Phases of URu_2Si_2

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The tetragonal heavy-fermion system URu_2Si_2 may well be an unconventional superconductor, as evidenced by its upper critical field and its low-temperature thermodynamic properties. Under the assumption that it belongs to the E representation of the D_{4h} point group, we enumerate the phase diagrams which are possible in the H-T plane. We take into account the possible coupling of superconductivity to the antiferromagnetic order parameter.

Unconventional superconductivity in heavy-fermion systems continues to be a rich field of research. Much activity has concentrated recently on UPt_3, because it possesses a very interesting phase diagram in the H-T plane.[1,2] In particular, a good explanation of the phases of UPt_3 can be obtained under the assumption that its gap parameter belongs to the E_1 or E_2 representations of the point group though many details remain to be worked out. Recently, upper critical field measurements on a different heavy fermion system, URu_2Si_2, have been interpreted successfully using the same hypothesis.[3] Thus it is possible that URu_2Si_2 may also have a complex phase diagram in the presence of a magnetic field. From the point of view of symmetry, it differs from UPt_3 in two important respects. It belongs to the tetragonal group D_{4h} rather than the hexagonal group D_{6h}. Its antiferromagnetic moment $\mathbf{M}_s$ is directed along the tetragonal axis (c axis) rather than in the basal plane. Both of these affect the Ginzburg-Landau theory of the E_1 (which becomes the E representation in D_{4h}) representation and change the analysis of the phases. In this Brief Report we enumerate the possible phase diagrams of URu_2Si_2 taking these facts into account.

We may write the free energy of the system[4] as follows:

$$F = F_u + F_m + F_g \ ,$$

where

$$F_u = \alpha \boldsymbol{\psi}\cdot\boldsymbol{\psi}^* + \beta_1(\boldsymbol{\psi}\cdot\boldsymbol{\psi}^*)^2 + \beta_2|\boldsymbol{\psi}\cdot\boldsymbol{\psi}|^2 + \beta_3(|\psi_x|^4 + |\psi_y|^4) \ ,$$

$$F_m = -ib\mathbf{M}_s\cdot\boldsymbol{\psi}\times\boldsymbol{\psi}^* \ ,$$

$$F_g = \sum_{ij}(K_1 p_i\psi_j p_i^*\psi_j^* + K_2 p_i\psi_i p_j^*\psi_j^* + K_3 p_i\psi_j p_j^*\psi_i^*) + \sum_i (K_4|p_i\psi_i|^2 + K_5|p_z\psi_i|^2 \ .$$

In these formulas $\boldsymbol{\psi}=(\psi_x,\psi_y)$ is a two-dimensional complex vector. $p_i=-i\partial/\partial x_i+(2e/\hbar c)A_i$, and the sums run over $i,j=x,y$. The constants α, β, b, and K are regarded in this paper as unknown parameters. $\mathbf{M}_s=M_s\hat{\mathbf{z}}$ is the antiferromagnetic order parameter. The admissibility of the coupling term is discussed in more detail below.

Let us consider the minimization of F when $H=0$. The $\boldsymbol{\psi}$ is uniform and $F_g=0$. If $b=0$, then we have three possibilities,[4] depending on the values of β_2 and β_3: $\psi_y=\pm i\psi_x$ (A phase); $\psi_x=0$, $|\psi_y|>0$ or $\psi_y=0$, $|\psi_x|>0$ (C phase); and $\psi_x=\pm\psi_y$ (D phase). When $b\neq 0$, $F_m=bM_s(|\psi_+|^2-|\psi_-|^2)$, where $\psi_\pm=(\psi_x\pm i\psi_y)/\sqrt{2}$. It is physically reasonable to treat F_m as a perturbation. From its form, one may see that it shifts T_c. More importantly, however, it may split the transition at zero field. Only if the A phase is the stable phase at zero field and low temperature does this *not* occur, for then $\psi_y=\pm i\psi_x$ minimizes *both* F_u and F_m. A splitting is caused by competition of second- and fourth-order terms. Note that this is the opposite of the situation in UPt_3 where the splitting occurs *only* if the A phase is the one stabilized by the fourth-order terms. For the C and D cases then, one should find a double specific heat anomaly. Early measurements have seen a very broad peak[5] which is somewhat reminiscent of early measurements on UPt_3. There have been two recent measurements. In one of these there is evidence for a double transition in the specific heat.[6] In the other, this splitting is believed to be due to the presence of two distinct phases of the starting material.[7] Measurements in a magnetic field would help to distinguish the various possibilities.

Along the $H_{c2}(T)$ curve, the fourth-order terms in F_u can be neglected and the resulting quadratic form may be completely diagonalized. The result for $\mathbf{H}\|c$ is that one has phase A once more, or phase U, a more complicated phase described elsewhere.[8] For $\mathbf{H}\perp c$, one finds the C phase. An experiment at constant temperature varying the field will detect a transition.

