

Metamagnetism and the fifth-order susceptibility in UPt_3

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An enhanced susceptibility is a natural consequence of the “heavy fermion” (HF) state, rendering the possibility of measurably large nonlinear susceptibilities. In recent work a universal behavior of the peaks observed in the linear (χ_1) and the third-order (χ_3) susceptibility in HF metamagnets has been identified. This universality is well accounted for by a single energy scale model considering on-site correlations only. A prediction of this model is a peak in the fifth-order susceptibility χ_5 as well. In measurements on a HF metamagnet, UPt_3 , reported herein, we find that χ_5 , rather than attaining a peak, saturates at low temperatures and is positive. The thermodynamic implications of these towards the stability of the metamagnetic HF state are discussed.

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The heavy electron metals exhibit a fascinating diversity of magnetic properties [1,2]. A significant fraction of them order antiferromagnetically, some even order as ferromagnets, and others remain nonmagnetic, yet they harbor within them other possible exotic types of magnetic order. Proposals of such exotic order include multipolar order such as quadrupolar (ferromagnetic and antiferromagnetic) [3], hexadecapolar [4], hidden order [5,6], etc. Higher-order susceptibilities can be an indicator of multipolar order. At the same time, heavy electron materials also contain physics of Kondo compensation, and, conceivably, nonlinear susceptibilities, caused by the response of Kondo-like compensation to a larger magnetic field. Irrespective of which of the scenarios above is valid, the experimental measurement of successively higher-order dc magnetic susceptibilities is useful.

The Wilson ratio, a measure of the strong correlations, is often close to unity in heavy electron materials. That ensures that the heavy effective mass also translates to a large susceptibility of the conduction electron system [7]. Beginning with such enhanced susceptibilities, it may be surmised that the nonlinear components of the susceptibility might similarly be very large and easily accessed experimentally. This is indeed the case, and a measurement of the third-order susceptibility has been reported in a number of heavy electron systems [8–11]. We have recently reported measurements of χ_3 in UPt_3 where we observe a peak in χ_3 at a temperature approximately half the temperature where a peak in χ_1 is observed [12]. We also noted that this scaling feature is observed in a number of other measurements reported in the literature. We have proposed a single energy scale model (see below) which accounts for the peak in χ_3 , as well as arriving at the correct relationship between the temperatures T_3 and T_1 . Experimentally the ratio T_3/T_1 is close to 1/2 whereas the model gets the ratio to be 0.4. This model also predicts that there is a peak in the fifth-order susceptibility χ_5 at a temperature lower than T_3 .

In this Rapid Communication we present measurements of the fifth-order susceptibility in a heavy electron material. These measurements were performed at the National High Magnetic Field Laboratory, Florida on a single crystal of UPt_3 employing a capacitance torque magnetometer in fields up to 30 T. Capacitance isotherms were measured as the

magnetic field was swept for both orientations of the field with respect to the hexagonal crystallographic axes. The two capacitance isotherms were then deconvoluted to compute the magnetization separately for the field parallel to the c axis as well as the a axis of UPt_3 . This deconvolution procedure has been described in detail elsewhere [13]. The magnetization at low fields may be expanded in odd powers of the applied field H as

$$M = \chi_1 H + \chi_3 H^3 + \chi_5 H^5. \quad (1)$$

Thus, by plotting the ratio M/H as a function of H^2 , we can discern both the third-order and the fifth-order susceptibilities from a quadratic fit to the experimental data with the intercept yielding the linear susceptibility χ_1 .

