Magnetocaloric properties of a spin-1 anisotropic ferromagnetic Heisenberg system within the pair approximation

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Abstract
The thermodynamics properties of a ferromagnetic Heisenberg model with the exchange anisotropy of a spin-1 system in presence of single-ion anisotropy (D) and external magnetic field (h) are studied here using the pair approximation approach. We treat the model in three-dimensional lattice with coordination number z = 6. Other coordination numbers have been also examined. Within this approximation based on Gibbs free energy, we investigate the effect of both single-ion and exchange coupling anisotropies on the thermodynamic quantities such as magnetization, entropy, heat capacity, susceptibility and entropy change as in-process measures. The objective is to describe magnetocaloric effect (MCE) and relative cooling power aiming to optimize them. We find some interesting phenomena in these quantities due to external magnetic field, exchange anisotropy and single-ion anisotropy. For conflicting interactions, re-entrance behavior can occur. Interestingly, high spin size as well as strong exchange or single-ion anisotropy can improve the MCE performances.

1. Introduction
Ferromagnetic spin systems are interesting thanks to their important properties, such as magnetocaloric effects (MCE) and their promising applications in magnetic refrigeration especially around the room temperature [1–3]. Theoretically, these systems are usually studied in the Ising models for which all the spins lie along the same axis, i.e. z-axis. However, in real system, spins may be oriented in the three space directions leading to a more complicated configuration, which requires an accurate quantum treatment such as Heisenberg models. In addition, spins interaction can be stronger in one direction than the others. Thus, we need to add an exchange anisotropy term to explore its effects on the phase diagrams. It should also be mentioned that the exact solutions are usually unreachable. Therefore, it is often necessary to introduce valuable approximations. Ferromagnetic spin systems have been studied by using many techniques in line with their different applications. Blume–Capel model is among these approaches including nearest-neighbors (nn) interactions on the Bethe lattice by using recursion equations for the coordination numbers z = 1–6 [4]. Thermodynamic properties of ferromagnetic Heisenberg spin-1/2 system were also investigated and second-nn effects were analyzed in the Oguchi’s approximation showing Schottky-peaks of the specific heat [5].

The critical behaviors of Heisenberg ferromagnetic system on a square lattice were examined theoretically by the Green’s function technique taking into account spins quantum nature [6]. Recently, Mert et al have studied the spin-1 within pair approximation method (PA) in presence of a negative second-nn exchange interaction and revealed several anomalies at low temperature [7].

In this work, we aim to investigate spin-1 Heisenberg ferromagnetic system on a 3D lattice within the PA. This method has been introduced at first and applied to XYZ system by Bukman et al [8]. Later, other authors have developed and tested it in various systems such as Balcerzak [9]. The PA is considered as relatively simple
but more accurate than the mean field approximation (MFA). Besides, such approximation takes into account the local nn correlation functions, which appear in a natural way from the spin-pair Hamiltonian [9, 10].

Our main objective is to evaluate the influence of both exchange anisotropy and crystal field on the MCE in the above mentioned system, and we hope providing argued proposals to illuminate designers while improving refrigeration performances of their devices.

The paper is organized as follows: in section 2, the formalism of PA is described, and the basic self-consistent equations are obtained. In section 3, the numerical results of magnetic properties are presented and discussed. In section 4, conclusions are given.

2. General formalism

2.1. Spin Hamiltonian
We consider a three-dimensional ferromagnetic Heisenberg model with exchange anisotropy and single-ion anisotropy under an external magnetic field. The spin Hamiltonian is given by:

\[ H = -\sum_{<ij>} [J_{ij} (S_i^x S_j^x + S_i^y S_j^y) + J_{ij}^z S_i^z S_j^z] - D \sum_i (S_i^z)^2 - h_0 \sum_i S_i^z, \]  

