Amplification of Fluctuations in Superconductors above T_c

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We calculate the contribution of fluctuating Cooper pairs to the electronic spin susceptibility of a superconductor for temperatures larger than the transition temperature T_c. If paramagnetic impurities are added to the sample, this results in fluctuations in the impurity spin susceptibility. It is shown that through this mechanism, fluctuations in the electronic spin susceptibility are amplified to such an extent that in the dirty limit they should become the dominant correction to the total susceptibility.

I. INTRODUCTION

THE subject of fluctuations in superconductors above the superconducting transition temperature T_c has been of considerable interest over the last few years. After the experimental observations of fluctuations in the electrical resistivity by Glover,[1] it was shown by Aslamazov and Larkin[2] how fluctuations can be treated by microscopic theory, at least at temperatures where they are small and their mutual interaction can be neglected. Some aspects of a microscopic theory can be described by various phenomenological approaches.[3–6] However, all those approaches have the setback that they describe the dynamical aspects of the theory incompletely.[7] After the detection of fluctuations in the resistivity, they were predicted and found in such quantities as the diamagnetic susceptibility[5,8,9] and the tunneling density of states.[10,11] All these physical quantities have the property that, in the framework of the BCS theory, they change discontinuously by going from the normal to the superconducting state. The aim of the present paper is to show that, under certain circumstances, one can amplify the superconducting fluctuations and detect them, therefore, in quantities in which they would be unobservable otherwise. The special quantity which we will deal with is the electronic spin susceptibility, which changes continuously by going through the transition point T_c but which has a discontinuity in the derivative with respect to temperature at T_c. The temperature dependence of the fluctuations above T_c is very different from the one of the examples mentioned above. Especially, the fluctuations do not diverge at T_c in the approximation mentioned above but rather behave like $(T-T_c)^{1/2}$. This can be seen from the following simple argument. Below but near T_c, the ratio of the spin susceptibility in the superconducting and normal state, respectively, is given by[12]

$$\chi_s/\chi_n = 1 - bn_s\,, \tag{1}$$

where n_s is the density of superconducting electrons and b is some constant.

As is shown in Ref. 8, the density of superconducting electrons due to fluctuations for temperatures $T > T_c$ is given by

$$n_s = (m/\pi)k_B T/\xi_{\rm GL}\,, \quad T > T_c \tag{2}$$

where $\xi_{\rm GL}$ is the temperature-dependent Ginzburg-Landau coherence length proportional to $(T-T_c)^{-1/2}$. Thus, we expect that, above T_c, fluctuations lead to a temperature-dependent contribution $\chi_p{}^f \sim (T-T_c)^{1/2}$. As discussed in this paper, one should normally not see experimentally $\chi_p{}^f$. However, one can amplify $\chi_p{}^f$ by adding magnetic impurities to the sample. Since those impurities interact via conduction electrons, the fluctuations in the electronic spin susceptibility lead to fluctuations in the impurity spin susceptibility which are large enough to be observed.

In Sec. II, we compute the quantity $\chi_p{}^f$, while, in Sec. III, we give a discussion of $\chi_p{}^f$, its amplification by magnetic impurities and possible experimental detection.

II. COMPUTATION OF $\chi_p{}^f$

In order to calculate the fluctuations in the electronic spin susceptibility above T_c, we use the connection between the susceptibility and the spin-spin correlation function (Kubo's equation). A detailed presentation of

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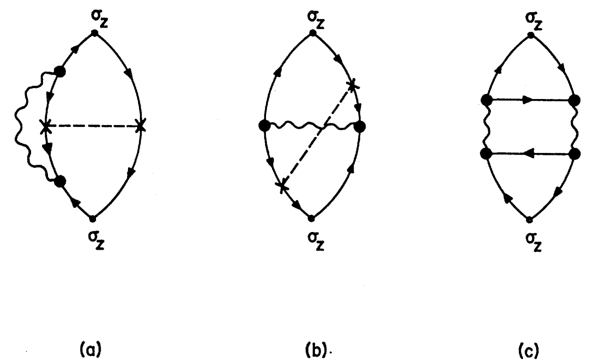

