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# Comparative Study of the Magnetocaloric Properties of Two Anisotropic Heisenberg Systems with Spin ½ and 1

**A. Boubekri, M.Y. El Hafidi*, M. El Hafidi**

*Laboratory of Condensed Matter Physics, Physics, Department, Faculty of Sciences Ben M'sik, Hassan II University of Casablanca, BP 7955, Av. Driss El Harty, 20643 Casablanca, Morocco.*

***Corresponding Author:** ***M.Y. El Hafidi,*** *Laboratory of Condensed Matter Physics, Physics, Department, Faculty of Sciences Ben M'sik, Hassan II University of Casablanca, BP 7955, Av. Driss El Harty, 20643 Casablanca, Morocco.*

**Abstract:** *The Pair Approximation Method (PAM) is applied to study the thermodynamic properties of a three-dimensional spin lattice in a cubic structure with coordination number z = 6. The used model consists of the anisotropic ferromagnetic Heisenberg with spins ½ and 1 under an external magnetic field.*

*Within this approximation based on Gibbs free-energy, we investigate the effect of both exchange coupling anisotropies and the spin magnitude on the thermodynamic quantities such as the magnetization, the entropy, the heat capacity and the entropy change. We also study the resulting Relative Cooling Power (RCP) for the two considered spin values.*

*This work shows that even small amount of exchange anisotropy and spin size have a significant influence on the magnetic properties and RCP.*

**Keywords:** *Heisenberg model, Spin Hamiltonian, Isothermal entropy change, Magnetocaloric effect, Pair approximation, Relative cooling power.*

## 1. INTRODUCTION

Initially, Ising spin models have attracted considerable attention because of their simplicity and solvability. However, in real systems, spins can also interact with different exchange integrals for different directions in the lattice [1]. To take into account the physical reality on one hand and to overcome the computational difficulties on the other hand, combinations of both Ising and Heisenberg models have been proposed [2]. In our turn, we will combine the two models by adopting the model of Ising-Heisenberg to study theoretically the magnetocaloric effect in a three-dimensional spin array where we perform a comparative approach of two selected spin values: ½ and 1. This phenomenon has regained interest in view of the hopes that it offers, in particular in magnetic refrigeration which aims to replace the traditional refrigeration of greenhouse gases and open the way for new designs of magnetic cooling [3,4]. This new technology based on the magnetocaloric effect (MCE) has recently drawn tremendous attention due to its good energy efficiency and environment friendliness. As a result, many materials with large MCE, especially around room temperature, have been developed and broadly investigated [5,6].

In this context, a wide variety of magnetocaloric materials has been suggested for low- and room-temperature applications including both oxides and intermetallics [7]. Particularly, the $RMnO_3$ (R = magnetic rare earth) manganites showed large satisfaction of low temperature magnetic cooling requirements [8]. In addition to their excellent magnetocaloric properties over the temperature range around 10 K, $RMnO_3$ oxides unveil better chemical and mechanical stabilities when compared with other materials such as intermetallics [9].

Up to now, many efforts have been made to elaborate composite refrigerant materials to satisfy the requirements of a magnetic refrigerator covering a large temperature span [10], whereas relatively few work has been reported on gadolinium Gd and/or $Gd_5(Si_xGe_1\text{-}x)_4$ alloys, which display large MCE near room temperature [11].

In this work, we aim to apply the PAM to an anisotropic Ising-Heisenberg ferromagnetic system with two spin values (½ and 1). In this sense, we will investigate the effects of exchange anisotropy and spin

magnitude with (or without) the magnetic field on the MCE and magnetocaloric properties in a cubic system with coordination number z=6. We aim also to elucidate several physical parameters that will participate to improve magnetic refrigeration efficiency.

It is worth to note that numerous theoretical investigations have recently been reported concerning PAM with different spin values [12, 13]. However, in the current research, the focus has been on quantitative and comparative study of anisotropic exchange system with the two selected spin values (½ and 1).

The paper is organized as follows:

In Section 2, we formulate the model and apply the pair approximation method (PAM) in the anisotropic Heisenberg ferromagnetic spin system. In Section 3, we present the results of numerical calculations for the thermal relevant thermodynamic quantities and analyze their influence on the MCE. Finally, section 4 is devoted to relevant conclusions and outlooks.

