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Specific heat of heavy-fermion URu_2Si_2 in high magnetic fields

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We have performed specific-heat measurements on the heavy-fermion superconductor URu_2Si_2 in magnetic fields up to 17.5 T. A sharp peak in the specific heat signals the antiferromagnetic transition at $T_N = 17.5$ K, which shifts to lower temperatures in applied magnetic fields. In order to describe the specific heat below T_N, we have used the characteristic features of the excitation spectrum measured by neutron scattering. The relative field dependence of the antiferromagnetic transition temperature T_N and the energy gap Δ in the magnetic excitation spectrum can be described by a single scaling relation of the form $[1-(B/B_0)^2]$. The scaling field of 48.5 T is close to the metamagnetic transition field $B^* = 40$ T, where the heavy-fermion state is suppressed. [S0163-1829(97)05746-9]

I. INTRODUCTION

The heavy-fermion superconductor URu_2Si_2 has attracted continuous attention in the last decade for its unusual ground-state properties. At $T_N = 17.5$ K the system undergoes an antiferromagnetic phase transition which is accompanied by sharp peaks in the specific heat[1,2] and the thermal expansion.[3] A second transition occurs at $T_c = 1.2$ K and indicates the onset of superconductivity which coexists with the antiferromagnetic order. Neutron-scattering measurements[4,5] revealed a simple antiferromagnetic structure with a tiny ordered moment of $(0.03 \pm 0.01)\mu_B$/U atom, oriented along the c axis of the tetragonal crystal structure. The formation of an energy gap in the magnetic excitation spectrum is reflected by an exponential temperature dependence of the specific heat,[1,2] the thermal expansion,[3] and the NMR and nuclear quadrupole-resonance (NQR) relaxation rates[6] in the ordered state. Electrical resistivity[7] and point-contact spectroscopy measurements[8] show a similar energy gap, indicating a strong scattering of the conduction electrons by the magnetic excitations. Magnetization measurements in high magnetic fields[9,10] show a suppression of the heavy-fermion state in three subsequent steps at 35.8, 37.3, and 39.4 T for fields along the easy axis ($B \| c$). These transitions have been confirmed in high-field measurements of the magnetoresistance and Hall coefficient.[11]

Although the magnetic excitation spectrum is known, the nature of the magnetic order is still unclear. The large anomalies in the specific heat and thermal expansion at T_N are hard to reconcile with the small ordered moment. The energy gap in the magnetic excitation spectrum can qualitatively be described with a singlet-singlet model[12,13] for the $5f^2$ configuration (U^{4+}), but the estimated ordered moment is considerably larger than the observed value. A comparison of the scaling behavior of the excitation gap[7] and the ordered moment[14] as function of magnetic field indicates that the ordered moment shows a stronger suppression than the excitation gap in magnetic fields up to 8 T. This suggests the existence of two distinctly different critical fields for the magnetic excitation gap and the ordered moment. However, recent neutron-scattering experiments[15] in a magnetic field of 12 T do not show a substantial difference in the critical temperatures of the magnetic excitation gap and the ordered moment.

The discrepancy in the description of the magnetic excitation gap and observed ordered moment has led to the suggestion that the dipolar-ordered moment may not be the (main) order parameter that drives the magnetic phase transition (see Ref. 16 for a recent overview). The order parameter could be of nondipolar nature (e.g., quadrupolar), as suggested by a strong signal in the nonlinear susceptibility at the magnetic phase transition.[17] Alternatively, Barzykin and Gor'kov[18] have proposed an order parameter which involves triple-spin correlators. However, recent high-field neutron experiments[14] have ruled out the possibility of the triple-spin correlators. Additional polarized neutron-scattering experiments[19] have shown that the antiferromagnetic Bragg peaks, observed in low fields, arise from purely dipolar order. It has been suggested that there may be two successive phase transitions,[19,20] where the first is quadrupolar and the second, at a slightly lower temperature, is dipolar.

