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Gap states in dilute-magnetic-alloy superconductors: A quantum Monte Carlo study

M. Jarrell
Department of Physics, The Ohio State University, Columbus, Ohio 43210

D. S. Sivia
Theoretical Division and Manuel Lujan Jr. Neutron Scattering Center, The Los Alamos National Laboratory, Los Alamos, New Mexico 87545

B. Patton
Department of Physics, The Ohio State University, Columbus, Ohio 43210

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Using a combination of quantum Monte Carlo simulation, perturbation theory, and maximum-entropy analytic continuation, we calculate the density of states for a dilute-magnetic-alloy superconductor. We find that the peak location of the gap states moves more quickly with increasing Kondo temperature than predicted by Zittartz *et al.*, and that the integrated intensity of the gap states is in extreme qualitative disagreement with that prediction.

It is well known that magnetic impurities can have a profound effect on the properties of superconductors. They can severely reduce T_c, change the specific-heat jump, and even result in states within the superconducting gap. When the impurities have a strong antiferromagnetic coupling with the superconducting host, these gap states have a peaked structure. This effect was described for classical-spin impurities by Shiba,[1] and for quantum Kondo-type impurities by Zittartz, Bringer, and Müller-Hartmann[2] (ZBM). Experimentally, this structure can be seen in measurements of the differential conductance from tunneling experiments into dilute magnetic superconducting alloys.[3] Such structure is also seen when tunneling into normal dilute magnetic alloys which are proximity coupled to a pure superconducting material such as lead.[4]

Using a self-consistent Monte Carlo method, in conjunction with a method of analytic continuation, we are able to calculate the superconducting density of states of a dilute magnetic superconducting alloy. This approach has the advantage of treating the impurity exactly, and thus is more accurate than previous theories. In Fig. 1, we demonstrate the temperature dependence of the location of the states in the superconducting gap, and in Fig. 2 we show that a gap can appear in the impurity spectral function of an impurity embedded in a superconducting host.

Many theoretical attempts have been made to try to understand the gap states of dilute-magnetic-alloy superconductors, of which we will mention a few. Abrikosov and Gorkov,[5] using a second-order Born approximation appropriate in the weak-coupling limit, found that magnetic impurities could fill in the superconduction gap. However, they found no discernible structure of the states within the gap. Shiba,[1] using a classical impurity model, found that the states could have structure. However, in this approximation the location of the states within the gap is temperature independent. ZBM (Ref. 2) treat the impurity with the Nagaoka-Suhl approximation, and the interaction between the impurity and the superconducting host with a self-consistent t-matrix approximation. They find that the peak location of the gap states is given approximately by y_0,

$$y_0=\left[\frac{\ln^2(T_c/T_K)}{\ln^2(T_c/T_K)+\pi^2S(S+1)}\right]^{1/2}, \tag{1}$$

where T_c is the superconducting transition temperature,

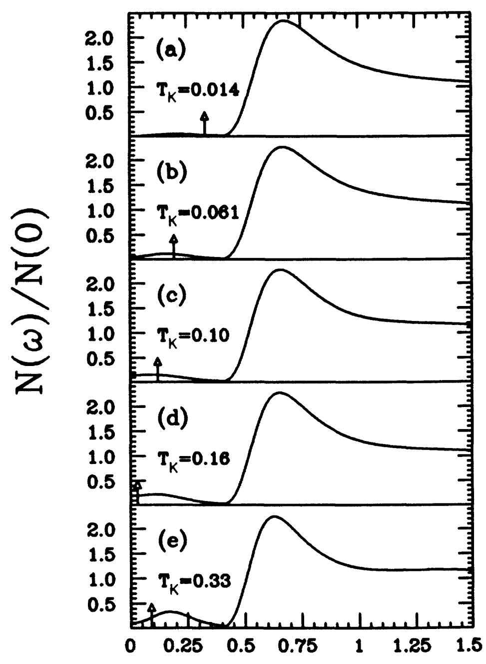

FIG. 1. Superconducting density of states for various values of T_K when $T=0.05$, $\lambda_0=2.8$, $\omega_0=0.75$, $\bar{c}=c/(2\pi)^2 \times N(0)T_{c0}=0.1$, and $T_c\approx T_{c0}=0.2$. $N(\omega)$ is normalized by the density of states at the Fermi surface of the normal metallic host $N(0)$. The arrow in each plot is the location of the gap states as predicted by Zittartz *et al.* [Eq. (1)]. Note that for large T_K the ZBM result disagrees with our result. Also, in contradiction to that predicted by ZBM, the integrated intensity of the gap states increases monotonically with T_K/T_{c0}.

