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Large refrigerant capacity of R Ga (R = Tb and Dy) compounds

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The magnetic properties and magnetocaloric effects (MCEs) of R Ga (R = Tb and Dy) compounds are investigated. The TbGa compound exhibits two successive magnetic transitions: spin-reorientation (SR) transition at $T_{SR} = 31$ K and second-order ferromagnetic (FM)–paramagnetic (PM) transition at Curie temperature $T_C = 154$ K, while the DyGa compound undergoes a SR transition with $T_{SR} = 25$ K and a FM–PM transition with $T_C = 113$ K. It is noteworthy that a broad distribution of the magnetic entropy change peak is observed. The values of the refrigerant capacity (RC) for TbGa and DyGa are found to be 620.6 and 381.9 $J/kg$ for a field change of 0–5 T, respectively. And for a field change of 0–7 T, the values are 900 and 584.2 $J/kg$, respectively. The large value of RC for TbGa and DyGa originates from the combined contribution from SR and FM–PM transitions, which enlarges the temperature span of large MCE. © 2012 American Institute of Physics. [doi:10.1063/1.3672842]

I. INTRODUCTION

Excellent magnetocaloric properties have been observed in some magnetic materials with first-order phase transitions.1–4 However, the first-order phase transition is usually accompanied by considerable thermal and magnetic hysteresis, thus reducing the RC of magnetic materials. It is of importance to explore magnetic refrigerant materials with large reversible MCEs as well as high RC values based on the second-order magnetic transition.

Binary R Ga (R = rare-earth) intermetallic compounds crystallize in the orthorhombic CrB type structure with the space group of Cmcm.5 They are ferromagnets that undergo the second-order ferromagnetic (FM)–paramagnetic (PM) transitions at respective Curie temperature $T_C$.6–8 A systematic investigation of the magnetic structure for R Ga (Pr-Er) was carried out by measuring the temperature dependencies of the hyperfine interactions and it was observed that the R Ga (R = Nd, Sm, Ho, and Er) compounds exhibit spin-reorientation (SR) transition below $T_C$.8 Recently, the magnetic and magnetocaloric properties of R Ga (R = Gd, Ho, and Er) have been studied.9–11 In this paper, we carefully investigate the magnetic properties and MCEs of R Ga (R = Tb and Dy).

II. EXPERIMENT

The R Ga (R = Tb and Dy) ingots were prepared in a purified Ar atmosphere by arc-melting of the stoichiometric amounts of the high purity (>99.9 wt. %) constituent elements on a water cooled copper hearth. To achieve composi-

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and second-order FM–PM transition at $T_C = 154$ K, and the DyGa compound undergoes a SR transition with $T_{SR} = 25$ K and a second-order FM–PM transition with $T_C = 113$ K. The $T_C$ values observed here are in good agreement with the previous results.\(^7\) No thermal hysteresis is observed in the ZFC and FC curves above $T_C$. However, it is noted that the ZFC and FC magnetization data show thermomagnetic irreversibility below $T_C$ as observed in Ref. \(^11\) and it is likely due to a domain wall pinning effect.

Magnetization isotherms of $R$Ga ($R$ = Tb and Dy) were measured in a wide temperature range under increasing and decreasing fields up to 7 T (Fig. 3). Both TbGa and DyGa are found to exhibit similar $M$-$H$ curves. The magnetization increases rapidly at low fields and shows a tendency to saturate with an increase of the field, exhibiting typical FM nature. For both TbGa and DyGa, the magnetization curves under increasing and decreasing fields are completely reversible in a wide temperature range around $T_C$ and no magnetic hysteresis is observed. From M-$H$ curves, we can also get the $M^2$ versus $H/M$ curves, i.e., Arrott plots of $R$Ga ($R$ = Tb and Dy) around $T_C$. Neither inflection nor negative slope in the Arrott plots is observed, indicating that the $R$Ga ($R$ = Tb and Dy) compounds exhibit second-order FM–PM transition.

The second-order phase transition usually means thermal and magnetic reversibility, which has been confirmed in Figs. 2 and 3. This is favorable to practical applications of the magnetic refrigeration materials.

