Possible superconductivity in nearly antiferromagnetic itinerant fermion systems

M. T. Béal-Monod and C. Bourbonnais
Laboratoire de Physique des Solides, Bâtiment 510, Université Paris-Sud, 91405 Orsay, France

V. J. Emery
Physics Department, Brookhaven National Laboratory, Upton, New York 11973
and Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France
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Strong spin fluctuations arising in itinerant fermion systems close to a magnetic instability may induce or inhibit superconductivity depending on the nesting wave vector $\mathbf{q}_0$ for which the instability occurs. If $|\mathbf{q}_0|$ is small but finite, triplet pairing is favored and singlet pairing is suppressed as efficiently as in nearly ferromagnetic systems ($q_0=0$). If $|\mathbf{q}_0|$ is large, there is a repulsive contribution from backward scattering by which triplet as well as singlet pairings are strongly depressed. The cases of UPt_3, $CePb_3$, and some organic compounds are considered.

It has been known for a long time that strong spin fluctuations ("paramagnons[1]") in nearly ferromagnetic (NF) fermion systems, prevent BCS singlet pairing superconductivity,[1b] but favor triplet pairing.[2] This result has been well illustrated in the case of the triplet superconductivity of liquid ^{3}He.[3] The present note considers the effects of a paramagnon-mediated interaction near to a magnetic instability at finite wave vector $\mathbf{q}_0$. It is found that if the system is nearly antiferromagnetic (NAF) with a large value of $|\mathbf{q}_0|$, then backward scattering from the paramagnon-mediated interaction is repulsive for both parallel and antiparallel spin states, so that both triplet and singlet pairings are suppressed. As the wave vector $\mathbf{q}_0$ is varied (increasing from zero), the paramagnon effect changes continuously from a qualitatively ferromagnetic to a qualitatively antiferromagnetic character, so that the effective interaction between parallel spins changes continuously from a strong attraction to a strong repulsion. These effects appear relevant for the study of possible mechanisms responsible for the observed superconductivity in the heavy-fermion systems UPt_3, $CePb_3$, or in some organic compounds under pressure such as the Bechgaard ditetramethyltetraselenafulvalenium salts $(TMTSF)_2X$.

We first briefly recall some known results. In the paramagnon model[1] for a single parabolic band of itinerant fermions, a strong Hubbard contact repulsion I among opposite spins [Fig. 1(a)] can induce a magnetic transition when it is strong enough for the Stoner criterion:

$$1-\bar{I}=0 \tag{1}$$

to be fulfilled. Here

$$\bar{I}=I\max\chi^0(\mathbf{q},0)=I\chi^0(\mathbf{q}_0,0) \tag{2}$$

and $\chi^0(\mathbf{q},\omega)$ is the dynamic spin-correlation function for free fermions as a function of the momentum transfer $\mathbf{q}$ and the frequency ω. When max $\chi^0(\mathbf{q},0)$ occurs for $\mathbf{q}=\mathbf{q}_0=0$, the system undergoes a ferromagnetic instability; when it occurs for one or several finite values of $\mathbf{q}$ a modulated magnetic structure is produced.

If $\bar{I}$ is slightly less than 1, the system remains paramagnetic, although "nearly magnetic", and exhibits strong spin fluctuations, the paramagnons, which renormalized all the properties of the system.[1,4,5] In particular, from the bare repulsive interaction I among opposite spins of Fig. 1(a), paramagnon-mediated effective interactions are generated between opposite spins [Figs. 1(b) and 1(c)] and among parallel spins [Fig. 1(d)]. Spin constraints impose an even number $(2n)$ of closed loops or elementary bubbles in the longitudinal paramagnon of Fig. 1(b) exchanged between two opposite spins; for the same reason, an odd number $(2n+1)$ of such closed loops is needed in the longitudinal paramagnon of Fig. 1(d). This means[6] that the contributions of the two diagrams are multiplied,

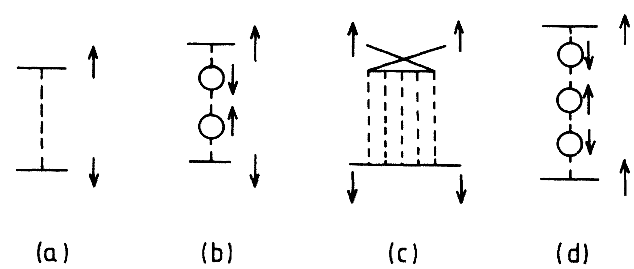

FIG. 1. (a) The bare interaction I, dashed line, of contact type among opposite spins, solid lines. (b) The paramagnon-mediated interaction among opposite spins; the longitudinal paramagnon exchanged between the two opposite spins must be read as containing an even $(2n)$ number of elementary bubbles as explained in the text. (c) The paramagnon-mediated interaction among parallel spins; an odd $(2n+1)$ number of bubbles is involved in the longitudinal paramagnon exchanged between parallel spins. (d) The paramagnon-mediated interaction among opposite spins involving a transverse paramagnon (an infinite ladder in the interaction I).

