Impurity-Induced Virtual Bound States in \textit{d}-Wave Superconductors

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Abstract

It is shown that a single, strongly scattering impurity produces a bound or a virtual bound quasiparticle state inside the gap in a \textit{d}-wave superconductor. The explicit form of the bound state wave function is found to decay exponentially with angle-dependent range. These states provide a natural explanation of the second Cu NMR rate arising from the sites close to Zn impurities in the cuprates. Finally, for finite concentration of impurities in a \textit{d}-wave superconductor, we reexamine the growth of these states into an impurity band, and discuss the Mott criterion for this band.
Effects of impurities on the properties of superconductors have been investigated in great detail for low-temperature [1], heavy-fermion [2], and high-temperature superconductors [3]. The main reason for the interest in the effects of impurities on the superconducting state is the fact that the superconducting properties are qualitatively modified by impurity atoms, depending, for example, whether they are magnetic or non-magnetic. In principle, this observation can be useful as a method of identifying the nature of the pairing state in superconductors. For example, any magnetic impurity will be a strong pair-breaker for (s-wave, d-wave, etc.) spin-singlet superconductors, in accord with the generalized Anderson theorem. On the other hand, even scalar (non-magnetic) impurities are pair-breakers for “higher-orbital-momentum” states, such as a d-wave pairing state.

The two main approaches in understanding the effects of impurities in conventional (s-wave) superconductors rely either on the strong- or on the weak-scattering limit. (a) The Abrikosov-Gor’kov (AG hereafter) formalism [4] treats impurities in the Born approximation. Any impurity problem is characterized by two physical parameters: the phase shift $\delta_0$ due to impurity scattering (which we assume to be s-wave) and the concentration of impurities $n_{\text{imp}}$. In the AG approach, the only parameter entering the formalism is the scattering rate, $\tau^{-1} = (2n_{\text{imp}}/\pi N_0) \sin^2 \delta_0$, proportional to the product of the concentration and $\sin^2 \delta_0$. Here $N_0$ is the normal-state density of states at the Fermi energy, and $\delta_0 = N_0 V$ is the s-wave phase shift for a weak impurity potential $V$. Therefore, in the limit of dilute concentration of strong magnetic impurities, the AG approach will yield a small average scattering rate. On the other hand, the local properties of the superconductor near an impurity site, such as the local density of states and the gap amplitude, will be modified dramatically. In this limit (b) the Yu-Shiba approach [6,7] should be used, which treats magnetic impurities in the unitary-scattering limit with the s-wave phase shift $\delta_0 \approx \pi/2$. It was shown by Yu and Shiba that, in the unitary limit, a localized magnetic impurity, interacting with the spin density of conduction electrons at the impurity site, produces a true bound state inside the energy gap, $|\omega| < \Delta_0$, where the density of states vanishes. Note that, in general, the overlap with the particle-hole continuum only allows virtual states to be formed with finite
lifetime. The relation between this approach and the AG treatment was established in [7], where it was shown that in the Born limit one recovers the AG results, and the bound state is indistinguishably close to the band edge.

Our work is partially motivated by the fact that non-magnetic impurities are known to be strong pair-breakers in a nontrivial superconductor. The Zn substitutions in cuprates are one example of this. Although Zn ions are nominally non-magnetic, $T_c$ is strongly suppressed by Zn substitution of Cu in the planes [8,9]. Therefore, it is reasonable to assume that Zn ions are non-magnetic unitary scatterers.

The purpose of this Letter is to address the question of virtual impurity-bound states in a $d$-wave superconductor and, within this framework, to explore possible implications of the assumption that the pairing in cuprates is in the $d_{x^2-y^2}$ channel. We shall generalize the original Yu-Shiba [3,4] approach to the case of arbitrary-strength of non-magnetic impurities in a $d_{x^2-y^2}$ superconductor. The results, summarized below, can be easily generalized for any nontrivial pairing state and may be relevant for heavy-fermion superconductors with impurities as well.

