Critical behavior of specific heat and isothermal magnetocaloric coefficient in the $UCuP_2$ ferromagnet

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The specific heat (C_p) and isothermal magnetocaloric (M_T) coefficient were measured (T=20–300 K, B=0–13 T) for the strongly anisotropic ferromagnet $UCuP_2$ with T_C=94.5 K. From the zero-field $C_p(T)$ curve the ratio of the critical amplitudes $A^+/A^-=0.50\pm0.05$ was derived. The isothermal magnetocaloric coefficient taken at $T=T_C$ was also found to follow simple power-law dependence with the critical exponent $\omega=0.48\pm0.05$ and the critical amplitude $A_m=0.270\pm0.007$. Comparison of these parameters with the respective theoretical values indicated that $UCuP_2$ belongs to the three-dimensional Ising universality class, in spite of its layered structure. This conclusion was confirmed by calculation of the full scaling functions for both measured quantities.

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I. INTRODUCTION

$UCuP_2$ is a representative of a numerous family of actinide- or lanthanide-based compounds of the general composition $(A,L)TX_2$, where T is a d-electron transition metal and X is a pnictogen (X=P, As, Sb, Bi).[1] Most of these materials crystallize with a tetragonal structure of the $HfCuSi_2$ type (space group $P4/nmm$), yet $UCuP_2$ is a rare example of a phase adopting a tetragonal unit cell of the $SrZnBi_2$ type (space group $I4/mmm$, a=3.803 Å, c=18.523 Å).[2] The two types of structures are closely related to one another (see discussion in Ref. 2), with the common feature of a typical layered character, with planes of A or L, T, and X atoms alternating along the fourfold axis (in the case of $UCuP_2$ the sequence of atomic sheets in a single unit cell is P-U-P-Cu-P-U-P-U-P-Cu-P-U-P). As a result, the $(A,L)TX_2$ compounds are expected to exhibit a strongly two-dimensional character of their physical properties, and indeed this type of magnetic and electrical behavior has been observed experimentally (for a recent review on actinide-based materials see Ref. 1).

As found from single-crystal studies,[3] $UCuP_2$ is a uniaxial ferromagnet with the Curie temperature $T_C\approx75$ K and the spontaneous magnetic moment of $0.98\mu_B$ aligned along the tetragonal c axis. In the ordered state the compound shows a high value of the magnetic anisotropy constant K_1, 9×10^5 J/m^3, which corresponds to the anisotropy field of about 13 T. As regards its electrical conductivity behavior, $UCuP_2$ has been found to be a semimetal with a strongly damped number of free carriers, which was estimated from the resistivity, Hall effect, and optical conductivity data to be $0.06e^-$–$0.15e^-$ per formula unit.[4,5] Interestingly, in the paramagnetic region the resistivity measured within the a-c plane of the tetragonal unit cell shows a Kondo-like effect,[3,4] which is an appealing issue in view of the ferromagnetic character of this particular compound.

In this paper we report on extensive calorimetric measurements of $UCuP_2$ performed on single-crystalline specimens in the temperature range 20–300 K and in magnetic fields up to B=13 T. Temperature dependencies of the specific heat C_p were measured at a number of constant fields and field dependencies of the isothermal magnetocaloric coefficient M_T were taken at several constant temperatures. The calorimetric data in the vicinity of the Curie temperature are discussed here in terms of the critical fluctuations of the order parameter. The critical properties of $UCuP_2$ have been properly described in terms of the three-dimensional (3D) Ising universality class, and some arguments are given to discriminate this picture from the 2D Ising universality class description that would seem more appropriate based on the structural and magnetic properties of the compound studied.

II. EXPERIMENT

High-quality single crystals of $UCuP_2$ were grown by the chemical vapor transport method, as described previously.[2,3] Their quality was checked by x-ray diffraction (Stoe powder diffractometer with Cu $K\alpha$ radiation and Xcalibur charge-coupled device Oxford Diffraction four-circle diffractometer with graphite-monochromated Mo $K\alpha$ radiation) and energy dispersive x-ray spectrometry (EDAX PV9800 microprobe attached to a Philips 515 scanning electron microscope). Moreover, the magnetic behavior of the single crystal selected for calorimetric studies was checked by magnetization measurements (Quantum Design MPMS-5 superconducting quantum device magnetometer). All the structural and magnetic data obtained are in good agreement with those reported previously.[2,3]

Both the specific heat C_p and the isothermal magnetocaloric coefficient M_T were measured using a heat-flow calorimeter.[6] In this method the sample is connected to a heat sink by means of a sensitive heat-flow meter of high thermal conductance. If the temperature of the sink T varies with a constant rate T then (neglecting some small corrections) C_p is directly proportional to the voltage U on terminals of the heat-flow meter

$$C_p \equiv \frac{dq}{dT} = \frac{q}{T} = \frac{U}{AT}, \qquad (1)$$

where q is the heat flux flowing from the sink to the sample

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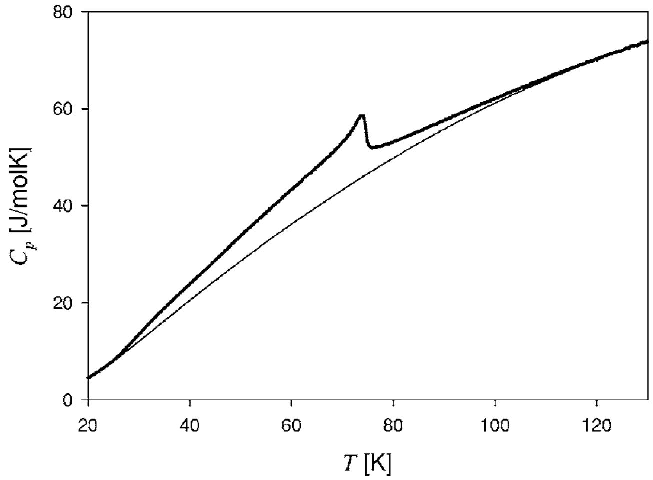

FIG. 1. Temperature dependence of the zero-field specific heat of single-crystalline $UCuP_2$. Thin line represents the nonmagnetic background described by Eq. (3).

