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LETTER TO THE EDITOR

d-wave superconductivity and antiferromagnetism in UPt_3

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Abstract. A general classification of the solutions of the Ginzburg–Landau equation for UPt_3 is presented. The free energy includes an E_{1g} superconducting order parameter, an antiferromagnetic order parameter and the coupling between them. There are four distinct regions of the parameter space. In one of these the coupling acts as a perturbation and the split transition in UPt_3 is essentially a superconducting transition. The strong coupling region gives a mixed character to the transition. Phenomenologically it appears that the first alternative is most likely to be the correct one.

Considerable interest has attached to UPt_3 since the discovery of superconductivity in this heavy-fermion material. It soon became evident that low-temperature thermodynamic properties were anomalous, and suggestive of lines of nodes in the superconducting gap function [1]. A leading candidate for the superconducting state is a two component spin singlet order parameter (OP) transforming according to the E_{1g} representation of the point group [2, 3]. This is written as $\boldsymbol{\psi} = (\psi_x, \psi_y)$. Neutron scattering experiments [4] have made it clear that antiferromagnetism is also present below $T_N = 5$ K, and that it couples to the superconductivity. It is therefore important to have a general form of the Ginzburg–Landau free energy for the coupled system, and to completely classify the solutions. Most importantly, it is necessary to understand all the possibilities for the split transition occurring at about $T_c \approx 0.5$ K.

The free energy density at zero external field may be written as $F = F_S + F_M + F_{SM}$, where

$$\begin{aligned} F_S &= \alpha_S(T - T_S)\boldsymbol{\psi} \cdot \boldsymbol{\psi}^* + \beta_1(\boldsymbol{\psi} \cdot \boldsymbol{\psi}^*)^2 + \beta_2|\boldsymbol{\psi} \cdot \boldsymbol{\psi}|^2 \\ F_M &= \alpha_M(T - T_M)M^2 - a(\boldsymbol{Q} \cdot \boldsymbol{M})^2 + b_M M^4 \\ F_{SM} &= b|\boldsymbol{M} \cdot \boldsymbol{\psi}|^2 + cM^2\boldsymbol{\psi} \cdot \boldsymbol{\psi}^* + dM^2|\hat{\boldsymbol{Q}} \cdot \boldsymbol{\psi}|^2. \end{aligned} \tag{1}$$

This free energy is correct to the fourth order in the space of $\boldsymbol{M}$ and $\boldsymbol{\psi}$. $\boldsymbol{M} = (M_x, M_y)$ is taken as a two-dimensional vector confined to the basal plane since it appears experimentally that spin–orbit coupling enforces this constraint. The $\hat{\boldsymbol{Q}}$ vector of the magnetism is taken as fixed in the x direction, in accordance with the experiment. For definiteness we take b, c, d all positive. Changing the signs makes no essential difference in what follows. Various pieces of this free energy have been written down previously [5–7].

As the temperature is reduced, the first transition takes place at $T_N = 5\,\text{K} = T_M + a/\alpha_M$. (We take $a > 0$.) Below this temperature we find $M_x^2 = \alpha_M(T_N - T)/2b_M$. This is the magnetic state. The next transition takes place at $T_{c1} = T_s - (b + c) M_x^2(T_{c1})/\alpha_S$, where experimentally $T_{c1} \approx 0.5$ K so $M_x^2(T_{c1}) \approx \alpha_m T_N/2b_M$. Below T_{c1} we find $|\psi_y|^2 \approx \alpha_s\,(T_{c1} - T)/2(\beta_1 + \beta_2)$. This is the superconducting transition. Experimentally, there is a third transition (the 'lower transition') at $T_{c2} \approx T_{c1} - 0.05$ K. So the crucial question about equation (1) is: what happens as T is further reduced? To understand this, it is convenient to rewrite F for $T \leqslant T_{c1}$ as

$$F_S(\psi_x) = [\alpha_S(T - T_S) + 2\beta'_S - 4\beta'_2 \sin^2\varphi]|\psi_x|^2 + (\beta_1 + \beta_2)|\psi_x|^4$$

$$F_M(M_y) = \alpha_M(T - T_M)M_y^2 + b_M M_y^4 + 2b'_M M_y^2 \tag{2}$$

$$F_{SM}(\psi_x, M_y) = (b' + c' + d')|\psi_x|^2 + c''M_y^2 + (c + d)|\psi_x|^2 M_y^2 + 2b''|\psi_x| M_y \cos\varphi.$$

