VNU Journal of Science: Mathematics – Physics, Vol. 40, No. 4 (2024) 64-71

VNU Journal of Science: Mathematics - Physics

Journal homepage: https://js.vnu.edu.vn/MaP

Original Article

Theoretical Study of the Bilayer Honeycomb Spin Lattice in Transverse Field with Competing Ferromagnetic and Antiferromagnetic Interactions

Nguyen Tu Niem*

VNU University of Science, 334 Nguyen Trai, Thanh Xuan, Hanoi, Vietnam

Received 28 May 2024
Revised 17 July 2024; Accepted 10 December 2024

Abstract: In this work we studied theoretically the thermodynamics and magnetization process of this bilayer honeycomb spin lattice structure for the transverse field case using the transverse Ising model (TIM) [1] and mean field approximation (MFA). Theoretical investigations of the magnetization process in the AF bilayer honeycomb spin lattice with FM order in each layer using the Ising spin model in longitudinal and transverse fields have been shown. The obtained results show that the AF exchange interaction coupling has the effect of decreasing the critical fields and magnitude of the longitudinal susceptibility of the film.

Keywords: Bilayer film, Transverse Ising model, dynamical susceptibility,

1. Introduction

Among two-dimensional (2D) layered magnetic materials, the bilayer honeycomb lattices have attracted a significant amount of interest due to their interesting magnetic properties [2-4]. The magnetic properties of bilayer honeycomb lattices depend on the stacking of the layers and are different from those of a monolayer lattice [5]. More interestingly, these properties can be controlled by using strain, doping, and external fields [6-8]. These behaviors have important implications for fundamental research and possible applications. Therefore, the bilayer honeycomb lattices have become the subject of numerous experimental and theoretical studies.

* Corresponding author.
E-mail address: nguyentuniem@gmail.com

https//doi.org/10.25073/2588-1124/vnumap.4944

We have studied the magnetization process of the AA-stacked bilayer honeycomb spin lattice with an applied longitudinal field and the competition between two different kinds (antiferromagnetic: AF, or ferromagnetic: FM) of intra-layer and inter-layer spin exchange couplings.

This work is to extend theoretical works about the magnetization process in honeycomb spin lattice [9]. Our aim is to investigate the thermodynamic properties and the magnetization process of two ferromagnetic honeycomb AA-stacking spin layers under applied transverse fields.

The paper is organized as follows: In Section 2, we define the Ising model in longitudinal and transverse fields, the mean field approximation is used. In Section 3, the numerical results for magnetization, susceptibility, and free energy are discussed in detail. Finally, conclusions are given in Section 4.

2. Method of Calculation

We consider the Ising model on a honeycomb lattice consisting of two spin layers and N spins in every layer (Fig. 1). The spins of each layer are distributed equally in two triangular a and b spin sub-lattices. The system described by Hamiltonian:

$$H = -\sum_{nvj}\left(h_0 s^z_{nvj} + \Omega_0 s^x_{nvj}\right) - \frac{J}{2}\sum_{n,j,\Delta}\left(s^z_{naj}s^z_{nb,j+\Delta} + s^z_{nbj}s^z_{na,j-\Delta}\right) - \frac{J'}{2}\sum_{n,v,j,\rho} s^z_{nvj}s^z_{nvj+\rho} - J_P\sum_{vj} s^z_{1vj}s^z_{2vj} \quad (1)$$

Here n = 1, 2 is the number of layers. h_0 (Ω_0) is the external magnetic field perpendicular and parallel to the monolayer spin plane and given in energy unit. J, J' are nearest neighbors (NN), second nearest neighbors (2nd NN) inplane exchange interaction couplings between spins, respectively. J_p is the exchange coupling between the spin NN pair at different layers but belonging to the same sub-lattice.

The in-plane position of a spin in the ν–sub-lattice is indicated by two-dimensional lattice vectors $\boldsymbol{j}$. s^{α}_{nvj} $(\alpha = x, y, z)$ denote the components of a spin operator in the crystallographic xyz frame.

