

Effect of Anderson magnetic impurities on superconductivity

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For the purpose of clarifying the Kondo effect in superconductors, in particular, at lower temperatures than a characteristic temperature T_K , a perturbation calculation is performed within the Anderson model. In a microscopic Fermi-liquid theory which accurately describes the low-energy excitations, we consider the transition temperature T_c and the jump in the specific heat ΔC of the alloys. The energy dependence of the magnetic scattering generally leads to a finite tail in the concentration dependence of T_c , which suggests the existence of the *third* transition back to a superconducting state at a low temperature in superconducting alloys showing a "reentrance" phenomenon. Alloys with high Kondo temperatures are found to be well described by a nonmagnetic Hartree-Fock solution with renormalized parameters. In addition, expansion formulas for the four-vertex parts of the electron scattering are derived near the Fermi surface.

I. INTRODUCTION

Dilute magnetic impurities which can be generally described by the Anderson model are known to suppress the superconductivity through their resonance nature as well as through the repulsive interaction acting on them. Early works based on the Hartree-Fock (HF) theory clarified their effect in the "nonmagnetic" case,¹⁻³ while in the "magnetic" case³ the spins carried by the impurities have been treated classically⁴ without taking the Kondo effect into account.

The critical boundary in the HF theory between the magnetic and the nonmagnetic states is a consequence of the approximation that completely neglects all fluctuations. By contrast, studies of the Kondo effect have shown gradual disappearance of the local moment of magnetic impurities at low temperatures. Therefore, one can expect a continuous change from the nonmagnetic resonant state to the collective singlet state of the s - d exchange system⁵ as the repulsive interaction increases beyond the HF boundary⁶; the antiferromagnetic s - d exchange system is nothing but the strong coupling limit of the Anderson Hamiltonian system⁷. This picture has become much clearer in a recent microscopic theory developed by Yamada and Yosida whose general considerations are based on a perturbation calculation in the Anderson model.^{8,9} Moreover an accurate numerical analysis carried out in Wilson's renormalization-group theory¹⁰ has also confirmed the smooth magnetic transition. Experimentally, worth mentioning is a typical superconducting alloy, $(La, Th)Ce$ which continuously changes the magnetic character according to the constituents of the matrix.¹¹

Theoretical investigations of the Kondo effect in superconductors have been mainly approached from the magnetic side, i.e., by discussing the s - d Ham-

iltonian. A remarkable success was a prediction by Müller-Hartmann and Zittartz (MZ)¹² as well as Ludwig and Zuckermann¹³ of the so called reentrance phenomenon from the superconducting state back into the normal state with decreasing temperature. The origin of the phenomenon, which is actually observed in certain alloys,^{11,14} can be understood as the increase of magnetic scattering of conduction electrons around the Kondo temperature T_K .

However, the effect of the nonmagnetic state at sufficiently lower temperatures than T_K has so far remained unclear, because of the difficulty in describing the low-energy excitations.¹⁵ Although the magnetic scattering vanishes at $T=0$ and on the Fermi surface, the scattering off the Fermi surface has always nonzero values, and is enhanced by the Kondo effect around $\omega \sim T_K$ (ω : energy measured from the Fermi surface). Therefore the energy dependence of the scattering is indispensable in describing the superconductivity whose properties are determined by the electronic states within the momentum shell $|\omega| < \omega_D$ (ω_D : the Debye frequency). If one replaces the energy-dependent magnetic scattering by the value at $\omega=0$, then the effect of impurities on the superconductors at $T=0$ disappears.^{12,13} This is the reason why the theories in Refs. 12 and 13 could not yield any critical impurity concentration above which $T_c=0$. In their theories the prediction of the third transition at low temperature is a direct consequence of the approximation. Recent attempts to take the energy dependence into account have not yet been successful for the $T \ll T_K$ region, because of the unsatisfactory description of the low-frequency, low-temperature region^{16,17} and/or the way of performing the frequency integration invoking an extra unknown parameter.¹⁷

These difficulties will be naturally overcome in

this paper by approaching the problem from the nonmagnetic side of the Anderson Hamiltonian.⁴⁸ We apply the Yamada-Yosida theory developed in the normal state^{8,9} to discuss the impurity effect on superconductors. This theory which takes the repulsive interaction as a perturbation has been formulated quite generally and is free from the HF singularity. It can be regarded as a microscopic version of the phenomenological Fermi-liquid theory proposed by Nozières for the s - d system.¹⁸ As the theory is exact for low-lying excitations, we can best describe the high- T_K superconducting alloys where discrepancies between experiments and existing theories have long been pointed out.^{11,23,34}

In this paper we concentrate on the nonmagnetic temperature region lower than a characteristic temperature (T_K , in the s - d limit) determined by the interaction strength. Our results are expected to be correct if $T \lesssim \frac{1}{3} T_K$. In our treatment the initial transition temperature T_∞ can be comparable to or even higher than the characteristic temperature, say T_K ; in this case we can reach the region $T_c(c) \ll T_K$ by increasing the impurity concentrations c . We try to clarify how the "nonmagnetic" state of magnetic impurities generally suppresses the superconductivity. Several controversial points will be discussed, in particular, the critical concentration at which the superconducting state terminates, the third transition of the "reentrant" alloys (again back to the superconducting state at a very low temperature), the BCS law of corresponding states and applicability of the nonmagnetic HF theory.²³

We will find that the repulsive interaction at the impurities strongly influences superconductivity through two essential effects; firstly, it causes narrowing of the impurity level related to the Kondo effect, and secondly, it scatters a pair of conduction electrons inelastically. In the language of the s - d system the latter may be called an effective interaction mediated by the virtual polarization of the singlet impurities.¹⁹ In a simple modification of the Abrikosov-Gorkov (AG) theory²⁰ or in a conventional spin fluctuation model²¹ the latter process is missing, while from the HF theory of the Anderson model no narrowing effect is expected. On the other hand, both effects have been taken into account in the Yamada-Yosida theory.^{8,9}

In the s - d limit, Matsuura, Ichinose and Nagaoka recently proposed an interpolation theory to discuss the magnetic and nonmagnetic regions.¹⁹ Since they used the same Fermi-liquid description for the low-energy excitations their results mainly agree with ours taken in the limit. In the present paper treating the Anderson model we do not consider the crossover into the magnetic region, $T > T_K$, because a quantitatively reliable theory

seems extremely difficult to construct. Qualitatively, the results of many authors in the s - d system agree in the magnetic region and even in the crossover region, and indeed some of them^{12,16,22} have provided reasonable explanations for the experimental data^{11,14} within these regions.

After some preliminaries in the following section we consider in Sec. III the dilute case, calculating the initial decrease in the transition temperature and the jump in the specific heat. The concentration dependence of the transition temperature $T_c(c)$ is discussed in Sec. IV. Existence of the critical concentration and the third transition associated with a finite tail of the $T_c(c)$ curve will be found. In the final section we show that our results can answer several problems posed by experimentalists.^{11,23} For example, we will point out the similarity in the functional form of our results to the nonmagnetic HF ones. In this connection, expressions for some other physical quantities of high- T_K superconducting alloys are presented. Useful formulas for the vertex parts in the Fermi-liquid expansion are given in Appendix A.

II. PRELIMINARIES

We consider a BCS superconductor containing dilute magnetic impurities characterized by the Anderson model without orbital degeneracy. The Hamiltonian is

$$\begin{aligned} H(\Delta) = & \sum_{k\sigma} \epsilon_k a_{k\sigma}^\dagger a_{k\sigma} - \left(\Delta \sum_{\mathbf{k}} a_{\mathbf{k}\uparrow}^\dagger a_{\mathbf{k}\downarrow}^\dagger + \text{H.c.} \right) \\ & + \sum_{\mathbf{k}i\sigma} (V e^{-i\mathbf{k}R_i} a_{k\sigma}^\dagger d_{i\sigma} + \text{H.c.}) \\ & + E_d \sum_{i\sigma} n_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \end{aligned} \quad (2.1)$$

where $a_{k\sigma}$ and $a_{k\sigma}^\dagger$ are, respectively, annihilation and creation operators for a conduction electron with momentum k , spin σ , and kinetic energy $\epsilon_k (= \epsilon_{-\mathbf{k}})$, and Δ is the order parameter of the superconducting state. The magnetic impurities are assumed randomly distributed at sites R_i , where a repulsive interaction U works between impurity electrons with opposite spins. The impurity electrons are represented by operators $d_{i\sigma}$, $d_{i\sigma}^\dagger$ ($n_{i\sigma} = d_{i\sigma}^\dagger d_{i\sigma}$) and are coupled with the conduction electrons through the mixing matrix V . We will choose the impurity level E_d to be $-\frac{1}{2}U$ so as to treat the symmetric case, in which the impurity system in the strong coupling limit, $U \rightarrow \infty$, becomes equivalent to the s - d exchange system with an antiferromagnetic coupling constant,

$$J = -8V^2/U. \quad (2.2)$$

We treat the repulsive interaction U perturbationally, by choosing as unperturbed state the nonmag-

netic resonant state at the Fermi level with width $\Gamma = \pi\rho V^2$ (ρ : the density of states of the conduction electrons at the Fermi level). Since we are not making any HF-type approximation on the impurity site, anomalous averages such as $\langle d^\dagger d^\dagger \rangle$ used in the Ratto-Blandin theory¹ are not invoked in (2.1) from the beginning.

