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 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 $Nd_2Ni_3Si_5$, $Gd_2Ni_3Si_5$, and $Dy_2Ni_3Si_5$, magnetization studies indicated only one magnetic transition, whereas, heat-capacity data show two transitions. T_N of the heavier rare-earth member, $Tb_2Ni_3Si_5$, showing significant deviation from de Gennes scaling is notable. Magnetic entropy, ΔS, estimated from heat-capacity data suggest that the magnetic ground state is a doublet in $R_2Ni_3Si_5$ (R=Pr, Nd, Sm, Dy, Ho) and a quartet in $Tb_2Ni_3Si_5$. In all the cases, ΔS, 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 $Gd_2Ni_3Si_5$, ΔS 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 $Gd_2Ni_3Si_5$ 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).[1–7] 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 $U_2Co_3Si_5$ structure[8] and order antiferromagnetically. $Tb_2Ni_3Si_5$ revealed a well-separated double magnetic transition (T_{N_1}=19.5 K and T_{N_2}=12.5 K).[6] 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).[10] The positive GMR in $Tb_2Ni_3Si_5$ at 4.4 K and at 45 kG (~85%) is the largest among polycrystalline materials.[9] 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.[10] 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.[3–6] The heat-capacity measurements were performed at T.I.F.R., using a home built, fully automated adiabatic calorimeter[11] 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.[4–6]

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 temperature, T_N, as seen in magnetic susceptibility data confirming the magnetic origin of the peaks in heat capacity. In the case of $Tb_2Ni_3Si_5$, two peaks are seen in magnetic susceptibility[6] as well as in resistivity[12] and they correspond to the peaks seen in heat-capacity data. In the case of $Dy_2Ni_3Si_5$, two clear peaks are seen in heat-capacity, but in the magnetic susceptibility, only the higher transition at ~9.5 K is seen clearly; the lower transition appears as a very

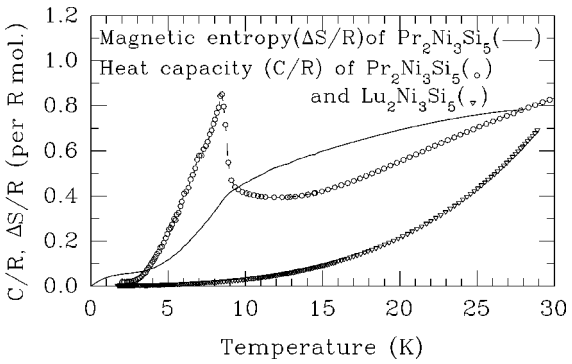

FIG. 1. Low-temperature heat capacity (C/R) of $Pr_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown.

small anomaly at ~4 K. In the case of $Nd_2Ni_3Si_5$ 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 $Gd_2Ni_3Si_5$ both the susceptibility and resistivity measurements show only a single transition: while the magnetic susceptibility exhibits a peak[4] at the higher transition temperature (14.7 K), the resistivity exhibits a change of slope[4] 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[13] 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 $Tb_2Ni_3Si_5$. 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 $Tb_2Ni_3Si_5$ is not clear.

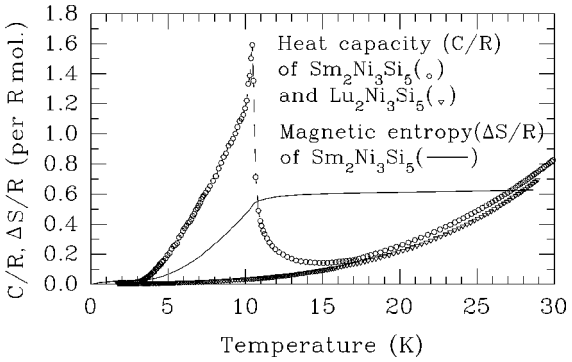

FIG. 2. Low-temperature heat capacity (C/R) of $Sm_2Ni_3Si_5$ (○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown.

