Pressure-Induced Magnetism and Hidden Order in $URu_2Si_2$

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Abstract

We discuss the discovery of pressure-induced antiferromagnetism in $URu_2Si_2$, in the context of neutron, NMR and $\mu$SR results. The identification of a critical pressure separating mean-field and Ising phase transitions leads us to propose that the system lies close to a bicritical point associated with magnetic and (non-magnetic) hidden order. We conclude that the recent observation of an isotropic, field-independent component in the silicon NMR line-width implies that the hidden order parameter breaks time-reversal invariance and present a preliminary discussion of the underlying nature of the hidden order parameter.

Key words:
$URu_2Si_2$, Hidden Order, Antiferromagnetism

The origin of the entropy loss in $URu_2Si_2$ at $T_0 = 17.5K$ is an outstanding problem in heavy fermion physics.[1] The observed staggered moment[2] ($m_0 = 0.03\mu_B$) cannot account for the sharp discontinuities in bulk properties [3–8] and the entropy change[9] that develop at this transition. The field-dependences of the magnetization and the gap[10,11] are dissimilar, implying the presence of a hidden order parameter $\psi$ that remains uncharacterized.[12,13]

Neutron experiments on $URu_2Si_2$ indicate that the staggered magnetic moment increases linearly with applied pressure,[15] $M \propto P$, up to 1 GPa. $M(T)$ is mean-field for $P < 1$ GPa but is Ising in character at higher pressures. In a parallel study, NMR measurements[16] on $URu_2Si_2$ show that at $T \leq T_0$ the silicon NMR line-width develops a field-independent, isotropic component.
whose temperature-dependent magnitude is proportional to that of the hidden order parameter. These results imply an isotropic field distribution at the silicon sites whose root-mean square value is proportional to the hidden order

\[ \langle B^\alpha(i)B^\beta(j) \rangle = a^2 \psi^2 \delta_{\alpha\beta}, \]  

(1)

and is \( \sim 10 \) gauss at \( T = 0 \). This field magnitude is too small to be explained by the observed moment which induces a field \( B_{\text{spin}} = \frac{\mu_0 M}{a^2} = 100 \) Gauss where \( a \) is the \( U-U \) bond length \( (a = 4 \times 10^{-10}) \). Furthermore this moment is aligned along the \( c \)-axis, and thus cannot account for the isotropic nature of the local field distribution detected by NMR.

Now we try to unify these two experiments within a common framework. It has been widely assumed that the magnetic and the hidden order coexist and are homogeneous.[12,13] However recent NMR studies of \( URu_2Si_2 \) under pressure[14] indicate that for \( T < T_0 \) there is coexistence of antiferromagnetic and paramagnetic regions, implying that the magnetic and the hidden order parameters are phase separated.[17] Then the change in character of the magnetic transition at \( P = P_c \) is naturally interpreted as originating from a bicritical point. This picture, supported by \( \mu SR \) data,[18] suggests that at ambient pressure the observed magnetization is a volume fraction effect which develops separately from the hidden order via a first order transition.

In order to study this scenario further, we assume that the free energy \( F[\psi, M, V] \) is function of the hidden order \( \psi \), the staggered magnetization \( M \) and the unit cell volume \( V \)

\[ F = F_\psi + F_M + g \psi^2 M^2 \]  

(2)

where \( F_X = (T_X(V) - T) X^2 + \frac{1}{2} u_X^2 X^4 \) with \( X = \{\psi, M\} \) and \( T_\psi = T_M \) at a critical volume \( V_c \). If \( g^2 \geq u_\psi^2 u_M^2 \), there exists a bicritical point[19] at \( V = V_c \) with an associated first order line as shown in Fig. 1a. For \( V > V_c \), the hidden order phase transition temperature is stable and \( T_\psi > T_M \).

In order to transform the \( T - V \) phase diagram (Fig. 1a) into one for \( T - P \) (Fig. 1b), we note that the pressure \( P = -\frac{\partial F}{\partial V} \) is discontinuous across the first-order line in Fig. 1a leading to two distinct pressure scales, \( P_\psi \) and \( P_M \), in the \( T - P \) plot (Fig. 1b). In the associated coexistence region (Fig. 1b), the fraction of magnetic phase \( x \) is given by the expression \( P(x) = (1-x)P_\psi + xP_M \) so that the net magnetization is then

\[ M = Mx = M \left( \frac{P - P_\psi}{P_M - P_\psi} \right) \]  

(3)
Equation (3) displays the linear development of $M(P)$ for $P > P_\psi$; here we attribute a small $P_\psi$ to a large pressure-change associated with the first-order line in Fig. 1a. In the hidden order phase, the magnetization will develop via a first order phase transition in qualitative agreement with $\mu$SR data.[18] Thus this simple approach can model the observed change of the magnetization at low $P$.

