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Low-Temperature Heat Capacity of LaPt$_2$ and CePt$_2$ and the Magnetic Susceptibility of CePt$_2$[†]

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The heat capacity of LaPt$_2$ and CePt$_2$ has been measured from 1.3 to 9 °K and the magnetic susceptibility of CePt$_2$ from 1.3 to 80 °K. LaPt$_2$ behaves as a normal metal with $\gamma = 1.17$ mJ/g atom °K^2 and $\Theta_D = 236$ °K. CePt$_2$ orders antiferromagnetically at (1.6 ± 0.1) °K. The magnetic-susceptibility behavior and the entropy of ordering can be explained on the basis of a crystal-field splitting of the $J = \frac{5}{2}$ multiplet into a ground-state doublet and an excited quartet lying 216 °K above the doublet.

INTRODUCTION

The Laves-phase compounds comprise one of the largest groups of intermetallic compounds known. This is primarily due to the formation of these compounds by the rare-earth group elements with 13 other metals. Rare-earth Laves-phase compounds, RM_2, where R is a rare-earth metal, are formed with Mg, Al, Mn, Fe, Co, Ni, Tc, Ru, Rh, Re, Os, Ir, and Pt. In an effort to better understand these materials, we have begun a study of some of their physical properties, since about the only information known is their crystal structures and in some instances their magnetic properties.

The cerium Laves phases are of particular interest since the valence of cerium is known to vary from three to four in these compounds.[1] CePt$_2$ was chosen since cerium is trivalent in this compound.[1-4] Magnetic studies[2-4] from 35 to 470 °K have confirmed this, and furthermore they indicate that CePt$_2$ might order antiferromagnetically near 0 °K [the Curie-Weiss parameters are $p_{\text{eff}} = 2.33\,\mu_B$ and $\theta = 4$ °K (Ref. 2); $p_{\text{eff}} = (2.57 \pm 0.07)\,\mu_B$ and $\theta = (-26 \pm 5)$ °K (Ref. 3); $p_{\text{eff}} = 2.50\,\mu_B$ and θ was not given (Ref. 4), but a Curie temperature of 7 °K was reported].

The LaPt$_2$ Laves phase was chosen to be studied because the outer electronic configurations of LaPt$_2$ and CePt$_2$ are expected to be nearly the same. And it would be useful to compare the low-temperature heat capacities of these two compounds, especially if the $4f$ electron of the cerium atom causes some unusual low-temperature behavior.

A little information is available on the La-Pt and Ce-Pt phase diagrams. In addition to the Laves-phase compounds the RPt[5] and RPt$_5$[6] compounds (where R is La or Ce) are also known to exist. There is some doubt[7] whether RPt$_3$ exists or is the terminus of a solid-solution region extending from RPt$_2$.[8,9] No information exists on the melting modes and melting temperatures of these compounds.

EXPERIMENTAL

Sample Preparation

Chemical analysis of the component metals used to prepare the LaPt$_2$ and CePt$_2$ compounds is listed in Table I. Weighed amounts of the two components corresponding to the RPt$_2$ stoichiometry were arc melted. The arc-melted samples of CePt$_2$ or LaPt$_2$ were not single phase, thus indicating that these compounds form peritectically. Homogenization of these compounds was accomplished by placing them in a water-cooled trough[10] inside a continuously evacuated quartz tube and inductively heating them to (1350 ± 50) °C for 15 min. They were then cooled at the rate of 10 deg/min until a temperature of 700 °C was reached. At this temperature the power was shut off and the sample cooled rapidly to room temperature. Metallographic analysis showed that

TABLE I. Chemical analysis of the starting materials, La, Ce, and Pt (all impurities listed as ppm by weight).

	La	Ce	Pt
H	2	2	21
N	350	33	145
O	395	665	103
Mg		<55	VFT[a]
Al			VFT
Si	<15	<30	FT[b]
Ca	<15	<90	VFT
Cr		<80	VFT
Fe	105	<20	...[c]
Ni	<10		VFT
Cu		<20	VFT
Zr		<200	
Ru			T(*X*)[d,e]
Rh			T
Pd			FT
Ag			VFT
La		<200	
Ce	<300		
Pr	<600	<200	
Nd	<200	<200	
Ta	660	<500	

[a] VFT—very faint trace.