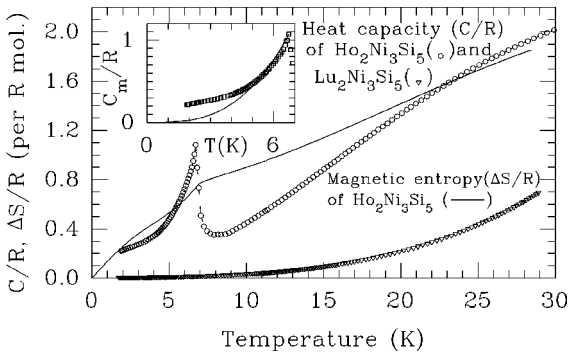

FIG. 3. Low-temperature heat capacity (C/R) of $Ho_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown. Inset: Magnetic contribution to heat capacity C_m as a function of temperature. The solid line represents the T^3 behavior. Note the deviation of C_m from the T^3 behavior at low temperature.

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[14]

$$T_N = \frac{2J(g_J-1)^2 \sum_{J_z} J_z^2 \exp(-3B_z^0 J_z^0/T_N)}{\sum_{J_z} (\exp(-3B_z^0 J_z^2/T_N))}, \qquad (3.1)$$

where J is the exchange constant for the $4f$ atoms and B_z^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

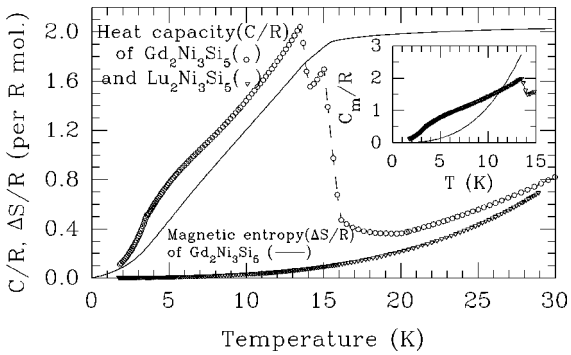

FIG. 4. Low-temperature heat capacity (C/R) of $Gd_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown. Inset: The magnetic contribution to heat capacity C_m as a function of temperature. The solid line represents the T^3 behavior and indicates that C_m does not follow a T^3 behavior.

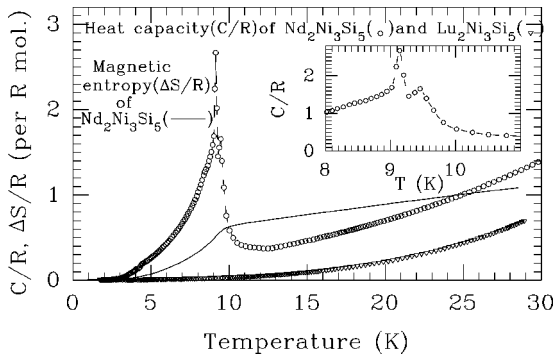

FIG. 5. Low-temperature heat capacity (C/R) of $Nd_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown. Inset: The expanded plot showing the occurrence double magnetic transition ($T_{N_1}=9.5$ K and $T_{N_2}=9.1$ K) in the compound.

conduction electrons and filled electron orbitals. C_l and C_e for a rare-earth member can be estimated from the nonmagnetic analog, La- or Lu-based member. Our attempts to synthesize $La_2Ni_3Si_5$ did not succeed and therefore, the heat capacity of $Lu_2Ni_3Si_5$ was taken to represent C_l and C_e. Though this material forms with a monoclinic variation of $U_2Co_3Si_5$-type structure, to a first-order approximation we assume that this distortion would not significantly alter the values of C_l and C_e.

Since $Lu_2Ni_3Si_5$ 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 $T\rightarrow 0$ 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 $R_2Ni_3Si_5$ was obtained by subtracting heat capacity of $Lu_2Ni_3Si_5$ from the heat capacity of $R_2Ni_3Si_5$. 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,[15] is made. In the calculations, the fact that one mole of $R_2Ni_3Si_5$ contains two moles of rare-earth ions has been taken into account.