(1)

where \( S_i \) is a spin-1 operator, \( D \) is the parameter of single-ion anisotropy and \( h_0 \) is the external field applied along the z-direction. The first summation denoting the exchange contribution is performed over all pairs of nn spins. The parameter that measures the ordering of the system (or the long-range order parameter) is given by the magnetization per site. Here, one must notice that the anisotropy is originated both by exchange coupling and crystalline field. Thus, for example, \( D > 0 \) favors the spin alignment in the z direction, while \( D < 0 \) tends to retain spins within the xy plane. In addition, it is worth noting that exchange couplings in the z-axis or the xy plane are not equivalent. They depend on the strength and the sign of the two constants \( J_{ij}^z \). Therefore, the special case \( J_\perp / J_\parallel = 0 \) corresponds to the pure Ising model, while the case \( J_\perp / J_\parallel = 0 \) is consistent with the isotropic Heisenberg model. Here we restrict ourselves to the limit \( 0 \leq J_\perp / J_\parallel \leq 1.2 \).

2.2. PA method
According to the cluster variational method with PA, the single site \( i \) effective Hamiltonian can be rewritten as follows [9]:

\[ H_i = -(z\lambda + h_0)S_i^z - D(S_i^z)^2. \]  

(2)

Thus, the single site partition function takes the following form:

\[ Z_i = 2e^{iD} \cosh[\beta(z\lambda + h_0)] + 1. \]

(3)

On the other hand, the Hamiltonian for a cluster with two spins \( (i,j) \) is given by:

\[ H_{ij} = -J_\perp [(S_i^x S_j^x + S_i^y S_j^y)] - J_\parallel (S_i^z S_j^z) - D[(S_i^z)^2 + (S_j^z)^2] - \Lambda'(S_i^z + S_j^z). \]  

(4)

In the PA, the last Hamiltonian can be written in the form of the 9 \( \times \) 9 following matrix:

\[
\begin{bmatrix}
-n_j - 2\Lambda' - 2D & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\Lambda' - D & 0 & -J_{ij} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -J_{ij} - 2D & 0 & -J_{ij} & 0 & 0 & 0 & 0 \\
0 & -J_{ij} & 0 & -\Lambda' - D & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -J_{ij} & 0 & -J_{ij} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -J_{ij} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -J_{ij} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -J_{ij} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -J_{ij} + 2\Lambda' - 2D & 0 \\
\end{bmatrix}
\]

(5)

in which \( \Lambda' \) is the effective field acting on two spins of cluster and taking the form \( \Lambda' = (z - 1)\lambda + h_0 \), where \( z \) is the coordination number of a given spin and \( \lambda \) is a common variational parameter playing a major role in the
PA method [8, 9]. It is clear from the above matrix elements that the matrix representation of $H_{ij}$ in this base is symmetric. Therefore, we need to determine the eigenvalues by diagonalizing the matrix of $H_{ij}$.

After performing calculations, we find the following eigenvalues:

$$
\begin{align*}
E_1 &= J_{ij} - 2D \\
E_2 &= -J_{ij} + 2\lambda' - 2D \\
E_3 &= -J_{ij} - 2\lambda' - 2D \\
E_4 &= J_L - \lambda' - D \\
E_5 &= J_L + \lambda' - D \\
E_6 &= -J_L + \lambda' - D \\
E_7 &= -J_L - \lambda' - D \\
E_8 &= \frac{1}{2}J_{ij} - D + \sqrt{(D - \frac{J_{ij}}{2})^2 + 2J_L^2} \\
E_9 &= \frac{1}{2}J_{ij} - D - \sqrt{(D - \frac{J_{ij}}{2})^2 + 2J_L^2}.
\end{align*}
$$

These eigenvalues will be used below to calculate partition functions. Thus, the corresponding partition function for double site is written:

$$Z_{ij} = e^{\beta(-J_{ij}+2D)} + 2e^{\beta(J_{ij}+2D)} \cosh(2\beta\lambda') + 4e^{3D} \cosh \beta \lambda \cosh(\beta \lambda') 
\times (\beta \lambda') \cosh(\beta J_L) + 2e^{\beta(-J_{ij}+D)} \cosh(\beta \lambda'),$$

where $w = \sqrt{(D - \frac{J_{ij}}{2})^2 + 2J_L^2}$, $\beta = 1/k_BT$ and $k_B$ is the Boltzmann constant.

