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THERMODYNAMIC PROPERTIES OF THE TERNARY COMPOUNDS CeM_2Ge_2

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Results of the susceptibility, specific heat and thermal expansion are reported for the compounds CeM_2Ge_2 (M: Ag, Au, Ru), which order antiferromagnetically below $T_N = 5$ K (Ag) and 15 K (Au) and ferromagnetically below $T_c = 7.5$ K (Ru). Distinct maxima in the electronic specific heat coefficient $\gamma(T)$ at 0.3 K for the Ag- and Au-based compounds hint at a coexistence of coherent electronic quasiparticles of "medium"-heavy mass with magnetic order.

Among Ce-based Kondo-lattice compounds that crystallize in the $ThCr_2Si_2$ structure, two have attracted considerable interest: $CeCu_2Ge_2$ orders antiferromagnetically below $T_N = 4.1$ K, a temperature which is comparable to the single-ion Kondo temperature $T^* = 6$ K [1]. The local 4f-derived magnetic moments are found to be slightly (Kondo-) reduced in the ordered state [2]. $CeNi_2Ge_2$ behaves as an enhanced Pauli paramagnet whose electronic specific heat $C_{el} = \gamma T$ (as $T \to 0$) classifies this system as a heavy-fermion compound: $\gamma \simeq 300$ mJ/K^{2}mol [3]. Both compounds have in common a pronounced maximum near 0.3 K in the "Sommerfeld coefficient" $\gamma(T)$. Very similar anomalies have been discovered earlier for $CeAl_3$, and normal-state $CeCu_2Si_2$ and have been ascribed to the existence (at low T) of a coherent heavy-fermion band at the Fermi energy [4]. $CeCu_2Ge_2$ is the first antiferromagnet exhibiting this "coherence-derived" $\gamma(T)$ peak, with $\gamma(T \to 0) \simeq 100$ mJ/K^{2}mol [1].

In this paper we discuss the magnetic dc-susceptibility $\chi(T)$, specific heat $C(T)$ and thermal expansion $\alpha(T)$ of new examples of CeM_2Ge_2 compounds (M: Ag, Au and Ru) and compare these results with earlier ones for $CeCu_2Ge_2$ and $CeNi_2Ge_2$ notably. We are particularly interested in two properties of these materials, namely (i) magnetic phase transitions (transition temperature and kind of magnetic order) and (ii) "coherence" effects (maxima in $\gamma(T)$). Recent neutron scattering studies by Loidl et al. [5] pinned down the Kondo temperatures of the Ce-compounds of interest to $T^* = 3$ K (M = Ag) and to $T^* < 2$ K (M = Au and Ru), respectively. Some selected results for these three materials are shown in figs. 1a–c and 2a–c.

Magnetic phase transitions. Like for M = Cu, but in contrast to M = Ni, magnetic order determined by dc-susceptibility develops at low T for all three of the new systems. This confirms the neutron-scattering results of ref. [5]. Whereas the Ag- and Au-based compounds are antiferromagnets, the Ru-based compounds exhibits ferromagnetic order. Their high-T susceptibilities follow a Curie–Weiss dependence with $\mu_{eff} = 2.41\mu_B$(Ag), $2.37\mu_B$ (Au) and $2.49\mu_B$ (Ru).

For $CeAg_2Ge_2$ both specific-heat and thermal-expansion data [6] locate the transition at $\simeq 5$ K. The downward shift of the $C(T)$ anomaly by an external magnetic field and a T^3-dependence of the $B = 4$ T data (200 mK $\leq T \leq$ 500 mK) prove the antiferromagnetic nature of the ordered state (see fig. 1a). Both, a shoulder in $\chi(T)$ [8] and a small, but real jump in $\alpha(T)$ at 7.5 K [6] suggest that a second phase transition of unknown origin exists for this compound.

The thermal expansion of $CeAu_2Ge_2$ exhibits a distinct mean-field type anomaly at $T_N = 15$ K (fig. 1b). A peak in $\chi(T)$ at the same temperature suggests that this material is an antiferromagnet, too.

Both the $\chi(T)$ (fig. 1c) and $\alpha(T)$ results for $CeRu_2Ge_2$ point to the existence of two closely spaced phase transitions at $T_{c1} = 8.5$ K and $T_{c2} = 7.5$ K, respectively. Though their definite nature should be the subject of future investigations, our $C(T)$ results hint at $CeRu_2Ge_2$ being an (anisotropic) ferromagnet below T_{c2}. In contrast to

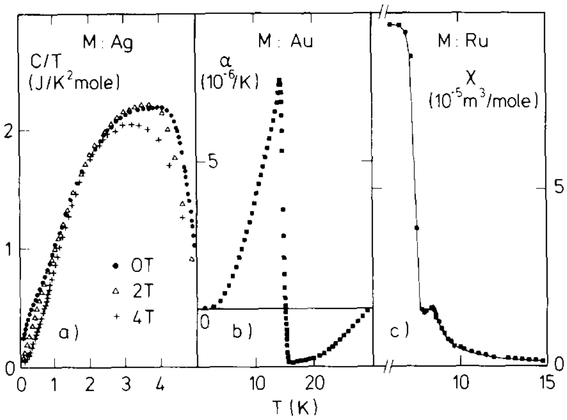

Fig. 1. Magnetic phase transitions of CeM_2Ge_2 monitored by the specific heat as C/T vs. T for M = Ag (a), thermal expansion $\alpha(T)$ for M = Au (b) and dc-susceptibility for M = Ru (c). Solid lines are guides to the eye.

