

**Theory of Kondo Effect in Superconductors. I<sup>\*)</sup>**—*Transition Temperature and Upper Critical Field*—Tamifusa MATSUURA, Shin'ichi ICHINOSE and Yosuke NAGAOKA  
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An approximation is proposed for the magnetic-impurity effect on superconductivity, where the many-body effect of impurities, i.e., the Kondo effect, is fully taken into account. It is an interpolation between the behavior in two limiting cases; i.e., the pair-breaking effect due to magnetic scattering of electrons in the high-temperature and energy region, and the effective repulsive interaction between the Cooper-pair electrons mediated by virtual polarization of impurities in the low-temperature and energy region. Using this approximation, the superconducting transition temperature and the upper critical field are calculated in the presence of impurities. Contrary to the Müller-Hartmann-Zittartz theory, a finite critical concentration is obtained for any value of the Kondo temperature, though it increases rapidly when the Kondo temperature increases.

**§ 1. Introduction**

As is well known, magnetic impurities have a number of striking effects on superconductivity. Abrikosov and Gor'kov<sup>2)</sup> (AG) investigated the problem in the Born approximation where dynamical effects of impurities are neglected. After the discovery of the Kondo effect,<sup>3)</sup> much attention was paid again to this problem, and much theoretical<sup>4)~11)</sup> as well as experimental<sup>12)~16)</sup> work was performed to clarify how the Kondo effect modifies the magnetic-impurity effect in superconductors.

Among them, Müller-Hartmann and Zittartz (MZ)<sup>5)</sup> studied the problem applying the Suhl-Nagaoka approximation to it. They found the temperature dependence of the pair-breaking parameter  $\alpha$  results in some interesting features of the transition temperature  $T_c$  as a function of the impurity concentration  $n$ . What they found is the following:

- 1) When  $T_K \ll T_{c0}$ , there appear two transition temperatures  $T_{c1}$  and  $T_{c2}$  above  $T_K$  for some region of  $n$ : i.e., with decreasing temperature a system first becomes superconducting at  $T_{c1}$  and then becomes normal again at  $T_{c2}$ .
- 2) The  $T_c$ - $n$  curve always has an infinite tail, and there appears no critical concentration  $n_c$  where  $T_c$  vanishes.

Here  $T_K$  and  $T_{c0}$  denote respectively the Kondo temperature and the transition temperature of a superconductor with no magnetic impurities. As far as correlation between impurities can be neglected, the first prediction seems unquestionable,

\*) A preliminary report of this work was given in Ref. 1).

since it is derived from the  $T$ -dependence of  $\alpha$  at  $T \gg T_K$  where the Suhl-Nagaoka approximation gives qualitatively correct results. It was also verified experimentally.<sup>12), 13)</sup> If we believe the impurity effects are completely included in the pair-breaking parameter, the second prediction also seems plausible, since impurities become nonmagnetic at  $T \rightarrow 0$  and  $\alpha$  should vanish there. However, experiments showed there always appears a finite critical concentration.<sup>11)~13)</sup> To solve this contradiction, we have to reexamine the MZ theory by the study of the impurity state at  $T \ll T_K$  in normal metals, where the Suhl-Nagaoka approximation is completely wrong.\*)

Concerning the impurity state at  $T=0$ , the Yosida theory showed it is a singlet bound state.<sup>13)</sup> The property of impurities at low but finite temperatures,  $T \ll T_K$ , was clarified by recent works of Wilson,<sup>19)</sup> Noziers,<sup>20)</sup> Yamada and Yosida<sup>21)</sup> and others.<sup>22)</sup> They found the property of impurities is normal, which is a quite different result from that of the Suhl-Nagaoka approximation. For instance, the magnetic susceptibility is finite at  $T=0$  and the specific heat is proportional to temperature at  $T \ll T_K$ .

Due to this recent development in theories, we can now clearly draw a physical picture of the so-called magnetic impurities in normal metals. What is essential in the Kondo effect is that impurities behave differently depending on temperature and energy  $\varepsilon$ : i.e.,

- 1) when  $T$  (and/or  $\varepsilon$ )  $\gg T_K$ , impurities behave as magnetic impurities with an effective interaction with conduction electrons which depends on  $T$  and  $\varepsilon$ ;
- 2) when  $T$  (and  $\varepsilon$ )  $\ll T_K$ , they behave as nonmagnetic ones with susceptibility  $\chi_{\text{imp}} \sim 1/T_K$ .

When temperature and energy change, the property of impurities gradually changes from one to the other.

When such impurities are put in superconductors, their effects on superconductivity are also expected to be different for  $T(\varepsilon) \gg T_K$  and for  $T(\varepsilon) \ll T_K$ . When  $T(\varepsilon) \gg T_K$ , they are mainly the pair-breaking effect due to magnetic scattering of electrons. In this region the Kondo effect can be taken into account as the temperature and energy dependence of the pair-breaking parameter. On the other hand, when  $T(\varepsilon) \ll T_K$ , the pair-breaking effect almost vanishes and instead there appears an effective repulsive interaction between Cooper-pair electrons with energy  $|\varepsilon| < T_K$ .\*\*\*) This repulsion results from the virtual polarization of impurities at

\*) There remain some other possibilities. Some authors<sup>8), 9)</sup> insist that, if the energy dependence of  $\alpha$  is taken into account, a finite critical concentration can be obtained. As will be discussed in §6, their argument is incorrect, since the energy dependence they assumed is inconsistent with the property of impurities in normal metals. Another possibility is the effect of correlation between impurities,<sup>17)</sup> which cannot be denied in actual experimental situations.

\*\*) The importance of this repulsive interaction in the problem of superconductivity was pointed out by Sakurai.<sup>10)</sup> It was also considered by Sólyom and Zawadowski<sup>4)</sup> and by Sundaram<sup>7)</sup> by perturbation of the  $s$ - $d$  model in the high-temperature region. Kim<sup>23)</sup> discussed a similar effect in the case of interacting impurities.

the singlet bound state, and is essentially the same effect as the paramagnon effect in transition-metal superconductors.<sup>24)</sup> If the effective exchange interaction between impurity and electrons is given by  $\tilde{J}$ , the strength of this repulsion is estimated as

$$\tilde{J}^2 \chi_{\text{imp}} \sim 1/T_K \rho^2 \quad (1.1)$$

per impurity, where we put  $\tilde{J} \sim 1/\rho$  which is the interaction strength in the unitarity limit,  $\rho$  being the density of states of conduction electrons per atom per spin.

From these considerations, we expect the following behavior of  $T_c$ :

1)  $T_{c0} \ll T_K < \omega_D$ : Due to the strong pair-breaking effect, electrons with energy  $T_K < |\varepsilon| < \omega_D$  cannot participate in superconductivity, and so the cutoff energy  $\omega_D$  is replaced by  $T_K$ . For electrons with energy  $|\varepsilon| < T_K$ , the superconducting interaction  $|g|$  is reduced by the repulsion (1.1) as

$$|g| \sim n/NT_K \rho^2, \quad (1.2)$$

where  $N$  is the number of atoms. Thus  $T_c$  is given by

$$T_c \sim T_K \exp[-1/(|g|N\rho - n/T_K \rho)] \quad (1.3)$$

and  $n_c$  by

$$n_c \sim T_K \rho \cdot |g|N\rho. \quad (1.4)$$

2)  $T_K \ll T_{c0}$ : When  $T_c > T_K$  the main effect is the pair-breaking one and the MZ theory is essentially correct, since the  $\varepsilon$ -dependence of  $\alpha$  plays only a secondary role here. Then  $T_c$  reduces to the order  $T_K$  at the concentration  $n \simeq n_c' (\geq T_{c0} \rho)$ . In this region of  $n$ , the interaction between electrons with energy  $|\varepsilon| < T_K$  is repulsive, as can be seen from Eq. (1.2). Therefore the critical concentration is essentially given by  $n_c'$ .

