We investigate superconductivity in a grand canonical ensemble with fixed number parity (even or odd). In the low temperature limit we find small corrections to the BCS gap equation and dispersion $E(k)$. The even-odd free energy difference in the same limit decreases linearly with temperature, in accordance with the behavior observed experimentally and previously arrived at from a quasiparticle model. The theory yields deviations from the BCS predictions for the specific heat, ultrasound attenuation, NSR relaxation rate, and electromagnetic absorption.

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I. INTRODUCTION

An extremely interesting recent development is the experimental study of capacitatively isolated superconducting metallic islands to which electrons can be added one by one. Two different experimental probes of such islands have demonstrated that even when the number of electrons is extremely large—of the order of $10^9$—a periodicity is observed for every two electrons added. Since the microscopic BCS theory of superconductivity is a pairing theory, it is natural to assume that the observed alternation is due to the evenness or oddness of the number of electrons on the island. At first sight one might have guessed that adding electrons would involve a localized state near the injection site, first filling it and then emptying it to form a pair. However, the temperature and magnetic field dependence of the effect confirm that for the reported experiments the relevant energies relate to superconducting correlations. Since these experiments probe fundamental properties of superconductors their precise analysis is important. What makes this problem particularly interesting from the point of view of theory is the fact that superconducting order is off-diagonal in number so that the treatment of fixed number is problematical.

Tuominen, Hergenrother, Tighe, and Tinkham have suggested that equilibrium properties can be calculated from the partition functions

$$Z_{e/o} = \frac{1}{2} \left\{ \prod_{k,\sigma} [1 + e^{-\beta E_{k,\sigma}}] \pm \prod_{k,\sigma} [1 - e^{-\beta E_{k,\sigma}}] \right\}, \quad (1.1)$$

where $e$ means even and $o$ odd, and $E_{k,\sigma}$ are the quasiparticle energies. The sign $\pm$ retains only configurations containing even or odd numbers of quasiparticles in a grand canonical ensemble. This approach has also been adopted by Lafarge et al. in interpreting their experimental results. Now, Eq.(1.1) cannot be quite right because the BCS theory is a self-consistent theory in which the energy is not simply a sum of quasiparticle energies. On the other hand, the number parity of a state with $N$ quasiparticles is the same as that of a state with $N$ electrons. In this paper we explore the consequences of treating the islands by considering the appropriate generalization of (1.1), namely

$$Z_{e/o} = \frac{1}{2} \text{Tr} \left\{ [1 \pm (-1)^N] e^{-\beta (H - \mu N)} \right\}.$$

(1.2)

We obtain the free energy for both even and odd case and show that in the low temperature limit it reduces to the ansatz (1.1). Using the same framework, we calculate the ultrasound attenuation, NSR relaxation rate, and electromagnetic absorption and find corrections to the standard BCS results at low temperature.

The method of Eq.(1.1) is normally used in another context where fixed number is important: nuclear physics. There energy-levels are calculated using an unrestricted grand canonical ensemble but only those with the appropriate quantum number for the nucleus in question are kept. Since we are here interested in thermodynamic properties which are related to fluctuations, it would seem more correct at the outset to suppress states with the wrong number parity in the statistical ensemble, as in (1.2).

In the BCS theory one fixes the average number of electrons $N$ by adjusting the chemical potential. The fluctuations in particle number (of order $\sqrt{N\Delta/E_F}$) at $T = 0$, where $\Delta$ is
the energy gap and $E_F$ the Fermi energy) are negligible for a macroscopic system. Since we are interested in a small free energy difference in the present work, we keep track of number fluctuations in both the even and the odd ensembles of (1.2), and explicitly check that they do not affect the physical properties we calculate.

This paper is organized as follows: in Sec. (II) we calculate the partition functions for the parity restricted ensembles; in Sec. (III) we minimize the grand potential and obtain the gap equation and dispersion for even or odd parity. We find only small corrections to the BCS results. Sec. (IV) is devoted to extracting thermodynamic properties from the grand potential. The differences between the results obtained with the method of Eq. (1.2) and the nuclear physics method, Eq. (1.1), turn out too small to be measurable with the experimental techniques presently available. All subsequent calculations are made within the approximation (1.1). We evaluate the even/odd free energy difference paying special attention to number fluctuations. The entropy and specific heat of the even and odd parity systems are also calculated: the results show deviations from BCS. In Sec. (V) we show how the number parity restriction affects the response of an even- or odd-N superconductor to various external probes. The final Sec. (VI) contains a summary and discussion.

II. CALCULATION OF PARTITION FUNCTION

In principle the most suitable framework for discussing the problem would be to consider $N$ particles interacting via the BCS interaction and construct a BCS-like trial ground-state wave function and excited states with a fixed number of particles. For our purposes, however, this scheme runs into technical difficulties. The fixed-$N$ BCS wave function cannot be normalized exactly, and a saddle point approximation yields the grand-canonical BCS results.

If one tries to use the even and odd ground states in the usual grand-canonical method, the difference between them will be practically removed by the parity-violating grand-canonical process of a single-particle exchange between the system and the particle reservoir.

In the partition function (1.2) the projection operator $[1 \pm (-1)^N/2]$ suppresses states with odd and even number parity, respectively. The expectation value of an operator in such an ensemble has the form

$$\langle O \rangle_{e/o} = Z_{e/o}^{-1} \text{Tr} \left\{ \frac{1 \pm (-1)^N}{2} O e^{-\beta (H-\mu N)} \right\}. \quad (2.1)$$

The reduced BCS Hamiltonian in the expressions above,

$$\mathcal{H} - \mu N = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} - \sum_{kk'} V_{kk'} c_{k\uparrow}^\dagger c_{k'\downarrow}^\dagger c_{-k'\downarrow} c_{k\uparrow}, \quad (2.2)$$

