PHYSICS TODAY VOL 19 NO 5

Some Remarks on the Theory of Superconductivity

When a metal becomes a superconductor the conduction electrons pair off, and quantum-mechanical effects can be seen macroscopically. However, theorists are still looking for a satisfactory microscopic theory to explain the phenomena.

by Felix Bloch

SUPERCONDUCTIVITY THEORY has been approached in several ways. The most direct but also the most difficult is the attempt to derive all facts from a detailed microscopic theory, and the most significant steps in this direction have been made since 1957 through the theory of John Bardeen, Leon N. Cooper and J. Robert Schreiffer.[1] Another approach, less fundamental but often very instructive, is based on suitably chosen simple models, of which the degenerate ideal Bose gas, suggested by M. R. Schafroth[2] in 1955, has proven to be particularly useful.

However, I shall mainly discuss a third way to deal with the problem—in which one tries to interpret some of the characteristic features of superconductivity in terms of general principles. It is probably the most modest and certainly the oldest approach, and I want to begin with an outline of its origins, not only in deference to their historical value but also because, to some extent, they are still relevant and bear upon recent developments.

These origins go back to the late twenties, shortly after quantim mechanics had furnished a satisfactory explanation of normal metallic conduction, and they particularly concern the nature of persistent currents, certainly one of the most striking aspects of superconductivity. The observation of this phenomenon convincingly demonstrates that the transitions of the conduction electrons caused by lattice vibrations and impurities, which lead to the resistance of a normal metal, are ineffective in the superconducting state. One might have been inclined to attribute this fact to certain novel selection rules, but they would have to be so strict that one could hardly expect to find good reasons for their existence. A far more appealing interpretation was suggested through analogy with ferromagnetism, where remanent magnetization had been explained by recognizing that parallel orientation of the magnetic moments of the atoms leads to a lower energy than random orientation. Similarly, it seemed plausible to interpret current flow in a superconductor as the result of a correlation between the velocities of the conduction electrons that is energetically favored and, therefore, manifests itself at sufficiently low temperatures.

Many of Felix Bloch's notable contributions have been in magnetism and superconductivity. He and E. M. Purcell shared a 1952 Nobel Prize for independently developing nuclear magnetic resonance techniques. Bloch is a professor of physics at Stanford. This article is based on an address that he gave, as retiring president, at the American Physical Society meeting last January.

The picture of independent conduction electrons, which has otherwise been so fruitful in the theory of metals, does not provide for such a correlation. This led to the conclusion that previously neglected interactions between the electrons must be essential for superconductivity just as interactions between individual atoms (in this case known to be due to exchange) are essential for ferromagnetism (in contrast to paramagnetism where atomic magnetic moments are considered to be independent). The problem was thus reduced to the question whether it is possible, by means of such an interaction, to explain the existence of an energy minimum in the presence of current flow. The affirmative answer to this question, however, soon ran into a characteristic hindrance. It is sometimes honored by the title of a theorem that says in its popular version that theories of superconductivity can be disproven; this statement was empirically found to be valid during a considerable period of time.

The difficulty is of a very general nature and exists for a system of arbitrarily interacting electrons described by a quite general Hamiltonian H. Its roots can best be seen from Hamilton's equations of classical mechanics by considering one of the components P of the total momentum of the electrons and the canonically conjugate coördinate X of their center of gravity. One has then

$$\frac{\partial H}{\partial P} = \dot{X} = V \qquad (1)$$

where V is the corresponding component of the drift velocity, proportional to the current. According to this equation, a minimum of the energy H is incompatible with an electric current since it requires the partial derivatives to vanish and, hence, the drift velocity to be zero. Thus if one believes in the suggested interpretation, as I think one should, there is no alternative to abandoning the framework of classical mechanics and saying that persistent currents must be a quantum phenomenon. It will be seen that this postulate is at least tenable in principle.

