

Quantum Phase in Nanoscopic Superconductors

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Using the pseudospin representation and the SU(2) phase operators we introduce a complex parameter to characterize both infinite and finite superconducting systems. While in the bulk limit the parameter becomes identical to the conventional order parameter, in the nanoscopic limit its modulus reduces to the number parity effect parameter and its phase takes discrete values. We evaluate the Josephson coupling energy and show that in bulk superconductor it reproduces the conventional expression and in the nanoscopic limit it leads to *quantized Josephson effect*. Finally, we study the *phase flow* or *dual Josephson effect* in a superconductor with fixed number of electrons.

KEY WORDS: mesoscopic superconductivity; order parameter; Josephson effect; quantum phase.

Recent experimental works on superconducting metallic islands at nanometer scale have established a link between bulk superconductors and atomic nuclei as regards pairing correlations [1–3]. The long-range order in a bulk superconductor can be described by an order parameter Δ which is complex and the equations have symmetry properties which ensure that if Δ is a solution, then $e^{i\theta}\Delta$ is also a solution [4,5]. On the other hand when size of a superconductor is reduced to nanometer scales so that the number of the electrons is fixed, the order parameter $\Delta = \langle c_{-k\downarrow}c_{k\uparrow} \rangle$ vanishes where $c_{-k\downarrow}$ and $c_{k\uparrow}$ are the annihilation operators for time-reversed states $|-\mathbf{k}\downarrow\rangle$ and $|\mathbf{k}\uparrow\rangle$, respectively. In this case, superconductivity manifests itself with nonvanishing number parity effect parameter Δ_P where the ground state energy of the system increases or decreases, depending upon whether the total number becomes odd or even, by addition of a new electron [6,7]. In this work, we propose a parameter, which unifies the order parameter Δ of the bulk limit and the number parity effect parameter Δ_P of the nanoscopic superconductors. Introducing the pseudospin or quasi-spin representation for the model Hamiltonian of the theory of superconductivity [8,9] and the SU(2) phase states [10] we define a quantum phase for a superconductor with discrete energy levels

along with modulus of the parameter which becomes equal to Δ_P . As we go from the nanoscopic limit to the bulk superconductor we show that the number parity effect parameter and the SU(2) phase go to the amplitude and the phase of the bulk order parameter, respectively.

We are going to start with a notation which is more proper for nanoscopic superconductors where energy levels are discrete and finite [11]. This reduced form of the BCS model was applied in nuclear physics and it has an exact solution [12]. The model Hamiltonian is

$$H = \sum_{j,\sigma} \epsilon_j c_{j\sigma}^\dagger c_{j\sigma} - g \sum_{j,j'} c_{j'}^\dagger c_{j\downarrow}^\dagger c_{j'\downarrow} c_{j'\uparrow} \quad (1)$$

where g is the pairing coupling constant for the time-reversed states $|j\uparrow\rangle$ and $|j\downarrow\rangle$, both having the energy ϵ_j . Here, $c_{j\sigma}^\dagger$ ($c_{j\sigma}$) is the creation (annihilation) operator for state $|j\sigma\rangle$ where $j \in \{1, \dots, \Omega\}$ and $\sigma \in \{\uparrow, \downarrow\}$.

Introducing the pseudospin variables [8,9]

$$s_j^z = \frac{1}{2}(c_{j\uparrow}^\dagger c_{j\uparrow} + c_{j\downarrow}^\dagger c_{j\downarrow} - 1), \quad s_j^- = c_{j\downarrow} c_{j\uparrow} = (s_j^+)^{\dagger} \quad (2)$$

which generate the SU(2) algebra

$$[s_i^+, s_j^-] = 2\delta_{ij}s_j^z, \quad [s_i^z, s_j^\pm] = \pm\delta_{ij}s_j^\pm, \quad (3)$$

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it is possible to rewrite the model Hamiltonian as

$$H = \sum_j 2\epsilon_j \left(s_j^z + \frac{1}{2} \right) - g \sum_{ij} s_i^+ s_j^-. \quad (4)$$

We note that the mapping from the Fermi operators to the pseudospin operators is possible as long as all single particle states are doubly occupied. However, since the original Hamiltonian (1) contains no terms which couple a singly occupied level to others, the only role of such states will be blocking from pairing interaction. Therefore, the summations in Eq. (4) are over doubly occupied or empty states. Both the above mapping and the BCS wave function [4] lack proper antisymmetrization due to separate treatment of singly occupied states, but since the model Hamiltonian (1) does not involve any scatterings into or out of such states, antisymmetrization with respect to inter-level pair exchange and intrapair electron exchange is sufficient.