[b] FT—faint trace.

[c] None found.

[d] T—trace.

[e] (*X*)—interference.

there was no second phase present. Spectrographic analysis showed that no silver was present, indicating no crucible contamination.

The lattice parameters of the actual $LaPt_2$ and $CePt_2$ alloy specimens which were used for the heat-capacity and magnetic-susceptibility measurements were 7.7762 ± 0.0001 and 7.7354 ± 0.0001 Å, respectively. Since $LaPt_2$ and $CePt_2$ are reported to exist over a large solid-solubility region (65.5 to 75.7 at. % Pt for Ce-Pt alloys) and since there is a discrepancy between the Néel temperature found in our study and that reported in another investigation, the samples upon which the experiments were carried out were chemically analyzed for their lanthanide and platinum contents. For $LaPt_2$ the chemical analyses show 26.56 ± 0.16 wt. % La and 73.78 ± 0.16 wt. % Pt, indicating a composition of 66.42 ± 0.19 at. % Pt (i.e., $LaPt_{1.978 \pm 0.006}$), and for $CePt_2$ 26.99 ± 0.12 wt. % Ce and 73.10 ± 0.13 wt. % Pt, indicating a composition of 66.05 ± 0.14 at. % Pt (i.e., $CePt_{1.946 \pm 0.004}$). The chemical analysis and the x-ray results, when used in conjunction with the lattice parameter vs composition data of Harris,[9] are in excellent agreement with each other and show that both of our samples contain an amount of rare earth which is slightly in excess of the theoretical 1:2 stoichiometry.

The $CePt_2$ and $LaPt_2$ samples were reactive in air. The samples would crumble into powder if left unprotected. A metallographic sample in the polished condition left exposed to the air would show many transgranular cracks after a few days. The samples were protected by sealing them in helium-filled vials. When sealed in the above manner, the sample had no tendency to crumble and a polished surface remained bright.

Heat-Capacity Measurements

Heat-capacity measurements[11] were made in an adiabatic heat-pulse type of calorimeter, which was designed to fit between the pole pieces of a 9-kOe magnet. A mechanical heat switch was used to isolate the sample from the helium bath. The temperature was measured with a Honeywell type-II germanium resistance thermometer (GRT). The GRT was calibrated in the range 1.4–4.2 °K against the vapor pressure of He^4 and in the range 4.2–22 °K against a second GRT which had been calibrated by gas-bulb thermometry.[12] The heat capacity of the addenda was determined in separate runs and subtracted from total heat capacity to give the heat capacity of the sample.

To check out the operation of the calorimeter and the validity of the thermometry, a Calorimetry Conference Copper Standard[13] was obtained. The heat capacity of a 1.1-mole piece of the copper standard was measured from 1.4 to 8 °K and is shown in Fig. 1. All but two of the points fall within 2% of the reference equation[13] with no systematic deviations. Furthermore, our results also agree quite well with critically evaluated low-temperature heat-capacity data for copper.[14] Thus our results for copper give additional confidence to the two thermometer calibrations.

Measurements of heat capacity in a magnetic field were made using an electromagnet with 10-cm-diam pole tips and a 5-cm pole gap. It was found that the change of resistance of the GRT above 4.2 °K was negligible ($< 5\ \Omega$) in fields of 9 kOe. Below 4.2 °K the change in resistance varied from 1.3% of R at 3.3 °K to 2.3% at 1.6 °K. The change in resistance of the GRT as a function of field was fitted to an equation and incorporated into the computer program to correct for the effect of field.

All heat-capacity data in zero field were taken in at least two separate runs with the sample warmed to room temperature between runs. No systematic deviation of the heat capacity from one run to the next was found. Since the heater current and Δt are known to 0.1% or better, it is felt that most of the errors associated with the measurement of the heat capacity are random in nature and associated with determining ΔT by extrapolation to the center of the heat pulse. The scatter in the data varied from 5% for the addenda and $LaPt_2$ to 2% for the copper standard. The heat capacity of the addenda amounted to about 20% of the total heat capacity of $CePt_2$ and 60% of $LaPt_2$. Systematic errors may exist from the thermometry but their magnitudes

