Simulation of the magnetocaloric effect by means of theoretical models in Gd_3Ni_2 and Gd_3CoNi systems

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Abstract In this work, using a scaling data method based on the mean-field theory (MFT), magnetization isotherms $M(H, T)$ and magnetic entropy change $-\Delta S_M(T)$ curves have been simulated using MFT combined with the Bean–Rodbell model which verified the second-order phase transition for Gd_3Ni_2 and Gd_3CoNi compounds. Under a low value of magnetic field H , $M(T)$ and $-\Delta S_M(T)$ curves have been successfully simulated and some magnetocaloric properties have been evaluated using the phenomenological model.

1 Introduction

The search for new materials with a large refrigerant capacity (RC) and high magnetocaloric effect (MCE) that can be applied in room and low-temperature magnetocaloric refrigerators is still one of the actual research topics advanced by the low-energy consumption and environmental impact of these devices [1, 2]. Among them, we cited intermetallic systems composed of rare earth (R) and transition metal (M) such as ‘ $\text{Nd}_2\text{Co}_{1.7}$ ’, ‘ Gd_3Ni_2 ’ and ‘ Lu_2Pd_5 ’ ... [3–5]. Recently, Provino et al. [6] have reported the crystal structure, thermal stability, magnetic behavior and MCE of Gd_3Ni_2 and Gd_3CoNi compounds.

As shown in the work of Provino et al. for Gd_3Ni_2 and Gd_3CoNi compounds [6], the magnetic entropy change ($-\Delta S_M$) increases proportionally to $H^{2/3}$ (H is the applied magnetic field). This is expected from the mean-field theory (MFT).

In this way, a scaling data method based on MFT [7, 8] and a phenomenological model proposed by Hamad [9] are exploited to study the MCE. First, following the scaling method [10] based on the MFT combined with the Bean–Rodbell model [11], we have estimated the exchange parameter (λ), the saturation magnetization (M_0), the total angular momentum (J) and the gyromagnetic factor (g) of Gd_3Ni_2 and Gd_3CoNi materials. These obtained parameters are used to simulate M vs. H , M vs. T and $-\Delta S_M$ vs. T curves. Second, the phenomenological model is exploited to simulate $M(T)$, $-\Delta S_M(T)$ curves and to calculate the full width at maximum in $-\Delta S_M(T)$ curve (δT_{FWHM}) giving the relative cooling power (RCP). Simulated results are compared to the experimental ones.

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2 Theoretical considerations

2.1 Mean-field theory

Based on the MFT, magnetization values can be calculated as a function of the applied field H , the temperature T and the exchange mean field H_{exch} as [8, 10]:

$$M(x) = M_0 B_J(x), \quad (1)$$

with $M_0 = NJg\mu_B$, the saturation magnetization, $B_J(x) = \frac{2J+1}{2J} \coth\left(\frac{2J+1}{2J}x\right) - \frac{1}{2J} \coth\left(\frac{x}{2J}\right)$, the Brillouin function, $x = \frac{Jg\mu_B}{k_B} \left(\frac{H+H_{\text{exch}}}{T}\right)$, $J = |L \pm S|$ is the total angular momentum and $g = 1 + \frac{J(J+1)+S(S+1)-L(L+1)}{2J(J+1)}$ is the gyromagnetic factor with L and S being, respectively, the orbital and the spin momentums.

By taking $x' = \frac{H+H_{\text{exch}}}{T}$, Eq. (1) can be written as:

$$\begin{aligned} M(H, T) &= f\left(\frac{H+H_{\text{exch}}}{T}\right) = f(x') \\ &= M_0 \left[\frac{2J+1}{2J} \coth\left(\frac{2J+1}{2J} \frac{Jg\mu_B}{k_B} x'\right) - \frac{1}{2J} \coth\left(\frac{1}{2J} \frac{Jg\mu_B}{k_B} x'\right) \right] \end{aligned} \quad (2)$$