(where the single-particle energies are measured from $\mu$) can be transformed, using the Bogoliubov-Valatin transformation

$$\gamma_{k0} = u_k c_{k\uparrow} - v_k c_{-k\downarrow}, \quad (2.3)$$
$$\gamma_{k1} = v_k c_{k\uparrow} + u_k c_{-k\downarrow}, \quad (2.4)$$
$$u_k^2 + v_k^2 = 1, \quad (2.5)$$

3
into the expression

$$\mathcal{H} - \mu N = \mathcal{H}_Q + \mathcal{H}_I + U. \quad (2.6)$$

Here $\mathcal{H}_Q$ corresponds to the kinetic energy of the Bogoliubov quasiparticles

$$\mathcal{H}_Q = \sum_k E_k (\gamma_{k0}^\dagger \gamma_{k0} + \gamma_{k1}^\dagger \gamma_{k1}). \quad (2.7)$$

where $E_k$ will be obtained later on by minimizing the grand potential. Note that if $(2.7)$ were substituted into $(1.2)$, Eq. $(1.1)$ would result. However, there are two additional terms in $(2.6)$. $\mathcal{H}_I$ describes the interaction between the elementary excitations.

$$\mathcal{H}_I = \sum_k \left[ \epsilon_k (u_k^2 - v_k^2) + 2u_kv_k \sum_{k'} u_{k'}v_{k'} V_{kk'} - E_k \right] (\gamma_{k0}^\dagger \gamma_{k0} + \gamma_{k1}^\dagger \gamma_{k1})$$

$$- \sum_{k,k'} V_{kk'} u_k v_k u_{k'} v_{k'} (\gamma_{k0}^\dagger \gamma_{k0} + \gamma_{k1}^\dagger \gamma_{k1}) (\gamma_{k0}^\dagger \gamma_{k0} + \gamma_{k1}^\dagger \gamma_{k1}). \quad (2.8)$$

Finally, $U$ is a c-number term depending on temperature :

$$U = 2 \sum_k \epsilon_kv_k^2 - \sum_{k,k'} V_{kk'} u_k v_k u_{k'} v_{k'}. \quad (2.9)$$

We now make the BCS mean field approximation by assuming that the low temperature dynamic properties of the system are governed by the independent quasiparticles described by $\mathcal{H}_Q$, and that the interaction term can be replaced by its thermal average

$$\mathcal{H} - \mu N \simeq \mathcal{H}_Q + C_{e/o}, \quad (2.10)$$

where the temperature dependent constant $C_{e/o}$ is defined as

$$C_{e/o} = \langle \mathcal{H}_I \rangle_{e/o} + U = \frac{\text{Tr}\left\{\frac{1+(-1)^N}{2} \mathcal{H}_I e^{-\beta \mathcal{H}_Q}\right\}}{\text{Tr}\left\{\frac{1+(-1)^N}{2} e^{-\beta \mathcal{H}_Q}\right\}} + 2 \sum_k \epsilon_k v_k^2 - \sum_{k,k'} V_{kk'} u_k v_k u_{k'} v_{k'}. \quad (2.11)$$

In this approximation it is possible to calculate the even-odd partition function explicitly. The result can be given in terms of the quantities

$$Z_{\pm} = \prod_{k,\sigma} (1 \pm e^{-\beta E_k}), \quad (2.12)$$

$$f_{\pm}(E_k) = \frac{\pm 1}{e^{\beta E_k} \pm 1}. \quad (2.13)$$

Here $f_{\pm}(E_k)$ is the usual Fermi function. Then the even-odd partition function has the form

$$Z_{e/o} = \frac{1}{2} (Z_+ \pm Z_-) e^{-\beta C_{e/o}}. \quad (2.14)$$

In the same mean field approximation $C_{e/o}$ becomes
\[ C_{e/o} = C_+ + \delta C_{e/o}, \]
\[ \delta C_{e/o} = \pm \frac{Z_-(C_- - C_+)}{Z_+ \pm Z_-}. \] (2.15)

The function \( C_+ \) in the above expression is the BCS result for the constant C,

\[
C_+ = 2 \sum_k \epsilon_k v_k^2 + 2 \sum_k \left[ \epsilon_k(u_k^2 - v_k^2) - E_k \right] f_+(E_k) - \sum_{k,k'} V_{kk'}u_k v_k u_{k'} v_{k'} [1 - f_+(E_k)] [1 - f_+(E_{k'})],
\] (2.16)

whereas \( C_- \) can be obtained from \( C_+ \) by a replacement of \( f_+(E_k) \) with \( f_-(E_k) \),

\[
C_- = C_+ [f_+(E_k) \to f_-(E_k)].
\] (2.17)

Inserting these relations into Eq. (2.15) we find

\[
\delta C_{e/o} = -\frac{2}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}} \left\{ \sum_k \frac{\epsilon_k(u_k^2 - v_k^2) - E_k}{\sinh \beta E_k} + 2 \sum_{k,k'} V_{kk'}u_k v_k u_{k'} v_{k'} \coth \frac{\beta E_k}{2} \sinh \frac{\beta E_k}{2} \right\}.
\] (2.18)

The even-odd grand potential can be directly obtained from the even-odd partition function via \( \Omega_{e/o} \equiv -k_B T \ln Z_{e/o} \). Casting \( \Omega_{e/o} \) in a form that makes the even-odd corrections explicit, we finally obtain

\[
\Omega_{e/o} = \Omega_{BCS} - \frac{1}{\beta} \ln \left[ \frac{1}{2} \left( 1 \pm \prod_{k,\sigma} \tanh \frac{\beta E_k}{2} \right) \right] + \delta C_{e/o}.
\] (2.19)

The last two terms in Eq. (2.19) represent corrections arising from even-odd effects. These corrections vanish for large enough temperature and/or volume. The easiest way to see how this happens when the temperature is increased is to calculate the product

\[
\prod_{k,\sigma} \tanh \frac{\beta E_k}{2} \equiv e^{\sum_{k,\sigma} \ln \tanh \frac{\beta E_k}{2}}.
\] (2.20)

