

Connection between Superfluidity and Superconductivity

M. R. SCHAFFROTH

The F. B. S. Falkiner Nuclear Research and Adolph Bassler Computing Laboratories, School of Physics,
The University of Sydney, Sydney, N.S.W., Australia*

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A general connection between equilibrium superfluidity and superconductivity is established. This connection allows us to apply to superconductivity a previously established theorem on the nonexistence of equilibrium superfluidity in actual physical systems. It follows that London's equations can only be approximately valid and have to be modified. An alternative equation proposed by Pippard is also in contradiction with the theorem.

1. INTRODUCTION

IT has long been suspected that there exist fundamental links between the two low-temperature phenomena of superfluidity and superconductivity.^{1,2}

Recently, such a connection has become more apparent through the exhibition of a common model for the two phenomena: the perfect Bose-gas. It was shown that a gas of charged bosons below condensation is a superconductor.³

Furthermore, it has been shown that for a small range of angular velocities, the perfect Bose gas also exhibits superfluid properties.⁴ It is the purpose of this paper to establish the validity of the connection between the two phenomena beyond the range of the Bose-gas model and to show that the two phenomena are related quite generally. More specifically, we shall define "perfect superconductivity" and "equilibrium superfluidity" of a physical system as being those properties which are actually exhibited by the Bose gas and then prove that for a system of identical spinless particles one of these properties entails the other. For systems of nonidentical particles cancellations can occur which destroy one effect, and therefore exceptions to the theorem are possible. Similar cancellations may occur in the case of particles with spin, namely when the contribution of the spins just cancels the contribution of the orbits. Such cancellations do not, however, affect the general connection between the two phenomena of superfluidity and superconductivity.

Beyond establishing the reason for the existence of a common model, the Bose gas, for the two phenomena, the above theorem yields important information about the superconductivity of real metals when viewed in connection with a recently established theorem on superfluidity in real physical systems.⁵ It was pointed out there that for every real physical system, (i.e., a

system of interacting particles) at a finite temperature there exists a "correlation length Λ " such that the momentum correlations of two particles farther apart than Λ decrease very rapidly. It was then proved⁵ that no system with finite Λ , i.e., no real physical system, can show equilibrium superfluidity. In conjunction with the present theorem this entails that no superconducting metal can be a "perfect superconductor." It will be shown in Sec. 2 that a system obeying London's phenomenological equations⁶ of superconductivity is a perfect superconductor. We can, therefore, conclude that the London equations are incompatible with the finiteness of the correlation length and can, therefore, only be approximately valid. Similarly, it can be shown that the modified equations proposed by Pippard⁷ are inadmissible for the same reason. A more detailed discussion of this point will be given in a subsequent publication.

A word must be added at this stage about the question of how far one is justified in drawing from our theorem any conclusions about the behavior of actual superconductors. The possibility of accidental cancellations mentioned earlier presents no difficulty here, because the proof that the London equations are incompatible with the existence of a finite correlation length is independent of the actual occurrence of such cancellations. Apart, therefore, from the remote possibility that superconductivity be an effect of the spins rather than orbital motions—which seems to be excluded both by the existence of charge-carrying supercurrents and by the essentially diamagnetic nature of the Meissner-Ochsenfeld effect—our theorem can be applied to actual superconductors.

2. DEFINITIONS

We first define our terms precisely.

A. Consider a system in thermal equilibrium rotating with angular velocity ω around the z -axis and denote its angular momentum by $L(\omega)$. The quantity

$$I \equiv (dL/d\omega)_{\omega=0} \quad (1)$$

⁶ See reference 2, paragraph 8.

⁷ A. B. Pippard, Proc. Roy. Soc. (London) **A216**, 547 (1953).

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¹ J. G. Daunt and K. Mendelssohn, Nature **150**, 604 (1942).

² F. London, *Superfluids I* (John Wiley and Sons, Inc., New York, 1950), Introduction.

³ M. R. Schafroth, Phys. Rev. **96**, 1149 (1954), and this issue [Phys. Rev. **100**, 463 (1955)].

⁴ J. M. Blatt and S. T. Butler, this issue [Phys. Rev. **100**, 476 (1955)].