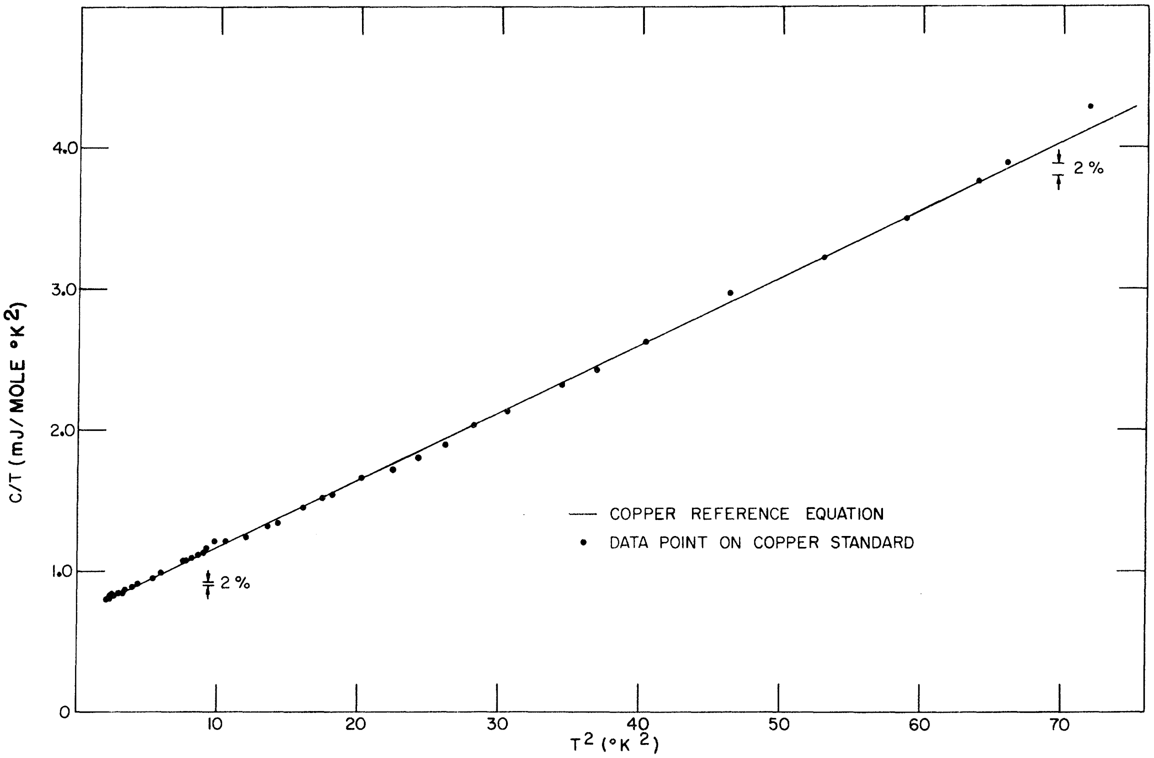

FIG. 1. Heat capacity of Calorimetry Conference Copper Standard.

are difficult to assess.

Magnetic-Susceptibility Measurements

The magnetic susceptibility of $CePt_2$ was measured by using a modified Foner-type vibrating-sample magnetometer for high-field measurements, 1–30 kOe.[15] The measurements were made by varying the field from 1 to 30 kOe while maintaining the sample at a constant temperature (within ±0.1 °K) over the entire temperature range. Three independent measurements were made on the susceptibility of $CePt_2$ over a 3-year period. The first set of data was taken on the apparatus described by Miller[15] and is designated Foner II. The second and third sets of data were taken on a modified version of this apparatus and are designated Foner III. The basic difference between these two apparatuses is that the detector system as described by Miller[15] was replaced by two ratio transformers and a dual-phase lock-in detector,[16] which enabled us to measure the pure-inductive component.

The sample temperature in both cases is measured with Au–Fe and Au–Co vs copper thermocouples. Measurements were made over the temperature range 1.3–80 °K.

