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The case for phase separation in URu_2Si_2

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Abstract

Motivated by experiment, we review the case for phase inhomogeneity in URu_2Si_2. In this scenario, the paramagnetic hidden order phase coexists with small distinct domains of antiferromagnetism whose volume fraction increases with pressure. The implications for the nature of the hidden order are discussed.

The heavy-fermion material URu_2Si_2 poses a unique challenge. Discovered almost two decades ago, it provides a classic example of a mean-field phase transition [1] at $T_c = 17$ K; yet there is still no consensus on the nature of the underlying order. More specifically, the transition is characterized by sharp anomalies in a number of bulk properties [2–4] and a gap [5–8] that each develop at T_c. Initially the ordered phase of this material was characterized as a spin density wave, but subsequent neutron scattering measurements [7, 8] indicated that the size of the staggered moment is too small ($\sim$0.02–0.04 μ_B/uranium atom) to account for the substantial entropy loss which occurs at the transition [9].

A sequence of recent experimental developments has led to new insight into the nature of the hidden order in URu_2Si_2. High-field measurements [10, 11] have revealed that the staggered magnetization and the gap have *different* field dependences, suggesting that there are two distinct order parameters, M and ψ. Initial theories assumed that the hidden order and the spin antiferromagnetism were coupled and spatially homogeneous [12]. Pressure-dependent neutron scattering studies [13] subsequently showed that the ordered antiferromagnetic moment M in URu_2Si_2 grows roughly linearly with applied pressure, $M \propto P$, up to $P_0 = 1$ GPa. Within the homogeneous scenario, this result requires a pressure-dependent coupling between the hidden order and the magnetism ($\Delta\mathcal{F} \sim -\psi M P$); such a linear coupling, required to nearly vanish at ambient pressure, is awkward to justify on symmetry grounds.

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Pressure-dependent NMR studies [14] provided a natural resolution of this dilemma by revealing that there exist distinct antiferromagnetic and paramagnetic regions whose relative volume fraction changes with applied pressure and temperature. More specifically, the single resonance associated with paramagnetism at high temperatures remains clearly at temperatures $T < T_c$, where at finite applied pressure it coexists with two symmetric satellite lines associated with antiferromagnetic ordering. The frequency shift associated with these additional resonances is independent of applied pressure, indicating the presence of a constant magnetic moment. However, the relative integrated intensities of the antiferromagnetic and the paramagnetic lines are temperature- and pressure-dependent, and are naturally interpreted as reflecting the relative volume fractions of spin ordered and disordered regions. Indeed, assuming a fixed magnetic moment, the pressure dependence of the magnetic Bragg peak observed in neutron scattering [13] is consistent with the antiferromagnetic volume fraction taken from the NMR work [14]. The natural conclusion from these studies, supported by earlier μSR data [15], is that the observed increase in the magnetization as a function of pressure is simply a volume fraction effect [14]. Moreover, these measurements indicate that at ambient pressure there exists a large pressure-independent moment that resides in less than 10% of the material. The majority phase therefore contains no conventional spin order, and theoretically the hidden order parameter is no longer accountable for the small but finite presence of antiferromagnetism.

In the pressure-dependent neutron scattering experiments [13], the character of the magnetic transition changes from mean field to Ising at $P = P_0$. This feature, combined with the observed linear pressure dependence of M for $P < P_0$, is naturally interpreted as originating from the presence of a bicritical point (figure 1(a)) [16]. In this scenario, at ambient pressure the observed magnetization is a volume fraction effect which develops *distinctly* from the hidden order via a first-order transition. We can study the phase behaviour of such a system using the free energy

$$F = F_\psi + F_M + g\psi^2 M^2 \tag{1}$$

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 $g^2 \geqslant u_\psi^2 u_M^2$, a bicritical point exists [17] at $V = V_c$ with an associated first-order line.

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

$$\mathcal{M} = Mx = M\left(\frac{P - P_\psi}{P_M - P_\psi}\right). \tag{2}$$

Equation (2) indicates the linear relation of the observed magnetization as a function of pressure for $P > P_\psi$ where P_ψ is small due to a large pressure change associated with the first-order line in figure 1(a).