For \( \Delta \ll k_B T \), so that \( E_k \simeq |\epsilon_k| \), one obtains

\[
\prod_{k,\sigma} \tanh \frac{\beta E_k}{2} = e^{-3\pi^2 N/2\beta \epsilon_F}.
\] (2.21)

where \( N \) is the total number of particles and \( \epsilon_F \) is the Fermi energy. Since all even-odd corrections are proportional to some power of this product, such effects are negligible at temperatures close to \( T_c \). As we will see, a much lower temperature limit for the onset of even-odd effects can be set if we start from the low temperature limit and calculate the temperature where the even-odd free energy difference vanishes. In fact, when \( \Delta \gg k_B T \), the relevant excitations are \( E_k \simeq \Delta \). Defining \( N_{eff} \) by

\[
N_{eff} = \frac{1}{\beta} \ln \left[ \frac{1}{2} \left( 1 \pm \prod_{k,\sigma} \tanh \frac{\beta E_k}{2} \right) \right] + \delta C_{e/o}.
\]
\[ \prod_{k, \sigma} \tanh \frac{\beta E_k}{2} \equiv \left( \tanh \frac{\beta \Delta}{2} \right)^{N_{eff}}, \]  

(2.22)

direct calculation shows that
\[ N_{eff} \simeq 4 N(0)V \int_\Delta^\infty dE E \sqrt{E^2 - \Delta^2} e^{-\beta(E-\Delta)} \simeq 2 N(0)V \sqrt{2\pi k_B T \Delta}, \]

(2.23)

where \( V \) is the volume of the system. Since \( \tanh x < 1 \) for all finite arguments \( x \), the product is again negligible for large enough \( N_{eff} \), i.e. large volume or particle number. For the samples used by Tuominen et al. and Lafarge et al. \( N_{eff} \sim 10^4 \).

III. MINIMIZATION OF THE GRAND POTENTIAL

Before performing the minimization it is useful to rearrange the terms in Eq.(2.19) as follows
\[ \Omega_{e/o} = \Omega_{BCS}(f_+(E_k) \rightarrow f(E_k)_{e/o}) - 4 \sum_{k, k'} V_{kk'} u_k v_k u_{k'} v_{k'} \left[ \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o} \right], \]

(3.1)

where \( n_k = \gamma_k^\dagger \gamma_k \) is the quasiparticle number operator and
\[ f(E_k)_{e/o} \equiv \langle n_k \rangle_{e/o} \frac{f_+(E_k) Z_+ \pm f_-(E_k) Z_-}{Z_+ \pm Z_-}. \]

(3.2)

Also
\[ \Omega_{BCS}(f_+(E_k) \rightarrow f(E_k)_{e/o}) = -\frac{1}{\beta} \ln \left[ \frac{1}{2} \left( Z_+ \pm Z_- \right) \right] + \sum_{k, \sigma} \epsilon_k v_k^2 + \sum_{k, k'} \left[ \epsilon_k (u_k^2 - v_k^2) - E_k \right] f_{e/o}(E_k) - \sum_{k, k'} V_{kk'} u_k v_k u_{k'} v_{k'} \left[ 1 - 2 f_{e/o}(E_k) \right] \left[ 1 - 2 f_{e/o}(E_{k'}) \right]. \]

(3.3)

As we shall see later on, the last term in Eq.(3.1) is small, but nonzero. This is an explicit manifestation of the fact that the thermal Wick’s theorem is weakly violated in the even/odd ensembles. The grand potential is a functional of two independent functions, say \( E_k \) and \( u_k, \Omega_{e/o} = \Omega_{e/o}(\{E_k, v_k\}) \). Minimization with respect to these parameters will give two variational equations, which in the BCS case yield the energy spectrum and the gap equation. Following the same strategy, we obtain
\[ 2\epsilon_k u_k v_k - (u_k^2 - v_k^2) \Delta_{k}^{e/o} = A_{k}^{e/o}, \]

(3.4)
\[ E_k - \epsilon_k (u_k^2 - v_k^2) - 2u_k v_k \Delta_k^{e/o} = B_k^{e/o}. \]

(3.5)

Here \( \Delta_k^{e/o} \) is defined as
\[ \Delta_k^{e/o} \equiv \sum_{k'} V_{k, k'} u_{k'} v_{k'} [1 - 2 f_{e/o}(E_{k'})]. \]

(3.6)

Note that \( u_k \) and \( v_k \) are also parity dependent. We have omitted the e/o labels for the sake of simplicity. We shall see later on that the gap in the spectrum is somewhat different from \( \Delta_k^{e/o} \).
Both $A^{e/o}_k$ and $B^{e/o}_k$ were generated by the residual, Wick’s theorem violating term in Eq.(3.1).

\[ A^{e/o}_k = 4 \frac{u_k^2 - v_k^2}{1 - 2f_{e/o}(E_k)} \sum_{k,k'} V_{kk'} u_k v_k u_{k'} v_{k'} \left( \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o} \right), \]  
(3.7)  

\[ B^{e/o}_k = 4 \frac{u_k v_k}{\partial f_{e/o}(E_k)/\partial E_k} \sum_{k,k'} V_{kk'} u_k v_k u_{k'} v_{k'} \frac{\partial}{\partial E_k} \left( \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o} \right). \]  
(3.8)  

These coefficients also vanish in the thermodynamic BCS limit, since then, for reasons similar to those given above for the grand potential, even/odd effects are small, and Wick’s theorem is restored. Also note that, since $A^{e/o}_k$ and $B^{e/o}_k$ are proportional to $u_k^2 - v_k^2$ and $u_k v_k$, respectively, they can be interpreted as changes in $\Delta^{e/o}_k$ due to Wick’s theorem violation.

