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Antiferromagnetism and superconductivity in UPt_3

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Abstract. The short-ranged antiferromagnetism recently seen in UPt_3 is proved incompatible with two-dimensional (2D) order parameter models that take the antiferromagnetism as a symmetry-breaking field. To adjust to the local moment direction, the order parameter twists over very long length scales as per the Imry–Ma argument. A variational solution to the Ginzburg–Landau equations is used to study the nature of the short-range superconducting order. Although there are still two transitions, the lower one is of first order—in contradiction to experiments. It is shown that the latent heat predicted by the 2D models at the lower transition is too large not to have been seen. A simple periodic model is numerically studied to show that the second transition cannot be a crossover either.

1. Introduction

The purpose of this paper is to discuss the implications of recent experiments [1] on antiferromagnetic (AFM) order in UPt_3 for the superconducting state. It is found that (i) a very weak AFM that sets in at 5 K is multi-domain in structure with a domain size of about 300 Å, and (ii) despite a magnetic moment per U atom of only 0.02 μ_B, the domains can neither be moved nor oriented by magnetic fields up to 3.2 T. We shall show that these findings pose great difficulty for certain widely studied models [2, 3] in which the superconducting order parameter transforms as one of the two-dimensional (2D) representations of the relevant point group, D'_{6h}, and in which the AFM (or even less plausibly, a structural modulation) provides a symmetry-breaking field that splits the superconducting transition into two just as an applied magnetic field splits the A transition in superfluid ^{3}He. We believe that these data make it overwhelmingly likely that superconducting UPt_3 is described by two order parameters belonging to independent representations [4, 5], although the precise representations involved are still debatable.

Two main experimental facts with which any theory of UPt_3 must agree are as follows.

(A) There are two superconducting transitions at T_{c+} and T_{c-} in zero field [6]; $T_{c+} \simeq 550$ mK, and $\Delta T_c = T_{c+} - T_{c-} \simeq 50$ mK. Both transitions appear to be of second order, with no observable latent heat.

(B) For basal-plane magnetic fields, $\boldsymbol{H} \perp \boldsymbol{c}$, the phase diagram is isotropic [7]; i.e., it is the same for all directions of the field‡.

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‡ We shall ignore an anisotropy of 4–5% [8] as it is too small to affect our arguments.

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We deliberately do not exploit other facts that also bear on the order parameter symmetry in this paper. Chief among these is that for $\boldsymbol{H} \perp \boldsymbol{c}$, there is a tetracritical point in the magnetic field–temperature (H–T) plane where four second-order lines meet [9]. The original 2D models [2] are incompatible with a tetracritical point unless $\boldsymbol{H} \perp \boldsymbol{c}$ [4, 10], which also argues against them, as the observed phase diagrams for $\boldsymbol{H} \parallel \boldsymbol{c}$ and $\boldsymbol{H}$ at 45° to $\boldsymbol{c}$ are similar to those for $\boldsymbol{H} \perp \boldsymbol{c}$ [9, 11], although the tetracritical points in these cases may only be apparent ones due to limited resolution of the phase boundaries [5, 12]. Secondly, a specific E_{2u} model *is* compatible with a tetracritical point for all field orientations [3]. It is therefore useful to have independent arguments that constrain the order parameter without relying on the presence or absence of tetracritical points.

With this motivation, we analyse in this paper the Ginzburg–Landau (GL) theory for 2D models [2, 3] when the AFM symmetry-breaking field is allowed to vary randomly in direction over a length scale $a = 300$ Å†. Since the problem involves an order parameter with a continuous symmetry in the presence of a random field, the Imry–Ma argument [14] implies that the long-ranged average of the order parameter must itself vanish. Other forms of order are not ruled out, however, and a balance of gradient and field orientation energies shows [15] that there are two transitions, the upper one of second order, and the lower one of first order. If one is only interested in global properties of the phase diagram and the universality class of the transitions, there is nothing more to be said. We are more interested in whether or not this scenario could describe the experimental data on UPt_3. To this end we calculate the latent heat at the lower transition assuming that the coupling to antiferromagnetism is weak. We find that this is too large to have been missed by the experiments [6] if the splitting is taken to equal the observed value. In other words, we can dismiss the possibility that the lower transition seen experimentally is of first order with an unobservably small latent heat. A second possible explanation for the data (which arises when the AFM coupling is not weak) is that the two transitions predicted by the theory are too close to be resolved, and that the lower specific heat jump is actually due to a crossover and not a true transition. We exclude this possibility too. Using a simple periodic model for the AFM domain structure, we find that the observed H_{c2}-slopes [9, 11] and the 300 Å domain size imply that a sufficiently sharp crossover must be accompanied with a ΔT_c of order 80 K, which is clearly absurd.