There are a number of experimental observations that are consistent with this scenario where the hidden order and the spin antiferromagnetism are phase separated. Within this framework the *spatially* inhomogeneous average 'moment' is taken to be

$$M^2 = \frac{1}{V}\int \langle M(x)M(0)\rangle \, \mathrm{d}^3x, \tag{3}$$

where $M(x)$ is the local staggered magnetization. For a fixed site-independent value of $M(x)$, equation (3) is simply proportional to the volume fraction of antiferromagnetic regions. Earlier μSR studies [15] found that the muon precession signal, sensitive to magnetic ordering,

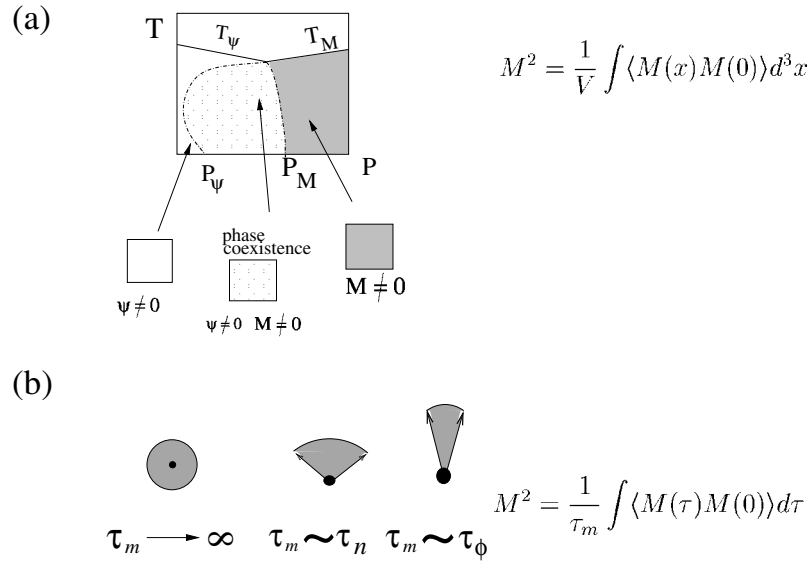

Figure 1. Schematic diagrams graphically contrasting (a) temporally and (b) spatially inhomogeneous moments that play key roles in the dynamical moment and the phase separation scenarios respectively of URu_2Si_2.

developed abruptly at the transition T_c, suggesting a first-order transition of the magnetization. Upon cooling, this precession frequency remained constant, indicating that the size of the moment is temperature independent. By contrast, the amplitude of the precessing signal increased with decreasing temperatures indicating a change in the underlying antiferromagnetic volume fraction. Recent μSR studies have extended this work, confirming the increase of the precession amplitude with applied pressure [18]; this result is consistent with the NMR data [14]. At ambient pressure the onset temperatures of the hidden order and the antiferromagnetism are very close, but they can be separated by both chemical [18] and applied [19] pressure. Finally such measurements [18, 19] indicate that the onset detection of the inhomogeneous antiferromagnetism is critically dependent on sample quality and history, particularly as a function of pressure. This is to be expected in a system of spatial inhomogeneities.

A proposal [20] alternative to phase inhomogeneity emphasizes the inferred presence of a *dynamical* order parameter whose time dependence is invoked to explain observed behaviour in URu_2Si_2. In particular resonant x-ray scattering, a probe with a timescale of $\tau_\phi \sim 10^{-14}$ s, indicates a moment of 0.3 μ_B/uranium atom which is consistent with the entropy lost at the transition [2, 9]. It is argued that there is temporal averaging of the moment over time-reversed Néel states on the timescales probed by neutron scattering ($\tau_n \sim 10^{-12}$ s), so only a fraction of it is observed (0.02 μ_B/uranium) (figure 1(b)). Similarly, it is noted that NMR and μSR measurements on URu_2Si_2, both of which have much longer observation timescales than do neutrons, indicate no long-range magnetic order at all. In this scenario, the *temporally* inhomogeneous average 'moment' is taken to be

$$M^2 = \frac{1}{\tau_m}\int \langle M(\tau)M(0)\rangle \, \mathrm{d}\tau \tag{4}$$

where $M(\tau)$ is the dynamical staggered magnetization. Here τ_m refers to the measurement time; because the moment fluctuates between different orientational states, longer and longer time averaging occurs as τ_m is increased, leading to a decreasing value of M. Application of pressure is argued to slow down the moment fluctuations, hence making the full amplitude of the magnetic order parameter 'accessible' to neutrons. Indeed the observed saturated moment measured under pressure by means of neutron scattering is consistent with the value observed by means of fast resonant x-rays [13, 20].

