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Superconductivity in Spin-Glasses

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It is shown that spin glass ordering does not affect the superconductivity as a result of total compensation of the paramagnetic effect and the effect of spin-flip scattering freezing out in a spin-glass phase.

Показано, что упорядочение спинов при переходе в состояние спинового стекла не оказывает влияния на сверхпроводимость, что является следствием взаимной компенсации парамагнитного эффекта и эффекта вымораживания процессов рассеяния с переворотом спина.

1. Introduction

Recently there has been a considerable growth of the literature on the coexistence of superconductivity and magnetic ordering [1, 2], due to the experimental discovery of such phenomena in some rare-earth compounds with regular positions of magnetic atoms [3 to 5]. Likewise it has been known for a long time that there is some experimental-evidence of such a coexistence in dilute alloys of transition metals in a superconducting matrix [1]. In such systems the type of magnetic ordering is unknown in most cases. In the theory of dilute alloys of magnetic impurities the concept of the spin-glass phase is preferred now due to the long-range and oscillating behaviour of the indirect exchange interaction via the conduction electrons [6, 7]. There is good experimental evidence for the coexistence of superonductivity and spin-glass ordering in $Gd_xTh_{1-x}Ru_2$ [8] and $Gd_xCe_{1-x}Ru_2$ [8], as well as some evidence for it in the amorphous alloy of $La_{80}Au_{20}$ with Gd impurities [9].

The influence of magnetic impurities upon superconductivity was first considered by Abrikosov and Gorkov [10]. Gorkov and Rusinov have considered a possibility of coexistence of superconductivity and ferromagnetism in such a system [11]. In the present paper we will attempt to analyze the influence of spin-glass ordering upon superconductivity.

2. General Formalism

To describe superconductivity in a system with some kind of magnetic ordering it is convenient to use a four-dimensional matrix formalism, defining the electron operators in spinor form [1, 2]:

$$\hat{\Psi}(\boldsymbol{r}) = \begin{pmatrix} \psi_\uparrow(\boldsymbol{r}) \\ \psi_\downarrow(\boldsymbol{r}) \\ \psi_\uparrow^+(\boldsymbol{r}) \\ \psi_\downarrow^+(\boldsymbol{r}) \end{pmatrix}; \qquad \hat{\Psi}^+(\boldsymbol{r}) = \left((\psi_\uparrow^+(\boldsymbol{r})\ \psi_\downarrow^+(\boldsymbol{r})\ \psi_\uparrow(\boldsymbol{r})\ \psi_\downarrow(\boldsymbol{r})\right), \tag{1}$$

where $\psi_\uparrow(\boldsymbol{r})$ is the ordinary electron destruction operator with spin directed upwards and so on.

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The zero-order Hamiltonian for a superconducting system takes the form

$$\mathcal{H}_0 = \int \mathrm{d}\boldsymbol{r}\, \hat{\Psi}^+(\boldsymbol{r})\, \hat{h}_0(\boldsymbol{r})\, \hat{\Psi}(\boldsymbol{r}) , \tag{2}$$

where

$$\hat{h}_0(\boldsymbol{r}) = \begin{pmatrix} \hat{H}_0(\boldsymbol{r}) & \hat{\Delta}(\boldsymbol{r}) \\ \hat{\Delta}^+(\boldsymbol{r}) & -\hat{H}_0^{\mathrm{tr}}(\boldsymbol{r}) \end{pmatrix} = H_0(\boldsymbol{r})\, \sigma_0\tau_3 + \Delta_1\sigma_2\tau_2 + \Delta_2\sigma_2\tau_1 . \tag{3}$$

$H_0(\boldsymbol{r})$ is the free-electron Hamiltonian, σ_i and τ_i are two independent sets of Pauli matrices, direct product of which can be used to represent any 4×4 matrix, $\Delta_1 = \mathrm{Re}\, \Delta$, $\Delta_2 = \mathrm{Im}\, \Delta$, where Δ is the gap function of superconductivity theory.