To orient the discussion further, let us review how the 2D models account for facts A and B when the AFM is uniform. The GL free energy f_{GL} for all these models can be written in terms of a vector $\boldsymbol{\eta} = (\eta_x, \eta_y)$ with complex components. In zero magnetic field f_{GL} is the sum of bulk, gradient‡, and symmetry-breaking field terms:

$$f_{\text{bulk}} = -(T - T_0)\eta_i^*\eta_i + \beta_1(\eta_i^*\eta_i)^2 + \beta_2|\eta_i\eta_i|^2 \tag{1.1}$$

$$f_{\text{grad}} = \kappa_1\,\partial_i\eta_j^*\,\partial_i\eta_j + \kappa_2\,\partial_i\eta_i^*\,\partial_j\eta_j + \kappa_3\,\partial_i\eta_j^*\,\partial_j\eta_i + \kappa_4\,\partial_z\eta_i^*\,\partial_z\eta_i \tag{1.2}$$

$$f_{\text{SBF}} = 2\epsilon(|\boldsymbol{n}_i\eta_i|^2 - \eta_i^*\eta_i/2). \tag{1.3}$$

† To avoid misunderstanding, we stress that we are not studying the AFM transition itself, or the reason for AFM disorder [13]. Since the AFM order sets in well above the superconducting transition, it is justified to treat it as an external field. The spatial variation of this field in no way invalidates the usual argument for using GL theory to interpret superconducting transitions. Briefly, the argument (also known as the Ginzburg criterion) is that the critical region is of width $(d/\xi_0)^4$ relative to T_c, where d is an inter-electron distance, and ξ_0 is the coherence length. For UPt_3, $d \sim 5$ Å, and $\xi_0 \sim 200$ Å, so this region is unobservably small. In particular, the question of whether the transition is sharp or smeared out by the random AFM is experimentally irrelevant.

‡ The gradient energy in the modified 2D model [3] is slightly different, as discussed below.

Here, $i, j \in \{x, y\}$, and the summation convention is used, $\boldsymbol{n}$ is the basal-plane unit vector along the direction of the magnetic moments, and ϵ, β_1, and β_2 are all positive. (If $\beta_2 < 0$, there is only one transition, and the model cannot describe UPt_3.) Suppose that $\boldsymbol{n} = \hat{\boldsymbol{y}}$ everywhere. Then the first transition occurs at $T_{c+} = T_0 + \epsilon$ to a phase with $\boldsymbol{\eta} = e^{i\phi}(1, 0)$. Ignoring the global phase ϕ, we refer to this as the real or (1, 0) phase. It breaks the rotational symmetry of the normal state. The second transition occurs at $T_{c-} = T_0 - \beta_1\epsilon/\beta_2$ to a phase with $\boldsymbol{\eta} = e^{i\phi}(1, iu)$ where u is real and grows smoothly from 0 to ± 1 as T is lowered further. This phase breaks rotational and time-reversal symmetry, and we shall refer to it as the axial or the (1, i) phase. This explains fact A. To explain fact B, it is necessary in the original 2D models [2] to assume that the symmetry-breaking field rotates in response to the applied magnetic field. A symmetry-breaking perturbation that was locked to the lattice would split the T_cs, but would not yield an isotropic basal-plane phase diagram. In particular, the tetracritical point would not exist for some $\boldsymbol{H}$-orientations†. This assumption was justified by hypothesizing [15] that the AFM anisotropy energy was so weak that the Néel vector reoriented itself to stay perpendicular to the magnetic field even for relatively small H, and this idea has even been refined [16] to explain the minute anisotropy of H_{c2} found by Keller *et al* [8].

The neutron scattering data of Lussier *et al* [1] directly refute the above hypothesis. They find that an AFM domain is of average size $a \simeq 300$ Å‡ and is associated with one of three choices for the spin-density wavevector $\boldsymbol{q}$, with $\boldsymbol{n} \parallel \boldsymbol{q}$. The scattering intensity under the peaks with different $\boldsymbol{q}$s is essentially unchanged by fields of up to 3.2 T, for either $\boldsymbol{H} \parallel \boldsymbol{q}$ or $\boldsymbol{H} \perp \boldsymbol{q}$, and in both field-cooled and zero-field-cooled experiments. In addition, application of a field causes no transfer of intensity to a scattering wavevector $\boldsymbol{Q}$ which is forbidden (because $\boldsymbol{Q} \times \boldsymbol{n} = \boldsymbol{0}$) when $H = 0$. This shows that the moments do not cant away from the $\boldsymbol{q}$-direction while preserving the wavevectors.

Why, given these data, should one further investigate the 2D models? One reason is the modified 2D model [3]. In this model, $\kappa_2 = \kappa_3 = 0$, and the AFM is assumed to split the κ_1-term into $\kappa_1^+|\partial_i\eta_\parallel|^2 + \kappa_1^-|\partial_i\eta_\perp|^2$, where $\eta_\parallel$ and $\eta_\perp$ are components of $\boldsymbol{\eta}$ parallel and perpendicular to $\boldsymbol{n}$, and $\kappa_1^+ - \kappa_1^- = \mathcal{O}(\epsilon)$. (A similar splitting is assumed for κ_4.) Note that the AFM continues to couple directly to the order parameter as described by equation (1.3). The linearized GL equations for $\eta_\parallel$ and $\eta_\perp$ then decouple even when $H \neq 0$, and an isotropic basal-plane phase diagram is recovered even if the AFM is rigidly locked to the lattice. However, the random AFM excludes this model too, as the scaling of gradient and field orientation energies with ϵ, and the overall balance between them, is unchanged. A second reason is that although the general thrust of the Imry–Ma argument [14] must continue to hold, the problem for UPt_3 is not directly equivalent to an n-component vector spin model, and we are unaware of any specific analysis for it. In particular we know of no careful comparison of the energetics of the axial phase versus a real state with a wandering axis of orientation.

