Antiferromagnetic ordering in heavy-fermion system Ce$_2$Au$_2$Cd

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Abstract

La$_2$Au$_2$Cd and Ce$_2$Au$_2$Cd were prepared from the elements by reactions in sealed tantalum tubes in a water-cooled sample chamber of an induction furnace. These intermetallics crystallize with the tetragonal Mo$_2$FeB$_2$ type, space group P4/mbm. While La$_2$Au$_2$Cd is Pauli paramagnetic, Ce$_2$Au$_2$Cd shows Curie-Weiss behaviour above 100 K with an experimental magnetic moment of 2.41(2) $\mu_B$/Ce atom, indicating trivalent cerium. Antiferromagnetic ordering is detected for Ce$_2$Au$_2$Cd at 5.01(2) K and magnetization measurements reveal a metamagnetic transition at 3 K at a critical field of around 20 kOe with a saturation moment of 1.50(2) $\mu_B$/Ce atom at 80 kOe. The low-temperature heat capacity properties characterize Ce$_2$Au$_2$Cd as a heavy fermion material with an electronic specific heat coefficient ($\gamma$) = 807(5) mJ/mol K$^2$ as compared to La$_2$Au$_2$Cd with $\gamma$ = 6(5) mJ/mol K$^2$.

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I. INTRODUCTION

The Ce$_2$T$_2$In indides and Ce$_2$T$_2$Sn stannides (T = late transition metal) with ordered U$_3$Si$_2$ type structure have intensively been investigated in recent years with respect to their greatly varying magnetic and electrical properties. and ref. therein Ce$_2$Ni$_2$In and Ce$_2$Rh$_2$In are intermediate-valence systems. Ce$_2$Pt$_2$In is a strongly temperature dependent paramagnet, and Ce$_2$Cu$_2$In, Ce$_2$Pd$_2$In, and Ce$_2$Au$_2$In order magnetically at 5.5, 4.3, and 3.2 K, respectively. Ce$_2$Pt$_2$In shows strong Kondo type interactions and a non-magnetic heavy fermion ground state with an electronic specific heat coefficient $\gamma$ = 500 mJ/mol K$^2$. Interesting behaviour was observed for the palladium compounds Ce$_2$Pd$_2$Sn and Ce$_2$Pd$_2$In which show small ranges of homogeneity Ce$_2$Pd$_{2-x}$Sn$_{1-x}$ and
Ce$_2$Pd$_{2+x}$In$_{1-x}$. The increase in palladium content leads to a decrease of the magnetic ordering temperature.$^5$ The cerium atoms in Ce$_2$Pt$_2$Sn are trivalent and this stannide shows two magnetic transitions at 2.5 and 6.5 K.$^7$

In parallel to the stannides, also some plumbides Ce$_2$T$_2$Pb ($T =$ Rh, Pd, Pt, Au) $^5$–$^10$ have been synthesized. Ce$_2$Pd$_2$Pb$^5$ is a 6.2 K antiferromagnet and Ce$_2$Rh$_2$Pb$^10$ behaves like a normal metal.

Recently, the first cadmium containing compounds have been reported,$^8$–$^{14}$ where the $X$ site is completely occupied by cadmium. This substitution leaves opportunities for varying the properties, since cadmium reduces the valence electron count per formula unit by two with respect to the stannides and plumbides. Two highly interesting compounds are Ce$_2$Ni$_{1.88}$Cd$^{11}$ and Ce$_2$Rh$_{1.86}$Cd$^{13}$ which both show intermediate valence of cerium. In view of these promising results we have started a more systematic study of the Ce$_2$T$_2$Cd intermetallics and their non-magnetic counterparts La$_2$T$_2$Cd. Herein we report on the magnetic properties and a specific heat study on La$_2$Au$_2$Cd and Ce$_2$Au$_2$Cd.

