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Suppression of heavy fermions by high fields in $CeCu_6$

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Measurement of the specific heat of $CeCu_6$ in fields to 24 T between 1.8 and 8 K and of the magnetization in fields to 20 T at 1.3 and 4.2 K is reported. The value of γ ($=C/T$) in 24 T is 350 mJ/mol K^2. The slope of the C/T-vs-T^2 data, proportional to the Debye temperature, in 24 T is the same as that for $LaCu_6$ in zero field, with no low-temperature upturn in the C/T data in 24 T observed. These data indicate that we achieve complete suppression of the heavy-fermion ground state in $CeCu_6$ with this high applied magnetic field. These results are in sharp contrast with results on uranium systems, indicating a fundamental difference in the heavy-fermion ground state.

I. INTRODUCTION

One of the more intriguing questions[1] of recent interest is what causes the formation of the heavy-fermion ground state in certain 4*f* and 5*f* electron intermetallic compounds. Since a number of the known heavy-fermion systems (HFS's) are (antiferro-) magnetic[2-4] (e.g., UCd_{11}, U_2Zn_{17}, and $NpBe_{13}$), and because all HFS's exhibit very large magnetic susceptibilities and, in some instances,[5-7] instability toward formation of a magnetic ground state, numerous measurements of the magnetic field dependence of the physical properties of HFS's have been carried out to try to answer this question.

Here we report measurements of the specific heat of polycrystalline $CeCu_6$ in several fields up to 24 T (a new record high field for specific-heat measurements) between 1.8 and 8 K, as well as magnetizations measurements at 1.3 and 4.2 K in fields up to 20 T. These results significantly expand on the previous field range for such measurements (11 T for specific heat and 15 T for magnetization) and show a striking total suppression of the (temperature-dependent) mass enhancement upturn in the specific heat divided by temperature (C/T) at low temperatures such that C/T as $T\rightarrow 0$ falls from 1500 mJ/mol K^2 in zero field to 350 mJ/mol K^2 in 24 T. Accompanying this suppression of the zero-field upturn in C/T is a 40% decrease in the magnetization at 1.3 K in a 20 T field versus the linear extrapolation of the low-field magnetization (M) data.

These results will be compared to existing lower-field data for magnetoresistance,[8] specific heat,[9,10] and de Haas–van Alphen[11] (dHvA) measurements on $CeCu_6$, as well as the few other existing high-field measurements on other HFS's (M versus H for[12] $CeAl_3$, C/T for[13] UPt_3, for[14] UBe_{13}, and for[14,15] $UBe_{12.94}Cu_{0.06}$). Of particular interest is the great disparity in the size of the change in C/T with field between Ce and U HFS's, plus the fact that our 11 and 14.5 T data for C/T in $CeCu_6$ strongly call into question the tentative dHvA result that the effective mass in $CeCu_6$ is field insensitive to 13 T at low temperatures.

II. EXPERIMENT

The samples of $CeCu_6$ were prepared by arc-melting together in a zirconium-gettered argon atmosphere 99.9999% pure Cu and the highest available purity Ames Laboratory Ce in the correct proportions, allowing for slight weight losses. The resulting buttons were stored in vacuum dessicators until use.

Magnetic susceptibility from 1.7 to 400 K was measured in 5000 G in an automated superconducting quantum interference device (SQUID) susceptometer from Quantum Design. As has been noted before[16] in studies of single crystal $CeCu_6$, the susceptibility, χ, is extremely anisotropic, with χ for field in the *c* direction a factor of 10 higher than χ with field in the *b* direction. In our polycrystalline samples, there was some preferential orientation, such that the sample would readily be oriented to give $\chi(1.7\ K)=38\times10^{-3}$ emu/mol, implying[16] a significant partial *c*-axis orientation. This orientation was used for the field measurements of the specific heat. For the magnetization measurements, the (fixed) alignment was similar, such that $\chi(1.3\ K)=33\times10^{-3}$ emu/mol.

