



Magnetocaloric effect in $(Gd_{1-c}Tb_c)_5Si_2Ge_2$

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ABSTRACT

In this paper we theoretically discuss the magnetocaloric effect in the doped compound $(Gd_{1-c}Tb_c)_5Si_2Ge_2$. To this end we use a model Hamiltonian of localized interacting magnetic moments including the magnetoelastic coupling and the Zeeman interaction. In order to treat the two body interaction we use an extension of the mean field theory in which is considered the type of magnetic rare earth ions is occupying the neighborhood of a given site. The obtained results of the magnetocaloric potentials are in very good agreement with the available experimental data.

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1. Introduction

The magnetocaloric effect is characterized by the isothermal entropy change ΔS_{iso} and by the adiabatic temperature change ΔT_{ad} under magnetic field variation. It is well known that the temperature and the magnitude of the peaks of the magnetocaloric potentials ΔS_{iso} and ΔT_{ad} can be tuned by applying pressure [1–3] and doping [4,5]. A lot of experimental data of the magnetocaloric potentials in doped compounds can be found in the literature [6,7]. However, only a few papers theoretically discuss the magnetocaloric effect in doped compounds [8–10]. In the theoretical description of the magnetocaloric effect in doped compounds, it is necessary to consider the nature of the host and the impurities. For instance, rare earth based compounds doped with non-magnetic impurities such as $R_5Si_2(Ge_{1-c}M_c)_2$, where R is a rare earth element and M is a non-magnetic element may be described by a Heisenberg-like Hamiltonian with an effective exchange interaction parameter renormalized by the disorder introduced into the electron gas. This simplification in the theoretical description of the magnetocaloric effect in $R_5Si_2(Ge_{1-c}M_c)_2$ may be introduced because the magnetism in these doped compounds comes entirely from the rare earth ions and the impurities enter at the non-magnetic sublattice. On the other hand the theoretical description of the magnetocaloric effect in doped compounds with more than one type of magnetic ions

such as $(R^a_{1-c}R^b_c)_5Si_2Ge_2$, where R^a and R^b are rare earth elements is much more complex, because the impurities are introduced at the rare earth magnetic sublattice.

In this work we are only interested in the theoretical description of the magnetocaloric effect in the rare earth doped compound $(Gd_{1-c}Tb_c)_5Si_2Ge_2$, which undergoes a first order phase transition. To this end we use a Heisenberg-like Hamiltonian including the magnetoelastic coupling and the Zeeman interaction. The great difficulty to describe the magnetocaloric effect in this doped compound is associated with the existence of disorder in the rare earth sublattice. The effect of the disorder on the magnetocaloric effect in $(Gd_{1-c}Tb_c)_5Si_2Ge_2$ could be treated by improving the classical Monte Carlo simulation [11,12] to incorporate the nature of the first order phase transition observed in this doped compound. However, such an improvement is not so easy to be done. Besides, due to the great computer time needed to perform Monte Carlo simulation, it is interesting to search for alternative methods of calculations to deal with this type of disorder.

Hence, in order to treat this kind of disordered problem and get an insight in the physical mechanisms involved in the magnetocaloric effect in the doped compound $(Gd_{1-c}Tb_c)_5Si_2Ge_2$, we adopt in this work an extended mean field theory in which the type of rare earth ions is explicitly considered in the two body interaction. Our theoretical calculations of the magnetocaloric potentials are in a very good agreement with the available experimental data. Moreover, our calculations predict a systematic behavior of the magnetocaloric potentials in the doped compound $(Gd_{1-c}Tb_c)_5Si_2Ge_2$ with increasing Tb concentration. Based on this

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prediction, we also propose composite materials made up of some samples of the doped compound $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$ with different Tb concentration, which exhibit the table-like behavior, i.e., an almost constant value of the magnetocaloric potential ΔS_{iso} in a wide range of temperatures.

2. Formulation

In order to discuss the magnetocaloric effect in the doped compound $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$ we start with the following Hamiltonian:

$$\mathcal{H} = -\sum_{i,l} \mathcal{J}_0^{ab} \vec{J}_i^a \cdot \vec{J}_l^b - \sum_{ij} \mathcal{J}_1^{ab} \vec{J}_i^a \cdot \vec{J}_j^b - \sum_i g_i^a \mu_B \vec{B} \cdot \vec{J}_i^a \quad (1)$$

