Susceptibility of spin-fluctuation compounds in high magnetic fields

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The differential susceptibilities of the spin-fluctuation compounds $TiBe_2$, UAl_2, UCo_2, and UPt_3 have been measured at low temperatures as a function of magnetic field up to 40 T. A systematic relation is observed between the signs of the second derivatives of the susceptibility with respect to field and temperature. A characteristic field H_{sf} and a characteristic temperature T_{sf} are defined by the inflection points in the $\chi(H)$ and $\chi(T)$ curves, respectively. It turns out that $\mu_B H_{sf}$ is of the order of kT_{sf}.

I. INTRODUCTION

Actinide metals and compounds with large $5f$ density-of-states values at the Fermi level often exhibit spin-fluctuation behavior.[1,2] In general, spin-fluctuation effects manifest themselves in a $T^3\ln(T/T_{sf})$ term in the specific heat, a T^2 term in the low-temperature susceptibility with either a positive or a negative sign, and as a T^2 term in the resistivity at low temperatures in combination with a large increase of the resistivity (of the order of 100 $\mu\Omega$ cm) in going up to room temperature.

From the temperature dependences of the resistivity, the specific heat and the susceptibility, a characteristic temperature T_{sf} is defined. An unambiguous definition of this temperature is difficult. For UAl_2, for instance, values between 7 and 25 K have been reported.[3,4] The $T^3\ln(T/T_{sf})$ term in the specific heat is evident for UAl_2, UCo_2, and UPt_3 from an upturn in the C/T-versus-T^2 plot at low temperatures and is found to be only weakly field dependent in fields up to 5 T.

High-magnetic-field experiments offer unique possibilities to study spin-fluctuation phenomena in strongly exchange-enhanced paramagnetic materials. In selected materials the characteristic temperature for spin fluctuations is sufficiently low for the Zeeman energy in high-field experiments to become comparable with the thermal energy at the characteristic temperature. The suppression of spin fluctuations in high magnetic fields was already observed in $TiBe_2$ (Ref. 5) and in UAl_2.[6] We extended the study of this effect to a single crystal of UPt_3 and performed more precise experiments on UAl_2. In UPt_3 the magnetic susceptibility is anisotropic and exhibits a low-temperature maximum for field directions in the hexagonal plane.

Recently, UPt_3 has been discovered to become superconducting at a temperature of 0.54 K,[7] making this material the first compound that shows a combination of superconductivity and spin-fluctuation effects. Naturally, the first question to be answered is whether the low-temperature anomalies in the magnetic and other electronic properties of exchange-enhanced paramagnetic metals are due to many-body effects (a low spin-fluctuation temperature) or to very fine details in the one-electron band structure near the Fermi level (a low effective degeneracy temperature). Answers to this question can only be given by a thorough study of these phenomena in experimental and theoretical investigations. High-magnetic-field experiments are one way in which this problem can be approached experimentally.

In the present paper detailed information on the magnetization curves of three intermetallic uranium compounds, UCo_2, UAl_2, and UPt_3, and on that of $TiBe_2$ are presented. These results are compared with high-magnetic-field studies on UCo_2 and UAl_2 performed earlier.[6,8] In addition we report on the temperature dependence of the susceptibility of UPt_3 and on the specific heat of UCo_2, UAl_2, and UPt_3 in zero field and in applied fields of 5 T. Part of these latter results have been previously published.[8,9]

II. EXPERIMENTAL RESULTS

Before giving the details of our magnetization curves we present briefly our results on the specific heat of UCo_2, UAl_2, and UPt_3 and on the temperature dependence of the susceptibility of UPt_3.

A. Specific-heat measurements

Since a $T^3\ln(T/T_{sf})$ term in the specific heat is regarded as the most reliable indication for spin-fluctuation behavior, our specific-heat measurements on UCo_2, UAl_2, and a polycrystalline sample of UPt_3 are presented in Figs. 1, 2, and 3, respectively, in order to elucidate the presence of this term in these compounds. The experimental data are described with the usual phonon and electronic terms and with the additional $T^3\ln(T/T_{sf})$ term due to spin fluctuations:[10,11]

$$C/T=\gamma+\beta^*T^2+\delta T^2\ln T+\epsilon T^4 . \quad (1)$$

In this expression β^* has been introduced instead of β since the coefficient of the T^2 term deviates from the coefficient of the T^2 term in the usual expression without the logarithmic term. This fit is represented by the broken lines in Figs. 1–3 and gives rise to the values for the coefficients as shown in Table I. The term ϵT^4 in Eq. (1) is added to allow for a fit up to higher temperatures. For