The rest of the paper is organized as follows. The GL free energy is analysed in section 2. We perform a variational analysis with the aim of achieving an understanding of the nature of the short-range order at the upper transition, and also calculate the latent heat at the lower transition. The simple periodic model is analysed in section 3. A brief discussion in section 4 concludes the paper.

† It follows that a structural modulation which should, *a priori*, be insensitive to the orientation of $\boldsymbol{H}$, also cannot be a viable symmetry-breaking field.

‡ The absence of anomalies in the heat capacity [17] or NMR linewidth, Knight shift, or $1/T_1$ [18], raises doubts as to whether the AFM is static long-range order; we set these doubts aside here as they make the 2D models even more problematic.

2. Analysis of the GL free energy

We wish to minimize the GL free energy, which is still given by equations (1.1)–(1.3), but $\boldsymbol{n}(\boldsymbol{r})$ is now $\boldsymbol{r}$-dependent with a correlation length $a = 300$ Å. We first simplify the gradient energy to

$$f_{\text{grad}} = \kappa |\nabla \eta_i|^2 \tag{2.1}$$

as we are interested only in the competition between the gradient and the symmetry-breaking energies, and not in the precise slopes of the H_{c2}-lines and whether they cross or not†. It is convenient to write $\boldsymbol{n} = (\cos\theta, \sin\theta)$, and introduce the gradient energy scale $V_d = \kappa/a^2$, and circular polarization components, $\eta_{L,R} = 2^{-1/2}(\eta_x \pm \mathrm{i}\eta_y)$.

The first step is to find the critical temperature T_c, and the nature of the critical order parameter. To do this we drop the quartic terms in f_{GL}, and minimize the rest. This yields an eigenvalue problem given by the linearized GL equations:

$$-\kappa\, \nabla^2 \eta_L + \epsilon \mathrm{e}^{2\mathrm{i}\theta} \eta_R = -t\eta_L \tag{2.2}$$

$$-\kappa\, \nabla^2 \eta_R + \epsilon \mathrm{e}^{-2\mathrm{i}\theta} \eta_L = -t\eta_R \tag{2.3}$$

where $t = T_c - T_0$ is the T_c-*enhancement*. The critical solution is that with the *largest* value of t.

Two qualitatively different types of solution to equations (2.2) and (2.3) are as follows. The first, or local solution, is motivated by the answer for uniform θ [2]. Then $\boldsymbol{\eta}$ is also uniform, the gradient terms vanish, and we get $\eta_L = -\mathrm{e}^{2\mathrm{i}\theta}\eta_R$, $t = \epsilon$. If we now require $\boldsymbol{\eta} \perp \boldsymbol{n}$, i.e., $\eta_L = -\mathrm{e}^{2\mathrm{i}\theta}\eta_R$, even when θ is varying, then we pick up a gradient energy of the order of V_d, and $t_{\text{local}} = \epsilon - \alpha V_d$, where $\alpha = 2a^2\langle(\nabla\theta)^2\rangle$ is a number of order one, and $\langle\cdots\rangle$ denotes a spatial average. This solution does well only when $\epsilon \gg V_d$, i.e., when the symmetry-breaking energy is dominant. The second, or real solution, has the form $\boldsymbol{\eta} = \mathrm{e}^{\mathrm{i}\phi}\boldsymbol{m}$, $\boldsymbol{m}$ being a real unit vector varying on a length $L \gg a$. The gradient energy is clearly of order $V_d(a/L)^2$. A random-walk argument for the symmetry-breaking energy leads to the estimate $-\epsilon(a/L)^{3/2}$. Minimizing the sum of these two energies, we obtain $L \sim a(V_d/\epsilon)^2$, and $t_{\text{real}} \simeq \epsilon(\epsilon/V_d)^3$. This solution requires $V_d \gg \epsilon$ for consistency, and the T_c-enhancement is strongly reduced compared to the uniform case.

A correct estimate of the symmetry-breaking energy for the real solution is more subtle, however. Although long-range order in $\boldsymbol{\eta}$ is forbidden, it is not obvious that the estimate of the energy itself is correct, as one could try to lower it by short-range adjustment to the local value of $\boldsymbol{n}$. Indeed, we will find that though the T_c-*splitting* is governed by the ϵ^4/V_d^3 energy gain, this gain is subdominant to that due to the short-ranged order, and the *enhancement* in T_c itself is larger, of order ϵ^2/V_d.

The above results follow from a variational argument, which we present in three stages. In the first stage, let us consider the trial wave function $\eta_L = 1$, $\eta_R = \zeta \mathrm{e}^{-2\mathrm{i}\theta(\boldsymbol{r})}$, with ζ as a variational parameter. We call this the axial solution. (See the reasons below.) The optimal value of ζ is to be found by minimizing

$$W = \frac{\langle\eta|H_{GL}|\eta\rangle}{\langle\eta|\eta\rangle} \tag{2.4}$$

where H_{GL} is the linear operator, or GL 'Hamiltonian', in equation (2.2) and (2.3), $|\eta\rangle$ is the trial solution, and we use an obvious quantum mechanical notation for the averages.