We have performed specific-heat measurements on a single crystal of URu_2Si_2 in magnetic fields up to 17.5 T in order to study the field dependence of the energy gap in the magnetic excitation spectrum. Previous specific-heat measurements in magnetic fields were performed in magnetic fields up to 5 T for single-crystalline samples[3] and 7 T for polycrystalline samples.[21] We will use the characteristic features of the magnetic excitation spectrum to describe the specific heat in the antiferromagnetically ordered state.

II. SPECIFIC HEAT IN HIGH MAGNETIC FIELDS

The specific-heat measurements were performed on a single crystal of URu_2Si_2, grown in a tri-arc furnace with the Czochralski method and annealed for 8 days at a temperature

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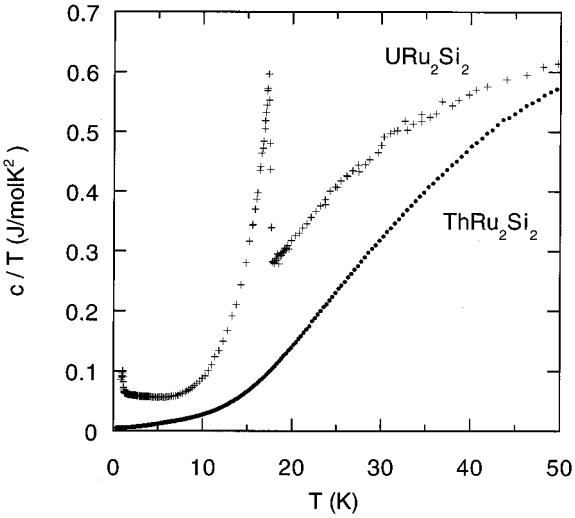

FIG. 1. Specific heat of URu_2Si_2 in zero field. The anomaly at $T_N = 17.5$ K signals the antiferromagnetic transition, while superconductivity sets in at $T_c = 1.2$ K. For comparison, the specific heat of the nonmagnetic reference system $ThRu_2Si_2$ (Ref. 24) is shown.

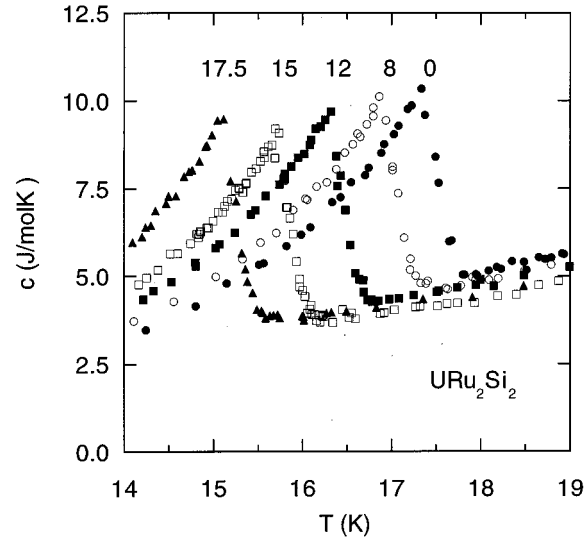

FIG. 2. Specific heat of URu_2Si_2 around the antiferromagnetic phase transition in magnetic fields of 0, 8, 12, 15, and 17.5 T ($B \| c$). The transition temperature T_N decreases, while the jump in the specific heat is roughly constant in magnetic field.

of 950 °C. The sample, with a mass of 1.34 g, was mounted with the c axis of the tetragonal crystal structure along the field direction of a 17.5-T superconducting magnet. We have measured the specific heat of URu_2Si_2 with an adiabatic technique[22] in constant magnetic fields (0, 8, 12, 15, and 17.5 T) in the temperature range from 0.5 to 50 K.)

In Fig. 1, the zero-field specific heat is shown. The data are in good agreement with earlier specific-heat measurements[23] on the same sample. At $T_N = 17.5$ K, a sharp peak signals the antiferromagnetic transition, while at low temperatures the electronic specific heat shows a continuous increase until superconductivity sets in at $T_c = 1.2$ K. For comparison, the specific heat of the nonmagnetic reference system $ThRu_2Si_2$ (Ref. 24) is shown.