⁵ Blatt, Butler, and Schafroth, this issue [Phys. Rev. **100**, 481 (1955)].

is the moment of inertia. Its classical value is, for a system of N identical particles with mass M

$$I_0 = NM \langle x^2 + y^2 \rangle, \tag{2}$$

where $\langle \rangle$ denotes the average in thermal equilibrium.

Definition I: We call "equilibrium superfluid" a system for which $(I - I_0)/I_0$ does not decrease indefinitely as $\langle x^2 + y^2 \rangle$ is increased, i.e.,

$$(I - I_0)/I_0 \geq \eta > 0 \text{ for all } \langle x^2 + y^2 \rangle. \tag{3}$$

Note 1: It is known⁴ that the ideal Bose gas is an equilibrium superfluid in this sense. It is also known⁴ that equilibrium superfluidity is not enough to ensure actual superfluid behavior.

B. Perfect Superconductivity

Consider a system of charged particles in a fixed cylindrical volume V in a homogeneous magnetic field H parallel to the axis of the cylinder. Denote the total magnetic moment this system acquires in thermal equilibrium by $M(H)$. We shall define a quantity

$$\chi'(V) \equiv \frac{1}{V} \left(\frac{\partial M}{\partial H} \right)_{H=0}. \tag{4}$$

For any system with paramagnetic or diamagnetic behavior, χ' is independent of V .

Note 2: The quantity χ' here is not necessarily identical with the magnetic susceptibility χ as usually defined. The latter is

$$\chi = \lim_{H \rightarrow 0} \lim_{\Delta H \rightarrow 0} \lim_{V \rightarrow \infty} \left(\frac{1}{V} \frac{\Delta M}{\Delta H} \right), \tag{5}$$

whereas (4) means, for large volumes

$$\chi'(\infty) = \lim_{V \rightarrow \infty} \left(\frac{1}{V} \lim_{H \rightarrow 0} \lim_{\Delta H \rightarrow 0} \frac{\Delta M}{\Delta H} \right). \tag{6}$$

The limiting processes are not necessarily interchangeable, and so χ' and χ may turn out to be different. However, we are not concerned here with the value of the susceptibility; we only ask whether the last limiting process in (6) exists or not; i.e., whether χ' approaches a finite limit or whether it increases indefinitely with the dimensions of the container. This question, however, makes clearly no sense in relation to the susceptibility χ .

Definition II: A system for which

- (a) $\chi' < 0$
- (b) $-\chi'$

increases indefinitely, $\propto R^2$, with the radius R of the cylinder, shall be called a "perfect superconductor."

Definition IIa: A system for which

- (a) $\chi' > 0$
- (b) $\chi' \propto R^2$

shall be called a "perfect ferromagnet."

Theorem I: A system for which London's equation for the current density⁸

$$-\lambda c \text{curl} \mathbf{i} = \mathbf{B} \tag{7}$$

⁸ See reference 2, paragraph 3.

holds, is a perfect superconductor. In actual superconductors, the only source of \mathbf{B} is the supercurrent \mathbf{i} itself, so that in Maxwell's equation

$$\text{curl} \mathbf{H} = (4\pi/c) \mathbf{i} \tag{8}$$

one has to put $\mathbf{H} = \mathbf{B}$. (7) and (8) together, then, do not allow any homogeneous field to penetrate. However, as has been pointed out earlier,⁹ it is a consistent "gedanken-experiment" to allow for a magnetic polarization $\mathbf{P}(x)$ inside the superconductor (arising e.g., out of a polarization of the nuclear moments) which has no other interaction with the superconducting particles than the magnetic one. The result is that, since now $\mathbf{B} = \mathbf{H} + 4\pi \mathbf{P}$, a homogeneous \mathbf{B} is possible, and (8) can be disregarded, its only function being to determine the necessary polarization \mathbf{P} .

