Bethe Ansatz Approach to the Thermodynamics of Superconducting Magnetic Alloys

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We derive thermodynamic Bethe ansatz equations for a model describing a $U \to \infty$ Anderson impurity embedded in a BCS superconductor. The equations are solved analytically in the zero-temperature limit, $T = 0$. It is shown that the impurities depress superconductivity in the Kondo limit; however, at $T = 0$ the system remains in the superconducting state for any impurity concentration. In the mixed-valence regime, an impurity contribution to the density of states of the system near the Fermi level overcompensates a Cooper pairs weakening, and superconductivity is enhanced. [S0031-9007(98)05846-3]

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Since the pioneering work of Abrikosov and Gor'kov (AG) [1], the problem of superconducting magnetic alloys has been the subject of many early [2] and more recent [3] studies. Almost all of the theoretical methods developed to attack the Kondo problem in normal metals, from perturbative approaches to Wilson's numerical renormalization group, have been generalized to the case of superconductors. Perhaps the only exception is the Bethe ansatz (BA) technique, which solves the Kondo problem in normal metals exactly, but cannot be straightforwardly generalized to the case of dilute superconducting alloys. The basic theoretical models describing magnetic impurities in normal metals, such as the *s-d* (Kondo) and Anderson models, are integrable under *two additional conditions*: (i) An electron-impurity coupling is assumed to be energy independent, and (ii) a band electron dispersion E_k can be linearized around the Fermi level $E_k \simeq v_F(k - k_F)$, where k_F and v_F are the Fermi momentum and velocity, respectively [4]. Since a carrier dispersion in the superconducting state cannot be linearized near the Fermi level, these conditions eliminate superconductivity from the BA analysis of the behavior of magnetic alloys.

However, it has recently been discerned [5] that the basic "impurity" models of quantum optics, describing a system of Bose particles with a nonlinear dispersion coupled to two-level atoms, exhibit *hidden integrability* and are thus exactly diagonalized by BA. One of the most exciting potential applications of the approach developed may be an extension of the BA method to the Kondo problem in superconductors and other Fermi systems (e.g., gapless Fermi systems [6]) with an essentially nonlinear dispersion of charge carriers.

In this Letter, we employ hidden integrability of a model describing an Anderson impurity with an infinitely large Coulomb repulsion on an impurity orbital embedded in a BCS superconductor [7] to study the thermodynamic properties of the system. In the standard manner [4], we find a set of basic thermodynamic equations for the energies of elementary excitations of the system. In terms of these equations, we derive an exact equation for the order parameter of the superconducting phase transition [8], Δ, minimizing the thermodynamic potential of the system, Ω, with respect to Δ, $\delta\Omega/\delta\Delta = 0$. While at finite temperatures the basic equations require a numerical analysis, in the zero-temperature limit $T = 0$ they are solved analytically, giving an exact expression for the impurity contribution to the parameter Δ.

At $T = 0$, the order parameter is given by the expression $\Delta = \Delta_0 \exp(-\mu_{\text{imp}})$, where Δ_0 is the order parameter in the absence of impurities, and the parameter μ_{imp} [see Eq. (15)] describes the impurity contribution. The magnitude and sign of μ_{imp} are determined completely by the position of the impurity energy level ϵ_d with respect to the Fermi energy of the host metal ϵ_F. In the Kondo limit, where ϵ_d lies much below ϵ_F, μ_{imp} is positive, and hence magnetic impurities depress superconductivity. However, the system remains in the superconducting state at any concentration of impurities. In the mixed-valence regime, where ϵ_d lies near the Fermi level, the parameter μ_{imp} becomes negative. An impurity contribution to the density of states of the system near the Fermi level dominates over a Cooper pairs weakening, and the Anderson impurities enhance superconductivity.

Throughout this paper, the BA technique and many results of the exact solution of the Anderson model in normal metals are often used with no special references. All needed details can be found in excellent comprehensive reviews [4] and references therein.