The calculation of partition function has a key role since all thermodynamic quantities can be derived from it.

Thus, the magnetization $m_i$ for single site defined by:

$$m_i = \langle S_i^z \rangle = \frac{\text{Tr}[S_i^z \exp(-\beta H_i)]}{Z_i}$$

takes the following expression:

$$m_i = \frac{2 \sinh[\beta(z\lambda + h)]}{2 \cosh[\beta(z\lambda + h)] + \exp(-\beta D)}.$$

Similarly, the magnetization $m_{ij}$ for pair of sites defined by:

$$m_{ij} = \langle S_i^z + S_j^z \rangle = \frac{\text{Tr}_{ij}[S_i^z + S_j^z \exp(-\beta H_{ij})]}{Z_{ij}}$$

becomes:

$$m_{ij} = \frac{4e^{\beta(J_{ij}+2D)} \sinh[2\beta(\lambda' + h)] + 4e^{3D} \cosh(\beta J_L) \sinh[\beta(\lambda' + h)]}{Z_{ij}}.$$

In order to find measureable thermodynamic variables, we calculate first the total Gibbs free energy which is given by the general expression:

$$G = \langle H \rangle - \sigma T,$$

where $\sigma$ is the entropy of the system and $\langle H \rangle$ is thermodynamic average of the Hamiltonian in equation (1) given by [8]:

$$\langle H \rangle = U - hM.$$
$U$ stands for the internal energy and $M$ is the total magnetization expressed respectively by:

$$U = -\frac{Nz}{2} \left[ J_\perp \langle S_i^z S_j^z + S_i^z S_j^z \rangle + J_\parallel \langle S_i^z S_j^z \rangle \right],$$

$$M = N \langle S_i^z \rangle.$$  \hspace{1cm} (13)

Here, $N$ is the total number of spins and $z$ is the number of the nn. Note that the internal energy $U$ includes both longitudinal $\langle S_i^z S_j^z \rangle$ and transverse $\langle S_i^z S_j^z + S_i^z S_j^z \rangle$ spin correlations.

Using the method presented in [9, 11], the total Gibbs free energy per one lattice site can be finally presented in the form:

$$G = \frac{z}{2} G_{ij} - (z - 1) G_i,$$  \hspace{1cm} (15)

where the Gibbs free energy $G_i$ for single site is written:

$$G_i = -k_B T \ln \{2e^{\beta D} \cosh [\beta (\Lambda + h)] + 1\},$$

and the pair Gibbs free energy $G_{ij}$ is defined by:

$$G_{ij} = -k_B T \ln \{Z_{ij}\}. \hspace{1cm} (17)$$

Thus, in further calculations, $m_i$ and $m_{ij}$ can be treated as the only two variational parameters determining the Gibbs energy. With respect to these parameters the Gibbs energy functional should then be minimized [9].

2.3. Thermodynamic properties

The variational parameter $\lambda$ can be found by minimizing the total Gibbs free energy [14]

$$\frac{\partial G}{\partial \lambda} = 0.$$ \hspace{1cm} (18)

This condition leads to the equation:

$$m_i = \frac{1}{2} m_{ij}.$$ \hspace{1cm} (19)

or, to the explicit form:

$$m_i = \frac{2 \sinh[\beta(z\Lambda + h)]}{2 \cosh[\beta(z\Lambda + h)] + \exp(-\beta D)} = \frac{2e^{\beta D(z\Lambda') \sinh[\beta(\Lambda' + h)]}}{Z_{ij}},$$ \hspace{1cm} (20)

The last equation (20) means that the magnetization for single spins and the pair spins are the same in order to keep the symmetry of the model.