Such behavior is qualitatively in good agreement with experiments. If we want to carry out more quantitative calculations, we immediately encounter some difficulties. As discussed above, theories can give analytic expressions for the behavior of impurities only in limiting cases,  $T \gg T_K$  and  $T \ll T_K$ . In the intermediate region  $T \sim T_K$ , we have no such theories except numerical calculations.<sup>22)</sup> Separation of the impurity effect into the pair-breaking and the effective repulsion has no clear meaning in this region. Therefore it seems inevitable to choose some kind of interpolation. Fortunately no sharp transition between two limiting behaviors is expected in the intermediate region, and so we can believe such interpolation gives semi-quantitatively correct results. In this paper we shall carry out such calculations based on an interpolation expression which is constructed so as to coincide with theoretical expressions in limiting cases. Recently Sakurai<sup>10)</sup> performed a similar investigation in the region  $T \ll T_K$ . Some of his results are essentially the same with ours given in the previous note<sup>1)</sup> and in this paper.

So far we have discussed the impurity effect on the transition temperature.

If the effect of an applied magnetic field on impurities is neglected, a similar calculation is possible for the upper critical field  $H_{c2}$ . It is also given in this paper. As far as  $T_c$  and  $H_{c2}$  are concerned, we have only to study the self-consistent equation of the order parameter  $\Delta$  in its lowest order. To calculate the specific heat jump  $\Delta C$ , we have to study higher-order terms, which requires a much more complicated analysis. If we further want to study the properties of the superconducting states, we have to calculate Green's functions with finite  $\Delta$ . Such problems will be discussed in forthcoming papers. Throughout this paper we consider the so-called  $s$ - $d$  limit and neglect the correlation between impurities.

In §2, we give a general formulation of the self-consistent equation for  $\Delta$ , taking account of dynamical properties of impurities. In §3 we discuss the self-energy and the vertex of the effective interaction due to impurities, and construct an interpolation expressions for them. Using them, we calculate the transition temperature in §4 and the upper critical field in §5. In §6 we give summary and comments on some other works.

## § 2. Formulation

We first study the self-consistent equation for the order parameter  $\Delta$ , taking account of the dynamical behavior of impurities. Following AG, we take the average of  $\Delta$  over the random distribution of impurities, independently of the Green's functions. Then the self-consistent equation is given to the first order of  $\Delta$  by

$$\Delta = |g|Q(T)\Delta, \quad (2.1)$$

$$Q(T) = \int_0^{1/T} d\tau' \int d^3r' \langle T_\tau \{ \tilde{\psi}_\uparrow(r, \tau) \tilde{\psi}_\downarrow(r, \tau) \tilde{\psi}_\uparrow^\dagger(r', \tau') \tilde{\psi}_\downarrow^\dagger(r', \tau') \} \rangle, \quad (2.2)$$

where  $\tilde{\psi}_\sigma^\dagger$  and  $\tilde{\psi}_\sigma$  are respectively the Heisenberg representation of the creation and annihilation operators of conduction electrons with spin  $\sigma$  with respect to imaginary time  $\tau$  and  $\tau'$ , and  $\langle \dots \rangle$  denotes both the statistical average and the average over the impurity distribution.

To calculate Eq. (2.2), we take the Fourier transform of the two-particle Green's function with respect to  $r-r'$  and  $\tau-\tau'$ . Then, introducing the vertex correction  $\gamma(\omega)$  and the renormalized one-particle Green's function  $G_{\mathbf{k}}(\omega)$ , we can rewrite Eq. (2.2) as

$$Q(T) = T \sum_\omega \sum_{\mathbf{k}} \gamma(\omega) G_{\mathbf{k}}(\omega) G_{-\mathbf{k}}(-\omega), \quad (2.3)$$

$$G_{\mathbf{k}}(\omega) = \frac{1}{i\omega - \xi_{\mathbf{k}} - \Sigma(\omega)}, \quad (2.4)$$

where  $\omega = (2n+1)\pi T$ ,  $\xi_{\mathbf{k}}$  is the one-electron energy of conduction electrons and  $\Sigma(\omega)$  is the self-energy correction due to impurities. Here we took  $\gamma(\omega)$  and  $\Sigma(\omega)$  independent of  $\mathbf{k}$ , assuming the size of impurities is sufficiently small. Equation (2.3) is illustrated in Fig. 1. If we further assume the electron-hole symmetry,

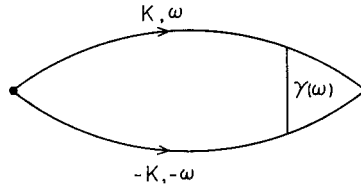


Fig. 1. Diagram corresponding to the two-particle Green's function in Eq. (2.3).

then  $\gamma(\omega)$  and  $\Sigma(\omega)$  satisfy the relations

$$\gamma(\omega) = \gamma(-\omega), \tag{2.5}$$

$$\Sigma(\omega) = -\Sigma(-\omega). \tag{2.6}$$

In this case Eq. (2.3) becomes

$$Q(T) = \pi N \rho T \sum_{\omega} \frac{\gamma(\omega)}{|\omega| + |\Sigma(\omega)|}. \tag{2.7}$$

When the impurity concentration  $n$  is sufficiently low,  $\Sigma(\omega)$  is proportional to it. Under the same condition,  $\gamma(\omega)$  is given by solving the following equation:

$$\begin{aligned} \gamma(\omega) &= 1 + T \sum_{\omega'} \sum_{\mathbf{k}} \Gamma_{\uparrow\downarrow}(\omega, \omega') G_{\mathbf{k}}(\omega') G_{-\mathbf{k}}(-\omega') \gamma(\omega') \\ &= 1 + \pi N \rho T \sum_{\omega'} \Gamma_{\uparrow\downarrow}(\omega, \omega') \frac{\gamma(\omega')}{|\omega'| + |\Sigma(\omega')|}, \end{aligned} \tag{2.8}$$

which is illustrated in Fig. 2. Here  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$ , defined graphically in Fig. 3, is the irreducible vertex of the effective interaction between electrons due to impurities. It is also proportional to  $n$ . Thus, if  $\Sigma(\omega)$  and  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  are given,  $Q(T)$  can be calculated by solving Eq. (2.8) for  $\gamma(\omega)$  and substituting it in Eq. (2.7).

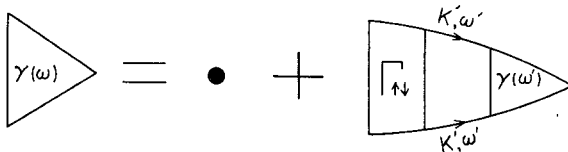


Fig. 2. Diagram corresponding to the vertex function in Eq. (2.8).

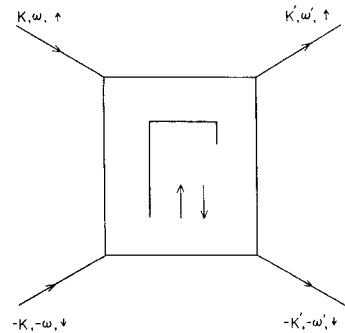


Fig. 3. Diagram of the effective interaction between electrons due to impurities.

### § 3. Approximation for selfenergy and vertex

In this section we examine the behavior of  $\Sigma(\omega)$  and  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  for various

limiting cases, and propose approximate expressions for them which are expected to be valid in the wide range of  $\omega$  and  $T$  in the  $s$ - $d$  limit of impurities.

1) *Nonmagnetic limit of the Anderson model*<sup>25)~27)</sup>

The Green's function of  $d$ -electrons is given by

$$G_d(\omega) = \frac{1}{i\omega + i\Gamma \operatorname{sgn} \omega}, \quad (3.1)$$

where

$$\Gamma = \pi V^2 N \rho \quad (3.2)$$

and  $V$  is the matrix element of the  $s$ - $d$  mixing. In Eq. (3.1) we have assumed the electron-hole symmetry as before.