The electron interaction with magnetic atoms can be described by the ordinary s–d exchange model and the interaction Hamiltonian in the four-dimensional matrix formalism takes the form [1]

$$\mathcal{H}_{\mathrm{int}} = \tfrac{1}{2} \int \mathrm{d}\boldsymbol{r}\, \hat{\Psi}^+(\boldsymbol{r})\, V(\boldsymbol{r})\, \hat{\Psi}(\boldsymbol{r}) , \tag{4}$$

where

$$\hat{V}(\boldsymbol{r}) = \sum_i J(\boldsymbol{r} - \boldsymbol{R}_i)\, \boldsymbol{\alpha} \cdot \boldsymbol{S}_i , \tag{5}$$

$$\alpha_\mu = \begin{pmatrix} \sigma_\mu & 0 \\ 0 & -\sigma_\mu^{\mathrm{tr}} \end{pmatrix}. \tag{6}$$

$\frac{1}{2}\boldsymbol{\sigma}$ is the electron spin operator, $J(\boldsymbol{r} - \boldsymbol{R}_i)$ is the s–d exchange integral, $\boldsymbol{S}_i$ is the spin of the magnetic atom at the site $\boldsymbol{R}_i$.

To consider superconductivity with any kind of magnetic ordering it is useful to isolate the mean-field effects. The Hamiltonian of electron interaction with a mean magnetic field, following from (5) is

$$\mathcal{H}_{\mathrm{int}}^{\mathrm{MF}} = \tfrac{1}{2} \int \mathrm{d}\boldsymbol{r}\, \hat{\Psi}^+(\boldsymbol{r})\, \boldsymbol{H}(\boldsymbol{r})\, \boldsymbol{\alpha}\hat{\Psi}(\boldsymbol{r}) , \tag{7}$$

where

$$\boldsymbol{H}(\boldsymbol{r})\, \boldsymbol{\alpha} = \begin{pmatrix} \boldsymbol{H}(\boldsymbol{r})\, \boldsymbol{\sigma} & 0 \\ 0 & -\boldsymbol{H}(\boldsymbol{r})\, \boldsymbol{\sigma}^{\mathrm{tr}} \end{pmatrix} = H^\mu\alpha_\mu , \tag{8}$$

$$\boldsymbol{H}(\boldsymbol{r}) = \sum_i J(\boldsymbol{r} - \boldsymbol{R}_i)\, \langle \boldsymbol{S}_i \rangle \tag{9}$$

is the mean magnetic field at the point $\boldsymbol{r}$, $\langle \boldsymbol{S}_i \rangle$ the thermodynamic average of the impurity spin. The mean field $\boldsymbol{H}(\boldsymbol{r})$ leads to the paramagnetic effect suppressing superconductivity.

We must also consider a perturbation (fluctuations) over the mean-field:

$$\tilde{\mathcal{H}}_{\mathrm{int}} = \mathcal{H}_{\mathrm{int}} - \hat{\mathcal{H}}_{\mathrm{int}}^{\mathrm{MF}} = \int \mathrm{d}\boldsymbol{r}\, \Psi^+(\boldsymbol{r}) \sum_i J(\boldsymbol{r} - \boldsymbol{R}_i)\, \boldsymbol{\alpha}(\boldsymbol{S}_i - \langle \boldsymbol{S}_i \rangle)\, \hat{\Psi}(\boldsymbol{r}) . \tag{10}$$

The perturbation theory over $\mathcal{H}_{\mathrm{int}}$ produces the Green's function

$$D_{ij}^{\mu\nu}(\tau, \tau') = - \langle T_\tau (S_i^\mu(\tau) - \langle S_i^\mu \rangle)\, (S_j^\nu(\tau') - \langle S_j^\nu \rangle) \rangle , \tag{11}$$

where τ is the Matsubara "time".

