Simultaneous Onset of Ferromagnetism and Superconductivity

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In a few compounds, magnetic ordering and superconductivity appear to arise at the same value of a critical parameter. Assuming a model of magnetic ordering based on localized spins, this simultaneous onset may be explained as a coupling of *two* conduction electrons via *one* localized spin. This is analogous to magnetic order arising from coupling of *two* localized spins via *one* conduction electron.

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1. Introduction.—The compound UGe_2 is paramagnetic at high pressures. As the pressure is reduced, a ferromagnetic transition occurs at a certain critical value p_c [1–3]. The transition temperature rises from zero at p_c to a saturation value at low pressure. At the same pressure p_c, a superconducting transition occurs, with transition temperature initially rising with decreasing pressure, but eventually declining to zero at a considerably lower pressure. Thus, ferromagnetism and superconductivity not only coexist over a certain pressure range, but appear to arise together. It is proposed that the exchange interaction of individual spins assumed localized on the uranium ions with pairs of conduction electrons is responsible for this effect. This problem has been studied with some success in a series of papers [4,5] in terms of diagrammatic many-body theory, on the basis of the itinerant electron model. These authors make the following argument: When magnetic ordering first arises, it is weak, and the attendant spin wave spectrum is so "flat" and weak, that it cannot result in mediating an attractive electron pair interaction. Spin waves do not fully describe the dynamic degrees of freedom of the spin system. Otherwise stated, the corresponding transverse susceptibility is ineffective. However, the longitudinal susceptibility along the molecular field is still large. It evidently describes the part of the local spin density not tied up in collective motion. In this paper, we adopt this viewpoint, but discuss it in terms of localized spins, rather than an itinerant model. We assume that the magnetic transition occurs by some mechanism not described here. Once it occurs, a small average polarization of each local spin in a net mean exchange field arises, which facilitates binding of electron pairs.

2. Cooper pairing.—Consider just two electrons entering with equal and opposite momenta above the Fermi sea. Suppose that the electrons interact with a particular local spin $\vec{S}$ at lattice position $\vec{R}$ by a contact interaction

$$J\omega\vec{S}[\vec{\sigma}_1\delta(r_c + \vec{\rho}/2 - \vec{R}) + \vec{\sigma}_2\delta(r_c - \vec{\rho}/2 - \vec{R})], \tag{1}$$

where ω is an atomic volume, J the exchange energy, r_c the center of mass, and ρ the relative coordinate of the two electrons. For any given state of the local spin $\vec{S}$, in the absence of the interaction (1), the singlet state of the pair, normalized in volume Ω,

$$\psi_k^S = \frac{1}{\sqrt{\Omega}}\cos(\vec{k}\cdot\vec{\rho})[\alpha(1)\beta(2) - \alpha(2)\beta(1)]$$

is degenerate with all three triplet states

$$\psi_k^T = \frac{1}{\sqrt{\Omega}}\sin(\vec{k}\cdot\vec{\rho})[\alpha(1)\alpha(2), \alpha(1)\beta(2) + \alpha(2)\beta(1), \beta(1)\beta(2)]$$

of the pair, and the degeneracy is removed only to second order in the interaction (1). (The spatial factors ensure overall antisymmetry.) $\vec{k}$ is the relative momentum of the pair. Both wave functions are independent of r_c, which affects only their normalization. An average exchange field will lift the degeneracy partly, but the singlet and the triplet with the zero z component of spin remain degenerate in lowest order. Resolution of the degeneracy is conveniently discussed in terms of second quantization, for which

$$\psi_k^S = (c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger - c_{k\downarrow}^\dagger c_{-k\uparrow}^\dagger)|F\rangle/\sqrt{2} \tag{2}$$

and ψ_k^T = one of the three states,

$$\{c_{k\uparrow}^\dagger c_{-k\uparrow}^\dagger, (c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger + c_{k\downarrow}^\dagger c_{-k\uparrow}^\dagger)/\sqrt{2}, c_{k\downarrow}^\dagger c_{-k\downarrow}^\dagger\}|F\rangle, \tag{3}$$