The argument that if the phases along the H_{c2} curve and the $H=0$ curve are different then one should have a transition in between is due originally to Volovik,[9] who presented it without proof. It may be objected that a smooth evolution from the low-field configuration to the high-field configuration is possible, given that the configuration of the order parameter in the intermediate region is spatially inhomogeneous. A detailed analysis of the Ginzburg-Landau equations shows that a transition does in fact occur, however, if the field is in the basal plane. This case (that corresponding to the first diagram in Fig. 1) is worked out in a forthcoming publication.[10] No proof that a transition must occur exists for other directions of the field, and a smooth evolution cannot be completely ruled out. The example of UPt_3 suggests that if a transition exists for one direction of **H**, it should exist for all directions. Strong arguments for the existence of a transition for $\mathbf{H}\|c$, based on numerical work, have been given by Tokuyasu, Hess, and Sauls.[11]

This analysis leads, in sum, to the possible phase diagrams pictured in Fig. 1. The simplest case is $H\perp c$. Then the three possibilities at zero field all give a nontrivial phase diagram. This will have a different topology when the A phase is stable at low fields, as discussed above. In all cases, the C phase is stable at high fields, but the second case, where C is also stable at low fields, requires some further comment. When the coupling to the magnetization is included, the C phases along the $H=0$ lines and the $H_{c2}(T)$ curve are perturbed, but generally in a different fashion. Thus we may still expect a transition between the two phases, one of which has (arbitrarily) been labeled C'.

For $H\|c$ the situation is complicated by the possibility of two high-field phases. However, the analysis is straightforward, being similar to UPt_3, and leads to a phase transition in all cases except when the A phase is stable at all fields. In this situation, there is also no splitting of the transition at zero field. This particular case is also interesting for another reason, namely that the phase diagram changes its topology as the direction of the field is rotated: for $H\perp c$ there are two superconducting phases, for $H\|c$ there is only one. A detailed solution of the upper critical field problem shows that the $H\|c$ case is a critical point.[12] What happens, therefore, is that as **H** is rotated away from the C axis a second phase "peels off" from the $H_{c2}(T)$ curve. Therefore, in the generic case there are two phases, and $H\|c$ is a special direction for which there is only one phase.

The phase diagrams in Fig. 1 are based on the assumption that the (T) curve is greater in absolute magnitude than the magnitude of the slope of the second transition line. This simplifies the pictures. However, it is the opposite of what happens in UPt_3, where the two curves appear to cross. Such a crossing is actually forbidden in the simple Ginzburg-Landau theory outlined above except when $H\perp c$. In this special direction there is a "selection rule" which means that the two curves do not influence one another and a tetracritical point is possible.[13] For other directions of the field, the crossing is avoided and the topology, though not the shape, of the phase boundaries, is as pictured here. Another possible case for which crossing can occur for all directions of the applied field is the "glassy" superconducting state.[13]

In this picture of the system, therefore, there is no parameter regime in which the phase diagram is trivial for all directions of the applied field. This conclusion is independent of any coupling to the antiferromagnetic order parameter. It is certainly worthwhile to search the H-T plane using all the experimental probes available, particularly ultrasonic absorption and specific heat measurements. It is the coupling to antiferromagnetism which could produce a zero field splitting of the transition, according to the analysis given above. This conclusion is subject to one important caveat. Translation through

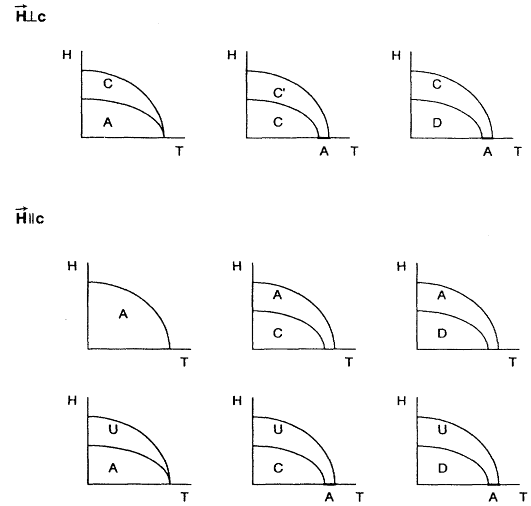

FIG. 1. Phase diagrams allowed by Ginzburg-Landau theory for a tetragonal superconductor whose order parameter belongs to the E representation. For **H** along the tetragonal axis ($\mathbf{H}\|c$), there are normally two phases. The only exception is when the A phase is stable at all fields. As the field is rotated into the basal plane, the A or U phase changes continuously into the C phase. The low-field phase must remain the same. Each column corresponds to a different choice of the β parameters in the Ginzburg-Landau free energies. The two possibilities for $\mathbf{H}\|c$ correspond to different choices of the K parameters. For details, see Refs. 4 and 8. The H_{c1} line is not shown on the diagrams.

$(a/2,a/2,c/2)$ is a symmetry operation of the system above T_N. If this operation combined with time reversal is a symmetry operation below T_N, then the coupling free energy term F_m is not allowed.[14,15] In other words, if the transition at T_N is to the simple nearest-neighbor Néel state, then no splitting should occur. There is reason to suppose from the specific heat that this is not the case and that the magnetic transition is a secondary manifestation of another transition.[16] The analysis at zero field given above would hold, for example, if the transition is ferromagnetic. The analysis apart from the small zero-field splitting is always valid independent of these considerations.

In conclusion, the hypothesis that URu_2Si_2 is a superconductor belonging to the E representation leads to the result that it should show a field-induced transition. It may or may not show a split transition at zero field.

Note added in proof. Recent specific-heat measurements on refined samples with $\mathbf{H}_{\perp}c$ show only a single transition [results of E. Knetsch, A. Menovsky, M. Meisel, G. Nieuwenhuys, and J. Mydosh (unpublished)]. According to Fig. 1, this proves unambiguously that URu_2Si_2 is *not* a multicomponent superconductor. It could still be an unconventional one-component system.

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