Neglecting the spatial dependence, the order parameter is determined self-consistently through

$$\Delta^\dagger = g \sum_k \langle a_{k\dagger}^\dagger a_{-k\dagger}^\dagger \rangle_s = g \sum_k T \sum_{\omega_n} \mathcal{F}_k(i\omega_n), \quad (2.3)$$

where g is the effective attractive interaction be-

$$\Delta^\dagger = g \sum_{k_1 k_2} \left(\int_0^\beta d\tau_1 \langle T_\tau a_{k_1\dagger}^\dagger(0) a_{-k_1\dagger}^\dagger(0) a_{-k_1\dagger}(\tau_1) a_{k_1\dagger}(\tau_1) \rangle + \sum_{k_2 k_3} \int_0^\beta d\tau_1 d\tau_2 d\tau_3 \langle T_\tau a_{k_1\dagger}^\dagger(0) a_{-k_1\dagger}^\dagger(0) a_{-k_1\dagger}(\tau_1) a_{k_1\dagger}(\tau_1) \right. \\ \left. \times a_{k_2\dagger}^\dagger(\tau_2) a_{-k_2\dagger}^\dagger(\tau_2) a_{-k_3\dagger}(\tau_3) a_{k_3\dagger}(\tau_3) \right) |\Delta|^2 + \dots \Delta^\dagger, \quad (2.5)$$

with $a(\tau) = e^{H\tau} a e^{-H\tau}$ and $H_n \equiv H(\Delta=0)$. Hereafter, the average is taken in the normal state of the alloy.

After extracting terms referring to the pure superconductor we can represent the impurity effect by two quantities $A(c, T)$ and $B(c, T)$ in the following equation,

$$\ln \frac{T_{c0}}{T} = A(c, T) + \lambda(3) \left(\frac{| \Delta(T) |}{\pi T} \right)^2 [1 + B(c, T)], \quad (2.6)$$

where $\lambda(n) = \sum_{l=0}^{\infty} (2l+1)^{-n}$ and $T_{c0} = (2\gamma/\pi)\omega_D e^{-1/\lambda}$, with $\gamma = 1.781$ and $\lambda = g\rho$.

For alloys the transition temperature T_c and the jump in the specific heat ΔC at the phase boundary are given by

$$\ln(T_{c0}/T_c) = A(c, T_c) \quad (2.7)$$

and

$$\frac{\Delta C}{\Delta C_0} = \frac{T_c}{T_{c0}} \frac{1}{1+B(c, T_c)} \left(1 + T_c \frac{\partial A(c, T_c)}{\partial T_c} \right)^2, \quad (2.8)$$

respectively³. Here T_{c0} and ΔC_0 are the initial values of the pure system, where $\Delta C_0 = 1.42\gamma_n T_{c0}$, $\gamma_n = (2\pi^2/3)k^2\rho_s$ holds according to the BCS theory (k is the Boltzmann constant, $\rho_s \equiv \rho/N$, N is the total number of the atomic sites).

The quantities $A(c, T)$ and $B(c, T)$ are obtained in this paper through a perturbational analysis of (2.5) with respect to the s - d mixing V and the repulsive interaction U as well as the impurity concentration c ($=N_i/N$; N_i is the number of the impurities). At this point it is convenient to introduce some functions needed in the perturbational treatment⁸: (i) *The thermal Green's function (GF) of the unperturbed conduction electron,*

$$G_k(i\omega_n) = (i\omega_n - \epsilon_k)^{-1}; \quad (2.9)$$

tween the conduction electrons in the pure system, $\langle a_{k\dagger}^\dagger a_{-k\dagger}^\dagger \rangle_s$ is the average taken in the superconducting state and $\mathcal{F}_k(i\omega_n)$ is the anomalous Green's function defined by

$$\mathcal{F}_k(i\omega_n) = \int_0^\beta d\tau \langle T \bar{a}_{k\dagger}^\dagger(\tau) \bar{a}_{-k\dagger}^\dagger(0) \rangle_s e^{i\omega_n \tau} \quad (2.4)$$

with $\bar{a}(\tau) = e^{H\tau} a e^{-H\tau}$, $\omega_n = (2n+1)\pi T$, n integer. The summations over k in (2.3) are taken within a momentum shell, $|\epsilon_k| < \omega_D$ (Debye frequency). The self-consistent equation (2.3) is expanded near the transition temperature in terms of Δ up to third order

(ii) *the thermal GF of the perturbed d electron,*

$$G_d(i\omega_n) = (i\omega_n + i\Gamma \operatorname{sgn} \omega_n - \Sigma(i\omega_n))^{-1}, \quad (2.10)$$

where $\Sigma(i\omega_n)$ is the self-energy due to the repulsive interaction U . In the symmetric case of the Anderson model, it is known that $G_d(i\omega_n)$ is a pure-imaginary odd function on the imaginary axis [cf. Eq. (2.11)].

(iii) *the vertex part $\Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m)$, which represents the inelastic scattering of a pair of d electrons with opposite spins from one energy state $(i\omega_n, -i\omega_n)$ to another $(i\omega_m, -i\omega_m)$ (see Fig. 1). It is defined including the reducible parts. We will choose the sign of the vertex function so that $\Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m) = U + \dots$ holds in the lowest approximation. The exact expressions for the self-energy and the vertex part have been obtained by Yamada and Yosida^{8,9} near the Fermi surface in frequency*

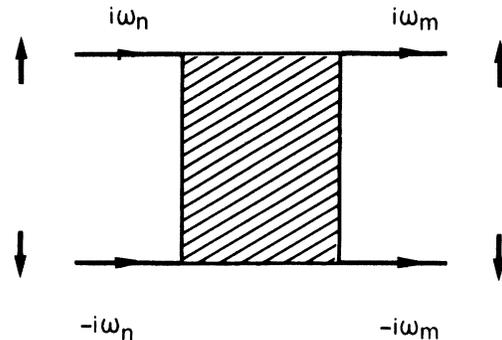


FIG. 1. Vertex part $\Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m)$ representing the inelastic scattering of a d -electron pair with different spins.

expansions:

$$\Sigma(i\omega_n) = -(\tilde{\chi}_{\uparrow\uparrow} - 1)i\omega_n - \frac{i \operatorname{sgn}\omega_n}{2\Gamma} \tilde{\chi}_{\uparrow\downarrow}^2 [(i\omega_n)^2 + (\pi T)^2] + \dots, \quad (2.11)$$

$$\Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m) = \pi\Gamma \tilde{\chi}_{\uparrow\downarrow} \left(1 - \frac{\tilde{\chi}_{\uparrow\uparrow}}{\Gamma} |\omega_n + \omega_m| + \dots\right). \quad (2.12)$$

The ω -linear term in (2.12) is new and the derivation is given in Appendix A. The coefficients $\tilde{\chi}_{\sigma\sigma'}$ are generalized susceptibilities defined by the following correlation functions between two electrons with spins σ and σ' ($= \pm\sigma$),

$$\tilde{\chi}_{\sigma\sigma'} = \pm\pi\Gamma \int_0^\beta d\tau \langle [n_\sigma(\tau) - \langle n_\sigma \rangle] [n_{\sigma'}(0) - \langle n_{\sigma'} \rangle] \rangle. \quad (2.13)$$

They have been perturbationally calculated up to the fourth order in $u \equiv U/\pi\Gamma$ in the symmetric case of the Anderson model.⁸ $\tilde{\chi}_{\uparrow\uparrow}(\tilde{\chi}_{\uparrow\downarrow})$ starts from 1(u) and increases with u smoothly but rapidly approaching $\pi\Gamma/4T_K$, that is half of the normalized susceptibility ($\tilde{\chi}_{\uparrow\uparrow} + \tilde{\chi}_{\uparrow\downarrow}$) of the s - d interacting sys-

tem. Here, T_K the Kondo temperature is, by definition, the inverse of the susceptibility at $T=0$, in the units of $g=2$, $\mu_B=1$, and coincides with the binding energy of the singlet ground state in the Yosida-Yoshimori theory.⁵ T_K is proportional to $\exp(-\frac{1}{8}\pi^2 u)$ for large u if we take the result, $T_K \propto \exp(-1/|J\rho|)$, which is known in the s - d model as the leading approximation.

III. INITIAL DECREASE IN THE TRANSITION TEMPERATURE AND THE SPECIFIC-HEAT JUMP

Let us first look at the dilute limit keeping only linear terms with respect to impurity concentration.

A. Transition temperature T_c

As shown in (2.7), T_c is determined by $A(c, T)$, which is given by the two-particle Green's function of a pair of electrons in (2.5). The effect of a single impurity is perturbationally analyzed as

$$\begin{aligned} \frac{\rho_s A(c, T)}{c} = & -V^2 \left[2T \sum_n \sum_k F_k(i\omega_n) G_k(i\omega_n) G_d(i\omega_n) + V^2 T \sum_n \left(\sum_k F_k(i\omega_n) \right)^2 G_d(i\omega_n) G_d(-i\omega_n) \right. \\ & \left. - V^2 T^2 \sum_{nm} \left(\sum_k F_k(i\omega_n) \right) \left(\sum_{k'} F_{k'}(i\omega_m) \right) G_d^2(i\omega_n) G_d^2(i\omega_m) \Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m) \right], \end{aligned} \quad (3.1)$$

where

$$F_k(i\omega_n) = G_k(i\omega_n) G_{\bar{k}}(-i\omega_n) = (\omega_n^2 + \epsilon_k^2)^{-1}. \quad (3.2)$$

The first two terms of the right-hand side (rhs) of (3.1) represent the mixing of a conduction electron with a d electron which is perturbed by another d electron. The third term indicates a process in which both of the paired conduction electrons jump into a d -level and repulsively interact with each other. The former scatterings are elastic, while the latter is generally inelastic.