Specific heat results in zero field agreed with previous data[8] to within 10% at 1.8 K, consistent with some slight sample variation. The high-field calorimeter was similar to a previous design,[17] with the improvement that sapphire was used as the reference heat sink and thermometer reference block. This change avoided the noise heating from the ripple on the Bitter magnet current in the old copper block design. Thus, measurements of specific heat can be made at Francis Bitter National Magnet Laboratory in a 24 T, 1-in. bore magnet. The high-field magnetization data were taken with a vibrating sample magnetometer modified for high-field use and described elsewhere.[18]

III. RESULTS AND DISCUSSION

The magnetization of $CeCu_6$ as a function of field at 1.3 and 4.2 K is shown in Fig. 1. This type of data shows qualitatively quite clearly that the mechanism responsible for the very large magnetic susceptibility[1,8] ($\chi = M/H$) at low temperatures in $CeCu_6$ begins to be suppressed already by 5 T. As the slope of the M versus H data continues to decrease as the field H increases, we eventually find a 40% decrease in χ at 1.3 K and 20 T compared with zero field. This indeed is a large change—Pd, one of the most strongly enhanced paramagnetic elements, has[19] a linear M versus H curve to 30 T. Also, $CeAl_3$—the HFS most like $CeCu_6$ (γ and χ are the same within 10–20 % at low temperatures)—has a 33% decrease in χ from 0 to 20 T. Recent neutron studies[20] have indicated the presence of antiferromagnetic fluctuations in $CeCu_6$. It is worthwhile to note here as well that the fluctuation systems UAl_2 and UPt_3 have[21,22] much more linear M versus H behavior to high fields.

As a prelude to discussing the specific heat results, let us first discuss magnetoresistance results for $CeCu_6$. In zero field at $T < 0.3$ K, the resistivity[8] $\rho = \rho_0 + AT^2$, where the coefficient of the T^2 term $A = 120\ \mu\Omega\,\mathrm{cm/K^2}$. There exists a simple model[8] that $A \propto N(E_F)$, the electronic density of states at the Fermi energy, and that $C/T \propto N(E_F)$, implying that one can use A versus field as a (more easily measured) substitute for C/T versus field. However, it has already been observed that $A(H)$ decreases by about a factor of 6 at low temperature at 5 T, whereas less than a 40% decrease in C/T is observed[10] at 5 T at 0.3 K. (A question also raised by this discussion, which we will address below, is that of the temperature dependence of $\Delta C(H)/T$—one must be careful to compare results at the same temperature.) A more recent model[23] for $A(H)$ discusses a correlation of $A(H)$ with (C/T) squared, which still leaves a wide disparity between the model and experiment.

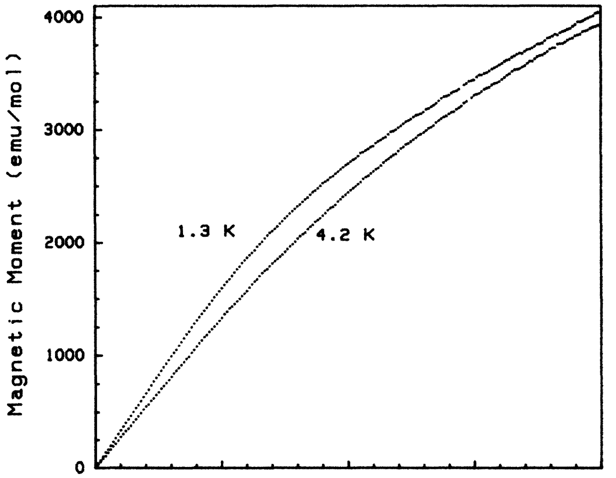

FIG. 1. Magnetization vs field for $CeCu_6$ at 1.3 and 4.2 K. The absolute accuracy of the data is better than ±5%, based on measurement of a Ni standard in the same run. The slope of these data give χ, the magnetic susceptibility, vs field. The difference in slope at low field at the two temperatures is just the observed 20% difference in χ. At the highest field, this difference in χ at 1.3 and 4.2 K is absent, consistent with the absence of any observed temperature dependence in C/T at the highest field. The single crystal M vs H data to 15 T of Ref. 16 showed about a 5% decrease or less in χ (15 T) vs $\chi(0)$ in the a- and b-axis directions, with a 47% decrease in the c-axis direction.