II. EXPERIMENTAL DETAILS
Starting materials for the synthesis of La$_2$Au$_2$Cd and Ce$_2$Au$_2$Cd were lanthanum and cerium ingots (Johnson Matthey), a gold bar (Heraeus), and a cadmium rod (Johnson Matthey, Ø 8 mm), all with stated purities better than 99.9%. Because of the low boiling point of cadmium (1083 K),$^{15}$ the samples were prepared in sealed high-melting tubes. Pieces of the rare earth elements, the gold bar, and the cadmium rod were weighed in the ideal 2:2:1 atomic ratio and arc-welded$^{16}$ in small tantalum tubes (ca. 1 cm$^3$ tube volume) under an argon pressure of ca. 600 mbar. The argon was purified over silica gel, molecular sieves, and titanium sponge (900 K). The sealed tantalum tubes were then placed in a water-cooled quartz sample chamber of a high-frequency furnace (Hüttinger Elektronik, Friburg, Type TIG 1.5/300) under flowing argon.$^{17}$ The elements were brought to reaction through inductive annealing at ca. 1500 K for about one minute and the products were subsequently annealed at ca. 900 K for another two hours. The products could easily be separated from the tantalum tubes via mechanical fragmentation. No reaction of the samples with the crucible material was detected. For further details concerning the sample preparation we refer to Ref. 12.
The purity of the samples was checked through Guiner powder patterns using CuK$_{α1}$ radiation and α-quartz ($a = 491.30$, $c = 540.46$ pm) as an internal standard. The magnetic and calorimetric measurements were performed on a Quantum Design PPMS with ACMS and specific heat options in the temperature range of 3 to 300 K, with magnetic flux densities up to 80 kOe. For magnetization measurements, the samples were enclosed in thin-walled gelatin capsules. The specific heat measurements were carried out with a relaxation technique. The samples were mounted on the sample holder with Apizeon N grease.

### III. RESULTS AND DISCUSSIONS

**Structure**

La$_2$Au$_2$Cd and Ce$_2$Au$_2$Cd crystallize with the tetragonal Mo$_2$FeB$_2$ type structure,\(^\text{18}\) space group $P4/mbm$. In Figure 1, we show the XRD patterns for both compounds along with the theoretically expected peak positions (shown by vertical ticks). Both compounds were obtained as X-ray pure materials. The cell constants were calculated by least-square refinements of the powder data. The correct indexing was ensured through intensity calculations.\(^\text{19}\) Cell constants and volumes as given in Fig. 1 agree with in experimental error with the values of $a = 809.2(1)$ pm, $c = 400.27(9)$ pm for La$_2$Au$_2$Cd and $a = 804.93(7)$ pm, $c = 393.36(6)$ pm for Ce$_2$Au$_2$Cd previously reported in Refs. [8, 12].

**Magnetic measurements**

Figure 2(a) shows the $dc$ magnetic susceptibility measured in a stable field of 10 kOe for Ce$_2$Au$_2$Cd. The samples were zero field cooled to the lowest temperature, measurement was performed while warming the sample up to room temperature. La$_2$Au$_2$Cd does not show any magnetic behavior and is paramagnetic to the lowest temperature measured. The room temperature susceptibility of $2.93(3) \times 10^{-4}$ emu/mol is consistent with Pauli paramagnetism.

The $\chi(T)$ curve of Ce$_2$Au$_2$Cd exhibits an definite peak at $T \sim 5K$ indicating antiferromagnetic ordering. The inverse susceptibility ($\chi^{-1}$) follows Curie-Weiss behavior above 100 K. However, the deviation of $\chi^{-1}$ below 100 K, can be attributed to a combination of crystal field effects and magnetic ordering. From the CW fit of the linear region in $\chi^{-1}$ in the temperature region $100 < T(K) < 300$, the paramagnetic Curie
temperature ($\theta_p$) and the effective Bohr magneton number ($\mu_{\text{eff}}$) for Ce$_2$Au$_2$Cd is -3.3(2) K and 2.41(2) $\mu_B$/mol-Ce respectively. The $\mu_{\text{eff}}$ obtained experimentally is in close agreement with value of the free Ce$^{3+}$ ion (2.54 $\mu_B$) indicating that unlike some of the Ce$_2$T$_2$Cd compounds (T = Ni, Rh), cerium is in trivalent state in Ce$_2$Au$_2$Cd. This is in excellent agreement with the course of the lattice parameters (lanthanoid contraction) for the series $RE_2Au_2Cd$, $RE$ = La, Ce, Pr, Nd and Sm, where Ce$_2$Au$_2$Cd shows no anomaly.

The negative sign of $\theta_p$ indicates the magnetic interactions are antiferromagnetic. There is no effect of H on the ordering temperature, while measuring $\chi(T)$. The insert in Fig 2a, shows $\chi(T)$ for Ce$_2$Au$_2$Cd measured in $H = 0.1$, 1 and 10 kOe applied fields. It is interesting to observe that though the susceptibility depends upon the excitation field but the ordering temperature is not affected by it. In the ZFC (symbols)-FC(continuous lines) $\chi(T)$ for $H = 0.1$ and 1 kOe, FC curve follows the ZFC curve and there is no bifurcation between them.