Using  $G_d(\omega)$ , the selfenergy is given by

$$\Sigma(\omega) = nV^2 N G_d(\omega). \quad (3.3)$$

Taking account of the repulsive interaction  $U$  between  $d$ -electrons in the lowest approximation,  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  is given by

$$\Gamma_{\uparrow\downarrow}(\omega, \omega') = nV^4 N \left[ \frac{1}{T} |G_d(\omega)|^2 \delta_{\omega, \omega'} - |G_d(\omega)|^2 U |G_d(\omega')|^2 \right]. \quad (3.4)$$

If the ladder approximation is taken for the  $d$ -electron interaction,  $U$  in Eq. (3.4) is replaced by the effective interaction  $\tilde{U}$ . Then Eqs. (3.3) and (3.4) give the result of Ratto-Blandin<sup>26)</sup> and Kaiser.<sup>27)</sup>

2) *Born approximation in the  $s$ - $d$  limit (AG theory)*

To the second order of the  $s$ - $d$  exchange interaction  $J$ , the selfenergy is given by

$$\Sigma(\omega) = -in \frac{3\pi}{16} J^2 \rho \operatorname{sgn} \omega, \quad (3.5)$$

where the magnitude of the impurity spin is assumed 1/2. To get the expression for  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$ , it should be noticed that in our definition of  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  the spin-flip scattering is included in the exchange part  $\omega = -\omega'$ . We get

$$\Gamma_{\uparrow\downarrow}(\omega, \omega') = -\frac{3nJ^2}{16NT} \left\{ \frac{1}{3} \delta_{\omega, \omega'} + \frac{2}{3} \delta_{\omega, -\omega'} \right\}. \quad (3.6)$$

Substituting Eqs. (3.5) and (3.6) in Eqs. (2.7) and (2.8), and noting the relation (2.5), we get the result of the AG theory.

3) *Kondo effect in the most divergent approximation*

In the most divergent approximation, the Kondo effect can be taken into account by replacing  $J$  in Eqs. (3.5) and (3.6) by the spin-flip part of the  $t$ -matrix. It is given by

$$\tau(\omega) = -\frac{1}{\rho} \left( \ln \frac{|\omega|}{T_K} \right)^{-1}, \quad (3.7)$$

which is valid only when  $|\omega| \gg T_K$ . For  $T > T_K$ , Eqs. (3.5) ~ (3.7) are sufficient for calculating  $Q(T)$ , at least qualitatively. In order to study the behavior at  $T < T_K$ , they should be supplemented by the expressions of  $\Sigma(\omega)$  and  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  for  $|\omega| < T_K$ , which were given by Yamada and Yosida.<sup>21)</sup>

#### 4) Yamada-Yosida theory of the Anderson model

According to them,  $G_d(\omega)$  is given by

$$G_d(\omega) = \frac{1}{i\omega + i\Gamma \operatorname{sgn} \omega - \Sigma_d(\omega)}, \quad (3.8)$$

where  $\Sigma_d(\omega)$  is the selfenergy of  $d$ -electrons, and is given for  $|\omega| \ll T_K$  by

$$\Sigma_d(\omega) = -i\omega \left( \frac{\pi\Gamma}{4T_K} - 1 \right) - \frac{i}{2} \Gamma \left( \frac{\pi}{4T_K} \right)^2 [(i\omega)^2 + (\pi T)^2]. \quad (3.9)$$

Substitution of Eq. (3.9) in Eq. (3.8) gives

$$G_d(\omega) = -\frac{i}{\Gamma} \frac{\operatorname{sgn} \omega}{1 + (\pi|\omega|/4T_K)}, \quad (3.10)$$

where the second term of Eq. (3.9) was neglected. Using the vertex of the  $d$ -electron interaction  $\Gamma_d(\omega, \omega')$ ,  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  is expressed as

$$\Gamma_{\uparrow\downarrow}(\omega, \omega') = nV^4N \left[ \frac{1}{T} |G_d(\omega)|^2 \delta_{\omega, \omega'} - |G_d(\omega)|^2 \Gamma_d(\omega, \omega') |G_d(\omega')|^2 \right]. \quad (3.11)$$

For  $T=0$  and  $\omega=\omega'=0$ ,  $\Gamma_d$  is given by

$$\Gamma_d(0, 0) = \frac{\pi^2 \Gamma^2}{4T_K}. \quad (3.12)$$

On the basis of these expressions for limiting cases, we now propose an approximation for  $\Sigma$  and  $\Gamma_{\uparrow\downarrow}$  by interpolation. We expect  $\Gamma_d$  not to depend on  $\omega$  and  $\omega'$  so strongly, as far as  $T \ll T_K$  and  $|\omega| \sim |\omega'| \ll T_K$ . Then, if we approximate it by a constant given by Eq. (3.12), the second term of Eq. (3.11) becomes

$$-\frac{n}{N} \frac{1}{4T_K \rho^2} \left( 1 + \frac{\pi|\omega|}{4T_K} \right)^{-2} \left( 1 + \frac{\pi|\omega'|}{4T_K} \right)^{-2}. \quad (3.13)$$

At the first glance, this seems to be a reasonable approximation for the whole region of  $\omega$  and  $\omega'$ , since the factor  $|G_d(\omega)|^2$  reduces contributions from the region  $|\omega|, |\omega'| > T_K$  where the above approximation for  $\Gamma_d$  is incorrect. However, an important contribution drops off in this approximation, as will be shown shortly.

Since Eq. (3.11) is a general expression for  $\Gamma_{\uparrow\downarrow}$ , it should reduce to Eq. (3.6) for  $|\omega|, |\omega'| \gg T_K$  in the  $s$ - $d$  limit  $U \gg \Gamma$ . Comparing Eq. (3.11) with Eq. (3.6), we find  $\Gamma_d(\omega, \omega')$  should have singularities at  $|\omega \pm \omega'| \lesssim T_K$  when  $|\omega|, |\omega'| \gg T_K$ .

When Eq. (3.11) is substituted in Eq. (2.8), contributions from this singular part can readily be taken into account in the following way. When Eq. (3.11) is substituted in Eq. (2.8), we expect the factor other than  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  in the summand of the second term to be a slowly varying function of  $\omega'$  compared with  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  for  $|\omega| \gg T_K$ . Then this term can be approximated as

$$\frac{\pi N \rho \gamma(\omega)}{|\omega| + |\Sigma(\omega)|} T \sum_{|\omega - \omega'| \lesssim T_K} \Gamma_{\uparrow\downarrow}(\omega, \omega') + \frac{\pi N \rho \gamma(-\omega)}{|-\omega| + |\Sigma(-\omega)|} T \sum_{|\omega + \omega'| \lesssim T_K} \Gamma_{\uparrow\downarrow}(\omega, \omega').$$

In other words, singular contributions of  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  can be included in the elastic part by putting

$$\Gamma_{\uparrow\downarrow}(\omega, \omega') \cong \left\{ \sum_{|\omega - \omega'| \lesssim T_K} \Gamma_{\uparrow\downarrow}(\omega, \omega') \right\} \delta_{\omega, \omega'} + \left\{ \sum_{|\omega + \omega'| \lesssim T_K} \Gamma_{\uparrow\downarrow}(\omega, \omega') \right\} \delta_{\omega, -\omega'} \quad (3.14)$$

for  $|\omega| \gg T_K$  and  $|\omega'| \gg T_K$ . Thus we get an approximate expression for  $\Gamma_{\uparrow\downarrow}(\omega, \omega')$  as

$$\Gamma_{\uparrow\downarrow}(\omega, \omega') = \frac{n}{N} \left[ \frac{1}{T} \Gamma_1(\omega) \delta_{\omega, \omega'} - \frac{1}{4T_K \rho^2} f(\omega) f(\omega') \right], \quad (3.15)$$

where

$$f(\omega) = \left( 1 + \frac{\pi |\omega|}{4T_K} \right)^{-2} \quad (3.16)$$

and  $\Gamma_1(\omega)$  is given for  $|\omega| \ll T_K$  by

$$\Gamma_1(\omega) = V^4 N^2 |G_d(\omega)|^2 = \frac{1}{(\pi \rho)^2} f(\omega), \quad (3.17)$$

and for  $|\omega| \gg T_K$  by

$$\Gamma_1(\omega) \cong -\frac{3}{16} \frac{1}{\rho^2} \left( \ln \frac{|\omega|}{T_K} \right)^{-2}. \quad (3.18)$$

Here the exchange term proportional to  $\delta_{\omega, -\omega'}$  was included in the term proportional to  $\delta_{\omega, \omega'}$ , noting the relations (2.5) and (2.6), and the Kondo effect was taken into account in the most divergent approximation.

