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Parity effect in mesoscopic and nanoscopic superconducting particles

I.O. Kulik *, H. Boyaci, Z. Gedik

Department of Physics, Bilkent University, Bilkent 06533, Ankara, Turkey

Abstract

Superconductivity in small metallic specimens is studied with regard to the size dependence of the *parity gap* (Δ_P), a parameter distinguishing between the energy of even and odd number of electrons in the granule. Δ_P is shown to be an *increasing* function of level spacing δ . The energy gap of superconductor Δ , on the other hand, *decreases* with increasing δ and vanishes at $\delta = \delta_c$ which is of the order of Δ . However, non-zero value of Δ_P persists above δ_c in a *gapless* superconducting–insulating state. Level degeneracy in small specimens having perfect geometry changes the size dependence of the parity gap, the Josephson effect, and flux quantization. Parity gap is evaluated using an interpolation procedure between the continuum limit ($\delta \ll \Delta$), the moderate mesoscopic regime ($\delta \sim \Delta$), and the nanoscopic scale ($\delta \gg \Delta$), for which an exact solution to the pairing problem is provided with the numeric diagonalization of system Hamiltonian in a small metallic cluster. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Superconductivity in metals is related to condensation of electrons into pairs which are extended objects with a size of order $\xi \sim \hbar v_F / \Delta_0$. A fundamental question [1] is whether pairing can survive in small specimens with size d less than ξ . In energy scale this corresponds to condition that $\delta_1 > \Delta_0$ where $\delta_1 = \hbar v_F / d$ is typical scale of dimensional energy quantization. The energy levels can remain highly degenerate in a perfect sample (see Fig. 1) but in a disordered sample, level spacing $\delta_2 \sim \epsilon_F / N_s$, where N_s is the total number of

sites (which is of the order of total number of electrons, N), is much smaller than δ_1 and can put a limit to superconducting pairing at $\delta_2 \sim \Delta_0$. In recent experiments [2,3], signature of superconducting pairing was traced in ultrasmall specimens down to sizes of 10 nm. The superconductivity reveals itself through the number-parity effect, the dependence of superconducting energy on whether the number of electrons in sample is even or odd. Matveev and Larkin [4] introduced a *parity gap* $\Delta_P = (1/2)(E_{2n+1} + E_{2n-1}) - E_{2n}$ where E_n is superconducting energy vs number of electrons n , to characterize that the paired state is preferable over the state with odd number of electrons. In a number of papers, in particular in the paper of Braun and von Delft [5], E_n was calculated assuming that in an odd-number sample, electron

* Corresponding author. Fax: +90-312-2664579.

E-mail address: kulik@fen.bilkent.edu.tr (I.O. Kulik).

Fig. 1. Degeneracy of energy levels for parabolic energy vs momentum dependence of total energy $E_n = Cn^2 = C(n_1^2 + n_2^2 + n_3^2)$.

state near the Fermi energy is blocked from pairing, similar to the way in which finite-number systems are treated in nuclear physics [6]. In present paper, we investigate the suppression of superconductivity in a finite size system by using an approximation similar to that of [5], and also in the opposite limit, by starting from atomic-scale system (small atomic cluster) for which an exact solution of pairing problem is provided, and trying to match small- and large-size approximations through an interpolation procedure.

2. Large-scale approximation

In the Bardeen–Cooper–Schrieffer (BCS) approximation [7] a variational density matrix $\rho_{\alpha\beta} = (1/Z)\phi_{\alpha}^* \exp(-(H - \mu N)/kT)\phi_{\beta}$ is exploited to obtain the ground state energy with trial wave functions

$$\begin{aligned}\phi_{\alpha}^{(1)} &= \prod_{\mathbf{k}} (u_{\mathbf{k}} + v_{\mathbf{k}} a_{\mathbf{k}\uparrow}^{\dagger} a_{-\mathbf{k}\downarrow}^{\dagger}) |0\rangle, \\ \phi_{\alpha}^{(2)} &= \prod_{\mathbf{k}\mathbf{k}_F} (u_{\mathbf{k}} + v_{\mathbf{k}} a_{\mathbf{k}\uparrow}^{\dagger} a_{-\mathbf{k}\downarrow}^{\dagger}) a_{\mathbf{k}_F}^{\dagger} |0\rangle\end{aligned}\quad (1)$$

for even and odd number of electrons, respectively. In the BCS pairing scheme (case 1), assuming

equally spaced levels with distance δ , we receive $\Delta_{2m} = \Delta_{2m+1} = \Delta_0$ whereas in the blocked pairing state (case 2) $\Delta_{2m} = \Delta_0$ and $\Delta_{2m+1} = \Delta_0 - \delta/2$. The energy of superconductor at $T = 0$ is calculated as $\langle H - \mu N \rangle = \sum_n (\xi_n - (\Delta^2 + \xi_n^2)^{1/2})$ resulting in a parity gap value $\Delta_P^{(1)} = \delta/4$ (for $\delta \ll \Delta_0$) in the first case, and $\Delta_P^{(2)} \simeq \Delta_0 \ln(\omega_D \delta / \Delta_0^2) + (1/2)\delta$ in the second. Energy of the blocked state is larger than that of the BCS state and therefore there is no reason to believe that the former is a better approximation for pairing. There is no direct relation of the above calculation to the question that there is a large uncertainty in the number of particles, and assertion that number of particles in $\phi_{\alpha}^{(2)}$ is odd is merely an illusion.