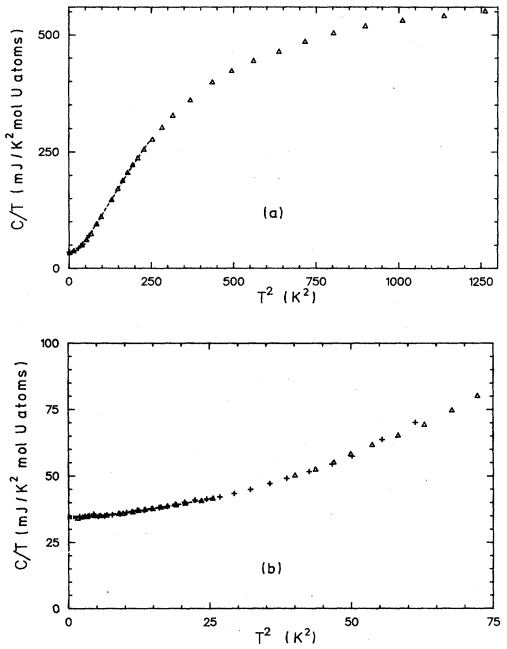

FIG. 1. Specific heat of UCo_2 with (+) and without (△) a magnetic field of 5 T. The dashed curve in (b) represents the three-parameter fit of the zero-field data to Eq. (1); see Table I.

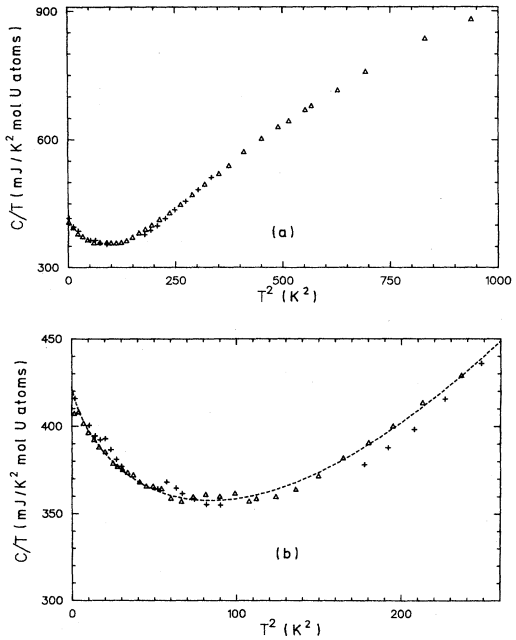

FIG. 3. Specific heat of UPt_3 with (+) and without (△) a magnetic field of 5 T. The dashed curve in (b) represents the three-parameter fit of the zero-field data to Eq. (1); see Table I.

comparison, data by Trainor *et al.*[1] and data by Stewart *et al.*[12] on UAl_2 and data by De Visser *et al.*[13] on UPt_3 are also given.

We stress the very small field effect on the specific heat of the order of 1(1)% at 5 T; it excludes any contribution to the specific heat of weakly interacting local moments as origin for the upturn in the specific heat.

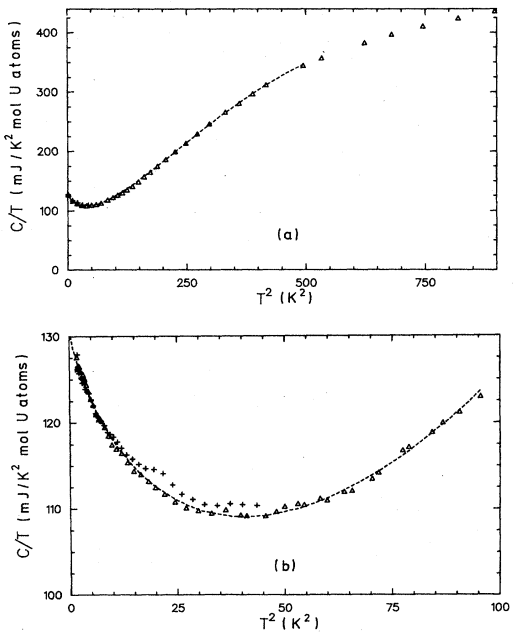

FIG. 2. Specific heat of UAl_2 with (+) and without (△) a magnetic field of 5 T. The dashed curves in (a) and (b) represent the four- and three-parameter fits of the zero-field data to Eq. (1), respectively; see Table I.

