

Andreev bound states in the Kondo quantum dots coupled to superconducting leads

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Received 26 May 2008, in final form 4 August 2008

Published 22 September 2008

Online at stacks.iop.org/JPhysCM/20/415225

Abstract

We have studied the Kondo quantum dot coupled to two superconducting leads and investigated the subgap Andreev states using the NRG method. Contrary to the recent NCA results (Clerk and Ambegaokar 2000 *Phys. Rev. B* **61** 9109; Sellier *et al* 2005 *Phys. Rev. B* **72** 174502), we observe Andreev states both below and above the Fermi level.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

When a localized spin (in an impurity or a quantum dot) is coupled to BCS-type *s*-wave superconductors [1], two strong correlation effects compete with each other. On the one hand, the superconductivity tends to keep the conduction electrons in singlet pairs [1], leaving the local spin unscreened. The spin state of the total wavefunction is thus a doublet. On the other hand, the Kondo effect tends to screen the local spin with the spins of the quasi-particles in the superconductors at the expense of the quasi-particle excitation energies. The total spin state is then a singlet [2]. This competition gives rise to a quantum phase transition from the doublet to singlet state, and the transport properties change dramatically across this transition [3, 4]. For example, as demonstrated directly in a recent experiment [5], the Josephson current through a quantum dot coupled to two superconducting leads has a π -shift in its current–phase relation (a so-called π -junction behavior) for the doublet state, while its current–phase relation is similar to the usual one (a 0-junction behavior) for the singlet state [6–13]. For this reason, the doublet–singlet transition is also called the 0– π transition.

Previous studies of the transition between the doublet and singlet state all focused on the current–phase relation $I_S(\phi)$ of the Josephson current. However, there are other non-trivial issues to be addressed for a deeper understanding of the doublet–singlet transition in such a system, i.e. about the Andreev bound states: (1) how many subgap Andreev states are there and (2) are the subgap Andreev states true bound states or quasi-bound states with finite level broadening?

Using the non-crossing approximation (NCA), Clerk and Ambegaokar [8] investigated the close relation between the 0– π transition in $I_S(\phi)$ and the Andreev states. They found that there is only one subgap Andreev state and that the Andreev state is located below (above) the Fermi energy E_F in the doublet (singlet) state. They provided an intuitively appealing interpretation that, in the doublet state, the impurity level well below E_F is singly occupied and due to the strong on-site interaction energy U only hole-like excitations are allowed; and that in the singlet state, due to a small probability of finding the impurity empty, only electron-like excitations are allowed. This result was supported further by a more elaborate NCA method by Sellier *et al* [13]. As stressed by Clerk and Ambegaokar [8], this observation has a strong contrast with the non-interacting case, where bound states always occur in pairs (below and above E_F) [14, 15]. Moreover, NCA predicted that the Andreev state has a finite broadening.

In contrast, the Hartree–Fock approximation (HFA) [16] predicts two Andreev states, below and above E_F in the Kondo regime. This was in agreement with the numerical renormalization group (NRG) calculations by [17], who studied the Andreev states as a function of the impurity level position. Slave-boson mean-field approximation (SBMFA) [18] also predicts both electron-like (above E_F) and hole-like (below E_F) Andreev states. Further, they both predict infinitely sharp Andreev states. However, HFA and SBMFA are effectively non-interacting theories and may not be a strong argument against the NCA results. A perturbative approach beyond HFA predicted both Andreev states [19]. In a previous work [9], we also observed both Andreev states in the NRG

result. However, the model in these works had the particle–hole symmetry, and cannot rule out the NCA results either.

As shown above, despite its importance, the nature of the subgap Andreev states has remained controversial among different theoretical methods. In this work, we report a systematic study of the issues using the NRG method. Contrary to the NCA results, we find both electron-like and hole-like Andreev states (section 2), except in the region deep inside the doublet phase, where the superconducting gap is even bigger than the hybridization and which is not relevant near the $0-\pi$ transition. We provide supporting arguments based on the variational wavefunctions (section 3) suggested by Rozhkov and Arovas [7] and on an effective Fermi-liquid model (section 4). Our NRG results also strongly suggest that the Andreev states are true bound states with vanishing broadening (section 5), another difference from the NCA results.