These processes are shown diagrammatically in Figs. 2(a)–2(c), where the thin (thick) lines represent unperturbed conduction- (perturbed d -) electron Green's functions and the hatched square represents the vertex part of the d -electron scattering. The summations over k within $|\epsilon_k| \leq \omega_D$ yield

$$\sum_k F_k(i\omega_n) = \frac{2\rho}{\omega_n} \tan^{-1} \frac{\omega_D}{\omega_n} \sim \frac{\pi\rho}{|\omega_n|}, \quad \omega_D > |\omega_n|, \quad (3.3)$$

and

$$\sum_k F_k(i\omega_n) G_k(i\omega_n) \sim -i \frac{\pi\rho}{2} \frac{\operatorname{sgn}\omega_n}{|\omega_n|^2}, \quad \omega_D > |\omega_n|. \quad (3.4)$$

Then

$$\begin{aligned} \frac{\rho_s A(c, T)}{c} = & T \sum_n \frac{1}{\omega_n^2} \tilde{G}_d(i\omega_n) [1 - \tilde{G}_d(i\omega_n)] \\ & + \frac{\pi}{\Gamma} T^2 \sum_{nm} \frac{\tilde{G}_d^2(i\omega_n) \tilde{G}_d^2(i\omega_m)}{|\omega_n| |\omega_m|} \\ & \times \tilde{\Gamma}_{\uparrow\downarrow}^d(i\omega_n, i\omega_m), \end{aligned} \quad (3.5)$$

with the frequency summations taken within the Debye cut off, $|\omega_n| \lesssim \omega_D$. Here we have introduced normalized functions \tilde{G}_d and $\tilde{\Gamma}_{\uparrow\downarrow}^d$ by

$$\tilde{G}_d(i\omega_n) = G_d(i\omega_n) / G_d(i0 \operatorname{sgn}\omega_n) \quad (3.6)$$

and

$$\tilde{\Gamma}_{\uparrow\downarrow}^d(i\omega_n, i\omega_m) = \Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m) / (\pi\Gamma), \quad (3.7)$$

respectively.

Because of the singular nature of the summand, the major contribution to $A(c, T)$ at low temperatures comes from the low-frequency region. Therefore we substitute the expansions for the self-energy and the vertex part near the Fermi surface, (2.11), (2.12) into (3.5). Then one of

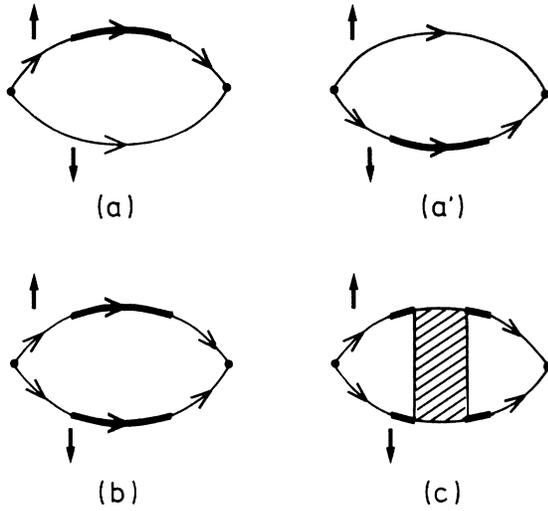


FIG. 2. Diagrammatic representations of the terms in Eq. (3.1), which contribute to the initial decrease in T_c . The processes (a), (a'), and (b) contain mixing effect and elastic scatterings while (c) does inelastic one.

the singularities in (3.5) is cancelled by

$$\bar{\alpha}(i\omega) \equiv \bar{G}_d(i\omega)[1 - \bar{G}_d(i\omega)] \sim |\omega_n|/\bar{\Gamma} + \dots, \quad |\omega_n| < \bar{\Gamma}, \quad (3.8)$$

where $\bar{\Gamma} \equiv \Gamma/\bar{\chi}_{\uparrow\uparrow}$. The result for $\Delta T_c \equiv T_c - T_{\infty}$ is

$$\frac{|\Delta T_c|}{cT_{\infty}} = \frac{1}{\pi\rho_s\bar{\Gamma}} \left\{ \frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \left[\ln \frac{2\gamma\omega_D}{\pi T_{\infty}} - \left(2 + \frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \right) \frac{\omega_D}{\bar{\Gamma}} \right]^2 + \left\{ \ln \frac{2\gamma\omega_D}{\pi T_{\infty}} - \left[2 - \frac{3}{2} \left(\frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \right)^2 \right] \frac{\omega_D}{\bar{\Gamma}} \right\} + \frac{\pi^3}{8} \left(\frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \right)^2 \frac{T_{\infty}}{\bar{\Gamma}} \right\}, \quad (3.9)$$

where we assume $\bar{\Gamma} \gg \omega_D, T_{\infty}$; and $\gamma = 1.78\dots$. This is exact up to the linear term of $\omega_D/\bar{\Gamma}$ and $T_{\infty}/\bar{\Gamma}$. The first and the second terms in (3.9) represent the inelastic and elastic scattering, respectively, while the last one comes from the explicit temperature dependence of the self-energy (2.11), which gives a small contribution unless $\bar{\Gamma} \sim T_{\infty}$.

With increasing u , $\bar{\chi}_{\uparrow\uparrow}$ and $\bar{\chi}_{\downarrow\downarrow}$ become large; then, if $\omega_D \sim \Gamma/\bar{\chi}$ the expansions up to the first few terms in (2.11) and (2.12) are insufficient for making precise calculations. In this case, however, we can estimate the integration in (3.5) from a general consideration on the shape of $G(i\omega)$. It has a peak at the Fermi surface $\omega = 0$, and as shown by Yamada,⁸ this peak narrows rapidly with interaction strength U . The effective width of $G(i\omega)$ is estimated as $\bar{\Gamma}$. When $u \geq 3$, two side peaks corresponding to the local moment begin to appear at $\omega \approx \frac{1}{2}U$; however, since they are far

from the Debye cut off, i.e., $\omega_D \ll \Gamma < U$ in usual situation, we can neglect them at low temperatures $T_{\infty} < \bar{\Gamma}$, where low frequencies dominate the integration. By taking into account just the central peak we can replace the Debye cutoff by $\bar{\Gamma}$ if $\bar{\Gamma} < \omega_D$. Then we are led to a result for $T_{\infty} \ll \bar{\Gamma}$,

$$\frac{|\Delta T_c|}{cT_{\infty}} = \frac{1}{\pi\rho_s\bar{\Gamma}} \left[\frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \ln^2 \frac{2\gamma[\omega_D, \bar{\Gamma}]}{\pi T_{\infty}} + \ln \frac{2\gamma[\omega_D, \bar{\Gamma}]}{\pi T_{\infty}} + \frac{\pi^3}{8} \left(\frac{\bar{\chi}_{\uparrow\uparrow}}{\chi_{\uparrow\uparrow}} \right)^2 \frac{T_{\infty}}{\bar{\Gamma}} \right], \quad (3.10)$$

where

$$[\omega_D, \bar{\Gamma}] \equiv (\omega_D^{-1} + \bar{\Gamma}^{-1})^{-1} \approx \min[\omega_D, \bar{\Gamma}]. \quad (3.11)$$

In the following the numerical constants within the argument of the logarithm should only be considered to be approximately correct. The initial decrease of T_c given by (3.10) is plotted in Fig. 3 as a function of the interaction strength u or the effective width $\bar{\Gamma}$. In obtaining this result we have related u to $\bar{\chi}_{\uparrow\uparrow}$ using the perturbation result⁸ if $u \leq 3$ and putting $\bar{\chi}_{\uparrow\uparrow} = \cosh u$ if $u > 3$. We have also approximated for simplicity, $\bar{\chi}_{\uparrow\uparrow}/\chi_{\uparrow\uparrow} = \tanh u$ everywhere.⁸ One observes a rapid increase in $|\Delta T_c|$ with u which is in strong contrast to the nonmagnetic HF result.¹⁻³

If we take the s - d limit, (3.10) becomes

$$\Delta T_c = -\frac{cT_{\infty}}{4\rho_s T_K} \left(\ln \frac{8\gamma e^{1/2}}{\pi^2} \frac{T_K}{T_{\infty}} \right)^2, \quad (3.12)$$

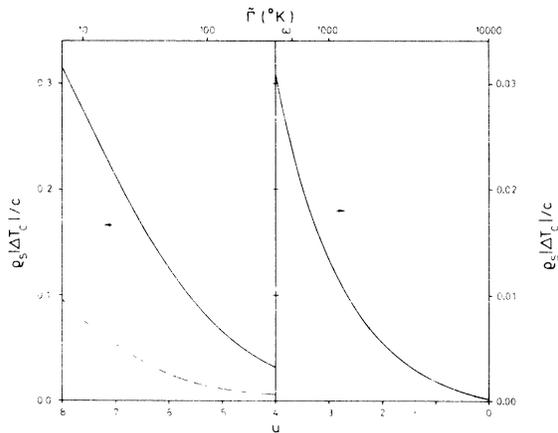


FIG. 3. Initial decrease in T_c [Eq. (3.10)] as a function of the interaction strength $u [u = U/(\pi\Gamma)]$ or the effective width $\bar{\Gamma}$ of the d -electron state density. The initial values are chosen $T_{c0} = 1^\circ\text{K}$, $\Gamma = 10000^\circ\text{K}$ with $\omega_D = 500^\circ\text{K}$. Dash-dot curve shows the contribution from the elastic scattering. Dotted curve shows a result of the nonmagnetic HF theory. Different scales are used for $u \geq 4$.

for $T_\infty \ll T_K < \omega_D$. The T_K/T_∞ dependence in (3.12), which we think is exact, is in contrast to the expression obtained within the Suhl-Nagoaka approximation.²⁴ The latter is inversely proportional to $\ln^2(T_K/T_\infty)$ for high- T_K superconducting alloys. Essentially the same result as (3.12) was recently obtained by Matsuura, Ichinose and Nagaoka.¹⁹

