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Letter

Large magnetocaloric effect in re-entrant ferromagnet $PrMn_{1.4}Fe_{0.6}Ge_2$

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ABSTRACT

Magnetocaloric effects (MCE) at multiple magnetic phase transition temperatures in $PrMn_{1.4}Fe_{0.6}Ge_2$ were investigated by heat capacity and magnetization measurements. $PrMn_{1.4}Fe_{0.6}Ge_2$ is of a re-entrant ferromagnet and performs multiple magnetic phase transitions in the temperature range from 5 to 340 K. A large magnetic entropy change ($-\Delta S_M$) 8.2 J/kg K and adiabatic temperature change (ΔT_{ad}) 4.8 K are observed for a field change of 0–1.5 T around 25.5 K, associated with the field-induced first order magnetic phase transition (FOMT) from the antiferromagnetic to the ferromagnetic state with an additional Pr magnetic contribution. These results suggest that a re-entrant ferromagnet is probably promising candidate as working material in the hydrogen and nature gas liquefaction temperature range magnetic refrigeration technology.

1. Introduction

Cooling technology based on the magnetocaloric effect is considered extremely beneficial for the future in helping to address the two key issues of energy shortages and global warming. As such, it is highly desirable to explore new materials which offer prospects for a giant magnetocaloric effect (GMCE) especially under low applying field. The GMCE is generally associated with a first-order magnetic transition (FOMT) because of the large difference in magnetization between two adjacent magnetic phases. Thus, Pecharsky and Gschneidner discovered a GMCE in $Gd_5Ge_2Si_2$ originating from a first-order field induced structural and magnetic transition in this compound at 276 K [1] (see also [2,3]) with other GMCE ferromagnetic materials such as $MnFeP_{0.45}As_{0.55}$ [4], MnAs [5], and $La(FeSi)_{13}$ [6], subsequently having been discovered. These materials display a GMCE due to the first-order magnetic disorder-to-order transition from the paramagnetic to the ferromagnetic state near room temperature. There are also studies of other properties which are important for fundamental mechanism understanding and practice application [7–9], and there are also studies of paramagnetic salts, garnets, and molecular clusters which can be utilized for low temperature cooling purposes [10].

Ternary rare-earth (R) compounds of the type RT_2X_2, where T = transition metal, X = Si, Ge, exhibit a large variety of structural and physical properties and have been studied extensively over the years. The interesting phenomenon of re-entrant ferromagnetism has been found in several of these RT_2X_2 compounds by controlling the interplay between the R–T and T–T exchange interactions through elemental substitution [11–13]. While there are several reports on MCE in RT_2X_2 [14,15], few reports [16] exist in the literature on an MCE associated with re-entrant ferromagnetic transitions. This may be due to the fact that most re-entrant phase transitions are order-to-order phase transitions between antiferromagnetic (AFM) and ferromagnetic (FM) states, and these kinds of transitions are not expected to show a very high MCE. The magnetic structures and properties of $PrMn_{2-x}Fe_xGe_2$ have been studied recently over the entire Mn and Fe concentration ranges e.g. [13] We have investigated the magnetic phase transitions and determined magnetic phase diagrams in last paper [11], in this paper, interest is focussed on the magnetocaloric properties of the re-entrant ferromagnet $PrMn_{1.4}Fe_{0.6}Ge_2$. It has been found that a first-order temperature-induced and field-induced order (AFmc) to order (Fmc + F(Pr)) magnetic phase transition around 25.5 K can induce giant MCE with low magnetic and thermal hysteresis. At the same time, the MCE in the whole experimental temperature range from 5 to 340 K has been observed in this re-entrant ferromagnet.

2. Experimental

The polycrystalline $PrMn_{1.4}Fe_{0.6}Ge_2$ compound was prepared by arc melting the high purity elements on a water-cooled Cu hearth under purified argon gas. The microstructure characterisation and results was reported in Refs. [11,13]. The magnetization and heat capacity were measured under applied magnetic field of 0–5.0 T over the temperature range 5–300 K. The experiments were carried out on a physical properties measurement system (PPMS–Quantum Design). The isothermal entropy change, corresponding to a magnetic field change ΔH starting from zero field, was derived from the magnetization data by means of the following expression

