PHYSICAL REVIEW B VOLUME 28, NUMBER 1 1 JULY 1983

Microscopic theory of coexistence of superconductivity and antiferromagnetism

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(Received 3 January 1983)

A theory of the coexistence of superconductivity and antiferromagnetism is presented. We study the role of the "diagonal" exchange coupling between magnetic ions and conduction electrons, using Eliashberg's formalism. This coupling generates a spatial displacement of the Cooper-paired states, and thus reduces the pairing strength. The reduction is linear in the exchange integral and the staggered magnetization. The theory agrees well with experiment for $Dy_{1.2}Mo_6S_8$ and $Tb_{1.2}Mo_6S_8$.

Several rare-earth ternary compounds have been reported,[1,2] in which[3] antiferromagnetism (AF) and superconductivity (SC) coexist. The rare-earth ions form an ordered lattice of magnetic spins, interacting weakly with the conduction electrons. The Néel temperature T_N is usually below the SC critical temperature T_c; it is thus possible to see the influence of AF on SC. In Fig. 1, we show the temperature dependence of the critical field H_{c2} in $Dy_{1.2}Mo_6S_8$ and in $Tb_{1.2}Mo_6S_8$. H_{c2} is seen to behave anomalously near T_N, and $H_{c2}(0)$ is considerably smaller than expected from extrapolation of the high-temperature behavior.[1,4]

Baltensperger and Strässler[5] show that in an AF, the Cooper pairing must be between time-reversed states displaced by a lattice vector. They argue that in an AF the pairing strength is reduced, by a magnon-mediated repulsive electron-electron interaction. Ramakrishnan and Varma[6] study the effect both of magnons and of the Abrikosov-Gorkov pair-breaking mechanism. Using a sum rule, they show that for nonpathological band structures these effects tend to compensate; moreover, their net contribution is the same above and below T_N. To explain the strong influence of AF on SC, Ref. 6 postulates strong Fermi surface (FS)[3] nesting. This postulate needs justification.

We consider here a more immediate influence of the magnetic ions on the conduction electrons: the "diagonal" part JS_zs_z of the exchange interaction. Below T_N, where the Fourier component $S_z(\vec{q}_0)$ becomes the macroscopic staggered magnetization, we show that this term weakens the phonon-induced attraction between the displaced electron states of Ref. 5. The reduction is linear in the exchange integral. Machida *et al.*[7] also find a linear reduction, but only by making the restrictive assumption $q_0 = 2k_F$. (Here $\vec{q}_0$ is the wave vector of the AF order.)

The present paper is similar in spirit to Zwicknagl and Fulde.[8] However, our formulation is more transparent, and we derive analytic expressions for the essential features. We explicitly relate the strength of SC to the AF order. Reference 8, on the other hand, finds numerical solutions for a particular model, i.e., an assumed band structure, phonon spectrum, and exchange and electron-phonon interactions.

We start from the Hamiltonian

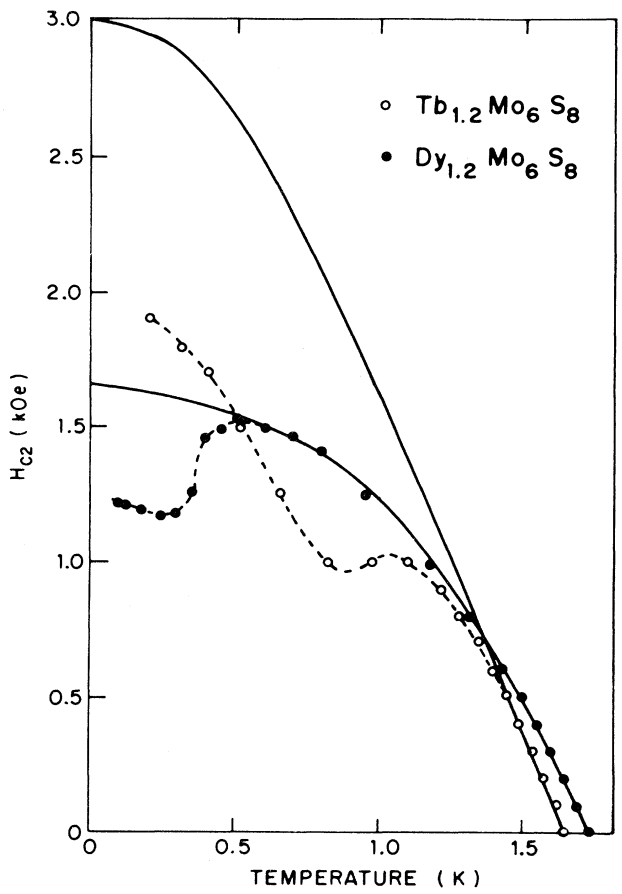

FIG. 1. Temperature dependence (Ref. 1) of H_{c2} (broken curve) and its extrapolation from the paramagnetic region (solid curve) for $Dy_{1.2}Mo_6S_8$ and $Tb_{1.2}Mo_6S_8$.