FIG. 1. Lowest-order contributions of fluctuations to the electronic spin susceptibility. Solid lines denote electron propagators, wavy lines denote the particle-particle t matrix, heavy dots denote renormalization factors due to impurity scattering, and dashed lines connect impurity scattering events (crosses) at the same impurity site.

this connection is given, for example, in Refs. 13 or 14. The lowest-order contributions of fluctuations to the spin susceptibility are given by the three types of diagrams shown in Figs. 1(a)–1(c). We proceed now in calculating these diagrams. Let us first consider diagram 1(a). The solid lines which describe electrons correspond to the propagator

$$G(\mathbf{p},\omega_n)=(i\tilde{\omega}_n-\epsilon_p)^{-1}. \tag{3}$$

Here, $\epsilon_p=v_F(p-p_F)$ and $\tilde{\omega}_n$ is the frequency, renormalized for impurity scattering. We have

$$\tilde{\omega}_n=\omega_n+(1/2\tau)n/|\omega_n|\,, \tag{4}$$

where τ is the mean free time due to impurity scattering and $\omega_n=2\pi T(n+\frac{1}{2})$.

The tilde in the above diagram represents the particle-particle t matrix in the presence of nonmagnetic and magnetic impurities. One can obtain this t matrix very easily from the linear part of the corresponding generalized Ginzburg-Landau equation (see, for example, Ref. 15). Since we will restrict our following considerations to the dirty limit in which the mean free path is much smaller than the coherence distance, we will give an expression for the t matrix in that limit only. One obtains

$$t(Q,\Omega_0)=\frac{N(0)^{-1}}{1-\rho_c\psi^{(1)}(\frac{1}{2}+\rho_c)}\times\frac{1}{\eta+(\pi/8T)C|\Omega_0|+(\pi D/8T_c)CQ^2}. \tag{5}$$

Here we have introduced the following quantities: $\eta=(T-T_c)/T_c$, $\Omega_0=2\pi Tn_0$, D is the diffusion constant given by $D=\tau v_F^2/3$, $N(0)$ is the electronic density of states for one spin, and

$$C=\frac{\psi^{(1)}(\frac{1}{2}+\rho_c)}{\psi^{(1)}\frac{1}{2}[1-\rho_c\psi^{(1)}(\frac{1}{2}+\rho_c)]}. \tag{6}$$

$\psi^{(1)}(z)$ is the trigamma function and ρ_c denotes the pair-breaking parameter due to the presence of magnetic impurities. It can be obtained by measuring the ratio T_c/T_{c0}, in which T_c and T_{c0} are the transition temperatures in the presence and absence of magnetic scattering centers, respectively. The following relation holds:

$$\ln(T_c/T_{c0})+\psi(\tfrac{1}{2}+\rho_c)-\tfrac{1}{2}\psi=0. \tag{7}$$

It should be noted that C is the same factor which enlarges the fluctuations above T_c in the resistivity due to pair breaking.[16,17] The dots in Fig. 1(a) at the end of the fluctuation line denote vertex renormalization factors due to impurity scattering. One obtains, in the standard fashion,[15]

$$\begin{aligned}\eta[Q,\tfrac{1}{2}(2\omega_n-\Omega_0)]&=\frac{1/\tau}{|2\omega_n-\Omega_0|+4\pi T\rho_c+DQ^2}, \quad \omega_n(\omega_n-\Omega_0)>0\\ &=1,\quad \omega_n(\omega_n-\Omega_0)<0.\end{aligned} \tag{8}$$