It is instructive to compare the situation in URu_2Si_2 with that for the pseudobinaries $U(Pt_{1-x}Pd_x)_3$. For $x \leqslant 0.01$, neutron [21, 22] and magnetic x-ray scattering [23] experiments reveal a small moment (0.02 μ_B/uranium atom), comparable in magnitude to that observed in analogous measurements on URu_2Si_2 at ambient pressure. However, NMR [24] and μSR experiments [25, 26] on these Pd-doped UPt_3 materials do not detect this moment at all, leading to the suggestion that it fluctuates on timescales intermediate between the observation time windows of the two sets of probes. The development of this small-moment antiferromagnetic state occurs via a crossover [27] rather than a transition and is *not* accompanied by any thermodynamic anomalies. By contrast, at higher dopings ($0.02 \leqslant x \leqslant 0.08$), both neutrons and muons see a large moment ($\sim$0.6 μ_B) and there are associated discontinuities in bulk properties [27]. Here the key point is that fluctuating moments do exist and their development is associated with a crossover and *not* a true phase transition; they can enhance pre-existing thermodynamic anomalies [28] but they cannot produce such discontinuities purely by themselves. In contrast to the situation in $U(Pt_{1-x}Pd_x)_3$ for low doping ($x \leqslant 0.01$), in URu_2Si_2 there are dramatic thermodynamic signatures of a true phase transition coexisting with the presence of a small static moments [9], that *cannot* be *solely* explained by dynamical fluctuations. Similarly the pressure-independent frequency shift of the antiferromagnetic resonance lines detected in NMR measurements [14] at temperatures $T < T_0$ is difficult to reconcile with a homogeneous dynamical moment whose fluctuating timescale is reduced with applied pressure.

Recent high-field measurements (figure 2) of the antiferromagnetic moment and the magnetic excitations with neutron scattering [11] also yield insight into the question of spatial inhomogeneity of the spin magnetism. These experiments indicate that the field dependence of the moment has a distinctive inflection point at 7 T, and remains finite but small up to fields of order 17 T. Such behaviour is strongly suggestive of a *local* linear coupling term for M and ψ of the form

$$\Delta\mathcal{F} = g \int \mathrm{d}^3x\, M(x)\psi(x). \tag{5}$$

Indeed it was shown earlier [12] that the presence of such a term in the Landau–Ginzburg free energy leads to a field dependence of the staggered magnetization, $M[h]$, of the form

$$M^2[h] = M_0^2 \frac{[1-h^2]}{(1+\delta h^2)^2}, \tag{6}$$

where $h = \frac{H}{H_c}$ is the ratio of the external and the measured critical magnetic fields ($H_c = 40$ T) and δ is defined through the relation $T_m(V, h) - T = [T_m(V) - T](1 + \delta h^2)$. We note that this expression has a point of inflection around the field value $H_m \sim H_c h_m$ where $h_m = \frac{1}{\sqrt{1+2\delta}}$; qualitatively this is because M decreases with decreasing h but, due to its coupling with ψ, must maintain a nonzero value up to $h = 1$ (figure 2).

At first sight, the presence of the observed inflection point [11] is rather puzzling, particularly as the original phenomenology [12] was developed for homogeneously coexisting magnetic and hidden order parameters. Certainly it appears to confirm the existence of a linear

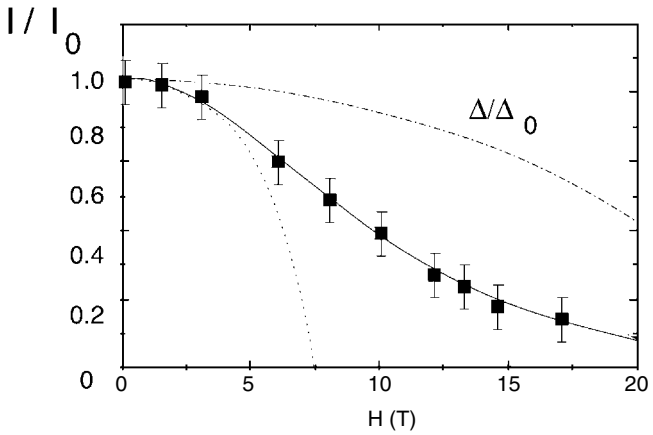

Figure 2. A sketch of the data [11], showing the field variation of the neutron scattering intensity from the staggered moment, compared with the predictions (6) of Landau–Ginzburg theory. The dashed curve shows the variation of the hidden order gap with the field, while the dotted curve shows the quadratic extrapolation of the low-field dependence of the moment.