RESULTS

Heat Capacity of $LaPt_2$

For the compound $LaPt_2$ the heat-capacity results (Fig. 2 and Table II) are straightforward in that this compound acts like a normal metal. The specific heat, when plotted as C/T vs T^2 is a straight line at temperatures below 4.2 °K with $\gamma = 1.17 \pm 0.02$ mJ/g atom $°K^2$ and $\beta = 0.149 \pm 0.002$ mJ/g atom $°K^4$ [$\Theta_D = (236 \pm 1)$ °K].[17] The scatter in this data is somewhat larger than that present in the data for Cu and $CePt_2$. Presumably this is due to the difficulty in obtaining rapid thermal equilibrium with this compound. A higher density of points was taken on this compound to counteract the larger amount of scatter.

Magnetic Susceptibility of $CePt_2$

The magnetic susceptibility of $CePt_2$ has been determined from 1.3 to 80 °K (see Fig. 3). Two independent runs were made over the entire temperature range, and a third run from 1.3 to 3.0 °K. The poorest agreement between runs is ~ 8.5% in the 15–35 °K temperature range; but over the rest of the temperature span the agreement is a few percent.

The unusual curvature of the susceptibility between 10 and 60 °K is due to the crystal-field splitting of the $J = \frac{5}{2}$ level of cerium into a doublet (lowest state) and a quartet. The temperature dependence of the magnetic susceptibility χ for this case has been derived by White *et al.*[18] and Jones,[19] as

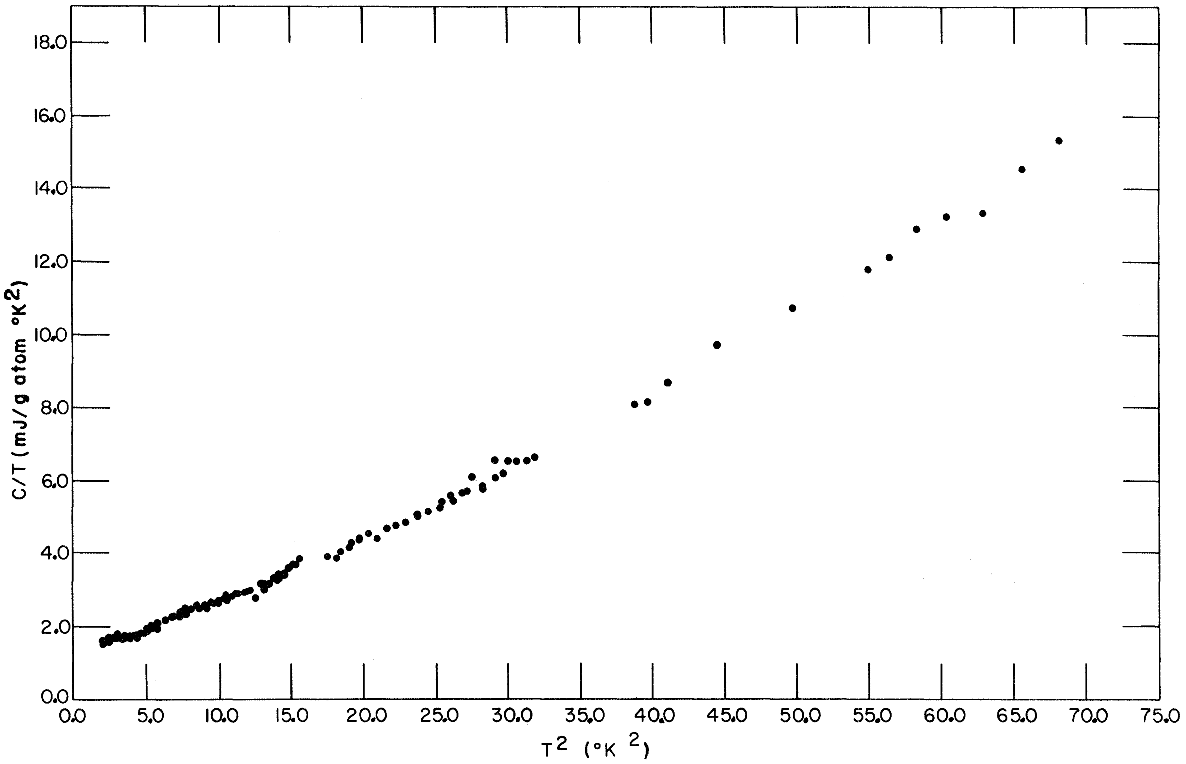

FIG. 2. Heat capacity of $LaPt_2$.

