Nodal phase correlation and macroscopic quantum effects in superconducting rings

J. Riess*
Laboratoire de Physique Mathématique, Collège de France, Paris 5e, France
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We introduce the new concept of "nodal phase correlation" (NPC) into many-particle quantum mechanics. NPC is a particular quantum correlation among particles, which is important for current-carrying states of isolated systems. Mathematically it is characterized by a definite relation in configuration space between the phase and the modulus of the many-particle wave function. We illustrate the significance of NPC by showing the crucial role it plays in magnetic flux quantization and in certain macroscopic interference effects in superconducting rings. To this end we partly rederive the theory of these phenomena. Our method is based on the BCS theory. We critically examine previous work, in particular the single-particle pairing theory, which is considered to be a standard approach to magnetic flux quantization. This theory does not give rise to NPC. We are able to show that the corresponding many-particle wave function is in fact unrealistic and not consistent with the BCS theory of superconductivity.

I. INTRODUCTION

We introduce a new concept into many-particle quantum mechanics: *nodal phase correlation.* This concept gives a useful and detailed mathematical characterization of the many-particle wave function of a number of special quantum-mechanical correlation phenomena which occur in current-carrying systems. In this paper we shall discuss the many-particle wave function of a superconducting ring, which is a particularly interesting example of a system where a nodal phase correlation structure can be realized. We shall see that this structure is responsible for the effect of magnetic-flux quantization[1] and for certain macroscopic interference effects depending on the magnetic flux enclosed in the ring (for a review see Ref. 2).

In the literature, however, the relevance of nodal phase correlation to the macroscopic quantum effects mentioned above has not been recognized.[3] In our opinion this can be explained by the fact that nodal phase correlation does not appear in a simple way in the formalism of field theory, which governs modern many-particle theory.

In Sec. II of this paper we develop the concept of nodal phase correlation (NPC) and the more specified concept of n-particle NPC. We then show the relevance of this concept to the general structure of currents in quantum mechanics. In Secs. III and IV we illustrate the significance of NPC by rederiving the effect of quantized magnetic flux and the macroscopic interference effects mentioned above. In particular, our analysis shows that the decisive structure of the many-particle wave function, which is responsible for these effects, is a two-particle NPC. It appears that this fact has so far not been fully recognized. We give a critical discussion of previous standard work about flux quantization, covering in particular the contributions of Brenig,[4] Byers and Yang,[5] and Yang.[6]

II. NODAL PHASE CORRELATION (NPC)

We consider an isolated system of N particles which can be described by a spin-free time-independent Schrödinger Hamiltonian. In an earlier paper[7] we have shown that the phase $\varphi(x)$ of an eigenfunction

$$\psi(x) = R(x)e^{i\varphi(x)} = a(x) + ib(x), \quad x \in R^{3N}$$
$$(R, \varphi, a, b \text{ real}) \tag{1}$$

depends in a critical way on the $(3N-2)$-dimensional nodal[8] hypersurfaces (denoted by M_{3N-2}) and their circulation numbers m_k,

$$m_k = \frac{1}{2\pi} \oint_{P_k} \frac{\partial}{\partial x_\alpha} \varphi(x)\, dx_\alpha, \tag{2}$$

where P_k is a closed path which encircles once the kth $(3N-2)$-dimensional nodal hypersurface, but no other $(3N-2)$-dimensional nodal hypersurface. The circulation numbers m_k are integers.

Simple one-particle examples ($N=1$) are given by the hydrogen states

$$\psi_{n,l,m}(\vec{r}) = q_{n,l,m}(r, \theta)[1 - (\cos\theta)^2]^{m/2} e^{im\alpha} \tag{3}$$

which have a single one-dimensional nodal line (since $N-2=1$) along the z axis ($\theta = 0, \pi$) around which the phase function $\varphi(r) = m\alpha$ (α is the azimuthal angle) is multivalued and where the circulation number is just the familiar magnetic quantum number. For systems with more than one particle the phase function $\varphi(x)$ may be the sum of two real functions h and h', such that h depends

only on n definite particle coordinates $\vec{r}_i$, and h' does not depend on these n coordinates, and where h itself cannot be decomposed into a sum of functions depending on different particle coordinates. Now let us further assume that h is multivalued "around" at least one nodal hypersurface M_{3N-2}. Then the position in configuration space, R_{3N}, of the hypersurface M_{3N-2} can only depend on the n-particle coordinates $\vec{r}_i$, which are arguments of h. We call such a function h a nodal phase. In the case where n is equal to one, i.e., where, e.g., $h=h(\vec{r}_k)$, there is no phase correlation between the kth particle and the rest of the particles. But in the case where n is greater than one, there is a phase correlation between the n particles whose coordinates appear as arguments of h. We call this correlation *n-particle nodal phase correlation.*

The importance of nodal phase structures is illustrated by the following theorem[7]: Consider a spin-free time-independent Schrödinger Hamiltonian which does not contain a magnetic vector potential, and whose coefficients are sufficiently regular (for details see Ref. 7). Let the Hamiltonian be defined in the space of square integrable functions $\mathfrak{L}_2(G)$, where G is a simply connected domain in the configuration space R^{3N}. Let $\psi(x)$ be an eigenfunction. Further let the corresponding N-current density

$$j_\alpha(x)=-\frac{i\hbar}{2M_\alpha}\left(\psi^*(x)\frac{\partial}{\partial x_\alpha}\psi(x)-\psi(x)\frac{\partial}{\partial x_\alpha}\psi^*(x)\right)$$
$$=\frac{\hbar}{M_\alpha}R^2(x)\frac{\partial}{\partial x_\alpha}\varphi(x) \qquad (4)$$