The subgap Andreev bound states are important from an experimental point of view as well because they are directly related to the transport properties of the superconductor–quantum dot–superconductor systems as in the recent experiment [20–23]. Recent developments in mesoscopic transport experiments may allow direct measurement of these Andreev states through tunneling spectroscopy. So far, the NCA/SNCA and the NRG method are the only methods that can treat rather systematically the many-body correlations of the Anderson-type impurity coupled to superconductors. The discrepancy between the two methods may motivate further theoretical efforts for better understanding of the many-body states of the system.

2. The NRG results

We consider a quantum dot (or magnetic impurity) coupled to two superconducting leads. The Hamiltonian $H = H_C + H_D + H_T$ consists of three parts: H_C describes two, left (L) and right (R), BCS-like s-wave superconductors with the superconducting gap $\Delta_{L(R)}$ and bandwidth D :

$$H_C = \sum_{\ell=L,R} \left[\sum_{k\sigma} \xi_{\ell k} c_{\ell k\sigma}^\dagger c_{\ell k\sigma} - \left(\Delta_{\ell} c_{\ell k\uparrow}^\dagger c_{\ell \bar{k}\downarrow} + \text{h.c.} \right) \right] \quad (1)$$

where we have used the shorthand notation $\bar{k} \equiv -k$. We assume identical superconductors, $\xi_{Lk} = \xi_{Rk} = \xi_k$ and $\Delta_L = \Delta_R^* = \Delta e^{+i\phi/2}$ where ϕ is the phase difference between the two superconductors. H_D describes an Anderson-type localized level in the quantum dot:

$$H_D = \sum_{\sigma} \epsilon_d d_{\sigma}^\dagger d_{\sigma} + U n_{\uparrow} n_{\downarrow} \quad (2)$$

with $n_{\sigma} = d_{\sigma}^\dagger d_{\sigma}$. ϵ_d is the single-particle energy of the level and U is the on-site interaction. Here we consider the particle–hole *asymmetric* case ($\epsilon_d \neq -U/2$). To compare our results directly with the NCA results, we will take $U = \infty$, preventing double occupancy. Finally, H_T is responsible for the tunneling of electrons between the quantum dot and the superconductors:

$$H_T = \sum_{\ell k\sigma} \left(V_{\ell k} c_{\ell k\sigma}^\dagger d_{\sigma} + \text{h.c.} \right). \quad (3)$$

For simplicity we will assume the symmetric junctions with tunneling elements insensitive to the energy, $V_{Lk} = V_{Rk} = V$. The broadening of the level is given by $\Gamma = \pi \rho_L(E_F) |V_L|^2 + \pi \rho_R(E_F) |V_R|^2 = 2\pi \rho(E_F) |V|^2$. For the calculation, we followed the standard NRG method [24–28] extended to superconducting leads [17, 29, 30].

There are two competing energy scales in the system. The superconductivity is naturally governed by the gap Δ . The Kondo effect is characterized by the Kondo temperature T_K , given by [9, 11, 31, 32]

$$T_K = \sqrt{\frac{\Gamma W_0}{2}} \exp \left[\frac{\pi \epsilon_d}{2\Gamma} \left(1 + \frac{\epsilon_d}{U} \right) \right] \quad (4)$$

where $W_0 \equiv \min\{D, U\}$. For $T_K \gg \Delta$, the ground state is expected to be a singlet and the Josephson current is governed by the Kondo physics. In the opposite limit $T_K \ll \Delta$, the ground state is a doublet and the transport can be understood perturbatively in the spirit of the Coulomb blockade (CB) effect [4, 33]. The transition happens at $T_K \sim \Delta$. See figure 3.

