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Origin of the Large Anisotropy in the χ_3 Anomaly in URu_2Si_2

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Abstract. Motivated by recent quantum oscillations experiments on URu_2Si_2, we discuss the microscopic origin of the large anisotropy observed many years ago in the anomaly of the nonlinear susceptibility in this same material. We show that the magnitude of this anomaly emerges naturally from hastatic order, a proposal for hidden order that is a two-component spinor arising from the hybridization of a non-Kramers Γ_5 doublet with Kramers conduction electrons. A prediction is made for the angular anisotropy of the nonlinear susceptibility anomaly as a test of this proposed order parameter for URu_2Si_2.

The character of the long-range "hidden order" (HO) in the heavy fermion material URu_2Si_2 continues to fascinate and elude theorists and experimentalists [1]. Despite a large entropy released at the HO transition temperature ($T_0 = 17.5K$) accompanied by many sharp thermodynamic anomalies there[1, 2, 3, 4], there is no consensus on the important local degrees of freedom and more importantly how they form the hidden order state.

A revisit to the nonlinear susceptibility (χ_3) and its sharp anomaly at T_0 [3, 4] may provide insight into the mysterious nature of the hidden order. Originally the behavior of this mean-field-like anomaly inspired a density-wave description. A simple Landau theory, $f = f(T - T_c(B^2))$, was constructed that yielded the relation

$$\Delta\left(\frac{c_v}{T}\right)\Delta\chi_3 = 12\left[\Delta\left(\frac{\partial\chi}{\partial T}\right)\right]^2 \tag{1}$$

that was tested successfully experimentally[5], suggesting underlying itinerant order; here we recall that $M = \chi_1 B + \frac{\chi_3}{3!}B^3$ where M is the magnetization and χ_1 and χ_3 are the linear and nonlinear susceptibilities respectively. However the large c-axis anisotropy of this χ_3 anomaly was then emphasized and ascribed to local Ising f-moment behavior [6, 7]. The appropriate Landau theory is then

$$f(T, B_z) = [\alpha(T_c - T) - \eta_z B_z^2]|\Psi|^2 + \beta|\Psi|^4 \tag{2}$$

where Ψ is the hidden order parameter; here a key question arises: what is the microscopic origin of the large anisotropy coefficient η_z?

The anisotropic field-response of the U-ions in URu_2Si_2 is most naturally understood if their low-energy configurations are $5f^2$, and this point has been noted by several authors[6, 7, 8, 9, 10]. Such strong Ising anisotropy was also observed in dilute $U_xTh_{1-x}Ru_2Si_2$, thus indicating that it is a key property of the U ions [8]. It was originally described by quadrupolar ordering of the local f-moments[6, 7, 9], but unfortunately recent resonant X-ray measurements see no signature for such multipolar order[11, 12]. Furthermore there is experimental evidence from scanning tunnelling microscopy[17, 18], and de Haas-van Alphen (dHvA) and Shubnikov-de Haas (SdH)[13, 14, 15, 16] measurements for itinerant behavior below T_o, so a local-moment scenario cannot capture the full picture at the transition.

Recent quantum oscillations provide a clue towards resolving this dilemma, as they reveal that the quasiparticles deep in the HO phase exhibit a large Ising anisotropy[16]. More specifically their Zeeman splitting depends only on the c-axis component of the magnetic field, $\Delta E = g(\theta)\mu_B B$ with a g-factor $g(\theta) = g\cos\theta$. The size of the measured g-factor anisotropy is resolution limited and exceeds thirty, corresponding to an anisotropy of the Pauli susceptibility in excess of 900 that is too large to be attributed simply to spin-orbit coupling. These experiments suggest that the anisotropy of the U $5f^2$ moments is transferred to the mobile quasiparticles in the HO phase via hybridization. Angle-dependence of the Pauli-limited critical field of the superconducting state[19, 20] confirms the g-factor anisotropy; furthermore the presence of Ising quasiparticles in a superconductor with $T_c \sim 1.5K$ indicate that the U $5f^2$ configuration is degenerate to within an energy resolution of $g\mu_B B \sim 5K$. The tetragonal symmetry of URu_2Si_2 protects the non-Kramers doublet Γ_5 that is quadrupolar in the basal plane and magnetic along the c-axis, and it has been proposed as the origin of the magnetic anisotropy in both dilute and dense URu_2Si_2[8, 9, 16, 21].

