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Enhancement of the magnetocaloric effect in the $Lu_2Fe_{17-x}Mn_x$ system

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ABSTRACT

Non-monotonic variation of the magnetocaloric effect (MCE) with a maximal value for the composition $x = 0.7$ has been observed in the $Lu_2Fe_{17-x}Mn_x$ system by means of isothermal magnetic measurements. The $Lu_2Fe_{17-x}Mn_x$ compounds with $x = 0$–0.5 are ferromagnets at low temperatures and antiferromagnets at high temperatures, with a near-constant Néel temperature $T_N \sim 278$ K. The temperature of the "ferromagnet–antiferromagnet" phase transition, Θ_T, increases rapidly with increasing content of Mn, and the compounds from the range $x = 0.7$–2 are ferromagnets only, with the practically unchanged Curie temperature, $T_C(0.7) = 287$ K. Thus, first- and second-order magnetic transitions merge for the composition $Lu_2Fe_{16.3}Mn_{0.7}$, where the associated entropy change $|\Delta S_M|$ is 3.6 J/kg K at 300 K in a field of 5 T. This is a way of enhancing the MCE in such multicomponent systems.

1. Introduction

Room-temperature magnetic refrigeration utilizing the magnetocaloric effect (MCE) is currently a topic of growing interest due to refrigeration efficiency, reliability and environmental safety [1–6]. The discovery of a giant MCE due to a magnetic transition in Gd [1], $Gd_5Si_2Ge_2$ [2], $MnFeP_{1-x}As_x$ [3], Ni_2MnGa [4] and $LaFe_{13-x}Si_xH_y$ [5,6] opened up the possibility of magnetic refrigeration near room temperature. The R_2Fe_{17} compounds, where R represents a rare-earth element, have attracted interest as magnetocaloric materials due to their large magnetization, magnetic ordering temperature close to 300 K, low cost of their principal component, easy fabrication and there being no hysteresis problem in a magnetic field [7,8]. The peak entropy change is modest in the R_2Fe_{17} binaries [9,10], but it can be increased by means of compositional fine tuning involving small amounts of other elements [10–13]. The temperature interval over which a substantial entropy change occurs is significant, approaching 150 K, an important criterion for improving the overall effectiveness of such materials for magnetic refrigeration [9]. Large MCE in $R_2Fe_{17-x}Co_x$, $Ce_{2-x}R_xFe_{17}$, $Pr_{10+x}Fe_{90-x}$ and other series has been demonstrated [11–13]. In $Pr_{1.5}Ce_{0.5}Fe_{17}$, the MCE is about one-half that of Gd [12].

In this paper, a study of the MCE in $Lu_2Fe_{17-x}Mn_x$ ($x = 0$–2) compounds is presented. The compounds exhibit a magnetic phase transition close to room temperature and thus they are promising for applications [14].

2. Experimental details

The Lu_2Fe_{17-x} Mn_x compounds, $x = 0$, 0.5, 0.7, 1, 2 were prepared by argon arc melting from starting materials. The ingots were homogenized at 1450 K for 9 h and then quenched in water. The X-ray diffraction measurements were carried out with a DRON diffractometer using Fe Kα radiation. According to the X-ray analysis, all the $Lu_2Fe_{17-x}Mn_x$ compounds have a hexagonal crystal structure of the Th_2Ni_{17}-type. Some amount of α-Fe has been detected in the compound with $x = 1$. The temperature dependences of magnetization $M(T)$ were measured for the polycrystalline samples in a field of 0.01 T in the temperature range 2–400 K. The temperatures of the magnetic phase transitions were determined from the magnetization $M(T)$ dependences: T_N – as a maximum, T_C and Θ_T – from the kink on $M(T)$. The $M(H)$ dependences were measured by a SQUID device on free powder samples in fields up to 5 T at various temperatures in the vicinity of the transition temperatures. The saturation magnetization M_{sat} was determined by extrapolation of the experimental dependences $M(1/H)$ to $1/H \to 0$.

