Theory of Superconductors Containing Magnetic Impurities*

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We have studied in detail and extended the recent theory of Gor'kov and Rusinov which describes superconductors with ferromagnetically aligned impurities, under the assumption that the electron mean free path due to spin-orbit interaction is small as compared with the coherence distance. We show that all usually considered physical quantities of such a system are identical to those of a superconductor containing magnetic impurities with randomly oriented spins, and point out those vertices for which differences should arise. In addition, we investigate the magnetic properties of such systems. The upper critical field as a function of impurity concentration and temperature is calculated, and it is shown that the specific heat at the transition point in the presence of a field is a function of temperature only and does not depend on the magnetic-impurity concentration. The last results can explain the recent experimental findings by Finnemore *et al.*

I. INTRODUCTION

THE question of whether or not ferromagnetic alignment of impurities can occur in a superconductor if it occurs in the normal state of the same metal has been of considerable interest. It is well known that the momentum-dependent electron-spin susceptibility $\chi(q)$ vanishes for $q=0$ at zero temperature if the system is in the BCS-superconducting state. Since $\chi(0)$ is related to the spatial average of the electron spin density it seemed difficult to understand some experimental results,[1] which indicate that ferromagnetic alignment of the impurity spins in certain superconducting systems. It is well known, however, that the ability of magnetic impurities to flip the electron spins gives rise to a finite spin susceptibility in superconductors. Furthermore, spin-orbit interactions, which seem to be important in the materials of interest, can lead to a spin susceptibility in the superconducting state which is almost identical to that in the normal state. This was first pointed out by Ferrell and Anderson in connection with the Knight-shift problem. Under these circumstances it seems plausible that the impurity spins will line up also in the superconducting state if they do so in the normal state. Gor'kov and Rusinov,[2] in fact, showed in a detailed analysis that there will always be a region in the temperature-versus-impurity-concentration plane in which the superconducting state will have nonvanishing average electron-spin polarization. While, in the absence of spin-orbit interaction, this region is limited to low temperatures and small concentrations, the above authors obtained the result that in the case of large spin-orbit interaction (i.e., the mean free path l_{so} between spin flips is much smaller than the coherence distance ξ_0), this region can be extended considerably. In the limit $l_{so}\to 0$, the alignment of the impurities does not reflect itself in the dependence of the transition temperature on impurity concentration, which is the same as for impurities with randomly oriented spins.[3]

The aim of this communication is twofold. First, we want to extend Gor'kov and Rusinov's formalism so that the spin-orbit interaction is included explicitly in the Green's function. For short spin-orbital mean free path the calculations simplify considerably and by including terms of order l_{so}/ξ_0, but no higher, the problem of ferromagnetically aligned impurities can be reduced to that of impurities with random spin orientation for practically all physical quantities. This will be done in the next section. Second, we want to investigate the magnetic properties of superconductors with magnetic impurities and compare them with the experimental findings by Finnemore *et al.*[4] This is done in Sec. III. In Sec. IV the conclusions of this work are assembled.

II. FERROMAGNETIC SUPERCONDUCTORS WITH LARGE SPIN-ORBIT INTERACTION

In the following calculations we will assume that in the solvent material (e.g., lanthanum) the electrons have a mean free path between spin-flips due to spin-orbital interaction which is at least one order of magnitude smaller than the coherence distance. This interaction can be caused by nonmagnetic impurities, dislocations, and inhomogeneities in the sample. The mean free path l between collisions which do not involve an electron spin-flip, will be somewhat smaller than l_{so}.

We calculate now the temperature Green's function

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[1] H. Suhl, B. T. Matthias and E. Corenzwit, J. Phys. Chem. Solids **11**, 346 (1959); see also N. E. Phillips and B. T. Matthias, Phys. Rev. **121**, 105 (1961).
[2] L. P. Gor'kov and A. I. Rusinov, Zh. Eksperim. i Teor. Fiz. **46**, 1363 (1964) [English transl.: Soviet Phys.—JETP **19**, 922 (1964)].
[3] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **39**, 1781 (1960) [English transl.: Soviet Phys.—JETP **12**, 1243 (1961)].
[4] D. K. Finnemore, D. L. Johnson, J. E. Ostenson, F. H. Spedding, and B. J. Beaudry, Phys. Rev. **137**, A550 (1965).

