

## Proximity effects between superconducting and magnetic films

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The effect of magnetic impurities in the normal-metal side of a normal-metal-superconductor ( $N$ - $S$ ) "dirty" sandwich on the transition temperature of the sandwich is considered. We derive a sum rule for the kernel of the gap equation, which enables us to obtain the transition temperature in the Cooper limit. The effect of a magnetic order in the normal-metal side at temperatures close to the Curie temperature is discussed, both in the Cooper limit and in thicker films, using in the latter case Werthamer's approximation.

### I. INTRODUCTION

It is well known<sup>1</sup> that magnetic impurities reduce the transition temperature of a bulk superconductor. In the case of a normal-metal-superconductor ( $N$ - $S$ ) sandwich, Hauser, Theuerer, and Werthamer<sup>2</sup> showed that magnetic impurities in the  $N$  side of the sandwich reduce the penetration depth of the superconducting pairs into the  $N$  side. They used Werthamer's approximation<sup>3</sup> which is suitable at long distances from the boundary compared to the coherence length. Thus Werthamer's method cannot be applied in cases where the thicknesses of the normal and the superconducting slabs are small compared to their respective coherence lengths. This limit, known as the Cooper limit, is interesting as it is amenable to a rather complete solution.

The calculation of the transition temperature of an  $N$ - $S$  sandwich (in the absence of magnetic impurities) in the Cooper limit was presented by de Gennes.<sup>4</sup> The calculation relies heavily upon a sum rule satisfied by the kernel of the linearized integral equation for the gap function near the transition temperature. This equation is<sup>1</sup>

$$\Delta(\vec{r}) = V(\vec{r}) T \sum_{\omega} \int d\vec{r}' \Delta(\vec{r}') H(\vec{r}\vec{r}', \omega), \quad (1)$$

where  $\Delta(\vec{r})$  is the gap function,  $V(\vec{r})$  is the BCS electron-electron potential,  $\omega = \pi T(2n+1)$ , and the sum runs over all integers. (We use a unit system in which  $\hbar = c = k_B = 1$ .) The sum rule satisfied by  $H(\vec{r}\vec{r}', \omega)$  is<sup>5</sup>

$$\int d\vec{r}' H(\vec{r}\vec{r}', \omega) = \frac{\pi}{|\omega|} N(\vec{r}), \quad (2)$$

where  $N(\vec{r})$  is the local density of states. Using this sum rule and the condition that  $H(\vec{r}\vec{r}', \omega)/N(\vec{r}')$  is continuous across the boundary it is almost a trivial matter to find the transition temperature of the sandwich in the Cooper limit where  $\Delta$  and  $H$  can be assumed to be constant in space.

The sum rule has the simple form (2) only when there is no symmetry breaking perturbation in either the  $S$  or the  $N$  side, which acts differently

on the two electrons of the Cooper pair. In an  $N$ - $S$  sandwich containing magnetic impurities in the  $N$  side, the  $N$  and  $S$  metals have different time reversal properties<sup>5</sup> and therefore the sum rule (2) does not hold. Thus, in order to find the transition temperature of such a sandwich in the Cooper limit, by the elegant method of de Gennes,<sup>4</sup> we first have to find a sum rule analogous to (2). We derive this sum rule in Sec. II. The result is not surprising: If both  $N$  and  $S$  would contain magnetic impurities the equation analogous to (2) is<sup>5</sup>

$$\int d\vec{r}' H(\vec{r}\vec{r}', \omega) = \frac{\pi}{|\omega| + 1/\tau_s} N(\vec{r}), \quad (3)$$

where  $1/\tau_s$  is the exchange scattering rate of the electrons from the magnetic impurities. In the case where only  $N$  contains magnetic impurities we show that:

$$\left(|\omega| + \frac{1}{\tau_s}\right) \int_{-\infty}^0 dx' H(xx', \omega) + |\omega| \int_0^{\infty} dx' H(xx', \omega) = \pi N(x). \quad (4)$$

Here we assumed that  $\Delta(\vec{r})$  depends only on one space coordinate  $x$ , the boundary between the  $S$  and the  $N$  slabs is at the plane  $x=0$ , and the  $N, S$  sides are at  $x < 0$ ,  $x > 0$ , respectively. Using (4) we find in Sec. II the transition temperature of the sandwich in the Cooper limit.

In Sec. III we discuss the effect of a ferromagnetic ordering in the  $N$  side on the transition temperature. The magnetic order of the impurities may be due to their indirect exchange interaction through the conduction electrons. (For a discussion of the dependence of the Curie temperature of such systems on the concentration of the impurities see Gorkov and Rusinov.<sup>6</sup>) We show that at temperatures close to the Curie temperature it is very easy to take into account the spontaneous magnetization in the calculation of the transition temperature in the Cooper limit. We also consider in Sec. III the case of thick films. By using Werthamer's approximation<sup>3</sup> we find the effect of the spontaneous magnetization on the penetration

depth of superconducting pairs. Section IV includes our concluding remarks and a discussion of some limitations of our results.

As previously discussed by Gorkov and Rusinov<sup>6</sup> and Werthamer,<sup>7</sup> the spin-orbit scattering plays an important role when the magnetic impurities become ordered. This comes about since an increased rate of spin-orbit scattering tends to counteract the effect of the spontaneous magnetization. For the sake of completeness, we give in the Appendix an alternative derivation of  $H(\vec{r}\vec{r}', \omega)$ , which includes both spin-orbit and exchange scatterings.