Conventionally in heavy fermion materials, hybridization involves valence fluctuations between a ground-state Kramers doublet and an excited singlet; it develops via a crossover, leading to mobile heavy quasiparticles. However if the ground-state is a non-Kramers doublet, the Kondo effect will involve an excited Kramers doublet[8]. Hybridization now carries spin and coherence; its development breaks time-reversal and spin rotational invariance at a true phase transition. Furthermore because it mixes states of half-integer (non-Kramers doublets) and integer (conduction electrons) it breaks Kramers parity and thus also breaks double-time reversal[21]. In URu_2Si_2 optical[22] and tunnelling probes[17, 18] indicate that hybridization develops abruptly at the HO transition, and thus we have proposed that the hybridization is a two-component order parameter ("hastatic" order) that transforms as a spinor[21].

Let us briefly digress to discuss the microscopic valence fluctuations of a non-Kramers doublet. The valence fluctuation physics of a non-Kramers doublet are described by an Bolech-Andrei model[23] (ABM) $H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \sum_j H_{ABM}(j)$, where the valence fluctuations and atomic physics of each U ion are described by

$$H_{ABM}(j) = V\left[c^\dagger_{\sigma\alpha}(j) X_{\sigma\alpha}(j) + \text{H.c}\right] + \epsilon X_{\sigma\sigma}(j) \quad (3)$$

where the $X_{\sigma\alpha} = |5f^3, \sigma\rangle\langle 5f^2, \alpha|$ is the Hubbard operator between the Kramers and non-Kramers state, $X_{\sigma\sigma} = |5f^3, \sigma\rangle\langle 5f^3, \sigma|$ is the projection onto the excited Kramer's doublet, and $\epsilon = E[5f^3] - E[5f^2]$ is the energy difference between the $5f^3$ excited Kramers doublet and the $5f^2$ non-Kramers ground-state. The operator $c^\dagger_{\sigma\alpha}(j)$ creates a conduction *hole* with channel and spin index α and σ at site j. Valence fluctuations out of a non-Kramer's doublet involve at least two different crystal symmetry channels, the Γ_7 and Γ_6 channels. The ABM results from the projection of an Anderson model into the low-lying $5f^2$, $5f^3$ subspace. $H_{ABM}(j) = \mathcal{P} H_{Anderson}(j) \mathcal{P}$. Typically the $c^\dagger_{\sigma\alpha}$ is expanded in terms of the two valence

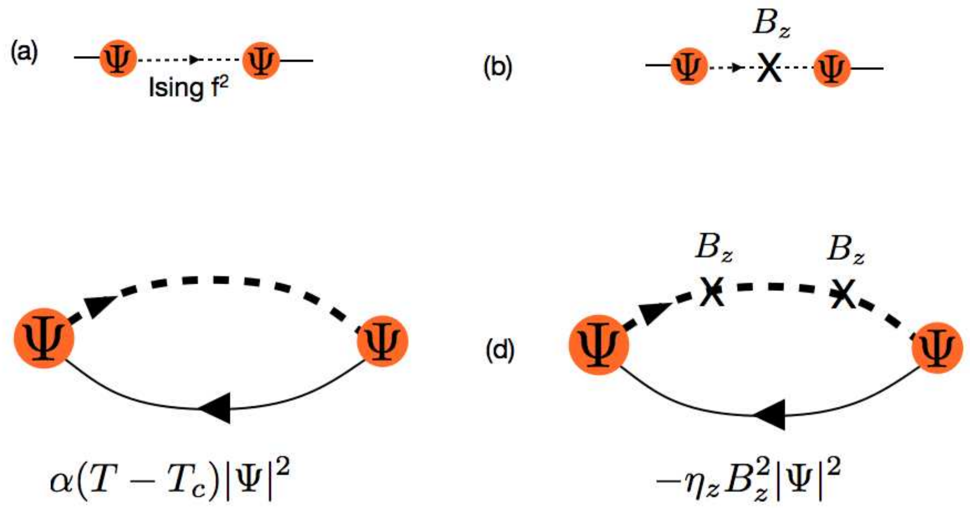

Figure 1. Showing (a) resonant Kondo scattering via hybridization off non-Kramers $5f^2$ doublet, (b) coupling to magnetic field via f-state is purely Ising like (c) zero field quadratic contribution to the Landau energy (d) Ising field-dependent contribution $-B_z^2\Psi^2$ term in the Landau theory.

fluctuation channels $\lambda = 1, 2 \equiv \Gamma_{6,7}$ as follows

$$c^\dagger_{\sigma\alpha} = \sum_{\lambda=1,2} c^\dagger_{\lambda\sigma'}\Gamma^{\sigma'}_{\sigma\alpha}(\lambda), \qquad \Gamma^{\sigma'}_{\sigma\alpha}(\lambda) = \langle 5f^2\alpha|f_{\lambda\sigma'}|5f^3\sigma\rangle. \tag{4}$$

where $c^\dagger_{\lambda\sigma} = \sum_{\mathbf{k},\sigma'}\gamma^\lambda_{\sigma\sigma'}(\mathbf{k})c^\dagger_{\mathbf{k}\sigma}e^{-i\mathbf{k}\cdot\mathbf{R}_j}$ defines the spin-orbit coupled Wannier states of the conduction holes in terms of the crystal-field form-factors $\gamma^\lambda_{\sigma\sigma'}(\mathbf{k})$ and plane wave conduction states.