The real part of ac susceptibility ($\chi'$), shown in Figure 3, exhibits a prominent peak at $T_N$ with no frequency dependence. The imaginary part ($\chi''$) is essentially featureless with a broad feature seen only at higher frequencies, thus ruling out any spin-glass anomalies. There are no features in $\chi_{ac}$ measured for second and third harmonics (and hence not shown here), thus ruling out any ferromagnetic impurities also. These observations clearly establish long-range magnetic ordering of the antiferromagnetic type at 5 K in Ce$_2$Au$_2$Cd.

The magnetization as a function of applied magnetic field at different temperatures spanning $T_N$ are shown in Figure 4. The magnetization at temperatures 300 and 100 K ($>> T_N$) varies linearly with the application of field. However for $T = 10$ K, the magnetization increases linearly up to a field of 60 kOe and deviates slightly at higher fields. For $M(H)$ at $T = 4.5$ K (i.e., just below $T_N$), $M$ varies sluggishly with $H$ without saturating up to 80 kOe. The $M(H)$ at 3 K, ($T < T_N$), exhibits a metamagnetic transition starting around 20 kOe and increases with $H$ without saturating, up to the highest field measured. It may be recalled that an isostructural compound, Ce$_2$Au$_2$In also exhibits such a step-like increase in magnetization$^{2, 3}$ at $T < T_N$, but the metamagnetic transition already takes place around 5 kOe. However unlike Ce$_2$Au$_2$In, magnetization for Ce$_2$Au$_2$Cd at 3 K does not saturate. The moment value for Ce$_2$Au$_2$Cd
at 80 kOe and 3 K is 1.53(3) $\mu_B$/Ce atom, smaller than the maximum value of 2.14 $\mu_B$/Ce atom according to $g \times J$. An even smaller saturation magnetization of only 0.97 $\mu_B$/Ce atom was observed for Ce$_2$Au$_2$In at 1.7 K and 50 kOe.$^2$

**Specific heat studies**

Heat capacity, $C(T)$, for Ce$_2$Au$_2$Cd and its isostructural non-magnetic counterpart La$_2$Au$_2$Cd were measured by relaxation method using the PPMS heat capacity option in the temperature range 3–100 K on a puck calibrated for temperature and magnetic field. We have also measured the heat capacity for Ce$_2$Au$_2$Cd in presence of applied dc fields of 10, 20, 30 and 50 kOe.

Figure 5 (a and b) shows $C(T)$ and $CT^{-1}$ vs. $T$ respectively for both Ce$_2$Au$_2$Cd and La$_2$Au$_2$Cd. The heat capacity for the lanthanum compound varies with temperature and essentially shows contributions from the lattice. However, the cerium compound exhibits a sharp peak in $C(T)$ at 5 K undergoing magnetic transition. This is also a confirmation for the ordering temperature from magnetization measurements.

The magnetic part of heat capacity ($C_{\text{mag}}$) was deduced after subtracting the lattice part (i.e., the heat capacity of non-magnetic La$_2$Au$_2$Cd) from the total heat capacity of Ce$_2$Au$_2$Cd. The peak at 5 K can be clearly seen in $C_{\text{mag}}$ also. It is interesting to observe that from the bottom of the peak (~ 5.5 K) up to 29 K (which is equal to $\Theta_D$) there is hardly any change in $C_{\text{mag}}$ but beyond 29 K, $C_{\text{mag}}$ increases rapidly with increasing temperature.