Applying the reciprocal function f^{-1} of f on the first term of Eq. (2), we can get:

$$\frac{H}{T} = f^{-1}(M) - \frac{H_{\text{exch}}}{T}. \quad (3)$$

The magnetic entropy is given by the thermo-magnetic relations as [12]:

$$\left(\frac{\partial S}{\partial H}\right)_T = \left(\frac{\partial M}{\partial T}\right)_H \quad (4)$$

$$\left(\frac{\partial S}{\partial M}\right)_T = \left(\frac{\partial H}{\partial T}\right)_M. \quad (5)$$

From the $M(H, T)$ data and using Eq. (4), the magnetic entropy change arising when the applied magnetic field changes from 0 to H can be calculated as [13]:

$$\Delta S_M(T, \Delta H) = S_M(T, H) - S_M(T, 0) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH. \quad (6)$$

In the case of magnetization measurements at small discrete field and temperature intervals, numerical approximation to the integral in Eq. (6) could be expressed as [13, 14]:

$$\Delta S_M(T, \Delta H) = \sum \frac{M_i - M_{i+1}}{T_i - T_{i+1}} \Delta H_i, \quad (7)$$

where M_i and M_{i+1} are the magnetization values measured under a magnetic field ΔH_i , at T_i and T_{i+1} , respectively.

Equations (3) and (5) allow us to estimate the magnetic entropy change:

$$-\Delta S_M(T)_{H_1 \rightarrow H_2} = \int_{M|_{H_1}}^{M|_{H_2}} \left(f^{-1}(M) - \left(\frac{\partial H_{\text{exch}}}{\partial T}\right)_M \right) dM. \quad (8)$$

On the other hand, the Bean–Rodbell model appends the magneto-volume effects to the classical Weiss mean-field theory [11]. According to this model, the exchange interactions

depend powerfully on the interatomic distances. The critical temperature T_C is given as follows:

$$T_C = T_0(1 + \beta\omega), \quad (9)$$

where $\omega = \frac{v-v_0}{v_0}$ with v and v_0 being, respectively, the sample volume with and without exchange interactions and T_0 is the transition temperature. The parameter β controls the spin–lattice coupling strength. The Gibbs free energy is given then as [8]:

$$G = -\frac{3}{2} \left(\frac{J}{J+1} \right) N k_B T_C \sigma^2 - H g J \mu_B N \sigma + \frac{1}{2K} \omega^2 - T S, \quad (10)$$

with $\sigma(x) = B_J(x)$, S and K being, respectively, reduced magnetization, the magnetic entropy and the compressibility.

The above free energy is minimized ($\frac{dG}{d\omega} = 0$) at:

$$\omega = \frac{3}{2} \left(\frac{J}{J+1} \right) N k_B K T_0 \beta \sigma^2. \quad (11)$$

The following magnetic state equation can be obtained after minimizing G , with respect to σ , [11, 12]:

$$\sigma(Y) = B_J(Y), \quad (12)$$

with

$$Y = \frac{1}{T} \left[3T_0 \left(\frac{J}{J+1} \right) \sigma + \frac{g J \mu_B}{k_B} H + \frac{9}{5} \frac{(2J+1)^4 - 1}{[(2J+1)]^4} T_0 \eta \sigma^3 \right], \quad (13)$$

where the parameter η checks the nature of the magnetic phase transition: a first order for $\eta > 1$ and a second order for $\eta < 1$ [15, 16].

2.2 Phenomenological model

Based on the phenomenological model [9, 17], the dependence of magnetization on temperature under a low magnetic field is represented as:

$$M(T) = \left(\frac{M_i - M_f}{2} \right) \tanh[A(T_C - T)] + BT + C, \quad (14)$$

with M_i and M_f being, respectively, the initial and final values of magnetization at ferro-magnetic–paramagnetic transition, $A = \frac{2(B-S_c)}{M_i - M_f}$, B is the magnetization sensitivity $\frac{dM}{dT}$ in the ferromagnetic region before transition, S_c is the magnetization sensitivity $\frac{dM}{dT}$ at T_C and $C = \frac{M_i - M_f}{2} - BT_C$; and the magnetic entropy change of a magnetic system under arising magnetic field from 0 to final value $H_{\max}$ is given by:

$$\Delta S_M(T) = \left[-A \left(\frac{M_i - M_f}{2} \right) \operatorname{sech}^2(A(T_C - T)) + B \right] H_{\max}. \quad (15)$$

