

# Self consistent study of the quantum phases in a frustrated antiferromagnet on the bilayer honeycomb lattice.

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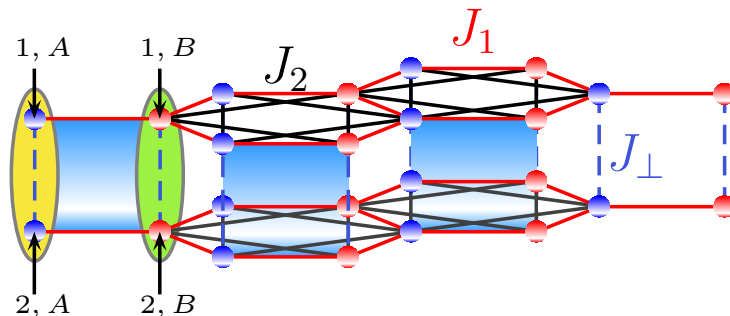
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**Abstract.** We study the frustrated Heisenberg model on the bilayer honeycomb lattice. The ground-state energy and spin gap are calculated, using different bosonic representations at mean field level and numerical calculations, to explore different sectors of the phase diagram. In particular we make use of a bond operator formalism and series expansion calculations to study the extent of dimer inter-layer phase. On the other hand we use the Schwinger boson method and exact diagonalization on small systems to analyze the evolution of on-layer phases. In this case we specifically observe a phase that presents a spin gap and short range Néel correlations that survives even in the presence of non-zero next-nearest-neighbor interaction and inter-layer coupling.

## 1. Introduction

The study of the possible disordered ground states on two-dimensional antiferromagnets has received a great interest in the last years. In particular, the existence of quantum disordered phases has been studied in the phase diagram of antiferromagnets in a single layer honeycomb lattice [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. However, the study of the influence of possible interlayer coupling on these phases is scarce [11, 12, 13, 14]. From the experimental side, a significative progress on the study of the bismuth oxynitrate,  $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$ , has been made by Smirnova *et al.*[15]. In this material the  $\text{Mn}^{4+}$  ions form a honeycomb lattice and two layers of such honeycomb lattices are separated by bismuth atoms, forming a bilayer structure. The study of the magnetic susceptibility indicates two-dimensional magnetism and no long-range ordering down to 0.4 K, suggesting a nonmagnetic ground state[15, 16]. In addition, density functional studies indicate that dominant interactions are the interlayer interaction  $J_{\perp}$  and the nearest-neighbor interaction  $J_1$  on each layer[17].





**Figure 1.** (Color online) Schematic representation of the coupling interactions in  $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$ . Colored areas correspond to the unit cells. The sites in each unit cell are labeled as  $(1, A)$ ,  $(2, A)$ ,  $(1, B)$  and  $(2, B)$ .

The aim of this paper is to study the zero temperature ground state of the frustrated spin-1/2 Heisenberg model on the bilayer honeycomb lattice. We study the  $S = 1/2$  case where the quantum fluctuations becomes more important in order to characterize the quantum phases in the model. On the other hand, although the material  $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$  has  $S = 3/2$ , the substitution of  $\text{Mn}^{4+}$  in  $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$  by  $\text{V}^{4+}$  may lead to the realization of the  $S = 1/2$  Heisenberg model on the honeycomb lattice. We use two different mean field self-consistent approaches based on bosonic representations of the spin operators to study this system, combined with Lanczos and series expansion methods to support the mean field results.