($M_\alpha=M_j$ for $\alpha=3j-2,\ 3j-1,\ 3j$; M_j is the mass of the jth particle; $j=1,2,\ldots,N$), be such that the surface integral of the $3N$-dimensional vector $\varphi(x)j_\alpha(x)$ over the total boundary hypersurface of the domain of the system in the configuration space R^{3N} vanishes. (This is true, e.g., if, in three dimensions, the particle density vanishes at the boundary of a finite domain, e.g., in an isolated piece of solid, as it is considered in Sec. III.) Then the phase function $\varphi(x)$ of the eigenfunction $\psi(x)$ is a constant [apart from possible jumps across nodal $(3N-1)$-dimensional hypersurfaces] unless there exists at least one $(3N-2)$-dimensional nodal hypersurface with nonzero circulation number m. In other words, whenever the phase $\varphi(x)$ differs from a constant, the wave function $\psi(x)$ is zero on at least one $(3N-2)$-dimensional hypersurface M_{3N-2}, and the phase $\varphi(x)$ is multivalued, such that its value changes by $2\pi m$ along any closed path in the $3N$-dimensional configuration space which encircles once a particular hypersurface M_{3N-2} [but no other $(3N-2)$-dimensional nodal hypersurface with nonzero circulation number]. *Other phase structures do not exist under the conditions stated above.* This theorem is a direct consequence of the mathematical structure of the Schrödinger equation in configuration-space representation. It gives the most general structural relation between the phase $\varphi(x)$ and the modulus $R(x)$ of the wave function $\psi(x)$, saying that the mere existence of a nonzero phase gradient $\partial_\alpha\varphi(x)$ implies that the modulus $R(x)$ vanishes on a $(3N-2)$-dimensional hypersurface.

If the vector potential is different from zero, the phase $\varphi(x)$ may be different from a constant without the simultaneous existence of nodal hypersurfaces M_{3N-2}. However, if the Hamiltonian is sufficiently regular, the following is always true[7]: Whenever the phase changes its value along a closed path in the space R^{3N}, then this path encloses a nodal hypersurface of type M_{3N-2}. Thus from the behavior of the phase on the closed path in R^{3N} some information follows about the behavior of the wave function in the "interior" of the closed path.

NPC can occur in solutions of the Schrödinger equation of free particles. This shows that it is a typical quantum correlation in the sense of the Einstein-Podolsky-Rosen experiment,[9] i.e., a correlation which is possible among particles which do not interact dynamically. From particular examples of NPC (e.g., the one treated in Sec. III) it becomes clear that NPC describes a correlation between the measured values of the positions and the orbital angular momenta around different axes of the individual particles.

In the absence of a magnetic field the nodal phase structure [Eq. (2)] does in general not give any observable effects because of time-reversal degeneracy of the states with opposite equal circulation numbers. In the presence of a magnetic field $\vec{B}(r)=\mathrm{curl}\,\vec{A}(r)$ this degeneracy is removed under favorable constellations of the $3N$-dimensional vectors

$$\mathfrak{A}_\alpha(x)=(\vec{A}(\vec{r}_1),\vec{A}(\vec{r}_2),\ldots,\vec{A}(\vec{r}_N))_\alpha \quad \text{and} \quad \frac{\partial}{\partial x_\alpha}\varphi(x)$$

(cf. Secs. III and IV).

III. MAGNETIC FLUX QUANTIZATION

We consider a superconductor shaped as a long, hollow cylinder (centered by the z axis) of inner and outer radii a and b, respectively, and we assume the radius a and the wall thickness $b-a$ to be much greater than the penetration depth. The Hamiltonian in the space of N superconducting electrons has the form

$$H = \frac{-\hbar^2}{2m_0}\sum_{j=1}^{N}\left(\frac{\partial}{\partial \vec{r}_j}\right)^2 + V. \tag{5}$$

Here m_0 is the electron mass and V is the sum of two terms, the first one representing the sum of the single-electron potential seen by each conduction electron, the second one representing the electron phonon interaction. According to BCS the superconducting states are described by approximate eigenfunctions $\psi(x, s)$ of the general form[10]

$$\psi(x, s) = \mathfrak{A}[\psi(\vec{r}_1, \vec{r}_2)\chi_{12}\psi(\vec{r}_3, \vec{r}_4)\chi_{34} \cdots \psi(\vec{r}_{N-1}, \vec{r}_N)\chi_{N-1,N}]. \tag{6}$$

Here χ_{ij} is the spin function $\uparrow(i)\downarrow(j)$ and $\mathfrak{A}$ is the antisymmetrizing operator. For our purpose it is sufficient to consider the system as being translationally invariant in the bulk of the superconductor. In this case the functions $\psi(\vec{r}_i, \vec{r}_j)$ have the form

$$\psi(\vec{r}_i, \vec{r}_j) = f(\vec{\kappa}_{ij})e^{i\vec{k}\cdot\vec{R}_{ij}}, \tag{7}$$

with

$$\vec{\kappa}_{ij} = \vec{r}_i - \vec{r}_j, \quad \vec{R}_{ij} = \tfrac{1}{2}(\vec{r}_i + \vec{r}_j). \tag{8}$$

[Equation (7) is only valid if the vector $\vec{k}$ is sufficiently small (see, for example, Ref. 10, p. 57).] The function $f(\vec{\kappa})$ (which itself depends on $\vec{k}$) is symmetric with respect to the origin, and it extends only over the microscopic range of the order of 10^{-4} cm, i.e., of Pippard's coherence length (see Ref. 10, p. 43).