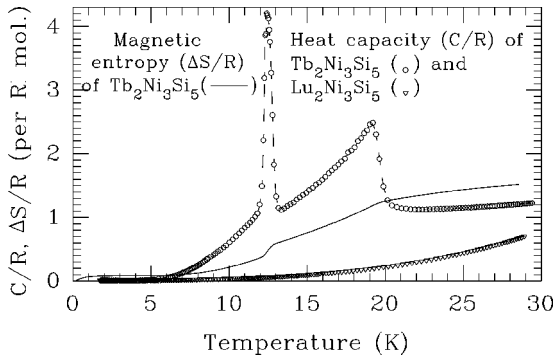

FIG. 6. Low-temperature heat capacity (C/R) of $Tb_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown.

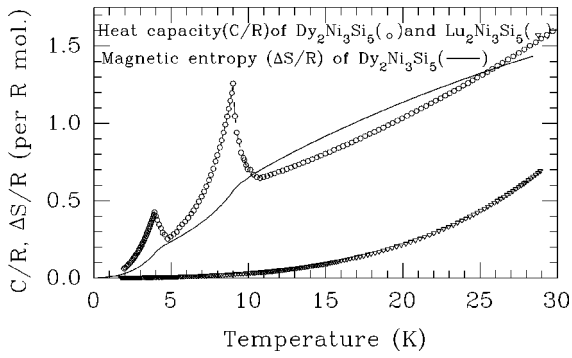

FIG. 7. Low-temperature heat capacity (C/R) of $Dy_2Ni_3Si_5$(○). The solid line represents the magnetic contribution of the entropy, ΔS. For comparison, C/R of $Lu_2Ni_3Si_5$(▽) is also shown.

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 ΔS at the T_N and at 28 K (the maximum temperature of our measurement) for different members of the $R_2Ni_3Si_5$ series, along with the total ΔS 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 ΔS at the higher T_N.

In the case of $Gd_2Ni_3Si_5$ where no crystal-field effects (CEF) would be present, ΔS at $T_N(\sim 1.88R)$, is slightly less than the expected total ΔS, $[J=7/2,\ R\ln 8(=\sim 2.08R)]$ which it very closely reaches as a plateau at 28 K $(\sim 2.02R)$. For $Tb_2Ni_3Si_5$ ΔS $(\sim 1.13R)$ 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 $R\ln 4$

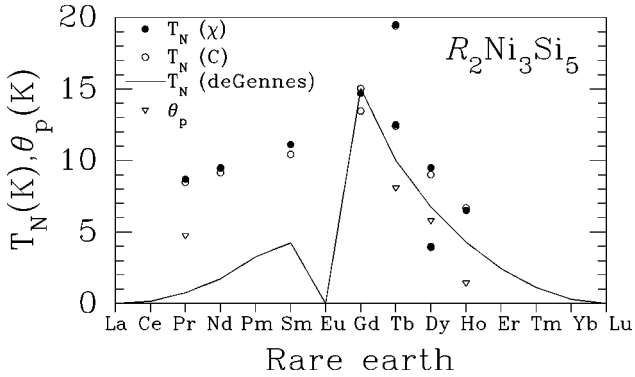

FIG. 8. Magnetic ordering temperatures for $R_2Ni_3Si_5$ compounds. (●) symbols represent the ordering temperatures as the occurrence of a peak in the magnetic-susceptibility measurement and (○) represents the ordering temperatures as the occurrence of a peak in the heat-capacity data. The solid line represents the ordering temperatures expected from the de Gennes scaling when J is a good quantum number. The higher ordering temperature (15 K) of $Gd_2Ni_3Si_5$ is used as the reference temperature. The paramagnetic Curie temperature Θ_p whenever positive, are represented by ▽. Since Θ_p is negative for $Nd_2Ni_3Si_5$, $Gd_2Ni_3Si_5$, and $Ho_2Ni_3Si_5$, they are not shown in this figure. Note that the de Gennes scaling breaks down severely in the case of lighter rare-earth members as well as in the case of T_{N_1} of $Tb_2Ni_3Si_5$.

TABLE I. Magnetic properties, including the magnetic entropies, of different members of the $R_2Ni_3Si_5$ series. T_N values from de Gennes scaling, $(g_J-1)^2J(J+1)$, were obtained with $Gd_2Ni_3Si_5$ as a reference material and J as good quantum number.