coupling between M and Ψ. In a homogeneous system, such a term can only occur in the free energy if (i) M and ψ have the same ordering wavevector and (ii) ψ breaks time-reversal symmetry. From neutron scattering measurements [7, 8] it is known that M is commensurate, whereas there are many indications that the hidden order is not. In particular, we expect ψ to be incommensurate due to the fact that the observed entropy loss and the accompanying gap suggest that it results from a Fermi surface instability. Furthermore, the observed insensitivity of the elastic response [29] at T_c is consistent with the presence of an incommensurate density wave that couples weakly to uniform strain. Thus it appears that in a homogeneous system the presence of a linear coupling term for the magnetization and the hidden order is unlikely, due to the dissimilarity of their respective wavevectors. However, in a phase-separated scenario, it may be easier to motivate the presence of such a coupling between M and ψ. The spatial inhomogeneity of the spin order and the distribution of antiferromagnetic domain sizes mean that translational invariance is lost, making it possible for a *local* coupling to develop between the two order parameters. The presence of stacking faults and other defects will tend to enhance the strength of this coupling. Indeed, in the presence of disorder it is difficult [30] to avoid a local linear coupling between random fields and a coexisting order parameter if such a term in the free energy is allowed by time-reversal invariance.

The presence of an inflection in the field-dependent magnetization is an indication that the hidden order parameter *breaks* time-reversal symmetry, consistently with previous NMR measurements [31, 32]. Motivated both by experiment and by symmetry considerations, we have argued elsewhere [33] that the two leading candidates for producing the hidden order are a quadrupolar charge density wave and an orbital antiferromagnet. The key factor distinguishing these contenders is the presence or absence of time-reversal breaking. Thus the recent high-field measurements [11] point towards orbital antiferromagnetism. Naturally it would be optimal to have a direct experimental test of this conjecture. Neutron scattering could provide such a probe, particularly since the form factor associated with the extended current loops of the orbital antiferromagnet is different from that of point spins [32]. More specifically, we have used the spatial distribution of the orbital fields consistent with the NMR results [31] to determine the positions, the intensities and the form factor associated with the peaks anticipated

in neutron scattering measurements. Perhaps most important, we find [32] that the maximum scattering intensity is predicted to lie in a ring $\vec{Q} = \vec{Q}_0 + \vec{q}$ of radius $|\vec{q}| \sim 0.2$ centred around wavevector $\vec{Q}_0 = (001)$, where $\vec{q}$ lies in the a–b plane. It should be noted that scattering in the vicinity of $\vec{Q}_0$ is *forbidden* for the case of ordered spins aligned along the c-axis, for their dipole form factor is proportional to $(\hat{Q} \times \vec{M})^2$, and thus vanishes for $\hat{Q} \parallel \vec{M}$. Hence the presence of neutron scattering at this particular wavevector would be a 'smoking gun' confirmation of incommensurate orbital antiferromagnetism as the enigmatic hidden order.

In summary, motivated by experiment, we have presented the case for phase separation of spin magnetism and hidden order in URu_2Si_2. We argue that pressure-dependent neutron [13] and nuclear magnetic resonance [14] studies are naturally interpreted in terms of a *spatially* inhomogeneous moment whose volume fraction increases with applied pressure. The alternative proposal of a *temporally* inhomogeneous moment that is spatially homogeneous cannot, to our knowledge, account for the marked entropy loss and the bulk discontinuities associated with the transition at T_c. Furthermore, recent observation of a marked inflection in the field dependence of the magnetization indicates an underlying linear coupling between M and ψ, which is difficult to understand in a homogeneous scenario due to the disparity of their wavevectors. By contrast, such a term could be realized as a local coupling in a phase-segregated system where the microscopic spatial inhomogeneities of the order parameters break translational symmetry. It is important to emphasize that, even within the phase separation scenario, such a coupling can *only* exist if the hidden order parameter breaks time-reversal invariance. Thus the field-dependent magnetization studies [11], like earlier NMR measurements [31], point towards incommensurate orbital antiferromagnetism as a key contender for being the hidden order. A direct test of this conjecture would be neutron measurements at a particular wavevector where scattering is forbidden for point spins. We eagerly anticipate the results of these measurements.

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