Our main results are as follows: (i) A strongly-scattering scalar impurity is a requirement for a localized, virtual or marginally real, bound state to exist in a $d$-wave superconductor. It is intuitively obvious that any strong enough pair-breaking impurity — magnetic or non-magnetic — will induce such a state. Indeed, the low-lying quasiparticle states close to the nodes in the energy gap will be strongly influenced even by a non-magnetic impurity potential, resulting in a well-defined bound state in the unitary limit. This should be compared to the fact that, in $s$-wave superconductors, both magnetic and resonant non-magnetic impurities produce bound states inside the energy gap [10]. (ii) The energy $\Omega'$ and the decay rate $\Omega''$ of this state are given by

$$\Omega \equiv \Omega' + i\Omega'' = \Delta_0 \frac{\pi c/2}{\log(8/\pi c)} \left[ 1 + \frac{i\pi}{2} \frac{1}{\log(8/\pi c)} \right]$$

where $c = \cot \delta_0$. We have assumed impurity scattering to be close enough to the unitary limit so that the result can be computed to logarithmic accuracy with $\log(8/\pi c) \gg 1$. It
is only in this limit that the bound state is well-defined. In the unitary limit, defined as $\delta_0 \to \pi/2 (c \to 0)$, the virtual bound state becomes a marginally bound one at $\Omega \to 0$ with $\Omega''/\Omega' \to 0$. In the opposite case of weak scattering with $c \lesssim 1$, the energy of the virtual bound state formally approaches $\Omega' \sim \Delta_0$ and the state is ill-defined because $\Omega'' \sim \Omega'$ (see Fig. 1). The wave function of the bound state is found to decay exponentially, except along the directions of the vanishing gap. (iii) For a finite impurity concentration $n_{\text{imp}}$, we recover the results obtained earlier for non-conventional superconductors [2,3]. While generally one finds that an impurity band is formed after averaging over impurity positions, the Mott criterion for the formation of the impurity band should be modified because of highly anisotropic impurity states; we suggest that the metal-insulator transition can be observed in the impurity band.

**Single-Impurity Problem.** Consider the single scalar impurity problem with

$$H_{\text{int}} = \sum_{\vec{k} \vec{k}'} V_{\vec{k} \sigma} c^\dagger_{\vec{k} \sigma} c_{\vec{k}' \sigma},$$

where $V$ is the strength of the scalar impurity potential at $\vec{r} = 0$, resulting in $s$-wave phase shift $\delta_0$.

The scattering of quasiparticles off the impurity is described by a $T$-matrix, $\hat{T}(\omega)$, which is independent of wavevector. The Green’s function in the presence of an impurity is

$$\hat{G}_{\vec{k} \vec{k}'}(\omega) = \hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega) + \hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega) \hat{T}(\omega) \hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega),$$

where both $\hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega)$ and $\hat{T}(\omega)$ are matrices in Nambu space. Here $[\hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega)]^{-1} = \omega \hat{\tau}_0 - \Delta_{\vec{k}} \hat{\tau}_1 - \xi_{\vec{k}} \hat{\tau}_3$, where $\xi_{\vec{k}}$ is the quasiparticle energy, $\Delta_{\vec{k}} = \Delta_0 \cos 2\varphi$ is the gap function with $d_{x^2-y^2}$-symmetry, $\hat{\tau}_i (i = 1, 2, 3)$ are the Pauli matrices, and $\hat{\tau}_0$ is the unit matrix in Nambu particle-hole-spinor space.

