Interplay between particle-hole splitting and the Kondo effect in quantum dots

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We discuss the influence of proximity effect on the quantum dot located between metallic and superconducting electrodes and consider its interplay with the Kondo state arising from the on-dot correlations. We show that upon increasing the hybridization to superconducting lead the Kondo resonance is gradually washed out. This behavior indirectly affects the subgap charge transport suppressing the zero-bias enhancement of the Andreev conductance.

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I. MOTIVATION

The issue of quantum impurity immersed in a continuum of fermion states has a fundamental importance for the solid-state[1] and nanoscopic physics.[2] Such problem is particularly interesting for a discrete state embedded near the Fermi energy of an isotropic superconductor where the low-energy single-particle states are not allowed to exist. One may then ask whether impurity is going to be inactive or it would interfere with itinerant fermions and eventually build up the singlet Kondo state.

We address here a similar question investigating impurity located between the metallic and superconducting leads. The recent rapid development of nanotechnology enables controllable manipulation of the hybridization between such "quantum dot" (QD) and the external reservoirs of itinerant fermions. On practical side, one can measure the nonequilibrium charge current applying a source-drain voltage V across the junction. Since the low-bias (subgap) current is transmitted solely through the Andreev reflections[3,4] [which are sensitive both to the effective electronic structure and to the superconducting order induced in QD (Refs. 5–9)] the Andreev spectroscopy might be a useful tool for exploring the interplay between superconductivity and the Kondo effect in quantum dots.

In this report we briefly recollect signatures of the particle-hole splitting showing up in a subgap Andreev current due to the proximity effect. We furthermore analyze under what conditions there can appear the low-temperature enhancement of zero-bias Andreev conductance driven by the Kondo effect. For this purpose we focus on the limit of large energy gap $\Delta \to \infty$ and treat approximately the on-dot correlations. In particular, we try to clarify why some authors have predicted existence (although weak) of the zero-bias Kondo anomaly[7] while the other ones reported its absence[10] apparently due to specific choice of the model parameters. We hope that a controversial issue of the zero-bias Andreev feature could be soon resolved experimentally.

II. PARTICLE-HOLE SPLITTING

Strongly correlated quantum dot hybridized with the normal (N) and superconducting (S) electrodes can be described by the single impurity Anderson model

$$\hat{H} = \hat{H}_N + \hat{H}_S + \sum_{\sigma} \epsilon_d \hat{d}^{\dagger}_{\sigma} \hat{d}_{\sigma} + U \hat{n}_{d\uparrow} \hat{n}_{d\downarrow} + \sum_{\mathbf{k},\sigma} \sum_{\beta=N,S} (V_{\mathbf{k}\beta} \hat{d}^{\dagger}_{\sigma} \hat{c}_{\mathbf{k}\sigma\beta} + V^{*}_{\mathbf{k}\beta} \hat{c}^{\dagger}_{\mathbf{k}\sigma,\beta} \hat{d}_{\sigma}). \tag{1}$$

Operators d_σ ($d^{\dagger}_{\sigma}$) denote annihilation (creation) of QD electron with spin σ and energy level ε_d, U is the on-dot repulsion (or charging) energy between opposite spin electrons, and the last term describes hybridization of the QD with external leads. We represent the normal electrode by $\hat{H}_N = \Sigma_{\mathbf{k},\sigma} \xi_{\mathbf{k}N} \hat{c}^{\dagger}_{\mathbf{k}\sigma N} \hat{c}_{\mathbf{k}\sigma N}$ while superconducting lead is described by the BCS Hamiltonian $\hat{H}_S = \Sigma_{\mathbf{k},\sigma} \xi_{\mathbf{k}S} \hat{c}^{\dagger}_{\mathbf{k}\sigma S} \hat{c}_{\mathbf{k}\sigma S} - \Sigma_{\mathbf{k}} \Delta (\hat{c}^{\dagger}_{\mathbf{k}\uparrow S} \hat{c}^{\dagger}_{-\mathbf{k}\downarrow S} + \hat{c}_{-\mathbf{k}\downarrow S} \hat{c}_{\mathbf{k}\uparrow S})$. Energies $\xi_{\mathbf{k}\beta} = \varepsilon_{\mathbf{k}\beta} - \mu_\beta$ are measured with respect to the chemical potentials which can be detuned by external voltage $\mu_N = \mu_S + eV$. We shall restrict to the wide band limit $|V_{\mathbf{k}\beta}| \ll D$, where $-D \leq \varepsilon_{\mathbf{k}\beta} \leq D$.