Figure 1 summarizes the results. Figure 1(a) shows the positions, E_e and E_h , of the subgap Andreev states for $U = \infty$ and $\phi = 0$. We observe two Andreev states, below and above E_F , are observed in a wide range of Δ/T_K (in particular on both sides of the transition point $\Delta_c/T_K \sim 1$). More important are figures 1(b) and (c), the spectral weights A_e (A_h) of the electron-like (hole-like) Andreev states, defined by

$$G_{dd}^R(E) \approx \frac{A_p}{E - E_p + i0^+} \quad (5)$$

near $E \simeq E_p$ ($p = e, h$). Except for very large Δ ($\Delta \gg \Gamma$), the spectral weights of both E_e and E_h are the same in order of magnitude. These observations are consistent with the behavior of the occupation of the dot level shown in figure 1(d). Unlike the intuitive interpretation by Clerk and Ambegaokar [8], the occupation does not change much across the transition point, although there is a small jump (emphasized in the blue circle in figure 1(d)). The results in figure 1 remain qualitatively the same for finite U ; an example is shown in the inset of figure 1(c). Finite phase difference (not shown in the figure) does not make any qualitative change (about the existence of the Andreev states both above and below E_F), either.

3. Variational theory

The main features of the results presented above can be understood *qualitatively* in terms of the variational wavefunctions [7]. For the singlet state in the $U = \infty$ limit we take the trial function of the form

$$|S\rangle = \left\{ \mathcal{A} + \frac{1}{\sqrt{2}} \sum_{q \in L,R} B_q (\gamma_{q\uparrow}^\dagger d_{\downarrow}^\dagger - \gamma_{q\downarrow}^\dagger d_{\uparrow}^\dagger) + \sum_{qq'} C_{qq'} \gamma_{q\uparrow}^\dagger \gamma_{q'\downarrow}^\dagger \right\} |0\rangle \quad (6)$$