From the previous analysis [2,4,7], it is known that $\hat{T} = T_0 \hat{\tau}_0$ for s-wave scattering and a particle-hole symmetric system. Therefore, the only relevant term in the $T$-matrix takes the form

$$T_0(\omega) = G_0(\omega)/[c^2 - G_0(\omega)^2],$$

where $G_0(\omega) = \frac{1}{2\pi N_0} \sum_{\vec{k}} \text{Tr} \hat{G}^{(0)}_{\vec{k} \vec{k}'}(\omega) \hat{\tau}_0$. The virtual and bound states in the single-impurity problem are given by the poles of the $T$-matrix

$$c = \pm G_0(\Omega),$$

which is an implicit equation for $\Omega$ as a function of $c$, the strength of impurity scattering. The two signs in Eq. (2) are a result of the particle-hole symmetry. Choosing the gap function
at the Fermi surface so that \( \Delta(\varphi) = \Delta_0 \cos 2\varphi \), one finds \( G_0(\omega) = \left\langle \omega/\sqrt{\Delta(\varphi)^2 - \omega^2} \right\rangle_{FS} \), where the angular brackets denote averaging over the Fermi surface; for simplicity, we take \( \langle \bullet \rangle_{FS} = \int \bullet \, d\varphi/2\pi \). For \( |\omega| \ll \Delta_0 \), one finds
\[
G_0(\omega) = \frac{2\omega}{\pi \Delta_0} \left( \log \frac{4\Delta_0}{\omega} - \frac{i\pi}{2} \right).
\]
(3)

In principle, the solution of Eq. (2) is complex, indicating a resonant nature of the quasiparticle state, better described as a virtual state. This is easily seen from Eq. (1), which solves Eq. (2) to logarithmic accuracy. However, as \( c \to 0 \), the resonance can be made arbitrarily sharp. For \( c = 0 \), the virtual state becomes a marginally well-defined state bound to the impurity. Exact numerical solution of Eq. (2) as a function of \( c \) is shown in Fig. 2. To our knowledge, this result has not been claimed before. As \( c \to 1^- \), \( \Omega' \) and \( \Omega'' \) increase without bound so that \( \Omega''/\Omega' \to 1^- \), and the solution becomes unphysical. For \( c > 1 \), no solution has been found for \( \Omega \).

There are important physical implications of these bound states in a \( d \)-wave superconductor. Consider the most interesting case of unitary impurities in the dilute limit, separated by a distance greater than the coherence length \( \xi \). Before averaging over impurities, these bound states are nearly localized close to the impurity sites (see below) and can substantially modify the local characteristics of the superconductor: for example, the local density of states and the local NMR relaxation rates of atoms close to the impurities.