We now discuss the nature of the hidden order parameter, emphasizing the observed isotropic field distribution at the silicon sites. The magnetic fields at these nuclei have two possible origins:[20] the electron-spin interaction and the orbital shift due to current densities. In $URu_2Si_2$, the electron fluid exhibits a strong Ising anisotropy along the c-axis, as measured by the Knight shift[16]; thus the electron-spin interaction cannot be responsible for the isotropic fields at the silicon sites. Alternatively we propose that these local fields are induced by currents that develop inside the crystal as the hidden order develops; thus the observed isotropic line-width is attributed to the orbital shift. We are therefore suggesting that for $T < T_0$, $URu_2Si_2$ is an orbital antiferromagnet[21]. Such states have been been studied extensively in the context of the two-dimensional Hubbard model, [22–25], particularly in connection with staggered flux phases[26,27]. More recently commensurate current density wave order has been proposed as an explanation of the spin-gap phase in the underdoped cuprate superconductors.[28]

As a simple check on the applicability of orbital antiferromagnetism to $URu_2Si_2$, we estimate local fields at the silicon sites due to orbital currents circulating around the square uranium plaquettes in the a-b plane. On dimensional grounds, the current along the $U - U$ bond is given by $I = e \Delta / h$ where $\Delta$ is the gap associated with hidden order formation. A microscopic derivation of this expression can be obtained from the Hubbard model[29], assuming that the hidden order parameter is the current along a $U - U$ bond. If this current loops around a plaquette of side length $a$, then the field induced at a height $a$ above the plaquette is approximately $B = \left( \frac{e a}{2 \pi a} \right) \left( \frac{e \Delta}{h} \right)$. Using the values $a = 4 \times 10^{-10}m$, $\Delta = 110K$, we obtain $I = 2.3 \mu A$ and $B = 11$ Gauss, in good agreement with the local field strength detected in NMR and $\mu$SR measurements.[16,18]

In order to test whether this proposal will yield local isotropic fields, we allow the circulating current around a plaquette (cf. Fig. 2) centered at site $X$ to develop staggered order $I(X) = \psi e^{iQ \cdot X}$. The current along a bond is then the difference of the circulating currents along its adjacent plaquettes. The field at a silicon site can be computed using Ampere’s law, where the relevant
The silicon atoms in $URu_2Si_2$ are located at low-symmetry sites, so that the fields do not cancel; they reside above and below the centers of the uranium plaquettes. The proposed orbital antiferromagnet must have $Q \neq (\pi, \pi)$ in order to produce isotropic field distributions at the silicon sites as displayed in Fig. 3a. Using the vector potential in (4), our initial study finds that an incommensurate $Q$ vector in the vicinity of $Q = (\frac{1}{4}, \frac{1}{4}, 1)$ (Fig 3b) produces an isotropic field distribution at the silicon sites. Such a configuration is staggered between the $U$ layers, with a periodicity of four unit cells in the basal plane.

We end with a brief discussion about the microscopic nature of the underlying hidden order. First, the presence of isotropic local fields[16] at the silicon sites, implies that $\psi$ must break time-reversal invariance. Second, we believe that the magnitude of the observed fields indicates that they are current-induced; this proposal is also compatible with the observed robustness of $\psi$ to application of high magnetic fields[10]. A simple possibility[30] is to identify the hidden order parameter directly with a charge current, corresponding to the imaginary part of an electron-hopping operator

$$\bar{I} \propto -i\langle c_\sigma^\dagger(x + \hat{i}/2) c_\sigma(x - \hat{i}/2) - H.c. \rangle.$$  

This bond current order would involve a significant fraction of the entire gap $\Delta = 110K$ and thus could account for the observed local fields; furthermore the associated entropy $\frac{S}{R \ln 2} = \frac{\Delta}{2E_F}$ at $T_0$ could easily account for the experimentally observed value of $\frac{S}{\ln 2} \sim 0.2R$.

Our proposal of incommensurate orbital antiferromagnetism in $URu_2Si_2$ can be tested by experiment. $\mu SR$ measurements should confirm that the magnetic volume fraction of the sample increases with pressure. The incommensurate current ordering can be probed by neutrons, whose scattering off the fields produced by the orbital antiferromagnetism should produce (i) a small incommensurate Bragg peak with a rapidly decaying form-factor characteristic of an extended object and (ii) a dispersing gapless mode centered around the incommensurate Bragg peak, associated with collective translations of the orbital antiferromagnet.