of a superconductor with magnetic impurities using a molecular field approach. It will be advantageous to introduce a four-dimensional space in which the electron field operators are written as[5]

$$\Psi(\mathbf{x})=\begin{bmatrix}\psi_\uparrow(\mathbf{x})\\ \psi_\downarrow(\mathbf{x})\\ \psi_\uparrow^\dagger(\mathbf{x})\\ \psi_\downarrow^\dagger(\mathbf{x})\end{bmatrix}\quad \Psi^\dagger(\mathbf{x})=[\psi_\uparrow^\dagger(x),\psi_\downarrow^\dagger(\mathbf{x}),\psi_\uparrow(\mathbf{x}),\psi_\downarrow(\mathbf{x})]. \tag{1}$$

In the presence of a spin-exchange field set up by the ferromagnetically aligned impurities, the Green's function is written as

$$G_0(\mathbf{p},\omega_n)=(i\omega_n-I\rho_3\sigma_3-\xi_p\rho_3-\Delta\rho_1\sigma_2)^{-1}. \tag{2}$$

Here $\omega_n=\pi T(2n+1)$ with n being an integer. $\xi_p=p^2/2m-\mu$ where μ is the chemical potential. Furthermore I is given by $I=n_a u_2(0)\bar{S}_z$ where n_a is the concentration of the magnetic impurities and $\bar{S}_z$ is the thermal average of their spins assuming that the impurity alignment is in the z-direction. The quantity $u_2(0)$ is the spatial average of the exchange potential defined more accurately below. $\rho_1\cdots\rho_3$ and $\sigma_1\cdots\sigma_3$ denote the Pauli spin matrices acting on different spaces. The products $\rho_i\sigma_j$ are defined as, for example,[6]

$$\rho_3\sigma_j=\begin{pmatrix}\sigma_j & 0\\ 0 & -\sigma_j\end{pmatrix}. \tag{3}$$

The scattering effect of the magnetic as well as nonmagnetic impurities, the inhomogeneities, dislocations, etc., is taken into account by renormalizing the electron self-energy.[7] We write

$$G^{-1}(\mathbf{p},\omega_n)=G_0^{-1}(\mathbf{p},\omega_n)-\Sigma(\mathbf{p},\omega_n), \tag{4}$$

where the self-energy $\Sigma(\mathbf{p},\omega_n)$ is given by

$$\Sigma(\mathbf{p},\omega_n)=\sum_a\int[d^3p'/(2\pi)^3]V_a(\mathbf{p}',\mathbf{p})G(\mathbf{p}',\omega_n)V_a(\mathbf{p},\mathbf{p}')$$

$$+n_b\int[d^3p'/(2\pi)^3]V_b(\mathbf{p}',\mathbf{p})G(\mathbf{p}',\omega_n)V_b(\mathbf{p},\mathbf{p}'). \tag{5}$$

The summation index a sums over all magnetic impurities. The scattering effect of the nonmagnetic impurities, dislocations, and inhomogeneities is characterized by a scattering potential $V_b(\mathbf{p},\mathbf{p}')$ and an impurity concentration n_b. The scattering potential $V_b(\mathbf{p},\mathbf{p}')$ has a spin-flip part from the spin-orbital interaction as well as a nonspin-flip part. We write

$$V_b(\mathbf{p},\mathbf{p}')=v_1(\mathbf{p}-\mathbf{p}')\rho_3+i(v_{so}/p_F^2)[\mathbf{p}\times\mathbf{p}']\boldsymbol{\alpha}\rho_3. \tag{6}$$

The matrix vector $\boldsymbol{\alpha}$ is defined as

$$\boldsymbol{\alpha}=\tfrac{1}{2}(1+\rho_3)\boldsymbol{\sigma}+\tfrac{1}{2}(1-\rho_3)\sigma_2\boldsymbol{\sigma}\sigma_2. \tag{7}$$

[5] V. Ambegaokar and A. Griffin, Phys. Rev. **137**, A1151 (1965).
[6] K. Maki and P. Fulde, Phys. Rev. **140**, A1586 (1965).
[7] A. A. Abrikosov and L. P. Gorkov, Zh. Eksperim. i Teor. Fiz. **35**, 1558 (1958) [English transl.: Soviet Phys.—JETP 8, 1090 (1959)].

