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Theoretical studies on magnetic superconductors

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Abstract. The discovery of magnetic superconductors has posed the problem of the coexistence of two kinds of orders (magnetic and superconducting) in some temperature intervals in these systems. New microscopic mechanisms developed by us to explain the coexistence and reentrant behaviour are reported. The mechanism for antiferromagnetic superconductors which shows enhancement of superconductivity below the magnetic transition is found relevant for rare-earth systems having less than half-filled f-atomic shells. The theory will be compared with the experimental results of $SmRh_4B_4$ system. A phenomenological treatment based on a generalized Ginzburg-Landau approach will also be presented to explain the anomalous behaviour of the second critical field in some antiferromagnetic superconductors.

These magnetic superconductors provide two kinds of Bose fields, namely, phonons and magnons which interact with each other and also with the conduction electrons. Theoretical studies of the effects of the excitations of these modes on superconducting pairing and magnetic ordering in these systems will be discussed.

Keywords. Magnetic superconductors; second critical field; coexistence; enhancement of superconductivity.

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1. Introduction

The discovery of a large number of magnetic superconductors in the last ten years has led to considerable amount of theoretical and experimental investigations. It was believed that these two kinds of cooperative states were mutually exclusive. In the singlet superconductors we have ordering of conduction electrons in the momentum space in the time-reversed state ($\mathbf{k}\uparrow$; $-\mathbf{k}\downarrow$). In magnetic cooperative phenomena, we have ordering of magnetic moments in the ordinary space. It was thought that the generation of large internal fields in magnetic superconductors would destroy time reversal symmetry and hence suppress singlet superconductivity completely.

The appearance of both kinds of orders in some temperature intervals and even their coexistence in some rare earth compounds had posed theoretical challenges and opened avenues for new experiments. A vast amount of literature has been gathered in the last few years. We shall not review here the present status of experimental results and various theoretical models developed. These are now available in two review articles (Shrivastava and Sinha 1984; Bulaevskii *et al* 1985). We shall, therefore, address ourselves to some specific systems and specific properties. The coexistence is most pronounced in antiferromagnetic superconductors (RRh_4B_4, R = Nd, Sm, Tm; RMO_6S_8, R = Gd, Tb, Dy). The system RMO_6S_8 exhibits anomalous depression of H_{C2} (second critical field) near but below T_N. For $SmRh_4B_4$, H_{C2} increases below T_N

after a break in slope, suggesting an enhancement of superconductory in this system with the onset of antiferromagnetic ordering (Maple 1982). The mechanism involving suppression of magnetic scattering below T_N alone will not be adequate to explain the observed enhanced superconducting pair density. For example in $TmRh_4B_4$ one gets a depression of H_{C2} below T_N. In fact a bell-shaped curve for H_{C2} versus temperature (T) is obtained (Hamaker *et al* 1981). It would appear therefore that the state of rare-earth ions in the crystal is important in determining the mechanism of enhancement. The most obvious difference is that for all those systems where depression of H_{C2} below T_N occurs the rare earth ions have half-filled or more than half-filled *f*-shells. The systems showing enhancement (e.g., $SmRh_4B_4$) have rare earth ion (e.g. Sm) containing less than half-filled *f*-shell. This situation makes the emergence of a new mechanism possible which can be strong enough to counter the exchange scattering mechanism and, in effect, lead to the enhancement of the strength of superconducting pairing interaction below the antiferromagnetic ordering temperature. In what follows, we shall discuss the role of one such mechanism (Sinha 1979).

Furthermore, in magnetic superconductors we can produce real boson fields e.g. magnons and phonons by the application of appropriate fields. The presence of such Bose fields will be explored in the context of enhancement or suppression of superconducting pairing and magnetic order.