In calculating  $Q(T)$ , we need<sup>\*)</sup>

$$n\alpha(\omega) = |\Sigma(\omega)| - n\pi\rho\Gamma_1(\omega) \quad (3.19)$$

instead of  $|\Sigma(\omega)|$  and  $\Gamma_1(\omega)$  separately. It is given for  $|\omega| \ll T_K$  by

$$\begin{aligned} \alpha(\omega) &= V^2 N |G_d(\omega)| - n\pi\rho V^4 N^2 |G_d(\omega)|^2 \\ &\cong \frac{|\omega|}{4T_K \rho} \end{aligned} \quad (3.20)$$

<sup>\*)</sup>  $\alpha(\omega)$  should not be considered as a pair-breaking parameter in the usual sense, for it contains the mass renormalization effect. For  $|\omega| \ll T_K$ , the part of  $\alpha$  proportional to  $|\omega|$  gives the mass renormalization, while the lifetime effect arises from the second term of Eq. (3.9) and is proportional to  $|\omega|^2$ .



and for  $|\omega| \gg T_K$  by

$$\alpha(\omega) \cong \frac{3\pi}{8\rho} \left( \ln \frac{|\omega|}{T_K} \right)^{-2}. \tag{3.21}$$

In the intermediate region, we should take interpolation of these expressions. Though interpolation is rather arbitrary, we choose the following expression for convenience:\*)

$$\alpha(\omega) = \begin{cases} \frac{1}{\pi\rho} \left[ \left( \frac{\pi|\omega|}{4T_K} \right) - \frac{1}{2} \left( \frac{\pi|\omega|}{4T_K} \right)^2 \right], & \frac{\pi|\omega|}{4T_K} < 1, \\ \frac{1}{2\pi\rho} \frac{(3/4)\pi^2}{(\ln(\pi|\omega|/4T_K))^2 + (3/4)\pi^2}, & \frac{\pi|\omega|}{4T_K} > 1. \end{cases} \tag{3.22}$$

It is shown in Fig. 4.  $\alpha(\omega)$  takes the maximum value  $1/2\pi\rho$  at  $\pi|\omega|/4T_K=1$ . The expression for the high frequency region is similar to the expression of the pair-breaking parameter in the MZ theory. There remains some ambiguity in the form of  $\alpha$  in the region  $\omega \sim T_K$ . However, the refinement of Eq. (3.22) in this region seems meaningless, since separation of the impurity effect into two parts, the pair-breaking and the effective repulsion, loses a clear meaning there.

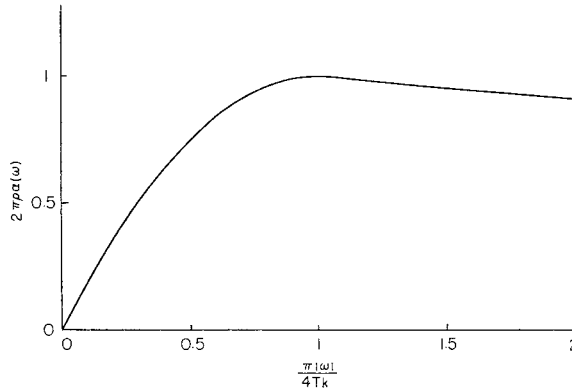


Fig. 4. The frequency dependence of the parameter  $\alpha(\omega)$  given in Eq. (3.22).

#### § 4. Transition temperature

From Eq. (2.1), the transition temperature  $T_c$  is determined by

$$|g|Q(T_c) = 1, \tag{4.1}$$

where  $Q(T)$  is calculated by using Eqs. (2.7), (2.8), (3.15) and (3.19). We obtain

\*) The coefficient of the quadratic term in  $|\omega|$  does not coincide with the exact one arising from the second term of Eq. (3.9). It was determined so that  $\alpha(\omega)$  becomes a smooth, continuous function at  $|\omega|=4T_K/\pi$ .

$$Q(T) = N\rho \left\{ \Phi_0(T, n) - \frac{n}{4T_{K\rho}} \frac{[\Phi_1(T, n)]^2}{1 + (n/4T_{K\rho})\Phi_2(T, n)} \right\}, \quad (4.2)$$

where

$$\Phi_k(T, n) = 2\pi T \sum_{\omega > 0} \frac{f(\omega)^k}{\omega + n\alpha(\omega)}. \quad (4.3)^*$$

From Eqs. (4.1) and (4.2) we get the equation to determine  $T_c$  as

$$\Phi_0(T_c, n) - \frac{n}{4T_{K\rho}} \frac{[\Phi_1(T_c, n)]^2}{1 + (n/4T_{K\rho})\Phi_2(T_c, n)} = \frac{1}{|g|N\rho}, \quad (4.4)$$

or, using  $T_{c0}$ , as

$$\ln\left(\frac{T_{c0}}{T_c}\right) = -\Phi(T_c, n) + \frac{n}{4T_{K\rho}} \frac{[\Phi_1(T_c, n)]^2}{1 + (n/4T_{K\rho})\Phi_2(T_c, n)}, \quad (4.5)$$

where

$$\Phi(T, n) = 2\pi T \sum_{\omega > 0} \left\{ \frac{1}{\omega + n\alpha(\omega)} - \frac{1}{\omega} \right\}. \quad (4.6)$$

When  $T_c \gg T_K$ ,  $\Phi_1$  and  $\Phi_2$  can be neglected. Then Eq. (4.5) reduces to

$$\ln\left(\frac{T_{c0}}{T_c}\right) = -\Phi(T_c, n). \quad (4.7)$$

This is a well-known formula for the case where the pair-breaking parameter  $\alpha$  depends on  $\omega$ . If we replace  $\alpha(\omega)$  by  $\alpha(T_c)$ , we obtain the result of the MZ theory. On the other hand, if  $T_K \gtrsim \omega_D$ , we can put

$$\Phi_0(T_c, n) \cong \Phi_1(T_c, n) \cong \Phi_2(T_c, n). \quad (4.8)$$

In this case Eq. (4.4) reduces to

$$2\pi T_c \sum_{\omega > 0} \frac{1}{\omega} = \frac{1}{|\tilde{g}|N\rho}, \quad (4.9)$$

where

$$|\tilde{g}| = |g| - \frac{n}{4NT_{K\rho}^2}. \quad (4.10)$$

Here we have used the relation  $n \ll T_{K\rho}$ , which will be shown later. Equation (4.9) is the BCS formula to determine  $T_c$  when the effective interaction is given by  $|\tilde{g}|$ .

Differentiating both sides of Eq. (4.5) with respect to  $n$ , we get the initial decrease of  $T_c$  as<sup>\*)</sup>

\*) Strictly speaking, the  $\omega$  sum in the definition of  $\Phi_2$  is different from that of  $\Phi_0$  and  $\Phi_1$ . In the latter case it is restricted by  $|\omega| < \omega_D$  while it is not in the former. This difference, however, becomes appreciable only when  $T_K > \omega_D$ .

\*\*) Details of these calculations is given in the Appendix.