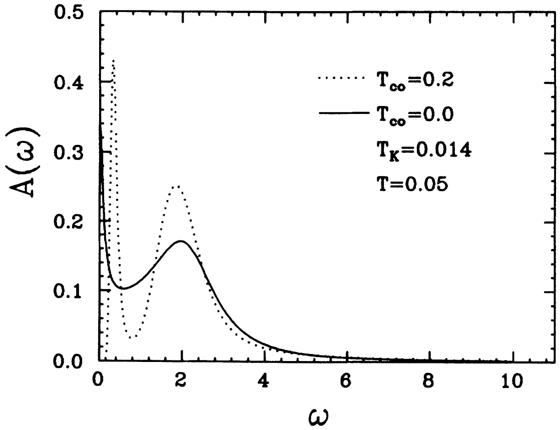

FIG. 2. The spectral function of the impurity site in a normal host (solid line) and in a superconducting host (dashed line) when $T_{c0}=0.2$, $T_K=0.014$, $\bar{c}=0.1$, and $T=0.05$. $A(\omega)=-(1/\pi)\mathrm{Im}G_d(\omega+i\delta)$ where G_d is the impurity single-particle Green's function. A gap opens in the spectral function of an impurity embedded in a dilute-magnetic-alloy superconductor when $T_K \ll T_{c0}$.

and S is the spin. In the ZBM theory both the shape and location of the gap states are entirely symmetric in $\ln(T_c/T_K)$. This symmetry is broken in the theory of Matsuura[6] which is an approximate interpolation scheme between the ZBM result at high temperatures $(T>T_K)$ and a Fermi-liquid theory at low temperatures $(T<T_K)$. However, to our knowledge only the pole location y_0 was calculated with this theory, not the superconducting density of states. In any case, a more fundamental approach is desirable.

We use a combination of quantum Monte Carlo and perturbation theory to obtain the host and impurity Matsubara Green's functions. These are analytically continued to real frequencies using the maximum-entropy method.

We model the magnetic impurities with a symmetric Anderson model in the limit of infinite metallic bandwidth. This model is characterized by a hybridization width $\Gamma=\pi N(0)V^2$ [where V is the hybridization matrix element, and $N(0)$ is the density of states at the Fermi surface], an on-site repulsion U, and a Kondo temperature T_K. In the limit $U \gg \Gamma$, a spin-$\frac{1}{2}$ magnetic moment forms on the impurity orbital. This moment couples antiferromagnetically to the conduction electrons with an exchange $J=-8\Gamma/\pi N(0)U$. We choose our definition of the Kondo temperature such that the Kondo resistivity at $T=T_K$ is half its maximum value, $\rho(T_K)/\rho(T=0)=\frac{1}{2}$. This is consistent with how T_K is defined in the Nagaoka-Suhl formalism used by ZBM. From a previous calculation[7] we found that

$$T_K \approx 0.91(1+\pi\Gamma/2U)\sqrt{2\Gamma U/\pi}\exp(-\pi U/8\Gamma)$$

when defined in this way.

We model the superconducting host with a Holstein model in which the conduction electrons interact with Einstein phonons with a coupling strength λ_0, and frequency ω_0, resulting in a transition temperature T_{c0} of the pure system. This model is well described by the Eliashberg eqautions.

We assume that a small finite concentration c of uncorrelated magnetic impurities are embedded in the superconducting host, but that the distance between the impurities is much smaller than the superconducting coherence length. This we believe is the physically interesting limit since, even at the 1% doping level, the impurities are tens of angstroms apart, whereas typical superconducting coherence lengths are thousands of angstroms. Thus, we will assume that the superconducting order parameter is spatially constant, and average over all possible spacial configurations of the impurities. This step restores momentum conservation, and allows us to use a standard perturbation theory to describe the superconducting state. In addition, in the dilute limit, the impurities are uncorrelated so that each impurity makes an independent contribution to the impurity diagrams. The net contribution is simply cN times the contribution of a single impurity (where N is the number of lattice sites), which is equivalent to using an average t-matrix approximation in the dilute limit.