## 2. THEORY

The Hamiltonian of the spin-S system located at the corners of the cubic lattice is given by:

$$H = -\sum_{<i,j>}\left[J_{xy}\left(S_i^x S_j^x + S_i^y S_j^y\right) + J_z\left(S_i^z S_j^z\right)\right] - b\sum_i S_i^z \tag{1}$$

where the first term is the exchange part related to the interaction of the atomic spin (i) with the atomic spin (j), $J_{xy}$ and $J_z$ refer respectively to the in-plane *xy* and along the z-axis anisotropy exchange integrals. The second term of the Hamiltonian stands for the Zeeman energy, where $b = g\mu_B B$ denoting the external magnetic field, g being the Landé factor, $\mu_B$ the Bohr magneton and B the effective applied field along the z-axis.

In the pair approximation method, we introduce the single site and the pair Hamiltonians respectively $H_i$, $H_{ij}$ which take the following forms [14]:

$$H_i = -(\Lambda + b)S_i^z \tag{2}$$

$$H_{ij} = -J_{xy}(S_i^x S_j^x + S_i^y S_j^y) - J_z S_i^z S_j^z - (\Lambda' + b)(S_i^z + S_j^z) \tag{3}$$

where the molecular field parameters $\Lambda$ and $\Lambda'$ can be expressed by these equations:

$$\Lambda = z\lambda \quad \text{and} \quad \Lambda' = (z-1)\lambda \tag{4}$$

Here, z is the coordination number of a given spin. In the considered lattice, each spin has six nearest neighbors, $\lambda$ parameter will be calculated by minimization of total Gibbs free-energy.

The variational parameters are determined by minimization of the total free-energy per site with respect to the following rule:

$$\frac{G}{N} = \frac{z}{2}G_{ij} - (z-1)G_i \tag{5}$$

where N is the total number of spin sites in the system, $G_i$ is the single-site Gibbs energy calculated with the single-site density matrix, and $G_{ij}$ is the two sites Gibbs energy calculated with the pair-site density matrix.

Besides, since the partition function $Z$ for single-site and pair-site is formally given by:

$$Z = Tr\exp(-\beta H) \tag{6}$$

we can get the magnetization $m_i$ with the following relationship:

$$m_i = \left\langle S_i^z \right\rangle = \frac{Tr\left[S_i^z \exp(-\beta H)\right]}{Z} \tag{7}$$

where $\beta = 1/k_B T$ and $k_B$ is the Boltzmann constant.

The single-site and double-site Gibbs energy $G$ will be determined from the common definition:

$$G = -\frac{1}{\beta}\ln(Z) \tag{8}$$

Using the above formulas, we get the expression of partition function for the single-site for the two spin values (S= ½ and 1) respectively:

$$Z_i^1 = 2\{\cosh[\beta(z\lambda + b)]\} + 1 \tag{9}$$

$$Z_i^{1/2} = \left\{2\cosh\left[\frac{\beta(z\lambda + b)}{2}\right]\right\} \tag{10}$$

Thus, the magnetization $m_i$ for single site is given by:

$$m_i^1 = \frac{2\sinh[\beta(z\lambda + b)]}{2\cosh[\beta(z\lambda + b)] + 1} \tag{11}$$

$$m_i^{1/2} = \frac{1}{2}\tanh\left[\frac{\beta(\Lambda + b_0)}{2}\right] \tag{12}$$

Similarly, we can get the expression of partition function and magnetization for double site:

$$Z_{ij}^1 = e^{\beta(-J_z + 2D)} + 2e^{\beta(J_z + 2D)}\cosh(2\beta\Lambda') + 4e^{\beta D}\cosh(\beta\Lambda')\cosh(\beta J_{xy}) + 2e^{\beta\left(\frac{-J_z}{2} + D\right)}\cosh(\beta w) \tag{13}$$

where $w = \sqrt{\left(\frac{J_z}{2}\right)^2 + 2J_{xy}^2}$

$$m_{ij}^1 = \frac{4e^{\beta(J_z + 2D)}\sinh[2\beta(\Lambda' + b)] + 4e^{\beta D}\cosh(\beta J_{xy})\sinh[\beta(\Lambda' + b)]}{Z_{ij}^1} \tag{14}$$