### A. Exact solution of the minimization equations

It is possible to give a formal exact solution to (3.4) and (3.5) by making the usual change of variables $u_k = \cos \theta_k$, $v_k = \sin \theta_k$. We obtain from the first equation of minimization, (3.4) a solution similar to the BCS case

\[ u_k = \sqrt{\frac{1}{2} \left( 1 + \frac{\epsilon_k}{(\epsilon_k^2 + \Delta^{e/o}_k)^{1/2}} \right)}, \]  
(3.9)  

\[ v_k = \sqrt{\frac{1}{2} \left( 1 - \frac{\epsilon_k}{(\epsilon_k^2 + \Delta^{e/o}_k)^{1/2}} \right)}, \]  
(3.10)  

but now $\Delta^{e/o}_k$ is given by

\[
\Delta^{e/o}_k = \sum_{k'} V_{k,k'} \Delta^{e/o}_{k'} \frac{1}{2(\epsilon_k^2 + \Delta^{e/o}_k)^{1/2}} \left( [1 - 2f_{e/o}(E_{k'})] \right) 
+ \frac{1}{1 - 2f_{e/o}(E_k)} \left[ \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o} \right], \]  
(3.11)  

The spectrum $E_k$ can be obtained from (3.5) by using the above results.

\[
E_k = (\epsilon_k^2 + \Delta^{e/o}_k)^{1/2} \left( \frac{\Delta^{e/o}_k}{(\epsilon_k^2 + \Delta^{e/o}_k)^{1/2}} \right) \sum_{k'} V_{k,k'} \Delta^{e/o}_{k'} \frac{1}{2(\epsilon_k^2 + \Delta^{e/o}_k)^{1/2}} 
\times \left( \frac{2}{1 - 2f_{e/o}(E_k)} + \frac{1}{\partial f_{e/o}(E_k)/\partial E_k} \right) \left[ \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o} \right]. \]  
(3.12)  

The expressions above contain the quantity

\[ \rho_{e/o}(k,k') \equiv \langle n_k n_{k'} \rangle_{e/o} - \langle n_k \rangle_{e/o} \langle n_{k'} \rangle_{e/o}. \]  
(3.13)  

This can be evaluated explicitly. For $k \neq k'$
\[
\rho_{e/o}(k, k') = \frac{f_+(E_k)f_+(E_{k'})Z_+ \pm f_- (E_k)f_- (E_{k'})Z_-}{Z_+ \pm Z_-}.
\]

Inserting the expressions for \( f_{\pm} \) and \( Z_{\pm} \) [Eq. (2.13 - 2.12)], we get

\[
\rho_{e/o}(k, k') = \text{cosech}\beta E_k \text{cosech}\beta E_{k'} \left( \frac{1}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}} \right) \left( 1 \pm \prod_{k,\sigma} \tanh \frac{\beta E_k}{2} \right). \tag{3.15}
\]

On the other hand, for \( k = k' \) \( \rho(k, k) \) has the usual form for noninteracting fermions

\[
\rho_{e/o}(k, k') = f_{e/o}(E_k) - f_{e/o}(E_k)^2 \tag{3.16}
\]

The results obtained so far are exact. The expressions for \( \Delta_{e/o} \) [Eq. (3.11)] and the dispersion \( E_k \) [Eq. (3.12)] constitute a closed system of two self-consistent equations which in principle can be solved iteratively. It is, however, possible to give approximate analytic result for the low temperature limit.

**B. Low temperature approximation of the exact solution**

The temperature range where the experiments revealed even/odd effects is 50mK < \( T < T^* \approx 200\text{mK} \). In this temperature range, for the given island parameters \( N_{eff} \sim 10^4 \). Then the following inequalities hold:

\[
\exp(-\beta \Delta) \ll \frac{1}{N_{eff}} \ll 1. \tag{3.17}
\]

Here \( \Delta \) is the smallest value of the dispersion \( E_k \) for both the even and odd cases. As we shall see the zero temperature limit of the gap in the even and the odd case differs, but by a small amount only. So in first approximation \( \Delta \) is the same for both parities. These inequalities enable us to systematically keep track of the terms in an expansion of the exact result and to make a controlled approximation. In this regime

\[
\frac{1}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}} \approx \frac{1}{1 \pm (1 + 2N_{eff}e^{-\beta \Delta_{e/o}})}, \tag{3.18}
\]

\[
\frac{1}{1 \pm \prod_{k,\sigma} \tanh \frac{\beta E_k}{2}} \approx \frac{1}{1 \pm (1 - 2N_{eff}e^{-\beta \Delta_{e/o}})}. \tag{3.19}
\]

Here \( \Delta_{e/o}^0 \equiv \Delta_{e/o}(T = 0) \) The parity dependent average occupation number \( f_{e/o}(E_k) \) then becomes

\[
f_{e/o}(E_k) = f_+(E_k) - \frac{\text{cosech}\beta E_k}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}} \approx \exp(-\beta E_k) - \frac{2 \exp(-\beta E_k)}{1 \pm (1 + 2N_{eff}e^{-\beta \Delta_{e/o}})}. \tag{3.20}
\]
For the even parity we have
\[ f_e(E_k) = N_{eff} \exp[-\beta(E_k + \Sigma^0_e)], \] (3.21)

whereas for the odd case
\[ f_o(E_k) = \exp(-\beta E_k \left(1 + \frac{\exp(\beta \Delta^0_e)}{N_{eff}}\right)). \] (3.22)

Similar calculation leads to approximate results for \( \rho_{e/o}(k, k') \). We obtain for \( k \neq k' \)
\[ \rho_e(k, k') = \exp[-\beta(E_k + E_{k'})] \sim O \exp(-2\beta \Delta) \] (3.23)
\[ \rho_o(k, k') = \exp[-\beta(E_k + E_{k'} - 2\Sigma^0_o)] \sim O(1/N_{eff}^2). \] (3.24)

We see that \( \rho_{e/o}(k, k') \) is smaller than the even/odd corrections coming from \( f_{e/o}(E_k) \) by at least a factor of \( 1/N_{eff} \), and therefore can be safely neglected in Eq. (3.11) and Eq. (3.12).