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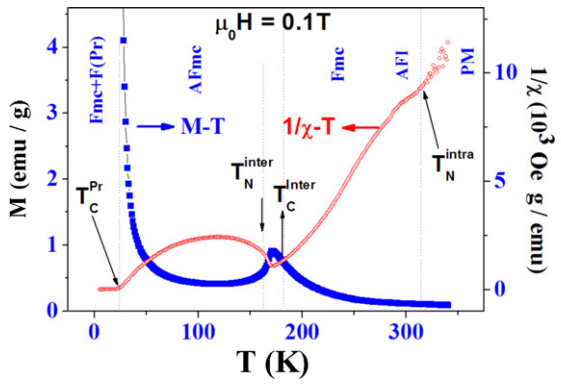

Fig. 1. Magnetic behaviours of $PrMn_{1.4}Fe_{0.6}Ge_2$, M–T and $1/\chi$–T curves.

which can be obtained from the Maxwell relation:

$$\Delta S_M(T,H) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H \partial H \quad (1)$$

3. Results and discussion

The temperature dependences of the $PrMn_{1.4}Fe_{0.6}Ge_2$ magnetization on cooling and heating from 5 to 340 K in magnetic fields 0.1 T are presented in Fig. 1, the $1/\chi$–T curve is presented as well. As shown in Fig. 1, the M–T and $1/\chi$–T curves of $PrMn_{1.4}Fe_{0.6}Ge_2$ reveal the presence of four magnetic transitions, similar to the case of $SmMn_2Ge_2$ [17–19]. Based on the magnetic structures of $PrMn_2Ge_2$ [20,21] and the similarity in magnetic behaviour of $PrMn_{1.4}Fe_{0.6}Ge_2$ to that of $SmMn_2Ge_2$, it can be seen that with decreasing temperature from 340 K, $PrMn_{1.4}Fe_{0.6}Ge_2$ exhibits a transition from paramagnetism (PM) to an *ab*-plane antiferromagnetic state (AFI) [13] at Néel temperature, $T_N^{intra} \approx 333$ K. Around Curie temperature, $T_C^{inter} \approx 168$ K, this AFI structure gives way to a canted ferromagnetic structure Fmc-type. With further decrease in temperature, a canted antiferromagnetic structure (AFmc) state occurs around $T_N \approx 157$ K and, finally, there is a transition to an Fmc state plus an additional Pr-sublattice magnetic contribution (Fmc + F(Pr)) at $T_C^{Pr} \approx 25.5$ K. Both $PrMn_{1.4}Fe_{0.6}Ge_2$ and $SmMn_2Ge_2$ exhibit similar magnetic behaviour [13] even though the values of the room temperature *a*-parameters differ by ~1% ($a = 0.4088$ nm and $a \sim 0.4045$ nm [19] respectively). This common magnetic behaviour for disparate values of the *a*-parameter may be related to electronic effects as suggested in the case of $NdMn_{1.6}Fe_{0.4}Ge_2$ for which the ferromagnetic to antiferromagnetic transition takes place at a larger d_{Mn-Mn} distance than in the pure $NdMn_2Ge_2$ compound [12].

It is well known that in RMn_2Ge_2 compounds, even slight variations in the unit cell parameters due to external factors, such as pressure, field, temperature or chemical substitution, are sufficient to modify the interlayer Mn–Mn spacing, leading to multiple magnetic phase transitions (e.g. Refs. [16,19]). Such transitions are likely to be accompanied by an anomaly in the thermal expansion (magneto-volume effect) and can therefore be controlled by application of pressure or field. It has been reported that the width of the lower temperature antiferromagnetic phase in $SmMn_2Ge_2$ can be extended by applied hydrostatic pressure [16]. The sharp transition of AFmc to Fmc + F(Pr) at low field implies the unexpected large magnetocaloric effect as outlined in the following analysis. All these also imply that the type of transitions are likely to be 1st order [11] and to exist thermal and magnetic hysteresis, but as our previous reported [11], the thermal hysteresis at $T_C^{Pr} \simeq 25.5$ K was about 4.5 K and magnetic hysteresis is small as shown in Fig. 2(a) of our previous reports Ref. [11].