Here φ is the phase angle between ψ_x and ψ_y, i.e., $\boldsymbol{\psi} = (|\psi_x| e^{-i\varphi}, |\psi_y|)$. Furthermore we have defined $\beta'_S = (\beta_1 + \beta_2)|\psi_y|^2$, $\beta'_2 = \beta_2|\psi_y|^2$, $b'_M = b_M M_x^2$, $b' = bM_x^2$, $c' = cM_x^2$, $d' = dM_x^2$, $c'' = c|\psi_y|^2$ and $b'' = b|\psi_y|M_x$. The point is that in a small range of temperature near T_{c2} we can now regard ψ_y and M_x as effective fields acting on ψ_x and M_y. The notation may be further simplified if we write

$$F = \varepsilon_S|\psi_x|^2 + \varepsilon_M M_y^2 + 2t|\psi_x|M_y + O(\psi_x, M_y)^4 \tag{3}$$

where $\varepsilon_S(T) = \alpha_S(T - T_S) + 2\beta'_S - 4\beta'_2 \sin^2\varphi + b' + c' + d'$, $\varepsilon_M(T) = \alpha_M(T - T_M) + b'_M + c''$ and $t = b'' \cos\varphi$. Equations (2) and (3) are the same as those written down in [7], except for inessential details.

The quadratic form (3) may now be diagonalised, and the final form for F is

$$F = \lambda_-\Phi_-^2 + \lambda_+\Phi_+^2 + \beta_-\Phi_-^4 + \beta_+\Phi_+^4 + \beta_{+-}\Phi_-^2\Phi_+^2 + \beta_+^1\Phi_+^3\Phi_- + \beta_-^1\Phi_-^3\Phi_+. \tag{4}$$

$\Phi_\pm$ are linear combinations (to be determined) of $|\psi_x|$ and M_y, and $\lambda_\pm = \frac{1}{2}(\varepsilon_S + \varepsilon_M) \pm \frac{1}{2}[(\varepsilon_S - \varepsilon_M)^2 + 4t^2]^{1/2}$. The next transition occurs [7] when $\lambda_-(T) = 0$ or

$$\varepsilon_S\varepsilon_M = t^2. \tag{5}$$

This defines T_{c2}. Below this temperature $\Phi_-^2 \sim (T_{c2} - T)$ and

$$F(\psi_x, M_y) = -\tfrac{1}{4}\lambda_-^2(\varphi)/B(\varphi). \tag{6}$$

$B(\varphi)$ is a complicated function of the λs and βs. We now wish to classify all the solutions of equations (5) and (6). There are four regimes of parameter space.

(i) *Lower transition is magnetic; weak coupling.* This part of the parameter space is characterised by the inequalities

$$|\varepsilon_S(T = 0)| \gg t(T = 0) \qquad |\varepsilon_M(T = 0)| \gg t(T = 0). \tag{7}$$

That is, the energy scales for both superconductivity and magnetism are much larger than the coupling energy between the two. In addition $\varepsilon_M(T)$ crosses zero near T_{c2}. Then the transition is essentially magnetic, and the critical temperature is given by

$$T_{c2} = T_N - (b'_M + c'')/\alpha_M + t^2/\varepsilon_S\alpha_M \tag{8}$$

and the order parameter $\Phi_- \sim M_y$.