The specific heat of $CeCu_6$ in 0 and 11 T (from Ref. 9) is shown in Fig. 2; the specific heat of $CeCu_6$ in 0, 14.5, and 24 T is shown in Fig. 3. For comparison, results in 0 and 10 T for[24] $CeCu_2Si_2$ and 0 and 22.5 for[14] $UBe_{12.94}Cu_{0.06}$ are shown in Figs. 4 and 5. Further, results for $\Delta C(H)/T$ for these systems, as well as for $CeAl_3$, are tabulated in Table I.

As seen clearly from Figs. 2–5, all the field data are lower than the zero-field data below some temperature of order several kelvins, although the change in C/T with field is quite small in the uranium case. If data are measured to higher temperature, then the field data uniformly show an increase versus the zero field C/T data. (Data[13] for UPt_3 are the exception to this; see Table I.)

From Figs. 2 and 3 one can estimate C/T as $T \rightarrow 0$ in 13 T, the highest field used in the recent de Haas–van Alphen (dHvA) study,[11] as being of order 600 $\mathrm{mJ/mol\,K^2}$ (see also Table I). Since the field specific-heat data shown here do not extend below 1.8 K, the possibility of a change in slope of the data below this temperature causing a different zero-temperature value must be considered. The lowest-temperature, highest field-specific heat data available, Ref. 10, imply that, if anything, our estimate is too *high* since in 5.5 T Fujita *et al.* measure $C/T \simeq 700$ $\mathrm{mJ/mol\,K^2}$, i.e., they observe a bending over

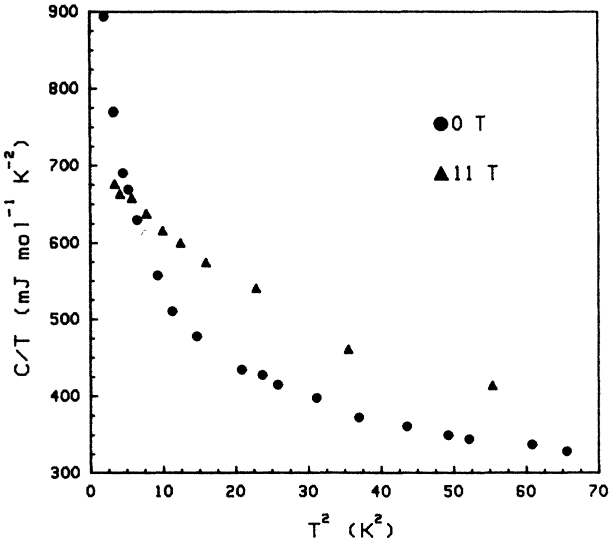

FIG. 2. Specific heat divided by temperature vs temperature squared in 0 (circles) and 11 T (triangles) for $CeCu_6$, from Ref. 8. Note the crossing of the two sets of data at $T^2 = 9\ \mathrm{K}^2$, and the significantly lower rate of rise of the 11-T data as $T \rightarrow 0$. These data were taken in a superconducting magnet and have an accuracy of ±3%, with a ±1% precision.