In Fig. 1 we show the magnetization along the a axis (parallel to the basal plane) for nine different temperatures, as identified in the individual panels. The magnetic isotherms at high temperatures start out similar to those of a conventional paramagnet, with the moment tending to saturation gradually as the field is increased. This “normal” paramagnetic response means that $M - \chi_1 H < 0$, making the third-order susceptibility negative with a negative sign also for the fifth-order susceptibility. As the temperature is lowered, the isotherms “stiffen,” implying χ_3 is becoming less negative, and with a further decrease in temperature an upward curvature in the response develops, thus signaling a positive trend in the nonlinear susceptibilities. At the lowest temperatures a sharp upward curvature in the magnetization at the metamagnetic field is apparent, suggesting that either χ_3 or χ_5 or both are strongly positive. In order to separate out these two susceptibility components we follow the procedure explained above with plots of M/H vs H^2 . These plots are shown in the series of nine panels in Fig. 2. At high temperatures the curves in Fig. 2 have a downward slope and curvature and yield negative values for χ_3 and χ_5 . This negative slope quickly gives way to an almost flat response at $T \sim 14$ K, below which the opposite tendency starts to develop. At the lowest temperatures an upward curvature is clearly visible, with the data for 2 K in particular being a pure quadratic, thus indicating that χ_3 is nearly zero but χ_5 is positive. The individual values of χ_5 obtained through such plots are shown

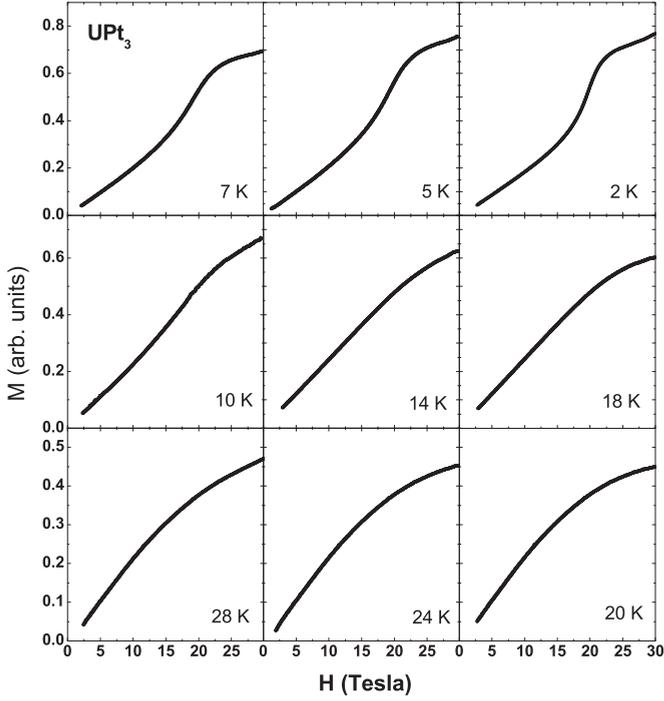


FIG. 1. Magnetization isotherms of UPt_3 with field applied along the a axis. A “normal” paramagnetic response observed at high temperatures with a negative bending gives way to an upward curvature at lower temperatures, signaling the rise of higher-order susceptibilities to positive values prior to the metamagnetic transition itself at 20 T.

in Fig. 3. Here the data have been fit to H^2 values up to 250 T^2 , which represents an order of magnitude increase in scale compared to that employed in our earlier work on χ_3 . The values of χ_5 are negative at high temperatures, turn positive at lower temperatures, crossing zero at approximately 10 K. χ_5 continues to remain positive at the lowest temperatures in this study with no indication of a peak. For purposes of comparison and to establish a scale we show in the figure included in the Supplemental Material [14] the χ_1 as given by the intercept as well as χ_3 obtained from the initial slopes of the fits.

In the work that was reported in Ref. [12] the peak temperatures T_3 and T_1 followed the relation $T_3 \cong T_1/2$. Seeking a simple understanding of this relation (as well as the earlier correlations between the critical field and T_1 and the critical field and inverse of the peak linear susceptibility) we were led to a minimal one energy scale model which contains (a) a metamagnetic transition and (b) an opportunity to calculate $\chi_3(T)$, which seemed to capture the essential details of the observed temperature dependence. The materials we considered are primarily Kondo lattices, likely with incompletely compensated local moments which are also in a crystalline lattice and therefore with electronic energy levels subject to the crystalline electric fields. Our model at the current stage does not depend on the exact crystalline structure. We also cannot assert with any certainty that the metamagnetism is a form of shakeoff of the Kondo cloud. However, it seems certain that we need only one energy scale

