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Nuclear ferromagnetism induced Fulde-Ferell-Larkin-Ovchinnikov state

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Abstract

We present a theoretical study of the influence of the nuclear ferromagnetism on superconductivity in the presence of the electron-nuclear spin interaction. It is demonstrated that in some metals, e.g. Rh, W, the BCS condensate imbedded in a matrix of ferromagnetically ordered nuclear spins should manifest the FFLO (Fulde-Ferell-Larkin-Ovchinnikov) state. We outline that the optimal experimental conditions for observation of FFLO could be achieved by creation, via adiabatic nuclear demagnetisation, of the negative nuclear spin temperatures. In this case the nuclear polarisation points in the opposite to the external magnetic field direction and the electromagnetic part of the nuclear spin magnetisation compensates the external magnetic field, while the exchange part creates the non-homogeneous superconducting order parameter.

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The problem of coexistence of the superconducting and magnetic ordering, in spite of its long history [1], is still among the enigmas of the modern condensed matter physics. Most of the theoretical and experimental efforts were devoted to studies of the coexistence of the electron ferromagnetism and superconductivity.

Recently a growing interest to the physics of the superconducting state in the presence of the nuclear magnetism has appeared both theoretically [2 - 6] and experimentally [7 - 12]. In [7] the magnetic critical field $H_c(T)$ of a metallic compound $AuIn_2$ with $T_{ce} = 0.207K$ was studied up to the temperatures lower than the temperature of the nuclear spin ferromagnetic ordering T_{cn}. It was observed that the magnetic critical field $H_{c0} = 14.5$ G has fallen by almost a factor of two at $T < T_{cn} = 35\mu K$. The possibility of such a reduction of $H_c(T)$ by the nuclear ferromagnetism was outlined in [2]. Later on it was theoretically considered in more details [3, 5, 6].

The critical magnetic field in the presence of magnetic moment of nuclear spins is defined in the first approximation by the standard expression [1]

$$H_c\,(T) = H_{co}\,(T) - 4\pi\,(1-n)\,M_n(H_c). \tag{1}$$

Here M_n is the nuclear magnetization and n is the demagnetizing factor, depending on the sample form. It follows from the Eq. (1) that the difference between $H_c\,(T)$ and $H_{co}\,(T)$ is maximal in cylindric samples ($n = 0$) and vanishing in thin plate samples ($n = 1$).

In the very low temperature limit, $T << T_{ce}$ the temperature dependence of the magnetic critical field is quite weak

$$H_{co}\,(T) = H_{co}\left(1 - \frac{T^2}{T_{ce}^2}\right). \tag{2}$$

The difference $H_c - H_{co}$ therefore depends mostly on the initial conditions at the adiabatic nuclear demagnetizing procedure: the applied magnetic field H_i and magnetic nuclear spin moment $M_n\,(H_i)$. In the limit when the initial magnetic field H_i is sufficiently larger than the local nuclear field h, $M_n\,(H_i)$ is defined by the following expression

$$M_n(H_i) = M_{no}B_s(x), \tag{3}$$

where $x = \mu_n H_i/sT$, $B_s(x)$ is the Brillouin function and s is the nuclear spin, M_{no} is the saturation value of the nuclear spin magnetization $M_{no} = \mu_n n_n$, μ_n and n_n are the nuclear magnetic moment and the nuclear spin density, respectively.

The final nuclear magnetization $M_n(H_f)$ in the field H_f could be found from [13]

$$M_n(H_f) = M_n(H_i)\frac{H_f}{\sqrt{h^2 + H_f^2}}. \tag{4}$$

Eqs. (1)-(4) define the influence of the "electromagnetic" part of the nuclear spin ordering on the superconducting critical field H_c.

Recently the shift of $H_c(T)$ as a function of M_n was measured in several metals: Al [8], Sn [9], In [11], and Rh [12]. It was found that the experimental data for Al and In do not fit the Eq. (1), i.e. the shift in the critical field is not linear in M_n when M_n is approaching to its saturated value M_{no}.