2. Mechanism and enhancement of superconducting order

The Hamiltonian for the system comprising conduction electron derived from Rh 4*d* orbitals, localized *f* electrons and various interactions can be written as

$$\begin{aligned} H = & \sum_{\mathbf{k}\sigma} (\varepsilon_{\mathbf{k}} - \mu) C^+_{\mathbf{k}\sigma} C_{\mathbf{k}\sigma} + \sum_n E_n C^+_{n\sigma} C_{n\sigma} \\ & - V \sum_{\mathbf{k}\mathbf{k}'} C^+_{\mathbf{k}'\uparrow} C^+_{-\mathbf{k}'\downarrow} C_{-\mathbf{k}\downarrow} C_{\mathbf{k}\uparrow} \\ & + \left(\sum_{\mathbf{k}lm} g^{lm}_{\mathbf{k}} C^+_{l\uparrow} C^+_{m\downarrow} C_{-\mathbf{k}\downarrow} C_{\mathbf{k}\uparrow} + \text{h.c.} \right) + \sum_{lm} H_{lm}(\text{ex}), \end{aligned} \tag{1}$$

where the first two terms are the conduction electron (creation, annihilation) operators $C^+_{\mathbf{k}\sigma}$, $C_{\mathbf{k}\sigma}$ for the Bloch state $|\mathbf{k}\sigma\rangle$, and rare-earth site localized (creation, annihilation) operators ($C^+_{n\sigma}$, $C_{n\sigma}$), at site $\mathbf{R}_n$ with single particle energies ($\varepsilon_{\mathbf{k}}$ = conduction electron energy, E_n = rare earth localized electron energy). The third term is the usual phonon mediated BCS pairing interaction between conduction operators, V being the interaction coefficient. The fourth term is the new interaction, involving conduction electron pairs making transition to localized states at sites l and m respectively; $g^{lm}_{\mathbf{k}}$ being the interaction constant. The last term $\sum_{lm} H_{lm}(\text{ex})$ represents the effective exchange interaction between rare-earth magnetic ions which may arise from various mechanisms (direct and indirect). Here l and m run over two magnetic sublattices respectively. The effect of the new interaction (cf fourth term of equation (1)) can be taken account either in the gap function (Sinha 1979), or in giving an additional pairing interaction which depends on antiferromagnetic order. In the present paper, we shall follow the latter procedure. This term in the first order can be eliminated by a suitable canonical transformation. This leads to an interaction term which gives BCS like pairing of two conduction electrons but the coefficient depends on the antiferromagnetic order. In fact, it can be shown to depend on sub-lattice magnetization and hence the strength of this interaction will increase with sublattice magnetization as the temperature is decreased below T_N (the Ne'el temperature). The new pairing interaction can be explicitly written as (Jagadish and Sinha 1986),

$$-\sum_{\mathbf{k}\mathbf{k}'}\left(\sum_{lm}\frac{g_{\mathbf{k}}^{lm}\,g_{\mathbf{k}}^{lm*}}{\Delta E_0}\,(|S_l^Z|+|S_m^Z|)\,C_{\mathbf{k}'\uparrow}^{+}\,C_{-\mathbf{k}'\downarrow}^{+}\,C_{-\mathbf{k}\downarrow}\,C_{\mathbf{k}\uparrow}\right. . \tag{2}$$

Combining this with BCS pairing interaction, the effective pairing interaction can be recast in the form

$$\lambda_{\rm eff}=\lambda_{\rm BCS}[1+\alpha_m\, m(T)]$$

or

$$\bar{V}N(0)=VN(0)\,[1+\alpha\, m(T)]=V\bar{N}(0), \tag{3}$$

where $m(T)$ is the reduced sub-lattice magnetization $m(T)=M(T)/M(0)$ and α_m is the coefficient. The magnetic exchange scattering considered by Machida (1980) (who extended the treatment of Abrikosov and Gorkov 1961), namely, $(I/2N)\,(g_J-1)\,\mathbf{J}\cdot\sigma$ where $\mathbf{J}$ is the rare-earth ion angular momentum, g_J the Lande factor, I the exchange constant for scattering between conduction electrons and localized magnetic moments, also leads to a similar expression but with opposite sign.

$$-\alpha m(T), \tag{4}$$

where

$$\alpha=\frac{1}{4\pi}(g_J-1)\,[J(J+1)]^{1/2}\,\frac{|I|}{\varepsilon_F\,VN(0)}, \tag{5}$$