is given by

$$1/\chi = T/Cf(T) - \theta/C, \qquad (1)$$

where C is the Curie constant, θ the Weiss constant, and $f(T)$ is given by

$$f(T) = \frac{0.238e^{-x} + 1.542(1 - e^{-x})/x(1 - e^{-x})/x}{1 + 2e^{-x}}, \qquad (2)$$

where $x = \Delta/kT$ and Δ is the energy separation between the doublet and quartet. The best fit of the experimental data was obtained for $\Delta = 150$ cm^{-1} $(\Delta/k) = 216$ °K), $\theta = 5$ °K, and $C = 0.6775$ ($p_{\text{eff}} = 2.33$ μ_B). This is shown as the solid line drawn through the data points in the upper left-hand portion of Fig. 3. Reasonable fits to the data (within ±10%) could be obtained as long as the parameters were kept between the limits of 140 and 160 cm^{-1} for Δ (201 to 230 °K for Δ/k), − 5 to + 10 °K for θ, and 0.632 to 0.749 for C (2.25 to 2.45μ_B for p_{eff}). However, as one deviates from the best-fit parameters the curvature of the experimental data becomes more difficult to match.

The upper end of our data connects reasonably well with the lower data points of Olcese.[2] Our data are also in fair to good agreement with the results of van Daal and Buschow.[4] The effective magnetic moment derived from our data ($p_{\text{eff}} = 2.33$ μ_B) is the same as Olcese's value derived from data obtained over the temperature range 100–470 °K, but is only in fair agreement with the values of 2.57 and 2.50μ_B reported by Vijayaraghavan *et al.*[3] and van Daal and Buschow,[4] respectively. The theoretical value for the effective magnetic moment of Ce^{+3} is 2.54μ_B. The conduction-electron contribution to the susceptibility is thought to account for the fact that our value for the moment is ~10% smaller than the theoretical value. The paramagnetic Curie temperature (Weiss parameter) varies widely from + 5 °K for our results to − 4 °K for Olcese and − 26 °K for Vijayaraghavan *et al.* The actual Néel temperature deduced from susceptibility data is (1.5 ± 0.1) °K, and is (1.7 ± 0.1) °K as deduced from heat-capacity data (to be discussed next). A Curie temperature of 7 °K was reported by van Daal and Buschow,[4] but because of the lack of details in their paper it is difficult to determine how this value was obtained. Presumably, their conclusions are based on the drop in resistivity at 7 °K and the fact that the susceptibility deviates from a Curie-Weiss behavior in a manner which suggests ferromagnetic ordering (if the crystal-field effect is not considered to give this curvature). The increase in the susceptibility below 1.5 °K and the field dependence of the susceptibility clearly indicate that $CePt_2$ orders antiferromagnetically. Furthermore, the curvature of the susceptibility is only explainable by crystal-field effects, since the entropy as-

TABLE II. Heat capacity of $LaPt_2$.

Temp. (°K)	C (mJ/g atom °K)	Temp. (°K)	C (mJ/g atom °K)	Temp. (°K)	C (mJ/g atom °K)
1.425	2.16	2.870	6.98	4.522	20.3
1.437	2.26	2.931	7.42	4.589	20.0
1.466	2.26	2.964	7.42	4.656	21.6
1.479	2.30	3.035	7.74	4.720	22.2
1.512	2.38	3.047	7.49	4.803	23.0
1.528	2.44	3.094	8.10	4.879	24.5
1.561	2.62	3.124	8.23	4.884	24.2
1.574	2.55	3.134	8.30	4.956	25.3
1.624	2.71	3.180	8.54	4.959	25.3
1.680	2.79	3.180	8.31	5.040	26.2
1.684	2.84	3.230	8.84	5.049	27.0
1.733	2.87	3.260	9.26	5.114	28.3
1.739	3.06	3.277	8.75	5.129	27.7
1.787	3.00	3.327	9.30	5.186	29.0
1.846	3.14	3.360	9.65	5.222	29.6
1.848	3.04	3.388	9.79	5.255	31.7
1.881	3.26	3.452	10.04	5.322	30.8
1.903	3.17	3.488	10.26	5.325	30.5
1.972	3.29	3.505	10.38	5.403	32.5
1.983	3.40	3.560	9.78	5.405	35.1
2.064	3.59	3.616	11.2	5.455	33.4
2.114	3.49	3.640	10.8	5.486	35.5
2.163	3.80	3.690	11.5	5.539	35.8
2.172	3.87	3.730	12.1	5.603	36.3
2.216	4.03	3.738	12.0	5.653	37.8
2.235	4.11	3.768	12.2	5.653	37.1
2.266	4.22	3.778	12.7	6.220	50.4
2.300	4.35	3.825	12.9	6.299	51.0
2.329	4.59	3.826	13.0	6.410	55.3
2.336	4.47	3.866	13.7	6.671	64.5
2.415	4.64	3.908	14.2	7.052	75.1
2.431	5.04	3.928	14.3	7.412	86.9
2.526	5.40	3.961	15.0	7.514	90.7
2.626	5.95	4.194	16.2	7.636	97.8
2.646	6.05	4.274	16.3	7.767	102.3
2.660	5.96	4.300	17.1	7.930	105
2.722	6.41	4.370	18.0	8.095	117
2.734	6.10	4.388	18.6	8.256	126
2.789	6.87	4.444	19.1		
2.801	6.47	4.455	19.6		