The effect of an applied magnetic field on the antiferromagnetic transition is shown in Fig. 2. For increasing magnetic fields, the transition temperature T_N is gradually suppressed, while the jump in the specific heat at the transition $\Delta c(T_N)$ is nearly independent of the magnetic field, as listed in Table I. The field-dependent values of T_N and $\Delta c(T_N)$ have been determined by an equal-entropy construction[25] with an ideal step transition.

III. EVALUATION OF THE EXCITATION GAP AND DISCUSSION

The phonon contribution to the specific heat of URu_2Si_2 is determined from the specific heat of the nonmagnetic reference system $ThRu_2Si_2$. At low temperatures, the specific heat of $ThRu_2Si_2$ (Ref. 24) is well described by an electronic term of $\gamma = 5.53$ mJ/mol K^2 and three acoustical modes with a Debye temperature of $\Theta_D = 184$ K. A clear deviation of the Debye temperature dependence for the specific heat is observed above 10 K, which can accurately be described up to 80 K with six additional optical modes with an Einstein temperature of $\Theta_E = 206$ K. After subtraction of the linear electronic term, the phonon contribution is obtained. Specific-heat measurements of both URu_2Si_2 and $ThRu_2Si_2$ at higher temperatures[26] show that the specific-heat curves nearly coincide above 100 K, confirming the close similarity of the phonon spectrum for both systems.

For temperatures far below the antiferromagnetic transition, the specific heat is mainly determined by the electronic contribution. In Fig. 3, the low-temperature specific heat is shown in zero field and in a magnetic field of 17.5 T after subtraction of the phonon contribution. The electronic specific heat of URu_2Si_2 shows a moderate mass enhancement by the electron correlations which increases with decreasing temperatures and is insensitive to the applied magnetic field up to 17.5 T (for $T > T_c$). This is consistent with the situation in other heavy-fermion systems as UPt_3 (Ref. 27) and $CeRu_2Si_2$ (Ref. 28), where far below the metamagnetic transition, the electronic specific heat is independent of the applied magnetic field. The metamagnetic transition in URu_2Si_2 occurs in three subsequent steps near 40 T,[9,10] which is far above the field range of interest in this experiment. The superconductivity is suppressed at a relatively low critical field of $B_{c2} = 3$ T for fields along the c axis.[29]

The low-temperature specific heat for systems close to an antiferromagnetic instability can be described by the spin-fluctuation model of Moriya and Takimoto.[30] The electronic specific heat related to the spin fluctuations depends on a characteristic temperature T_0, a cutoff wave vector q_c, and parameters y_0 and y_1, which describe the reduced inverse staggered susceptibility y. As the magnetic excitations in URu_2Si_2 are predominantly found along the c axis,[5,24] we use a cutoff wave vector of $q_c = 2\pi/c$. Good agreement with the specific heat at low temperatures is obtained for $T_0 = 73$ K, $y_0 = 0.02$, and $y_1 = 10$, as shown in Fig. 3. The characteristic spin-fluctuation temperature T_0 is of the same order of magnitude as the coherence temperature which is obtained from the maxima in the resistivity[2] and the susceptibility.[1,2] The temperature dependence of the electronic specific heat at

TABLE I. Experimental values of the antiferromagnetic transition temperature T_N, the jump in the specific heat $\Delta c(T_N)$, and the magnetic entropy $s_m(T_N)/R$ of the magnetic order for different magnetic fields B, applied along the c axis. The magnetic entropy in the paramagnetic state is fitted to a model of crystal-field excitations with a Lorentzian broadening. The fitting parameters E_0 and W correspond to the size and the full width at half maximum of the broadened level splitting.