The contribution from the case \( k = k' \) is also negligible, since it represents only one term in a summation over \( k' \). Furthermore
\[ \frac{1}{-\partial f_{e/o}(E_k)/\partial E_k} \frac{\partial \rho_{e/o}(k, k')}{\partial E_k} = \pm \exp[-\beta(E_{k'} - \Sigma^0_{e/o})] \frac{1}{N_{eff}}. \] (3.25)

Finally, using the results we obtained above, it is possible to give a somewhat simpler, approximate expression for \( \Sigma^0_{e/o} \) and \( E_k \).
\[ \Sigma^0_{e/o} = \sum_{k'} V_{k,k'} \frac{\Sigma^0_{k'}}{2(\epsilon_k^2 + \Sigma^0_{e/o})^{1/2}} [1 - 2f_{e/o}(E_{k'})] + O\left(\frac{1}{N_{eff}^2}\right). \] (3.26)
\[ E_k = (\epsilon_k^2 + \Sigma^0_{e/o})^{1/2} \sum_{k'} V_{k,k'} \exp[-\beta(E_{k'} - \Sigma^0_{e/o})] + O\left(\frac{1}{N_{eff}^2}\right). \] (3.27)

Notice that within this approximation \( \Sigma^0_{e/o} = \Delta^0_{e/o} \). From Eq. (3.26), we can now extract the low temperature behavior of \( \Delta_{e/o} \). Using \( E_k \approx (\epsilon_k^2 + \Delta^2_{e/o})^{1/2} \), we obtain for the even case
\[ \Delta_e(T) \simeq \Delta_{BCS}(0) \left[1 - (4\pi k_B T N(0) V) \exp(-2\beta \Delta_e(0))\right]. \] (3.28)

Here \( \Delta_{BCS}(0) \) is the zero temperature limit of the BCS gap for the same interaction \( V_{k,k'} \). Note that \( \Delta_e(0) = \Delta_{BCS}(0) \). We used the expression (2.23) for \( N_{eff} \) to obtain the prefactor of the exponential. The most significant difference between the above expression and the usual BCS result is the \( \exp(-2\beta \Delta) \) decrease of the gap, to be contrasted with the \( \exp(-\beta \Delta) \) behavior predicted by the conventional BCS theory. Furthermore the prefactor of the exponential is linear in \( T \), whereas in the BCS is proportional to \( T^{1/2} \). For odd parity
\[ \Delta_o(T) \simeq \Delta_{BCS}(0) \left[ 1 - \frac{1}{2\Delta_o(0)N(0)V} - \left( \frac{2\pi k_B T}{\Delta_o(0)} \right)^{1/2} \exp(-\beta \Delta_o(0)) \right]. \quad (3.29) \]

Note that in the odd case \( \Delta_o(T) \) saturates to a value that is less than the BCS gap:

\[ \Delta_o(0) \simeq \Delta_{BCS}(0) - \frac{1}{2N(0)V}. \quad (3.30) \]

The difference, however, is very small, \( \sim 10^{-4} \Delta_{BCS} \). Finally, let us examine Eq. (3.27) which gives the approximate dispersion. We can evaluate the sum in the second term and obtain

\[ \sum_{k'} V_{k,k'} \exp\left[ -\beta \left( E_{k'} - \Delta_{e/o}^0 \right) \right] \simeq \lambda \left( \frac{2\pi k_B T}{\Delta_{e/o}(0)} \right)^{1/2}. \quad (3.31) \]

where \( \lambda = |V_{k,k'}|_{av} N(0)V \simeq 1/4 \) is the superconducting coupling constant. The smallest possible value of the dispersion, \( \delta_{e/o} \equiv E(k = k_F) \), is slightly different from \( \Delta_{e/o} \). In fact, from Eq. (3.27) we get

\[ \delta_{e/o} = \Delta_{e/o} \left[ 1 \pm \frac{\lambda}{N_{eff}} \left( \frac{2\pi k_B T}{\Delta_{e/o}(0)} \right)^{1/2} \right]. \quad (3.32) \]

This means that low temperature behavior of the spectral gap \( \delta_{e/o} \) will be somewhat different from that of \( \Delta_{e/o} \)

\[ \delta_e(T) \simeq \Delta_{BCS}(0) \left[ 1 - \frac{\lambda}{2\Delta_o(0)N(0)V} - \left( 4\pi k_B T N(0)V \right) \exp(-2\beta \Delta_e(0)) \right], \quad (3.33) \]

\[ \delta_o(T) \simeq \Delta_{BCS}(0) \left[ 1 - \frac{1 - \lambda}{2\Delta_o(0)N(0)V} - \left( \frac{2\pi k_B T}{\Delta_o(0)} \right)^{1/2} \exp(-\beta \Delta_o(0)) \right]. \quad (3.34) \]

Clearly, both \( \delta_e(0) \) and \( \delta_o(0) \) is smaller than \( \Delta_{BCS}(0) \)

\[ \delta_o(0) \simeq \Delta_{BCS}(0) - \frac{\lambda}{2N(0)V}, \quad (3.35) \]

\[ \delta_o(0) \simeq \Delta_{BCS}(0) - \frac{1 - \lambda}{2N(0)V}. \quad (3.36) \]

Nevertheless, since \( \lambda < 1 \), the odd gap is depleted somewhat more than the even gap. The deviations from the BCS results found in this section are probably too small to be experimentally observable in the quantities we investigate later on. Therefore, in the subsequent calculations we ignore any deviation from the BCS gap equation and dispersion.
IV. THERMODYNAMIC PROPERTIES

Now we can estimate the grand potential itself and the even-odd grand potential difference in the low temperature limit $k_B T \ll \Delta$, by using the standard BCS relations for $E_k$ and $v_k$ in eq. (2.19). In this limit

$$\delta C_{e/o} \simeq \frac{-2}{1 \pm (1 + 2N_{eff} e^{-\beta \Delta})} \times \sum_{kk'} \frac{4\Delta_k \Delta_{k'}}{E_k E_{k'}} e^{-\beta(E_k + 2E_{k'})}$$

(4.1)

is exponentially small for both even and odd parity. In fact, after neglecting the parity dependence of $C$, the results obtained within this approximation do not differ from those obtained from Eq. (1.1). For the even-odd grand potential difference, calculated at a fixed chemical potential $\mu$, we obtain

$$\delta \Omega_{e/o}(\mu) \equiv \Omega_o(\mu) - \Omega_e(\mu) = \Delta - k_B T \ln N_{eff}.$$  