as required in the following; $|F\rangle$ denotes the Fermi state. (Note that in superconductivity theory, the pairing $c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger$ is usually described as singlet pairing; in fact, it is a superposition of singlet and triplet with a zero z-spin component. This is normally of no great consequence, except in magnetic problems of the kind discussed here.) In this notation, the form of the interaction (1) is

$$V = J\frac{\omega}{\Omega}\sum_R \sum_{k,k';\alpha,\beta} \vec{S}\cdot\vec{\sigma}_{\alpha\beta}c_{k\alpha}^\dagger c_{k'\beta}e^{i(\vec{k}-\vec{k}')\cdot\vec{R}} \tag{4}$$

with plane wave states normalized to total volume Ω. In terms of projection operators P^S, P^T of the singlet and triplet manifolds and the states $\Psi^S = P^S\Psi$, $\Psi^T = P^S\Psi$, the Schrödinger equation reads

$$(P^S H_0 - E)\Psi^S + P^S V\Psi^T = 0, \tag{5}$$

$$P^T V\Psi^S + (P^T H_0 - E)\Psi^T = 0. \tag{6}$$

Eliminating Ψ^T gives

$$(P^S H_0 - E)\Psi^S - P^S V P^T \frac{1}{E - P^T H_0 P^T} P^T V \Psi^S = 0\,, \tag{7}$$

Ψ^S is expanded in a series of pair states with zero net momentum such as (2). However, the intermediate triplet states in Eq. (7) will have all possible net momenta, since V does not conserve momentum. We write $\Psi^S = \sum_k a_k \psi_k^S$. With g denoting the propagator, a_k satisfies

$$(2\epsilon_k - E)a_k = \sum_{k'} (\psi_k^S, VgV\psi_{k'}^S)a_{k'}\,, \tag{8}$$

and VgV is evaluated in stages. We have, keeping only "contracted" terms,

$$V\psi_{k'}^S = J\frac{\omega}{\Omega}\sum_i\sum_l [S_i^+(c_{l\downarrow}^\dagger c_{-k'\downarrow}^\dagger e^{i(\vec{l}-\vec{k}')\cdot\vec{R}_i} + c_{-l\downarrow}^\dagger c_{k'\downarrow}^\dagger e^{-i(\vec{l}-\vec{k}')\cdot\vec{R}_i}) - S_i^-(c_{-l\uparrow}^\dagger c_{k'\uparrow}^\dagger e^{-i(\vec{l}-\vec{k}')\cdot\vec{R}_i} + c_{l\uparrow}^\dagger c_{-k'\uparrow}^\dagger e^{i(\vec{l}-\vec{k}')\cdot\vec{R}_i}) + \tfrac{1}{2}S_i^z(c_{l\uparrow}^\dagger c_{-k'\downarrow}^\dagger + c_{l\downarrow}^\dagger c_{-k'\uparrow}^\dagger)e^{i(\vec{l}-\vec{k}')\cdot\vec{R}_i}]|F\rangle\,. \tag{9}$$