B (T)	T_N (K)	$\Delta c(T_N)$ (J/mol K)	$s_m(T_N)/R$	E_0 (K)	W (K)
0.0	17.54(1)	6.3(4)	0.152(3)	59(1)	2(1)
8.0	17.06(1)	6.6(4)	0.148(3)		
12.0	16.46(1)	6.1(4)	0.152(3)	59(1)	11(2)
15.0	15.85(1)	5.9(4)	0.148(3)		
17.5	15.26(1)	6.4(4)	0.147(3)	60(1)	21(2)

higher temperatures is shown in the inset of Fig. 3 for the obtained model parameters.

In order to analyze the magnetic contribution of the specific heat, we have to consider the magnetic excitation spectrum of URu_2Si_2, which has been determined from inelastic neutron-scattering measurements.[5,23] The observed magnetic excitations are polarized along the tetragonal c axis and show two characteristic minima in the dispersion curve, which are both located on the zone boundary. The first minimum has an excitation gap of 2.5 meV (29 K) and is found at $\mathbf{Q}=(1\ 0\ 0)$, which corresponds to the magnetic Bragg point of the antiferromagnetic structure. Around this minimum a linear dispersion is observed along all principal directions for the relevant energy range of the thermal excitations ($\Delta E \leqslant k_B T_N$). The second minimum is located at $\mathbf{Q}=(1.4\ 0\ 0)$ and shows an excitation gap of 4.5 meV (52 K). The intensity of the longitudinal magnetic excitations rapidly decreases away from the two minima in the dispersion curve.

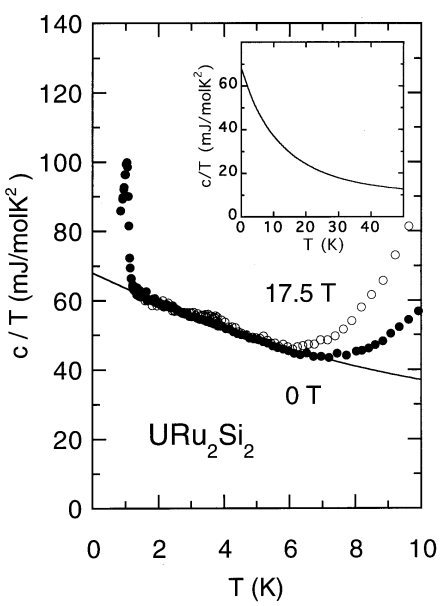

FIG. 3. Low-temperature specific heat of URu_2Si_2 in zero field and in a magnetic field of 17.5 T after subtraction of the phonon contribution. The solid curve corresponds to a fit of the electronic contribution with the spin-fluctuation model (Ref. 30) for a spin-fluctuation temperature of $T_0=73$ K and a reduced susceptibility described by $y_0=0.02$ and $y_1=10$. The inset shows the model prediction at higher temperatures.

The specific heat in the antiferromagnetically ordered state ($T<T_N$) is frequently described by an empirical fitting function of the form[21]

$$c_m(T)=A\ \exp(-\Delta/k_B T). \tag{1}$$

The estimated values of the energy gap Δ obtained with this function are typically in the order of 140 K for the specific heat of URu_2Si_2.[21] The electrical resistivity[1,7] has been analyzed with a simple model[31] for the scattering of conduction electrons by magnetic excitations which show an energy gap with a quadratic dispersion. In the low-temperature limit ($k_B T \ll \Delta$), the electrical resisistivity is described by a considerably smaller and strongly anisotropic energy gap of $\Delta=73$ K for $I \| a$ and $\Delta=51$ K for $I \| c$.[1,7] Inelastic neutron-scattering measurements[5,23] indicate an even smaller gap. The magnetic excitation spectrum of URu_2Si_2 shows a minimum of 29 K at $\mathbf{Q}=(1\ 0\ 0)$ and a second minimum of 52 K at $\mathbf{Q}=(1.4\ 0\ 0)$.