B. Specific-heat jump ΔC

The jump in the specific heat ΔC at the phase transition is given relative to T_c through the relation (2.8). We calculate the relative ratio $(\Delta C/\Delta C_0)/(T_c/T_{c0})$ up to leading terms neglecting higher order ones which vanish when $T_c \rightarrow 0$. We assume the dilute limit and $T_c \ll \tilde{\Gamma}$. One of the factors in (2.8) is easily calculated from the obtained result (3.9):

$$\begin{aligned} & \left(1 + T_c \frac{\partial A(c, T_c)}{\partial T_c}\right)^2 \\ &= 1 - \frac{4c}{\pi \rho_s \tilde{\Gamma}} \left\{ \frac{\tilde{\chi}_{\uparrow\uparrow}}{\tilde{\chi}_{\uparrow\downarrow}} \left[\ln \frac{2\gamma \omega_D}{\pi T_{c0}} - \left(2 + \frac{\tilde{\chi}_{\uparrow\uparrow}}{\tilde{\chi}_{\uparrow\downarrow}}\right) \frac{\omega_D}{\tilde{\Gamma}} \right] + \frac{1}{2} \right\}. \end{aligned} \quad (3.13)$$

The other factor in (2.8) contains $B(c, T_c)$ defined by (2.6), which is examined by means of a perturbation expansion of the four-particle Green's function in (2.5). For convenience we describe perturbed Green's functions of conduction electrons in the presence of a single impurity by

$$\begin{aligned} g_{kk'}(i\omega_n) &= G_k(i\omega_n) \delta_{kk'} + V^2 G_R(i\omega_n) \\ &\quad \times G_d(i\omega_n) G_{k'}(i\omega_n). \end{aligned} \quad (3.14)$$

$$\begin{aligned} T \sum_\omega \left(\sum_{k_1 \sim k_4} g_{k_1 k_2}(i\omega) g_{k_2 k_3}(-i\omega) g_{k_3 k_4}(i\omega) g_{k_4 k_1}(-i\omega) - \sum_k F_k^2(i\omega) \right) \\ = -T \sum_\omega \frac{1}{\omega^4} \tilde{G}_d(i\omega) [1 - \tilde{G}_d(i\omega)] [\tilde{G}_d^2(i\omega) - \tilde{G}_d(i\omega) + \frac{3}{2}] = \frac{\rho\lambda(3)}{(\pi T)^2} \left(-\frac{3}{\pi\rho\tilde{\Gamma}} + O(T) \right). \end{aligned} \quad (3.15)$$

(b) Terms including self-energy corrections and one vertex correction between a pair of electrons with opposite spins:

$$\begin{aligned} & -4V^4 T^2 \sum_{\omega, \omega'} \sum_{k_1 \sim k_4} F_{k_1}(i\omega) G_d^2(i\omega) \Gamma_{\uparrow\downarrow}^d(i\omega, i\omega') G_d^2(i\omega') G_{k_2}(i\omega') g_{k_2 k_3}(-i\omega') g_{k_3 k_4}(i\omega') G_{k_4}(-i\omega') \\ &= -\frac{\pi}{\tilde{\Gamma}} T^2 \sum_{\omega, \omega'} \frac{\tilde{\Gamma}_{\uparrow\downarrow}^d(i\omega, i\omega')}{|\omega||\omega'|^3} \tilde{G}_d^2(i\omega) \tilde{G}_d^2(i\omega') [\tilde{G}_d^2(i\omega) - \tilde{G}_d(i\omega') + \frac{1}{2}] \\ &= \frac{\rho\lambda(3)}{(\pi T)^2} \left\{ \frac{-4}{\pi\rho\tilde{\Gamma}} \frac{\tilde{\chi}_{\uparrow\downarrow}}{\tilde{\chi}_{\uparrow\uparrow}} \left[\ln \frac{2\gamma \omega_D}{\pi T} - \left(2 + \frac{\tilde{\chi}_{\uparrow\downarrow}}{\tilde{\chi}_{\uparrow\uparrow}}\right) \frac{\omega_D}{\tilde{\Gamma}} + O(T \ln T) \right] \right\}. \end{aligned} \quad (3.16)$$

Diagram representations of these terms are shown in Fig. 4(a) and 4(b). The terms in the parentheses in (3.15) and (3.16) give $B(c, T)$. Other terms in the expansion contain either more than two inde-

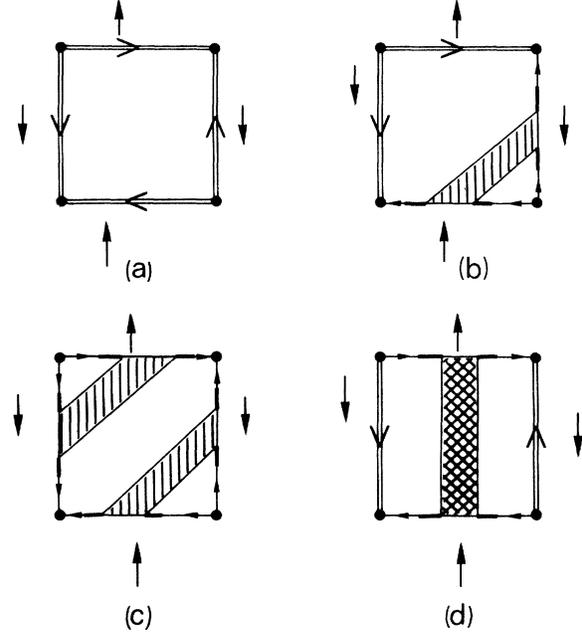


FIG. 4. Diagrammatic representations of the terms considered in the initial decrease in ΔC : (a) and (b) corresponding to Eqs. (3.15) and (3.16), respectively, make relevant contributions. (c) and (d) give higher-order corrections. Double lines are the perturbed Green's functions of conduction electrons, thin lines are for unperturbed conduction electrons and thick lines are for perturbed d electrons.

Among many terms in $-\langle T_\tau a^\dagger a^\dagger a a a^\dagger a^\dagger a a \rangle$ the following ones are found to give the dominant contributions:

(a) Terms including self-energy correction only:

pendent frequencies like $T \sum \omega_1^{-1} \omega_2^{-1} \omega_3^{-2}$ [Fig. 4(c)] or vertices of parallel-spin electron scattering, $\Gamma_{\uparrow\downarrow}^d(i\omega, i\omega')$ like $T \sum \omega_1^{-2} \omega_2^{-2} \Gamma_{\uparrow\downarrow}^d(i\omega_1, i\omega_2)$ [Fig. 4(d)]. Since this vertex vanishes on the Fermi surface⁹

(see also Appendix A) both terms make higher-order corrections to $B(c, T)$ and are at most of the order of $T^2 \ln T$. From (3.15) and (3.16) we obtain

$$B(c, T_c) = -\frac{4c}{\pi\rho_s\bar{\Gamma}} \left\{ \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \left[\ln \frac{2\gamma\omega_D}{\pi T_{c0}} - \left(2 + \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \right) \frac{\omega_D}{\bar{\Gamma}} \right] + \frac{3}{4} \right\}. \quad (3.17)$$

After partial cancellation with the similar term in (3.13) we finally get cutoff independent results, which is valid also for $\bar{\Gamma} < \omega_D$,

$$\frac{\Delta C}{\Delta C_0} \Big/ \frac{T_c}{T_{c0}} = 1 + \frac{c}{\pi\rho_s\bar{\Gamma}} = 1 + \frac{c\bar{\chi}_{\uparrow\uparrow}\rho_d}{\rho_s}, \quad (3.18)$$

where $\rho_d = (\pi\Gamma)^{-1}$ is the density of states of d electrons at the Fermi level. As the vertex corrections have cancelled, the ratio, $\Delta C/T_c$, is expressed exclusively by the self-energy correction at low frequencies, namely, the enhancement factor of the specific heat, $\bar{\chi}_{\uparrow\uparrow}$.⁸ We can rewrite the result (3.18) in a familiar form in the BCS theory,

$$\Delta C = 1.42 C_n(T_c) = 1.42 \gamma_{\text{alloy}} T_c, \quad (3.19)$$

where C_n is the electronic specific heat of the alloy in the normal state with an enhanced coefficient,

$$\gamma_{\text{alloy}} = \frac{2}{3} \pi^2 k^2 (\rho_s + c\bar{\chi}_{\uparrow\uparrow}\rho_d). \quad (3.20)$$

In this respect the law of the corresponding states holds in the alloy with $\bar{\Gamma} \gg T_{c0}$.

The initial slope of the ratio is given by

$$C^* \equiv \lim_{c \rightarrow 0} \frac{d\left(\frac{\Delta C}{\Delta C_0}\right)}{d\left(\frac{T_c}{T_{c0}}\right)} = 1 - \left[\ln \frac{2\gamma[\omega_D, \bar{\Gamma}]}{\pi T_{c0}} \left(1 + \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \ln \frac{2\gamma[\omega_D, \bar{\Gamma}]}{\pi T_{c0}} \right) \right]^{-1}. \quad (3.21)$$

The value C^* is known to be 1.436 for a well-defined magnetic moment (AG theory²⁰) and 1 for BCS-like superconducting alloys. The present result shows the nonmagnetic nature of the high- T_K impurities which is again in contrast to the MZ theory.²² Ichinose found the approximately same result for C^* in the s - d limit.²⁵

IV. CONCENTRATION DEPENDENCE OF THE CRITICAL TEMPERATURE

For finite concentrations the nature of the deviation from the linear concentration dependence reflects the magnetic properties of impurities:

well-defined magnetic moments are known to give rise to a negative curvature in $T_c(c)$, nonmagnetic impurities a positive one.

In this section we treat primitively the many-impurity problem neglecting possible interactions or interferences between impurities which are randomly distributed in the matrix. By taking an average over the impurity distribution we recover the momentum conservation for conduction electrons after repeated scattering.

