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Interaction between magnetic impurities in superconductors*

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The Ruderman-Kittel theory of the interaction of magnetic impurities with each other is combined with Shiba's theory of the interaction between magnetic impurities and conduction electrons in superconductors. The result explains the experimental observation that, for a given ratio of the transition temperature of the alloy to that of the pure host metal, the effect of the impurity-impurity interaction is typically no larger for alloys with magnetic impurity concentration $n \approx 1$ at.% than for those in which n is three orders of magnitude smaller.

I. INTRODUCTION

It is well known that magnetic-impurity atoms in a superconductor interact with the conduction electrons and tend to break Cooper pairs,[1] drastically decreasing the superconducting transition temperature T_c. For the most part, theories of this pair-breaking effect assume that the interaction between magnetic impurities are small and can be ignored. In this paper, we will derive an expression for the impurity-impurity interaction from which one can determine the importance of this interaction for various materials.

T_c is depressed from a value T_{c0} toward 0 as the ratio n of the number of impurity atoms to the total number of atoms is increased from 0 to a critical value n_{cr}. For small and moderate impurity concentrations, the data for T_c/T_{c0} vs n/n_{cr} follow the curve predicted by the theory of Shiba[2,3] (the same curve as that predicted by the earlier, classic, but less exact theory of Abrikosov and Gor'kov[4]).[5] This curve describes the relation

$$\ln \frac{T_c}{T_{c0}} = \psi(\tfrac{1}{2}) - \psi\left(\frac{1}{2} + 0.1404\,\frac{n}{n_{cr}}\,\frac{T_{c0}}{T_c}\right), \qquad (1)$$

where ψ is the digamma function.

Sometimes the T_c data which are obtained deviate gradually from the theoretical curve and show reentrant behavior, which is apparently related to the Kondo effect. This phenomenon is predicted by the theory of Müller-Hartmann and Zittartz[6] for materials in which the Kondo temperature is smaller than T_c, but is not very small compared with the lowest temperature obtained in the experiment. We are not concerned here with this gradual deviation from the theoretical curve, but rather with a deviation which frequently sets in[7-9] much more sharply as n approaches n_{cr}. Bennemann has shown theoretically that this sharp deviation can be understood as a result of an impurity-impurity interaction.[10]

Pair breaking affects not only T_c, but the various thermodynamic and transport properties as well. It is not surprising that an impurity-impurity interaction affects these properties as well as T_c. An example is furnished by the thermal conductivity of Zn-Mn alloys at $n \approx 10^{-3}$ at.%.[11-13] (The T_c data[11] for these alloys also deviate from Shiba's predictions at about the same value of n.) However, Shiba's theory successfully accounts for experimental results on T_c and on tunneling[14] (sometimes),[15] electromagnetic absorption,[12] and thermal conductivity[16,17] in samples for which $n \lesssim 1$ at.%. It is paradoxical that, for a given value of T_c/T_{c0}, the interaction between impurities seems to be no more important in materials with n on the order of 1 at.% than for those with n three orders of magnitude smaller. The result of our calculation will explain this paradox.

We will assume that the pair-breaking phenomenon can be examined in the framework of Shiba's theory, which treats the impurity spin classically. The theory of Müller-Hartmann and Zittartz treats it quantum mechanically, taking proper account of the commutation relations for the different spin components, but their theory is less rigorously self-consistent than Shiba's theory.[6,18,19]

For the electron concentrations of interest here, the influence of impurities on each other is dominated by the Ruderman-Kittel interaction.[20] As a result of the exchange interaction, an impurity atom polarizes the conduction-electron spins in its neighborhood, and they in turn interact with other impurity atoms. The same exchange interaction is responsible for the pair-breaking depression of T_c. One might wonder, then, whether the impurity-impurity interaction will become appreciable at a value of n which corresponds to the same value of T_c/T_{c0} for all materials. Our calculation shows that it does, qualitatively.

The strength of the Ruderman-Kittel interaction should decrease below T_c because of the reduction in the electron-spin polarizability introduced by Cooper pairing.[1] We will ignore that effect, so the strength of the impurity-impurity interaction which we calculate will be an upper limit.

II. CALCULATION

We are concerned with the effect of the interaction between two impurity atoms of the same kind. The Ruderman-Kittel interaction between spin i and spin j is of the form $-J_{ij}S_iS_j$. The interaction constant J_{ij} depends on the exchange constant J for the interaction between the impurity atom and each conduction electron. J_{ij} also depends on the Fermi energy E_F, the Fermi wave number k_F, the atomic volume Ω, the distance between the impurity atoms R_{ij}, and the electron mean free path λ. J_{ij} is given by[21]

$$J_{ij}=-\frac{J^2k_F^6\Omega^2}{128\pi^3E_F}f(2k_FR_{ij})\,e^{-R_{ij}/\lambda}\,, \tag{2}$$

where

$$f(x)=(\sin x-x\cos x)/x^4\,. \tag{3}$$

We will give an expression for a typical value of R_{ij} later in Eq. (9). For impurity concentrations $\lesssim 10^{-2}$, $2k_FR_{ij}$ is typically much larger than one, so we will drop the $\sin x$ term in Eq. (3).