## 2. Self consistent calculations on the bilayer Model

We study the following Heisenberg model on the bilayer honeycomb lattice

$$H = \sum_{\vec{r}, \vec{r}', \alpha, \beta} J_{\alpha, \beta}(\vec{r}, \vec{r}') \vec{S}_{\alpha}(\vec{r}) \cdot \vec{S}_{\beta}(\vec{r}') \quad (1)$$

where,  $\vec{S}_{\alpha}(\vec{r})$  is the spin operator on site  $\alpha$  corresponding to the unit cell  $\vec{r}$ .  $\alpha$  takes the values  $\alpha = (1, A)$ ,  $(2, A)$ ,  $(1, B)$ ,  $(2, B)$  corresponding to the four sites on each unit cell as depicted in Fig. 1, together with the couplings  $J_{\alpha, \beta}(\vec{r}, \vec{r}')$ . The coupling  $J_{\perp}$  does not introduce frustration in the system and then, at the classical level and  $T = 0$ , it does not affect the classical Néel phase, present for  $J_2/J_1 < 1/6$ . In the quantum case the situation is much subtle, increasing  $J_{\perp}$  Néel order is likely to melt giving rise to a non-magnetic phase.

For large values of  $J_{\perp}$  we expect the ground state to be an interlayer valence bond crystal (IVBC) with corresponding spins from both layers forming dimers (as illustrated in Figure 1). This limit represents an excellent starting point for the bond operators formalism and series expansion calculations.

On the other hand, starting from the magnetically ordered phase, the Néel order can be destroyed both by increasing the frustration on each layer or increasing the coupling between layers. The destruction of Néel order in a frustrated single layer honeycomb lattice has been studied by means of various approaches [3, 4, 8, 11, 14, 19, 20, 21, 22]. In the following we use two different bosonic representations of the spin operators to study the influence of the interlayer coupling in the ground state of the bilayer model.

### 2.1. Bond operators Mean field approach.

First, we use the well known bond-operator method to study the bilayer antiferromagnet described by Hamiltonian (1). We start by introducing a bond-operator representation of spin

operators

$$\vec{\mathbf{S}}_{\eta,A}^{\alpha}(\vec{r}) = \frac{1}{2} \left( (-1)^{\eta+1} \left[ \mathbf{s}_A^{\dagger}(\vec{r}) \mathbf{a}_{\alpha}(\vec{r}) + \mathbf{a}_{\alpha}^{\dagger}(\vec{r}) \mathbf{s}_A(\vec{r}) \right] - i \epsilon_{\alpha\beta\gamma} \mathbf{a}_{\beta}^{\dagger}(\vec{r}) \mathbf{a}_{\gamma}(\vec{r}) \right) \quad (2)$$

$$\vec{\mathbf{S}}_{\eta,B}^{\alpha}(\vec{r}) = \frac{1}{2} \left( (-1)^{\eta+1} \left[ \mathbf{s}_B^{\dagger}(\vec{r}) \mathbf{b}_{\alpha}(\vec{r}) + \mathbf{b}_{\alpha}^{\dagger}(\vec{r}) \mathbf{s}_B(\vec{r}) \right] - i \epsilon_{\alpha\beta\gamma} \mathbf{b}_{\beta}^{\dagger}(\vec{r}) \mathbf{b}_{\gamma}(\vec{r}) \right), \quad (3)$$

where  $\eta = 1, 2$  is the layer index,  $\vec{\mathbf{S}}_{\eta,A}^{\alpha}(\vec{r})$  is the spin operator on the sublattice  $A$  of layer  $\eta$  corresponding to the unit cell located at  $\vec{r}$  (See figure 1). Operators  $\mathbf{s}_A^{\dagger}(\vec{r})$  and  $\mathbf{a}_{\alpha}^{\dagger}(\vec{r})$  create singlet and triplets states (out of a vacuum  $|0\rangle$ ) in the vertical bond placed in sub-lattice  $A$  and are defined as:  $\mathbf{s}_A^{\dagger}|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ ,  $\mathbf{a}_x^{\dagger}|0\rangle = -\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle)$ ,  $\mathbf{a}_y^{\dagger}|0\rangle = \frac{i}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle)$ ,  $\mathbf{a}_z^{\dagger}|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ , and similar expressions for sublattice  $B$ . These kind of representations were proposed by Sachdev[24] in order to treat quantum phase transitions between Néel and dimerized phases. Operators belonging to the same unit cell satisfy the bosonic commutation relations whereas operators belonging to different unit cells commute. The restriction that the physical states are either singlets or triplets leads to the constraints in each unit cell,  $\mathbf{s}_A^{\dagger} \mathbf{s}_A + \sum_{\alpha} \mathbf{a}_{\alpha}^{\dagger} \mathbf{a}_{\alpha} = 1$  and  $\mathbf{s}_B^{\dagger} \mathbf{s}_B + \sum_{\alpha} \mathbf{b}_{\alpha}^{\dagger} \mathbf{b}_{\alpha} = 1$ . Introducing the bond-operators representation of the spin operators in (1) we obtain a bosonic version of the Hamiltonian. We transform Fourier and retain terms up to second order to write  $\mathbf{H} = \mathbf{H}_{\perp} + \mathbf{H}_{\mathbf{1}} + \mathbf{H}_{\lambda}$ , where