We use a Nambu-Gorkov matrix representation for both the host and impurity Green's functions. Within this representation the Eliashberg equations are represented in Fig. 3(a). Here, Σ is the host self-energy, the double solid line is the host Green's function, the cross represents the hybridization matrix element V, and the triple dotted line is the fully dressed impurity propagator as determined from the Monte Carlo simulation. The Monte Carlo simulation requires as an input the impurity Green's function with $U=0$ (or equivalently, the Green's function corresponding to a constant Hubbard-Stratonovich field configuration), which is represented as the double dotted line. It is determined from the Dyson equation [Fig. 3(b)]. The Monte Carlo algorithm of Hirsch and Fye[8] is

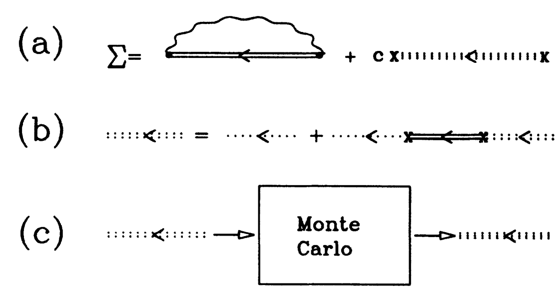

FIG. 3. Feynman graphs used. A Nambu 2×2 matrix form is used for all fermion Green's functions. The Eliashberg equations are represented in (a). The double solid line is the host Green's function, the X represents the hybridization matrix element V, c is the concentration of impurities, and the triple dotted line is the fully dressed impurity propagator as determined from the Monte Carlo simulation. The Monte Carlo simulation requires as an input the impurity Green's function with $U=0$ (or equivalently, a constant Hubbard-Stratonovich field configuration). It is determined from the Dyson equation, (b). The Monte Carlo simulation is then used to dress the double dotted line to all orders is U, (c), producing the fully dressed impurity Green's function which is the triple dotted line.

then used to dress the double dotted line to all orders is U [Fig. 3(c)], producing the triple dotted line which is the fully dressed impurity Green's function. The set of equations represented in Fig. 3 are iterated until convergence is reached, usually less than four complete iterations.

The host and lattice Green's functions must then be analytically continued to real frequencies in order to obtain the density of states and the impurity spectral function. We use the maximum-entropy method[9-11] to analytically continue our data. Starting with a default model (the solution to which the analysis will default in the absence of any data), it finds the real frequency image which has the most entropy and is in statistical agreement with the data. For the density of states, our default model is the density of states of the pure host superconductor, and for the impurity spectral function it is the perturbation-theory result of Horvatić, Sǒkčević, and Zlatić.[12] It should be noted that the resolution of our images (in this case the superconducting density of states) deteriorates rapidly with ω. This is due to the rapidly decreasing sensitivity of the Monte Carlo data to change in the density of states as ω increases. Therefore, whereas we are able to make quantitative statements about the approximate location and integrated intensity of the structure (but not the detailed shape) of the gap, we are unable to distinguish reliably features at frequencies above the gap. More details about the maximum-entropy method of analytic continuation, and its ability to propagate error, will be discussed in a future publication.[11]