The dashed line in Fig. 1(a) represents the scattering of electrons by nonmagnetic as well as magnetic impurities. In calculating $\chi_p{}^f$, a whole ladder of impurity lines has to be summed. This leads to an additional renormalization factor. We are now in the position of writing an analytic expression I_1 for diagram 1(a). We obtain

$$\begin{aligned}I_1=4T^2\sum_{\Omega_0}\sum_{\omega_n}\int\frac{d^3Q}{(2\pi)^3}\int\frac{d^3p}{(2\pi)^3}G^3(\mathbf{p},\omega_n)\\ \times G(-\mathbf{p}+\mathbf{Q},\,-\omega_n+\Omega_0)t(Q,\Omega_0)\\ \times\{\eta[Q,\,\tfrac{1}{2}(2\omega_n-\Omega_0)]\}^3.\end{aligned} \tag{9}$$

The factor 4 results from the spin sum and from taking into account that the tilde can also appear on the right-hand side of the diagram. We perform first the d^3p integration and obtain

$$\begin{aligned}I_1=-8\pi T^2N(0)\sum_{\Omega_0}\sum_{\omega_n}\int\frac{d^3Q}{(2\pi)^3}t(Q,\Omega_0)\\ \times\{\eta[Q,\,\tfrac{1}{2}(2\omega_n-\Omega_0)]\}^3\,\mathrm{sgn}\omega_n\,\theta(\omega_n(\omega_n-\Omega_0))\\ \times\frac{\tilde{\omega}_n+\tilde{\Omega}_n}{[(Qv_F)^2+(\tilde{\omega}_n+\tilde{\Omega}_n)^2]^2},\end{aligned} \tag{10}$$

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where $\Omega_n \equiv \omega_n - \Omega_0$ and $\theta(x)$ is defined by

$$\theta(x)=1,\quad x>0$$
$$=0,\quad x<0. \tag{11}$$

Before doing the d^3Q integration, we note that in the last factor of Eq. (10) we can neglect $(Qv_F)^2$ compared with $(\tilde{\omega}_n+\tilde{\Omega}_n)^2$, since the range of integration is determined by the renormalization factor η. After performing the integration, we obtain

$$I_1=-\frac{32T_c^3}{\pi^2 CD}\frac{1}{1-\rho_c\psi^{(1)}(\frac{1}{2}+\rho_c)}\sum_{\Omega_0}\sum_{\omega_n}\theta(\omega_n(\omega_n-\Omega_0))\times\left\{\frac{3\pi}{16D^{1/2}}\frac{1}{(|2\omega_n-\Omega_0|+4\pi T\rho_c)^{5/2}}-\left(\frac{2\pi T_c}{CD}\right)^{1/2}\times\frac{1}{(|2\omega_n-\Omega_0|+4\pi T\rho_c)^3}\left(\eta+\frac{\pi}{8T}C|\Omega_0|\right)^{1/2}\right\}. \tag{12}$$

From now on, we shall be interested only in that term of the expression I_1 which is strongly temperature-dependent near T_c. It is given by

$$I_1=-\frac{1}{32\pi^3 T_c}\frac{\psi^{(2)}(\frac{1}{2}+\rho_c)}{1-\rho_c\psi^{(1)}(\frac{1}{2}+\rho_c)}\times\left(\frac{8T_c}{\pi CD}\right)^{3/2}\sqrt{\eta}+\cdots. \tag{13}$$

The other terms are constant near T_c and, thus, lead to a renormalization of the zeroth-order diagram.

We proceed now in calculating the diagram shown in Fig. 1(b). Again, we have to take a sum over the ladder of dashed impurity lines. However, this time a summation does not just lead to a renormalization factor, since there are two vertices between the ends of the impurity lines. Instead, we can write the contribution of diagram 1(b) in the form

$$I_2=-2T^2\sum_{\Omega_0}\sum_{\omega_n}\int\frac{d^3Q}{(2\pi)^3}\int\frac{d^3p}{(2\pi)^3}t(Q,\Omega_0)\times\left[\eta\left(Q,\frac{2\omega_n-\Omega_0}{2}\right)\right]^2 L(\mathbf{p},\mathbf{Q},\omega_n,\Omega_0). \tag{14}$$

