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Effect of Nonlocality on the Spin Correlations in Ferromagnetic Superconductors

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The dipole and Ruderman-Kittel-Kasuya-Yosida interactions between impurity spins in a superconductor are suppressed at long wavelengths. This results in a maximum in the neutron scattering at a temperature-independent wave number. It is essential to use nonlocal theory for the diamagnetic screening by the superconducting electrons. The local London approximation affects the neutron scattering cross section and overestimates the spin fluctuation energy by one order of magnitude.

In this note we wish to report results on the theory of spin correlations in ferromagnetic superconductors, which we have obtained in mean-field theory, taking into account the effect on the spin fluctuations of the superconducting electrons. At the time that our report was being prepared for publication, the work of Blount and Varma[1] came to our attention. In this paper we will, therefore, in addition to presenting our own results, discuss the ways in which they differ from those of Blount and Varma. The principal difference is that the diamagnetic response of the superconducting electrons to the spin impurities is drastically affected by nonlocality. This reduces the change in free energy by an order of magnitude and makes the spin fluctuations more compatible with the superconducting state. The nonlocality also affects the "shape" of the spin correlations. Furthermore, we include the Ruderman-Kasuya-Kittel-Yosida[2] (RKKY) interaction in our calculation. Both of these aspects of our theory ought to be verifiable by neutron scattering.

A strong indication of the important role played by the RKKY interaction has been noted by Redi and Anderson[3] who find that the assumption of purely magnetic dipole interaction in the rare-earth Chevrel compounds leads to antiferromagnetic rather than to the observed ferromagnetic ordering. They conclude that the favoring of the ferromagnetic state "...is due to conduction electron effects." Therefore we regard any treatment of the spin correlations which neglects the RKKY interaction as unrealistic. Neverthrless, in spite of the relative importance of the RKKY interaction compared to the dipole interaction, we can treat it on an absolute scale as a weak perturbation on the superconducting electrons. It is the experimental fact of the weak coupling of the spins to the electrons and the occurrence of a first-order rather than a second-order transition that permits us to adopt a perturbation theory approach, using linear-response functions for the superconducting electrons.

The Weiss mean-field theory gives χ^{-1}, the reciprocal of the spin susceptibility, in terms of the Curie susceptibility, $\chi_C = C/T$, as

$$\chi^{-1} = \chi_C^{-1} - \lambda, \tag{1}$$

where λ is the Weiss molecular-field coefficient. λ is the ratio of the molecular field to the magnetization density. For a transverse magnetic polarization density of very long wavelength, there is no depolarizing effect and the Weiss field is 4π times the magnetization density. Thus, $\lambda = 4\pi$. At the finite wave number q, λ is reduced by the fraction $(q/q_D)^2$, where q_D is a Debye cutoff, inversely proportional to the average interspin spacing. Therefore,

$$\lambda(q) = 4\pi(1 - q^2/q_D^2). \tag{2}$$

Substitution of Eq. (2) into Eq. (1) gives the Ornstein-Zernike result,[4]

$$\chi_n^{-1}(q,\kappa) = (4\pi/q_D^2)(q^2 + \kappa^2). \tag{3}$$

The square of the inverse correlation length is

$$\kappa^2 = (q_D^2/4\pi C)(T - T_C). \tag{4}$$

The Curie temperature is $T_C = 4\pi C$. The critical exponents have their mean-field values, $\nu = \frac{1}{2}$ and $\gamma = 2\nu = 1$.

The transverse magnetic field of wave number q which is produced by the spin fluctuations will be shielded in the superconducting state by the diamagnetic response of the superconducting electrons. If we should use the local London equation and the local London penetration depth, λ_L, the screening factor would be $(1 + q^{-2}\lambda_L^{-2})^{-1}$. But both from the sum rule[5,6] and from BCS[7] theory it is known that local London screening is a gross overestimate in the large-q range. This is particularly true for weak superconductors, where the correction factor for nonlocality is $(q\xi_D)^{-1}$. This serves to define the "diamagnetic coherence length" ξ_D, which in weak superconductors is generally an order of magnitude larger than λ_L, under the assumption that the electron mean free path is not especially small. The Weiss coefficient becomes, consequently, in the superconducting state,

$$\lambda_S(q) = \frac{\lambda_n(q)}{1 + (q\xi_D)^{-1}(q^2\lambda_L^2)^{-1}} \approx \lambda_n(q) - \frac{4\pi}{q^3\xi_D\lambda_L^2}. \tag{5}$$

