Nonlinear susceptibility measurements in heavy fermion systems

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Abstract

We discuss the nonlinear susceptibility ($\chi_3$) as a probe of higher-order spin correlations in heavy fermion materials, with particular emphasis on $\text{URu}_2\text{Si}_2$ and $\text{UBe}_1\text{3}$. The sharp discontinuity of $\chi_3$ in $\text{URu}_2\text{Si}_2$ is contrasted with two other antiferromagnets $\text{U}_2\text{Zn}_{1.7}$ and $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$ with larger moments. Nonlinear magnetic properties of $\text{URu}_2\text{Si}_2$ can be understood simply in terms of Landau-Ginzberg theory for a large hidden, itinerant order parameter. In $\text{UBe}_1\text{3}$ we discuss how $\chi_3$ can be used to probe the presence of quadrupolar fluctuations.

Heavy fermion materials display a rich variety of "strong correlation" phenomena, and have attracted much interest as candidates for unconventional electron pairing [1]. These compounds contain a dense lattice of magnetic rare earth or actinide ions embedded in a conducting host. At low temperatures, the spin degrees of freedom in the local moments correlate with the surrounding conduction electrons to severely modify the properties of the metal. A feature shared by several of these heavy fermion systems is the presence of distinctive low-energy antiferromagnetic spin fluctuations, often accompanied by a very small staggered magnetization [2, 3]. Clearly, further characterization of this novel spin ordering is a crucial step towards a better understanding of the many-body ground-state. In this paper we discuss the nonlinear susceptibility ($\chi_3$) as a probe of multispin correlations with particular emphasis on two cases ($\text{URu}_2\text{Si}_2$ and $\text{UBe}_1\text{3}$) where $\chi_3$ measurements can provide constraints on the microscopic nature of the spin ordering process.

Traditionally, the leading-order nonlinear contribution to the magnetization expansion

$$M = \chi_1 H + \frac{1}{3!} \chi_3 H^3 + \cdots$$

$$= \sum_{n=1}^{\infty} \frac{1}{(2n - 1)!} \chi_{2n-1} H^{2n-1}$$

has been used to probe Edwards-Anderson order in spin glasses [4]. Morin and Schmitt [5] pioneered the

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generalization of this method to nonrandom spin systems, using it to probe multipolar interactions in the paramagnetic phases of rare-earth compounds. We note that the development of irreducible two-spin order is signalled by a positive divergence in $\chi_3$, analogous to the behavior of $\chi_1$ in the conventional dipolar case; nonlinear susceptibility measurements are thus particularly useful in materials where the possibility of unconventional spin-ordering already exists.

The heavy-fermion compound $\text{URu}_2\text{Si}_2$ is a particularly clear-cut case that defies description in terms of traditional antiferromagnetism, and thus serves as a natural starting point for our discussion here. In this system anomalies in the susceptibility, specific heat and the resistivity at $T_N = 17.3\, \text{K}$ indicate a magnetic transition [6–8], the accompanying gap in the magnetic excitation spectrum suggests the formation of a spin density wave at this temperature [9]. However, the tiny observed moment ($\mu_0 = 0.04\, \mu_B$) cannot account for the large entropy loss [2] at the transition and thus suggests multispin ordering [10].

Nonlinear susceptibility measurements on $\text{URu}_2\text{Si}_2$, discussed in detail elsewhere [11, 12] reveal a sharp anomaly in $\chi_3$ at $T_N = 17.3\, \text{K}$ that tracks closely with the structure of the mean-field specific heat at this transition (see Fig. 1). This result can be derived from a simple mean-field scaling hypothesis for the free energy

$$F(t, H) = F(T - T_N(H^2)).$$

(2)

where the only field-dependence enters through the transition temperature. This is most clearly seen within the framework of a generic Landau-Ginzburg theory

$$F = \alpha (t - 1)\psi^2 + \frac{\beta}{2} \psi^4 + F_c,$$

(3)

where $t \equiv T/T_N$, and $\psi$ and $H$ are the magnetic order parameters and field, respectively; a finite $\Delta\chi_3(T_N)$ is generated by a coupling $F_c = -\eta H^2$, which implies the development of an induced quadrupolar moment $Q_2 \sim \eta \psi^2$. Minimizing Eq. (3) with respect to the order parameter ($\partial F/\partial \psi^2 = 0$), we find the free energy

$$F = -\frac{x}{2\beta} \left\{ t - \left( 1 + \frac{\eta H^2}{2x} \right) \right\}^2,$$

(4)

which has the same form as Eq. (2). Near the transition, this approach leads to the expression

$$\chi_3 \sim \frac{\chi_1}{T},$$

(5)

where $\chi_1 \equiv \hat{\chi}_1/\gamma T$ and $\gamma \equiv \epsilon_s(T)/T$; we expect the quantity $a = 3x_1/\gamma$ to vary smoothly near the transition.