In [2, 3] we have suggested that apart from the influence of the "electromagnetic" part of the polarized nuclear spins on the superconducting order, the hyperfine coupling between the nuclear spins and conduction electron spins may play a crucial role on the coexistence between superconducting state and nuclear ferromagnetism. It was shown also [4] that creation of the negative nuclear spin temperatures, (NNST), may result in enhancement of the superconducting ordering. In this work we demonstrate that the hyperfine part of the nuclear-spin-electron interaction may result, in some metals, in appearance of the nonuniform superconducting order parameter, the so called Fulde-Ferell-Larkin-Ovchinnikov state (FFLO) [14, 15]. It follows, that the range of parameters where FFLO can exist is much wider under the conditions of NNST.

The FFLO state was thought originally to take place in superconductors with magnetically ordered magnetic impurities [14, 15]. The main difficulty, however, in the observation of the FFLO in this case is in simultaneous action of the "electromagnetic" and "exchange" parts of the magnetic impurities on the superconducting order. In most of the known superconductors the "electromagnetic" part is destroying the superconducting order before the "exchange" part modify the BCS condensate to a nonuniform FFLO state.

The situation may change drastically in the case of the nuclear spin ferromagnetic ordering. Indeed, the nuclear magnetic moment $\mu_n = \hbar e/M_i c$, is at least three orders of magnitude smaller than the electron Bohr magneton $\mu_e = \hbar e/m_o c$, so that the "electromagnetic" part of the nuclear spin fields is quite low, compared to that of the magnetic impurities. On the other hand the "exchange" part is strongly dependent on the nuclear charge Z. In what follows, we define the conditions and materials where the interplay between these two contributions can be in favor for the "exchange" part, thus providing the necessary conditions for appearance of the FFLO.

As in the case of the magnetic impurities [14, 15], the polarized nuclear spins also remove the spin degeneracy of conduction electrons

$$E_{\pm} = \sqrt{\Delta^2 + (p - p_F)^2 v_F^2} \pm J, \tag{5}$$

where Δ is the gap in the electron spectrum, p_F and v_F the electron Fermi momentum and velocity. The parameter $J \equiv \mu_e H_{hyp}$ defines the Zeeman splitting of the electron spins, due to the hyperfine interaction between the polarized nuclear spins and the conduction electron spins. We note that H_{hyp} is proportional to the magnetization M_n produced by the nuclear spins [3] and $H_{hyp}^{\max} \equiv H_{no}$, the maximal value of H_{hyp}, is achieved when $M_n = M_n^{\max}$ $\equiv M_{no}$.

By introducing the reduced nuclear magnetization $M_n^* \equiv \frac{M_n}{M_{no}}$, we can write $H_n = H_{no} M_n^*$; $J = J_o M_n^*$where $J_o \equiv \mu_e H_{no}$. H_{no} and $J_o$could be found from the expression [16]

$$H_{no} = \frac{\kappa}{\chi} M_{no}, \tag{6}$$

where κ is the Knight shift constant and χ is the conduction electron paramagnetic susceptibility. κ was measured for most of metals and can be as large as 10^{-2} for nuclei with large Z since $\kappa \sim Z$ [16]. At present χ is experimentally defined only for Li and Na to be of order of 10^{-6}.

Using the Fermi liquid theory, the electronic susceptibility is defined by the relation $\chi = \chi_o/(1+Z_o)$, where Z_o is the Fermi liquid constant, $\chi_o = \mu_e^2 \nu$ and ν is the density of electronic states which could be defined from the low-temperature value of the specific heat in a normal metal [17], $C(T) = \frac{\pi^2}{3} \nu T$. In superconductors, however, there exist an alternative definition of ν via the gap Δ and the critical magnetic field H_{co}

$$\nu \Delta^2 = \frac{H_{co}^2(0)}{2\pi}; \ \Delta = 1.76 T_{ce}. \tag{7}$$

Unfortunately, the Fermi liquid constant Z_o in a superconducting state is not known.