Consider a local density of states, defined as \( N(\vec{r}, \omega) = -\frac{1}{\pi} \text{Im} G(\vec{r}, \vec{r}'; \omega + i0^+) \), with the total Green’s function in the presence of the impurity \( \hat{G}(\vec{r}, \vec{r}'; \omega) = \hat{G}^{(0)}(\vec{r} - \vec{r}', \omega) + \hat{G}^{(0)}(\vec{r}, \omega) \hat{T}(\omega) \hat{G}^{(0)}(\vec{r}', \omega) \), the second term describing the local distortion due to the impurity. Using the eigenstates representation of \( G(\vec{r}, \vec{r}'; \omega) = \sum_n \langle \psi_n^*(\vec{r}) \psi_n(\vec{r}') \rangle / (\omega - E_n) \), we find two terms in the local density of states \( N(\vec{r}, \omega) = N(\omega) + N_{\text{imp}}(\vec{r}, \omega) \approx \sum_n \langle \psi_n^*(\vec{r}) \psi_n(\vec{r}) \rangle \delta(\omega - E_n) \), for well defined states. The first term originates from the bulk quasiparticles, which are described by plane-wave eigenstates with \( E_{\vec{k}} = \sqrt{\xi^2_{\vec{k}} + \Delta_{\vec{k}}^2} \), \( G^{(0)}(\vec{r}, \omega) = \sum_{\vec{k}} [u_{\vec{k}}^2 / (\omega - E_{\vec{k}}) + v_{\vec{k}}^2 / (\omega + E_{\vec{k}})] \), where \( u_{\vec{k}} \) and \( v_{\vec{k}} \) are the standard Bogoliubov factors. The bulk density of states is constant in the system with \( N(\omega)/N_0 = \omega/\Delta_0 \), for \( \omega \ll \Delta_0 \). The second term, \( N_{\text{imp}}(\vec{r}, \omega) = \)
\[ -\frac{1}{\pi}\text{Im} \left[ \hat{G}^{(0)}(\vec{r}, \omega)\hat{T}(\omega)\hat{G}^{(0)}(\vec{r}, \omega) \right]_{11}, \]
originates from the virtual quasiparticle state created at the impurity:
\[ N_{\text{imp}}(\vec{r}, \omega) = -\frac{1}{\pi}\text{Im} \sum_n (\psi_{\text{imp},n}^*(\vec{r})\psi_{\text{imp},n}(\vec{r})) / (\omega - E_{\text{imp},n} + i0^+). \]
As an important example, consider the limit of unitary scattering for which the resonant state is formed at \( E_{\text{imp},n} \equiv \Omega \rightarrow 0 \). Because \( \text{Im} G^{(0)}(\vec{r}, \omega = 0) = -\pi N(\omega = 0) = 0 \), only the imaginary part of the T-matrix contributes to \( N_{\text{imp}} \) and the bound-state probability density is found to decay as the inverse second power of the distance from the impurity along the nodes of the gap function,
\[ N_{\text{imp}}(\vec{r}, \omega = 0) = \text{Re} \left[ \hat{G}^{(0)}(\vec{r}, \omega = 0) \right]^2 \propto r^{-2}, \quad (4a) \]
and exponentially in the vicinity of the extrema of the gap function,
\[ N_{\text{imp}}(\vec{r}, \omega = 0) \propto [\xi(\varphi)/r]^{-1} e^{-2r/\xi(\varphi)}, \quad (4b) \]
where \( \xi(\varphi) \) is the angle-dependent coherence length of the superconductor, naturally defined as \( \xi(\varphi) = \hbar v_F / |\Delta(\varphi)| \). The fact that the impurity state is marginally bound is reflected in the logarithmically divergent normalization. This divergence should be cut off at an average distance between impurities at any finite concentration. More generally, for an arbitrary position of the resonance, taking into account that only one state has been produced with \( E_{\text{imp},n} = \Omega' + i\Omega'' \), we find \( N_{\text{imp}}(\vec{r}, \omega) = \frac{1}{\pi}\sum_i F(\vec{r} - \vec{r}_i) \Omega_i'' / \left[ (\omega - \Omega_i')^2 + \Omega_i''^2 \right] \), where we have introduced the sum over different impurities, located at \( \vec{r}_i \), and \( F(\vec{r} - \vec{r}_i) = \langle \psi_{\text{imp}}^*(\vec{r} - \vec{r}_i)\psi_{\text{imp}}(\vec{r} - \vec{r}_i) \rangle \) is the probability density of the \( i \)-th impurity state.

The local variations of the density of states can be probed directly, in principle, by scanning-tunneling microscopy. However the NMR experiments on Cu in Zn-doped cuprates are quite revealing as well. From Eq. (4a) and below, one concludes immediately that the local NMR signal would show two distinct relaxation rates (or even the hierarchy of rates): one coming from the Cu sites, far away from the impurities, and another from the sites, close to the impurities. The Cu sites near the impurities will be sensitive to the higher local density of states and will have a higher relaxation rate at low temperatures \([13]\). At finite impurity concentration (\( \sim 2\% \)), the volume-averaged density of states will have a finite limit
at $\omega \to 0$, as follows from Eq. (1a). The relaxation rates of Cu atoms close to and away from an impurity will, therefore, have the same temperature dependence $(T_1 T)^{-1} = \text{const}$, but will be of a different magnitude.

This behaviour has been observed experimentally: Ishida et al. [8] have measured two NMR relaxation rates for Cu in Zn-doped YBa$_2$Cu$_3$O$_{7-\delta}$. The second NMR signal with higher relaxation was inferred arising from the near-impurity Cu sites. A direct comparison of our prediction for local quantities, as probed by NMR, will require a specific model and is left for a future publication.

We would like to contrast our picture of the dilute limit of strongly scattering centers to the usual approach of averaging over impurities at finite concentration. If one considers averaging over impurities, two NMR relaxation rates, arising from unequivalent sites, cannot be resolved; the local inhomogeneous aspect of the localized states is lost after averaging over impurity positions.