3. Spin-Glass Ordering and Superconductivity

At present there is no complete spin-glass theory even in the mean field approximation. The most popular Edwards-Anderson model of spin-glass behaviour [12, 13] is based on the so-called replica method and the limit of replica number $n \to 0$ and faces some basic difficulties (such as negative entropy) [7]. Some other models were proposed not using the replica method [14 to 16]. All of these models try to describe the spin-glass phase via the order-parameter $q = \langle \langle \boldsymbol{S}_i \rangle^2 \rangle_{\mathrm{c}}$ [12], where $\langle ... \rangle_{\mathrm{c}}$ denotes the configurational averaging, and lead to a practically equivalent behaviour of physical quantities, though not in complete agreement with the experiment [6]. There is even some doubt in the existence of the spin-glass transition itself [17].

Our aim is to consider the influence of the Edwards-Anderson order-parameter upon superconductivity. The main results will be in fact independent of any specific model of spin-glass in the mean-field approximation. Thus we consider the simplest model of [14], which leads to the same main results as the Edwards-Anderson model, but is free from the unphysical artefacts of the replica method.

In the Medvedev-Zaborov model and analogous models of [15, 16] it is supposed that the chaotic orientations of impurity spins lead to a random magnetic mean field at every site $\boldsymbol{h}_i = \boldsymbol{h}(\boldsymbol{R}_i)$. The distribution function of this field can be shown to be Gaussian [14]:

$$P(\boldsymbol{h}_i) = (\tfrac{2}{3}\,\pi A q)^{-3/2} \exp\left(-\frac{|\boldsymbol{h}_i|^2}{\frac{2}{3}\,Aq}\right), \tag{12}$$

where q is the Edwards-Anderson order-parameter defined by

$$q = \int_0^\infty \mathrm{d}h\; P(h)\, b_S^2\left(\frac{h}{T}\right), \tag{13}$$

where

$$P(h) = 4\pi h^2\, (\tfrac{2}{3}\,\pi q A)^{-3/2} \exp\left(-\frac{h^2}{\frac{2}{3}\,qA}\right) \tag{14}$$

is the distribution function for the absolute value of the mean field, $b_S(x)$ is the Brillouin function,

$$A = \frac{c}{v_0} \int_{v_0} \mathrm{d}\boldsymbol{R}\; I^2(\boldsymbol{R}) \equiv c I^2\,, \tag{15}$$

where $I(\boldsymbol{R})$ is the indirect exchange integral (for example of the RKKY type), c the concentration of magnetic atoms, v_0 the volume per one such an atom, T the absolute temperature. The integration in (15) goes over the whole volume of the system except the volume v_0 around the origin.

The solution of (13) for $q(T)$ leads to dependences similar to that of the Edwards-Anderson theory, $q(T) \neq 0$ for $T < T_\mathrm{f}$, where T_f is the spin-glass "freezing" temperature:

$$T_\mathrm{f} = \tfrac{1}{3}\, S(S+1)\, A^{1/2} = \tfrac{1}{3}\, S(S+1)\, c^{1/2}\, I^{1/2}\,, \tag{16}$$

where S is the magnitude of the impurity spin.

The distribution of molecular fields is factorized over the sites:

$$P\{\boldsymbol{h}_i\} = \prod_i P(\boldsymbol{h}_i) \tag{17}$$

and there is no short-range magnetic order:

$$\langle \boldsymbol{h}_i \boldsymbol{h}_j \rangle_\mathrm{c} = qA\delta_{ij}\,. \tag{18}$$