B. Susceptibility measurements

The susceptibility measurements with improved accuracy on a single crystal of hexagonal UPt_3 are presented in Fig. 4. We note that a maximum in the susceptibility-versus-temperature curve is clearly observable for field directions in the basal plane. Along the *c* axis this maximum is absent. These results are summarized in Table III, together with relevant information for UCo_2 and UAl_2 as derived from literature data.

C. Magnetization measurements

Magnetization measurements on $TiBe_2$, UAl_2, UCo_2, and UPt_3 were performed in the Amsterdam High-Field

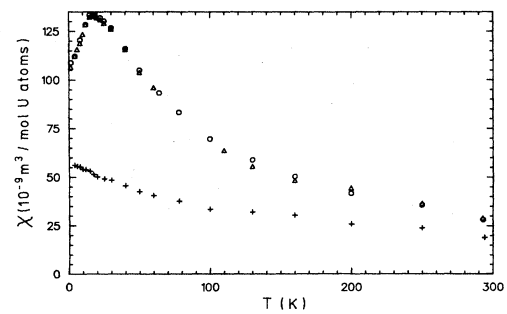

FIG. 4. $\chi(T)$ of UPt_3 measured in fields up to 1.3 T along *a* axis (○), *b* axis (△), and *c* axis (+).

TABLE I. Specific heat of UAl_2, UCo_2, and UPt_3; C/T fitted to $\gamma+\beta^*T^2+\delta T^2\ln T+\epsilon T^4$ (C in mJ K^{-1}/mol U-atoms, T in K).

Compound	Temperature range	γ	β^*	δ	$\epsilon\times10^3$	Ref.
UAl_2	1.3–23	132.7(2)	−3.04(3)	1.35(1)	−1.46(2)	a
	1.3–8	131.3(1)	−2.60(3)	1.11(2)		a
	1.3–10	131.1(1)	−2.55(2)	1.08(1)		a
	0.8–6	143	−4.38	1.94		b
	2 –6.3	142.1	−3.44	1.41		c
	2 –23	142.3	−3.64	1.566	−1.7	c
	0.3–2.2	~126				d
UPt_3						
Polycrystalline	1.3–16.5	422.4(5)	−4.18(5)	1.54(2)		a
Single crystal	1.2–20	422	−3.27	1.38		e
	1.2–15	423	−3.80	1.41		e
	1.2–10	421	−3.80	1.41		e
	6 –10	416	−3.13	1.15		e
	8 –20	413	−3.45	1.30		e
UCo_2	1.3–6	35.4(1)	−0.42(1)	0.43(1)		f
	1.3–6	34.6(4)	0.0(2)	0.21(9)		a
	1.3–8.7	36.8(4)	−0.9(1)	0.69(4)		a

[a]This work.
[b]Trainor *et al.* (Ref. 1).
[c]Stewart *et al.* (Ref. 12).
[d]Armbrüster *et al.* (Ref. 3).
[e]De Visser *et al.* (Ref. 13).
[f]Franse *et al.* (Ref. 6), from a graphical analysis.

Installation.[14] In normal operation this installation has the advantage that the field can be kept constant long enough to reduce the influence of the eddy currents to the magnetization to a negligible part.

For precise measurements of the differential susceptibility, however, it is better to obtain as much data as possible during the limited time of one field run. This was accomplished by measuring the magnetization in a magnetic field linearly changing in time at a constant rate of 40 or 56 T s^{-1}. In one field run about 80 field and magnetization data points are collected. This type of measurement induces, of course, a spurious effect in the magnetization due to eddy currents. For a magnetization curve linear in the field, a field-independent resistivity, and a constant rate of change of the field, this effect is field independent in first order and thus of negligible influence on the differential susceptibility.

For a bulk sample of polycrystalline UAl_2 the difference in magnetization determined with increasing and decreasing field amounted to 2% in a field of 21 T. The values measured at constant fields were, within the experimental accuracy, equal to the average of the magnetizations measured with increasing and decreasing field (see Fig. 5). For experimental reasons (to reduce heating of the magnet) all subsequent measurements were performed with decreasing fields. With the experimental technique described above it was possible to evaluate the differential susceptibility by taking the quotient of the difference in magnetization and the difference in field between a small number (of the order of four) of subsequent data points. The differential susceptibilities of UAl_2, of UPt_3 along the basal-plane directions, and of $TiBe_2$ are shown in Figs. 6, 7, and 8, respectively.