The magnetic impurities have a scattering potential

$$V_a(\mathbf{p},\mathbf{p}')=u_1(\mathbf{p}-\mathbf{p}')\rho_3+u_2(\mathbf{p}-\mathbf{p}')\mathbf{S}_a\boldsymbol{\alpha}, \tag{8}$$

where $\mathbf{S}_a$ is the spin of the ath impurity.

We decompose $\Sigma(\mathbf{p},\omega_n)$ into

$$\Sigma(\mathbf{p},\omega_n)=-i\Sigma_0(\mathbf{p},\omega_n)-i\Sigma_1(\mathbf{p},\omega_n)\rho_2\sigma_1+\Sigma_2(\mathbf{p},\omega_n)\rho_1\sigma_2$$
$$+\Sigma_3(\mathbf{p},\omega_n)\rho_3\sigma_3+\Sigma_4(\mathbf{p},\omega_n)\rho_3+i\Sigma_5(\mathbf{p},\omega_n)\sigma_3 \tag{9}$$

and introduce six new quantities defined by

$$\begin{aligned}\tilde{\omega}&=\omega_n+\Sigma_0(\mathbf{p},\omega_n),\\ \tilde{I}&=I+\Sigma_3(\mathbf{p},\omega_n),\\ \tilde{\Delta}&=\Delta+\Sigma_2(\mathbf{p},\omega_n),\\ \tilde{\Omega}&=\Sigma_1(\mathbf{p},\omega_n),\\ \tilde{\xi}_p&=\xi_p+\Sigma_4(\mathbf{p},\omega_n),\\ \tilde{\Lambda}&=\Sigma_5(\mathbf{p},\omega_n).\end{aligned} \tag{10}$$

Expressed in those quantities, G^{-1} can be written as

$$G^{-1}(\mathbf{p},\omega_n)=i(\tilde{\omega}_n+i\tilde{I}\sigma_3\rho_3)-\rho_3(\tilde{\xi}_p+i\tilde{\Lambda}\sigma_3\rho_3)$$
$$-\rho_1\sigma_2(\tilde{\Delta}+i\tilde{\Omega}\sigma_3\rho_3). \tag{11}$$

With this form for $G(\mathbf{p},\omega_n)$, Eqs. (4) and (5) can be solved and equations for $\tilde{\omega}$, $\tilde{\Delta}$, can be obtained. It is useful to introduce $\tilde{\omega}_\pm$, $\tilde{\Delta}_\pm$ defined by

$$\tilde{\omega}_\pm=\tilde{\omega}\pm\tilde{I}i;\quad \tilde{\Delta}_\pm=\tilde{\Delta}\pm\tilde{\Omega}i. \tag{12}$$

Furthermore, as is usually done, we set $\tilde{\omega}_\pm/\tilde{\Delta}_\pm=u_\pm$. In this notation we obtain

$$\begin{aligned}\tilde{\omega}_\pm&=\omega\pm iI+\frac{u_\pm}{(u_\pm^2+1)^{1/2}}\left[\frac{1}{2\tau}+\frac{\langle S_z^2\rangle}{S^2}\frac{1}{2\tau_2}\right]\\ &\quad+\frac{u_\mp}{(u_\mp^2+1)^{1/2}}\left[\frac{\langle S_x\rangle^2}{S^2}\frac{1}{\tau_2}+\frac{1}{\tau_{so}}\right],\\ \tilde{\Delta}_\pm&=\Delta+\frac{1}{(u_\pm^2+1)^{1/2}}\left[\frac{1}{2\tau}-\frac{\langle S_z^2\rangle}{S^2}\frac{1}{2\tau_2}\right]\\ &\quad+\frac{1}{(u_\mp^2+1)^{1/2}}\left[-\frac{\langle S_x\rangle^2}{S^2}\frac{1}{\tau_2}+\frac{1}{\tau_{so}}\right].\end{aligned} \tag{13}$$

Here the following quantities have been introduced:

$$\begin{aligned}\frac{1}{\tau}&=n_aN(0)\int d\Omega|u_1(\theta)|^2+n_bN(0)\int d\Omega|v_1(\theta)|^2+\frac{3}{\tau_{so}},\\ \frac{1}{\tau_{so}}&=\tfrac{1}{3}n_bN(0)\int d\Omega|v_{so}|^2\sin^2\theta,\\ \frac{1}{\tau_2}&=n_aN(0)S^2\int d\Omega|u_2(\theta)|^2.\end{aligned} \tag{14}$$

Furthermore, $\langle S_x^2\rangle=\langle S_y^2\rangle$ has been assumed.