Compound	T_{N_1} (K)	T_{N_2} (K)	T_N (K) de Gennes	CW fit of χ data μ_{eff}/R (μ_B)	Θ_p (K)	Range (K)	μ 6 K, 5.5 T (μ_B)	$\Delta S_{\text{mag}}/R$ T_{N_1}	$\Delta S_{\text{mag}}/R$ T_{N_2}	$\Delta S_{\text{mag}}/R$ 28 K	$\Delta S_{\text{mag}}/R$ $\ln(2J+1)$
$Pr_2Ni_3Si_5$	8.5		0.8	3.67	4.7	9–300	1.15	0.38		0.78	2.20
$Nd_2Ni_3Si_5$	9.5	9.1	1.7	3.75	−14.3	30–300	1.21	0.57	0.50	1.07	2.30
$Sm_2Ni_3Si_5$	10.4		4.2				0.03	0.52		0.63	1.79
$Gd_2Ni_3Si_5$	15.0	13.5	15.0	8.11	−15.0	16–300	2.73	1.88	1.70	2.02	2.08
$Tb_2Ni_3Si_5$	19.4	12.4	10.0	9.89	8.1	80–300	2.50	1.21	0.45	1.51	2.56
$Dy_2Ni_3Si_5$	9.0	3.9	6.8	10.79	5.8	60–300	6.36	0.562	0.16	1.42	2.77
$Ho_2Ni_3Si_5$	6.7		4.3	10.47	1.4	7–300	5.99	0.73		1.83	2.83

(=1.39), and therefore, it is likely to be a quartet state. In the case of $Ho_2Ni_3Si_5$, $\Delta S(\sim 0.73)$ at T_N is very close to $R\ln 2$ and hence the ground state could be a doublet. In the cases of $R_2Ni_3Si_5$ (R=Nd, Sm, Dy), the ΔS at $T_N(\sim 0.57, 0.52, 0.56$, respectively) is less but closer to that expected for a doublet ground state. In the case of $Pr_2Ni_3Si_5$, ΔS (~ 0.38) is considerably less than $R\ln 2$. ΔS attains this value at ~ 20 K, which is much above T_N.

One mechanism which leads, at T_N, to a value of $\Delta S < R\ln 2$, is the Ising model. A calculation based on a three-dimensional Ising model for spin-1/2 system on a diamond lattice ($z=4$) predicts a magnetic entropy, $\Delta S = 0.511R$.[16] A similar value of entropy at $T_N(\sim 0.5R)$ had earlier been seen in a few members of $R_2Fe_3Si_5$ series which have been described as Ising systems.[15] It may be noted that ΔS at T_N for $Pr_2Ni_3Si_5$ is less than even this value.

A more likely alternate reason, which is applicable to all the cases discussed here, (as a reduced entropy at T_N than that expected from the suggested respective ground states, is seen in all the cases except that of $Ho_2Ni_3Si_5$) is due to a precursor effect of magnetic order arising from the short-range magnetic correlations that may occur just above T_N. We had suggested, from our magnetoresistance (MR) measurements on these compounds, the existence of short-range ferromagnetic correlations in all the compounds discussed here.[9,10,12] Precursor effects have earlier been observed in other systems such as $Gd_{1-x}Y_xNi_2Si_2$ also.[17] The case of $Ho_2Ni_3Si_5$, where also, MR has shown the existence of precursor effects of magentic order does not fit in this picture. However, as discussed later, ΔS below T_N in this case is anomalous and the actual ΔS may be higher than our extrapolated ΔS estimate.

In all these cases, except for $Gd_2Ni_3Si_5$, ΔS at 28 K is considerably less than the calculated saturation value of ΔS, for the respective cases. This suggests crystalline electric field (CEF) splitting of electronic levels in these compounds. The continuous increase of ΔS above T_N in the cases of $R_2Ni_3Si_5$ (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 $Sm_2Ni_3Si_5$ the well-defined plateau of ΔS 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 $\Delta S = 0.63R \approx R\ln 2$, confirming a doublet ground state. A closer value of ΔS in the case of $Gd_2Ni_3Si_5$ 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, $Nd_2Ni_3Si_5$, $Sm_2Ni_3Si_5$, and $Tb_2Ni_3Si_5$ 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 $Gd_2Ni_3Si_5$ (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 $Gd_2Ni_3Si_5$,[9] below its magnetic ordering temperature.