For description of the proximity-effect spread on the QD from superconducting lead it is convenient to introduce the retarded Green's function $\boldsymbol{G}(\tau) = \langle\langle \hat{\Psi}(\tau); \hat{\Psi}^{\dagger} \rangle\rangle$ using the Nambu spinor notation $\hat{\Psi}^{\dagger} = (\hat{d}^{\dagger}_{\uparrow}, \hat{d}_{\downarrow})$, $\hat{\Psi} = (\hat{\Psi}^{\dagger})^{\dagger}$. Its Fourier transform can be written as

$$\boldsymbol{G}(\omega)^{-1} = \begin{pmatrix} \omega - \varepsilon_d & 0 \\ 0 & \omega + \varepsilon_d \end{pmatrix} - \boldsymbol{\Sigma}^{0}_{d}(\omega) - \boldsymbol{\Sigma}^{U}_{d}(\omega), \tag{2}$$

where $\boldsymbol{\Sigma}^{0}_{d}$ denotes the self-energy of noninteracting ($U=0$) case and the last part Σ^{U}_{d} accounts for the correlation effects. The effect of $\boldsymbol{\Sigma}^{0}_{d}$ (known exactly) has been so far discussed by a number of authors.[5–14] Below we briefly recall the features of particle-hole mixing observable in the spectral function and in subgap Andreev conductance.

For simplicity we shall consider the QD whose energy level ε_d is located deep inside the gap of superconductor focusing on energies $|\omega| \ll \Delta$. The self-energy $\boldsymbol{\Sigma}^{0}_{d}$ becomes then a static (ω independent) quantity[7,12,13]

$$\boldsymbol{\Sigma}^{0}_{d}(\omega) = -\frac{1}{2} \begin{pmatrix} i\Gamma_N & \Gamma_S \\ \Gamma_S & i\Gamma_N \end{pmatrix}, \tag{3}$$

where $\Gamma_\beta = 2\pi \Sigma_{\mathbf{k}} |V_{\mathbf{k}\beta}|^2$. Under such circumstances QD can be treated as a superconducting grain characterized by a nonvanishing off-diagonal order parameter $\langle \hat{d}_{\downarrow} \hat{d}_{\uparrow} \rangle$. The spectral function $\rho_d(\omega) \equiv -\frac{1}{\pi} \mathrm{Im}\, \boldsymbol{G}_{11}(\omega + i0^{+})$ acquires then effectively BCS-type structure

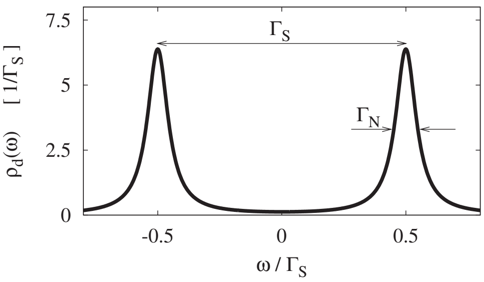

FIG. 1. Spectral function $\rho_d(\omega)$ of the uncorrelated ($U=0$) quantum dot obtained for $\Gamma_S=10\Gamma_N$ and the energy level located in a center of superconductor's gap $\varepsilon_d=0$ assuming $\Delta\to\infty$.

