Monte Carlo calculations of the magnetocaloric effect in RAl$_2$ (R=Dy,Er)

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In this work we calculate the magnetocaloric effect in the Laves phase compound RAl$_2$ by using a model Hamiltonian of interacting spins where the spin-spin interaction is treated in the Monte Carlo simulation. The isothermal entropy change and the adiabatic temperature change upon magnetic field variations, for the compounds DyAl$_2$ and ErAl$_2$ are in good agreement with the available experimental data. © 2006 American Institute of Physics. [DOI: 10.1063/1.2150815]

I. INTRODUCTION

The magnetocaloric effect,[1–3] is characterized by the isothermal entropy change (ΔS) and the adiabatic temperature change (ΔT_{ad}) upon magnetic field variation. From the theoretical point of view, the description[4–9] of the magnetocaloric effect in rare earth based compounds has been made by using a Heisenberg model Hamiltonian in which the spin-spin interaction is treated in the molecular field approximation. Although the mean field approximation explains many experimental data of the magnetocaloric quantities ΔS and ΔT_{ad}, it fails in explaining the magnetic part of the heat capacity around the magnetic ordering temperature. This discrepancy between theory and experiment, which occurs because the molecular field theory does not take into account short range interactions, points out that we should go beyond the mean field approximation to understand the real physical mechanisms involved in the magnetocaloric effect in rare earth based compounds.

II. THEORETICAL CALCULATION

In this work, we use classical Monte Carlo simulation[10,11] to calculate the magnetocaloric effect in the compounds RAl$_2$ (R=Er and Dy). In order to do so, we start with the following energy:

$$E = -\sum_{il} \lambda_{il} \mathbf{J}_i \cdot \mathbf{J}_l + \sum_i \zeta_i (\mathbf{r}_i \cdot \mathbf{J}_i)^2 - \sum_i g\mu_B \mathbf{J}_i \cdot h^{\text{ext}}, \quad (1)$$

where λ_{ij} is the exchange interaction parameter between neighboring sites, $\mathbf{J}_i$ is the total angular momentum of the R ions and h^{ext} is the external magnetic field. The second term represents the single ion anisotropy.

In order to calculate the energy given in Eq. (1), via the Monte Carlo simulation, we use a Potts-like model,[10,11] where the z components of the total angular momentum are

considered as quantum quantities, which can assume discrete values in the interval $-J \leq J^Z \leq J$. Within this consideration the maximum value of the magnetic entropy $S_{\text{mag}} = \Re \ln(2J+1)$, where $\Re$ is the gas constant. For a given temperature, the mean energy $\langle E \rangle$ is calculated by

$$\langle E \rangle = \frac{1}{(N_c - N_0)} \sum_{i>N_0}^{N_C} E_i, \quad (2)$$

where E_i is the energy of the last spin configuration of a given Monte Carlo step. N_c represents the total number of Monte Carlo steps and N_0 is the number of Monte Carlo steps used for thermalization. A similar relation holds for the mean square energy $\langle E^2 \rangle$. The mean value of the total angular momentum per lattice site is calculated by

$$\langle J \rangle_\alpha = \frac{1}{(N_c - N_0)} \sum_{i>N_0}^{N_C} \left[\frac{1}{N_S} \sum_{k=1}^{N_S} J_k \right]_\alpha, \quad (3)$$

where $\alpha = x, y, z$. In Eq. (3), the label "i" represents the Monte Carlo step and the label "k" represents the lattice sites. N_S represents the number of lattice sites and J_k is the spin at each lattice site. The total heat capacity is given by: $C(T, h^{\text{ext}}) = C_{\text{mag}}(T, h^{\text{ext}}) + C_{\text{el}}(T) + C_{\text{lat}}(T)$. The magnetic heat capacity (C_{mag}), per magnetic ions, is given by

$$C_{\text{mag}}(T, h^{\text{ext}}) = \frac{\langle E^2 \rangle - \langle E \rangle^2}{k_B T^2}. \quad (4)$$

$C_{\text{el}} = \gamma T$ is the contribution from the conduction electrons, where γ is the Sommerfeld coefficient. C_{lat} is the contribution from the crystalline lattice which is taken here in the Debye approximation as