In this work, rather than the exact solution of the problem, we are interested in the qualitative result which has also been obtained numerically. The ground state energy for even number of electrons is lower in comparison to neighboring odd number states [13–16] including degenerate case [17]. Parity dependence of the condensation energy and pairing parameters in nanoscopic superconductors was first emphasized by von Delft *et al.* [11] but the first correction to the bulk limit had been obtained by Jankó *et al.* [18] and Golubev and Zaikin [19].

Phase operators and phase states have been studied mainly in quantum optics and possible connection of quantum phase and the mean field treatment of the BCS Hamiltonian has been pointed out by Shumovsky [20]. Given SU(2) algebra, for example the one generated by the components of the total spin operator $s = \sum_i s_i$, we can introduce [10] the radial operator defined by

$$s_r = \sqrt{s^+ s^-} \quad (5)$$

and the exponential of the phase operator given by

$$E = \sum_{m=-s}^{m=s} |S; sm+1\rangle \langle S; sm|. \quad (6)$$

Here, $|S; sm\rangle$ is simultaneous eigenstate of s^2 and s_z operators with eigenvalues $s(s+1)$ and m , respectively. In order to simplify the notation, m is defined modulo $2s+1$ so that $|S; ss+1\rangle = |S; s-s\rangle$. The label S has been introduced to distinguish them from the phase states to be defined below. We are going to

make use of the cosine and the sine operators

$$\text{Cos} = \frac{1}{2}(E + E^{-1}), \quad \text{Sin} = \frac{1}{2i}(E - E^{-1}). \quad (7)$$

For integer s or on the so called Bose sector, the eigenstate of E with eigenvalue $\exp(-i2\pi\mu/(2s+1))$ is evaluated to be

$$|\theta; s\mu\rangle = \frac{1}{\sqrt{2s+1}} \sum_{m=-s}^{m=s} \exp\left[i \frac{2\pi\mu}{2s+1} m\right] |S; sm\rangle \quad (8)$$

and a similar expression holds for half integer s or in the Fermi sector.

In terms of the radial and the exponential of the phase operators for the total spin, it is possible to rewrite the interaction part of the Hamiltonian (4) as $-gs_r E E s_r^\dagger$. Since E is unitary, we obviously have $E E^\dagger = I$ but our aim in keeping E and E^\dagger is to define the phase properly. Now, we introduce the superconductivity criterion as $\langle s_r \rangle \neq 0$. We are going to prove that this definition agrees with existing criteria for both grand canonical and canonical superconducting systems. We are going to show that $\langle s_r \rangle$ becomes identical to the modulus of the BCS order parameter in the bulk limit while in the nanoscopic limit it reduces to the number parity effect parameter Δ_p in units of g . There have been several suggestions for a canonically meaningful pairing parameter [11,15,21–24]. Our definition is equivalent to that of Tian *et al.* [24], which has been proposed by Penrose and Onsager [25] and Yang [26] as a measure of the strength of the spontaneous symmetry breaking field. Amico and Osterloh [27] and Zhou *et al.* [28], have calculated the pairing correlation function $\langle s_i^- s_j^+ \rangle$ analytically by extending Richardson's results [29]. We further introduce $\langle E \rangle$ as exponential of the phase. This definition is justified by the observation that in the grand canonical ensemble $\langle E \rangle$ turns out to be exponential of phase of the BCS order parameter.

We first note that $[s_r, s_z] = 0$ and hence s_r gives a good quantum number even for a finite system. Secondly, $\langle s_r \rangle$ is filling dependent even for a single, d -fold degenerate level in contrast to Δ_p . While $\langle s_r \rangle = \sqrt{v(d-v+1)}$ for v pairs, the number parity effect parameter Δ_p is $gd/2$, independent of v . For $v = d/2$, i.e. half-filling or $m = 0$, the two results become identical.

