Local pairing at U impurities in BCS superconductors can enhance T_c

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We analyze here the role electrons on Anderson U impurities play in superconductivity in a metal alloy. We find that phonon coupling at impurities counteracts the traditional effects which dominate T_c suppression in the nonmagnetic limit. In some cases, we find that nonmagnetic impurities can enhance T_c. Qualitative agreement is found between the predicted increase and the experimental data for IV-VI degenerate semiconductors doped with Tl or In. In the Kondo limit, a Fermi-liquid analysis reveals that it is the enhancement in the density of states arising from the Kondo resonance that counteracts pair weakening. [S0163-1829(97)07946-0]

When a nonmagnetic Anderson-U impurity[1] is placed in a superconductor, two distinct mechanisms can operate to suppress the superconducting transition temperature, T_c. First, resonant scattering between the U impurity and the conduction electrons leads to a broadening of the impurity levels. Such broadening increases the amplitude for binding conduction electrons on the impurity thereby inhibiting pair formation.[2] Second, the on-site Coulomb repulsion leads to a weakening of the pairing interaction that keeps two electrons bound in a Cooper pair. As a result, T_c is suppressed.[3,4]

In the nonmagnetic limit, Kondo[5] impurities also lead to pair weakening. When $T<T_K$, the formation of a Kondo singlet state at each impurity quenches the local moment.[6] However, the conduction electrons forming the many-body resonance around each impurity are spin polarized. Consequently, conduction electrons of opposite spin experience a net Coulomb repulsion when they visit a Kondo impurity,[7] thereby weakening the pair interaction that holds a Cooper pair together.

In theoretical treatments of the pair-weakening effect,[8–11] it is generally assumed that electrons on the impurities do not participate in superconductivity. That this view might not be entirely consistent can be seen from the early work of Ratto and Blandin (RB).[3] Within an Anderson-U model in a BCS superconductor, Ratto and Blandin[3] showed that the Cooper pair amplitude on a U impurity is nonzero. Hence, electron pairs annihilated on a U-impurity reemerge in the conduction band as a Cooper pair. In addition, Suhl also suggested that local impurities should give rise to local regions of superconductivity.[12]

In this work, we consider explicitly phonon-induced pairing on nonmagnetic Anderson-U impurities in a BCS superconductor. First, we show that the phonon coupling constants involving the impurity are at least as large as $\lambda_{kk'}$, the standard phonon coupling constant for the Cooper pairs in the conduction band. As a result, such local processes can lead to an enhancement, relative to previous treatments[3,4,8] of T_c. While it is well-known that pure potential scattering can enhance T_c in low-T_c materials[13] through coupling to transverse phonon modes, the present work suggests that in the case of nonmagnetic U impurities, an additional channel is available to enhance T_c.

The starting point for our analysis is a collection of identical noninteracting (dilute limit) Anderson-U impurities

$$H_0=\sum_{k,\sigma}\epsilon_k a^\dagger_{k\sigma}a_{k\sigma}+\epsilon_d\sum_{i\sigma}a^\dagger_{i\sigma}a_{i\sigma}+\sum_{k,i\sigma}V_{ik}(a^\dagger_{k\sigma}a_{i\sigma}+a^\dagger_{i\sigma}a_{k\sigma})+U\sum_i n_{i\uparrow}n_{i\downarrow}. \quad (1)$$

In Eq. (1), ϵ_d is the defect energy of the impurity, V_{ik} the overlap integral between a band state with momentum k and the ith impurity, $a^\dagger_{k\sigma}$ creates an electron in the band, $a^\dagger_{i\sigma}$ creates an electron with spin σ on the ith impurity and $n_{i\sigma}=a^\dagger_{i\sigma}a_{i\sigma}$. In the Hartree-Fock approximation, each impurity level is broadened with a width $\Gamma=\rho_0\langle|V_{ik}|^2\rangle$ where ρ_0 is the density of states at the Fermi level. As a result of the hybridization of the localized level with electrons in the conduction band, the on-site Coulomb repulsion is felt by all electrons in the system. To include the pairing interactions in the superconducting state, we write the total Hamiltonian as $H=H_0+H_{\rm pair}$ where $H_{\rm pair}$ contains the BCS interactions among all the electrons:

$$H_{\rm pair}=\frac{1}{2}\sum_{k,k'}\lambda_{kk'}a^\dagger_{k\uparrow}a^\dagger_{-k\downarrow}a_{-k'\downarrow}a_{k'\uparrow}+\lambda_d\sum_i n_{i\uparrow}n_{i\downarrow}+\sum_{ik}\lambda_{ik}(a^\dagger_{i\uparrow}a^\dagger_{i\downarrow}a_{-k\downarrow}a_{k\uparrow}+\text{H.c.}), \quad (2)$$

where the λ's are determined by the electron-phonon interaction. The last two terms in Eq. (2) account for local pairing on the U impurity as well as scattering of Cooper pairs between the impurity and band states. In the nonmagnetic limit, this problem has been solved previously without the last two terms.[3,4,9]

It is instructive at the outset to establish the magnitude of the coupling constants in the last two terms in Eq. (2). To evaluate λ_d and λ_{ik} we expand the impurity states $|i\sigma\rangle=\Sigma_k\alpha_{i\sigma k\sigma}|k\sigma\rangle$ in terms of the k states, $|k\sigma\rangle$, in the band. In the expansion for the impurity states, we relied on the completeness of the k basis. If the bandwidth D is finite, the k states do not form a complete set. However, what is essential here is that the band contain the states $|\epsilon_k-\epsilon_F|<\Gamma$. As is typically done, we assume that the matrix ele-

ment $\lambda_{kk'}=\lambda_0$ is a constant for k states with $|\epsilon_k-\epsilon_F|$ less than ω_D, the Debye frequency of the metal. In the estimates that follow, we will assume that $\omega_D>\Gamma$. Using the standard form for the electron-phonon interaction, $V_{\rm ph}=\lambda_0 a^\dagger_{k+q\uparrow}a^\dagger_{k'-q\downarrow}a_{k'\downarrow}a_{k\uparrow}$, we find that

$$\lambda_d=\langle i\uparrow,i\downarrow|V_{\rm ph}|i\uparrow,i\downarrow\rangle$$

$$=\lambda_0\sum_{k,k',q}\alpha^*_{i\uparrow,k+q\uparrow}\alpha^*_{i\downarrow,k'-q\downarrow}\alpha_{i\uparrow,k\uparrow}\alpha_{i\downarrow,k'\downarrow}=\lambda_0\sum_q g(q).$$

From the orthogonality of the k states, it follows that $\Sigma_k|\alpha_{i\sigma k\sigma}|^2=1$; hence, $g(q=0)=1$. From the continuity of $g(q)$ it follows that $\lambda_d=\tilde{N}\lambda_0$, where $\tilde{N}$ is proportional to the number of electrons in the conduction band. Hence, the on-site phonon interaction for the impurity electrons, is enhanced over the k-state pairing value. Consequently, the effective on-site Coulomb repulsion is reduced to $\tilde{U}=U+\lambda_d$. Similarly, the scattering matrix element

$$\lambda_{ki}=\lambda_{ik}=\langle i\uparrow,i\downarrow|V_{\rm ph}|k\uparrow,-k\downarrow\rangle$$

$$=\lambda_0\sum_q\alpha^*_{i\uparrow,k+q\uparrow}\alpha^*_{i\downarrow,-k-q\downarrow}\approx\lambda_0$$

is also related to λ_0. An exact equality obtains if two conditions are true, namely $\langle x|d\rangle$ is real and $\alpha_{i\uparrow,k\uparrow}=\alpha_{i\downarrow,k\downarrow}$. As we will see, the presence of the mixing term λ_{ki} enhances the density of electron states participating in superconductivity. We will assume that λ_{ki} is a constant. Both effects, reduction of the on-site Coulomb repulsion and the enhancement of density of states at the Fermi level, play a positive role in superconductivity. *We show ultimately that they can conspire to increase T_c in the nonmagnetic limit.*