$$\rho_d(\omega) = \frac{1}{2}\left[1+\frac{\varepsilon_d}{E_d}\right]\frac{\frac{1}{\pi}\Gamma_N/2}{(\omega-E_d)^2+(\Gamma_N/2)^2} + \frac{1}{2}\left[1-\frac{\varepsilon_d}{E_d}\right]\frac{\frac{1}{\pi}\Gamma_N/2}{(\omega+E_d)^2+(\Gamma_N/2)^2}, \quad (4)$$

with $E_d=\sqrt{\varepsilon_d^2+\Delta_d^2}$ and $\Delta_d=\Gamma_S/2$. The spectrum of noninteracting QD consists thus of the particle and hole peaks at $\omega=\pm E_d$ whose spectral weights depend on ε_d and broadening is controlled by the coupling Γ_N to metallic lead. In Fig. 1 we illustrate the spectral function for $\varepsilon_d=0$ and $\Gamma_S=10\Gamma_N$ when the particle-hole (p-h) peaks are well separated. Otherwise, for $|\varepsilon_d|\geq\Gamma_S$ the p-h features are less pronounced and for $\Gamma_S\leq\Gamma_N$ they practically merge into the single Lorentzian.

Consequences of the proximity effect responsible for the p-h splitting can be detected in the tunneling current. At low bias $|eV|<\Delta$ (of the order of millivolts) the charge current $I(V)=\frac{e}{2}\Sigma_{\mathbf{k},\sigma}\frac{d}{dt}\langle\hat{c}^\dagger_{\mathbf{k}\sigma N}\hat{c}_{\mathbf{k}\sigma N}-\hat{c}^\dagger_{\mathbf{k}\sigma S}\hat{c}_{\mathbf{k}\sigma S}\rangle$ is transferred only through the anomalous Andreev channel.[4] Using the Keldysh formalism one can express the Andreev current by the following Landauer-type formula[7,8]

$$I_A(V) = \frac{2e}{h}\int d\omega T_A(\omega)[f(\omega+eV,T)-f(\omega-eV,T)], \quad (5)$$

with $f(\omega,T)=[1+\exp(\omega/T)]^{-1}$ and the corresponding transmittance $T_A(\omega)$ determined by the off-diagonal part of the Green's function $T_A(\omega)=\Gamma_N^2|\boldsymbol{G}_{12}(\omega)|^2$.[8] Andreev transmittance of the uncorrelated QD simplifies in the limit $\Delta\to\infty$ to

$$T_A(\omega) = \frac{\Gamma_N^2(\Gamma_S/2)^2}{[(\omega-E_d)^2+(\Gamma_N/2)^2][(\omega+E_d)^2+(\Gamma_N/2)^2]}, \quad (6)$$

hence at low temperatures the differential conductance $G_A(V)=\frac{d}{dV}I_A(V)$ should exhibit the p-h features. Figure 2 shows that for $\Gamma_S\geq\Gamma_N$ the differential Andreev conductance indeed achieves the optimal value $4e^2/h$ (Ref. 4) near the quasiparticle energies $eV=\pm E_d$.

Expression for the corresponding zero-bias conductance $G_A(0)=\frac{4e^2}{h}(\frac{2\Gamma_N\Gamma_S}{4E_d^2+\Gamma_N^2})^2$ is at low temperatures identical with Eq. (9) obtained by Beenakker[4] from the Breit-Wigner formula. For $\varepsilon_d=0$ this reproduces the well-known result $G_A(0)=\frac{4e^2}{h}(\frac{2\Gamma_N\Gamma_S}{\Gamma_S^2+\Gamma_N^2})^2$ (Ref. 12) being invariant under replacement $\Gamma_N\leftrightarrow\Gamma_S$. Let us remark that the zero-bias conductance diminishes upon increasing asymmetry between the couplings Γ_β and is further suppressed whenever QD level ε_d moves away from the center of energy gap. From these introductory considerations we conclude that in order to observe any particular features at low voltages one must restrict to nearly the symmetric case $\Gamma_S\sim\Gamma_N$.