$$H = H_e + H_{\text{ph}} + H_{e\text{-ph}} \ , \tag{1}$$

$$H_e = \sum_{\vec{k},\sigma} E(\vec{k}) c_\sigma^\dagger(\vec{k}) c_\sigma(\vec{k}) + \tfrac{1}{2} J(\vec{q}_0) S_z(\vec{q}_0) \sum_{\vec{k},\sigma} \sigma c_\sigma^\dagger(\vec{k}) c_\sigma(\vec{k}+\vec{q}_0) \ , \tag{2a}$$

$$H_{\text{ph}} = \sum_{\vec{q},j} \Omega_j(\vec{q}) b_j^\dagger(\vec{q}) b_j(\vec{q}) \ , \tag{2b}$$

$$H_{e\text{-ph}} = \sum_{j,\sigma,\vec{k},\vec{k}'} g_j(\vec{k},\vec{k}') c_\sigma^\dagger(\vec{k}) c_\sigma(\vec{k}') [b_j(\vec{k}-\vec{k}') + b_j^\dagger(\vec{k}'-\vec{k})] \ , \tag{2c}$$

where $c_\sigma(\vec{k})$, $c_\sigma^\dagger(\vec{k})$; $b_j(\vec{q})$, $b_j^\dagger(\vec{q})$ are, respectively, annihilation and creation operators for conduction electrons and phonons, $\sigma = \pm 1$ is a spin index, $\Omega_j(\vec{q})$, $E(\vec{k})$ are the phonon (jth branch) and electron spectra, and $g_j(\vec{k},\vec{k}')$ are the electron-phonon matrix elements. For simplicity, the discussion is limited to a single electron band containing the Fermi level E_F.

We assume that the spin-flip frequency of the magnetic ions is so slow compared to typical electron and phonon frequencies that in (2a) we may replace $S_z(\vec{q}_0)$ by its expectation value $S_M = \langle S_z(\vec{q}_0)\rangle$ (adiabatic approximation). References 7 and 9 make a similar approximation, but start from a BCS Hamiltonian rather than Eq. (1). They thus fail to find the correct effect of AF on the electron-phonon interaction. Nass *et al.*[9] make the further assumption that AF affects the Cooper pairing of an electron $\vec{k}$ only if both $\vec{k}$ and $\vec{k}+\vec{q}_0$ are within the BCS cutoff from the FS. This assumption is shown here to be invalid.

Whether the lattice is magnetically ordered or not, $2\vec{q}_0$ is always a reciprocal-lattice vector, and hence $\vec{k}+2\vec{q}_0$ is equivalent to $\vec{k}$. Hence H_e can be diagonalized by the transformation

$$c_\sigma(\vec{k}) = \cos\alpha_{\vec{k}}\, \bar{c}_\sigma(\vec{k}) - \sigma \sin\alpha_{\vec{k}}\, \bar{c}_\sigma(\vec{k}+\vec{q}_0) \ , \tag{3}$$

where $\alpha_{\vec{k}} = -\alpha_{\vec{k}+\vec{q}_0}$ and

$$\cos 2\alpha_{\vec{k}} = \left[1 + \left(\frac{J(\vec{q}_0) S_M}{E(\vec{k}) - E(\vec{k}+\vec{q}_0)}\right)^2\right]^{-1/2} \ . \tag{4}$$

The transformation (3) transforms $H_{e\text{-ph}}$ to

$$H_{e\text{-ph}} = \sum_{j,\sigma,\vec{k},\vec{k}'} \bar{c}_\sigma^\dagger(\vec{k}) \bar{c}_\sigma(\vec{k}') \{\bar{g}_j(\vec{k},\vec{k}')[b_j(\vec{k}-\vec{k}') + b_j^\dagger(\vec{k}'-\vec{k})] + \sigma \tilde{g}_j(\vec{k},\vec{k}')[b_j(\vec{k}-\vec{k}'+\vec{q}_0) + b_j^\dagger(\vec{k}'-\vec{k}-\vec{q}_0)]\} \ , \tag{5}$$

where $\bar{g}$ and $\tilde{g}$ depend on g and $\alpha_{\vec{k}}$.