$$Z_{ij}^{1/2} = \left\{2\exp\left(\beta\frac{J_z}{4}\right)\cosh[\beta(\Lambda' + b)] + 2\exp\left(-\frac{\beta J_z}{4}\right)\cosh\left(\frac{\beta J_{xy}}{2}\right)\right\} \tag{15}$$

$$m_{ij}^{1/2} = \frac{\sinh[\beta(\Lambda' + b)]}{2\left\{\cosh[\beta(\Lambda' + b)] + \exp\left(-\frac{\beta J_z}{4}\right)\cosh\left(\frac{\beta J_{xy}}{2}\right)\right\}} \tag{16}$$

Note that we can establish the value of the variational parameter $\lambda$ from the minimum condition for the Gibbs free energy:

$$\frac{\partial G}{\partial \lambda} = 0 \tag{17}$$

On this basis, all the thermodynamic properties can be found. In this work, we focus on the magnetization, entropy, entropy change and RCP. Thus, the corresponding formulas for the magnetization and entropy are given respectively in the following way:

The first-order derivative of the Gibbs energy (Eq. 5) over the field gives the magnetization per one lattice site:

$$m = -\frac{1}{N}\left(\frac{\partial G}{\partial b}\right)_T \tag{18}$$

while the entropy per one lattice site can be found from the general formula:

$$S = -\frac{1}{N}\left(\frac{\partial G}{\partial T}\right) \tag{19}$$

The Curie temperature $T_C$ can be calculated by linearization of self-consistent Eq. (17) from the condition: b=0 and $\lambda \to 0$; hence, one gets:

In the case of spin- ½ :

$$\exp\left(\frac{\beta_C J_z}{2}\right) = \frac{z}{z-2}\cosh\left(\frac{\beta_C J_{xy}}{2}\right) \tag{20}$$

while, in the case of spin-1:

$$\frac{z}{3(z-1)} = \frac{2e^{\beta_C(J_z)} + \cosh(\beta_C J_{xy})}{e^{\beta_C(-J_z)} + 2e^{\beta_C(Jz)} + 4\cosh(\beta_C J_{xy}) + 2e^{\frac{-\beta_C Jz}{2}}\cosh(\beta_C w)} \tag{21}$$

Finally, all these analytic results will then be programmed and incorporated for a numerical computation enabling us to calculate the various physical variables mentioned above and analyze their behavior in different situations.

## 3. Numerical results and discussions

The numerical resolution of relevant equations in our proposed model has been performed thanks to a computational program that we developed specially for this aim.

As a first step, we plot the Curie temperature versus different anisotropy exchange $J_{xy}/J_z$ from 0 up to 1, on the basis of equations (20-21) with coordination number z=6. The results are displayed in Fig.1 where curves show that with increase of $J_{xy}$ or decrease of anisotropy, the Curie temperature decreases slowly with a non-linear line; it is also seen that the spin magnitude enhances significantly the critical temperature.

It is worth to note that for the pure Ising model, the system with z=6 and S= ½ exhibits a phase transition at Curie temperature $k_BT_c/J_z$=1.23 which is on the one hand identical to that determined by Balcerzak [15], and on the other hand, is between the value predicted by the Effective Field Theory (EFT) (1.268) [16, 17] and that given by the expended Bethe-Peirels model (1.20) [18].

Otherwise, for the isotropic Heisenberg model with z=6, we get the value $k_BT_c/J_z$ =0.91 which is below the value given by the EFT (1.21) [19].

