Superconducting Gap Renormalization around two Magnetic Impurities: From Shiba to Andreev Bound States

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We study the renormalization of the gap of an s-wave superconductor in the presence of two magnetic impurities. For weakly bound Shiba states, we analytically calculate the part of the gap renormalization that is sensitive to the relative orientation of the two impurity spins. For strongly exchange coupled impurities, a quantum phase transition from a sub-gap Shiba state to a supra-gap Andreev state is identified and discussed by solving the gap equation self-consistently by numerics.

PACS numbers: 74.25.Ha, 73.20.Hb, 74.45.+c

Introduction. The study of magnetic impurities in superconductors has a long history. Important effects include the renormalization (reduction) of the gap in the host superconductor by the impurities1,2 and the induced Yu-Shiba-Rusinov, or Shiba, bound states3,4. In recent years, renewed interest in chains of magnetic impurities in superconductors was furthermore driven by their potential of hosting Majorana zero modes.5–7

Using scanning tunneling microscopy, the Shiba states induced by individual magnetic impurities on the surface of a superconductor have been shown to be strongly localized at the impurity sites8,9. This can be understood as a consequence of their sub-gap energy. When this energy is below the Fermi level, the Shiba state is occupied by a single electron, whose spin direction is dictated by the impurity spin.8,9 Shiba states are to be contrasted with Andreev bound states in extended superconductor-normal-superconductor (S-N-S) heterostructures, or in heterostructures composed of superconductors with different gaps (S-S’-S). Andreev bound states can be viewed as standing waves of electrons and holes inside the gapless region that result from Andreev reflections on the S-N-S and S-S’-S interfaces.20,21

In this work, we address a scenario intermediate between single impurities and impurity chains, and specifically study the renormalization of the superconducting order parameter in the presence of two magnetic impurities. We quantify the inter-impurity scattering analytically and discuss how the Shiba states are affected by the gap renormalization. In particular, we observe that sub-gap Shiba states can transmute into supra-gap Andreev states at the impurity sites, which we show by a simple analytical model and by self-consistent numerics.

Model. In the analytical part of our study, we analyze two (weakly bound) magnetic impurities in a three-dimensional s-wave superconductor. The corresponding Hamiltonian \( H = H_{\text{kin}} + H_{\text{SC}} + H_{\text{imp}} \) can be decomposed into the kinetic energy, \( H_{\text{kin}} \), the BCS mean field superconductivity, \( H_{\text{SC}} \), and the impurity contribution, \( H_{\text{imp}} \). If there were no impurities, the system could be modeled by the Hamiltonian \( H_0 = H_{\text{kin}} + H_{\text{SC},0} \), where \( H_{\text{kin}} = \sum_{k,\sigma} E_k c_{k\sigma}^\dagger c_{k\sigma} + \text{H.c.} \) (\( E_k \) denotes the kinetic energy dispersion), and \( H_{\text{SC},0} = \int d\mathbf{r} \Delta_0 c_{\mathbf{r}\uparrow}^\dagger c_{\mathbf{r}\downarrow} + \text{H.c.} \) with a superconducting order parameter of constant value \( \Delta_0 \) (chosen to be positive). Here, \( c_{\mathbf{r}\sigma} \) is the annihilation operator for an electron of three-dimensional momentum \( \mathbf{k} \) and spin \( \sigma \), while \( c_{\mathbf{r}\sigma}^\dagger \) creates a spin \( \sigma \) electron at position \( \mathbf{r} \) (in the calculations, we use \( \uparrow \equiv +, \downarrow \equiv - \)). Due to the impurities, however, the superconducting part of the Hamiltonian is promoted from \( H_{\text{SC},0} \) to

\[
H_{\text{SC}} = \int d\mathbf{r} \Delta(\mathbf{r}) c_{\mathbf{r}\downarrow}^\dagger c_{\mathbf{r}\uparrow} + \text{H.c.} \tag{1}
\]