Following the methods of [14] it is easy to show that the mean magnetic field $\boldsymbol{H}(\boldsymbol{r})$ acting upon a conduction electron is also Gaussian:

$$\mathcal{P}(\boldsymbol{H}(\boldsymbol{r})) = (\tfrac{2}{3}\,\pi q \mathcal{A})^{-3/2} \exp\left\{-\frac{|\boldsymbol{H}(\boldsymbol{r})|^2}{\frac{2}{3}\,q\mathcal{A}}\right\}, \tag{19}$$

$$\mathcal{P}\{\boldsymbol{H}(\boldsymbol{r})\} = \prod_{\boldsymbol{r}} \mathcal{P}(\boldsymbol{H}(\boldsymbol{r}))\,, \tag{20}$$

$$\langle \boldsymbol{H}(\boldsymbol{r})\, \boldsymbol{H}(\boldsymbol{r}') \rangle_\mathrm{c} = q\mathcal{A}\delta(\boldsymbol{r} - \boldsymbol{r}')\,, \tag{21}$$

where

$$\mathcal{A} = \frac{c}{v_0} \int \mathrm{d}\boldsymbol{R}\; J^2(\boldsymbol{R}) \equiv c J^2 \tag{22}$$

and $q(T)$ is defined by (13).

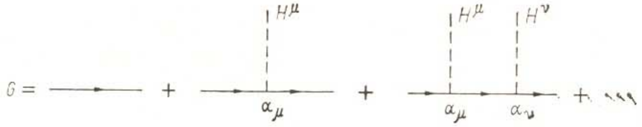

Fig. 1

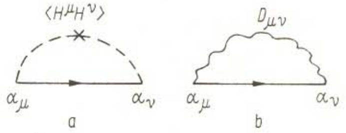

Fig. 2

Now we have to consider the superconductivity of electrons under the influence of the random magnetic field $\boldsymbol{H}(\boldsymbol{r})$ distributed according to (19) to (21). The interaction given by (7) can be analyzed by perturbation theory, which leads to the summation of graphs for the electrons Green's function shown in Figure 1.

Here the continuous line represents the matrix Green's function defined by the equation of motion

$$\left\{-\frac{\partial}{\partial\tau}\sigma_0\tau_0-\hat{h}_0(\boldsymbol{r})\right\}g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')=\delta(\boldsymbol{r}-\boldsymbol{r}')\,\delta(\tau-\tau')\,. \tag{23}$$

The dashed line describes the interaction with the random field $\boldsymbol{H}(\boldsymbol{r})$. Averaging over (19), (20) we obtain that the second graph in Fig. 1 is equal to zero, while the third one gives the ordinary electron self-energy in the random field (see Fig. 2a). It is equal to

$$\begin{aligned}\Sigma_{\mathrm{MF}}(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau')&=\int\{\delta\boldsymbol{H}(\boldsymbol{r})\}\,\mathcal{P}\{H(\boldsymbol{r})\}\,H^\mu(\boldsymbol{r})\,H^\nu(\boldsymbol{r}')\,\alpha_\mu\,g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu=\\&=\langle\sum_{ij}J(\boldsymbol{r}-\boldsymbol{R}_i)\,J(\boldsymbol{r}'-\boldsymbol{R}_j)\,\langle S_i^\mu\rangle\,\langle S_j^\nu\rangle\rangle_{\mathrm{c}}\,\alpha_\mu g_0(\boldsymbol{r}\tau\,;\,\boldsymbol{r}'\tau')\,\alpha_\nu\end{aligned} \tag{24}$$

or, using (21),

$$\Sigma_{\mathrm{MF}}(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau')=\tfrac{1}{3}\,\mathcal{A}q\,\delta(\boldsymbol{r}-\boldsymbol{r}')\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\mu \tag{25}$$

or, in the momentum representation,

$$\Sigma_{\mathrm{MF}}(\boldsymbol{p}\varepsilon_n)=\mathcal{A}qN_0\,\tfrac{1}{3}\int\mathrm{d}\xi_{\boldsymbol{p}}\,\alpha_\mu g_0(\boldsymbol{p}\varepsilon_n)\,\alpha_\mu\,, \tag{26}$$

where N_0 is the free-electron density of states at the Fermi level. Equation (26) coincides with the appropriate expression of the Abrikosov-Gorkov theory [1, 10] with the substitution of the ordinary spin-flip scattering rate by $\Gamma_{\mathrm{sf}}'=2\pi\mathcal{A}q(T)\,N_0=2\pi cJ^2q(T)\,N_0$. Thus the paramagnetic effect (random molecular field) in spin-glasses influences the superconductivity in the same way as magnetic impurities in the Abrikosov-Gorkov theory.

