Magnetic Impurities and Superconductivity below the Kondo Temperature. Nagaoka's Model*

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The Green's-function method and decoupling scheme due to Nagaoka for dilute magnetic alloys in the normal state is extended to include superconductivity. A general expression for the superconducting critical temperature T_c is found for low-impurity concentrations n_I. This expression involves unknown parameters which occur in Nagaoka's equations for the normal state. When $T_c \gg$ the Kondo temperature T_K, the expression for the initial change in T_c with respect to n_I contains two terms: (i) the decrease in T_c of Abrikosov and Gor'kov as corrected by Maki and Griffin for the Kondo anomaly, and (ii) a change in T_c due to the polarization of the impurity spin which depends on the sign of the coupling constant J of the s-d exchange interaction. To order J^3, the term (ii) corresponds to the Solyom and Zawadowski's result. These results are obtained by use of Nagaoka's solution of his equations in the range $T_c \gg T_K$. A solution of Nagaoka's equations due to Hamann is next used to find an expression for T_c valid for all T_K. This expression is analyzed and compared with experimental data in the range $T_c \ll T_K$.

1. INTRODUCTION

IN a recent letter,[1] referred to as I, it was shown that the decrease in superconducting critical temperature T_c in a dilute magnetic alloy may be due to an anomalous resonance at the Fermi level when $T_c \ll T_K$, T_K being the Kondo temperature.[2] The interacting Hamiltonian was taken to be the s–d exchange interaction between the spin of the conduction electrons and the impurity spin.[2] The Green's-function method used to obtain an equation for T_c was similar to that of Takano and Ogawa.[3] There are many objections to this method, in particular the nonconservation of the number of electrons and the requirement of off-diagonal long-range order in the normal state. However, the equation for T_c so obtained seemed reasonable in the light of Nagaoka's approximate solution[4] for the normal state of dilute magnetic alloys at temperatures $T \ll T_K$. The purpose of this paper is to improve the results of I by extending Nagaoka's Green's-function method[4] to include superconductivity. The reason is that Nagaoka's decoupling scheme for the higher-order Green's functions does not suffer from the difficulties of Takano and Ogawa[3] and the resultant equation for T_c will be correct for all T_K. The equation for T_c obtained in I is only useful when $T_c \ll T_K$.

Nagaoka's method[4,5] has been subjected to a large amount of careful analysis recently. In his first paper[4], Nagaoka obtains an expression for the one-electron Green's function involving two unknown averages (see Sec. 2) which must be determined self-consistently. He first attempts a perturbation expansion of the Green's function in powers of the coupling constant J of the s–d exchange interaction at temperatures $T > T_K$. The residual resistivity calculated from the approximate Green's function is found to disagree with the expression obtained by Abrikosov.[6] For $T \ll T_K$, Nagaoka solves the self-consistent equation for the averages by an ansatz which lead to a resonance at the Fermi level with a temperature-dependent width $\Gamma(T)$. $\Gamma(T)$ is the solution of a nonlinear integral equation. Analytically this solution corresponds to the description of the conduction electron self-energy or t matrix by a single pole. Hamann[7] reduces Nagaoka's equations to a single nonlinear integral equation for the t matrix $t(\omega)$ and is able to find an approximate solution for $t(\omega)$ valid at all temperatures. For $T \gg T_K$ this solution gives rise to the same expression for the resistivity that Abrikosov[6] obtains. For $T \ll T_K$ Hamann's solution for $t(\omega)$ does not have the same analytic structure as Nagaoka's solution. In fact, the only similarity between the two solutions is that both give rise to the complete unitarity limit for s-wave scattering at zero temperature. Hamann further predicts that there is almost complete cancellation of the magnetic moment on the impurity at zero temperature for a spin-$\frac{1}{2}$ impurity. He also states that the contact s–d exchange interaction may not be applicable to magnetic impurities of spin greater than $\frac{1}{2}$. In consequence, we restrict our considerations to spin-$\frac{1}{2}$ impurities in this paper. Similar work to Hamann's has been done by Nagaoka,[5] for $T \gg T_K$, Falk and Fowler,[8] and Fischer.[9] Bloomfield and Hamann have been able to solve Nagaoka's equations exactly and are in the process of investigating the thermodynamic

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[5] Y. Nagaoka, Progr. Theoret. Phys. (Kyoto) **37**, 13 (1967).
[6] A. A. Abrikosov, Physics **2**, 5 (1965).
[7] D. R. Hamann, Phys. Rev. **158**, 570 (1967).
[8] F. Falk and M. Fowler, Phys. Rev. **158**, 570 (1967).
[9] K. Fischer, Phys. Rev. **158**, 567 (1967).

properties of dilute magnetic alloys in the normal state on the basis of this solution.[10]

As mentioned above, the Hamiltonian used to describe the interaction between the conduction electrons and a magnetic impurity of spin $\frac{1}{2}$ is the s–d exchange Hamiltonian.[2] Superconductivity is introduced into the formalism by the inclusion of the BCS interaction in the total Hamiltonian. In Sec. 2, the Green's-function formalism of Zubarev[11] is described and the equations of motion of the one-particle Green's functions are written down. The extension of Nagaoka's decoupling scheme[4] to superconductivity is discussed and used to obtain a closed form of the equations. Expressions for the one-particle Green's functions of the system are then written down in terms of four unknown averages which must be determined self-consistently. One of these averages is the order parameter Δ of the superconducting state. In Sec. 3 the equations of Sec. 2 are linearized with respect to Δ. A general expression for the superconducting critical temperature T_c is obtained in terms of the solutions of Nagaoka's equation in the normal state.

Nagaoka's approximate solutions of his equations[4,5] are substituted into this expression in Sec. 4(a) for $T_c \gg T_K$. Then the expression for the change in T_c with respect to the concentration of impurities n_I contains two terms. The first term represents a decrease in T_c due to spin-exchange scattering by a localized moment and corresponds to the results of Abrikosov and Gor'kov,[12] Griffin,[13] and Maki.[14] The second term gives rise to an increase in T_c for $J<0$. To order J^3 this term is identical to the result of Solyom and Zawadowski[15] who ascribe the increase in T_c to the virtual polarization of the impurity spin. Hamann's solution[7] for the t matrix is substituted into the general expression for T_c in Sec. 4(b). The resultant equation for T_c is valid for all temperatures and gives a decrease in T_c with n_I when $T_c \ll T_K$. Section 5 contains a comparison of the results of Sec. 4 with experimental data and a discussion of further extensions.