Equation (7) describes the local, microscopic structure of the ground state ($\vec{k} = 0$) and of current carrying lower excited states ($\vec{k} \neq 0$). For a discussion of a macroscopic ring we will have to consider the behavior of the wave function on a macroscopic scale. This behavior will be determined by the shape of the macroscopic domain. The local structure of $\psi(\vec{r}_i, \vec{r}_j)$ [Eq. (7)] is valid only in the interior of the solid, i.e., in the neighborhood of all pairs $(\vec{r}_i, \vec{r}_j)$ for which both $\vec{r}_i$ and $\vec{r}_j$ are contained in the volume of the hollow cylinder. Now $f(\vec{\kappa}_{ij})$ extends only over a microscopic range of the order of 10^{-4} cm. This means that, on a macroscopic scale, the range of $\vec{\kappa}_{ij}$ is confined to the origin $|\vec{\kappa}|_{ij} \approx 0$, i.e., $\vec{r}_i \approx \vec{r}_j$. As a consequence, the macroscopic support of the function $\psi(\vec{r}_i, \vec{r}_j)$ with respect to the variable $\vec{R}_{ij}$ is the whole volume of the hollow cylinder. (Physically, the support in $\vec{\kappa}$ space originates essentially in the electron-phonon interaction, whereas the support in $\vec{R}$ space is a consequence of the effective crystal potential extending over the volume of the ring, together with the support in $\vec{\kappa}$ space.) The local structure of $\psi(\vec{\kappa}, \vec{R})$ [Eq. (7)] corresponds to a separation of the variables $\vec{\kappa}$ and $\vec{R}$,

$$\psi(\vec{\kappa}, \vec{R}) = f(\vec{\kappa})\, g(\vec{R}), \tag{9}$$

where $g(\vec{R})$ is an eigenfunction of the kinetic energy operator

$$E_{\mathrm{kin}}(\vec{R}) = \frac{-\hbar^2}{4m_0}\left(\frac{\partial}{\partial \vec{R}}\right)^2, \tag{10}$$

such that $g(\vec{R})$ does not change appreciably over a coherence length. Since the support in $\vec{R}$ space is the volume of the hollow cylinder, the eigenfunction $g(\vec{R})$ is adapted to this cylindrical symmetry, i.e., we have

$$g(\vec{R}) = a(R_z, R_{xy})e^{i\lambda\alpha_+(\vec{R})} \quad (\lambda = m_+ = \text{integer}). \tag{11}$$

Here α_+ is the azimuthal angle in $\vec{R}$ space and $R_{xy} = (R_x^2 + R_y^2)^{1/2}$. For small values of m_+ this global structure [Eq. (11)] is consistent with the local structure [Eq. (7)], since, as a consequence of the macroscopic dimension of the ring, neither $a(R_z, R_{xy})$ nor the $\vec{R}$ gradient of $\alpha_+(\vec{R})$ vary appreciably over a microscopic region. [Remark: From the mathematical point of view λ can actually be any real number, since we consider a doubly connected region (the volume of the ring), where the Schrödinger equation is not fully defined. However, for physical reasons, the Schrödinger equation of the doubly connected region must be consistent with a covering Schrödinger equation which is defined in a larger, simply connected region (e.g., in the whole three-dimensional $\vec{R}$ space), containing our doubly connected ring volume. It can be shown[11] that under quite general assumptions the covering Schrödinger equation has only single-valued solutions. Therefore we must restrict the value of λ to integers.]

For the following the dependence of $\psi(\vec{r}_i, r_j)$ on the angle $\alpha_+(\vec{R}_{ij})$ is important. The angle $\alpha_+(\vec{R}_{ij})$ is related to the azimuthal angles $\alpha(\vec{r}_i)$ and $\alpha(\vec{r}_j)$ of the spaces of the ith and jth particles, respectively, by the following important relation:

$$\alpha(\vec{r}_i) \approx \alpha(\vec{r}_j) \approx \alpha_+(\vec{R}_{ij}), \tag{12}$$

whence

$$2\alpha_+(\vec{R}_{ij}) \approx \alpha(\vec{r}_i) + \alpha(\vec{r}_j). \tag{13}$$

Equation (12) is valid for all points $(\vec{r}_i, \vec{r}_j)$ for which $\psi(\vec{r}_i, \vec{r}_j)$ is appreciably different from zero. This is easily verified, since these points obey the relations $\vec{r}_i \approx \vec{r}_j$, together with $|\vec{r}_i| \gg 0$, $|\vec{r}_j| \gg 0$, the latter being a consequence of the macroscopic inner radius of the cylinder.