(4.2)

The experimentally relevant quantity is the free energy difference of two isolated (canonical) systems with $2N + 1$ and $2N$ particles respectively. This can be related to $\delta \Omega_{e/o}$ through the relation

$$e^{-\beta \Omega} = \sum_N e^{-\beta F_N(T)} e^{\beta N}.$$  

(4.3)

where $F_N(T)$ is the free energy energy for a canonical ensemble. Using a Gaussian approximation about the peak of the summand, one obtains

$$\Omega_o(\mu_o) - \Omega_e(\mu_e) \simeq F(2N + 1) - \mu_o(2N + 1) + F(2N) - \mu_e 2N - \frac{1}{2\beta} \ln \left(\frac{\Delta N_{e/o}^2}{\Delta N_e^2}\right).$$  

(4.4)

where $\Delta N_{e/o}^2$ are the number fluctuations in the grand ensembles (1.1). The investigation of how the number fluctuations depend on the number parity of the ensemble is an important test of the present model, since one must check that the fluctuations in the parity restricted grand canonical ensembles do not wash out the free energy difference one is attempting to calculate.

The number operator written in terms of the Bogoliubov creation and annihilation operators has the form

$$N = \sum_k v_k^2 + (u_k^2 - v_k^2)[\gamma_{k0}^\dagger \gamma_{k0} + \gamma_{-k1}^\dagger \gamma_{-k1}] + 2u_k v_k[\gamma_{k0}^\dagger \gamma_{-k1}^\dagger - \gamma_{k0} \gamma_{-k1}].$$  

(4.5)

The last two terms are off diagonal in quasiparticle number, and therefore they do not contribute to the average particle number. (They, however, turn out to be crucial for the mean square fluctuations $\Delta N_{e/o}^2$.) The average quasiparticle number is

$$\langle N \rangle_{e/o} = \sum_k v_k^2 + (u_k^2 - v_k^2) \frac{f_+ Z_+ \pm f_- Z_-}{Z_+ \pm Z_-}.$$  

(4.6)
Now we can fix the chemical potentials for the even/odd systems in (1.4), since they satisfy the above equation with $<N>_e = 2N$ and $<N>_o = 2N + 1$, respectively. From (4.6) we obtain that $\mu_o = \mu_e + \delta \mu$, where $\delta \mu \propto \epsilon_F/2N$ is of the order of the level spacing (much smaller than $\Delta$).

When calculating the mean square fluctuations $\Delta N_{e/o}^2 = <N^2>_e - <N>_e^2$ by using the operator expression in eq. (4.5, we see that the off-diagonal terms now give a nonzero contribution, proportional to $(\Delta/E_k)^2$

$$\Delta N_{e/o}^2 = \sum_k \left\{ 2\left(\frac{\epsilon_k}{E_k}\right)^2[f_+^2(1 - f_+) - f_+^2 - f_-^2] - 4\left(\frac{\Delta}{E_k}\right)^2[f_+^2 + (1 - f_+)^2 - \frac{f_+^2 - f_-^2}{1 \pm Z_-/Z_+} - \frac{(1 - f_+)^2 - (1 - f_-)^2}{1 \pm Z_-/Z_+}] \right\}.$$ (4.7)

At low temperatures, when $f_\pm \rightarrow \pm \exp(-\beta \Delta)$, direct calculation shows that:

$$\Delta N_{e/o}^2 = 4 \sum_k (\Delta/E_k)^2 + O(1/N_{eff}),$$ (4.8)

regardless of the even or odd nature of the ensemble. This very important result reassures us that thermal and quantum fluctuations in the particle number do not dominate the difference between the grand canonical ensembles of even and odd parity in Eq.(1.4). Therefore this modified grand canonical method can be used to calculate even/odd differences in canonical quantities. One should note that this property is intrinsic to the superconducting state: at $T = 0$ the fluctuations are of a purely quantum nature $\Delta N_{e/o} \propto \sqrt{N\Delta/E_F}$, and decrease as the superconducting gap $\Delta$ is depleted. In contrast, the same quantity for the normal state is $\Delta N_{e/o} \propto \sqrt{NT/T_F} \rightarrow 0$ as $T \rightarrow 0$.

Expanding $\Omega_e(\mu_e) \simeq \Omega_e(\mu_o) + \delta \mu 2N$ on the left-hand side of Eq.(1.4), and using (4.8) and (4.12), we obtain:

$$F(2N + 1) - F(2N) - \mu = \Delta - k_B T \ln N_{eff}.$$ (4.9)

This expression, previously given in refs.13, has been used to provide a reasonable account of the highest temperature at which the 2e periodicity of the I-V characteristics for the single-electron-transistor is experimentally observed by Tuominen and collaborators13. $T^*$ = $\Delta/k_B \ln N_{eff} \sim 300$ mK. It also agrees with the temperature dependence of $\delta F_{e/o}$ measured by Lafarge et al.13.

The method presented here allows us to calculate other possibly measurable properties of the superconducting state with restricted particle number parity. For example, we can calculate the entropy from the expression

$$S_{e/o} = -k_B Tr\{\rho_{e/o} \ln \rho_{e/o}\},$$ (4.10)

where the density matrix is given by

$$\rho_{e/o} = Z_{e/o}^{-1} \frac{1 \pm (-1)^N}{2} e^{-\beta(H - \mu N)},$$ (4.11)

After performing the parity restricted trace and using the results we obtained previously for $Z_{e/o}$ (Eq. (2.14)) we get
\[ S_{e/o} = S_{BCS} + \frac{1}{T} \sum_k E_k \delta f_{e/o} + k_B \ln \left[ \frac{1}{2} \left( 1 \pm \prod_{k,\sigma} \tanh \frac{\beta E_k}{2} \right) \right], \tag{4.12} \]

where \( S_{BCS} \) is the entropy of a BCS superconductor
\[ S_{BCS} = -k_B \sum_{k,\sigma} \{(1 - f_+) \ln(1 - f_+) + f_+ \ln f_+ \}, \tag{4.13} \]

and \( \delta f_{e/o} \) is the even/odd change in the quasiparticle occupation \( f_+ \)
\[ \delta f_{e/o} = \frac{f_+ - f_-}{1 \pm \mathcal{Z}_+ / \mathcal{Z}_-} = \frac{1}{\sinh \beta E_k} \frac{1}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}}. \tag{4.14} \]