The plot of $C_{\text{mag}}/T$ vs. $T^2$ is linear below $T_N$ and is shown in the insert of Figure 6a. The $T^3$ behavior of $C_{\text{mag}}$ in the ordered region is a typical feature of heavy fermions exhibiting antiferromagnetic magnetic ordering.$^{20, 21}$ It is interesting to observe that the value of the coefficient of the electronic specific heat ‘$\gamma$’ (Sommerfeld coefficient) obtained from the linear fit below $T_N$, is 807(5) mJ/mol K$^2$. The values of the coefficient of thermal expansion ($\beta$) and the Debye temperature ($\Theta_D$) are 80 mJ/mol K$^4$ and 29 K, respectively. For comparison, the isostructural counterpart of the title compound is Ce$_2$Au$_2$In. It exhibits antiferromagnetic ordering at around 3 K, but has a $\gamma$ of 37 mJ/mol K$^2$. Among other isostructural indides, the highest $\gamma = 500$ mJ/mol K$^2$ is observed for non-magnetic heavy fermion system Ce$_2$Pt$_2$In.$^4$ To the best of our knowledge, Ce$_2$Au$_2$Cd is the first compound among cadmium based 221 intermetallics exhibiting such a high value of $\gamma$ and hence be called a heavy fermion compound exhibiting
antiferromagnetic ordering. At this point it is worthwhile to note, that in the family of actinide (An) intermetallics $An_2T_2In$, $U_2Pt_2In$ shows an even larger $\gamma$ value of 850 mJ/mol K$^2$, while $U_2Pt_2Sn$ (334 mJ/mol K$^2$) and $U_2Pd_2In$ (393 mJ/mol K$^2$) have slightly smaller Sommerfeld coefficients. In the context of cerium intermetallics $Ce_2Au_2Cd$ can be discussed in line with the equiatomic compounds $CePdIn$ (700 mJ/mol K$^2$) and $CePtIn$ (> 500 mJ/mol K$^2$).

In Fig. 6 (b) we show the variation of magnetic entropy ($S_{mag}$) as a function of temperature. At the ordering temperature, $S_{mag}$ reaches the value of $\sim 7.93$ J/mol K, which is about 75% of the expected $R\ln 2$ value. The entropy reaches the 100% value of $R\ln 2$ at $T \sim 29$ K (which incidentally is equal to $\Theta_D$). Beyond $\Theta_D$, $S_{mag}$ increases linearly with $T$.

We have also studied the effect of the external magnetic field on $C(T)$. Figure 7 (a) and (b) shows $C$ and $C/T$ vs. $T$ for $Ce_2Au_2Cd$ measured in zero field and the applied dc fields of 10, 30 and 50 kOe. As expected for an antiferromagnet, the peak temperature ($T_p$) and magnitude of $C$, shifts to lower values with increasing field for $Ce_2Au_2Cd$. There is a small observable change in $T_p$ for $H = 10$ kOe. But with increasing the field to 30 kOe, $T_p$ shifts to lower temperature and exhibits two broad transitions. The magnitude of $C$ however is further lowered. The appearance of a secondary peak at lower temperatures with application of 30 kOe field may imply field induced changes in the magnetic structure. At 50 kOe the peak is completely smeared out. The $C_H(T)$ curves crosses each other just above the ordering temperature. A qualitative discussion about the crossing points in specific heat curves is given by Vollhardt et al. Though the observation of crossing of $C(T)$ curves for low values of $H$ were initially made in normal-liquid $^3$He, they can also be seen in heavy fermion systems with and without Fermi liquid behavior. Hence it will be interesting to correlate the high $\gamma$ value and crossing of $C_H(T)$ curves in the vicinity of second-order magnetic transition for $Ce_2Au_2Cd$ in the context of heavy fermion behavior.

Crossing of specific heat curves, as discussed above, can also be linked to the change in entropy with respect to the applied field. A close look at Fig. 7(b) shows that $C/T$ is linear below $T_p$. We have calculated the total entropy of $Ce_2Au_2Cd$ for measurements performed in $H = 0$, 10 and 30 kOe fields.
In figure 8 (a) we plot the variation of total entropy of the system ($S_{\text{tot}}$) with temperature and the effect of magnetic field on it. We have also plotted, for clarity, the change in total entropy ($\Delta S_{\text{tot}}$) with application of field, as a function of temperature in Fig. 8(b). The curve S1 shows $S_{10\text{kOe}}-S_0$, i.e., difference in entropy measured at 0 and 10 kOe. Similarly S2 shows the difference of entropy in 0 and 30 kOe. S1 changes sign of $\Delta S$ around $T_N$, and becomes negative. However S2 is quite interesting. It exhibits a peak like feature around 3.6(2)K and decreases up to $T_N$, then again rises at higher temperatures. Recalling the M(H) curve at 3K in this context, we have seen a metamagnetic transition around 30 kOe, which is clearly seen as a peak in the plot of $dM/dH$ vs. H. Such field induced behavior indicates towards possible modification of the Fermi surface, also supported by the enhancement of $\gamma$ value. We strongly feel that a detailed investigation of the (magneto) transport properties would be quite rewarding.