In the presence of a magnetic field $\vec{B}(\vec{r}) = \mathrm{curl}\,\vec{A}(\vec{r})$ the N-particle Schrödinger equation reads ($-e$ is the electric charge):

$$\left[\frac{1}{2m_0}\sum_{k=1}^{N}\left(\frac{\hbar}{i}\frac{\partial}{\partial\vec{r}_k}+\frac{e}{c}\vec{A}(\vec{r}_k)\right)^2+V-E\right]\psi'(x,s)=0. \tag{14}$$

If a magnetic flux ϕ is present in the hole of the cylinder, then in the volume of the ring the vector potential takes the form

$$\vec{A}(\vec{r})=(\phi/2\pi)\mathrm{grad}\alpha(\vec{r}), \tag{15}$$

and Eq. (14) is equivalent to

$$\left[\frac{-\hbar^2}{2m_0}\sum_{k=1}^{N}\left(\frac{\partial}{\partial\vec{r}_k}\right)^2+V-E\right]\psi'(x,s)$$

$$\times\exp\left(\frac{ie\phi}{hc}\sum_{k=1}^{N}\alpha(\vec{r}_k)\right)=0. \tag{16}$$

Now we claim that the solutions $\psi'(x,s)$ have the same structure as the functions $\psi(x,s)$ in the flux-free case [Eqs. (6), (9), and (11)]. Let us use the ansatz for the pair functions

$$\psi'(\vec{r}_i,\vec{r}_j)=f'(\vec{\kappa}_{ij})\,g'(\vec{R}_{ij}). \tag{17}$$

Then the total eigenfunction of the field-free Hamiltonian in Eq. (16) consists of sums of products of the functions

$$\psi''(\vec{r}_i,\vec{r}_j)=\psi'(\vec{r}_i,\vec{r}_j)\times\exp\{i(e\phi/hc)[\alpha(\vec{r}_i)+\alpha(\vec{r}_j)]\}. \tag{18}$$

If now the support of the function $f'(\vec{\kappa})$ is approximately the same as in the flux-free case, we can apply relation (13), whence

$$\psi''(\vec{r}_i,\vec{r}_j)\approx\psi(\vec{r}_i,\vec{r}_j)\exp[i(2e\phi/hc)\alpha_+(\vec{R}_{ij})]\equiv f'(\vec{\kappa}_{ij})\,g''(\vec{R}_{ij}). \tag{19}$$

Here the $\vec{R}$-dependent function $g''(\vec{R})$ is an eigensolution of the operator (10) considered in the (doubly connected) volume of the ring. But now the eigenfunction

$$g''(\vec{R})=a'(R_z,R_{xy})e^{i\lambda\alpha_+(\vec{R})} \tag{20}$$

has to obey the subsidiary condition

$$\lambda=m_++2e\phi/hc,\quad m_+\text{ integer}, \tag{21}$$

taking into account the fact that $\psi'(x)$ is single valued [see the remark made in connection with Eq. (11)]. From Eqs. (19) and (20) we obtain

$$\psi'(\vec{\kappa},\vec{R})=f'(\vec{\kappa})\,a'(R_z,R_{xy})e^{im_+\alpha_+(\vec{R})},\quad m_+\text{ integer}. \tag{22}$$

If we consider solutions where λ in Eq. (20) is of the same order of magnitude as λ in Eq. (11), then $\psi'(\vec{\kappa},\vec{R})$ has in fact the same structure as $\psi(\vec{\kappa},\vec{R})$ in Eqs. (9) and (11), since owing to the same separation of variables the two functions obey similar equations.

The expectation value of the azimuthal part $(-\hbar^2/4m_0|\vec{R}|^2)(\partial/\partial\alpha_+)^2$ of the kinetic energy operator (10), times $\frac{1}{2}N$ (the number of pairs), represents the dominant part E_ϕ of the flux dependent total energy of the N-electron system. For the ground state we obtain

$$E_\phi=(N\hbar^2/8m_0l^2)[m_++(2e/hc)\phi]^2, \tag{23}$$

where l is the average radius of the cylinder. E_ϕ is a periodic function of the total flux ϕ, with minima (equal to zero) for

$$\phi=(-hc/2e)m_+, \tag{24}$$

and differences between the minima

$$\Delta\phi=hc/2e. \tag{25}$$

This periodicity leads to the phenomena of trapped magnetic flux and of flux quantization in units of $hc/2e$ (Ref. 13; see also Refs. 4 and 5).

IV. DISCUSSION

A. Nodal phase correlation

We consider a total N-particle wave function $\psi(x,s)$ which is a solution of Eq. (16) (where we omit the prime). The corresponding *space* functions $\psi(x)$ are linear combinations of functions of the type

$$\psi_\nu(x)=\psi(i_1,j_1)\,\psi(i_2,j_2)\cdots\psi(i_{N/2},j_{N/2}). \tag{26}$$

Here the index ν represents a definite partition of N into $\frac{1}{2}N$ pairs ij, and $\psi(ij)$ stands for a pair function $\psi(\vec{r}_i,\vec{r}_j)$ defined by Eq. (22) (where we omit the primes). The functions $\psi(\vec{r}_i,\vec{r}_j)$ and their N-particle product functions $\psi_\nu(x)$ clearly show two-particle NPC whenever the circulation number m_+ is different from zero. The phase of each pair function $\psi(\vec{r}_i,\vec{r}_j)$ appearing in a particular function $\psi_\nu(x)=R_\nu(x)e^{i\varphi_\nu(x)}$ gives rise to a class of paths P_ν^{ij} defined by

$$m_+=\frac{1}{2\pi}\oint_{P_\nu^{ij}}\frac{\partial}{\partial x_\beta}\varphi_\nu(x)\,dx_\beta, \tag{27}$$