Although the empirical fitting function of Eq. (1) can reproduce the shape of the specific heat in the ordered state, it gives unrealistic values for the gap in the magnetic excitation spectrum and is therefore inadequate. The electrical resistivity measurements are analyzed in terms of a physical model, but the assumption of the low-temperature limit is not fulfilled for the temperature range of the fitted data. In order to describe the magnetic specific heat in the ordered state, we will use the essential features of the magnetic excitation spectrum determined by inelastic neutron-scattering measurements. As the energy difference between the two minima in the magnetic excitation spectrum is larger than the thermal energy at T_N, we will only consider the lowest minimum at $\mathbf{Q}=(1\ 0\ 0)$.

The magnetic specific heat of a system with independent excitations can be described as

$$c_m=\int_0^\infty E g(E)\{df(E)/dT\}dE, \tag{2}$$

where $g(E)$ is the density of states and $f(E)=1/[\exp(E/k_B T)-1]$ is the Bose factor for magnetic excitations. The density of states is directly related to the excitation spectrum. For an isotropic excitation spectrum of the form $E(k)=\Delta+\alpha k^\beta$ the density of states is

$$g(E)=(V_m/2\pi^2\beta\alpha^{3/\beta})(E-\Delta)^{3/\beta-1}, \tag{3}$$

TABLE II. Fitting parameters for the measured specific heat of URu_2Si_2 for different magnetic fields B, applied along the c axis. The energy gap Δ_0 and the linear dispersion α are fitted with a temperature-dependent gap function of the form $\Delta(T)=\Delta_0(1-(T/T_N)^x)$ with $x=3$.

B (T)	$T_N(B)$ (K)	$T_N(B)/T_N(0)$	$\Delta_0(B)$ (K)	$\Delta_0(B)/\Delta_0(0)$	$\alpha(B)$ (KÅ)	$\alpha(B)/\alpha(0)$
0.0	17.54(1)	1	26.7(4)	1	105.4(2)	1
8.0	17.06(1)	0.973(1)	25.6(5)	0.96(3)	103.5(2)	0.982(3)
12.0	16.46(1)	0.938(1)	24.1(5)	0.90(3)	100.0(2)	0.949(3)
15.0	15.85(1)	0.904(1)	23.8(6)	0.89(3)	99.5(2)	0.944(3)
17.5	15.26(1)	0.870(1)	23.6(4)	0.88(3)	94.2(2)	0.894(3)

with $g(E)=0$ for $E<\Delta$ and a molar volume of $V_m=4.91\times10^{-5}$ m^3 for URu_2Si_2. At low temperatures ($k_BT \ll \Delta$), the leading term in the specific heat of a system with a magnetic excitation spectrum of the form $E(k)=\Delta+\alpha k^\beta$ is given by

$$c_m(T)=(V_m k_B/2\pi^2\beta\alpha^{3/\beta})\Gamma(3/\beta)\Delta^2(k_BT)^{3/\beta-2} \times\exp(-\Delta/k_BT), \quad (4)$$

where $\beta\neq0$ and $\Gamma(z)$ is the gamma function. For an excitation spectrum with a dispersionless gap ($\beta=0$), the Einstein function of optical phonons is obtained with the following low-temperature behavior:

$$c_m(T)=R\Delta^2(k_BT)^{-2}\exp(-\Delta/k_BT), \quad (5)$$

where R is the gas constant. The frequently used exponential relation for the low-temperature specific heat of URu_2Si_2 [Eq. (1)] corresponds in this scheme to a value of $\beta=3/2$. The lowest minimum in the measured excitation spectrum at $\mathbf{Q}=(1\,0\,0)$, however, shows a linear dispersion ($\beta=1$) for all principal directions. This leads to an additional temperature dependent prefactor for the exponential temperature dependence of the specific heat at low temperatures. The measured specific heat of URu_2Si_2 at low temperatures is dominated by the large electronic contribution. For a physical description of the magnetic excitations, we need to extend the specific heat to higher temperatures and use the explicit form of Eq. (2). The temperature dependence of the gap in the excitation spectrum can be taken into account by introducing a temperature-dependent gap function of the form $\Delta(T)=\Delta_0(1-(T/T_N)^x)$. For an excitation spectrum of the form $E(k)=\Delta+\alpha k^\beta$, the specific heat is described by

