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Magnetic and magnetocaloric properties of $Sm_xGd_{1-x}Mn_2Si_2$

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Abstract

The magnetic and magnetocaloric properties of $Gd_{1-x}Sm_xMn_2Si_2$ compounds with $0 \leq x \leq 1$ have been studied to determine their suitability as magnetic refrigerant materials. The rare earth ordering temperatures are found to decrease from 60 K for $x = 0$ to 37 K for $x = 1$. The temperature variations of magnetization under 'field-cooled' and 'zero-field-cooled' conditions are found to differ slightly, indicating a small thermomagnetic irreversibility. Magnetocaloric effect is calculated in terms of isothermal magnetic entropy change (ΔS_M) using the magnetization isotherms. The maximum values of ΔS_M are found to be 5.9 J kg^{-1} K^{-1} in $GdMn_2Si_2$ and 3.4 J kg^{-1} K^{-1} in $SmMn_2Si_2$.

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Keywords: Magnetocaloric effect; Magnetic entropy; Ferrimagnets; Spin fluctuations

1. Introduction

Recently, a lot of attention is being given to the magnetocaloric studies in an effort to identify potential magnetic refrigerants [1–5]. The magnetocaloric effect (MCE) is defined as the heating or cooling of magnetic materials due to the application of a magnetic field. Fundamentally, this effect is similar to the adiabatic demagnetization, which is commonly applied to paramagnetic materials to produce cooling at low temperatures. The success in developing giant MCE materials is expected to simplify the refrigerator design, thereby making them more viable. During the last few years, many rare earth (R)-transition metal (TM) intermetallic compounds have been shown to possess giant MCE [6–9]. Based on the various reports available, it is certain that giant MCE can be obtained only in materials which show first-order transitions, magneto-structural transitions or field-induced metamagnetic transitions [10–12]. In an effort to identify potential refrigerant materials from the reservoir of R-TM intermetallics, many novel materials are being probed to study their MCE behavior. In this context, we find that ternary RMn_2Si_2 compounds which crystallize in the tetragonal $ThCr_2Si_2$-type structure have not been exploited very much till now.

RMn_2Si_2 compounds are characterized by the layered structure obtained by the stacking of atomic planes in the sequence Mn–Si–R–Si–Mn, perpendicular to the *c*-axis [13,14]. In this series, both R and Mn atoms possess magnetic moments. The interlayer and intra layer Mn–Mn exchange interactions are very sensitive to the lattice parameter, leading to ferromagnetic or antiferromagnetic ordering of the Mn sublattice. Since the lattice parameters change with rare earth element, the magnetic ordering within the Mn sublattice is also dependent on the rare earth. It has been reported that in most of the RMn_2Si_2 compounds, the Mn sublattice shows the antiferromagnetic ordering above the room temperature while R sublattice remains disordered. As temperature is reduced, the R moments also order magnetically at T_C^R, below which Mn and R sublattices couple ferromagnetically or antiferromagnetically depending on whether the rare earth is light or heavy. Therefore, the magnetic properties of these compounds are quite interesting and they show exotic magnetic phase transitions [13,14]. However, except $NdMn_2Si_2$ compounds [15], not many reports are available on the magnetocaloric studies on this series. Since MCE is closely related to the nature of the magnetic state, we have studied the magnetic and magnetocaloric properties of polycrystalline $Gd_{1-x}Sm_xMn_2Si_2$ compounds with

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$x = 0$, 0.4, 0.6 and 1 and the results are presented in this paper.

2. Experimental details

Polycrystalline samples of $Gd_{1-x}Sm_xMn_2Si_2$ with $x = 0$, 0.4, 0.6, and 1 were synthesized by arc melting the constituent elements in a water-cooled copper hearth under purified argon atmosphere. The purity of the starting elements was 99.9% for Sm, Gd and 99.99% for Mn and Si. The ingots were remelted four to five times to ensure homogeneity. The as-cast samples were characterized by power X-ray diffraction (XRD) using Cu Kα radiation. The magnetization (M) was measured using a PPMS (Quantum design) in the temperature (T) range of 10–150 K, up to a field (H) of 50 kOe. Thermomagnetic analysis was performed both under zero-field-cooled (ZFC) and field-cooled (FC) modes. In the former case, the samples were cooled in the absence of a field and the magnetization was measured during warming, by applying a nominal field of 200 Oe. In the FC mode, the sample was cooled in presence of a field and the magnetization was measured during warming, under the same field. The magnetocaloric effect has been calculated in terms of isothermal magnetic entropy change, using the magnetization isotherms collected near the magnetic transition temperatures.