The order parameter is set by the self-consistent equation

\[
\Delta(\mathbf{r}) = -g \langle c_{\mathbf{r}\downarrow} c_{\mathbf{r}\uparrow} \rangle_H, \tag{2}
\]

where \( g > 0 \) is the microscopic attractive interaction between electrons in the superconductor, and where the expectation value is taken with respect to the full \( H \). The magnetic impurities, finally, are treated as classical spins \( \mathbf{S}_i \), residing at positions \( \mathbf{r}_i \), and modeled by a purely magnetic point-like scattering potential. We address impurities polarized along the \( \hat{z} \) axis in spin space, thus covering both parallel and antiparallel impurity spin alignments. The corresponding impurity Hamiltonian reads

\[
H_{\text{imp}} = \sum_{i,\sigma} J_i S_{iz} \sigma c_{\sigma}^\dagger(\mathbf{r}_i) c_{\sigma}(\mathbf{r}_i), \tag{3}
\]

where \( S_{iz} = \pm |\mathbf{S}_i| \) is the \( \hat{z} \) component of the spin of impurity \( i \), and \( J_i \) is the exchange coupling between this impurity and the electrons in the superconductor. This model comes with the Debye frequency \( \omega_D \) as a natural high-energy cutoff.

If the renormalization of the superconducting gap is small, \( |\delta \Delta(\mathbf{r})| = |\Delta(\mathbf{r}) - \Delta_0| \ll \Delta_0 \), one can approximate \( \Delta(\mathbf{r}) \) by evaluating the right-hand side of the gap equation for an unrenormalized gap,

\[
\Delta(\mathbf{r}) \approx -g \langle c_{\mathbf{r}\downarrow} c_{\mathbf{r}\uparrow} \rangle_H. \tag{4}
\]

with \( H' = H_0 + H_{\text{imp}} \). This is the approach we employ in the remainder of the analytical calculation. When

...
$[\delta \Delta(r)]$ becomes of order $\Delta_0$, this approximation ceases to be valid, and we make use of numerical simulations. The tight-binding Hamiltonian $\hat{H}$ is defined as

$$\hat{H} = -t \sum_{\langle i, i' \rangle} \sum_{\sigma = \pm 1} c^\dagger_{i\sigma} c_{i'\sigma} + \sum_{i = 1}^{N_x - N_y} \sum_{\sigma = \pm 1} [\Delta_i \sigma c^\dagger_{i\sigma} c_{i\sigma} - (\mu - J_i \sigma) c^\dagger_{i\sigma} c_{i\sigma}] + \text{H.c.},$$

where $c^\dagger_{i\sigma}$ is the annihilation operator acting on an electron with spin $\sigma$ at lattice site $i$, and the first sum runs over neighboring sites $i$ and $i'$ located in a two-dimensional square lattice of size $N_x \times N_y$ with lattice constant $a$. The chemical potential $\mu$ is taken from the bottom of the energy band, and the local order parameter $\Delta_i$ is determined self-consistently in an iterative procedure for fixed value of the exchange coupling $J_i$ at the site $i$ starting from the uniform superconducting order parameter $\Delta_0$ until convergence is reached.  

**Analytical $T$-matrix approach.** We begin with calculating the full imaginary time Nambu Green's function $\hat{G}(r, r', \tau, \tau') = -\langle T_\tau \Psi^\dagger(r', \tau') \Psi(r, \tau) \rangle_{\mu'}$ using the 'bare' Green's function $\hat{G}_0(r, r', \tau, \tau') = -\langle T_\tau \Psi^\dagger(r', \tau') \Psi(r, \tau) \rangle_{\mu_0}$, and where the Nambu spinor is defined as $\Psi_k = (c^\dagger_{k\uparrow}, c^\dagger_{k\downarrow})^T$. For equal positions $r = r'$, a Fourier transformation from imaginary time to Matsubara frequencies $\omega_n$ yields

$$\hat{G}_0(r, r, \omega_n) = -\frac{\pi \nu_F}{\sqrt{\omega_n^2 + \Delta_0^2}} (i\omega_n \tau_0 + \Delta_0 \tau_x),$$

