Weak itinerant ferromagnetism in YCo$_9$Si$_4$

H. Michor$^a$, M. El-Hagary$^a$, S. Özcan$^a$, A. Horyn$^a$, E. Bauer$^a$, M. Reissner$^a$, S. Khmelevskyi$^b$, P. Mohn$^b$, P. Rogl$^c$

$^a$Institut für Festkörperphysik, T.U. Wien, A-1040 Wien, Austria
$^b$Center for Computational Materials Science, T.U. Wien, A-1040 Wien, Austria
$^c$Institut für Physikalische Chemie, Universität Wien, A-1090 Wien, Austria

Abstract

Weak itinerant ferromagnetism in YCo$_9$Si$_4$ below about 25 K is studied by means of magnetisation, specific heat, and resistivity measurements. Single crystal X-ray Rietveld refinements at room temperature reveal a fully ordered distribution of Y, Co and Si atoms within the tetragonal space group $I4/mcm$ isostructural with LaCo$_9$Si$_4$. The latter exhibits itinerant electron metamagnetism with an induced moment of about 1 $\mu_B$/f.u. above 6 T, whereas YCo$_9$Si$_4$ exhibits a spontaneous magnetisation $M_0 \approx 12$ Am$^2$/kg at 2 K which corresponds to an ordered moment of about 1.6 $\mu_B$/f.u. indicating weak itinerant ferromagnetism.

Key words:
YCo$_9$Si$_4$, itinerant magnetism, specific heat

Recent interest on weak itinerant ferromagnetism e.g. in ZrZn$_2$ [1] in the context with quantum critical phenomena motivated the search for new materials showing weak itinerant ferromagnetism or being close to a ferromagnetic (FM) instability. An interesting system in this respect is the solid solution LaCo$_{9-x}$Si$_x$ where ferromagnetism vanishes near the stoichiometric composition LaCo$_9$Si$_4$ [2] where full translational symmetry (space group $I4/mcm$) is confirmed by single crystal X-ray diffractometry [3]. LaCo$_9$Si$_4$ is a strongly exchange enhanced Pauli paramagnet and exhibits an itinerant electron metamagnetic phase transition at about 3.5 T for $H || c$ and 6 T for $H \perp c$, which is the lowest value ever found for rare earth intermetallic compounds [3]. In this paper, we report on low temperature measurements on the isostructural and isoelectronic compound YCo$_9$Si$_4$ which was initially reported in Refs. [4,5] to be FM with $T_C \approx 848$ K.

Polycrystalline samples of YCo$_9$Si$_4$ were synthesized by induction melting of pure elements (Y 3N, Co 4.5N, Si 6N) under protective argon atmosphere and subsequent annealing at 1050$^\circ$C for one week. The phase purity and composition has been verified by means of electron microprobe analysis. The crystal structure was determined by means of single crystal X-ray diffraction ($R_F = 2\%$) revealing a fully ordered distribution of Y, Co and Si atoms with the LaFe$_9$Si$_4$-type [6] with a single rare earth site, three cobalt sites and again a single Si site. The lattice parameters are $a = 7.754(1)$ Å and $c = 11.487(1)$ Å at room temperature (see Ref. [3] for experimental details). Crystallographic order is also corroborated by a reasonably low residual resistivity $\rho_0 = 7\Omega$cm (see below).

Temperature and field dependent magnetisation measurements $M(T, H)$ on YCo$_9$Si$_4$ depicted in Fig. 1 as an Arrott plot $M^2$ versus $H/M$ reveal weak ferromagnetism below about 25 K with an extrapolated spontaneous magnetisation $M_0 \approx 12$ Am$^2$/kg at 2 K (see the dashed line in Fig. 1) corresponding to...
associated with the second order phase transition towards $YCo\_9$ field value both compounds a relatively large electronic Sommer-land value $LaCo\_1$. 

Fluctuation (Fermi liquid) regime for the latter compound $\rho = 0$.

$A\mu_0$ with FM state, $\chi_{FM}$ regime, $LaCo\_1$. $

The Curie-temperature $T_C$ is around 25 K in reasonable agreement with specific heat and transport anomalies (see below).

The temperature dependent resistivity $\rho(T)$ of $YCo\_9$ shown in Fig. 2 reveals a significant change of slope around about 25 K which is associated with the onset of ferromagnetism. Below about 15 K, in the FM state, $\rho(T)$ is well described by a power law behavior $\rho(T) = \rho_0 + AT^\alpha$ (see the solid line in Fig. 2) with $\rho_0 = 7\mu\Omega cm$, $A = 0.176\mu\Omega cm/K^{-\alpha}$ and $\alpha = 1.72$. The corresponding fit for nearly ferromagnetic $LaCo\_9$ (dashed line in Fig. 2) yields $\rho_0 = 16\mu\Omega cm$, $A = 0.085\mu\Omega cm/K^{-\alpha}$ and $\alpha = 1.9$ indicating a spin fluctuation (Fermi liquid) regime for the latter compound.

The specific heat of $YCo\_9$ and (for comparison) $LaCo\_9$ is shown in Fig. 3 as $C/T$ vs. $T$ revealing for both compounds a relatively large electronic Sommerfeld value $\gamma$ close to 200 mJ/mol K$^2$ and in the case of $YCo\_9$ a small somewhat broadened anomaly associated with the second order phase transition towards weak itinerant ferromagnetism with a jump $\Delta C/T$ of the order of 100 mJ/mol K$^2$ in approximate agreement with the Stoner-Wohlfarth model (see e.g. Ref. [7]) yielding $\Delta C/T_C = M^2_h/\chi_0 T_C^2 \sim 70 mJ/mol K^2$. In the case of exchange enhanced Pauli paramagnetic $LaCo\_9$ the value of $\gamma \sim 200 mJ/mol K^2$ can be compared with the density of states obtained from ab-initio electronic structure calculations, $N(E_F) \sim 19 states/eV f.u.$, revealing a spin-fluctuation mass enhancement $\lambda_{spin} \sim 3.3 [3]$.

For $YCo\_9$ band calculations have been performed in the same manner as described in Ref. [3] for $LaCo\_9$ yielding practically the same picture with respect to the Co $d$-bands as for $LaCo\_9$ and within the numerical accuracy the same density of states at the Fermi level. The spin-fluctuation mass enhancement $\lambda_{spin}$ is thus very similar in $YCo\_9$ and $LaCo\_9$. Band calculations at the experimental lattice constant yield a FM ground state for both compounds, which is experimentally confirmed only for $YCo\_9$ while $LaCo\_9$ shows a paramagnetic ground state and metamagnetism. In analogy to the conclusions drawn for $LaCo\_9$ we expect also for $YCo\_9$ in the FM state the largest moments of about 0.3–0.4 $\mu_B$/Co to be at the 16$k$ Co-sites and significantly smaller moments at the 4$d$ and 16$l$ Co-sites.

References
[1] C. Pfleiderer et al., Nature (London) 412 (2001) 58.
[2] M. El-Hagary et al., J. Alloys Compounds 367 (2004) 239.
[3] H. Michor et al., Phys.Rev. B 69 (2004) 081404(R).
[4] R.V. Skolozdra et al., J. Alloys Compounds 296 (2000) 272.
[5] Yu. Gorelenko et al., Visnyk of the Lviv University, Series Chemical 43 (2003) 62.
[6] W.H. Tang et al., J. Appl. Phys. 76 (1994) 4095.
[7] P. Mohn and G. Hilscher, Phys.Rev. B 40 (1989) 9126.