The $\Delta S_M$ of $R$Ga ($R$ = Tb and Dy) was calculated based on isothermal magnetization data by using the Maxwell relation $\Delta S_M = \int_0^T (\partial M/\partial T)_H dH$. The $\Delta S_M$ as a function of temperature is shown in Fig. 4. For TbGa, there are two sharp peaks in the $\Delta S_M$-$T$ curves around $T_{SR}$ and $T_C$, respectively, corresponding to the two transitions detected in the $M$-$T$ curves. The maximal values of $-\Delta S_M$ are 10.8 and 8.1 J/kg K at $T_{SR}$ and $T_C$ for a field change of 0–7 T, respectively. Similar results are observed in HoGa,\(^10\) ErGa,\(^11\) HoAl\(_2\),\(^13\) and Gd\(_4\)Co\(_3\).\(^14\) It can be seen from Fig. 4(a) that the two peaks partly overlaps between $T_{SR}$ and $T_C$, which enlarges greatly the temperature range of MCE. For DyGa, its maximal values of $-\Delta S_M$ are 5.8

### Table I. The crystallographic, magnetic, and magnetocaloric data for the $R$Ga ($R$ = Gd-Er) compounds.

<table>
<thead>
<tr>
<th>Compounds</th>
<th>a (Å)</th>
<th>b (Å)</th>
<th>c (Å)</th>
<th>$T_{SR}$ (K)</th>
<th>$T_C$ (K)</th>
<th>$\Delta S_M$ at $T_{SR}$ (J/kg K)</th>
<th>$\Delta S_M$ at $T_C$ (J/kg K)</th>
<th>RC (J/kg)</th>
<th>Refs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>GdGa</td>
<td>4.338(2)</td>
<td>11.0104(3)</td>
<td>4.1058(2)</td>
<td>~100</td>
<td>183</td>
<td>~3.3(^a)</td>
<td>4.8(^a)</td>
<td>~438(^a)</td>
<td>9</td>
</tr>
<tr>
<td>TbGa</td>
<td>4.317(8)</td>
<td>10.929(1)</td>
<td>4.088(1)</td>
<td>31</td>
<td>154</td>
<td>8.3(^b)/10.8(^b)</td>
<td>5.9(^b)/8.1(^b)</td>
<td>620.6(^b)/900(^b)</td>
<td>this work</td>
</tr>
<tr>
<td>DyGa</td>
<td>4.297(4)</td>
<td>10.856(5)</td>
<td>4.066(4)</td>
<td>25</td>
<td>113</td>
<td>4.3(^b)/5.8(^b)</td>
<td>5.8(^b)/8.1(^b)</td>
<td>381.9(^b)/584.2(^b)</td>
<td>this work</td>
</tr>
<tr>
<td>HoGa</td>
<td>4.280(9)</td>
<td>10.781(9)</td>
<td>4.042(4)</td>
<td>20</td>
<td>69</td>
<td>7.8(^b)</td>
<td>17.1(^b)</td>
<td>455(^b)</td>
<td>10</td>
</tr>
<tr>
<td>ErGa</td>
<td>4.263(9)</td>
<td>10.728(8)</td>
<td>4.030(6)</td>
<td>15</td>
<td>30</td>
<td>16.5(^b)</td>
<td>21.3(^b)</td>
<td>494(^b)</td>
<td>11</td>
</tr>
</tbody>
</table>

\(^a\)The result is estimated from the temperature dependence of $\Delta S_M$ in Ref. \(^9\).

\(^b\)The values are obtained for a field change of 0–5 T.

\(^b\)The values are obtained for a field change of 0–7 T.
and 8.1 J/kg K at $T_{SR}$ and $T_C$, respectively, for a field change of 0–7 T.

As another important criterion to evaluate the magnetocaloric properties of magnetic refrigerant materials, the RC of $R$Ga ($R$ = Tb and Dy) compounds were estimated based on the $\Delta S_M-T$ curves by using the approach suggested by Gschneidner et al. The RC value is defined as

$$RC = \int_{T_1}^{T_2} |\Delta S_M|dT,$$

where $T_1$ and $T_2$ are the temperatures corresponding to both sides of the half-maximum value of $\Delta S_M$ peak, respectively. It is noteworthy that a broad distribution of the $\Delta S_M$ peak is observed. The value of RC for TbGa is found to be 900 J/kg with $T_1 = 22.7$ K (temperature of the cold end) and $T_2 = 183$ K (temperature of the hot end) for a field change of 0–7 T. For the DyGa compound, the RC value is 584.2 J/kg with $T_1 = 19.6$ K and $T_2 = 136.9$ K. The magnetic and magnetocaloric data for the $R$Ga compounds with $R$ = Gd-Er are also listed in Table I. Although the TbGa compound has a moderate $\Delta S_M$, its RC value is much larger than those of other magnetic refrigerant materials in a wide temperature range. The large RC of TbGa originates from the combined contribution from SR and FM–PM transitions, which enlarges the temperature span of the MCE. It is noted that the value of $\Delta S_M$ for $R$Ga increases monotonically with the decrease of $T_C$ as the $R$ atomic number increases. The results indicate that the working temperature and temperature span of the MCE could be tuned by the substitution between different $R$ elements. Therefore, the $R$Ga compounds are expected to be good candidates for the magnetic refrigeration.

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