Transition from weak ferromagnetism to metamagnetism in the itinerant-electron system $Y_{1-x}La_xCo_9Si_4$

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Abstract. The magnetism of solid solution $Y_{1-x}La_xCo_9Si_4$ between strongly enhanced Pauli paramagnetic LaCo$_9$Si$_4$ and weakly ferromagnetic YCo$_9$Si$_4$ has been investigated. The Curie temperature $T_C$ in the ferromagnetic region and the metamagnetic transition field $H_M$ in the paramagnetic region change continuously against $x$ and approach zero at the same composition $x \approx 0.15$, suggesting the presence of a critical point from spontaneous to field-induced ferromagnetism.

1. Introduction

Strongly enhanced Pauli paramagnetic LaCo$_9$Si$_4$ exhibits an itinerant electron metamagnetic transition at a low field of $H_M \approx 4.5$ T [1, 2], while YCo$_9$Si$_4$ is a weak itinerant ferromagnet with the Curie temperature $T_C \approx 25$ K and spontaneous moment 1.6 $\mu_B$/f.u. [3]. They form the same LaFe$_9$Si$_4$-type crystal structure (space group $I4/mcm$). In the structure, there are three inequivalent Co sites, $16k$, $16l$ and $4d$. Among them, only the $16k$ site, which forms a two-dimensional network in the $c$ plane, is magnetic [1]. It is interesting to note that the lattice volume of LaCo$_9$Si$_4$ is larger than that of YCo$_9$Si$_4$. This fact is in discordance with the general tendency in 3d itinerant electron systems, where the Stoner criterion is satisfied by narrowing of 3d bands associated with volume expansion. On the other hand, as expected, the ferromagnetism of YCo$_9$Si$_4$ is destabilized under pressure; a ferromagnetic quantum critical point is expected to be present under pressure of the order of 6 GPa, and non-Fermi liquid behaviors have been observed near the critical point [4]. In this study, we have prepared specimens of $Y_{1-x}La_xCo_9Si_4$ systematically and measured evolution of the magnetism to make a magnetic phase diagram.

2. Experimental procedures

Polycrystalline samples of $Y_{1-x}La_xCo_9Si_4$ with different $x$ were prepared by arc melting under an argon atmosphere. Y (3N pure) and La (3N) ingots were first melted to make an alloy, which was melted together with Co (3N) and Si (6N) chips. The total mass of ingots was controlled to be less than 3 g to improve homogeneity. Taking account of the loss of Si and Y-La during the melting, the starting composition of Co was fixed to 8.9. The obtained alloys were sealed in evacuated quartz tubes and annealed at 1050°C for 1 week. The pure compounds ($x = 0$ and 1) prepared with the same method reproduce well the magnetic properties.
Figure 1. (a) Temperature dependences of magnetization measured for $Y_{1-x}La_xCo_9Si_4$ under 1 T. Blue and red marks represent ferromagnetic and paramagnetic materials. (b) Concentration dependences of the Curie temperature $T_C$ and the metamagnetic transition field $H_M$ of $Y_{1-x}La_xCo_9Si_4$. Reported in the literatures [1, 3]. All the samples were characterized by x-ray powder diffraction with Cu $K\alpha$ radiation. The Rietvelt refinement analysis reproduced satisfactorily the x-ray patterns by assuming fully ordered Co and Si sites. The magnetization was measured using a SQUID magnetometer (Quantum Design MPMS-5) in the temperature range 1.8–290 K and the magnetic field range 0–5 T.

3. Results and discussion

Temperature dependences of magnetization for $Y_{1-x}La_xCo_9Si_4$ with different $x$ are shown in figure 1. They were measured under a relatively high field of 1 T. Y-rich compounds exhibit typical ferromagnetic behaviors. As the La content increases, the low-temperature magnetization is reduced systematically and nearly continuously. As reported, LaCo$_9$Si$_4$ shows a broad

Figure 2. (a) The magnetization curve and its field-derivative of $Y_{0.3}La_{0.7}Co_9Si_4$ at 2 K. (b) Temperature dependences of magnetization (under a field of 0.01 T) and inverse susceptibility (measured at 1 T) for $Y_{0.9}La_{0.1}Co_9Si_4$. The inset shows the magnetization curve at 5 K.
maximum at ~20 K. In cases of La-rich compounds, the susceptibility $\chi(T)$ obeys the Curie-Weiss law at high temperatures, but tends to be suppressed at low temperatures, suggesting that the maximum in $\chi(T)$ shifts to lower temperatures from ~20 K of pure LaCo$_9$Si$_4$.