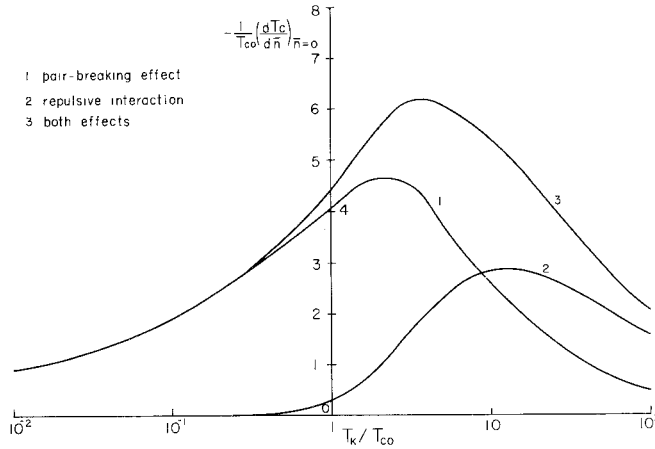


Fig. 5. The initial decrease of the transition temperature as a function of  $T_K/T_{c0}$ . Numbers attached to each curve denote 1) the contribution from the pair-breaking effect, 2) the effective repulsive interaction and 3) the total value, respectively.

$$\frac{1}{T_{c0}} \left( \frac{dT_c}{dn} \right)_{n=0} = \left[ \frac{\partial \Phi(T_{c0}, n)}{\partial n} \right]_{n=0} - \frac{1}{4T_K \rho} [\Phi_1(T_{c0}, 0)]^2. \quad (4.11)$$

Here two contributions have a clear physical meaning. The first is the pair-breaking effect due to magnetic scattering, while the second is the contribution from the effective repulsive interaction between electrons. When  $T_{c0} \gg T_K$ , we can neglect the second one and obtain the AG result

$$\frac{1}{T_{c0}} \left( \frac{dT_c}{dn} \right)_{n=0} \approx - \frac{3\pi^2 J^2 \rho}{32T_{c0}}. \quad (4.12)$$

On the other hand, when  $T_{c0} \ll T_K$ , the second contribution is larger than the first one and we obtain

$$\frac{1}{T_{c0}} \left( \frac{dT_c}{dn} \right)_{n=0} \approx - \frac{1}{4T_K \rho} \left( \ln \frac{T_K}{T_{c0}} \right)^2. \quad (4.13)$$

In order to calculate Eq. (4.11) in the whole region of  $T_K/T_{c0}$ , we should carry out numerical calculations. Results are given in Fig. 5, where the two contributions are also shown separately. As can be seen, the pair-breaking effect is important when  $T_K/T_{c0} < 1$ , while the effective repulsive interaction is important when  $T_K/T_{c0} > 1$ . The initial decrease becomes maximum where  $T_K/T_{c0} = 0(1)$ . The numerical value of  $T_K/T_{c0}$  at the maximum is not so much meaningful, since it depends on the detail of our interpolation.

We next consider the limit  $T_c \ll T_K$ . In this limit, Eq. (4.4) becomes (see the Appendix)

$$\left[ \psi_1(0, n) + \frac{4T_K \rho}{n} - \frac{1}{|g|N\rho} \right] - \frac{1}{\Phi(T_c, n)} \left[ \psi_2(0, n) - \frac{4T_K \rho}{n} \right]^2 = 0, \quad (4.14)$$

where

$$\Psi_1(T, n) = \Phi_0(T, n) - 2\Phi_1(T, n) + \Phi_2(T, n), \quad (4.15)$$

$$\Psi_2(T, n) = \Phi_1(T, n) - \Phi_2(T, n), \quad (4.16)$$

$$\phi(T, n) = \frac{\ln(\beta T_K/T)}{1 + n/4T_K\rho}, \quad (4.17)$$

$\beta$  being a constant of the order unity. It should be remarked that the  $\Phi_k$ 's diverge for  $T \rightarrow 0$ , but that the  $\Psi_k$ 's do not.

The critical concentration  $n_c$  where  $T_c$  vanishes is determined by

$$\Psi_1(0, n_c) + \frac{4T_K\rho}{n_c} - \frac{1}{|g|N\rho} = 0. \quad (4.18)$$

In the limit  $T_K \ll T_{c0}$ , Eq. (4.18) approximately reduces to

$$\int_{T_K}^{\infty} \left( \frac{1}{\omega} - \frac{1}{\omega + n_c \alpha(\omega)} \right) d\omega = \ln \left( \frac{T_{c0}}{T_K} \right). \quad (4.19)$$

This implies that  $n_c$  is nearly equal to the concentration where  $T_c$  becomes  $T_K$ , as discussed in § 1. In this case, the lower and upper bounds of  $n_c$  can be estimated by replacing  $\alpha(\omega)$  in Eq. (4.19) by its maximum and minimum values,  $\alpha(\omega \sim T_K) \sim 1/\rho$  and  $\alpha(\omega \gg T_K) \sim J^2\rho$ . Thus we obtain

$$T_{c0}\rho \lesssim n_c \lesssim T_{c0}\rho / (J\rho)^2. \quad (4.20)$$

The value of  $n_c$  given in the previous note [Eq. (5) of Ref. 1)] is this lower bound. Though the upper bound is realized in the limit  $T_K \rightarrow 0$ , numerical calculation shows that  $n_c$  is of the order  $T_{c0}\rho$  as far as  $T_K/T_{c0}$  is not so small. On the other hand, when  $\omega_D > T_K \gg T_{c0}$ , we find

$$n_c \simeq \frac{4T_K\rho}{\ln(T_K/T_{c0})}, \quad (4.21)$$

which is approximately equal to Eq. (1.4). The numerical result of the critical concentration is given in Fig. 6.

In the region  $n_c - n \ll n_c$ , Eq. (4.14) gives

$$T_c = \beta T_K \exp \left[ -\frac{pn_c}{n_c - n} \right], \quad (4.22)$$

where

$$p = \frac{(1 + (n_c/4T_K\rho)) [\Psi_2(0, n_c) - (4T_K\rho/n_c)]^2}{n_c [-(d\Psi_1(0, n_c)/dn_c) + (4T_K\rho/n_c^2)]}. \quad (4.23)$$

Equation (4.21) shows that  $(dT_c/dn)_{n=n_c} = 0$ . The existence of this finite tail was pointed out by Sakurai.<sup>10)</sup> The length of the tail depends on the value of  $T_K/T_{c0}$ . Equation (4.23) is approximately estimated as

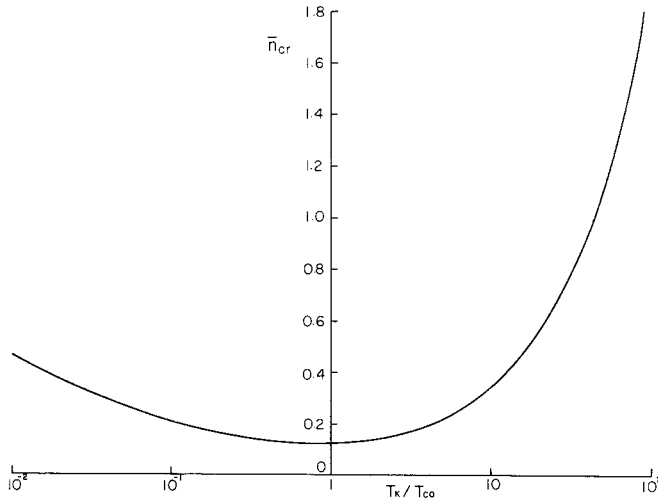


Fig. 6. The critical concentration  $\bar{n}_{cr} \equiv n_{cr}/(2\pi)^2 T_{c0} \rho$  as a function of  $T_K/T_{c0}$ .

$$p \sim \frac{T_K \rho}{n_c} \tag{4.24}$$

Then we find  $p \ll 1$  for  $T_K/T_{c0} \ll 1$  and  $p \gg 1$  for  $T_K/T_{c0} \gg 1$ . For  $T_K/T_{c0} \ll 1$ , using Eqs. (4.20), (4.22) and (4.24), we find  $T_c$  becomes of the order  $T_K$  when

$$\frac{n_c - n}{n_c} \sim \frac{T_K \rho}{n_c} \leq \frac{T_K}{T_{c0}} \ll 1. \tag{4.25}$$

This shows the tail is very short in this case. Therefore its presence does not contradict our estimate of  $n_c$  by Eq. (4.19). For  $T_K/T_{c0} \gg 1$ , Eq. (4.22) with Eqs. (4.21) and (4.24) gives

$$\frac{n_c - n}{n_c} \sim \frac{\ln(T_K/T_{c0})}{\ln(T_K/T_c)}. \tag{4.26}$$

When  $T_K/T_{c0} \sim 10^2$  and  $T_c/T_{c0} \sim 10^{-3}$ , for instance, this gives  $2/S$ ; i.e.,  $T_c$  reduces to  $10^{-3} T_{c0}$  at  $n = (3/S)n_c$ . Since the tail is very long in this case, it seems difficult to determine  $n_c$  precisely by experiments.