We will, therefore, estimate χ_o using the Eqs. (7) and known experimental values for $H_{co}(0)$ and T_{ce}. In Table 1 we present the values of χ_o and nuclear spin field H_{no}, Eq. (6). For most of superconductors the hyperfine nuclear spin field H$_{no}$ is up to four orders of magnitude larger than the magnetic moment of the polarized nuclei M_{no}: $\frac{\kappa}{\chi} \sim 10^4$. As it was discussed previously [4], the nuclear field H_{no} and the parameter Z_o in superconductors could be defined from experimental data on $H_c(T)$ and $H_{co}(T)$ using the expression

$$[H_c(T) + 4\pi(1-n) M_n]^2 = H_{co}^2(T) - 2\frac{J^2}{\Delta^2} H_{co}^2(0). \tag{8}$$

At low enough temperatures $H_c(T) \simeq H_c(T)$ and Eq. (8) can be written in reduced variables in the form

$$[H_c^* + (1-n)\varepsilon M_n^*]^2 = 1 - 2\lambda^2 M_n^{*^2}, \tag{9}$$

where the reduced variables are defined as follows:

$$H_c^* = \frac{H_c}{H_{co}}, M_n^* \equiv \frac{M_n}{M_{no}}, \varepsilon = \frac{4\pi M_{no}}{H_{co}}, \lambda = (1+Z_o)\lambda_o \text{ and } \lambda_o = \varepsilon\frac{\Delta\kappa}{2\mu_e H_{co}}.$$

The values of ε and λ_o for several superconductors are given in the Table 1. The values of M_{no} for all the known superconductors are given in [10].

Elements	$\mathrm{H}_{co}(G)$	$\mathrm{H}_{no}(G)$	$\chi_o 10^6$	ε	λ_o
$AuIn_2$	14.5	5720	1.14	0.7	1.06
Al	105	957	1.85	0.13	0.03
Mo	95	703	2.47	0.04	0.03
Rh	0.049	22	6.2	8.2	2.8
Cd	30	615	0.8	0.06	0.05
Ta	830	902	8.0	0.01	0.008
W	1.2	347	1.48	0.52	0.88
Ir	19	185	4.27	0.04	0.05
Tl	171	3970	1.19	0.02	0.06

Table 1: The values of χ_o and nuclear spin field H_{no}, ε and λ_o for several superconductors.

While analyzing the Eq. (9) one should bear in mind the possibility of nuclear spin system having an either positive or negative temperature, as it was outlined by us in [4].

Consider first the **positive nuclear spin temperature** $T_n > 0$. In this case, the nuclear magnetization is directed along the applied field and Eq. (9) has a single valued solution

$$H_c^* = \sqrt{1 - 2\lambda^2 M_n^{*2}} - (1-n)\varepsilon M_n^*. \tag{10}$$

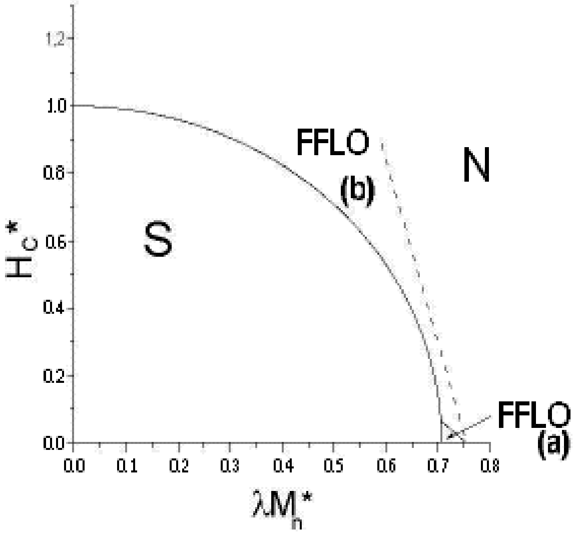

Figure 1: The phase diagram for the Fulde-Ferell-Larkin-Ovchinnikov state for two types of non-homogeneous order parameter. a) an order parameter $\Delta = \Delta_o \exp\{iq \cdot r\}$ and b) $\Delta = \Delta_o \cos\{q \cdot r\}$. Positive nuclear temperatures, $T_n > 0$, and the demagnetising factor $n = 1$.