2. THE GREEN'S-FUNCTION FORMALISM FOR DILUTE MAGNETIC ALLOYS IN THE SUPERCONDUCTING STATE

We begin by considering a single spin-$\frac{1}{2}$ impurity in a metal matrix. The impurity position is taken as the origin of configuration space and the metal is assumed to be an isotropic superconductor with transition temperature T_{c0} in the absence of impurities. The Hamiltonian for the system is given by

$$H = H_0 + H_{sd} + H_{\mathrm{BCS}}. \tag{2.1}$$

H_0 is the unperturbed Hamiltonian for the conduction electrons and is written

$$H_0 = \sum_{k,\sigma} \epsilon_k C_{k\sigma}^{\dagger} C_{k\sigma}. \tag{2.2}$$

$C_{k\sigma}^{\dagger}$ and $C_{k\sigma}$ are creation and destruction operators for a conduction electron of momentum k, energy ϵ_k, and spin σ. ϵ_k is measured from the Fermi level of the pure metal. H_{sd} is the s–d exchange interaction between the conduction electron and impurity spins and is given by

$$H_{sd} = -(J/2N) \sum_{kl} \{S_Z(C_{k\uparrow}^{\dagger} C_{l\uparrow} - C_{k\downarrow}^{\dagger} C_{l\downarrow}) + S_- C_{k\uparrow}^{\dagger} C_{l\downarrow} + S_+ C_{k\downarrow}^{\dagger} C_{l\uparrow}\}. \tag{2.3}$$

S_Z and $S_{\pm}$ are the components of the spin operator of the impurity, N is the total number of atom in the crystal and J is the coupling constant of the s–d exchange interaction. H_{BCS} is the model Hamiltonian of BCS[16] which describes the superconducting state of the conduction electrons and is written

$$H_{\mathrm{BCS}} = -|g| \sum_{k,k'} a_{k\uparrow}^{\dagger} a_{-k\downarrow}^{\dagger} a_{-k'\downarrow} a_{k'\uparrow}. \tag{2.4}$$

g is the coupling constant of the BCS interaction and is always negative.

Let $A(t)$, $B(t)$ be two fermion operators in the Heisenberg representation. Then the retarded double-time temperature Green's function associated with A and B is written[11]

$$G_{AB}(t) = \langle\langle A \mid B\rangle\rangle_t = -i\theta(t)\langle[A(t), B(0)]_+\rangle. \tag{2.5}$$

$\theta(t)$ is the Heaviside step function and $\langle\cdots\rangle$ denotes the statistical average for the system. $G_{AB}(\omega) = \langle\langle A \mid B\rangle\rangle_\omega$ is the Fourier transform of $G_{AB}(t)$ with respect to time t. The equation of motion of $G_{AB}(\omega)$ is written[11]

$$\omega G_{AB}(\omega) = (1/2\pi)\langle[A(0), B(0)]_+\rangle + \langle\langle[A, H]_- \mid B\rangle\rangle_\omega. \tag{2.6}$$

H is the total Hamiltonian of the system given by (2.1) in our problem.

We define the following Green's functions:

$$G_{kk'}(\omega) = \langle\langle C_{k\uparrow} \mid C_{k'\uparrow}^{\dagger}\rangle\rangle_\omega, \tag{2.7}$$

$$F_{kk'}^{\dagger}(\omega) = \langle\langle C_{-k\downarrow}^{\dagger} \mid C_{k'\uparrow}^{\dagger}\rangle\rangle_\omega, \tag{2.8}$$

$$\Gamma_{kk'}(\omega) = \langle\langle S_Z C_{k\uparrow} + S_- C_{k\downarrow} \mid C_{k'\uparrow}^{\dagger}\rangle\rangle_\omega, \tag{2.9}$$

$$\Phi_{kk'}^{\dagger}(\omega) = \langle\langle S_Z C_{k\downarrow}^{\dagger} - S_- C_{k\uparrow}^{\dagger} \mid C_{k'\uparrow}^{\dagger}\rangle\rangle_\omega. \tag{2.10}$$

[10] P. Bloomfield and D. R. Hamann, Phys. Rev. (to be published).
[11] D. N. Zubarev, Usp. Fiz. Nauk. **71**, 71 (1960); [English transl.: Soviet Phys.—Usp. **3**, 320 (1966)].
[12] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **39**, 1781 (1960) [English transl.: Soviet Phys.—JETP **12**, 1243 (1961)].
[13] A. Griffin, Phys. Rev. Letters, **15**, 703 (1965).
[14] K. Maki, Phys. Rev. **153**, 428 (1967).
[15] J. Solyom and A. Zawadowski (to be published).
[16] J. Bardeen L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

Using (2.1) through (2.6) we obtain equations of motion for the Green's functions defined in (2.7) through (2.10). These are

$$(\omega-\epsilon_k)G_{kk'}-\Delta F_{kk'}=(\delta_{kk'}/2\pi)-(J/2N)\sum_l \Gamma_{lk'}, \tag{2.11}$$

$$(\omega+\epsilon_k)F_{kk'}-\Delta G_{kk'}=-(J/2N)\sum \Phi_{lk'}{}^\dagger, \tag{2.12}$$

$$(\omega-\epsilon_k)\langle\langle S_Z C_{k\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega=-(J/2N)\{\sum_l[\langle\langle S_Z{}^2 C_{l\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\tfrac{1}{2}\langle\langle S_- C_{l\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]$$
$$+\sum_{ll'}[-\langle\langle S_- C_{k\uparrow}C_{l\uparrow}{}^\dagger C_{l'\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\langle\langle S_+ C_{k\uparrow}C_{l\downarrow}{}^\dagger C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]\}+\mid g\mid \sum_{l'}\langle\langle S_Z C_{-k\downarrow}{}^\dagger C_{-l'\downarrow}C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega, \tag{2.13}$$

$$(\omega-\epsilon_k)\langle\langle S_- C_{k\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega=-(J/2N)\{\sum_l[\tfrac{3}{4}G_{lk'}-\langle\langle S_Z{}^2 C_{l\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\tfrac{1}{2}\langle\langle S_- C_{l\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\langle\langle S_Z C_{l\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]$$
$$+\sum_{ll'}[\langle\langle S_- C_{k\downarrow}C_{l\uparrow}{}^\dagger C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega-\langle\langle S_- C_{k\downarrow}C_{l\downarrow}{}^\dagger C_{l'\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega-2\langle\langle S_Z C_{k'\downarrow}C_{l\downarrow}{}^\dagger C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]\}$$
$$-\mid g\mid \sum_{l'}\langle\langle S_- C_{-k\uparrow}{}^\dagger C_{-l'\downarrow}C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega, \tag{2.14}$$

