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On the orientation of the magnetization vector and the associated entropy in $Nd_2Fe_{14}B$

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ABSTRACT

We have used the methods of classical statistical mechanics in calculating the temperature and field-dependence of specific magnetic and magnetothermal properties of $Nd_2Fe_{14}B$. For example, we correlate the orientation of the magnetization vector, relative to a specific crystallographic axis, to the angular coordinates of the most probable location in a probability landscape of this anisotropic system. Further, we correlate the probability distribution to the associated magnetic entropy. The field dependence of the canting angle, either off the *c*-axis or off the basal plane, is also discussed in particular for critical magnetic fields at which first-order magnetization process takes place. Specific features of magnetization curves, at certain temperatures, are discussed in the light of results based on models involving crystal field and/or exchange interaction effects.

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1. Introduction

The compound $Nd_2Fe_{14}B$ has attracted much attention as an important candidate for permanent magnets [1]. Numerous experimental and theoretical work has been devoted to either improve the key magnetic properties e.g. Curie temperature and coercivity, or to understand the origin of its high magnetic quality. For example, studies on the magnetization, magnetostriction and torque measurements [2–12], magnetic susceptibility [13,14], heat capacity [15,16], size-dependent spin reorientation [17], and ab initio calculations [18] are only representatives of the vast literature on this important material. In addition, extensive theoretical efforts have been done to understand the magnetic properties of rare-earth transition metal compounds (RT) e.g. $Nd_2Fe_{14}B$. Those efforts have resulted in constructing a Hamiltonian which considers both the crystal field and/or the exchange energy to describe those systems, where the itinerant 3d electrons of the T atoms are treated differently from the more localized 4f electrons of the R atoms. Examples of studies, using such involved Hamiltonian are calculation of the spin-reorientation temperature of $Nd_2Fe_{14}B$ [19] and calculation of the magnetization curves at selected temperatures along specific crystallographic directions in the tetragonal $Nd_2Fe_{14}B$ crystal [3,20–22].

The aim of this work is to use the temperature dependence of experimentally determined anisotropy constants [5] in order to calculate a host of magnetic and magneto thermal properties, using the fundamentals of classical statistical mechanics in a simple model. In the discussion section it is hoped to demonstrate that the results of our calculation are not in disagreement with more involved methods where the specific nature of 4f and 3d electrons, and consequently the relative strength of the interactions present, were taken into consideration.

We have already reported, mainly, on calculating the magnetization curves of $Nd_2Fe_{14}B$ in the temperature range 77–293 K using a statistical mechanics-based model [7]. In the present paper we extend our work on this system, in particular we calculate the probability of having the magnetization vector orientated either along or perpendicular to the *c*-axis, for a range of temperatures and magnetic fields. This study helps us to correlate the angular location of the magnetization vector to both of the Zeeman and thermal energies in this anisotropic system. We also report on the temperature dependence of magnetic susceptibility, magnetic heat capacity and magnetic entropy in magnetic fields of different strength and directions within the tetragonal system of $Nd_2Fe_{14}B$.

2. Model and analysis

We treat the magnetization vector as a classical vector whose orientation in space is dictated by an interplay between anisotropy, Zeeman and thermal energies. For such a vector the laws of classical statistical physics are appropriate. The angular states, of such a vector should assume continuous, rather than discrete

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orientation, i.e. the polar and azimuthal angles θ and φ should assume all values in their respective ranges 0–π and 0–2π, and hence an integration rather than a summation should be used.

The classical partition function, therefore is given by [7]

$$Z(T, H) = \frac{1}{4\pi}\int_0^{2\pi}\int_0^{\pi} e^{-E(T,H,\theta_m,\varphi_m)V\beta}\sin\theta_m\ d\theta_m\ d\varphi_m \tag{1}$$

where V is the volume of a spherical magnetic particle taken to be 10^{-19} cm^3 ($\sim$28 Å in radius) throughout this work unless otherwise specified, and $\beta = 1/k_BT$, where k_B is the Boltzmann constant and T is the absolute temperature.

The classical partition function includes the total energy density of this tetragonal magnetic system [5,23], i.e. the sum of the anisotropy and Zeeman energy densities. The total energy, however, may include other types of energy e.g. magnetostrictive for specific systems, but in this work we focus only on the magnetocrystalline anisotropy, besides of course, the Zeeman term, therefore the energy density takes the following form:

$$E(T, H, \theta_m, \varphi_m) = K_1\sin^2\theta_m + K_2\sin^4\theta_m + K_3\sin^4\theta_m\cos 4\varphi_m + K_4\sin^6\theta_m + K_5\sin^6\theta_m\cos 4\varphi_m - \vec{H}\cdot\vec{M_s} \tag{2}$$

where θ_m is the angle between the magnetization vector and the c-axis, φ_m is the azimuthal angle (chosen in this work to be the angle between the projection of the magnetization vector in the basal plane, and the [110] direction, unless otherwise specified) and K_i's are the anisotropy constants. The temperature dependence of the five anisotropy constants, and the saturation magnetization M_s were approximated by simple polynomials using the best fit to the tabulated data of $K(T)$ and $M_s(T)$ in the temperature range 78–293 K reported by Blozoni et al. [5].