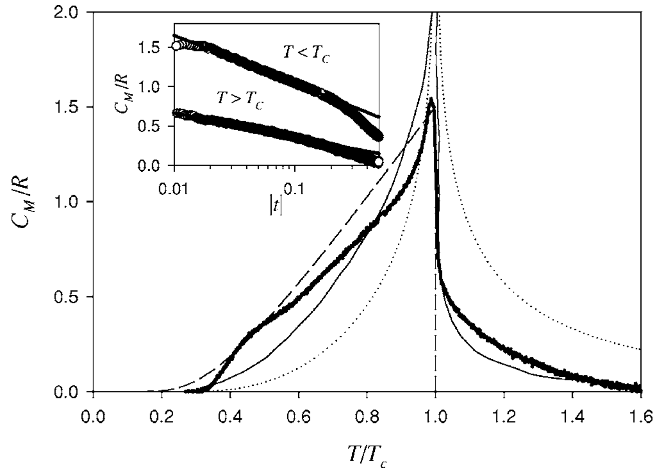

FIG. 2. The magnetic specific heat C_M of $UCuP_2$ for $B=0$ (thick solid line). Thin lines represent specific heat for the 3D Ising (solid), 2D Ising (dotted), and mean-field approximation (dashed) models. Both Ising models are shown in arbitrary units. The inset displays the fitting according to Eq. (5).

and A is the heat-flow meter sensitivity. The C_p measurements could be performed both upon cooling and heating, at any value of constant magnetic field B.

On the other hand, it is possible to stabilize the sink temperature and sweep the magnetic field at some constant rate B. This way the magnetocaloric effect may be studied in quasi-isothermal conditions

$$M_T \equiv \frac{dq}{dB} = \frac{-q}{B} = \frac{-U}{AB}, \tag{2}$$

where M_T is the isothermal magnetocaloric coefficient. The minus sign was introduced to keep the convention of the heat-flux direction used in the formula for C_p. The M_T measurements could be performed upon both increasing and decreasing field. More technical details are given in Ref. 6. Some supplementary information can also be found in our previous publications describing isothermal magnetocaloric measurements of antiferromagnetic compounds UAs (Ref. 7) and $UNi_{0.5}Sb_2$ (Ref. 8).

A single crystal of $UCuP_2$ (32 mg) was glued (using GE varnish) to the heat-flow meter in such a way that the easy magnetization axis (i.e., the c axis) was oriented parallel to the magnetic field. Several temperature runs $300 \to 20 \to 300$ K with the rate $T \approx 1$ K/min were performed at various magnetic fields up to $B=13$ T in order to determine the in-field specific heat. Next, a series of magnetic sweeps $0 \to 13 \to 0$ T were performed with the rate $B=0.2$ T/min. These runs gave the magnetocaloric data.

III. SPECIFIC HEAT

A. General features

The zero-field specific heat curve (Fig. 1) agrees well with the data presented in Ref. 9. No differences between the C_p data taken upon cooling and heating were noticed. The ferromagnetic transition manifests itself by the λ-shaped anomaly at $T_C=74.5$ K. Using the relation $\mu_B B_C = k_B T_C$ one estimates the characteristic magnetic field for scaling analysis (see below) as $B_C=111$ T.

In order to analyze the magnetic component C_M in the total specific heat, it was assumed that the nonmagnetic contributions can be described by a simple formula composed of three terms

$$C_{NM} = 2C_D + 2C_E + \gamma T. \tag{3}$$

The two first terms stand for the phonon contributions. The acoustical phonons connected mainly with the heavier atoms (U, Cu) are described simply by two Debye modes (C_D) with a common, averaged Debye temperature (Θ_D). The optical phonons connected mainly with the two lighter P atoms are represented by two Einstein modes (C_E) with the same Einstein temperature (Θ_E). The third term described by the γ parameter contains both the electronic and dilatation contributions. The values of $\Theta_D=195\pm6$ K, $\Theta_E=377\pm5$ K, and $\gamma=26\pm4$ mJ/mol K^2 were obtained by fitting the $C_{NM}(T)$ model to the measured data with the exclusion of the ±50 K region around T_C. We stress that the applied approach should be treated merely as a rough approximation for the nonmagnetic contributions. However, within a relatively narrow window around T_C the error made in the determination of $C_M(T)$ can be treated as a constant that does not disturb the analysis performed below.

B. Zero-field C_p fluctuation amplitude ratio

The magnetic contribution to the specific heat of $UCuP_2$ is presented in Fig. 2. The total magnetic entropy was estimated to be $S_{magn} \approx 5.7$ J/mol K, which is close to the value of $R \ln 2 = 5.76$ J/mol K (R is the gas constant) expected for a doubly degenerate magnetic ground state. This finding stands in opposition to the previous, much smaller estimation of $S_{magn} \approx 1.2$ J/mol K given in Ref. 9. The large discrepancy probably arises from different definitions of nonmagnetic background applied in the two studies.

