Spin susceptibility and spin-spin relaxation in superconductors

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(Received 9 May 1984)

It is shown that the conduction-electron spin susceptibility $\chi(r)$ and thus the indirect interaction between magnetic impurities in superconductors can lead to new behavior in very dilute systems that is absent at higher impurity concentrations and that is compatible with recent experiment. In the superconducting state we find that $\chi(r)$ decays exponentially at *all* r, the spin-spin coupling can be antiferromagnetic at intermediate r, and the spin-spin relaxation rate depends markedly on Fermi-surface anisotropy.

It is generally accepted that the presence of the energy gap in a superconductor leads to exponential decay of the conduction-electron spin susceptibility $\chi(r)$, and, therefore, of the electron-mediated indirect interaction between magnetic moments, at distances greater than the coherence length $\xi(T)$.[1,2] Since most studies of magnetic impurity interactions have been made on concentrated samples in which the separations of magnetic ions are *smaller* than ξ, this and other effects of the gap have been generally neglected. Recently, however, Kumagai and Fradin[3] have performed NMR experiments on very dilute samples of $Y_{1-c}R_cRh_4B_4$ ($R=$Er, Gd) in which impurity separations are comparable to $\xi(T)$. They find longitudinal relaxation rates of the paramagnetic ions which are *linear* in concentration in the normal state, and which decrease rapidly with T for $T<T_c$ by factors 10–100 times any previously observed. These facts lead us to consider the indirect interaction between rare-earth (RE) ions as the responsible mechanism. Accordingly, we have studied $\chi(r,\xi)$ in detail and summarize the results here. We confirm the exponential decay[2] and find it to be present at *all* r; and we find antiferromagnetic coupling at r/ξ of order unity. Furthermore, the suppression of the relaxation rate below T_c by this exponential damping factor is found to be much too weak to account for the data if $\chi(r)$ is assumed to be isotropic. However, on invoking anisotropy of the Fermi surface, hence, also of $\chi(r)$ and the magnetic interactions, we find a pronounced dependence of this relaxation rate on the degree of anisotropy for very low concentrations ($c\sim10^{-4}$).

We start from the general expansion of the isotropic susceptibility χ in a series of thermal-Green's-function products.[1] We then replace fermion occupation factors by their $T=0$ limits while retaining the temperature dependence of the gap Δ, and obtain an *analytic* result. For finite coherence length ξ, we define a funtion $\Phi(r,\xi)$ by

$$\chi(r,\xi)=-[A/(2k_Fr)^3]\Phi(r,\xi)\ , \tag{1a}$$

where

$$\Phi(r,\infty)=\cos(2k_Fr)-\sin(2k_Fr)/(2k_Fr)$$

is the usual normal-state oscillatory factor and A a constant. The analysis then gives

$$\Phi(r,\xi)=\Phi(r,\infty)[zK_1(z)] -\tfrac{1}{2}[\sin(2k_Fr)/(2k_Fr)]z^2K_0(z) +[1-\cos(2k_Fr)]z\int_z^\infty K_0(x)dx\ , \tag{1b}$$

with

$$z=(2/\pi)(r/\xi)\ , \tag{1c}$$

where K_0, K_1 are modified Bessel functions. For $r<<\xi$, $\Phi(r,\xi)\approx\Phi(r,\infty)$. For $r>>\xi$,

$$\Phi(r,\xi)\sim\Phi(r,\infty)\exp[-(2/\pi)(r/\xi)]$$

exhibits the anticipated suppression of the range of χ. Plots of Eq. (1) in Fig. 1 display this damping. The plots also show that for r/ξ of order unity, $\chi(r,\xi)$ oscillates without vanishing about a finite value corresponding to antiferromagnetic coupling. The distances at which these interesting features occur are comparable to the magnetic ion separations in the samples of Ref. 3.