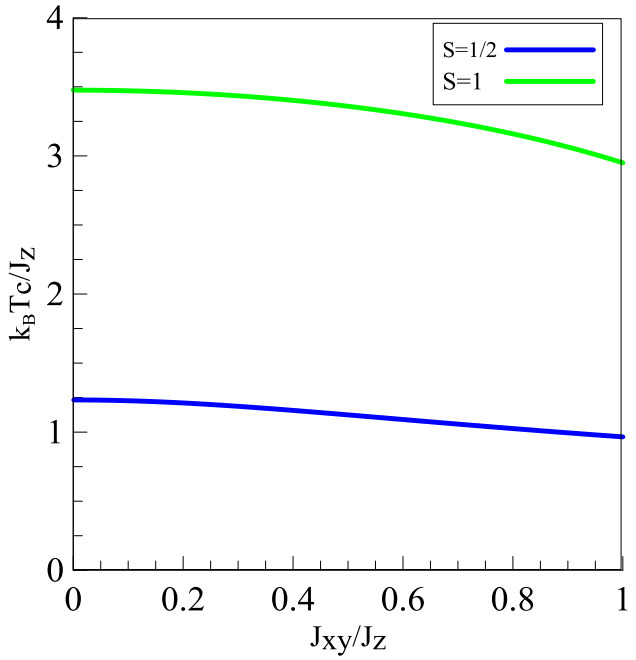


**Fig1.** *The critical temperature versus the exchange anisotropy $J_{xy}/J_z$= for two spin values (1 and ½ )*

The thermal evolution of the magnetization is depicted in Figures (2a and 2b) for several selected values of the exchange anisotropy $J_{xy}/J_z$ for the two spin values (1 and ½). In absence of applied field (Fig. 2a), the spontaneous magnetization vanishes at the Curie temperature. In the presence of an applied field (Fig. 2b), the magnetization persists beyond $T_C$ with an inflection point centered at the critical temperature. For high magnetic fields, m (b, T) continues its declining beyond $T_C$ since correlations between spin pairs persist well above the Curie point. The ferromagnetic behavior is guaranteed by the exchange which translates the mutual interaction between spins of neighboring atoms. This exchange energy tends to align the magnetic moments of neighboring atoms creating the saturated magnetic domains.

The figures show also that the Curie temperature shifts and increases as the exchange anisotropy decreases.

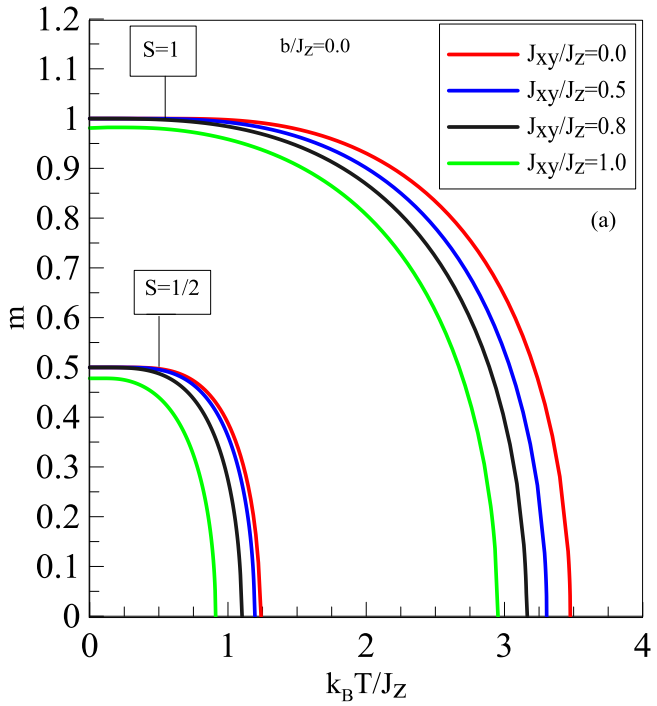


**Fig2a.** *The temperature dependence of the magnetization m in absence of magnetic field b for various values of the anisotropy ratio Jxy/Jz with spin values ½ and 1.*