The behavior of  $T_c$  as a function of  $n$  is given by numerical calculation for the wide region of  $T_K/T_{c0}$ . Results are given in Fig. 7. When  $T_K \ll T_{c0}$ , the finite tail discussed above cannot be seen in this figure as it is too short.

Concluding this section, we should give a remark on the definition of the Kondo temperature. In the Yamada-Yosida theory, it is defined as the inverse of the impurity susceptibility. On the other hand, in the most divergent approximation, there is ambiguity by a factor of the order unity. Since we made an interpolation between them, our calculation also contains some ambiguity about  $T_K$ . Therefore, quantitative comparison of our results with experiments does not seem

so much meaningful.

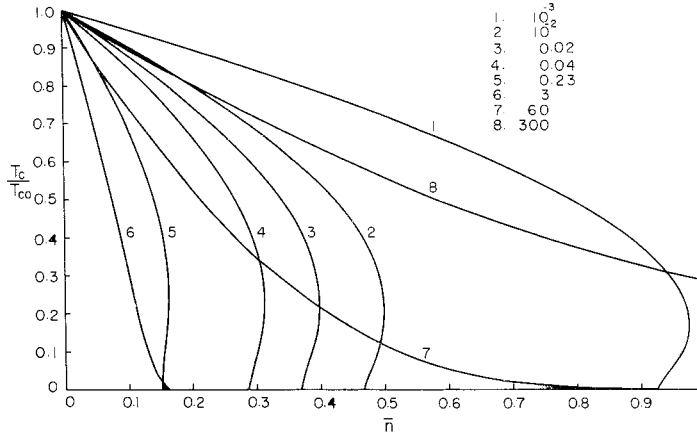


Fig. 7. The superconducting critical temperature as a function of the impurity concentration:  $\bar{n} \equiv n/(2\pi)^2 T_{c0} \rho$ . Numbers attached to each curve denote the ratio  $T_K/T_{c0}$ .

### § 5. Upper critical field

To calculate the upper critical field  $H_{c2}$ , we assume the additivity of the pair-breaking mechanisms, and consider the dirty limit. This is the same approximation as taken by Maki.<sup>9</sup> Then the equation to determine  $H_{c2}$  can be obtained by a slight modification of Eq. (4.4); i.e., it is given by

$$\tilde{\Phi}_0(T_c, H_{c2}, n) - \frac{n}{4T_K \rho} \frac{[\tilde{\Phi}_1(T_c, H_{c2}, n)]^2}{1 + (n/4T_K \rho) \tilde{\Phi}_2(T_c, H_{c2}, n)} = \frac{1}{|g|N\rho}, \quad (5.1)$$

where

$$\tilde{\Phi}_k(T, H, n) = 2\pi T \sum_{\omega > 0} \frac{f(\omega)^k}{\omega + n\alpha(\omega) + DeH} \quad (5.2)$$

and  $D$  is the diffusion constant, which we assume is determined by nonmagnetic impurities. In this approximation the effect of the magnetic field on impurities is completely neglected. If it is taken into account, both the pair-breaking parameter  $\alpha$  and the effective repulsion depend on  $H$ . Since the magnetic field characteristic of the Kondo effect is given by  $H_K = k_B T_K / \mu_B$ , this approximation is allowed only when  $H_{c2} \ll H_K$ .

In limiting cases  $T_K \ll T_{c0}$  and  $T_K \gg T_{c0}$ , Eq. (5.1) reduces to simple forms similar to Eqs. (4.7) and (4.9). In general cases it should be solved numerically. Results are given in Fig. 8. In Maki's theory,  $H_{c2}$  tends to  $H_{c2}^{(0)}$ , the value at  $T=0$  and  $n=0$ , when  $T$  tends to zero, independently of  $n$ . This behavior arises because  $\alpha$  vanishes at  $T=0$ , and corresponds to the infinite tail of the  $T_c$ - $n$  curve in the MZ theory. Our results are quite different from this;  $H_{c2}$  at  $T=0$  depends on  $n$ . The behavior characteristic of the Kondo effect is seen in curves with

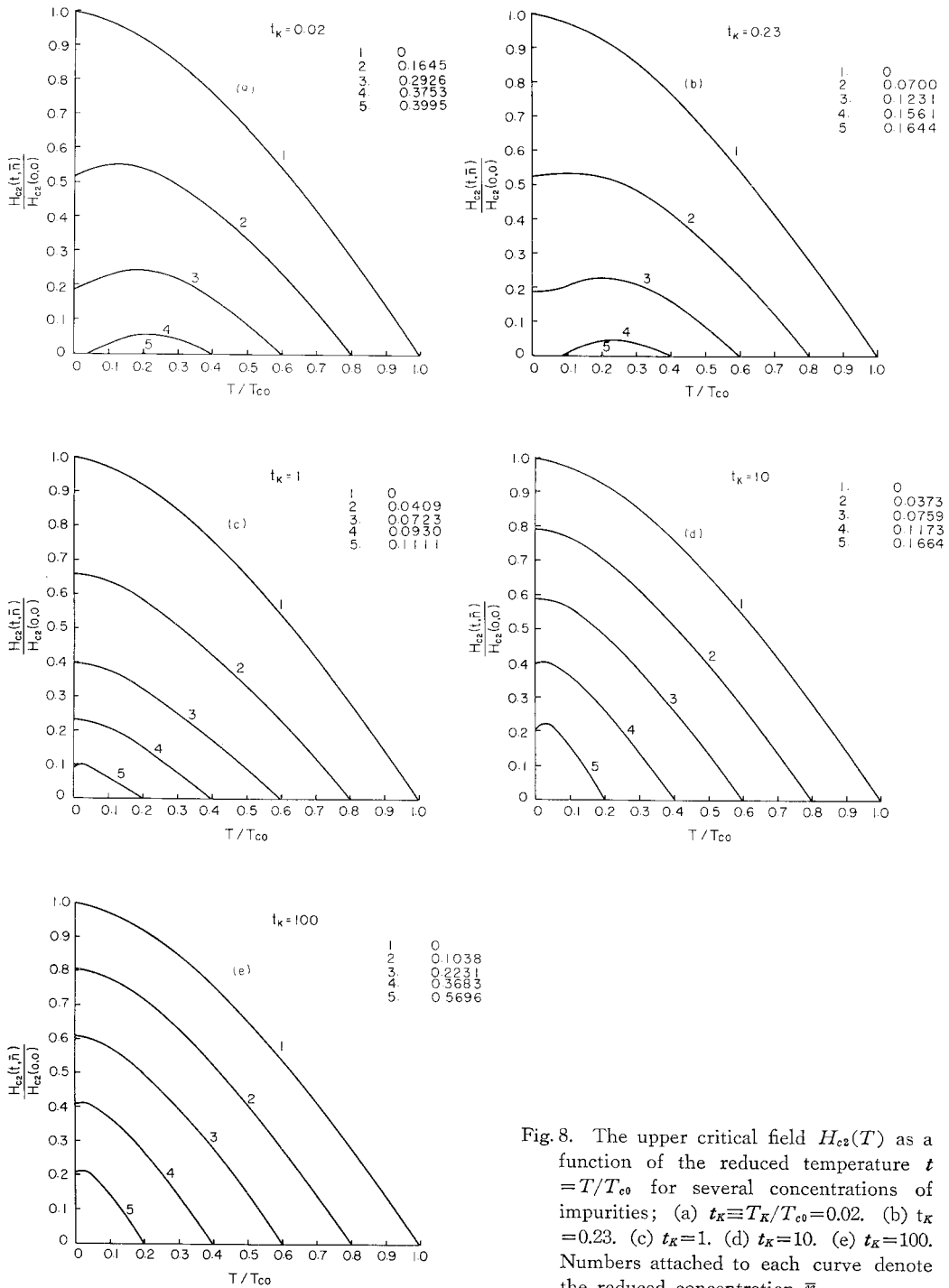


Fig. 8. The upper critical field  $H_{c2}(T)$  as a function of the reduced temperature  $t = T/T_{c0}$  for several concentrations of impurities; (a)  $t_K \equiv T_K/T_{c0} = 0.02$ . (b)  $t_K = 0.23$ . (c)  $t_K = 1$ . (d)  $t_K = 10$ . (e)  $t_K = 100$ . Numbers attached to each curve denote the reduced concentration  $\bar{n}$ .

