Nonlinear Susceptibility as a Probe of Tensor Spin Order in URu_2Si_2

A. P. Ramirez,[1] P. Coleman,[2] P. Chandra,[3] E. Brück,[4] A. A. Menovsky,[4] Z. Fisk,[5] and E. Bucher[1,6]

[1]*AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974*
[2]*Serin Physics Laboratory, Rutgers University, P.O. Box 849, Piscataway, New Jersey 08854*
[3]*NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540*
[4]*Natuurkundig Laboratorium Der Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands*
[5]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*
[6]*Universität Konstanz, D7750 Konstanz, Germany*

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The nonlinear susceptibility is discussed as a probe of multispin correlation functions. In the heavy-fermion compound URu_2Si_2, measurements of the leading nonlinear susceptibility (χ_3) reveal a sharp anomaly at 17.3 K closely matching that of the specific heat at the same temperature. We attribute this result to the development of anisotropic triplet pairing in the particle-hole channel of the heavy electron fluid.

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Heavy-fermion compounds have attracted much interest as candidates for unconventional electron pairing [1]. These systems exhibit marked low-energy antiferromagnetic spin fluctuations often accompanied by a very small staggered magnetization [2,3]. URu_2Si_2 is a particularly dramatic example; here specific-heat and susceptibility anomalies [4–6] indicate a magnetic transition at 17.3 K despite a tiny observed moment ($\mu_0 \sim 0.04\mu_B$) that cannot account for the entropy loss at this temperature [2]. In this Letter we present nonlinear susceptibility measurements on URu_2Si_2 that probe *tensor* spin order; they suggest that an itinerant quadrupolar order parameter drives this mysterious transition.

The nonlinear susceptibility was first introduced as a direct probe of Edwards-Anderson order-parameter fluctuations in spin glasses [7]. In nonrandom systems this method can be generalized to multipolar moment fluctuations; to date, it has been predominantly used as a probe of quadrupolar interactions in rare-earth compounds [8]. These studies focused on the paramagnetic phase, with the aim of probing the uncorrelated multispin fluctuation spectrum. By contrast, here we examine the phase transition behavior of the nonlinear susceptibility in a manifestly itinerant magnet. In general terms, the magnetization in the direction of the applied field in the paramagnetic state is

$$M = \chi_1 H + \frac{1}{3!}\chi_3 H^3 + \cdots = \sum_{n=1} \frac{1}{(2n-1)!}\chi_{2n-1} H^n , \quad (1)$$

where χ_{2n-1} is proportional to the $2n$th irreducible (I) moment of the magnetization

$$\chi_{2n-1} = \langle M^{2n}\rangle_I / T^{2n-1} \quad (2)$$

and T is the temperature. The development of uniform spin order involving the n-spin (irreducible) order parameter

$$q_n = \langle Q_n \rangle = \frac{1}{V}\int d1 \cdots dn \langle S(1)S(2)\cdots S(n)\rangle_I \quad (3)$$

(S is the spin component in the direction of the applied field) is signaled by a positive divergence in the corresponding nonlinear susceptibility

$$\chi_{2n-1} \sim \langle(\delta Q_n)^2\rangle / T^{2n-1} \to \infty \quad (4)$$

in the absence of singularities in lower-order terms.

The leading nonlinear contribution in the magnetization expansion, χ_3, is of particular interest. In a collinear antiferromagnet, χ_3 passes through zero near T_N: $\chi_3 < 0$ for $T > T_N$ reflecting the negative curvature of the Brillouin function in finite field, whereas $\chi_3 > 0$ for $T < T_N$ due to the presence of quadrupolar fluctuations induced by a finite magnetization. Alternatively, the development of *quadrupolar* spin order is signalled by a positive divergence in χ_3, analogous to the behavior of χ_1 in the dipolar case. We note that this discussion can be easily extended to spins with continuous symmetry where a positive divergence in χ_3 would signal the development of *tensor* magnetism [9–11].