ΔS_M reaches its maximum at $T = T_C$. Such a maximum can be evaluated as:

$$\Delta S_M^{\max} = \left[-A \left(\frac{M_i - M_f}{2} \right) + B \right] H_{\max}. \quad (16)$$

Fig. 1 Isotherms M vs. H at different temperatures T for **a** Gd_3Ni_2 and **b** Gd_3CoNi [6]

The RCP is given by:

$$\text{RCP} = -\Delta S_M^{\max} \times \delta T_{FWHM} = \left[M_i - M_f - 2 \frac{B}{A} \right] H_{\max} \times \cosh^{-1} \sqrt{\frac{2A(M_i - M_f)}{A(M_i - M_f) + 2B}}, \quad (17)$$

with $\delta T_{FWHM} = \frac{2}{A} \cosh^{-1} \sqrt{\frac{2A(M_i - M_f)}{A(M_i - M_f) + 2B}}$ being the full width at half maximum in $(-\Delta S_M(T))$ curve.

3 Results and discussion

Figure 1a, b show experimental isotherms $M(H, T)$, respectively, for Gd_3Ni_2 and Gd_3CoNi compounds reported by Provino et al. [6]. Measures have been taken near Curie temperature, $T_C = 147$ K for Gd_3Ni_2 and $T_C = 176$ K for Gd_3CoNi [6]. It is clear from Fig. 1a, b that the substitution of Ni in Gd_3Ni_2 by Co in Gd_3CoNi favors magnetization values. Based on the work of Pecharsky et al. [18], the saturation moment reaches $7.5 \mu_B$ per Gd for Gd_3Al_2 under 5.5 T applied field and at 5.04 K temperature. However for Gd_3Ni_2 , it has been found $7 \mu_B$ per Gd under 6.5 T applied field and at 5 K temperature [6]. As known, Ni is magnetic but Al is not. So, the substitution of Al by Ni contributes to decrease magnetization values.

To determine the mean-field exchange (H_{exch}) for Gd_3Ni_2 and Gd_3CoNi compounds, we have used the scaling data method [7]. This method consists of plotting the evolution of $\frac{H}{T}$ vs. $\frac{1}{T}$, taken at constant values of magnetization M (10 emu g^{-1} step) from 110 to 200 K, using Fig. 1a for Gd_3Ni_2 , and from 125 to 250 K, using Fig. 1b for Gd_3CoNi . The evolution of $\frac{H}{T}$ vs. $\frac{1}{T}$ is shown in Fig. 2a, b for Gd_3Ni_2 and Gd_3CoNi , respectively. From Eq. (3), the slope of each isomagnetic line in Fig. 2a, b will give H_{exch} . The values of H_{exch} are plotted vs. M in Fig. 3a, b for Gd_3Ni_2 and Gd_3CoNi , respectively. These figures show the evolution of H_{exch} vs. M , fitted by $H_{\text{exch}} = \lambda_1 M + \lambda_3 M^3$ equation [18]. Each adjustment shows a negligible dependence on M^3 value ($\lambda_3 = -3.610^{-5}$ and $-2.810^{-5} (\text{T emu}^{-1} \text{g})^3$ for Gd_3Ni_2 and Gd_3CoNi , respectively), so we can assume that $H_{\text{exch}} = \lambda_1 M \approx \lambda M$, with $\lambda_1 = 0.23$ and $0.31 \text{ T emu}^{-1} \text{g}$ for Gd_3Ni_2 and Gd_3CoNi , respectively. The second step of this method consists of building the scaling plot of M vs. $\frac{H+H_{\text{exch}}}{T}$ which is shown in Fig. 4a, b for Gd_3Ni_2 and Gd_3CoNi , respectively, with black symbols. All curves of Fig. 4a,

Fig. 2 $\frac{H}{T}$ vs. $\frac{1}{T}$ curves with constant values of M per curve for **a** Gd_3Ni_2 and **b** Gd_3CoNi

Fig. 3 H_{exch} vs. M fitted by $\lambda_1 M + \lambda_3 M^3$ function for **a** Gd_3Ni_2 and **b** Gd_3CoNi

Fig. 4 Scaled data in M vs. $\frac{H+H_{\text{exch}}}{T}$ and Brillouin function fit for **a** Gd_3Ni_2 and **b** Gd_3CoNi

b, converging in one curve, are fitted by Eq. (2), using MATLAB software to determine M_0 , J and g . A good agreement between adjusted and theoretical parameters J and g is observed (Table 1). This agreement confirms the coupling state between sublattices suggested above.