To introduce the effects of anistropy, imagine an extreme limit of Fermi-surface nesting in which the constant energy surfaces consist of three pairs of parallel planes normal to the k_x, k_y, k_z axes. The normal-state susecptibility for such a system is readily found to be

$$\chi(x,y,z)=\delta_{x,o}\delta_{y,o}\chi_{1D}(z)+\cdots\ ,$$

where the ellipsis stands for cyclic terms. In this limit, a magnetic ion at the origin can interact only with those other magnetic ions that lie on the coordinate axes. Similarly, if the one-electron energy surfaces are three cylinders along k_x, k_y, k_z, a magnetic ion at the origin can interact significantly only with other magnetic ions lying in the three coordinate planes, via a two-dimensional (2D) susceptibility function.

We now concentrate on effects of the exponential damping factor, and recast $\chi(r)$ to exhibit clearly that the exponential dependence on r is analytically exact for *all* r, not just asymptotically. The form of $\chi(r)$ that we display was not derived from Eq. (1b), but from the general Green's function expression[1] by using a particular sequence of ana-

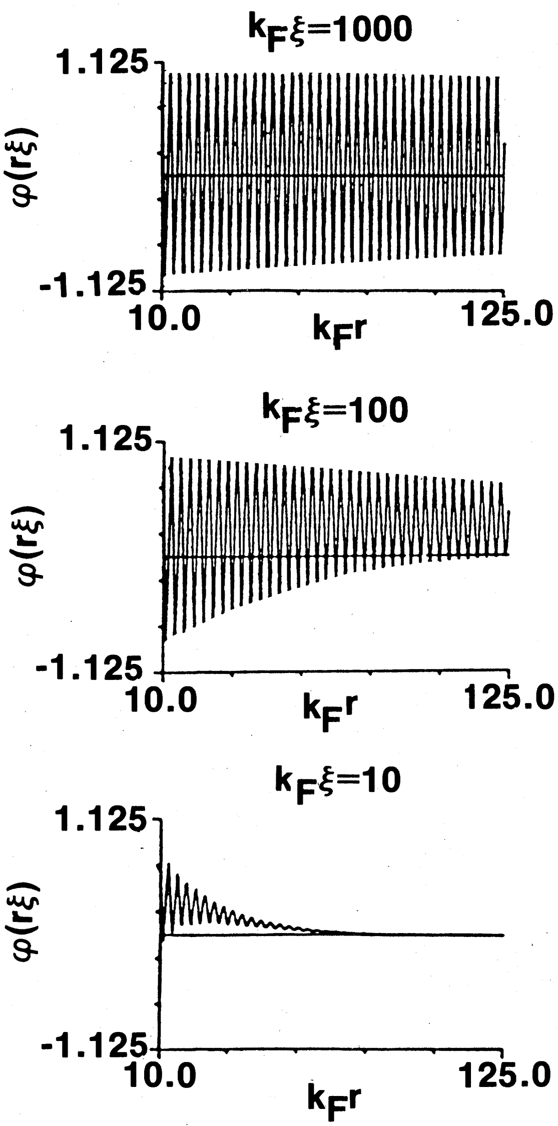

FIG. 1. Function $\Phi(r,\xi)$ of Eq. (1) for the isotropic susceptibility as a function of r for three values of ξ as indicated. For $k_F\xi = 1000$, essentially normal-state behavior persists to the largest r of the plot. For $k_F\xi = 100$, the exponential decay and antiferromagnetic bias are clear. The plot of $k_F\xi = 10$ is an extreme example of both the damping and the bias. Note that the range of k_Fr shown corresponds to different orders of magnitude of r/ξ in these three plots.

lytic continuations. We thereby can show that for any dimension and for all r

$$\chi(r,\xi) \cong \exp(-2r/\pi\xi)\chi(r,\infty), \qquad (2)$$

where $\chi(r,\infty)$ is the normal-state susceptibility.[4] Equation (2) omits the antiferromagnetic bias seen in Fig. 1, while fully retaining the damping. The spin-spin interaction of the magnetic ions vanishes and ceases to contribute to relaxation when $\xi(T)$ becomes small compared to the mean impurity separation.