A phenomenological band-structure approach for understanding the magnetic ordering characteristics of ternary intermetallics containing Ce ($4f$) and U ($5f$) as magnetic ions has been given by Endstra et.al.\textsuperscript{29} In order to consistently understand the magnetic behavior of Ce$_2$Au$_2$Cd, one needs to carry out a systematic study of Ce$_2$T$_2$Cd series. More insight into the mechanism underlying the absence or presence of magnetic-ordering of these ternary intermetallics can be gained by applying proper experimental and theoretical approach. The band-structure approach proposed by Endstra et.al however explains the phenomenon within the Doniach phase diagram\textsuperscript{29} of the Kondo lattice. The magnetic properties of Ce$_2$Au$_2$Cd give indication of the Kondo effect. Since $\theta_p$ is comparable to $T_N$, the entropy reduction due to the Kondo screening competes with the entropy reduction due to antiferromagnetic transition of the hybridized states. However, contrary to the usual Doniach picture, it seems that the antiferromagnetic transition here is due to the formation of the spin-density waves (SDW), which destabilizes a part of the Fermi surface. Hence, in order to understand the structural, magnetic and physical properties of Ce$_2$Au$_2$Cd, an estimate of the $f$-$d$ hybridization strength by theory and detailed photoemission spectroscopy results will be quite rewarding.

To conclude, heavy fermion behavior was observed for the 5 K antiferromagnetic Ce$_2$Au$_2$Cd while La$_2$Au$_2$Cd is a simple Pauli paramagnet. The
Ce$_2$T$_2$Cd intermetallics are an interesting family of compounds with promising ground-state properties. Further investigations on these materials are currently in progress.

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FIG. 1 XRD patterns (Guinier technique, Cu K\textsubscript{α1} radiation) for La\textsubscript{2}Au\textsubscript{2}Cd and Ce\textsubscript{2}Au\textsubscript{2}Cd. The theoretically calculated peak positions are given as ticks in the lower part of each diagram.
FIG.2 (a) dc susceptibility ($\chi$) as a function of temperature (T) for Ce$_2$Au$_2$Cd, measured in applied field of 10 kOe. The insert shows $\chi$(T) measured in different applied fields of H = 0.1, 1 and 10 kOe. The ZFC-FC $\chi$(T) curves are distinguished by open circles (ZFC) and continuous line (FC). (b) $\chi^{-1}$ vs. T for Ce$_2$Au$_2$Cd is shown by open circles. The continuous line is the fit from high temperature linear region. The arrow near the origin indicates $\theta_p$ extrapolated from the CW fit.
FIG. 3 The real and imaginary part of the ac susceptibility (χ’ and χ’’) vs. T respectively measured at various frequencies (ν = 111, 197, 341, 607, 1057, 1847, 3247, 5697 and 9999 Hz) and ac amplitude of 1 Oe for Ce$_2$Au$_2$Cd.
**FIG. 4** Magnetization (M) vs. applied field (H) at temperatures spanning $T_N$ for Ce$_2$Au$_2$Cd. The line passing through the data points shows a straight-line behavior of the magnetization below about 15 kOe.
FIG. 5 (a) Heat capacity (C) vs. T and (b) C/T vs. T for $RE_2Au_2Cd$ ($RE = Ce$ and La) shown by data points (Ce) and continuous line (La). The ordering temperature for $Ce_2Au_2Cd$ is indicated by the vertical arrow.
FIG. 6 (a) The magnetic part of heat capacity ($C_{\text{mag}}$) for Ce$_2$Au$_2$Cd obtained by subtracting the lattice part ($C_{\text{Latt}}$) from the total heat capacity of Ce$_2$Au$_2$Cd. The ordering temperature and Debye temperature are indicated by vertical arrows. The insert shows $C_{\text{mag}}/T$ vs. $T^2$ for Ce$_2$Au$_2$Cd highlighting the $T^3$ behavior of $C_{\text{mag}}$ in the ordered state. The continuous line passing through the data points is a guide for the eyes only. (b) The magnetic entropy for Ce$_2$Au$_2$Cd. At the $T_N$, $S_{\text{mag}}$ reaches about 75% of $\ln 2$ only (dotted line).
FIG. 7 (a) Field dependence of heat capacity ($C_H$) and (b) $C_H/T$ vs. $T$, for Ce$_2$Au$_2$Cd. The crossing over point (see text) is indicated by vertical arrows.
FIG. 8 (a) Total entropy ($S_{tot}$) for Ce$_2$Au$_2$Cd under applied fields. (b) The change in total entropy with the change in applied field, where $S_1 = S_{10kOe} - S_0$ and $S_2 = S_{30kOe} - S_0$. 