where each path P_ν^{ij} encircles once the region $V(ij)=\{x\,|\,R_{xy}^{ij}<a\}$. According to Sec. II this implies that a nodal hypersurface M_{3N-2} associated with the particular class of paths P_ν^{ij} must lie somewhere in the interior of the region $V(ij)$. Its exact position is not known, since the detailed form of $\psi(x)$ is unknown in $V(ij)$. If $\psi(x,s)$ had the structure [Eqs. (6) and (22)] also in the volume $V(ij)$, then this nodal hypersurface (denoted by $M_{3N-2}^{\nu ij}$) would be explicitly known and defined by

$$(\vec{r}_i+\vec{r}_j)_x=\vec{0}=(\vec{r}_i+\vec{r}_j)_y. \tag{28}$$

There are $\frac{1}{2}N$ hypersurfaces $M_{3N-2}^{\nu ij}$ associated with each function $\psi_\nu(x)$. The analysis of Sec. II shows that *flux quantization in superconductors*, and in particular *the factor $\frac{1}{2}$ appearing in the flux quantum ($hc/2e$), originate in this two-particle NPC structure* of the functions $\psi_\nu(x)$. {Remark: In Sec. II we have defined NPC as being associated with a total space function $\psi(x)$, and not with functions $\psi_\nu(x)$ into which it may be decomposed. Now the dominant support of a function $\psi(x)$ is the union of branches each of which represents the dominant support of a function $\psi_\nu(x)$. In our case the different functions $\psi_\nu(x)$ have almost disjoint dominant supports in the space R^{3N}. [The dominant support of a function $\psi_\nu(x)$ is the region where $\psi_\nu(x)$ is appreciably different from zero. It consists of all points $x=(\vec{r}_1,\dots\vec{r}_i,\dots\vec{r}_j,\dots\vec{r}_N)$, such that $|\vec{r}_i-\vec{r}_j|<\xi$ (where ξ is of the order of 10^{-4} cm, for all pairs $(\vec{r}_i,\vec{r}_j)$ which are arguments of a pair function $\psi(r_i,r_j)$ appearing in the product function (26).] Therefore $\psi(x)$ is equal to some function $\psi_\nu(x)$ on almost the whole of each branch of its proper dominant support. That is, apart from the intersections of the branches (which are negligible for our macroscopic considerations) the NPC structure of $\psi(x)$ is the same as that of the functions $\psi_\nu(x)$.}

It is instructive to compare our system of superconducting electrons with a system of identical, spinless, interaction-free bosons with charge $-q$. Here the ground-state function $\psi(x)$ is a product of N equal single-particle functions $u(\vec{r}_k)\sim e^{im\alpha(\vec{r}_k)}$, i.e., the NPC index n is equal to one and $\psi(x)$ does not show NPC. In a method analogous to Sec. III one finds that the azimuthal part of the ground-state energy of the bosons in the ring is proportional to $(m+q/hc)^2$, whence a flux quantum equal to hc/q is obtained. We see that the factor $\frac{1}{2}$, which occurs for superconductors as a consequence of two-particle NPC ($n=2$), is here replaced by one, as a consequence of n being equal to one.

The effect of two-particle NPC is further illustrated by the fact that owing to Eq. (13) applied to Eq. (22) (we omit the primes), the functions $\psi_\nu(x)$ can approximately be written

$$\psi_\nu(x)\approx\exp\left(\sum_{k=1}^{N} i[\tfrac{1}{2}\alpha(\vec{r}_k)]m_+\right)\prod_{(ij)} f(\vec{\kappa}_{ij})\,a(R_z^{ij},R_{xy}^{ij}). \tag{29}$$

Equation (29) makes one think of $\psi_\nu(x)$ being an eigenfunction of the one-particle orbital angular momentum operators

$$L_z^k=-i\hbar\left(x_k\frac{\partial}{\partial y_k}-y_k\frac{\partial}{\partial x_k}\right),$$

with possible half-integer orbital angular momenta (since m_+ is integer). This is in fact true if we consider only points $x\in R^{3N}$, which are situated on the dominant support of $\psi_\nu(x)$. For points x which lie outside the dominant support of $\psi_\nu(x)$, however, the phase structure of $\psi_\nu(x)$ is in general different from that given in Eq. (29). [This is certainly the case if $|\vec{r}_i|\approx|\vec{r}_j|\approx 0$, or if $|\vec{r}_i|\approx|\vec{r}_j|$ with $(\vec{r}_i-\vec{r}_j)_{xy}\gg 10^{-4}$ cm, since in these cases relation (13) is not valid.] Further, the projection of the dominant support into a one-particle subspace depends on the position of other particles. This means, that $\psi_\nu(x)$ is not an eigenfunction of a properly defined single-particle orbital angular momentum operator L_z^k.

Despite the phase structure appearing in Eq. (29), the function $\psi_\nu(x)$ is still single valued. In fact, consider a vector x in the dominant support of $\psi_\nu(x)$ and let a single-particle vector $\vec{r}_k$ go once around the hole $[(\vec{r}_k)_{xy}<a]$ of the macroscopic cylinder; then x remains in the dominant support only if another single-particle vector $\vec{r}_{k'}\approx\vec{r}_k$ (the mate of $\vec{r}_k$) simultaneously goes around the hole $[(\vec{r}_{k'})_{xy}<a]$. Hence the phase of $\psi_\nu(x)$ changes by $2\pi(\frac{1}{2}m_+)+2\pi(\frac{1}{2}m_+)=2\pi\times$ integer. We further remark that, on the dominant support of $\psi_\nu(x)$, the total angular momentum is always integer, since the simulated half-integer one-particle angular momenta always occur in pairs.