If we represent $X_{\sigma\alpha} = \hat{\Psi}^\dagger_\sigma f_\alpha$ using a slave boson decomposition, where $\Psi^\dagger_\sigma$ is a slave boson describing the excited Kramers' doublet $\hat{\Psi}^\dagger_\sigma|0\rangle \equiv |5f^3,\sigma\rangle$ while $f^\dagger_\alpha$ is an Abrikosov pseudo-fermion describing the low-lying non-Kramer's doublet $f^\dagger_\alpha|0\rangle \equiv |5f^2,\alpha\rangle$. Hastatic order corresponds to the condensation of the slave boson,

$$X_{\sigma\alpha} \to \langle\hat{\Psi}^\dagger_\sigma\rangle f_\alpha \equiv \Psi^*_\sigma f_\alpha \tag{5}$$

The condensed slave boson defines the two-component Hastatic order parameter, giving rise to hybridization terms of the form shown in Fig. 1 (a). Although this term looks like a conventional hybridization in a heavy fermion system, it breaks time reversal symmetry in a new way, by mixing an integer spin non-Kramers doublet with a half-integer spin conduction electron. The coherent, broken symmetry physics of the HO phase is is determined by the hybridization Hamiltonian

$$H^*(j) = V\left[\Psi^*_\sigma c^\dagger_{\sigma\alpha}(j)f_\alpha(j) + \text{H.c}\right] + \epsilon|\Psi|^2 \tag{6}$$

Let us now return to the anomaly in χ_3. In the field-dependent Landau theory of hastatic order, we get the first temperature-dependent term (Fig. 1 a) from the hybridization of the conduction electrons and the Ising f-moments. We can obtain the Landau theory from a Feynman diagram expansion of the Free energy in powers of $\langle\Psi\rangle$, as illustrated in Fig. 1. Applying a magnetic field, the conduction electrons couple to it isotropically, whereas the non-Kramers doublet couples linearly only to B_z(Fig. 1b). Thus the resulting field-dependent Landau theory, depicted schematically in Fig1c and Fig1d, is

$$f[\psi] = [\alpha(T - T_c) - \eta_z B_z^2 - \eta_\perp B_\perp^2]\Psi^2 + \beta\Psi^4 \tag{7}$$

where $B_z = B\cos\theta$ and $B_\perp = B\sin\theta$. Minimizing the free energy with respect to Ψ, we obtain

$$f = -\frac{[\alpha(T_c - T) + B^2(\eta_z \cos^2\theta + \eta_\perp \sin^2\theta)]^2}{4\beta} \tag{8}$$

from which

$$\Delta\chi_3 = -\frac{\partial^4 f}{\partial B^4} = \frac{6}{\beta}(\eta_z \cos^2\theta + \eta_\perp \sin^2\theta)^2$$

We can calculate η_z and $\eta_\perp$ within a two-channel Anderson lattice model of hastatic order [21], and we find that $\frac{\eta_\perp}{\eta_z} = \frac{T_0^2}{D^2}$ where T_o and D are the hidden order transition temperature and the bandwidth respectively. Taking a conservative $\frac{T_0}{D} \sim \frac{1}{30}$, leads to an anisotropy of 10^3 in $\frac{d\chi_1}{dT}$ and thus a factor of 10^6 in $\Delta\chi_3$. Of course our model is simplified and there will be many material features (e.g. f-electron contributions to η_z arising from fluctuations to excited states) that will reduce the anisotropy. The crucial point here is that the anisotropy in the χ_3 anomaly will be several orders of magnitude greater than the single-ion anisotropy in χ_1, and that $\chi_3 \propto \cos^4\theta$ where θ is the angle from the c-axis. This enormous Ising anisotropy in the anomaly of χ_3 is a natural consequence of hastatic order and is one of its many testable consequences. Careful new measurements to determine the angular dependence of $\Delta\chi_3$ would verify the Ising nature of the quasiparticles.

In conclusion we have shown that hybridization between half-integer conduction electrons and spin-half non-Kramers doublets provides a natural microscopic explanation for the large anomaly observed in the nonlinear susceptibility anomaly[3, 4] in URu_2Si_2. Here the f-moments couple only to the c-axis component of the applied field and, via hybridization, transfer their Ising anisotropy to the conduction electrons. These mobile Ising quasiparticles form Cooper pairs at lower temperatures, and the observable consequences of their hastatic nature in the superconducting state remain to be explored.

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