question whether quantization of the flux is a new physical principle or not. Furthermore, sometimes the discussions seem<sup>2</sup> to be based on the assumption that the wave function of the superconductor in the presence of the flux is proportional to that in its absence, an assumption which is not correct. We shall show in this Letter that (i) no new physical principle is involved in the requirement of the quantization of magnetic flux through a superconducting ring, (ii) the Meissner effect is closely related to the requirement that the flux through any area with a boundary lying entirely in superconductors is quantized, and (iii) the quantization of flux is an indication of the pairing of the electrons in the superconductor.

<u>Macroscopic discussion</u>. Consider a multiply connected superconducting body P with a tunnel O (Fig. 1). We shall only discuss macroscopic



FIG. 1. Multiply connected superconductor.