$$\mathbf{H}_{\perp} = -\frac{3}{2} J_{\perp} s^2 N + \frac{J_{\perp}}{4} \sum_{\vec{k}, \alpha} \left\{ \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) \mathbf{a}_{\alpha}(\vec{k}) + \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) \mathbf{b}_{\alpha}(\vec{k}) \right\}, \quad (4)$$

$$\begin{aligned} \mathbf{H}_{\mathbf{1}} &= \frac{J_1}{4} \sum_{\vec{k}, \alpha} \left\{ \gamma(\vec{k}) \left( \mathbf{a}_{\alpha}(\vec{k}) \mathbf{b}_{\alpha}(-\vec{k}) + \mathbf{a}_{\alpha}(\vec{k}) \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) + \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) \mathbf{a}_{\alpha}(\vec{k}) + \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) \mathbf{a}_{\alpha}^{\dagger}(-\vec{k}) \right) \right. \\ &\quad \left. + \gamma(\vec{k}) \left( \mathbf{b}_{\alpha}(\vec{k}) \mathbf{a}_{\alpha}(-\vec{k}) + \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) \mathbf{b}_{\alpha}(\vec{k}) + \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) \mathbf{b}_{\alpha}^{\dagger}(-\vec{k}) + \mathbf{b}_{\alpha}(\vec{k}) \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) \right) \right\}, \end{aligned} \quad (5)$$

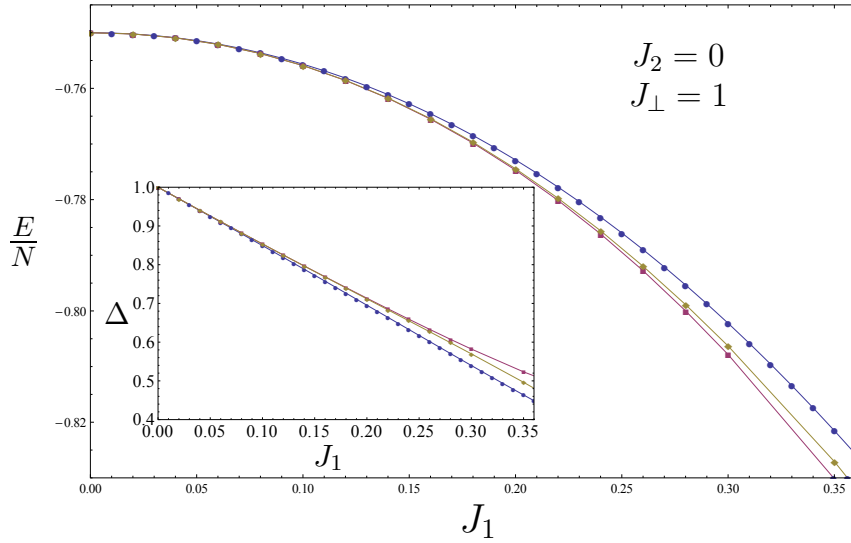
$$\mathbf{H}_{\lambda} = (2s^2 - 5)N\lambda + \lambda \sum_{\vec{k}, \alpha} \left\{ \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) \mathbf{a}_{\alpha}^{\dagger}(\vec{k}) + \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) \mathbf{b}_{\alpha}^{\dagger}(\vec{k}) \right\}, \quad (6)$$