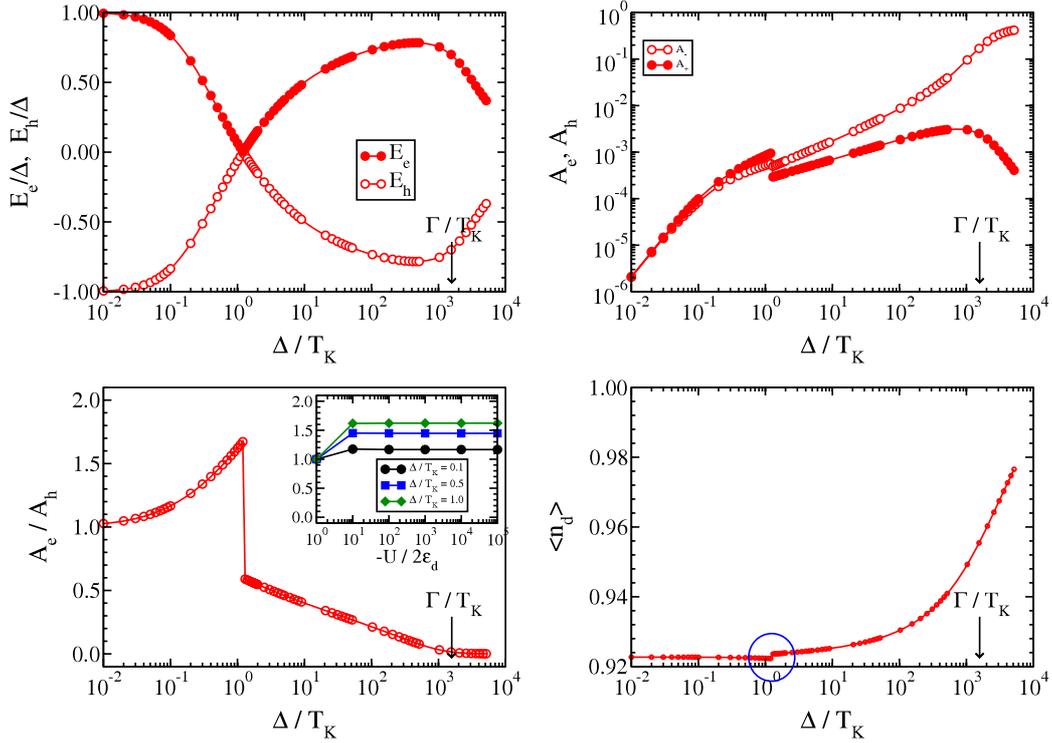


Figure 1. The NRG results on the Andreev states. The energies (a) and the corresponding spectral weights (b) of the subgap Andreev states for the $U = \infty$ Anderson model. (c) The ratio A_e/A_h of the spectral weights of the Andreev states. Inset: A_e/A_h as a function of $-U/2\epsilon_d$. (d) Average occupation of the quantum dot level. $\epsilon_d = -0.1D$, $U = \infty$, $\Gamma = 0.02D$ ($T_K \approx 3.88 \times 10^{-5}$).

with $C_{qq'} = C_{q'q}$. For the doublet state we take

$$|D_\uparrow\rangle = \left\{ \tilde{\mathcal{A}}d_\uparrow^\dagger + \sum_q \tilde{\mathcal{B}}_q \gamma_{q\uparrow}^\dagger + \sum_{qq'} \tilde{\mathcal{C}}_{qq'} \gamma_{q\uparrow}^\dagger \gamma_{q'\downarrow}^\dagger d_\uparrow^\dagger - \frac{1}{\sqrt{3}} \sum_{qq'} \tilde{\mathcal{D}}_{qq'} \gamma_{q\uparrow}^\dagger (\gamma_{q'\uparrow}^\dagger d_\downarrow - \gamma_{q'\downarrow}^\dagger d_\uparrow) \right\} |0\rangle \quad (7)$$

with $\tilde{\mathcal{C}}_{qq'} = \tilde{\mathcal{C}}_{q'q}$ and $\tilde{\mathcal{D}}_{qq'} = -D_{q'q}$, and analogously for $|D_\downarrow\rangle$. In equations (6) and (7), we have used the collective index $q \equiv (\ell, k)$ to simplify the expressions. The quasi-particle operators $\gamma_{\ell k\sigma}$ and $\gamma_{\ell k\sigma}^\dagger$ in equations (6) and (7) are related to the bare electron operators $c_{\ell k\sigma}$ and $c_{\ell k\sigma}^\dagger$ in superconductors by a unitary transform:

$$\begin{bmatrix} \gamma_{\ell k\uparrow} \\ \gamma_{\ell k\downarrow}^\dagger \end{bmatrix} = \begin{bmatrix} u_{\ell k}^* & -v_{\ell k}^* \\ v_{\ell k} & u_{\ell k} \end{bmatrix} \begin{bmatrix} c_{\ell k\uparrow} \\ c_{\ell k\downarrow}^\dagger \end{bmatrix}, \quad (8)$$

where $u_{\ell k}$ and $v_{\ell k}$ are coherence factors of the quasi-particle excitations with energies

$$E_{\ell k} = \sqrt{\xi_{\ell k}^2 + |\Delta_\ell|^2}, \quad (9)$$

and satisfy the Bogoliubov–de Gennes equation [1]:

$$\begin{bmatrix} \xi_{\ell k} & \Delta_\ell \\ \Delta_\ell^* & -\xi_{\ell k} \end{bmatrix} \begin{bmatrix} u_{\ell k} \\ v_{\ell k} \end{bmatrix} = E_{\ell k} \begin{bmatrix} u_{\ell k} \\ v_{\ell k} \end{bmatrix}. \quad (10)$$