The λ shape of the specific heat anomaly at T_C suggests that the transition is governed by short-range interactions, i.e., fluctuations of the order parameter. The strong magnetic anisotropy of $UCuP_2$ (Ref. 3) implies a uniaxial (Ising) model as the appropriate description of the compound studied. Indeed, a comparison of the experimental C_p data with the predictions of the 2D and fcc 3D Ising models[10] (see Fig. 2) reveals a general similarity (especially for the 3D case). On the other hand, the height of the peak at T_C is nearly equal to that expected from the mean-field model with spin $S=1/2$ (dashed line), i.e., $\Delta C_M/R=1.5$. In the view of further analysis this coincidence appears to be fully accidental, caused by smearing the C_M divergence due to the experimental conditions.

Another point to be mentioned is a pronounced bump in $C_M(T)$ visible at $\sim 0.45T_C$ (i.e., at $T \sim 30-35$ K). This feature was found to be hardly field dependent up to $B=13$ T (not shown). Interestingly, the position of this anomaly seems to coincide with an inflection point in the temperature-dependent electrical resistivity[4,5] as well as with a bump in the temperature variation of the thermal conductivity,[11] the latter being interpreted as a manifestation of the magnon contribution to the heat transport. However, presently we cannot definitely exclude the possibility that the observed bump in $C_M(T)$ is an experimental artifact; therefore we leave this issue for further studies.

In some critical region around T_C a simple power-law dependence of the specific heat is expected:

$$C_M^{\pm} \sim \frac{A^{\pm}}{\alpha}|t|^{-\alpha}, \tag{4}$$

where $t=(T-T_C)/T_C$ is the reduced temperature, α stands for the critical exponent, $A^{\pm}$ are the fluctuation amplitudes, while the "+" and "−" superscripts refer to high- and low-temperature side of the transition, respectively. For the uniaxial systems, $\alpha=0$ is expected for the 2D case, and $\alpha=0.119$ is appropriate for the 3D model. However, for the latter case Eq. (4) is obeyed only within a very narrow region around the critical temperature, $|t| < 5\times 10^{-3}$,[12] which is not accessible by our experimental setup. Outside this range the divergence weakens and can be effectively described by $\alpha \to 0^+$. Therefore, one is allowed to expand Eq. (4) into series with respect to α and neglect all terms above the first-order one:

$$C_M^- \sim C_0 + \Delta C - A^- \ln|t|, \quad C_M^+ \sim C_0 - A^+ \ln t. \tag{5}$$

C_0 is a constant which also comprises the possible error made in the determination of C_{NM}. The approach separates the critical anomaly into a weak logarithmic divergence and a finite jump of the height ΔC. It is exact for the 2D Ising model ($\alpha=0$). One can easily prove[13] that in this case the applicability of Eq. (5) embraces the much wider region of $|t| < 10^{-1}$. The important point is that the critical amplitude ratio A^+/A^-, which is another fingerprint of the type of the universality class, is expected to be preserved even in the temperature region of weakened divergence.

TABLE I. Parameters (in R units) of the analysis of the magnetic specific heat of $UCuP_2$ in terms of Eq. (5). $A^{\pm}$ are the fluctuation amplitudes and ΔC is the height of the specific heat jump.

A^+	0.132 ± 0.01
A^-	0.264 ± 0.02
A^+/A^-	0.50 ± 0.05
ΔC	0.37 ± 0.05

As it is apparent from the semilogarithmic plots presented in the inset to Fig. 2, the magnetic specific heat of $UCuP_2$ follows the logarithmic dependence of Eq. (5) within the window $0.01 < |t| < 0.2$. The fitting parameters are summarized in Table I. We have to stress that the observed behavior cannot be regarded as an indication for $\alpha=0$ and thus it does not attest to the 2D Ising model. The reason is that the analysis was made outside the temperature region, where the ideal power-law dependence [i.e., Eq. (4)] could be expected. Similarly, the value of ΔC is worthless. On the contrary, as stated above, we can make use of the amplitude ratio A^+/A^-. Surprisingly, the measured value is very close to $A^+/A^-=0.52-0.54$, the value predicted for the 3D Ising model[14] and quite far from the value of $A^+/A^-=1$ expected for the 2D version of this model. Thus, it seems that $UCuP_2$ belongs to the 3D Ising universality class, in spite of its layered structure. This interesting finding will be discussed in more detail in the following sections.

C. Scaling function for in-field C_p

Figure 3(a) presents the magnetic part of the specific heat measured in an applied magnetic field. As expected for a ferromagnet (e.g., as for a Gd single crystal[15]), the anomaly in $C_M(T)$ is gradually smeared by rising field and the position of the steepest slope (the inflection point) moves toward higher temperatures. This behavior may be better understood by proper scaling analysis. According to the Widom hypothesis, the Gibbs free energy density of the magnetic system governed by the fluctuations of the order parameter can be expressed by some homogeneous function of the reduced variables of temperature (t) and magnetic field ($b=B/B_C$)[16,17]

$$F(t,b) = RTt^{d/y}\psi(z), \quad z = b/t^{x/y}, \tag{6}$$

where d is the dimensionality of the system, while x and y are some scaling factors for magnetic field and temperature, respectively. By the substitution $\tilde{z}=z^{-y/x}=t/b^{y/x}$ one can express the free energy in a more convenient form as