Two experimental methods of nuclear spin cooling should be considered: the adiabatic demagnetization and the dynamic polarization [16, 18].

a) Nuclear polarization is created by the adiabatic demagnetization. In this case

$$M_n^* = \frac{H_c^*}{\sqrt{h^{*2} + H_c^{*2}}} B_S \left(\frac{\mu_n H_i}{sT} \right); h^* = \frac{h}{H_{co}}. \tag{11}$$

It is easy to see that the solution $H_c^* = 0$ do not satisfy the Eqs. (10) and (11) simultaneously. This means that the nuclear spins polarized by adiabatic demagnetization, while reducing H_c, do not completely destroy the superconductivity.

b) Nuclear polarization is created by the methods of dynamic polarization. In this case the Eq. (11) is not valid and the $H_c^* = 0$ solution can be obtained at suitable choice of parameters. Indeed, even in the case of a thin plate, $n = 1$, the BCS superconducting state vanishes at $\lambda M_n^* > 1/\sqrt{2} = 0.707$. Since $M_n^* < 1$, a first order phase transition from the superconducting phase to a normal one will occur in superconductors with $\lambda > 1/\sqrt{2}$.

In [14, 15] it was conjectured, however, that even at $\lambda M_n^* > 1/\sqrt{2}$ a superconducting state with a non-homogeneous order parameter may appear, the long searched FFLO state. In [14, 15] it was found that the difference in the free energies of a normal and non-homogeneous superconducting states, at $J \lesssim J_c$, is: $f_s - f_n = -C_o^2 \nu \left(J_c - J\right)^2$; $J_c \simeq 0.755$. Using the general thermodynamic arguments [17] the critical field $\overline{H}_c^*$ can be defined from an equation

$$4C_o^2 \lambda^2 \left(M_n^* - M_{nc}^*\right)^2 = \left[\overline{H}_c^* + (1-n)\, \varepsilon M_n^*\right]^2 . \qquad (12)$$

Here $\overline{H}_c^* \equiv \frac{\overline{H}_c}{H_{co}}$, where $\overline{H}_c$ is the critical field of the FFLO phase and M_{nc}^* is the critical value of the nuclear magnetization: $\lambda M_n^* = 0.754$.

In [15] different forms of the non-homogeneous order parameter were considered. It was found there that the FFLO state with an order parameter $\Delta = \Delta_o \exp\{i \overrightarrow{q} \cdot \overrightarrow{r}\}$ can exist in a narrow interval $0.707 > \lambda M_n^* > 0.755$ with a constant $C_o^2 = 0.44$, see Fig. 1, the solid line. It is interesting to note that for an order parameter $\Delta = \Delta_o \cos\{i \overrightarrow{q} \cdot \overrightarrow{r}\}$ the constant $C_o^2 = 7.35$ and the FFLO state may exist in a much wider interval of values of λM_n^*, see Fig. 1, the dashed line. One should bear in mind, however, that this wide interval is limited by the made above assumption: $J \lesssim J_c$.

At positive nuclear temperatures, $T_n > 0$, the Eq. (12) has one solution

$$\overline{H}_c^* = 2C_o \lambda \left(M_n^* - M_{nc}^*\right) - (1-n)\, \varepsilon M_n^* . \qquad (13)$$

Eq. (10) and Eq. (13) define the H_c^*,$\overline{H}_c^*$ vs M_n^* phase diagram for two superconducting and one normal metal phases, see Fig. 1.

Among the possible candidates for the observation of nuclear spin polarization induced FFLO state are Rh and W. Rh was studied in details in [12]. It has rather high $\varepsilon = 8.2$ and $\lambda_o = 2.8$. The transition to a superconducting phase with a nonuniform order parameter is possible when the reduced magnetization $M_n^* > 0.25$. The local field h is however also rather high: $h^* = \frac{h}{H_{co}} = 6.9$, [12]. This makes it difficult to get sufficiently high values of M_n^* using the adiabatic demagnetization method.

It follows from the Eq. (10) and Eq. (11) that in superconductors with $h^* >> 1$

$$H_c^* \simeq \left[\left(1 + \frac{\varepsilon}{h^*}\,(1-n)\, B_S\right)^2 + 2\lambda^2 \frac{B_S}{h^{*2}}\right]^{-\frac{1}{2}} . \qquad (14)$$

For W, with $\varepsilon = 0.52$ and $\lambda_o = 0.88$, the nuclear spin relaxation time is very long (the Korringa's const $\kappa = 39Ks$). This makes it feasible, in W, to get nuclear temperatures $T >> T_n$ by adiabatic demagnetization.