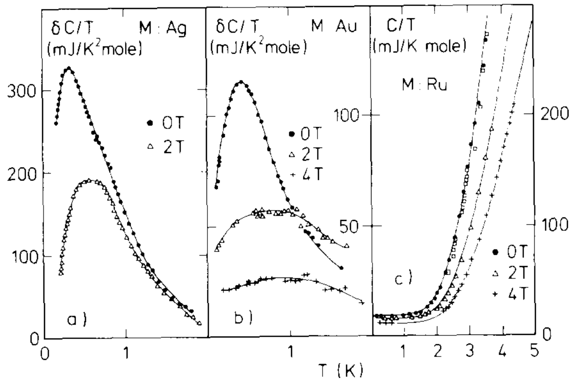

Fig. 2. Low-temperature specific heat of CeM_2Ce_2 as $\delta C/T$ vs. T for M = Ag, Au (a, b) and C/T vs. T for M = Ru (c). $\delta C = C - C_{\mathrm{nuclear}} - C_{\mathrm{magnon}}$, where C_{magnon} is obtained from an extrapolation of the measured $C(T)$ data [9] above $T = 3$ K. Square in (c) are taken from Felten [7]. Solid lines are fits of eq. (1) (see text) to the data (c) or guides to the eye (a, b).

$CeAg_2Ge_2$ (fig. 2a) and $CeAu_2Ge_2$ (fig. 2b) as discussed below, $CeRu_2Ge_2$ (fig. 2c) exhibits a low-T specific heat which is well described by

$$C = \gamma T + \beta T^{3/2} \exp(-\Delta/kT) \tag{1}$$

where $\gamma(\mathrm{mJ/K^2mol})$, $\beta(\mathrm{J/K^{5/2}mol})$ and Δk_B^{-1} (K) amount to 20, 2.6, 10.5 (for $B = 0$ T), 17, 1.5, 10.5 ($B = 2$ T) and 13, 1.2, 11.3 ($B = 4$ T), respectively. Apart from an enhanced electronic contribution γT the low-T specific heat is apparently due to a ferromagnetic magnon spectrum with an energy gap of order $k_B \times 10$ K.

Coherence effects in $CeAg_2Ge_2$ and $CeAu_2Ge_2$. In figs. 2a and 2b we have plotted the difference δC, shown as $\delta C/T$ vs. T, between the measured specific heat and both the nuclear and magnon contribution [9] for the Ag- and Au-compounds. $\delta C(T)$ is interpreted as the electronic specific heat. Like for $CeCu_2Ge_2$ [1] a peak is found in $\gamma(T) = \delta C(T)/T$ near $T = 350$ mK (Ag) and 500 mK (Au). We attribute this anomaly to *coherent* electronic quasi-particles of medium heavy mass that coexist with antiferromagnetic order. In the case of $CeAg_2Ge_2$, our interpretation is corroborated by the discovery of quasi-elastic magnetic neutron scattering below T_N [5]. For the Ag- and Au-based compound the "coherence structure" reacts much more strongly to a magnetic field, compared with earlier findings for $CeCu_2Ge_2$ [1], $CeAl_3$ and $CeCu_2Si_2$ [4], in that the $\gamma(T)$ peak is considerably depressed and shifted to higher temperatures. The absence of a similar T-dependence as well as the rather low value of $\gamma(T \to 0)$ for $CeRu_2Ge_2$ can be explained by the large ratio of T_{c1}/T^*. In other words, since Ce^{3+} moments become fixed at $T \geqq T_{c1} = 8.5\ \mathrm{K} \gg T^*$, the mass renormalization of the electronic quasiparticles due to the Kondo effect is not significant.

We would welcome renormalized band-structure calculations in magnetically ordered Kondo lattices of $ThCr_2Si_2$ structure in order to check our hypothesis [1] that coherent (medium) heavy quasiparticles can coexist with long range magnetic order.

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References

[1] F.R. de Boer, J.C.P. Klaasse, P.A. Veenhuizen, A. Böhm, C.D. Bredl, U. Gottwick, H.M. Mayer, L. Pawlak, U. Rauchschwalbe, H. Spille and F. Steglich, J. Magn. Magn. Mat. 63 & 64 (1987) 91.

[2] G. Knopp, H. Spille, A. Loidl, K. Knorr, U. Rauchschwalbe, R. Felten, G. Weber, F. Steglich and A.P. Murani, J. Magn. Magn. Mat. 63 & 64 (1987) 88.

[3] G. Knopp, A. Loidl, K. Knorr, L. Pawlak, M. Duczual, R. Caspary, U. Gottwick, H. Spille, F. Steglich and A.P. Murani, Z. Physik B, to be published.

[4] C.D. Bredl, S. Horn, F. Steglich, B. Lüthi and R. Martin, Phys. Rev. Lett. 52 (1984) 1982.

[5] A. Loidl, G. Knopp, H. Spille, F. Steglich and A.P. Murani, Proc. Intern. Conf. on Neutron Scattering, Grenoble (1988), to be published.
[6] A. Zuber, Diploma thesis, TH Darmstadt (1988), unpublished.
[7] R. Felten, Ph. D. thesis, TH Darmstadt (1987), unpublished.
[8] A. Böhm, Diploma thesis, TH Darmstadt (1988), unpublished.
[9] R. Caspary, Diploma thesis, TH Darmstadt (1988), unpublished.