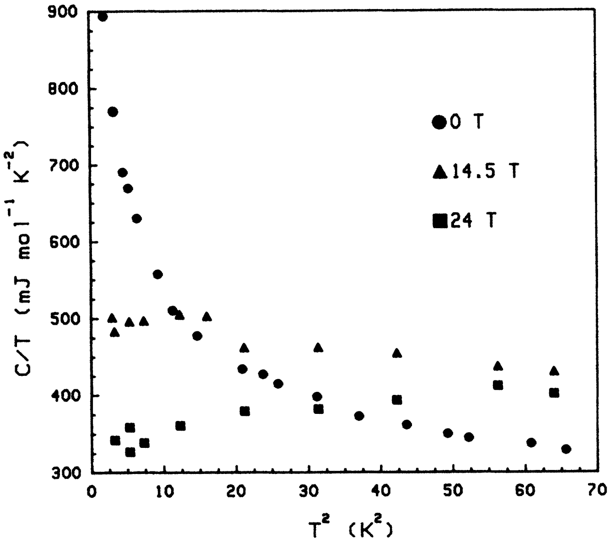

FIG. 3. Specific heat in 0 (circles), 14.5 (triangles), and 24 T (squares) fields divided by temperature vs temperature squared for $CeCu_6$. There may be a peak in C/T at around $T^2=12$ K^2 in the 14.5 T data, followed by a decrease at lower temperature. The 24 T data has a slope vs T^2 very similar to that of $LaCu_6$. The absolute accuracy of the field data is ±5%, with a precision below 4 K of ±4%, improving to ±2% at 8 K.

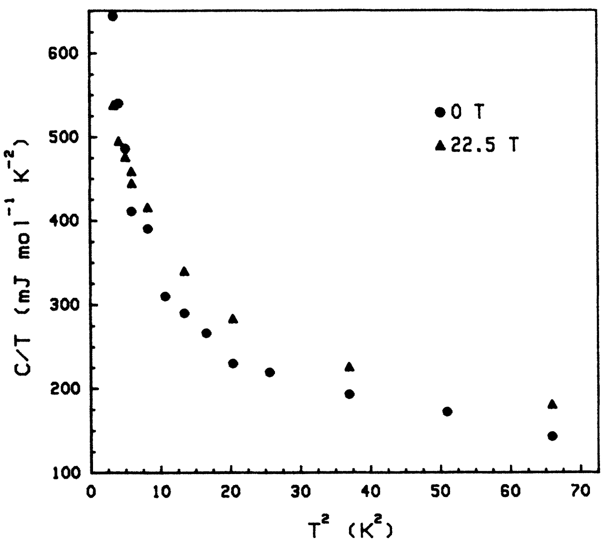

FIG. 5. Specific heat divided by temperature vs temperature squared in 0 (circles) and 22.5 T (triangles) for $UBe_{12.94}Cu_{0.06}$ from Ref. 14. Note that even at this high value of applied field that the specific heat is little altered by field at low temperatures.

of C/T in field at low temperature. Therefore, our estimate of C/T in 13 T may be used as a conservative estimate of the decrease. Thus, the measurement described in Ref. 11 of the field dependence of one dHvA frequency ($\propto m^*$, the effective mass), with $m^*=6m_e$, may be an indication that the lower effective mass portions of the Fermi surface are not field sensitive. However, the 11 and 14.5 T C/T of the present work at least strongly indicate the need for further dHvA work.

A second conclusion derived from the data in Figs. 2–5 and Table I is that there is a sharp dichotomy between changes in C/T with field in Ce versus U HFS's. This greater "stiffness" of the electron-electron correlations in U systems is also consistent with another observed major difference between Ce and U HFS's, that of the observed behavior upon doping. For Ce HFS's single-ion behavior is observed [i.e., C/T and χ for $(Ce_{1-x}M_x)B_y$ scale quite well[28,29] with amount of Ce, implying essentially noninteracting Ce], whereas doping[5–7,30] of U HFS's, even at small dopant concentrations, leads to rapid changes in C/T and χ. Therefore, any sort of rigid band model to explain the behavior of the specific heat of Ce and U HFS's in field, based on the results presented in Figs. 2–5 and Table I, must be viewed as too simplistic. Clearly, U ions interact much more strongly in U HFS's with the same C/T $(T\to 0)$ and therefore the same effective bandwidth, W, as a corresponding Ce system (e.g., UBe_{13} compared with $CeCu_2Si_2$).