with $\nu_F$ being the density of states (per spin) at the Fermi energy, and with the Pauli matrices $\tau_i$ acting in Nambu space ($\tau_0 = \text{I}_2 \times 2$), while we obtain

$$\hat{G}_0(r, r', \omega_n) = -\frac{\pi \nu_F}{\sqrt{\omega_n^2 + \Delta_0^2}} \frac{e^{-\sqrt{\omega_n^2 + \Delta_0^2} |\delta r| / \nu_F}}{k_F |\delta r|}$$

$$\times \left( \sin(k_F |\delta r|) (i\omega_n \tau_0 + \Delta_0 \tau_x) + \cos(k_F |\delta r|) \sqrt{\omega_n^2 + \Delta_0^2} \tau_z \right),$$

at distances $|\delta r| = |r - r'|$ larger than the Fermi wavelength ($k_F$ is the Fermi momentum). Using the imaginary time-dependent Dyson equation, the full Green’s functions can be expressed as

$$\hat{G}(r, r', \omega_n) = \hat{G}_0(r, r', \omega_n)$$

$$+ \sum_{i,j} \hat{G}_0(r_i, r_i, \omega_n) T_n^{ij} \hat{G}_0(r_j, r', \omega_n),$$

where the $T$-matrix $T_n = V \left[ \text{I}_{4 \times 4} - G_{0,n} V \right]^{-1}$ involves the $4 \times 4$-matrix $G_{0,n}$ with entries $G_{0,n}^{ij} = \hat{G}_0(r_i, r_j, \omega_n)$, and $V = \text{diag}(J_1 S_{1z}, J_2 S_{2z}, J_3 S_{3z}, J_4 S_{4z})$.

**Single impurity physics.** When the impurities are far apart from each other, $(k_F r^{(12)})^{-1} \ll 1$, the gap near a given impurity is predominantly renormalized by scattering processes off this impurity. We can then for instance focus on impurity $1$. Next to this impurity, and using $\alpha_i = \pi \nu_F J_i S_{iz}$, the dominant contribution to the gap renormalization at $r_1$ reads

$$\delta \Delta^{(1)}(r_1) = \frac{g \pi |\alpha_1| |\Delta_0 \nu_F|}{(1 + |\alpha_1|^2)^2}$$

$$- \frac{g a^2 \Delta_0}{2} \int_{-\infty}^{\infty} \frac{d \omega}{|\omega|^2 + \Delta_0^2} \frac{\omega^2 (3 + |\alpha_1|^2)}{\omega^2 (1 + |\alpha_1|^2)^2} + 4 a^2 \Delta_0^2.$$

A Gaussian high-energy cutoff at the scale $\omega_J$ regularizes the logarithmic UV divergence of this integral. For small $|\alpha_1|$, we can further expand this expression to second order in $\alpha_1$. Since the self-consistent equation \(8\) yields a bare gap of $\Delta_0 \approx 2 \omega_J e^{-1/\nu_F}$, we find

$$\delta \Delta(r_1) \approx -3 |\alpha_1|^2 \Delta_0.$$
ψ

0

\alpha

(\text{blue dots}) is suppressed for small ¯\alpha Andreev state transitions. The gap between the impurities hence, in ∆

occupied, accompanied by jumps in bound state energies and, E

states have finite overlap, but restores at larger ¯\alpha.

FIG. 1: The energy spectrum (black, green, and brown dots) and the size of the superconducting gap ∆1 directly under the impurity (red dots) for a square lattice of size 17a × 13a with (a) a single impurity (b) two identical impurities (\bar J1 = \bar J2) as function of exchange \bar J1/\bar t found numerically. ∆1 gets suppressed with increasing \bar J1 and an effective S-S’ junction builds up. After the phase transition at the critical value \bar Jc, ∆1 changes its sign giving rise to a π-junction. Sub-gap Shiba states (green dots) evolve into supra-gap Andreev states (brown dots) at values of \bar JSA where the bound state energy coincides with ∆1. (b) For two impurities, separated by a distance x12/a = 4, the hybridization between the two bound states lifts their degeneracy. There are thus two phase transitions, one at \bar J1c and one at \bar J2c, where these states become occupied, accompanied by jumps in bound state energies and, hence, in ∆1 and ∆2. There are also two distinct Shiba to Andreev state transitions. The gap between the impurities (blue dots) is suppressed for small \bar J1, when the two bound states have finite overlap, but restores at larger \bar J1, see also Fig. 2. The chosen parameters are ∆0/\bar t = 0.4 and μ/\bar t = 0.9.