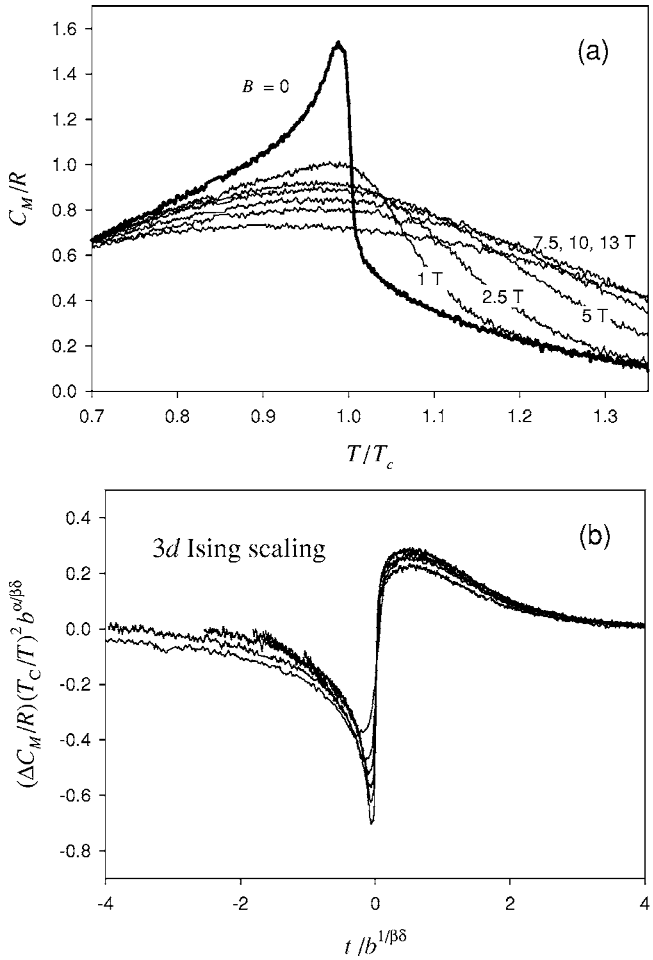

FIG. 3. (a) In-field magnetic specific heat of $UCuP_2$ measured in the vicinity of T_C. The data were taken upon cooling in a magnetic field applied along the easy magnetization axis. (b) Scaling plot for $\Delta C_M = C_M(B) - C_M(0)$ according to Eq. (10) for B=1, 2.5, 5, 7.5, 10, and 13 T. The values of the critical parameters α, β, and δ were taken as for the 3D Ising model.

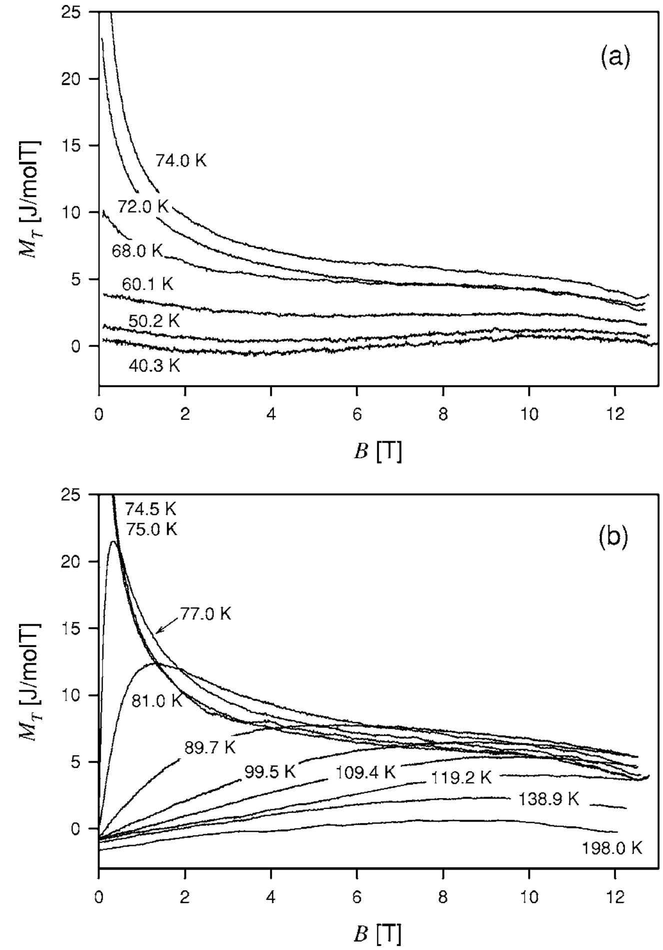

FIG. 4. Isothermal magnetocaloric coefficient for $UCuP_2$ measured at (a) $T < T_C$, (b) $T \geq T_C$=74.5 K. All the M_T data were taken with decreasing magnetic field applied along the easy magnetization axis.

$$F(t,b) = RTb^{d/x}\psi_2(\tilde{z}), \qquad (7)$$

where ψ_2 is another scaling function. The specific heat $C = -T(\partial^2 F/\partial T^2)_B$ may be given in a differential form,

$$\Delta C_M \equiv C_M(b) - C_M(0) = \frac{-RT^2}{T_C^2} b^{(d-2y)/x}[\psi_2''(\tilde{z}) - \psi_2''(\infty)], \qquad (8)$$

which leads to the dimensionless scaling relation

$$\frac{\Delta C_M}{R}\left(\frac{T_C}{T}\right)^2 b^{(2y-d)/x} = -[\psi_2''(\tilde{z}) - \psi_2''(\infty)] = \psi_3(\tilde{z}), \qquad (9)$$

where ψ_3 is yet another scaling function of the scaling variable $\tilde{z}$. Finally, using the formulas $\delta = x/(d-x)$ and $\nu = 1/y$ as well as the set of scaling laws one obtains

$$\frac{\Delta C_M}{R}\left(\frac{T_C}{T}\right)^2 b^{\alpha/\beta\delta} = \psi_3(t/b^{1/\beta\delta}), \qquad (10)$$

where α, β, δ, and ν are the usual critical exponents. As shown in Fig. 3(b), the above relation causes all the in-field $C_M(T)$ curves of $UCuP_2$ to collapse into a universal scaling function. Here, we adopted the values of the critical parameters for the 3D Ising model (α=0.119, β=0.326, and δ=4.8). We also tested the 2D Ising exponent values (not shown), yet the overlay of the curves was clearly worse than for the 3D case.