For this purpose, let Ψ denote a rigorous solution of the Schrödinger equation depending on all the coördinates of the electrons and E the corresponding eigenvalue of their energy. For sufficiently small values of ΔP and with X still denoting the coördinate of the center of gravity, it is possible to construct a "neighboring" solution

$$\Psi' = \Psi \exp\left(2\pi i \Delta P X/h\right) \qquad (2)$$

signifying an increment ΔP of the total momentum in the x direction. The corresponding increment of the energy is obtained by a perturbation treatment and is given by

$$\Delta E = V\Delta P + O\left(\Delta P^2\right) \qquad (3)$$

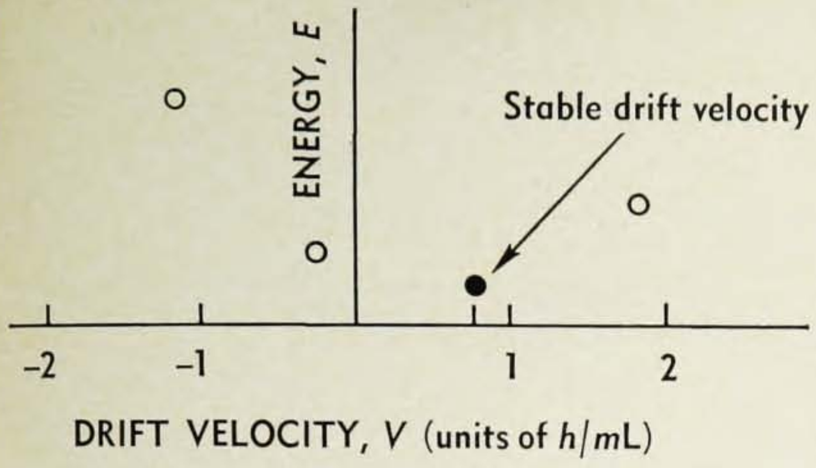
V stands for the expectation value of the x component of the drift velocity in the original state of the electron system described by the wave-function Ψ. The second term in equation 3 is to the lowest order quadratic in ΔP so that one would obtain the result $\Delta E/\Delta P = V$, equivalent to that of equation 1, in the limit in which ΔP reaches zero by continuous variation. The essential change from classical mechanics, however, is that this limiting process is not generally permissible. Indeed, with the electrons confined in a narrow wire loop of circumference L and with the x coördinate measured along the loop, a change by the amount L of this coördinate for any one of the electrons brings it back to the same position and, hence, may not affect the value of either Ψ or Ψ'. For a total number N of electrons this implies a change of the coördinate X of the center of gravity by the amount L/N; since this change must leave the exponential in equation 2 unaltered, it follows that ΔP can only assume the discrete set of values

$$\Delta P = n(Nh/L) \qquad (4)$$

where n is an integer. The corresponding increment of the drift velocity is obtained by dividing ΔP by the total mass $M = Nm$, where m is the mass of the electron, and is thus given by

$$\Delta V = n(h/mL) \qquad (5)$$

Going back to equation 3, requiring the original state to be that with the lowest possible energy E of the system means that the increment ΔE cannot become negative for either sign of ΔP. Instead of necessarily demanding that the drift velocity V be zero, this condition can also be satisfied as long as V does not exceed a value of order of magnitude h/mL. It is important to notice that this value goes to zero not only for h going to zero (that is, in the classical limit) but also for L going to infinity. This already indicates that the finite macroscopic dimensions of the system are essential to the explanation of persistent currents, a circumstance that has been overlooked in early theories. Taking finite dimensions into account, however, one can envisage a finite drift velocity at the lowest energy in the manner schematically indicated in figure 1. The circles in this figure represent the energy E for discrete values of the drift velocity V spaced (according to equation 5) by the amount h/mL. The filled circle indicates the lowest energy at the correspondingly stable



ALLOWED ELECTRON ENERGY E near a minimum for discrete values of drift velocity V, separated by amounts h/mL. Filled circle shows lowest energy at correspondingly stable drift velocity. —FIG. 1

drift velocity, chosen to be finite and of order of magnitude h/mL. It is of course quite arbitrary to assume that the lowest energy corresponds to a positive value of V since it could equally well occur at negative values. Either choice implies the existence of a preferred direction and we shall directly come back to this point.