Figure 2(a) shows, as an example of La-rich compounds, the magnetization curve $M(H)$ and its field-derivative $dM/dH$ of Y$_{0.3}$La$_{0.7}$Co$_9$Si$_4$ measured at 2 K. The magnetization varies linearly in a low field range, indicating no spontaneous magnetization, and shows an S-shape anomaly near 4 T, namely a metamagnetic transition as in pure LaCo$_9$Si$_4$, although the transition is broadened by inhomogeneity due to the alloying. From the peak position of $dM/dH$, the transition field $H_M$ is estimated to be ~3.7 T, which is reduced from ~4.5 T of pure LaCo$_9$Si$_4$.

As an example of Y-rich compounds, $M(T)$ and $1/\chi(T)$ is shown for Y$_{0.9}$La$_{0.1}$Co$_9$Si$_4$ in figure 2(b). The inset shows the $M(H)$ curve at 5 K. These are of a typical ferromagnet. For this compound, $T_C$ was estimated to be 15 K from the Arrott plot. The spontaneous moment at 2 K is estimated to be 1.2$\mu_B$/f.u., which is a little smaller than that of YCo$_9$Si$_4$.

Figures 3(a) and (b) show $\chi(T)$ and $dM/dH$, respectively, for Y$_{0.85}$La$_{0.15}$Co$_9$Si$_4$, a compound close to the ferromagnetic-paramagnetic boundary. Here $\chi(T)$ was measured under a small field of 0.01 T, which is less than $H_M \approx 0.02$ T as seen in figure 3(b). The $\chi(T)$ peak is reduced down to ~4 K.

We made similar measurements on other specimens and determined $T_C$ and $H_M$, which are plotted in figure 1(b) as a function of $x$. Both $T_C$ and $H_M$ change monotonically, and approach zero at the identical concentration $x \approx 0.15$, indicating the presence of a phase transition from spontaneous to field-induced ferromagnetism at 0 K, namely, a ferromagnetic quantum critical point. In spite of several trials, however, we have not succeeded in making the sample with both $T_C$ and $H_M$ equal to 0 at the same time.

It is of interest to compare the variation of magnetism with that of lattice volume. Figure 4(a) shows concentration dependences of lattice constants $a$ and $c$ at room temperature evaluated in the Rietveld refinement analysis. Both $a$ and $c$ show similar dependences, but interestingly, do not vary monotonically. The lattice volume $\omega = a^2c$ is plotted in figure 4(b). (Note that all the variation of $\omega$ does not correspond to uniform volume expansion because the lattice expansion is, exactly speaking, not isotropic, leaving ambiguities in quantitative discussions.) In the Y-rich side, the ferromagnetism is destabilized in spite of the slight expansion of $\omega$ at $x \leq 0.15$. This means that the narrowing of 3d bands associated with the volume expansion is not the origin of the ferromagnetism. In general, $\omega$ can be decomposed into lattice and magnetic terms as $\omega = \omega_L + \omega_M$. Here we assume that $\omega_M$ is a function of $S_L^2$, where $S_L$ is longitudinal spin fluctuations at the atomic site. If electrons are well delocalized both in paramagnetic
Figure 4. (a) Concentration dependences of lattice parameter $a$ and $c$ of $Y_{1-x}La_xCo_9Si_4$ at room temperature. (b) The concentration dependence of lattice volume $\omega = a^2c$ at room temperature. The broken line is the guide for eyes.

and ferromagnetic regions, and the electronic state varies continuously at the critical point, we expect moderate increase of $S_L$ with temperature well above $T_C$, and hence weak concentration dependence of $\omega_M$ at room temperature. On the other hand, as actually seen in figure 4(b), large additional volume expansion deviating from nearly linear variation (the broken line) was observed in the ferromagnetic region, which should be ascribed to additional spontaneous volume magnetostriction. (Note that the broken line does not correspond to $\omega_L$ but include both $\omega_L$ and the temperature-induced term of $\omega_M$.) This indicates that $S_L^2$ at room temperature of the Y-rich ferromagnetic compounds is much larger than that of paramagnetic phases at the critical region ($x \sim 0.3$), and suggests that Co local moment is stabilized not only below $T_C$ but also in all the temperature range. In other words, the magnetism of $YCo_9Si_4$ is rather close to the local moment limit. The localized nature of Co moment in the ferromagnetic phase and the transition to Fermi-liquid-like state seem to characterize the critical point in the present system.

In summary, we measured concentration dependences of the Curie temperature $T_C$ and the metamagnetic transition field $H_M$ for $Y_{1-x}La_xCo_9Si_4$ and confirmed that both the quantities change continuously and approach zero at $x \simeq 0.15$, revealing the presence of a critical point from spontaneous ferromagnetism to field-induced ferromagnetism at the composition. Although the concentration dependence of magnetic properties looks continuous, some of quantities such as lattice volume change discontinuously at the critical point, which seems to characterize the nature of the $(Y-La)Co_9Si_4$ system.

References
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