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$$C_{\text{lat}}(T) = 9\Re N_i \left[4 \left(\frac{T}{\theta_D} \right)^3 \int_0^{\theta_D/T} \frac{x^3}{(e^x - 1)} dx \right.$$

$$\left. - \left(\frac{\theta_D}{T} \right) \frac{1}{(e^{\theta_D/T} - 1)} \right], \tag{5}$$

where N_i is the number of ions per formula unit and Θ_D is the Debye temperature. The total entropy of the compound is given by: $S(T,h^{\text{ext}}) = S_{\text{mag}}(T,h^{\text{ext}}) + S_{\text{el}}(T) + S_{\text{lat}}(T)$. The contribution from the magnetic ions (S_{mag}) is given by

$$S_{\text{mag}}(T,h^{\text{ext}}) = \int_0^T \frac{C_{\text{mag}}(T,h^{\text{ext}})}{T} dT, \tag{6}$$

where C_{mag} is the magnetic heat capacity calculated in Eq. (4). The contribution from the conduction electrons is taken as $S_{\text{el}} = \gamma T$. The contribution from the crystalline lattice, also taken in the Debye approximation, is given by

$$S_{\text{lat}}(T) = N_i \left[-3R \ln \left(1 - e^{-\frac{\Theta_D}{T}} \right) \right.$$

$$\left. + 12R \left(\frac{T}{\Theta_D} \right)^3 \int_0^{\Theta_D/T} \frac{x^3}{e^x - 1} dx \right]. \tag{7}$$

III. NUMERICAL RESULTS AND CONCLUSIONS

In order to calculate the mean energy and consequently the magnetocaloric quantities ΔS and ΔT_{ad} in the Laves phase compounds DyAl$_2$ and ErAl$_2$ we use a tridimensional cluster of $5 \times 5 \times 5$ cubic unit cells with 8 R ions per cell and consider only first next neighbors interactions. The total angular momentum for both Dy and Er ions is $J=15/2$. The Landè factor for Dy and Er are $g=4/3$ and $g=6/5$, respectively. The exchange interaction parameters λ_{ij}, were chosen to correctly reproduce the experimental data of the magnetic ordering temperature. These parameters are about $\lambda_{ij} = 0.064$ meV for the compound DyAl$_2$ and $\lambda_{ij} = 0.0137$ meV for the compound ErAl$_2$. The anisotropic coefficients for the compounds DyAl$_2$ and ErAl$_2$ were taken as $\zeta_i = 0.0043$ meV and $\zeta_i = 0.0017$ meV, respectively. In order to establish the magnitude of the z components of the total angular momentum of the Dy or Er ions at each lattice site we proceed as follows: We draw a random number r such that $0 < r < 1$ and fix the value of J^z according to the following scheme. If the random number (r) lies in the interval $[0, 1/(2J+1)]$ we take $J^z = -J$. If the random number (r) lies in the interval $[1/(2J+1), 2/(2J+1)]$ we take $J^z = -J+1$; if the random number (r) lies in the interval $[2/(2J+1), 3/(2J+1)]$ we take $J^z = -J+2$; and so forth. The simulation was performed using 2000 Monte Carlo steps for thermalization of the system and more 3000 Monte Carlo steps to compute the average values of the physical quantities. The magnetic part of the heat capacity was calculated using Eq. (4) and the magnetic entropy was obtained from Eq. (6). The electronic heat capacity and electronic entropy were calculated using $\gamma = 5.4$ mJ/(mol K^2). The lattice heat capacity and entropy were calculated using $\Theta_D = 330$ K for the compound DyAl$_2$
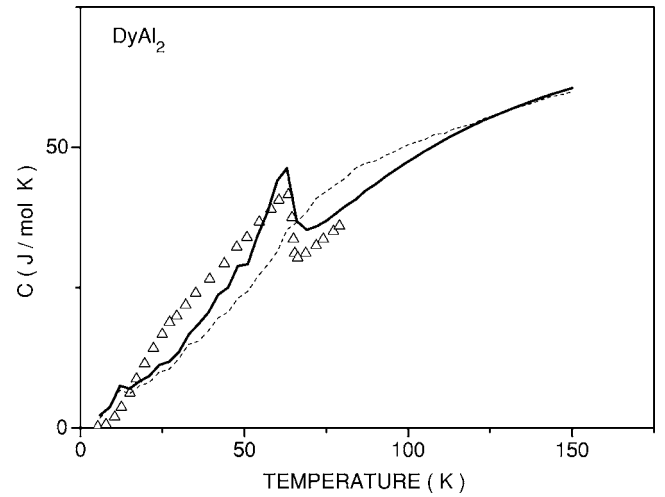
FIG. 1. Temperature dependence of the total heat capacity of the compound DyAl$_2$. The solid and dotted lines represent the calculations within the Monte Carlo simulation for $h^{\text{ext}}=0$ and $h^{\text{ext}}=5$ T, respectively. Open triangles are experimental data (see Ref. 12).

and $\Theta_D = 220$ K for the compound ErAl$_2$. The parameters γ and Θ_D are in the usual range of values used in the literature.[4]