In the limit of a short spin-orbital mean free path, l_{so}, the expressions simplify considerably. In that case we introduce two new quantities u, v defined by

$$u=\tfrac{1}{2}(u_+ + u_-), \quad v=\tfrac{1}{2}(u_+ - u_-) \tag{15}$$

and expand in powers of v. It is shown in the Appendix that up to terms linear in v, a combination of Eqs. (13) gives

$$\frac{\omega}{\Delta}=u\left[1-\frac{1}{(1+u^2)^{1/2}}\left(\frac{1}{\tau_2\Delta}+\frac{I^2\tau_{so}}{2\Delta}\right)\right]+O(\tau_{so}{}^2I^2). \tag{16}$$

This equation is of the same form as the one found by Abrikosov and Gor'kov[3] for the case of magnetic impurities with random spin orientation. Those authors showed that the most important effect of such impurities is to break electron pairs and that the strength of the depairing effect is characterized by a depairing parameter α which equals $\alpha=1/\tau_2\Delta$. Equation (16) shows that the same features hold also for ferromagnetically aligned impurities with the only difference that the depairing parameter is now of the form

$$\alpha=1/\tau_2\Delta+I^2\tau_{so}/2\Delta. \tag{17}$$

Thus the alignment of the impurities leads to an enhancement of the pairbreaking effect.

In order to show that all physical quantities of interest will be unaffected by the alignment of the impurity spins, except for enhancement of the depairing parameter, we have to investigate the influence of the term $i\tilde{\Lambda}\sigma_3$ which appears in $G^{-1}(\mathbf{p},\omega_n)$ [see Eq. (11)]. For this purpose we introduce two functions $\tilde{\xi}_\pm$ which are defined by

$$\tilde{\xi}_\pm=\tilde{\xi}\pm i\tilde{\Lambda}. \tag{18}$$

They can be readily calculated from Eqs. (10) and are given by

$$\tilde{\xi}_\pm=\xi\pm(i\bar{S}_z/2\tau_{12})[u_\pm/(u_\pm{}^2+1)^{1/2}] \tag{19}$$

where

$$1/\tau_{12}=2n_bN(0)\int d\Omega\, u_1(\theta)u_2(\theta).$$

Thus we see that expressions which involve an integration of $G(\mathbf{p},\omega_n)$ over $d\xi$ as, for example, the tunneling density of states, will not depend on τ_{12} and hence will be the same as for impurities with random spin orientations. Physically and mathematically the situation is similar to the one met in the case of anisotropic superconductors, where the anisotropy is washed out by the addition of impurities, which leads to an isotropic gap and to a sharp tunneling density of states as given by BCS.

In our case the difference in the density of states for spin-up and spin-down electrons is washed out by the short electron-spin relaxation time and we are left (because of the depairing effect) not with a BCS type of density of states but with a type which applies to many pairbreaking situations.[6]

Two-particle correlation functions which can be related to vertex functions as discussed in Ref. 6 will depend on τ_{12} only if the vertex contains components which do not commute with $\rho_3\sigma_3$. In that case, the terms proportional to $\tau_{12}{}^{-1}$ enter with different signs into the two Green's functions which belong to every vertex. Inspection of the vertices in Ref. 6 corresponding to the commonly considered physical quantities shows that the only quantity which has a vertex which does not commute with $\rho_3\sigma_3$ is the nuclear-spin relaxation rate. Since in that case the main contribution to the relaxation rate comes from large momentum transfer, the τ_{12} dependence can be neglected here, and we are again back to the problem of random impurity spin distribution. Thus we conclude that all physical quantities of interest are identical for ferromagnetic and paramagnetic impurities if the depairing parameter is modified in the former case.