We start with our proof by examining the canonical system. In analogy to the pairing energy in nuclear physics [30], Matveev and Larkin [6] introduced the

parity effect parameter

$$\Delta_P = E_{2n+1} - \frac{E_{2n} + E_{2n+2}}{2} \quad (9)$$

for nanoscopic superconductors where E_n is the ground state energy for n electrons. Assuming that the expectation value of the single-particle energy part (the first term in Eq. (4)) follows a monotonic behavior so that $T_{2n+1} \simeq (T_{2n} + T_{2n+2})/2$, the main contribution to the ground state energy will come from the interaction part so that

$$\Delta_P = -g \left(\langle s_r^2 \rangle_{2n+1} - \frac{\langle s_r^2 \rangle_{2n} + \langle s_r^2 \rangle_{2n+2}}{2} \right). \quad (10)$$

Now, the eigenstates and in particular the ground state of the model Hamiltonian will be of the form

$$\sum_s C_{sm} |S; sm\rangle \quad (11)$$

because the interaction term commutes with s^2 and s_z while the single-particle part commutes with the latter only and hence m is a good quantum number. We note that since s is the total spin in general it is multiply degenerate. It is possible to calculate the expectation value of the radial operator $s_r = \sqrt{s^+ s^-}$ as

$$\langle s_r \rangle_n = \sum_s |C_{sm}|^2 \sqrt{s(s+1) - m(m-1)}. \quad (12)$$

Here, the number of electrons n is a function of m . In BCS theory the single-particle states participating in pairing interaction are assumed to be those in a shell of thickness $\sim 2\hbar\omega_D$, ω_D being the Debye frequency, symmetric around the Fermi level. In this case, half of the states are full while the half is empty and hence $m = 0$. Near half-filling where $m \simeq 0$ and for $s \gg 1$, we can approximate the square root as s to give $\langle s_r \rangle_n \simeq \sum_s |C_{s0}|^2 s$. Similarly, for s_r^2 with the same approximations we find that

$$\langle s_r^2 \rangle_{2n} \simeq \langle s_r^2 \rangle_{2n+2} \simeq \sum_s |C_{s0}|^2 s^2. \quad (13)$$

For $2n + 1$ electrons, the mere effect of the unpaired electron is to block one of the single-particle energy levels from pairing which in our notation means that the corresponding spin value becomes $s - 1/2$. However, using Eq. (10) this simply gives that $\Delta_P \simeq g \langle s_r \rangle$. Therefore, the parameter we introduced $\langle s_r \rangle$ (multiplied by the pairing coupling constant g) is identical to the number parity effect in the proper limit.

Next, we examine the grand canonical case or the thermodynamic limit. In its present form the model Hamiltonian (4) commutes with s_z and therefore m is a good quantum number or equivalently the number

of electrons is a conserved quantity. To make a connection with the BCS order parameter we are going to replace the interaction part of the Hamiltonian by

$$-g(s_r E \Delta^* + \Delta E^\dagger s_r - |\Delta|^2) \quad (14)$$

which is nothing but the standard mean field approximation since $s_r E = s^+$ and $E^\dagger s_r = s^-$. The BCS wave function describes a state with totally indefinite number of particles but with a definite phase. We can project the BCS states onto states of definite particle number by taking the Fourier transform with respect to the phase [31] and that is why particle number N and phase ϕ are conjugate variables with an uncertainty relation $\delta N \delta \phi \simeq 1$. It has been shown that in the thermodynamic limit the ground state of the BCS Hamiltonian (4) is also the ground state of the mean field Hamiltonian whose interaction part is given by Eq. (14) [32]. These are nothing but the phase states which we have defined above. In our case this result can be verified by observing that near half-filling and at large s , we have $[s_r, E] \simeq 0$. Therefore, we evaluate the expectation value of $s_r E$ in state $|\theta; s\mu\rangle$ and find that

$$\frac{\exp(-i2\pi\mu/(2s+1))}{2s+1} \sum_m \sqrt{s(s+1) - m(m-1)}. \quad (15)$$

We identify the phase $-2\pi\mu/(2s+1)$ as ϕ and the factor in front (the sum divided by $2s+1$) as the modulus of the order parameter $|\Delta|$. This completes our argument on the relation of $\langle s_r \rangle$ and $\langle E \rangle$ to Δ_P and Δ except one point: What happens to $\langle E \rangle$ for a system with discrete energy levels but yet with indefinite number of electrons? We note that this is not the thermodynamic limit. The system is finite but yet the number of electrons is not fixed. Such a situation can be realized through a Josephson junction.