Substitution of Eq. (5) into Eq. (1) yields for the inverse spin susceptibility in the superconducting state

$$\chi_S^{-1}(q,\kappa) = \frac{4\pi}{q_D^2}\left(q^2 + \frac{q_D^2}{q^3\xi_D\lambda_L^2} + \kappa^2\right) = 4\pi(q_c/q_D)^2\overline{\chi}_S^{-1}(x,\overline{\kappa}), \tag{6}$$

where the scaled dimensionless inverse susceptibility is

$$\overline{\chi}_S^{-1}(x,\overline{\kappa}) = x^2 + x^{-3} + \overline{\kappa}^2, \tag{7}$$

with $\overline{\kappa} = \kappa/q_c$ and $x = q/q_c$. Here we have introduced the characteristic wave member q_c defined by

$$q_c^5 = q_D^2/\xi_D\lambda_L^2. \tag{8}$$

With $\xi_D/\lambda_L \sim 10$ and $q_D\lambda_L \sim 10^2$, Eq. (8) shows q_c to be somewhat more than one order of magnitude smaller than q_D, which justifies the approximation in Eq. (5).

In the above treatment of the nonlocal electrodynamics we have ignored l, the finite mean free path of the conduction electrons. This is justified because we are probing the electron system at neutron-scattering wavelengths of the order of q_c^{-1}. As noted above, this is only one order of magnitude greater than the interatomic spacing. As better samples become available, with larger l values, the approximation $ql \gg 1$ will become even better. It might be objected that the samples are generally type-II superconductors, while the treatment given here would be appropriate for a type-I superconductor. But this would be incorrect and in the present context the usual classification scheme is irrelevant. By probing at distances less than l we can effectively set l equal to infinity.

Equation (7) determines the stiffness of the system with respect to spin fluctuations at the scaled wave number $x = q/q_c$. A further effect on the magnetic stiffness is produced by the RKKY[2] interaction between spins, mediated by the conduction electrons. This can be visualized as a polarization cloud of conduction-electron spin surrounding each localized impurity spin. In the superconducting state a second polarization cloud appears,[8] of opposite sign and of range many orders of magnitude larger (ξ_D instead of q_F^{-1}, the reciprocal of the Fermi wave number). The strength of the second, extended cloud is such that the two clouds cancel exactly (in the absence of spin-orbit effects), in the $q \to 0$ limit. But most of the extended negative polarization cloud becomes ineffective[9] as soon as q gets larger than ξ_D^{-1}. In that case only the fraction $(q\xi_D)^{-1}$ of the RKKY interaction is lost in passing to the superconducting state. This means that, if the RKKY interaction is contributing to the Weiss field, we must add to $\overline{\chi}_S^{-1}$ the additional stiffness K/x, giving

$$\overline{\chi}_S^{-1}(x, \overline{\kappa}, K) = x^2 + (K/x) + x^{-3} + \overline{\kappa}^2, \tag{9}$$

where K is a dimensionless RKKY strength parameter.

The upper part of Fig. 1 shows Eq. (9) plotted versus x^2 for the two values $K = 0$ and $K = 1$. Also shown is the normal-state Ornstein-Zernike stiffness. All curves are drawn for $\overline{\kappa} = 0$. Nonzero values of $\overline{\kappa}$ simply shift the function up and do not change its shape. The minimum stiffness remains at the same wave number, independent of temperature. The neutron-scattering intensity is determined by the mean-square spin fluctuation and is thus proportional to the reciprocal of the stiffness, i.e., to $\overline{\chi}_S$ itself. This function is plotted in the lower part of Fig. 1. It will be seen that the maximum neutron-scattering intensity occurs at a certain temperature-independent value of the wave number. The temperature dependence serves to raise or lower the intensity at the maximum. The dashed curves are obtained from the London local approximation [x^{-2} instead of x^{-3} in Eq. (9)], with the additional neglect of the RKKY interaction, and correspond to the Blount-Varma calculation. As can be seen, nonlocality causes the neutron-scattering intensity to drop more abruptly on the low-q side of the maximum, while inclusion of the RKKY interaction makes the intensity fall off more slowly