A simple way to make these heuristic arguments rigorous is through the Hartree-Fock decoupling of the Green function equations of motion method used by Ratto and Blandin (RB).[3] While more sophisticated methods exist,[11,14,15] the work of RB is sufficient to describe the nonmagnetic limit of the Anderson model. The linearized Hartree-Fock equations of motion for the creation operators can be written succinctly

$$[H,a^\dagger_{k\sigma}]=\epsilon_k a^\dagger_{k\sigma}+\sum_i V_{ik}a^\dagger_{i\sigma}-\Delta^\dagger_k a_{-k-\sigma},$$

$$[H,a^\dagger_{i\sigma}]=Ea^\dagger_{i\sigma}+\sum_k V_{ik}a^\dagger_{k\sigma}-\Delta_i a^\dagger_i \tag{3}$$

in terms of matrix elements of the gap,

$$\Delta_k=-\lambda_0\sum_{k'}\langle a_{k'\uparrow}a_{-k'\downarrow}\rangle-\sum_i\lambda_{ki}\langle a_{i\uparrow}a_{i\downarrow}\rangle,$$

$$\Delta_i=-\tilde{U}\langle a_{i\uparrow}a_{i\downarrow}\rangle-\lambda_{ki}\sum_{k'}\langle a_{k'\uparrow}a_{-k'\downarrow}\rangle. \tag{4}$$

The Hartree-Fock on-site energy is $E=\epsilon_d+\tilde{U}\langle n_i\rangle$, with $\langle n_i\rangle=\langle n_{i\uparrow}\rangle=\langle n_{i\downarrow}\rangle$. The presence of λ_{ki} causes the gap equations to become coupled. In fact, it is through this coupling that the single-particle density of states becomes enhanced.

Let us define $\eta=\lambda_{ki}/\lambda_0$ and introduce the Green functions $G(p,q;t)=-\langle T[a_{p\sigma}(t)a^\dagger_{q\sigma}(0)]\rangle$ and $F^\dagger(p,q;t)=\langle T[a^\dagger_{-p\downarrow}a^\dagger_{q\uparrow}(0)]\rangle$. Here p or q represent either a local impurity or a band state. In terms of the discrete frequencies $\omega=\omega_n\equiv(2n+1)\pi T$, the Fourier components of the Green functions are defined as $G(p,q;t)=T\Sigma_\omega e^{-i\omega t}G_\omega(p,q)$. The gap equations, Eq. (4), are then linear combinations

$$\Delta^\dagger_k=-\lambda_0 T\sum_\omega\left(\sum_{k'}F^\dagger_\omega(k,k')+\eta\sum_i F^\dagger_\omega(i,i)\right),$$

$$\Delta^\dagger_i=-T\tilde{U}\sum_\omega F^\dagger_\omega(i,i)-T\lambda_0\eta\sum_{k',\omega}F^\dagger_\omega(k',k') \tag{5}$$

of the $F^\dagger_\omega$ Green functions. The sum over k' in Eq. (5) is restricted over a momentum shell around the Fermi surface of width ω_D. Equation (5) must be solved to obtain T_c. To facilitate this, we introduce the Hartree-Fock approximation to the Hamiltonian in the normal metal, $\tilde{H}_0$, as well as the corresponding Green function, $\tilde{G}$. From the Hartree-Fock equation of motion, $(i\omega-\tilde{H}_0)\tilde{G}_\omega=1$ and the Gor'kov equations,[16] $(i\omega-\tilde{H}_0)\tilde{G}_\omega+\Delta F^\dagger_\omega=1$ and $(i\omega+\tilde{H})F^\dagger_\omega+\Delta^\dagger G_\omega=0$, it follows that to linear order in the gap, $F^\dagger_\omega(p,q)=\tilde{G}_{-\omega}(-\ell,-p)\Delta^\dagger_\ell\tilde{G}_\omega(\ell,q)$, where ℓ is summed over the k and i states. This approximation is valid at and slightly below the critical temperature T_c where the gap first appears. If we now substitute this expression into the self-consistent gap equations [Eq. (5)] and average over the random position of the impurities as well as average products of Green functions, we obtain a quadratic equation,

$$1+T_c\lambda_0\sum_{\omega,k}\left[\sum_{k'}S_\omega(k,k')+\eta n_s(S_\omega(k,i)+S_\omega(i,k))\right]+T_c\tilde{U}\sum_{\omega,j}S_\omega(i,j)$$