III. MAGNETIC PROPERTIES

We want to study in this section the behavior of superconductors containing magnetic impurities in the presence of a magnetic field. This is of particular interest in view of recent experiments by Finnemore *et al.* on Gd-La alloys. It is advantageous to consider first the case of a uniform current flow in the superconductor which is quite similar to the magnetic field case.[8] Thus we start with a zeroth-order Green's function $G_0(\mathbf{p},\omega_n)$ which can be written as

$$G_0{}^{-1}(\mathbf{p},\omega_n)=(i\omega_n+\mathbf{q}\cdot\mathbf{v}-I\rho_3\sigma_3-\xi_p\rho_3-\Delta\rho_1\sigma_2)^{-1} \tag{20}$$

where $\mathbf{q}$ is the pairing momentum in the presence of a current and $\mathbf{v}=\mathbf{p}/m$. The self-energy due to impurity scattering is again given by Eq. (5) so that the Green's function G as defined by Eq. (4) can be calculated as easily as in the zero-current case if we restrict ourselves to the dirty limit $\tau\Delta\ll1$. It is convenient to introduce a quantity ζ defined by

$$\zeta=\tfrac{2}{3}\tau_{tr}(qv_F)^2/\Delta \tag{21}$$

which characterizes the strength of the depairing effect due to the current. In this, τ_{tr} denotes the transport mean free time. We obtain in analogy to Eq. (13) the

[8] K. Maki, Progr. Theoret. Phys. (Kyoto) 31, 378 (1964).

following set of equations.

$$\tilde{\omega}_\pm=\omega\pm iI+\frac{u_\pm}{(u_\pm^2+1)^{1/2}}\left[\frac{1}{2\tau}+\frac{\langle S_z\rangle^2}{S^2}\frac{1}{\tau_2}+\frac{3}{2}\frac{\zeta\Delta}{(u_\pm^2+1)^{1/2}}\right]+\frac{u_\mp}{(u_\mp^2+1)^{1/2}}\left[\frac{\langle S_x^2\rangle}{S^2}\frac{1}{\tau_2}+\frac{1}{\tau_{so}}\right],$$
$$\tilde{\Delta}_\pm=\Delta+\frac{1}{(u_\pm^2+1)^{1/2}}\left[\frac{1}{2\tau}-\frac{\langle S_z^2\rangle}{S^2}\frac{1}{\tau_2}-\frac{\zeta\Delta(2u_\pm^2-1)}{2(u_\pm^2+1)}\right]+\frac{1}{(u_\mp^2+1)^{1/2}}\left[-\frac{\langle S_x^2\rangle}{S^2}\frac{1}{\tau_2}+\frac{1}{\tau_{so}}\right]. \quad (22)$$

Here we have dropped all terms of higher order in $(\tau\Delta)$ and also terms of the order τ/τ_{so} which we assume to be much smaller than unity. In order to obtain an equation for the maximum pairing momentum (later upper critical field) as a function of temperature and impurity concentration, we make use of the equation for the order parameter Δ which we write as

$$\Delta=\tfrac{1}{4}|g|T\sum_n\int\frac{d^3p}{(2\pi)^3}\mathrm{Tr}\{\rho_1\sigma_2G(\mathbf{p},\omega_n)\} \quad (23)$$

with g denoting the strength of the electron-electron interaction potential due to phonons. We rewrite Eq. (23) as

$$\ln\frac{T}{T_{c0}}=\pi T\sum_n\left\{\frac{1}{\Delta}-\mathrm{Re}\frac{1}{(u_\pm^2+1)^{1/2}}-\frac{1}{|\omega_n|}\right\} \quad (24)$$

where T_{c0} is the transition temperature in the absence of a current and impurities. "Re" means that the real part has to be taken. If the transition to the normal state is of second order,[9] we can use Δ as an expansion parameter near the transition point. In that case Eqs. (22) simplify to

$$\omega_+=X_+-a+bX_+/X_-,\qquad \omega_-=X_--a+bX_-/X_+ \quad (25)$$

where we have set $u_\pm=X_\pm/\Delta$ and

$$a=\frac{1}{\tau_2}\frac{\langle S_z^2\rangle+\langle S_x^2\rangle}{S^2}+\frac{1}{\tau_{so}}+\zeta\Delta,\quad b=\frac{1}{\tau_{so}}-\frac{1}{\tau_2}\frac{\langle S_x^2\rangle}{S^2}. \quad (26)$$