All the preceding considerations apply not only to arbitrary interactions between the electrons but also to an arbitrary scalar and vector potential acting upon each of them. In this context it was pointed out by Fritz and Heinz London[3] in 1935 that it is particularly the presence of a vector potential that is essential to provide the preferred direction implied in figure 1. This idea may not seem unfamiliar if one thinks of diamagnetism in which the vector potential is likewise responsible for the persistent Amperian current in the atom. There is, however, the very significant difference that the length L in diamagnetism is of atomic size whereas the persistent currents of superconductivity require L to be a macroscopic dimension. In fact, it was London who first stated that superconductivity represents a quantum phenomenon on macroscopic scale.

To recall briefly the use of the vector potential in London's phenomenological theory, one of its components will be considered to have the magnitude A. The relation between the corresponding component of velocity v and momentum p of an electron is then given by

$$v = \frac{1}{m}\ \left(p - \frac{e}{c}A\right) \tag{6}$$

where e is the charge of the electron and c the velocity of light. In the absence of a preferred direction, that is, for $A = 0$, the average values $\overline{v}$ and $\overline{p}$ of velocity and momentum, respectively, must both vanish. An assumed adiabatic change of the vector potential from zero to the finite value A leaves the average momentum at its original value $\overline{p} = 0$ but results according to equation 6 in a drift velocity

$$v = V = -\,eA/mc \tag{7}$$

and, hence, in a current density

$$i = \rho V = -\,\frac{\rho e}{mc}\,A \tag{8}$$

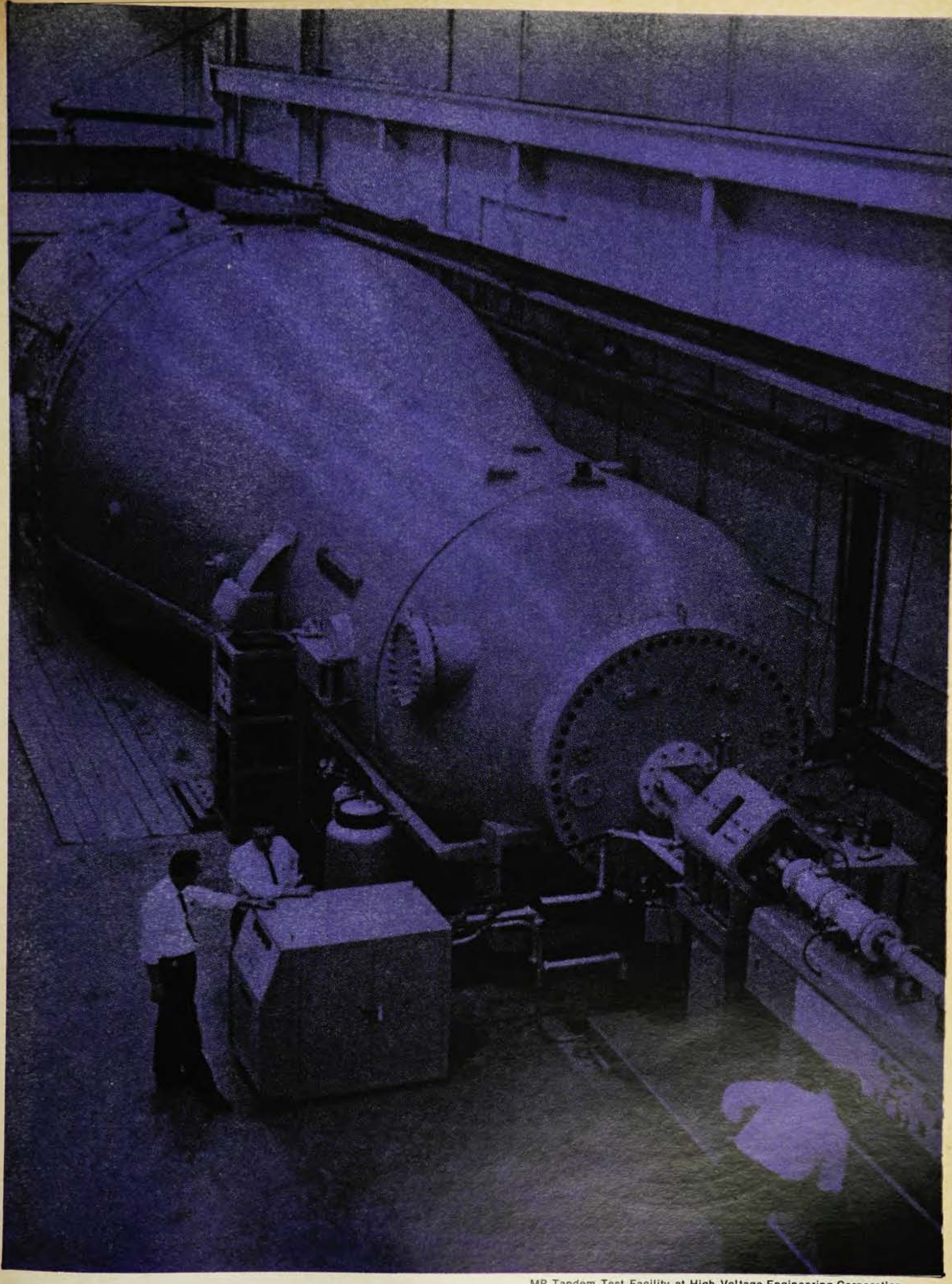
where ρ denotes the charge density of the electrons. With this equation, valid for each component of current density and vector potential, one thus obtains for the curl of these vectors London's famous relation between the current density and the magnetic field $\mathbf{H} = \text{curl}\ \mathbf{A}$

