Magnetic entropy change and refrigerant capacity in GdFeAl compound

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Magnetic properties and magnetocaloric effect of GdFeAl compound have been investigated. The small saturated magnetization of GdFeAl compound is caused by the antiferromagnetic coupling between the magnetic moments of Gd and Fe atoms. A second-order magnetic phase transition is confirmed around 265 K. The maximum magnetic entropy change for GdFeAl compound is 3.7 J kg^{-1} K^{-1} under the field change of 0–5 T. However, a large refrigerant capacity of 420 J kg^{-1} is obtained, which is due to the large full width at half peak of the magnetic entropy change versus temperature curve in GdFeAl compound. © *2009 American Institute of Physics*.
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The magnetocaloric effect (MCE) of a solid is widely studied through the adiabatic temperature change or the isothermal magnetic entropy change (ΔS_M) in a variable magnetic field.[1,2] The magnetic refrigeration near room temperature, based on the MCE, is expected to replace conventional gas compression in the near future because of its higher efficiency and lower environmental impact.[2] Currently, the large MCE has been found in materials with a first-order phase transition, such as $Gd_5(Si,Ge)_4$,[3] $La(Fe,Si)_{13}$,[4,5] MnAs,[6,7] MnFe(P,As),[8] Ni_2MnGa,[9] etc. However, the remarkable hysteresis that appears in some materials associated with a first-order phase transition may reduce the actual efficiency of the cooling process.[10] Although the materials with a magnetic second-order phase transition usually show a lower value of ΔS_M than those with a first-order phase transition, their magnetization exhibits excellent reversibility for the temperature/magnetic field cycling, which is highly desired in the practical application of the magnetic refrigeration technique.

It is well known that there are a great number of rare-earth intermetallic compounds with a composition ratio of 1:2, such as the series RM_2. The crystallographic structures of RM_2 depend on the elements R (rare-earth element) and M (metal element).[11] The MCE of cubic Laves phase compounds RM_2 have been extensively investigated.[12–15] In this work, we will report the magnetic entropy change and refrigerant capacity (RC) for GdFeAl compound. The maximum $-\Delta S_M$ of 3.7 J kg^{-1} K^{-1} has been obtained in GdFeAl compound under the field change of 0–5 T. Although the value of $-\Delta S_M$ is not very large, the full width at half peak of the $-\Delta S_M \sim T$ curve, which is defined as the temperature interval corresponding to the half maximum of $-\Delta S_M$, reaches about 159 K. This leads to a large RC (420 J kg^{-1} for the field change of 0–5 T).

GdFeAl ingot was prepared by arc melting Al, with the purity of 99.99%, and Gd and Fe, with the purity of 99.9%, in a high-purity argon atmosphere. The ingot was turned over and remelted several times in order to ensure homogeneity. The as-cast bulk alloy was sealed in a quartz tube in vacuum and annealed at 1073 K for 8 days, and then quenched to room temperature. X-ray powder diffraction indicates that the sample was approximately composed of a single $MgZn_2$-type phase (space group $P6_3/mmc$) (see Fig. 1). The lattice parameters a and c were determined to be 5.42 and 8.79 Å, respectively, consistent with the report in Ref. 16. Magnetic measurements were carried out using a superconducting quantum interference device magnetometer and a commercial Physical Property Measurement System (Quantum Design). The magnetic entropy change was calculated from the isothermal magnetization data by the Maxwell relation.

Figure 2 shows the thermal-magnetic M-T curve under a field of 0.1 T for GdFeAl compound. A magnetic phase transition takes place at Curie temperature T_C=265 K, corresponding to the maximum slope of M-T curve. A kink in the M-T curve is also found around 220 K. Perhaps the kink is caused by the existence of the small impurity in the sample. The inset of Fig. 2 shows the magnetization curve at 5 K.