respectively, by $(-1)^{2n}=+1$ and $(-1)^{2n+1}=-1$, and the two paramagnon-mediated interactions thus exhibit opposite signs. On the other hand the transverse paramagnon of Fig. 1(c) does not contain any closed loop, so no extra factor enters. Let us consider the effective interactions involved in Fig. 1(b), 1(c), and 1(d), for a pair scattering from $(\mathbf{k},\sigma;-\mathbf{k},\sigma')$ into $(\mathbf{k}',\sigma;-\mathbf{k}',\sigma')$ with the spin indices σ and σ' being $\uparrow$ or $\downarrow$. One gets the well-known results:[7] from Fig. 1(b):

$$V_{\uparrow\downarrow}(\mathbf{k}'-\mathbf{k})=\frac{I^3[\chi^0(|\mathbf{k}'-\mathbf{k}|)]^2}{1-I^2[\chi^0(|\mathbf{k}'-\mathbf{k}|)]^2}\ , \tag{3a}$$

from Fig. 1(c):

$$V_{\uparrow\downarrow}(\mathbf{k}'+\mathbf{k})=\frac{I^2\chi^0(|\mathbf{k}'+\mathbf{k}|)}{1-I\chi^0(|\mathbf{k}'+\mathbf{k}|)}\ , \tag{3b}$$

from Fig. 1(d):

$$V_{\uparrow\uparrow}(\mathbf{k}'-\mathbf{k})=V_{\downarrow\downarrow}(\mathbf{k}'-\mathbf{k})=-\frac{I^2\chi^0(|\mathbf{k}'-\mathbf{k}|)}{1-I^2[\chi^0(|\mathbf{k}'-\mathbf{k}|)]^2}\ . \tag{3c}$$

It should be noted that the signs of the various contributions to Eqs. (3) are determined by spin constraints and are independent of the wave vector $\mathbf{q}_0$ at which χ^0 has a maximum.

To make a comparison between (NF) and nearly antiferromagnetic (NAF) systems, we shall first assume rotational invariance in real space and spin space, and consider the two extremes $\mathbf{q}_0=\mathbf{0}$ and $|\mathbf{q}_0|=2k_F$, where k_F is the Fermi wave vector. Since the important states are close to the Fermi surface, the momentum transfer may be written

$$|\mathbf{k}'\pm\mathbf{k}|^2\simeq 2k_F^2(1\pm\mu)\ , \tag{4}$$

where $\mu=(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')$ is the cosine of the scattering angle. Now the singlet and triplet scattering amplitudes may be expanded in partial waves:

$$V_s(\mu)=\sum_{\text{even}\,l}(2l+1)V_lP_l(\mu)\ , \tag{5}$$

$$V_t(\mu)=\sum_{\text{odd}\,l}(2l+1)V_lP_l(\mu)\ , \tag{6}$$

where $P_l(\mu)$ are Legendre polynomials. Denoting the contributing from Eqs. (3a), (3b), and (3c) by $F_1(1-\mu)$, $F_2(1+\mu)$, and $-F_3(1-\mu)$, respectively, the coefficients in Eqs. (5) and (6) are given by

$$V_l=\tfrac{1}{2}\int_{-1}^{1}d\mu\,P_l(\mu)[F_1(1-\mu)+F_2(1+\mu)]\ . \tag{7}$$

For l even and equal to $2n$, this may be rewritten as

$$V_{2n}=\tfrac{1}{2}\int_{-1}^{1}d\mu\,P_{2n}(\mu)[F_1(1-\mu)+F_2(1-\mu)]\ , \tag{8}$$

whereas for odd l

$$V_{2n+1}=\tfrac{1}{2}\int_{-1}^{1}d\mu\,P_{2n+1}(\mu)[F_1(1-\mu)-F_2(1-\mu)] \tag{9}$$

$$=\tfrac{1}{2}\int_{-1}^{1}d\mu P_{2n+1}(\mu)[-F_3(1-\mu)]\ . \tag{10}$$

Equation (10) follows from Eqs. (3) and (9) for triplet $m=0$ states or directly from Eq. (3b) for triplet $m=\pm1$, so the result is invariant under spin rotations.