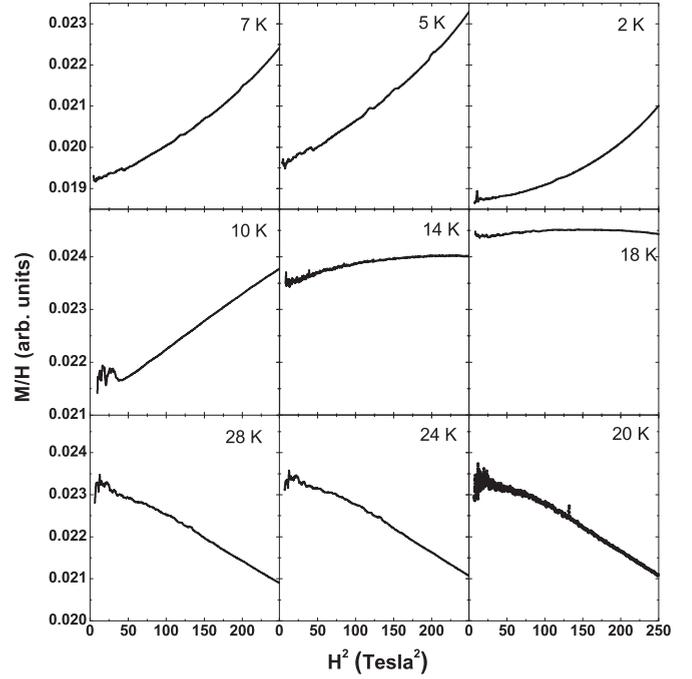


FIG. 2. Nonlinear part of the magnetic response after subtracting out the linear term. The behavior at high temperatures (lower panels) is almost linear and sloping down, thus implying a negative χ_3 . Around 14 K χ_3 turns positive with a definite indication of a negative χ_5 . The response at 10 K is largely a straight line, indicating a large positive χ_3 and χ_5 about to cross over to the positive side. The response at lower temperatures clearly shows both χ_3 and χ_5 are positive, with the exception of the data at 2 K, which is a perfect parabola, indicating a near zero χ_3 .

and the minimal model produces the suggestive functional forms of all measured quantities.

In the minimal model we start with the Hamiltonian $H = \Delta S_z^2 - \gamma S_z B$, where Δ sets the energy scale and could represent the separation between the singlet ground state and the lowest excited state. In this model the quantum spins are

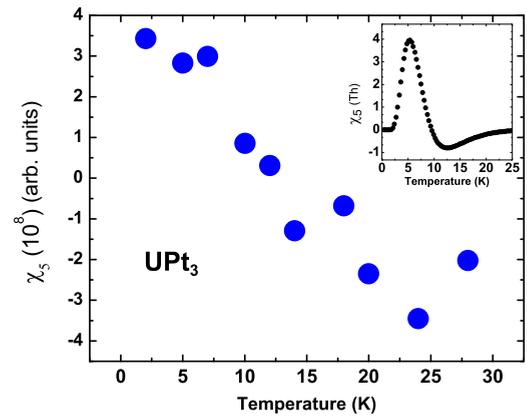


FIG. 3. (Color online) Fifth-order susceptibility χ_5 in UPt_3 obtained from the quadratic term in fits to the data shown in Fig. 2. The vertical scale is derived from that in Fig. 2. χ_5 is negative at high temperatures and increases monotonically as $T \rightarrow 0$. The inset shows the calculated χ_5 as derived in the single energy scale model.