First, let us introduce another Green's function associated with the conduction electrons

$$L(i\omega_n) = \lim_{\Delta \rightarrow 0} \sum_k \mathfrak{F}_k(i\omega_n) / \Delta, \quad (4.1)$$

in terms of which $A(c, T)$ given in (2.6) is expressed by

$$A(c, T) = -\rho^{-1} T \sum_{\omega_D > |\omega_n|} [L(i\omega_n) - L_0^0(i\omega_n)], \quad (4.2)$$

where $L_0^0(i\omega_n)$ refers to the Green's function of the pure system. Following the Abrikosov-Gorkov treatment^{19,20} we set up an integral equation for $L(i\omega)$ to describe the successive scattering of paired electrons by many impurities:

$$L(i\omega) = L_0(i\omega) \left(1 + N_i T \sum_{\omega'} K(i\omega, i\omega') L(i\omega') \right), \quad (4.3)$$

where N_i is the number of impurities and $L_0(i\omega)$ is defined including the self-energy correction due to d electron as

$$L_0(i\omega) = \sum_k [i\omega - \epsilon_k - N_i V^2 G_d(i\omega)]^{-1} \times [-i\omega - \epsilon_{-k} - N_i V^2 G_d(-i\omega)]^{-1} = \pi\rho [|\omega| + N_i V^2 |G_d(i\omega)|]^{-1}. \quad (4.4)$$

On the other hand, the integral kernel $K(i\omega, i\omega')$ is given by

$$K(i\omega, i\omega') = -V^4 G_d^2(i\omega) [T^{-1} \delta_{\omega, \omega'} + \Gamma_{\uparrow\uparrow}^d(i\omega, i\omega') G_d^2(i\omega')], \quad (4.5)$$

in which both the elastic and inelastic scattering of a pair of electrons at one impurity site are taken into consideration. To make the mathematical treatment easier we approximate the frequency-dependent vertex part by

$$\Gamma_{\uparrow\uparrow}^d(i\omega, i\omega') \approx \Gamma_{\uparrow\uparrow}^d(i0, i0) = \pi\Gamma\bar{\chi}_{\uparrow\uparrow}, \quad (4.6)$$

since we expect the major contribution to come from the vicinity of the Fermi surface. Then a formal solution of (4.3) is

$$\begin{aligned}
(\pi\rho)^{-1}L(i\omega) &= (\pi\rho)^{-1}L(i|\omega|) \\
&= H(i\omega) - \frac{c\bar{\chi}_{\uparrow\uparrow}}{\rho_s\Gamma} H^{(2)}(i\omega) T \sum_{\omega'} H^{(2)}(i\omega') \left(1 + \frac{c\bar{\chi}_{\uparrow\uparrow}}{\rho_s\Gamma} T \sum_{\omega'} H^{(4)}(i\omega') \right)^{-1}, \quad (4.7)
\end{aligned}$$

where

$$\begin{aligned}
H(i\omega) &= H(i|\omega|) \\
&= (\pi\rho)^{-1} [L_0^{-1}(i\omega) - N_{\uparrow} V^4 |G_d(i\omega)|^2]^{-1} \\
&= \{ |\omega| + c\alpha(i|\omega|) \}^{-1}, \quad (4.8) \\
\alpha(i|\omega|) &= (\pi\rho_s)^{-1} \bar{\alpha}(i|\omega|) \\
&\equiv (\pi\rho_s)^{-1} \bar{G}_d(i\omega) [1 - \bar{G}_d(i\omega)] \quad (4.9)
\end{aligned}$$

and

$$H^{(n)}(i\omega) \equiv H(i\omega) [\bar{G}_d(i\omega)]^n. \quad (4.10)$$

Due to the partial cancellation between the self-energy part in (4.4) and the first term of the integral kernel (4.5), $H(i\omega)$ is inversely proportional to $|\omega|$ near the Fermi surface. This fact turns out important in determining low-temperature phenomena. The transition temperature T_c is then calculated from (2.7), (4.2), and (4.7) as

$$\ln \frac{T_{c0}}{T_c} = -2\pi T_c \sum_{\omega_n > 0}^{\omega_D} \left(H(i\omega_n) - \frac{1}{\omega_n} \right) + \frac{c\bar{\chi}_{\uparrow\uparrow}}{\pi\rho_s\Gamma} \left(2\pi T_c \sum_{\omega_n > 0}^{\omega_D} H^{(2)}(i\omega_n) \right)^2 \left/ \left(1 + \frac{c\bar{\chi}_{\uparrow\uparrow}}{\pi\rho_s\Gamma} \times 2\pi T_c \sum_{\omega_n > 0}^{\omega_D} H^{(4)}(i\omega_n) \right) \right. \quad (4.11)$$

After carrying out the summations over the frequencies as shown in Appendix B, we obtain in the region $T_c \ll \bar{\Gamma}$, irrespective of the ratio of $\bar{\Gamma}/T_{c0}$,

$$\ln \frac{T_{c0}}{T_c} = \frac{\bar{c}}{1 + \bar{c}} \left(\ln \frac{2\gamma[\omega_D, W(\bar{c})]}{\pi T_c} + \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \frac{(\ln(2\gamma[\omega_D, \bar{\Gamma}]/\pi T_c))^2}{1 + \bar{c}(1 + (\bar{\chi}_{\uparrow\uparrow}/\bar{\chi}_{\uparrow\uparrow}) \ln(2\gamma[\omega_D, \bar{\Gamma}]/\pi T_c))} \right), \quad (4.12)$$

where

$$\bar{c} = c/(\pi\rho_s\bar{\Gamma}) \quad (4.13)$$

and a cutoff frequency, $W(\bar{c})$, is introduced by a condition

$$(\bar{c} + 2)^{-1} \approx \bar{\alpha}(i\omega)\bar{\Gamma}/\omega|_{\omega=W(\bar{c})}. \quad (4.14)$$

The frequency dependence of the impurity scattering at higher frequencies $\bar{\Gamma} < |\omega|$ has been phenomenologically taken into account. With the help of the Hamann's result²⁶ for the t matrix at high frequencies in the s - d system,²⁷ a reasonable approximation for the rhs of (4.14) is

$$\begin{aligned}
\bar{\alpha}(i\omega)\bar{\Gamma}/\omega &= 1/(1+x)^2, \quad x \leq 1 \\
&= 1/4x[(4/3\pi^2)\ln^2 x + 1], \quad x \geq 1, \quad (4.14')
\end{aligned}$$

with $x = \omega/\bar{\Gamma}$. The rhs of (4.14) and the concentration dependence of the cutoff $W(\bar{c})$ is plotted in Figs. 5 and 6, respectively. As long as $T_{c0} \leq \bar{\Gamma}$, \bar{c} is less than 10 (as seen later in Fig. 7), then $W(\bar{c})$ remains of the order of $\bar{\Gamma}$. If $T_{c0} \gg \bar{\Gamma}$, \bar{c} becomes extremely large and much higher frequencies region ($\omega \gg \bar{\Gamma}$) are involved. In the classical limit of the s - d system $\bar{\Gamma} \rightarrow 0$, we can show that $W(\bar{c})$ becomes $O(c(JM)^2\rho_s)$, which turns out to be $O(T_{c0})$ at the critical concentration.

At low temperatures the leading term in Eq. (4.12), $\ln T_c$, cancels out from both sides. Then the next leading term, the temperature-indepen-

dent one, fixes the critical concentration c_0 [$T_c(c_0) = 0$],²⁸ while the following term which is inversely proportional to $\ln T_c$ determines the concentration dependence of the transition temperature.

The situation might be compared with the corresponding expansion in the AG theory,²⁰

$$\ln \frac{T_{c0}}{T_c} = \ln \frac{2\gamma}{\pi T_c \tau_s} + \frac{\pi^2}{6} (\tau_s T_c)^2, \quad (4.15)$$

where

$$1/\tau_s = \frac{1}{2} c \rho_s \pi S(S+1)(JM)^2. \quad (4.16)$$

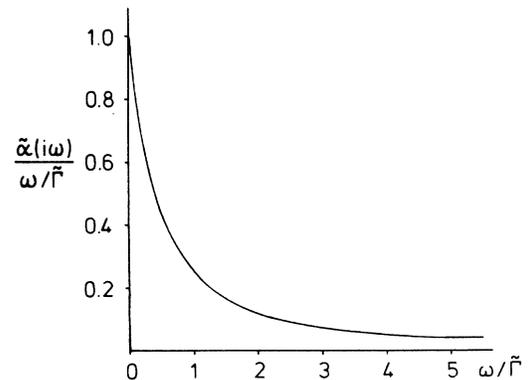


FIG. 5. Right-hand side of Eq. (4.14) as a function of the normalized frequencies $\omega/\bar{\Gamma}$.

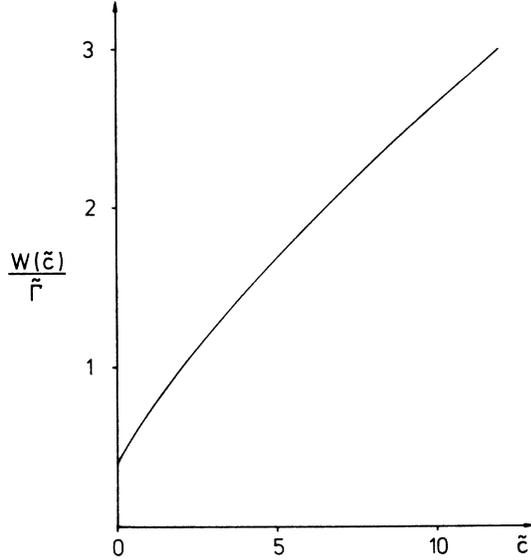


FIG. 6. Cutoff frequencies $W(\tilde{c})$ determined by the condition (4.14) as a function of the normalized concentration $\tilde{c} [=c/(\pi\rho_s\tilde{\Gamma})]$.

