Magnetostriction studies on the itinerant electron metamagnet LaCo$_9$Si$_4$

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Abstract. The magneto-volume coupling of the itinerant electron metamagnetic compound LaCo$_9$Si$_4$ was studied by measuring the forced magnetostriction of a single crystal utilizing a capacitive miniature dilatometer cell. The volume changes and the anisotropy with respect to different crystal orientations were measured as a function of the applied magnetic field for different temperatures ranging from 4.2 K up to 50 K. The forced volume-magnetostriction coefficient $\lambda_V(H) = V^{-1} \partial V/\partial H$ is positive up to 9 T with a pronounced maximum at the critical field $\mu_0 H_c \simeq 3.5$ T for $H \parallel c$ and $\mu_0 H_c \simeq 5.85$ T for $H \perp c$, respectively.

1. Introduction

The family of rare earth cobalt silicides $R$Co$_9$Si$_4$ with tetragonal crystal structure, space group $I4/mcm$, exhibits extraordinary electronic ground states like weak itinerant ferromagnetism in YCo$_9$Si$_4$ [1], ferrimagnetism in GdCo$_9$Si$_4$ [2], intermediate valence in CeCo$_9$Si$_4$ [3] and most interestingly strongly exchange-enhanced Pauli paramagnetism and itinerant electron metamagnetism (IEMM) in LaCo$_9$Si$_4$ [4]. Earlier reports [5] on the onset of ferromagnetism in the solid solution LaCo$_{9-x}$Si$_x$ near the critical concentration $x = 4$ attracted our attention and motivated a careful re-investigation of LaCo$_9$Si$_4$ and the solid solution with compositions nearby [6]. Contrary to reports on weak ferromagnetism with $T_c = 900$ K in LaCo$_9$Si$_4$ [5], a paramagnetic ground state has been established [6] for stoichiometric, single phase LaCo$_9$Si$_4$. The exchange-enhanced paramagnetic ground state of LaCo$_9$Si$_4$ becomes instable towards a ferromagnetic one for fields larger than 3 T and exhibits an IEMM transition [4].

Ab initio electronic structure calculations of LaCo$_9$Si$_4$ and YCo$_9$Si$_4$ revealed essentially the same Co 3$d$ band features and suggested a weakly ferromagnetic ground state at the experimental lattice constants for both compounds [1, 4]. Experimentally, however, a ferromagnetic ground state is observed only for YCo$_9$Si$_4$ [1], whereas strongly exchange-enhanced Pauli paramagnetic LaCo$_9$Si$_4$ slightly falls short of the Stoner limit for ferromagnetism, i.e. both compounds are located nearby, but on opposite (para- and ferromagnetic) sides of a ferromagnetic quantum critical point. YCo$_9$Si$_4$ exhibits a 2.75 % smaller unit cell volume than LaCo$_9$Si$_4$ and, thus, motivates studies of the magneto-volume coupling in these isoelectronic compounds which are both close to the Stoner limit. The application of hydrostatic pressure on YCo$_9$Si$_4$ results in a decrease of the Curie temperature $T_c$, i.e. reveals a positive magneto-volume coupling, and indicates that a ferromagnetic quantum critical point may be reached at 6 GPa [7]. In this paper, we report on low temperature magnetostriction studies on LaCo$_9$Si$_4$ and discuss the magneto-volume coupling in this IEMM material.
2. Experimental details

The starting alloy for crystal growth is prepared by inductive levitation melting of highest purity elements lanthanum (Ames MPC [8], 3N8), cobalt (Umicore, 5N) and silicon (Alfa Aesar, 6N) under argon (Linde, 6N) atmosphere. The single crystal was grown via the Czochralski pulling method using a tri-arc furnace by Centorr Vacuum Industries. In order to obtain stoichiometric LaCo$_9$Si$_4$ some excess of cobalt has been added to the starting alloy. The magnetic properties of LaCo$_{9-x}$Si$_x$ are extremely sensitive to the stoichiometry [6] (deviations from $x = 4$ by only $\pm 0.05$ result either in the appearance of a ferromagnetic component or in the disappearance of the IEMM), thus, offering a rather sensitive option to check the composition of the crystal.

X-ray studies of fragments of the crystal were carried out on a Siemens D-5000 diffractometer to check the lattice parameters and phase purity. Optical techniques and x-ray Laue photos were utilized for the selection and orientation of the largest single crystal domain with dimensions after preparation $685 \times 820 \times 1045 \mu m$. Subsequent investigations on the crystal quality in terms of the residual resistivity ratio (RRR) revealed a RRR $\sim 3$.

A capacitive dilatometer based on the tilted plate principle was used for magnetostriction measurements [9]. The cell was designed and tested for a wide temperature range (0.3-300 K), high magnetic fields (up to 11 T) and the use of small (active length $\sim 1$ mm) and irregular shaped samples. The compact design permits the mounting parallel and perpendicular to a superconducting coil and therefore it is possible to measure the full magnetostriction tensor of anisotropic materials.