For practical purposes the distinction between the impurity bound states and continuum in our case is not as well defined as in $s$-wave superconductors. Any finite temperature will produce a finite lifetime for these bound states, and they will be hybridized with the continuum of low-energy quasiparticles.

**Finite Concentration of Impurities.** Consider the growth of the impurity band with finite concentration of strongly scattering impurities. As was mentioned above, scalar (non-magnetic) impurities are pair-breakers for any nonconventional superconductor, and they substantially change the low-energy spectrum of superconducting quasiparticles. This problem has been addressed earlier in great detail (for example, see [2,3]). Here we will repeat the main steps and give results for the quasiparticle scattering rate and low-energy density of states for completeness.

For finite impurity concentration, the self-consistent Green’s function, averaged over impurity positions, obeys the Dyson equation $\langle \hat{G}_k(\omega) \rangle^{-1} = \langle \hat{G}^{(0)}_k(\omega) \rangle^{-1} - \hat{\Sigma}(\omega)$ with $\hat{\Sigma}(\omega) = n_{\text{imp}} \hat{T}(\omega)$ in the single-site approximation. The $T$-matrix has to be determined
self-consistently with \((G_0(\omega)) = \frac{1}{2\pi N_0} \sum_k \text{Tr} \langle \hat{G}_k(\omega) \rangle \hat{\tau}_0\). As above [see the remark preceding Eq.(2)], only \(\Sigma_0 \equiv \frac{1}{2} \text{Tr} \hat{\Sigma} \hat{\tau}_0\) is nonzero. The algebra is straightforward, and for unitary scattering yields
\[
\gamma \simeq \sqrt{n_{\text{imp}}(\Delta_0/\pi N_0)}, \quad (5)
\]
where \(\gamma = -\text{Im} \Sigma(\omega \to 0)\) is the scattering rate for low-energy quasiparticles. For \(|\omega| \lesssim \gamma\), the density of states is determined by impurities and is finite: \(N_{\text{imp}}(0)/N_0 = 2\gamma/\pi \Delta_0\). The characteristic width of the impurity-dominated region is \(\omega^* \simeq \gamma \propto \sqrt{n_{\text{imp}}}\).

The origin of the finite density of states is the impurity band, grown from the impurity-induced bound states (consider \(c = 0\)). Scaling of the impurity bandwidth \(\gamma \propto \sqrt{n_{\text{imp}}}\) has been obtained earlier for the case of paramagnetic impurities in an \(s\)-wave superconductor \[7\]. The fact that \(\gamma \propto \sqrt{n_{\text{imp}}}\) is obeyed in the case of a \(d\)-wave superconductor with scalar impurities as well supports our claim that the low-energy states in a disordered \(d\)-wave superconductor are indeed formed from the bound states at finite concentration.

Here we shall comment on the condition for the formation of the impurity band with extended states, i.e., the Mott criterion. Consider the case of a (3D) \(s\)-wave superconductor. A magnetic impurity generates a bound state with the impurity wave function \(|\Psi_{\text{imp}}| \sim \exp(-\alpha r), \alpha \sim \xi^{-1}\) \[3,4\]. For the true conduction band to be formed, the overlap between localized states should be large enough. This leads to the Mott criterion for the minimum concentration \(n_{\text{imp}}^{1/3} \alpha^{-1} \geq 0.2\) \[14\]. In practice, for conventional superconductors this implies a very low critical concentration \(n_{\text{imp}}^c \sim (a/\xi)^3\) (\(a\) being the lattice constant), as the coherence length \(\xi\) is relatively large \[15\]. The situation is qualitatively different in a \(d\)-wave superconductor. The wave function of impurities has lobes sticking out in the directions of vanishing gap, i.e., when \(\cos 2\varphi = 0\) [see Eq. (1a)], and the overlap between impurities is larger along these directions. The Mott transition in a system of strongly anisotropic localized states is an interesting problem and it should be expected that the Mott criterion should be modified with a somewhat lower constant in the r.h.s. of \(n_{\text{imp}}^{1/2} \xi = \text{const}\) and it follows that \(n_{\text{imp}}^c \propto (a/\xi)^2 \sim 1\%,\) which is within the current experimental range. A short
coherence length of high-$T_c$ superconductors ($\xi \sim 20\text{Å}$) suggests the possibility of the experimental observation of the metal-insulator transition in the impurity band in a $d$-wave superconductor (assuming that localization effects are small).

