The International Journal Of Engineering And Science (IJES)
||Volume||2 ||Issue|| 8 ||Pages|| 17-23 ||2013||
ISSN(e): 2319 – 1813 ISSN(p): 2319 – 1805

Coexistence of Superconductivity and Ferromagnetism in a Magnetic Superconductor

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ABSTRACT
A model is presented utilizing a Hamiltonian with equal spin singlet and triplet pairings based on quantum field theory and green function formalism, to show the correlation between the superconducting and ferromagnetic order parameters. The model exhibits a distinct possibility of the coexistence of superconductivity and ferromagnetism, which are two usually incompatible cooperative phenomena. The work is motivated by the recent experimental evidences of a long-range magnetic order below the superconducting phase temperature in a number of ternary or pseudo-ternary compounds of the lanthanides. The theoretical results are then applied to show the coexistence of superconductivity and ferromagnetism in the lanthanide compound $HoMo_6S_8$.

KEYWORDS: *Spin singlet and triplet state,Superconducting and ferromagnetic order parameters, Green function.*

Date of Submission: 12 Aug. 2013, Date of Acceptance: 30 Aug 2013,

I. INTRODUCTION

The study of magnetic superconductors has gained tremendous importance since the discovery of the ternary compounds of the lanthanides, in which a long-range magnetic order is observed below the superconducting phase temperature [1, 2]. In fact, $ErRh_4B_4$ was the first ferromagnetic superconductor in which superconductivity was found to exist in a small temperature interval with modulated ferromagnetic phase as had been observed in a detailed study by Sinha *et al*. [3]. Later ferromagnetic superconductivity was experimentally confirmed in other compounds such as URhGe, UCoGe and $ZrZn_2$ [2, 4, 5].

The first observation of a zero resistance in the ferromagnetic state of $HoMo_6S_8$ was reported by Lynn *et al*.[6] and then by Genicon *et al*.[7]. Subsequently, neutron diffraction experiments carried out on singlet crystals of $HoMo_6S_8$ by Rossat *et al*.[8], further confirmed the above observations. They found that for T<0.60k, with slow cooling, a ferromagnetic phase appears and then the magnetic intensity increases and afterwards saturates at above T=0.4k, indicating that below this temperature, the thermal fluctuations are negligible. The material has a superconducting transition at T=1.82k and a magnetic transition at T=0.67k in the vicinity of the reentry to the normal conducting state (Lynn *et al*.)[6].

The above exciting discovery stimulated a lot of interest in the study of coexistence of superconductivity and ferromagnetism. The two cooperative phenomena have been considered until very recently to be incompatible in view of the fact that conventional superconductivity favors spin singlet

The problem of understanding the true nature and mechanism of superconductivity coexisting with ferromagnetism is so complex that a complete solution is still lacking [13, 14]. There has been suggestion that the coupling of longitudinal and transverse spin fluctuations plays an important role and should be included in the study of the problem [15]. Abrikosov [16] and Mineev *et al*. [17] have pointed out that S-wave superconductivity may result from the electron interaction mediated by ferromagnetically aligned localized moments. In the present paper, we start with a model Hamiltonian which incorporates not only terms of the BCS type but also an additional new term representing interaction between conduction electrons and localized electrons. The results of the calculations clearly show that over a certain temperature range superconductivity and ferromagnetism can co-exist.

II. MODEL HAMILTONIAN OF THE SYSTEM

The system under consideration consists of conduction electrons and localized electrons, between which exchange interaction exists. Within the framework of the BCS model, the Hamiltonian of the system can be written as:

$$H=H_1+H_2+H_3 \tag{1}$$

where,

$$H_1 = \sum_{\kappa,\sigma} \epsilon_\kappa \, \hat{a}^{\dagger}_{\kappa,\sigma}\hat{a}_{\kappa,\sigma} + \sum_{l,\sigma} \epsilon_l \, \hat{b}^{\dagger}_{l,\sigma}\hat{b}_{l,\sigma} \tag{2}$$

represents, the single particle energies of the conduction and the localized electrons, measured relative to the chemical potential. $\hat{a}^{\dagger}_{\kappa,\sigma}(\hat{a}_{\kappa,\sigma})$ and $\hat{b}^{\dagger}_{l,\sigma}$ $(\hat{b}_{l,\sigma})$ are the creation (annihilation) operators for conduction and localized electrons respectively.