† This simplification is equally justified for the modified 2D model [3]. The gradient energy cost for variations of $\boldsymbol{\eta}$ along and perpendicular to $\boldsymbol{n}$ is now slightly different, but it is, nevertheless, always positive. Any suitable average for the gradient energy scale V_d therefore suffices for the scaling arguments that follow.

A simple calculation gives

$$W = 2\frac{\epsilon\,\mathrm{Re}(\zeta) + \alpha V_d|\zeta|^2}{1+|\zeta|^2}. \tag{2.5}$$

(The quantity $\alpha = 2a^2\langle(\nabla\theta)^2\rangle$ as before.) Minimization gives a T_c-enhancement $t_{\mathrm{axial}} = -\alpha V_d + (\epsilon^2 + \alpha^2 V_d^2)^{1/2}$, which is $\approx\epsilon^2/2\alpha V_d$ for $\epsilon \ll V_d$, and $\approx\epsilon - \alpha V_d$ when $\epsilon \gg V_d$. We have thus obtained a solution that is better than both the local and the real ones.

In the second stage, we generalize the above solution by taking

$$\begin{aligned} \eta_L &= 1 \\ \eta_R(\boldsymbol{r}) &= \int \mathrm{d}^3\boldsymbol{r}'\ Q(\boldsymbol{r}-\boldsymbol{r}')\mathrm{e}^{-2\mathrm{i}\theta(\boldsymbol{r}')} \equiv h(\boldsymbol{r}) \end{aligned} \tag{2.6}$$

where $Q(\boldsymbol{r})$ is a variational kernel. We take $\mathrm{e}^{2\mathrm{i}\theta(\boldsymbol{r})}$ to be Gaussian distributed, with $\langle\mathrm{e}^{2\mathrm{i}\theta(\boldsymbol{r})}\rangle = 0$, and a correlation function $g(\boldsymbol{x}) = \langle\mathrm{e}^{2\mathrm{i}\theta(\boldsymbol{r})}\mathrm{e}^{-2\mathrm{i}\theta(\boldsymbol{r}+\boldsymbol{x})}\rangle$ decaying on a length scale a. Denoting the volume of the system by Ω, the average of the symmetry-breaking energy is found to be

$$2\,\mathrm{Re}\,\frac{\epsilon}{\Omega}\int \mathrm{d}^3r\ \eta_L^*\eta_R\mathrm{e}^{2\mathrm{i}\theta(\boldsymbol{r})} = 2\epsilon F_1[Q] \tag{2.7}$$

where $F_1[Q]$ is the functional

$$F_1[Q] = \int \frac{\mathrm{d}^3k}{(2\pi)^3}\ Q(\boldsymbol{k})g(\boldsymbol{k}) \tag{2.8}$$

where $Q(\boldsymbol{k})$ and $g(\boldsymbol{k})$ are Fourier transforms of $Q(\boldsymbol{r})$ and $g(\boldsymbol{r})$. Similarly, the average of the gradient energy equals

$$\frac{\kappa}{\Omega}\int \mathrm{d}^3r\ |\nabla\eta_R|^2 = \kappa F_2[Q] \tag{2.9}$$

where

$$F_2[Q] = \int \frac{\mathrm{d}^3k}{(2\pi)^3}\ k^2Q^2(\boldsymbol{k})g(\boldsymbol{k}). \tag{2.10}$$

Finally, the spatial average of $|\eta_R|^2$, needed for normalization, is given by

$$\frac{1}{\Omega}\int \mathrm{d}^3r\ |\eta_R|^2 = F_3[Q] \equiv \int \frac{\mathrm{d}^3k}{(2\pi)^3}\ Q^2(\boldsymbol{k})g(\boldsymbol{k}). \tag{2.11}$$

The expectation of H_{GL} is therefore given by

$$W = (2\epsilon F_1[Q] + \kappa F_2[Q])/(1 + F_3[Q]) \tag{2.12}$$

which must be minimized with respect to $Q(\boldsymbol{k})$. Setting the variation of W to zero, we obtain

$$(1+F_3)(2\epsilon\,\delta F_1 + \kappa\,\delta F_2) - (2\epsilon F_1 + \kappa F_2)\delta F_3 = 0. \tag{2.13}$$

Using $\delta F_i/\delta Q(\boldsymbol{k}) = g(\boldsymbol{k})$, $2k^2Q(\boldsymbol{k})g(\boldsymbol{k})$, and $2Q(\boldsymbol{k})g(\boldsymbol{k})$ for $i = 1$, 2, and 3, respectively, and cancelling $g(\boldsymbol{k})$ in this equation, we get

$$(1+F_3)[\epsilon + \kappa k^2 Q(\boldsymbol{k})] - (2\epsilon F_1 + \kappa F_2)Q(\boldsymbol{k}) = 0. \tag{2.14}$$

This yields the Yukawa-like form

$$Q(\boldsymbol{k}) = -\frac{\epsilon}{\kappa(k^2 + k_0^2)} \tag{2.15}$$

where k_0^2 is given self-consistently by

$$k_0^2 = -\frac{2\epsilon F_1 + \kappa F_2}{\kappa(1+F_3)} = -\frac{W_{\min}}{\kappa}. \tag{2.16}$$

In the last form, which follows by comparison with equation (2.12), $W_{\min}$ denotes the minimum value of W.