To demonstrate the consequences of combining anisotropy and damping, we have made calculations for limiting models in which the magnetic interactions are governed by a one-dimensional or by a two-dimensional exponentially damped $\chi(r)$, as well as for the isotropic case. The paramagnetic relaxation as a function of $\xi(T)$ is determined by the Fourier transform of the spin-spin autocorrelation function

$$(J_z(t),J_z(o))/(J_z(o),J_z(o))$$

for the magnetic system, which we denote by $P_n(t)$ for the case of n-dimensional interactions. Following standard techniques and averaging procedures[5,6] we obtain

$$\ln P_n(t) = -(c/a_0^n)\alpha_n \int_0^\infty d\,r r^{n-1}\{1-\exp[-t^2\omega_n^2(r)]\}, \qquad (3a)$$

where α_n is a purely geometric factor, a_0 the linear spacing of substitutional sites, and c the fraction of substitutional sites that is occupied by magnetic impurities. The averaging gives

$$\omega_n(r) = (\lambda_n/r^n)\exp(-2r/\pi\xi), \qquad (3b)$$

where λ_n is a constant independent of c and proportional to the strength of the interaction. When $\xi \to \infty$, $\ln P_n(t)$ reduces to $[-t/\tau_n(\infty)]$ with a normal-state relaxation time $\tau_n(\infty)$ inversely proportional to c and independent of temperature.

Introducing the mean separation of magnetic impurities $d \equiv (a_0/c^{1/3})$ and changing the variable of integration, we obtain

$$\ln P_n(t) = -[t/\tau_n(\infty)]I_n\{A_n c^{n/3-1}(d/\xi)^n[t/\tau_n(\infty)]\}, \qquad (4a)$$

where

$$I_n(z) = (1/\sqrt{\pi})\int_0^\infty dx\,(1-\exp\{-(1/x^2)\exp[-(zx)^{1/n}]\}) \qquad (4b)$$

and A_n is a constant weakly dependent on n and independent of temperature. All temperature dependence arises from the coherence length $\xi(T)$ appearing explicitly, and the spin-spin relaxation rate is independent of T above T_c. In general, $I_n(z)$ drops rapidly at small z, followed by a very slow asymptotic decrease to zero. For a given ξ of experimental interest both the asymptotic reduction and the rapidity of the initial drop are *much* greater for $n=1$ than for $n=3$ because of the n-dependent factors multiplying $[t/\tau_n/\infty)]$ in the argument of I_n. At the low concentrations used by Kumagai and Fradin, one expects d/ξ of order unity at $T=0$. The crucial factor $c^{n/3-1}$ in the argument of I_n clearly arises because the interactions have an effective dimensionality lower than that of the system. For $c=2\times10^{-4}$, the factors $c^{n/3-1}$ for $n=1,2,3$ are in the ratios 292:17:1. Illustrative plots of the I_n as functions of $t/\tau_n(\infty)$ are presented in Fig. 2 for two values of d/ξ. Since the argument of I_n is scaled by $(d/\xi)^n$, plots for other values of (d/ξ) are similar but with the initial drop occurring in an expanded or contracted interval of $t/\tau_n(\infty)$.

Kumagai and Fradin have fitted their $T < T_c$ data using a Lorentzian (Fourier transform) $P_n(\omega)$ with a T-dependent relaxation time, and find reductions of the relaxation rate by factors of up to 100 and more. As Fig. 2 shows, I_n can have an asymptotic tail which varies quite slowly on the time scale $2\pi/\omega_0$ where ω_0 is an NMR frequency. Accordingly, $P_n(\omega)$ can closely resemble a Lorentzian with a relaxation rate reduced by an average factor $\langle I_n\rangle$ typical of the near asymptotic region. For this to be true, the initial drop of I_n must be, of course, sufficiently rapid. Inspection of