## II. SUM RULE AND THE TRANSITION TEMPERATURE IN THE COOPER LIMIT

Here we first derive the sum rule (4) for an  $N$ - $S$  sandwich with magnetic impurities in the  $N$  side, and then use it to calculate the transition temperature of the sandwich in the Cooper limit. The sum rule (4) is obtained for the case where there is no ferromagnetic ordering in the  $N$  side. Its extension to include the magnetic order (in the Born approximation) is straightforward and is discussed in Sec. III.

The derivation of the sum rule is as follows: We assume that the  $N$ ,  $S$  sides of the sandwich are semi-infinite and occupy the regions  $x < 0$ ,  $x > 0$ , respectively. We then construct a differential equation for  $H(xx', \omega)$  and solve it with the requirement that  $H(xx', \omega)/N(x)$  is continuous across the boundary. Using the solution, we prove the sum rule (4).

The first step is to find a differential equation for  $H(xx', \omega)$  in the case where the  $N$  metal occupies all the space, i. e.,  $H(xx', \omega) = H_N(xx', \omega)$ . From (A18) and (A22) of the Appendix we have that in the dirty limit (in the absence of a spontaneous magnetization)

$$H_N(xx', \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dq e^{iq(x-x')} \times 2\pi N_N \frac{1}{2|\omega| + 2/\tau_s + D_N q^2}, \quad (5)$$

where  $N_N$  is the density of states at the Fermi level of the  $N$  metal and  $D_N$  is the diffusion coefficient of the  $N$  metal:

$$D_N = \frac{1}{3} \tau_N (v_{FN})^2. \quad (6)$$

From (5) we find that

$$\left[ 2 \left( |\omega| + \frac{1}{\tau_s} \right) - D_N \frac{d^2}{dx'^2} \right] H_N(xx', \omega) = 2\pi N_N \delta(x - x'). \quad (7)$$

When all the space is occupied by the  $S$  metal (which does not contain magnetic impurities)  $H(xx', \omega) = H_S(xx', \omega)$  and thus

$$\left( 2|\omega| - D_S \frac{d^2}{dx'^2} \right) H_S(xx', \omega) = 2\pi N_S \delta(x - x'). \quad (8)$$

Therefore, in order to find  $H(xx', \omega)$  of the sandwich, we have to solve the following set of equations<sup>4</sup>:

$$\begin{aligned} \left[ 2 \left( |\omega| + \frac{1}{\tau_s} \right) - D_N \frac{d^2}{dx'^2} \right] H(xx', \omega) &= 2\pi N(x) \delta(x - x'), & x' < 0 \\ \left( 2|\omega| - D_S \frac{d^2}{dx'^2} \right) H(xx', \omega) &= 2\pi N(x) \delta(x - x'), & x' > 0 \end{aligned} \quad (9)$$

with appropriate boundary conditions at  $x = 0$ .

It should be emphasized that in writing Eqs. (9) we used the diffusion approximation developed by de Gennes<sup>4</sup> and Werthamer.<sup>3</sup> That is, we took into account only the diffusive part of the kernel  $H$  in (1). The effect of a correction term to the diffusive part of  $H$  is small for very dirty systems,<sup>8</sup> in which the coherence length  $(D/2\pi T)^{1/2}$  is much greater than the mean free path  $l$ . Thus our treatment is valid in the extreme dirty limit. In solving Eqs. (9) we will assume, like in Ref. 4, that the transmission coefficient of the barrier between the  $N$  and  $S$  slabs is much greater than the ratio of  $l$  to the coherence length.

The effect of a barrier (in the absence of magnetic impurities) was considered by McMillan.<sup>9</sup> He showed that it tightly couples the two sides only for energies up to a certain energy determined by the transmission coefficient, rather than for all energies up to the cutoff energy. We shall present the extension of McMillan's calculation to include the magnetic impurities in a future article.

The general solution of (9) which vanishes when  $x, x' \rightarrow \pm\infty$  is

$$H(xx', \omega) = \begin{cases} A e^{-|x-x'|\lambda_S} + B e^{-(x+x')\lambda_S}, & x, x' > 0 \\ C e^{-x\lambda_S + x'\lambda_N}, & x' < 0, x > 0 \\ D e^{-x'\lambda_S + x\lambda_N}, & x' > 0, x < 0 \\ E e^{-|x-x'|\lambda_N} + F e^{(x+x')\lambda_N}, & x', x < 0 \end{cases} \quad (10)$$

where the coefficients  $A, B, C, D, E, F$  depend on  $\omega$  and

$$\lambda_S^2 = 2|\omega|/D_S, \quad \lambda_N^2 = 2(|\omega| + 1/\tau_s)/D_N. \quad (11)$$

We now determine the coefficients in (10). By integrating the first equation of (9) over  $x$ , using (10), we find:

$$\begin{aligned} \frac{4E}{\lambda_N} \left( |\omega| + \frac{1}{\tau_s} \right) &= 2\pi \int_{-\infty}^{\infty} dx N(x) \delta(x - x') \\ &= 2\pi N(x') = 2\pi N_N, \end{aligned} \quad (12)$$

which gives us the value of  $E$ . Similarly, integra-

tion of the second equation of (9) over  $x$  gives us

$$A = \frac{\pi N_S \lambda_S}{2|\omega|} = \frac{\pi N_S}{D_S \lambda_S}. \quad (13)$$