Following the variational principle, one can determine the coefficients \mathcal{A} , \mathcal{B} , and \mathcal{C} in the trial wavefunction (6) by

minimizing the singlet-state energy:

$$E = \frac{\langle S|H|S\rangle}{\langle S|S\rangle} \quad (11)$$

and the coefficients $\tilde{\mathcal{A}}$, $\tilde{\mathcal{B}}$, $\tilde{\mathcal{C}}$ and $\tilde{\mathcal{D}}$ in the trial wavefunction (7) by minimizing the doublet-state energy:

$$\tilde{E} = \frac{\langle D_\sigma|H|D_\sigma\rangle}{\langle D_\sigma|D_\sigma\rangle}. \quad (12)$$

More explicitly, upon variation of the singlet energy E in equation (11) with respect to the coefficients \mathcal{A} , \mathcal{B} and \mathcal{C} one obtains the variational equations:

$$\mathcal{A} = \frac{\sqrt{2}V}{E} \sum_q v_q^* \mathcal{B}_q, \quad (13)$$

$$(E - \epsilon_d - E_q) \mathcal{B}_q = \sqrt{2}V \sum_{q'} C_{qq'} u_{q'}^* + \sqrt{2}V \mathcal{A} v_q, \quad (14)$$

and

$$C_{qq'} = \frac{V}{\sqrt{2}} \frac{\mathcal{B}_q u_{q'} + \mathcal{B}_{q'} u_q}{E - (E_q + E_{q'})}. \quad (15)$$

Similarly, the variation of \tilde{E} in equation (12) with respect to $\tilde{\mathcal{A}}$, $\tilde{\mathcal{B}}$, $\tilde{\mathcal{C}}$ and $\tilde{\mathcal{D}}$ gives another set of variational equations for the doublet state:

$$\tilde{\mathcal{A}} = \frac{V}{\tilde{E} - \epsilon_d} \sum_q u_q^* \tilde{\mathcal{B}}_q, \quad (16)$$

$$(\tilde{E} - E_q) \tilde{\mathcal{B}}_q = V \tilde{\mathcal{A}} u_q - V \sum_{q'} (\tilde{\mathcal{C}}_{qq'} + \sqrt{3} \tilde{\mathcal{D}}_{qq'}) v_{q'}^*, \quad (17)$$

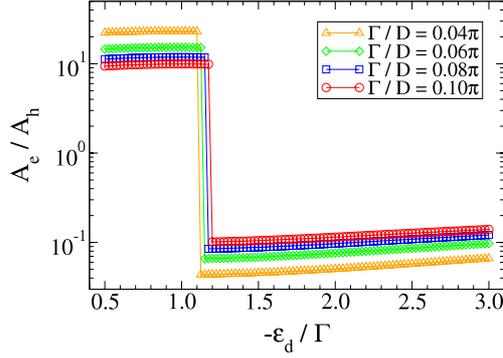


Figure 2. Results from the variational calculations. Plotted is the ratio A_e/A_h of the spectral weights as a function of $-\epsilon_d/\Gamma$ for various values of Γ . $\Delta/D = 0.1$.

$$\tilde{C}_{qq'} = -\frac{V}{2} \frac{\tilde{B}_q v_{q'} + \tilde{B}_{q'} v_q}{\tilde{E} - \epsilon_d - (E_q + E_{q'})}, \quad (18)$$

and

$$\tilde{D}_{qq'} = -\frac{\sqrt{3}V}{2} \frac{\tilde{B}_q v_{q'} - \tilde{B}_{q'} v_q}{\tilde{E} - \epsilon_d - (E_q + E_{q'})}. \quad (19)$$

The variational equations (13)–(19) are solved numerically. To this end, one has first to discretize the conduction bands of the superconductors. Since the quasi-particle density of states in superconductors has a square-root singularity ($\sim 1/\sqrt{E_q}$; see equation (9)), the discretization spacing δE should be sufficiently smaller than Δ . Moreover, in order to investigate the Kondo correlations one has to span in equations (13)–(19) energies well above the Kondo temperature T_K as well as the gap energy Δ . It means that, for $T_K \gg \Delta$, a huge number of energy slices are required to solve the variational problem. For this reason, the variational method is practically limited to the regime where T_K is not too large compared with Δ .

