Impurity-induced virtual bound states in *d*-wave superconductors

A. V. Balatsky,^{*} M. I. Salkola, and A. Rosengren[†]

Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

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It is shown that a single, strongly scattering impurity produces a bound or a virtual-bound quasiparticle state inside the gap in a *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 density of impurities in a *d*-wave superconductor, we re-examine the growth of these states into an impurity band, and discuss the Mott criterion for the metal-insulator transition in this band.

Effects of impurities on the properties of superconductors have been investigated in great detail for lowtemperature,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 nonmagnetic. 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 (nonmagnetic) 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) formalism⁴ treats impurities in the Born approximation. Any impurity problem is characterized by two physical parameters: the phase shift δ_0 due to impurity scattering (which we assume to be s wave) and the density of impurities n_{imp} . In the AG approach, the only parameter entering the formalism is the scattering rate, $\bar{\tau}^{-1} = (2n_{\rm imp}/\pi N_0) \sin^2 \delta_0$, proportional to the product of the density 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 density of strong magnetic impurities, the AG approach will yield a small average scattering rate.⁵ 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 unitaryscattering limit with the s-wave phase shift $\delta_0 \simeq \pi/2$. 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 Ref. 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 nonmagnetic impurities are strong pair breakers in a nontrivial superconductor. The Zn substitutions in cuprates are one example of this.^{8,9} Although Zn ions are nominally nonmagnetic, T_c is strongly suppressed by Zn substitution of Cu in the planes.¹⁰ Therefore, it is reasonable to assume that Zn ions behave as nonmagnetic unitary scatterers.

Our purpose 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 generalize the original Yu-Shiba^{6,7} approach to arbitrary-strength nonmagnetic impurities in a $d_{x^2-y^2}$ superconductor. The results, summarized below, can be easily applied 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 nonmagnetic — 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 nonmagnetic impurity potential, resulting in a well-defined bound state in the unitary limit. This should be compared with the fact that, in *s*-wave superconductors, both magnetic and resonant nonmagnetic impurities produce bound states inside the energy gap.¹¹ (ii) The energy Ω' and the decay rate Ω'' of this state are given by

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

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 $\ln(8/\pi c) \gg 1$. It is only in this limit that the bound state is well defined. In the unitary limit, defined as $\delta_0 \rightarrow \pi/2$ $(c \rightarrow 0)$, the virtual bound state becomes a marginally bound one at $\Omega \rightarrow 0$ with $\Omega''/\Omega' \rightarrow 0$. 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) While generally one finds that an impurity band for quasiparticles is formed after averaging over impurity positions, disorder and quasiparticle correlations can lead to qualitatively new phenomena of which one example is a metal-insulator transition.

Single-impurity problem. Consider the single scalar impurity problem with $H_{\text{int}} = \sum_{\vec{k}\vec{k}'\sigma} V c^{\dagger}_{\vec{k}\sigma} c_{\vec{k}'\sigma}$, where V is the strength of the scalar impurity potential at $\vec{r} = 0$, resulting in s-wave phase shift δ_0 .

The scattering of quasiparticles from the impurity is described by a T matrix, $\hat{T}(\omega)$, which is independent of wave vector. The Green's function in the presence of an impurity is $\hat{G}_{\vec{k}\vec{k}'}(\omega) = \hat{G}_{\vec{k}}^{(0)}(\omega)\delta_{\vec{k}\vec{k}'} + \hat{G}_{\vec{k}}^{(0)}(\omega)\hat{T}(\omega)\hat{G}_{\vec{k}'}^{(0)}(\omega)$, where both $\hat{G}_{\vec{k}}^{(0)}(\omega)$ and $\hat{T}(\omega)$ are matrices in Nambu space. Here $[\hat{G}_{\vec{k}}^{(0)}(\omega)]^{-1} = \omega\hat{\tau}_0 - \Delta_{\vec{k}}\hat{\tau}_1 - \xi_{\vec{k}}\hat{\tau}_3$, 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 particlehole-spinor space.