Our results for the superconducting density of states are shown in Fig. 1 for various T_K when $T=0.05$, $T_{c0}=0.2$, $\lambda_0=2.8$, $\omega_0=0.75$, and $\bar{c}=c/(2\pi)^2N(0)T_{c0}=0.1$. The concentration of impurities is normalized in this way to be consistent with ZBM. The gap states develop and move toward the center of the gap as T_K/T_c increases from zero [Figs. 1(a) and 1(b)], have almost reached the center of the gap when $T_K\approx 0.5T_c$ [Fig. 1(c)], and then move back toward the center of the gap as T_K/T_c increases further [Figs. 1(d) and 1(e)]. Finally, when T_K/T_c becomes so large that the impurity becomes nonmagnetic ($U/\Gamma<\pi$), perturbation theory in U becomes exact and our results agree with those of Kaiser[13] (not shown). The arrow in each plot is the location of the gap states as predicted by ZBM [Eq. (1)]. This result roughly agrees with our calculations in the limit $T_c>T_K$ [in Fig. 1(a), the integrated intensity of the gap states was so small that locating the peak is not possible within error bars], which is to be expected since their result is based upon the Nagaoka approximation. The Nagaoka approximation, a self-consistent perturbation theory in J, is only correct in the high-temperature limit. For $T_c<T_K$, the states move as a function of T_K/T_c much more quickly than predicted by ZBM. This discrepancy is also seen in the tunneling experimental of Dumoulin *et al.*[4] They measure the tunneling conductance into normal dilute magnetic alloys proximity coupled to a pure-lead superconducting host. Here, T_K/T_c can be adjusted by changing the type of magnetic impurity, or the strength of the proximity coupling. They find that the states move more quickly with increasing T_K/T_c than that predicted by ZBM.[4,14] Nevertheless, Eq. (1) does a good job of predicting the qualitative behavior of the motion of the gap states with T_K/T_c.

ZBM also predict that the shape and intensity of the gap states should be symmetric in T_K/T_c for the same $\bar{c}$. However, we find that for $\bar{c}=0.1$ the integrated intensity of states within the gap increases monotonically with T_K/T_c. From Figs. 1(a)-1(e), the integrated intensity of the gap states increases roughly 5 times [0.015 ± 0.004, Fig. 1(a); 0.033 ± 0.008, Fig. 1(b); 0.044 ± 0.009, Fig. 1(c); 0.059 ± 0.012, Fig. 1(d); 0.081 ± 0.013, Fig. 1(e)]. Thus an experimental search for superconducting gap states might be more fruitful in materials for which $T_K>T_c$. In fact, this increase in prominence of the gap states with T_K/T_c has also been seen experimentally.[4]

We also analytically continued the impurity spectral function $A(\omega)$ for the impurity embedded in a superconducting host. As shown in Fig. 2, in a pure metallic host (the solid line) $A(\omega)$ has a central $\omega=0$ peak, and a high-frequency peak associated with charge transfer on the impurity site. In a superconducting host (the dashed line) the central peak can develop a gap. In a normal metal, the central peak signals the formation of a resonant singlet of energy approximately T_K which screens the impurity moment. In a superconductor, all the electrons within a gap frequency of the Fermi surface are paired into Cooper singlets. Spin-flip scattering of such electrons costs an energy of order of the gap energy; thus a gap opens in $A(\omega)$ if $T_K\ll T_c$. As $\bar{c}$ increases (and the superconducting gap is reduced) or as T_K increases, the gap in $A(\omega)$ disappears (not shown here). Thus the gap in $A(\omega)$ is only seen in dilute limit when $T_K\ll T_c$.

We note that this gap in the impurity spectrum could have experimental ramifications for NMR measurements of the nuclear relaxation rate $1/T_1\propto\chi''(\omega_n)/\omega_n$, where $\chi''(\omega)$ is the dynamic susceptibility of the impurity, and ω_n is the nuclear gyrofrequency. If a gap opened in $\chi''(\omega)/\omega$, then $1/T_1$ would be exponentially suppressed in a superconductor. Unfortunately, whenever we found a gap in $\chi''(\omega)$, it was not statistically significant within the propagated error. Hence we do not report results for $\chi''(\omega)$.

In conclusion, using a combination of self-consistent Monte Carlo, perturbation theory, and maximum-entropy analytic continuation, we have demonstrated the temperature dependence of the impurity states within the superconducting gap. We find that the peak location of the gap states is consistent with that predicted by ZBM when $T_K/T\ll 1$, but that the peak location moves more quickly with increasing T_K/T_c than that predicted by ZBM. We also find that the integrated intensity of the gap states is qualitatively different than that predicted by ZBM in that it is temperature dependent, and increases monotonically with T_K/T_c. Both the rapid motion of the gap states with T_K/T_c, and the temperature dependence of their integrated intensity are consistent with experimental observations.

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