Since  $H(xx', \omega)$  must be symmetric when we change  $x \rightleftharpoons x'$ ,  $C = D$ . It is now very convenient to express the coefficients in terms of the boundary conditions at  $x = 0$ . We write the general boundary conditions<sup>4</sup> in terms of two constants,  $\mu$  and  $\nu$  which will be determined shortly:

$$H(x0^+, \omega) = \mu H(x0^-, \omega), \quad (14)$$

$$\left. \frac{d}{dx} H(xx', \omega) \right|_{x=0^+} = \nu \left. \frac{d}{dx} H(xx', \omega) \right|_{x=0^-}.$$

From (10) and (14) we find that

$$B = A \frac{\mu \lambda_S - \nu \lambda_N}{\mu \lambda_S + \nu \lambda_N}, \quad C = A \frac{2\lambda_S}{\mu \lambda_S + \nu \lambda_N}, \quad (15)$$

$$D = E \frac{2\mu \nu \lambda_N}{\mu \lambda_S + \nu \lambda_N}, \quad F = E \frac{\nu \lambda_N - \mu \lambda_S}{\mu \lambda_S + \nu \lambda_N},$$

and since  $C = D$  we have

$$\mu \nu = \frac{N_S}{N_N} \frac{D_N}{D_S}. \quad (16)$$

Now from the continuity of  $\Delta/NV$  across the boundary<sup>3,4</sup> we have that  $H(xx', \omega)/N(x)$  is continuous, i. e.,

$$\mu = N_S / N_N, \quad (17)$$

and therefore

$$\nu = D_N / D_S. \quad (18)$$

These boundary conditions are the same as those found by de Gennes<sup>4</sup> in a system without magnetic impurities, and used by Hauser *et al.*<sup>2</sup> to describe a sandwich containing magnetic impurities. de Gennes finds the second condition [Eq. (18), Eq. (3.16) in his article] from the sum rule (2) which does not hold in a system containing magnetic impurities. Nevertheless, de Gennes's Eq. (3.16) must hold in our sandwich as well: It is a boundary condition for  $x'$  and cannot depend on  $x$ . Once calculated for a certain  $x$ , say  $x \rightarrow \infty$ , it must hold for any other  $x$ . For example, we could apply de Gennes's Eq. (3.16) to our system for  $x \rightarrow \infty$ . In that case the  $N$  side of the sandwich has almost no influence on the  $S$  side and de Gennes's condition holds.<sup>10</sup>

Now that we have the boundary conditions for  $H(xx', \omega)$  of the sandwich, we integrate (9) over  $x'$

$$2 \left( |\omega| + \frac{1}{\tau_s} \right) \int_{-\infty}^0 dx' H(xx', \omega) + 2|\omega| \int_0^{\infty} dx' H(xx', \omega) - \left( D_N \int_{-\infty}^0 dx' \frac{d^2}{dx'^2} H(xx', \omega) + D_S \int_0^{\infty} \frac{d^2}{dx'^2} H(xx', \omega) \right) = 2\pi N(x). \quad (19)$$

From (14) and (18) we see that the last term on the left-hand side vanishes and we obtain the sum rule (4).

The Cooper limit is reached when the thicknesses  $d_N, d_S$  of the two slabs of the sandwich are much smaller than the respective coherence lengths. The reciprocal coherence lengths of the  $N$  and  $S$  metals are the smallest  $\lambda_N, \lambda_S$ , respectively [Eq. (11)]

$$\xi_N = [D_N / (2\pi T + 1/\tau_s)]^{1/2}, \quad (20)$$

$$\xi_S = (D_S / 2\pi T)^{1/2}.$$

In this limit  $H(xx', \omega)$  is almost constant when  $x$  or  $x'$  are varied in one of the slabs. Thus we have to find  $H(NN, \omega)$ ,  $H(SS, \omega)$  and  $H(NS, \omega) = H(SN, \omega)$  in order to get the gap function of the sandwich. The equations for these quantities are derived from (4), (14), and (17):

$$(|\omega| + 1/\tau_s) d_N H(SN, \omega) + |\omega| d_S H(SS, \omega) = \pi N_S,$$

$$(|\omega| + 1/\tau_s) d_N H(NN, \omega) + |\omega| d_S H(NS, \omega) = \pi N_N, \quad (21)$$

$$N_N H(SS, \omega) = N_S H(SN, \omega), \quad N_N H(NS, \omega) = N_S H(NN, \omega).$$

Solving these equations and inserting the results

into the gap equation (1) we find

$$\Delta_S = V_S N_S (d_N \Delta_N N_N + d_S \Delta_S N_S) \gamma, \quad (22)$$

$$\Delta_N = V_N N_N (d_N \Delta_N N_N + d_S \Delta_S N_S) \gamma,$$

where  $\gamma = \gamma(T)$  is

$$\gamma = T\pi \sum_{\omega} [N_N (|\omega| + 1/\tau_s) d_N + N_S |\omega| d_S]^{-1}$$

$$= (N_N d_N + N_S d_S)^{-1} \sum_n \left( |2n + 1| + \frac{1}{\pi T \tau_s} \frac{N_N d_N}{N_N d_N + N_S d_S} \right)^{-1}. \quad (23)$$