$$c_m(T)=(V_m k_B/2\pi^2\beta)(k_BT/\alpha)^{3/\beta}\int_0^\infty d\varepsilon(\varepsilon+\delta)\varepsilon^{3/\beta-1} \times(\varepsilon+\delta-\delta')\exp(\varepsilon+\delta)/[\exp(\varepsilon+\delta)-1]^2, \quad (6)$$

where $\varepsilon=(E-\Delta)/k_BT$, $\delta=\Delta/k_BT$, and $\delta'=d\Delta/d(k_BT)$. The maximum value of the specific heat is observed at the antiferromagnetic transition. At T_N the excitation gap vanishes and Eq. (6) reduces to

$$c_m(T_N)=(V_m k_B/2\pi^2\beta)(k_BT_N/\alpha)^{3/\beta} \times\{\Gamma(3/\beta+2)\zeta(3/\beta+1)-\delta_N'\Gamma(3/\beta+1)\zeta(3/\beta)\}, \quad (7)$$

where $\delta_N'=-x\Delta_0/k_BT_N$ is the slope of the gap function at T_N and $\zeta(z)$ is Riemann's zeta function. The best fit of the experimental data with the measured magnetic excitation spectrum is obtained for a temperature-dependent gap function with $x=3$. The values of T_N, Δ_0, and α for the different applied magnetic fields are listed in Table II. In Fig. 4 the magnetic contribution to the specific heat is compared with the corresponding fit to the magnetic excitation spectrum [Eq. (6)] in zero field and in a magnetic field of 17.5 T.

In Fig. 5 the relative field dependences of T_N, Δ_0, and α are shown. The field dependence of the transition temperature closely follows a simple scaling relation $T_N(B)/T_N(0)=[1-(B/B_0)^2]$, where $T_N(0)=17.54(1)$ K and $B_0=48.5(1)$ T. The estimated scaling field is comparable to the value of $B_0=40$ T obtained by magnetoresistance measurements[7] in magnetic fields up to 25 T. Recent measurements of the thermal expansion in magnetic fields up to 25 T (Ref. 32) and the magnetization up to 20 T (Ref. 33) confirmed the existence of this simple scaling relation for T_N with scaling fields of $B_0=37$ T and $B_0=44$ T, respectively.

The energy gap in the magnetic excitation spectrum, estimated from the specific-heat measurements (Table II), is close to the value of 29 K observed in neutron-scattering measurements at $\mathbf{Q}=(1\,0\,0)$. In high magnetic fields, the energy gap shows a small reduction with roughly the same

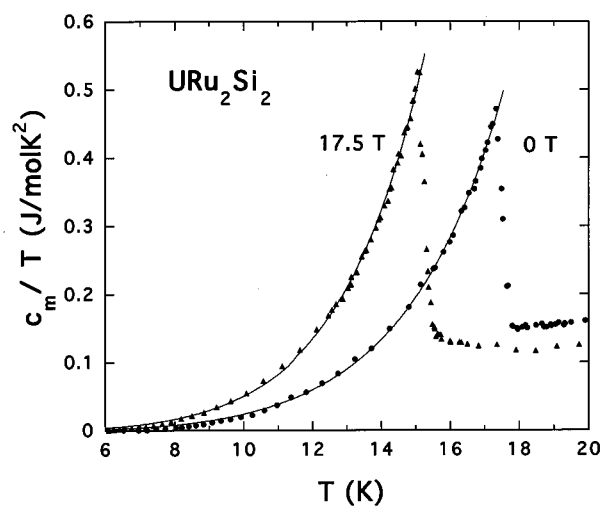

FIG. 4. Magnetic specific heat c_m/T of URu_2Si_2 in zero field and in a magnetic field of 17.5 T. The solid curves correspond to a fit of magnetic excitations in the antiferromagnetically ordered state (see text).