Approximation (29) is valid for each function $\psi_\nu(x)$ on its proper dominant support. Therefore, a total space function $\psi(x)$ [which is a linear combination of functions $\psi_\nu(x)$], in very good approximation contains the factor

$$\exp\left(\sum_{k=1}^{N} i[\alpha\tfrac{1}{2}(\vec{r}_k)]m_+\right)$$

on its dominant support. Brenig[4] uses (as an ansatz) a wave function with such a factor. However, he finally constructs his wave function such that it is also single valued in each $\alpha(\vec{r}_k)$ individually. Our present analysis justifies the idea of the general phase structure in Brenig's ansatz and shows its origin. On the other hand, it also shows that a phase structure according to Eq. (29) occurs only on the dominant support of the wave function, and that here, i.e., on this particular subset of the configuration space R^{3N}, the wave function is not single valued in each angle variable $\alpha(\vec{r}_k)$ separately. [This is not in contradiction to the fact that the wave function is single valued in each particle variable individually, i.e., single valued in the whole of the configuration space R^{3N}. In other words our statement means the following: if we let one angular variable $\alpha(\vec{r}_k)$ vary around the hole of the cylinder, while all other variables

are kept fixed, then after a microscopic variation (of the order of the coherence length divided by the radius of the cylinder) we come to a region in configuration space R^{3N} which lies outside the dominant support, and there the wave function has no longer the form (29).] We therefore find that Brenig's wave function is not equivalent to our wave function, but rather to the single-particle pairing function which we discuss in Sec. III B.

B. Examination of the single-particle pairing approximation

A standard derivation of flux quantization[5] (cf. Ref. 10, p. 240ff) is based on solutions of the one-electron Schrödinger equation for the ring. Following Byers and Yang,[5] and Schrieffer[10] the single-particle states are then paired up according to a pairing scheme to form the N-particle state. (We remark that such a state, before antisymmetrization, does not show NPC, in contrast to the functions $\psi_\nu(x)$ discussed previously.) *The method works as follows*: First the single-particle functions are determined by solving the azimuthal part of the one-electron Schrödinger equation

$$\frac{1}{2m_0 r^2}\left(\frac{\hbar}{i}\frac{\partial}{\partial\alpha}+\frac{e\phi}{2\pi c}\right)^2\psi_M(\alpha) = \frac{\hbar^2}{2m_0 r^2}\left(M+\frac{e\phi}{hc}\right)^2\psi_M(\alpha), \quad (30)$$

where

$$r=(x^2+y^2)^{1/2} \text{ and } \psi_M(\alpha)=e^{iM\alpha}, \ M \text{ integer}. \quad (31)$$

The angular kinetic energy of the electron is given by

$$E_\phi=(\hbar^2/2m_0 l^2)(M+e\phi/hc)^2. \quad (32)$$

The pairing condition for pairing two single-particle functions ψ_M and $\psi_{\bar M}$ reads[10]

$$M+e\phi/hc=-(\bar M+e\phi/hc), \quad (33)$$

that is,

$$M\equiv m-e\phi/hc \text{ and } \bar M\equiv -m-e\phi/hc \quad (34)$$

are paired, with m and $e\phi/hc$ being both integer or both half-odd integer. As a consequence the minima of the azimuthal part of the two-particle energies are equal to

$$E_{\min}=\hbar^2 m^2/m_0 l^2, \quad (35)$$

where $m=0, \pm\frac{1}{2}, \pm 1, \ldots$.

Now let us *compare the azimuthal part*,

$$e^{iM\alpha(\vec r_i)}e^{i\bar M\alpha(\vec r_j)}\equiv e^{i(m-e\phi/hc)\alpha(\vec r_i)} \times e^{i(-m-e\phi/hc)\alpha(\vec r_j)}, \quad (36)$$

of a particular two-particle function (described by a definite value of m) *in the single-particle pairing theory with the azimuthal part*

$$e^{im_+\alpha_+(\vec R_{ij})}=e^{i(m_+/2)\alpha(\vec r_i)}e^{i(m_+/2)\alpha(\vec r_j)} \quad (\vec r_i\approx\vec r_j), \quad (37)$$

of our two-particle function $\psi(\vec r_i,\vec r_j)=f(\vec\kappa_{ij})g(\vec R_{ij})$ [Eqs. (17) and (22), where we omit the primes], both representing a state where the energy has a minimum. This means that

$$\tfrac{1}{2}m_+=-e\phi/hc, \quad (38)$$

and hence

$$\psi(\vec r_i,\vec r_j)\sim g(\vec R_{ij})\sim e^{im_+\alpha_+(\vec R_{ij})} = e^{-i(e\phi/hc)\alpha(\vec r_i)}e^{-i(e\phi/hc)\alpha(\vec r_j)}. \quad (39)$$