The magnetization, magnetic susceptibility, magnetic heat capacity and entropy are well known to be derived from the partition function (e.g. [24,25]). The magnetization is calculated using the following relation:

$$M = \frac{1}{\beta}\frac{\partial \ln Z(T, H)}{\partial H} \tag{3}$$

Another method for calculating the isothermal magnetization is to minimize Eq. (2), in order to find the coordinates θ_m and φ_m of the magnetization vector at equilibrium. These coordinates are calculated for a given temperature and different magnetic fields, applied along a certain crystallographic direction, in order to produce the magnetization curves. There are several methods to minimize a function of two variables (e.g. Davidon–Fletcher and Powell method) [26], however, we have used the Mathematica software package [27], to perform this task.

The magnetic susceptibility per unit volume is given by

$$\chi = \frac{\partial M}{\partial H} = k_BT\frac{\partial^2 \ln Z(T, H)}{\partial H^2} \tag{4}$$

The thermally-averaged energy $\langle E\rangle$ is obtained [24] via the standard relation

$$\langle E\rangle = -\ \partial \ln Z\ /\partial\beta$$

The magnetic heat capacity and the change in the magnetic entropy, as a function of temperature and field, are given by Eqs. (5) and (6) respectively:

$$C_{mag} = \frac{\partial\langle E\rangle_{mag}}{\partial T} = k_B\beta^2\frac{\partial^2 \ln Z(T, H)}{\partial\beta^2} \tag{5}$$

$$\Delta S_{mag} = \int_{T_1}^{T_2}\frac{C_{mag}(T)}{T}\,dT \tag{6}$$

where the quantity $\Delta S_{mag} = S(T_2) - S(T_1)$ is the change in entropy in a certain temperature range, for a given field.

The Helmholtz free energy F is given by $F=E-$TS, where E is referred to as internal energy or mean energy $\langle E\rangle$. The entropy, at constant field H, is given by $S = -(\partial F/\partial T)_H$ or equivalently by $S = k_B[\ln Z + \beta\langle E\rangle] = -k_B\sum_i P_i \ln P_i$, where Z is the sum-over-states (or an integral-over-states). For a system of a definite energy spectrum, the probability of having a state with an energy E_i is given by $P_i = e^{-E_i\beta}/Z$.

The probability, for the magnetization vector, to have angular coordinates θ_m and φ_m, at a given temperature and field is given by

$$P(T, H, \theta_m, \varphi_m) = \frac{e^{-E(T,H,\theta_m,\varphi_m)V\beta}}{Z} \tag{7}$$

Studying the probability angular dependence is important for an understanding of the behavior of magnetic systems. In particular, it enables one to study the evolution of the probability as a function of temperature and field [28,29].

We have used the cgs system of units and the Mathematica software package for numeric, symbolic, fitting analysis and plotting our results throughout this work.

3. Results and discussion

3.1. The magnetization curves of $Nd_2Fe_{14}B$

We have already reported on our calculation of the magnetization curves at T=77, 125, 176 and 270 K [7]. We report here on the magnetization curves at four additional temperatures, namely at T=4.2, 95, 150 and 293 K, as shown in Figs. 1–4 respectively. The absence of in-plane anisotropy is evident at T=293 K, in contrast to lower temperatures $\leq$ 176 K.

We have calculated the magnetization curves at T=95 K either by evaluating the partition function, and consequently determining the magnetization from Eq. (3) as shown in Fig. 2, or by finding the equilibrium value of θ_m at which the total energy density is minimum for a certain field. The isothermal dependence of the magnetization on field, along a specific crystalline direction, is calculated by determining the magnetization component along the field direction for different field values. The agreement between the results of Figs. 2 and 5 is obvious, and indicates that the component of the magnetization vector along the field direction is of such a magnitude that minimizes the energy. In Section 3.4, we will discuss the consistency between the energy and probability landscapes.

We discuss, now, specific features of magnetization curves calculated using our method, in the light of more involved calculations e.g. [5,12,21,22]. As we mentioned before, we have reported on the field dependence of magnetization in the 77–293 K temperature range along the [001], [100] and [110] directions [7]. Therefore we are able to compare between our results at 95 and 293 K, with those of Refs. [21,22]. Yamada et al. [21] suggested a model which takes into consideration both of the molecular field and crystalline electric field CEF. On the other hand, the Fe sublattice anisotropy was treated phenomenologically. According to their calculation, the magnetization curves at 290 K (Fig. 10 in Ref. [21]) exhibit an absence of anisotropy in the hard [100] and [110] directions and almost a field-independent easy [001] direction, up to 300 kOe. Our results in Fig. 4 are fairly in good agreement with the calculation of Yamada et al., in particular regarding the absence of an in-plane anisotropy and the value of the anisotropy field of $\sim$100 kOe. However, the experimental data reported by Yamada et al. shows a slight in-plane anisotropy close to

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