ε_F being the Fermi energy. Owing to the appearance of this form it is convenient to define α_m as the sum total of the new attractive interaction suggested here and the magnetic exchange scattering effect. More explicitly

$$\alpha_m=\alpha_c-\alpha, \tag{6}$$

where α_c arises from the new interaction (pair healing). For rare earth ions having more than half-filled f-shells, the energy denominator ΔE_0 is very high (owing to the existence of two electrons in the same orbital) and α_c will be much weaker than α. However, for less than half-filled shells, this is not so and α_c is likely to be larger than (or comparable with) α. In such situations, α_m is positive and one will have enhancement of superconducting pairing. In figure 1, we compare the experimental data on $SmRh_4B_4$ with the calculated curve for the second critical field H_{C2} against temperature. For this purpose we compute the gap function $\Delta(T)$ in the regions $T_C>T>T_N$ and $T_N>T>0$. In the latter portion the role of the new mechanism is taken into account. Having computed the gap function, H_{C2} is calculated via H_C, namely,

$$H_{C2}=\sqrt{2}\,\kappa\, H_C, \tag{7}$$

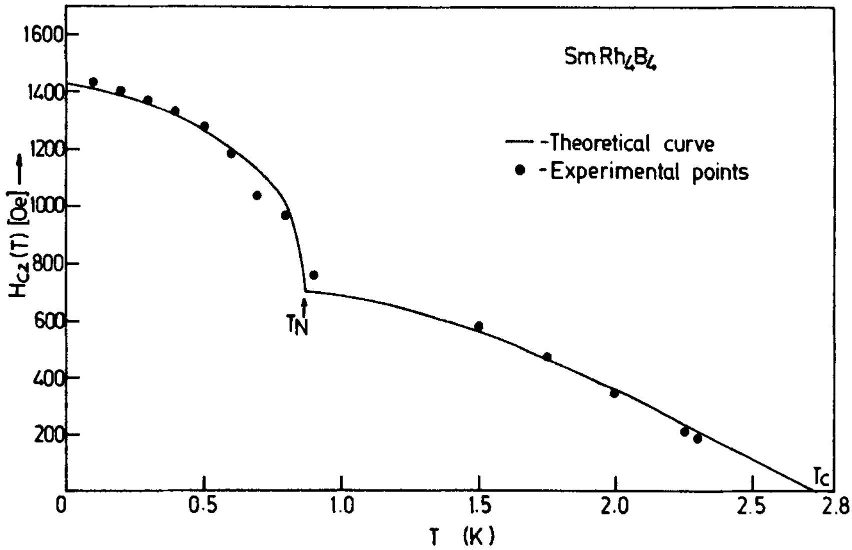

Figure 1. Temperature dependence of the critical field H_{C2} in $SmRh_4B_4$. The enhancement of H_{C2} below T_N is a direct result of the additional positive contribution to the interaction strength or equivalently to the density of states $N(0)$. Below T_N the coefficient of the BCS term in the Hamiltonian is $N(0)\,V(1+\alpha_m\, m(T))$. The value used for $N(0)$ is 0·57 states/eV-atom-spin direction, that for α_m is 0·2. The form $[1-(T/T_N)]^{0\cdot 5}$ is used for $m(T)$. A constant value of $\kappa = 2{\cdot}1$ is assumed for the region above T_N. Since κ depends on the density of states, which changes below T_N, the average value of $\kappa(=1{\cdot}55)$ below T_N is different.

$$\frac{-H_C^2}{8\pi} = -\frac{1}{2}N(0)\,\Delta^2 - N(0)\,\Delta^2 \ln\left|\frac{\Delta_0}{\Delta}\right| - 4\,N(0)\,k_B T\int_0^{\hbar\omega_D} \mathrm{d}\varepsilon \ln\left[1+\exp(-\beta\varepsilon)\right] + \frac{1}{3}\pi^2 N(0)\,(k_B T)^2. \tag{8}$$

The agreement between calculated and experimental values is satisfactory.

3. Phenomenological treatment

The phenomenological treatment for the temperature dependence of the second critical field H_{C2} for an antiferromagnetic superconductor is given in terms of a generalized Ginzburg-Landau (GGL) theory (Mahanti *et al* 1981) involving two order parameters $\psi(\mathbf{r})$ and $M(\mathbf{r})$. The Gibbs free energy of the system

$$G(T, \mathbf{H}; \psi, \mathbf{h}, \mathbf{M}) = F_S(T, \psi, \mathbf{h}) + F_m(T, \mathbf{M}) + F_{Sm}(T, \psi, \mathbf{M}) - \frac{1}{4\pi}\int (\mathbf{h}\cdot(\mathbf{H}+4\pi\,\mathbf{M}))\,\mathrm{d}r, \tag{9}$$