In this Hamiltonian, the first term describes the interaction between rare earth magnetic moments, where $\mathcal{J}_0^{ab}$ ($a, b = \text{Gd}$ or Tb) is the exchange interaction parameter and $\vec{J}_i$ (J_i) is the total angular momentum of the rare earth ions. The second term somehow describes the magnetoelastic interaction where $\mathcal{J}_1^{ab}$ is a model parameter [13]. The last term represents the Zeeman interaction between the rare earth angular momentum and the external magnetic field (B), where g_i^a is the Landé factor. In the mean field approximation, the previous Hamiltonian can be explicitly written in the form $\mathcal{H} = \mathcal{H}_1^{\text{Gd}} + \mathcal{H}_2^{\text{Tb}}$, where

$$\begin{aligned} \mathcal{H}_1^{\text{Gd}} = & -\sum_i \sum_{\alpha=x,y,z} [\tilde{\mathcal{J}}_0^{\text{GdGd}} \langle J_\alpha^{\text{Gd}} \rangle + \tilde{\mathcal{J}}_0^{\text{GdTb}} \langle J_\alpha^{\text{Tb}} \rangle \\ & + \tilde{\mathcal{J}}_1^{\text{GdGd}} \langle J_\alpha^{\text{Gd}} \rangle^3 + \tilde{\mathcal{J}}_1^{\text{GdTb}} \langle J_\alpha^{\text{Gd}} \rangle \langle J_\alpha^{\text{Tb}} \rangle^2 + g^{\text{Gd}} \mu_B B \cos \theta_\alpha] J_{ix}^{\text{Gd}} \end{aligned} \quad (2)$$

and

$$\begin{aligned} \mathcal{H}_2^{\text{Tb}} = & -\sum_i \sum_{\alpha=x,y,z} [\tilde{\mathcal{J}}_0^{\text{TbTb}} \langle J_\alpha^{\text{Tb}} \rangle + \tilde{\mathcal{J}}_0^{\text{TbGd}} \langle J_\alpha^{\text{Gd}} \rangle \\ & + \tilde{\mathcal{J}}_1^{\text{TbTb}} \langle J_\alpha^{\text{Tb}} \rangle^3 + \tilde{\mathcal{J}}_1^{\text{TbGd}} \langle J_\alpha^{\text{Tb}} \rangle \langle J_\alpha^{\text{Gd}} \rangle^2 + g^{\text{Tb}} \mu_B B \cos \theta_\alpha] J_{ix}^{\text{Tb}}. \end{aligned} \quad (3)$$

Here $\mathcal{H}_1^{\text{Gd}}$ describes an effective subsystem of Gd ions in which the exchange interaction between magnetic moments depends whether Gd or Tb ions are in the neighborhood of a given site. Similarly $\mathcal{H}_2^{\text{Tb}}$ describes a subsystem of Tb ions, in which the exchange interaction between magnetic moments depends on the type of ions are occupying the neighboring sites. In these Hamiltonians θ_α ($\alpha = x, y, z$) are the angles between the applied magnetic field and the crystallographic axes x , y and z . The parameters $\tilde{\mathcal{J}}_0^{ab} = \mathcal{J}_0^{ab} Z_{ab}$, and $\tilde{\mathcal{J}}_1^{ab} = \mathcal{J}_1^{ab} Z_{ab}$ ($a, b = \text{Gd}, \text{Tb}$) are effective exchange interaction parameters where Z_{ab} represents the number of neighboring ions. For instance, Z_{GdTb} represents the number of Tb ions are in the neighborhood of a site occupied by Gd ions.

The mean values $\langle J_\alpha^a \rangle$ ($a = \text{Gd}$ or Tb and $\alpha = x, y, z$) can be obtained by the following equation:

$$\langle J_\alpha^a \rangle = \frac{\sum_j \langle \Psi_j^a | J_\alpha^a | \Psi_j^a \rangle e^{-\beta E_j^a}}{\sum_j e^{-\beta E_j^a}}, \quad (4)$$

where $\beta = 1/k_B T$ and k_B is the Boltzmann constant. $E_j^a; |\Psi_j^a\rangle$ are the energy eigenvalues and eigenvectors of the mean field Hamiltonians $\mathcal{H}_1^a$. The total magnetization of the compound, $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$, can be calculated by

$$M(T, B, c) = (1 - c)M^{\text{Gd}}(T, B, c) + cM^{\text{Tb}}(T, B, c),$$

where $M^a(T, B, c) = g^a \mu_B \langle J^a \rangle$ ($a = \text{Gd}$ or Tb) is the magnetization of the effective system of Gd or Tb ions described by the effective Hamiltonian $\mathcal{H}_1^{\text{Gd}}$ or $\mathcal{H}_2^{\text{Tb}}$. In the same way, the total entropy in

the compound, $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$ can be calculated by

$$S(T, B, c) = (1 - c)S^{\text{Gd}}(T, B, c) + cS^{\text{Tb}}(T, B, c),$$

where $S^a(T, B, c)$ ($a = \text{Gd}$ or Tb) given by

$$S^a(T, B, c) = S_{\text{mag}}^a(T, B, c) + S_{\text{lat}}^a(c, T) + S_{\text{el}}^a(T) \quad (5)$$