3. Experimental results and discussion

The magnetic susceptibility $\chi(T)$ of LaCo$_9$Si$_4$ obtained from low field SQUID magnetometry shown in the insert of figure 1b is strongly exchange-enhanced and exhibits a maximum at 13 K and 22 K for $H \parallel c$ and $H \perp c$, respectively, in good agreement with previous studies [4]. The position of the maximum $T_{max}$ is field dependent and gets continuously suppressed to zero at the critical field $H_c$ (not shown for brevity).
A vibrating sample magnetometer (Quantum Design PPMS) was used for additional field dependent isothermal magnetization studies carried out both, with rising and falling field. At low temperatures $T < 25$ K the isothermal magnetization displayed in figure 1a with $H \parallel c$ and figure 1b with $H \perp c$ shows a steplike increase of $M(H)$, the IEMM, at critical fields $\mu_0 H_c \approx 3.5$ T and $\mu_0 H_\perp \approx 5.85$ T, respectively. For elevated temperatures $T > 25$ K the IEMM disappears in the isothermal magnetization $M(H)$. The observed anisotropy of the magnetic susceptibility, of the temperature of the maximum in $\chi(T)$ and of the critical field of IEMM are supposed to originate from a small but finite orbital contribution to the overall induced spin-dominated magnetic moment [4].

Magnetostriiction measurements carried out for the orientations in $c$- and $a$-direction, both with $H \parallel c$ at 4.2 K, are displayed in figure 2a together with the corresponding relative volume change $\omega = \Delta l_c/l_c + 2 \times \Delta l_a/l_a$. The latter, $\omega(H)$, as well as the magnetostriction coefficient $\lambda(H) = V_0^{-1} \partial V(H)/\partial \mu_0 H$ (insert in figure 2a) are positive. Length changes with respect to zero-field are measured in 0.1 T steps ranging from 0 to 9 T, the magnetostriction coefficients are calculated via $\lambda_i(B) = l_i^{-1}(0)[l_i(B + \Delta B) - l_i(B)]/\Delta B$. In the vicinity of the critical field $\lambda(H)$ passes a sharp maximum at $\mu_0 H_c = 3.5$ T. A tetragonal distortion of the crystal is observed, though absolute values of relative length changes, $|\Delta l/\mu_0(H)|$, still follow the trend of $\omega(H)$. Thereby, the $c$-axis increases its length approximately 3 times as large as the $a$-axis length decreases. The shape of the curves $\Delta l/\mu_0(H)$ differ from each other just by a signed scaling factor.

Applying the magnetic field along the $a$-direction of the crystal (figure 2b) shifts the position of the step-like increase in $\omega(H)$ to the value of the critical field $\mu_0 H_a \approx 5.85$ T and enhances the volume-coupling with respect to $H \parallel c$. Two different cases have to be distinguished for the $a$-axis, the one with $H \parallel a$ and the other with $H \perp a$. Again the $c$ lattice parameter $(H \perp c)$ increases while the length in both $a$-directions shrinks. The $a$-axis parallel to the magnetic field changes its length less than the perpendicular oriented one leading to a distortion of the crystal’s basal plane, which is not observed for $H \parallel c$, and an overall orthorhombic distortion of the crystal.

**Figure 2.** Volume magnetostriction $\omega = \Delta V/V$ and linear magnetostriction $\Delta l_c/l_c$, $\Delta l_a/l_a$ of LaCo$_5$Si$_4$ for $H \parallel c$ in (a) and $\omega$, $\Delta l_c/l_c$, $\Delta l_a/l_a|H|$, $\Delta l_{a,H}/l_{a,H}$ and the basal plane distortion for $H \perp c$ in (b) measured at $T = 4.2$ K. Inserts in both (a) and (b) show the volume magnetostriction coefficient $\lambda_V = \partial \omega/\partial \mu_0 H$ and the linear coefficients $\lambda_i = l_i^{-1}\partial l_i/\partial \mu_0 H$. 


The relative volume changes $\omega$ versus $M^2$ for $H\parallel c$ and $H \perp c$ are shown in figure 3. The magneto-volume coupling roughly follows the relation $\omega(H) = \kappa CM^2(H)$ with a coupling factor $\kappa C_c \simeq 2.5 \times 10^{-5}(\text{f.u.}/\mu B)^2$ for $H\parallel c$ and $\kappa C_a \simeq 3.4 \times 10^{-5}(\text{f.u.}/\mu B)^2$ for $H \perp c$. Interestingly, the coupling $\kappa C$ exhibits a slight increase when passing the IEMM transition which is different from archetypal IEMM system LuCo$_2$ where a significant reduction of $\kappa C$ has been observed when entering in the field induced ferromagnetic state [10].

4. Conclusions

Investigations of LaCo$_9$Si$_4$ by means of magnetostriction studies revealed a positive magneto-volume coupling at low temperatures accompanied with a tetragonal ($H\parallel c$) and orthorhombic ($H \perp c$) distortion of the crystal. A sharp maximum in the volume magnetostriction coefficient $\lambda_V(H)$ right at the critical field of IEMM is observed for both $H\parallel c$ and $H \perp c$. Despite of the fact that a ferromagnetic ground state is observed for YCo$_9$Si$_4$ with smaller unit cell volume and a paramagnetic one for LaCo$_9$Si$_4$ with larger unit cell volume, both compounds exhibit a positive magneto-volume coupling, i.e. pressure applied to LaCo$_9$Si$_4$ is expected to decrease the paramagnetic susceptibility and to increase the metamagnetic critical field.

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