(ii) *Lower transition is superconducting; weak coupling.* Again we have the inequalities $|\varepsilon_S(T=0)| \gg t(T=0)$ and $|\varepsilon_M(T=0)| \gg t(T=0)$, but now $\varepsilon_S(T)$ crosses zero near T_{c2}. Then we find

$$T_{c2} = T_{c1} - (2\beta_S' + b' + c' + d')/\alpha_S + t^2/\varepsilon_M\alpha_S \qquad \Phi_- \sim |\psi_x|. \tag{9}$$

(iii) *Strong coupling.* Here the opposite inequality holds:

$$t^2(T=0) > |\varepsilon_S(T=0)\varepsilon_M(T=0)|. \tag{10}$$

Note that near T_{c1}, we have $t^2 = \tilde{t}^2(T_{c1} - T)$, where $\tilde{t}^2 = b^2 M_x^2 \alpha_S \cos^2\varphi/2(\beta_1 + \beta_2)$. Therefore $t^2(T_{c2}) \ll t^2(T=0)$ and we have a solution of (5) in spite of the inequality (10). The critical temperature is given by

$$T_{c2} = T_{c1} - \varepsilon_S\varepsilon_M/\tilde{t}^2 \tag{11}$$

and Φ_- is a mixture of $|\psi_x|$ and M_y.

(iv) *No lower transition.* Equation (5) may be rewritten as a quadratic equation in T. If all solutions have $T < 0$, then there is no further transition. The criterion which determines this is algebraically complicated and so we do not give it explicitly here. Physically, the conditions are easily understood. They are (*a*) $T_S < 2\beta_S/\alpha_S$; (*b*) $T_M < b_M'/\alpha_M$; (c) $4b''^2/\alpha_S\alpha_M < 2\beta_S' - \alpha_S T_S + b_M' - \alpha_M T_M$. These merely say that the underlying critical temperatures for superconductivity and magnetism are small enough that non-linear couplings do not induce any transitions. This part of parameter space is clearly ruled out by experiment and therefore is not discussed further.

The above four solutions are limiting cases which give the possible extremes of behaviour of the system. Intermediate cases between these solutions are in principle also possible, but do not seem to be realised experimentally, with one possible exception to be mentioned below. Intermediate regimes would give a lower critical temperature T_{c2} which is not related to T_{c1} and therefore the smallness of the splitting would be accidental. This is also true of case (i) above and therefore this solution can also be eliminated from consideration.

This leaves only cases (ii) and (iii) as candidate solutions to be investigated in more detail. One issue is the value of φ. If $e^{i\varphi}$ is complex, then the superconducting state breaks time reversal symmetry with a number of interesting consequences [8]. The free energy (6) is a function of φ, and minimisation of this expression for cases (ii) and (iii) will give the equilibrium value of φ. For case (ii), at T_{c2} we have $|\varepsilon_M| > |\varepsilon_S|$, $\varepsilon_M > 0$, $\varepsilon_S < 0$, and $\lambda_- \approx \varepsilon_S - t^2/\varepsilon_M = 4\beta_2' \cos^2\varphi - 4b''^2 \cos^2\varphi/\varepsilon_M + \text{constant}$. Maximising λ_- alone taking into account the inequality (7) gives $\varphi = \pm\pi/2$ as long as $\beta_2 > 0$. (Note β_2' has the same sign as β_2.) $\beta_2 > 0$ is the weak coupling result [9]. For case (iii), $\lambda_- \approx -|t|$. This is maximised by $\varphi = 0$ or $\varphi = \pi$. Hence case (ii) leads to a maximally complex superconducting state, while case (iii) gives a real state [7]. Also very important is that in case (ii) the equilibrium value of t is zero, i.e. there is no coupling between $|\psi_x|$ and M_y, so $\Phi_- = |\psi_x|$. Hence $M_y = 0$ at all temperatures. In case (iii) the coupling t is maximised and $|\psi_x|$ and M_y appear simultaneously, as already mentioned.