where, we have assumed that condensation of singlets occurs, *i.e.*  $\langle \mathbf{s}_{\alpha}(\vec{r}) \rangle = s$ ,  $\gamma(\vec{k}) = s^2(1 + e^{i\vec{k} \cdot \vec{e}_1} + e^{i\vec{k} \cdot \vec{e}_2})$ ,  $\vec{e}_1$  and  $\vec{e}_2$  are the primitive vectors on a triangular lattice and  $\lambda$  is a Lagrange multiplier related to the constraint in the number of bosons. Diagonalization by using a Bogoliubov transformation allows us to write the following expression for the ground state energy

$$\frac{E}{N} = (2s^2 - 5)\lambda - \frac{3}{4} J_{\perp} (2s^2 + 1) + \sum_{\alpha} \int \frac{d^2k}{V} \left( \omega_{\alpha}^{(A)}(\vec{k}) + \omega_{\alpha}^{(B)}(\vec{k}) \right), \quad (7)$$

where  $\omega_{\alpha}^{(A)}(\vec{k})$  and  $\omega_{\alpha}^{(B)}(\vec{k})$  are triplet energies. The parameters  $s^2$  and  $\lambda$  are determined by solving self-consistently the saddle point conditions  $\frac{\partial E}{\partial \lambda} = 0$  and  $\frac{\partial E}{\partial s^2} = 0$  which are used to evaluate the energy and gap of the system and compare with numerical techniques.

In order to complement our study, we have performed series expansion (SE) calculations, starting from the limit of isolated dimers connecting spins from both layers via  $J_{\perp}$ . Notice that, this kind of expansions remain valid in the same limit that the bond operator approach (*i.e.* in the limit of strong inter-layer coupling.) To this end we have performed a continuous unitary transformation on the original Hamiltonian, using the flow equation method. This technique allows to obtain perturbatively an effective Hamiltonian that keeps the block diagonal structure of decoupled dimers. We refer for details of the method to ref.[25].