A spin at site j has three inter-sublattice nearest neighbors of the different type and six intra-sublattice second nearest neighbors of the same type. The position of the NN spins are denoted by vectors $\boldsymbol{\Delta}$: $\boldsymbol{\Delta}_1 = \frac{a_0}{2}\left(3,\sqrt{3}\right)$, $\boldsymbol{\Delta}_2 = \frac{a_0}{2}\left(3,-\sqrt{3}\right)$, $\boldsymbol{\Delta}_3 = a_0\left(-1,0\right)$ and the position of the 2nd NN spins are denoted by vectors $\boldsymbol{\rho}$: $\boldsymbol{\rho}_1 = \frac{a_0}{2}\left(3,\sqrt{3}\right) = -\boldsymbol{\rho}_4$, $\boldsymbol{\rho}_2 = \frac{a_0}{2}\left(3,-\sqrt{3}\right) = -\boldsymbol{\rho}_5$, $\boldsymbol{\rho}_3 = a_0\left(0,-\sqrt{3}\right) = -\boldsymbol{\rho}_6$. a_0 is the length of the hexagonal edge.

The spin operators s^{α}_{nvj} can be written in form $\delta s^z_{nvj} = s^z_{nvj} - m_{nvz}$, $\delta s^x_{nvj} = s^x_{nvj} - m_{nvx}$, where δs^z_{nvj} is the spin fluctuation operator.

In the mean field approximation as zero approximation, by neglecting fluctuation operators we can write Hamiltonian (1) in the form:

$$H_0 = 3N\sum_n\left[J m_{1az}m_{2bz} + J'\left(m^2_{naz} + m^2_{nbz}\right)\right] + NJ_p\sum_v m_{1vz}m_{2vz} - \sum_{nvj}\left(h_{nv}s^z_{nvj} + \Omega_0 s^x_{nvj}\right) \quad (2)$$

$$h_{1a(1b)} = h_0 + 3J\, m_{1bz(1az)} + 6J'\, m_{1az(1bz)} + J_p m_{2az(2bz)},$$

$$h_{2a(1b)} = h_0 + 3J\, m_{2bz(2az)} + 6J'\, m_{2az(2bz)} + J_p m_{1az(1bz)} \quad (3)$$

Where m_{nvx}, m_{nvz} (the index v takes values a or b) are thermodynamic average of the spin moment components per site. For example, $m_{nvz} = \langle s^z_{nv}\rangle$ and $\langle\ldots\rangle = Tr[\exp(-\beta H)\ldots]/\,Tr[\exp(-\beta H)]$, and $m_{nv} = \sqrt{m^2_{nvx} + m^2_{nvz}}$.

h_{nv} and Ω_0 are the longitudinal and transversal components of the total field $\gamma_{nv} = \sqrt{h^2_{nv} + \Omega^2_0}$ acting on the ν-sub-lattice spin. Hamiltonian H_0 can be diagonalizable by the unitary transformation:

$$s_{nv\boldsymbol{j}}^{x} = \frac{h_{nv}}{\gamma_{nv}} S_{nv\boldsymbol{j}}^{X} + \frac{\Omega_0}{\gamma_{nv}} S_{nv\boldsymbol{j}}^{Z},\ s_{nv\boldsymbol{j}}^{z} = -\frac{\Omega_0}{\gamma_{nv}} S_{nv\boldsymbol{j}}^{X} + \frac{h_{nv}}{\gamma_{nv}} S_{nv\boldsymbol{j}}^{Z}\ . \tag{4}$$

Hamiltonian of the system in new form:

$$H_0 = 3N\sum_n\left[Jm_{1az}m_{2bz} + J'\left(m_{naz}^2 + m_{nbz}^2\right)\right] + NJ_p\sum_v m_{1vz}m_{2vz} - \sum_{nv\boldsymbol{j}}\gamma_{nv}S_{nv\boldsymbol{j}}^{Z} \tag{5}$$