$$(\omega+\epsilon_k)\langle\langle S_Z C_{k\downarrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega=(J/2N)\{\sum_l[-\langle\langle S_Z{}^2 C_{l\downarrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\tfrac{1}{2}\langle\langle S_- C_{l\uparrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]$$
$$-\sum_{ll'}[-\langle\langle S_- C_{k\downarrow}{}^\dagger C_{l\uparrow}{}^\dagger C_{l'\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\langle\langle S_+ C_{k\downarrow}{}^\dagger C_{l\downarrow}{}^\dagger C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]\}+\mid g\mid \sum_{ll'}\langle\langle S_Z C_{l'\uparrow}{}^\dagger C_{-l'\downarrow}{}^\dagger C_{-k\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega, \tag{2.15}$$

$$(\omega+\epsilon_k)\langle\langle S_- C_{k\uparrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega=(J/2N)\{\sum_l[-\tfrac{1}{2}\langle\langle S_- C_{l\uparrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega-\langle\langle S_Z{}^2 C_{l\downarrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega$$
$$+\tfrac{3}{4}\langle\langle C_{l\downarrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega+\langle\langle S_Z C_{l\downarrow}{}^\dagger \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]-\sum_{ll'}[\langle\langle S_- C_{k\uparrow}{}^\dagger C_{l\uparrow}{}^\dagger C_{l'\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega-\langle\langle S_- C_{k\uparrow}{}^\dagger C_{l\downarrow}{}^\dagger C_{l'\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega$$
$$-2\langle\langle S_Z C_{k\uparrow}{}^\dagger C_{l\downarrow}{}^\dagger C_{l'\uparrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega]\}-\mid g\mid \sum_{l'}\langle\langle S_- C_{l'\uparrow}{}^\dagger C_{-l'\downarrow}{}^\dagger C_{-k\downarrow} \mid C_{k'\uparrow}{}^\dagger\rangle\rangle_\omega. \tag{2.16}$$

In the derivation of (2.11) to (2.16) we have used the following relations for spin-$\frac{1}{2}$ operators[4]

$$S_\pm S_Z=\mp S_\pm, \qquad S_Z S_\pm=\pm\tfrac{1}{2}S_\pm,$$
$$S_+S_-=\tfrac{3}{4}+S_Z-S_Z{}^2, \tag{2.17}$$

and we require $\langle S_Z\rangle=0$ in the absence of a magnetic field. The decoupling scheme due to Gor'kov[17] has been used to derive (2.11) and (2.12). Δ is the order parameter defined by

$$\Delta=\mid g\mid \sum\langle C_{k\uparrow}{}^\dagger C_{-k\downarrow}{}^\dagger\rangle. \tag{2.18}$$

The next step is to make the following approximation: Certain sets of operators occurring inside the Green's functions in Eqs. (2.13) to (2.16) are replaced by their average values. This is done after the manner of Nagaoka[4] and the only nonvanishing averages are taken to be those which conserve spin. The relations between these averages are

$$\langle C_{l\uparrow}{}^\dagger C_{l'\uparrow}\rangle=\langle C_{l\downarrow}{}^\dagger C_{l'\downarrow}\rangle, \tag{2.19a}$$

$$\langle C_{l\uparrow}{}^\dagger C_{l'\downarrow}{}^\dagger\rangle=-\langle C_{l'\downarrow}{}^\dagger C_{l\uparrow}{}^\dagger\rangle, \tag{2.19b}$$

$$\langle S_- C_{l\uparrow}{}^\dagger C_{l'\downarrow}\rangle=\langle S_+ C_{l\downarrow}{}^\dagger C_{l'\uparrow}\rangle$$
$$=2\langle S_Z C_{l\uparrow}{}^\dagger C_{l'\uparrow}\rangle$$
$$=-\langle S_Z C_{l\downarrow}{}^\dagger C_{l'\downarrow}\rangle, \tag{2.20}$$

$$\langle S_- C_{l\downarrow}C_{l'\downarrow}\rangle=-\langle S_+ C_{l\uparrow}C_{l'\uparrow}\rangle$$
$$=-2\langle S_Z C_{l\uparrow}C_{l'\downarrow}\rangle$$
$$=2\langle S_Z C_{l\downarrow}C_{l'\uparrow}\rangle, \tag{2.21a}$$

$$\langle S_- C_{l\uparrow}{}^\dagger C_{l'\uparrow}{}^\dagger\rangle=-\langle S_+ C_{l\downarrow}{}^\dagger C_{l'\downarrow}{}^\dagger\rangle$$
$$=-2\langle S_Z C_{l\downarrow}{}^\dagger C_{l'\uparrow}{}^\dagger\rangle$$
$$=2\langle S_Z C_{l\uparrow}{}^\dagger C_{l'\downarrow}{}^\dagger\rangle. \tag{2.21b}$$

Use of the decoupling scheme and the relationships between averages in (2.19) through (2.21) enables us to derive a set of equations in closed form from (2.11) through (2.16). These equations are more simply expressed in terms of the Nambu-Schrieffer matrix formalism[18] for superconductivity. To this purpose we define the following matrices:

$$\hat{G}_{kk'}=\begin{bmatrix} G_{kk'} & F_{kk'} \\ F_{kk'}{}^\dagger & G_{kk'}{}^\dagger \end{bmatrix}, \tag{2.22}$$

$$\hat{\Gamma}_{kk'}=\begin{bmatrix} \Gamma_{kk'} & \Phi_{kk'} \\ \Phi_{kk'}{}^\dagger & \Gamma_{kk'}{}^\dagger \end{bmatrix}. \tag{2.23}$$

Then the equations for $\hat{G}_{kk'}$ and $\hat{\Gamma}_{kk'}$ are

$$(\omega-\epsilon_k\tau_3-\Delta\tau_1)\hat{\Gamma}_{kk'}+(J/N)[\tau_3(n_k-\tfrac{1}{2})+\tau_1\Delta_k]\hat{\Gamma}_{k'}$$
$$=(J/2N)[(m_k-\tfrac{3}{4})-\tau_1\vartheta_k]\hat{G}_{kk'}, \tag{2.24}$$

$$(\omega-\epsilon_k\tau_3-\Delta\tau_1)\hat{G}_{kk'}=(\delta_{kk'}/2\pi)-(J/2N)\hat{\Gamma}_{k'}. \tag{2.25}$$

[17] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **34**, 735 (1958) [English transl.: Soviet Phys.—JETP **34**, 505 (1958).

[18] J. R. Schrieffer, *Theory of Superconductivity* (W. A. Benjamin and Co., Inc., New York, 1964).