We start with a Hamiltonian that includes the Hamiltonians of the BCS and Anderson models,

$$H = \sum_{k,\sigma} E_k a_{k\sigma}^{\dagger} a_{k\sigma} - \sum_k (\Delta a_{k\uparrow}^{\dagger} a_{k\downarrow}^{\dagger} + \Delta^* a_{k\downarrow} a_{k\uparrow}) + \Delta^2/g + \sum_{k,\sigma} v_k (a_{k\sigma}^{\dagger} d_\sigma + d_\sigma^{\dagger} a_{k\sigma}) + \epsilon_d \sum_\sigma d_\sigma^{\dagger} d_\sigma + U d_\uparrow^{\dagger} d_\uparrow d_\downarrow^{\dagger} d_\downarrow , \quad (1)$$

where we have used the standard spherical harmonic representation for band electron operators. The Fermi

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operator $a^\dagger_{k\sigma}$ creates a conduction electron with the momentum modulus k, spin $\sigma = \uparrow, \downarrow$, and the energy $E_k = \epsilon_k - \epsilon_F$, where ϵ_k is the kinetic energy. Only the s wave is assumed to be coupled to the impurity, therefore all other partial waves have been dropped. An electron localized in the impurity orbital is described by the Fermi operators d_σ. The fourth term of Eq. (1) represents the hybridization of the band and impurity level electrons with the matrix element v_k, while the Coulomb repulsion on the impurity orbital is described by the last term. The parameter $\Delta = g\sum_k \langle a_{k\downarrow} a_{k\uparrow}\rangle$ is assumed to result from the Cooper pairing phenomenon with positive coupling constant g.

Diagonalization of the BCS part of Eq. (1) by the Bogoliubov-Valatin unitary transform [9] gives

$$H = E_{\mathrm{BCS}} + \sum_{k\sigma} \omega_k c^\dagger_{k\sigma} c_{k\sigma} + v\sum_{k\sigma}(d^\dagger_\sigma c_{k\sigma} + c^\dagger_{k\sigma} d_\sigma) + \epsilon_d \sum_\sigma d^\dagger_\sigma d_\sigma + U d^\dagger_\uparrow d_\uparrow d^\dagger_\downarrow d_\downarrow , \tag{2}$$

where $\omega_k = -\sqrt{k^2 + \Delta^2}$ for $k < 0$, $\omega_k = \sqrt{k^2 + \Delta^2}$ for $k > 0$, and $E_{\mathrm{BCS}} = \sum_k (k - \omega_k) + \Delta^2/g$. For simplicity, we have linearized the band spectrum of the host metal in the normal state around the Fermi level and set $v_F = 1$. The electron momentum and energy are taken relative to the Fermi values. We have also set $k = k_F$ in both the hybridization matrix element $v = v(k_F)$ and in the coefficients of the unitary transform.

Moreover, to apply the BA method to the Hamiltonian (2), we have omitted the terms $d^\dagger_\sigma c^\dagger_{k\sigma}$ and $d_\sigma c_{k\sigma}$, which do not conserve the number of excitations in the system; these terms are assumed to lead only to insignificant corrections. The bare vacuum of the model is then defined by $c_{k\sigma}|0\rangle = d_\sigma|0\rangle = 0$. To obtain the ground state of the system in the absence of the impurity, one thus needs to fill all states with $k < 0$. In the normal state, $\Delta = 0$, Eq. (2) reduces to the integrable version of the Anderson model diagonalized by Wiegmann [10].