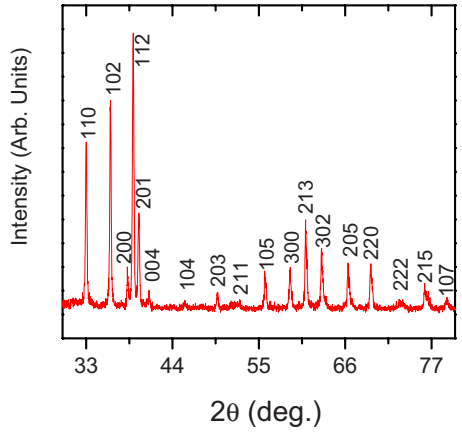

FIG. 1. (Color online) X-ray powder diffraction pattern of GdFeAl compound at room temperature.

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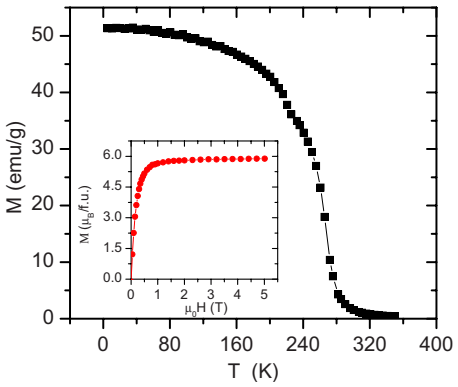

FIG. 2. (Color online) Temperature dependence of magnetization under a field of 0.1 T. The inset gives the magnetization curve at 5 K for GdFeAl compound.

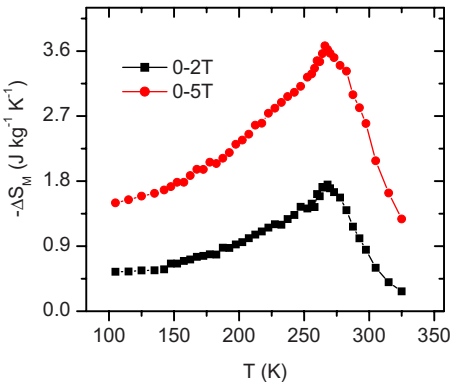

FIG. 4. (Color online) Magnetic entropy change as a function of temperature under field changes of 0–2 and 0–5 T for GdFeAl compound.

The saturated magnetization $\mu_s = 5.8\mu_B$ per GdFeAl formula is obtained, which is in accord with the result in Ref. 11. It is the antiferromagnetic coupling between the magnetic moments of Gd and Fe sublattice that reduces the total moment per unit cell.[17] Moreover, a small average Fe moment size of $0.80\mu_B$ at low temperature has been calculated from ^{57}Fe Mössbauer measurement result of GdFeAl.[17]

Figure 3(a) displays the isothermal magnetization curves measured in a temperature range of 100–330 K under the magnetic fields up to 5 T for GdFeAl compound. In the vicinity of T_C, from 257 to 271 K, the temperature step is 2 K; in far regions of 145–255 and 275–300 K, the temperature step is 5 K; in other temperature regions, the 10 K step is chosen. The sweep rate of the field is slow enough to ensure that the magnetization curves are obtained in an isothermal process. Figure 3(b) shows the Arrott plots[18] of GdFeAl compound. The inflection or negative slope as an indication of metamagnetic transition above T_C is not observed in Fig. 3(b), which indicates a nature of the second-order phase transition in GdFeAl compound.

Figure 4 shows the magnetic entropy change as a function of temperature for the field changes of 0–2 and 0–5 T. The peak value of ΔS_M, for a field change of 0–5 T, is 3.7 J kg^{-1} K^{-1} for GdFeAl compound. This value is comparable with or even higher than those of many previously published results for manganese oxides with a second-order phase transition.[19] Although this value is not very large, the full width at half peak of the ΔS_M-T curve reaches about 159 K. This is very useful for obtaining a large RC.[20]

In the literatures, a variety of methods are used to calculate the RC.[20,21] In the present work the RC values are determined by numerically integrating the area under the ΔS_M-T curve, using the temperatures at half maximum of the

