Low-temperature heat-capacity studies of $R_2Ni_3Si_5$ ($R =$ Pr, Nd, Sm, Gd, Tb, Dy, Ho)

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We report here our low temperature ($2–30 \text{ K}$) heat capacity, $C_p$ measurements of $R_2Ni_3Si_5$ ($R =$ Pr, Nd, Sm, Gd-Ho). Large peaks in heat capacity data at magnetic transition temperatures ($T_N$) confirm the bulk nature of magnetic order in these compounds. In Nd$_2$Ni$_3$Si$_5$, Gd$_2$Ni$_3$Si$_5$, and Dy$_2$Ni$_3$Si$_5$, magnetization studies indicated only one magnetic transition, whereas, heat-capacity data show two transitions. $T_N$ of the heavier rare-earth member, Tb$_2$Ni$_3$Si$_5$, showing significant deviation from de Gennes scaling is notable. Magnetic entropy, $\Delta S$, estimated from heat-capacity data suggest that the magnetic ground state is a doublet in R$_2$Ni$_3$Si$_5$ ($R =$ Pr, Nd, Sm, Dy, Ho) and a quartet in Tb$_2$Ni$_3$Si$_5$. In all the cases, $\Delta S$ at $T_N$ is slightly less than that expected due to the suggested states, which we attribute to the occurrence of precursor effects of magnetic order above $T_N$. Except for Gd$_2$Ni$_3$Si$_5$, $\Delta S$ of the compounds does not reach the saturation limit of $R \ln(2J+1)$ even at 30 K, indicating the presence of crystalline electric field (CEF) effects. A hump in $C_p$ is observed below $T_N$ in Gd$_2$Ni$_3$Si$_5$ which is interpreted in terms of a possible amplitude-modulated magnetic spin structure. [S0163-1829(99)12605-5]

I. INTRODUCTION

Physical properties of rare-earth intermetallic materials have been of interest due to the effect of hybridization of 4f orbitals with the 3d orbitals resulting in anomalous phenomena such as, valence fluctuation, heavy fermion behavior, Kondo effect, magnetic transition temperature higher than expected, etc. In this context we had investigated the series $R_2Ni_3Si_5$ ($R =$ Y, rare earth). Our synthesis and magnetic susceptibility studies of $R_2Ni_3Si_5$ ($R =$ Pr, Nd, Sm, Gd, Tb, Dy, Ho) (Refs. 4–7) showed that they crystallize in U$_2$Co$_3$Si$_5$ structure and order antiferromagnetically. Tb$_2$Ni$_3$Si$_5$ revealed a well-separated double magnetic transition ($T_{N1} = 19.5 \text{ K}$ and $T_{N2} = 12.5 \text{ K}$). We had found positive giant magnetoresistance (GMR) in R$_2Ni_3Si_5$ ($R =$ Tb, Sm, Nd) (Ref. 9) and anomalous magnetoresistance in R$_2Ni_3Si_5$ ($R =$ Pr, Dy, Ho). The positive GMR in Tb$_2$Ni$_3$Si$_5$ at 4.4 K and at 45 kG ($\sim 85\%$) is the largest among polycrystalline materials. From our studies of the temperature dependence of magnetoresistance, we had suggested the possibility of the existence of short-range ferromagnetic correlations above $T_N$ in these materials. In order to get further insight into the nature of the magnetic ordering in these materials, we have studied the low-temperature heat capacity of the series R$_2Ni_3Si_5$ ($R =$ Pr, Nd, Sm, Gd, Tb, Dy, Ho), the results of which are reported here.

II. EXPERIMENT

The compounds, R$_2Ni_3Si_5$ ($R =$ rare earth), were prepared by the standard arc melting procedure. Details of sample preparation and unit-cell parameters are given elsewhere. The heat-capacity measurements were performed at T.I.F.R., using a home built, fully automated adiabatic calorimeter in the temperature range 1.5–30 K. The absolute accuracy of our heat-capacity measurements is better than 1% in this temperature range. The measurements reported here have been performed on samples obtained from the same batch on which the magnetic measurements were reported earlier.