In (2.24) and (2.25) m_k, $\mathfrak{p}_k$, n_k, Δ_k are defined as follows:

$$m_k=3\sum_l\langle S_-C_{l\uparrow}{}^\dagger C_{k\downarrow}\rangle, \tag{2.26a}$$

$$n_k=\sum_l\langle C_{l\uparrow}{}^\dagger C_{k\uparrow}\rangle, \tag{2.26b}$$

$$\Delta_k=\sum_l\langle C_{l\downarrow}{}^\dagger C_{k\uparrow}{}^\dagger\rangle, \tag{2.26c}$$

$$\mathfrak{p}_k=\begin{bmatrix}0 & \sum_l\langle S_+C_{l\downarrow}{}^\dagger C_{k\downarrow}{}^\dagger\rangle\\ \sum_l\langle S_-C_{l\downarrow}C_{k\downarrow}\rangle, & 0\end{bmatrix}. \tag{2.26d}$$

τ_1 and τ_3 are Pauli spin matrices and

$$\hat\Gamma_k=\sum_l\hat\Gamma_{lk};\qquad \hat G_k=\sum_l\hat G_{lk}. \tag{2.27}$$

Nagaoka's equations for the normal state[4] are recovered from (2.24) and (2.25) if we take $\mathfrak{p}_k=\Delta_k=\Delta=0$.

The following results are obtained for $\hat\Gamma_k$ and $\hat G_{kk'}$ from (2.24) and (2.25):

$$\hat G_{kk'}(\omega)=\hat G_{0k}(\omega)[(\delta_{kk'}/2\pi)+\hat t(\omega)\hat G_{0k'}(\omega)], \tag{2.28a}$$

$$\hat\Gamma_k(\omega)=-(2N/J)\hat t(\omega)\hat G_{0k}(\omega), \tag{2.28b}$$

$$G_{0k}(\omega)=(\omega-\epsilon_k\tau_3-\Delta\tau_1)^{-1}. \tag{2.28c}$$

$t(\omega)$ is the t matrix for the superconducting state and is given by

$$t(\omega)=-(J^2/4N^2)\hat A(\omega)\,(1+(J^2/4N^2)\hat F(\omega)\hat A(\omega))^{-1}, \tag{2.29a}$$

where

$$\hat A(\omega)=[1+(J/N)(\tau_3\hat G(\omega)+\tau_1\hat\Delta(\omega))]^{-1}\times[\hat\Gamma_1(\omega)-\hat\Gamma_2(\omega)] \tag{2.29b}$$

and

$$\hat G(\omega)=\sum_k(n_k-\tfrac12)\hat G_{0k}(\omega), \tag{2.30}$$

$$\hat\Delta(\omega)=\sum_k\Delta_k\hat G_{0k}(\omega), \tag{2.31}$$

$$\hat\Gamma_1(\omega)=\sum_k(m_k-\tfrac34)\hat G_{0k}(\omega), \tag{2.32}$$

$$\hat\Gamma_2(\omega)=\sum_k\mathfrak{p}_k\hat G_{0k}(\omega), \tag{2.33}$$

$$\hat F(\omega)=\sum_k\hat G_{0k}(\omega). \tag{2.34}$$

Equations (2.26) through (2.34) form a complete set of equations for the problem of a single spin-$\frac12$ magnetic impurity in a superconducting matrix. These equations are generalized to the case of a finite concentration of impurities in Sec. 3. An expression for T_c is then derived by linearizing with respect to the order parameter Δ.

3. THE GENERAL EQUATION FOR T_c

We first perform the transformation $\omega\to i\omega_n$, where

$$\omega_n=(2n+1)\pi kT. \tag{3.1}$$

The conclusion of a finite concentration n_I of impurities[19] gives the following form for the one-electron Green's function $\hat G_{kk'}$:

$$\hat G_{kk'}=\hat G_k\delta_{kk'} \tag{3.2a}$$

$$\hat G_k=[i\omega_n-\epsilon_k\tau_3-\Delta\tau_1-n_Il(i\omega_n)]^{-1}. \tag{3.2b}$$

We next assume a solution of the form

$$\hat G_k=[iZ_n\omega_n-\epsilon_k\tau_3-\Delta_n\tau_1]^{-1}. \tag{3.3}$$

Z_n and Δ_n are functions of ω_n. Finally $\hat G_{0k}$ which occurs in the definition (2.30) to (2.34) must be replaced by $\hat G_k$ in (3.3). From (2.18) and (3.2) the equation for Δ becomes

$$\Delta=\pi N(0)\mid g\mid kT\sum_n\frac{\Delta_n}{[Z_n{}^2\omega_n{}^2+\Delta_n{}^2]^{1/2}} \tag{3.4}$$

after integration over momentum space. To find the equation for T_c, we begin by linearizing (3.4) with respect to Δ_n. The result is

$$\Delta=\pi N(0)\mid g\mid kT\sum_n(\Delta_n/Z_n\mid\omega_n\mid), \tag{3.5}$$

where Z_n is independent of Δ. In order to obtain expressions for Z_n and Δ_n from (3.2b) and (3.3) it is necessary to obtain an expression for $\hat t\ (i\omega_n)$ which is linearized with respect to Δ. Now

$$n_k=n_k{}^0+O(\Delta^2),$$

$$m_k=m_k{}^0+O(\Delta^2),$$

$$\mathfrak{p}_k,\ \Delta_k=O(\Delta). \tag{3.6}$$

$n_k{}^0$ and $m_k{}^0$ are the solutions of Nagaoka's equations in the normal state.[4] $m_k{}^0$ is an even function of ϵ_k and $(n_k{}^0-\frac12)$ is an odd function of ϵ_k. The use of these considerations give the following linearized forms for $\hat G$, $\hat\Gamma_1$, and $\hat F$ in (2.30), (2.32), and (2.34) using (3.3) and (3.6):

$$\tau_3\hat G(i\omega_n)=-G_n{}^0, \tag{3.7}$$

$$\hat\Gamma_1(i\omega_n)=i\Gamma_n{}^0-\Delta_n{}^{(2)}\tau_1, \tag{3.8}$$

$$\hat F(i\omega_n)=iF_{1n}-F_{2n}\tau_1, \tag{3.9}$$

where

$$G_n{}^0=\sum_k(n_k{}^0-\tfrac12)\epsilon_k/[Z_n{}^2\omega_n{}^2+\epsilon_k{}^2]; \tag{3.10}$$

$$\Gamma_n{}^0=-Z_n\omega_n\sum_k(m_k{}^0-\tfrac34)/[Z_n{}^2\omega_n{}^2+\epsilon_k{}^2]; \tag{3.11a}$$

$$\Delta_n{}^{(2)}=-(\Delta_n/Z_n\omega_n)\Gamma_n{}^0; \tag{3.11b}$$

$$F_1=-\pi N(0)\omega_n/\mid\omega_n\mid;$$

$$F_2=-\pi N(0)\Delta_n/(Z_n\mid\omega_n\mid). \tag{3.12}$$

The calculation of $\hat\Delta$ and $\hat\Gamma_2$ in (2.31) and (2.33) is somewhat more complex. From (2.26c) and (3.3),

[19] See, for example, G. Rickayzen, *International Spring School of Physics, 1961* (W. A. Benjamin and Co., Inc., New York, 1964).