As shown by Krasnow and Stanley[18] for the case of the 3D Ising and Heisenberg classes, the general shapes of the C_p scaling functions are quite similar for different models. Therefore, because of limited accuracy of the experimental data, in model discrimination it is more convincing to use as an argument the quality of the data collapse. On this basis we conclude that the above scaling analysis of the in-field specific heat of $UCuP_2$ can be regarded as a further argument in favor of the 3D Ising model.

IV. ISOTHERMAL MAGNETOCALORIC EFFECT

A. General features

The field dependencies of the isothermal magnetocaloric coefficient M_T measured at different temperatures are presented in Fig. 4. It is worth noting that M_T does not contain any nonmagnetic contributions. Therefore, there is no need to subtract any background data, as is unavoidable in the analysis of the magnetic specific heat, and the thermodynamical information can be derived directly from the "as-measured" data.

The isothermal magnetocaloric coefficient of $UCuP_2$ is positive in both the paramagnetic and ordered regions. This is a simple consequence of decreasing entropy (ΔS) upon increasing external field, as expected for a ferromagnet,

$$\Delta S(B) = -\int_0^B \frac{M_T}{T} dB. \tag{11}$$

Far below T_C (T=40.3 and 50.2 K) the coefficient M_T is close to zero and hardly field dependent [see Fig. 4(a)]. The visible small variations with B should rather be attributed to experimental errors. With rising temperature M_T clearly tends to increase (note the isotherms T=60.1 and 68.0 K), especially for low fields (T=72.0 K). For temperatures close to T_C [T=74.0, 74.5, and 75.0 K—see also Fig. 4(b)] M_T diverges in weak fields reaching values above 25 J/mol T for $B<0.5$ T. At the Curie temperature T_C we estimated the entropy decrease between 0 and 13 T to be $\Delta S(B=13\text{ T}) \approx -0.17R$ only. This small value is not surprising, if one takes into account that our maximum available laboratory field is only a small fraction of the characteristic magnetic field B_C=111 T. Only for $B \gg B_C$ does one expect ΔS to reach the theoretical value $-R \ln 2$.

For temperatures above T_C the overall features of the $M_T(B)$ curves change dramatically. In contrast to those taken in the ordered state, which start from a finite value, all the $M_T(B)$ curves measured in the disordered phase (note the isotherms T=77.0 K and above) begin from the zero value (to be precise we should note that the apparent curves start from $M_T \approx -1$ J/mol T, yet we assume this small shift to be a systematic measurement error). Next, all curves pass through a maximum. With increasing temperature this maximum shifts toward higher fields and broadens. Such a fundamental difference in the behavior of the isothermal magnetocaloric coefficient above and below T_C can be explained based on the thermodynamic relation between M_T and the magnetization M,

$$M_T = -T\left(\frac{\partial M}{\partial T}\right)_B. \tag{12}$$

There is no spontaneous magnetization above T_C and hence $dM/dT=0$ at $B=0$ which yields $M_T(B=0)=0$. On the other hand, below T_C the slope of the temperature dependence of the spontaneous magnetization at zero field is finite and negative. This results in finite and positive values of M_T.

TABLE II. The critical exponent ω for the isothermal magnetocaloric coefficient M_T for some important universality classes (d is the spatial dimensionality, n is the dimensionality of the order parameter, SAW denotes the self-avoiding walks model).

Class	ω
MFA	0.333(1/3)
2D Ising (n=1)	0.467(7/15)
3D SAW n=0	0.479
3D Ising (n=1)	0.433
3D XY (n=2)	0.392
3D Heisenberg (n=3)	0.364

B. Critical parameters for M_T

When the critical temperature is approached both the slope $(dM_T/dB)_{B\to 0}$ on the high-temperature side and the limiting value of $M_T(B\to 0)$ strongly increase. Eventually, at $T=T_C$ the dependence $M_T(B)$ diverges for $B\to 0$. As for other thermodynamical quantities one expects that at T_C the isothermal magnetocaloric coefficient follows a scaling law in the form

$$M_T \approx A_m b^{-\omega}, \tag{13}$$

where ω is the critical exponent, A_m is the magnetocaloric critical amplitude, and $b=B/B_C$ is the reduced magnetic field normalized by the characteristic field B_C. Following the usual way of reasoning in scaling theory one can derive the relation between ω and other critical exponents. According to the Widom hypothesis, the free energy satisfies the scaling relation

$$\tilde{F}(L^y t, L^x b) = L^d F(t,b), \tag{14}$$

where $\tilde{F}$ is the free energy of a volume expanded by some scaling factor L and d stands for the dimensionality of the system, while x and y are the scaling factors for the magnetic field and the temperature, respectively. Taking into account the relation $M=(\partial F/\partial B)_T$ and Eq. (12), the critical exponent for M_T can be expressed as a function of the respective exponents for the temperature dependencies of the specific heat (α) and the magnetization (β) as well as the field dependence of magnetization (δ):

$$\omega = \frac{1-\beta}{2-\alpha-\beta} = \frac{1-\beta}{\beta\delta}. \tag{15}$$

In Table II the critical exponent ω is given as calculated from Eq. (15) for a few important systems using the corresponding α and β values.[19] It is characteristic that for all the systems considered ω varies within the window of 0.4 ± 0.1. Thus, for all universality classes a strong divergence in $M_T(B)$ for $B\to 0$ is expected, even for the mean-field approximation or XY systems, for which, respectively, only a finite step or a finite peak in the specific heat is predicted.