For a cylindrical container with \mathbf{B} parallel to the axis (7) at once yields

$$i_z = i_r = 0; \quad i_\phi = -\frac{1}{\lambda c} \frac{r}{2} B. \tag{9}$$

Therefore,

$$\mathbf{M} = \frac{1}{2} \int d^3x (\mathbf{r} \times \mathbf{i}), \tag{10}$$

which yields

$$M_x = M_y = 0,$$

and

$$M_z = -(1/8\lambda c) B V R^2, \tag{11}$$

and therefore,

$$\chi' = -(1/8\lambda c) R^2. \tag{12}$$

This proves our assertion.

3. CONNECTION THEOREM

We wish now to prove the following theorem, which forms the main body of this paper:

Theorem II. If a system of identical spinless particles is an equilibrium superfluid, then the same system, when the particles are given a test charge ϵ , is also a perfect superconductor; and, conversely, any perfect superconductor consisting of identical spinless particles, is an equilibrium superfluid.

More in detail, we shall show that the moment of inertia I (1) and the quantity χ' (4) are related by

$$I = I_0 \left(1 + \frac{2\chi'}{n(x^2 + y^2)} \cdot \frac{M c^2}{\epsilon^2} \right) \tag{13}$$

where n is the density of particles. (Note that χ' is proportional to ϵ^2 , so that the test charge ϵ actually drops out.)

Note 3: The restriction to spinless particles may easily be removed, at least as long as one neglects any spin-orbit coupling. One can then define uniquely an orbital part of χ' , χ'_l and a spin

⁹ M. R. Schafroth, *Helv. Phys. Acta* **24**, 645 (1951).

part χ_2' . In terms of these one finds that (13) is replaced by

$$I = I_0 \left[1 + \frac{2(\chi_1' + g^{-1}\chi_2')}{n(x^2 + y^2)} \frac{Mc^2}{e^2} \right], \quad (13')$$

where g is the g -factor of the particles. This can lead to an exception from the theorem, when $\chi_1' \propto \langle x^2 + y^2 \rangle$, $\chi_2' \propto \langle x^2 + y^2 \rangle$, but $\chi_1' + g^{-1}\chi_2' = o(\langle x^2 + y^2 \rangle)$; i.e., when the system is at the same time a perfect superconductor and a perfect ferromagnet, in such a ratio that the two susceptibilities just cancel. This case is clearly too exceptional to warrant the complication of carrying through the proof of (13') in detail.

Note 4: Similarly, the restriction to identical particles is not very serious. Taking r kinds of particles with charges ϵ_i , masses M_i , densities n_i and g -factors g_i , (13') has to be replaced by

$$\frac{I - I_0}{I_0} = \sum_{i,j=1}^r (C_{ij}^{(1)} + C_{ij}^{(2)}),$$

$$\chi' = \frac{1}{2Vc^2} \sum_{i,j=1}^r (C_{ij}^{(1)} + g_i g_j C_{ij}^{(2)}) \frac{\epsilon_i \epsilon_j}{M_i M_j}, \quad (13'')$$

where $C_{ij}^{(1)}$, $C_{ij}^{(2)}$ are certain coefficients. For the system to be either an equilibrium superfluid, a perfect superconductor or a perfect ferromagnet, at least one of the C 's has to be $\propto \langle x^2 + y^2 \rangle$. It can, however, happen that for certain values of ϵ_i , m_i , g_i one of the two expressions $(I - I_0)/I_0$ and χ' is $\propto \langle x^2 + y^2 \rangle$, whereas in the other the corresponding terms just cancel. Accidental cancellations, therefore, are possible which invalidate the theorem.

We now proceed to prove (13). The Hamiltonian for N identical spinless particles of mass M and charge ϵ in a volume V under the influence of a homogeneous magnetic field H_{\parallel} is

$$H = \sum_{i=1}^N \frac{1}{2M} \left(\mathbf{p}_i - \frac{\epsilon}{2c} \mathbf{H} \times \mathbf{r}_i \right)^2 + V(\mathbf{r}_1, \dots, \mathbf{r}_N). \quad (14)$$

The magnetic moment operator is

$$\mathbf{u} = \sum_{i=1}^N \frac{\epsilon}{M c} \mathbf{r}_i \times \left(\mathbf{p}_i - \frac{\epsilon}{2c} \mathbf{H} \times \mathbf{r}_i \right). \quad (15)$$