In summary, we have discussed the implications of three recent experiments
on $URu_2Si_2$. We conclude that the observed pressure-induced antiferromagnetism is probably due to phase separation. We argue that the development of isotropically distributed magnetic fields at the silicon sites indicates that the hidden s order parameter breaks time-reversal invariance. Based on the size and isotropy of the measured local fields, we propose that $URu_2Si_2$ is an incommensurate orbital antiferromagnet and make a number of predictions for experiment.

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References

[1] c.f. W.J.L. Buyers, Physica B 223 & 224, 9 (1996).
[2] C. Broholm et al., Phys. Rev. Lett. 58, 1467 (1987).
[3] U. Walter et al. Phys. Rev. B33, 7875 (1986).
[4] M.B. Maple et al., Phys. Rev. Lett. 56, 185 (1986).
[5] T.E. Mason and W.J. Buyers, Phys. Rev. B43, 11471 (1991).
[6] Y. Miyako et al., J. Appl. Phys. 76, 5791 (1991).
[7] A.P. Ramirez et al., Phys. Rev. Lett. 68, 2680 (1992).
[8] T.E. Mason et al., J. Phys.: Condens. Matt. 7, 5089 (1995).
[9] T.T.M. Palstra et al., Phys. Rev. Lett. 55, 2727 (1985).
[10] S.A.M. Mentink et al., Phys. Rev. B 53, 6014 (1996).
[11] N.H. van Dijk et al, Phys. Rev. B 56, 14493 (1997).
[12] N. Shah et al, Phys. Rev. B 61, 564 (2000).
[13] Y. Miyako et al., RIKEN Review 27, 54 (2000).
[14] K. Matsuda et al., Phys. Rev. Lett. 87, 087203, (2001).
[15] H. Amitsuka et al., Phys. Rev. Lett. 83 5114 (1999).
[16] O. Bernal et al, cond-mat/0106479.
[17] c.f. S.A. Kivelson et al., cond-mat/0105201 for a similar discussion in the context of the cuprates.
[18] G.M. Luke et al, Hyperfine Inter. 85, 397 (1994).
[19] P.M. Chaikin and T.C. Lubensky, Principles of Condensed Matter Physics (Cambridge University Press, Cambridge, 1994).

[20] C.P. Schlichter, Principles of Magnetic Resonance, (Springer-Verlag, Berlin 1978).

[21] B.I. Halperin and T.M. Rice, Solid State Phys. 21, eds. by F. Seitz, D. Turnbull and H. Ehrenreich, (Academic Press, New York, 1968).

[22] I. Affleck and J.B. Marston, Phys. Rev B 37, 3774 (1988).

[23] G. Kotliar, Phys. Rev. B 37, 3664 (1988).

[24] A.A. Nersesyan and G.E. Vachnadze J. Low Temp. Phys. 77, 293 (1989).

[25] H. Schultz, Phys. Rev. B 39, 2940 (1989).

[26] X.-G. Wen and P.A. Lee, Phys. Rev. Lett. 76, 503 (1996).

[27] D.A. Ivanov et al., Phys. Rev. Lett. 84, 3958 (2000).

[28] S. Chakravarty et al., Phys. Rev. B, 63, 9450311 (2001); S. Chakravarty et al., cond-mat/0101204; S. Tewari et al., cond-mat/0101027.

[29] T.C. Hsu, J.B. Marston and I. Affleck, Phys. Rev. B 43, 2866 (1991).

[30] P. Chandra, P. Coleman and J. A. Mydosh, to be published.

[31] V. Barzykin and L.P. Gorkov, Phys. Rev. Lett. 70, 2479 (1993); V. Barzykin and L.P. Gorkov, Phys. Rev. Lett. 74, 4301 (1995).

[32] G. Aeppli and C. Broholm, Handbook of the Physics and Chemistry of the Rare Earths 19 (North-Holland, Amsterdam, 1994) p. 123.
Fig. 1. (a) Proposed temperature-volume phase diagram for $URu_2Si_2$, with a first order line emanating from the bicritical point where $T_\psi$ and $T_M$ are equal. (b) In the temperature pressure diagram the first order line is broadened into a region of phase co-existence, in which the staggered moment is approximately linear in the applied pressure.

Fig. 2. Schematic drawing showing how current along a bond is the difference of the circulating currents around neighboring Uranium plaquets.
Fig. 3. Schematic field distribution of (a) commensurate and (b) incommensurate orbital antiferromagnet. Figure shows “edge on” view along the a-axis of the unit cell. (a) gives rise to an anisotropic field distribution along the c-axes at the silicon sites. The $Q$ vector in (b) has been chosen to give an isotropic field distribution at the silicon sites. (c) Showing field distributions at Silicon sites for a sequence of four evenly spaced $Q$ vectors between cases (a) and (b).