$$\text{curl}\ \mathbf{i} = -\,\frac{\rho e}{mc}\,\mathbf{H} \tag{9}$$

In contrast to this relation, there is evidently something peculiar about equation 8, from which it was derived, since equation 8 is not gauge invariant. For example, it can have no physical consequence to add a constant to the vector potential and yet this would lead to a corresponding additive change in the current density if the equation were taken too literally. Nevertheless, it is seen to contain a grain of truth when one considers a closed conductor.

Such a conductor is indicated in figure 2 as a circular loop of mean radius R and circumference $L = 2\pi R$, v and A denoting tangential velocity and vector potential respectively. For simplicity the loop shall be assumed sufficiently thin to neglect the radial variation of A. The line integral of the vector potential along a closed path within and around the loop has then the value

$$LA = \phi \tag{10}$$



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due to Stokes' theorem; the quantity ϕ signifies the magnetic flux through the loop. Since the angular momentum of an electron is given by

$$pR = n(h/2\pi) \tag{11}$$

for integral n, the eigenvalues of the momentum p are given by

$$p_n = n(h/2\pi R) = n(h/L) \tag{12}$$

and, in view of equation 6, the eigenvalues of velocity v are given by

$$v_n = \frac{1}{m}\left(p_n - \frac{e}{c}A\right) = \frac{h}{mL}\left(n - \frac{eLA}{hc}\right)$$

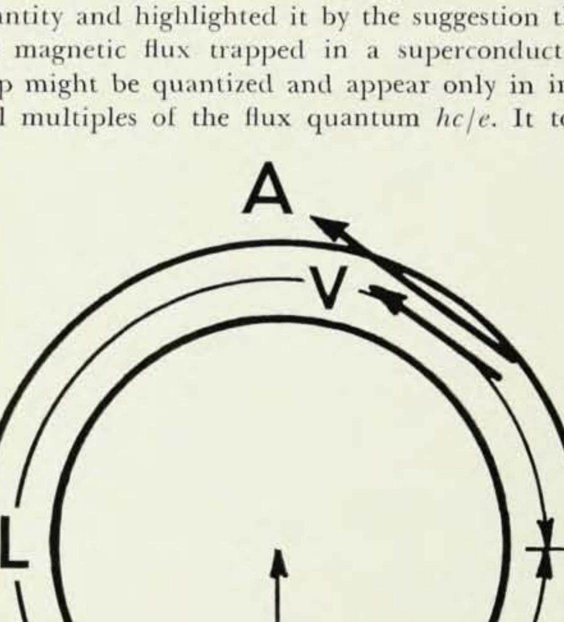
Substituting by means of equation 10, we can write the preceding equation in the form

