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

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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 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 $M_0 \approx 12 \text{Am}^2/\text{kg}$ at 2 K which corresponds to an ordered moment of about 1.6$\mu_B$/f.u. indicating weak itinerant ferromagnetism.

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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$_{13-x}$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 $H \parallel c$ 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 $T_C \simeq 848$ 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°C for 1 week. The phase purity and composition has been verified by means of electron microprobe analysis. A small crystal ($\sim 50 \mu$m$^3$ extracted from polycrystalline material was used to determine the crystal structure 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 details). Crystallographic order is also corroborated by a reasonably low residual resistivity $\rho_0 = 7 \mu\Omega\text{cm}$ (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 $M_0 \simeq 12$ Am$^2$/kg at 2 K (see the dashed line in Fig. 1) corresponding to $1.6 \mu_B$/f.u. and a longitudinal susceptibility in the FM regime, $\chi_0 \sim 0.25$ Am$^2$/kgT, determined from the $\mu_0H/M$ axis intercept of the dashed line extrapolation in Fig. 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$_9$Si$_4$ 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 $\rho_0 = 7 \mu\Omega\text{cm}$, $A = 0.176 \mu\Omega\text{cm}/K^{-\alpha}$ and $\alpha = 1.72$. The corresponding fit for nearly ferromagnetic LaCo$_9$Si$_4$ (dashed line in Fig. 2) yields $\rho_0 = 16 \mu\Omega\text{cm}$, $A = 0.085 \mu\Omega\text{cm}/K^{-\alpha}$ and $\alpha = 1.9$ indicating a spin fluctuation (Fermi liquid) regime for the latter compound.

The specific heat of YCo$_9$Si$_4$ and (for comparison) LaCo$_9$Si$_4$ 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$_9$Si$_4$ 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 $\Delta C/T_c = M_0^2/\chi_0 T_C^2 \simeq 70$ mJ/mol K$^2$. In the case of exchange enhanced Pauli paramagnetic LaCo$_9$Si$_4$, the value of $\gamma \simeq 200$ mJ/mol K$^2$ can be compared with the density of states

![Fig. 1. Arrott plot $M^2$ vs. $H/M$ of isothermal magnetization data of YCo$_9$Si$_4$.](image1)

![Fig. 2. Temperature-dependent resistivity $\rho(T)$ of YCo$_9$Si$_4$ and LaCo$_9$Si$_4$; dashed and solid lines are fits, see text.](image2)
obtained from ab initio electronic structure calculations, \(N(E_F) \sim 19 \) states/eV f.u., revealing a spin-fluctuation mass enhancement \(\lambda_{\text{spin}} \sim 3.3\) [3].

For \(\text{YCo}_9\text{Si}_4\) band calculations have been performed in the same manner as described in Ref. [3] for \(\text{LaCo}_9\text{Si}_4\) yielding practically the same picture with respect to the Co d-bands as for \(\text{LaCo}_9\text{Si}_4\) and within the numerical accuracy the same density of states at the Fermi level. The spin-fluctuation mass enhancement \(\lambda_{\text{spin}}\) is thus very similar in \(\text{YCo}_9\text{Si}_4\) and \(\text{LaCo}_9\text{Si}_4\). Band calculations at the experimental lattice constant yield a FM ground state for both compounds, which is experimentally confirmed only for \(\text{YCo}_9\text{Si}_4\), while \(\text{LaCo}_9\text{Si}_4\) shows a paramagnetic ground state and metamagnetism. In analogy to the conclusions drawn for \(\text{LaCo}_9\text{Si}_4\) we expect for \(\text{YCo}_9\text{Si}_4\) also in the FM state the largest moments of about 0.3–0.4\(\mu_\text{B}/\text{Co}\) to be at the 16k Co-sites and significantly smaller moments at the 4d and 16l Co-sites.

**Fig. 3.** Temperature-dependent specific heat as \(C/T\) vs. \(T\) of \(\text{YCo}_9\text{Si}_4\) and \(\text{LaCo}_9\text{Si}_4\).