sociated with the magnetic ordering at 1.7 °K is ~$R\ln 2$ (to be discussed shortly) and not $R\ln 6$, if no crystal-field splitting occurred.

The difference between the ordering temperature found here (1.5 or 1.7 °K) and that found by van Daal and Buschow (7 °K) is not understood, especially since they gave no information characterizing their sample with respect to impurities and the presence of second phase. In addition to these two possibilities, it may be that $CePt_2$ exists over a solid-solubility range,[8,9] and the transition temperatures (ours and theirs) may correspond to different compositions in the solid-solution region. As noted earlier, our sample contains a slight excess of Ce.

The low-temperature data points from 1.5 to 10 °K lie very close to a straight line (see lower right-hand curve, Fig. 3). A least-squares fit of the data yielded a slope of 3.00 ± 0.04, an intercept of $1/\chi = 2.21 \pm 0.25$, and a value of $T = (-0.74 \pm 0.10)$ °K when $1/\chi = 0$. It is easily shown that the low-temperature limit of Eq. (1) becomes

$$\lim_{T \to 0} \frac{1}{\chi} \to \frac{T}{0.2381C} - \frac{\theta}{C}, \qquad (3)$$

but a quick calculation shows that even at 5 °K for

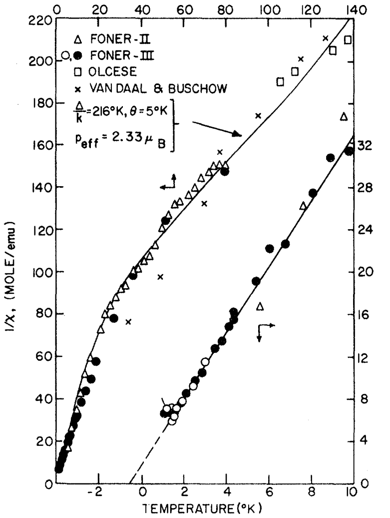

FIG. 3. Magnetic susceptibility of $CePt_2$.

$\Delta = 150$ cm^{-1}, the value of $f(T)$ is still about 20% larger than the limiting value of 0.2381. A splitting of two or three times larger would be required for $f(T)$ to be within a few percent of 0.2381 at 5 °K. Equation (3) thus cannot be used to evaluate these low-temperature parameters. The most reasonable explanation for this is that, as the Néel temperature is approached, the Curie-Weiss law no longer holds for $CePt_2$, as has been noted for most materials which order magnetically.

Recently Wang and Cooper[20] evaluated the behavior of the magnetic susceptibility of antiferromagnetically ordering cerium compounds below their Néel temperatures. Because the Néel temperature of our $CePt_2$ sample lies just above the lower temperature limit of our susceptibility apparatus (1.2 °K), it is not possible to see if $CePt_2$ follows their model. Presumably, because the crystal field (216 °K) is large compared to the exchange field (1.5 °K), $CePt_2$ would behave analogously to CeP below the ordering temperature.