III. RESULTS AND DISCUSSION

The heat-capacity results of our measurements on R$_2Ni_3Si_5$ ($R =$ Pr, Sm, Ho, Gd, Nd, Tb, Dy) are given in Figs. 1, 2, 3, 4, 5, 6, and 7. In R$_2Ni_3Si_5$ ($R =$ Pr, Sm, Ho), a single large peak (at 8.5, 10.4, and 6.7 K, respectively) and in R$_2Ni_3Si_5$ ($R =$ Nd,Gd,Tb,Dy) two peaks (at 9.2 and 9.5K, 13.5 and 15.0 K, 12.4 and 19.2 K, and 3.9 and 9.0 K, respectively) are seen in the heat capacity. The temperature at which the peaks occur (the higher temperature peak where two peaks are seen) agree with the magnetic ordering temperatures, $T_N$, as seen in magnetic susceptibility data confirming the magnetic origin of the peaks in heat capacity. In the case of Tb$_2$Ni$_3$Si$_5$, two peaks are seen in magnetic susceptibility as well as in resistivity and they correspond to the peaks seen in heat-capacity data. In the case of Dy$_2$Ni$_3$Si$_5$, two clear peaks are seen in heat-capacity, but in the magnetic susceptibility, only the higher transition at $\sim 9.5 \text{ K}$ is seen clearly; the lower transition appears as a very
small anomaly at ~4 K. In the case of Nd$_2$Ni$_3$Si$_5$ also, two well-resolved peaks are seen (at ~9.5 and ~9.1 K) in the heat-capacity data but are not well resolved in the magnetic-susceptibility data, probably due to the ordering temperatures being very close to each other. In the case of Gd$_2$Ni$_3$Si$_5$ both the susceptibility and resistivity measurements show only a single transition: while the magnetic susceptibility exhibits a peak at the higher transition temperature (14.7 K), the resistivity exhibits a change of slope at the lower transition temperature (13.5 K). Absence of the resistivity anomaly at $T_N$ = 15 K may be due to competing effects of increase of resistivity due to formation of superzone energy gaps and decrease of resistivity due to ordering of spins.

Normally, the $T_N$'s of rare-earth members of a series are expected to follow the well-known de Gennes scaling. In Fig. 8, $T_N$'s of the different members of the series are plotted along with those expected (with respect to Gd) on the basis of de Gennes scaling. We find that $T_N$'s of the lighter members of the series do not follow the de Gennes scaling. Often this is found to be the case with other rare-earth series of compounds also. However, even among the heavier rare-earth members in this series, taking the higher $T_N$, the agreement is only marginal. Particularly significant is the deviation of $T_N$ of Tb$_2$Ni$_3$Si$_5$. Though the lower $T_N$'s fall closer to that expected from de Gennes scaling, we believe that the scaling is applicable only for the higher magnetic ordering temperature. At present, the reason for the anomalously high $T_{N_1}$ of Tb$_2$Ni$_3$Si$_5$ is not clear.

Since it is known that the crystalline electric field can enhance the magnetic transition temperature,$^{14}$ possibly the crystalline field effect is responsible for the enhancement of $T_N$ for lighter rare-earth members of this series (Fig. 8). If one adds the CEF terms to the exchange Hamiltonian, the ordering temperature then can be expressed as

$$T_N = \sum_j (\exp(-3B_j^0J_j^2/T_N))^{1/2} \sum_j (\exp(-3B_j^0J_j^2/T_N))^{-1/2}.$$  

where $J$ is the exchange constant for the 4f atoms and $B_j^0$ is the crystal-field parameter. Since Gd is the S-state ion, its ordering temperature can be used to fix the value of exchange constant. At present there is no experimental data on the crystal-field parameters of these compounds. Therefore, it is difficult to decide if CEF effects alone are responsible for the observed deviation from de Gennes scaling.

In order to obtain the magnetic contribution $C_m$ to the heat capacity, we need to subtract from the total heat capacity $C_p$ the lattice contribution $C_l$, arising from phonon excitations, and electronic contribution $C_e$, originating from...
conduction electrons and filled electron orbitals. \( C_t \) and \( C_e \) for a rare-earth member can be estimated from the nonmagnetic analog, La- or Lu-based member. Our attempts to synthesize La\(_2\)Ni\(_3\)Si\(_5\) did not succeed and therefore, the heat capacity of La\(_2\)Ni\(_3\)Si\(_5\) was taken to represent \( C_t \) and \( C_e \). Though this material forms with a monoclinic variation of U\(_2\)Co\(_3\)Si\(_5\)-type structure, to a first-order approximation we consider the value of \( C_t \) for the region below 3 K, the experimental data was used down to 3 K and \( T_N \).