Δ_k becomes

$$\Delta_k = -kT \sum_{n'} \Delta_{n'}/[Z_{n'}{}^2\omega_{n'}{}^2+\epsilon_k{}^2+\Delta_{n'}{}^2]. \tag{3.13}$$

Substitution of (3.13) into (2.31), linearization with respect to Δ_n, and integration over momentum space, yields

$$\tau_1\hat{\Delta}(i\omega_n) = -i\tau_1\Delta_n{}^{(1)}, \tag{3.14}$$

where

$$\Delta_n{}^{(1)} = \sum_{n'} A_{nn'}\Delta_{n'}, \tag{3.15}$$

and

$$A_{nn'} = -\pi N(0)kT\omega_n/[|\,\omega_n\,|\,|\,\omega_{n'}\,|\,(|\,\omega_{n'}\,|+|\,\omega_n\,|)]. \tag{3.16}$$

To obtain $\hat{\Gamma}_2$, we must first calculate $\hat{v}_k$ which is defined by (2.26d). From (2.23) and (2.26d) $\hat{v}_k$ becomes

$$\hat{v}_k = kT(\sum_{n'} \hat{\Gamma}_k)_{\text{od}}, \tag{3.17}$$

where od implies the retaining of off-diagonal matrix elements only. $\hat{\Gamma}_k$ is given by

$$\hat{\Gamma}_k = -(2N/J)[(1-Z_n)i\omega_n+(\Delta_n-\Delta)\tau_1]\hat{G}_{0k} \tag{3.18}$$

from (2.28c), (3.2b), and (3.3). Then from (3.18) and (3.19)

$$\hat{v}_k = -kT(2/J) \sum_{n'} \epsilon_k\tau_3(\Delta_{n'}-\Delta)/[Z_{n'}{}^2\omega_{n'}{}^2+\epsilon_k{}^2+\Delta_{n'}{}^2]. \tag{3.19}$$

Substitution of (3.19) into (2.33) and linearization with respect to Δ and Δ_n yields

$$\hat{\Gamma}_2(i\omega_n) = \Delta_n{}^{(3)}, \tag{3.20}$$

where

$$\Delta_n{}^{(3)} = \sum_{n} B_{nn'}\Delta_{n'} - B_n\Delta \tag{3.21}$$

and

$$B_{nn'} = -(2_s/J)kT \sum_{k} \epsilon_k{}^2/[(Z_n{}^2\omega_n{}^2+\epsilon_k{}^2)(Z_{n'}{}^2\omega_{n'}{}^2+\epsilon_k{}^2)], \tag{3.22a}$$

$$B_n = \sum_{n'} B_{nn'}. \tag{3.22b}$$

We now have enough results to evaluate the t matrix defined in (2.29a) to first order in Δ. We begin by evaluating $\hat{A}(i\omega_n)$ defined in (2.29b). Substitution of (3.7), (3.8), (3.14), and (3.20) into (2.29b), linearization with respect to Δ and some tedious matrix algebra yields

$$\hat{A}(i\omega_n) = iA_n{}^0+A_{2n}\tau_1, \tag{3.23}$$

where

$$A_n{}^0 = [1-(J/N)G_n{}^0]^{-1}\Gamma_n{}^0, \tag{3.24}$$

$$A_{2n} = -[1-(J/N)G_n{}^0]^{-2}[(J/N)\Gamma_n{}^0\Delta_n{}^{(1)}+[1-(J/N)G_n{}^0](\Delta_n{}^{(2)}+\Delta_n{}^{(3)})]. \tag{3.25}$$

This substitution of (3.23) and (3.9) into (2.29a) and linearization with respect to Δ yields the following result for the t matrix:

$$t(i\omega_n) = it_n{}^0+\tau_1 t_{2n}, \tag{3.26}$$

where

$$t_n{}^0 = -(J/2N)^2[1-(J^2/4N^2)F_{1n}A_n{}^0]^{-1}A_n{}^0, \tag{3.27}$$

$$t_{2n} = -(J/2N)^2[1-(J^2/4N^2)F_{1n}A_n{}^0]^{-2}[A_2+(J^2/4N^2)F_2(A_n{}^0)^2]. \tag{3.28}$$

$t_n{}^0$ is the t matrix for the normal state.[20] Substitution of (3.26) into (3.2b) and comparison with (3.3) yields

$$Z_n\omega_n = \omega_n - n_I t_n{}^0, \tag{3.29}$$

$$\Delta_n = \Delta + n_I t_{2n}. \tag{3.30}$$

[20] H. Suhl, Phys. Rev. **138**, A515 (1965).

Finally substitution of (3.24), (3.25), (3.7), and (3.28) into (3.30) yields the following linear integral equation for Δ_n:

$$\Delta_n = G_n^{(1)}\Delta + n_I G_n^{(2)} \sum_{n'} C_{nn'}\Delta_{n'}. \tag{3.31}$$

The coefficients $G_n^{(1)}$ and $G_n^{(2)}$ are determined by further substitution of (3.11b), (3.12), (3.15), and (3.21) into (3.25). The result is

$$G_n^{(1)} = [(1-(J^2/4^2)F_{1n}A_n^0)^2 - n_I J^2 C_n^{(1)}/4N^2]^{-1}[(1-(J^2/4N^2)F_{1n}A_n^0)^2 + n_I J^2 C_n^{(2)}/4N^2], \tag{3.32}$$

$$G_n^{(2)} = [(1-(J^2/4^2)F_{1n}A_n^0)^2 - n_I J^2 C_n^{(1)}/4N^2]^{-1} n_I J^2/4N^2. \tag{3.33}$$

$C_n^{(1)}$, $C_n^{(2)}$ and $C_{nn'}$ are given by

$$C_n^{(1)} = -(\Gamma_n^0/Z_n\omega_n)(1-(J/N)G_n^0)^{-1} + (J^2/4N^2)[\pi N(0)(A_n^0)^2/Z_n \mid \omega_n \mid], \tag{3.34}$$

$$C_n^{(2)} = -(1-(J/N)G_n^0)^{-1}B_n, \tag{3.35}$$

$$C_{nn'} = (1-(J/N)G_n^0)^{-2}[(J/N)\Gamma_n^0 A_{nn'} + (1-(J/N)G_n^0)B_{nn'}]. \tag{3.36}$$