an equation for the wave function at the impurity site, ψ(0), in terms of the bound state energy E,

\psi(0) = \int \frac{d\mathbf{p}}{(2\pi)^3} \left( E + \xi_p \tau_z + \Delta_0 \tau_x \right) \left( \sqrt{E^2 + \xi_p^2 - \Delta_0^2} \right) \psi(0),

where \xi_p = p^2/2m - \mu, and \tau corresponds to the parallel and antiparallel orientation of the bound state spin as compared to the impurity spin, respectively. This equation can be rewritten as

\begin{equation}
1 - \tau_z (\alpha' E \pm \alpha \Delta_0) + (\alpha' \Delta_0 \pm \alpha E)^2 \sqrt{\Delta_0^2 - E^2} \psi(0) = 0,
\end{equation}

where \alpha' = \nu F \pi a^3 \Delta', and \alpha = \pi \nu F J S. We find that bound state energies E within the bulk gap ∆0 satisfying Eq. (11) are given by

\begin{equation}
\frac{E}{\Delta_0} = \tau \frac{1 - w^2}{1 + w^2},
\end{equation}

where \tau is the eigenvalue of \tau_z, and w = τα±α. Plugging this expression for E back into Eq. (11), we find that a necessary condition is |w| = τw = 0. Because consistency with BCS theory requires ∆0, ∆′ < \EF, and since we are primarily interested in the regime where |α| is of order unity, we find that |α| ≫ |α′|. Therefore, τ and ±α must be of the same sign, and there are two solutions with opposite spin like for the uniform case. Defining as above the bound state as a Shiba state when its energy is within the renormalized gap, |E| < |∆0 − ∆′|, and as an Andreev state when its energy is between the renormalized gap and the bulk gap, |∆0 − ∆′| < |E| < ∆0, the critical value of the exchange coupling for the transition from a Shiba to an Andreev state reads

\begin{equation}
|\nu F J S_0| = -\alpha' + \sqrt{\Delta^2 - 2\Delta_0 \Delta'} \approx \sqrt{\Delta^2 - 2\Delta_0 \Delta'},
\end{equation}

for ∆ > ∆0, |α| + α′ > 1 (as in the numerics), and for ∆′ < ∆0, |α| + α′ < 1, while otherwise the fraction under the square root has to be inverted.

Two impurity physics. When the impurities are further apart than the Fermi wavelength, r_{12} = |\mathbf{r}_1 - \mathbf{r}_2| \gg k_F^{-1}, we can expand the gap renormalization in orders of (k_F r_{12})^{-1}. To second order, we find

\begin{equation}
G(\mathbf{r}_1, \mathbf{r}_1', \omega_n) \approx G_{00}^{11} + G_{01}^{11} \tau_n^{(1)} G_{01}^{11} + (1 + G_{00}^{11} \tau_n^{(1)}) G_{00}^{12} T_n^{(2)} G_{01}^{21} \left( 1 + T_n^{(1)} G_{00}^{11} \right),
\end{equation}

where T_n^{(i)} = J_i \tau_z \left( I_{2 \times 2} - J_i \tau_z G_{01,2}^{ii} \right)^{-1} is the \text{T}-matrix for scattering off impurity i = 1, 2. The terms