Having obtained the parameter $\lambda$ after solved numerically the above relationship, now one should derived all thermodynamic properties from the total Gibbs free energy as the function of temperature $\left(\beta = \frac{1}{k_B T}\right)$ and the external magnetic field.

The first-order derivative of the Gibbs energy (equation (15)) over the field gives the magnetization per one lattice site:

$$m = -\frac{1}{N} \left( \frac{\partial G}{\partial h} \right)_T.$$ \hspace{1cm} (21)

and the second-order derivative of the Gibbs energy over the field leads to expression of the magnetic susceptibility:
In turn, the entropy per one lattice site can be found from the general formula:

\[
\sigma = -\frac{1}{N} \left( \frac{\partial G}{\partial T} \right)_h.
\]  

(23)

We can similarly get the expression of the specific heat as a first-order derivative of the Gibbs free energy over temperature:

\[
C_h = -\frac{1}{N} T \left( \frac{\partial^2 G}{\partial T^2} \right)_h = T \left( \frac{\partial \sigma}{\partial T} \right)_h.
\]  

(24)

To study the MCE in the 3D ferromagnetic system with exchange and crystal field anisotropies \((S = 1)\), we should calculate the entropy change as a function of the temperature and external magnetic field which is given by the Maxwell relation \([12]\):

\[
\Delta S_T = -\int_0^h \left( \frac{\partial m}{\partial T} \right) dh
\]  

(25)

and the relative cooling power (RCP) which has been often used as a parameter measuring the performance of the magnetic refrigeration is defined as \([2]\):

\[
\text{RCP}(h_2 \rightarrow h_1) = -\Delta S_{m \text{ max}} \times \Delta T_{1/2},
\]  

(26)

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

2.4. Critical temperature

The Curie temperature \(T_C\) can be found by linearization of self-consistent equation \((20)\) when \(\lambda \rightarrow 0\) and external magnetic field \(h = 0\), leading to:

\[
\frac{z}{(z - 1)(2 + e^{-\beta_C D})} = \frac{2e^{\beta_C (J_{1/} + D)}}{e^{\beta_C (-J_{1/} + D)} + 4\cosh(\beta_C J_{1/}) + 2e^{-\beta_C J_{1/}} - \frac{\beta_C J_{1/}}{2}},
\]  

(27)

where \(\beta_C = 1/k_B T_C\).

Then, the detailed phase diagrams can be obtained on the \((D/J, k_B T/J_{1/})\) and \((k_B T/J_{1/})\) planes by investigating the thermal variations of the order-parameters and free energy. The obtained phase lines are classified in terms of their stabilities by comparing the free energy values. Some examples of the numerical calculations, and the discussion of the theoretical results will be given in section 3.

3. Numerical results and discussion

In this section, we use the above formulas to calculate all thermodynamic properties of the spin-1 ferromagnetic Heisenberg model on the 3D lattice with coordination number \(z = 6\).

Firstly, we have to get the thermal behavior of the variational parameter \(\lambda\) because all thermodynamic quantities are expressed by this parameter \([13, 14]\).

In this purpose, by using equation \((20)\) and developing a numerical program, we were able to plot the curve of thermal variational of the parameter \(\lambda\) for different values of exchange anisotropy ratio \(J_{1/} / J_{1/} = 0.0; 0.6; 0.8; 1.0; 1.1; 1.2\) as shown in figure 1. Notice that the cases \(J_{1/} / J_{1/} = 0\) and \(J_{1/} / J_{1/} = 1\), correspond respectively to the Ising and the isotropic ferromagnetic Heisenberg models.