$$=T_c^2\lambda_0(\tilde{U}-\lambda_0\eta^2)\sum_{\omega,\omega',k,k'}\left[n_sS_\omega(i,k)S_{\omega'}(k',i)-\sum_j S_\omega(k,k')S_\omega(i,j)\right], \tag{6}$$

for the transition temperature where n_s is the impurity concentration. We have introduced the average $S_\omega(p,q) = \langle \widetilde{G}_\omega(p,q)\widetilde{G}_{-\omega}(-p,-q)\rangle_{\rm av}$. In obtaining Eq. (6) we decoupled the gap from the average of the product of Green functions.

To facilitate a solution for T_c, we note that the on-site repulsion $\widetilde{U}$ and the phonon coupling strength λ_0 are of quite different magnitudes. Typically, $\widetilde{U} \gg |\lambda_0|$. In this limit, Eq. (6) simplifies to an equation linear in the phonon coupling,

$$\frac{1}{|\lambda_0|} = T_c \sum_{\omega,k} \left[\sum_{k'} S_\omega(k,k') + 2\eta_{\rm eff} n_s S_\omega(k,i) - n_s T_c \widetilde{U}_{\rm eff} \sum_{\omega',k'} S_{\omega'}(k,i) S_\omega(i,k') \right], \tag{7}$$

where the subscript ''eff'' indicates division by $(1 + \widetilde{U} T_c \Sigma_{\omega,j} S_\omega(i,j))$.

The averages appearing in Eq. (7) can be evaluated straightforwardly following the ladder summation techniques. For example,[3]

$$\sum_{k,k'} S_\omega(k,k') \approx \frac{2\rho_0}{|\omega|} \tan^{-1}\frac{\omega_D}{|\omega|} - n_s \frac{\Gamma}{E^2 + (|\omega| + \Gamma)^2} + O(n_s^2). \tag{8}$$

The other averages are computed analogously. If we use these expressions for S_ω coupled with the standard BCS expression for the transition temperature, $(|\lambda_0|\rho_0)^{-1} = \ln(2e\gamma\omega_D/(\pi T_{c0}))$, we obtain that

$$\ln\frac{T_c}{T_{c0}} = n_s A \frac{\rho_d(\epsilon_F)}{\rho_0} [2\eta_{\rm eff} - 1 - A\rho_d(\epsilon_F)\widetilde{U}_{\rm eff}], \tag{9}$$

where

$$A = \ln(2\gamma\sqrt{E^2+\Gamma^2}/\pi T_{c0}) - \frac{\Gamma}{E}\tan^{-1}\frac{E}{\Gamma},$$

$$\eta_{\rm eff} = \frac{\eta}{1 + (U/\pi E)\tan^{-1}(E/\Gamma)}, \tag{10}$$

with γ the Euler-Mascheroni constant. The corresponding expression for $U_{\rm eff}$ can be obtained from Eq. (10) by replacing η with U. The local density on the impurity, $\rho_d(\epsilon_F) = \Gamma/(\pi(E^2+\Gamma^2))$, is given by the standard Lorentzian form.[1] Recall the η dependence arises from the scattering from a Cooper pair between the band and localized states. Also $\widetilde{U} < U$ as a result of the phonon coupling on U impurities. The importance of these terms should now be clear. When $\eta_{\rm eff} = 0$ and $\widetilde{U} = U$, the correction to T_c is precisely the negative correction of RB.[3] For $\eta \neq 0$ and $\widetilde{U} < U$, the transition temperature is enhanced relative to the predictions in earlier treatments of this problem.[8–11] In fact, we compare in Fig. 1 the predictions of the present theory for the initial slope of T_c with the earlier predictions of RB.[3] For modest values of Γ and $\widetilde{U}$, we find that nonmagnetic impurities can actually enhance T_c in contrast to the suppression indicative of pair weakening. The magnitude of the increase in T_c is of $O(n_s\epsilon_F/(n_0\Gamma))$, where n_0 is the conduction electron density.