The differential susceptibility as presented here is not likely to be affected by magnetic impurities. Impurities. with a moment of the order of $1\mu_B$ are expected to be saturated at 4.2 K in fields above 5 T. Moreover, magnetization curves on UAl_2 and UPt_3 at 1.5 K are almost identical with the ones observed at 4.2 K.

III. ANALYSIS OF THE EXPERIMENTAL RESULTS

Before attributing the low-temperature anomalies in susceptibility and specific heat to spin-fluctuation effects,

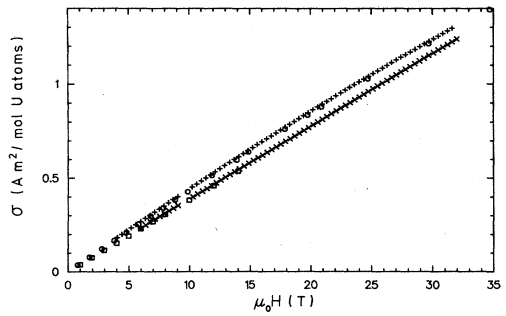

FIG. 5. Magnetization vs field of UAl_2 at 4.2 K (+,○) and 20 K (×,□).

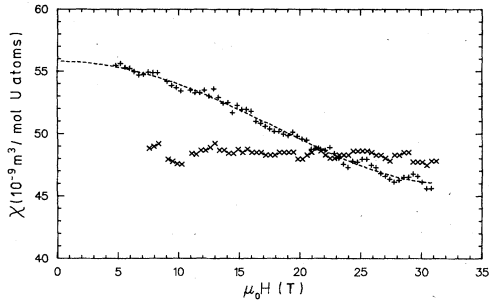

FIG. 6. Differential susceptibility for UAl_2 at 4.2 K (+) and 20 K (×). The dashed curve represents a description with the parameters given in Table II.

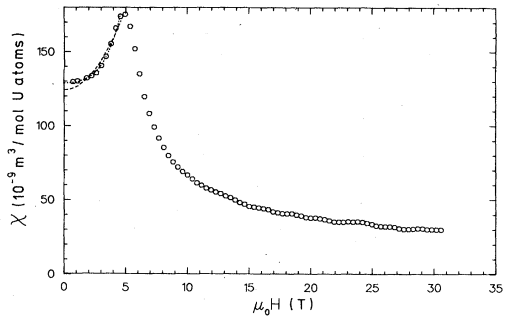

FIG. 8. Differential susceptibility for $TiBe_2$ at 1.4 K (○). The dashed and dotted curves represent a fit for three and two parameters, respectively; see Table II.

more straightforward interpretations of these anomalies in terms of magnetic impurities, complex magnetic ordering phenomena, crystal-field effects, or details in the band structure must be considered. By inspecting the specific-heat data of UPt_3 we will point out the inappropriateness of these other possibilities. For UAl_2 and $TiBe_2$ we refer to discussions in previous publications (UAl_2, Trainor *et al.*,[1] $TiBe_2$, Stewart *et al.*[5]).

Local magnetic moments in an internal field, indeed, can give rise to an upturn in the specific heat at low temperatures. Assuming that the upturn in the specific heat of UPt_3 is due to this mechanism a molecular field equivalent to a temperature less than a few kelvin is calculated. With impurity moments larger than $0.2\mu_B$, it must be expected that an external field of 5 T clearly influences the specific heat, which is apparently not the case.

Magnetic ordering below 16 K, which might be concluded from the maximum in the susceptibility-versus-temperature curves, is not consistent with the specific-heat data either. The superconducting transition that is evident from specific-heat measurements below 0.5 K is another indication for the absence of magnetic order in UPt_3. An interpretation of the high-field magnetization curve at 4.2 K in terms of a special type of spin-flop transition turns out to be inappropriate for those reasons.

Crystal-field effects that could be responsible for the anistropy in the susceptibility and for the maxima in the

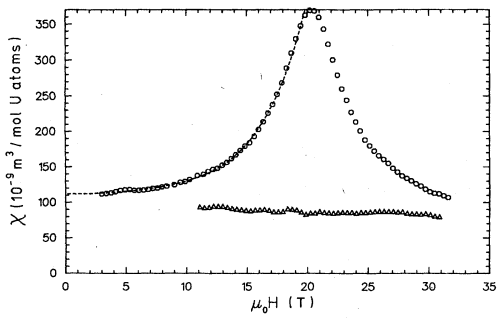

FIG. 7. Differential susceptibility for UPt_3 at 4.2 K along the *b* axis (○) and at 77 K along the *a* axis (△). The dashed curve represents a description with the parameters given in Table II.