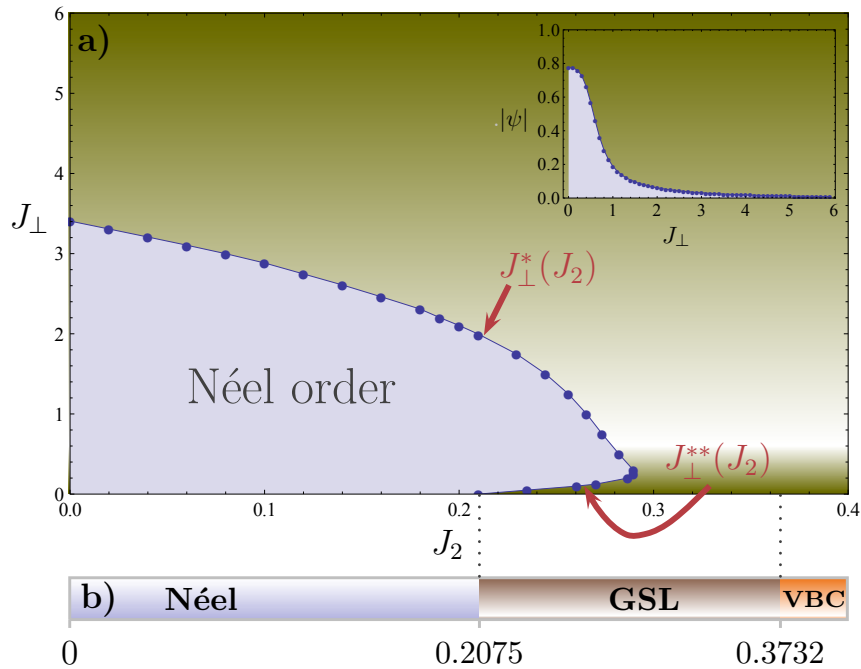


**Figure 2.** (Color online) Ground state energy per dimer as a function of  $J_1$  obtained by the self consistent bond-operator approach (blue circles), Lanczos (ED) on a 24 sites system (red squares) and  $O(5)$  Series Expansion (SE) (yellow rhombi). Inset: triplet gap (same set of parameters as main panel) BO (blue circles) ED (red squares) and SE (yellow rhombi).

For the present model we have performed  $O(5)$  and  $O(4)$  SE in  $J_{1,2}$  for ground state energy and for triplet dispersion, respectively. Explicit expressions are too long to be printed explicitly but are available electronically upon request. In Fig.2 we show the ground state energy per site as a function of  $J_1$  and  $J_2 = 0$ , obtained by BO (blue circles),  $O(5)$  SE (yellow rhombi) and ED (red squares) on a system of 24 sites. As it can be observed, all the techniques predict an energy decreasing with the coupling of interlayer-dimer via  $J_1$ . Furthermore, there is an excellent quantitative agreement between the three methods for small values of  $J_1$ . On the other hand, triplet gap is shown in the inset of Fig.2 for the same set of parameters as the ground state energy. Here we also observe that all the techniques predict a tendency to a closure of the gap, when  $J_1$  is turned on. Our calculations shows that BO, ED and SE predict the same behavior.

## 2.2. Self consistent Schwinger Boson Mean-Field Theory

As we have seen previously, bond operator and series expansion methods are both suitable to study the interlayer-dimer phase. In order to investigate the evolution of on-layer phases as a function of inter-layer coupling we apply a representation of the spin operators in terms of Schwinger bosons [27],  $\vec{S}_\alpha(\vec{r}) = \frac{1}{2}\vec{b}_\alpha^\dagger(\vec{r}) \cdot \vec{\sigma} \cdot \vec{b}_\alpha(\vec{r})$ . Here  $\vec{b}_\alpha(\vec{r})^\dagger = (\mathbf{b}_{\alpha,\uparrow}^\dagger(\vec{r}), \mathbf{b}_{\alpha,\downarrow}^\dagger(\vec{r}))$  is a bosonic spinor corresponding to the site  $\alpha$  in the unit cell at position  $\vec{r}$ ,  $\vec{\sigma}$  are Pauli matrices, and the constraint in the number of bosons  $\sum_\sigma \mathbf{b}_{\alpha,\sigma}^\dagger(\vec{r})\mathbf{b}_{\alpha,\sigma}(\vec{r}) = 2S$  has to be satisfied on each site. In order to perform a mean field decomposition, we define the following  $SU(2)$  invariants,  $\mathbf{A}_{\alpha\beta}(\vec{x}, \vec{y}) = \frac{1}{2} \sum_\sigma \sigma \mathbf{b}_{\alpha,\sigma}(\vec{x})\mathbf{b}_{\beta,-\sigma}(\vec{y})$  and  $\mathbf{B}_{\alpha\beta}(\vec{x}, \vec{y}) = \frac{1}{2} \sum_\sigma \mathbf{b}_{\alpha,\sigma}^\dagger(\vec{x})\mathbf{b}_{\beta,-\sigma}(\vec{y})$ . This decomposition allow us to treat ferromagnetism and antiferromagnetism on equal footing and has been successfully used to describe a number of quantum frustrated antiferromagnets[1, 2, 3, 14, 28, 29, 30]. We perform a Hartree-Fock decoupling where the mean field parameters are the expectation values of the  $SU(2)$  invariants  $\mathbf{A}$  and  $\mathbf{B}$ . The mean field equations for these parameters  $A_{\alpha\beta}$  and  $B_{\alpha\beta}$  and the constraints in the number of bosons must be solved self-consistently (see refs. [1], [3] and [14] for further details). To obtain the phase boundary between the magnetically ordered and disordered phases using the self consistent Schwinger