$$H_2 = -\sum_{\kappa,\kappa'} V_{\kappa\kappa'} \, \hat{a}^{\dagger}_{\kappa\uparrow}\hat{a}^{\dagger}_{-\kappa\downarrow}\hat{a}_{-\kappa'\downarrow}\hat{a}_{\kappa'\uparrow} \tag{3}$$

is the BCS type electron-electron pairing interaction term due to exchange of phonons. And,

$$H_3 = \sum_{l,m,\kappa} u^{k}{}_{l,m} \hat{a}^{\dagger}_{\kappa\uparrow}\hat{a}^{\dagger}_{-\kappa\downarrow}\hat{b}_{l\uparrow}\hat{b}_{m\uparrow} + h.c \tag{4}$$

(h.c.= hermitian conjugate)

is the new term describing the interaction between conduction and localized electrons with the coupling constant u. the Hamiltonian in (1) will be used to determine the equations of motion in terms of the Green function.

III. COUPLING OF SUPERCONDUCTING AND FERROMAGNETIC ORDER PARAMETERs

The double-time temperature dependent retarded Green function is given by (Zubarev) [18]:

$$G_r(t-t') \equiv \langle\langle \hat{A}(t);\hat{B}(t')\rangle\rangle$$
$$= -i\theta(t-t') < [\hat{A}(t),\hat{B}(t')] > \tag{5}$$

Where $\hat{A}$ and $\hat{B}$ are Heisenberg operators and $\theta(t-t')$ is the Heaviside step function, defined as,

$$\theta(t-t') = \begin{cases} 1 \ , if\ t > t' \\ 0 \ , if\ t < t' \end{cases} \tag{6}$$

which is related to the Dirac delta function as, $\frac{d}{dt}[\theta(t-t')] = \delta(t-t')$ (7)

Using the Heisenberg equation of motion (with $\hbar = 1$),

$$i\frac{d\hat{A}(t)}{dt} = [\hat{A}(t), H]$$

along with the property (7), it is straightforward to obtain the following equation of motion for $G_r(t-t')$

$$i\frac{d}{dt}G_r(t-t') = \delta(t-t') < [\hat{A}(t),\hat{B}(t')] > + \langle\langle [\hat{A}(t),H],\hat{B}(t')] \rangle\rangle \tag{8}$$

The Fourier transformation $G_r(\omega)$ is given by

$$G_r(t-t') = \int G_r(\omega) \exp\,[-i\omega(t-t')]d\omega \tag{9}$$

Taking the Fourier transform of (8), we get:

$$\omega G_r(\omega) = < [\hat{A}(t),\hat{B}(t')] >_\omega + \langle\langle [\hat{A}(t),H],\hat{B}(t')] \rangle\rangle_\omega \tag{10}$$

From (10), it follows that

$$\omega \langle\langle \hat{a}_{\kappa\uparrow}\, \hat{a}^{\dagger}{}_{\kappa\uparrow} \rangle\rangle = 1 + \langle\langle [\hat{a}_{\kappa\uparrow}, H], \hat{a}^{\dagger}{}_{\kappa\uparrow} \rangle\rangle \tag{11}$$

where the anti-commutation relation,

$$\{\hat{a}_{\kappa\sigma}, \hat{a}^{\dagger}{}_{\kappa'\sigma'}\} = \delta_{\kappa\kappa'}\delta_{\sigma\sigma'}, \tag{12}$$

has been used. To derive an expression for $\ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg$, we have calculate the commutator $[\hat{a}_{\kappa\uparrow}, H]$, using (2), (3), and (4). After some lengthy but straightforward calculations and using the identities $[A, BC] = \{A, B\}C - B\{A, C\}$ and $[AB, C] = A\{B, C\} - \{A, C\}B$ (13)
we arrive at the following results:

$$[\hat{a}_{\kappa\uparrow}, H_1] = \epsilon_{\kappa}\hat{a}_{\kappa\uparrow}$$

$$[\hat{a}_{\kappa\uparrow}, H_2] = -\sum_p V_{kp}\, \hat{a}^{\dagger}_{-\kappa\downarrow}\hat{a}_{-p\downarrow}\hat{a}_{p\uparrow} \tag{14}$$

$$[\hat{a}_{\kappa\uparrow}, \hat{H}_3] = \sum_{l,m} u^{k}{}_{l,m}\hat{a}^{\dagger}_{-\kappa\downarrow}\, \hat{b}_{l\uparrow}\hat{b}_{m\uparrow}$$

Plugging (14) in to (11), we get

$$\omega \ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg = 1 + \epsilon_{\kappa} \ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg - \sum_p V_{kp} \ll \hat{a}^{\dagger}_{-\kappa\downarrow}\hat{a}_{-p\downarrow}\hat{a}_{p\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg$$

$$+ \sum_k u^{k}{}_{l,m} \ll \hat{a}^{\dagger}_{-\kappa\downarrow}\, \hat{b}_{l\uparrow}\hat{b}_{m\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg \tag{15}$$

Using Wick's theorem for factorization (Schwabl) [19], we can reduce (15) into the following form,

$$(\omega - \epsilon_{\kappa}) \ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg = 1 - (\Delta - \gamma) \ll \hat{a}^{\dagger}_{-\kappa\downarrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg \tag{16}$$

where

$$\Delta = V\sum_p \ll \hat{a}_{-p\downarrow}, \hat{a}_{p\uparrow} \gg = V\sum_p \ll \hat{a}^{\dagger}_{-p\downarrow}, \hat{a}^{\dagger}_{p\uparrow} \gg \tag{17}$$

$$\gamma = u\sum_k \ll \hat{b}_{l\uparrow}, \hat{b}_{m\uparrow} \gg = u\sum_k \ll \hat{b}^{\dagger}_{l\uparrow}, \hat{b}^{\dagger}_{m\uparrow} \gg \tag{18}$$

are related to the superconducting and ferromagnetic order parameters respectively, and are assumed to be real.

To determine $\ll \hat{a}^{\dagger}_{-k\downarrow}, \hat{a}^{\dagger}_{k\uparrow} \gg$ in (16), we use (10), to obtain

$$\omega \ll \hat{a}^{\dagger}_{-\kappa\downarrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg = \ll [\hat{a}^{\dagger}_{-\kappa\downarrow}, H], \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg \tag{19}$$

where (12) has been used,

Proceeding in the same manner, we can reduce (19) into the form:

$$(\omega + \epsilon_{\kappa}) \ll \hat{a}^{\dagger}_{-\kappa\downarrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg = -(\Delta - \gamma) \ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}_{\kappa\uparrow} \gg \tag{20}$$

Eliminating $\ll \hat{a}_{\kappa\uparrow}, \hat{a}^{\dagger}_{\kappa\uparrow} \gg$ from (16) and (20), we obtain,

$$\ll \hat{a}^{\dagger}_{-\kappa\downarrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg = \frac{-(\Delta - \gamma)}{(\omega^2 - \epsilon_{\kappa}^2 - (\Delta - \gamma)^2)} \tag{21}$$

To take into account the temperature dependence of order parameters, we shall rewrite (17) and (18) as:

$$\Delta = \frac{V}{\beta}\sum_{\kappa} \ll \hat{a}^{\dagger}_{-\kappa\downarrow}, \hat{a}^{\dagger}{}_{\kappa\uparrow} \gg \tag{22}$$

$$\gamma = \frac{u}{\beta}\sum_{\kappa} \ll \hat{b}^{\dagger}_{p\uparrow}, \hat{b}^{\dagger}{}_{m\uparrow} \gg \tag{23}$$

where $\beta = \frac{1}{K_B T}$

For mathematical convenience, we replace the summation in (22) by integration. Thus

$$\sum_{\kappa} \equiv \int_{-\epsilon_F}^{\omega} N(0)\, d\epsilon \tag{24}$$

where $N(0)$ is the density of states at the Fermi level.