Cases (ii) and (iii) are limiting cases which can be handled analytically. In general, the original free energy (1) must be minimised numerically to investigate intermediate cases. When this is done, the result is qualitatively the same as is given by the minimisation of (6): cases (ii) and (iii) are stable minima separated by a boundary in parameter space of first-order transitions. On this boundary the free energy is independent of φ. Because of this degeneracy, higher-order terms not included in (1) would broaden the boundary into a finite region (but most likely small) where $0 < |\varphi| < \pi/2$ and $M_y \neq 0$. We call this case (ii*a*). Even though its occurrence is *a priori* improbable it is included for completeness.

This completes the analysis of the possible equilibrium solutions of (1). Cases (ii) and (iii) are well known (see [3–5, 10], and [7] respectively). Only case (ii*a*) is new. The point of the present work is to establish that these are the only three theoretical candidates for the state of UPt_3 and to compare their properties.

The most important phenomenological distinction between cases (ii) and (iii) is the node structure of the energy gap function $|\Delta(\boldsymbol{k})|$. If we choose basis functions k_zk_x and k_zk_y for the E_{1g} representation, then $|\Delta(\boldsymbol{k}) = |\psi_xk_zk_x + \psi_yk_zk_y|$. For case (ii), we find $|\Delta(\boldsymbol{k})| = |\psi_x||k_z|(k_x^2 + k_y^2)^{1/2}$, which has zeros in the basal plane $k_z = 0$ and on the z axis $k_x = k_y = 0$. The intersection of these sets with the Fermi surface gives point nodes and 'horizontal' lines of nodes. (Note that the translation group symmetry will also require a line of nodes at $k_z = \pi/c$, where c is the lattice constant along the z direction [11].) For case (iii), $\Delta(\boldsymbol{k}) = |k_z(\psi_xk_x + \psi_yk_y)|$ and ψ_x and ψ_y may be taken to be real. This function has the horizontal line of nodes $k_z = 0$ and 'vertical' lines of nodes on the intersection of the plane $\psi_xk_x + \psi_yk_y = 0$ with the Fermi surface. More generally, a complex $\boldsymbol{\psi}$ always guarantees lines of horizontal nodes and point nodes, while a real $\boldsymbol{\psi}$ always gives both horizontal and vertical lines of nodes. Hence case (ii*a*) falls in the former category and in this respect is similar to case (ii). With respect to the existence of the transverse component of the magenetism M_y, however, it more resembles case (iii).

We now turn to a discussion of experiments in UPt_3, concentrating on low-field properties.

Specific heat. The two salient features here are: first, the closeness of the two jumps at T_{c1} and T_{c2}, and second, the comparable size of the jumps [12]. The first point has already been commented upon: it eliminates cases 1 and 4 but does not distinguish between cases (ii) and (iii). The second observation, however, clearly does distinguish between (ii) and (iii). The specific heat jump at T_{c2} is given by $\Delta C_v(T_{c2})/T_{c2} = -\partial^2 F(\psi_x, M_y)/\partial T^2|_{T_{c2}}$, and $F(\psi_x, M_y)$ is written out in (6). The jump for case (ii) has been calculated previously [5, 6]: $\Delta C_v(T_{c2})/T_{c2} = \alpha_S^2/2\beta_1$. For case (iii) the result is $\Delta C_v(T_{c2})/T_{c2} = \tilde{t}^2/2B$. The specific heat jump at the upper superconducting transition (for both cases) is $\Delta C_v(T_{c1})/T_{c1} = \alpha_S^2/2(\beta_1 + \beta_2)$. Experimentally the two jumps are certainly comparable in magnitude. This would argue for case (ii) since β_1 and β_2 would not be expected to be very different, at least in weak coupling theory. There is no particular relation between $\tilde{t}$ and α_S, however, so in case (iii) there is no reason to expect similar-sized jumps. The same reasoning would tend to rule out case (ii*a*). It is important to note that the split transition was *predicted* theoretically on the assumption of case (ii) [10].