In what follows, we confine ourselves to very large values of U, $\epsilon_d + U > \mathcal{D}^{(+)}$, where $\mathcal{D}^{(+)}$ is the upper edge of the band, so that double occupancy of the impurity level is excluded. The eigenvalues of the model Hamiltonian (2) are then found from the following Bethe ansatz equations (BAE) [7]:

$$\exp(ik_j L)\frac{h_j - \epsilon_d/2\Gamma - i/2}{h_j - \epsilon_d/2\Gamma + i/2} = \prod_{\alpha=1}^{M} \frac{h_j - \lambda_\alpha - i/2}{h_j - \lambda_\alpha + i/2}, \tag{3a}$$

$$\prod_{j=1}^{N} \frac{\lambda_\alpha - h_j - i/2}{\lambda_\alpha - h_j + i/2} = -\prod_{\beta=1}^{M} \frac{\lambda_\alpha - \lambda_\beta - i}{\lambda_\alpha - \lambda_\beta + i}, \tag{3b}$$

where $E = \sum_j \omega(k_j)$ is the eigenenergy, N is the total number of particles in the interval L, and M is the number of particles with spin down. The function $h_j \equiv h(k_j)$ is defined by $h(k) = \frac{k}{2\Gamma} + \frac{\epsilon_d}{2\Gamma}(1 - \frac{k}{\omega(k)})$, where $\Gamma = \pi\rho v^2 = v^2/2$ and $\rho = 1/2\pi$ is the density of states of the host metal in the normal state.

In the thermodynamic limit, spin "rapidities" λ_α are grouped into bound spin complexes of size n,

$$\lambda^{(n,j)}_\alpha = \lambda^n_\alpha + i(n + 1 - 2j)/2, \qquad j = 1, \ldots, n . \tag{4}$$

The simplest bound states of charge excitations are associated with real spin rapidities λ_α, and their charge rapidities $k^{(\pm)}_\alpha$ are found from the equation

$$h(k^{(\pm)}_\alpha) = \lambda_\alpha \pm i/2 . \tag{5}$$

In the normal state, Eq. (5) has a single solution, $k^{(\pm)}_\alpha = 2\Gamma(\lambda_\alpha \pm i/2)$. Since $\Delta \ll \Gamma$, this solution acquires in the superconducting state a small correction of the order of $(\Delta/\Gamma)^2$,

$$k^{(\pm)}(\lambda) \simeq 2\Gamma(\lambda \pm i/2) - \frac{1}{2}\left(\frac{\Delta}{2\Gamma}\right)^2 \frac{\epsilon_d}{(\lambda \pm i/2)^2} . \tag{6a}$$

The energy of such "normal" charge complexes, $\omega_n(\lambda) = \omega(k^{(+)}) + \omega(k^{(-)})$, is found to be

$$\frac{\omega_n(\lambda)}{4\Gamma} \simeq \lambda + \frac{1}{2}\left(\frac{\Delta}{2\Gamma}\right)^2 \frac{\lambda - \epsilon_d/2\Gamma}{\lambda^2 + 1/4}, \tag{6b}$$

where the second term describes the "gap" correction. The sign of this correction is different for different λ, therefore the appearance of the energy gap can either increase or decrease the total energy of normal charge complexes. In what follows, this fact will play a crucial role in the interplay between the magnetic and superconducting properties of the system.

Equation (5) also admits gap charge complexes with rapidities $p^{(\pm)}_\alpha = \pm i\Delta \cos z^{(\pm)}_\alpha$ and the energy $\omega_g(\lambda_\alpha) = 2\Delta \mathrm{Re}\, \sin z_\alpha$. Here,

$$z^{(+)}(\lambda_\alpha) = z^{(-)*}(\lambda_\alpha) = \arctan \frac{-i\epsilon_d/2\Gamma}{\lambda_\alpha - \epsilon_d/2\Gamma + i/2}, \tag{7}$$

and the terms of order Δ/Γ are omitted. In what follows, we consider the case of negative ϵ_d. The gap complexes can then be shown to exist only for $\lambda > \epsilon_d/2\Gamma$, and their energies are positive, $\omega_g > 0$. Bethe ansatz equations admit also "long" charge complexes associated with spin complexes (4). We, however, do not consider such excitations, because, as in the normal state, they do not contribute to the low-temperature thermodynamics of the system. Finally, the subgap spectrum of the model contains a discrete mode (DM) [7], which is naturally treated as a particle-impurity bound state. In the BA approach, a DM is first found as a solution of a single particle problem rather than as a multiparticle discrete mode predicted by Shiba [11]. It can be shown that for $\epsilon_d < 0$ the renormalized energy of a DM in the multiparticle spectrum of the model (2) is much bigger than Δ, and hence a DM does not contribute to the thermodynamics of the system in the temperature range of

physical interest $T \leq T_c$, where T_c is the superconducting critical temperature.