Consider now the rest of the interaction given by the Hamiltonian (10). The simplest self-energy corresponding to this interaction is shown in Fig. 2b:

$$\tilde{\Sigma}(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')=-\sum_{ij}J(\boldsymbol{r}-\boldsymbol{R}_i)\,J(\boldsymbol{r}'-\boldsymbol{R}_j)\,D_{ij}^{\mu\nu}(\tau,\tau')\,\alpha_\mu\,g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu\,. \tag{27}$$

We use now the static approximation for $D_{ij}^{\mu\nu}(\tau,\tau')$,

$$D_{ij}^{\mu\nu}(\tau,\tau')\to-\langle S_i^\mu S_j^\nu\rangle+\langle S_i^\mu\rangle\,\langle S_j^\nu\rangle\,. \tag{28}$$

Then

$$\begin{aligned}\Sigma(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')&=\sum_{ij}J(\boldsymbol{r}-\boldsymbol{R}_i)\,J(\boldsymbol{r}'-\boldsymbol{R}_j)\,\langle S_i^\mu S_j^\nu\rangle\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu-\\&-H^\mu(\boldsymbol{r})\,H^\nu(\boldsymbol{r}')\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu\,.\end{aligned} \tag{29}$$

After the configurational averaging we get

$$\begin{aligned}\tilde{\Sigma}(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau')&=\langle\sum_{ij}J(\boldsymbol{r}-\boldsymbol{R}_i)\,J(\boldsymbol{r}'-\boldsymbol{R}_j)\,\langle S_i^\mu S_j^\nu\rangle\rangle_{\mathrm{c}}\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu-\\&-\tfrac{1}{3}\,\mathcal{A}q\,\delta(\boldsymbol{r}-\boldsymbol{r}')\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu\,.\end{aligned} \tag{30}$$

In the following we use the standard assumption of the spin-glass theory [7, 13], corresponding to the absence of short-range magnetic order:

$$\langle\langle S_i^\mu S_j^\nu \rangle\rangle_c \approx \delta_{\mu\nu}\delta_{ij}\,\tfrac{1}{3}\,S(S+1)\,. \tag{31}$$

Then the total electron self-energy is equal to

$$\begin{aligned}\Sigma(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau') &= \Sigma_{\mathrm{MF}}(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau') + \tilde{\Sigma}(\boldsymbol{r}-\boldsymbol{r}',\tau-\tau') \approx \\ &\approx \langle \sum_i J(\boldsymbol{r}-\boldsymbol{R}_i)\,J(\boldsymbol{r}'-\boldsymbol{R}_i)\rangle_c\,\tfrac{1}{3}\,S(S+1)\,\alpha_\mu\,g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\nu \approx \\ &\approx \tfrac{1}{3}\,cJ^2S(S+1)\,\alpha_\mu g_0(\boldsymbol{r}\tau,\boldsymbol{r}'\tau')\,\alpha_\mu\delta(\boldsymbol{r}-\boldsymbol{r}')\,,\end{aligned} \tag{32}$$

where the last equality is valid for the point-like s–d exchange. In the momentum representation

$$\Sigma(\boldsymbol{p}\varepsilon_n) = \frac{\Gamma_{\mathrm{sf}}}{2\pi}\,\frac{1}{3}\int \mathrm{d}\xi_{\boldsymbol{p}}\,\alpha_\mu g_0(\boldsymbol{p}\varepsilon_n)\,\alpha_\mu\,, \tag{33}$$