From the previous analysis,^{2,4,7} it is known that $\hat{T} = T_0 \hat{\tau}_0 + T_3 \hat{\tau}_3$ for s-wave scattering and a particle-hole symmetric system. Therefore, the only relevant terms in the T matrix are $T_0(\omega) = G_0(\omega)/[c^2 - G_0(\omega)^2]$ and $T_3(\omega) =$



$$c = \pm G_0(\Omega), \tag{2}$$

which is an implicit equation for Ω 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 $G_0(\omega) = \int (d\varphi/2\pi)/\sqrt{[\Delta(\varphi)/\omega]^2 - 1}$.¹² For $|\omega| \ll \Delta_0$, we obtain

$$G_0(\omega) = \frac{2\omega}{\pi\Delta_0} \left(\ln \frac{4\Delta_0}{\omega} - \frac{i\pi}{2} \right).$$
(3)

In principle, the solution of Eq. (2) is complex, indicating a resonant nature of the quasiparticle state and 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 welldefined state bound to the impurity. Exact numerical solution of Eq. (2) as a function of c is shown in Fig. 2. As $c \to 1^-$, Ω' and Ω'' increase without bound so that $\Omega''/\Omega' \to 1^-$, and the solution becomes unphysical. For c > 1, no solution has been found for Ω .¹³ Indeed, in the Born limit $c \gg 1$, no resonance structure is generated and the density of states is modified only very weakly by the impurity potential $[\hat{T} = \mathcal{O}(c^{-1})]$.

There are important physical implications of these bound states in a d-wave superconductor. The most interesting case is unitary impurities in the dilute limit,



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 ω 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 Ω' (solid line), Ω'' (dashed line), and Ω''/Ω' (chain-dashed line). A spherical Fermi surface and $\Delta_{\vec{k}} = \Delta_0 \cos 2\varphi$ have been assumed. The width Ω'' of the virtual bound state is always smaller than its energy Ω' in the neighborhood 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 Ω .

separated by a distance greater than the coherence length ξ . 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 $N(\vec{r}, \omega) =$ $-\frac{1}{\pi} \operatorname{Im} G(\vec{r}, \vec{r}; \omega + i0^+)$, 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 $G(\vec{r},\vec{r}';\omega) = \sum_n \psi_n^*(\vec{r})\psi_n(\vec{r}')/(\omega - E_n)$, we find two terms in the local density of states $N(\vec{r},\omega) = N(\omega) +$ $N_{\rm imp}(\vec{r},\omega) = \sum_n \psi_n^*(\vec{r})\psi_n(\vec{r})\delta(\omega - E_n)$, for well-defined states. The first term originates from the bulk quasiparticles, which are described by plane-wave eigenstates with $E_{\vec{k}} = \sqrt{\xi_{\vec{k}}^2 + \Delta_{\vec{k}}^2}$, $G^{(0)}(\vec{r} = \phi, \omega) = \sum_{\vec{k}} [u_{\vec{k}}^2/(\omega - E_{\vec{k}}) + v_{\vec{k}}^2/(\omega + E_{\vec{k}})]$, where $u_{\vec{k}}$ and $v_{\vec{k}}$ are the standard Bogoliubov factors. In a *d*-wave superconductor, the bulk density of states is uniform with $N(\omega)/N_0 = \omega/\Delta_0$, for $\omega \ll \Delta_0$. The second term, $N_{\rm imp}(\vec{r},\omega) = -\frac{1}{\pi} {
m Im} \, [\hat{G}^{(0)}(\vec{r},\omega) \hat{T}(\omega) \hat{G}^{(0)}(-\vec{r},\omega)]_{11}$, originates from quasiparticle states created at the impurity: $N_{imp}(\vec{r}, \omega) = \sum_{n} \psi^*_{imp,n}(\vec{r}) \psi_{imp,n}(\vec{r}) \delta(\omega - E_{imp,n}).$ 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 $\hat{G}^{(0)}(\vec{r},\omega=0)\hat{G}^{(0)}(-\vec{r},\omega=0)$ is real only the imaginary part of the T matrix contributes to $N_{\rm imp}$ 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,