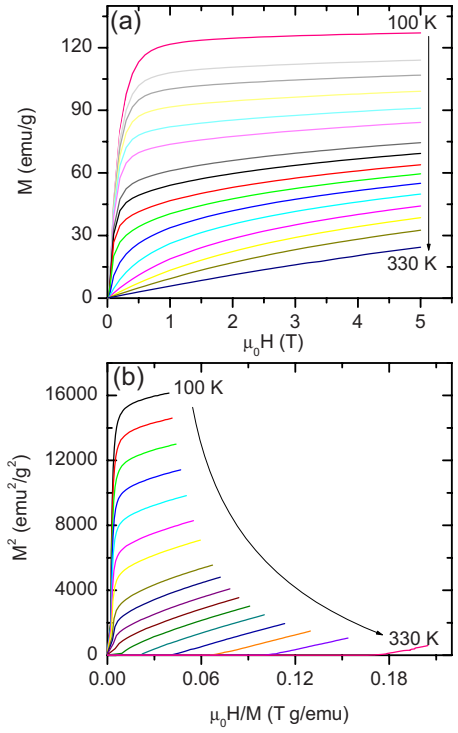

FIG. 3. (Color online) Isothermal magnetization curves (a) and the Arrott plots (b) for GdFeAl compound.

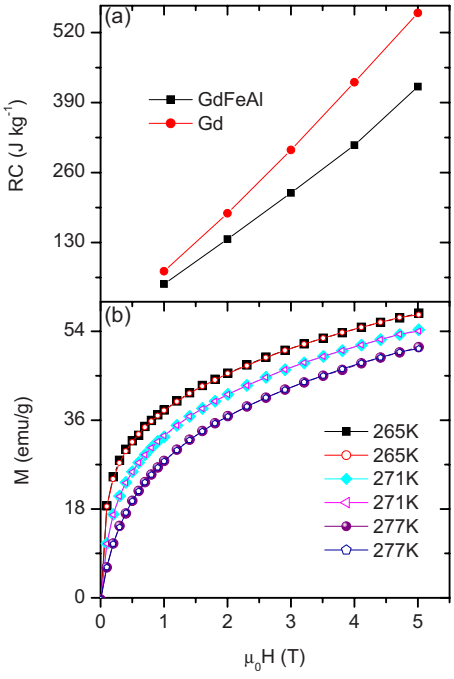

FIG. 5. (Color online) Field dependence of RC for GdFeAl compound as well as for pure Gd for comparison (a) and isothermal magnetization curves in the vicinity of T_C for GdFeAl compound measured in increasing field (closed symbols) and decreasing field (open symbols) (b).

ΔS_M peak as the integration limits: $RC = \int_{T_1}^{T_2} \Delta S_M(T) dT$. Figure 5(a) shows the field dependence of RC for GdFeAl compound. For comparison, the results of pure Gd are also shown in Fig. 5(a). The value of RC for GdFeAl compound is 420 J kg^{-1} for a field change of 0–5 T, which is smaller than that of pure Gd (556 J kg^{-1}). However, it is much larger than those of $Gd_5Si_2Ge_2$ (305 J kg^{-1}) and $Gd_5Si_2Ge_{1.9}Fe_{0.1}$ (360 J kg^{-1}).[22] Furthermore, a reversible behavior for the field increasing/decreasing cycles around T_C is observed [see Fig. 5(b)] due to the second-order phase transition in GdFeAl compound. Thus, detrimental effects for fast-cycling refrigerators of hysteresis losses and slow kinetics do not exist in GdFeAl compound.

In summary, the peak value of ΔS_M (3.7 J kg^{-1} K^{-1}) at 266 K is observed under the magnetic field change of 0–5 T for GdFeAl compound. A magnetic second-order transition is justified by the Arrott plot. A large RC, 420 J kg^{-1} under the field change of 0–5 T, is found in GdFeAl compound. A large full width at half peak of the ΔS_M-T curves and no hysteresis loss are the advantages of GdFeAl compound.

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