In Fig. 1, we plot the total heat capacity for the intermetallic DyAl$_2$, calculated in the absence of an applied magnetic field and for an applied magnetic field of 5 T. For the compound ErAl$_2$, we obtain a similar curve not shown in this article. From Fig. 1, we observe, a good agreement between our calculations for $h^{\text{ext}}=0$ (solid line) and the corresponding experimental data[12] (open triangles). Further experimental data are necessary to compare with our theoretical calculations for $h^{\text{ext}}=5$ T (dotted line). From the total heat capacity curves, we calculate the total entropy for $h^{\text{ext}}=0$ and $h^{\text{ext}}=5$ T. From the total entropy curves, we calculate the isothermal entropy change and the adiabatic temperature change for a magnetic field variation from 0 to 5 T. In Figs. 2 and 3, we plot the isothermal entropy change and the adiabatic temperature change for the compounds DyAl$_2$ and ErAl$_2$ upon
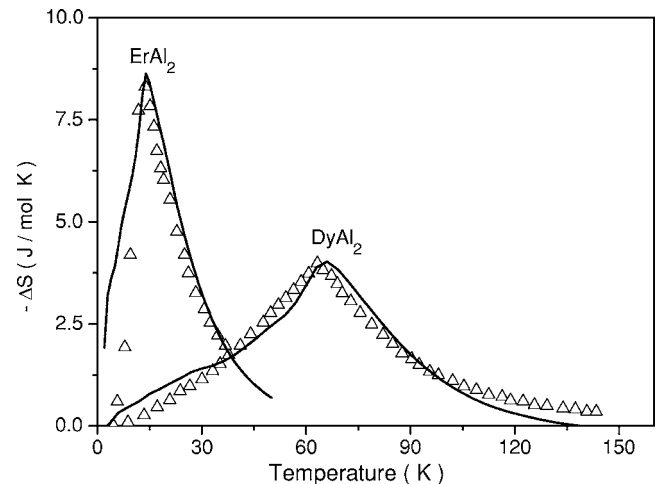
FIG. 2. Isothermal entropy change in the intermetallic compounds DyAl$_2$ and ErAl$_2$ for magnetic field variation from 0 to 5 T. Solid lines correspond to our Monte Carlo calculations whereas open triangles are experimental data (see Ref. 4).
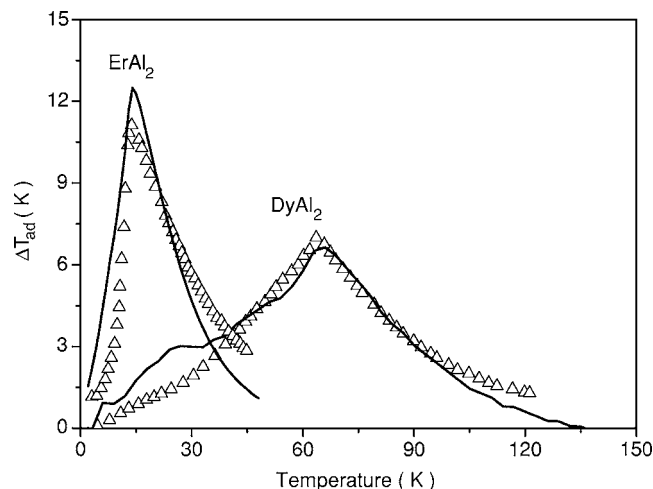
FIG. 3. Adiabatic temperature change in the compound $DyAl_2$ and $ErAl_2$ for magnetic field variation from 0 to 5 T. Solid lines correspond to our Monte Carlo calculations whereas open triangles are experimental data (see Ref. 4).

magnetic field variation from 0 to 5 T. From these figures, we can observe that our theoretical calculations are in good agreement with experimental data.[4]

In conclusion, in this work we discuss the magnetocaloric effect in the laves phase compound RAl_2 by using a model Hamiltonian of interacting spins, where the spin-spin interaction is treated in the Monte Carlo simulation. The Monte Carlo simulation is a powerful tool to study the magnetocaloric effect in rare earth compounds because it takes into account short range interactions which are not well described in the molecular field theory. Our theoretical calculations for the intermetallic compounds $DyAl_2$ and $ErAl_2$ show that the isothermal entropy change and the adiabatic temperature change upon magnetic field variation from 0 to 5 T are in good agreement with experimental data.[4] The present Monte Carlo simulations can be used to calculate the magnetocaloric effect in other compounds of RAl_2 series as well as in, e.g., the RNi_2 series. Besides, the Monte Carlo simulations enable us to calculate the magnetocaloric effect of doped rare earth compounds, such as $(R_{1-x}R'_x)Al_2$ and $(R_{1-x}R'_x)Ni_2$, where the molecular field theory is no longer a good approximation.

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