The average magnetic moment in thermal equilibrium is, therefore,

$$\mathbf{M} = \langle \mathbf{u} \rangle = \text{Trace} \left\{ \left[\sum_k \frac{\epsilon}{M c} \mathbf{r}_k \times \left(\mathbf{p}_k - \frac{\epsilon}{2c} \mathbf{H} \times \mathbf{r}_k \right) \right] \right.$$

$$\left. \times \exp \left[-\beta \left(\sum_i \frac{1}{2M} \left(\mathbf{p}_i - \frac{\epsilon}{2c} \mathbf{H} \times \mathbf{r}_i \right)^2 \right) + V \right] \right\}, \quad (16)$$

where the trace has to be extended over the space of all properly symmetrized states, and where $\beta = 1/kT$.

Introducing the Larmor angular velocity

$$\omega_0 = e\mathbf{H}/2Mc \quad (17)$$

(16) can be written

$$\mathbf{M} = \frac{\epsilon}{M c} \text{Trace} \left\{ \left[\sum_k (\mathbf{r}_k \times \mathbf{p}_k - M \mathbf{r}_k \times (\omega_0 \times \mathbf{r}_k)) \right] \right.$$

$$\left. \times \exp \left[-\beta \sum_i \left(\frac{1}{2M} (\mathbf{p}_i - M \omega_0 \times \mathbf{r}_i)^2 + V \right) \right] \right\}. \quad (18)$$

On the other hand, the angular momentum in thermal equilibrium of the same system without magnetic field, but rotating with angular velocity ω_0 , is given by taking the average of the angular momentum operator

$$\mathbf{L} = \sum_k \mathbf{r}_k \times \mathbf{p}_k \quad (19)$$

over a canonical ensemble with Hamiltonian¹⁰

$$H' = \sum_{i=1}^N \frac{1}{2M} \mathbf{p}_i^2 + V(\mathbf{r}_1, \dots, \mathbf{r}_N) - \omega_0 \cdot \mathbf{L}, \quad (20)$$

or

$$H' = \sum_{i=1}^N \left[\frac{1}{2M} (\mathbf{p}_i - M \omega_0 \times \mathbf{r}_i)^2 - \frac{M}{2} (\omega_0 \times \mathbf{r}_i)^2 \right]$$

$$+ V(\mathbf{r}_1, \dots, \mathbf{r}_N). \quad (20')$$

One finds, therefore

$$\mathbf{L}(\omega) = \text{Trace} \left\{ \sum_k (\mathbf{r}_k \times \mathbf{p}_k) \exp(-\beta H') \right\}. \quad (21)$$

Comparing (21) with (18) we see that, neglecting terms of second order in ω_0 , one has

$$\mathbf{M} = \frac{\epsilon}{M c} \mathbf{L}(\omega_0) - \frac{\epsilon}{M c} \text{Trace} \left\{ \left[\sum_k \mathbf{r}_k \times (\omega_0 \times \mathbf{r}_k) \right] \right.$$

$$\left. \times \exp \left[-\beta \sum_i \frac{1}{2M} \mathbf{p}_i^2 + V \right] \right\}. \quad (22)$$

Taking ω_0 to lie in the z -direction this reads

$$\mathbf{M} = \frac{\epsilon}{M c} \mathbf{L}(\omega_0) - \frac{\epsilon N}{c} \omega_0 \langle x^2 + y^2 \rangle, \quad (23)$$

where $\langle \rangle$ is the average over the equilibrium distribution at $\omega_0 = 0$. Differentiating with respect to ω_0 and using (1), (2), (4), and (17) this readily gives (13).

The essential step in this proof is the expansion of (16) and (21) in powers of ω_0 , keeping only linear terms. This is only justified by our particular definition of I , (2) and χ' , (4) which is restricted to finite volumes. The expansions in power of ω_0 will, as a rule, be justified only for very small angular velocities, $\omega_0 < \omega_1 \sim \hbar/MR^2$. Therefore this procedure cannot be used for computing the actually measured quantities χ , (5) and

$$I' = \frac{\Delta L(\omega)}{\Delta \omega}, \quad (24)$$

with

$$\Delta \omega \gg \omega_1.$$

However, for the purpose of this paper, the foregoing procedure is justified.