Since Lu\(_2\)Ni\(_3\)Si\(_5\) superconducts below 2 K,\(^3\) for our considerations, the experimental data was used down to 3 K and for the region below 3 K, the experimental data in the region 3 - 6 K was fit to a polynomial and the values extrapolated to \( T = 0 \) K was used. The same procedure was adopted for other materials also to get the extrapolated values below the lower limit of our measuring temperature. The magnetic contribution to heat capacity \( C_m \) of the magnetic members of the series R\(_2\)Ni\(_3\)Si\(_5\) was obtained by subtracting heat capacity of Lu\(_2\)Ni\(_3\)Si\(_5\) from the heat capacity of R\(_2\)Ni\(_3\)Si\(_5\). Numerical integration was performed on the magnetic heat capacity thus obtained, to calculate the magnetic entropy of the compounds (shown as solid lines in Figs. 1–7). In this procedure the reasonable assumption, of the nuclear specific heat and nuclear Schottky effect being negligibly small,\(^1\) is made. In the calculations, the fact that one mole of R\(_2\)Ni\(_3\)Si\(_5\) contains two moles of rare-earth ions has been taken into account.

We discuss now the nature of the ground state of the energy level of the rare-earth ions in these materials on the basis of their magnetic entropy. The magnetic entropies \( \Delta S \) at the \( T_N \) and at 28 K (the maximum temperature of our measurement) for different members of the R\(_2\)Ni\(_3\)Si\(_5\) series, along with the total \( \Delta S \) expected on the basis of Hund’s rule total angular momentum \( J \) of the energy level of the trivalent rare-earth ion, are summarized in Table I. In these discussions, wherever two magnetic transitions are observed, we consider the value of \( \Delta S \) at the higher \( T_N \).

In the case of Gd\(_2\)Ni\(_3\)Si\(_5\) where no crystal-field effects (CEF) would be present, \( \Delta S \) at \( T_N (\sim 1.88R) \), is slightly less than the expected total \( \Delta S \), \([J = 7/2, R \ln 8 (\sim 2.08R)]\) which it very closely reaches as a plateau at 28 K \((\sim 2.02R)\). For Tb\(_2\)Ni\(_3\)Si\(_5\) \( \Delta S (\sim 1.13R) \) at \( T_N \) is much larger than that expected for a doublet ground state \((R \ln 2 = 0.69R)\) and therefore, the ground state of the Tb ion is not a doublet. The value is only slightly less than \( R \ln 4 \)
The continuous increase of measurements on these compounds, the existence of short-range field for the respective cases. This suggests crystalline electric field in a few members of R$_2$Ni$_3$Si$_5$, other systems such as Gd$_2$Ni$_3$Si$_5$, ~R~ the cases discussed here, similar value of entropy at $T_N$ however, as discussed later, cursor effects of magnetic order does not fit in this picture.

However, as discussed later, $\Delta S$ below $T_N$ in this case is anomalous and the actual $\Delta S$ may be higher than our extrapolated $\Delta S$ estimate.

In all these cases, except for Gd$_2$Ni$_3$Si$_5$, $\Delta S$ at 28 K is considerably less than the calculated saturation value of $\Delta S$, for the respective cases. This suggests crystalline electric field (CEF) splitting of electronic levels in these compounds. The continuous increase of $\Delta S$ above $T_N$ in the cases of R$_2$Ni$_3$Si$_5$ ($R$=Pr, Nd, Tb, Dy, Ho) indicates that the electronic levels of $R$ ions in these compounds are CEF split with closely spaced sublevels. In the case of Sm$_2$Ni$_3$Si$_5$ the well-defined plateau of $\Delta S$ above $T_N$ indicates that the next higher CEF level is at a considerably higher energy. A few degrees above the transition temperature, the magnetic entropy reaches a plateau with $\Delta S=0.63R=\ln 2$, confirming a doublet ground state. A closer value of $\Delta S$ in the case of Gd$_2$Ni$_3$Si$_5$ is consistent with the fact that since the Gd$^{3+}$ ion is an $S$-state ion, there would no CEF effect.

Considering that the three compounds, Nd$_2$Ni$_3$Si$_5$, Sm$_2$Ni$_3$Si$_5$, and Tb$_2$Ni$_3$Si$_5$ have different type of ground states, we believe that the GMR exhibited by these materials is unlikely to be due to CEF effects.