Next, the propagator g, applied to this expression, multiplies the first, second, and third terms in this sum by $g_\downarrow = (E - \epsilon_l - \epsilon_k - 2h_{\rm ex})^{-1}$, $g_\uparrow = (E - \epsilon_l - \epsilon_k + 2h_{\rm ex})^{-1}$, and $g_0 = (E - \epsilon_l - \epsilon_k)^{-1}$, respectively. $P^S H_0 \psi_k^S$ is simply $2\epsilon_k = \hbar^2/(mk^2) - 2\epsilon_f$. On the other hand, for the intermediate triplet states $\psi_{l,k'}^T = c_{l\uparrow}^\dagger c_{-k'\uparrow}^\dagger$, etc., carrying net momentum $\vec{l} - \vec{k}'$, we have $P^T H_0 \psi_{l,k}^T = (\epsilon_k^T + \epsilon_l^T)\psi_{l,k}^T$, with $(\epsilon_k^T + \epsilon_l^T) = \epsilon_k + \epsilon_l - h_{\rm ex} \times$ (z component of the triplet spin of $\Psi_{l,k}^T$). Here, $h_{\rm ex}$ is the (weak) average exchange field, evidently proportional to J and to the mean value of S_z, considered small just below the transition. In the formation of $VgV\psi_{k'}^S$, only a few terms are found that survive the following criteria: (i) correlation between different localized spins is neglected in accordance with the neglect of spin wave modes, and (ii) terms with exponentials that still depend on k'. These eventually require $k' = k$ in the formation of the final matrix element $(\psi_k^S, VgV\psi_{k'}^S)$, and contribute negligibly to the sum in (8), and (iii) terms such as S^+S^z, etc. that do not have a finite average in the weakly ferromagnetic state. The surviving terms in $VgV\psi_{k'}^S$ are proportional to

$$\sum_{i,l,l'} g_\uparrow(l,k')S_i^+S_i^- c_{l\downarrow}^\dagger c_{-l'\uparrow}^\dagger e^{i(\vec{l}-\vec{l}')\cdot\vec{R}_i} - g_\downarrow(l,k')S_i^-S_i^+ c_{l\uparrow}^\dagger c_{-l'\downarrow}^\dagger e^{i(\vec{l}-\vec{l}')\cdot\vec{R}}.$$

Since $\psi_k^{S\dagger} = \langle F|(c_{-k\downarrow}^\dagger c_{k\uparrow}^\dagger - c_{-k\uparrow}c_{k\downarrow})$, this finally gives

$$(\psi_k^S, VgV\psi_{k'}^S) = -J^2\frac{\omega^2 N}{\Omega^2}[g_\uparrow(k,k')S^+S^- + g_\downarrow(k,k')S^-S^+] \tag{10}$$

with N the number of lattice sites. The g's may be written

$$g_{\uparrow,\downarrow} = \frac{E - (\epsilon_k + \epsilon_{k'})^+_-2h_{\rm ex}}{[E - (\epsilon_k + \epsilon_{k'})]^2 - 4h_{\rm ex}^2}\,.$$

In the sense of perturbation theory, $E - (\epsilon_k + \epsilon_{k'})$ may be neglected in the numerator, since E is small and close to the Fermi surface $(\epsilon_k + \epsilon_{k'})$ is small. Hence, and since $(S^+, S^-) = 2S^z$, Eq. (10) becomes, using $\omega/\Omega \approx 1/N$,

$$(\psi_k^S, VgV\psi_{k'}^S) = -J^2\frac{4\omega^2 N}{\Omega^2}\frac{h_{\rm ex}S^z}{[E - (\epsilon_k + \epsilon_{k'})]^2 - 4h_{\rm ex}^2} \tag{11}$$

$$= \frac{1}{N}W_{kk'}\,, \tag{12}$$

say, whose average is finite if $\langle S^z\rangle$ is finite. For the simplest mean field models there should be a thermodynamic relation between that average and $h_{\rm ex}$, but this is doubtful in the present case of UGe_2 with its pressure-driven magnetic transition. [Apart from the E dependence, expression (11) has the form of the pair interaction mediated, to lowest order, by Einstein phonons. In strict perturbation theory, E should be equated to zero. For the case of $S = 1/2$, the S operators can be replaced by an extra fermion, and then it should become possible to sum the entire ladder diagram [6]. We do not pursue this matter here.] Equation (8) may be written

$$a_k = \frac{1}{E - 2\epsilon_k}\frac{1}{N}\sum_{k'}(-W_{k,k'})a_{k'}\,. \tag{13}$$

With E equated to zero inside W, and the ϵ close to the Fermi surface, W is positive for positive h, and weakly dependent on k, k'. Treating it as constant, Eq. (13) then gives the usual relation of the BCS theory in the weak coupling limit

$$1 = (-W)\frac{1}{N}\sum\frac{1}{E - 2\epsilon_k}\,,$$

which gives one bound state at negative E. This corresponds to the "bonding orbital" of the resolved degeneracy. The antibonding orbital can be found by writing $\Psi = \sum a_k\psi_k^S + b_k\psi_k^T$, and solving (5) and (6) together. However, the result is obvious from the diagonal sum rule: since the first order energies are equal (and taken equal to zero), the sums of the bonding and antibonding energies must be zero. This means that the antibonding energy value is submerged in the continuum.