Equation (4.12) reduces to this result, in the classical limit $T_K \rightarrow 0$, by fixing $W(\tilde{c})$ as $1/\tau_s$,²⁹ and by including the higher-order temperature-dependent terms. In the AG theory T_c is proportional to $(c_0$

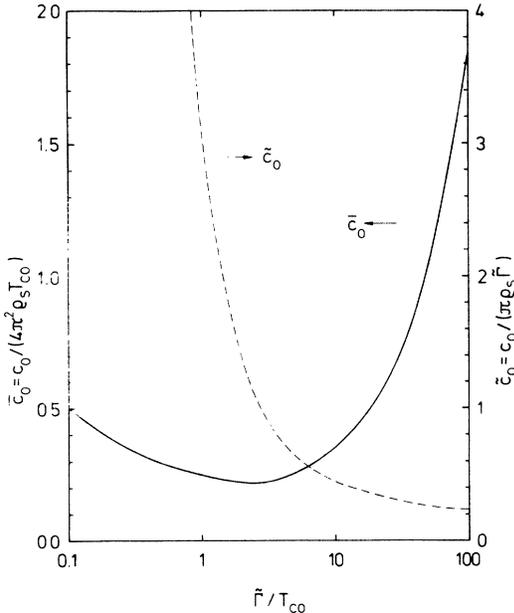


FIG. 7. Normalized critical concentration \tilde{c}_0 (solid curve) as a function of $\tilde{\Gamma}/T_{c0}$ ($\approx T_K/T_{c0}$). Dashed curve shows \tilde{c}_0 , the same critical concentration normalized by $\tilde{\Gamma}$. As long as \tilde{c}_0 remains small, low-frequency region is dominant in the integration (4.11). $T_c = 1^\circ\text{K}$, $\Gamma = 10\,000^\circ\text{K}$ and $\omega_D = 500^\circ\text{K}$.

$-c)^{1/2}$ near c_0 . On the other hand, our formula (4.12), which is rewritten more explicitly as

$$\frac{\pi T_c(c)}{2\gamma[\omega_D, \tilde{\Gamma}]} = \exp \left[\frac{\tilde{c}}{1+\tilde{c}} \frac{\tilde{\chi}_{\uparrow\uparrow}}{\tilde{\chi}_{\uparrow\downarrow}} - \left(\ln \frac{2\gamma[\omega_D, \tilde{\Gamma}]}{\pi T_{c0}} + \tilde{c} \ln \frac{2\gamma[\omega_D, W(\tilde{c})]}{\pi T_{c0}} \right)^{-1} \right]^{-1}, \quad (4.17)$$

gives, as shown later, a monotonically decreasing function $T_c(c)$ with positive curvature as long as $T_c(c) \ll \tilde{\Gamma}$.

The critical concentration c_0 is obtained from the zero of the parentheses in the rhs of (4.17) of (4.17).

In the limiting cases, (i) $\tilde{\Gamma} \gg T_{c0}$

$$c_0 \approx \frac{\pi\rho_s\tilde{\Gamma}}{\chi_{\uparrow\uparrow}} \ln^{-1} \frac{2\gamma[\omega_D, \tilde{\Gamma}]}{\pi T_{c0}}, \quad (4.18)$$

and (ii) $\tilde{\Gamma} \rightarrow 0$ (the AG limit)

$$c_0 \rightarrow T_{c0} / [\gamma\rho_s(JN)^2 S(S+1)]. \quad (4.19)$$

A numerical result for c_0 is plotted in Fig. 7 as a function of $\tilde{\Gamma}/T_{c0}$. The critical concentration c_0 has its minimum value around $\tilde{\Gamma}/T_{c0} \approx 2$, while the normalized one $\tilde{c}_0 (=c_0/(\pi\rho_s\tilde{\Gamma}))$ is a decreasing function of $\tilde{\Gamma}/T_{c0}$ except when u becomes small enough.

Equation (4.17) describes quite generally how the "nonmagnetic" state of the magnetic impurities suppresses the superconductivity. In particular, (i) for $T_{c0} \ll \omega_D < \tilde{\Gamma}$, Eq. (4.17) is valid in the whole range of the concentration, $0 < c < c_0$. We can rewrite it in a simpler form as

$$\frac{T_c}{T_{c0}} = \exp \left(\frac{-(A+B)c}{\lambda(1-Bc)} \right) \quad (4.20)$$

with $A = \tilde{\chi}_{\uparrow\uparrow} \rho_d/\rho_s$, $B = \tilde{\chi}_{\uparrow\uparrow} \rho_d/(\lambda\rho_s)$, and $\lambda = gN\rho_s$ (the coupling constant of the superconductor). If one puts $\tilde{\chi}_{\uparrow\uparrow} = 1$ and $\tilde{\chi}_{\uparrow\downarrow} = U_{\text{eff}} \rho_d$ [where $U_{\text{eff}} = U/(1+U\rho_d)$] (4.20) reduces to Kaiser's HF result,^{2,23} while the replacement, $\tilde{\chi}_{\uparrow\uparrow} = 1$ and $\tilde{\chi}_{\uparrow\downarrow} = U_{\text{eff}} \rho_d \exp(U\rho_d)$, corresponds to Schlottmann's recent result³⁰ obtained by a renormalization-group method, in which the self-energy correction has been neglected. The pair weakening effect caused by the inelastic repulsive interaction is the main mechanism suppressing the superconductivity and determines the critical concentration, at which the effective electron interaction changes its sign.¹⁹

(ii) For $T_{c0} < \tilde{\Gamma} < \omega_D$, as long as $\ln \tilde{\Gamma} \approx \ln W(\tilde{c})$,³¹ we can proceed parallel to case (i). Equation (4.20) approximately holds if one replaces λ by

$$\lambda' = \ln^{-1}(2\gamma[\omega_D, \tilde{\Gamma}]/\pi T_{c0}). \quad (4.21)$$

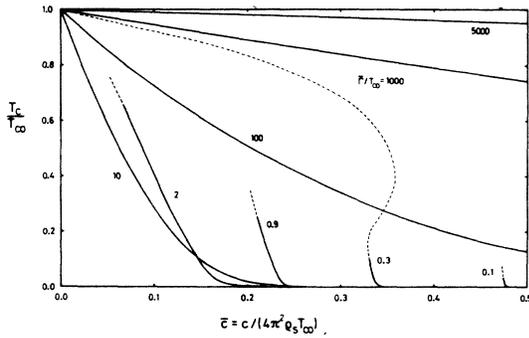


FIG. 8. Concentration dependence of the critical temperature for several values of $\bar{\Gamma}/T_{c0}$. Solid curves are results of our theory while dotted curves are their natural extension. In particular for $\bar{\Gamma}/T_{c0}=0.3$ an expected $T_c(c)$ curve showing three transitions is schematically illustrated. The same parameters are used as in Fig. 3 and 7.

(iii) For $\bar{\Gamma} \approx T_{c0} \ll \omega_D$, Eq. (4.17) is applicable only in a limited concentration range satisfying $T_c(c) \ll \bar{\Gamma}$. In this case, \bar{c} becomes larger with decreasing $\bar{\Gamma}$ and the \bar{c} dependence of the cut-off $W(\bar{c})$ cannot be neglected. [$\ln W(\bar{c}) \neq \ln \bar{\Gamma}$].

Figure 8 shows some numerical results of $T_c(c)$ given by (4.17) for various parameters of $\bar{\Gamma}/T_{c0}$. In all cases $T_c(c)$ has a finite tail decreasing exponentially near the critical concentration as

$$T_c(c) = \frac{2\gamma}{\pi} [\omega_D, \bar{\Gamma}] \exp\left(\frac{a}{\bar{\chi}_{\uparrow\downarrow}} \frac{\pi\rho_s\Gamma}{c-c_0}\right) \quad (4.22)$$

with constant a of the order of 1 except when $\Gamma \ll T_{c0}$. Therefore once a superconducting alloy happens to show a reentrance phenomenon at $T_c \sim T_K$, by cooling, it should show the *third* transition from the normal to the superconducting state

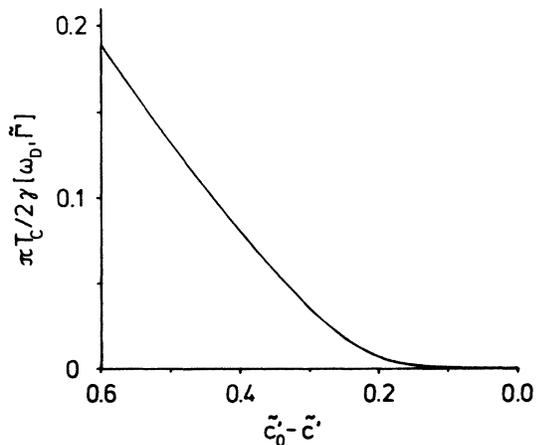


FIG. 9. Finite tail in the $T_c(c)$ curve near the critical concentration. The concentrations are normalized by $\pi\rho_s\Gamma/\bar{\chi}_{\uparrow\downarrow}$, i.e., $\bar{c}' = (\bar{\chi}_{\uparrow\downarrow}/\bar{\chi}_{\uparrow\downarrow})\bar{c}$.

in a certain range of concentration ($|c - c_0| \lesssim \pi\rho_s T_K \ll 1$). As seen from (4.22) and also from Fig. 8 the tail becomes shorter and steeper for small $\Gamma/\bar{\chi}_{\uparrow\downarrow} \sim T_K$, but if the concentration is scaled by $\pi\rho_s\Gamma/\bar{\chi}_{\uparrow\downarrow}$ then it is approximated by a universal function which is plotted in Fig. 9.

V. DISCUSSION

[A] The finite tail in $T_c(c)$ shown in the Sec. IV is a general feature of superconducting alloys containing magnetic impurities in the *nonmagnetic* state and occurs irrespective of the interaction strength $U/(\pi\Gamma)$. It is a consequence of the frequency dependence of the elastic impurity scattering $\alpha(\omega)$, which is linear in ω near the Fermi surface. Even in the case $T_K < T_{c0}$, where the scattering in a wider range of frequency contributes to the determination of the critical concentration c_0 , the temperature variation of the transition concentration $c(T_c)$ near c_0 [equivalent to $T_c(c)$] is dominated by the low-energy excitations.