(3.31) cannot be solved analytically. However, to lowest order in the impurity concentration n_I, only the first iteration need be retained, i.e.,

$$\Delta_n = [G_n^{(1)} + n_I G_n^{(2)} \sum_{n'} C_{nn'} G_{n'}^{(1)}]\Delta. \tag{3.37}$$

The equation for the superconducting critical temperature T_c is given from (3.5) and (3.7) by

$$1 = \pi N(0) \mid g \mid kT_c \sum_n (Z_n \mid \omega_n^c \mid)^{-1} \times [G_n^{(1)} + n_I G_n^{(2)} \sum_{n'} C_{nn'} G_{n'}^{(1)}]. \tag{3.38}$$

ω_n is replaced by $\omega_n^c = (2n+1)\pi kT_c$ on the right-hand side of (3.38) and Z_n is given by (3.29). From (3.38) the equation for T_c may be written in terms of T_{c0}, the critical temperature of the pure metal, using the BCS formula $\ln(1.14\ kT_{c0}/\omega_D) = -N(0) \mid g \mid$. Here ω_D is the Debye frequency. Then, (3.38) becomes

$$\ln(T_{c0}/T_c) = \pi kT_c \sum_n (Z_n \mid \omega_n^c \mid)^{-1} \times [Z_n - G_n^{(1)} - n_I G_n^{(2)} \sum_{n'} C_{nn'} G_{n'}^{(1)}]. \tag{3.39}$$

From (3.11b), (3.16), (3.22), (3.27), (3.35), (3.36), and (3.39) we obtain the following expression for T_c to lowest order in n_I:

$$\ln(T_{c0}/T_c) = \pi n_I kT_c (J/2N)^2 \sum_n (\omega_n^c \mid \omega_n^c \mid)^{-1} \times (1-(J^2/4N^2)F_{ln}A_n^0)^{-2}A_n^0\{2+\pi N(0)(J/N) \times (1-(J/N)G_n^0)^{-1} \mid \omega_n \mid kT_c \times \sum_{n'} [\mid \omega_{n'}^c \mid (\mid \omega_n^c \mid + \mid \omega_{n'}^c \mid)]^{-1}\}. \tag{3.40}$$

Here $A_{n'}{}^0 F_{ln}$ and G_n^0 are defined in (3.12), (3.24), and (3.10), respectively. (3.40) is a general expression for T_c. The only unknowns are m_k^0 and n_k^0 and they have to be determined by solving Nagaoka's equations in the normal state. This problem is considered in Sec. 4.

4. EVALUATION OF T_c FOR PARTICULAR SOLUTIONS OF NAGAOKA'S EQUATIONS

In this section the approximate solutions of Nagaoka's equations[4] due to Nagaoka[4,5] and Hamann[7] for the normal state are substituted into (3.40). The results are then analyzed in the light of previous results for T_c in dilute magnetic alloys.[21]

A. Nagaoka's Solution for $T_c \gg T_K$

In this limit the function A_n^0 defined in (3.12) is given by[5]

$$A_n^0 = [3\pi N(0)\omega_n/4 \mid \omega_n \mid](1-(J/N)G_n^0)^{-2}. \tag{4.1}$$

Here G_n^0 is given by

$$G_n^0 = \sum_k (f_k - \tfrac{1}{2})\epsilon_k/(\omega_n^2 + \epsilon_k^2). \tag{4.2}$$

The form (4.2) for G_n^0 is obtained from (3.10) by replacing n_k^0 in (3.10) by the Fermi distribution function f_k. The term $J^2F_{ln}A_n^0/4N^2$ on the right-hand side of (3.40) is neglected. We define the dimensionless quantity h_n as follows:

$$h_n^{-1} = \pi N(0)(J/N)(1-(J/N)G_n^0)^{-1}. \tag{4.3}$$

Then, from (4.1) and (4.3), (3.40) becomes

$$\ln(T_{c0}/T_c) = [3n_I kT_c/16N(0)] \sum_n (\mid \omega_n^c \mid h_n^2)^{-1} \times \{(2/\mid \omega_n^c \mid) + (kT_c/h_n) \times \sum_{n'} [\mid \omega_{n'}^c \mid (\mid \omega_n^c \mid + \mid \omega_{n'}^c \mid)]^{-1}\}. \tag{4.4}$$

[21] P. Hohenberg, Zh. Eksperim. i Teor. Fiz. **45**, 1208 (1963); [English transl.: Soviet Phys.—JETP **18**, 834 (1964)].

Now

$$f_k-\tfrac{1}{2}=-kT\sum_n \epsilon_k/[\omega_n^2+\epsilon_k^2]. \quad (4.5)$$

Then from (4.2) and (4.5)

$$G_n^0=-\pi N(0)kT\sum_n [|\omega_n|+|\omega_{n'}|]^{-1}. \quad (4.6)$$

At low temperatures we may replace the sum over n by an integral, i.e.,

$$kT\sum_n [|\omega_n|+|\omega_{n'}|]^{-1}=\pi^{-1}\int_0^{+D}\frac{dx}{x+|\omega_n|}$$
$$=\pi^{-1}\ln\left(\frac{D}{|\omega_n|}\right), \quad (4.7)$$

where D is the width of the conduction band. From (4.3), (4.6), and (4.7)

$$h_n^{-1}=\pi N(0)(J/N)[1+N(0)(J/N)\ln(D/|\omega_n|)]^{-1}. \quad (4.8)$$

Also

$$kT\sum_n [|\omega_n|(|\omega_n|+|\omega_{n'}|)^{-1}=\Psi(2n+1)/\pi|\omega_n|, \quad (4.9)$$

where

$$\Psi(x)=\psi(\tfrac{1}{2}+\tfrac{1}{2}x)-\psi(\tfrac{1}{2}) \quad (4.10)$$

and $\psi(x)$ is the digamma function. Then from (4.4), (4.8), and (4.9) the equation for T_c becomes

$$\ln\left(\frac{T_{c0}}{T_c}\right)=\frac{3n_IN(0)J^2}{16kT_cN^2}\sum_{n=0}^{\infty}(2n+1)^{-2}\left[1+\frac{N(0)J}{N}\ln\left\{\frac{D}{(2n+1)\pi kT_c}\right\}\right]$$
$$\times\left\{2+\frac{N(0)J}{N}\Psi(2n+1)\left[1+\frac{N(0)J}{N}\ln\left\{\frac{D}{(2n+1)\pi kT_c}\right\}\right]^{-1}\right\}. \quad (4.11)$$

The decrease in T_c with respect to n_I arising from the first term in the curly brackets in (4.11) is essentially that obtained by Griffin[13] and Maki.[14] To lowest order in J this term reduces to the classical result of Abrikosov and Gor'kov[12] for the initial decrease in T_c due to s-d exchange scattering in the Born approximation. The remaining term in (4.11) represents an increase in T_c for $J<0$ and a decrease in T_c for $J>0$. To third order in J this term reduces to the change in T_c obtained by Solyom and Zawadowski[15] and its significance is discussed in their paper. The important point is that the general expression (3.40) reduces to the known changes in T_c found by previous investigations in the range $T_c\gg T_K$.