The local spectral density $A(E) = A^>(E) + A^<(E)$ of quasi-particle excitations on the quantum dot is given in the Lehmann representation (see [34] and equation (5)) as

$$A^>(E) = \sum_{\sigma} \sum_n |\langle n | d_{\sigma}^{\dagger} | G \rangle|^2 \delta(E - E_n) \quad (20)$$

and

$$A^<(E) = \sum_{\sigma} \sum_n |\langle n | d_{\sigma} | G \rangle|^2 \delta(E + E_n) \quad (21)$$

where $|G\rangle$ is the ground state and $|n\rangle$ are single-particle or single-hole excited states with energies E_n . $A^>(E)$ describes electron-like excitations while $A^<(E)$ describes hole-like excitations. From the form of the trial wavefunctions in equations (6) and (7), it is clear that the spectral weight of the hole-like Andreev state in the singlet phase depends on how much the quantum dot is occupied (B_q) in $|S\rangle$ and how much the quantum dot is empty (\tilde{B}_q) in $|D_{\sigma}\rangle$; namely, on the matrix element

$$\langle D_{\sigma} | d_{\bar{\sigma}} | S \rangle = -\frac{1}{\sqrt{2}} \sum_q \tilde{B}_q^* B_q \quad (22)$$

up to normalization constant $\sqrt{\langle S | S \rangle \langle D_{\sigma} | D_{\sigma} \rangle}$. Here $\bar{\sigma}$ denotes the spin direction opposite to σ . The weight of the electron-like

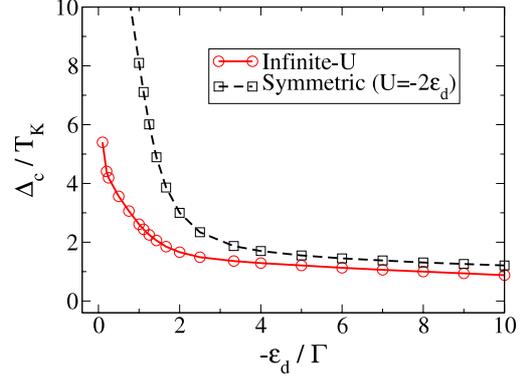


Figure 3. Phase diagram of the Anderson impurity model with superconducting leads (for $\phi = 0$) deduced from the NRG results. The phase boundaries for the infinite- U model (red solid line with circles) and for the particle–hole symmetric model (black dashed line with squares), respectively, have been calculated by the NRG method.

Andreev state in the doublet phase also depends on $\langle D_{\sigma} | d_{\bar{\sigma}} | S \rangle$. Likewise, the weight of the electron-like (hole-like) Andreev state in the singlet (doublet) phase is determined by the matrix element

$$\langle D_{\sigma} | d_{\sigma}^{\dagger} | S \rangle = \tilde{A}^* A + \sum_{qq'} \tilde{C}_{qq'}^* C_{qq'}. \quad (23)$$

According to the NCA results [8, 13], $\langle D_{\sigma} | d_{\bar{\sigma}} | S \rangle$ ($\langle D_{\sigma} | d_{\sigma}^{\dagger} | S \rangle$) should vanish in the singlet (doublet) phase. However, as shown in figure 2, neither of them vanishes, and the spectral weights A_e and A_h are similar in order of magnitude on both sides of the transition point, in agreement with the NRG results. We must point out that the agreement between the variational and NRG results is only at a qualitative level. The ratio A_e/A_h from the variational method is about five times bigger than the NRG result. However, this is not surprising because the variational method is limited in the region where T_K is not too large compared with Δ ; see the discussion below equation (19).

There is another interesting point to be noticed in the variational wavefunctions in equations (6) and (7). The lowest-energy solution to the variational equations (13)–(15) for $|S\rangle$ is well separated from the continuum. This is also the case for the doublet state $|D_{\sigma}\rangle$. It suggests that the subgap Andreev state is a true bound state without broadening. We will come back to this point in section 5 below.

