

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

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

<sup>2</sup>L. Onsager, *Proceedings of the International Conference on Theoretical Physics, Kyoto and Tokyo, September, 1953* (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was mentioned by Lars Onsager to one of us (WMF) at the conference on superconductivity in Cambridge, England, 1959 (unpublished).

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<sup>5</sup>E. Burton, H. Grayson-Smith, and J. Wilhelm, *Phenomena at the Temperature of Liquid Helium* (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* **7**, 50 (1961)].

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

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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 require-

ment 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.

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

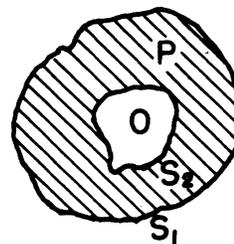


FIG. 1. Multiply connected superconductor.

dimensions much larger than the penetration depth. The Meissner effect then states that inside the superconductor  $P$  the magnetic field is zero, and the current is zero. Surface currents, however, do exist and persist on the surfaces  $S_1$  and  $S_2$ . The surface currents and the external sources of magnetic fields together produce no magnetic flux in the interior  $P$  of the superconductor. They in general, however, produce a net magnetic flux through  $O$ , to be denoted by  $\Phi$ .

The energy eigenfunction  $\psi$  of the electrons in the superconductor satisfies

$$\sum_j \frac{1}{2m} \left[ -i\hbar \vec{\nabla}_j + \frac{e}{c} \vec{A}(\vec{r}_j) \right]^2 \psi + V\psi = E\psi, \quad (1)$$

where  $\vec{A}$  is the vector potential due to the surface currents and external magnetic sources. Inside  $P$ ,

$$\vec{\nabla} \times \vec{A} = 0.$$

Hence  $\vec{A} = \vec{\nabla}\chi$ , where  $\chi$  is not single valued in  $P$  but increases by

$$\Delta\chi = \oint \vec{A} \cdot d\vec{l} = \iint \vec{H} \cdot d\vec{\sigma} = \Phi, \quad (2)$$

whenever one goes around the tunnel  $O$  once. Defining

$$\psi' = \psi \exp \sum_j i \frac{e}{c\hbar} \chi(\vec{r}_j), \quad (3)$$

we see that (1) reduces to

$$\sum_j \frac{1}{2m} (-i\hbar \vec{\nabla}_j)^2 \psi' + V\psi' = E\psi'. \quad (4)$$

The vector potential  $\vec{A}$  is eliminated from this equation. However, the boundary condition for  $\psi'$  is that when all electron coordinates are fixed, except for one,  $\vec{r}_l$ , and  $\vec{r}_l$  is brought around  $O$  once,  $\psi'$  changes by a constant factor

$$\psi' \rightarrow \psi' e^{i(e/c\hbar)\Phi}. \quad (5)$$

[To prove (5) we use (3) and (2) and the fact that  $\psi$  is single valued.]

The eigenvalues  $E$  are determined by the differential equation (4) and the boundary condition (5). It is thus obvious that we have:

**Theorem 1.** The energy levels are periodic in the magnetic flux  $\Phi$  with a period  $ch/e$ .

If the surfaces  $S_1$  and  $S_2$  are concentric cylinders on which  $\psi = 0$ , and  $V$  is put equal to zero, the energy levels  $E$  can be explicitly solved for, illustrating this theorem. One notices that  $\psi'$  is not simply proportional to  $\psi(\Phi = 0)$ , as is sometimes<sup>2</sup> assumed in the literature.

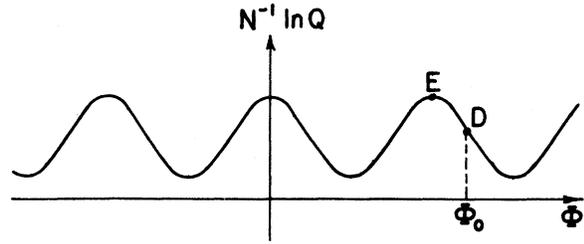


FIG. 2. Periodic variations in  $N^{-1} \ln Q$  as a function of trapped flux  $\Phi$ .

If  $V$  is a real function of  $r_j$ , by taking the complex conjugate of (4) and (5), we have:

**Theorem 2.** The energy levels are even functions of  $\Phi$ .

It is clear that theorems 1 and 2 remain valid if we introduce the lattice coordinates of the metal and if we introduce the spin of the electrons. The operation of complex conjugation in the proof of theorem 2 has then to be replaced by the time reversal operation and the proof depends on the time reversal invariance of the interactions.

From these theorems it follows trivially that:

**Theorem 3.** The partition function  $Q$  of the system is an even periodic function of  $\Phi$  with period  $ch/e$ .

At  $\Phi = (ch/2e) \times \text{integer}$ , this theorem shows that

$$\partial \ln Q / \partial \Phi = 0, \quad (6)$$

and that  $\ln Q$  has the general form shown in Fig. 2.