Like other scaling laws, Eq. (15) leads to some associated universal combination of critical amplitudes, which depends only on the type of the universality class. Using the proce-

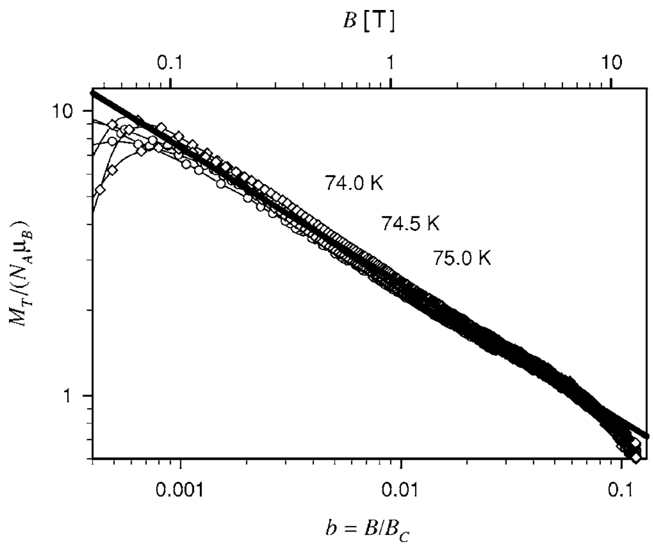

FIG. 5. Log-log plot of the isothermal magnetocaloric coefficient measured for $UCuP_2$ at temperatures close to T_C=74.5 K. The data for increasing (diamonds) and decreasing (circles) magnetic field are shown. The thick solid line represents the fit according to Eq. (13).

dure described in Sec. 1.3 of Ref. 14 it is easy to prove that the following combination, containing A_m, is universal:

$$R_{m1} \equiv A_m \xi_c{}^d \left(\frac{D}{A^+}\right)^{1/\beta\delta}, \tag{16}$$

where ξ_c is the amplitude for the field dependence of the correlation length ($\xi \approx \xi_c b^{-\nu/\beta\delta}$) and D is the amplitude for the temperature dependence of the magnetization ($M \approx D|t|^\beta, t<0$). Equivalently, another combination is also universal,

$$R_{m2} \equiv A_m \xi_c{}^d \left(\frac{D_c}{D}\right)^{1/\beta}. \tag{17}$$

Here, the symbol D_c denotes the amplitude for the field dependence of the magnetization $[M \approx (1/D_c) b^{1/\delta}]$.

Figure 5 presents the log-log plot of the $M_T(B)$ data of $UCuP_2$ measured at three different temperatures close to T_C. The values of M_T were normalized by the factor $N_A\mu_B$ =5.584 J/mol T (N_A is the Avogadro number, μ_B is the Bohr magneton), which is the magnetic analog of the gas constant. It is worth emphasizing that even at B=13 T the system remains in the low-field regime ($b \ll 1$), because our highest available external field is much smaller than the characteristic field B_C=111 T, derived for $UCuP_2$. In consequence, as is apparent from Fig. 5, the isothermal magnetocaloric coefficient follows pretty well the scaling law across two orders of magnitude of the magnetic field. Some deviations appearing in weak fields presumably result from rounding the M_T anomaly due to experimental conditions and/or because of the effect of ferromagnetic domains. In turn, some deviations in high-field region come probably from a rising error-to-signal ratio.

TABLE III. Parameters of fitting Eq. (13) to the experimental $M_T(B)$ data for $UCuP_2$: ω is the critical exponent and A_m is the magnetocaloric fluctuation amplitude. The latter parameter is given in $N_A\mu_B$ units.

ω	0.48±0.05
A_m	0.270±0.007

The fitting parameters are given in Table III. The value of ω is close to that calculated for the 2D Ising system (compare Table II); however, the 3D Ising model cannot be ruled out due to the experimental uncertainty. Interpretation of the amplitude A_m in terms of Eqs. (16) or (17) is quite troublesome, since we do not know the respective universal constant R_{m1} or R_{m2}. We can only notice that $A_m \sim 1/\xi_c{}^d$, i.e., the magnetocaloric amplitude is inversely proportional to the magnetic correlation volume ($\xi_c{}^d$). This is in analogy to the well-known relation for the specific heat amplitude $A^+ \sim 1/\xi_0^{+d}$, where ξ_0^{+d} is the usual correlation volume derived from the temperature dependence of the correlation length.

C. Scaling function for M_T

To derive the full scaling function for the isothermal magnetocaloric coefficient M_T we start from Eq. (6), as for the specific heat. First, we calculate the magnetization $M = (\partial F/dT)_T$,

$$M = \frac{RT}{B_C} t^{(d-x)/y} \psi_1'(z). \tag{18}$$

As in the analysis of the specific heat, the scaling factors x and y can be expressed in terms of the usual critical exponents. Next, we transform the above equation into its dimensionless form (we recall that $\mu_B B_C = k_B T_C$ and $b = B/B_C$)

$$\frac{M}{N_A\mu_B}\frac{T_C}{T}|t|^{-\beta} = \psi_4(b/|t|^{\beta\delta}), \tag{19}$$

where ψ_4 is some homogeneous function describing the scaling of the magnetization.