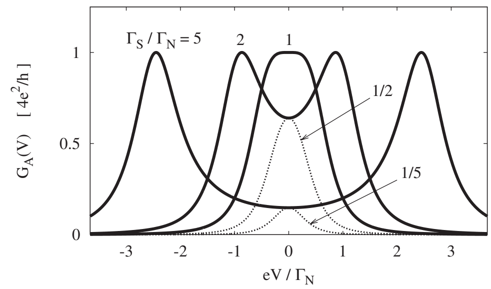

FIG. 2. The subgap Andreev conductance $G_A(V)$ obtained at several values of the ratio Γ_S/Γ_N for the uncorrelated QD $U=0$ with $\varepsilon_d=0$, $\Gamma_N=0.03D$. Notice that the zero-bias conductance $G_A(0)$ is invariant under replacement $\Gamma_S\leftrightarrow\Gamma_N$ and it is optimal for $\Gamma_S\sim\Gamma_N$.

III. INTERPLAY WITH THE KONDO EFFECT

In the limit $\Delta\to\infty$ the self-energy $\boldsymbol{\Sigma}_d^0(\omega)$ becomes a static quantity, therefore Oguri *et al.*[15] remarked that the effect of superconducting lead could be exactly modeled by the auxiliary *superconducting* QD described by

$$\hat{H} = \sum_\sigma \epsilon_d\hat{d}^\dagger_\sigma\hat{d}_\sigma + (\Delta_d\hat{d}^\dagger_\uparrow\hat{d}^\dagger_\downarrow + \mathrm{H.c.}) + U\hat{n}_{d\uparrow}\hat{n}_{d\downarrow} + \sum_{\mathbf{k},\sigma}(V_{\mathbf{k}N}\hat{d}^\dagger_\sigma\hat{c}_{\mathbf{k}\sigma\beta} + V^*_{\mathbf{k}\beta}\hat{c}^\dagger_{\mathbf{k}\sigma,\beta}\hat{d}_\sigma) + \hat{H}_N. \quad (7)$$

Let us emphasize that Eq. (7) is completely equivalent to Eq. (1) irrespective of the Coulomb interaction U. In other words, no approximations have been imposed so far on correlations.

A clear virtue of the auxiliary Hamiltonian (7) can be seen analyzing the on-dot Coulomb repulsion $U\hat{n}_{d\uparrow}\hat{n}_{d\downarrow}=\frac{U}{2}(\hat{n}_d-1)^2+\frac{U}{2}(\hat{n}_d-1)$. The operator $(\hat{n}_d-1)^2$ turns out to be invariant under the Bogoliubov-Valatin transformation (see Appen-

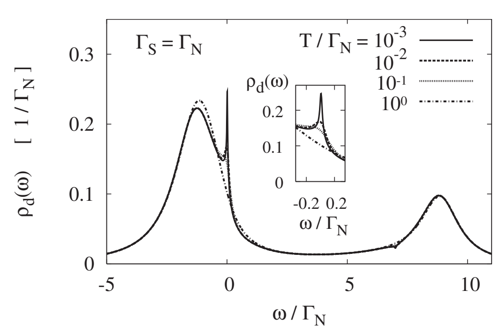

FIG. 3. Temperature dependence of the spectral function $\rho_d(\omega)$ of the correlated QD ($U=10\ \Gamma_N$) in the equilibrium situation ($V=0$) for $\varepsilon_d=-1.5\Gamma_N$, $\Gamma_S=\Gamma_N$. Notice formation of the Kondo resonance at $\omega=0$ whereas the p-h splitting (not visible here) appears for stronger hybridizations Γ_S (see Fig. 4). Kondo temperature for the present case is $T_K=\frac{\sqrt{U\Gamma_N}}{2}e^{\pi\varepsilon_d(\varepsilon_d+U)/U\Gamma_N}\simeq0.029\Gamma_N$.

dix B in Ref. 12), therefore the term Σ_d^U accounting for the correlations must have a diagonal structure. With this rigorous constraint in mind we shall adopt some approximation because exact solutions are not available at this level (besides the one-dimensional case).