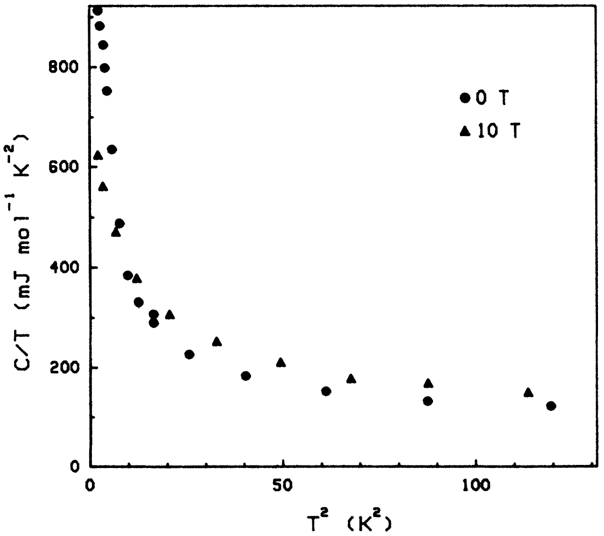

FIG. 4. Specific heat divided by temperature vs temperature squared in 0 (circles) and 10 T (triangles) for $CeCu_2Si_2$ from Ref. 24. Note the crossing of the two sets of data at $T^2=9$ K^2.

Finally, what is of special note in the results presented in Fig. 3 is that, in $CeCu_6$ (and, as a prediction, presumably also in $CeCu_2Si_2$ and $CeAl_3$, see Table I) at high enough field the upturn in C/T below 8 K is *totally suppressed.* This observation is based not just on the lack of any upturn in C/T in the 24 T data in Fig. 3, but also based on the value of the slope of the C/T versus T^2 data in 24 T:

$$C/T=\gamma(T)+\beta T^2 \ . \tag{1}$$

This slope, β in Eq. (1), is related to the lattice specific heat and the Debye temperature thereof via

$$\Theta_D=\left[\frac{1944\times 7}{\beta}\right]^{1/3}\times 10 \ , \tag{2}$$

with β in units of mj/[(mol $CeCu_6$) K^4], and the number 7

TABLE I. Specific heat vs field for selected HFS's.

System	Reference	C/T ($T \rightarrow 0$) (mJ/mol K^2)	H (T)	Crossover temperature (K)
CeCu$_6$	25	1500	0	
	9	700	11	3.07
	This work	510	14.5	3.5
	This work	350	24	5.8
	10	1450 (at peak in C/T at 0.3 K)	0	
	10	780 (at peak of C/T)	6	~2.3 (field data stop at 2 K)
CeAl$_3$	26	1650 (at peak in C/T at 0.5 K)	0	a
	26	1320 (at 0.5 K)	6	a
	26	1210 (at 0.5 K)	8	a
	27	2050 (at 0.5 K)	0	a
	27	1725 (at 0.5 K)	4	a
CeCu$_2$Si (nonsuperconducting)	24	1050	0	
	24	700	10	2.95
UBe$_{12.94}$Cu$_{0.06}$ (nonsuperconducting)	15	1430 (at 0.2 K)	0	
	15	1070 (at 0.2 K)	8	
	15	at $T > 0.7$ K, change with 8 T field is insignificant		
	14	540 (2 K)	0	
	14	495 (2 K)	22.5	2.3
UBe$_{13}$	14	in 18 T, $T > 2$ K, changes in C/T are small, consistent with data for UBe$_{12.94}$Cu$_{0.06}$		
UPt$_3$	13	450	0	
	13	485 (note increase with field)	11^b	~6
	13	560	19^b	4
	13	~520	22.5^b	3

[a] Data not taken to high enough temperature.