A nontrivial solution for  $\Delta_N, \Delta_S$  is obtained when

$$\gamma = (N_N^2 d_N V_N + N_S^2 d_S V_S)^{-1}. \quad (24)$$

Assuming that both  $N$  and  $S$  metals have the same frequency cutoff we get from (23) and (24)

$$\frac{N_N d_N + N_S d_S}{N_N^2 d_N V_N + N_S^2 d_S V_S} = \ln \frac{1.14 \Theta_D}{T} - \chi \left( \frac{1}{\pi T \tau_s} \frac{N_N d_N}{N_N d_N + N_S d_S} \right), \quad (25)$$

where  $\chi(x) = \psi(\frac{1}{2} + \frac{1}{2}x) - \psi(\frac{1}{2})$ , and  $\psi$  is the digamma function. The left-hand side of (25) is the "effective  $(NV)^{-1}$ " in the BCS formula for the transition

temperature in the absence of magnetic impurities.<sup>4</sup> Denoting this term by  $\ln(1.14\Theta_D/T_{c0})$ , where  $T_{c0}$  is the transition temperature of the sandwich in the absence of magnetic impurities, the transition temperature  $T_c$  is given by

$$\ln \frac{T_{c0}}{T_c} = \chi \left( \frac{1}{\pi T_c \tau_s} \frac{N_N d_N}{N_N d_N + N_S d_S} \right). \quad (26)$$

When the transition temperature of the  $N$  metal is very close to zero and in the absence of magnetic

impurities Eq. (25) gives

$$\ln \frac{T_{cs}}{T_{c0}} = \frac{N_N d_N}{N_S d_S} \ln \frac{1.14\Theta_D}{T_{cs}}, \quad (27)$$

where  $T_{cs}$  is the transition temperature of the bulk  $S$  metal. Thus from (26) and (27) we see that at low magnetic impurity concentrations where  $1/\pi T_c \tau_s \ll 1$ , the transition temperature decreases proportionally to the concentration:

$$\begin{aligned} T_c &= T_{c0} e^{-\chi} \approx T_{c0} \exp \left( -\frac{\pi}{4T_c \tau_s} \frac{d_N N_N}{d_N N_N + d_S N_S} \right) \approx T_{c0} \left( 1 - \frac{\pi}{4T_c \tau_s} \frac{d_N N_N}{d_N N_N + d_S N_S} \right) \\ &= T_{cs} \left( \frac{1.14\Theta_D}{T_{cs}} \right)^{-R} \left( 1 - \frac{\pi}{4T_c \tau_s} \frac{R}{R+1} \right), \quad R = \frac{d_N N_N}{d_S N_S}. \end{aligned} \quad (28)$$

It is worth noting that the spin-orbit scattering time does not appear at all in these calculations. In Sec. III we shall see that it becomes important when there is magnetic order in the  $N$  side.

### III. EFFECT OF FERROMAGNETIC ORDERING IN THE $N$ SIDE

As was pointed out by Gorkov and Rusinov,<sup>6</sup> the magnetic order in a superconducting alloy containing magnetic impurities may come about because of the indirect exchange interaction via the conduction electrons. When the magnetic impurities become ordered, the spin-orbit interaction plays an effective role since it counteracts the effect of the spontaneous magnetization.

In this section we consider the effect of the magnetic order on the transition temperature of a dirty sandwich in two cases: (i) when the thicknesses of the two slabs are small enough so that the Cooper limit is reached and (ii) when the films are thick compared to their respective coherence lengths so that Werthamer's approximation may be used. As can be expected, in the first case the spontaneous magnetization reduces further the transition temperature. In the second case we show that it decreases the depth of penetration of superconducting pairs into the normal metal.

From Eq. (A22) in the Appendix it is seen that the effect of the magnetization on  $Q(\vec{q}, \omega)$  and therefore on  $H_N(\chi\chi', \omega)$  of (5) is just to change  $1/\tau_s$  into  $1/\tau_s + h^2\tau_a$ . Here  $h$ , defined in (A5), is proportional to the spontaneous magnetization and  $1/\tau_a = 1/\tau_s^2 + \frac{2}{3}(1/\tau_{s0}) \rightarrow 1/\tau_{s0} \rightarrow 0$  [see (A8) and (A17)].

It is worth noting that this simple result holds only in the extreme dirty limit and in the first Born approximation  $h\tau_s \ll 1$ ,<sup>6</sup> at temperatures close to the Curie temperature. Outside this region, where  $h$  is larger than a certain critical value, the superconducting transition may be of the first

order.<sup>11</sup> Replacing  $1/\tau_s$  by  $1/\tau_s + h^2\tau_a$ , the transition temperature in the Cooper limit, in the presence of the spontaneous magnetization, is

$$\ln \frac{T_{c0}}{T_c} = \chi \left[ \frac{1}{\pi T_c} \left( \frac{1}{\tau_s} + h^2\tau_a \right) \frac{N_N d_N}{N_N d_N + N_S d_S} \right]. \quad (29)$$

The reason why the magnetic order has such a simple effect on  $T_c$  is as follows: As can be seen from the Appendix, we have treated the exchange scattering in the Born approximation at temperatures close to the Curie temperature.<sup>6</sup> Therefore, we just add to  $1/\tau_s$  a term which describes the scattering due to the  $\vec{q}=0$  Fourier transform of the exchange interaction. In the molecular-field approximation this term is connected with the internal magnetic field produced by the spontaneous magnetization. From (29) it is seen that an increased rate of spin-orbit scattering tends to counteract the effect of the spontaneous magnetization (by decreasing  $\tau_a$ ). Thus the spin-orbit scattering, which has no effect on  $T_c$  when the  $N$  side is in the paramagnetic phase, becomes important when the  $N$  side is ferromagnetic.