In practice, interaction between impurities would have an influence in hindering the Kondo effect and preventing the impurity from falling down into the non-magnetic state. Probably, compared to the noninteracting case, the magnetic scattering would be (a) relatively reduced at $|\omega| \approx \bar{\Gamma}$, but (b) enhanced at $|\omega| \ll \bar{\Gamma}$ and would remain finite at $\omega=0$ and $T=0$. Consequently the interaction has a tendency to shorten the finite tail [it may increase the absolute value of c_0 but even if so, the $T_c(c)$ curve near c_0 would be steeper reflecting the magnetic character of the impurities].

Because of the shortness of the tail, precise measurements in a narrow concentration range are required to find out the third transition.³² Although the third transition has so far not been reported in the literature, a recent experiment by Winzer on $(La, Y):Ce$ supports the existence.³³ It would be desirable to invest more effort to reexamine other candidate alloys [such as $(La, Th):Ce$, $(LaAl_2):Ce$] systematically.

[B] In Fig. 8 we plot the $T_c(c)$ curves within the part for which our calculation is meaningful. Although the results are limited it would be natural to conclude the following from the figure:

(i) The maximum of the initial decrease in T_c occurs at a higher value of T_K/T_{c0} (≈ 5) compared to $(T_K/T_{c0})_m$ (≈ 2), the value which gives the minimum critical concentration c_0 .

(ii) The reentrant phenomenon (as well as the third transition) could be observed in superconducting alloys with relatively smaller value of T_K/T_{c0} (≈ 0.5) compared to $(T_K/T_{c0})_m$. Similar tendencies have been theoretically obtained by Schlottmann.¹⁶ Conclusion (ii) is in accord with the experimental

observation on $(La, Th):Ce$.

[C] Besides the third transition this calculation can answer several questions which have been raised by experimentalists.²³ Firstly, on $Al-Mn$, in the analysis of the early experiments Aoki and Ohtsuka³⁴ needed an unusually small width of the impurity level to explain the specific-heat coefficient and the rapid decrease in T_c . Their result is now understood in terms of the narrowing effect, $\Gamma \rightarrow \bar{\Gamma} = \Gamma/\bar{\chi}_{\uparrow\uparrow}$. If we take $u \approx 3$, then a reasonable reduction of Γ from 1 to 0.1 eV is expected.

Another point concerns the $T_c(c)$ curves observed by the Maple *et al.*³⁵ group on ThU and⁴¹ $(La, Th):Ce$ alloys. They found the applicability of the HF formula given by Kaiser² in a wider range than expected (not only purely nonmagnetic but including the high- T_K case).³⁶ The fact can be clarified by our general result, (4.20) and (4.21) for $\bar{\Gamma} > T_{c0}$, which has the same functional form as Kaiser's.

Moreover, as for the thermodynamic properties we can understand why the BCS law of corresponding states holds in high- (T_K/T_{c0}) alloys like ThU (Ref. 37) and $(La_{1-x}Th_x):Ce$ ($x \gtrsim 0.7$).⁴¹ In our theory, the initial slope $d(\Delta C/\Delta C_0)/d(\Delta T/\Delta T_{c0})$ approaches the BCS value instead of the AG value in the high- T_K limit. It should be noted that the resistance minimum, a typical Kondo effect of magnetic impurities observed at $T \gtrsim T_K$, is compatible with the BCS law at $T_c \ll T_K$.

[D] As shown in the text our results for T_c and ΔC , (3.10), (3.18), and (4.20) have the same functional forms as those obtained in the nonmagnetic HF theory. That is because, as long as we are in the low-temperature and the low-frequency region, we can start with a simple approximation of taking the term in the self-energy which is linear in ω and the constant term of the vertex part near the Fermi surface. Then, after a frequency renormalization in the Green's function, $G_d = (i\bar{\chi}_{\uparrow\uparrow}\omega + i\Gamma \text{sgn}\omega)^{-1}$, most of the calculation can be carried out parallel to the HF theory. Therefore, just the following replacement of the parameters in the HF results leads us to valid expressions for physical quantities in high- T_K alloys: (a) The repulsive interaction:

$$U_{\text{eff}} \rho_d \rightarrow \bar{\chi}_{\uparrow\uparrow}; \quad (5.1)$$

(b) the effective width of the d level at the Fermi level:

$$\Gamma = (\pi\rho_d)^{-1} - \Gamma/\bar{\chi}_{\uparrow\uparrow} = \bar{\Gamma}; \quad (5.2)$$

(c) the density of states of d electrons at the Fermi level:

$$\rho_d \rightarrow \rho_d \text{ (unchanged)}. \quad (5.3)$$

It is noted here that the effective interaction (a) which was reduced in the HF theory is now, by contrast, much enhanced and $1/\rho_d$ (c) should be

distinguished from the width of the d level (b); the former remains unchanged but the latter is narrowed to cause the enhancement of the specific heat.³⁸ It is remarkable that the Kondo effect can be brought into the result of the nonmagnetic HF theory by renormalizing parameters through $\bar{\chi}_{\uparrow\uparrow}$ and $\bar{\chi}_{\uparrow\uparrow}$.

[E] The above consideration leads us to the following results for several physical quantities of high- T_K alloys. We assume in the following $\bar{\Gamma} \gg T_{c0}$.

(i) *Thermodynamic critical field* $H_c(T)$.^{39,40} At $T=0$,

$$H_c(0)/H_{c0}(0) = (1 + c\bar{\chi}_{\uparrow\uparrow}\rho_d/\rho_s)^{1/2} T_c/T_{c0}, \quad (5.4)$$

or equivalently written as

$$H_c^2(0) = 5.95 T_c C_n(T_c). \quad (5.5)$$

This is the BCS law of corresponding states observed in $AlMn$ (Ref. 41) and ThU (Ref. 42) alloys. Near the critical temperature $T \sim T_c$, we have

$$\frac{d(H_c(T)/H_c(0))}{d(T/T_c)} = 1.73. \quad (5.6)$$

The initial decrease at low concentrations is obtained as

$$h^* \equiv \left. \frac{d(H_c/H_{c0})}{d(T_c/T_{c0})} \right|_{c \rightarrow 0} = 1 - \frac{1}{2} \left[\frac{1}{\lambda'} \left(1 + \frac{1}{\lambda'} \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \right) \right]^{-1}, \quad (5.7)$$

with λ' defined in (4.21).

(ii) *The lowest excited state in the superconductor gap*.^{3,43} For a single impurity a doublet state is found at ω_0

$$\frac{\omega_0}{\Delta} = 1 - 2 \left(\frac{\Delta}{\bar{\Gamma}} \right)^2 \left(1 + \frac{1}{\lambda''} \frac{\bar{\chi}_{\uparrow\uparrow}}{\bar{\chi}_{\uparrow\uparrow}} \right)^2, \quad (5.8)$$

where

$$\lambda''^{-1} = \ln \left(\frac{2\gamma[\omega_D, \bar{\Gamma}]}{\pi\Delta} \right), \quad (5.9)$$

and we have assumed $\omega_0 \sim \Delta$, $T=0$. This result has been recently obtained by Matsuura⁴⁴ in the $s-d$ limit.

(iii) *The average order parameter at $T=0$* .^{2,3} The result is

$$\frac{\Delta(0)}{\Delta_0(0)} = \frac{1 + c\bar{\chi}_{\uparrow\uparrow}\rho_d/\rho_s}{1 - c\bar{\chi}_{\uparrow\uparrow}\rho_d/(\rho_s\lambda'')} \frac{T_c}{T_{c0}}, \quad (5.10)$$

that is, the BCS correspondence law does not hold in this case. The initial decrease at small concentrations is given by

$$\delta^* = \left. \frac{d(\Delta(0)/\Delta_0(0))}{d(T_c/T_{c0})} \right|_{c \rightarrow 0} = 1 - \lambda''. \quad (5.11)$$

Small differences between the expressions for C^* (3.21), h^* (5.7), and δ^* (5.11) are noted.

[F] In this paper we have considered a special

case (no degeneracy and the symmetric Anderson model). We can discuss the general case as well in a parallel way and confirm the usefulness of the nonmagnetic HF results. However, at present, it is not clear how to make the necessary modification of the parameters in the HF solution. To our knowledge the orbital degeneracy introduces a factor of $2l+1$ which multiplies the concentration c , while the nonsymmetry makes the density of state ρ_d at E_F smaller than in the symmetric case by an amount which depends on the position of the level E_d and on the occupation number of d electrons. Moreover, the quantities represented by $\tilde{\chi}_{\uparrow\uparrow}$ and $\tilde{\chi}_{\uparrow\downarrow}$ in our results would also be changed. In the strong limit they are still of the order of Γ/T_K . In general, we do not know how they depend on E_d , U , and other exchange interactions between degenerate d electrons, and thus we defer further discussion to future study.

[G] The magnetic impurities in the alloys in the superconducting state below the finite tail of the $T_c(c)$ curve is certainly in a singlet state, which is strongly coupled to the quasiparticle excitations of the pure superconductor by a binding energy of the order of $-T_K$ (Ref. 45). (Δ is now small.) At a fixed temperature, for smaller impurity concentrations the superconducting state is more stable and the formation of the singlet state requires more energy because of the quasiparticle excitations. If T_∞ is larger than T_K , at a fixed low temperature for lower concentrations, the impurity cannot stay in the singlet state, then, magnetic moments will appear in the superconducting state. Therefore, we can expect a discontinuous change in the ground state with changing the impurity concentrations. This phenomenon is related to the excited levels within the gap of the superconductors^{3,4,45}; as the level in the gap crosses the Fermi level, the transition takes place⁴⁶ from the singlet to a doublet state. The behavior of the magnetic moments in superconducting state⁴⁷ is one of the important problems to be further studied both theoretically and experimentally.