Thus

$$X_+=\omega_++a-b(\omega_++a+b)/(\omega_-+a+b) \quad (27)$$

and Eq. (24) becomes

$$\ln\frac{T}{T_{c0}}=2\pi T\sum_{n>0}\left\{\frac{\omega_n+a+b}{(\omega_n+a)^2-b^2+I^2}-\frac{1}{\omega_n}\right\}. \quad (28)$$

The right-hand side can be expressed in terms of polygamma functions and we obtain

$$\ln\frac{T}{T_{c0}}+\frac{1}{2}\left[\left(1+\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_-)+\left(1-\frac{b}{(b^2-I^2)^{1/2}}\right)\psi(\tfrac{1}{2}+\rho_+)\right]-\psi(\tfrac{1}{2})=0. \quad (29)$$

Here $\psi(z)$ is the digamma function and $\rho_\pm$ are defined as

$$\rho_\pm=(2\pi T)^{-1}\{a\pm(b^2-I^2)^{1/2}\}. \quad (30)$$

Equation (29) can be used to determine the maximum pairing momentum q. It also applies for the determination of the critical field parallel to a thin film and the upper critical field H_{c2} of a bulk sample. In the former case we simply have to replace q^2 in Eq. (21) by

$$q^2\rightarrow\langle[e\mathbf{A}(x)]^2\rangle$$

where $\mathbf{A}(x)$ denotes the vector potential and the brackets indicate that a spatial average has to be taken. In the latter case we have to replace as shown by one of the authors (K.M.)[10]

$$q^2\rightarrow\tfrac{1}{4}(i\nabla+2eA)^2, \quad (31)$$

where ∇ operates on the space-dependent order parameter, and to multiply both sides of Eq. (29) from the right with $\Delta(\mathbf{r})$.[10] Equation (29) simplifies considerably if we restrict ourselves to certain limiting cases.

a. Random Impurity Spin Orientation

If the magnetic impurities have randomly oriented spins, it follows that $I=0$. In that case

$$\ln(T/T_{c0})+\psi(\tfrac{1}{2}+\rho_0)-\psi(\tfrac{1}{2})=0, \quad (32)$$

where

$$\rho_0=(1/2\pi T)\{1/\tau_2+2\tau_{tr}v_F^2q^2/3\} \quad (33)$$

for the current and parallel magnetic-field case, while

$$\rho_0=(1/2\pi T)\{1/\tau_2+\tau_{tr}v_F^2eH/3\} \quad (34)$$

for the upper critical field case of a bulk sample.

[9] While in the case of a magnetic field parallel to a thin film or perpendicular to a type-II superconducting sample the transition to the normal state is of second order, the situation considered here, namely that of a uniform current flowing, can lead to a first- or second-order phase transition depending on the geometry [see P. Fulde and R. A. Ferrell, Phys. Rev. **131**, 2457 (1963)]. Since we are actually interested in the magnetic-field case we assume in our calculations a second-order transition.

[10] K. Maki, Physics **1**, 21 (1964); **1**, 201 (E) (1964).

b. Short Spin-Orbital Mean Free Path

If the impurities are ferromagnetically aligned and the spin-orbital mean free path is short, we can expand in terms of $b^{-1}=\tau_{so}$ and obtain

$$\ln(T/T_{so})+\psi(\tfrac{1}{2}+\rho_0+I^2\tau_{so}/4\pi T)-\psi(\tfrac{1}{2})=0$$

and from that by expansion in terms of $I^2\tau_{so}/4\pi T$

$$\ln(T/T_{c0})+\psi(\tfrac{1}{2}+\rho_0)-\psi(\tfrac{1}{2})+(I^2\tau_{so}/4\pi T)\psi'(\tfrac{1}{2}+\rho_0)=0. \quad (35)$$

Here $\psi'(z)$ is the derivative of the digamma function and terms of the order $(\tau_{so}\Delta)^2$ have been dropped. At low enough temperature or high enough impurity concentrations the correction terms may become important even if τ_{so} is very small.