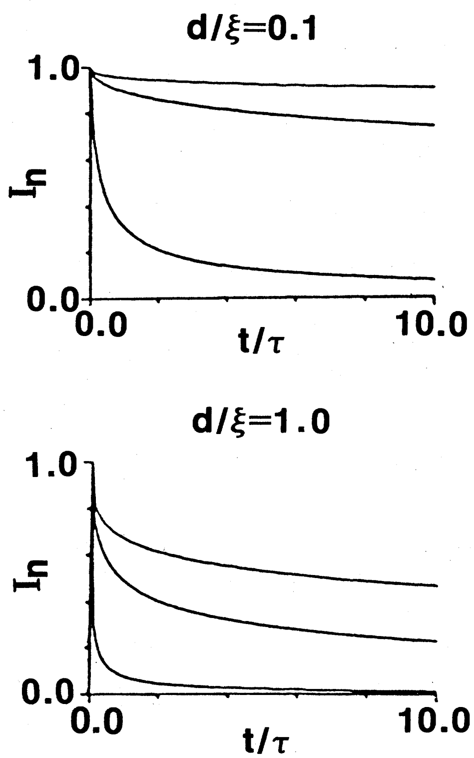

FIG. 2. Functions I_n of Eq. (4) which govern suppression in the superconducting state of the spin-spin relaxation as given by the correlation function $P_n(t)$. Plotted vs time in units of the normal-state relaxation time $\tau=\tau_n(\infty)$ for the indicated ratios (d/ξ) of magnetic impurity spacing to coherence length, and for an impurity fraction $c=2\times10^{-4}$. In each plot, the upper curve is that for the isotropic model $(n=3)$, the middle curve is for the anisotropic model with effectively two-dimensional magnetic interactions $(n=2)$, and the lower one is for $n=1$. For $(d/\xi)\geqslant2$, I_1 is, at $t/\tau\geqslant5$, too small on the scale of the plots to be discernible from the axis.

the asymptotic tails in Fig. 2 shows that one can anticipate a reduction of the relaxation rate by a factor of only 4 or 5 in the isotropic model $(n=3)$. In contrast, for the $n=1$ anisotropic model one anticipates reductions by factors which are already 20 to 30 for $d/\xi=0.1$ and become well over 100 at lower temperatures. Numerical evaluations of $P_n(\omega)$ bear out these expectations, and show that suppression of the spin-spin relaxation below T_c is much too weak in the isotropic model to account for the data, while the $n=1$ anisotropic model overdoes the suppression. Lack of any Fermi-surface information precludes a more detailed model. Despite the simplicity of our model calculations this variation with n is sufficiently dramatic to suggest a strongly anisotropic Fermi surface in these systems.

We have obtained an analytic expression for the isotropic susceptibility in the superconducting state that exhibits both exponential decay at large r/ξ and a bias toward antiferromagnetic coupling for r/ξ of order unity. We further found the exponential dependence on r to be analytically exact in any dimension, and at *all r*, not just asymptotically. It is to be noted that the occurrence of an exponential factor depends only on the existence of the gap, not on the strength of the pairing interaction. It was pointed out that strong anisotropy of the Fermi surface can lead to indirect magnetic interactions of, effectively, a dimensionality lower than that of the system. This, in turn, was shown to have a pronounced effect on the suppression of the spin-spin relaxation rate by the presence of an energy gap. This suppression and any effects of anisotropy on it are experimentally detectable only at very small spin concentrations. Conditions favorable to detection were realized in the experiments of Kumagai and Fradin, and comparison of our results with their data suggests strong Fermi-surface anisotropy in the systems they studied.

We wish to thank B. D. Dunlap, F. Y. Fradin, and M. Tachiki for many conversations covering all aspects of this work. This work was supported by the U.S. Department of Energy.

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[4]The exact analytic result for the 1D case has the form

$$\Phi(r,\xi)=\exp(-2k_F r\sinh x_0)F(r,x_0) \ ,$$

where x_0 is defined by $\sinh(2x_0)\equiv(\Delta/E_F)=$(energy gap)/(Fermi energy) and $F(r,0)=\Phi(r,\infty)$. Expansion of the integral $F(r,x_0)$ for $x_0<<1$ produces the antiferromagnetic terms as the leading corrections to $F(r,0)$. On expanding $\sinh x_0$ to lowest order in (Δ/E_F) and neglecting (Δ/E_F) as compared to 1 in $F(r,x_0)$, we obtain Eq. (2). The two- and three-dimensional cases are similar.
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