Heat Capacity of $CePt_2$

The specific heat of an 11-g sample of $CePt_2$ was determined from 1.4 to 9 °K at zero field and from 1.4 to 4 °K at 9.0 kOe. The results are shown in Fig. 4 and tabulated in Table III. The peak at 1.7 °K is due to antiferromagnetic ordering of the Ce atoms in $CePt_2$. Because of the large heat-capacity contribution from the antiferromagnetic ordering in $CePt_2$, it is impossible to evaluate the usual lattice and electronic contributions. Presumably they are essentially the same as those of the $LaPt_2$ compound.

The effect of an applied field of 9.0 kOe is to lower the transformation temperature about 0.1 °K, which is what might be expected for an antiferro-

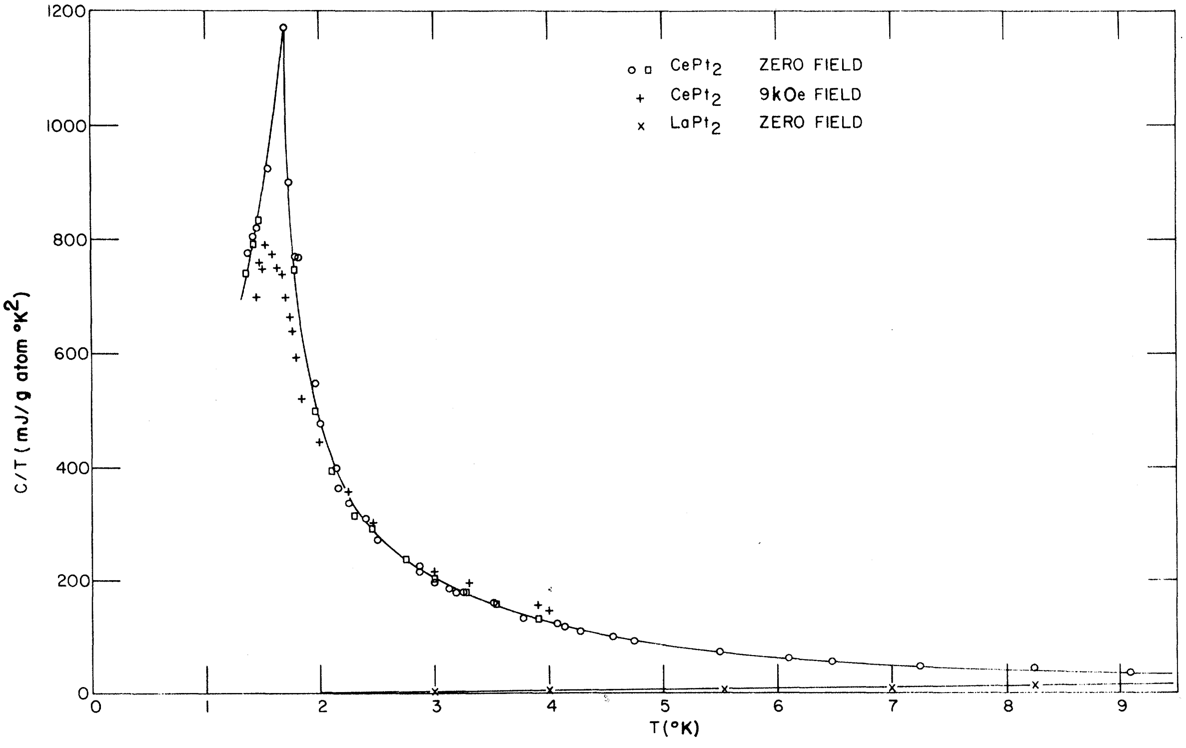

FIG. 4. Heat capacity of $CePt_2$. For comparison the heat capacity of $LaPt_2$ is also shown.

TABLE III. Heat Capacity of $CePt_2$.