We now examine the effect of the magnetic order on the superconducting features of a thick sandwich, using Werthamer's<sup>3</sup> approximation. This approximation is valid when the thicknesses of the  $N$  and  $S$  slabs are greater than the respective coherence lengths.

Hauser *et al.*,<sup>2</sup> considered this system in the absence of a ferromagnetic order. We therefore omit the details and consider only the effect of the ferromagnetic order. As before, the effect of the spontaneous magnetization is to change  $1/\tau_s$  into  $1/\tau_s + h^2\tau_a$ . Thus the equations from which  $T_c$  of the sandwich is found are

$$\ln \frac{T_c}{T_{cs}} = \pi T_c \sum_{\omega} \left( \frac{1}{|\omega| + D_S q^2/2} - \frac{1}{|\omega|} \right), \quad (30)$$

$$\ln \frac{T_c}{T_{cN}} = \pi T_c \sum_{\omega} \left( \frac{1}{|\omega| + 1/\tau_s + h^2\tau_a - D_N q_N^2/2} - \frac{1}{|\omega| + 1/\tau_s + h^2\tau_a} \right), \quad (31)$$

$$N_N D_N q_N \tanh q_N d_N = N_S D_S q_S \tanh q_S d_S. \quad (32)$$

(For the details of the calculations see Refs. 2, 3.) Here  $T_{cN}$  is the transition temperature of the bulk  $N$  metal.

When  $T_{cN}/T_c \ll 1$  we obtain from (31) a measure of the depth of penetration,  $q_N^{-1}$ , of superconducting pairs into the normal metal:

$$q_N^2 \frac{D_N}{2\pi T_c} = 1 + \frac{1}{\pi T_c} \left( \frac{1}{\tau_s} + h^2\tau_a \right). \quad (33)$$

In the limit  $1/\pi T_c \tau_s \gg 1$  and above the Curie temperature, this result reduces to that of Hauser *et al.*<sup>2</sup>

#### IV. DISCUSSION

We have considered a sandwich of "dirty" superconducting and magnetic films in two cases: (i) in the Cooper limit where the thicknesses of the two slabs are smaller than their respective coherence lengths and (ii) in the limit where the thicknesses are greater than the coherence lengths so that Werthamer's approximation may be used. The magnetic properties of the  $N$  side of the sandwich enter into the expressions for the transition temperature and for the penetration depth (of superconducting pairs into the normal metal) through two quantities: the exchange scattering time  $\tau_s$  and the internal field  $h$  due to the spontaneous magnetization. The exchange scattering time has a peculiar behavior near the Curie temperature: its derivative according to the temperature diverges at the Curie temperature.<sup>12</sup> This feature influences both the superconducting transition temperature  $T_c$  and the penetration depth when  $T_c$  is close to the Curie temperature.

The internal field  $h$  of the spontaneous magnetization comes into play together with the spin-orbit

scattering. An increased rate of spin-orbit scattering tends to lower the effect of  $h$ . However, we have treated the spontaneous magnetization only in the Born approximation valid close to the Curie temperature, where the magnetization is small. Outside this region where  $h$  is not small a more careful analysis is needed. Probably the spontaneous magnetization will have a more complicated effect on the superconducting properties of the sandwich. For example, the internal field may cause the superconducting transition to become of the first order.<sup>11</sup>

The validity of our results is restricted to the extreme dirty limit, where the mean free path is much shorter than the impurity-scattering-limited coherence length  $\xi$ .<sup>8</sup> Another restriction is the assumption that the transmission coefficient of the barrier between the two slabs is much greater than  $l/\xi$ .<sup>4</sup> Using these approximations we obtained simple results which are the generalization of de Gennes<sup>4</sup> calculation for the Cooper limit. More accurate approximations may be obtained by the use of a tunneling Hamiltonian to describe the effect of the barrier.<sup>9</sup> These will be presented in a future article.