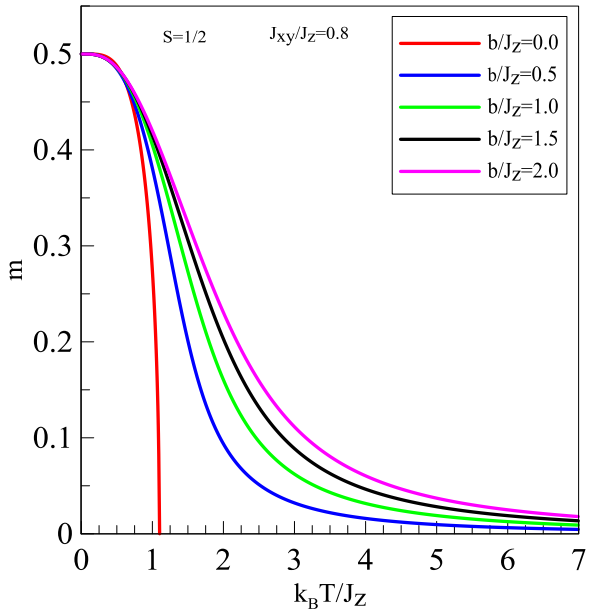


**Fig2b.** *The temperature dependence of the magnetization m for several values of the magnetic field ($b/J_z$=0; 0.5; 1.0; 1.5; 2.0) in the case of $J_{xy}/J_z$=0.8 and spin ½ .*

Figs. 3a – 3b stand to the magnetic entropy versus the reduced temperature $k_BT/J_z$ without and in presence of an external magnetic field for different values of exchange anisotropy. Below $T_c$, the curves show a non-linear and a monotone behavior with a clear tendency to saturation near $T_c$ , particularly in absence of the magnetic field. However, above $T_c$, the entropy evolves in a quasi-constant profile around its maximal value because of the thermal agitation which becomes dominant and disrupts the arrangement of the magnetic moments.

Note that the break point (see fig. 3a) of the entropy coincides with $T_c$ whose value is in perfect agreement with that determined by the phase diagram in fig1. This confirms that $T_c$ decreases gradually with the $J_{xy}/ J_z$ exchange anisotropy ratio.

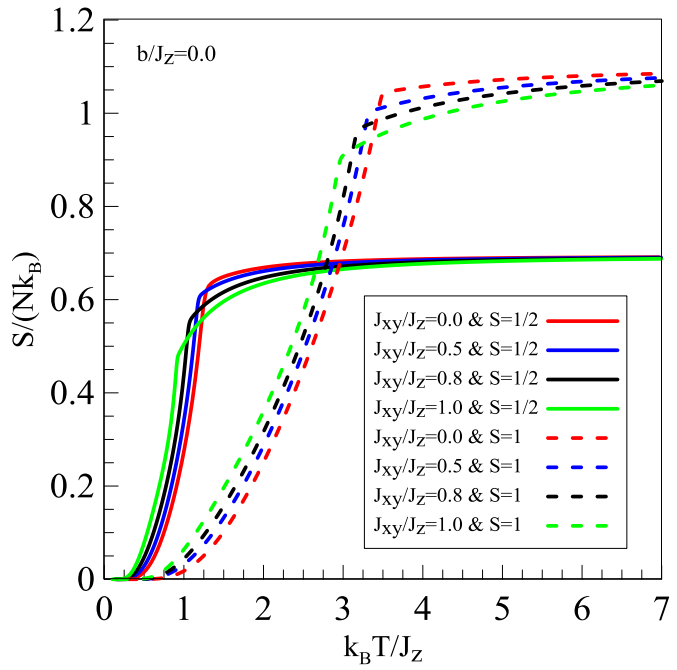


**Fig3a.** *Entropy per spin vs temperature without the external field for various $J_{xy}/J_z$ ratios with spin values ½ and 1.*