Fig. 2(a) shows the magnetization curves between 20 K and 60 K measured at 2 K intervals and the M–H verves over the range 120–200 K (5 K intervals) shown in Fig. 2(b). It is clear that above $T_C^{Pr} \approx 25.5$ K and below $T_N^{inter} \approx 157$ K, while the magnetization increases slowly with magnetic field in the low-field range and before increasing sharply at a critical field, the magnetization is not saturated at 5 T between 25.5 K and 157 K. The step in the magnetization curves indicates a field-induced AFM to FM or FM to AFM phase transition, which implies the first order nature of those transitions. It has been reported [22] that the order of a magnetic transition is related to the sign of the Landau coefficient factors. A transition is expected to be first order when the factor is negative, whereas it will be second-order for a positive value. The Arrott plots of H/M versus M^2 at 2 K intervals between 20 K and 60 K and at selected temperatures between 120 K and 200 K are shown in Ref. [11]. It has been determined that the magnetic transitions from AFI to Fmc above 168 K, from Fmc to AFmc below 157 K, and from AFmc to Fmc + F(Pr) below 25.5 K are first order transitions in applied magnetic field.

The temperature dependence of the isothermal magnetic entropy change $-\Delta S_M(T, H)$ calculated from Eq. (1) for different magnetic-field changes around the three first order magnetic phase transition temperatures – 25.5 K, 157 K, and 168 K – are presented in Fig. 3(a) and (b). The values of $-\Delta S_M^{max}$ at 25.5 K are 8.2 and 11.8 J kg^{-1} K^{-1} for magnetic-field changes from 0 to 1.5 and 5 T, respectively. These values are larger than those reported for Gd (5.1 J kg^{-1} K^{-1} for 2 T) and comparable with $Gd_5(Si_2Ge_2)$ [2]. The giant MCE value $-\Delta S_M^{max} = 8.2$ J kg^{-1} K^{-1} obtained for the relatively small field change from 0 to 1.5 T is very beneficial for applications. The maxima of $-\Delta S_M^{max}$ at 25.5 K for the different magnetic-field

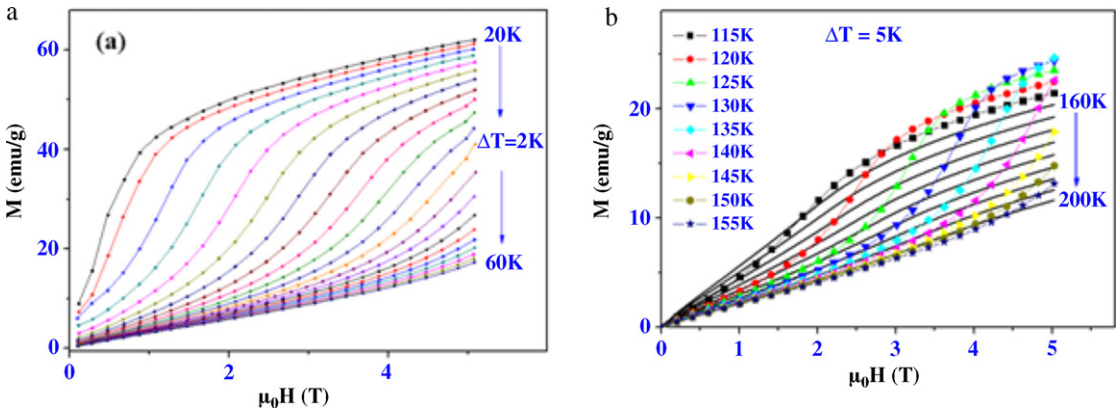

Fig. 2. Magnetization curves (a) at 2 K intervals between 20 and 60 K and (b) at 5 K intervals between 120 and 200 K.