3. Experimental results

The values of T_C and M_{sat} for the $Lu_2Fe_{17-x}Mn_x$ compounds, $x = 0$, 0.5, 0.7, 1, 2, as obtained in [14,15] and then confirmed in this paper, are listed in Table 1. The $Lu_2Fe_{17-x}Mn_x$ compounds with $x = 0$–0.5 are ferromagnets at low temperatures and antiferromagnets at high temperatures. The temperature of the "ferromagnet–antiferromagnet" phase transition, Θ_T, increases quickly from 135 to 253 K with growing content of Mn, whereas the Néel temperature remains practically unchanged ($T_N \sim 278$ K)

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Table 1
Transitions temperatures, saturation magnetization at 2 K [14,15] and magnetic entropy change in 5 T field of $Lu_2Fe_{17-x}Mn_x$.

x	Θ_T (K)	T_N (K)	T_C (K)	M_{sat} (μ_B)	$-\Delta S_M$ (J/kg K)
0.0	153	277	-	33.9	2.8
0.5	249	276	-	31.8	3.4
0.7	-	-	287	30.6	3.6
1.0	-	-	301	25.5	2.7
2.0	-	-	292	26.8	2.5

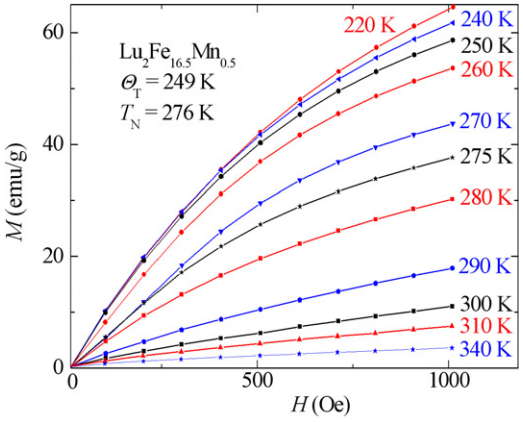

Fig. 1. (Colour on-line) Magnetization isotherms for the free powder samples of $Lu_2Fe_{16.5}Mn_{0.5}$ at $T = 220$–350 K.

and is close to $T_C = 287$ K for the compound with $x = 0.7$. The compounds from the range $x = 0.7$–2 are ferromagnets only, with a near-constant $T_C(x)$ value.

The magnetization $M(H)$ curves of the Lu_2Fe_{17} and $Lu_2Fe_{16.5}Mn_{0.5}$ crystals or powder samples at different $T < T_N$ and in magnetic fields up to 5 T have been presented in [14,16]. The magnetization isotherms of $Lu_2Fe_{16.5}Mn_{0.5}$ and $Lu_2Fe_{16.3}Mn_{0.7}$ at several temperatures between 220 and 350 K in the vicinity of the magnetic phase transitions and in external fields up to 1 T are shown in Figs. 1 and 2. The $M(H)$ dependences for other samples studied are similar and are not shown here. All curves have a shape typical of ferromagnets at $T < \Theta_T$ ($Lu_2Fe_{16.5}Mn_{0.5}$) or $T < T_C$ ($Lu_2Fe_{16.3}Mn_{0.7}$). The value of the spontaneous magnetic moment decreases monotonically with increasing temperature. The field-induced magnetic phase transition from a helical antiferromagnet to a ferromagnet occurs in the basal plane of the $Lu_2Fe_{17-x}Mn_x$ compounds at $\Theta_T < T < T_N$. The analogous effect for Lu_2Fe_{17} was measured in [14–19]. The critical field H_{cr} of this metamagnetic transition is maximal just below T_N. In the case of the single-crystalline Lu_2Fe_{17} and $Lu_2Fe_{16.5}Mn_{0.5}$ compounds, the values of $H_{cr} = 0.23$ T at $T = 250$ K and $H_{cr} = 0.02$ T at $T = 272$ K, respectively, were obtained in [16]. The metamagnetic transition is clearly observed on powdered samples of $Lu_2Fe_{16.5}Mn_{0.5}$ in the temperature range $T = 260$–275 K in Fig. 1. There are no metamagnetic transitions in the ferromagnetic composition $Lu_2Fe_{16.3}Mn_{0.7}$ in Fig. 2.