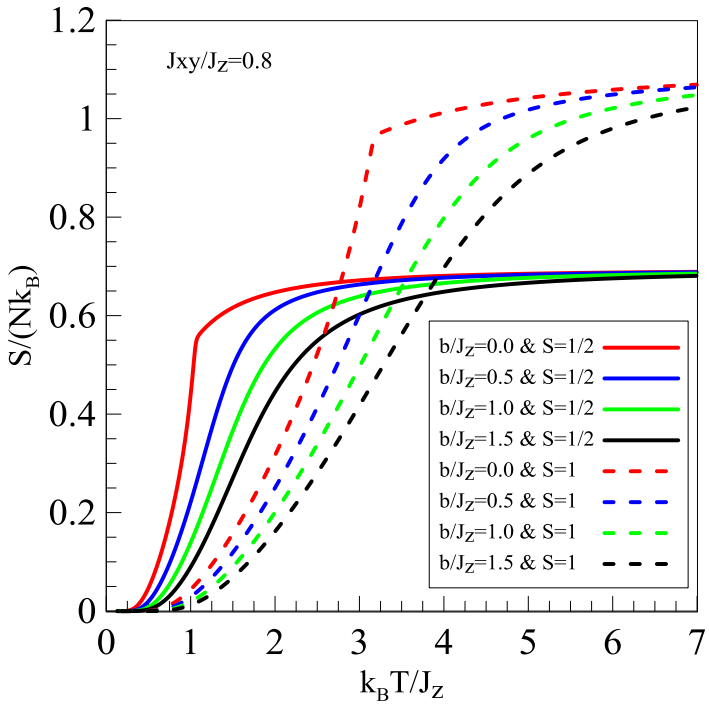


**Fig3b.** *Entropy per spin upon temperature under various values of the external field ($b/J_z$=0.0-0.5-1.0-1.5) in the case of $J_{xy}/J_z$ =0.8 with spin values of ½ and 1.*

It is worth remembering that the specific heat can be obtained from the second-order derivative of the Gibbs energy over temperature:

$$C_b = -T\left(\frac{\partial^2 G}{\partial T^2}\right)_b = T\left(\frac{\partial S}{\partial T}\right)_b \tag{22}$$

The thermal evolution of $C_b$ is represented for some selected values of exchange anisotropy, without the field in Fig. 4a and for a fixed value of anisotropy and several field values (fig. 4b). As well seen, all curves start from zero at lower temperature and display a sharp maximum at the critical temperature which decreases and broadens when a field is applied (Fig. 4b). This critical point shifts down while increasing the anisotropy parameter $J_{xy}/J_z$ as shown in Fig. 4a. Besides, the specific heat curves extend over a large range of temperature by increasing magnetic field.

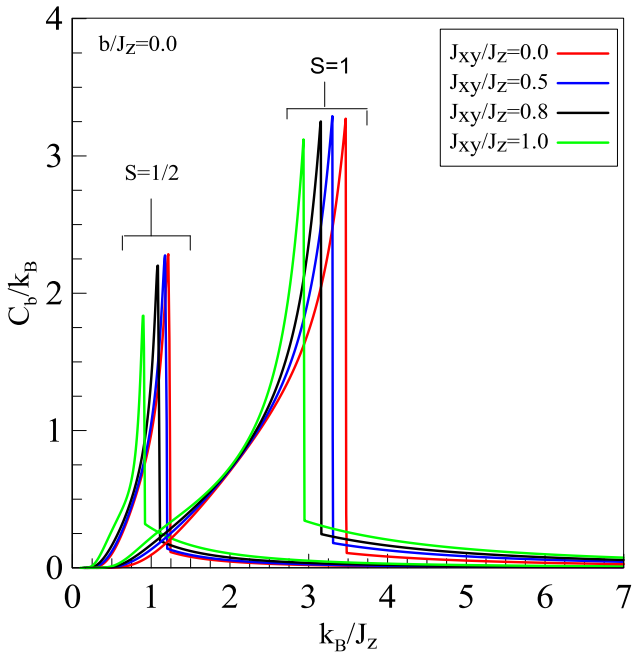


**Fig4a.***The specific heat versus the temperature for different anisotropy parameter in absence of magnetic field for spin values (½ and 1)*