Substituting (24) and (22) in (21), we obtain,

$$\Delta = -\frac{VN(0)}{\beta}\int_{-\epsilon_F}^{\infty}\frac{(\Delta-\gamma)}{(\omega^2-\epsilon^2-(\Delta-\gamma)^2)}d\epsilon \qquad (25)$$

For further simplification, we use the Matsubara frequency (Allen and Dynes[20]),

$$\omega \to i\omega_n \text{ where, } \omega_n = \frac{(2n+1)\pi}{\beta} \qquad (26)$$

Using (26) and the fact that for effective pairing interactions, $-\hbar\omega_b < \epsilon < \hbar\omega_b$, we have from (25) ,

$$\Delta = 2VN(0)\beta\sum_n\int_0^{\hbar\omega_b}\left[\frac{(\Delta-\gamma)}{(2n+1)^2\pi^2+\beta^2E^2}\right]d\epsilon, \qquad (27)$$

where $E^2 = \epsilon^2 + (\Delta-\gamma)^2$.

Finally, using the Poisson summation

$$\frac{1}{2x}\tanh\left(\frac{x}{2}\right) = \sum_n\frac{1}{(2n+1)^2\pi^2+x^2} \qquad (28)$$

And defining $\alpha = VN(0)$, we can rewrite (27) as:

$$\frac{\Delta}{\alpha} = \int_0^{\hbar\omega_b}\frac{(\Delta-\gamma)}{\sqrt{\epsilon^2+(\Delta-\gamma)^2}}\tanh\left(\beta\sqrt{\epsilon^2+(\Delta-\gamma)^2}/2\right)d\epsilon \qquad (29)$$

From (29), it clearly follows that the order parameters Δ and γ , for superconductivity and magnetism are interdependent.

We now consider the equations of motion for the Green function for the localized holmium f-electrons, which are responsible for magnetism. From (10), we can write,

$$\omega \langle\langle \hat{b}_{l\uparrow}, \hat{b}^{\dagger}{}_{l\uparrow} \rangle\rangle = 1 + \langle\langle [\hat{b}_{l\uparrow}, H], \hat{b}^{\dagger}{}_{l\uparrow} \rangle\rangle \qquad (30)$$

Substituting for H and following exactly the same procedure, we arrive at the relation,

$$\langle\langle \hat{b}^{\dagger}{}_m, \hat{b}^{\dagger}_l \rangle\rangle = \frac{\Delta_{lm}}{\omega^2-\epsilon_l^2-\Delta_{lm}^2} \qquad (31)$$

where $$\Delta_{lm} = \sum_k u_k^{lm} \langle\langle \hat{a}^{\dagger}_{k\uparrow}, \hat{a}^{\dagger}{}_{-k\downarrow} \rangle\rangle \qquad (32)$$

substituting (31) in (23), and using (26) and (28), we arrive at the following result

$$\frac{\gamma}{\alpha} = \Delta_{lm}\int_0^{\hbar\omega_D}\frac{1}{\sqrt{\epsilon^2+\Delta_{lm}^2}}\tanh\left(\beta\sqrt{\epsilon^2+\Delta_{lm}^2}/2\right)d\epsilon \qquad (33)$$

From (33), it is again evident that the order parameters Δ and γ are interdependent, as was the case from (29).

It is, therefore, possible that in some temperature interval, ferromagnetism and superconductivity can co-exist, although one phase has a tendency to suppress the critical temperature and the order parameter of the other phase.