The origin of the Josephson interaction is single-particle tunneling electron pairs. At low energies, single-particle tunneling interaction lead to two contributions both of which are second-order processes. The first one, where an electron goes from one superconductor to the other and returns, leads to proximity effect. The second one is the Josephson tunneling of two electrons from one superconductor to the other. The only effect of the first process is to renormalize the single-particle energies. Furthermore, there is no net current associated with this process. We can evaluate the explicit contributions of these two processes by considering two superconductors, both of which are described by the model Hamiltonian (4) so that

we are going to denote, the total Hamiltonian as H_0 . Let us consider a tunneling interaction of the form

$$V = t \sum_{j,j',\sigma} (c_{2j'\sigma}^\dagger c_{1j\sigma} + c_{1j\sigma}^\dagger c_{2j'\sigma}) \quad (16)$$

where $c_{1j\sigma}$ ($c_{2j\sigma}$) is the annihilation operator for state $|j\sigma\rangle$ of the first (second) superconductor. One way to introduce V perturbatively is to use the unitary transformation method [33] where the second-order Hamiltonian takes the form $H_0 + [V, \Omega]/2$. Here, the anti-Hermitian operator Ω is given by

$$\Omega = \sum_{m_1, m_2, n_1, n_2} \frac{|m_1 m_2\rangle \langle m_1 m_2| V |n_1 n_2\rangle \langle n_1 n_2|}{\epsilon_{m_1 m_2}^{(0)} - \epsilon_{n_1 n_2}^{(0)}} \quad (17)$$

where $|n_1 n_2\rangle$ denotes the ground state of H_0 with n_1 electrons in the first superconductor and n_2 electrons in the second and $\epsilon_{n_1 n_2}^{(0)}$ is the corresponding energy eigenvalue of the combined system. Since we are interested in low-energy excitations, at each step we project the system into its ground state. The two contributions we discussed above, the proximity and Josephson processes, can easily be calculated. Repeating the approximations we did in Eq. (13), we find that the strength of both terms are given by $-t^2/(\Delta_{1P} + \Delta_{2P}) = \epsilon_J$. In particular the Josephson interaction term can then be written as $\epsilon_J(E_1 E_2^{-1} + E_1^{-1} E_2)/2$ where E_i is the exponential of the phase operator in the i th superconductor. We immediately observe that for phase state $|\phi_1 \phi_2\rangle$, expectation value of this term is simply $\epsilon_J \cos(\phi_1 - \phi_2)$. To simplify our final analysis let us assume that one of the superconductors is large so that it can be described by the BCS state with a fixed phase ϕ which we can assume to be zero without loss of generality. Then for the other we can write down an effective Hamiltonian

$$H_{\text{eff}} = \sum_j 2\tilde{\epsilon}_j \left(s_j^z + \frac{1}{2} \right) - g s^+ s^- + 4\epsilon_C (s_z - \langle s_z \rangle)^2 + \epsilon_J \text{Cos} \quad (18)$$

where ϵ_C is single-electron charging energy of the island and $\tilde{\epsilon}_j$ is renormalized single-particle energy. The Josephson current $I_J = 2e\langle \dot{s}_z \rangle$ can be easily calculated as $I_J = 2e\epsilon_J \langle \text{Sin} \rangle / \hbar$ where Sin is the sine operator. Therefore, in the bulk limit where eigenstates are nearly phase states, we recover the conventional expression for the Josephson current [34,35]. Eigenstates of the Hamiltonian composed of the first three terms of H_{eff} are still given by (11) where m is a good quantum number. Hence, H_{eff} is nothing but tight-binding Hamiltonian with nearest neighbor ($m \pm 1$) hopping matrix element $\epsilon_J/2$. The nature of the eigenstates depend upon the on-site energies. For example,

for quadratic dependence of energy eigenvalues (in the absence of $\epsilon_J \text{Cos}$ term) on m , which would be the case for flat $\tilde{\epsilon}_j$, we can find the exact eigenvalues and eigenstates of H_{eff} . In this case we obtain a tight-binding Hamiltonian with on-site energies having quadratic dependence on site index and we can find the solution by observing that the expansion coefficients of the Mathieu function ce_{2n} satisfy a recursion relation which is identical to the characteristic equation of H_{eff} [36].