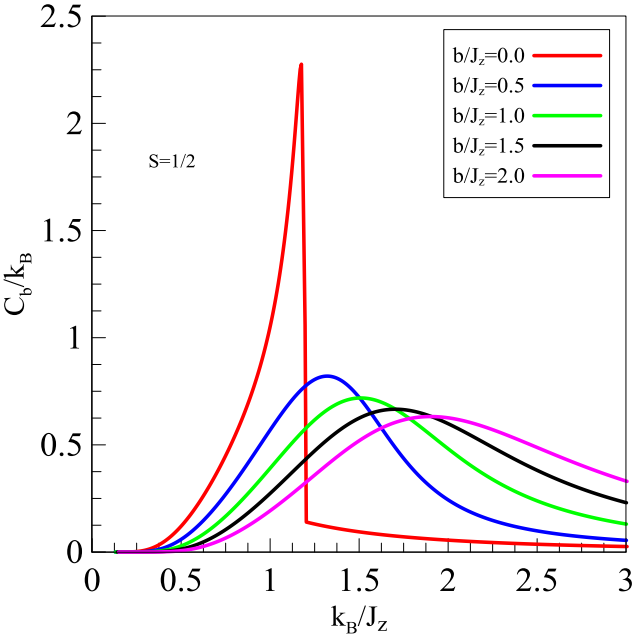


**Fig4b.***The specific heat versus the temperature for external magnetic field: 0, 0.5, 1, 1.5, and 2 while the anisotropy parameter $J_{xy}/J_z$ takes 0.5.*

Based on the magnetization curves reported in Figures 2a and 2b, the change of isothermal magnetic entropy caused by variation of magnetic field external from *0* to *b* can be determined using the well-known Maxwell relation:

$$\Delta S_T = -\int_0^b \left(\frac{\partial m}{\partial T}\right) db \tag{23}$$

The variation of magnetic entropy as a function of temperature for different anisotropy values $J_{xy}/J_z$: 0, 0.5, 0.8 and 1.0 is shown in Figures 5a and 5b. Clearly, the entropy change increases abruptly for low temperature until it reaches a maximum value that matches with the transition point.

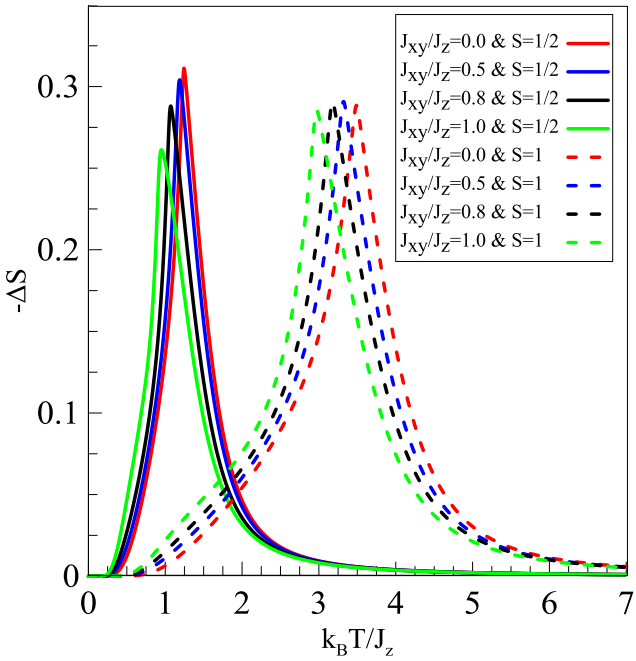


**Fig5a.** *The varition of magnetic entroy behavior for different values of the relative exchange ainsotropy $J_{xy}/J_z$ for the spin values (½ and 1) and an isothermal field change $\Delta b/J_z=0.5$*

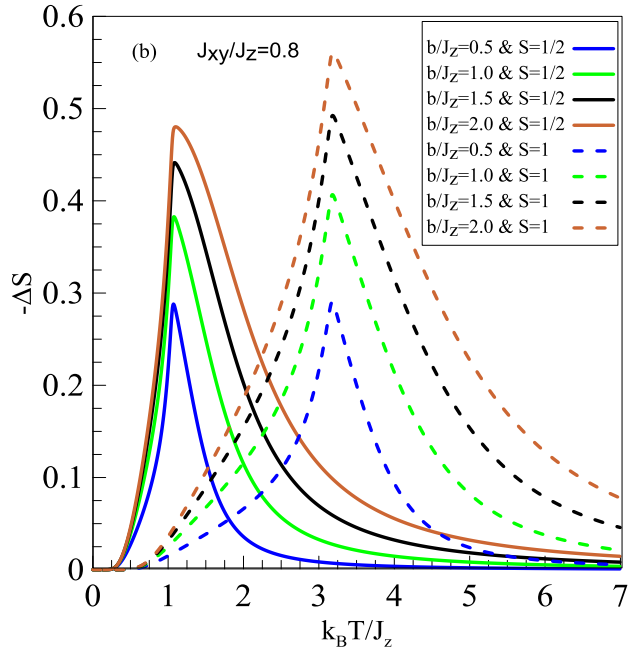


**Fig5b.** *The variation of magnetic entroy behavior under different values of the external field ($h/J_z$=0.5-1.0-1.5-2.0) with spin values 1and ½ in the case of anisotropy value $J_{xy}/J_z=0.8$.*

In addition, it can be seen that the Curie temperature goes up with the spin magnitude increase.