IV. DEPENDENCE OF THE MAGNETIC ORDER PARAMETER ON THE TRANSITION TEMPERATURE FOR SUPERCONDUCTIVITY AND FERROMAGNETISM

To study how γ depends on the superconducting transition temperature T_C, we consider the case, when $T \to 0$, or $\beta \to \infty$

We can then replace
$\tanh(\beta\sqrt{\epsilon^2 + (\Delta-\gamma)^2}/2) \to 1$
in (29) and get,

$$\frac{\Delta}{\alpha} = \int_0^{\hbar\omega_b} \frac{(\Delta-\gamma)}{\sqrt{\epsilon^2 + (\Delta-\gamma)^2}} d\epsilon$$

or

$$\frac{1}{\alpha} = \left(1 - \frac{\gamma}{\Delta}\right) sinh^{-1}\left(\frac{\hbar\omega_b}{\Delta-\gamma}\right) \tag{34}$$

since $\left(\frac{\hbar\omega_b}{\Delta-\gamma}\right)^2 \gg 1$, the above equation reduces to

$$\frac{1}{\alpha} = \left(1 - \frac{\gamma}{\Delta}\right) \ln\left(\frac{2\hbar\omega_b}{\Delta-\gamma}\right)$$

from which it immediately follows that,

$$\Delta - \gamma = (2\hbar\omega_b)\exp\left(-\frac{1}{\alpha(1 - \gamma/\Delta)}\right) \tag{35}$$

from the BCS theory, the order parameter Δ, at T=0 for a given superconductor with transition temperature T_C is given by

$$2\Delta(0) = 3.5\, k_B T_c \quad (36$$

using this result in (35), we obtain

$$\gamma = 1.75 k_B T_c - 2\hbar\omega_b exp\left(-\frac{1}{\alpha(1 - \frac{\gamma}{1.75 k_B T_c})}\right) \tag{37}$$

Once we know α, we can solve (37) numerically to draw the phase diagram for γ and T_C.

To estimate α, we consider the case,

$T \to T_C$

which implies, $\Delta \to 0$

From (29), we then have

$$\frac{1}{\alpha} = \int_0^{\hbar\omega_b} \frac{1}{\sqrt{(\epsilon^2+\gamma^2)}} \tanh(\beta\sqrt{\epsilon^2+\gamma^2}/2) d\epsilon - \lim_{\Delta\to 0}\int_0^{\hbar\omega_b} \frac{\gamma}{\Delta\sqrt{(\epsilon^2 + (\Delta-\gamma)^2)}} \tanh(\beta\sqrt{\epsilon^2+(\Delta-\gamma)^2}/2) d\epsilon$$
$$= I_1 - I_2 \tag{38}$$

Putting $\epsilon^2 + \gamma^2 = E^2$,
we can write

$$I_1 = \int_0^{\hbar\omega_b} \frac{1}{E}\tanh(\beta\, E/2) dE \tag{39}$$

Using the result

$$\int_0^x \frac{tanh x}{x} dx = (\ln x)(\tanh x)\,\Big|_0^x - \int_0^x \frac{\ln x}{cosh^2 x} dx$$

and the fact that, at low temperature, $\tanh\left(\frac{\hbar\omega_b}{2k_B T_c}\right) \to 1$

we have from (39),

$$I_1 = \ln\left(\frac{\hbar\omega_b}{2k_B T_c}\right) - \ln\left(\frac{\pi}{4\delta}\right) \tag{40}$$

where δ is the Euler constant having the value $\delta = 1.78$ (Hsian) [21]

we can write (40) as,

$$I_1 = \ln(1.14\,\frac{\hbar\omega_b}{k_B T_c}) \tag{41}$$

Using L' Hospital's rule, it is easy to show that

$$I_2 = -\int_0^{\hbar\omega_b} (\gamma^2\beta) \frac{sech^2\left(\beta\sqrt{\epsilon^2+\gamma^2}/2\right)}{2(\epsilon^2+\gamma^2)} d\epsilon$$

which can be neglected since γ^2 is very small.

Substituting (41) in (38), we then obtain

$$\frac{1}{\alpha} = \ln\left(1.14\,\frac{\hbar\omega_b}{k_B T_c}\right)$$

This implies,

$$T_c = \frac{1.14\hbar\omega_b}{k_B}\exp\left(-\frac{1}{\alpha}\right) \tag{42}$$

which can be used to estimate $\exp\left(-\frac{1}{\alpha}\right)$ for $HoMo_6S_8$, using the experimental value T_C=1.82 k for this compound (Lynn et al.[6]).