In the standard manner, we then find in the thermodynamic limit a set of equations for the renormalized energies of the elementary excitations $\varepsilon(k)$, $\xi(\lambda)$, and $\kappa_n(\lambda)$, corresponding to unpaired charge excitations with real k_j, normal charge complexes and spin complexes, respectively:

$$\varepsilon(k) = \omega(k) + \int_{-\infty}^{\infty} d\lambda a_1[h(k) - \lambda]F[-\xi(\lambda)] + \int_{\epsilon_d/2\Gamma}^{\infty} d\lambda a_1[h(k) - \lambda]F[-\eta(\lambda)] - \sum_{n=1}^{\infty}\int_{-\infty}^{\infty} d\lambda a_n[\lambda - h(k)]F[-\kappa_n(\lambda)], \quad (8a)$$

$$\xi(\lambda) = \omega_n(\lambda) + \int_{-\infty}^{\infty} dk h'(k) a_1[\lambda - h(k)]F[-\varepsilon(k)] + \int_{-\infty}^{\infty} d\lambda a_2(\lambda - \lambda')F[-\xi(\lambda')] + \int_{\epsilon_d/2\Gamma}^{\infty} d\lambda a_2(\lambda - \lambda')F[-\eta(\lambda')], \quad (8b)$$

$$F[\kappa_n(\lambda)] = \sum_{m=1}^{\infty}\int_{-\infty}^{\infty} d\lambda A_{nm}(\lambda - \lambda')F[-\kappa_m(\lambda')] + \int_{-\infty}^{\infty} dk h'(k) a_n[\lambda - h(k)]F[-\varepsilon(k)]. \quad (8c)$$

The renormalized energy of gap complexes is given by

$$\eta(\lambda) = \omega_g(\lambda) + \xi(\lambda) - \omega_n(\lambda), \qquad \lambda > \epsilon_d/2\Gamma. \quad (8d)$$

Here, $F[f(x)] \equiv T\ln[1 + \exp(f(x)/T)]$, $h' = dh/dk$, $a_n(x) = (2n/\pi)(n^2 + 4x^2)^{-1}$, and $A_{nm}(x) = \delta_{nm}\delta(x) + (1 - \delta_{nm})[a_{|n-m|} + a_{n+m} + 2\sum_{k=1}^{\min(n,m)-1} a_{|n-m|+2k}(x)]$. The thermodynamic potentials of the host superconductor Ω_h and the impurity Ω_i are found to be

$$\frac{\Omega_h}{L} = \frac{E_{\rm BCS}}{L} - 2\int_{-\infty}^{\infty}\frac{dk}{2\pi}F[-\omega(k)], \quad (9a)$$

$$\Omega_i = 2\epsilon_d - \xi(\epsilon_d/2\Gamma). \quad (9b)$$

Therefore, the equation for the order parameter $\delta(\Omega_h + \Omega_i)/\delta\Delta = 0$ takes the form

$$1 = \frac{gL}{2}\int\frac{dk}{2\pi}\frac{\tanh[\omega(k)/2T]}{\omega(k)} - \frac{g}{2\Delta}\frac{\delta\Omega_i}{\delta\Delta}, \quad (10)$$

where the first term is the standard BCS term, while the second term describes the impurity contribution. The low-temperature thermodynamics of the $U \to \infty$ Anderson impurity embedded in a BCS superconductor is thus described completely by Eqs. (8)–(10).