We now discuss on the features of double magnetic transition in some of these materials. [The heat-capacity result of Gd$_2$Ni$_3$Si$_5$ (Fig. 4) is interesting due to the double magnetic transition (which is not usually seen in Gd-based materials) as well as due to a significant nearly linear term below $T_N$ in $C_m$ (discussed later). Considering that the Gd ion is an $S$-state ion, the double magnetic transition must be due to spin orientation arising from anisotropy of exchange coupling, and not due to CEF effect. The possibility of a spin reorientation is suggested from the observation of a change in slope in the isothermal magnetization curve of Gd$_2$Ni$_3$Si$_5$, below its magnetic ordering temperature.

With respect to the double magnetic transition observed in Tb$_2$Ni$_3$Si$_5$, we point out that double magnetic transition has also been seen in TbFe$_2$Si$_2$ (Ref. 18) and in TbNi$_2$Si$_2$. From neutron-scattering experiments on TbFe$_2$Si$_2$ (Ref. 20) and on TbNi$_2$Si$_2$, the transition at higher temperature is found to be due to an onset of magnetic order which is incommensurate with the lattice, and the transition at the lower temperature is found to be due to a change of the magnetic structure from incommensurate to commensurate structure with the lattice. It is quite likely that the same is possible in our case of Tb$_2$Ni$_3$Si$_5$. A noticeable feature of the two transitions in our case is the differences in shape of the peaks at the two transition temperatures. While the peak at 19.2 K is nearly $\lambda$ type and broad, the peak at 12.4 K is very sharp with a half-width of around 0.5 K. The sharpness of the second peak indicates the possibility of a first-order transition.

In the case of Dy$_2$Ni$_3$Si$_5$ which also shows double magnetic transition (Fig. 7) both the peaks appear to be $\lambda$-like which is unlike that of Tb$_2$Ni$_3$Si$_5$ and indicates that the nature of the transition at lower magnetic transition temperature is different in these two materials. Another notable feature in our results, is the unusual behavior of $C_m$ in Gd$_2$Ni$_3$Si$_5$ and Ho$_2$Ni$_3$Si$_5$ below $T_N$. We find that $C_m$ in the present materials, except those of Ho and Gd compounds, nearly follow a $T^3$ behavior as expected for a typical antiferromagnet. In the case of Ho$_2$Ni$_3$Si$_5$, we observe an upward deviation from $T^3$ behavior below 5 K. $C_m$.

### Table I. Magnetic properties, including the magnetic entropies, of different members of the R$_2$Ni$_3$Si$_5$ series. $T_N$ values from de Gennes scaling, $(g_J-1)^2J(J+1)$, were obtained with Gd$_2$Ni$_3$Si$_5$ as a reference material and $J$ as good quantum number.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$T_{N_1}$ (K)</th>
<th>$T_{N_2}$ (K)</th>
<th>$T_N$ (K)</th>
<th>$\mu_{eff}/R$</th>
<th>$\Theta_p$ (K)</th>
<th>$\Delta S_{mag}/R$ at 6 K &amp; 5.5 T</th>
<th>$\Delta S_{mag}/R$ at 28 K</th>
<th>$\ln(2J+1)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pr$_2$Ni$_3$Si$_5$</td>
<td>8.5</td>
<td>0.8</td>
<td>3.67</td>
<td>1.15</td>
<td>0.38</td>
<td>0.78</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td>Nd$_2$Ni$_3$Si$_5$</td>
<td>9.5</td>
<td>9.1</td>
<td>1.7</td>
<td>1.21</td>
<td>0.57</td>
<td>0.50</td>
<td>1.07</td>
<td>2.30</td>
</tr>
<tr>
<td>Sm$_2$Ni$_3$Si$_5$</td>
<td>10.4</td>
<td>4.2</td>
<td>0.03</td>
<td>2.50</td>
<td>1.21</td>
<td>0.45</td>
<td>1.51</td>
<td>2.56</td>
</tr>
<tr>
<td>Gd$_2$Ni$_3$Si$_5$</td>
<td>15.0</td>
<td>13.5</td>
<td>8.11</td>
<td>2.73</td>
<td>1.88</td>
<td>1.70</td>
<td>2.02</td>
<td>2.08</td>
</tr>
<tr>
<td>Tb$_2$Ni$_3$Si$_5$</td>
<td>19.4</td>
<td>12.4</td>
<td>10.0</td>
<td>6.36</td>
<td>0.562</td>
<td>0.16</td>
<td>1.42</td>
<td>2.77</td>
</tr>
<tr>
<td>Dy$_2$Ni$_3$Si$_5$</td>
<td>9.0</td>
<td>3.9</td>
<td>10.79</td>
<td>5.99</td>
<td>0.73</td>
<td>1.83</td>
<td>2.83</td>
<td></td>
</tr>
<tr>
<td>Ho$_2$Ni$_3$Si$_5$</td>
<td>6.7</td>
<td>4.3</td>
<td>10.47</td>
<td>5.99</td>
<td>0.73</td>
<td>1.83</td>
<td>2.83</td>
<td></td>
</tr>
</tbody>
</table>
of Ho$_2$Ni$_3$Si$_5$ at 2 K (the lowest limit of our measurement) is the largest among that observed in all the other materials in this series. An upward deviation in $C_m$ has been reported in the case of Ho$_2$Fe$_2$Si$_5$ also. The origin of this deviation is not clear at present.\textsuperscript{24}