We recognize that *Eqs.* (36) *and* (39) *are not identical.* The only situation in which they could be formally identical would be in the presence of excited states of the relative coordinate function $f(\vec\kappa_{ij})$, such that $f(\vec\kappa_{ij})$ would have the form

$$f(\vec\kappa_{ij})=b(\kappa_z^{ij},\kappa_{xy}^{ij})e^{im_-\alpha_-(\vec\kappa_{ij})}, \quad m_- \text{ integer}. \quad (40)$$

[Here κ_z^{ij}, κ_{xy}^{ij}, and $\alpha_-(\vec\kappa_{ij})$ are cylindrical coordinates in $\vec\kappa_{ij}$ space.] In fact, if in analogy to Eq. (13) we had

$$2\alpha_-(\vec\kappa_{ij})\equiv 2\alpha_-(\vec r_i-\vec r_j)\approx\alpha(\vec r_i)-\alpha(\vec r_j) \quad (41)$$

(α_- is the azimuthal angle in $\vec\kappa$ space), then we could replace Eq. (39) by

$$\psi(\vec r_i,\vec r_j)=f(\vec\kappa_{ij})g(\vec R_{ij}) \sim e^{i(m_-/2-e\phi/hc)\alpha(\vec r_i)}e^{i(-m_-/2-e\phi/hc)\alpha(\vec r_j)} \quad (42)$$

which is now equal to Eq. (36) if $\frac{1}{2}m_-$ equals m. (This latter condition is consistent with m_- being integer and m being integer or half-odd integer.) Thus we obtain the result that the single-particle pairing formalism is formally equivalent to our approach presented in Sec. III, provided Eqs. (40) and (41) are valid, and provided that $\frac{1}{2}m_-$ is pairwise equal to m. However, Eq. (41) is, in general, not valid on the dominant support of $\psi(\vec r_i,\vec r_j)$, where $\vec r_i\approx\vec r_j$. Hence the two sets of pair functions cannot be formally identical.

Even if only Eq. (40) was valid [but not Eq. (41)], the two methods would not even approximately be equivalent. This becomes most evident, if we *compare the N-particle states of the two approaches in the presence of the flux* ϕ.

a. In the single-particle pairing method the ground state has the form

$$\psi_N(x,s)\sim\mathfrak{A}\,(e^{iM_1\alpha(\vec r_1)+i\bar M_1\alpha(\vec r_2)} \times\chi_{12}\cdots e^{iM_k\alpha(\vec r_{2k-1})+i\bar M_k\alpha(\vec r_{2k})} \times\chi_{2k-1,2k}\cdots e^{iM_{N/2}\alpha(\vec r_{N-1})+i\bar M_{N/2}\alpha(\vec r_N)}\chi^{N-1,N}). \quad (43)$$

Here M_k are integers which are *all different* from each other. The total energy is given by

$$E_N(\phi)=\frac{\hbar^2}{2m_0 l^2}\sum_{k=1}^{N}\left[\left(M_k+\frac{e\phi}{hc}\right)^2+\left(\overline{M}_k+\frac{e\phi}{hc}\right)^2\right]. \tag{44}$$

It has (almost equal) minima at $\phi=(hc/2e)\times$ integer. (The minima are not equal to zero.) In the absence of the flux we have $M_k=-\overline{M}_k$. When the flux increases, the energy changes continuously, and the numbers M_k and $\overline{M}_k$ to be paired change whenever $2e\phi/hc$ passes a value $\frac{1}{2}+$ integer [cf. Eqs. (33) and (34), or Fig. 8.8 in Ref. 10].

b. In our case, on the other hand, the ground state has the form [Eq. (6)], where the orbital pair functions

$$\psi(\vec{r}_i,\vec{r}_j)=f(\vec{r}_i-\vec{r}_j)g(\tfrac{1}{2}(\vec{r}_i+\vec{r}_j))$$

are highly correlated, and are *all equal.* The azimuthal energy, which is due to the $\vec{R}_{ij}$ spaces, is given by Eq. (23). It has minima (equal to zero) whenever ϕ is equal to $(-hc/2e)m_+$ (m_+ integer). In the absence of the flux, the function g has no azimuthal phase factor in the ground state, i.e., m_+ is equal to zero. With increasing flux ϕ the energy changes continuously, and the quantum number m_+ changes (in the same way for all pairs) whenever $2e\phi/hc$ passes a value $\frac{1}{2}+$ integer. Now if in fact $f(\vec{\kappa})$ had exited states of the form (40), then the quantum number m_- would nevertheless be zero, despite the presence of the magnetic flux, because otherwise the energy would increase by the azimuthal part E^{κ}_{az} of the energy corresponding to the $\vec{\kappa}_{ij}$ spaces

$$E^{\kappa}_{az}=\frac{m_-^2 N\hbar^2}{2m_0}\int\frac{\psi^*(\vec{\kappa},\vec{R})\,\psi(\vec{\kappa},\vec{R})}{\kappa_{xy}^2}\,d\vec{\kappa}\,d\vec{R}, \tag{45}$$

and the system would no longer be in its ground state, since this additional energy is independent of the flux. [That is, unlike the phase $m_+\alpha_+(\vec{R})$, the phase $m_-\alpha_-(\vec{\kappa})$ does not contribute to minimize the magnetic energy. This follows from the fact that on the dominant support of a function $\psi_\nu(x)$ the N-dimensional vector potential $\mathfrak{A}_\alpha(x)$ in very good approximation depends only on the variables $\alpha_+(\vec{R}_{ij})$.]