3. Josephson effect in mesoscopic metal particles

Consider two identical small superconducting grains interacting through the Hamiltonian $H_T = \sum_{nm} T_{nm} a_n^{\dagger} b_m + \text{h.c.}$ where $a_n^{\dagger} (a_n)$ creates (annihilates) an electron in the first grain in state n . Similarly $b_n^{\dagger}$ and b_n are operators for the second grain. By introducing the phases ϕ_1 and ϕ_2 to the Bogoliubov u, v -coefficients in Eq. (1), we calculate coupling energy due to H_T as a function of ϕ_1 and ϕ_2 in a standard way [7] $\Delta E = \Delta E'_S - \Delta E_S \cos(\phi_1 - \phi_2)$ where

$$\Delta E_S = \langle |T|^2 \rangle \Delta^2 \sum_{|\xi_n|, |\xi_m| < \omega_D} \frac{1}{(\varepsilon_n + \varepsilon_m) \varepsilon_n \varepsilon_m}. \quad (2)$$

The stationary Josephson current at zero temperature is $J_c \sin(\phi_1 - \phi_2)$ where $J_c = 2e\Delta E_S/\hbar$. Fig. 2 shows dependence of Josephson critical current and energy gap as a function of gap suppression parameter δ/Δ_0 . The curves show that superconductivity is suppressed at level spacing $\delta_c = 3.56\Delta_0$. This however is a conclusion within the mean-field approximation. It does not exclude the possibility that pairing may survive for $\delta > \delta_c$ as a *gapless* superconducting state or as a state of *superconducting insulator* with an effective insulator gap δ larger than the superconducting pseudogap.

Fig. 2. Josephson critical current vs energy level spacing δ (—) and gap parameter Δ vs δ (---). The latter has been scaled by a factor of 1/2 for convenience.

4. Richardson–Sherman approximation

A finite size solution to the pairing problem was suggested by Richardson and Sherman [8] (and recently revived by von Delft and Braun [9]) assuming that states with energy within the interac-

tion shell $-\omega_D < \xi_n < \omega_D$ can be approximated as hard-core bosons whereas states away from this shell are treated exactly as non-interacting particles. The interaction Hamiltonian for hard-core bosons is

$$H = 2\delta \sum_{nz} n c_{nz}^\dagger c_{nz} - \frac{V\delta}{d} \sum_{|\xi_n|, |\xi_m| < \omega_D, \alpha\beta} c_{nz}^\dagger c_{m\beta} \quad (3)$$

where boson operators are $c_{nz} = a_{n\bar{\alpha}\downarrow} a_{nz\uparrow}$, d is level degeneracy, and α labels states within the degenerate energy level $\xi_n = n\delta$. Hamiltonian (3) is solved [10] by iteration scheme proposed by Richardson and Sherman [8]. Dependence of Δ_P on δ/Δ_0 is shown in Fig. 3. In non-degenerate case ($d = 1$) $\Delta_P(\delta)$ shows a behavior similar to previous results with smaller number of levels [11,12]. In case of degenerate energy levels, pairing gap is multi-valued depending upon which three successive states are used in calculation of Δ_P . The *true pairing gap* is possibly the largest one in Fig. 3(b), and others represent a new kind of mesoscopic effect which may or may not be related to superconductivity. Small δ of $\Delta_P(\delta)$ does not match with any of pairing gaps calculated in Section 3, which means possibly that neither of the approximations

Fig. 3. Parity gap vs level spacing for non-degenerate case (a) and for double degenerate levels (b). In (a), numbers 1–4 label dependences received with a number of levels in the interaction shell $n_D = 30, 60, 120$, and 360. In (b), numbers 1–4 correspond to index i in various pairing gaps defined according to $\Delta_P^{(i)} = 1/2(E_{4m+i} + E_{4m+i-2}) - E_{4m+i-1}$.

Fig. 4. (a) Energy vs number of electrons in cubic cluster with attractive interaction at sites. (1) $U/t = 2$, (2) $U/t = 3$, (3) $U/t = 4$. (b) Parity gap vs level spacing in cubic cluster.

(original BCS or BCS with one level blocked) are good for small systems.

5. Parity effect in atomic clusters

We calculate the exact eigenstates of the negative- U Hubbard Hamiltonian

$$H = -t \sum_{\langle i,j \rangle, \sigma} a_{i\sigma}^\dagger a_{j\sigma} - U \sum_{i=1}^N a_{i\uparrow}^\dagger a_{i\downarrow}^\dagger a_{i\downarrow} a_{i\uparrow} + \sum_{i\sigma} V_i a_{i\sigma}^\dagger a_{i\sigma} \quad (4)$$

(with $U > 0$) in small system of N sites. V_i is random potential at site i . The fact that Hamiltonian (4) commutes with the total number of electrons and also with the total spin leads to lowering of dimensionality $2^{2N} \times 2^{2N}$ of the matrix. Interaction is non-retarded (unlike BCS Hamiltonian) and momentum dependence of energy is non-parabolic. We do not believe however that these are crucial differences, and indeed the small- U limit mean-field solution [13] corresponds to an energy gap $\Delta_0 \simeq 12t \exp(-12t/U)$. Fig. 4a shows minimal energy E_N of cubic cluster as a function of number of electrons N . Parity effect clearly, manifests itself in lower E_N values for even N and,

when plotting Δ_P against δ/Δ_0 (Fig. 4b), displays a behavior very similar to one received in the Richardson–Sherman model.

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