Thus in principle a finite $\Delta\chi_3$ could occur at a conventional spin density wave transition; in this case $\eta = \mu_0^2$ and the Landau-Ginzburg relation (5) cannot be satisfied by the observed coexistence of a small moment and large specific heat and susceptibility anomalies [12, 13].

The nonlinear susceptibility measurements on $\text{URu}_2\text{Si}_2$ lead us to the conclusion that the magnetic transition at $T_N = 17.3\, \text{K}$ is mean-field in character, thus suggesting itinerant spin ordering in agreement with the observed behavior of $\rho(T)$, To reemphasize this point, we contrast the excellent $\chi_3$ mean-field fit for $\text{URu}_2\text{Si}_2$ with that of $\text{U}_2\text{Zn}_{17}$ and $\text{U}_{0.05}\text{Th}_{0.05}\text{Pt}_3$ (Fig. 1); in these two latter heavy fermion antiferromagnets the spin ordering involves strong fluctuations that are inconsistent with the underlying assumptions leading to the simple scaling form (2). By contrast, the mean-field nature of the experimental data for $\text{URu}_2\text{Si}_2$ is consistent with nonlocal itinerant spin ordering at $T_N$ of the general form

$$\langle \Psi^*(x)\sigma^z \Psi(x') \rangle = f(x, x').$$

(6)
where \(f(x, x')\) is a spin-pairing wave function that has an effective node at the origin and must break time-reversal symmetry [12]. The observed divergence in \(\chi_3\) rules out the formation of isotropic singlets as a possible explanation of the spin-order [14]; it is also difficult to reconcile with an irreducible three-spin order parameter [10] which would have its dominant singularity in \(\chi_5\).

\(\chi_3\) is another good candidate for unconventional magnetic correlations. Unlike the situation in the rare-earth compounds, photoemission does not provide an unambiguous identification of the valence of the uranium ion [15] and quasielastic crystal-field features are difficult to resolve in inelastic neutron scattering experiments [16]. More specifically two rival models associated with two distinct ground-state configurations, \(5f^2 (U^4+)\) and \(5f^3 (U^5+)\), have been proposed and this controversy remains unresolved. In the conventional magnetic picture (\(5f^2\)), a low-lying Kramers doublet provides the driving force for the heavy-fermion physics, whereas the even–even \(5f^2\) configuration yields nonmagnetic low-lying crystal-field excitations in this cubic environment [17]. Emphasizing several anomalous experimental results, Cox [18] has proposed a quadrupolar ground-state \(I_3\) in this \(J = 4\) manifold for UBe\(_{13}\); fluctuations within this non-Kramers doublet would then result in the quadrupolar Kondo effect and the possibility of a non-Fermi-liquid ground-state.

The nonlinear susceptibility \(\chi_3\) is an ideal means of discerning between these two very different scenarios for UBe\(_{13}\). For a quadrupolar doublet with energy splitting

\[
\Delta E = \{zW\} H^2 = H_{\text{eff}},
\]

the resulting free energy,

\[
F = F_0 - \frac{1}{2T} (H_{\text{eff}})^2,
\]

yields

\[
\chi_3 \sim \frac{(zW)^2}{T}.
\]

analogous to the standard Curie susceptibility for a dipolar magnet. In general \(W\) will be anisotropic in a cubic environment; by contrast the nonlinear susceptibility associated with a Kramer doublet is negative \([\chi_3 \sim -1/T^3]\) and isotropic. We note that in heavy fermion systems we expect the single-ion divergences in \(\chi_3\) to be quenched at low temperatures by the Kondo effect but the qualitative features distinguishing the two models should remain. Detailed nonlinear susceptibility measurements are currently in progress on single-crystal samples of UBe\(_{13}\), and the results will be reported in a future publication.

In conclusion, we have discussed the nonlinear susceptibility as a tool to probe multispin order in heavy fermion systems with specific applications to URu\(_2Si_2\) and UBe\(_{13}\). In URu\(_2Si_2\), \(\chi_3\) measurements provide us with constraints on the form of the itinerant spin-order parameter and allow us to rule out several proposed models. In UBe\(_{13}\) the possibility of a quadrupolar Kondo effect is intriguing; careful \(\chi_3\) studies will allow us to unambiguously identify the nature of the low-lying crystal-field doublet as a first step towards characterizing its complex many-body ground-state.

References


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