Now the body current in the superconductor around  $O$  is

$$I = kTc \partial \ln Q / \partial \Phi. \quad (7)$$

( $c$  = velocity of light.) In the differentiation we keep the temperature  $T$  constant.

The Meissner effect requires that  $I = 0$ . Thus the equilibrium states are given by the maxima and minima on the curve in Fig. 2. We shall now give an argument to show that the maxima, not the minima, are the equilibrium states realized. A point  $D$  in Fig. 2 is not an equilibrium state so the calculation of the partition function at that point is strictly speaking meaningless. But the slope of the curve at that point indicates that if a flux  $\Phi_0$  is made to pass through  $O$ , a body current would be induced, the sense of the current being negative according to (7). The additional flux due to this body current causes the flux through  $O$  to decrease. The equilibrium state reached would therefore be  $E$

which is a maximum of the curve. We state this as:

**Theorem 4.** The superconducting state is given by the maxima of  $\ln Q$  as a function of  $\Phi$ .

If the external flux does not assume a value for which  $\ln Q$  is a maximum, surface currents will flow on  $S_1$  and  $S_2$  to make up a total flux  $\Phi$  for which  $\ln Q$  is a maximum.

The experiment of reference 1 and theorem 4 together prove that  $\ln Q$  has maxima at integral values of  $\Phi/(ch/2e)$ . Whether a microscopic theory yields these maxima will be discussed in the next section. If it does, then theorem 4 shows the following: The flux through any surface whose boundary loop lies entirely in superconductors is quantized in units  $ch/2e$ . The requirement of this quantization in turn clearly implies that the flux through any small area in a superconductor is zero; hence it implies the Meissner effect.

In a loose sense the above argument can be used to "derive" the Meissner effect itself: If the magnetic flux in a superconductor is not zero, body currents will flow around all loops through which the flux is not quantized. The system cannot reach a steady state until all magnetic flux is expelled from the interior of the superconductor.

**Microscopic considerations.** We now want to see whether a microscopic calculation does or does not lead to maxima of  $\ln Q$  at  $\Phi/(ch/2e) = \text{integer}$ .

To investigate this point we first take a collection of noninteracting spinless electrons between two concentric cylinders  $S_1$  and  $S_2$ . The electrons at a point at a distance  $r$  from the axis have momenta  $p_r$ ,  $p_\theta$ , and  $p_z$  in the radial, azimuthal, and  $z$  directions. Clearly

$$p_\theta r = n\hbar. \quad (n = \text{integer})$$

The energy of the electron is

$$\frac{1}{2m} \left[ p_r^2 + p_z^2 + \frac{\hbar^2}{r^2} \left( n + \frac{e}{ch} \Phi \right)^2 \right]. \quad (8)$$

The partition function  $Q$  can be computed from such an energy spectrum. The resultant  $N^{-1} \ln Q$  [to the order  $N^0$ ] does not depend on  $\Phi$ . Thus, according to theorem 4, for a collection of noninteracting electrons, the flux in the tunnel does not have to be quantized.

It is not difficult to understand why for such a model  $N^{-1} \ln Q$  does not depend on  $\Phi$ . To see this we suppress the  $p_r$  and  $p_z$  degrees of freedom and take the temperature  $T = 0$ . The one-dimen-

sional Fermi sea problem,

$$\frac{\hbar^2}{2mr^2} \sum \left( n + \frac{e}{ch} \Phi \right)^2,$$

gives an average energy per particle of

$$\frac{E}{N} = \text{constant} + \frac{\hbar^2}{2mr^2} \frac{e^2}{c^2 \hbar^2} \Phi^2, \quad \left| \frac{e\Phi}{ch} \right| \leq \frac{1}{2}, \quad (9)$$

if  $N \equiv$  the number of particles is odd. But if  $N$  is even, then

$$\begin{aligned} \frac{E}{N} &= \text{constant} + \frac{\hbar^2}{2mr^2} \left( \frac{e\Phi}{ch} - \frac{1}{2} \right)^2, \quad \frac{1}{2} \geq \frac{e\Phi}{hc} \geq 0 \\ &= \text{constant} + \frac{\hbar^2}{2mr^2} \left( \frac{e\Phi}{ch} + \frac{1}{2} \right)^2, \quad 0 \geq \frac{e\Phi}{hc} \geq -\frac{1}{2}. \end{aligned} \quad (10')$$

Thus, depending on the evenness or oddness of  $N$ , the energy has a minimum at  $e\Phi/ch = \frac{1}{2}$  or 0 (modulo 1). The three-dimensional problem at  $T = 0$  is decomposable into many one-dimensional problems with varying values of  $N$ . Thus the above-discussed fluctuation leads to a cancellation for the three-dimensional problem, resulting in an  $N^{-1} \ln Q$  versus  $\Phi$  curve that is flat. (An  $N^{-1} \ln Q$  curve that is flat applies to the case of a metal in its normal rather than superconducting state.) A similar cancellation obtains for  $T \neq 0$ .