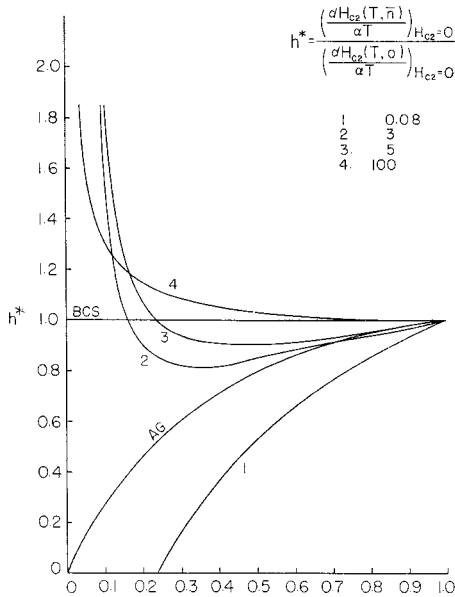


Fig. 9. The initial slope of the  $H_{c2}$ - $T$  curve,  $(dH_{c2}/dT)_{H_{c2}=0}$  as a function of reduced temperature  $t_c \equiv T_c/T_{c0}$  for several values of  $T_K$ .

showed only one curve with  $T_K/T_{c0} = 0.08$ . Since curves obtained by numerical calculations are unstable in the low-temperature region for  $T_K/T_{c0} \sim 1$ , we did not show them in the figure. This seems to be related to the behavior of  $T_c$  which has a large derivative  $(dT_c/dn)$  in the wide region of  $T_c$ . For  $T_K/T_{c0} > 1$ , the derivative  $(dH_{c2}/dT)$  increases with decreasing  $T$ , though it is finite in the limit  $T \rightarrow 0$ .

## § 6. Discussion

In this paper we proposed an interpolation approximation for the effect of magnetic impurities on superconductivity. In this approximation, the effect is separated into the pair-breaking effect and the effective repulsive interaction. Expressions for both effects are taken so that they coincide with theoretical results in limiting cases. What is essential here is that the effect depends not only on temperature  $T$  but on energy  $\epsilon$ . The first effect is important when  $T > T_K$  and/or  $\epsilon > T_K$ , while the second one is when  $T < T_K$  and  $\epsilon < T_K$ . Electrons with energy  $|\epsilon| < \omega_D$  participate in superconductivity, and the impurity effect on them appear differently depending on their energy.

In most of previous theories, only one of the two effects has been taken into account. Theories where the pair-breaking effect is considered are valid only when

small  $T_K/T_{c0}$  and relatively large  $n$ , where  $H_{c2}$  vanishes at temperatures lower than the second critical temperature.

As remarked above, these results become meaningless when  $H_{c2} \gtrsim H_K$ . The magnetic-field effect on impurities is expected to appear as  $(H/H_K)^2$  in the lowest order. Therefore, the initial slope of the  $H_{c2}$ - $T$  curve,  $(dH_{c2}/dT)_{H_{c2}=0}$ , is not affected by this effect, and our approximation of neglecting it is justified. It is plotted in Fig. 9, where the curve calculated by the AG theory is also illustrated. As is seen in Fig. 7, the temperature where  $(dT_c/dn)$  diverges does little depend on  $T_K$  as far as  $T_K \ll T_{c0}$  in our approximation. Corresponding to this behavior of  $T_c$ , curves of  $(dH_{c2}/dT)$  versus  $(T/T_c)$  are almost independent of the parameter  $T_K/T_{c0}$  if it is smaller than unity. Therefore we



$T_c > T_K$ . On the other hand, theories where only the repulsive interaction is taken into account are applicable when  $T_K > \omega_D$ , though  $G_d(\omega)$  and  $U$  in Eq. (3.4) should be replaced by Eqs. (3.10) and (3.12) if  $U \gg \Gamma$ . In general cases we showed both effects should be taken into account.

It should be emphasized here that the effect of the repulsive interaction cannot be included in the pair-breaking parameter even if its energy dependence is taken into account. In the pair-breaking theory, the transition temperature is determined by Eq. (4.7), i.e.,

$$\ln\left(\frac{T_{c0}}{T_c}\right) = -2\pi T_c \sum_{\omega>0} \left\{ \frac{1}{\omega + n\alpha(\omega)} - \frac{1}{\omega} \right\}. \quad (6.1)$$

Some authors insist that this equation gives a finite critical concentration. In order that Eq. (6.1) gives a finite  $n_c$ , the r.h.s. should diverge at  $T_c \rightarrow 0$ . This is the case if  $\omega/\alpha(\omega)$  tends to zero at  $T=0$  and  $\omega \rightarrow 0$ . The pair-breaking parameter used by Schlottmann,<sup>9)</sup> and Müller-Hartmann, Schuh and Zittartz<sup>9)</sup> satisfies this criterion. Therefore it is not surprising that they found a finite  $n_c$ . What is important here, however, is that the  $\omega$ -dependence of  $\alpha$  should be consistent with theories and experiments of the Kondo effect in normal metals. Such a singular behavior of  $\alpha$  is not consistent with them.

Here we have to mention the recent work of Sakurai,<sup>10)</sup> where he calculated the transition temperature, taking account of both effects. Starting from the Anderson model, he included the effect of magnetic scattering by introducing an effective cutoff. Some of his results are in agreement with ours. It seems to the present authors, however, that his treatment of magnetic scattering is rather phenomenological and that the physical meaning of his cutoff parameter is not so clear. We have also to give a remark about the relationship between Shiba's theory and ours. Shiba developed a Hartree-Fock theory which covers the nonmagnetic and magnetic cases of the Anderson model,<sup>20)</sup> i.e., two limiting cases  $U \ll \Gamma$  and  $U \gg \Gamma$ . We have restricted ourselves to the case  $U \gg \Gamma$ , and attempted to cover the cases  $T_K \gg T_{c0}$  and  $T_K \ll T_{c0}$ . As was shown by Yamada and Yosida,<sup>21)</sup> there is no essential difference between the case  $U \ll \Gamma$  and the case  $U \gg \Gamma$  and  $T_K \gg T$ . Therefore a similarity can be seen between Shiba's theory and ours, though the Kondo effect is not taken account of in the former.

Throughout this paper we have neglected the correlation between impurities. In actual situations this approximation is not always allowed. When  $T \gg T_K$ , a random molecular field works on each impurity which arises from the RKKY interaction between impurities. It is of the order  $nJ^2\rho/\mu_B$ . If it is higher than  $H_K$ , the correlation gives an important effect on the behavior of impurities. The criterion that it can be neglected is given by

$$n \ll \frac{T_K}{J^2\rho}. \quad (6.2)$$

When  $T_K/T_{c0}$  is sufficiently small, it is not satisfied near  $n \sim n_c$ . Therefore, to

examine experimental results in this region, we should carefully study how the correlation effect modifies the results given in this paper. In the molecular field approximation, the problem will be reduced to the study of the magnetic-field effect on impurities, which is also required in the calculation of  $H_{c2}$  when  $H_{c2} \gtrsim H_K$ .