The essentially *new feature* is the term in $\tilde{g}$ in (5), a new umklapp term associated with the reduced translational symmetry of the magnetized lattice. Its spin-dependent phase [arising from the phase factor σ in (3)] is related, via Bloch's theorem, to the spatial displacement of the different spin states. This term thus represents an interaction between the phonons and the *spin* density of the conduction electrons. Although explicitly an electron-phonon term, it resembles the electron-magnon interaction *formally*. And, like the electron-magnon interaction, it induces a pair-weakening electron-electron repulsion.

In Nambu's representation the $\bar{g}$ term is described by the usual electron-phonon vertex, containing the Pauli matrix τ_3, but for the $\tilde{g}$ vertex, the corresponding factor is the unit matrix. This introduces additional terms into Eliashberg's equations.[10] One can show that by assuming isotropy, and taking the limit $T \to T_c$, where Eliashberg's equations become linear, the equation for the SC gap function Δ can be written as

$$\Delta(i\omega_n)\left[1 + \frac{1}{|2n+1|}\sum_{n'} \lambda(i\nu_{n'-n})\right] = \sum_{n'} \frac{\Delta(i\omega_{n'})}{|2n'+1|} \lambda(i\nu_{n'-n}) \langle \cos 2\alpha_{\vec{k}} \rangle_{\vec{k}\in \text{FS}} \ , \tag{6}$$

where $\nu_n = 2\pi k_B T n$ and $\omega_n = 2\pi k_B T(n+\frac{1}{2})$ are Matsubara frequencies; the average $\langle \cos 2\alpha_{\vec{k}} \rangle$ is taken over the FS and λ is defined as usual:

$$\lambda(i\nu_n) = 2N(E_F) \sum_j \left\langle \frac{|g_j(\vec{k},\vec{k}')|^2 \Omega_j(\vec{k}-\vec{k}')}{[\Omega_j(\vec{k}-\vec{k}')]^2 + \nu_n^2} \right\rangle_{\vec{k},\vec{k}'} \ . \tag{7}$$

$N(E_F)$ is the density of states at E_F: in three dimensions we may neglect the AF renormalization of $N(E)$. Dispersion in the electron-phonon coupling can cause deviations from Eq. (6); such deviations might perhaps account for the enhancement of SC by AF order in $SmRh_4B_4$.

The AF-induced reduction of the average electron-electron attraction is described by the factor $\langle \cos 2\alpha_{\vec{k}} \rangle^2_{\vec{k} \in FS}$ in (6). To obtain an analytic expression for its magnitude, we make the approximation (rigorous for free electrons) that $E(\vec{k}) - E(\vec{k}+\vec{q}_0)$ is linear in $\vec{k}$ when $\vec{k}$ is on the FS. It is reasonable for more general band structures, because by (4) the main contribution to $\langle \cos 2\alpha_{\vec{k}} \rangle_{\vec{k} \in FS}$ comes from the small regions where $E(\vec{k}) - E(\vec{k}+\vec{q}_0)$ is small and a linear expansion in $\vec{k}$ is valid. Hence, by (4),

$$\langle \cos 2\alpha_{\vec{k}} \rangle_{\vec{k} \in FS} \cong (E_2 - E_1)^{-1} \int_{E_1}^{E_2} dE / \{1 + [J(\vec{q}_0) S_M / E]^2\}^{1/2} , \tag{8}$$

where E_1 and E_2 are the extreme values of $E(\vec{k}) - E(\vec{k}+\vec{q}_0)$ when $E(\vec{k}) = E_F$. We write $B = E_2 - E_1$, assume that $E_1 < 0 < E_2$, and evaluate the integral (8), to get

$$\langle \cos 2\alpha_{\vec{k}} \rangle^2_{\vec{k} \in FS} \cong 1 - 4|J(\vec{q}_0) S_M / B| + O(J^2) . \tag{9}$$

For application to real materials, we approximate B by the width of the nondegenerate band around E_F.