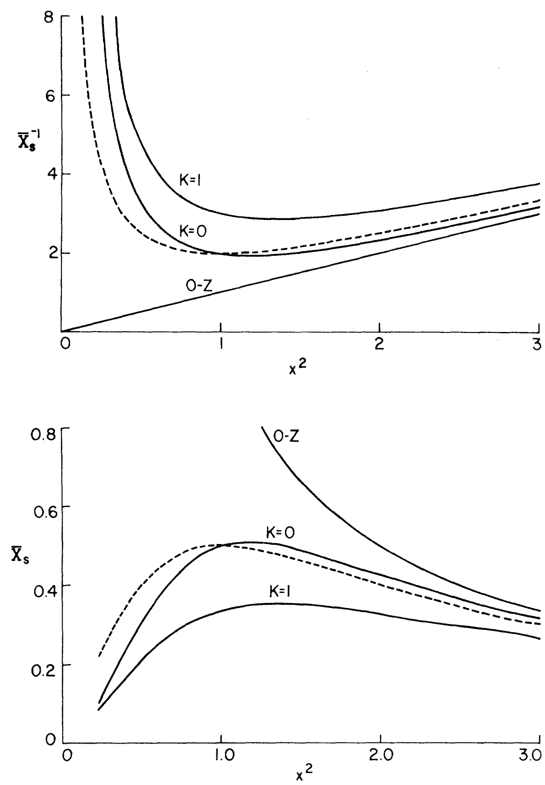

FIG. 1. Critical spin susceptibility $\overline{\chi}_S$ (lower half) and inverse susceptibility (upper half), vs square of wave number, with and without RKKY interaction ($K=1$ and 0, respectively). $x = q/q_c$, where q_c is a characteristic wave number determined by the superconductivity parameters. The curve labeled O-Z shows the normal-state Curie-point susceptibility. The dashed curve neglects RKKY and nonlocality.

on the high-q side. Detailed experimental studies ought to be able to verify the nonlocal effect and to fix the RKKY parameter K.

It remains to study the stability of the superconducting state with respect to the spin fluctuations. The impurity spins will tend to destroy the superconducting state by raising its free energy density relative to the normal state. This rise is

$$\begin{aligned}\Delta F &= F_s(\overline{\kappa}, K) - F_n(\overline{\kappa}, K)\\ &= \frac{k_B T}{(2\pi)^3}\int d^3k \ln \frac{\overline{\chi}_S^{-1}}{\overline{\chi}_n^{-1}}\\ &= q_c^3 (k_B T/2\pi^2) I(\overline{\kappa}, K),\end{aligned} \tag{10}$$

where

$$I(\overline{\kappa}, K) = \int_0^{q_D/q_c} dx\, x^2 \ln\left(1 + \frac{x^{-3} + K/x}{x^2 + \overline{\kappa}^2}\right). \tag{11}$$

It will be noted that the RKKY constant plays a key role in the integration. A relatively small value has a large effect on the value of the integral. We therefore consider the behavior of Eq. (11) in the vicinity of $K=0$. Linearizing within the integrand, we obtain

$$I(\bar{\kappa},K)\approx I(\bar{\kappa},0)+K\ln\frac{q_{\rm D}}{\max(\kappa,q_c)}, \qquad (12)$$

where the first term can now be evaluated with the upper limit set equal to infinity. We observe that the integral in Eq. (11) is not well behaved at $x=0$. This is because in this extreme limit the approximation of Eq. (5) is no longer valid. However, as noted in the discussion following Eq. (5), the approximation is good even for values of q smaller than q_c by about an order of magnitude. This means that the integral is in trouble in the very small range $0\leq x\lesssim\frac{1}{10}$, where a more accurate χ_s^{-1} has to be used. For present purposes we ignore this fine detail and cut off the integral at a lower limit. The second term has been evaluated to logarithmic accuracy only.

A rough estimate of the stability of the superconducting state can easily be obtained from Eq. (10). Not shown is the superconducting coherence energy, which lowers the superconducting state by an amount of the order of $q_{\rm F}^3(k_{\rm B}T)^2/\epsilon_{\rm F}$. The ratio of Eq. (10) to the unperturbed superconducting coherence energy can therefore be estimated, by taking I to be ~ 1, as

$$\frac{\Delta F}{q_{\rm F}^3}\frac{\epsilon_{\rm F}}{(k_{\rm B}T)^2}\approx\left(\frac{q_c}{q_{\rm F}}\right)^3\frac{\epsilon_{\rm F}}{k_{\rm F}T}=\frac{q_{\rm D}^3}{q_{\rm F}^3}\left(\frac{q_c}{q_{\rm D}}\right)^3\frac{\epsilon_{\rm F}}{k_{\rm F}T}$$
$$\approx 10^{-4}\epsilon_{\rm F}/k_{\rm B}T. \qquad (13)$$

One order of magnitude has come from $q_{\rm D}^3/q_{\rm F}^3$, the ratio of spin to conduction-electron densities. The others follow from the estimate for $q_c/q_{\rm D}$ given above. As $\epsilon_{\rm F}/k_{\rm B}T$ is $\sim 10^4$, Eq. (13) indicates that the competition between superconductivity and ferromagnetism is quite close. With further lowering of the temperature, the ratio in Eq. (13) will rise above unity, corresponding to the observed first-order phase transition.[10-12] If the nonlocality of the diamagnetic response is neglected and local London theory used, as done by Blount and Varma, the competition in Eq. (13) is an order of magnitude more unfavorable for the superconducting state. Thus the local approximation might suggest incorrectly a first-order transition into the paramagnetic state.

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