In the low temperature limit the entropy for the even-parity system is
\[ S^e \simeq k_B \beta \Delta N_{eff}^2 e^{-2\beta \Delta}, \tag{4.15} \]

whereas for the odd-parity system we have
\[ S^o \simeq k_B \ln N_{eff} + S_{BCS}. \tag{4.16} \]

Here \( S_{BCS} = k_B N_{eff} (\beta \Delta) e^{-\beta \Delta} \). The specific heat \( C \) can be obtained directly from the above expression via
\[ C_{e/o} = T \frac{\partial S_{e/o}}{\partial T}. \tag{4.17} \]

More specifically, for the even case
\[ C^e_v \simeq k_B (N_{eff} \beta \Delta)^2 e^{-2\beta \Delta}. \tag{4.18} \]

Note the \( \exp(-2\beta \Delta) \) dependence of the heat capacity for this case, different from the usual \( \exp(-\beta \Delta) \) in the BCS case. This discrepancy is the direct consequence of the number parity restriction applied to the system. The odd-parity system has a specific heat that is finite at all temperatures due to the presence of an unpaired electron:
\[ C^o_v \simeq \frac{1}{2} k_B + C_{vBCS}. \tag{4.19} \]

Finally, we would like to make some technical remarks regarding the calculations presented in this paragraph. The result in Eq. (4.13) can be obtained in another way as well, by using the thermodynamic relation \( <N>_{e/o} = -\partial \Omega_{e/0} / \partial \mu \) keeping in mind the requirements from minimization, \( \partial \Omega_{e/0} / \partial E_k = 0 \) and \( \partial \Omega_{e/0} / \partial v_k = 0 \). In contrast to this, the thermodynamic relationship \( \Delta N^2_{e/o} = \frac{1}{\beta} \partial <N> / \partial \mu \) does not give the correct result for the mean square fluctuations: the crucial off-diagonal terms do not contribute to \( <N> \) and as a result are missed in the expression for \( \Delta N^2_{e/o} \) derived in this way. This reflects the peculiarity of the superconducting state. Yet another possible source of error is encountered if one tries to calculate the entropy by directly applying the thermodynamic relation \( S = -(\partial \Omega / \partial T)_V \) instead of using the expression (4.10). The reason is the following: the temperature dependence of the function \( C = <H_I> \) is an artifact of the mean field approximation. Therefore, when calculating the entropy in this way, one should not take the derivative of \( C \). In fact, by evaluating \( S_{e/o} \) with the help of Eq. (4.10), one can explicitly show that within the mean field approximation all contributions from \( C_{e/o} \) drop out. Exactly the same situation arises in the ordinary BCS calculation.
V. TRANSPORT PROPERTIES

Here we calculate the response of the parity restricted superconducting state to various external probes. The acoustic attenuation rate of ultrasound in a superconductor, within the framework of the BCS theory, is given by the expression

\[ \alpha_S(T) = \frac{2\alpha_N(T)}{\hbar \omega} \int_{-\Delta}^{\infty} dE \frac{E}{\sqrt{E^2 - \Delta^2}} \frac{E'}{\sqrt{(E')^2 - \Delta^2}} \left(1 - \frac{\Delta^2}{EE'}\right) [f_+(E) - f_+(E')], \quad (5.1) \]

where \( E' = E + \hbar \omega \). In the parity restricted ensemble the thermal factor is replaced by

\[ f_+(E) - f_+(E') \to \frac{[f_+(E) - f_+(E')]Z_+ \pm [f_-(E) - f_-(E')]Z_-}{Z_+ \pm Z_-}. \quad (5.2) \]

As a result, we obtain for the ratio of the rates in the superconducting and the normal phase

\[ \left( \frac{\alpha_S(T)}{\alpha_N(T)} \right)_{e/o} = 2f(\Delta) - \frac{2}{1 \pm \prod_{k,\sigma} \coth \frac{\beta E_k}{2}} \frac{1}{\sinh \beta \Delta}. \quad (5.3) \]

In the low temperature limit, for the even case the ratio drops faster than in the BCS case

\[ \left( \frac{\alpha_S(T)}{\alpha_N(T)} \right)_e = 2N_{eff}e^{-2\beta \Delta}. \quad (5.4) \]

This should be contrasted with the \( \exp(-\beta \Delta) \) dependence of the BCS ratio. The odd case also shows a deviation

\[ \left( \frac{\alpha_S(T)}{\alpha_N(T)} \right)_o = 2e^{-\beta \Delta} + \frac{2}{N_{eff}}. \quad (5.5) \]

Both terms are of the same order of magnitude at \( T^* \), the onset temperature of even-odd effects. As we will see later on the low temperature response functions to other external probes, like the NSR relaxation rate and electromagnetic absorption, will have similar behavior as the ultrasound attenuation calculated above.