In Fig. (1) we have made a comparison between the result of using Eqs. (32) and (34) and experimental data obtained by Finnemore *et al.* In those experiments the upper critical field was determined as a function of temperature and impurity concentration. Since no independent measurement of the resistivity was made for the samples involved, we don't know the mean free path in them. Therefore we have to assume a relationship of the form

$$1/\tau_{tr}=1/\tau_{tr}^{(0)}+\eta n_a \quad (36)$$

where $\tau_{tr}^{(0)}$ is the transport mean free time in the absence of magnetic impurities and to determine $\tau_{tr}^{(0)}$ and η by fitting the slopes of the curves for the La 0.2 at.% Gd and La 0.5 at.% Gd samples. The data for the pure La sample given in Ref. (3) cannot be used in such an analysis because the material was swaged and hence had an especially short mean free path. Figure 1 shows that it would be desirable to have the measurements extended to lower temperatures.

In order to investigate the jump in the specific-heat curve and the slope of the magnetization curve near H_{c2}, we need to know the free energy. For this purpose we set

$$H_{c2}^*(T)=H_{c2}(T)-3/\tau_{tr}v_F^2e\tau_2 \quad (37)$$

where $H_{c2}(T)$ is the upper critical field in the absence of magnetic impurities. Then we can use the expressions derived by one of the authors (K.M.) for the Helmholtz and Gibbs free energy F and G of a type-II superconductor in the immediate subcritical region with depairing taken into account.[10] We only have to replace H_{c2} in Maki's expressions by $H_{c2}^*(T)$. Thus we obtain for the difference of Gibbs free energy between superconducting and normal state

$$\Delta G=-\frac{1}{8\pi}\left[H_0^2+\frac{(H_{c2}^*-H_0)^2}{(2\kappa_2^2(T)-1)\beta}\right] \quad (38)$$

where H_0 is the applied field and $\kappa_2(T)$ and β are defined

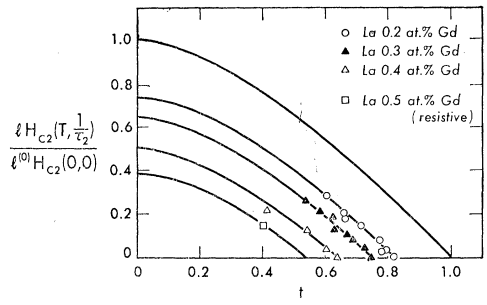

FIG. 1. Upper critical field H_{c2} as a function of reduced temperature $t=T/T_{c0}$ for different magnetic impurity concentrations $n_a\sim 1/\tau_2$. The experimental results were obtained by Finnemore *et al.* and are compared with the present theory. Since the mean free paths l in the presence of magnetic impurities and $l^{(0)}$ in the absence of magnetic impurities are not known, an adjustment has been made as explained in the text.

in Ref. 10. Furthermore, use has been made of a relation between ΔG and ΔF namely

$$\Delta G=\Delta F-H_0B/4\pi \quad (39)$$

where B is the magnetic induction.

The slope of the magnetization as well as the jump in the specific heat at the transition point can be obtained from Eq. (38) directly. We find for the slope of magnetization

$$4\pi\frac{\partial M}{\partial H_0}=\frac{1}{8\pi}\frac{\partial^2}{\partial H_0^2}\left\{\frac{(H_{c2}^*-H_0)^2}{(2\kappa_2^2(T)-1)\beta}\right\}=\frac{1}{(2\kappa_2^2(T)-1)\beta}. \quad (40)$$

It is interesting to notice that the parameter $\kappa_2(T)$ depends on the temperature T only and is independent of τ_2. This is a consequence of the additivity of the different pairbreaking mechanisms involved. Since $\kappa_2(T)$, however, also depends on τ the above statement is correct only as long as τ does not change appreciably if magnetic impurities are added. It would be interesting to check the independence of the slope of the magnetization on the magnetic impurity concentration experimentally.

The jump in the specific heat at the transition point can be calculated from

$$\Delta C_s=-T\frac{\partial^2}{\partial T^2}(\Delta G)\Big|_{H_0=H_{c2}^*}=\frac{T}{4\pi}\frac{1}{(2\kappa_2^2(T)-1)\beta}\left(\frac{\partial H_{c2}^*}{\partial T}\right)^2$$
$$=\frac{T}{4\pi}\frac{1}{(2\kappa_2^2(T)-1)\beta}\left(\frac{\partial H_{c2}(T)}{\partial T}\right)^2. \quad (41)$$

Thus also ΔC_s is independent of magnetic impurity concentration and a function of temperature only. Again this is a consequence of the additivity of the different pairbreaking mechanisms involved.