$$N_{
m imp}(\vec{r},\omega=0) \propto {
m Re} \, [\hat{G}^{(0)}(\vec{r},\omega=0)]_{11}^2 \propto r^{-2}, ~~(4{
m a})$$

and exponentially in the vicinity of the extrema of the gap function,

$$N_{\rm imp}(\vec{r},\omega=0) \propto [\xi(\varphi)/r] e^{-2r/\xi(\varphi)}, \tag{4b}$$

where $\xi(\varphi)$ is the angle-dependent coherence length of the superconductor, naturally defined as $\xi(\varphi) = \hbar v_F / |\Delta(\varphi)|$. 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 density. 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 $N_{\text{imp}}(\vec{r}, \omega) = \frac{1}{\pi} \sum_i F(\vec{r} - \vec{r_i}) \Omega_i'' / [(\omega - \Omega_i')^2 + \Omega_i''^2]$, where we have introduced the sum over different impurities, located at $\vec{r_i}$, and $F(\vec{r} - \vec{r_i}) = \psi^*_{\text{imp}}(\vec{r} - \vec{r_i}) \psi_{\text{imp}}(\vec{r} - \vec{r_i})$ 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 Zndoped 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.¹⁴ At finite impurity density (~ 2%), the volume-averaged density of states will have a finite limit at $\omega \to 0$, as follows from Eq. (4a). The relaxation rates of Cu atoms close to and away from an impurity will, therefore, have the same temperature dependence $(T_1T)^{-1} = \text{const}$, but will be of a different magnitude.

This behavior has been observed experimentally: Ishida *et al.*⁸ have measured two NMR relaxation rates for Cu in Zn-doped YBa₂Cu₃O_{7- δ}. 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 density. If one considers averaging over impurities, distinct NMR relaxation rates, arising from unequivalent sites, cannot be resolved;¹⁵ the local inhomogeneous aspect of the localized states is lost.

For practical purposes the distinction between the impurity bound states and the 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 quasiparticle states.

Finite density of impurities. Consider the growth of the impurity band with finite density of strongly scattering impurities. As was mentioned above, scalar (nonmagnetic) 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 Refs. 2 and 3). Here we will first repeat the usual arguments leading to the quasiparticle scattering rate and low-energy density of states and then point out our critique¹⁶ of why this approach may be misleading.

For finite impurity density, the self-consistent Green's function, averaged over impurity positions, obeys the Dyson equation $\langle \hat{G}_{\vec{k}}(\omega) \rangle^{-1} = \langle \hat{G}_{\vec{k}}^{(0)}(\omega) \rangle^{-1} - \hat{\Sigma}(\omega)$ with $\hat{\Sigma}(\omega) = n_{\rm imp}\hat{T}(\omega)$ in the single-site approximation. The T matrix is determined self-consistently with $\langle G_0(\omega) \rangle = \frac{1}{2\pi N_0} \sum_{\vec{k}} \operatorname{Tr} \langle \hat{G}_{\vec{k}}(\omega) \rangle \hat{\tau}_0$. As above [see the remark preceding Eq. (2)], only $\Sigma_0 \equiv \frac{1}{2} \operatorname{Tr} \hat{\Sigma} \hat{\tau}_0$ and $\Sigma_3 \equiv \frac{1}{2} \operatorname{Tr} \hat{\Sigma} \hat{\tau}_3$ are nonzero. The algebra is straightforward, and for unitary scattering yields $\gamma \simeq \sqrt{n_{\rm imp}(\Delta_0/\pi N_0)}$, where $\gamma = -\operatorname{Im} \Sigma_0(\omega \to 0)$ is the scattering rate for low-energy quasiparticles. For $\omega \lesssim \gamma$, the density of states is determined by impurities and is finite: $N_{\rm imp}(0)/N_0 = 2\gamma/\pi \Delta_0$. The characteristic width of the impurity-dominated region is $\omega^* \simeq \gamma \propto \sqrt{n_{\rm imp}}$.