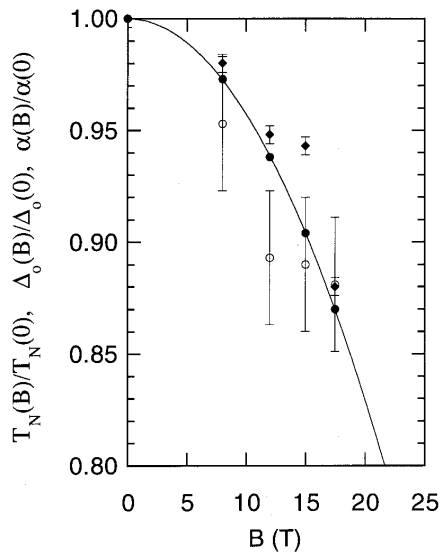

FIG. 5. Relative field dependence of the transition temperature T_N (solid circle), the magnetic excitation gap Δ_0 (open circle), and the dispersion α (solid diamond). The curve represents a scaling function of the form $[1-(B/B_0)^2]$ with a scaling field of $B_0 = 48.5(1)$ T.

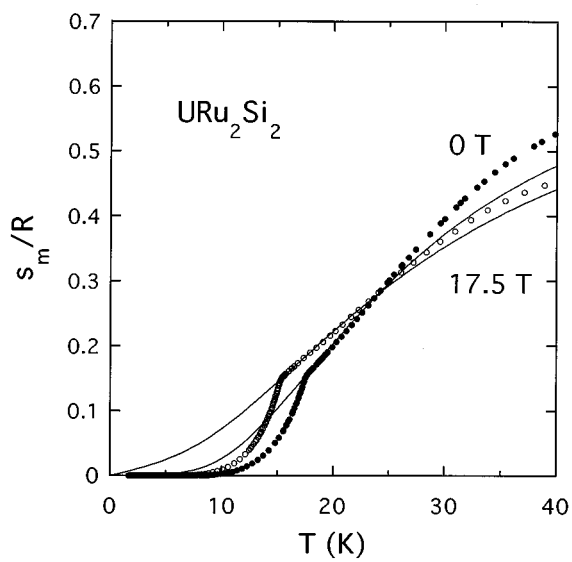

FIG. 6. Magnetic entropy s_m/R of URu_2Si_2 as a function of temperature in zero field and in a magnetic field of 17.5 T. The pronounced kink signals the antiferromagnetic transition. The solid lines correspond to a fit of the broadened crystal-field excitations in the paramagnetic state (see text).

scaling behavior as T_N as a function of magnetic field. This is in good agreement with far-infrared spectroscopy measurements[34] which show a small reduction of the energy gap in magnetic fields up to 20 T. Additionally, we find that the dispersion weakens in high magnetic fields and follows the same scaling behavior as T_N, indicating that the magnetic excitations can be described with a single energy scale. From Eq. (7) it is easy to see that if T_N, Δ_0, and α have the same scaling behavior with magnetic field, the magnetic specific heat at T_N is independent of the magnetic field. This is consistent with the measured values of $\Delta c(T_N)$ as a function of magnetic field (Table I). The derived values of the dispersion are slightly below the values of the magnetic excitation spectrum measured with inelastic neutron-scattering measurements. This is probably caused by the additional contribution of the second minimum at $\mathbf{Q}=(1.4\ 0\ 0)$, which has not been taken into account in the model.