Equations (8) and (10) are in a suitable form to compare the consequences of near ferromagnetic and near antiferromagnetic instabilities. The main contribution to the integrals comes from $\mu\approx\mu_0$ corresponding to the critical value $\mathbf{q}_0$ of the momentum transfer. The functions $F_j(1-\mu)$ are large and positive at that point. In the (NF) case, $\mu_0\simeq1$ and hence $V_{2n}>0$, $V_{2n+1}<0$ since $P_l=1$. This is the usual result that ferromagnetic fluctuations enhance triplet pairing but suppress singlet pairing. On the other hand, for (NAF) systems, $\mu_0\cong-1$ (backward scattering), $P_{2n+1}(\mu_0)<0$, so both V_{2n} and V_{2n+1} are positive and both kinds of pairing are suppressed.

This analysis does not apply when there is a spatial asymmetry or a strong spin-orbit coupling. However, the conclusion only depends on the parity of the wave function, and parity remains a good symmetry. Thus the general conclusion is that backward scattering from antiferromagnetic spin fluctuations suppresses odd-parity superconductivity. The outcome for an arbitrary modulation vector will depend on the details of the system but there will be a crossover from qualitatively (NF) behavior to qualitatively (NAF) behavior, as $|\mathbf{q}_0|$ increases.

Given this result, it is interesting to ask how a nearly antiferromagnetic itinerant fermion system can avoid the repulsive effects of spin fluctuations and become a superconductor. One possibility has been suggested elsewhere:[8] for finite $\mathbf{q}_0$, the induced interaction between fermions oscillates in real space and may be weak or attractive if the impact parameter is chosen appropriately. In a rotationally invariant system, this is equivalent to choosing the angular momentum so that $P_l(\mu_0)$ has the appropriate sign: in the case of organic superconductors it amounts to pairing electrons on different stacks of organic molecules.[8] The effect is stronger for even-parity states because all three components of the spin-one boson, which constitutes the spin fluctuation, are exchanged. Although this mechanism could be responsible for pairing, it would be sufficient if the pair wave function were able to avoid the repulsion and allow some other attractive interaction to be effective.

In considering the experimental consequences of these remarks, it is useful to keep in mind the effects of spin fluctuations on equilibrium properties[6] For a (NF) system, the dependences on the temperature T and the Stoner enhancement factor $(1-\bar{I})^{-1}$ are quite universal, and in particular the coefficient γ of the linear terms in the specific heat $C(T)$ is proportional to $\ln|(1-\bar{I})|$. On the other hand, the properties of a NAF system vary with nesting and especially on whether $\mathbf{q}_0$ is a single point[5a][γ is proportional to $(1-\sqrt{1-\bar{I}})$ and not strongly enhanced] or lies on a line or surface. There may be a strong enhancement[5c] for some shapes of Fermi surface. It is possible also to imagine a mixed situation in which the susceptibility is large for $\mathbf{q}_0=\mathbf{0}$ and for finite $\mathbf{q}$ or, alternatively, a strong anisotropy in which for example $\partial\chi^0/\partial q_x=0$ for $q_x=0$ but $\partial\chi^0/\partial q_y=0=\partial\chi^0/\partial q_z$ at finite $\mathbf{q}$. Then the properties of the system may display a mixture of NF and NAF characters.[9] In such cases, one of them may dominate the equilibrium properties, while the other would contribute to the main features of the dynamical properties. As far as superconductivity is concerned, either both characters will combine to help triplet pairing, if the modulated tendency corresponds to a forward scattering, or, if it corresponds to a backward scattering, the NF and the NAF tendencies will be in competition, so that triplet superconductivity will be weakened or even suppressed.

We turn now to an examination of the experimental information.

(a) UPt_3 is an anisotropic heavy-fermion system which becomes a superconductor around $T_c = 0.5$ K.[9] Above the superconducting temperature, this system exhibits several characteristic features[1,4] of a NF tendency, in the T dependence of $C(T)$:[10] a strong γ value[11] and a clear cut $T^3 \ln T$ contribution which is not found in any of the known NAF cases.[5] Neutron scattering experiments have been recently performed: low-energy excitations found by Buyers *et al.*[12a] identified with NAF spin fluctuations appeared spurious later on;[12b] "local" spin fluctuations (structureless in $\mathbf{q}$) with no direct evidence for spatial coherence were found by Johnson *et al.*;[12c] antiferromagnetic correlations in the basal planes, below 30 K, were observed by Goldman *et al.*[12d] It can be hoped that a complete set of inelastic scattering data $S(q,\omega)$ versus ω for varying values of the scattering angle will become available in the near future. Then, if our simplified model applies qualitatively, there are the following possibilities.