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 $\gamma \propto \sqrt{n_{\rm imp}}$ has been obtained earlier for the case of para-

magnetic impurities in an s-wave superconductor.⁷ The fact that $\gamma \propto \sqrt{n_{\rm imp}}$ 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 density.

However, the self-consistent T-matrix approximation has strong limitations because it does not take into account that the single-impurity states are highly anisotropic and decay as a power law along diagonal directions. Furthermore, the slow decay along these directions will imply long-range impurity-impurity tunneling amplitudes for quasiparticles. These properties will modify qualitatively the conventional picture of disorderinduced localization (i.e., the Anderson localization).¹⁶

Finally, there is an intriguing possibility to observe a Mott-Hubbard transition in the impurity band. While the long-range Coulomb interaction is screened on the length scales of a mean-impurity separation, the quasiparticles experience a strong Coulomb repulsion near each impurity site. The strength of the Coulomb repulsion will be dictated by the renormalized μ^* ($k \sim \xi^{-1}$, $\omega \sim v_F \xi^{-1}$) at the scale of the coherence length ξ , which is the typical size of the bound state both in s-wave and d-wave superconductors. Two limiting cases can therefore occur: (i) $\mu^* \gg t$ (strong repulsion) and (ii) $\mu^* \ll t$ (weak repulsion), where t is the average impurities overlap integral. For strong repulsion and at low impurity densities, standard arguments lead to the Mott transition in the half-filled band. For weak repulsion, doubly occupied sites will be allowed and the Mott transition is unlikely to occur, in particular at realistic impurity densities. The superfluid condensate, suppressed in the vicinity of the impurity, cannot completely screen the Coulomb interaction between quasiparticles in the bound state. For the high- T_c superconductors, the coherence length is small (~ 20 Å) and the bare Coulomb repulsion will be substantial at this length scale, which suggests that the high- T_c superconductors belong to the first limiting case and might thus exhibit the Mott transition in the impurity band.

Assuming a strong quasiparticle-quasiparticle repulsion, the impurity band is half filled leading to an insulating behavior. In order to derive a condition for the formation of the impurity band with extended states —

- * Also at the Landau Institute for Theoretical Physics, Moscow, Russia.
- [†] Permanent address: Dept. of Theoretical Physics, Royal Institute of Technology, Stockholm, Sweden.
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the Mott criterion - consider first the case of a halffilled impurity band in a (three-dimensional) s-wave superconductor. A magnetic impurity at $\vec{r} = 0$ generates a bound state with a impurity wave function $|\psi_{\rm imp}(\vec{r})| \sim$ $e^{-\alpha r}$, $\alpha \sim \xi^{-1.6,7}$ 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 density $n_{\rm imp}^{1/3} \alpha^{-1} \ge 0.2$.¹⁷ In practice, for conventional superconductors this implies a very low critical density $n_{\rm imp}^c \sim (a/\xi)^3$ (a being the lattice constant), as the coherence length ξ is very large.¹⁸ 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. (4a)], 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 lower constant in the right-hand side of $n_{imp}^{1/2}\xi = \text{const.}$ The questions of localization and the Mott transition in the impurity band for the strongly anisotropic impurity states will be addressed in a forthcoming publication.¹⁶

In conclusion, we find that a strongly scattering impurity potential 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 $N_{\rm imp}(\vec{r},\omega)$ dramatically, which could be probed experimentally, e.g., in NMR. We also argue that the Anderson localization in a *d*-wave superconductor in the presence of strongly anisotropic impurity states qualitatively differs from that in a conventional superconductor with finite-range impurity states.

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purity scattering has not been addressed previously.

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