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#### APPENDIX: CALCULATION OF $H(\vec{r}\vec{r}', \omega)$ INCLUDING SPIN-ORBIT, EXCHANGE SCATTERINGS, AND THE SPONTANEOUS MAGNETIZATION

Here we calculate  $H(\vec{r}\vec{r}', \omega)$  appearing in Eq. (1). Our starting point is the expression:

$$H(\vec{r}\vec{r}', \omega) = \frac{1}{2} \text{Tr} \left[ \langle \hat{g}^{-1} \hat{G}^n(\vec{r}'\vec{r}, -\omega) \hat{g} \hat{G}^n(\vec{r}\vec{r}', \omega) \rangle \right]. \quad (A1)$$

(For its derivation see, for example, Abrikosov and Gorkov<sup>1</sup> or Fischer.<sup>13</sup>) Here  $\hat{g} = i\sigma_y$ ,  $\sigma_y$  is the second Pauli matrix,  $\hat{G}^n$  is Green's function of the normal metal, and  $\langle \rangle$  denotes an average over the impurities positions and their spin orientations.  $\hat{G}^n(\vec{r}\vec{r}', \omega)$  satisfies

$$\left( i\omega + \frac{1}{2m} \frac{\partial^2}{\partial \vec{r}^2} + \mu \right) G_{\alpha\beta}^n(\vec{r}\vec{r}', \omega) - \int d\vec{r}_1 \sum_{\gamma} V_{\alpha\gamma}(\vec{r}\vec{r}_1) G_{\gamma\beta}^n(\vec{r}_1\vec{r}', \omega) = \delta_{\alpha\beta} \delta(\vec{r} - \vec{r}'), \quad (A2)$$

where  $\alpha, \beta$  are the spin indices and  $\hat{V}$  is the scattering potential:

$$\begin{aligned} \hat{V}(\vec{r}\vec{r}') &= \frac{1}{(2\pi)^6} \int d\vec{p} d\vec{p}' e^{i\vec{p}\cdot\vec{r} - i\vec{p}'\cdot\vec{r}'} \hat{V}(\vec{p}\vec{p}'), \\ \hat{V}(\vec{p}\vec{p}') &= \sum_i e^{i(\vec{p}' - \vec{p}) \cdot \vec{R}_i} [v_{a1}(\vec{p} - \vec{p}') + v_{a2}(\vec{p} - \vec{p}') i \vec{\sigma} \cdot \hat{p} \times \hat{p}'] \\ &\quad + \sum_j e^{i(\vec{p}' - \vec{p}) \cdot \vec{R}_j} [v_{b1}(\vec{p} - \vec{p}') + v_{b2}(\vec{p} - \vec{p}') i \vec{\sigma} \cdot \hat{p} \times \hat{p}' + u(\vec{p} - \vec{p}') \vec{\sigma} \cdot \vec{S}_j]. \end{aligned} \quad (A3)$$

$\vec{R}_i, \vec{R}_j$  are the position vectors of the nonmagnetic and the magnetic impurities, respectively;  $\vec{S}_j$  is the spin of the  $j$ th impurity ion;  $v_{a1}, v_{b1}$  are the non-exchange scattering potentials of the nonmagnetic and magnetic impurities, respectively;  $v_{a2}, v_{b2}$  are the spin-orbit scattering potentials where  $\vec{p} = \vec{p}/p_F$  ( $p_F$  is the Fermi momentum), and  $u$  is the Fourier transform of the exchange interaction between the localized magnetic impurities and the conduction electrons.

The technique for averaging over the impurities positions and their spin orientations was developed by Abrikosov and Gorkov.<sup>1</sup> Using their method it can be shown that the Green's function averaged over all impurities configurations and spin orientations is<sup>6</sup>

$$\langle \hat{G}^n(\vec{r}\vec{r}', \omega) \rangle = \frac{1}{(2\pi)^3} \int d\vec{p} e^{i\vec{p}\cdot(\vec{r}-\vec{r}')} \langle \hat{G}^n(\vec{p}, \omega) \rangle, \quad (\text{A4})$$

$$\langle \hat{G}^n(\vec{p}, \omega) \rangle = \begin{pmatrix} (i\omega\eta_+ - h - \epsilon_p)^{-1} & 0 \\ 0 & (i\omega\eta_- + h - \epsilon_p)^{-1} \end{pmatrix}.$$

Here  $\epsilon_p = (p^2 - p_F^2)/2m$ ,  $h$  is proportional to the spontaneous magnetization

$$h = n_j u(0) \langle S_j^z \rangle \quad (\text{A5})$$

(the spontaneous magnetization is in the  $z$ -direction) and  $n_j$  is the number of magnetic impurities per unit volume;

$$\eta_{\pm} = 1 + \frac{1}{2|\omega|} \left( \frac{1}{\tau_1} + \frac{1}{\tau_{so}} + \frac{1}{\tau_s} \pm \frac{1}{\tau_{12}} \right). \quad (\text{A6})$$

$\tau_1$  is the nonexchange scattering time:

$$\frac{1}{\tau_1} = \frac{m}{2\pi p_F} \int dp p [n_i |v_{a1}(p)|^2 + n_j |v_{b1}(p)|^2], \quad (\text{A7})$$

where  $n_i$  is the number of non-magnetic impurities per unit volume.  $\tau_{so}$  is the spin-orbit scattering time:

$$\begin{aligned} \frac{1}{\tau_{so}} &= \frac{m p_F}{(2\pi)^2} \int d\Omega [n_i |v_{a2}(\Omega)|^2 + n_j |v_{b2}(\Omega)|^2] \sin^2\theta \\ &\approx 2\pi N(0)^{\frac{2}{3}} (n_i |v_{a2}|^2 + n_j |v_{b2}|^2). \end{aligned} \quad (\text{A8})$$