(i) A low-energy peak is observed in $S(q,\omega)$ versus ω, whose value is maximum for a finite value $\mathbf{q}_0$; then UPt_3 would tend toward a modulated state. According to the present note, triplet superconductivity may arise if $|\mathbf{q}_0|$ is small. If such is the case, it would remain to check whether a $T^3 \ln T$ term may still arise in the computation of $C(T)$ (beyond the linear γT term), when $|\mathbf{q}_0|$ is finite but small or whether it occurs only when $|q_0|$ is strictly equal to zero.

(ii) The peak value of $S(q,\omega)$ is maximum for a vanishing value of $\mathbf{q}$ in agreement with the NF tendency suggested by the $T^3 \ln T$ term of $C(T)$; the observed superconductivity could then be consistent with a triplet pairing.

(iii) The peak in $S(q,\omega)$, plotted versus q, is maximum for two values of $\mathbf{q}$, one equal to zero and one finite; then NF and NAF tendencies would coexist. The NF one may be responsible for the $T^3 \ln T$ in $C(T)$. As far as superconductivity is concerned, the two tendencies would combine or compete depending on the magnitude of the finite $\mathbf{q}_0$.

Of the few heavy-fermion systems which become superconductors, UPt_3 might be the best candidate for triplet superconductivity. Recent upper critical field H_{c2} experiments[13] on three heavy-fermion superconductors showed that, although $CeCu_2Si_2$ and UBe_{13} are "Pauli limited", UPt_3 is not, and it was concluded[13] that either a strong spin-orbit coupling suppresses the Pauli limiting process or UPt_3 is a triplet superconductor. Group-symmetry arguments[14] appear, at present, to disfavor certain kinds of triplet pairings for the heavy-fermion superconductors. However it has been suggested[15] that such arguments might be turned around in the presence of spin anisotropy of the pairing interactions, possibly more likely to arise in hexagonal UPt_3 than, for instance, in cubic UBe_{13}. Thus the question of triplet pairing for UPt_3 is still open. Another point which is of importance is that triplet pairing can easily be destroyed by normal impurities.[16] However, it has been shown[17] that in the weakly localized regime of electron scattering on a small amount of normal impurities, quantum effects weaken the pair breaking parameter due to impurity scattering so that triplet superconductivity is less sensitive to impurities than one could imagine, *a priori*. Furthermore UPt_3 can be obtained in a rather pure state compared to the other heavy-fermion superconductors, having a superconducting coherence length smaller than the average impurity mean free path.[18] Thus UPt_3 could be triplet superconductor of nophononic origin.[19]

(b) Another heavy-fermion compound $CePb_3$ becomes a superconductor when a finite magnetic field H is applied.[20] In zero field this system is an antiferromagnetic heavy fermion[(20)] with a Néel temperature $T_N \sim 1.1$ K. For increasing field, T_N decreases and eventually vanishes. At still higher fields, $CePb_3$ exhibits superconductivity and, in particular, $T_c \sim 0.20$ K for $H = 14T$. This has been attributed[20] to the Jaccarino-Peter mechanism[21] for a magnetic field induced BCS superconductivity. An alternative explanation would be that under the influence of the increasing field:

(i) either the wave vector moves from a large $\mathbf{q}_0$ value to small $\mathbf{q}_0$;

(ii) or there are two peaks, one at $\mathbf{q}_0 = 0$, the other at large q_0; the latter one diminishes while the former one is enhanced.

In both cases the field ultimately suppresses antiferromagnetic spin fluctuations, the system becomes nearly ferromagnetic and then strong paramagnons induce triplet superconductivity in the usual way. Evidently, more data are needed before any firm conclusion can be reached; in particular, it would be interesting to have some measurements of $C(T)$, say for $H = 14T$, and $T > 0.20$ K.