At finite temperatures, the thermodynamic BA equations require a numerical analysis. However, at $T = 0$ they are significantly simplified and can be solved analytically. Indeed, one can show that the energies $\varepsilon(k)$, $\eta(\lambda)$, and $\kappa_n(\lambda)$ are positive. Therefore, as in the normal alloys, the ground state of the system contains only normal charge complexes and is described by a single equation for the function $\xi(\lambda) = \xi_-(\lambda)\theta(Q - \lambda) + \xi_+(\lambda)\theta(\lambda - Q)$, where Q is a single zero of the function $\xi(\lambda)$, $\xi(Q) = 0$,

$$\xi(\lambda) = d(\lambda) + \int_Q^{\infty} d\lambda' R(\lambda - \lambda')\xi_+(\lambda'). \quad (11a)$$

Here, $R(x) = \int(d\omega/2\pi)\exp(-i\omega x)/(1 + \exp|\omega|)$, and

$$d(\lambda) = \omega_n(\lambda) - \int_{-\infty}^{\infty} d\lambda' R(\lambda - \lambda')\omega_n(\lambda'). \quad (11b)$$

Equations (11) determine completely the physical properties of the ground state of the system, but for our purposes it is more convenient to derive equations describing the ground state directly from BAE. Introducing the "particle" and "hole" densities of normal charge complexes, $\sigma(\lambda) = 0$, $\lambda > Q$, and $\tilde{\sigma}(\lambda) = 0$, $\lambda < Q$, respectively, we find in the continuous limit of BAE,

$$\frac{1}{2\pi}\frac{dk(\lambda)}{d\lambda} + \frac{1}{L}a_2(\lambda - \epsilon_d/2\Gamma) = \int_{-\infty}^{Q} d\lambda' a_2(\lambda - \lambda')\sigma(\lambda') + \sigma(\lambda) + \tilde{\sigma}(\lambda), \quad (12)$$

where $k(\lambda) = k^{(+)}(\lambda) + k^{(-)}(\lambda)$ is the momentum of complexes. The functions $\sigma(\lambda)$ and $\tilde{\sigma}(\lambda)$ are divided into the host and impurity parts, i.e., $\sigma(\lambda) = \sigma_h(\lambda) + L^{-1}\sigma_i(\lambda)$, $\tilde{\sigma}(\lambda) = \tilde{\sigma}_h(\lambda) + L^{-1}\tilde{\sigma}_i(\lambda)$. The gap-dependent part of the impurity energy is then given by

$$E_i(\Delta) = \frac{\Delta^2}{2\Gamma}\Phi, \qquad \Phi = \int_{-\infty}^{Q} d\lambda\phi(\lambda)\sigma_i(\lambda), \quad (13a)$$

where $\phi(\lambda) \equiv (\lambda - \epsilon_d/2\Gamma)/(\lambda^2 + 1/4)$, while the function $\sigma_i(\lambda)$ is determined by the same equation as in the normal state,

$$\sigma_i(\lambda) + \tilde{\sigma}_i(\lambda) = R(\lambda - \epsilon_d/2\Gamma) + \int_Q^{\infty} d\lambda' R(\lambda - \lambda')\tilde{\sigma}_i(\lambda'). \quad (13b)$$

At $T = 0$, Eq. (10) reads

$$1 = \frac{gL}{2\pi}\int_0^{\omega_D}\frac{dk}{\sqrt{\Delta^2 + k^2}} - \frac{g}{2\Delta}\frac{\delta E_i}{\delta\Delta}, \quad (14a)$$

where ω_D is the Debye frequency. By inserting the well-known solution of the Wiener-Hopf equation (13b) into Eq. (13a), we finally obtain

$$E_i(\Delta) = -\frac{\Delta^2}{2\Gamma}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\frac{\phi(-\omega)\exp(i\omega Q)}{G^{(-)}(\omega)} \times \int_{-\infty}^{\infty}\frac{d\omega'}{2\pi i}\frac{R(\omega')G^{(-)}(\omega')}{\omega' - \omega + i0}\exp(i\omega'\epsilon_d^*/2\Gamma). \quad (14b)$$

Here, $\phi(\omega)$ and $R(\omega)$ are the Fourier images of the functions $\phi(\lambda)$ and $R(\lambda)$, and $\epsilon_d^* = \epsilon_d - 2\Gamma Q$

is the renormalized impurity level. The functions $G^{(+)}(\omega) = G^{(-)}(-\omega)$, $G^{(-)}(2\pi\omega) = \sqrt{2\pi} \times [(i\omega + 0)/e]^{i\omega}/\Gamma(1/2 + i\omega)$ are analytical functions in the upper $(+)$ and lower $(-)$ half-planes.