The above equation was used to analyze the experimental magnetization data for $UCuP_2$. As for C_p, we tested two sets of the critical parameters, for the 3D and 2D Ising models. To give an example, Fig. 6 presents the results of both trials. The difference in the quality of the data collapse is quite spectacular. Only with the 3D Ising scaling (top panel) do the data form two well-defined branches, separately for the low- and high-temperature sides of the transition. As expected, for high values of the scaled magnetic field ($b/|t|^{\beta\delta} > 10$), i.e., far away from the critical point, both branches merge asymptotically.

In the next step we use Eq. (12) to obtain the scaling function for the isothermal magnetocaloric coefficient,

$$\frac{M_T}{N_A\mu_B}\left(\frac{T_C}{T}\right)^2 |t|^{1-\beta} = \psi_5(b/|t|^{\beta\delta}), \tag{20}$$

where ψ_5 represents the magnetocaloric scaling function. Before and after the temperature differentiating, respectively, we applied the $z = \tilde{z}^{-x/y}$ and $\tilde{z} = z^{-y/x}$ variable transformations.

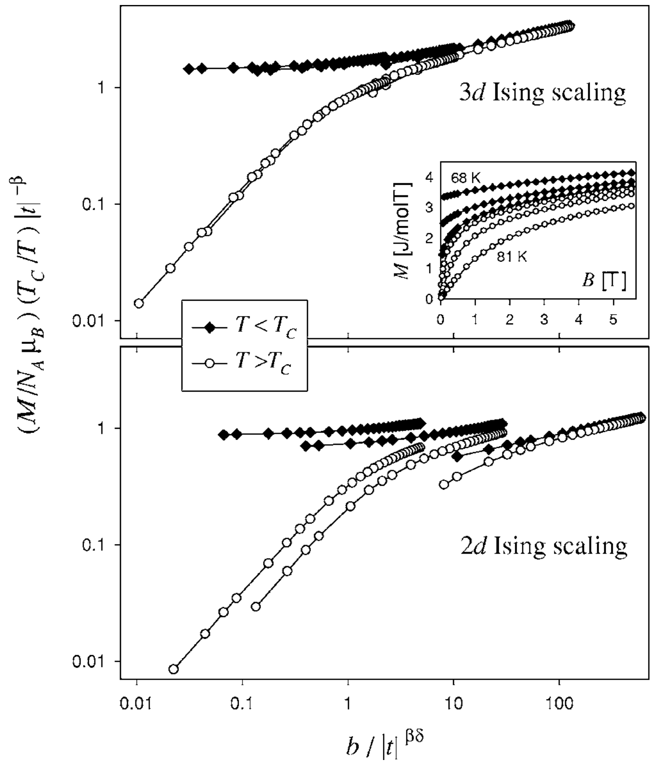

FIG. 6. Magnetization scaling plot for $UCuP_2$ according to Eq. (19) for magnetic fields up to $B=5.5$ T. The data for $T<T_C$ ($T=68$, 72, and 74 K, solid diamonds) and $T>T_C$ ($T=75$, 77, and 81 K, empty circles) are included. The values of the critical parameters α, β, and δ, were taken as for the 3D (top panel) and 2D Ising models (bottom panel). The inset in the top panel presents the raw magnetization data versus magnetic field at all six temperature values.

Figure 7 displays the scaling of the magnetocaloric data of $UCuP_2$ with the critical exponents appropriate for the 3D Ising model. Apparently, the data for $T<T_C$ and $T>T_C$ excellently collapse onto two different universal curves. The low-temperature branch monotonically descends, whereas the high-temperature branch exhibits a maximum at $b/|t|^{\beta\delta}\approx 0.5$. The latter expression describes the temperature dependence of the position of the maximum in the $M_T(B)$ curves observed for $T>T_C$. As for the scaled magnetization, the two branches converge for $b/|t|^{\beta\delta}>10$. A similar analysis was performed for the 2D Ising model, but the data collapse was of much worse quality, especially for low magnetic fields (not shown).

V. SUMMARY

The heat-flow calorimetric method,[6] developed recently, was applied to investigate the thermodynamics of the ferromagnetic compound $UCuP_2$ with $T_C=74.5$ K. The specific heat at constant magnetic field and the isothermal magnetocaloric coefficient were measured over the temperature interval $20<T<300$ K and the magnetic field range $0<B<13$ T.

The zero-field specific heat qualitatively agrees with the previous data;[9] however the estimations of the magnetic entropies differ substantially ($S_{magn}\approx 5.7$ J/mol K $\approx R\ln 2$ has been obtained from our data versus $S_{magn}\approx 1.2$ J/mol K given in Ref. 9). The analysis of the $C_p(T)$ anomaly near the critical temperature in terms of the asymptotic formula for $\alpha\to 0^+$ [Eq. (5)] yielded the critical amplitude ratio $A^+/A^-=0.50\pm 0.05$. The comparison of this value with the theoretical results[14] clearly indicates that $UCuP_2$ belongs to the 3D Ising universality class, in spite of its layered crystal structure[2] and very strong anisotropy of the magnetic properties.[3] This conclusion has been corroborated by the scaling analysis of the in-field specific heat curves: only with the set of the critical exponents predicted by the 3D Ising model all the $C_p(T,B)$ curves follow a universal scaling function [Eq. (10)].