Ultrasonic attenuation. Here the most important result is the polarisation dependence of transverse ultrasound propagating in the basal plane. It was observed that the absorption is considerably stronger for a polarisation vector lying in the plane than for polarisation perpendicular to the plane [13]. This anisotropy would strongly suggest that there exist horizontal lines of nodes but no vertical lines of nodes. This would agree for case (ii), which has this node structure.

Electromagnetic response. The response of UPt_3 to electromagnetic radiation at 10 MHz is also anisotropic [14]. Calculations indicate that this anisotropy is also indicative of a state whose line nodes lie only in the basal plane, with the line probably broadened to a strip by resonant impurity scattering [15]. This would again be an indication that case (ii) or (ii*a*) is realised in UPt_3.

Lower critical field. Recent measurements of the lower critical field for $\boldsymbol{H}$ in the basal plane have been interpreted as being in confirmation of case (ii) [5, 16]. dH_{c1}/dT increases by roughly a factor of two as T is lowered through T_{c2}. Since H_{c1} is proportional to the superfluid density, this is a strong suggestion that the transitions at T_{c1} and T_{c2} have essentially the same character in the sense that $|\psi_y|^2 \sim (T_{c1} - T)\alpha_S/(\beta_1 + \beta_2)$ and $|\psi_x|^2 \sim (T_{c2} - T)\alpha_S/\beta_1$. Thus the conclusion from these H_{c1} measurements is consistent with the specific heat measurements and points to case (ii).

On the other hand, $H_{c1}(T)$ for $\boldsymbol{H}$ along the c axis shows no such kink at T_{c2}. However, one should note that, for zero-field-cooled samples, case (ii) does not make a simple prediction. There will be two equivalent domains for this case, corresponding to (1, i) and (1, −i) states. These states have different H_{c1} values [5]. If the magnetisation is measured for $T < T_{c2}$, then $M(H)$ would show a two-kink structure rather than the conventional single kink. (This last statement assumes that full equilibrium is reached, which is doubtful in practice because of flux pinning.) There appears to be some evidence of such a two-kink structure at 300 mK [14].

Case (iii) does not have such a domain structure and would therefore predict similar behaviour for $H_{c1}(T)$ for the two directions of $\boldsymbol{H}$, presumably with only a small kink at T_{c2}.

Neutron scattering. Experiments have recently been done which measure the intensity of a single magnetic Bragg peak as a function of field and temperature [17]. At zero field, this intensity decreases by about 5% as the temperature is lowered from T_{c1} to zero, with a kink occurring near T_{c1} or T_{c2}. This may be attributed to (*a*) a decrease in $|\boldsymbol{M}|$ or (*b*) a rotation of $\boldsymbol{M}$ with $|\boldsymbol{M}|$ remaining roughly constant. Case (*a*) would be consistent with case (ii) if the coefficient c were large, i.e. when there is strong competition between the *magnitudes* of $\boldsymbol{\psi}$ and $\boldsymbol{M}$ but no rotation of $\boldsymbol{M}$. Case (iii) is consistent with either (*a*) or (*b*), as is case (ii*a*). Further experimentation is necessary to decide between (*a*) and (*b*).

We have so far concentrated on low-field properties. However, further information can be obtained by examining the phase diagram in the entire H–T plane. According to [7], case (iii) leads to the conclusion that there are three distinct superconducting phases (at least when $\boldsymbol{H}$ is in the a–b plane), while case (ii) is consistent with either two or three such phases. The recent specific heat [18] and neutron scattering [17] experiments suggest that the zero-field transition at T_{c2} behaves similarly to the low-temperature transition at $H \approx H_{c2}/2$. This suggests that only two phases are present, since in this case there would be a single phase boundary reaching from the point $H = 0$, $T = T_{c2}$ to the $T = 0$ axis.

In summary, there appears to be strong evidence for the node structure of cases (ii) or (ii*a*). Additional support for case (ii) in particular is the similar appearance of the transitions at T_{c1} and T_{c2} in specific heat and lower critical field measurements. If there is a rotation of the magnetisation vector at T_{c2}, however, this is not consistent with (ii), but only with (ii*a*) or (iii).

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