(c) Finally, the very close proximity between antiferromagnetic and superconducting ground states in the series of organic compounds $(TMTSF)_2X$ ($X = PF_6$, AsF_6, $ClO_4, \ldots$) is a well-established phenomenon.[22] On a general phase diagram of these compounds, the crossover between superconductivity and antiferromagnetism is obtained by varying external parameters such as pressure,[22] alloying,[23] cooling rate,[23] or applied magnetic field.[24] Recent NMR studies[25] for many superconductors of this series clearly show that strong antiferromagnetic correlations characterize the metallic regime which is a precursor to organic superconductivity, and they induce clear-cut deviations of the nuclear relaxation rate from the Fermi-liquid prediction. It has been shown[25] that, owing to the very strong anisotropy in these systems, these correlations are mostly one-dimensional in nature over a large range of temperature before the dimensionality crossover ($1D \rightarrow 3D$) is reached [at $T_x \sim 8$ K (Ref. 25)]. Below T_x, the growth of antiferromagnetic correlations saturates and at still lower temperature ($T_c \sim 1$ K) superconductivity takes place. It is therefore tempting to infer that spin fluctuations enhanced by the proximity to an antiferromagnetic instability play a role in the pairing mechanism. However, it is rather easy to show that the application of the simple model used here also leads to the suppression of both types of superconductivity. The usual decomposition of the repulsive Coulomb interaction for quasi-one-dimensional systems retains only the two relevant coupling constants g_1 for backward scattering and g_2 for forward scattering of electrons at the Fermi wave vector. In the Hubbard limit, $g_1 = g_2 = I$. The condition for pairing is that $g_2 - g_1$ (triplet) or $g_2 + g_1$ (singlet) are negative. The corrections due to spin fluctuations may be obtained from the work of Prigodin and Firsov[26] and are given by

$$\tilde{g}_2 \pm \tilde{g}_1 = \pm \frac{1}{2} \left[\frac{2g_1 - g_2}{1 + (2g_1 - g_2)\chi^0(\mathbf{q}_0, \omega)} + \frac{g_2}{1 - g_2 \chi^0(\mathbf{q}_0, \omega)} \right] + \frac{g_2}{1 - g_2 \chi^0(\mathbf{q}_0, \omega)} . \tag{11}$$

This result is simply Eqs. (3) applied to the case of backward scattering and generalized to allow for the possibility of two distinct coupling constants. It reduces to Eqs. (3) when $g_1 = g_2 = I$. As before, in the repulsive sector g_1 and g_2 greater than zero, when the antiferromagnetic Stoner factor $[1 - g_2 \chi^0(\mathbf{q}_0, \omega)]^{-1}$ dominates, both types of superconductivity are suppressed. It can be shown[27] that the inclusion of umklapp processes (g_3), which favor antiferromagnetic correlations and can given appreciable effects in these compounds,[28] does not modify the result. This discussion applies to pairing between carriers on the same organic stack. As mentioned above, antiferromagnetic correlations can favor pairing between carriers on different stacks.[8] According to a recent work,[29] when deviations from perfect three-dimensional nesting are sizable, such a pairing can result from interchain electron-hole pair tunneling (exchange) processes generated before the occurrence of the dimensionality crossover T_x.

It is worth mentioning that there is no experimental evidence[22] so far concerning whether the observed superconductivity in organic compounds is of triplet or singlet, or both, types; and thus many authors[22,28,30] have infered that triplet organic superconductivity cannot be discarded.

To conclude, we have gathered a few different examples to illustrate the main property of the simplified model used in our paper which is that strong spin fluctuations near a magnetic instability may induce or suppress triplet pairing superconductivity depending on the magnitude of the wave-vector characterizing the instability.

Note Added in Proof. T. Matsuura *et al.* [J. Magn. Magn. Mater. **52**, 239 (1985)] found that spin fluctuation exchange does not give an attractive potential for triplet pairing in a dense Kondo system in a simple cubic crystal. J. E. Hirsch [Phys. Rev. Lett. **54**, 1317 (1985)] showed that there is an enhancement of anisotropic singlet pairing in Hubbard and Anderson lattice models in the limit of large on-site electron-electron repulsion. After this paper was accepted for publication, we received preprints from D. J. Scalapino, E. Loh, Jr., and J. E. Hirsch and from K. Miyake, S. Schmitt-Rink, and C. M. Varma showing that exchange of AF spin fluctuations gives an attractive interaction for anisotropic pairing. The mechanism is the same as proposed in Ref. 8 for organic superconductors and it is consistent with the conclusions of the present paper, since the gap vanishes where the exchange of AF spin fluctuations gives rise to backward scattering. We thank Dr. J. E. Hirsch for discussion of this point.

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