$$v_n = \frac{h}{mL}(n - \alpha) \tag{13}$$

with the dimensionless quantity

$$\alpha = \phi / (hc/e) \tag{14}$$

There are two points that should be particularly noticed. The first is that in the expression for v_n there appears again the characteristic velocity h/mL that we met before. The second is that the quantity hc/e enters here as a natural unit in which to measure the flux. It was again Fritz London[4] who in 1948 emphasized the importance of this quantity and highlighted it by the suggestion that the magnetic flux trapped in a superconducting loop might be quantized and appear only in integral multiples of the flux quantum hc/e. It took more than ten years for this suggestion to be taken seriously and a full understanding of flux quantization was not reached until after it had been experimentally demonstrated by Bascom S. Deaver and William M. Fairbank[5] as well as by R. Doll and M. Naebauer[6] in 1961. These experiments, in fact, mark the starting point of the recent theoretical developments which will now be discussed.



CIRCULAR CURRENT LOOP with tangential vector potential A and velocity component v of an electron. —FIG. 2

Much of these developments is due to C. N. Yang[7,8] and a greatly simplified part of his ideas can be applied to the case of a thin circular loop considered before.[9] It is suitable for this purpose to introduce the velocity distribution function $f(v)$ with the significance that $f(v_n)$ represents the number (or, more rigorously, the statistical expectation value of the number) of electrons with the velocity v_n given by equation 13. The drift velocity is then

$$\bar{v} = V = \frac{1}{N}\sum_{n=-\infty}^{+\infty} v_n\, f(v_n) \tag{15}$$

and the current, $I = NeV/L$, around the loop can be written in the form

$$I = I(\alpha) \tag{16}$$

where, in view of equations 13 and 15

$$I(\alpha) = \frac{eh}{mL^2}\sum_{n=-\infty}^{+\infty} (n-\alpha)\, f\left[\frac{h}{mL}(n-\alpha)\right] \tag{17}$$

For a given function $f(v)$ this quantity depends only on α and has two important general properties: In the first place, the sign of the current is determined by the sense of rotation, implied by the sign of the flux ϕ or of α, and must reverse upon a reversal of the flux so that

$$I(-\alpha) = -I(\alpha) \tag{18}$$

and, in particular

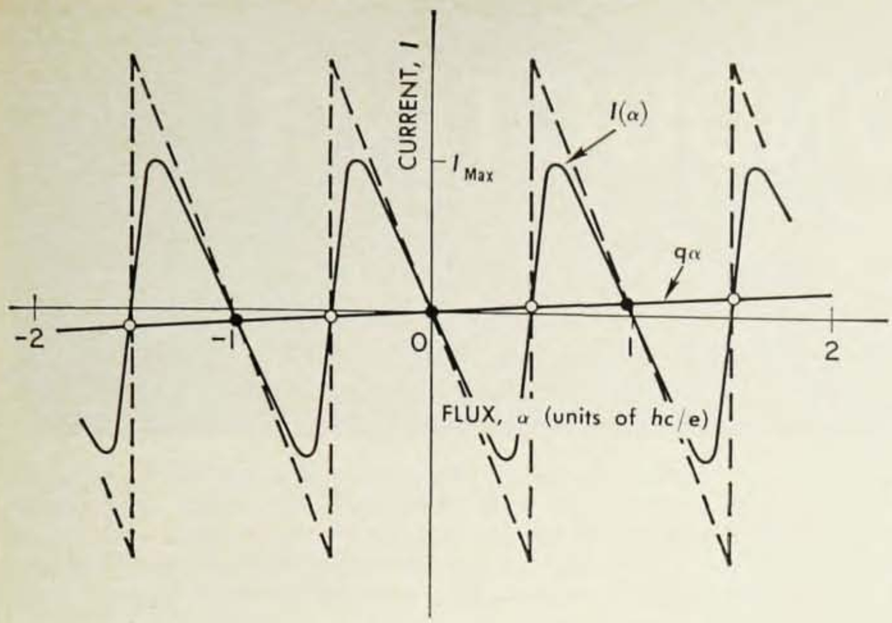
$$I(0) = 0 \tag{19}$$

In the second place $I(\alpha)$ is a periodic function of α with period unity so that

