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Thermodynamic properties of $Ce(Ru_xRh_{1-x})_3B_2$

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Abstract

Investigations of the magnetic phase diagram of $Ce(Ru_xRh_{1-x})_3B_2$ were complemented by detailed specific heat and susceptibility measurements revealing complex magnetic order without spontaneous magnetisation. This regime follows the ferromagnetic phase on Rh/Ru substitution for $x \gtrsim 0.06$.

Keywords: $CeRh_3B_2$; $Ce(Ru_xRh_{1-x})_3B_2$; Phase diagram; Ferromagnetism

Cerium, ytterbium or uranium intermetallics based on the $CaCu_5$-type structure exhibit a wide range of ground state properties due to the interplay of the Kondo effect, RKKY-interaction and crystal field splitting. In the course of such studies, $Ce(Ru_xRh_{1-x})_3B_2$ has attracted our attention, because its low-temperature magnetic phase boundary is hitherto uncertain. Moreover, the unusual large Curie temperature of $CeRh_3B_2$ ($T_c \approx 115$ K) is still subject of controversial discussions. Very recently, this extraordinary feature was associated with a substantial Ce 5d spin moment, whereas the Kondo effect does not play a key role in this compound [1]. With respect to $Ce(Ru_xRh_{1-x})_3B_2$, a rather drastic decay of ferromagnetic order from $T_c = 115$ K at $x = 0$ to $T_c \approx 80$ K at $x = 0.06$ was obtained, followed by a paramagnetic phase (see e.g. Refs. [2–4]). Careful investigations of the concentration range $0.1 < x < 0.45$ indicated [5], that a second magnetic phase appears with complex magnetic order and a maximum ordering temperature $T_M \cong 70$ K at $x = 0.2$. In addition to our former studies, this paper is concerned with the thermodynamic properties of the series $Ce(Ru_xRh_{1-x})_3B_2$ in both magnetic regimes.

In Fig. 1 we present specific heat measurements for alloys with $x = 0.2$, 0.4 and RE = Ce, La as representatives for this series. The C_p/T vs. T dependency clearly reveals the magnetic phase transition around 70 K. This phase transition stays almost constant, even in magnetic fields of 9 T. The increase of ruthenium to $x = 0.4$ results in the complete disappearance of magnetic order in the temperature range studied. In the inset we present the C_p/T vs. T^2 relation in order to deduce the Sommerfeld value γ of the specific heat for all alloys studied. The γ-value rises from about 13(2) at $x = 0$ to about 33(2) mJ/mol K^2 at $x = 0.1$ in line with the gradual decay of ferromagnetic order. For $x > 0.1$ the γ-values decrease reaching a minimum at $\gamma = 23(2)$ mJ/mol K^2 for $x = 0.2$ which shows the largest transition temperature of the complex magnetic regime. On further Rh/Ru substitution γ raises again up to 60(2) mJ/mol K^2 at $x = 0.4$ consistent with the onset of magnetic fluctuations in the NFL (non-Fermi liquid) region adjoining the complex magnetic domain. The latter value coincides with $\gamma = 67(1)$ mJ/mol K^2 for $x = 0.4$, derived from measurements down to 400 mK [5]. The extrapolated γ-values for the isotypic La-alloys are 11.7(2),13.5(2) and 5.3(2) mJ/mol K^2, respectively, for $x = 0$, 0.2, 0.4. The deduced Debye temperatures ($\Theta_D = 425(5)$, 435(5) and 440(10) K) indicate a rather stiff atomic lattice, most likely due to the boron metal bonds. Values of this order of magnitude have been reported earlier [6].

In Fig. 2 magnetisation data for $x \leqslant 0.085$ (left part) and susceptibility data for $x > 0.085$ (right part) are shown. T_C drops rapidly from 105 K ($x = 0.02$) to 80 K at

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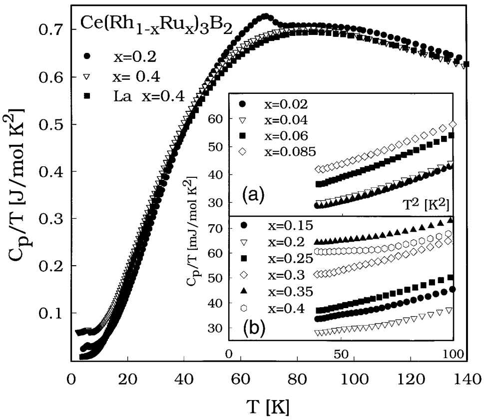

Fig. 1. Temperature dependent specific heat C_p plotted as C_p/T vs. T for $Ce(Ru_xRh_{1-x})_3B_2$, $x = 0.2$, 0.4 and for $La(Ru_{0.4}Rh_{0.6})_3B_2$. The insets show the low temperature behaviour of C_p/T vs. T^2 for various compositions x; (a) ferromagnetic phase and (b) complex order.