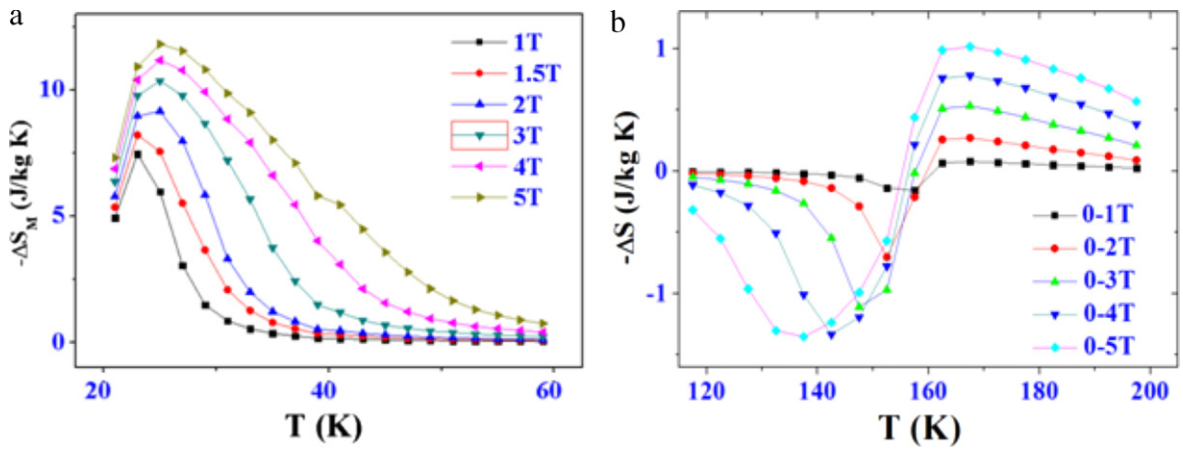

Fig. 3. Temperature dependence of the isothermal magnetic entropy change $-\Delta S_M(T, H)$ around three 1st order magnetic phase transition temperatures: (a) T_C^{Pr}; and (b) T_N^{inter} and T_C^{inter}.

changes correspond to the temperature and field induced first order AFmc to Fmc phase transition.

The curves of $-\Delta S_M$ versus T shown in Fig. 3(b), provide valuable information about the nature of the two other magnetic ordering transitions in $PrMn_{1.4}Fe_{0.6}Ge_2$. Around $T_N^{inter} \approx 157$ K, $-\Delta S_M$ is negative (inverse MCE) corresponding to the magnetic transition from the Fmc to the AFmc state, but it changes to positive values with increasing temperature around $T_C \approx 168$ K, corresponding to the magnetic transition from the AFl to the Fmc state (Fig. 1). An inverse MCE has been often observed in systems displaying first-order magnetic transitions and the origin is the same for all of them. Due to the presence of mixed exchange interactions, the applied magnetic field leads to a further spin-disordered state near the transition temperature, increasing the configurationally entropy [9]. As shown in Fig. 3(b), the $-\Delta S_M^{max}$ peak position moves to the low temperature side with increasing magnetic field. This behaviour, which is commonly observed in field-induced first order phase transition materials [23,24], is due to the large shift of transition critical field to high field with decreasing temperature around T_N^{inter} for the Fmc to AFmc transition.

In order to confirm the observed large MCE in this compound, we performed the heat capacity measurements in the fields of $H = 0$, 1.5 T, and 5 T (Fig. 4(a)). Obvious thermal anomalies at temperature ranges 15 K to 55 K, 120 K to 160 K corresponds to the magnetic transitions. The effect of magnetic field on the heat capacity of $PrMn_{1.4}Fe_{0.6}Ge_2$ is quite pronounced. These are more clearly seen in C_p/T–T curves (Fig. 4(b)). From the second law of thermodynamics, we know that the entropy (at constant pressure) of a system is related to the heat capacity as $(\delta S/\delta T)_{Hi} = C_p(T)_{Hi}/T$. The isothermal magnetic entropy change ΔS_{HC} can be calculated from the heat capacity