$$I(\alpha + 1) = I(\alpha) \tag{20}$$

since an increase of α by unity is equivalent to a relabelling of the summation index n to $n + 1$, thus leaving the sum in equation 17 unchanged.

Besides the relation of equation 16 between current and flux, the latter measured in units hc/e by the quantity α, there exists the linear relation



PLOT OF CURRENT VS FLUX α in a thin circular loop. Periodic curve $I(\alpha)$ represents current caused by vector potential; straight line $q\alpha$ results from Ampere's law. —FIG. 3

$$I = q\,\alpha \tag{21}$$

This follows from Ampere's law, provided that external sources of the magnetic field are excluded, with the constant q determined by the dimensions of the loop.

From the two equations 16 and 21 for the two quantities I and α, the solutions for both can be obtained. This is graphically demonstrated in figure 3 where the periodic curve, schematically representing the function $I(\alpha)$, and the straight line, representing $q\alpha$, are indicated. Their intercepts yield the solutions, that is, the possible values of α and the corresponding values of I. One can show that these values occur at extrema of the free energy and that the filled circles refer to a minimum and, hence, to a stable equilibrium whereas the open circles refer to a maximum and are not significant since they pertain to an unstable situation. The broken periodic curve represents a special case to be mentioned later. Since the slope q of the straight line diminishes with increasing radius R of the loop and is already quite small for any sizable value of R, it is seen that the stable values of α are practically intergers and that, according to equation 14, this fact explains the quantization of the flux ϕ. That the permissible values of ϕ and, hence, of the magnetic field are limited by the indicated finite amplitude I_{max} of the current suggests an explanation of the observed critical field beyond which superconductivity ceases to exist.

These explanations look deceptively simple; no more than the most elementary quantum theory is apparently required to understand both persistent currents and flux quantization, and one may well wonder why this was not already known about 40 years ago. In the first place, however, hindsight is always much better than foresight, and, in the second place, the difficulties mentioned before are in reality still there although they are quite hidden. In fact, it is not even evident how the particular properties of a superconductor enter at all into the preceding considerations. The reasoning seems so general that it is understandable why the idea has cropped up, at some stage, that every magnetic flux surrounded by a closed curve might be quantized under all circumstances. While it may seem plausible, however, to assume a finite value of I_{max} as shown in figure 3, it is by no means easy, on second thought, to justify this assumption. One must not forget that the amplitude of a periodic function could also vanish and the following considerations will show that this is just what one normally would have to expect.

It is necessary for this purpose to examine closely the orders of magnitude involved. Assuming for example the radius of the loop to be $R = 1$ cm, the velocity h/mL is found to be about 1 cm/sec. Although a comparable drift velocity, combined with the high density of conduction electrons in a metal, can lead to sizable current densities, it is important to notice that this value is extremely small compared to the typical range of electron velocities. This range, which characterizes the spread of the velocity distribution function $f(v)$, is of the order of magnitude of the Fermi velocity $v_F \approx 10^8$ cm/sec and with a function of such a large spread the replacement of the sum in equa-