We express J^2 in terms of Shiba's pair-breaking parameter α, which is defined by

$$\alpha=n(1-\epsilon_0^2)/2\pi N_0\,. \tag{4}$$

N_0 is the density of electron state per atom for one spin direction, and ϵ_0 is given by

$$\epsilon_0=|(1-\beta^2)/(1+\beta^2)|\,, \tag{5}$$

where $\beta=\frac{1}{2}\pi JSN_0$. Therefore

$$J^2=8\alpha/\pi nN_0S^2(1+\epsilon_0)^2\,. \tag{6}$$

Using the relations between k_F, E_F, and N_0, we find that

$$J_{ij}=A\cos(2k_FR_{ij})\,e^{-R_{ij}/\lambda}\,, \tag{7}$$

where

$$A=\Omega\alpha/32\pi^2nS^2(1+\epsilon_0)^2R_{ij}^3 \tag{8}$$

Dimensional analysis indicates that the proper value of R_{ij}^3 to use in this equation is a constant times Ω/n. The exact value of the proportionality constant is not critical here, but if we take R_{ij} to be the average distance from an impurity to its nearest impurity neighbor, then for $n\ll 1$ one can show that

$$R_{ij}^3=3[\Gamma(4/3)]^3\Omega/4\pi n\,, \tag{9}$$

where Γ is the gamma function.

The critical concentration n_{cr} can be related to T_{c0} by putting into Eq. (4) the corresponding critical value of α, which is $0.882\,k_BT_{c0}$. Combining this value of n_{cr} with Eqs. (4), (8), and (9), we find that

$$A=\frac{0.016\,43k_BT_{c0}}{S^2(1+\epsilon_0)^2}\,\frac{n}{n_{cr}}\,. \tag{10}$$

With the value of R_{ij} given by Eq. (9), the factor $e^{-R_{ij}/\lambda}$ is close to unity for all the materials under discussion. (A recent statement to the contrary is incorrect.[22]) We therefore drop this factor in Eq. (7) and find that

$$J_{ij}=A\cos(2k_FR_{ij})\,. \tag{11}$$

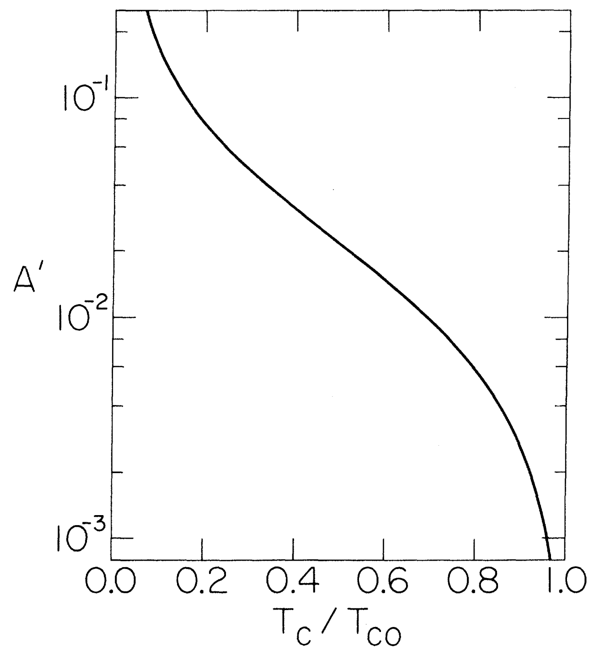

FIG. 1. A', the amplitude of the function $J_{ij}S^2/k_BT_c$ multiplied by $(1+\epsilon_0)^2$, as a function of T_c/T_{c0}. $A'/(1+\epsilon_0)^2$ determines the amount of impurity spin ordering at T_c.

III. DISCUSSION

Equations (10) and (11) are convenient expressions for the impurity-impurity interaction J_{ij} and its amplitude A.

Equation (5) shows that ϵ_0 must lie between 0 and 1. Therefore $(1+\epsilon_0)^2$ must be between 1 and 4. For most cases, it lies between 1.6 and 4. The amplitude A therefore depends only weakly on the impurity-conduction electron exchange constant J through the factor $(1+\epsilon_0)^2$. The ratio n/n_{cr} in Eq. (10) is a function of T_c/T_{c0}, according to Eq. (1). The amount of impurity-spin ordering which occurs at T_c will depend on $J_{ij}S^2/k_BT_c$; except for the factor $(1+\epsilon_0)^2$, the amplitude of the oscillating function $J_{ij} S^2/k_B T_c$ is $A' = 0.0164\,(n/n_{cr})/(T_c/T_{c0})$, which is a function only of T_c/T_{c0} (see Fig. 1). This explains why, for approximately the same value of T_c/T_{c0}, superconductors in which the concentration n of magnetic impurities is on the order of 1 at.% are typically no more susceptible to the effects of the impurity-impurity interaction than materials in which n is three orders of magnitude smaller. Figure 1 shows that A' rises rapidly as T_c/T_{c0} decreases. This rapid rise is reflected in the observed sharp onset of the effects of spin ordering as n approaches n_{cr}.

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