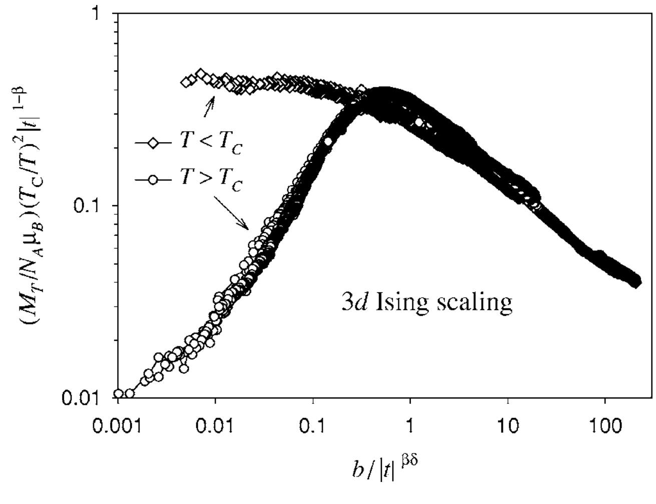

FIG. 7. Scaling plot for the isothermal magnetocaloric coefficient in $UCuP_2$ according to Eq. (20). The data for $T<T_C$ ($T=50.2$, 60.1, 68, 70, and 72 K, diamonds) and $T>T_C$ ($T=77$, 79.8, 81, 89.7, 109.4, and 119.2 K, circles) are included.

The behavior of the isothermal magnetocaloric coefficient $M_T(B)$ has been found to be substantially different in the ordered and the paramagnetic states. Far away from the critical temperature, M_T is positive and small both below and above T_C. However, when $t\to 0^-$ the $M_T(B)$ dependence strongly bends upward for $B\to 0$ (still being finite at $B=0$), and finally develops into a divergence at T_C. In contrast, just above T_C the $M_T(B)$ curve starts from $M_T=0$ at $B=0$, initially rises steeply, then reaches a maximum, and eventually slowly falls down. The critical curve for $T=T_C$ has been found to follow a simple power law [Eq. (13)] over two decades of the magnetic field. According to the derived scaling law for the magnetocaloric critical exponent, its experimental value $\omega=0.48\pm 0.05$ is close to those expected for the 2D Ising (0.467) and the 3D Ising (0.433) models. In order to discriminate between these two universality classes a full scaling analysis of the $M_T(B)$ curves [Eq. (20)] has been performed. Only with the set of the 3D Ising critical parameters do all the magnetocaloric data form two well-defined branches, separately for the low- and high-temperature sides of the transition. A similar result, unambiguously pointing to the 3D Ising model, was obtained via the scaling analysis of the magnetization data [Eq. (19)].

The present example of ferromagnetic $UCuP_2$ confirms that the recently developed method of measuring the isothermal magnetocaloric coefficient is a useful tool for studying the critical phenomena in magnetic materials. The two main advantages of investigating M_T are the absence of any nonmagnetic contributions and the high value of the magnetocaloric critical exponent ($0.3<\omega<0.5$ versus $-0.1<\alpha<0.2$). The latter feature ensures large and easily measurable divergence in $M_T(B)$ for small magnetic fields. Additional merits are the possibility to measure the $C_p(T,B)$ dependencies using the same experimental setup (upon both heating and cooling), and, last but not least, the simplicity of the method.

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[1] D. Kaczorowski, *Ternary Actinide Pnictides and Chalcogenides*, Landolt-Börnstein, New Series, Group III, Vol. 27, pt. B8 (Springer-Verlag, Berlin, 2004).
[2] H. Noël, Z. Zołnierek, D. Kaczorowski, and R. Troć, J. Less-Common Met. **132**, 327 (1987).
[3] D. Kaczorowski, R. Troć, and H. Noël, J. Phys.: Condens. Matter **3**, 4959 (1991).
[4] N. Korner, J. Schoenes, and D. Kaczorowski, Helv. Phys. Acta **62**, 207 (1989).
[5] P. Fumagalli, J. Schoenes, and D. Kaczorowski, Solid State Commun. **65**, 173 (1988).
[6] T. Plackowski, Y. Wang, and A. Junod, Rev. Sci. Instrum. **73**, 2755 (2002).
[7] T. Plackowski, A. Junod, F. Bouquet, I. Sheikin, Y. Wang, A. Jeżowski, and K. Mattenberger, Phys. Rev. B **67**, 184406 (2003).
[8] T. Plackowski, D. Kaczorowski, and Z. Bukowski, Phys. Rev. B **72**, 184418 (2005).
[9] A. Böhm, D. Kaczorowski, G. Weber, and F. Steglich, J. Alloys Compd. **196**, L11 (1993).
[10] C. Domb, Adv. Phys. **9**, 149 (1960); see also *The Critical Point: A Historical Introduction to the Modern Theory of Critical Phenomena* (Taylor & Francis, London, 1996).
[11] J. Mucha, H. Misiorek, D. Kaczorowski, and A. Jeżowski, J. Alloys Compd. **189**, 217 (1992).
[12] J. A. Lipa, C. Edwards, and M. J. Buckingham, Phys. Rev. A **15**, 778 (1977).
[13] The analytical formula for C_p for the 2D Ising model is given, e.g., in J. J. Binney, N. J. Dowrick, A. J. Fisher, and M. E. J. Newman, *Theory of Critical Phenomena. An Introduction to the Renormalization Group* (Clarendon Press, Oxford, 1992).
[14] V. Privman, P. C. Hohenberg, and A. Aharony, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic Press, New York, 1991), Vol. 14.
[15] D. S. Simons and M. B. Salamon, Phys. Rev. B **10**, 4680 (1974).
[16] M. A. Fisher, Rev. Mod. Phys. **70**, 653 (1998).
[17] H. E. Stanley, Rev. Mod. Phys. **71**, S358 (1999).
[18] R. Krasnow and H. E. Stanley, Phys. Rev. B **8**, 332 (1973).
[19] P. Chaikin and T. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, England, 1995).