After integration of c_m/T, we obtain the magnetic entropy s_m (Fig. 6). In an applied magnetic field T_N is slowly suppressed, while the entropy related to the magnetic order $s_m(T_N)$ is found to be field independent, as indicated in Table I. In Fig. 6 the field dependence of the magnetic entropy is shown. Above T_N a strong field dependence is observed with a crossing of the s_m/R curves in zero field and in a magnetic field of 17.5 T. The magnetic entropy in the paramagnetic state ($T>T_N$) does not correspond to a simple Schottky curve for two dispersionless crystal-field levels. Inelastic neutron-scattering measurements[5,23,35] confirm the absence of well-defined crystal-field levels. For $T>T_N$ the magnetic excitations at $\mathbf{Q}=(1\ 0\ 0)$ become overdamped, while the magnetic excitations at $\mathbf{Q}=(1.4\ 0\ 0)$ persist around 4.5 meV (52 K). The strong field dependence of the magnetic entropy as function of temperature cannot be explained by a simple shift of the level splitting, but requires a substantial broadening in applied magnetic field in order to explain the crossing of the s_m/R curves in Fig. 6.

An estimate of the level broadening can be obtained using a generalized crystal-field model with a Lorentzian broadening of the level splitting. The corresponding magnetic entropy can be described as

$$s_m(T)=R\ \ln Z+RTd\ \ln Z/dT,$$

with

$$Z=1+\left\{\int_0^\infty dE\ L(E)\exp(-E/kT)\right\}\Big/\left\{\int_0^\infty dE\ L(E)\right\}, \tag{8}$$

where $L(E)=(2/\pi W)[1+4(E-E_0)^2/W^2]^{-1}$ is a Lorentzian function with a peak position E_0 and a full width at half maximum W. We have assumed that the ground state and the excited state are singlets which leads to a total magnetic entropy of $s_m(T\rightarrow\infty)=R\ \ln 2$. In Fig. 6 the fitted curves of the magnetic entropy in the paramagnetic state are shown. It is difficult to obtain an accurate description of the crystal-field contribution at high temperatures because the specific heat becomes increasingly dominated by the phonon contribution, which has been estimated from the specific-heat measurements on $ThRu_2Si_2$.[24] The fitted parameters for the size and width of the broadened level splitting are listed in Table I. In applied magnetic fields the energy E_0 of the level splitting is nearly field independent and shows a value that roughly corresponds to the minimum in the excitation spectrum at $\mathbf{Q}=(1.4\ 0\ 0)$. The broadening of the level splitting W shows a substantial increase in high magnetic fields.

A possible explanation for the broadening of the crystal-field levels in field may be related to a change in the admixture of quantum states that form the two singlet states in applied magnetic fields. For the proposed singlet-singlet model,[13] the ground state corresponds to $|\Gamma_{t1}(1)\rangle=\gamma|0\rangle+\varepsilon(|-4\rangle+|4\rangle)$ and the first excited state to $|\Gamma_{t2}\rangle=(1/\sqrt{2})(|-4\rangle-|4\rangle)$. A change in ε/γ may cause a broadening of the level splitting in high magnetic fields.

IV. CONCLUSIONS

Specific-heat measurements of the heavy-fermion superconductor URu_2Si_2 show an antiferromagnetic transition ($T_N = 17.5$ K) followed by a superconducting transition ($T_c = 1.2$ K). The antiferromagnetic transition is signaled by a sharp peak, indicating the formation of a magnetic excitation gap. The specific heat in the paramagnetic state ($T > T_N$) can qualitatively be described by a broadening of the crystal-field excitations. This broadening strongly increases in applied magnetic fields up to 17.5 T. In order to describe the specific heat below T_N, we have used the characteristic features of the excitation spectrum measured with neutrons. The magnetic energy gap is reduced while the dispersion weakens for magnetic fields up to 17.5 T. The transition temperature T_N, the magnetic energy gap Δ, and the dispersion α show the same scaling behavior with a scaling field of 48.5 T. This scaling field is close to the metamagnetic transition where the heavy-fermion state is suppressed. The presence of a large jump in the specific heat at the antiferromagnetic transition seems hard to reconcile with the small dipolar-ordered moment. This suggests that magnetic order parameter in URu_2Si_2 may involve both dipolar and quadrupolar components, where the well-defined magnetic excitations are related to the quadrupolar component.

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