**Figure 3.** (Color online) a) Phase diagram in the  $J_2 - J_\perp$  plane obtained with SBMFT. Gray region corresponds to the Néel phase whereas green region corresponds to magnetically disordered phases. b) Phase diagram of the single layer case corresponding to Ref. [3]. Inset:  $Z_3$  order parameter corresponding to the line  $J_2 = 0.38$

boson mean field theory we study the boson spectrum. In the gapless region the excitation spectrum is zero at  $\vec{k} = \vec{0}$ , where the boson condensation occurs, this is characteristic of the Néel ordered phase. On the other hand, in the gapped region, the absence of Bose condensation indicates that the ground state is magnetically disordered.

In Fig. 3-a) we show the phase diagram in the  $J_2 - J_\perp$  plane. For  $J_\perp \gg J_2$  one can expect a IVBC ground state adiabatically connected with the limit of decoupled dimers, i.e. two singlets per unit cell, between spins 1,  $A(1, B)$  and 2,  $A(2, B)$  (see Fig. 1). In the region  $0.2075 \lesssim J_2 \lesssim 0.289$  there is a reentrant effect. In this range, Néel phase separates from  $J_2$  axis, leaving a tiny space for a magnetically disordered phase. In this way, Néel phase is here not only limited by some value  $J_\perp^*(J_2)$  from above, but also by a second value  $J_\perp^{**}(J_2)$  from below (See Figure 3).

On the other hand, in the range  $0.3732 \lesssim J_2 \lesssim 0.398$  ( $J_\perp = 0$ ), there is evidence of the existence of an on-layer valence bond phase [3] (see Figure 3-b). In this phase,  $SU(2)$  and translational symmetries are preserved, but  $Z_3$  symmetry is broken. By turning on  $J_\perp$  the system moves to the IVBC where the  $Z_3$  symmetry is recovered. In the inset of Figure 3-a, we depict the  $Z_3$  directional symmetry-breaking order parameter  $\rho$  (defined in [18]) vs  $J_\perp$ . The behavior of this parameter suggest a  $Z_3$  symmetry restoring. Finally, in the region  $0.289 \lesssim J_2/J_1 \lesssim 0.3732$  the ground state preserves  $SU(2)$ , lattice translational and  $Z_3$  symmetries and the spin-spin correlations are short ranged[14]. This agrees with the evidence of a spin liquid phase in the phase diagram corresponding to  $J_\perp = 0$  [3, 8].

### 3. Conclusions

In summary, we have studied the phase diagram corresponding to a frustrated Heisenberg model on the bilayer honeycomb lattice, by means of bosonic mean field theories, complemented with exact diagonalization and series expansion, and described the behavior of the quantum phases as the interlayer coupling is increased. Using the Schwinger boson description we have determined the region where the system is Néel ordered and the lines where the Néel order is destroyed. We have determined an intermediate region where the phase diagram shows signatures of a reentrant behavior and we observe that for values of the interlayer coupling between  $(0.289 \lesssim J_2/J_1 \lesssim 0.3732)$  the Néel order is absent at  $J_\perp = 0$  and the system presents a nonzero spin gap, whereas in the region  $(0.3732 \lesssim J_2/J_1 \lesssim 0.398)$  each layer presents a nematic disordered phase[3]. In all the range of  $J_2$  studied, the system presents signatures of an interlayer-valence bond crystal (IVBC) phase for  $J_\perp/J_1 > 4$ . This phase evolves adiabatically from the limit of decoupled interlayer-dimers. This is corroborated by bond operators self-consistent calculations and series expansions starting explicitly from the limit of isolated interlayer dimers.

### ACKNOWLEDGMENTS

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