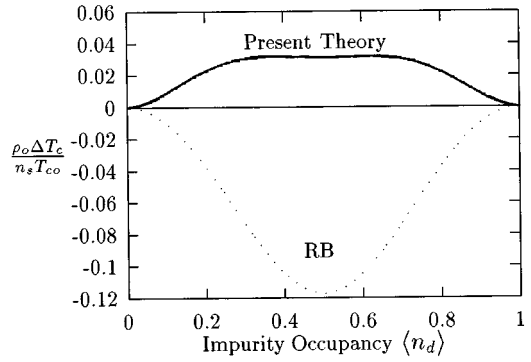

FIG. 1. Theoretical values for the initial slope of T_c predicted from Eq. (9) as a function of the filling, $\langle n_d \rangle$, on the impurity. ρ_0 is the density of states, n_s is the impurity concentration and $\Gamma = 2.0$ eV and $\widetilde{U} = 0.5$ eV.

Experimentally, T_c has been observed to increase when transition metals were doped into Ti.[17] Anderson[18] has suggested that transition metals such as Fe are nonmagnetic in Ti and hence might possibly increase T_c. While the present theory is consistent with the experimental trends, the agreement should not be taken as a confirmation because the experimental samples contained unusually high dopant concentrations.[17] Further experiments are needed on such samples in the dilute impurity regime to determine if nonmagnetic impurities do in fact increase T_c. However, in the context of degenerate semiconductors such as PbTe and SnTe doped with Tl and In, respectively, the observed superconductivity has been attributed to arise solely from impurity states.[19] In SnTe doped with In, T_c was increased by an order of magnitude with a 1% In-impurity level. More striking is the behavior in PbTe. In this material, superconductivity with a transition temperature of $T_c = 1-2$ K was observed only upon doping with Tl. Dopants such as Na yield no superconductivity down to temperatures of $T = 0.009$ K. Experimentally and theoretically,[20] it is now well-accepted that local-phonon coupling at the dopant impurities is largely responsible for superconductivity in these semiconductors. In addition, the impurities are thought to be in the extreme mixed-valence regime as the on-site repulsion is much less than the hybridization energy.[20] The large dielectric constant ($\epsilon^{\rm PbTe} \approx 33$) is primarily responsible for the lowering of the on-site Coulomb repulsion. For the experimentally relevant carrier concentrations and an impurity doping level of 1%, we estimate that $n_s/n_0 \sim 1$ and $\epsilon_F \sim 0.8$ eV. Also, Γ has been estimated[19] to range between 0.01 to 0.1 eV. For $\Gamma = 0.1$ eV, we estimate the magnitude of the relative increase in T_c to be $O(n_s\epsilon_F/(n_0\Gamma)) \approx 10$ which is qualitatively consistent with the increase seen experimentally.

We can extend this analysis to the nonmagnetic limit, $T < T_K$, of the Kondo problem. In this limit $\langle n_i \rangle = 1/2$. Below T_K, a Kondo system is described by a screened impurity in a Landau Fermi liquid with relatively weak quasiparticle interactions.[7] Sakurai[9] has shown that the nonmagnetic limit of the Hartree-Fock treatment of an Anderson impurity can be used to describe a Kondo system for $T < T_K$ by making the following transformation: (1) $\Gamma \to \Gamma/\widetilde{\chi}_{\uparrow\uparrow}$ and (2) $\widetilde{U}_{\rm eff} \to \Gamma^d_{\uparrow\downarrow} = \pi\Gamma\widetilde{\chi}_{\uparrow\downarrow}$. We have introduced the vertex function $\Gamma^d_{\uparrow\downarrow}$ for the inelastic scattering of a pair of d electrons of

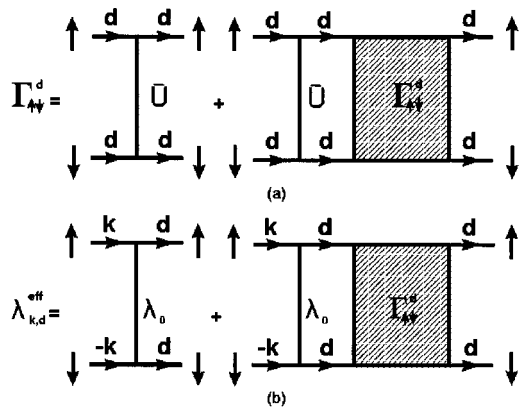

FIG. 2. (a) The vertex part of $\Gamma^d_{\uparrow\downarrow}$ in the ladder approximation. (b) The corresponding vertex for the scattering of a pair of k electrons into a pair of d electrons.