It remains to find the $F_i[Q]$ with the form (2.15) and substitute in equation (2.16) to obtain k_0 and $W_{\min}$. Since $g(x)$ decays on a length scale a, we expect $g(k)$ to decay on a scale a^{-1}. Since $g(\boldsymbol{x}=0)=1$, this decay must be faster than $1/k^3$ as $k \to \infty$; this fact will guarantee convergence of the integrals below. For F_1 we obtain

$$F_1 = -\frac{\epsilon}{\kappa}\frac{1}{2\pi^2}\int_0^\infty \frac{1}{k^2+k_0^2} g(k) k^2 \, \mathrm{d}k. \tag{2.17}$$

Now, by the second equality in equation (2.16), we expect k_0 to vanish as $\epsilon \to 0$. It is therefore valid to put k_0 to zero in the integrand above. This yields

$$F_1 \approx -c_1(\epsilon/V_d) \tag{2.18}$$

where

$$c_1 \equiv \frac{1}{2\pi^2 a^2}\int_0^\infty g(k) \, \mathrm{d}k \tag{2.19}$$

is a dimensionless number of order unity. In the same way, we get

$$F_2 = \frac{\epsilon^2}{\kappa^2}\frac{1}{2\pi^2}\int_0^\infty \frac{k^2}{(k^2+k_0^2)^2} g(k) k^2 \, \mathrm{d}k \approx c_1(\epsilon a/\kappa)^2 \tag{2.20}$$

where the last result follows from again setting $k_0 = 0$ in the integrand. Lastly, F_3 is given by

$$F_3 = \frac{\epsilon^2}{\kappa^2}\frac{1}{2\pi^2}\int_0^\infty \frac{1}{(k^2+k_0^2)^2} g(k) k^2 \, \mathrm{d}k. \tag{2.21}$$

It is impermissible to set $k_0 = 0$ now as the resulting integral is divergent. Instead, we note that the integrand is sharply peaked near $k = 0$, and use this fact to replace $g(k)$ by $g_0 \equiv g(\boldsymbol{k}=0)$. This yields

$$F_3 \approx c_2 \frac{1}{k_0 a}\left(\frac{\epsilon}{V_d}\right)^2 \tag{2.22}$$

where $c_2 = g_0/8\pi a^3$ is another dimensionless number of order unity.

We will show shortly that $F_3 \ll 1$. (This is also to be expected from the fact that $F_3 = \langle|\eta_R|^2\rangle$.) Assuming this to be the case, using equations (2.18) and (2.20), and neglecting F_3 in comparison with unity in equation (2.16), we obtain

$$k_0 a = \sqrt{c_1}(\epsilon/V_d) \tag{2.23}$$

$$t_{\text{axial}} = -W_{\min} = c_1 \epsilon^2/V_d. \tag{2.24}$$

It then follows that $F_3 \sim \epsilon/V_d \ll 1$ self-consistently. The assumption that $k_0 a \ll 1$ is also seen to be true. Although the T_c-enhancement is still of order ϵ^2/V_d, we can see that it exceeds that obtained with the earlier simple trial solution by noting that the quantity $2\alpha c_1$ can be written as the product $\langle k^2\rangle\langle k^{-2}\rangle$, where $\langle\cdots\rangle$ is a mean defined with respect to the distribution $g(\boldsymbol{k})\, \mathrm{d}^3k/(2\pi)^3$. The desired result then follows from noting that $2\alpha c_1 > 1$, i.e., $c_1 > 1/2\alpha$, either as a consequence of Hölder's inequality, or by recalling that the arithmetic mean of positive numbers with a non-zero spread always exceeds the harmonic

mean. Furthermore, the solution is non-perturbative since $|\eta_R| \sim (\epsilon/V_d)^{1/2}$, which is not an integer power of ϵ.

The improvement over the real solution is illusory, however. First, note that the axial solution is degenerate, since if $(\eta_L, \eta_R) = (u, v)$ is a solution to equations (2.2) and (2.3), so is (v^*, u^*). (This is merely complex conjugation.) Since $\eta_R \neq \eta_L^*$ for equation (2.6), these are distinct solutions. On the other hand, the GL equations are seen to be completely real when written in terms of η_x and η_y, suggesting that the ground state is also real and unique†. Thus a linear combination of the solution just found with its complex conjugate might be even lower in energy. The second point is that the length scale $k_0^{-1} \sim \epsilon^{-1}$ is much less than the Imry–Ma length scale $\sim \epsilon^{-2}$. Thus the axial order found above is only short ranged, and we can multiply the variational answer by a phase factor $\gamma(\boldsymbol{r})$ varying on a length scale greater than k_0^{-1} and try to further reduce the energy in this way. This forms the third stage of our argument. We denote the solution (2.6) by $\boldsymbol{\eta}^{(1)}$, its complex conjugate by $\boldsymbol{\eta}^{(2)}$, and examine the combination

$$\boldsymbol{u}(\boldsymbol{r}) = \frac{\mathrm{i}}{2}(\mathrm{e}^{\mathrm{i}\gamma(\boldsymbol{r})}\boldsymbol{\eta}^{(1)} - \mathrm{e}^{-\mathrm{i}\gamma(\boldsymbol{r})}\boldsymbol{\eta}^{(2)}). \tag{2.25}$$