In order to calculate the specific heat jump at  $T_c$ , we have to obtain the four-particle Green's function instead of the two-particle one of Eq. (2.2). It requires some generalization of the present calculation, which will be a straightforward but complicated task. To study the behavior of the superconducting state, such as impurity levels within the gap, the gapless behavior and so on, we have to calculate Green's functions with finite  $\mathcal{L}$ . In this case we need the knowledge of impurity properties in superconducting states. These problems will be discussed in forthcoming papers.

### Acknowledgements

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### Appendix

In this appendix we give order-of-magnitude discussion to solve Eq. (4.4) in limiting cases, and derive some equations given in § 4. To estimate  $\Phi_k$ , we use the following approximate relations: For  $|\omega| < 4T_K/\pi$ ,

$$\alpha(\omega) \simeq \frac{\omega}{4T_K\rho}, \quad 1-f(\omega) \simeq \frac{\pi|\omega|}{2T_K}; \quad (\text{A}\cdot 1)$$

for  $|\omega| > 4T_K/\pi$ ,

$$\frac{1}{2\pi\rho} > \alpha(\omega) > \frac{3\pi}{8}J^2\rho, \quad f(\omega) \simeq \left(\frac{4T_K}{\pi\omega}\right)^2; \quad (\text{A}\cdot 2)$$

for  $|\omega| \gg T_K$ , in particular, the lower bound of  $\alpha(\omega)$  is realized.

#### 1) Initial decrease

Each term of Eq. (4.11) is calculated by

$$\left(\frac{\partial\Phi}{\partial n}\right)_{n=0} = -2\pi T_{c0} \sum_{\omega>0} \frac{\alpha(\omega)}{\omega^2}, \quad (\text{A}\cdot 3)$$

$$\Phi_1 = 2\pi T_{c0} \sum_{\omega>0} \frac{f(\omega)}{\omega}. \quad (\text{A}\cdot 4)$$

When  $T_K \ll T_{c0}$ , they become

$$\left(\frac{\partial\Phi}{\partial n}\right)_{n=0} \simeq -\frac{3\pi^2 J^2 \rho}{32T_c}, \quad (\text{A}\cdot 5)$$

$$\Phi_1 \simeq \left(\frac{T_K}{T_{c0}}\right)^2. \tag{A.6}$$

Thus we find the second term of Eq. (4.11) can be neglected, and obtain Eq. (4.12). When  $T_K \gg T_{c0}$ , Eqs. (A.3) and (A.4) are estimated as

$$\frac{\partial \Phi}{\partial n} \Big|_{n=0} \simeq \frac{1}{4T_K \rho} \ln\left(\frac{T_K}{T_{c0}}\right), \tag{A.7}$$

$$\Phi_1 \simeq \ln\left(\frac{T_K}{T_{c0}}\right). \tag{A.8}$$

In this case the first term of Eq. (4.11) can be neglected, and we get Eq. (4.13).

2) Derivation of Eq. (4.14)

In the limit  $T_c \rightarrow 0$ , the singular part of  $\Phi_k (k=1, 2)$  is estimated as

$$\int_{T_c}^{T_K} \frac{d\omega}{\omega(1+n/4T'_K \rho)} = \frac{\ln(T_K/T_c)}{1+n/4T'_K \rho}. \tag{A.9}$$

Separating this singular part from  $\Phi_k$ , we put

$$\Phi_k = \phi + \Phi'_k, \tag{A.10}$$

where  $\phi$  is given by Eq. (4.17) and  $\Phi'_k$  is finite at  $T_c \rightarrow 0$ . Then, expanding the second term of Eq. (4.4) in an inverse power series of  $\phi$ , we obtain

$$\begin{aligned} \frac{n}{4T'_K \rho} \frac{\Phi_1^2}{1+(n/4T'_K \rho)\Phi_2} &= \phi + 2\Phi'_1 - \Phi'_2 - \frac{4T'_K \rho}{n} + \frac{1}{\phi} \left( \Phi'_1 - \Phi'_2 - \frac{4T'_K \rho}{n} \right)^2 \\ &+ \dots \end{aligned} \tag{A.11}$$

Substitution of Eq. (A.11) in Eq. (4.4) gives Eq. (4.14), where  $\Phi'_k$  is rewritten again by using  $\Phi_k$ .

3) Critical concentration

In the limit  $T_c \rightarrow 0$ , we get

$$\begin{aligned} \Psi_1 &= \int_0^{\omega_D} \frac{[1-f(\omega)]^2}{\omega+n\alpha(\omega)} d\omega \\ &= \ln\left(\frac{\beta\omega_D}{T_K}\right) - n \int_0^\infty \frac{\alpha(\omega)[1-f(\omega)]^2}{\omega[\omega+n\alpha(\omega)]} d\omega, \end{aligned} \tag{A.12}$$

$\beta$  being a constant of the order unity. Then Eq. (4.18) reduces to

$$n_c \int_0^\infty \frac{\alpha(\omega)[1-f(\omega)]^2}{\omega[\omega+n_c\alpha(\omega)]} d\omega = \frac{4T_K \rho}{n_c} + \ln\left(\frac{\beta T_{c0}}{T_K}\right). \tag{A.13}$$

When  $T_K \gg T_{c0}$ ,  $n_c \ll T_K \rho$  holds good, as verified in the following. In this case the l.h.s. of Eq. (A.13) is estimated as  $n_c/T_K \rho$ , and can be neglected compared with the terms on the r.h.s. Then  $n_c$  is given by Eq. (4.21). When  $T_K \ll T_{c0}$ ,

$n_c \gg T_{K\rho}$  is obtained. The l.h.s. of Eq. (A·13) is estimated as

$$\int_{T_K}^{\infty} \left\{ \frac{1}{\omega} - \frac{1}{\omega + n_c \alpha(\omega)} \right\} d\omega. \quad (\text{A} \cdot 14)$$

Then, neglecting the first term of the r.h.s. of Eq. (A·13), we obtain Eq. (4·19).

#### 4) Behavior near $n \simeq n_c$

We expand the first term of Eq. (4·14) with respect to  $n - n_c$ :

$$\Psi_1(0, n) + \frac{4T_{K\rho}}{n} - \frac{1}{|g|N\rho} = \left\{ \left( \frac{\partial \Psi_1}{\partial n} \right)_{n=n_c} - \frac{4T_{K\rho}}{n_c^2} \right\} (n - n_c). \quad (\text{A} \cdot 15)$$

Substitution of Eq. (A·15) in Eq. (4·14) gives Eqs. (4·22) and (4·23). The first term of Eq. (A·15) is calculated by

$$\left( \frac{\partial \Psi_1}{\partial n} \right)_{n=n_c} = - \int_0^{\infty} \frac{\alpha(\omega) [1 - f(\omega)]^2}{[\omega + n_c \alpha(\omega)]^2} d\omega \quad (\text{A} \cdot 16)$$

and the numerator of Eq. (4·23) by

$$\Psi_2(0, n_c) = \int_0^{\infty} \frac{f(\omega) [1 - f(\omega)]}{\omega + n_c \alpha(\omega)} d\omega. \quad (\text{A} \cdot 17)$$

They are estimated for  $T_K \gg T_{c0}$  as

$$\left( \frac{\partial \Psi_1}{\partial n} \right)_{n=n_c} \simeq \frac{1}{T_{K\rho}}, \quad \Psi_2(0, n_c) \simeq O(1) \quad (\text{A} \cdot 18)$$

and for  $T_K \ll T_{c0}$  as

$$\left( \frac{\partial \Psi_1}{\partial n} \right)_{n=n_c} \simeq \frac{1}{n_c}, \quad \Psi_2(0, n_c) \simeq \frac{T_{K\rho}}{n_c}. \quad (\text{A} \cdot 19)$$

Using these estimations, we obtain in both cases

$$\rho \simeq \frac{T_{K\rho}}{n_c}. \quad (\text{A} \cdot 20)$$

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