B. Hamann's Solution

The advantage of Hamann's solution is that it is valid over the whole range of T_K. Hamann's expression for t_n^0 as defined in (3.27) is given by

$$t_n^0=\frac{-\omega_n}{2\pi N(0)|\omega_n|}\left\{1-\ln\left(\frac{|\omega_n|}{kT_K}\right)\left[\ln^2\left(\frac{|\omega_n|}{kT_K}\right)+\tfrac{3}{4}\pi^2\right]^{-1/2}\right\}, \quad (4.12)$$

when $J<0$. From (3.12), (3.27), and (4.12) we obtain

$$(1-(J^2/4N^2)F_{ln}A_n^0)=(1+\pi N(0)\omega_nt_n^0/|\omega_n|)^{-1}. \quad (4.13)$$

We make the following approximation[13] for ease of calculation:

$$\frac{N}{|J|N(0)}\left(1+\frac{|J|}{N}G_n^0\right)\approx\left[\ln^2\left(\frac{|\omega_n|}{kT_K}\right)+\tfrac{3}{4}\pi^2\right]^{1/2}. \quad (4.14)$$

From (3.27), (3.40), (4.12), (4.13), and (4.14), we obtain

$$T_c-T_{c0}=-[n_I/kN(0)\pi]\sum_{n=0}^{\infty}(2n+1)^{-2}\phi(x_n)$$
$$\times[1-\phi(x_n)/2]\{1-\Psi(2n+1)[\ln^2x_n+\tfrac{3}{4}\pi^2]^{-1/2}\}, \quad (4.15)$$

where

$$x_n=(2n+1)\pi T_{c0}/T_K, \quad (4.16)$$

$$\phi(x)=\{1-\ln x[\ln x^2+\tfrac{3}{4}\pi^2]^{-1/2}\}. \quad (4.17)$$

We note that the change in T_c is a function of T_{c0}/T_K only. $(T_{c0}-T_c)N(0)k/n_I$ versus T_{c0}/T_K is plotted in Fig. 1, which indicates that there is an initial decrease in T_c for all values of T_K. For $T_c\gg T_K$, $\ln x_n\gg 1$. Then (4.23) reduces to the expression (4.11) for T_c if only terms of lowest order in $(\ln x_n)^{-1}$ are retained and if T_K is defined as in Ref. 4. For $T_c\ll T_K$, $|\ln x_n|\gg 1$ but $\ln c_n$ is negative for values of n for which $(2n+1)\pi T_{c0}>T_K$. Since the convergence of the series in (4.15) is very rapid, only a negligible correction is introduced if we assume $\ln x_n$ is negative for all n in (4.23). Then to lowest order in $(\ln x_n)^{-1}$ (4.15) becomes

$$T_c-T_{c0}=-\frac{3n_I}{4N(0)k\pi}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2|\ln x_n|^2}$$
$$\times\left\{1+\frac{\Psi(2n+1)}{|\ln x_n|}\right\}, \quad (4.18)$$

where $x_n=(2n+1)\pi T_c/T_K$. The relationship of this result to experimental data is discussed in Sec. 5.

5. CONCLUSION

We have been able to derive a general expression for the superconducting critical temperature T_c of a

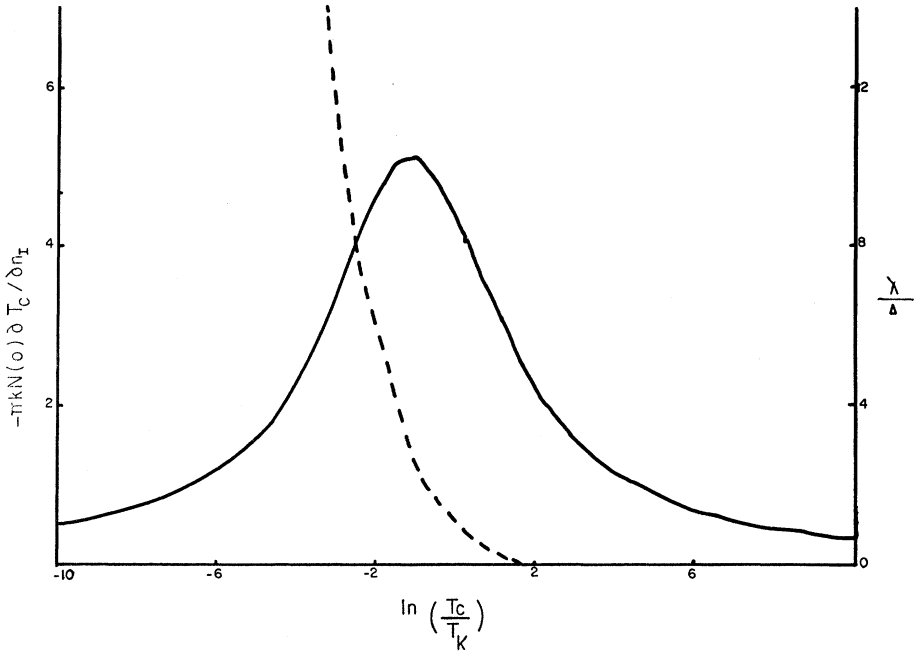

FIG. 1. The initial decrease in T_c as a function of the Kondo temperature T_K. The solid curve shows $-kN(0)(\partial T_c/\partial n_I)_0$ versus $\ln(T_c/T_K)$. The dashed curve shows the binding energy λ of the singlet state in units of the order parameter Δ versus $\ln(T_c/T_K)$ after the work of Hone (Ref. 22).

dilute magnetic alloy for any value of the Kondo temperature T_K. This expression depends on the averages n_k^0 and m_k^0 (see Sec. 2), which are determined by solving Nagaoka's equations for the normal state.[4] Nagaoka's solution[5] for $T>T_K$ was substituted into the expression for T_c and the result was found to agree with previous theoretical work. Hamann's solution was next used because of its validity for all T with respect to T_K, and a simple equation for T_c was obtained in the limit $T_c \ll T_K$. Hone in a recent paper[22] calculates the ground-state energy of a superconductor containing one magnetic impurity. His work compared favorably with our results obtained by use of Hamann's model[7] (see Fig. 1).