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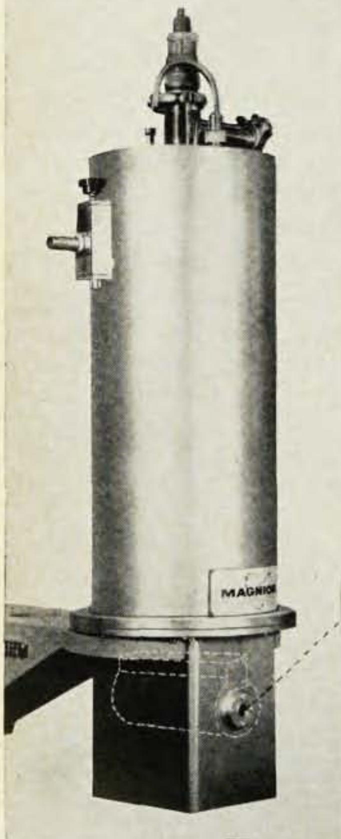

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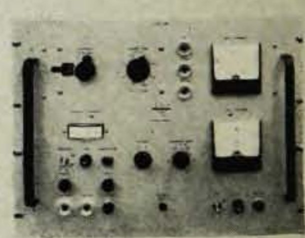

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tion 17 by an integral would normally be permitted as an excellent approximation. Yet, to concede the admissibility of this approximation is fatal to the proposed explanation of persistent currents. Indeed, the replacement of equation 17 by

$$I(\alpha) = \frac{eh}{mL^2} \int_{-\infty}^{+\infty} (x - \alpha)\, f\left[\frac{h}{mL}\,(x - \alpha)\right] dx$$

leads by the mere change of the integration variable x into $x + \alpha$ to

$$I(\alpha) = \frac{eh}{mL^2} \int_{-\infty}^{+\infty} x\ f\left[\frac{hx}{mL}\right]\, dx$$

an expression that is independent of α. On the other hand this expression must vanish for $\alpha = 0$, as stated in equation 19, and it follows therefore, that

$$I(\alpha) \equiv 0$$

This is actually the situation that one meets in the normal state of a metal, and the conclusion that no other than a vanishing current can occur at a stable equilibrium is here perfectly correct. The velocity distribution function evidently has to undergo some drastic change at the transition to the superconductive state to avoid this conclusion. In fact, quite peculiar properties of this function are required to introduce a serious error in the integral approximation of the sum in equation 17 and the finite value of the characteristic velocity h/mL, although very small for macroscopic dimensions, must somehow be reflected in these properties.

The usefulness of the degenerate ideal Bose gas as a model rests largely on the circumstance that it furnishes an example of such a peculiar velocity distribution. Because of Einstein-Bose condensation, one single term in the sum gives a contribution here that is comparable to or larger than that of all other terms together whereas replacement of the sum by the integral demands that consecutive terms differ only inappreciably from each other. Although Schafroth deduced only the Meissner effect (that is, expulsion of the magnetic field from the interior of the superconductor) from his model, it is not surprising that his model yields at the same time persistent currents and flux-quantization. The broken curve in figure 3 represents the special result at zero temperature, thus derived, when only one term in the sum does not vanish. Another equivalent way of describing this feature of a degenerate Bose gas is to say that the phase relation of the particle wavefunctions is maintained over the whole circumference of the loop or, in rather technical terms, that the reduced density matrix exhibits off-diagonal long-range order. This property has been thoroughly investigated in an important paper by Yang[8] in 1962.

The situation, however, is radically different for a Fermi gas where the exclusion principle prohibits dominance of any single term in the sum of equation 17. Indeed, an *ideal* Fermi gas is very well suited to the replacement of this sum by an integral and the resulting absence of persistent currents can equivalently be deduced from absence of off-diagonal long-range order in this case. On the other hand, Yang has shown that a *nonideal* Fermi gas can exhibit a type of long-range order that, instead of referring to single particles, as in the Bose gas, refers to *pairs* of particles. In fact the greatest progress in the theory of superconductivity lies in the recognition, originally built into the theory of Bardeen, Cooper and Schrieffer, that the interaction of the conduction electrons leads to their pairing in the superconductive state. This has been brilliantly borne out in the previously mentioned experiments on flux-quantization by the result that the flux quantum is not hc/e but $hc/2e$, which shows that the carriers of electricity in a superconductor have twice the charge of the electron.

