Magnetovolume anomalies in $Ce_2Fe_{17-x}Mn_x$

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The small nonmonotonic changes of the lattice parameters upon the substitution of Mn for Fe in Ce_2Fe_{17} lead to significant changes of the magnetic properties. In order to determine the role of the volume, we made ac susceptibility and thermal expansion measurements on the polycrystalline compounds $Ce_2Fe_{16.65}Mn_{0.35}$ and $Ce_2Fe_{16}Mn_1$ under hydrostatic pressure up to 10 kbar in the temperature range 10−300 K. The substitution of Mn for Fe leads to significant narrowing of the range of stability of the low-temperature ferromagnetic phase characterized by $\Theta_T = 48$ K ($x = 0.35$) in comparison with binary Ce_2Fe_{17}, where $\Theta_T = 94$ K. Applying a relatively small pressure of 1.5 kbar leads to suppression of both the ferromagnetic order and large positive spontaneous volume magnetostriction in $Ce_2Fe_{16.5}Mn_{0.35}$. The pressure effect on Θ_T is significant, $d\Theta_T/dp = -30$ K/kbar. The effect of pressure on the Néel temperature is moderate and slightly increases with increasing Mn content ($dT_N/dp = -2.2$ K/kbar for $Ce_2Fe_{16.65}Mn_{0.35}$). No ferromagnetic order was observed in $Ce_2Fe_{16}Mn_1$ at ambient pressure, but an unexpected reentering increase of the ac susceptibility was detected at pressures higher than 1.5 kbar below 50 K. It indicates the existence of a pressure-induced ferromagnetic phase at low temperature. The results show not only a strong correlation between the magnetic properties and the lattice volume but also a significant role of the Mn content.

Introduction

Ce_2Fe_{17} crystallizes in a rhombohedral Th_2Zn_{17} type of crystal structure. A number of unusual features for the set of R_2Fe_{17} compounds (R = rare earth element) can be singled out. Among them is an about 4% smaller lattice cell volume of Ce_2Fe_{17} than the regular cell volume of R_2Fe_{17} compounds extrapolated from its dependence on the rare earth elements. It has been established that cerium ions are in an intermediate valence state Ce^{3+}–Ce^{4+} [1] in this compound and that they have the smallest radius among the lanthanides.

Contrary to the majority of the R_2Fe_{17} compounds, the behavior of Ce_2Fe_{17} is peculiar below the ordering temperature. A susceptibility peak was observed at $T_N = 206$ K and characterizes a Néel temperature. Below $\Theta_T = 94$ K, Ce_2Fe_{17} exhibits spontaneous magnetization and anomalous thermal expansion. A noncollinear ferromagnetic arrangement (a fan structure parallel to the basal plane) and a helimagnetic structure have been determined by neutron diffraction below Θ_T and in the interval $\Theta_T < T < T_N$, respectively. These peculiarities were explained in the model of localized moments as being the result of a competition of positive and negative exchange interactions between iron atoms at different interatomic distances. Due to the small interatomic Fe−Fe distances in Ce_2Fe_{17}, the contribution of the negative exchange interactions becomes larger. Consequently, a noncollinear ferromagnetic or antiferromagnetic arrangement of the magnetic moments of the Fe atoms appears [2].

A giant negative pressure effect on Θ_T has been observed: $d\Theta_T/dP = -(38 \pm 2)$ K/kbar. The ferromagnetic state is suppressed by a critical pressure of $P_c = 2.5$ kbar. The pressure effect on the Néel temperature $T_N = 206$ K is significantly smaller: $dT_N/dP = -(1.7 \pm 0.2)$ K/kbar [3]. The large positive spontaneous volume magnetostriction observed below T_N is suppressed by application of high pressure and disappears at $P > P_c$. The pressure effect on the magnetic ordering was confirmed by neutron diffraction experiments [4].

The substitution of Mn for Fe leads to small changes of the lattice parameters but to significant

changes of the magnetic properties of the Ce_2Fe_{17} compound. The ferromagnetic phase disappears in $Ce_2Fe_{17-x}Mn_x$ compounds for $x \geq 0.5$, but a new, probably noncollinear ferromagnetic phase appears again for $x > 1$ [5]. To characterize the role of the volume in more detail, we performed high-hydrostatic-pressure studies (up to 10 kbar) of the ac susceptibility and thermal expansion of polycrystalline $Ce_2Fe_{16.65}Mn_{0.35}$ and $Ce_2Fe_{16}Mn_1$ compounds in the temperature range 10–300 K.