¹⁰ See the detailed discussion of this in Blatt, Butler and Schafroth, this issue [Phys. Rev. **100**, 481 (1955)].

4. DISCUSSION

It has been shown⁵ that no real physical system can be an equilibrium superfluid. The reason for this is that for all systems of interacting particles there exists a correlation length Λ which is an intrinsic property of the system, independent of the size of the volume, such that particles which are apart by a distance $>\Lambda$ have strongly decreasing momentum correlations.

The theorem of Sec. 3 together with the above statement that no equilibrium superfluid exists yields at once:

Theorem III: No perfect superconductors exist in nature.

In conjunction with the result of Sec. 2 that a system obeying London's equation is a perfect superconductor, this implies:

Corollary: No physical system can obey London's equation (7) exactly.

The ideal Bose-Einstein gas is not a physical system, since it does have an infinite correlation length⁴; indeed, it obeys London's equation.³

Note 5: The theorem of reference 5 is restricted to particles without spin and does, therefore, not exclude an anomalous moment of inertia I which is larger than the classical value I_0 ; this means that we cannot make a statement about ferromagnets similar to the one about superconductors.

Theorem II therefore leads us to the conclusion that London's equations can only be approximately valid for actual superconductors and that they have to be modified to comply with the finiteness of the correlation length Λ . We shall leave the task of finding an appropriate form of this modification to a later paper.

At present, we only wish to recall that the necessity for modifying London's equations has already been inferred by Pippard⁷ from experimental results on the penetration depth with impurity contents. Pippard proposes the following equation [reference 7, Eq. (7)] to replace London's equation (7):

$$\mathbf{i}(\mathbf{x}) = -\text{const} \int_V d^3x' \frac{e^{-r/\xi}}{r^4} \mathbf{r} [\mathbf{r} \cdot \mathbf{A}(\mathbf{x}')], \quad (25)$$

where $\mathbf{r} = \mathbf{x} - \mathbf{x}'$, ξ is a characteristic length of the material, and \mathbf{A} is the vector potential of the field \mathbf{B} , gauged to $\text{div} \mathbf{A} = 0$ and $(\mathbf{A} \cdot \mathbf{n}) = 0$ at the surface. Equation (25) is, however, also inadmissible from our present

point of view since it still describes a perfect superconductor. This can be seen qualitatively by noticing that Eq. (25) expresses the current density \mathbf{i} as a certain average of the vector potential \mathbf{A} over a region of space of the dimension of the characteristic length ξ . If one considers now a cylinder of radius R in a homogeneous field H_0 , then the vector potential $\mathbf{A} = \frac{1}{2} \mathbf{i} \times \mathbf{H}_0$ increases linearly from the axis to the surface of the cylinder. If the radius of the cylinder is large compared to ξ the current density increases essentially linearly, too, and therefore the argument in Sec. II for the London equations goes through essentially unchanged, leading to a $\chi' \propto R^2$.

Indeed, Pippard's modification of the London equation goes in the opposite direction from the one required by our theorem. The London equation is a strongly nonlocal relation between field and current. The current density is determined only by knowledge of the field \mathbf{B} over the whole volume of superconductor. The fact that it can be brought into the local-looking form

$$\mathbf{i} = -(1/\lambda c) \mathbf{A} \quad (26)$$

[where \mathbf{A} is the vector potential in the same gauge as in (25)] only shows that the vector potential in this gauge is a nonlocal description of the magnetic field. Pippard's procedure of smearing (26) over small volumes tends to make the relation between current and field even less local, whereas the finiteness of the correlation Λ requires that the relation in question should be more local than (26), namely such that the current density is determined by the field distribution within a volume of order Λ^3 .

We therefore conclude that London's equation (7) as well as Pippard's equation (25) are incompatible with a finite correlation length Λ . In a later paper we shall propose modified phenomenological equations for superconductors which are consistent with a finite Λ , and it will be shown then that Pippard's experimental facts become naturally understandable on this new basis, the correlation length Λ taking the role of his parameter ξ .

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