Temp. (°K)	C (mJ/g atom °K)	Temp. (°K)	C (mJ/g atom °K)	Temp. (°K)	C (mJ/g atom °K)
		Zero-field data			
1.361	1006	2.163	794	3.774	499
1.373	1066	2.258	754	4.077	509
1.397	1110	2.316	730	4.137	488
1.415	1147	2.360	735	4.285	468
1.437	1134	2.447	715	4.564	452
1.456	1192	2.504	674	4.764	445
1.470	1226	2.767	640	5.095	432
1.565	1485	2.869	613	5.484	407
1.672	1962	3.013	616	6.084	409
1.718	1548	3.045	598	6.223	382
1.780	1332	3.124	583	6.478	370
1.797	1388	3.192	612	7.265	344
1.821	1398	3.211	580	7.417	347
1.951	974	3.246	578	7.691	342
1.958	1073	3.262	580	8.286	338
1.989	940	3.271	583	9.100	334
2.116	833	3.533	555		
		At 9.0 kOe			
1.410	1055	1.631	1250	2.222	821
1.432	1067	1.642	1248	2.276	797
1.454	1117	1.655	1264	2.335	772
1.462	1108	1.666	1232	2.405	763
1.476	1135	1.671	1224	2.481	747
1.478	1133	1.687	1248	2.873	700
1.498	1159	1.700	1223	2.969	688
1.521	1205	1.721	1235	3.075	671
1.551	1213	1.737	1164	3.176	651
1.575	1217	1.766	1138	3.270	635
1.584	1226	1.791	1072	3.790	597
1.601	1239	1.817	1032	3.878	558
1.613	1236	1.842	973	3.967	589
1.628	1234	1.993	890		

magnetic material.

Assuming the heat-capacity curve of $LaPt_2$ represents the electronic and lattice contributions to the heat capacity of $CePt_2$, the entropy due to the ordering of $CePt_2$ was determined by extrapolating the C/T-vs-T curve to zero degrees. Then the lattice and electronic contributions were subtracted off and the area under the curve determined. The entropy was found to be $S = R\ln 1.75$. This indicates that the $J = \frac{5}{2}$ level ($S = R\ln 6$) is split by the crystal-field environment around the Ce atoms into a ground-state doublet ($S = R\ln 2$) and an excited state quartet ($S = R\ln 4$). These results are consistent with the magnetic-susceptibility data.

DISCUSSION

$LaPt_2$ has been reported to be a superconductor,[8] but the transition temperature of 0.46 °K is lower than can be measured with our calorimeter. The low value of γ (1.17 mJ/g atom °K^2) for this compound, when compared to those of the pure elements ($\gamma_{La} = 10.1$ mJ/g atom °K^2 and $\gamma_{Pt} = 6.68$ mJ/g atom °K^2),[21] is not too surprising, since the band structure of this cubic Laves phase is expected to be considerably different from that of the pure metals.[22] This γ value is quite low compared to the γ values found for other rare-earth Laves phases (which range from 1.81 mJ/g atom °K^2 for YAl_2[23] to 13.6 mJ/g atom °K^2 for $CeRu_2$[24]), but is comparable to that of $MgCu_2$ (1.03 ± 0.01 mJ/g atom °K^2)[22,25] and $MgZn_2$ (0.85 mJ/g atom °K^2).[25]

The Debye temperature of $LaPt_2$ (236 °K) is low when compared to those of other Laves-phase compounds.[22–26] The values for $CeNi_2$ (227),[26] $LaRu_2$ (158),[24,26] $CeRu_2$ (147),[24] and $CeRu_{2-x}Pt_x$ (143.5 for $x = 0.1$ and 144.5 for $x = 0.2$)[24] are smaller. The Debye temperatures for eight other Laves phases lie in the range 327–473 °K. As noted by

Joseph and Gschneidner,[26] the Debye temperature of the AB_2 Laves phase depends a great deal on the type of contacts, whether AA or BB, that are observed in the compound, and this in part may account for the wide variation observed.

Figure 4 shows the heat capacity for $LaPt_2$ plotted on the same scale as that of $CePt_2$. It is difficult to draw any conclusions about the γ value of $CePt_2$ from the comparison. But if the Θ_D value for $CePt_2$ were much lower than that of $LaPt_2$, the specific heat of $CePt_2$ would have started to rise at the high-temperature range of the heat capacity shown in Fig. 4.

Instead, even at 9 °K, the specific heat is still decreasing from the ordering peak. Thus it appears that Θ_D for $CePt_2$ is at least as large as that measured for $LaPt_2$. Also, the Θ_D values for pure cerium and lanthanum are about the same,[21] so that it is not expected that Θ_D for the two compounds would be very different. Based on the similarity of constituent atoms, outer electronic configurations of La and Ce, and lattice parameter and crystal structure, the γ value for $CePt_2$ would be expected to have about the same value as that of $LaPt_2$.

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