[b] Perpendicular to c axis.

is from the seven atoms per formula unit of CeCu$_6$. Lattice specific heats are chiefly determined by the atomic structure and the masses of the atoms in the unit cell. Thus, the Θ_D for[10] LaCu$_6$ (230 K) should be comparable to that for CeCu$_6$. Therefore, upon complete suppression of the temperature-dependent upturn in C/T caused by the formation of the heavy-fermion ground state in CeCu$_6$, the slope β of the C/T versus T^2 data should give a value for Θ_D comparable to that for LaCu$_6$. The value obtained from the 24-T CeCu$_6$ data in Fig. 3 for Θ_D is 240 K, fairly conclusive evidence that this field completely suppresses the upturn in C/T. (Incomplete 19-T data not shown in Fig. 3 indicates still some remnant upturn in this lower field.) What is of note from a theoretical perspective is that, although a noninteracting "Kondo-lattice" approach or, equivalently, a "Kondo-resonance" model[31] gives a good fit to C ($H=0$) for Ce systems,[32] and in particular CeCu$_6$,[33] this scheme of scaling noninteracting, single-ion results to a concentrated, Kondo-lattice system such as CeCu$_6$ does *not*[33] fit our high-field data, Fig. 3. Thus, interaction effects in nonzero magnetic field must be added to the existing theories.

Finally, it would be of interest to follow further the behavior of C/T for CeCu$_6$ with field, since presumably the observed C/T ($T \rightarrow 0$) value in 24 T, 350 mJ/mol K^2, would further decrease, albeit more slowly, according to a rigid band model. However, this rigid band, or "background" value for γ ($=C/T$) is small enough that rather large additional fields might be required to observe significant further decrease.

The field for total suppression of heavy fermions in CeCu$_6$, ~24 T, tells us the characteristic energy of the formation of the heavy-fermion ground state ($\sim\mu_{\text{eff}}H$). If we take[1] μ_{eff} from the high- (low-) temperature low-field χ data, in units of kelvin ($k_B T=\mu_{\text{eff}}H$) this characteristic formation energy is 43 (35) K.

IV. CONCLUSIONS

Measurement of the high-field specific heat of $CeCu_6$ has been performed up to 24 T, as well as magnetization measurements up to 20 T. The slope of the C/T versus T^2 data expected for a non-heavy-fermion $CeCu_6$ (i.e., the same slope as $LaCu_6$), with the heavy-fermion-associated upturn in C/T suppressed, is observed by 24 T. By comparison to high-field specific-heat data for other U and Ce systems, an essential difference between U and Ce HFS's clearly exists which must be addressed with new theoretical initiative.[34] Finally, the results presented here suggest that a more thorough dHvA investigation of the field dependence of the effective mass, m^*, in $CeCu_6$ up to 13 T will find strong, greater than 50% decreases in m^* for the heavier mass orbits.

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[1] G. R. Stwart, Rev. Mod. Phys. **56**, 755 (1984).

[2] Z. Fisk, G. R. Stewart, J. O. Willis, H. R. Ott, and F. Hulliger, Phys. Rev. B **30**, 6360 (1984).

[3] H. R. Ott, H. Rudigier, P. Delsing, and Z. Fisk, Phys. Rev. Lett. **52**, 1551 (1984).

[4] G. R. Stewart, Z. Fisk, J. L. Smith, J. O. Willis, and M. S. Wire, Phys. Rev. B **30**, 1249 (1984).

[5] A. de Visser, J. C. P. Klaasse, M. van Spang, J. J. M. Franse, A. Menovsky, and T. T. M. Palstra, J. Magn. Magn. Mater. **54-57**, 375 (1986).

[6] G. R. Stewart, A. L. Giorgi, J. O. Willis, and J. O'Rourke, Phys. Rev. B **34**, 4269 (1986).

[7] A. P. Ramirez, B. Batlogg, E. Bucher, and A. S. Cooper, Phys. Rev. Lett. **1072**, 57 (1986).

[8] A. Amato, D. Jaccard, E. Walker, J. Sierro, and J. Flouquet, J. Magn. Magn. Mater. **63-64**, 300 (1987).