It is clear that I_J vanishes for any state which can be written as a linear combination of $|S; sm\rangle$ states with real expansion coefficients. These are nothing but bound states in S -space. On the other hand for propagating states, like $|\theta; s\mu\rangle$, I_J is nonzero. For small enough systems a very interesting situation may arise because discreteness of μ and hence quantized I_J values might be observed. In other words, if the number of the single-particle energy levels and hence s is not too big, we can measure a *quantized Josephson current*. A single-electron transistor with a small enough superconducting island can be used to see quantization effect. Another possibility is to measure the Josephson plasma oscillations between a bulk and nanoscopic superconductor [37,38]. Recent first principle calculations for structural and electronic properties of aluminum covered single-wall carbon nanotubes show that a stable metallic ring can be formed [39]. These structures can also allow us to observe effect of phase quantization. Coulomb interaction works in the direction to suppress the current but using an external electric field relative strength of the Josephson interaction can be increased. One possibility is to measure the Josephson current through one-dimensional array of aluminum rings formed around a carbon nanotube. In general, any physical quantity depending upon the phase is a candidate to observe quantization. For BCS gap $\Delta = 2\hbar\omega_D e^{-1/\lambda}$ and level spacing δ satisfying $\delta \lesssim \Delta$, assuming for example that we can resolve discreteness of the phase angle for $2\hbar\omega_D/\delta \simeq 1000$ states in the Debye shell at the Fermi level, we evaluate λ to be $\gtrsim 0.14$. For larger λ , we can go to smaller sizes or less number of states and hence there is more chance to observe quantization effects.

If N and ϕ are conjugate variables and the Josephson effect is a phenomenon relevant to fixed ϕ and indefinite N , what is its dual effect where N is fixed but ϕ is indefinite? When a nanoscopic superconductor is coupled by Coulomb interaction to another superconductor, there appears a second-order effect which is analogue or *dual of Josephson effect* where particle numbers are fixed but the phases are

not determined. To make the analogy complete let us consider an interaction term of the form $\varepsilon_D(F + F^{-1})$ where ε_D is the dual Josephson interaction energy and F is dual to the operator E and it is defined by

$$F = \exp \left[i \frac{2\pi}{2s+1} sz \right]. \quad (19)$$

It is easy to show that $F|\theta; s\mu\rangle = |\theta; s\mu + 1\rangle$ [10]. We can evaluate the phase current $\langle \dot{\theta}_z \rangle$ where

$$\theta_z = \sum_{\mu=-s}^s \mu |\theta; s\mu\rangle \langle \theta; s\mu|, \quad (20)$$

in complete analogy to Josephson current as $i\varepsilon_D(F^{-1} - F)/\hbar$. This interaction is similar to the van der Waals force between two molecules which is a manifestation of discreteness of electronic energy levels. In the superconducting state, intragrain single-particle excitation spectra are modified due to the number parity effect and hence there appears an additional interaction due to pairing. In other words, dual Josephson effect refers to the attractive interaction between two superconductors due to virtual Cooper pair breaking (as a result of Coulomb interaction between the superconductors) where interaction energy ε_D is of the order of the ratio of Coulomb interaction squared to superconducting gap or number parity effect parameter. This effect might also have relevance to atomic nuclei when they approach close enough so that Coulomb force becomes appreciable.

In conclusion, we proposed a complex parameter to describe pairing correlations in a fermionic system. We showed that our definition agrees with the existing parameters in the canonical and grand canonical descriptions. We predicted possible quantization in Josephson effect in the nanoscopic limit. We further analyzed the dual Josephson effect in a nanoscopic superconductor and interpreted the resulting expression in terms of quantum phase flow. Recently, the complex parameter introduced in this work has been used to study quantum entanglement in a paired finite Fermi system [40].

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