The heavy-fermion compound URu_2Si_2 is a strong candidate for exotic spin ordering [12]. Here specific-heat, susceptibility, and resistivity anomalies [4–6] at 17.3 K are accompanied by a gap in the magnetic excitation spectrum [13], suggestive of a spin-density-wave transition. However, the observed moment below this temperature, $\mu_0 = 0.04\mu_B$ [2], cannot account for the large entropy reduction at the transition. Specifically, the moment loss parameter $\tilde{m}(\mu_0/\mu_{\rm eff})^2/[\gamma(T_N) - \gamma(0)/\gamma(T_N)]$ is the ratio of the mean-square fluctuating moment (normalized to the high-temperature value $\mu_{\rm eff}$) to the fraction of the Fermi surface participating in the gap [$\gamma \equiv C(T)/T$]; for complete dipole formation $\tilde{m} = 1$, but for URu_2Si_2 $\tilde{m} \sim 10^{-2}$, suggesting the possibility of higher-order multipolar order [2].

For the present measurement of χ_3 in URu_2Si_2, we used high-quality single crystals described elsewhere [14]. In particular, these crystals showed no sign of feromagnetism at 35 K, indicative of stacking fault defects. Measurements of M vs H up to 5 T were made with a commercial magnetometer. χ_3 was determined by fitting

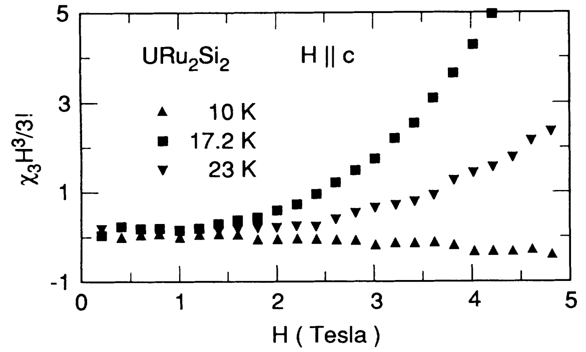

FIG. 1. The leading nonlinear term, $\chi_3 H^3/3!$, in the magnetization of URu_2Si_2, for H parallel to the tetragonal c axis, at three different temperatures spanning the transition at 17.3 K.

the data by Eq. (1), truncated at the cubic term. The deviation from linearity was small so higher-order terms could not be determined reliably. Figure 1 shows raw data (with the linear term subtracted) at different temperatures, indicating a strong temperature dependence of χ_3 in the vicinity of $T_c = 17$ K. The linear susceptibility χ_1 obtained from the fits is displayed in the lower frame of Fig. 2, and there is good agreement with previously reported data [4-6] at T_c. In contrast with the χ_1 results, χ_3, shown in the upper half of Fig. 2, exhibits a strong anomaly with the shape evocative of $\gamma = C(T)/T$. Also note the temperature *independence* of χ_3 above T_c (Fig. 2). This feature cannot be explained using local moments at finite temperatures, but instead suggests a quadrupolar analog of the Pauli susceptibility. Finally, the anisotropy of χ_3 reflects the Ising nature of spins, displayed already in χ_1.

In order to determine the possible generality of this effect to other heavy-fermion systems, we performed the same study on two other materials, U_2Zn_{17} and $U_{0.95}Th_{0.05}Pt_3$. U_2Zn_{17} was one of the first heavy-fermion compounds found to possess magnetic order, with a collinear antiferromagnetically ordered state with spins in the basal plane of the rhombohedral lattice [15]. Its ordered moment is $\mu_0 = (0.8 \pm 0.1)\mu_B$ per uranium atom, which is smaller than the free U moment of $\mu_{\text{eff}} = 2.25\mu_B$; nevertheless, $\tilde{m} = 0.6$ [15]. In $U_{0.95}Th_{0.05}Pt_3$, a spin-density-wave transition at 6 K was found to replace part of the superconducting Fermi-surface instability observed in the pure material. Its order is also collinear with spins lying in the hexagonal basal plane [16]. In $U_{0.95}Th_{0.05}Pt_3$, $\mu_0 = (0.65 \pm 0.1)\mu_B$, but like U_2Zn_{17}, the change in γ is correspondingly small [17]; $\tilde{m} = 0.5$, as expected for Fermi-surface instabilities to ground states with dipolelike ordered moments.