Substituting (9) into (6), we solve[10] to find T_c, the SC transition temperature which we *would* have if S_M were fixed at some temperature-independent value. To first order in $J(\vec{q}_0)/B$,

$$T_c / T_c^0 \cong 1 - 4\gamma |J(\vec{q}_0) S_M / B| , \tag{10}$$

where γ is a constant and $T_c^0 = T_c(S_M = 0)$. In McMillan's[11] "two-square-well" model, γ is approximately

$$\gamma \cong \lambda(1+\lambda)/(\lambda - \mu^*)^2 , \tag{11}$$

where $\lambda = \lambda(0)$, and μ^* is the previously omitted Coulomb repulsion parameter.

In $Dy_{1.2}Mo_6S_8$ and $Tb_{1.2}Mo_6S_8$, SC sets in while the magnetic ions are still disordered, i.e., the observed T_c is T_c^0. The effect of AF is to reduce $H_{c2}(T)$ below the value $H_{c2}^0(T)$ extrapolated from the paramagnetic region. Since we have seen that the primary effect of AF is to reduce the pairing interaction, we can express the reduction in H_{c2} as[4]

$$H_{c2}(T) = H_{c2}^0 [T T_c^0 / T_c(T)] T_c(T) / T_c^0 , \tag{12}$$

where $T_c(T)$ is a hypothetical SC transition temperature found from (10) by freezing S_M at its observed value $S_M(T)$ at temperature T. $[S_M(T)]^2$ is proportional to the intensity of the $\vec{q}_0$ neutron-diffraction peak[2] minus the background intensity, which are shown in Fig. 2 (the horizontal line is the background, found by extrapolation from $T > T_N$). We have tacitly assumed that the ratio of the spin and orbital contributions to the magnetic moments does not depend on T. Using one adjustable parameter to fix the ratio H_{c2}/H_{c2}^0 at $T = 0$, we can calculate $H_{c2}(T)$