Table 1 Adjusted parameters of Gd_3Ni_2 and Gd_3CoNi samples given by scaling data

Samples	g	J	$M_0(\text{emu g}^{-1})$	$(T \text{ emu}^{-1} \text{ g})$
Gd_3Ni_2	2.03	3.45	187.9	0.23
Gd_3CoNi	2.12	3.49	204.6	0.31

Fig. 5 Comparison between simulated and experimental M vs. H curves. Red solid lines are simulated results using Eq. (1) and black symbols are experimental data [6] for **a** Gd_3Ni_2 and **b** Gd_3CoNi

The adjusted values λ , M_0 , J and g are injected in Eq. (1) to generate simulated magnetization isotherms $M(T, H)$, indicated by red lines in Fig. 5a, b for Gd_3Ni_2 and Gd_3CoNi , respectively. As shown in this figure, generated curves correlated well with the experimental ones (black symbols).

The use of the Bean–Rodbell model started by replacing x in Eq. (1) with Y (see Eq. 13) and choosing an average value of η and T_0 to see a correlation between generated and experimental $M(T)$ curves. The generated $M(T)$ curves (red lines) correlated with the experimental ones [6] (black symbols), as shown in Fig. 6a, b, after taking average values: $\eta = 0.1$ and $T_0 = 147$ K for Gd_3Ni_2 , whereas for Gd_3CoNi , $\eta = 0.01$ and $T_0 = 176$ K. Since $\eta < 1$ for these two compounds, the second-order transition is confirmed. Using the Mean-Field Simulation Suite software [19], Eq. (8) may be resolved. Such resolution allows us to establish the evolution of $-\Delta S_M(T)$ curves (red lines), which are represented in Fig. 7a, b with experimental $-\Delta S_M(T)$ curves (black symbols), using Eq. (7), for Gd_3Ni_2 and Gd_3CoNi , respectively. A significant shift between theoretical (red lines) and experimental (black symbols) $-\Delta S_M(T)$ is observed in this figure, probably due to the roughness of the mean-field model to describe accurately well the magnetization of system having more than one magnetic system (Gd, Ni, Co). We should note that this model did not take into account the roles played by the Jahn–Teller effect and the charge ordering, which affect the MCE value even for the case of conventional ferromagnetic systems [20, 21]. Nonetheless, this model has been useful to describe qualitatively $-\Delta S_M(T)$, particularly at temperatures near to T_C [22]. Such discrepancy between the theoretical calculations and the experimental data was also reported by Balli et al. [23].

Figure 8a, b, reported in the work of Provino et al. [6], presents the variation in magnetization versus temperature, $M(T)$, for Gd_3Ni_2 and Gd_3CoNi , respectively, in 2–300 K temperature range, under 0.02 T applied magnetic field, in zero field cooling (ZFC) mode. The inset in each figure shows the derivative of M vs. T . These two figures are treated

Fig. 6 Comparison between simulated and experimental M vs. T curves. Red solid lines are simulated results using Eqs. (1 and 11) and black symbols are experimental data [6] for **a** Gd_3Ni_2 and **b** Gd_3CoNi

Fig. 7 Comparison between simulated and experimental $-\Delta S_M$ vs. T curves under 1, 2, 3, 4 and 5 T (from the bottom to the top). Red solid lines are simulated results using Eq. (6b) and black symbols are experimental data using Eq. (6a) for **a** Gd_3Ni_2 and **b** Gd_3CoNi

Fig. 8 M vs. T (the inset: $\frac{dM}{dT}$ vs. T) under 0.02 T applied magnetic field in ZFC for **a** Gd_3Ni_2 and **b** Gd_3CoNi [6]

Table 2 Experimental parameters served for the application of the phenomenological model