From the Hamilonian (5) we obtain the expression for the free energy $f = -\frac{1}{4N\beta} lnTr\left(e^{-\beta H_0}\right)$ and magnetic moment per spin:

$$f = \frac{3}{4}\sum_n \left[Jm_{naz}m_{nbz} + J'\left(m_{naz}^2 + m_{nbz}^2\right)\right] + \frac{J_p}{4}\sum_v m_{1vz}m_{2vz} - \frac{1}{4\beta}\sum_{nv}\ln\left\{\frac{sh\left[\left(s+\frac{1}{2}\right)y_{nv}\right]}{sh\left(\frac{y_{nv}}{2}\right)}\right\}, \tag{6}$$

$$m_{nvz} = \frac{h_{nv}}{\gamma_{nv}} b_s(y_{nv}), m_{nvx} = \frac{\Omega_0}{\gamma_{nv}} b_s(y_{nv}), m_{nv} = \sqrt{m_{nvx}{}^2 + m_{nvz}{}^2}\ . \tag{7}$$

$$y_{nv} = \beta\gamma_{nv} \tag{8}$$

The Brillouin function $b_s(y)$ figured in the equation (9) has the following form

$$b_s(y) = (s + 1/2)cth\big((s + 1/2)y\big) - \frac{1}{2}cth(y/2). \tag{9}$$

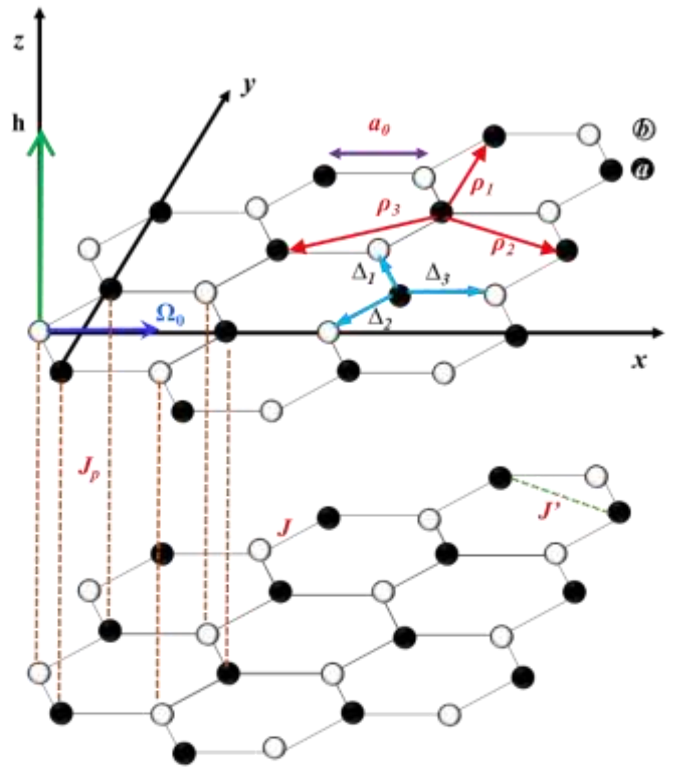

Figure 1. The structure of bilayer honeycomb lattice in longitudinal and transversal field.

3. Results and Discussions

In this part, we consider the case where the initial spin configuration without field is FM order in each layer and AF order of the spins in the two layers and the average spin moments of two sub-lattices in each layer are equal. In the numerical calculation, all quantities are non-unit, and the unit of energy is the magnitude of the NN exchange energy $|J|$. That means temperature $\tau = \frac{k_B T}{|J|} = \beta^{-1}$, and field $h = \frac{g\mu_B B}{|J|}$.

$$m_{1az} = m_{1bz} = m_{1z}; m_{2az} = m_{2bz} = m_{2z}; m_{n\nu x} = m_{nx};$$

$$m_{nz} = \frac{h_n}{\gamma_n} b_s(y_n); m_{nx} = \frac{\Omega_0}{\gamma_n} b_s(y_n); \qquad y_n = \beta\gamma_n, \gamma_n = \sqrt{h_n^2 + \Omega_0^2} \tag{10}$$

$$m_n = \sqrt{m_{nz}^2 + m_{nx}^2} \; ; \; m = \frac{1}{2}(m_1 + m_2). \tag{11}$$

The magnetic fields exerting on the spin at the first and the second layers and the free energy per spin are

$$h_1 = h_0 + 3(J + 2J')\, m_{1z} + J_p m_{2z}, h_2 = h_0 + 3(J + 2J')\, m_{2z} + J_p m_{1z}, \tag{12}$$

$$f = \frac{1}{4}\left\{3(J + 2J')(m_{1z}^2 + m_{2z}^2) + 2J_p m_{1z} m_{2z}\right\} - \frac{1}{2\beta}\sum_n ln\left\{\frac{sh\left[\left(s+\frac{1}{2}\right)y_n\right]}{sh\left(\frac{y_n}{2}\right)}\right\}, \tag{13}$$