4. Universality

Since the singlet–doublet transition in the system is a true quantum phase transition, the universality is also an important issue. With Δ and T_K being the only two low-energy scales in the system, physical quantities should depend only on the ratio of Δ/T_K but not on the details of the system.

In figure 4, we plotted the normalized spectral weights $A_{e(h)}/\Delta$ as a function of Δ/T_K for various values of ϵ_d/Γ . We observe that the curves of $A_{e(h)}$ overlap each other almost completely in the Kondo regime ($\Delta \ll T_K$) except for cases close to the mixed-valence regime ($|\epsilon_d| \lesssim \Gamma$). The

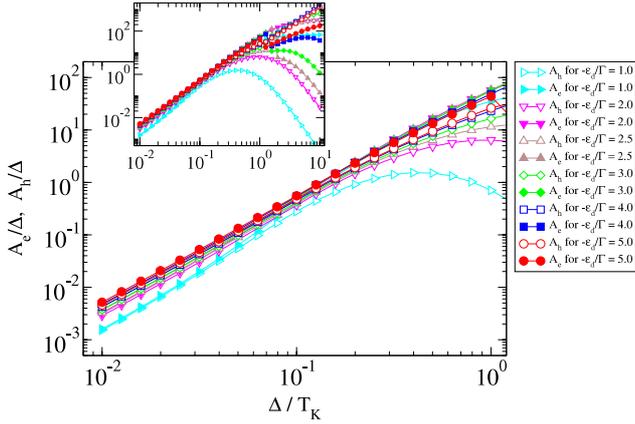


Figure 4. The NRG results of the spectral weights A_e (A_h) of the electron-like (hole-like) Andreev state as a function of Δ/T_K at $\epsilon_d = \phi = 0$ for different values of Γ . Inset: the same in a wider range of Δ/T_K including the doublet phase.

deviation from the universal behavior in the mixed-valence regime ($-\epsilon_d \lesssim \Gamma$) is not surprising because of strong charge fluctuations in the regime. This is also indicated in the phase diagram in figure 3: close to the mixed-valence regime ($-\epsilon_d/\Gamma \lesssim 1$), Δ_c/T_K becomes larger.

More interestingly, the universal curve in figure 4 can be fitted to a simple effective non-interacting model. To see this, let us consider a purely non-interacting localized level at ϵ_d . When it is coupled between two superconductors with phase difference ϕ , two Andreev bound states develop on the localized level, one being electron-like and the other hole-like. The energies, $\pm\omega_0$, of the electron-like and hole-like Andreev bound states are exactly opposite in sign. In other words, they are located symmetrically above and below the Fermi level of the superconductors. The value of ω_0 is determined by the zero of the function [14, 15]:

$$K(z) = z \left(\sqrt{1-z^2} + \Gamma \right) - \sqrt{\epsilon_d^2(1-z^2) + \Gamma^2 \cos^2(\phi/2)} \quad (24)$$

where Γ is the broadening of the level due to the coupling to the superconductors. The spectral weights of the Andreev states at energies $\pm\omega_0$ are respectively given by

$$\frac{A_{e(h)}}{\Delta} = \frac{(1-\omega_0^2)}{K'(\omega_0)K(\omega_0)} \left[z \left(1 + \frac{\Gamma}{\sqrt{1-\omega_0^2}} \right) \pm \epsilon_d \right], \quad (25)$$

where $K'(z) = dK/dz$. At the resonance ($\epsilon_d = 0$) and in the limit $\Gamma \gg \Delta$, the expression is reduced to

$$\frac{A_{e(h)}}{\Delta} \approx 2 \frac{\Delta^2}{\Gamma^2}. \quad (26)$$

Now let us try to construct an effective model for the Kondo resonance in the deep Kondo regime. The Kondo correlated state behaves like a Fermi liquid. Naturally, if the reservoirs are normal metal, the Kondo resonance can be regarded in effect as a non-interacting resonance level at the