In the case of Gd$_2$Ni$_3$Si$_5$, below $T_N$, the deviation of $C_m$ from $T^3$ behavior is rather severe. $C_m$ has a large linear component with a small hump. A similar behavior below $T_N$ has been seen in $C_m$ of the related material Gd$_2$Fe$_2$Si$_5$ (Ref. 15) (tetragonal structure) and in the cases of other materials, such as GdCu$_2$Si$_2$, GdNi$_2$Si$_2$, GdGa$_2$, and GdCu$_5$.\textsuperscript{25} In the case of Gd$_2$Fe$_2$Si$_5$, the large linear term has been suggested to be due to a two-dimensional ferromagnetic spin waves (where the magnon dispersion relation is proportional to $q^2$, where $q$ is the wave vector) or from one-dimensional antiferromagnetic spin waves (where the magnon dispersion relation is proportional to $q$).\textsuperscript{15}

Tsay et al.\textsuperscript{26} and Lai et al.\textsuperscript{27} have explained the observation of a hump in the heat capacity of several Gd-based antiferromagnetic oxide compounds, on the basis of the model proposed by Fishman and Liu.\textsuperscript{28} Fishman and Liu\textsuperscript{28} have shown that a hump could originate in a Heisenberg ferromagnet from quantum spin fluctuations induced by a transverse degree of freedom. They have also shown that as the total spin increases, the hump becomes more prominent and even may show up as a peak. The hump takes place at a temperature $T^* = 3T_N/(S+1)$ (where $S$ is the total spin quantum number). Since value of $S$ for Gd is maximum amongst rare earths (7/2), the effect may show up more in the case of Gd-based compounds and would occur at a temperature $T^* = 3T_N/5$. However, in the intermetallics compounds, such as above mentioned Gd$_2$Fe$_2$Si$_5$, GdCu$_2$Si$_2$, and in the present case of, Gd$_2$Ni$_3$Si$_5$, $T^*$ is much smaller ($T^*/T_N < 5$) than predicted by this theory. Moreover, the model is for ferromagnets and our materials are antiferromagnetic. Therefore, even though Tsay et al.\textsuperscript{26} and Lai et al.\textsuperscript{27} have applied the model successfully in the antiferromagnetic oxide system, we believe that this mechanism may not be applicable in our cases.

Another approach to explain such a behavior of $C_m$ below $T_N$ has been given by Blanco et al.\textsuperscript{29} They derived the expressions for $C_m$ for the case of the equal moment and the case of amplitude-modulated (where the amplitude of the magnetic moment exhibit a periodic variation, with a sine-wave type below and near $T_N$) antiferromagnetic systems, in the mean-field approximation. Under this approach they could successfully explain the heat-capacity behavior of Gd systems, such as, GdCu$_2$Si$_2$, GdNi$_2$Si$_2$, GdGa$_2$, and GdCu$_5$. Gd-based systems had been chosen because of the absence of crystal field in these compounds.