To study how γ depends on the magnetic transition temperature T_m, we consider (33). Neglecting Δ^2, and proceeding as before, it is easy to show that,

$$\gamma \approx -(\alpha\Delta)\ln(1.14\frac{\hbar\omega_b}{k_B T_m})$$

which gives,

$$T_m = \left(\frac{1.14\hbar\omega_b}{k_B}\right)\exp\left(\frac{\gamma}{\alpha\Delta}\right) \tag{43}$$

From (36), we can estimate $\Delta(0) \approx 0.28meV$ for $HoMo_6S_8$, using the known value of T_C.

we can use (43) to draw the phase diagram for γ and T_m.

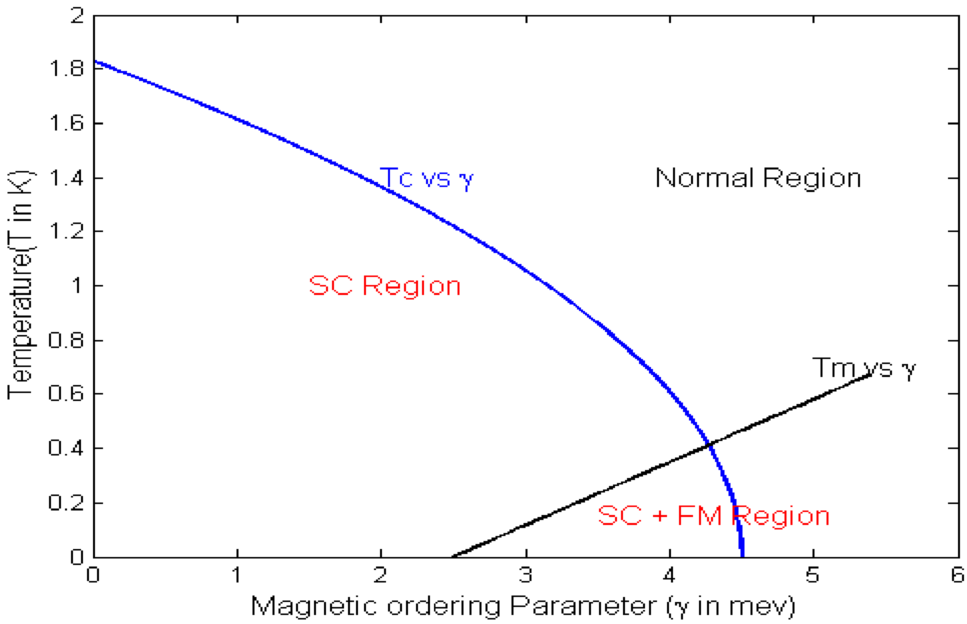

Fig. 1: Co-existence of superconductivity and ferromagnetism in $HoMo_6S_8$.

V. RESULTS AND DISCUSSION

In Fig. 1, we have presented the theoretical curve of the magnetic order parameter γ as a function of the superconducting temperature T_C. For this purpose, we have used (eqn. 37) which has been numerically solved using the relevant parameters for $HoMo_6S_8$. In the same figure, we have also plotted the curve of γ as a function of the magnetic transition temperature T_m, using (eqn. 43). This curve is found to be almost linear up to the temperature T_m = 0.67 k, which is the experimental value for $HoMo_6S_8$.

From Fig. 1, we observe that T_C decreases with increase in γ, whereas T_m increases with increase in γ. The superconducting and ferromagnetic phases, therefore, oppose each other. However, the present model shows that there is a small region of temperature, where both the phases may appear together. This is indicated by (SC+FM) in the figure.

We have thus demonstrated that a simple model based on a Hamiltonian which takes into account the spin interactions between conduction electrons and ferromagnetically ordered localized electrons, can explain the co-existence of singlet superconductivity and ferromagnetism in $HoMo_6S_8$.

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