$\chi(T)$ and $\chi(H)$ curves [see, for instance, the susceptibility data for hexagonal $PrNi_5$ (Ref. 15)] are inconsistent with the specific-heat data in which no Schottky type of contribution is observed.

Finally, the possibility that the low-temperature anomalies are due to details of the band structure must be discussed. To simplify matters we assume a peak in the density-of-states curve positioned near the Fermi level with relative height of $\Delta N/N$ and a width W. A rough estimate of the coefficient of the linear term in the specific heat from the specific-heat data at higher temperatures results in a value of 225 $mJ\,K^{-2}\,mol^{-1}$ and leads, by comparing the low- and higher-temperature γ values, to a relative height of this peak of 45% of the total density of states at the Fermi energy. The width of this peak can be estimated from the temperature where its influence on the specific heat is considerably reduced and amounts to an energy of the order of 1 meV. The effect of a magnetic field of 5 T on the specific heat would, of course, be enormous for such a sharp structure which apparently is not the case. In summary we conclude that the upturn in the C/T-versus-T^2 curve of UPt_3 below 10 K cannot be attributed to trivial contributions such as arising from magnetic impurities, crystal-field effects, or a sharp peak in the density of states curve close to the Fermi level, nor can it be considered as the precursor of a magnetic or superconducting transition. Since the temperature dependence of the specific heat can be represented in a satisfactory way by a $T^3 \ln T/T_{sf}$ term, we are motivated to apply the expressions from spin-fluctuation theories to our results for the field dependence of the differential susceptibility.

In the first approximation, spin-fluctuation effects on the susceptibility can be represented at low temperatures by a quadratic temperature and field dependence,[2,16] although Misawa[17] and Barnea and Edwards[18] derived logarithmic terms in the temperature and field dependences of the susceptibility. It should be stressed that the quadratic dependences, according to theory, are only first-order approximations for $H \ll H_{sf}$ and $T \ll T_{sf}$. In an attempt to fit our data in a field interval that clearly goes beyond this limit, we added terms containing higher powers of the field (see Table II). We mentioned previously that a

TABLE II. Fit of $\chi(H)$ to $\chi_0(1+aH^2+bH^4+cH^6)$ (χ in m^3/mol U-atoms, H in T).

Compound	Max. field	$10^9\chi_0$	10^5a	10^7b	10^8c
$TiBe_2$	5	124(2)	1800(100)		
	5	128(1)	550(200)	5700(900)	
UAl_2	31	55.8(1)	−34.7(8)	1.7(1)	
UPt_3 (basal plane)	20	112(1)	135(25)	0(16)	3.0(3)

unique definition of a spin-fluctuation temperature and field cannot be given on the basis of the observed anomalies in specific heat, susceptibility, resistivity, etc.

In this study we define the characteristic temperature T_{sf} and the characteristic field H_{sf} as the temperature and field at which the second derivatives of the susceptibility with respect to temperature or field are zero. The parameters Δ_T and Δ_H serve as an estimate for the amplitude of the anomalies in the $\chi(T)$ and $\chi(H)$ curves, respectively. These parameters are defined as the susceptibility difference between the value at the characteristic temperature (or field) and that at zero temperature (or field). These definitions of the characteristic temperature and field are certainly not identical to those used in theoretical studies. It cannot *a priori* be expected that the numerical values for these quantities obtained from the susceptibility data should be consistent with the characteristic temperature derived from a $T^3\ln T/T_{sf}$ analysis. In fact additional knowledge or assumptions concerning the phonon term have to be invoked before a value for T_{sf} can be derived from the specific-heat data.

According to the method given above we have found the values H_{sf} and Δ_H given in Table III for UPt_3, $TiBe_2$ and UAl_2. For comparison we also show values for T_{sf} and Δ_T deduced from susceptibility measurements. For reference γ values and Stoner factors ($S=5.85\chi/\gamma$; χ in $m^3\text{mol}^{-1}$; γ in $\text{mJ K}^{-2}\text{mol}^{-1}$) are also given, without making corrections for orbital and diamagnetic contributions to χ and for enhancement effects in γ.