The first and the second derivatives of free energy with respect to the magnetic field give us the spin moment *m* and the susceptibility χ of the bilayer film.

$$m_z = -\frac{\partial f}{\partial h_0} = \frac{1}{2}(m_{1z} + m_{2z}), \qquad m_x = -\frac{\partial f}{\partial \Omega_0} = \frac{1}{2}(m_{1x} + m_{2x}) \tag{14}$$

The longitudinal static susceptibility is given by

$$\chi^z = -\frac{\partial^2 f}{\partial h_0^2} = \frac{1}{2}\left\{\frac{Z_1 + Z_2 - 2[3(J+2J') - J_p]Z_1 Z_2}{1 - 3(J+2J')[Z_1 + Z_2] + [9(J+2J')^2 - J_p^2]Z_1 Z_2}\right\}, \tag{15}$$

$$Z_n = \frac{1}{\gamma_n^2}\left[b_s(y_n)\frac{\Omega_0^2}{\gamma_n} + \beta h_n^2 b_s'(y_n)\right]. \tag{16}$$

The first derivative of the Brillouin function presented in (16) is written as

$$b_s'(y) = \frac{1}{4sh^2(y/2)} - \frac{(s+1/2)^2}{sh^2[(s+1/2)y]}. \tag{17}$$

Similarly, the transverse static susceptibility, $\chi^x = -\frac{\partial^2 f}{\partial \Omega_0^2}$ is

$$\chi^x = \frac{1}{2}\sum_n Z_n + \frac{1}{2}\left\{\frac{3(J+2J')(k_1^2 + k_2^2) + 2J_p k_1 k_2 + [J_p{}^2 - 9(J+2J')^2](Z_1 k_2^2 + Z_2 k_1^2)}{1 - 3(J+2J')[Z_1 + Z_2] + [9(J+2J')^2 - J_p^2]Z_1 Z_2}\right\}, \tag{18}$$

where $k_n = \frac{\Omega_0 h_n}{\gamma_n^2}\left[\beta b_s'(y_n) - \frac{b_s(y_n)}{\gamma_n}\right]$. (19)

3.1. Magnetization

We investigate the magnetization process of the bilayer honeycomb lattice under the effect of the transverse field. Firstly, we study the magnetization of bilayer spin film with transverse field and temperature variation at a specified small longitudinal external field value h_0. The results are shown in Figure 2.

We found that while the effect of external longitudinal fields causes weak ferromagnetic properties of the film, the effect of the transverse field changes the magnitude of the total magnetic moment. The increase in transverse field leads to a decrease in the net spin moment and the reduction of the corresponding phase transition temperature of the film (Figure 2a).

Figure 2b shows the effect of the exchange interaction coupling between spins J_P on the magnetization. The increase in the magnitude of J_p leads to an increase in the phase transition temperature of the film.

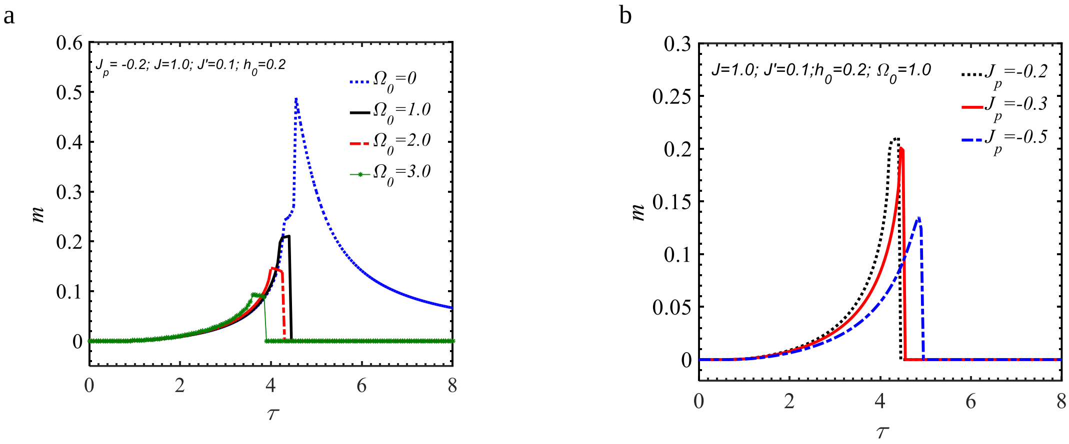

Figure 2. The dependence of the net spin moment on the temperature at a given value of the longitudinal field h_0 = 0.2 with different values of the transversal field Ω_0 (a) and the inter- exchange interaction J_p (b). The values of the parameters are: J = 1, J'= 0.1, s = 3/2 for all curves.