The samples used in the present study are from the same batches as described elsewhere [15,16]. In Figs. 3 and 4, χ_3 for these two materials is shown. Unlike χ_3 in URu_2Si_2, χ_3 for these compounds is well behaved; it follows the expected response for a normal antiferromagnet,

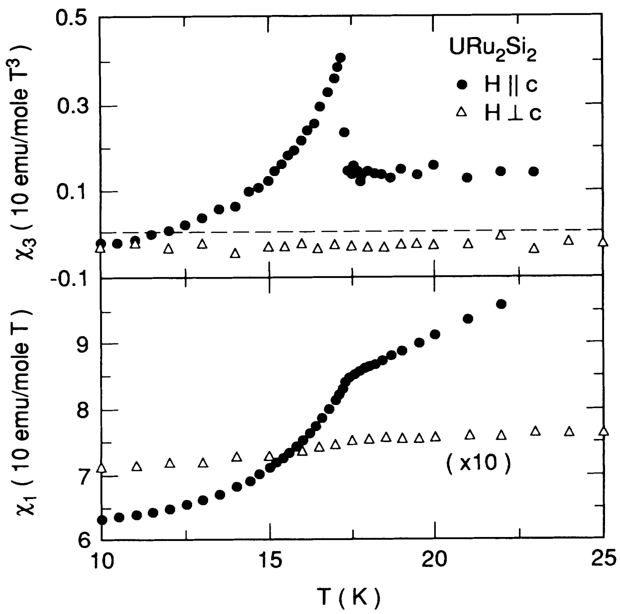

FIG. 2. Nonlinear susceptibility χ_3 and linear susceptibility χ_1 for URu_2Si_2. The response is shown for both the easy magnetic direction, parallel to the tetragonal c axis (solid symbols), and in the hard direction perpendicular to the c axis (open symbols). The peak in χ_3 at the same temperature where the specific heat peaks illustrates the predominance of high-order spin correlations for establishing magnetic order.

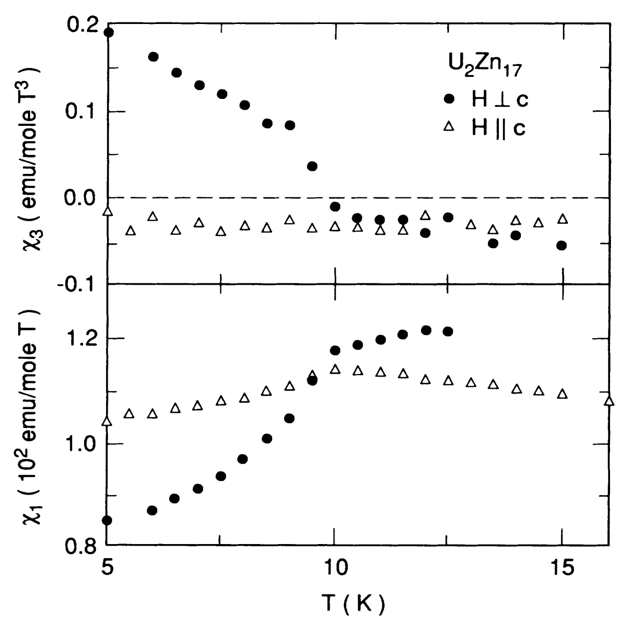

FIG. 3. Nonlinear susceptibility χ_3 and linear susceptibility χ_1 for U_2Zn_{17}. The response is shown for both the easy magnetic direction perpendicular to the rhombohedral c axis (solid symbols) and in the hard direction parallel to the c axis (open symbols). The vanishing of χ_3 near T_N is the expected behavior for a conventional antiferromagnet.