[9] G. R. Stewart, Z. Fisk, and M. S. Wire, Phys. Rev. B **30**, 482 (1984).

[10] T. Fujita, K. Satoh, Y. Onuki, and T. Komatsubara, J. Magn. Magn. Mater. **47-48**, 66 (1985).

[11] P. H. P. Reinders, M. Springford, P. T. Coleridge, R. Boulet, and D. Ravot, J. Magn. Magn. Mater. **63-64**, 297 (1987); Phys. Rev. Lett. **57**, 1631 (1986).

[12] F. R. deBoer, J. Klaasse, J. Aarts, C. D. Bredl, W. Lieke, U. Rauchschwalbe, F. Steglich, R. Felten, U. Umhofer, and G. Weber, J. Magn. Magn. Mater. **47-48**, 60 (1985).

[13] G. R. Stewart, Z. Fisk, J. L. Smith, B. L. Brandt, A. de Visser, A. Menovsky, and J. J. M. Franse (unpublished).

[14] G. R. Stewart, A. L. Giorgi, and B. L. Brandt (unpublished).

[15] H. M. Mayer, U. Rauchschwalbe, F. Steglich, G. R. Stewart, and A. L. Giorgi, Z. Phys. B **64**, 299 (1986).

[16] Y. Onuki, Y. Shimizu, and T. Komatsubara, J. Phys. Soc. Jpn. **54**, 304 (1985).

[17] G. R. Stewart, Rev. Sci. Instrum. **54**, 1 (1983).

[18] Y. Shapira, S. Foner, and E. J. McNiff, Jr. (unpublished).

[19] F. A. Muller, R. Gersdorf, and L. W. Roeland, Phys. Lett. **31A**, 424 (1970).

[20] G. Aeppli, H. Yoshizawa, Y. Endoh, E. Bucher, J. Hufnagl, Y. Onuki, and T. Komatsubara, Phys. Rev. Lett. **57**, 122 (1986).

[21] J. J. M. Franse, P. H. Frings, F. R. de Boer, A. Menovsky, C. J. Beers, A. P. J. van Deursen, H. W. Myron, and A. J. Arko, Phys. Rev. Lett. **48**, 1749 (1982).

[22] P. H. Frings, J. J. M. Franse, F. R. de Boer, and A. Menovsky, J. Magn. Magn. Mater. **31-34**, 240 (1983).

[23] K. Kadowaki and S. B. Woods, Solid State Commun. **58**, 507 (1986).

[24] G. R. Stewart, Z. Fisk, and J. O. Willis, Phys. Rev. B **28**, 172 (1983).

[25] H. R. Ott, H. Rudigier, Z. Fisk, J. O. Willis, and G. R. Stewart, Solid State Commun. **53**, 235 (1985).

[26] A. S. Edelstein, Solid State Commun. **56**, 271 (1985).

[27] C. D. Bredl, S. Horn, F. Steglich, B. Lüthi, and R. M. Martin, Phys. Rev. Lett. **52**, 1982 (1984).

[28] Y. Onuki and T. Komatsubara, J. Magn. Magn. Mater. **63-64**, 281 (1987).

[29] C. L. Lin, A. Wallash, J. E. Crow, T. Mihalisin, and P. Schlottmann, Phys. Rev. Lett. **58**, 1232 (1987).

[30] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. B **31**, 1651 (1985).

[31] K. D. Schotte and U. Schotte, Phys. Lett. **55A**, 38 (1975).

[32] C. L. Lin, A. Wallash, J. E. Crow, T. Mihalisin, and P. Schlottmann, Phys. Rev. Lett. **58**, 1232 (1987).

[33] G. R. Stewart, B. Andraka, and C. Quitmann, Phys. Scr. (to be published).

[34] One recent theoretical approach is that of D. Wohlleben, in the Proceedings of the Fifth International Conferene on Valence Fluctuations, edited by L. C. Gupta and S. K. Malik (unpublished).