treated as discrete. When the magnetic field is parallel to the quantization axis (z axis), the susceptibilities are calculated as

$$\chi_1 = \frac{\gamma^2}{\Delta} \frac{1}{\tau} \frac{1}{1+A}, \quad (2)$$

$$\chi_3 = \frac{\gamma^4}{3!\Delta^3} \frac{1}{\tau^3} \frac{A-2}{(1+A)^2}, \quad (3)$$

$$\chi_5 = \frac{\gamma^6}{5!\Delta^5} \frac{1}{\tau^5} \frac{A^2 - 13A + 16}{(1+A)^3}, \quad (4)$$

where $A = 0.5e^{1/\tau}$ and $\tau = k_B T/\Delta$. The principal features of $\chi_5(T)$ are similar to those of $\chi_3(T)$. At high temperatures, $\chi_5(T) < 0$. It has a peak at $T_5 = 0.17\Delta$. The value of Δ for UPt_3 (obtained from the measured values of H_m , T_1 , and T_3) is 30 K, and hence we can expect a peak at 5 K [15]. Our data clearly show a rise in χ_5 but no falloff below that temperature. There is no indication of a peak. It is noteworthy that both χ_1 and χ_5 saturate and are positive at the lowest temperatures measured in UPt_3 while the intermediate susceptibility χ_3 is close to zero.

For further analysis we begin with a general expansion of the magnetic free energy for B and M parallel to the z axis:

$$F = -BM + a_2 M^2 + a_4 M^4 + a_6 M^6 + \dots \quad (5)$$

In such an expansion the coefficients a_2 , a_4 , a_6 can be related to the susceptibilities

$$a_2 = \frac{1}{2\chi_1}, \quad (6)$$

$$a_4 = -\chi_3/4\chi_1^4, \quad (7)$$

$$a_6 = \frac{1}{\chi_1^7} [3\chi_3^2 - \chi_5\chi_1]. \quad (8)$$

When all coefficients are positive, the minimum of this free energy is $M = 0$. Since $\chi_1 \geq 0$, it follows that $a_2 > 0$ and there is no transition here driven by the quadratic term. From our measurements, though, we see that $\chi_3(T)$ changes sign and, starting from a negative value at high temperatures ($a_4 \geq 0$), it becomes positive, indicating a potentially discontinuous transition in M . The next coefficient, a_6 , if positive, would keep the free energy bounded. In terms of nonlinear susceptibilities, a_6 is expressed in Eq. (8) and is seen to be negative for $T < 12$ K. It is clear from the data that the positive and growing value of χ_5 below about 10 K has a critical effect on the condition expressed by Eq. (8). The fact that a_6 is about to vanish should have other consequences, and the situation is reminiscent of the B - A transition in superfluid ^3He [16]. The breakdown of the condition as shown in Fig. 4 implies that one or both of the following two scenarios can occur: (a) Higher-order terms such as a_8 are required for keeping the

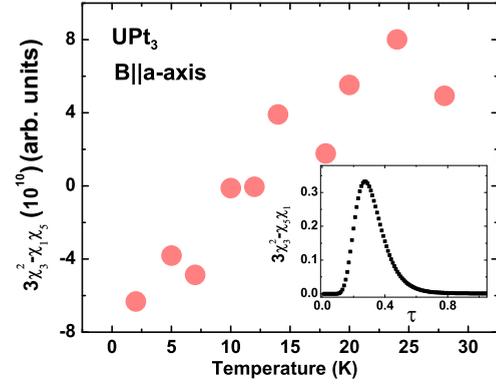


FIG. 4. (Color online) The quantity on the vertical axis (values derived from vertical scale of Fig. 2) is a measure of the stability of the metamagnetic state. At low temperatures (~ 10 K) this quantity assumes a negative value, thus implying that additional terms in the magnetic free energy expansion need to be considered in order to ensure thermodynamic stability. The inset shows the measure of stability [Eq. (8)] as computed from the minimal model. The computed response never crosses zero.

free energy bounded and (b) domains are formed below the instability temperature, thus necessitating gradient terms in the free energy [17]. We note that there is a precedence for the presence of antiferromagnetic domains in UPt_3 in particular [18–20] and in strongly correlated metamagnets in general [21]. Also as noted above, experimentally the $T \rightarrow 0$ value of χ_1 is greater than zero and this is at variance with the single energy scale model. It is quite likely that resolving this issue would also result in additional terms in the free energy, thus preserving a bounded free energy.