With respect to the double magnetic transition observed in $Tb_2Ni_3Si_5$, we point out that double magnetic transition has also been seen in $Tb_2Fe_3Si_5$ (Ref. 18) and in $TbNi_2Si_2$.[19] From neutron-scattering experiments on $Tb_2Fe_3Si_5$ (Ref. 20) and on $TbNi_2Si_2$,[19] 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 $Tb_2Ni_3Si_5$.[21] 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 λ 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.[15]

In the case of $Dy_2Ni_3Si_5$ which also shows double magnetic transition (Fig. 7) both the peaks appear to be λ-like which is unlike that of $Tb_2Ni_3Si_5$ and indicates that the nature of the transition at lower magnetic transition temperature is different in these two materials.[21]

Another notable feature in our results, is the unusual behavior of C_m in $Gd_2Ni_3Si_5$ and $Ho_2Ni_3Si_5$ 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 $Ho_2Ni_3Si_5$, we observe an upward deviation from T^3 behavior below 5 K. C_m

of $Ho_2Ni_3Si_5$ 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 $Ho_2Fe_3Si_5$ also.[15] The origin of this deviation is not clear at present.[24]

In the case of $Gd_2Ni_3Si_5$, 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 $Gd_2Fe_3Si_5$ (Ref. 15) (tetragonal structure) and in the cases of other materials, such as $GdCu_2Si_2$, $GdNi_2Si_2$, $GdGa_2$, and $GdCu_5$.[25] In the case of $Gd_2Fe_3Si_5$, 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).[15]

Tsay *et al.*[26] and Lai *et al.*[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.[28] Fishman and Liu[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 $T^*=\frac{2}{3}T_N$. However, in the intermetallics compounds, such as above mentioned $Gd_2Fe_3Si_5$, $GdCu_2Si_2$, and in the present case of, $Gd_2Ni_3Si_5$, T^* is much smaller ($T^*/T_N \sim 0.25$) than predicted by this theory. Moreover, the model is for ferromagnets and our materials are antiferromagnetic. Therefore, even though Tsay *et al.*[26] and Lai *et al.*[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.*[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, $GdCu_2Si_2$, $GdNi_2Si_2$, $GdGa_2$, 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[29]

$$C_m(T)=\Delta C_m+C_m'\frac{T-T_N}{T_N}+\cdots, \tag{3.2}$$

where the ΔC_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_m-T curve just below T_N. The shape of the hump would depend on the strength of the Fourier transform of the exchange integration.[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[29] for an amplitude-modulated system

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

and for the equal moment systems

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

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 $Gd_2Ni_3Si_5$ 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.[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., $Tb_2Fe_3Si_5$,[20] $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$.[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 $R_2Ni_3Si_5$ (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 $Tb_2Ni_3Si_5$ and $Dy_2Ni_3Si_5$ seen earlier through susceptibility studies, they have also revealed the occurrence of double magnetic transitions in $Nd_2Ni_3Si_5$ and $Gd_2Ni_3Si_5$ 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 $Tb_2Ni_3Si_5$ from de Gennes scaling is rather large.

The magnetic entropy estimate in these materials suggest that in all these materials, except for $Gd_2Ni_3Si_5$ (where there is no CEF) and $Tb_2Ni_3Si_5$, the ground state of the rare-earth

ion is a doublet. In the case of $Tb_2Ni_3Si_5$, it appears to be a quartet. In all these cases, ΔS 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.[10] These results also suggest that CEF effects are not likely to be the cause of GMR observed in $Nd_2Ni_3Si_5$, $Sm_2Ni_3Si_5$, and $Tb_2Ni_3Si_5$. Another important feature of our results is the observation of a hump in the heat capacity of $Gd_2Ni_3Si_5$ below its T_N, which may be due to amplitude-modulated magnetic spin structure.

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