The factor 2 comes from taking the spin sum. The over-all minus sign takes into account that the electronic spins at the two σ_3 vertices have opposite sign. The quantity $L(\mathbf{p},\mathbf{Q},\omega_n,\Omega_0)$ is calculated by summing the ladder of the impurity lines and is of the form

$$L=L_1+L_2$$
$$=G^2(\mathbf{p},\omega_n)G^2(-\mathbf{p}+\mathbf{Q},\ -\omega_n+\Omega_0)+\frac{1}{\pi\tau N(0)}\times G^2(\mathbf{p},\omega_n)G(-\mathbf{p}+\mathbf{Q},\ -\omega_n+\Omega_0)\int\frac{d^3p}{(2\pi)^3}G(\mathbf{p},\omega_n)\times G^2(-\mathbf{p}+\mathbf{Q},\ -\omega_n+\Omega_0)\{\eta[Q,\tfrac{1}{2}(2\omega_n-\Omega_0)]\}. \tag{15}$$

In L_2, we have taken into account that the impurity lines may run from left to right, as shown in Fig. 1(b), or from right to left. This leads to a factor of 2. The integral

$$\int\frac{d^3p}{(2\pi)^3}G(\mathbf{p},\omega_n)G^2(-\mathbf{p}+\mathbf{Q},\ -\omega_n+\Omega_0)=2\pi iN(0)\times\operatorname{sgn}\omega_n\,\theta(\omega_n(\omega_n-\Omega_0))\frac{1}{(Qv_F)^2+(\tilde{\omega}_n+\tilde{\Omega}_n)^2}. \tag{16}$$

If we perform in Eq. (14) the integration over d^3p, then we note that the function L_2 is identical with the function I_1 in Eq. (10). This connection can be established by replacing in Eq. (10) $\operatorname{sgn}\omega_n\,(\tilde{\omega}_n+\tilde{\Omega}_n)\simeq\tau^{-1}$. The contribution of L_1 to I_2 can be neglected in comparison to that of L_2 since it is smaller by a factor of τT_c than the latter. Thus, we obtain the result

$$I_2=I_1. \tag{17}$$

Finally, we have to compute the diagram shown in Fig. 1(c). However, it is easy to see that this diagram is equal to zero due to the fact that the spin sum has to be taken independently for the two triangles. Therefore, our final result is

$$\chi_p^f=-\mu_B^2\times 2I_1, \tag{18}$$

where I_1 is determined by Eq. (13). Thus, we have established that the temperature-dependent part of the contributions of fluctuations to the spin susceptibility behaves like $(T-T_c)^{1/2}$ in agreement with the simple physical argument given in the Introduction.

III. DISCUSSION

If we have a superconductor without any magnetic impurities, then the total susceptibility above the transition temperature T_c consists of four parts:

$$\chi_{\text{tot}}=\chi_d^{(0)}+\chi_p^{(0)}+\chi_d^f+\chi_p^f. \tag{19}$$

Here $\chi_d^{(0)}$, $\chi_p^{(0)}$ denote the normal-state diamagnetic and paramagnetic susceptibility.

We have

$$\chi_p^{(0)}=2\mu_B^2N(0) \tag{20}$$

and $\chi_d^{(0)}=-\frac{1}{3}\chi_p^{(0)}$ for the free-electron gas. The sum $\chi_d^{(0)}+\chi_p^{(0)}$ is different for different alloys but is approximately 10^{-6}–10^{-7}. The contribution of fluctuations to the diamagnetic susceptibility $\chi_d{}^f$ is given[8] by

$$\chi_d{}^f = -(1/6\pi)(e/\hbar c)^2 k_B T_c \xi_{\rm GL}. \tag{21}$$

The contribution of fluctuations to the paramagnetic susceptibility is given by [see Eqs. (18) and (13) with $\rho_c = 0$]

$$\xi_{\rm GL} = (\pi D/8T_c\eta')^{1/2} = \xi/\sqrt{\eta},$$

$$\chi_p{}^f = -\mu_B{}^2 \frac{1.05}{\pi^3} \frac{1}{k_B T_c \xi_{\rm GL}{}^3} \frac{1}{\eta}. \tag{22}$$

We emphasize again that Eq. (22) is valid only in the dirty limit. For example, for $T_c \simeq 5°$K and $\xi = 100$ Å (500 Å), we find $\chi_d{}^f \simeq 0.8(4)\times 10^{-8}/\sqrt{\eta}$, $\chi_p{}^f \simeq 0.5(0.004)\times 10^{-8}\times\sqrt{\eta}$.