3. Mean field theory.—We treat the many electron case as a scattering problem, with a T matrix defined by the relation $T\Phi_0 = V\Phi$, and satisfying the equation

$$T = V + V\frac{1}{E - H_0}T\,,$$

where Φ_0 and Φ are states of the Hamiltonians without and with interaction, respectively. The effective Hamiltonian is

$H = H_0 + T$, which, to second order in V, gives

$$H = H_0 + V + VgV, \tag{14}$$

with $g = 1/(E - H_0)$, as before. If this is written in a second quantized form, and the creation operators are written to the left of the annihilation operators, the propagator appears in the forms $g_\uparrow$, $g_\downarrow$, g_0, exactly as in Section 2. Retaining only terms with S^+ and S^- (for the same reason as in Section 2), VgV is found to be proportional to

$$\sum_{k,k'} g_\uparrow S^+ S^- c^\dagger_{k\downarrow} c^\dagger_{-k\uparrow} c_{-k'\uparrow} c_{k'\downarrow} + g_\downarrow S^- S^+ c^\dagger_{k\uparrow} c^\dagger_{-k\downarrow} c_{-k'\downarrow} c_{k'\uparrow}. \tag{15}$$

Processing the $g_{\uparrow,\downarrow}$ in the same way as before and relabeling the terms in the sum where needed, gives for the interaction

$$(-W) \sum_{k,k'} c^\dagger_{k\downarrow} c^\dagger_{-k\uparrow} c_{-k'\uparrow} c_{k'\downarrow}$$

as in BCS theory with s-wave symmetry of the order parameter.

4. Magnetic field dependence.—The exchange field h_{ex} used here acts on the electron spins only and, unless spin-orbit coupling becomes important, does not affect the electron orbits. Thus the Meissner effect should occur just as in the usual superconductors. However, one must still ask why the electrons should forego the gain of Zeeman energy in favor of antiparallel pairing (in this theory, they are polarized only in the intermediate states as described by the g's). In the present notation, the Zeeman energy of a pair is simply $2h_{\mathrm{ex}}$ and will have to be less than the energy gap, if the superconducting state is to prevail. Thus, using the usual formula for the gap (with $2h_{\mathrm{ex}}$ replacing the Debye energy ω_D), we must have

$$2h_{\mathrm{ex}} < \frac{2\langle h_{\mathrm{ex}} S^z \rangle}{\sinh \frac{1}{N(0)W}}$$

with $N(0)$ denoting the state density at ϵ_f. This is obviously impossible in the weak coupling limit. For stronger coupling, it will be satisfied if

$$\frac{1}{N(0)W} \lesssim \ln(1 + \sqrt{2}), \tag{16}$$

but then the theory may have to be carried to higher order. If, nevertheless, this inequality is taken seriously, and in Eq. (13), the k, k' dependence of W is neglected altogether, with $(E - \epsilon_k - \epsilon_{k'})$ equated to zero, then

$$W = J^2 \frac{\langle S^z \rangle}{h_{\mathrm{ex}}}.$$

But h_{ex} is proportional to $J\langle S^z \rangle$. Therefore $W \approx J$, so that a high density of states near the Fermi level and a large exchange coupling favor the superconductivity. Reference [1] provides strong evidence that UGe_2 is a heavy fermion superconductor with effective mass 50 times the free mass, consistent with the conclusions of this paper.

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[6] In fact, A. A. Abrikosov (private communication) has carried out such a summation, with similar results.