At the **negative nuclear spin temperatures**, $T_n < 0$, the nuclear magnetization M_n points in the direction opposite to the applied field H [4], and the direction of M_n can be reversed by a well known method of fast reversal of the external field [19]. In [4] we have shown that the NNST may strongly enhance the superconducting ordering. For the metals like Be, $TiH_{2.07}$, for example, the critical field H_c is order of magnitude higher than H_{co} at saturation of M_n.

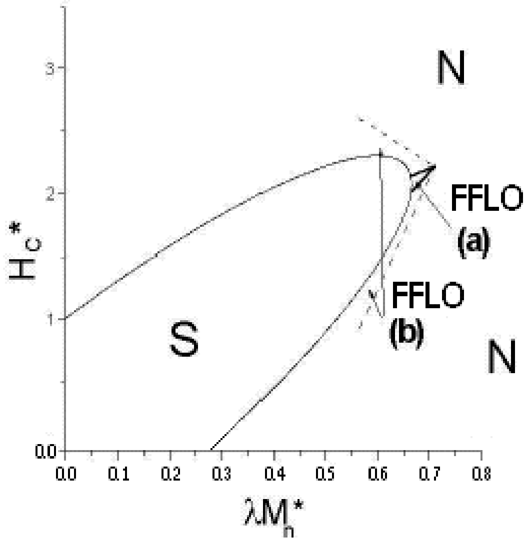

Figure 2: The phase diagram for the Fulde-Ferell-Larkin-Ovchinnikov state in Rh with $\varepsilon = 8.2$, $\lambda = 2.8$ and the demagnetising factor $n = 0$, at negative nuclear temperatures, $T_n < 0$, for two types of nonehomogeneous order prameter: a) $\Delta = \Delta_o \exp\{iq \cdot r\}$ and b) $\Delta = \Delta_o \cos\{q \cdot r\}$.

Let us show now that the NNST stimulate the appearance of the FFLO phase. As it was shown earlier in this paper, at $T_n > 0$, the Eq. (9) and Eq. (12) do not posses a solution with low H_c^*, where the FFLO state should appear, see Fig. 1. At $T_n < 0$, however, the Eq. (9) and Eq. (12) have two solutions for H_c^* and $\overline{H}_c^*$

$$H_c^* = (1-n)\,\varepsilon M_n^* \pm \sqrt{1 - 2\lambda^2 M_n^{*2}}, \tag{15}$$

$$\overline{H}_c^* = (1-n)\,\varepsilon M_n^* \pm 2C_o\lambda \left| M_n^* - M_{nc}^* \right|. \tag{16}$$

The phase diagram, in this case, is presented in Fig. 2. In this limit the FFLO phase can be realized in the high field region which makes its observation much easier than in the case of the positive nuclear temperatures, Fig. 1. We outline here that the application of a rather strong magnetic field at $T_n < 0$ will not destroy, as is the case at $T_n > 0$, but rather stabilize the FFLO state.

Let us discuss now the experimental feasibility of our model. The most suitable experimental conditions for observation of the FFLO state would be the case when the nuclear spin ordering appears first and than by lowering the external magnetic field the superconducting state is established [3]. In this limit the nuclear spin ordering is not modified substantially by superconductivity, since the nuclear spin relaxation times, at micro-Kelvin temperatures, are extremely long. This can be achieved by adiabatic nuclear demagnetization at nuclear temperatures T_n much higher than T_{cn}, the temperature of spontaneous nuclear ordering. In this case the sample is in a monodomain state. The superconducting ordering starts at the electron temperature T in the interval $T_n << T << T_{ce}$ and critical magnetic field $H_c(T)$,which is different from $H_{co}(T)$, the critical magnetic field in the absence of the nuclear spin ordering.

It was shown above that the FFLO state could be easier obtained under the conditions of negative nuclear spin temperature, since the electromagnetic part of the nuclear spin field reduces the influence of the external field and the exchange part would act to create the non-homogeneous superconducting ordering. Experimentally the NNST was achieved in single crystal Rh [12] employing the nuclear demagnetizing techniques. However, no study of possible superconducting order was performed. It follows, from the considerations presented above, that the FFLO state may appear under similar to [12] conditions at critical fields $H_c > H_{co} \simeq 0.05G$

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