[H] To summarize: in this paper by using a microscopic Fermi-liquid description, we have calculated the effect of magnetic impurities on superconductors. At sufficiently low temperatures the nonmagnetic states, the intrinsic nonmagnetic state ($u < 1$) and the singlet bound state caused by the Kondo effect ($u \gg 1$), have similar effects. The repulsive interaction at the impurity site has an essential role in suppressing the superconducting state. This interaction cannot properly be treated if we simply assume a virtual level with a width T_K at the Fermi level, leaving the repulsive scattering out of explicit consideration.

Extension of the theory to higher temperatures

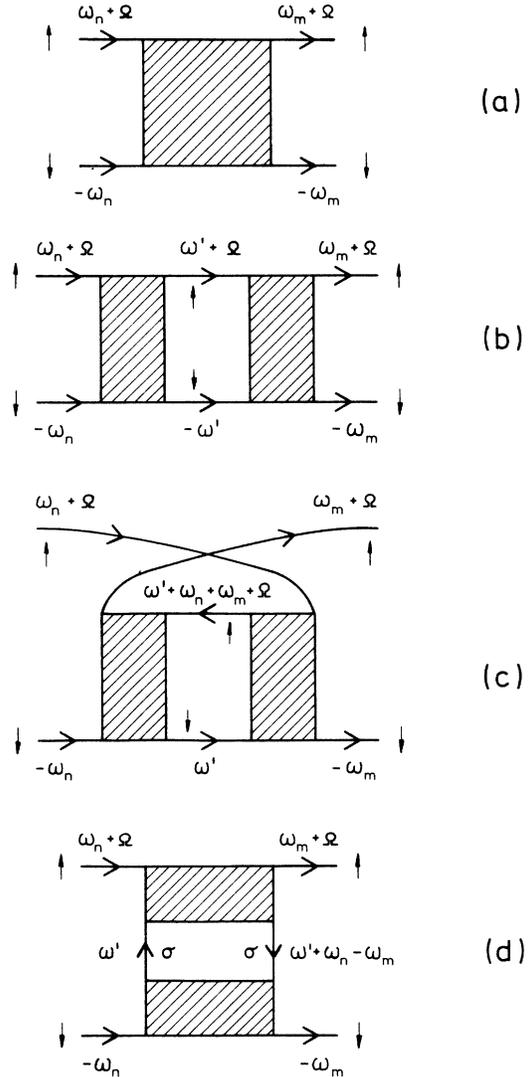


FIG. 10. Vertex part of the d electrons with opposite spins, $\Gamma_{d\uparrow\uparrow}$, (a). Linear dependence on the external frequencies comes from the scatterings with internal structures, (b), (c), (d).

seems difficult since this extension requires knowledge of the self-energy and the vertex part particularly at higher frequencies and higher temperatures, $|\omega|, T \gtrsim \bar{\Gamma}$. A possible approach is an interpolation method recently proposed for the s - d interacting system by Matsuura, Ichinose and Nagaoka.^{19,25,44} In their work they use the same method in the low-frequency region as discussed here, but employ the ω -dependent scattering matrix derived by Hamann²⁶ in the high-frequency region. They try to interpolate the two for intermediate frequencies. However, they neglect the explicit temperature dependence of the scattering amplitude, in direct contrast to the early treatment

of the MZ theory¹² which neglected the energy dependence. In light of the importance of the ω and T dependence around $T \sim T_K$ one has to consider how their results, especially in the intermediate region, might be modified by including the temperature dependence. As should be the case, for high- T_K alloys, $T_{c0} \ll T_K$, their results mainly agree with ours taken in the s - d limit $u \rightarrow \infty$. On the other hand, for $T_{c0} \gtrsim T_K$ alloys their result in the numerical calculation of $T_c(c)$ has not clearly shown the third transition.

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APPENDIX A

We consider, near the Fermi surface, a vertex part of d -electrons with opposite spins, $\Gamma_{\uparrow\downarrow}^d(i\omega_n, i\omega_m)$, corresponding to Fig. 1. To be more general we treat the case in which the total frequency of the coming electrons has a finite value Ω . Then the vertex part $\Gamma_{\uparrow\downarrow}^d(i\omega_n + i\Omega, -i\omega_n; i\omega_m + i\Omega, -i\omega_m)$ shown in Fig. 10 (a) depends on the external frequencies when internal lines of the vertex part contain them. Through perturbation consideration⁹ we can find that the linear dependence on the external frequencies comes only from the processes represented in Figs. 10(b), 10(c), and 10(d).

Now we take derivative of the internal lines with respect to the external frequencies by using a relation

$$i \frac{dG(i\omega - i\omega')}{d\omega} = G^2(i\omega - i\omega') \left(1 + i\partial \sum (i\omega - i\omega') / \partial \omega \right) + \frac{2}{\Gamma} \delta(\omega - \omega'). \quad (\text{A1})$$

After putting all the external frequencies to be zero, we obtain the derivative of the vertex part at the Fermi surface. The contributions of the three different processes are, respectively,

$$(b): \frac{\partial}{\partial \Omega} \Gamma_{\uparrow\downarrow}^d = -[\Gamma_{\uparrow\downarrow}^d(i0, i0)]^2 \frac{2}{i\Gamma} \frac{\text{sgn}\Omega}{2\pi i\Gamma} = \pi \tilde{\chi}_{\uparrow\downarrow}^2 \text{sgn}\Omega, \quad (\text{A2})$$

$$(c): \frac{\partial \Gamma_{\uparrow\downarrow}^d}{\partial (\omega_n + \omega_m + \Omega)} = [\Gamma_{\uparrow\downarrow}^d(i0, i0)]^2 \frac{2}{i\Gamma} \times \frac{\text{sgn}(\omega_n + \omega_m + \Omega)}{2\pi i\Gamma} = -\pi \tilde{\chi}_{\uparrow\downarrow}^2 \text{sgn}(\omega_n + \omega_m + \Omega), \quad (\text{A3})$$

$$(d): \frac{\partial \Gamma_{\uparrow\downarrow}^d}{\partial (\omega_n - \omega_m)} = \sum_{\sigma} \Gamma_{\uparrow\sigma}^d(i0, i0) \Gamma_{\sigma\downarrow}^d(i0, i0) \times \frac{2}{i\Gamma} \frac{\text{sgn}(\omega_n - \omega_m)}{2\pi i\Gamma} = 0, \quad (\text{A4})$$

where use has been made of the relations, $\Gamma_{\uparrow\downarrow}^d(i0, i0) = \pi \Gamma \tilde{\chi}_{\uparrow\downarrow}^d$ and $\Gamma_{\sigma\sigma}^d(i0, i0) = 0$.⁹ Therefore, the final expanded result is given by

$$\Gamma_{\uparrow\downarrow}^d(i\omega_n + i\Omega, -i\omega_n; i\omega_m + i\Omega, -i\omega_m) = \pi \Gamma \tilde{\chi}_{\uparrow\downarrow}^d \left(1 + \frac{\tilde{\chi}_{\uparrow\downarrow}^d (|\Omega| - |\omega_n + \omega_m + \Omega|)}{\Gamma} + \dots \right), \quad (\text{A5})$$

which reduces to (2.12) for $\Omega = 0$.

The same analysis on the vertex part of parallel-spin electrons leads to

$$\Gamma_{\sigma\sigma}^d(i\omega_n + i\Omega, -i\omega_n; i\omega_m + i\Omega, -i\omega_m) = -\pi \Gamma \tilde{\chi}_{\uparrow\downarrow}^2 |\omega_n - \omega_m| / \Gamma, \quad (\text{A6})$$

near the Fermi surface. The expansion (A6) is useful in examining the contribution of the terms corresponding to Fig. 4(d).

APPENDIX B

We estimate the frequency summations in (4.11). They are in general of the type

$$T \sum_n \frac{\varphi(i\omega_n)}{\omega_n} \quad (\text{B1})$$

with a function $\varphi(i\omega_n)$ smoothly decreasing from a finite value $\varphi(i0)$ at the Fermi surface to zero. The low-frequency region gives a singular $\ln T$ dependence, while the next-leading term which is temperature independent is estimated by introducing a cut-off frequency Ω , satisfying $\varphi(i\Omega) \sim \frac{1}{2} \varphi(i0)$ as

$$T \sum_{\omega_D > |\omega_n|} \frac{\varphi(i\omega_n)}{\omega_n} \approx \frac{\varphi(i0)}{\pi} \ln \frac{2\gamma[\omega_D, \Omega]}{\pi T}. \quad (\text{B2})$$

Then the respective terms in (4.11) become

$$T \sum_{\omega_D > |\omega_n|} \left(H(i\omega_n) - \frac{1}{\omega_n} \right) = -\frac{\tilde{c}}{1 + \tilde{c}} T \sum \frac{1/|\omega_n|}{1 + (1 + \tilde{c})^{-1} [(|\omega_n|/\tilde{\Gamma}) / \tilde{\alpha} (i|\omega_n|) - 1]} = -\frac{1}{\pi} \frac{\tilde{c}}{1 + \tilde{c}} \ln \frac{2\gamma[\omega_D, W(\tilde{c})]}{\pi T}, \quad (\text{B3})$$

and

$$T \sum_{\omega_D > |\omega_n|} H^{(n)}(i\omega) = \frac{1}{\pi} \frac{1}{1+\tilde{c}} \ln \frac{2\gamma[\omega_D, \tilde{\Gamma}]}{\pi T}, \quad n \geq 2 \quad (B4)$$

with $\tilde{c} = c/(\pi\rho_s\tilde{\Gamma})$ and $W(\tilde{c})$ determined by a cut-off condition,

$$(\tilde{c} + 2)^{-1} \simeq \tilde{\alpha}(i\omega)\tilde{\Gamma}/\omega \Big|_{\omega=W(\tilde{c})}. \quad (B5)$$

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