The BCS result for the nuclear spin relaxation rate is

\[ \frac{R_S(T)}{R_N(T)} = 2 \int_{-\Delta}^{\infty} \frac{[EE' + \Delta^2]f_+(E)(1 - f_+(E'))}{[E^2 - \Delta^2]^{1/2}[(E')^2 - \Delta^2]^{1/2}}. \quad (5.6) \]

Within the even/odd ensemble, the thermal factor is modified to

\[ f_+(E)(1 - f_+(E')) \to \frac{f_+(E)Z_+ \pm f_-(E)Z_-}{Z_+ \pm Z_-} - \frac{f_+(E)f_+(E')Z_+ \pm f_-(E)f_-(E')Z_-}{Z_+ \pm Z_-}. \quad (5.7) \]

An explicit estimation shows that only the first term will contribute significantly, and the second can be neglected. Denoting with \( F_{e/o}(E) \) the low temperature, \( E \to E' \) limit of the new parity dependent thermal factor, we obtain

\[ F_e(E) = N_{eff}e^{-\beta(E+\Delta)}, \quad (5.8) \]

\[ F_o(E) = e^{-\beta E} + \frac{e^{-\beta(E-\Delta)}}{N_{eff}}. \quad (5.9) \]
Using this function, the ratio of the relaxation rates can be written as
\[
\left( \frac{R_S(T)}{R_N(T)} \right)_{e/o} = 2F_{e/o}(\Delta) \int_{\Delta}^{\infty} \frac{[E^2 + \Delta^2]e^{-\beta(E-\Delta)}}{[E^2 - \Delta^2]} \, ,
\] (5.10)
where the integral now is only a weak function of temperature. All the important temperature and parity dependence is embodied in \( F_{e/o}(\Delta) \). Also note that \( F_{e/o}(\Delta) \) gives the low temperature behavior of the acoustic attenuation as well, since \( \frac{\alpha_S}{\alpha_N}_{e/o} \simeq 2F_{e/o}(\Delta) \).

Finally, let us discuss the infrared absorption. At very low temperatures there are only a few thermally excited quasiparticles available, and therefore substantial contribution to the absorption rate is given by the pair breaking process in which an incoming photon breaks a Cooper pair and creates two quasiparticles. In the case of the odd-parity system, however, there is always an unpaired quasiparticle present, which will lead to nonzero subgap absorption. Let us therefore consider the case when \( \hbar \omega < 2\Delta \), so that only single particle processes are allowed. We examine the real part of the conductivity - proportional to the rate of absorption - which has the following BCS form:

\[
\frac{\sigma_{1S}(T)}{\sigma_{1N}(T)} = 2 \left[ f_+ (\Delta) + \Delta^2 \int_{\Delta}^{\infty} \frac{[f_+ (E) (1 - f_+ (E))]}{E^2 - \Delta^2} \right] , \text{ for } \hbar \omega < 2\Delta.
\] (5.11)

A calculation similar to that presented above for the nuclear spin relaxation gives

\[
\frac{\sigma_{1S}(T)}{\sigma_{1N}(T)} = 2F_{e/o}(\Delta) \left[ 1 + \Delta^2 \int_{\Delta}^{\infty} \frac{e^{-\beta(E-\Delta)}}{E^2 - \Delta^2} \right] , \text{ for } \hbar \omega < 2\Delta.
\] (5.12)

The common feature of all these results obtained for the response of the parity-restricted superconductor to various probes is, that for low temperatures and energy transfers the \( \exp(-\beta \Delta) \) dependence - characteristic of a BCS superconductor - is replaced by a more rapidly decaying \( \exp(-2\beta \Delta) \) behavior for the \textit{even} case, and the appearance of a very small \( (\mathcal{O}(1/N_{eff})) \), but nonzero background due to a \textit{single}, unpaired quasiparticle, that can scatter, absorb, spin relax etc. Pictorially, the \( \exp(-2\beta \Delta) \) dependence comes from the exclusion of the parity-violating grand-canonical processes that involve a single particle exchange between the system and a particle bath (excitation energy \( \Delta \)), as well as from the important contribution of the momentum-conserving excitation process of a pair (excitation energy \( 2\Delta \)), in contrast to pair breaking with no momentum conservation (excitation energy \( \Delta \) per particle).

\[\text{VI. CONCLUSIONS}\]

In conclusion, we have presented a theoretical method to investigate number parity effects in the superconducting state by using a grand canonical ensemble in which the parity of the system is even or odd. This corresponds to a permeable wall between the superconductor and the particle bath that allows particle exchange only in pairs. We obtained the partition function which in turn yielded the grand potential, entropy and specific heat of the system. While the variational equations from the minimization of the grand potential gave essentially BCS results, the free energy difference between even and odd parity states obtained is in accordance with the experimental and theoretical findings of Tuominen et al. and the
measurements of Lafarge et al. The specific heat shows a more rapid \( \exp(-2\beta \Delta) \) decay at low temperatures; the same quantity in the odd-parity system has a finite zero temperature limit \( k_B/2 \), in contrast to the usual BCS result.

A simple and systematic low temperature approximation was made possible by the fact that for the temperature range and size of the islands investigated so far, \( N_{\text{eff}} \gg 1 \) and therefore an expansion was possible in powers of \( 1/N_{\text{eff}} \). In principle, this expansion breaks down at extremely low temperatures, since \( 1/N_{\text{eff}} \propto T^{-1/2} \). In order to observe such effects in the temperature range presently investigated (\( T \geq 50 \, \text{mK} \)), the volume of the island must be four orders of magnitude less, about \( 10^{-19} \, \text{cm}^3 \), which might be very difficult to achieve experimentally.

We have not addressed the problem of the magnetic field dependence of the even-odd free energy difference. The measurements of Tuominen et al. and Esteve et al. seem to suggest that the experimental results can be explained if one replaces in eq. (4.2) the gap in zero field \( \Delta \) with the gap edge \( \Delta(H) \) of the field dependent density of states provided by the Abrikosov - Gorkov theory. We believe that this replacement, although possibly correct, is not justified well enough. The present method cannot be easily generalized to the Green’s function technique, due to the lack of a Wick’s theorem: for the parity-conserving average defined in eq. (2.1), \( \langle \hat{n}_k \hat{n}_{k'} \rangle \neq \langle \hat{n}_k \rangle \langle \hat{n}_{k'} \rangle \) for a system of noninteracting fermions, where \( \hat{n}_k \) is the number operator (cf. Eq.(3.13)). There is, however, a Wick’s theorem for the two channels, with projection operator \( \frac{1}{2} \) and \( \pm (-1)^N/2 \), respectively, which add up to the parity-restricted average. Because of this, the normal-state properties and the Cooper instability need further investigation, to be performed in the future.

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