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PHYSICA B

Nonlinear susceptibility measurements in heavy fermion systems

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Abstract

We discuss the nonlinear susceptibility (χ_3) as a probe of higher-order spin correlations in heavy fermion materials, with particular emphasis on URu_2Si_2 and UBe_{13} . The sharp discontinuity of χ_3 in URu_2Si_2 is contrasted with two other antiferromagnets U_2Zn_{17} and $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$ with larger moments. Nonlinear magnetic properties of URu_2Si_2 can be understood simply in terms of Landau-Ginzberg theory for a large hidden, itinerant order parameter. In UBe_{13} we discuss how χ_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 (χ_3) as a probe of multispin correlations with particular emphasis on two cases (URu_2Si_2 and UBe_{13}) where χ_3 measurements can provide constraints on the microscopic nature of the spin ordering process.

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

$$\begin{aligned} M &= \chi_1 H + \frac{1}{3!} \chi_3 H^3 + \dots \\ &= \sum_{n=1} \frac{1}{(2n-1)!} \chi_{2n-1} H^{2n-1} \end{aligned} \quad (1)$$

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 χ_3 , analogous to the behavior of χ_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 URu_2Si_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 $T_c = 17.3$ 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 URu_2Si_2 , discussed in detail elsewhere [11, 12] reveal a sharp anomaly in χ_3 at $T_c = 17.3$ 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

$$F[T, H] = F[T - T_N(H^2)], \quad (2)$$

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

$$F = \alpha(t - 1)\psi^2 + \frac{\beta}{2}\psi^4 + F_c, \quad (3)$$

where $t \equiv T/T_N$, and ψ and H are the magnetic order parameters and field, respectively; a finite $\Delta\chi_3(T_N)$ is generated by a coupling $F_c = -\eta\psi^2H^2$, 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

$$F = -\frac{\alpha}{2\beta} \left\{ t - \left(1 + \frac{\eta H^2}{2\alpha} \right) \right\}^2, \quad (4)$$

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

$$\chi_3 = \left\{ \frac{3\chi'_1}{\gamma} \right\} \chi'_1, \quad (5)$$

where $\chi'_1 \equiv \partial\chi_1/\partial T$ and $\gamma \equiv c_v(T)/T$; we expect the quantity $a \equiv 3a\chi'_1/\gamma$ to vary smoothly near the transition.

Fig. 1. Comparison of χ_3 and $a\chi'_1$ for (a) URu_2Si_2 , (b) U_2Zn_{17} , and (c) $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$ with $a = 45.132$ and $126 \text{ mK}/T^2$, respectively ($a \equiv 3a\chi'_1/\gamma$, $\chi'_1 \equiv \partial\chi_1/\partial T$ and $\gamma \equiv c_v(T)/T$) contrasting the mean-field character of the transition in URu_2Si_2 with that in the fluctuation-dominated materials U_2Zn_{17} and $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$.

Thus in principle a finite $\Delta\chi_3$ could occur at a conventional spin density wave transition; in this case $\eta \sim \mu_0^2$ 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 URu_2Si_2 lead us to the conclusion that the magnetic transition at $T_N = 17.3$ 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 χ_3 mean-field fit for URu_2Si_2 with that of U_2Zn_{17} and $\text{U}_{0.95}\text{Th}_{0.05}\text{Pt}_3$ (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 URu_2Si_2 is consistent with nonlocal *itinerant* spin ordering at T_N of the general form

$$\langle \Psi^\dagger(x) \sigma^z \Psi(x') \rangle = f(x, x'), \quad (6)$$

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 χ_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 χ_5 .

UBe₁₃ 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 ($5f^3$), 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 Γ_3 in this $J = 4$ manifold for UBe₁₃; fluctuations within this *non-Kramers* doublet would then result in the quadrupolar Kondo effect and the possibility of a non-Fermi-liquid ground-state [19].

The nonlinear susceptibility (χ_3) is an ideal means of discerning between these two very different scenarios for UBe₁₃. For a quadrupolar doublet with energy splitting

$$\Delta E = \{\alpha W\} H^2 = H_{\text{eff}}, \quad (7)$$

the resulting free energy,

$$F \sim F_0 - \frac{1}{2T} (H_{\text{eff}})^2, \quad (8)$$

yields

$$\chi_3 \sim \frac{(\alpha W)^2}{T}, \quad (9)$$

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 χ_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 UBe₁₃, 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 URu₂Si₂ and UBe₁₃. In URu₂Si₂, χ_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 UBe₁₃ the possibility of a quadrupolar Kondo effect is intriguing; careful χ_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.

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