In the neighborhood of  $\Phi = +0$  the states with  $n > 0$  have energies that increase with  $\Phi$ , and those with  $n < 0$  have energies that decrease with  $\Phi$ . The average energy for the two states  $n$  and  $-n$ , however, increases with  $\Phi$  like

$$\text{constant} + (\hbar^2/2mr^2)(e\Phi/ch)^2. \quad (11)$$

If there is a "pair correlation" of the kind proposed by Bardeen, Cooper, and Schrieffer<sup>5</sup> for the superconductor so that states  $n$  and  $-n$  (or a pair of time-reversed states) are either both occupied or both unoccupied, (11) becomes the correct energy per particle for small  $\Phi$ . (In such a case the fluctuation and cancellation phenomena disappear.) This is represented in Fig. 3 by the parabolas at  $2e\Phi/ch = -2, 0, 2$ , etc.

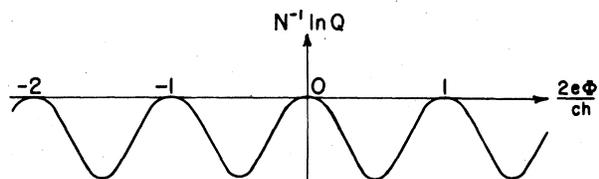


FIG. 3. A curve of  $N^{-1} \ln Q$  versus  $2e\Phi/ch$ , showing parabolic behavior near maxima at  $2e\Phi/ch = \text{integer}$ .

At  $2e\Phi/ch = 1$ , pairing between

$$n + \frac{e}{hc}\Phi = n + \frac{1}{2} = \frac{1}{2} \text{ and } -\frac{1}{2}, \frac{3}{2} \text{ and } -\frac{3}{2}, \text{ etc.},$$

occurs and the energy per particle remains the same as for the case  $\Phi = 0$  (to order  $N^0$ ). In the neighborhood of  $2e\Phi/ch = 1$ , the additional energy for each of these pairs is again twice

$$(\hbar^2/2mr^2)[(e\Phi/ch) - \frac{1}{2}]^2,$$

which give rise to the parabolas at  $2e\Phi/ch = \pm 1$  in Fig. 3. In the absence of a detailed theory, we draw a smooth curve in Fig. 3 to extrapolate between the maxima.

Thus the Bardeen, Cooper, Schrieffer pairs for the superconducting state give rise to the curves such as those depicted in Fig. 3, where the parabolas are repeated at periods  $\Delta(2e\Phi/ch) = 1$ , and the central parabola is given by

$$N^{-1} \ln Q = -\frac{f}{kT} \frac{\hbar^2}{2m \langle r^2 \rangle_{av}} \left( \frac{e\Phi}{ch} \right)^2 + \text{constant}, \quad (12)$$

where  $f$  = fraction of electrons that are paired.

It is interesting to estimate the magnitude of the body current at, say,

$$0 < 2e\Phi/ch < \frac{1}{2}. \quad (13)$$

It is, by (7),

$$I = -Nfc(e^2/mc^2)\Phi/(4\pi^2 \langle r^2 \rangle_{av}).$$

The flux induced by this current is, for a thin ring superconductor,

$$\Phi_{\text{induced}} = -f \times (\text{number of electrons in a length } e^2/mc^2 \text{ of the ring}) \Phi.$$

For the experiment of reference 1,  $-\Phi_{\text{induced}}/\Phi \gg 1$  if  $f$  is not too small, showing that the maxima in Fig. 3 are very pronounced.

From Fig. 3 and the argument preceding theorem 4, we conclude that the trapped flux  $\Phi$  and

the original flux  $\Phi_0$  are related in the following way:

$\Phi_0/(ch/2e)$	$\Phi$
$-\frac{1}{2} \rightarrow \frac{1}{2}$	0
$\frac{1}{2} \rightarrow \frac{3}{2}$	1
$\frac{3}{2} \rightarrow \frac{5}{2}$	2
etc.	

It is interesting to notice that the existence of the variation of the energy levels of the electrons in  $P$  with the flux  $\Phi$ , even when there is no magnetic field in  $P$ , is based on the same principle as the experiment proposed by Aharonov and Bohm.<sup>6,7</sup>

We wish to thank W. M. Fairbank and B. Deaver for informing us of the progress of their beautiful experiment and for many discussions. We also wish to thank F. Bloch for stimulating discussions. One of us (CNY) takes this opportunity to thank the members of the Physics Department of Stanford University for the hospitality extended him during his visit.

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<sup>1</sup>B. S. Deaver, Jr., and W. M. Fairbank, preceding Letter [Phys. Rev. Letters **7**, 43 (1961)].

<sup>2</sup>F. London, Superfluids (John Wiley & Sons, Inc., New York, 1950), p. 152.

<sup>3</sup>L. Onsager, Proceedings of the International Conference on Theoretical Physics, Kyoto and Tokyo, September, 1953 (Science Council of Japan, Tokyo, 1954), pp. 935-6.

<sup>4</sup>L. Onsager (private communication to W. M. Fairbank).

<sup>5</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

<sup>6</sup>Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).

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