Influence of the Coulomb interactions has been previously studied directly within the Hamiltonian (1) using various treatments such as the slave boson approach,[7,11] generalized noncrossing approximation,[10] perturbative scheme,[16] and the numerical renormalization group.[17] Following the lines[12,15] we revisit here the issue of interplay between the correlations and superconductivity using the auxiliary model (7) for which the exact matrix Green's function (2) can be written as

$$\boldsymbol{G}(\omega) = \begin{pmatrix} \omega - \varepsilon_d - \Sigma_N(\omega) & \frac{1}{2}\Gamma_S \\ \frac{1}{2}\Gamma_S & \omega + \varepsilon_d + \Sigma_N^*(-\omega) \end{pmatrix}^{-1}. \quad (8)$$

We estimate the value $\Sigma_N(\omega)$ using the equation of motion method[18]

$$\Sigma_N(\omega) = \omega - \varepsilon_d - \frac{[\omega - \varepsilon_d - \Sigma_0(\omega)]\{\omega - \varepsilon_d - [U + \Sigma_0(\omega) + \Sigma_3(\omega)]\} + U\Sigma_1(\omega)}{\omega - \varepsilon_d - [\Sigma_0(\omega) + \Sigma_3(\omega) + U(1 - n_{d,\sigma})]}, \quad (9)$$

where $\Sigma_0(\omega) = -\frac{i}{2}\Gamma_N$ and $\Sigma_{1,3}(\omega)$ are given by[18]

$$\Sigma_\nu(\omega) = \sum_{\mathbf{k}} |V_{\mathbf{k}N}|^2 \left(\frac{1}{\omega - \xi_{\mathbf{k}N}} + \frac{1}{\omega - U - 2\varepsilon_d + \xi_{\mathbf{k}N}} \right) \times [f(\omega,T)]^{\frac{3-\nu}{2}}. \quad (10)$$

This approach (9) qualitatively captures the following important aspects of Coulomb interactions: (i) the charging effect and (ii) appearance of the Kondo resonance at $\omega=0$ for low temperatures, smaller than T_K (see Fig. 3). In nonequilibrium conditions the Kondo resonance shifts to $\omega = \mu_N$ and this behavior differs from N-QD-N setup where there are two Kondo peaks at both chemical potentials μ_β.

Let us stress that the Kondo peak comes from a hybridization of the QD with the metallic lead. By increasing the coupling Γ_S to the superconducting lead we observe a gradual emergence of the particle-hole features simultaneously accompanied by a suppression of the Kondo resonance. Such competition between the p-h splitting and the Kondo effect is illustrated in Fig. 4 for a set of relative couplings Γ_S/Γ_N. To our knowledge a systematic study of this competition has not been discussed in the literature.

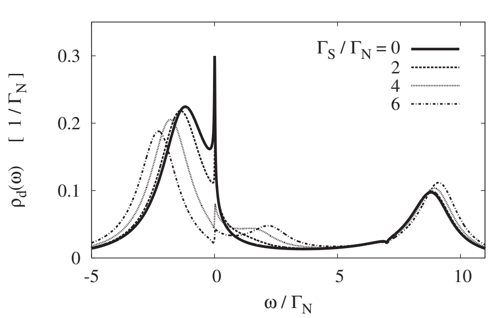

FIG. 4. Spectral function $\rho_d(\omega)$ of the correlated QD in the equilibrium situation ($V=0$) obtained for $\varepsilon_d = -1.5\Gamma_N$, $U=10\Gamma_N$, $T=0.001\ \Gamma_N$, and several representative values of the ratio Γ_S/Γ_N.

Since the differential Andreev conductance $G_A(V)$ is very sensitive to the available low-energy states of QD it does show up a characteristic zero-bias enhancement (see Fig. 5) arising from the Kondo peak. Besides this low-temperature zero-bias feature we notice also the quasiparticle peak at $|eV| = E_d$ and another weaker one at $|eV| = U$ coming from the upper Coulomb satellite in the spectral function.