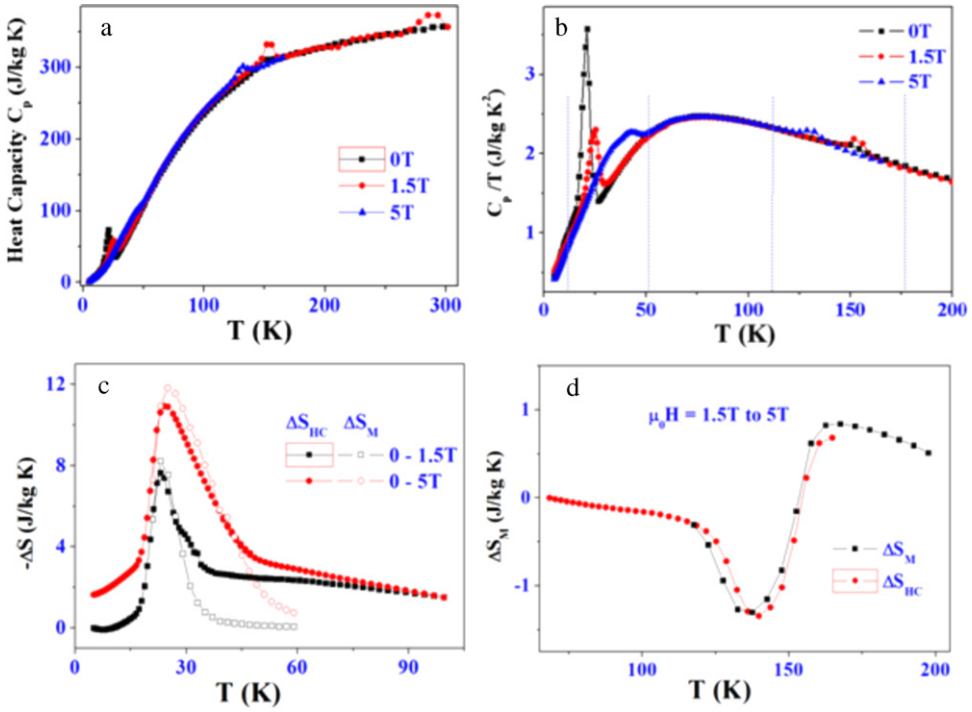

Fig. 4. (a) Heat capacity C_p of $PrMn_{1.4}Fe_{0.6}Ge_2$ measured under the fields of $H = 0$, 1.5 T, and 5 T; (b) Specific heat C_p/T–T curves of $PrMn_{1.4}Fe_{0.6}Ge_2$ under the fields of $H = 0$, 1.5 T, and 5 T. Comparison of isothermal magnetic entropy change $-\Delta S_M$ (obtained from magnetization data) with $-\Delta S_{HC}$ (obtained from heat capacity data) (c) at temperature around 25.5 K under field from 0 T to 1.5 T and from 0 to 5 T, and (d) at temperature from 120 K to 200 K under field from 1.5 T to 5 T.

Table 1
Maximum $-\Delta S_M$ (J/kg K) and ΔT_{ad} (K) values as well as RCP (J/kg) under 1.5 T, transition temperature T_m and transition types around 20 K for $PrMn_{1.4}Fe_{0.6}Ge_2$ and various giant MCE materials.

Material	$-\Delta S_M$	ΔT_{ad}	RCP (J/kg)	T_{tt} (K)	Transition order	Reference
$PrMn_{1.4}Fe_{0.6}Ge_2$	8.2	4.8	110	25.5	AFM-FM, 1st	Present
HoCuSi	~11	/~	77	8.3	AFM-FM, 1st	[25]
$GdCo_2B_2$	~7.8	4.1	96	25	AFM-FM, 1st	[26]
$HoMnO_3$	1.1	~1.9	/~	5.5	AFM-PM, 1st/2nd	[27]
$ErRu_2Si_2$	7.8	4.2	~80	5.5	AFM-FM, 1st	[15]
Ho_5Pd_2	6	/~	~110	28	AFM-FM, 1st	[15]
$TbCoC_2$	6.8	/~	~82	28	PM-FM, 2nd	[28]
$ErCo_2$	~7	4.2	56	32	PM-FM, 1st	[29]
$Gd_3Fe_5O_{12}$	~2	/~	/~	35	AFM-FM, 2nd	[30]
$La(Fe_{0.88}Si_{0.12})_{13}$	~12	5.5	~110	197	PM-FM, 1st	[6]
MnFePAs	~9	~5	~100	305	PM-FM, 1st	[4]
$Gd_5Si_2Ge_2$	~9	~5	90	278	PM-FM, 1st	[1,2]
Manganites	5.3	2.5	53	260	PM-FM, 1st/2nd	[31]
Pure Gd	~4	4.7	120	294	PM-FM, 2nd	[1,2]

data by

$$\Delta S_{HC}(T,H) = \int_0^T (Cp(T)_{Hi} - Cp(T)_{H0})\frac{\partial T}{T} \quad (2)$$

As shown in Fig. 4(c) and (d), the entropy change ΔS_{HC} exhibits a similar behaviour to ΔS_M (obtained from the Maxwell relation) by around three first order magnetic phase transitions, implying that the two techniques yield consistent results in this system. According to thermodynamics, the adiabatic temperature change (ΔT_{ad}) at an arbitrary temperature T can be expressed as:

$$\Delta T_{ad} = -\Delta S_{HC}(T, H_i) \times \frac{T}{C_p(T, H_0)} \quad (3)$$

where $Cp(T, H_0)$ is the heat capacity at temperature T and under field $H_0 = 0$ T. Based on the formula (3), the ΔT_{ad} at temperature range from 5 K to 60 K are presented in Fig. 5. The maximum ΔT_{ad} at temperature ~25.5 K are about 4.8 K and 7.7 K for magnetic field changes of 0–1.5 and 0–5 T, respectively.