Experimental

The polycrystalline samples of $Ce_2Fe_{16.65}Mn_{0.35}$ and $Ce_2Fe_{16}Mn_1$ were prepared by the induction melting method with subsequent annealing at 1000 °C for two weeks. The samples were tested by x-ray diffraction. The lattice parameters $a = 8.493$ Å, $c = 12.411$ Å and $a = 8.505$ Å, $c = 12.428$ Å of $Ce_2Fe_{16.65}Mn_{0.35}$ and $Ce_2Fe_{16}Mn_1$, respectively, were determined [6]. The ac-susceptibility and thermal expansion measurements were carried out in the temperature range 12–300 K in a Closed Cycle Refrigerator Cryostat (Cryophysics Ltd.) under hydrostatic pressure up to 10 kbar using a standard Cu–Be piston-cylinder cell. Measurements of the ac susceptibility were performed by the transformer method. The thermal expansion was measured by micro-strain gauges (Micro-Measurements Inc., SK-350).

Results

The temperature dependences of the ac susceptibility χ and thermal expansion $\Delta L/L$ of the $Ce_2Fe_{16.65}Mn_{0.35}$ compound at different pressures are visualized in Fig. 1.

At ambient pressure (Figs. 1,a, 2,a — 0 kbar curves) the drop of the susceptibility and thermal expansion around 50 K corresponds to a magnetic transition from the ferromagnetic to antiferromagnetic state (similar behavior of both the ac susceptibility and thermal expansion were observed in binary Ce_2Fe_{17} around 95 K). Both the small susceptibility peak and the change of the character of the temperature dependence $\Delta L/L$ around 200 K correspond to a magnetic transition from an antiferromagnetic to a paramagnetic state. The transition temperature $\Theta_T = 48$ K was determined as an inflection point of the $\chi(T)$ curve and the Néel temperature $T_N = 211$ K was defined as the maximum of $\chi(T)$. The thermal expansion behavior is rather complex. Anomalous positive spontaneous magnetostriction was observed below 100 K. The thermal expansion coefficient is negative below T_N. It
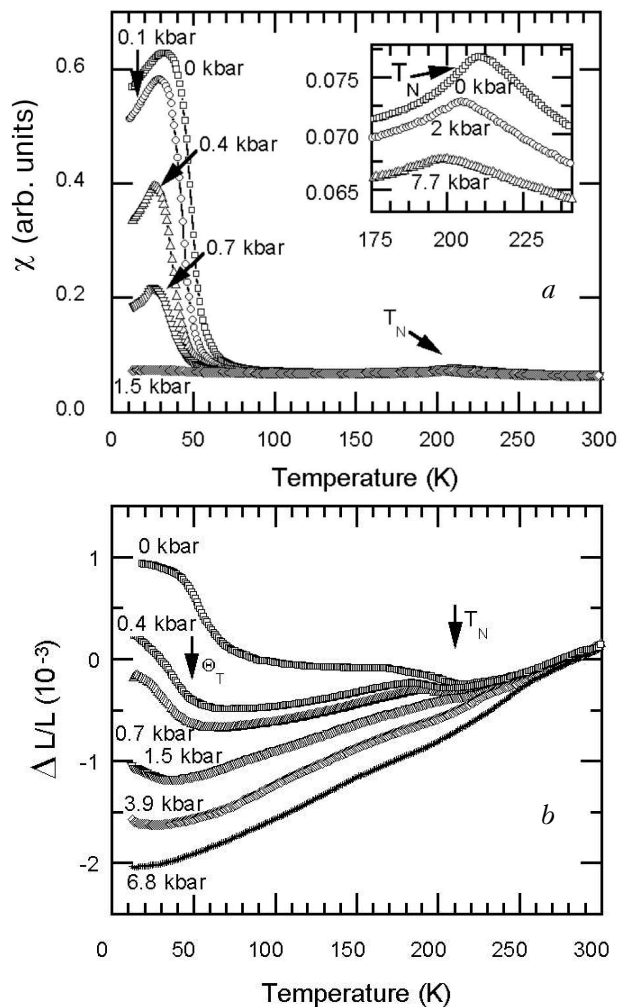

Fig. 1. Temperature dependence of the ac susceptibility (*a*) and thermal expansion (*b*) of $Ce_2Fe_{16.65}Mn_{0.35}$ under pressure.

equals zero at T_N and becomes positive at high temperatures.