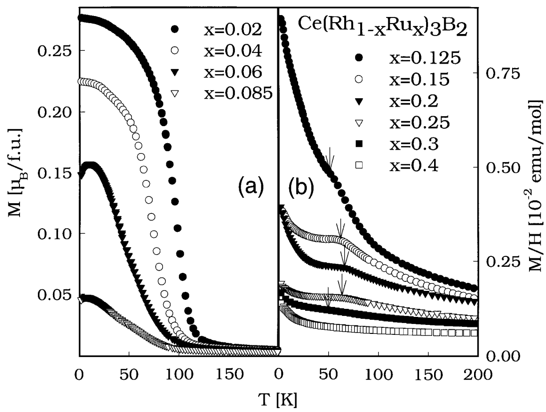

Fig. 2. Temperature dependent magnetisation M for $x \leqslant 0.085$ (a) and magnetic susceptibility M/H for $x > 0.085$ (b) obtained at 1 T. The arrows indicate the magnetic transition temperatures.

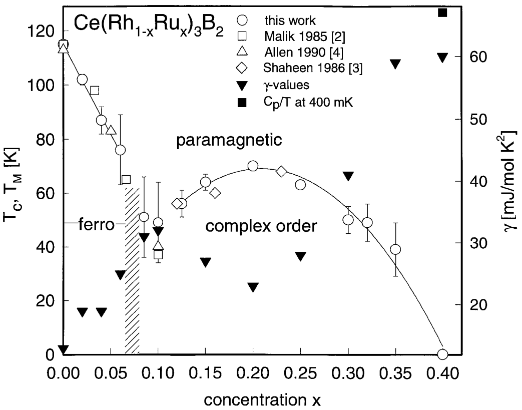

Fig. 3. Tentative magnetic phase diagram of $Ce(Ru_xRh_{1-x})_3B_2$.

$x = 0.06$ in good accordance with previous data [2–4]. Uncertainties in the determination of the ordering temperatures for $x \gtrsim 0.06$ arise due to extraordinary broad features in magnetisation, susceptibility and resistivity data. For the complex magnetic domain ($0.06 \lesssim x < 0.4$), T_M passes through a maximum at $x = 0.2$ (70 K) and finally vanishes for $x \geqslant 0.4$ (see arrows in right part of Fig. 2). Similarly the magnetisation M gradually decreases on Rh/Ru substitution while the absolute M values are extremely small.

Fig. 3 presents the magnetic phase diagram for the system $Ce(Ru_xRh_{1-x})_3B_2$ as derived from our measurements of the electrical resistivity, magnetic susceptibility and the specific heat. The results obtained from these independent methods are in excellent agreement. Furthermore we show a comparison with the data available in literature as far as the rhodium-rich, ferromagnetic region is concerned. The phase diagram reveals three distinct magnetic regions: the ferromagnetic range exhibits a steep drop of T_c as a function of Rh/Ru replacement, however gradually flattens out above $x \cong 0.06$ in a transition to the ordered complex magnetic regime. The present data thus presumably indicate a higher order critical point at the phase boundaries: paramagnetic, ferromagnetic and complex magnetic order. Therefore, a complete suppression of the magnetic order towards $T_c = 0$ is rather unlikely. It is interesting to note that the data points for $Ce(Ru_{0.1}Rh_{0.9})_3B_2$ published earlier [2,4] at $T_C \approx 40$ K would rather match our complex magnetic regime since an Arrott plot does not provide hints of a typical ferromagnet as do all the concentrations $x \leqslant 0.06$. In the light of this discussion we may point out that some of the hitherto published susceptibility data [2–4] for samples with $x \gtrsim 0.06$ now find a proper interpretation when the observed anomalies of $\chi(T)$ are associated with the complex magnetic order of yet unknown microscopic nature.

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References

[1] A. Yaounac et al., Phys. Rev. B 57 (1998) R681.
[2] S.K. Malik et al., Phys. Rev. B 31 (1985) 4728.
[3] S.A. Shaheen et al., J. Magn. Magn. Mater. 54–57 (1986) 357.
[4] J.W. Allen et al., Phys. Rev. B 41 (1990) 9013.
[5] E. Bauer et al., J. Appl. Phys. 83 (1998) 6423.
[6] S.A. Shaheen et al., J. Magn. Magn. Mater. 47/48 (1985) 285.