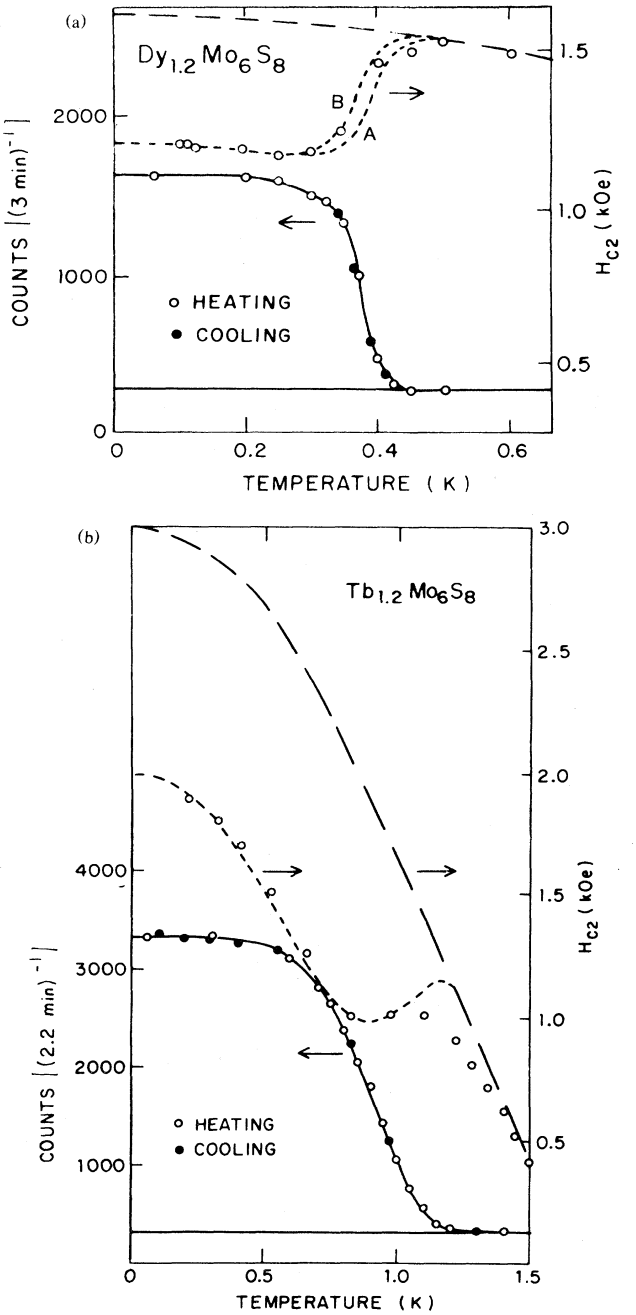

FIG. 2. Temperature dependence of the $\vec{q}_0$ neutron-diffraction peak intensity[2] (solid curve), the experimental and extrapolated H_{c2}^0 from Fig. 1 (open circles and coarse broken curve), and the present theoretical H_{c2} (fine broken curve) for (a) $Dy_{1.2}Mo_6S_8$, (b) $Tb_{1.2}Mo_6S_8$.

from the neutron-scattering data and the extrapolated $H_{c2}^0(T)$. The agreement[12] with experiment is good up to T_N. For $Dy_{1.2}Mo_6S_8$, $H_{c2}^0(T)$ is rather flat around T_N, and the results are not sensitive to the exact shape of $H_{c2}^0(T)$ [Fig. 2(a)]. But for $Tb_{1.2}Mo_6S_8$, this is not the case. The agreement of H_{c2} with experiment below T_N [Fig. 2(b)] depends on the shape chosen for $H_{c2}^0(T)$; the extrapolation which we have used appears reasonable.[1,4] Above T_N, Eq. (2a) breaks down and the theory will require modification.

For both compounds, our choice of the ratio $H_{c2}(0)/H_{c2}^0(0)$ fixes the ratio $4\gamma|J(\vec{q}_0)S_M(T=0)/B| \cong 0.3$. Now $S_M(T=0)$ is[1,2] about 2.5, B is[13] about 6×10^3 K, and, from (11), γ lies in the range 2 to 4. Hence we find $J(\vec{q}_0)$ in the range 50 to 100 K, consistent with other estimates.[1,4] [Note that Ruderman-Kittel-Kasuya-Yosida theory gives $T_N \propto J(\vec{q}_0)^2$.]

ACKNOWLEDGMENTS

One of us (J.A.) acknowledges stimulating discussions with Ø. Fischer and M. Peter, who initiated his interest in the problem. This work has been partly supported by the National Council of Research and Development of Israel, Kernforschungszentrum Karlsruhe, Germany, the Technion Vice President for Research Fund—Lawrence Deutsch Fund, and the Fund for Promotion of Research at the Technion.

[1]M. Ishikawa and Ø. Fischer, Solid State Commun. 24, 747 (1977); M. Ishikawa and J. Muller, *ibid.* 27, 761 (1978).
[2]D. E. Moncton, G. Shirane, W. Tomlinson, M. Ishikawa, and Ø. Fischer, Phys. Rev. Lett. 41, 1133 (1978); W. Tomlinson, G. Shirane, D. E. Moncton, M. Ishikawa, and Ø. Fischer, J. Appl. Phys. 50, 1981 (1979).
[3]We use the following abbreviations: AF for antiferromagnet (-ic), (-ism); SC for superconduct-or (-ing), (-ivity); and FS for Fermi surface.
[4]Ø. Fischer, Appl. Phys. 16, 1 (1978).
[5]W. Baltensperger and S. Strässler, Phys. Kondens. Mater. 1, 20 (1963).
[6]T. V. Ramakrishnan and C. M. Varma, Phys. Rev. B 24, 137 (1981).
[7]K. Machida, K. Nokora, and T. Matsubara, Phys. Rev. B 22, 2307 (1980).
[8]G. Zwicknagl and P. Fulde, Z. Phys. B 43, 23 (1981).
[9]M. J. Nass, K. Levin, and G. S. Grest, Phys. Rev. Lett. 46, 614 (1981); Phys. Rev. B 25, 4541 (1982).
[10]M. Peter, J. Ashkenazi, and M. Dacorogna, Helv. Phys. Acta 50, 267 (1977).
[11]W. L. McMillan, Phys. Rev. 167, 331 (1968).
[12]Curve A [Fig. 2(a)] deviates slightly from experiment near T_N. However, reducing T_N by 0.02 K (curve B) gives good agreement. This displacement is plausible because S_M and H_{c2} were measured on different samples, and T_c^0 was somewhat higher for the former one (Ref. 2).
[13]O. K. Anderson, W. Klose, and H. Nohl, Phys. Rev. B 17, 1209 (1978).