Next, we investigate the magnetization process of the bilayer honeycomb lattice under the effect of the transverse field.

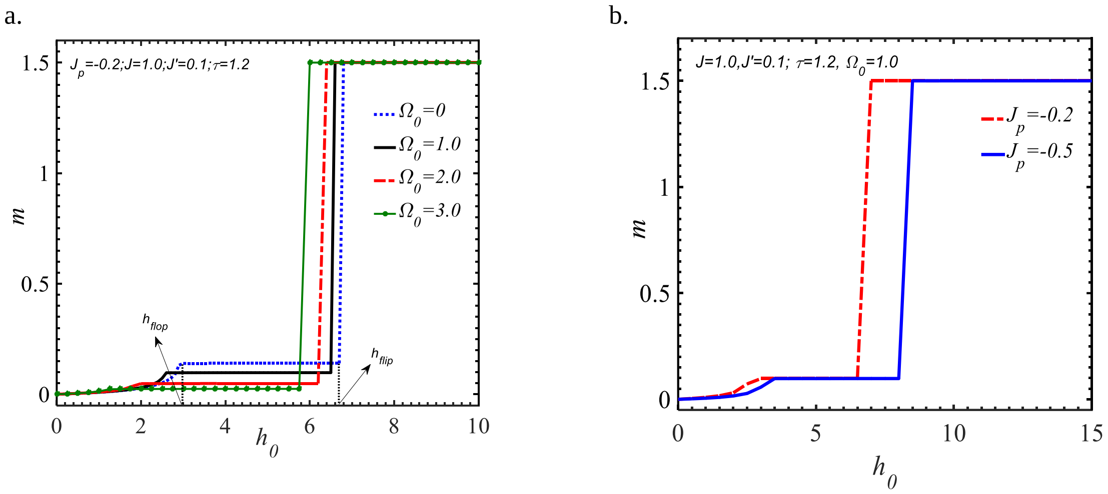

Figure 3. Magnetization process of the bilayer honeycomb spin lattice when changing the transverse field (a) and J_p (b). Here, the spins in the two layers are antiparallel. The chosen parameters are: s=3/2, τ=1.2.

In Figure 3, we plot the net spin moment of the film as a function of the external fields. The magnetization curves start at zero magnetic field. When the longitudinal magnetic field reaches a critical value, the interaction between the two material layers leads to a reversal of the magnetization of the second layer. The bilayer honeycomb film exhibits unusual behavior in the out-of-plane field: the first-order magnetization process with two critical longitudinal fields (or spin flop at low field and spin flip at high field). These critical values decrease with increasing transverse field and reducing the exchange interaction J_p.

The free energy of the antiparallel layer spin configuration changes discontinuously at two critical fields (Figure 4).

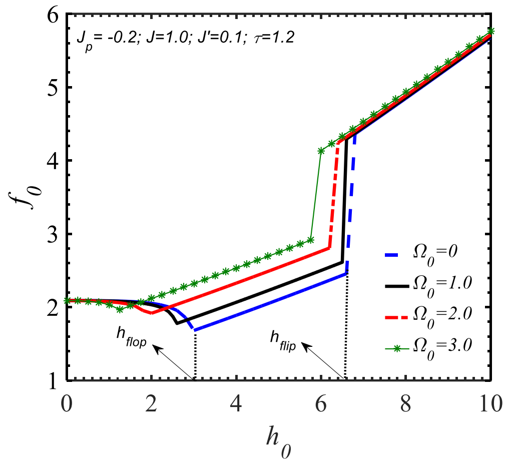

Figure 4. Field dependence of the free energy per spin of the bilayer honeycomb spin lattice. The initial spin configuration without fields is: FM order in each layer and AF order of the spins in the two layers. Here, s=3/2 and other parameters are shown in the figure.