This shows that the paramagnetic contribution of fluctuations is at least one order of magnitude smaller than the diamagnetic contribution (for $\eta \approx 0.1$).

In superconducting Knight-shift measurements, only the electronic spin susceptibility is detected. Measurements on thin tin films[18] which presumably have a very short mean free path show a characteristic dip in spin susceptibility just above T_c. It is not clear at present whether or not this dip has anything to do with the fluctuations calculated above.

As far as the relative size of the fluctuating contribution of the diamagnetic and paramagnetic susceptibility is concerned, the situation is changed drastically if the superconductor contains magnetic impurities. The diamagnetic contribution is now changed by a factor $\sqrt{C}$. Thus, we note that pair-breaking interactions do not influence the diamagnetic contribution of fluctuations very much. The change in the paramagnetic contribution can be read off immediately from Eq. (13). In addition to this, we now have to calculate the fluctuations of the impurity spin susceptibility which are caused by the changes in the electronic spin susceptibility. Here we use the Curie-Weiss law given by

$$\chi_{\rm imp} = C_0/[T - (T_K{}^{(0)} + T_K{}^{(1)})]. \tag{23}$$

Here C_0 is the Curie constant of the form

$$C_0 = (n_{\rm imp}/3k_B)(g_I \mu_B)^2 S(S+1)$$

and $T_K{}^{(0)}$ is the Curie temperature in the absence of fluctuations in the conduction-electron spin susceptibility. $n_{\rm imp}$ is the volume concentration of magnetic impurities. Let us assume that $T_K{}^{(0)} < T_c$. Near T_c, the fluctuations in the electronic spin susceptibility lead to a shift in the Curie temperature denoted by $T_K{}^{(1)}$. It is given by

$$T_K{}^{(1)}/T_K{}^{(0)} = \chi_p{}^f/\chi_p{}^{(0)}. \tag{24}$$

From Eqs. (23) and (24), it follows immediately that fluctuations in the electronic spin susceptibility lead to fluctuations in the impurity spin susceptibility of the size

$$\chi_{\rm imp}{}^f = \chi_p{}^f \frac{\chi_{\rm imp}{}^{(0)}}{\chi_p{}^{(0)}} \frac{1}{(T/T_K{}^{(0)} - 1)}, \tag{25}$$

where $\chi_{\rm imp}{}^{(0)}$ equals $\chi_{\rm imp}$ for $T_K{}^{(1)} = 0$. By using the above numbers for the susceptibility $\chi_p{}^f$, we can compute $\chi_{\rm imp}{}^f$. We assume, as an example, that the magnetic impurity concentration is 1% and that $g_I = 2$, $S = 1$, $\chi_p{}^{(0)} = 10^{-7}$, $T_{c0} = 5°$K and $T_c = 2.5°$K. With these numerical values, we find

$$\chi_{\rm imp}{}^f = 0.4\times 10^3 \frac{1}{(T/T_K{}^{(0)} - 1)^2} \chi_p{}^f. \tag{26}$$

This shows that the fluctuations in the impurity spin susceptibility are two orders of magnitude larger than the diamagnetic contribution $\chi_d{}^f$ if $\xi = 100$ Å. Equations (25) and (26) show that $\chi_{\rm imp}{}^f$ has two different temperature dependences. One results from $\chi_p{}^f$ and is given by $(T/T_c - 1)^{1/2}$ for temperatures larger but close to T_c. The other temperature dependence results from the Curie-Weiss law and is referenced to the Curie temperature which can be varied according to the impurity concentration. Thus, a measurement of the total susceptibility should show marked deviations from the Curie-Weiss law near the superconducting transition temperature T_c. From the order of magnitude of the effect, it seems possible to us to measure this predicted behavior.

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