Finally, we want to remark that for large κ_2 values ($\kappa_2 \geq 2$ is sufficient) the dependence of the slope of magnetization and the jump in the specific heat at the transition point on the mean free time τ is much simplified. We obtain in that case

$$-4\pi(\partial M/\partial H_0)=1/2\kappa_2^2(T)\beta \tag{40a}$$

which is proportional to τ^2 while

$$\Delta C_s=\frac{1}{4\pi}\frac{(\partial H_{c2}^*/\partial T)^2}{2\kappa_2^2(T)\beta} \tag{41a}$$

is independent of τ and depends on T only.

The independence of ΔC_s on the magnetic impurity concentration was shown by Finnemore *et al.* thus supporting our theory.

IV. CONCLUSIONS

The above calculations show that the problem of ferromagnetically aligned impurities in a superconductor can be reduced to the one of impurities with random spin orientation if the mean free path due to spin-orbit interaction is much shorter than the coherence distance. The only difference arising is an increase of the depairing parameter with impurity alignment. Thus all calculations for physical quantities such as tunneling density of states, specific heat, electrical and thermal conductivity, etc. which were done for the paramagnetic case apply also for the ferromagnetic case. As discussed in the text, that is somewhat accidental since physical quantities involving certain spin triplet vertices give different results in both cases even in the limit of short spin-orbit mean free path due to the appearance of certain mixing terms. But none of the usually considered quantities involves such vertices. The experiments by Finnemore *et al.* on Gd-La alloys are found in good agreement with the theory. Especially the additivity of the pairbreaking effects of the magnetic impurities and the field are nicely born out in those experiments. On the other hand, it would be desirable to have experiments performed which are designed to test the increase of the depairing parameter due to impurity spin alignment.

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APPENDIX

In order to derive Eq. (16) we rewrite Eq. (13) as

$$\begin{aligned}\frac{\omega}{\Delta}=u_\pm\mp\frac{Ii}{\Delta}-\frac{1}{\tau_2\Delta}\frac{\langle S_z^2\rangle}{S^2}\frac{u_\pm}{(u_\pm^2+1)^{1/2}}\\ -\left(\frac{1}{\tau_2\Delta}\frac{\langle S_x^2\rangle}{S^2}+\frac{1}{\tau_{so}\Delta}\right)\frac{u_\mp}{(u_\mp^2+1)^{1/2}}\\ +\left(-\frac{1}{\tau_2\Delta}\frac{\langle S_x^2\rangle}{S^2}+\frac{1}{\tau_{so}\Delta}\right)\frac{u_\pm}{(u_\mp^2+1)^{1/2}}.\end{aligned} \tag{A1}$$

We expand the sum and difference of Eqs. (A1) in terms of u, v and obtain

$$\begin{aligned}\frac{\omega}{\Delta}&=u\left(1-\frac{1}{\tau_2\Delta}\frac{1}{(u^2+1)^{1/2}}\right)+\frac{2v^2u}{\tau_{so}\Delta(u^2+1)^{3/2}}+O(v^2),\\ \frac{iI}{\Delta}&=\frac{2v}{(u^2+1)^{1/2}}\frac{1}{\tau_{so}\Delta}+O(v)\end{aligned} \tag{A2}$$

by making use of the following expansions

$$\begin{aligned}\frac{u_+}{(u_+^2+1)^{1/2}}+\frac{u_-}{(u_-^2+1)^{1/2}}&=\frac{2u}{(u^2+1)^{1/2}}\left(1-\frac{3v^2}{2(1+u^2)^2}\right),\\ \frac{u_+}{(u_+^2+1)^{1/2}}-\frac{u_-}{(u_-^2+1)^{1/2}}&=\frac{2v}{(u^2+1)^{3/2}},\\ \frac{u_+}{(u_-^2+1)^{1/2}}+\frac{u_-}{(u_+^2+1)^{1/2}}&=\frac{2u}{(u^2+1)^{1/2}}\left(1+\frac{v^2(4u^2+1)}{2(u^2+1)^2}\right),\\ \frac{u_+}{(u_-^2+1)^{1/2}}-\frac{u_-}{(u_+^2+1)^{1/2}}&=\frac{2v}{(u^2+1)^{3/2}}(1+2u^2).\end{aligned} \tag{A3}$$

From (A2), Eq. (16) of the text follows immediately.