The preceding comparison shows that the ground state of the single-particle pairing formalism corresponds to a highly excited state in our approach (which is more closely based on the BCS theory). For example, in the absence of the flux, the ground state (43) of the pairing theory corresponds to an excited state of our function $\psi(x,s)$ [Eqs. (6) and (9)], where all the relative coordinate functions $f(r_i-r_j)$ are in a *different* excited state (40), with quantum numbers m_- having different (and only even!) values for each pair [cf. Eqs. (33), (34), (36), (40)–(45)]. Thus our analysis shows, that *the single-particle pairing formalism leads to a rather physically unrealistic many-particle wave function of the superconducting ring system,* and that *it is not consistent with the* BCS *theory.*

c. Off-diagonial long-range order (ODLRO) It is well known[6,12] that a BCS function shows ODLRO of the reduced density matrix $\rho^{(2)}$. In fact, ODLRO [e.g., in the form given by Eq. (26c) of Ref. 6] can easily be verified for a BCS function by using the fact that the dominant supports of the different functions $\psi_\nu(x)$ are almost disjoint sets in the space R^{3N}, and that all pair functions in Eq. (6) are equal. Yang[6] gives an argument in favor of the statement that ODLRO implies a periodic free-energy behavior as a function of the flux ϕ (and hence implies magnetic-flux quantization). We emphasize, however, that the property of the energy, in order to be a periodic function of the magnetic flux, originates in the *phase structure* of the N-particle wave function, whereas ODLRO is also possible for real functions. Therefore Yang's statement has to be taken with precaution. As a counterexample imagine, e.g., a system, which is described by wave functions of the general type (6) discussed in Sec. III, but such that $g(\vec{R})$ is real (but still macroscopically extended), owing to some additional interaction in the system. In this case the magnetic energy is not periodic with the flux, but we still have ODLRO in $\rho^{(2)}$. Thus ODLRO is not a sufficient condition for magnetic-flux quantization. [Nor is it a necessary condition, since the wave function (43) of the single-particle pairing formalism, which leads to a periodic flux dependence of the energy, does not show ODLRO.] All we can say is that if the energy of the system is periodic with the flux, and if the system shows ODLRO, then the nature of ODLRO may determine the length of the period (compare Refs. 6 and 13).

V. SUPERCONDUCTING INTERFEROMETERS

While the single-particle pairing formalism, despite its unrealistic wave function (43), is able to approximately simulate the correct flux dependence of the energy of a superconducting ring, we expect it to be unsuccessful in describing physical effects which directly involve a measurement of a partial behavior of the wave function. On the other hand, we expect that the BCS-type function of Sec. III will be able to describe the effects. This is exactly what happens in the following example.

If one connects two Josephson junctions in parallel by superconducting links, one obtains a superconducting interferometer (for a review see Ref.

2). Such a system has the same topological structure as the ring discussed in Sec. III. The superconducting electron waves in the individual arms of the interferometer interfere with each other in the same way (apart from the Josephson effect) as in an electron interferometer for freely moving electron beams. Now if in this second case the two branches of the interferometer enclose a magnetic flux ϕ, the order of the interference pattern is shifted by unity whenever the magnetic flux increases by an amount $\Delta\phi = hc/e$. This is the (magnetic) Aharonov-Bohm effect.[14] (For a review see, e.g., Ref. 15.)

The theoretical derivation of the Aharonov-Bohm effect given in Ref. 14 (see also Ref. 16) consists in calculating stationary scattering states of the one-electron Schrödinger equation in the presence of the magnetic flux. (The system consists of uncorrelated electrons.) Analogously, in the case of a superconducting interferometer enclosing a magnetic flux one has to calculate stationary N-electron scattering states. In the framework of the single-particle pairing formalism this means that one has to build a Slater determinant out of N single-particle stationary scattering states, i.e., out of N single-particle states, each describing a one-electron Aharonov-Bohm effect. This method therefore leads to the same periodic flux dependence as in the Aharonov-Bohm effect for beams of free electrons, with a flux period $\Delta\phi = hc/e$. Experimentally, however, one observes[2] a flux period $\Delta\phi = hc/2e$.

On the other hand, if we use a BCS N-particle wave function as in Sec. III, we obtain at once the experimental value: Following the same arguments as given in Sec. III, the kinetic part of the Schrödinger equation here reduces to the scattering problem for the function $g(\vec{R})$. This is again the same problem as in the case of the Aharonov-Bohm effect for a single electron, except that now in $\vec{R}$ space, as a consequence of Eq. (13) (or more generally because $\vec{r}_i \approx \vec{r}_j \approx \vec{R}_{ij}$), the vector potential has twice the value it has in a single-particle space [cf. Eqs. (18), (19)], whence the flux period $\Delta\phi = hc/2e$.

The mathematical behavior of the scattering states is complicated. However, in the special case where $2\phi e/hc = m_+ =$ integer, the functions $g(\vec{R})$, for large radial distances from the flux carrying area, have the asymptotic form[14]

$$g(\vec{R}) \sim e^{-ikR_x}\, e^{-im_+\alpha_+(\vec{R})}. \tag{46}$$

Here we recognize the same two-particle NPC structure as in the closed-ring system discussed in Sec. III.

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*Correspondence regarding this article should be addressed as follows: J. Riess, Physikalisch-chemisches Institut der Universität Zürich, Rämistr. 76, 8001 Zürich, Switzerland.

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