As I stated in the beginning, I wanted mainly to discuss some of the features of superconductivity that can be treated in terms of rather general principles, and I hope that I have shown that

A Presidential Tradition

It seems to become one of the many cherished traditions of the American Physical Society that every third retiring presidential address somehow refers to the theory of superconductivity. Thus in 1960, George E. Uhlenbeck (PHYSICS TODAY 13, no. 7, 18, 1960) expressed in his address the thought that the theory of superconductivity "is still a bit controversial." In 1963, W. V. Houston (PHYSICS TODAY 16, no. 9, 36, 1963) stated on the same occasion his belief that "a simple physical picture of superconductivity still remains to be drawn from the theoretical work that has been carried out." Apparently it is indicated at this time to look again at the situation and to see what has happened along the lines desired by my predecessors. Although further progress has been made, I am afraid that I shall be unable to fulfill all their wishes; it is my hope, however, that the tradition will be maintained and that the retiring president will favor us in 1969 with a comprehensive account of the insights achieved.

one can go quite far in this manner. I certainly do not wish to imply, however, that the necessity of a detailed microscopic theory can thus be removed. Such an approach merely helps to clarify the goal to be reached, and I want to end with a brief discussion of how close, in my opinion, we are to this goal. Its ultimate attainment clearly hinges on a thorough understanding of interaction among the conduction electrons so that pairing and the ensuing off-diagonal long-range order, necessary to explain superconductivity, can be deduced without further assumptions. The difficulty of this undertaking must not be underestimated, particularly since it cannot be based on perturbation methods, and the great progress achieved by the theory of Bardeen, Cooper and Schrieffer rests indeed on an entirely different method. It can be characterized either as the use of trial functions, improved over those of the independent-particle picture by allowing correlations of two particles with opposite velocities, or as retaining in the Hamiltonian only those parts of the interaction that are responsible for these special correlations. I believe that the doubts in the finality of the theory expressed by my predecessors [10, 11] arise to a considerable extent from the somewhat arbitrary nature of this method. Although the results obtained from its application are strongly supported by the observed facts and are closely linked to the continuing and splendid experimental developments, I agree that a more thorough analysis of the foundations is required to make the procedure totally convincing to the theorists.

So far, such an analysis has been carried out only for the one-dimensional case. The interest in this case is not purely theoretical since it was suggested[12] that certain organic chains might exhibit superconductivity, possibly even at room temperature. One succeeds here in treating the full effect of the interactions as long as their range is long compared to the Fermi wavelength and their strength small compared to the Fermi energy. Both of these properties are usually accepted and they are certainly compatible with the orders of magnitude required by superconductivity. Just as in the three-dimensional case, it is found for attractive interactions in one dimension that a trial function that provides for pairing of electrons with opposite velocities yields a ground state of lower energy than the one obtained under the assumption of independent particles; thus in this sense, an improvement has been achieved. The correct and, in fact, still considerably lower energy, however, is obtained from an entirely different and well known feature of many-body problems. This feature consists of the collective modes (analogous to sound waves) sustained by the electron system; they contain the plasma oscillations as a particular consequence of Coulomb repulsion. The collective modes are equally suitable for taking attractive interactions into account, and the problem can be solved, at least for the low-lying states, quite similarly to that of lattice vibrations by the use of normal modes. In Houston's 1963 address[11] he raised the question whether these collective modes could not be gainfuly used to bring in the interactions of electrons; it seems, at least for the one-dimensional case, that the answer is emphatically, "Yes!" By doing just that, one finds that the interactions merely modify the frequency of a sound wave of given wavelength and fail to result in the far more drastic changes required for the existence of persistent currents.[13]

A thorough treatment of the interactions in the two- and three-dimensional cases has not been achieved yet and one knows that the role of collective modes is greatly altered here. For repulsive interactions, this is manifested by the appearance of the so-called "zero-sound," first pointed out by Lev D. Landau[14] and there can be little doubt that the quite different manifestation of attractive interaction in a three-dimensional Fermi gas will contain the pairing of particles as one of its essential attributes. Much progress toward the full understanding of this feature has already been made, but I believe that more is required before one can safely claim that the theory of superconductivity is complete. □

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