This has Cartesian components

$$u_x = -\sin\gamma(1 + \operatorname{Re} h) - \cos\gamma \operatorname{Im} h \tag{2.26}$$

$$u_y = \cos\gamma(1 - \operatorname{Re} h) + \sin\gamma \operatorname{Im} h. \tag{2.27}$$

Since $|h(\boldsymbol{r})| \ll 1$, this solution is close to $(-\sin\gamma, \cos\gamma)$, i.e., to a rotation of the $(1, 0)$ solution. If γ were a constant, the energy would be identical to that of the solution (2.6), i.e., given by equation (2.24). To calculate the difference, let us define

$$T_1 = T_0 + t_{\text{axial}} \tag{2.28}$$

and $A^2 = 1 + \langle |h(\boldsymbol{r})|^2 \rangle = 1 + F_3$. If we average the quadratic part of f_{GL} over a length scale of order k_0^{-1}, we obtain a coarse-grained free energy

$$f_{cg} \approx A^2((T_1 - T) + \kappa(\nabla\gamma)^2 - \epsilon \langle \cos 2[\theta(\boldsymbol{r}) - \gamma(\boldsymbol{r})] \rangle_{cg}) \tag{2.29}$$

where $\langle \cdots \rangle_{cg}$ denotes a coarse-grained average. Note that the first term is proportional to $T_1 - T$ and not $T_0 - T$ as in equation (1.1). We can now apply the Imry–Ma argument to equation (2.29). Suppose that the energy is minimized when γ varies on a length scale L. In a supercell of linear size L, there are approximately $(L/a)^3$ cells in each of which $\theta(\boldsymbol{r})$ can be regarded as constant and independent of the other cells. Thus the coarse-grained average of either $\cos 2\theta$ or $\sin 2\theta$ in a supercell is a number of order $(L/a)^{3/2}$ of either sign. By choosing $\cos 2\gamma \propto +\langle \cos 2\theta \rangle_{cg}$, and $\sin 2\gamma \propto +\langle \sin 2\theta \rangle_{cg}$, we obtain a symmetry-breaking energy which is systematically negative in every supercell. Since the gradient energy is of order L^{-2}, we recover the Imry–Ma length scale $L \sim \epsilon^{-2}$, and an energy lowering $\mathcal{O}(\epsilon^4)$. This energy is *subdominant* to the gain (2.24) from the short-ranged order of equation (2.6), but since it represents an additional gain, the true solution is indeed disordered and real. It is in this sense that the real solution must be understood.

Therefore, the transition temperature for the real solution is given by

$$T_{c+} = T_1 + \mathcal{O}(\epsilon^4/V_d^3). \tag{2.30}$$

(Actually, $T_1 - T_0$ could also contain a term of order ϵ^4, but that is immaterial.) Let us now consider what happens at lower temperatures. Now the quartic terms come into play, and the positive β_2-term in f_{GL} favours an axial solution. For $\epsilon \ll V_d$, we can compare the free

† Another way of saying this is that in a large but finite system, H_{GL} has no symmetries (including time reversal) with a random AFM. Thus this argument does not apply to the periodic model studied in section 3.

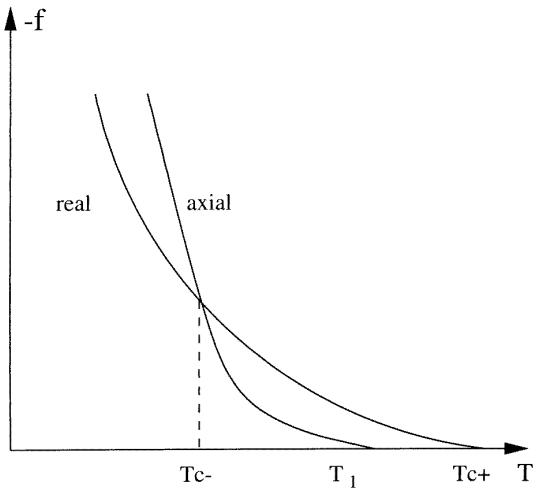

Figure 1. A sketch of the free energies (relative to that of the normal state) of the real and axial solutions, showing a change in slope, i.e., a first-order transition, at T_{c-}. The transition at T_{c+} is of second order.