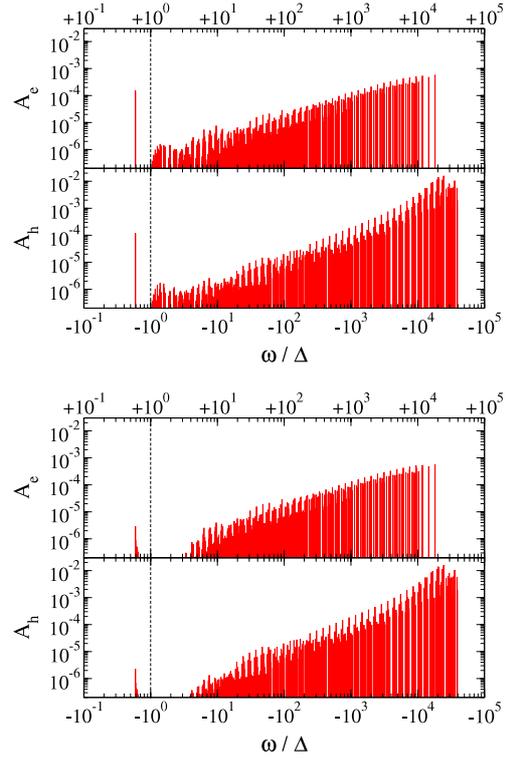


Figure 5. Raw data of the spectral weights of the discrete energy levels from the NRG calculation (a) at $T = 0.1\Delta$ and (b) at $T = 0.5$.

Fermi energy E_F . In other words, many physical properties are described pretty well by the effective impurity Greens function

$$G_d(z) = \frac{T_K/\Gamma}{z + iT_K} \quad (27)$$

with T_K playing the role of the level broadening. In the previous work [9], where the particle-hole symmetry was assumed, it was demonstrated that this may also remain to hold for superconducting reservoirs. To test this idea, let us consider an effective impurity model, where the impurity Greens function is given by equation (27) and the pairing potential [see equation (1)] is turned on the reservoir. Following the same lines leading to equation (25) in the case of a purely non-interacting resonance level, one can expect the spectral weights of the Kondo correlated Andreev states:

$$\frac{A_{e(h)}}{\Delta} \approx 2 \frac{\Delta^2}{T_K^2}, \quad (28)$$

which is the same as equation (26) except that Γ has been replaced by T_K . Indeed, the NRG data of the spectral weights in figure 4 fit very well to

$$\frac{A_{e(h)}}{\Delta} \sim \frac{\Delta^2}{T_K^2} \quad (29)$$

which is consistent with equation (28).

5. True bound state

Finally, we address whether the subgap Andreev state is a true bound state. Clerk and Ambegaokar [8] and Sellier *et al* [13]

found finite broadening of the Andreev states. This may come from the finite-temperature effects. NCA cannot go down to temperatures much lower than the Kondo temperature, and they worked at rather high temperatures [8, 13]. The spectrum from the NRG calculation is inherently discrete [35] and it is not easy to make a definite conclusion. However, as shown in figure 5, the subgap states are well separated from the continuum parts up to temperatures as high as the energy of the subgap states. At temperatures higher than the energy of the Andreev states, it is accompanied by other small spikes. It suggests that the subgap Andreev states are true bound states and that the finite broadening observed in the NCA results may be a finite-temperature effect.

6. Conclusion

We have studied the Kondo quantum dot coupled to two superconducting leads and investigated the subgap Andreev states using the NRG method. Contrary to the recent NCA results [8, 13], we observe Andreev states both below and above the Fermi level.

Acknowledgments

Special thanks are due to A Clerk for the helpful discussions and for initially drawing our attention to the issues. We also thank W Belzig for comments. This work was supported by the SRC/ERC program (R11-2000-071), the KRF grants (KRF-2005-070-C00055 and KRF-2006-312-C00543), the Second BK21 Program and the KIAS.

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