3.2. Susceptibility

In this part we present the results of the calculations for the susceptibility of the film.

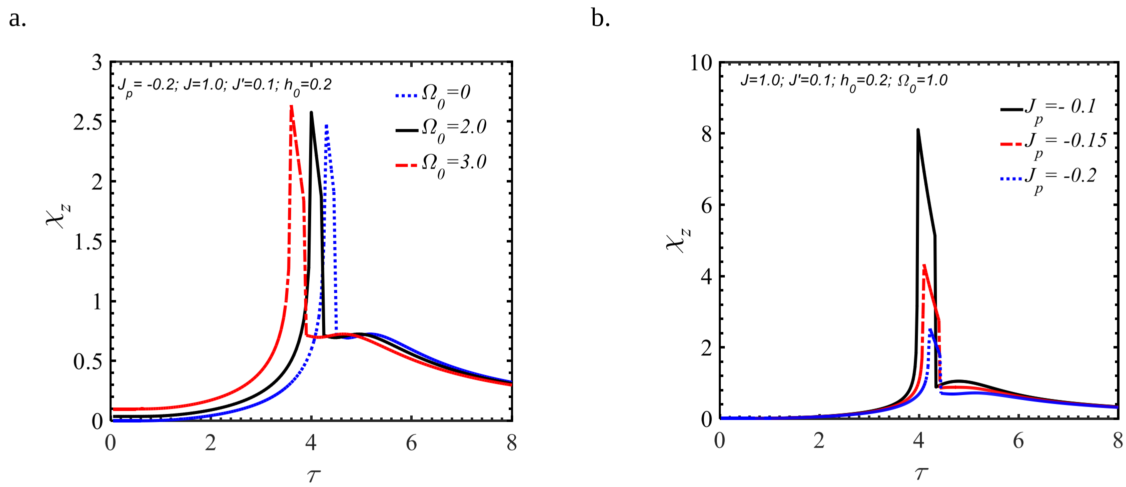

Figure 5. The dependence of the longitudinal susceptibility on the temperature at a given value of the longitudinal field h_0 = 0.2 with different values of the transversal field Ω_0 (a); and the inter- exchange interaction J_p (b). Here s=3/2 and chosen parameters are shown in the figure.

We have plotted the longitudinal susceptibility of the bilayer spin film as a function of the temperature at different values of the external fields and the inter-exchange interaction (Figure 5). We see the highest longitudinal susceptibility at the transition temperature for all values of the longitudinal and transversal fields. The longitudinal susceptibility decreases rapidly with increasing temperature of the system. With the increase in the transversal field, the highest point of the longitudinal susceptibility curve increases and shifts to the low-temperature region (Figure 5a). The inter-exchange interaction J_p impacts strongly on the highest longitudinal susceptibility. A slight decrease in the magnitude of the exchange interaction leads to a significant increase in the highest longitudinal susceptibility (Figure 5b).

a. b.

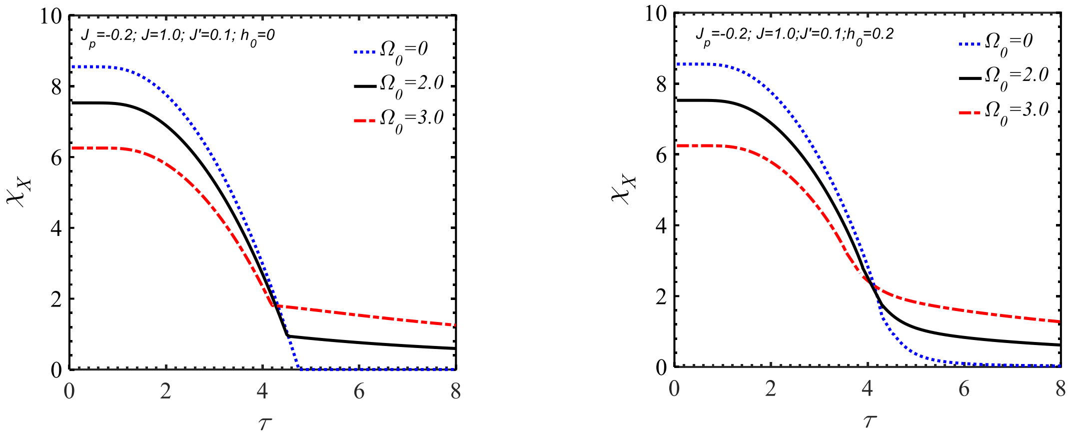

Figure 6. The effect of the longitudinal and transversal field on the transverse susceptibility of the bilayer spin film. The values of the parameters are: J = 1, J'=0.1, Jp=-0.2, s = 3/2.