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{The energy spectrum (black, green, and brown dots) and the size of the superconducting gap ∆1 directly under the impurity (red dots) for a square lattice of size 17a × 13a with (a) a single impurity (b) two identical impurities (\bar J1 = \bar J2) as function of exchange \bar J1/\bar t found numerically. ∆1 gets suppressed with increasing \bar J1 and an effective S-S’ junction builds up. After the phase transition at the critical value \bar Jc, ∆1 changes its sign giving rise to a π-junction. Sub-gap Shiba states (green dots) evolve into supra-gap Andreev states (brown dots) at values of \bar JSA where the bound state energy coincides with ∆1. (b) For two impurities, separated by a distance x12/a = 4, the hybridization between the two bound states lifts their degeneracy. There are thus two phase transitions, one at \bar J1c and one at \bar J2c, where these states become occupied, accompanied by jumps in bound state energies and, hence, in ∆1 and ∆2. There are also two distinct Shiba to Andreev state transitions. The gap between the impurities (blue dots) is suppressed for small \bar J1, when the two bound states have finite overlap, but restores at larger \bar J1, see also Fig. 2. The chosen parameters are ∆0/\bar t = 0.4 and μ/\bar t = 0.9.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{The cross-section of the spatial profile of the superconducting gap ∆ for two impurities located on the x axis for fixed \bar J2/\bar t = 2.3 and varying \bar J1: \bar J1/\bar t = 1 (yellow dotted line), \bar J1/\bar t = 1 (green dotted dashed line), \bar J1/\bar t = 2.3 (red dashed line), \bar J1/\bar t = 4 (blue solid line). The gap under the first impurity is increasing suppressed with increasing exchange coupling: After the phase transition \bar J1 > \bar Jc, π-junctions (negative gap value) under the impurity arise accompanied by restoration of the gap between the two impurities. The gap under the second impurity is suppressed the most when \bar J1 ≈ \bar J2 since then the two bound states hybridize strongly. All parameters are the same as in Fig. 1a.}
\end{figure}
\(G_{0,n}^{(i)} T_{n}^{(i)} G_{n}^{(i)}\) give rise to the single impurity renormalizations of the order parameter, which we denote as \(\delta \Delta^{(i)}\), while the other corrections to the bare Green’s function result in inter-impurity gap renormalizations \(\delta \Delta^{(12)}\). These different contributions are depicted in Fig. 3. The inter-impurity renormalizations have both terms that depend on the relative orientation of the two spins (parallel for \(\alpha_1 \alpha_2 > 0\), antiparallel for \(\alpha_1 \alpha_2 < 0\)), as well as orientation-independent contributions. All of these are of the order \((k_F r_{12})^{-2}\). Most interesting is the orientation-dependent contribution evaluated at the site of one of the impurities. As an example, \(\delta \Delta^{(12)}(r_1)\) reads at zero temperature

\[
\delta \Delta^{(12)}(r_1) = \mp |\alpha_1 \alpha_2| \Delta_0 \mu_F \int d\omega e^{-\frac{2r_{12}}{k_F r_{12}}} \sqrt{\omega^2 + \Delta_0^2} \\
\times \left\{ \left(1 - \alpha_1^2\right) \left(1 + \alpha_2^2\right) \omega^2 - \left(1 - \alpha_1^2\right)^2 \Delta_0^2 \right\} \\
- 2\left(1 - \alpha_1^2\right) \left(1 + \alpha_2^2\right) \omega^2 \cos(2k_F r_{12}),
\]

(15)

with \(A_i = (1 - \alpha_1^2)^2 \Delta_0^2 + (1 + \alpha_2^2)^2 \omega^2\). The upper (lower) sign applies for parallel (antiparallel) impurity spin alignment.

Besides addressing the gap renormalization at the sites of the impurities, it is also interesting to analyze the scaling of the different contributions to the gap renormalization as a function of the inter-impurity distance in their middle, that is at \(\mathbf{R} = (r_1 + r_2)/2\). The single impurity contributions are found to scale as

\[
\delta \Delta^{(i)}(\mathbf{R}) \sim \Delta_0 e^{-r_{12}/\xi} (k_F r_{12})^{-2},
\]