In conclusion, we find that a strongly scattering potential impurity produces a resonant or a marginally bound state inside the gap in a $d$-wave superconductor. The wave function of the impurity bound state is highly anisotropic with $1/r$ decay along the nodes of the gap and exponential with angle-dependent decay range otherwise. These bound states change the local density of states $N_{\text{imp}}(r,\omega)$ dramatically, which could be probed experimentally, e.g., in NMR. The short coherence length of high-$T_c$ superconductors may lead to a finite critical concentration $n_{\text{imp}}^c$ within the range of a few percents and allow a direct observation of the metal-insulator transition in the impurity band.

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[10] For example, see K.Machida and F.Shibata, Prog. Theor. Phys. 47, 1817 (1972); and references therein. Note, however, that simple potential scattering does not induce bound states in s-wave superconductors [7].

[11] We assume that the energy gap has line nodes in three dimensions with weak quasiparticle dispersion along the z axis; an extension to a general three-dimensional case is
The related model of the Anderson impurity in an unconventional superconductor has been considered by L. Borkowskii and P. Hirschfeld, Phys. Rev. B\textbf{46}, 9274 (1992), and\textit{unpublished}. The results found here for pure potential scattering require the generalization of the Anderson model to include the impurity potential phase shift, independent of the Kondo temperature. This aspect of impurity scattering has not been addressed previously.

The change of the matrix elements and antiferromagnetic correlations will also result in the change of the normal state relaxation rate for Cu sites close to the impurity. These effects are, we believe, less important at low temperatures, where the drastic increase of the local density of states will dominate and lead to an increased relaxation rate at sites near the impurity.

See, for example, Chapter 4 of N.F. Mott, \textit{Metal-Insulator Transitions}, (Taylor and Francis, 1974, London).

For the experimentally relevant concentrations $n_{\text{imp}} \simeq 0.5 - 1\%$, the impurity band with “metallic” conductivity is well established.
FIGURES

FIG.1 The relative density of states $N(\omega)/N_0$ for the $d$-wave superconductor in the absence of impurities (thin solid line), the impurity-induced bound state for $c \to 0$ (thick solid line), and the virtual bound state for $c > 0$ (dashed line); $N_0$ is the normal-state density of states at the Fermi energy. The finite lifetime ($\Omega'' \neq 0$) of the virtual bound state in the $d$-wave superconductor results from the finite density of states $N(\omega) \propto \omega$ for small $\omega$ from nodal quasiparticles, in contrast to the true bound state in the $s$-wave superconductor. Exactly at $\omega = 0$, the density of quasiparticle states is zero and the virtual bound state becomes marginally bound on the edge of the particle-hole continuum.

FIG.2 The energy $\Omega = \Omega' + i\Omega''$ of the virtual bound state in the one-impurity problem, given by Eq. (2), as a function of impurity strength $c$: the shown quantities are $\Omega'$ (solid line), $\Omega''$ (dashed line), and $\Omega''/\Omega'$ (chain-dashed line). A spherical Fermi surface and $\Delta \vec{k} = \Delta_0 \cos 2\varphi$ have been assumed. The width $\Omega''$ of the virtual bound state is always smaller than its energy $\Omega'$ in the neighbourhood of unitary scattering. In contrast, for weak scattering $c \sim 1$, $\Omega'' \sim \Omega'$ and an impurity-induced virtual state does not exist. The inset shows a comparison between the exact result and the asymptotic approximation (dotted lines), as computed to logarithmic accuracy by Eq. (1) for $\Omega$. 

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