energies of the real and axial solutions directly. Since $\langle|\eta_i\eta_i|^2\rangle$ vanishes for a purely axial solution and equals $\langle(\eta_i\eta_i^*)^2\rangle$ for a purely real one, these free energies will differ strongly in the contribution of the β_2-term. Approximating multiplicative factors such as $\langle|\eta|^4\rangle/\langle|\eta|^2\rangle^2$ by unity, and denoting real and axial solution quantities by subscripts r and a, the standard analysis of a Landau free energy with a second-order transition yields

$$f_r = -C_r(T - T_{c+})^2/2T_{c+} \tag{2.31}$$

$$f_a = -C_a(T - T_1)^2/2T_1 \tag{2.32}$$

where

$$C_r/T_{c+} = 1/2(\beta_1 + \beta_2) \tag{2.33}$$

$$C_a/T_1 = 1/2\beta_1. \tag{2.34}$$

The Cs are specific heats that would be obtained for the respective solutions. Since $C_a > C_r$, the free energies will intersect at a temperature $T_{c-} < T_1$ (see figure 1), giving a first-order transition at T_{c-}. Ignoring the small differences between T_0, T_{c+}, and T_1 compared to T_0 itself, we obtain

$$\Delta T_c = T_{c+} - T_{c-} = \frac{\mu}{\mu - 1}(T_{c+} - T_1) \tag{2.35}$$

where

$$\mu = (C_a/C_r)^{1/2}. \tag{2.36}$$

Differentiation of the free energies yields the entropies S_r and S_a, and the latent heat $\ell = T(S_r - S_a)$:

$$\ell = (\mu - 1)C_r\Delta T_c. \tag{2.37}$$

Let us ask whether the scenario that has emerged could describe the actual data. To this end, we take ΔT_c to be the observed T_c-splitting and C_r and C_a to be the specific heat jumps at the upper and lower transitions (relative to the normal state). Using data for sample 1 from reference [6], $\Delta T_c = 60$ mK, $C_r = 98$, and $C_a = 113$ mJ K^{-1}/(mol

UPt_3), which implies $\mu = 1.073$. Equation (2.37) then gives $\ell = 0.43$ mJ/(mol UPt_3). If we conservatively take the experimental temperature resolution to be 10 μK, then since the base specific heat is about 0.1 J K^{-1} mol^{-1}, we conclude that a latent heat of 1 μJ mol^{-1} would have been detected. The latent heat predicted by the 2D models is much larger. Furthermore, the measured [9] H_{c2}-slopes of $\sim$5 T K^{-1} imply values $\kappa = 6.6 \times 10^3$ K Å^2 and $V_d = 73$ mK. To obtain a ΔT_c of 60 mK we would therefore need $\epsilon \simeq 70$ mK, which is not much smaller than V_d.

3. The simple periodic model

The analysis so far has been done assuming $\epsilon \ll V_d$. It is also interesting to ask what happens when $\epsilon \gg V_d$. In this case we expect to obtain the local solution to a first approximation. Suppose that the solution is of the strictly local type near T_{c+}. Consider two nearby domains with $\boldsymbol{n} \parallel \boldsymbol{x}$, and $\boldsymbol{\eta} \parallel \boldsymbol{y}$, and an intervening domain with $\boldsymbol{n}$ at 120° to $\boldsymbol{y}$ and $\boldsymbol{\eta} \perp \boldsymbol{n}$. The system will lower its gradient energy by tunnelling of the η_y-component through the intervening domain. Since the $\boldsymbol{\eta}$-component parallel to $\boldsymbol{n}$ is exponentially small in the ratio ϵ/V_d, we expect the magnitude of the gradient energy gain to also be similarly small. However, the gradient energy favours relatively real η_x and η_y, while the β_2-term favours a relative phase of $\pi/2$ between them. General symmetry and continuity arguments would suggest that we still have two phase transitions with the lower one of first order, but now the T_c-splitting, and the degree of axiality at the lower transition will be exponentially small. It is then conceivable that an appreciable axiality only develops as a crossover at a lower temperature. This raises the possibility that the two true phase transitions are so close together that they appear to be one experimentally, and the crossover appears to be the phase transition at a lower temperature.

To investigate the above possibility, we have numerically studied a toy model with a periodic antiferromagnetic domain structure. The assumption of periodicity eliminates the possibility of a first-order transition altogether. This is because the model has an extra symmetry, and so is not in the same strict universality class as that studied in section 2. While the periodic model cannot capture the two very closely spaced transitions (it only yields one), it *can* capture the low-temperature crossover. Since we are only interested in the latter behaviour in this section, the periodic model is adequate. At the same time the periodicity simplifies the numerical problem enormously. We further take $\boldsymbol{n}(\boldsymbol{r})$ to vary only in one direction (x), and alternate between *two* orientations $\hat{\boldsymbol{x}}$ and $\hat{\boldsymbol{y}}$ with a periodicity of $2a$. More concretely, we have

$$F_{\mathrm{SBF}} = \int \mathrm{d}x \; 2\epsilon(x)(|\eta_x|^2 - |\eta_y|^2) \tag{3.1}$$

where $\epsilon(x) = \pm\epsilon$ for $0 < \pm x < a$, and $\epsilon(x) = \epsilon(x + 2a)$. The restrictions of one dimensionality and two orientations for $\boldsymbol{n}$ simplify the problem without changing its essential aspects. A closer analogue of the modified 2D model [3] would be obtained by allowing κ to vary periodically in addition to ϵ, but since the fit to the H_{c2}-data requires the two values of κ to be rather close, the gradient energies are not appreciably different from domain to domain, and nothing is gained from this refinement.