(16)

where \(\xi\) is the superconducting coherence length. This scaling has a simple geometrical interpretation: to leading order in \((k_F r_{12})^{-1} \ll 1\), the anomalous Green’s function (which determines the gap) is renormalized by the electron traveling from \(\mathbf{R}\) to impurity \(i\) at \(r_i\), scattering there, and coming back to \(\mathbf{R}\). Since a trip to, and back from, the impurity involves a propagator proportional to \((k_F r_{12})^{-1}\), see Eq. (8), the gap renormalization deriving from the single impurity scattering processes scales as \((k_F r_{12})^{-2}\). The total distance covered during these processes, precisely equal to \(r_{12}\), determines the argument of the exponential. The leading order inter-impurity terms, on the other hand, are found to scale as

\[
\delta \Delta^{(12)}(\mathbf{R}) \sim \Delta_0 e^{-2r_{12}/\xi} (k_F r_{12})^{-3},
\]

(17)

since they stem from an electron traveling first to impurity one, then to impurity two, and then coming back. This involves three trips, and a total distance of 2\(r_{12}\). The inter-impurity renormalization \(\delta \Delta^{(12)}\) is thus suppressed by an additional power of \(k_F r_{12} \gg 1\) in between the impurities as compared to its value at one of the impurities, see Eq. (15). Consequently, the inter-impurity gap renormalization \(\delta \Delta^{(12)}\) can be modeled as a function with well-defined peaks close to the two impurities, as shown in Fig. 3. We also confirm numerically that the renormalizations of the superconducting gap is the largest directly under the impurity, while, depending on parameters, \(\Delta(\mathbf{r})\) could even get larger than its initial value around the impurities, see Fig. 2.

With increasing exchange strengths, a similar transition as before between Shiba and Andreev states can also be observed in the case of two impurities, see Fig. 1b. First we note that when the two impurities are close and with comparable exchange strengths, the bound states overlap and their degeneracy gets lifted by hybridization \(\delta \Delta(\mathbf{r})\), which in turn gives rise to two separate quantum phase transitions in \(\Delta\), one at \(J_1\), and one at \(J_2\). As a consequence, we find now two distinct Shiba to Andreev state transitions around these values of exchange strengths, see Fig. 3b.

**Small impurity distances.** Let us finally address the case of small impurity distances \(k_F r_{12} \to 0\), where \(G_0(r_1, r_2, \omega_n) \to G_0(r_1, r_1, \omega_n) = G_0(r_2, r_2, \omega_n)\). Keeping all orders of \((k_F r_{12})^{-1} \gg 1\) in Eq. (7), we find that the gap renormalization is in this limit given by

\[
\delta \Delta(r_1) = \delta \Delta(r_2) = \delta \Delta^{(1)}(r_1) |_{\alpha_1 \to \alpha = \alpha_1 + \alpha_2},
\]

(18)

where \(\delta \Delta^{(1)}(r_1)\) is given in Eq. (8). Quite naturally, the gap renormalization resulting from two very close classical impurities with \(\alpha_1\) and \(\alpha_2\) is thus equal to the renormalization of a single impurity with \(\alpha = \alpha_1 + \alpha_2\). Provided that \(J_1\) and \(J_2\) have the same sign, the superconducting gap is reduced the least if the two spins are aligned antiparallel for short distances.

**Conclusions.** Analyzing two classical spins in a superconductor, we obtained the gap renormalization analytically for weakly and numerically for strongly coupled bound states. In addition, we found a transition for Shiba to Andreev states which is accompanied by two subsequent quantum phase transitions as the exchange coupling is varied. These predictions lead to dramatic spectral changes which could be observed, for instance, by STM techniques along the same lines as in previous experiments.7,13,23

![FIG. 3: Sketch of the renormalization of the superconducting gap, \(\delta \Delta(\mathbf{r}) = \Delta(\mathbf{r}) - \Delta_0\), in the presence of two spin-impurities localized at positions \(r_1\) and \(r_2\) along the \(x\) axis connecting the two impurities. The sketch shows the envelopes of three different contributions to the gap renormalization (see text).](image)
Acknowledgments

We thank L. Glazman and S. Gangadharaiah for valuable discussions. This work has been supported by the Swiss NF, NCCR QSIT, by the DFG through GRK 1621 and SFB 1143, and by the French Agence Nationale de la Recherche through the ANR contracts Dymesys and Mistral.

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