opposite spin. Below T_K, the susceptibilities are given by $\tilde{\chi}_{\uparrow\downarrow}=\tilde{\chi}_{\uparrow\uparrow}=\pi\Gamma/(4T_K)$. To calculate the transition temperature, we also need an expression for $\eta_{\rm eff}$. According to Eq. (9), η and U are rescaled in the same way. Hence, in the ladder approximation, the value of $\eta_{\rm eff}$ can be obtained by comparing two diagrams which correspond to $\Gamma^d_{\uparrow\downarrow}$ [see Fig. 2(a)], and the diagram for the scattering of a pair of k electrons into a pair of d electrons as shown in Fig. 2(b). We obtain that $\eta_{\rm eff}=\eta(\pi\Gamma)^2/(4T_K\tilde{U})$. Hence, the initial slope in T_c is

$$\frac{\Delta T_c}{T_{c0}}=\frac{n_s}{4\rho_0 T_K}A'\left(\frac{2\pi\Gamma\eta}{\tilde{U}}-A'\right), \tag{11}$$

where $A'=\ln(8\gamma(T_K/\pi^2T_{c0}))-1$.

To make contact with the Fermi-liquid picture of the Kondo problem, we rewrite this expression in the suggestive form

$$\frac{\Delta T_c}{T_{c0}}\approx\frac{n_s}{\rho_0 T_k}\ln\left(\frac{T_K}{T_{c0}}\right)\left(2\eta-\ln\left(\frac{T_K}{T_{c0}}\right)\right), \tag{12}$$

where we have used the fact that in the Kondo limit,[21] $\pi\Gamma=\tilde{U}=4T_K/w$ with w the Wilson number and we have dropped all irrelevant constants. Within the Fermi-liquid picture, $T_c\propto T_K\exp(\lambda^{-1})$ where λ is the dimensionless phonon coupling. In this expression, T_K replaces ω_D because electrons which are further away from the Fermi level than T_K are strongly scattered. In the presence of U impurities, there are two corrections to the dimensionless coupling constant λ. First, we must include the enhancement in the density of states arising from the Kondo resonance. This enhancement[7] scales as n_s/T_K. In addition, we must include the repulsion between quasiparticle states of opposite spin. The repulsion energy is essentially T_K below the Kondo temperature.[7,21] Within the quasiparticle picture, this repulsion is spread over $(\rho_0 T_K)^2$ states because there are two electrons participating in each scattering event. Hence, the change in the dimensionless coupling constant is given by[22]

$$\frac{\delta\lambda}{\lambda}=\frac{\delta\rho}{\rho_0}+\frac{\delta V}{V}=\frac{n_s}{\rho_0 T_K}+\frac{n_s}{\lambda\rho_0 T_K}. \tag{13}$$

However, $\Delta T_c=-T_c\delta\lambda/\lambda^2$. Consequently, the initial slope in T_c from the heuristic Fermi-liquid arguments

$$\frac{\Delta T_c}{T_{c0}}=\frac{n_s}{\rho_0 T_K}\ln\left(\frac{T_K}{T_{c0}}\right)\left(1-\ln\left(\frac{T_K}{T_{c0}}\right)\right) \tag{14}$$

is identical in form to the more exact expression derived in Eq. (12) because η is $O(1)$. The second term in both of these expressions is the standard pair-weakening effect, whereas the first is a positive correction arising from the enhancement in the density of states at a Kondo impurity. In the strong-coupling regime, $|\lambda|>1$, Eq. (13) predicts that Kondo impurities can enhance T_c. We conclude then that nonmagnetic impurities by virtue of local phonon pairing can counteract the standard T_c suppressing effects and in some cases actually enhance T_c. Experimental systems on which this prediction can be tested are the transition metal alloy Ti(Fe) and degenerate semiconductors.

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