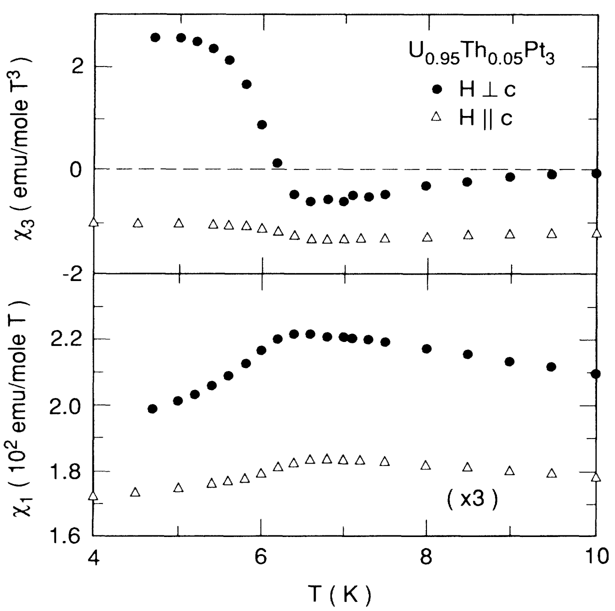

FIG. 4. Nonlinear susceptibility χ_3 and linear susceptibility χ_1 for $U_{0.95}Th_{0.05}Pt_3$. The response is shown for both the easy magnetic direction, perpendicular to the hexagonal c axis (solid symbols) and in the hard direction, parallel to the c axis (open symbols). The vanishing of χ_3 near T_N is the expected behavior for a conventional antiferromagnet.

passing through zero at T_N. We conclude, therefore, that the sharp anomaly in URu_2Si_2 is a result of the mismatch, as measured by $\tilde{m}$, between the ordered moment as observed by neutrons and the entropy loss at the transition.

We now turn to the interpretation of these results. Within a simple Landau-Ginzburg theory, a finite $\Delta\chi_3(T_N)$ is generated by a coupling $F_e = -\frac{1}{2}\eta\psi^2 B^2$ where ψ and B are the order parameter and the magnetic field, respectively; this additional term implies the development of a *quadrupolar* moment $Q_2 = -\partial^2 F/\partial B^2 = \eta\psi^2$. Thus, a finite $\Delta\chi_3$ could occur at a conventional spin-density-wave transition; in this case $(\eta \sim \mu_0^2)$ the Landau-Ginzburg theory predicts $(\Delta c/T_N)(T_N^3\Delta\chi_3) = 3(\mu_0^4)$. The observed specific heat and nonlinear susceptibilities anomalies are clearly *inconsistent* with the tiny observed moment [2] $(\mu_0 \sim 0.04\mu_B)$, and the data demand a new order parameter. Since the nonlinear susceptibility in the z direction scales with $\gamma = C(T)/T$, we conclude that the autocorrelation function for the energy fluctuations near the transition at $T = 17.3$ K is directly proportional to the fluctuations in the second moment of the magnetization:

$$\gamma = \langle \delta E^2 \rangle / T^3, \quad \chi_3 = \langle (\delta Q_2)^2 \rangle / T^3,$$
$$\gamma \sim \chi_3 \Rightarrow \langle \delta E^2 \rangle \sim \langle (\delta Q_2)^2 \rangle . \tag{5}$$

In other words, the dominant energy component responsible for the sharp specific-heat anomaly

$$H \sim \sum J_{ij} Q_{ij}^z ,$$
$$Q_{ij}^z = \langle S_i^z S_j^z \rangle_I = S_i^z S_j^z - \langle S_i^z \rangle \langle S_j^z \rangle , \tag{6}$$

is the *irreducible* (quadrupolar) part of the standard Ising coupling.