The isothermal magnetic entropy change ΔS_M can be obtained from the Maxwell relation [1]:

$$\Delta S_M(T, H) = \int_0^H (\delta M/\delta T)_H \, \mathrm{d}H. \quad (1)$$

Since $\delta M/\delta T$ peaks at the magnetic ordering temperature, a large MCE is expected close to the magnetic phase transition. Figs. 3–5 illustrate the magnetic entropy change for the $Lu_2Fe_{17-x}Mn_x$ compounds as a function of temperature and external field. For the $x = 0$ and 0.5 compounds, $|\Delta S_M(T)|$ is found to

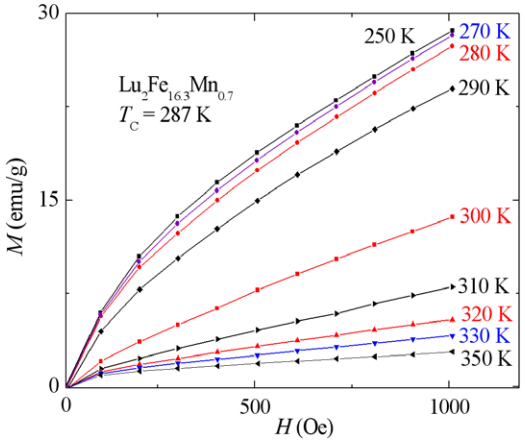

Fig. 2. (Colour on-line) Magnetization isotherms for the free powder samples of $Lu_2Fe_{16.3}Mn_{0.7}$ at $T = 250$–350 K.

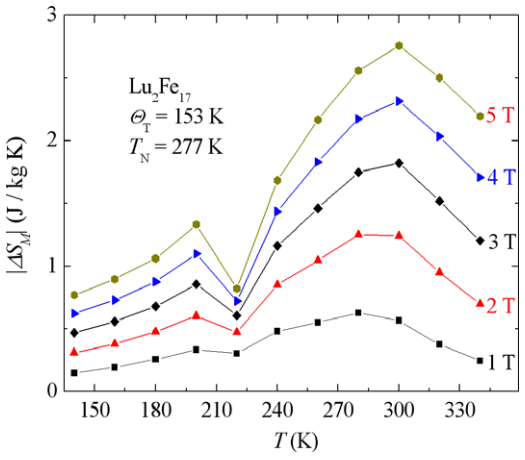

Fig. 3. (Colour on-line) Temperature dependence of magnetic entropy change $|\Delta S_M(T)|$ for Lu_2Fe_{17} under different external fields.

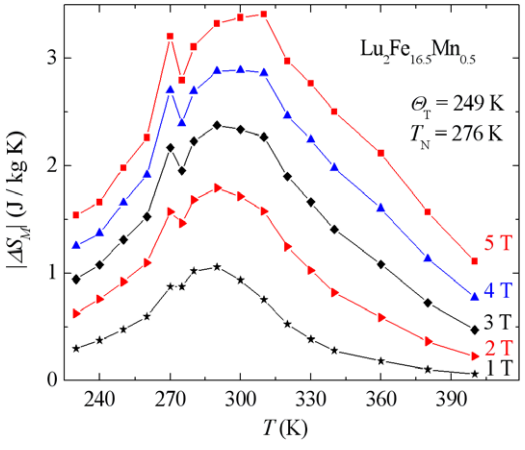

Fig. 4. (Colour on-line) Temperature dependence of magnetic entropy change $|\Delta S_M(T)|$ for $Lu_2Fe_{16.5}Mn_{0.5}$ under different external fields.

exhibit two peaks around approximately Θ_T and T_N (Figs. 3 and 4). These transitions display about the same entropy change for the $x = 0.5$ compound, but in the case of the binary compound a larger peak of $|\Delta S_M(T)|$ accompanies the higher-temperature transition at T_N. There is a single $|\Delta S_M(T)|$ peak near T_C for the ferromagnets with $x = 0.7$, 1, 2 (Fig. 5). The MCE depends strongly on the type of magnetic phase transition and the value of applied magnetic field, particularly, when we deal with antiferromagnetic materials.