where $F_S(T, \psi, \mathbf{h})$ is the Helmholtz free energy of a superconductor characterized by a microscopic field $\mathbf{h}(r)$. This includes the term $(1/8) \int h^2 \, \mathrm{d}r$. Explicitly

$$F_S(T, \psi, \mathbf{h}) = \int \mathrm{d}\mathbf{r} \, [\tfrac{1}{2} a|\psi|^2 + \tfrac{1}{4} b|\psi|^4 + p_0 |(\nabla - ir_0 \mathbf{A})\psi|^2 + |\mathbf{h}(r)|^2/8\pi]$$

$$p_0 = \hbar^2/2m^*, \quad r_0 = 2e/\hbar c, \tag{10}$$

$$\tilde{F}_m(T, \mathbf{M}) = F_m(T, \mathbf{M}) + \int 2\pi M^2 \, \mathrm{d}r, \tag{11}$$

$F_m(T, \mathbf{M})$ being the Helmholtz free energy of the rare earth magnetic system, and

$$F_{Sm}(T, \psi, \mathbf{M}) = \tfrac{1}{2} \int (\eta_1 |\mathbf{M}|^2 + \eta_2 |\nabla \mathbf{M}|^2) |\psi|^2 \, \mathrm{d}r. \tag{12}$$

A few words about the coefficients η_1 and η_2 will be appropriate here. Here η_1 takes into account the combined effect of exchange (pair-breaking) scattering and the new mechanism (pair-healing) discussed in § 2. Thus the overall sign of η_1 will depend on the fact as to which of the two mechanisms is the dominant one in a particular system. Similarly for η_2 as we can approximate $\eta_2 = \xi^2 \eta_1$, ξ being the superconducting coherence length. We get generalized G-L equations by minimizing G with respect to ψ^*, $\mathbf{h}$ and $\mathbf{M}$. The equations can be solved under various degrees of approximations. We shall write down only the important results. The effect of antiferromagnetic long range order on the superconducting order parameter to leading order in M turns out to be (on going over the q-space).

$$|\psi(T, \mathbf{M})| = |\psi(T, 0)| \left[1 - \frac{1}{b} \left\{ (\eta_1 + \eta_2 Q^2)/|\psi(T, 0)|^2 + \frac{4\pi Q^2 \lambda^2(T, 0)}{|\psi(T, 0)|^4 (1 + Q^2 \lambda^2(T, 0))^2} \right\} M^2 \right]^{1/2}, \tag{13}$$

where Q is the wave-vector defining antiferromagnetic order and λ is the London length. Thus we see that the effect of M on ψ can be of two types. First there is the electrodynamic effect whose strength depends on the quantity $Q\lambda$. This will act against the coexistence of non-zero $|\psi|$ and M. The second is the direct coupling effect whose strength depends on η_1 and η_2. This direct effect can go either way depending on the sign of η_1 and η_2.

If η_1 and η_2 are negative and adequately strong, then $\psi(T, M)$ will be enhanced below the magnetic transition. Now $\eta_1 = \eta_{1s} + \eta_{1c}$ where η_{1s} arises from pair-breaking and η_{1c} from the new mechanism. If the latter dominates then η_1 might become negative. This is expected to be the situation for $SmRh_4B_4$. When pair-breaking (exchange) scattering dominates we will have lowering of $\psi(T, M)$ below the magnetic transition.

These effects are experimentally seen clearly by measuring the second criterial field H_{C2} as a function of temperature (T). Accordingly, we shall give the highlights of these calculations on the basis of the generalized GL model for antiferromagnetic superconductors.

We consider the situation first without direct interaction between ψ and M (Mahanti *et al* 1981) i.e., $F_{sm} = 0$. For $H \approx H_{C2}$, the superconducting order parameter is small and one can linearize the GGL equations by keeping terms linear in ψ. The relevant equations are

$$a\psi - 2p_0(\nabla - ir_0\mathbf{A})^2\psi = 0. \tag{14}$$

$$\mathbf{h} = \mathbf{H} + 4\pi\mathbf{M} \equiv \nabla\times\mathbf{A}. \tag{15}$$