Applying pressure leads to a steep decrease of the magnetic susceptibility and transition temperature Θ_T, $d\Theta_T/dP = -30$ K/kbar. At a critical pressure $P_c > 1.5$ kbar the ferromagnetic phase disappears. The shift of Néel temperature T_N with pressure (see the inset in Fig. 1,*a*) is significantly smaller, $dT_N/dP = -2.2$ K/kbar, compared to the Θ_T decrease. The thermal expansion anomalies in the vicinity of Θ_T and T_N (Fig. 1,*b*) are strongly suppressed by pressure and practically disappear at pressures $P > 1.5$ kbar and $P > 4$ kbar, respectively.

We did not observe any presence of low temperatures anomalies (e.g., a ferromagnetic phase) in the temperature dependence of the ac susceptibility of $Ce_2Fe_{16}Mn_1$ at ambient conditions (Fig. 2,*a*, 0 kbar curve).

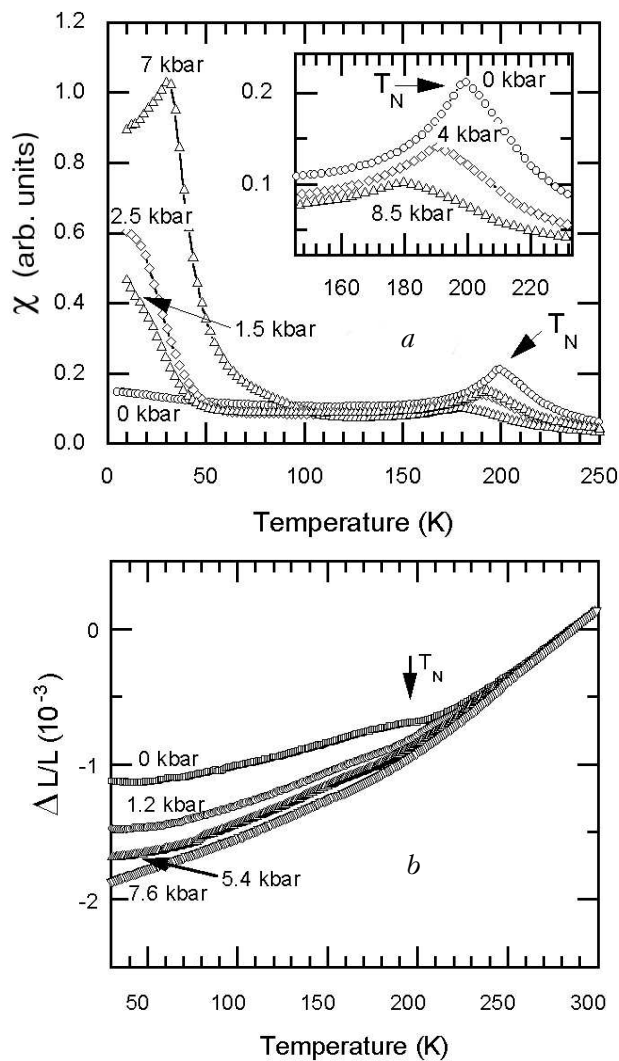A pronounced susceptibility peak was observed around 200 K. It should correspond to the transi-

Fig. 2. Temperature dependence of ac susceptibility (*a*) and thermal expansion (*b*) of $Ce_2Fe_{16}Mn_1$ under pressure.

Fig. 3. *P–T* magnetic phase diagrams of the compounds $Ce_2Fe_{16.65}Mn_{0.35}$ (*a*) and $Ce_2Fe_{16}Mn_1$ (*b*) (FM — ferromagnetic phase, AFM — antiferromagnetic phase, PM — paramagnetic phase).

tion from an antiferromagnetic to a paramagnetic state. The temperature of this transition $T_N = 196$ K was defined as the maximum of the $\chi(T)$ curve. Positive spontaneous magnetostriction was observed below T_N as an appreciable change of the character of $\Delta L/L$ dependence (Fig. 2,*b*, 0 kbar curve). However, the thermal expansion coefficient is positive in the whole temperature range.