Optimal conditions for observing the zero-bias anomaly occur when $\Gamma_S \sim \Gamma_N$ and QD energy level is being located close to the center of superconductor energy gap. With increase of the ratio Γ_S/Γ_N the particle-hole splitting forces the QD into the mixed-valence regime (see Fig. 4) and thereby the zero-bias enhancement gradually fades away as illustrated in Fig. 6. Such stringent conditions needed for the zero-bias anomaly of the Andreev current might explain why some authors have missed it by choosing inappropriate parameters.[10]

So far the experimental data indicated existence of the zero-bias enhancement in the Josephson current observed for S-QD-S junctions.[19] We hope that our study might be instructive for the experimentalists to find similar effect in case of

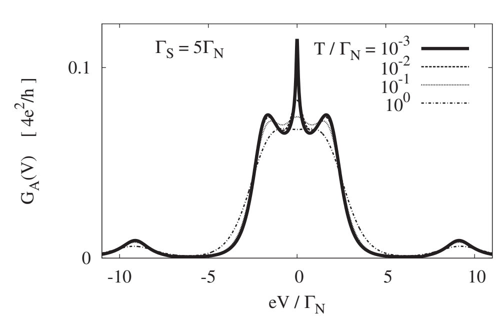

FIG. 5. Temperature evolution of the differential Andreev conductance $G_A(V)$ for the correlated QD with $U=10\ \Gamma_N$, $\varepsilon_d = -1.5\ \Gamma_N$, and $\Gamma_S = 5\ \Gamma_N$. This situation is outside the optimal conditions (corresponding to $\Gamma_S \sim \Gamma_N$) but it exemplifies the following features: (i) the zero-bias enhancement, (ii) the particle-hole peaks, and (iii) additional peaks at $|eV| \sim U$ due to the charging effect.

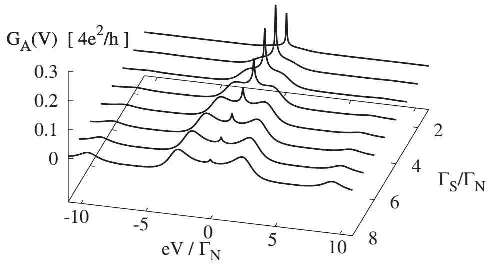

FIG. 6. The low-temperature (T=0.001 Γ_N) Andreev conductance as a function of external bias V/Γ_N and the asymmetry ratio Γ_S/Γ_N for the same set of parameters as used in Fig. 4.

the quantum dot located between the normal and superconducting leads.

IV. SUMMARY

We have investigated the quantum dot between the metallic and superconducting electrodes. Due to proximity effect the QD spectrum shows up the particle and hole peaks which split for $\Gamma_S \geq \Gamma_N$ if the energy level ε_d is located near the center of energy gap. These BCS-type features can be probed by the subgap Andreev current whose optimal conductance $4e^2/h$ is reached at $eV = \pm\sqrt{\varepsilon_d^2 + (\Gamma_S/2)^2}$. Practically such maxima could be manipulated experimentally by varying the coupling Γ_S with superconductor and adjusting the QD level ε_d by the gate potential.

The strong Coulomb correlations can further induce the Kondo resonance at $\omega = \mu_N$ owing to the QD hybridization with the normal lead. For $\Gamma_S \sim \Gamma_N$ and QD level located slightly below the center of the energy gap the Kondo resonance contributes an enhancement to the zero-bias Andreev conductance (although its magnitude is much smaller than the value $2e^2/h$ observed for the QD coupled with both metallic leads in the Kondo regime). For $\Gamma_S \geq \Gamma_N$ the p-h splitting drives the QD into the mixed-valence regime where the Kondo peak and the zero-bias anomaly are washed away. We hence conclude that in N-QD-S structures the particle-hole splitting related to the proximity effect competes with the singlet Kondo state in much the same way as superconductivity competes with magnetic ordering in the solid-state physics.

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