Extension to higher-impurity concentrations and to temperatures $T<T_c$ involves the solution of various integral equations [for example, (3.31)]. In addition use of the exact solution of Nagaoka's equations due to Bloomfield and Hamann[10] should give an improved result for T_c. These considerations will be dealt with in a future publication.

A comparison with experiment presents difficulties. In the case of dilute magnetic impurities, it has been shown[23] that the coupling constant g of the BCS Hamiltonian described by (2.4) must be replaced by an effective coupling constant which is dependent on the concentration of impurities. This results in a further decrease in T_c for low-impurity concentrations. Further, Schrieffer[24] has suggested that the Zn- and Al-iron group alloys which appear to be nonmagnetic, may have a very high Kondo temperature of the order of the Fermi temperature. In consequence, $T_c \ll T_K$ for these alloys since $T_c \sim 1°\mathrm{K}$. The initial decrease in T_c with n_I in the limit $T_c \ll T_K$ is approximately given by

$$T_c - T_{c0} = -[3n_I/32kN(0)][\ln(\pi T_K/T_c)]^{-2} - 2n_I T_{c0} V^2[E_d^{-1} + (E_d+U)^{-1}] \ln\left(\frac{\gamma\omega_D}{\pi k T_{c0}}\right). \quad (5.1)$$

The first term on the right-hand side of (5.1) is obtained from (4.18) under the assumption that the series in (4.18) converges very rapidly. The second term is the decrease in T_c due to the effective BCS Hamiltonian.[23] V, E_d, and U are the coupling constant of the s–d admixture interaction, the energy of the localized d-orbital on the impurity relative to the Fermi level, and the coupling constant of the d-d Coulomb interaction, respectively. All these parameters appear in Anderson's model.[25] V and E_d are of the same order of magnitude and $E_d<U$. Since $T_{c0} \sim 1°\mathrm{K}$ and $T_K > 10°\mathrm{K}$, these considerations imply that the dominant term is the first term on the right-hand side of (5.1).

It is important to comment on the nature of the result for T_c in Eq. (5.1). Hamann[7] argues that there should be complete cancellation of the magnetic moment on the impurity at $T=0$. His expression for the magnetic susceptibility χ of the localized spin-$\frac{1}{2}$ impurity at temperature $T_{c0}(\ll T_K)$ is

$$\chi(T_{c0}) = 2\chi_0(T_{c0})\ \ln^{-1}(T_K/T_{c0}). \quad (5.2)$$

Here χ_0 is the Curie-Weiss susceptibility of the alloy.

[22] D. Hone (to be published).
[23] J. Appelbaum, V. Celli, and M. J. Zuckermann, Phys. Letters **25A**, 24 (1967).
[24] J. R. Schrieffer, J. Appl. Phys. **38**, 1143 (1967).
[25] P. W. Anderson, Phys. Rev. **124**, 41 (1961).

From (5.2) we may infer that the effective magnetic moment μ_{eff} at temperature T_c is

$$\mu_{\text{eff}}(T_{c0}) = 2^{1/2}\mu_B \ln^{-1/2}(T_K/T_{c0}), \quad (5.3)$$

where μ_B is the Bohr magneton. Then the first term on the right-hand side of (5.1) gives rise to an initial decrease in T_c of the form

$$(\partial T_c/\partial n_I)_0 \approx -(3/128N(0)k)(\mu_{\text{eff}}(T_{c0})/\mu_B)^4. \quad (5.4)$$

We can therefore speculate that the initial decrease in T_c is related to the residual magnetic moment on the impurity at temperature T_{c0} when $T_{c0} \ll T_K$, but this statement requires further proof.

For general values of T_c with respect to T_K, the initial decrease is T_c with n_I, i.e., $(\partial T_c/\partial n_I)_0$ is given by Fig. 1. It is seen that $-(\partial T_c/\partial n_I)_0$ increases as the binding energy λ of the singlet state (between the conduction and localized electrons at $T=0$) decreases.[22] This is reasonable since the destruction of the singlet state gives rise to an increase in the effective impurity magnetic moment. When the singlet state is nearly destroyed, $-(\partial T_c/\partial n_I)_0$ reaches a maximal value and then decreases again as T_K increases further. This is due to the logarithmic character of the Kondo anomaly for $T_c > T_K$. Figure 1 may be used to obtain values of T_K for Zn–Iron group alloys from the data of Boato *et al.*[26] If we use $T_K \sim 1$°K for Zn–Mn, then $T_K \sim 10^{+5}$ °K for Zn–Fe. This is in agreement with Schrieffer's theory[24] since Zn–Fe is a "nonmagnetic" alloy.[27] A more detailed comparison with experiment will be included in a further publication when the calculations of Bloomfield and Hamann[10] become available. We conclude that the best result at present for T_c in dilute magnetic alloys is obtained by use of Hamann's solution[7] of Nagaoka's equations[4] in the normal state. This result bears no resemblance to the expression for T_c obtained in I by use of the method of Takano and Ogawa[3]. This is to be expected since Takano and Ogawa's solution for normal state of dilute magnetic alloys is very unreliable and in disagreement with every other solution. In addition to this, Hamann's approximate solution[7] does not appear to contain the anomalous resonance[4,5] in Nagaoka's solution for $T \ll T_K$.[10] The exact solution of Nagaoka's equation due to Hamann and Bloomfield[10] seems to include terms due to such a resonance. It is hoped that these terms will appear in our expression for T_c when the t-matrix of Ref. 10 is substituted into equation (3.40). Finally we reiterate that the s–d exchange interaction with momentum-independent matrix elements between conduction electron states may only be valid for a spin-$\frac{1}{2}$ impurity and that extension to higher spins requires inclusion at such momentum dependence in the formalism.[7]

[26] G. Boato, G. Galinaro, and C. Rizzuto, Phys. Rev. **148**, 353 (1966).

[27] It is also possible to obtain the effective magnetic moment μ_{eff} on the Fe impurity in dilute Al–Mn at temperature $T = T_{c0}$ from (5.4) and the data of Ref. 26. The result is that $\mu_{\text{eff}}(T_{c0}) = \mu/5$ where μ is the bare moment of the ion. There is no evidence at present for the existence of such a moment which is required if Schrieffer (Ref. 24) is correct. Measurements of the susceptibility of Al–Mn should resolve this question.

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