In conclusion, we have measured the fifth-order nonlinear magnetic susceptibility $\chi_5(T)$ in the heavy fermion compound UPt_3 . This is shown in Fig. 3, along with the expectation from a single energy scale model which we had developed earlier (shown in the inset). Whereas the model predicts a vanishing $\chi_5(T = 0)$, the experiments show saturation and no turnover into a peak. The experimental data, taken together with a polynomial expansion of the magnetic free energy in powers of the magnetization M confined to sixth order, imply a possible thermodynamic instability around $T = 10$ K. Detailed measurements of higher-order susceptibilities at low temperatures in other heavy fermion systems would be valuable in establishing whether the implied instability is a generic feature in itinerant metamagnets.

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[1] *Magnetism in Heavy Fermion Systems*, edited by H. B. Radousky (World Scientific, Singapore, 2000).

[2] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, UK, 1993).

- [3] P. Morin, J. Rouchy, D. Schmitt, and E. Delacheisserie, *J. Magn. Mater.* **90**, 105 (1990).
- [4] A. I. Tóth and G. Kotliar, *Phys. Rev. Lett.* **107**, 266405 (2011).
- [5] P. Chandra, P. Coleman, J. A. Mydosh, and V. Tripathi, *Nature (London)* **417**, 831 (2002).
- [6] C. M. Varma and L. Zhu, *Phys. Rev. Lett.* **96**, 036405 (2006).
- [7] P. A. Lee, T. M. Rice, J. W. Serene, L. J. Sham, and J. W. Wilkins, *Comments Condens. Matter Phys.* **12**, 99 (1986).
- [8] J. G. Park *et al.*, *J. Phys.: Condens. Matter* **6**, 9383 (1994).
- [9] A. P. Ramirez, P. Chandra, P. Coleman, Z. Fisk, J. L. Smith, and H. R. Ott, *Phys. Rev. Lett.* **73**, 3018 (1994).
- [10] A. Grauel *et al.*, *J. Appl. Phys.* **73**, 5421 (1993).
- [11] E. D. Bauer, P. C. Ho, M. B. Maple, T. Schauerer, D. L. Cox, and F. B. Anders, *Phys. Rev. B* **73**, 094511 (2006).
- [12] B. S. Shivaram, D. G. Hinks, M. C. de Andrade, M. B. Maple, and P. Kumar, [arXiv:1403.0742](https://arxiv.org/abs/1403.0742).
- [13] B. S. Shivaram, [arXiv:1403.1287](https://arxiv.org/abs/1403.1287).
- [14] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.89.161108> for Plots of χ_1 and χ_3 .
- [15] Various models for metamagnetism in heavy fermion systems have been proposed. It is possible to obtain the behavior of the magnetization all the way up to the metamagnetic field in many of these models. Thus the low field fits to the numerical predictions from these models should, in principle, produce plots of χ_5 against temperature. For examples, see M. Bercx and F. F. Assaad, *Phys. Rev. B* **86**, 075108 (2012); S. Viola Kusminskiy, K. S. D. Beach, A. H. Castro Neto, and D. K. Campbell, *ibid.* **77**, 094419 (2008); M. M. Wysokinski *et al.*, *J. Phys.: Condens. Matter* **26**, 055601 (2014); J. Bauer, *Eur. Phys. J. B* **68**, 201 (2009).
- [16] J. Sauls (private communication).
- [17] P. Kumar and B. S. Shivaram (unpublished)
- [18] E. Shung, T. F. Rosenbaum, and M. Sigrist, *Phys. Rev. Lett.* **80**, 1078 (1998).
- [19] A. Amann, A. C. Mota, M. B. Maple, and H. von Lohneysen, *Phys. Rev. B* **57**, 3640 (1998).
- [20] M. Sigrist and D. F. Agterberg, *Prog. Theor. Phys.* **102**, 965 (1999).
- [21] B. Binz, H. B. Braun, T. M. Rice, and M. Sigrist, *Phys. Rev. Lett.* **96**, 196406 (2006).