Additionally, in order to explore the phase diagram of the system, we calculate the Curie temperature on the basis of equation \((27)\). Typical results have been depicted in figure 2(a), showing the reduced critical temperature versus the exchange anisotropy ratio, for three selected values of the coordination number \(z \) \((z = 4–6)\) corresponding respectively to 2D, bilayer and 3D lattices. It is seen that \(T_C\) is higher for \(z = 6\) (bulk system) for all \(J_{1/} / J_{1/}\) values and decreases gradually when \(J_{1/} / J_{1/}\) is increased, while the MFA predicts a
linear decreasing [15]. Thus, our results are much closer to those obtained by field theory [16] or PA [17]. These results are equally in concordance with those obtained by Balcerzak et al [11] for $S = 1/2$ and $D = 0$. Figure 2(b) depicts the single-ion anisotropy effects on the phase diagram for $z = 6$. It is clear that positive values of $D$ enhance $T_C$, notably for low exchange anisotropy.

In figure 3(a), the spontaneous magnetization is depicted against reduced temperature $k_B T / J_{//}$ for selected values of exchange anisotropy parameter $J_\perp / J_{//} = 0.0 - 0.6 - 0.8 - 1.0 - 1.2$. In absence of magnetic field, the magnetization starts from its saturation value and decreases rapidly to zero at Curie temperature $T_C$. We can also see that the critical temperature shifts and rises with decreasing anisotropy parameter $J_\perp / J_{//}$. For $J_\perp / J_{//} = (1, 0.8, 0.6 \text{ and } 0)$, the corresponding values of Curie temperature are respectively:
While for \( J_{\perp}/J_{\parallel} \geq 1.1 \), an important feature is noticed, the spontaneous magnetization exhibits a re-entrant behavior at sufficiently low temperature where it completely vanishes. This is due to the conflicting effects of \( J_{\perp} \) and \( J_{\parallel} \). The first exchange coupling tends to remain the spins in \( z \)-axis while the second one attract them within the orthogonal plane. Consequently, the longitudinal magnetization has a minimum value over this low temperature range.

Whereas the figure 3(b) presents the thermal variation of magnetization in presence of an applied magnetic field. It is clear that when \( h \) is fixed, the magnetization decreases slowly from its saturation value to small but not vanishing values as the temperature increases because of the spin–spin correlations. Comparing the curves for \( h/J_{\parallel} = 0.5–2 \), we realize that with the increase of the external field, the derivatives of the Gibbs energy become smooth. For negative values of single-ion anisotropy (figure 3(c)), special feature of the spontaneous

\[ k_B T_c/J_{\parallel} = 2.949; 3.158; 3.303 \text{ and } 3.476. \]
magnetization occurs, where \( m(T) \) starts from the value 0.5 (for \( J_\perp/J_\parallel = 1 \), increases up to a maximum value before descending as the previous cases. This is the sign of reentrance as seen similarly for negative exchange anisotropy.

The thermal evolution of the susceptibility per site is depicted in figure 4, without and in presence of an applied field. The sharp peak that occurs in absence of field agrees well with the vanishing point of spontaneous magnetization \( T_C \), while it becomes smoother and moves to higher temperature when a field is applied. Similar observations have been reported by Balcerzak with a spin-\( 1/2 \) system [9].

The magnetic specific heat per one spin upon temperature in absence of the field \( h/J_\parallel = 0.0 \) for different exchange anisotropy strengths \( J_\perp/J_\parallel \) is reported in figure 5(a). As observed in the susceptibility, the peak of specific heat is related to the critical temperature.
The field effect on the specific heat profile is illustrated in figure 5(b) (for $J_\perp/J_\parallel = 0.8$). As we already mentioned, the transition becomes smoother as $h/J_\parallel$ increases, i.e. the peak amplitude decreases and the temperature at which the peak occurs moves to higher temperatures.

In figure 6, we plot the entropy curves versus the dimensionless temperature $k_B T / J_\parallel$, for various values of the exchange anisotropy $J_\perp/J_\parallel$, in absence of the field (figure 6(a)) and for some selected values of the applied reduced magnetic field $h/J_\parallel$ (figure 6(b)). We can see that the curves increase and reach their maximum values with increasing temperature at fixed magnetic field. For different magnetic fields, the value of entropy increases more slowly with increasing magnetic field. Positive values of single-ion anisotropy low down the entropy and reinforce the order (see figure 6(c)).