where

$$\Gamma_{\mathrm{sf}} = 2\pi cJ^2S(S+1)\,N_0 \tag{34}$$

is the standard electron spin-flip scattering rate (in Born approximation) coincides with the well-known result of the Abrikosov-Gorkov theory. In the sum of (25) and (30) the contributions dependent on the Edwards-Anderson order-parameter have cancelled each other completely. The physical meaning of such a cancellation is absolutely clear. We have seen that the paramagnetic effect in spin-glasses is equivalent to the spin-flip scattering rate $\Gamma'_{\mathrm{sf}} = 2\pi cJ^2q(T)\,N_0$. At the same time the "freezing" of spins during the spin-glass transition "freezes" out the ordinary mechanism of spin-flip scattering in such a way that the corresponding scattering rate becomes equal to $\Gamma''_{\mathrm{sf}} = \Gamma_{\mathrm{sf}} - 2\pi cJ^2q(T)\,N_0 \approx S(S+1) - \langle\langle S\rangle^2\rangle_c$. Both effects just compensate each other $\Gamma_{\mathrm{sf}} = \Gamma'_{\mathrm{sf}} + \Gamma''_{\mathrm{sf}}$. Superconductivity in the system of magnetic impurities is determined by the dependences of the Abrikosov-Gorkov theory despite the spin-glass ordering.

4. Discussion

The cancellation of the Edwards-Anderson order parameter demonstrated for the simplest graphs of Fig. 2 persists for all diagrams in higher orders of perturbation theory. This is quite obvious for diagrams without crossing interaction lines and also can be demonstrated directly for diagrams with crossing lines. This cancellation follows from the fact that the configurational average of the random molecular field is equal to zero and the Abrikosov-Gorkov behaviour is due to equation (31) holding both in paramagnetic and spin-glass phases. Note that we neglect the quantum nature of impurity spins which allows us to use the standard diagram technique.

Spin dynamics can be neglected [1] if the characteristic frequencies of spin motion in the spin-glass phase $\Omega_{\mathrm{SG}} \ll T_c \sim \Delta_0$ where T_c is the temperature of superconducting transition, and Δ_0 the superconductivity gap for $T = 0$. Ω_{SG} can be a characteristic frequency of a spin wave or the typical inverse time of change of the Edwards-Anderson order parameter when on the average it is equal to zero due to the slow relaxation processes [17]. Spin-glass dynamics can lead to a change in superconducting behavior in comparison with the Abrikosov-Gorkov theory. For example it is well known, that electron–electron interaction due to the exchange of spin-waves is repulsive, thus lowering the superconducting T_c.

Under the specific conditions [14] the system considered can undergo a transition not to a spin-glass phase but to that of a random ferromagnet (with a non-zero spontaneous magnetic moment). This leads to a change of the distribution function of the random molecular fields, particularly the average of the second graph in Fig. 1 as well as all graphs of odd power in the random field become non-zero. Then there is no compensation of the paramagnetic effect and spin-flip scattering freezing out, as in the case of ordinary ferromagnets [1, 2]. It is possible that such a situation was realized in the experiments with $Gd_xLa_{1-x}Ru_2$ [18], where two superconducting transition temperatures (re-entrant superconductivity) have been found for some concentrations of Gd.

Finally, note that we have neglected the influence of the superconducting transition upon a spin-glass transition. The appropriate analysis seems difficult due to the present status of spin-glass theory. The oscillating behaviour of the indirect exchange interaction via the conduction electrons remains in the superconducting phase and in fact this interaction is almost the same as in normal metals up to distances of the order of the superconducting coherence length [19]. This interaction is effectively cut off at distances of the order of the electron mean-free path, thus in the case of mean-free paths shorter than the superconducting coherence length the effective interaction of impurity spins is unchanged in a superconducting phase. In general, the interaction parameter (15) determining the spin-glass transition is apparently almost the same as in the case of a normal metal.

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