IV. DISCUSSION

High-magnetic-field studies on UAl_2,[6] on Pd,[20] and on $LuCo_2$ and YCo_2 (Ref. 21) revealed some systematics in the field and temperature dependence of the magnetic susceptibility of these strongly exchange-enhanced paramagnetic materials at low temperatures: The field and temperature dependences at low temperatures both have the same sign, being positive for Pd, YCo_2, $LuCo_2$, and $TiBe_2$ and negative for UAl_2.[22] The present experimental results for UPt_3 and UCo_2 support the hypothesis that there is a qualitative relation between the increase (decrease) of the differential susceptibility at 0 K as a function of the applied magnetic field and the increase (decrease) of the initial susceptibility as a function of the temperature. This qualitative similarity between $\chi(T)$ and $\chi(H)$ is even present over a temperature and field range several times T_{sf} and H_{sf}. Crystal-field effects that could provide an explanation for this similarity in the case of UPt_3 are inconsistent with the specific-heat data. In a simple one-electron band type of description such a systematic behavior could arise from a sharp structure in the density-of-states curve just above or below the Fermi level. For UPt_3, for instance, one would deduce from the differential susceptibility at 4.2 K that the center of a narrow peak with a width equivalent to 10 T and containing a few tenths of electron states per atom is positioned at a distance from the Fermi level equivalent to 20 T. The disappearance of the peak in the $\chi(H)$ curve above 20 K is

TABLE III. Values for χ_0, γ, and S for $TiBe_2$, UAl_2, and UPt_3; the effect of temperature and field on the susceptibility is expressed by means of the parameters T_{sf}, Δ_T, H_{sf}, and Δ_H; H_{sf} and Δ_H at 1.4 K for $TiBe_2$ and at 4.2 K for UAl_2 and UPt_3 (χ in m^3/mol U-atoms, γ in mJ K^{-2}/mol U-atoms, T in K and H in T).

Compound	$10^9\chi_0$	T_{sf}	Δ_T/χ_0	H_{sf}	Δ_H/χ_0	γ	S	Ref.
$TiBe_2$	100	5	1%			52	11	a
	122	5	~2%	4.8	25%			b
	124			4	30%			c
UAl_2	54.7	14	−10%					d
	55.8			18.4	−9.5%	132	2.5	c
UPt_3 (basal plane)	107	10	16%	18	140%	422	1.5	c

[a]Stewart *et al.* (Ref. 19).
[b]Acker *et al.* (Ref. 24).
[c]This work.
[d]Brodsky *et al.* (Ref. 2).

naturally explained in this way. It is not likely, however, that a peak in the density-of-states curve with the above given values for the characteristic parameters would cause the almost field-independent anomalies in the specific heat that are observed experimentally below 5 K. Therefore, we have to conclude that a single-particle description cannot account for all the presented data either.

By inspecting the temperature and field dependence of the magnetic susceptibility of UAl_2 it was found that an equivalent contribution to the susceptibility can be suppressed either by applying a magnetic field of about 20 T or by increasing the temperature to 20 K or higher.[6] It was concluded that for this material the characteristic field and temperature are related by $\mu_B H_{sf} \approx k T_{sf}$. Although our definition of T_{sf} and H_{sf} is an *ad hoc* one, we note also that for $TiBe_2$ and UPt_3 the values for $\mu_B H_{sf}$ and kT_{sf} are quite similar; see Table III. The amplitudes Δ_H and Δ_T always have the same sign but differ in magnitude, Δ_H being a few times larger than Δ_T except for UAl_2, where both quantities are almost equal.

Finally we mention that similar phenomena as we discussed for UPt_3 have been observed in the mixed-valence compound YbCuAl. The low-temperature susceptibility of this compound reaches its maximum value at 28 K and exceeds the zero-temperature value by almost 15%. In high-magnetic-field experiments at 1.4 and 4.2 K the magnetization is perfectly linear up to 10 T and exhibits a pronounced curvature upward above 15 T, the differential susceptibility increasing by 37%. A more extended study of spin- and valence-fluctuation systems has been published elsewhere.[23]

V. CONCLUSION

The electronic properties, i.e., the magnetic susceptibility and the specific heat, of UAl_2, UPt_3, $TiBe_2$, and UCo_2 reveal low-temperature anomalies that indicate spin-fluctuation phenomena. A qualitative analysis of the susceptibility data leads to an experimental definition of a characteristic temperature (T_{sf}) and field (H_{sf}) and of amplitudes of the susceptibility anomalies Δ_T and Δ_H. The similarity between the $\chi(H)$ and $\chi(T)$ curves is then expressed by $\mu_B H_{sf}$ being roughly equal to kT_{sf} and Δ_T and Δ_H having the same sign. The similarity even holds for temperatures or fields that clearly exceed the characteristic values.

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