Our results are qualitatively consistent with those obtained by the similar technique by Balcerzak for a spin-$1/2$ systems [9, 11].

**Figure 6.** (a) Entropy per one spin upon temperature without the external field for various $J_\perp/J_\parallel$ values. (b) Entropy per one spin upon temperature for some selected values of the external magnetic field with $J_\perp/J_\parallel = 0.8$. (c) Entropy per one spin upon temperature in absence of the magnetic field for some selected values of single-ion anisotropy with $J_\perp/J_\parallel = 1.0$. 
The magnetic entropy change \((\Delta S_T)\) which was calculated by using equation \((25)\), and plotted in figure 7(a) for a set of selected values of \(J_\perp / J_\parallel\). A large peak \((-\Delta S_{M\text{\_max}})\) together with a shoulder can be observed in the \(\Delta S_T\) curves around the FM and SR transition temperatures, respectively. Such two peaks (shoulder) behaviors in \((-\Delta S_T)\) curves were also observed in experimentally reported results with HoZn \([18]\) and ErGa materials \([19]\). As seen in figure 7(b), the applied field enhance the maximum of entropy change which is also moved towards higher temperatures. Similar effects can be obtained by increasing the single-ion anisotropy (figure 7(c)).

Finally, the relative cooling power is another key parameter permitting rigorous cataloging of the MCE for a given material \([1]\). Using equation \((26)\) for numerous change entropy curves under different fields, we have reported our summarizing results in figure 8.

It is clear that RCP increases progressively with field measuring the energy that can be transferred between the hot and cold reservoirs during the refrigeration process. Distinction between the curves for various values of the exchange begins to be detected only in high fields. To enlarge RPC, it is necessary to provide high magnetic field. On the other hand, we can conclude that by reinforcing the exchange, single-ion anisotropy or spin size, the RCP can be notably enhanced (figures 8(a)–(c)). This observation is in line with the experimental reality \([1, 20]\).

Figure 7. (a) The isothermal entropy change per site in the external field \(kT / J_\parallel = 0.5\) for different \(J_\perp / J_\parallel\) ratios. (b) The isothermal entropy change per site for \(J_\perp / J_\parallel = 0.8\) under different field values: \(\Delta h / J_\parallel = 0.5; 1; 1.5; 2\). (c) The isothermal entropy change per site in the external field \(\Delta h / J_\parallel = 0.5\) for single-ion anisotropy values \((D / J_\parallel = 1.5-0.0)\) by taken \(J_\perp / J_\parallel = 1.0\).
4. Conclusion

In this work, we have studied the spin-1 ferromagnetic Heisenberg system on a three dimensional lattice with PA method including the interaction anisotropy and external magnetic field into the Hamiltonian model.

The use of the PA shows that interaction anisotropy has a great influence on the all magnetic properties. The present study illustrates also that the critical temperature shifts and rises while decreasing anisotropy parameter $J_\perp/J_\parallel$. In fact, at the critical point, the fluctuations become very important and are affecting consequently the properties of the system, but the exchange anisotropy tends to cancel the critical fluctuations and therefore raises the critical temperature.

As a final note, we should mention that the entropy change peak shifts down, when the $J_\perp/J_\parallel$ anisotropy ratio increases. It means that the magnetocaloric properties will be influenced by exchange anisotropy parameter.

Our work is providing a clear answer on the role of exchange anisotropy and its impact on the MCE. Large RCP, large $\Delta T$ change, multiple exchange forms and wide operating temperature range make a material as an
attractive candidate for magnetic refrigeration in the suitable temperature region. Thus, by our contribution, we invite the scientific community to double their experiments efforts to synthesize new optimized materials for magnetic refrigeration.

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