Applying pressure, as in previous cases, results in a decrease of the ac susceptibility (inset in Fig. 2,*a*) and Néel temperature, $dT_N/dP = -2.3$ K/kbar. The thermal expansion anomaly below T_N (Fig. 2,*b*) decreases under pressure and practically disappears at $P > 1.2$ kbar. However, at pressures $P \geq 1.5$ kbar an unexpected increase of the susceptibility was observed at temperatures below 50 K (Fig. 2,*a*). This increase is not accompanied by any anomalies in the thermal expansion.
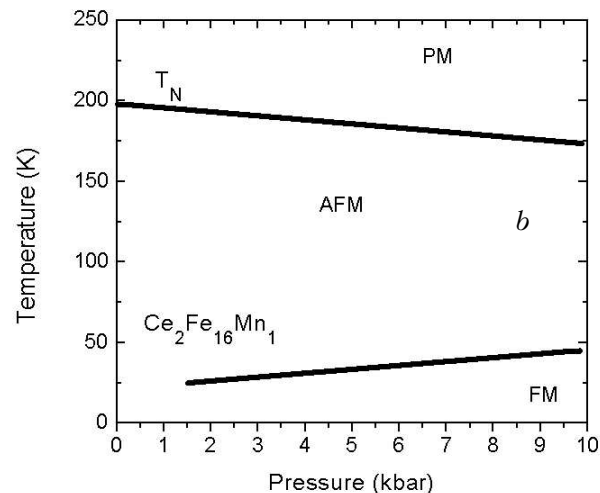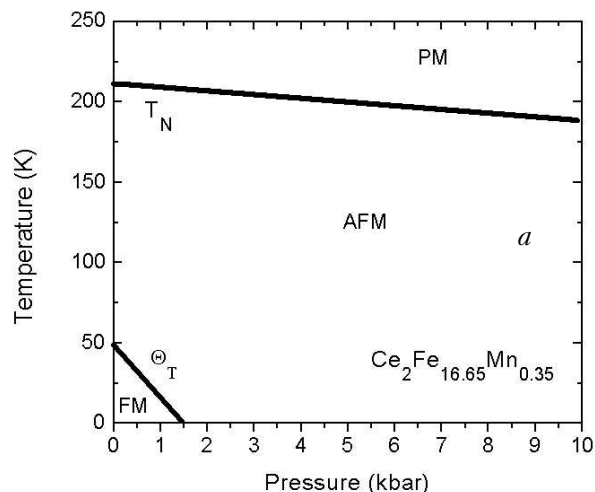
Using all available data (Table) the magnetic phase *P–T* diagrams of $Ce_2Fe_{16.65}Mn_{0.35}$ and $Ce_2Fe_{16}Mn_1$ compounds were created (Fig. 3).

Table

Transition temperatures and their pressure derivatives of $Ce_2Fe_{17-x}Mn_x$ compounds

$Ce_2Fe_{17-x}Mn_x$	Θ_T, K	$d\Theta_T/dP$, K/kbar	P_c, kbar	T_N, K	dT_N/dP, K/kbar
$x = 0$	94	−38	2.5	206	−1.7
$x = 0.35$	48	−30	1.5	211	−2.2
$x = 1$	—	—	—	196	−2.3

Conclusion

According to [7,8], the changes of the lattice parameter c due to the substitution of nonmagnetic elements (e.g., Al, Ga) for Fe seem to have a large influence on the magnetic properties of Ce_2Fe_{17}. In our case the substitution of Mn for Fe in Ce_2Fe_{17} leads not only to nonmonotonic changes of the lattice parameters but also to the appearance of Mn–Mn and Fe–Mn exchange interactions at different sites. As a result, a decrease of the transition temperature Θ_T in $Ce_2Fe_{17-x}Mn_x$ with the subsequent disappearance of the ferromagnetic state for $0.5 \leq x \leq 1$ and its reappearance for $x > 1$ was observed [5,6].

The appearance of the new, probably noncollinear ferromagnetic phase at low temperatures at pressures $P \geq 1.5$ kbar in $Ce_2Fe_{16}Mn_1$ compound was a rather unexpected result. Pressure usually destabilizes the ferromagnetic order, but in our case it leads to an increase of the transition temperature with increase of pressure. This behavior can hardly be explained using competition between positive and negative exchange interactions, as both the Fe–Fe and the Mn–Mn exchange interactions exhibit the same character of the distance dependence.

As the $Ce_2Fe_{16}Fe_1$ has the largest c lattice parameter among the set of $Ce_2Fe_{17-x}Mn_x$ compounds with $x < 2.5$ [6], one can suggest that the new pressure-induced phase in $Ce_2Fe_{16}Mn_1$ could be the same phase as the above-mentioned low-temperature phase in $Ce_2Fe_{17-x}Mn_x$ for $x > 1$. Its creation can be caused by the decrease of the c lattice parameter under pressure.

We can conclude that the observed complex magnetic behavior of the $Ce_2Fe_{17-x}Mn_x$ compounds under high pressures is the consequence of both the decrease of the lattice volume and the role of the Mn content.

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