Samples	M_i (emu g ⁻¹)	M_f (emu g ⁻¹)	T_C (K)	B (emu g ⁻¹ K ⁻¹)	S_C (emu g ⁻¹ K ⁻¹)
Gd ₃ Ni ₂	8.002	0.5	154.1	- 0.0003282	0.5041
Gd ₃ CoNi	13.24	0.62	175.9	- 0.002662	1.722

Fig. 9 Comparison between simulated and experimental M vs. T curves under 0.02 T applied magnetic field. Red solid lines are simulated results using Eqs. (12) and black symbols are experimental data [6] for **a** Gd₃Ni₂ and **b** Gd₃CoNi **Fig. 10** Comparison between simulated and experimental $-\Delta S_M$ vs. T curves under 0.02 T applied magnetic field. Red solid lines are simulated results using Eq. (13) and black symbols are experimental data using Eq. (5) for **a** Gd₃Ni₂ and **b** Gd₃CoNi

determine M_i , M_f (from $M(T)$ curves) and T_C , B , S_C (from $\frac{dM}{dT}$ vs. T curves) described above. Such determined parameters needed for the simulation of magnetic properties have been recapitulated in Table 2. Simulated curves (red lines) of M versus T , obtained using Eq. (14), agree well with the experimental ones (black symbols) as shown in Fig. 9a, b for Gd₃Ni₂ and Gd₃CoNi, respectively. In comparison, the correlation between simulated and experimental $M(T)$ curves under low magnetic field obtained from the phenomenological model is better than the ones deduced after using the mean-field theory combined by the Bean–Rodbell model. This may suggest that the phenomenological model can complete the MFT under low magnetic applied fields.

Table 3 Comparison between magnetocaloric properties of Gd₃Ni₂ and Gd₃CoNi systems with those of other compounds

Compound	H (T)	$-\Delta S_M^{\max}$ (J kg ⁻¹ K ⁻¹)	δT_{FWHM} (K)	RCP (J kg ⁻¹)
Gd ₃ Ni ₂ (this work)	0.02	0.009	12.03	0.108
Gd ₃ CoNi (this work)	0.02	0.022	12.11	0.266
(001)-oriented MnAs film [24]	0.03	0.01	20.03	0.2
La _{0.8} Sr _{0.05} Ca _{0.15} MnO ₃ [25]	0.05	0.028	17.192	0.475
Ge _{0.95} Mn _{0.05} film [26]	0.1	(3.2) 10 ⁻⁶	5.6	(17.2) 10 ⁻⁶
GdCaBaCo ₂ O _{5.5} [27]	0.1	(0.55) 10 ⁻⁶	4.08	(2.2) 10 ⁻⁶

Generated $-\Delta S_M(T)$ curves (red lines), in Fig. 10a, b for Gd₃Ni₂ and Gd₃CoNi, respectively, can be determined using Eq. (15). They are correlated with the experimental ones (black symbols) determined using Eq. (7). The $-\Delta S_M^{\max}$, the δT_{FWHM} and the RCP for Gd₃Ni₂ and Gd₃CoNi under 0.02 T are calculated using Eqs. (16) and (17). They are compared to those of other compounds in Table 3. We have noticed from Table 3 that these magnetic properties are comparable with those of (001)-oriented MnAs film [24] and La_{0.8}Sr_{0.05}Ca_{0.15}MnO₃ [25]. While they are significantly larger than the ones of Ge_{0.95}Mn_{0.05} film [26] and GdCaBaCo₂O_{5.5} [27].

4 Conclusion

Based on the MFT, the scaling data method allows us to investigate the coupling between sublattices of Gd₃Ni₂ and Gd₃CoNi systems. Simulated M vs. H , M vs. T and $-\Delta S_M$ vs. T curves are achieved using the MFT combined with the Bean–Rodbell model. A good agreement between simulated and experimental M vs. H , M vs. T curves is observed. While, a significant shift has been noted between simulated and experimental $-\Delta S_M$ vs. T curves probably due to the inability of the MFT to describe accurately well the magnetization of system having more than one magnetic (Gd, Co and Ni). Under low value of magnetic field H , the phenomenological model was exploited to simulate successfully M and $-\Delta S_M$ versus T and to evaluate some magnetocaloric properties.

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