Equations (14) determine explicitly the order parameter at $T = 0$. Neglecting a small gap correction to the normal value of $Q = -(1/2\pi)\ln(\mathcal{D}^{(+)}/\Gamma)$, one easily finds the solution of Eq. (14a) in the multi-impurity case

$$\Delta = \Delta_0 \exp(-\mu_{\rm imp}) . \tag{15}$$

The parameter $\mu_{\rm imp} = (c_i \epsilon_F/\Gamma)\Phi$, which is proportional to the impurity concentration c_i, describes the impurity contribution.

The qualitative behavior of the impurity contribution is clearly seen without a detailed study of the integral in Eq. (14b). Let us first consider the Kondo limit of the problem, where the renormalized impurity level lies much below the Fermi energy, $-\epsilon_d^*/2\Gamma \gg 1$. The function $\phi(\lambda)$ is then positive on an essential interval of integration in Eq. (13a), $\epsilon_d/2\Gamma < \lambda < Q$, and hence the parameter $\mu_{\rm imp}$ is also positive. The asymptotic estimate of the integral (14b) gives $\mu_{\rm imp} \simeq c_i \epsilon_F/|\epsilon_d|$. Thus, in the Kondo limit, magnetic impurities depress superconductivity.

If the impurity level is shifted to the vicinity of the mixed-valence regime $|\epsilon_d^*|/2\Gamma \leq 1$, the parameter $\mu_{\rm imp}$ changes the sign at some point $\epsilon_d = \tilde{\epsilon}_d < Q$. At $\epsilon_d > \tilde{\epsilon}_d$, $\mu_{\rm imp}$ is negative, and hence the Anderson impurities, which in the mixed-valence regime play the role of a nonmagnetic resonance energy level rather than that of a local magnetic moment, enhance superconductivity.

In summary, making use of the BA approach, we have derived the exact equations describing the low-temperature thermodynamics of the model (2). We have also derived an equation for the order parameter Δ, minimizing the thermodynamic potential of the system with respect to Δ [12]. Finally, at $T = 0$ we have evaluated the impurity part of the total energy of the system, and thus found an exact zero-temperature expression for the order parameter.

The results obtained have a clear physical meaning. The ground state of a normal magnetic alloy is well known to be composed of the charge complexes [4]. The appearance of a superconducting energy gap results in a gap correction to the energy of these complexes, and hence to the impurity part of the total energy of the system. The sign of the impurity contribution to an energy balance, $\delta E_i/\delta\Delta$, is different in the Kondo and mixed-valence regimes, leading, respectively, to either a depression or an enhancement of superconductivity. Equation (15) shows that at $T = 0$ the system remains in the superconducting state at any impurity concentration. Because of the well-developed Kondo screening of a local magnetic moment of impurities, the system exhibits a Cooper pairs weakening rather than a pairs breaking predicted by the AG theory [1], not accounting for the Kondo effect. Moreover, in the mixed-valence regime, an impurity contribution to the density of states of the system near the Fermi level even dominates over a Cooper pairs weakening, and superconductivity is enhanced.

Finally, it should be emphasized that the results obtained do not contradict a concept of gapless superconductivity suggested by Abrikosov and Gor'kov [1]. At $T = 0$ an energy gap in the spectrum of the system may vanish at some critical impurity concentration, while an order parameter along with the parameter Δ remains finite. The BA technique admits an analytical computation of an energy gap at $T = 0$ that can clarify this very important and interesting question.

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