The sharp mean-field specific-heat anomaly at the spin-ordering transition in URu_2Si_2 suggests that it involves a large number of electrons within the relevant coherence length (i.e., $n/\xi^3 \gg 1$). This transition is probably dominated by *itinerant* spin ordering in the heavy-fermion band of form

$$\langle \psi^\dagger(\mathbf{x}) \sigma^z \psi(\mathbf{x}') \rangle = f(\mathbf{x},\mathbf{x}') , \tag{7}$$

where $f(\mathbf{x},\mathbf{x}')$ is a spin-pairing wave function, closely analogous to that of anisotropic superconductors. In URu_2Si_2 it appears that the s-wave component of f, which determines the local vector magnetic order, is effectively absent. The nonlocal coupling (6) favors anisotropic pairing, possibly a staggered wave function of the form

$$f(\mathbf{x},\mathbf{x}') = g(\mathbf{x}-\mathbf{x}') \cos \mathbf{G} \cdot (\mathbf{x}+\mathbf{x}')/2 , \tag{8}$$

where $\mathbf{G} = (0,0,\pi)$; staggered order results in a gap in the excitation spectrum, accounting for the exponential behavior of the low-temperature specific heat [4], the loss of Fermi-surface density of states [13], and the large Fermi-surface reorganization suggested by the large increase in Hall constant below the transition [18]. The fundamental order parameter $f(x,x')$ *breaks* time-reversal symmetry, unlike the case at a conventional quadrupolar transition. This allows it to couple *linearly* to the staggered magnetic order $E_{LG} = -\eta(mf)$, resulting in a small induced moment $m \propto (\eta f) \sim (T_c - T)^{1/2}$ within mean-field theory. This order parameter will generate a quadrupolar moment $Q_2 \propto |f(x,x')|^2 \sim T_c - T$. We expect several physical variables to couple to Q_2; the magnetoresistance, the elastic constants, and the anisotropic magnetostriction should all display anomalies that increase linearly with decreasing temperature, consistent with experimental observation [19–21].

In principle, one could consider more complex multispin order parameters as driving the $T = 17.3$ K transition in URu_2Si_2 [12]; however, their associated correlation functions would not yield the observed signal in χ_3. In particular, the three-spin order parameter $\langle S(1)S(2)S(3)\rangle_I$ proposed by Gorkov [12] yields a leading singularity in χ_5, with small secondary effects in χ_1 and χ_3 comparable in magnitude to the observed vector moment $(\mu_0 \sim 0.04\mu_B)$ [2]. Similarly the formation of isotropic singlets [22], a proposed explanation for the spin ordering, cannot easily account for the observed divergence in χ_3. The development of staggered quadrupolar order is the most likely scenario; this permits the development of a perfect gap among the pairing electrons, consistent with the exponential form of the specific heat below 17.3 K [23].

In conclusion, we have presented the nonlinear susceptibility as a probe for examining multispin correlations. Measurements on the heavy-fermion metal URu_2Si_2 indicate an anomalous χ_3 at $T=17.3$ K that tracks with the mean-field specific-heat structure at this transition. Further measurements on this compound, including finite-field χ_3 and tunneling studies, would confirm the nature of this ordering. Multipolar order has been proposed for a number of rare-earth pnictides that display coupled magnetic and structural transitions [24,25]; the nonlinear susceptibility, measured in the ordered phases, would be an ideal means of confirming these suggestions. Similarly, anomalies in the low-temperature specific heat, susceptibility, and resistivity of the actinide compound UPd_3 [26] have been attributed to quadrupolar interactions; similar studies as presented here should be performed on this system. Finally muon-spin-resonance and specific-heat measurements indicate a moment loss value $\tilde{m} \leq 10^{-2}$ for $U_{1-x}Th_xBe_{13}$ at a transition below the normal-superconducting one [27]; as in URu_2Si_2 the enigmatic nature of the superconducting phase may be determined by its coexistence with exotic spin order.

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