The thermal variation of the transverse susceptibility for the bilayer honeycomb lattice is plotted in Figure 6. The graphs show a gradual decrease when the temperature increases. As the transerval field increases, the transverse susceptibility decreases. The longitudinal field which exerts on the film leads the transverse susceptibility to decrease more slowly.

4. Conclusions

In summary, we have theoretically investigated the magnetization process in the AF bilayer honeycomb spin lattice with FM order in each layer using the Ising spin model in longitudinal and transverse fields. In the frame of mean field approximation, the theoretical expressions for the free energy, the magnetization, and the susceptibility of the film are established as functions of the external fields, the temperature of the system, and the inter-layer exchange interaction. In the presence of the transversal field, the first order magnetization process happens at smaller values of the critical longitudinal fields. The AF exchange interaction coupling has the effect of decreasing the critical fields and magnitude of the longitudinal susceptibility of the film.

References

[1] B. T. Cong, N. T. Niem, B. H. Giang, Thermodynamic Properties of Ferroics Described by the Transverse Ising Model and Their Applications for CoNb2O6, Journal of Magnetism and Magnetic Materials, Vol. 483, 2019, pp. 136-142, https://doi.org/10.1016/j.jmmm.2019.03.093.

[2] R. Xu, X. Zou, Electric Field-Modulated Magnetic Phase Transition in Van der Waals CrI_3 Bilayers, Journal of Physical Chemistry Letters, Vol. 11, No. 8, 2020, pp. 3152-3158, https://doi.org/10.1021/acs.jpclett.0c00567.

[3] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, Y. Wang, Z. Q. Qiu, R. J. Cava, S. G. Louie, J. Xia, X. Zhang, Discovery of Intrinsic Ferromagnetism in Two-Dimensional Van der Waals Crystals, Nature, Vol. 546, No. 7657, 2017, pp. 265-269, https://doi.org/10.1038/nature22060.

[4] M. Gibertini, M. Koperski, A. F. Morpurgo, K. S. Novoselov, Magnetic 2D Materials and Heterostructures, Nature Nanotechnology, Vol. 14, No. 5, 2019, pp. 408-419, https://doi.org/10.1038/s41565-019-0438-6.

[5] B. Huang, G. Clark, E. Navarro-Moratalla, R. K. Dahlia, R. Cheng, L.S. Kyle, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, X. Xu, Layer-dependent Ferromagnetism in a Van der Waals Crystal Down to the Monolayer Limit, Nature, Vol. 546, No. 7657, 2017, pp. 270-273, https://doi.org/10.1038/nature22391.

[6] K. F. Mak, J. Shan, D. Ralph, Probing and Controlling Magnetic States in 2D Layered Magnetic Materials, Nature Reviews Physics, Vol. 1, No. 11, 2019, pp. 646-661, https://doi.org/10.1038/s42254-019-0110-y.

[7] S. Jiang, L. Li, Z. Wang, K. F. Mak, J. Shan, Controlling Magnetism in 2D CrI_3 by Electrostatic Doping, Nature Nanotechnology, Vol. 13, No. 7, 2018, pp. 549-553, https://doi.org/10.1038/s41565-018-0135-x.

[8] N. T. K. Oanh, N. H. Phong, N. T. Niem, B. T. Cong, N. D. Huy, B. H. Giang, Monte Carlo Investigation for an Ising Model with Competitive Magnetic Interaction Regime, Communications in Physics, Vol. 33, No. 2, 2023, pp. 205-212, https://doi.org/10.15625/0868-3166/18109.

[9] N. T. Niem, B. H. Giang, P. H. Thao, N. D. Huy, N. T. K. Oanh, B. T. Cong, Magnetization Process in Bilayer Honeycomb Spin Lattice, Materials Transactions, Vol. 64, No. 9, 2023, pp. 2118-2123, https://doi.org/10.2320/matertrans.MT-MG2022025.