To obtain the last equality we assumed that  $v_{a2}, v_{b2}$  are constants in Fourier space. The exchange scattering time  $\tau_s$  is given by

$$\frac{1}{\tau_s} = \frac{m}{2\pi p_F} \int dp p |u(p)|^2 n_j \Gamma(p), \quad (\text{A9})$$

$$\Gamma(p) = \sum_j e^{i\vec{p}\cdot\vec{R}_j} \langle \vec{S}_0 \cdot \vec{S}_j \rangle.$$

This scattering time includes the spin-spin correlation function  $\Gamma(p)$  which has a peculiar behavior near the Curie temperature,<sup>12</sup>

$$\frac{1}{\tau_{12}} = \frac{m}{\pi p_F} \int dp p n_j v_{b2}(p) u(p) \langle S_j^z \rangle. \quad (\text{A10})$$

This mixed term will not appear in the result for  $H(\vec{r}\vec{r}', \omega)$ . The results (A4)–(A10) were obtained in the Born approximation.<sup>1,6</sup>

The average appearing in (A1) is found in terms of  $\langle \hat{G}^n \rangle$ . Assuming that the spin-orbit scattering is comparatively infrequent,  $\tau_{so} \gg \tau_1$ , where  $1/\tau = 1/\tau_1 + 1/\tau_{so} + 1/\tau_s$ , we get

$$\langle \hat{g}^{-1} \hat{G}^n(\vec{r}\vec{r}', -\omega) \hat{g} \hat{G}^n(\vec{r}\vec{r}', \omega) \rangle \equiv \hat{Q}(\vec{r}\vec{r}', \omega) = \frac{1}{(2\pi)^6} \int d\vec{q} d\vec{q}' e^{i\vec{q}\cdot\vec{r} - i\vec{q}'\cdot\vec{r}'} \hat{Q}(\vec{q}\vec{q}', \omega), \quad (\text{A11})$$

$$\hat{Q}(\vec{q}\vec{q}', \omega) = \delta(\vec{q} - \vec{q}') \int d\vec{p} \langle \hat{g}^{-1} \hat{G}^n(-\vec{p} - \vec{q}, -\omega) \hat{g} \rangle \langle \hat{G}^n(\vec{p}, \omega) \rangle$$

$$+ \frac{1}{(2\pi)^6 d^2} \int d\vec{p} d\vec{p}_1 d\vec{p}_2 d\vec{p}_3 \langle \hat{g}^{-1} \hat{G}^n(-\vec{p} - \vec{q}, -\omega) \hat{g} \rangle \langle \hat{g}^{-1} \hat{V}^t(\vec{p}_1 \vec{p}_2) \hat{g} \hat{Q}(\vec{p}_3 \vec{q}', \omega) \hat{V}^t(-\vec{p}, -\vec{q} - \vec{p}_2 + \vec{p}_3) \rangle \langle \hat{G}^n(\vec{p}, \omega) \rangle, \quad (\text{A12})$$

where  $d = \int d\vec{p}$ . The average in the second term on right-hand side of (A12) is calculated as follows: Since the first term on the right-hand side is a combination of a unit matrix and  $\sigma_z$ , we write  $\hat{Q}$  as a similar combination:

$$\hat{Q}(\vec{q}\vec{q}', \omega) = Q_1(\vec{q}\vec{q}', \omega) + \sigma_z Q_2(\vec{q}\vec{q}', \omega). \quad (\text{A13})$$

We then calculate the average in the second term on the right-hand side of (A12) in the usual way,<sup>13</sup> and perform the integral in the first term. The result is

$$\hat{Q}(\vec{q}\vec{q}', \omega) = \hat{C}(\vec{q}, \omega) \left\{ \delta(\vec{q} - \vec{q}') + \frac{1}{(2\pi)^3 2\pi N(0)} \left[ \left( \frac{1}{\tau_1} + \frac{1}{\tau_{so}} - \frac{1}{\tau_s} \right) Q_1(\vec{q}\vec{q}', \omega) + \left( \frac{1}{\tau_1} + \frac{1}{\tau_{so}} + \frac{1}{\tau_s} \right) Q_2(\vec{q}\vec{q}', \omega) \sigma_z \right] \right\}. \quad (\text{A14})$$

Here

$$\begin{aligned} \hat{C}(\vec{q}, \omega) &= \int d\vec{p} \langle \hat{g}^{-1} \hat{G}^n(-\vec{p} - \vec{q}, -\omega) \hat{g} \rangle \langle \hat{G}^n(\vec{p}, \omega) \rangle \\ &= (2\pi)^3 2\pi N(0) \frac{1}{2v_F q} \left[ \left( \arctan \frac{\beta + v_F q}{\alpha} - \arctan \frac{\beta - v_F q}{\alpha} \right) + \frac{i}{2} \sigma_z \ln \frac{(v_F q - \beta)^2 + \alpha^2}{(v_F q + \beta)^2 + \alpha^2} \right], \end{aligned}$$

$$\beta = 2h \operatorname{sgn} \omega ,$$

$$\alpha = |\omega| (\eta_+ + \eta_-) = 2|\omega| + \frac{1}{\tau_1} + \frac{1}{\tau_{so}} + \frac{1}{\tau_s} , \quad (\text{A15})$$

and

$$\frac{1}{\tau_{so}} = \frac{m p_F}{(2\pi)^2 4\pi} \int d\Omega_p d\Omega_q [n_i |v_{a2}(\Omega_p)|^2 + n_j |v_{b2}(\Omega_p)|^2] (2|\hat{p} \times \hat{q} \cdot \hat{z}|^2 - |\hat{p} \times \hat{q}|^2) \approx -\frac{1}{3} \frac{1}{\tau_{so}} , \quad (\text{A16})$$

$$\frac{1}{\tau_s} = \frac{m}{2\pi p_F} \int dp p n_j |u(p)|^2 [\Gamma(p) - 2\Gamma^z(p)] = \frac{1}{\tau_s} - \frac{1}{\tau_s^z} , \quad (\text{A17})$$

where

$$\Gamma^z(p) = \sum_j e^{i\vec{p} \cdot \vec{R}_j} \langle S_0^z S_j^z \rangle$$

is the spin-spin correlation function of the  $z$  components.