Blanco et al. showed that the $C_m$ of an antiferromagnetic material, can be expressed as\textsuperscript{29}

$$C_m(T) = \Delta C_m + C'_m \frac{T-T_N}{T_N} + \ldots,$$  \hspace{1cm} (3.2)

where the $\Delta C_m$ is the amplitude of the discontinuity in $C_m$ at $T_N$, and the second term expresses the slope, $dC_m/dT$, of the $C_mT$ curve just below $T_N$. The shape of the hump would depend on the strength of the Fourier transform of the exchange integration.\textsuperscript{29} Since entropy has to be constant, any deviation from $T^3$ behavior in $C_m$, below $T_N$, will result in a decrease in the peak height of $C_m$ at $T_N$. Therefore, the magnitude of jump in $C_m$ at $T_N$ can yield information on the magnetic structure of the material.

Blanco et al. showed that\textsuperscript{29} for an amplitude-modulated system

$$\Delta C_{AM} = \frac{10}{3} \frac{J(J+1)}{2J^2+2J+1} k_B$$  \hspace{1cm} (3.3)

and for the equal moment systems

$$\Delta C_{EM} = \frac{5}{2J^2+2J+1} k_B.$$  \hspace{1cm} (3.4)

From the above two equations, one finds that the discontinuity (peak height) in $C_m$ at $T_N$ in equal moment systems is 20.15 J/K mol. for a Gd system and that for amplitude-modulated systems, the peak height is reduced to two-thirds of that for an equal moment system.

In our case, the maximum in the $C_m$ in Gd$_2$Ni$_3$Si$_5$ occurs at around 13.5 K with a value of about 17 J/K mol, which is considerably lower than that expected for a equal moment system (20.15 J/K mol). Since the system undergoes one more transition at 15 K which affects the magnitude of the jump at 13.5 K, one cannot immediately conclude that the magnetic structure is amplitude modulated. However, because of the presence of the hump along with the reduced maximum at 13.5 K, we speculate that below this temperature, the system has an amplitude-modulated magnetic structure. A neutron-diffraction study might help in this regard. We point out that in some cases, e.g., helical (which is equal moment) and amplitude-modulated magnetic structures may also produce a similar neutron-diffraction pattern.\textsuperscript{29}

All the above anomalous magnetic behavior points out that the magnetic structure of these materials is not a simple antiferromagnetic structure. Very likely, the magnetic coupling in the plane could be ferromagnetic whereas the planes are coupled antiferromagnetically. Such a magnetic structure has been observed in closely related materials, e.g., Tb$_2$Fe$_3$Si$_5$,\textsuperscript{20} U$_2$(Ru$_{0.65}$Rh$_{0.35}$)$_3$Si$_5$.\textsuperscript{30} Elastic and inelastic neutron-scattering experiments would be rewarding in these interesting materials.

### IV. CONCLUSION

Our heat-capacity measurements have confirmed the bulk magnetic ordering of R$_2$Ni$_3$Si$_5$ (R = Pr, Nd, Sm, Gd-Ho) at temperatures indicated by the magnetic-susceptibility results. These heat-capacity measurements have not only confirmed the occurrence of double magnetic transitions in Tb$_2$Ni$_3$Si$_5$ and Dy$_2$Ni$_3$Si$_5$ seen earlier through susceptibility studies, they have also revealed the occurrence of double magnetic transitions in Nd$_2$Ni$_3$Si$_5$ and Gd$_2$Ni$_3$Si$_5$ as well, which are not discernible in magnetic-susceptibility results. $T_N$’s of the lighter members of the series do not follow de Gennes scaling. The deviation of $T_N$ of Tb$_2$Ni$_3$Si$_5$ from de Gennes scaling is rather large.

The magnetic entropy estimate in these materials suggest that in all these materials, except for Gd$_2$Ni$_3$Si$_5$ (where there is no CEF) and Tb$_2$Ni$_3$Si$_5$, the ground state of the rare-earth
ion is a doublet. In the case of Tb$_2$Ni$_3$Si$_5$, it appears to be a quartet. In all these cases, $\Delta S$ at $T_N$ is slightly less than that expected from the suggested ground states. We attribute this to the occurrence of a precursor effect coming from the short-range magnetic correlations even above their $T_N$'s, as suggested from our earlier magnetoresistance measurements. These results also suggest that CEF effects are not likely to be the cause of GMR observed in Nd$_2$Ni$_3$Si$_5$, Sm$_2$Ni$_3$Si$_5$, and Tb$_2$Ni$_3$Si$_5$. Another important feature of our results is the observation of a hump in the heat capacity of Gd$_2$Ni$_3$Si$_5$ below its $T_N$, which may be due to amplitude-modulated magnetic spin structure.

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