The eigenvalue problem for T_c is identical to the Kronig–Penney model, and is analytically solvable. The solution is doubly degenerate corresponding to either η_x or η_y being zero everywhere. (Note that with $V_d = \kappa/a^2$ as before, $T_c - T_0 \approx \epsilon^2/12V_d$ for small ϵ, so even with $\epsilon \simeq 3V_d$, say, one is not in the strong-symmetry-breaking regime.) We solve the non-linear GL equation below T_c by discretizing it with a mesh size $h \geqslant 2^{-10}a$,

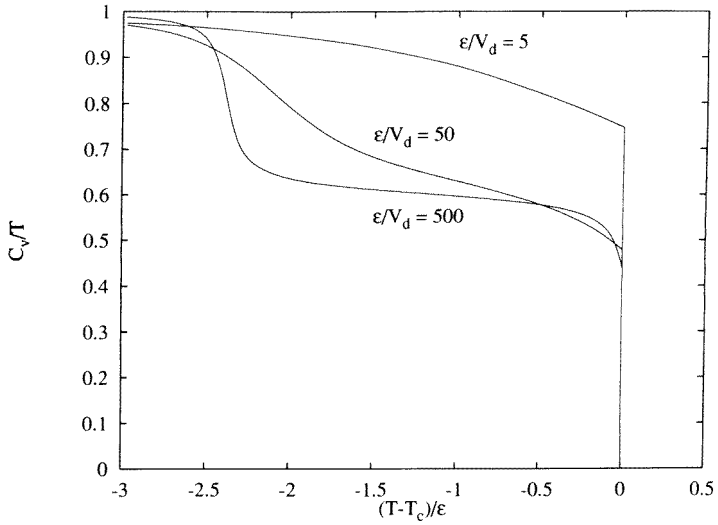

Figure 2. The computed specific heat for the simple periodic model. ϵ and V_d are the symmetry-breaking field and the gradient energies.

and using a Newton–Raphson method. The free energy is computed for a closely spaced set of T-values and numerically differentiated to obtain C_v/T. The calculations are done with $\beta_1 = 0.5V_d$ and $\beta_2 = 0.35V_d$. This ratio of β_2 to β_1 is chosen to yield comparable jumps in C_v/T at $T_{c\pm}$ in the uniform case. Our results for C_v/T are shown in figure 2 for various ϵ/V_d. There is clearly no second transition†. For $\epsilon/V_d = 500$ there is a crossover that could at first sight appear to mimic the data of reference [6], but this is not in fact the case. As noted earlier, the measured [9] H_{c2}-slopes imply that $V_d = 73$ mK. Since the temperature axis in figure 2 has been scaled by ϵ, and $\epsilon = 36$ K, the implied T_c-splitting would be $2.3\epsilon \simeq 80$ K. This is so far off the measured 50 mK splitting that the crossover scenario within a 2D model cannot be entertained for UPt_3.

The surprising result from this exercise is that one might have expected a crossover and an apparent T_c-splitting of order ϵ to appear for $\epsilon = 20V_d$, say. This is not the case, and the crossover is only manifest at a much larger value of ϵ/V_d.

4. Discussion

We have considered the effect of the short-ranged antiferromagnetic domain structure on the superconducting transition in UPt_3 within the 2D models, wherein the antiferromagnetism is the agency that splits the transition. It is known [15] that when the AFM is weak and random, one does obtain two transitions but the lower one is of first order. We have shown that this prediction is in *quantitative* conflict with observations by calculating the latent heat at this transition, and showing that if the T_c-splitting and the specific heats are to agree with the observed data, then this latent heat must be well above the experimental limits on detectability. In the course of this analysis we have studied a variational axial state to allow us to achieve a better understanding of the short-ranged order. We have also studied the

† This conclusion is supported by analytic work for $\epsilon \ll V_d$, in which T_c is found by keeping only one wavevector $q = \pi/a$. A linear stability analysis about this solution below T_c, and in the same approximation of one q, yields no further instabilities, *except* to the complex conjugate solution at T_c itself.

possibility that the lower transition may be a crossover and not a true phase transition. In this case, the observed T_c-splitting can not be reconciled with the gradient energy scale. In either case, the AFM domain structure rules out all 2D models [2, 3].

Finally, we briefly discuss experiments which show that the two transitions in UPt_3 merge into one at a pressure of $p_c \sim 4$ kbar [19]. The loose interpretation that this happens because the antiferromagnetism disappears at about the same pressure is thermodynamically unsustainable [4]. All mean-field models, whether based on one [12] or two [4, 5] primary order parameters, require either the re-emergence of a splitting for $p > p_c$, or a first-order line at $p \simeq p_c$. Neither possibility has been ruled out so far in our view, as the broad heat capacity anomalies seen in [19] could conceal two transitions, and a first-order transition has not been looked for. Careful experimental study of these possibilities is clearly highly desirable. At present, we believe that the two-order-parameter models, especially the AB model [4], are the only viable ones left for describing UPt_3.

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