In order to obtain (A14) we have neglected correlations between different spin components of different impurity ions, e. g.,  $\langle S_j^x S_{j_1}^y \rangle$ .

Since we are interested in the trace of  $\hat{Q}(\vec{q}, \vec{q}', \omega)$  [see (A1)], we need to know only  $Q_1(\vec{q}, \vec{q}', \omega)$  which is found from (A14). Thus from (A1), (A11), (A14), and (A15) we finally get

$$H(\vec{r}, \vec{r}', \omega) = \frac{1}{(2\pi)^3} \int d\vec{q} e^{i\vec{q} \cdot (\vec{r} - \vec{r}')} Q(\vec{q}, \omega) , \quad (\text{A18})$$

$$Q(\vec{q}, \omega) = 2\pi N(0) \left\{ \frac{a}{a^2 + b^2} - \left( \frac{1}{\tau_1} + \frac{1}{\tau_{so}} - \frac{1}{\tau_s} \right) + \frac{b^2}{(a^2 + b^2)^2} \left[ \frac{a}{a^2 + b^2} - \left( \frac{1}{\tau_1} + \frac{1}{\tau_{so}} - \frac{1}{\tau_s} \right) \right]^{-1} \right\}^{-1} , \quad (\text{A19})$$

where

$$a = \frac{1}{2v_F q} \left( \arctan \frac{\beta + v_F q}{\alpha} - \arctan \frac{\beta - v_F q}{\alpha} \right) , \quad (\text{A20})$$

$$b = \frac{1}{4v_F q} \{ \ln[(v_F q - \beta)^2 + \alpha^2] - \ln[(v_F q + \beta)^2 + \alpha^2] \} .$$

We consider only small values of  $q$  for which  $v_F q / \alpha \ll 1$ , i. e.,  $q \ll 2\pi T / v_F + 1/l$ , where  $l$  is the mean free path:

$$l = v_F \tau , \quad \frac{1}{\tau} = \frac{1}{\tau_1} + \frac{1}{\tau_{so}} + \frac{1}{\tau_s} . \quad (\text{A21})$$

Since we are dealing with temperatures close to the Curie temperature we have from (A5), (A15) that  $\beta/\alpha \ll 1$ . Thus we can expand  $a, b$  in (A19) and keep the lowest orders. Moreover, we shall assume that  $2\pi T \tau \ll 1$ , the "dirty limit." Under these conditions the expression for  $Q(\vec{q}, \omega)$  becomes much simpler:

$$Q(\vec{q}, \omega) = 2\pi N(0) \left[ 2|\omega| + \frac{2}{\tau_s} + \frac{1}{3} \tau (v_F q)^2 + 4h^2 \left( 2|\omega| + \frac{2}{\tau_a} + \frac{\tau (v_F q)^2}{3} \right)^{-1} \right]^{-1} , \quad (\text{A22})$$

where

$$\frac{1}{\tau_a} = \frac{1}{\tau_s} + \frac{2}{3} \frac{1}{\tau_{so}} . \quad (\text{A23})$$

Since  $1/\tau_s^z \approx \frac{1}{3} (1/\tau_s)$  we see from (A21), (A23) that  $\tau_a/\tau > 1$ . Thus in the dirty limit the last term in (A22) is approximately equal to  $2h^2 \tau_a$ .

<sup>1</sup>A. A. Abrikosov and L. P. Gorkov, Zh. Eksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. -JETP **12**, 1243 (1961)].

<sup>2</sup>J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev. **142**, 118 (1966).

<sup>3</sup>N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).

<sup>4</sup>P. G. de Gennes, Rev. Mod. Phys. **36**, 225 (1964).

<sup>5</sup>P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).

<sup>6</sup>L. P. Gorkov and A. I. Rusinov, Zh. Eksp. Teor. Fiz. **46**, 1363 (1964) [Sov. Phys. -JETP **19**, 922 (1964)].

<sup>7</sup>N. R. Werthamer, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969).

<sup>8</sup>W. Silvert and L. N. Cooper, Phys. Rev. **141**, 336 (1966).

<sup>9</sup>W. L. McMillan, Phys. Rev. **175**, 537 (1968).

<sup>10</sup>S. Alexander (private communication).

<sup>11</sup>G. Sarma, J. Phys. Chem. Solids **24**, 1029 (1963).

<sup>12</sup>O. Entin-Wohlman, G. Deutscher, and R. Orbach, Phys. Rev. B **11**, 219 (1975).

<sup>13</sup>O. H. Fischer, Helv. Phys. Acta **45**, 332 (1972).