3. Results and discussion

The powder XRD patterns show that all the $Gd_{1-x}Sm_xMn_2Si_2$ compounds have formed in single phase with the $ThCr_2Si_2$ structure (space group = $I4/mmm$). The lattice parameters were calculated by refining the XRD data using the Rietveld technique. Fig. 1 shows the Rietveld fitted XRD plots of the samples with $x = 0$, 0.4, 0.6 and 1. As can be seen from the difference plot between the observed and calculated patterns, the samples are single phase and free from any impurities. The structure is identical for all the compounds, except for the changes in the lattice parameters. The variation of lattice parameters, the unitcell volume and the Mn–Mn bond lengths along the a- and c-axis (d^a_{Mn-Mn} and d^c_{Mn-Mn}) with Sm content, is shown in Table 1. The increase in the lattice parameter and unit cell volume is attributed to the larger ionic radius of Sm compared to that of Gd. The fact that the intra later Mn–Mn spacing (d^a_{Mn-Mn}) is less than the critical value of 2.84 Å suggests that there is no Mn moment in the ab plane and also that the inter layer Mn–Mn coupling is antiferromagnetic in all the compounds in the present case [16].

Table 1
Lattice parameters (a and c), unit cell volume (V), Mn–Mn bond lengths (d_{Mn-Mn}) in $Gd_{1-x}Sm_xMn_2Si_2$ compounds

x	a (Å)	c (Å)	V (Å)^3	d^c_{Mn-Mn} (Å)	d^a_{Mn-Mn} (Å)
0	3.948	10.475	163.320	3.294	2.791
0.4	3.958	10.485	164.274	3.289	2.798
0.6	3.961	10.489	164.595	3.281	2.801
1.0	3.973	10.503	165.812	3.279	2.808

Fig. 2 shows the temperature dependence of the ZFC and FC magnetization data for all the compounds, collected in a field of 200 Oe. The sharp decrease in the magnetization seen in these compounds is attributed to the magnetic order–disorder transition occurring at the R sublattice. The corresponding transition temperature is termed as T_C^R and is calculated from the dM/dT versus T plots shown in the inset of Fig. 2. The variation of T_C^R with x is shown in Table 2. It may also be noted from the inset that the sharpness of the transition is more in the compounds with $x = 0$ and 1, as compared to the case of $x = 0.4$ and $x = 0.6$. It can also be seen from Fig. 2 that there is a small difference between the FC and ZFC magnetization data in all the compounds. This thermomagnetic irreversibility may be due to the domain wall pinning effect.

At low temperature the magnetization increases with decreasing temperature and below T_C^R, the alignment of the Gd moments

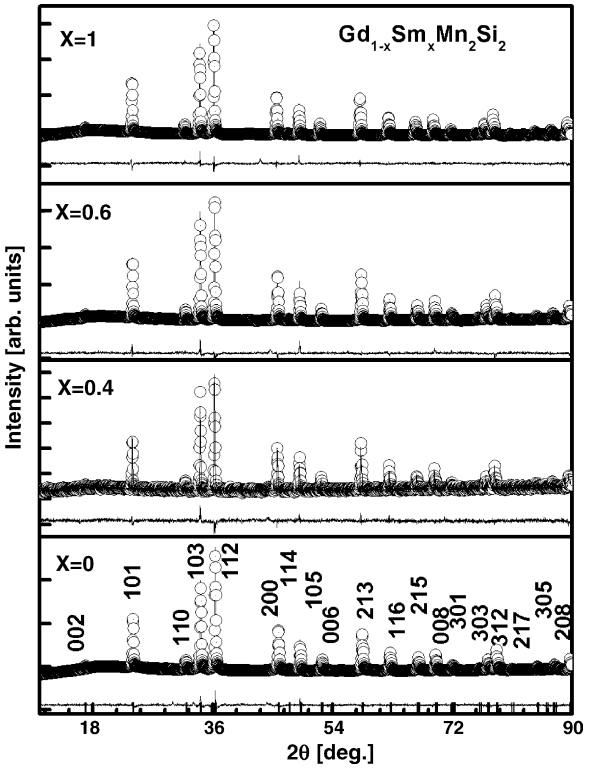

Fig. 1. Rietveld refined powder X-ray diffractograms of $Gd_{1-x}Sm_xMn_2Si_2$ compounds. The plots at the bottom show the difference between the calculated and experimental patterns in each case.

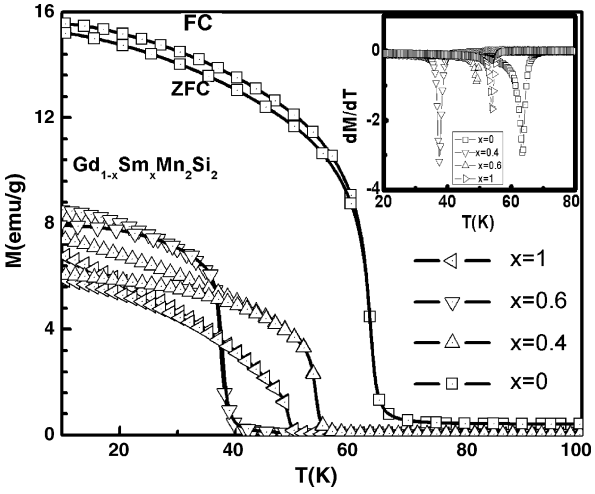

Fig. 2. Temperature variation of magnetization of $Gd_{1-x}Sm_xMn_2Si_2$ compounds in a field of 200 Oe. The inset shows the first derivative of magnetization with respect to temperature.