We have noticed the difference between entropy changes ($-\Delta S_M$) obtained from Maxwell equations and entropy changes ($-\Delta S_{HC}$) from C_p measurement especially when the temperature $T > 30$ K as shown in Fig. 3(c), which may due to two different measurements, since the isothermal magnetic measurement only picked up the magnetic moments, so the $-\Delta S_M$ obtained from Maxwell equations is pure magnetic entropy change. However, the heat capacity measurements picked up three parts of contributions (electrons, photons or lattice, and magnetic), normally the electrons contribution is smaller, and lattice and magnetic are the mainly contributions to the $-\Delta S_{HC}$, the magnetic contribution is picked up by magnetic measurement, e.g. the $-\Delta S_M$, therefore, the above mentioned difference mainly due to the magnetic field induced lattice changes contributed to entropy changes.

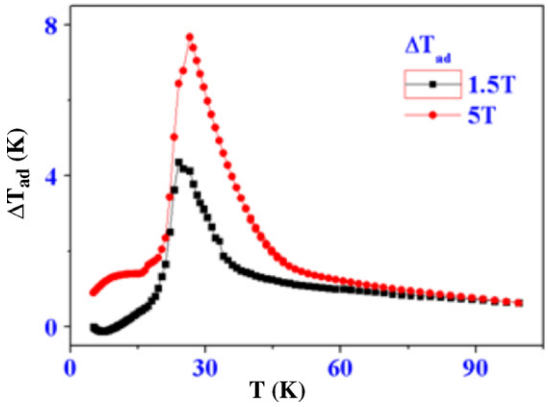

Fig. 5. Temperature dependence of adiabatic temperature rise ΔT_{ad} in $PrMn_{1.4}Fe_{0.6}Ge_2$ generated by a magnetic field change of 1.5 T and 5 T at temperature around 25.5 K.

The $-\Delta S_M$ spans in a temperature range, the full width of half maximum (δT) approaches to ~10 and ~20 K in the magnetic field changes from 0 to 1.5 T and to 5 T, respectively. The relative cooling power (RCP), evaluated by RCP $= -\Delta S_M^{max} \times \delta T$, reaches to ~110 and ~240 J/kg from 0 to 1.5 T and to 5 T, respectively. For comparison, the maximum $-\Delta S_M^{max}$ and ΔT_{ad} (K) values as well as RCP under field from 0 to 1.5 T, the transition temperature T_m and transition types around 20 K and near room temperatures for $PrMn_{1.4}Fe_{0.6}Ge_2$ and various recent discovered best giant MCE (GMCE) materials as shown in Table 1. These values are comparable with and some value even larger than best known GMCE materials near room temperature and around 20 K. Moreover, our observations indicate that systems which exhibit re-entrant magnetism also offer a route for GMCE, and the rare earth elements (Pr) add magnetic contribution accompany with the phase transition may uncover a new mechanism of GMCE, for which some confirmation works are on progressing through neutron diffraction experiments. In addition, this study provides significant opportunities for exploring new GMCE materials, such as the present layered $ThCr_2Si_2$-type structure, by controlling magnetic transitions in re-entrant magnetism through varying external factors, such as pressure, field, temperature, and chemical substitution, to vary the unit-cell parameters and to modify the interlayer Mn–Mn spacing.

4. Summary

To summarize, the unexpected exhibition of a low filed large MCE at around 25.5 K is associated with the field-induced FOMT from the AFmc to the Fmc + F(Pr) state, and both conventional and inverse magnetocaloric effects have been observed in polycrystalline $PrMn_{1.4}Fe_{0.6}Ge_2$. The MCE values are comparable to those reported for the best-known MCE materials. In particular, the MCE value $-\Delta S_{max} = 8.2$ J kg^{-1} K^{-1}, obtained for a small field change from 0 to 1.5 T, is very beneficial for applications. This suggests that $PrMn_{1.4}Fe_{0.6}Ge_2$ may be a promising candidate to be applied in magnetic refrigeration in the low temperature hydrogen or nature gas liquefaction range.

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