In the absence of an external field the magnetization density is taken to be of the form

$$\mathbf{M}(\mathbf{r}) = \hat{m}M_Q\cos\mathbf{Q}\cdot\mathbf{r} \tag{16}$$

where $\mathbf{Q}$ is the wave vector of AF order. In the presence of a field we have

$$\mathbf{M}(\mathbf{r}) = \hat{Z}\bar{\chi}(T)\,\mathbf{H} + \hat{m}M_Q(H,T)\cos\mathbf{Q}\cdot\mathbf{r}, \tag{17}$$

$\bar{X}$ being the average uniform ($q=0$) susceptibility. Also the vector potential is separated into two parts i.e., $\mathbf{A} = \mathbf{A}_0 + \mathbf{A}_1$, with

$$\mathbf{A}_0 = \hat{y}(1+4\pi\bar{\chi})Hx, \tag{18}$$

$$\mathbf{A}_1 = \frac{\hat{\mathbf{n}}\,4\pi M_Q(H,T)}{Q}\sin\mathbf{Q}\cdot\mathbf{r}. \tag{19}$$

The solution for the second critical field is found to be

$$\frac{H_{C2}(T)}{H^0_{C2}(T)} = \frac{1}{1+4\pi\bar{\chi}}\left[1-\left(\frac{4\pi M_Q}{H^0_{C2}}\right)^2\frac{1}{(Q\xi)^2}\right], \tag{20}$$

where $H^0_{C2} = \phi_0/2\pi\xi^2$, ($\phi_0 = eh/2c$, the flux quantum) is the critical field in the absence of screening and effects arising from magnetic order. The above expression takes into account uniforming screening effect proportional to $(1+4\pi\bar{\chi})^{-1}$ which tends to suppress H_{C2} and an electrodynamic effect which depends on M_Q and suppresses H_{C2} below T_N. Computation for some actual systems shows that apart from giving a dip around T_N in a few systems (e.g. $Dy(Mo)_6S_8$), the indirect effect is not adequate to explain the observed lowering or enhancements in other systems. It appears, therefore, that we must take into account the direct coupling F_{sm}. This will introduce additional terms such as $(\eta_1 M^2 + \eta_2|\nabla M|^2)\psi$ in (14); similarly for $M(r)$. Then the expected form for H_{C2} is given by

$$\frac{H_{C2}(T)}{H^0_{C2}} = \frac{1}{1+4\pi\bar{X}}\left[1-\left(\frac{4\pi M_Q}{H^0_{C2}}\right)^2\frac{1}{(Q\xi)^2} - (\eta_1 M_Q^2 + \eta_2 Q^2 M_Q^2)\,\xi^2\right]. \tag{21}$$

If η_1 and η_2 have positive signs i.e., they are dominated by exchange scattering and pair-breaking effects, the additional terms will lead to lowering of $H_{C2}(T)$ below T_N. This is actually so in systems such as $TmRh_4B_4$ and $Dy(MO)_6S_8$ etc. However, when they have negative signs owing to the domination of the new mechanism, the additional terms will enhance $H_{C2}(T)$ below T_N. This is the case for $SmRh_4B_4$ where the new mechanism not only quenches exchange scattering but leads to the enhancement of the superconducting order parameter and the second critical field below T_N. Thus the new mechanism, which operates in some systems effectively only when antiferromagnetic order sets in, leads to the enhancement.

4. Concluding remarks

In the foregoing sections, we have discussed the role of a new mechanism which involves transitions of a Cooper pair to two localized states with anti-parallel spins at two rare earth ion sites. This is found to be important for the enhancement for superconducting order in some systems ($SmRh_4B_4$). It can effectively quench exchange scattering and even lead to enhancement below T_N. Both microscopic and phenomenological treatments are discussed.

The gap function (both in the microscopic and phenomenological approaches) depends on the staggered magnetization below T_N in antiferromagnetic superconductors. The question naturally arises as to what effects will be produced when magnetization is changed by the excitation of spin wave modes. As the magnetization decreases on the excitation of these modes on the application of external perturbations one can expect either of the two possibilities. For systems dominated by exchange scattering, spin wave excitation will help superconducting order. On the other hand, when the new mechanism (pair-healing) dominates spin wave excitation will go in the opposite direction. It is worthwhile conducting experiments on magnetic superconductors and see what effects spin wave excitations produce on superconducting order-parameter or the second critical field.

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