represents the total entropy of the subsystem described by the effective Hamiltonian $\mathcal{H}_1^a$. In Eq. (5) $S_{\text{el}}^a = \gamma^a T$, where γ^a is the Sommerfeld coefficient, represents the electronic entropy. S_{mag}^a given by

$$S_{\text{mag}}^a(T, B, c) = N_m \Re \left[\ln \sum_i e^{-\beta E_i^a} + \frac{1}{k_B T} \frac{\sum_i E_i^a e^{-\beta E_i^a}}{\sum_i e^{-\beta E_i^a}} \right] \quad (6)$$

is the magnetic entropy, where $N_m = 5$ represents the number of magnetic ions per unit formula. S_{lat}^a given, in the Debye approximation, by

$$S_{\text{lat}}^a(T, B, P) = N_i \left[-3 \Re \ln \left(1 - e^{-\Theta_D^a/T} \right) + 12 \Re \left(\frac{T}{\Theta_D^a} \right)^3 \int_0^{\Theta_D^a/T} \frac{x^3}{e^x - 1} dx \right] \quad (7)$$

is the crystalline lattice entropy, where $N_i = 9$ represents the number of ions per unit formula.

3. Results

In order to apply the present model to calculate the magnetocaloric effect in $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$, we have to fix a set of model parameters. The total angular momentum and the Landé factors were taken from Hund's rule as $J^{\text{Gd}} = \frac{7}{2}$, $g^{\text{Gd}} = 2$ and $J^{\text{Tb}} = 6$, $g^{\text{Tb}} = \frac{3}{2}$. The Sommerfeld coefficient and the Debye temperature were taken as $\gamma^{\text{Gd}} = \gamma^{\text{Tb}} = 5$ (mJ/mol K²) and $\Theta_D^{\text{Gd}} = \Theta_D^{\text{Tb}} = 200$ K. The number of neighbor ions were taken as $Z_{\text{GdGd}} = 10$ and $Z_{\text{GdTb}} = 0$ for $c = 0$ and $Z_{\text{TbTb}} = 10$ and $Z_{\text{TbGd}} = 0$ for $c = 1$. For intermediate concentrations we take $Z_{\text{GdGd}} = (1 - c)10$, $Z_{\text{GdTb}} = 10c$, $Z_{\text{TbTb}} = 10c$ and $Z_{\text{TbGd}} = (1 - c)10$. The bare values of the exchange interaction parameters were taken as $\mathcal{J}_0^{\text{GdGd}} = 0.57$ meV, $\mathcal{J}_1^{\text{GdGd}} = \mathcal{J}_1^{\text{GdTb}} = 0.025$ meV, $\mathcal{J}_0^{\text{TbTb}} = 0.08$ meV and $\mathcal{J}_1^{\text{TbTb}} = 0$. These parameters were taken in order to reproduce the experimental data of the magnetic ordering temperatures in the pure compounds $\text{Gd}_5\text{Si}_2\text{Ge}_2$ and $\text{Tb}_5\text{Si}_2\text{Ge}_2$. For intermediate concentrations the exchange interaction parameters $\mathcal{J}_0^{\text{GdGd}}$ and $\mathcal{J}_0^{\text{TbTb}}$ should be self consistently calculated in order to incorporate the changes in the electron gas due to the presence of impurities. However, such a calculation is much complex and is not in the scope of the present work. Here, for the sake of simplicity, we just consider a very smooth linear dependence of these parameters when we go from $c = 0$ to 1. Moreover, the effective exchange interaction parameters $\tilde{\mathcal{J}}_0^{ab}$ and $\tilde{\mathcal{J}}_1^{ab}$ defined as $\tilde{\mathcal{J}}_0^{ab} = \mathcal{J}_0^{ab} Z_{ab}$, and $\tilde{\mathcal{J}}_1^{ab} = \mathcal{J}_1^{ab} Z_{ab}$ ($a, b = \text{Gd}, \text{Tb}$) already depend on the impurity concentration via the number of neighboring ions. Using these model parameters, we solve the magnetic state equations for several values of the impurity concentration.

In Fig. 1, we plot the temperature dependence of the magnetization in $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$ calculated in the absence of an external magnetic field. It can be observed from this figure that $\text{Gd}_5\text{Si}_2\text{Ge}_2$ undergoes a first order phase transition around 275 K. As the Tb concentration is increased the magnetic ordering temperature becomes smaller and the first order phase transition becomes weak. In Fig. 2, we plot the temperature dependence of the total specific heat capacity in $(\text{Gd}_{1-c}\text{Tb}_c)_5\text{Si}_2\text{Ge}_2$ calculated in the absence of an applied magnetic field. Notice that our calculations are in good agreement with the available experimental data [4] for $c = 0$. For the remaining concentrations are necessary further experimental data to

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