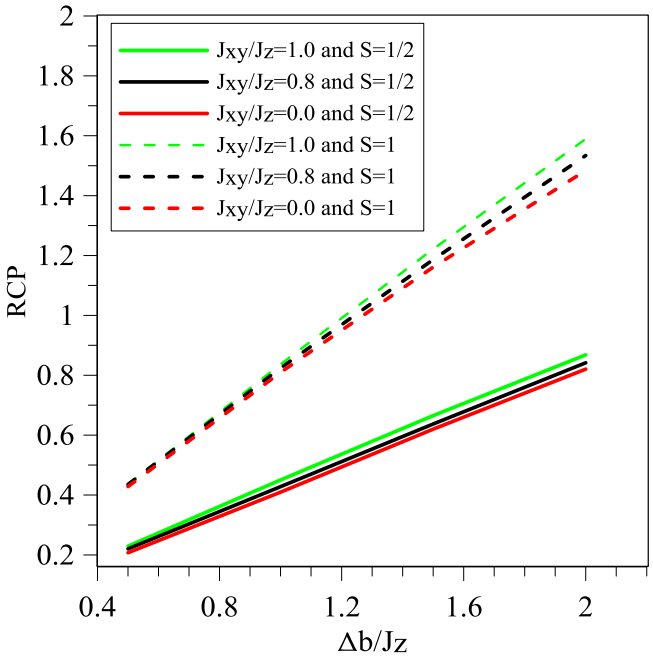


**Fig6.** *Relative Cooling Power (RCP) versus the external change field* $\Delta b/J_{xy}$ *for selected exchange anisotropy values* ($J_{xy}/J_z$*=0.0, 0.8, 1.0) and two spin values (1/2-1)*

In parallel with the isothermal entropy change, the relative cooling power (RCP) takes into account both the MCE amplitude and the material temperature range of work; it is also a crucial property for the evaluation of magnetocaloric performance and has been often used as a standard for good magnetic refrigeration materials. It is given by [20]:

$$RCP(b_2 \rightarrow b_1) = -\Delta S_{M\max} \times \Delta T_{1/2} \tag{24}$$

where $\Delta S_{M\max}$ and $\Delta T_{1/2}$ are the maximum value and the full width at half maximum of magnetic entropy change respectively.

Fig. 6 shows the RCP as a function of applied magnetic field for selected values of exchange anisotropy ratio (0, 0.8 and 1). As it can be seen from this figure, RCP curves increase linearly while increasing the magnetic field. In addition, anisotropy exchange has little influence on the RCP especially at weak fields. Thus, to reinforce the RCP, it is necessary to work under relatively high fields. Similar behavior of the influence of interaction anisotropy on Relative Cooling Power (RCP) by increasing the magnetic field exists in literature [21].

## 4. Conclusions and Proposals

In this paper, we have investigated the effect of anisotropy exchange and spins magnitude (½ and 1) on the ferromagnetic Ising-Heisenberg model with Pair approximation method in a three-dimensional lattice.

It is observed that the anisotropy parameter presents a great influence on all thermal dependencies of thermodynamic quantities, such as magnetization, entropy, and exchange entropy. Moreover, the critical temperature shifts and rises while decreasing anisotropy parameter; it is also noticed that the magnetocaloric effect with high spin (S> ½) becomes larger enhancing the performances (MCE and RCP) of the future magnetic refrigerators.

Our results suggest to the experimental researchers that materials with a strong ferromagnetic interaction could be synthesized by choosing high spin elements with significant exchange anisotropy. Such materials should lead to substantial improvements in the field of magnetic refrigeration, especially at room temperature.

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