Evidence of a spin liquid phase in the frustrated honeycomb lattice

D. C. Cabra,1, 2 C. A. Lamas,1 and H. D. Rosales1

1Instituto de Física de La Plata and Departamento de Física,
Universidad Nacional de La Plata, C.C. 67, 1900 La Plata, Argentina
2Facultad de Ingeniería, Universidad Nacional de Lomas de Zamora,
Cno. de Cintura y Juan XXIII, (1832) Lomas de Zamora, Argentina.

In the present paper we present some new data supporting the existence of a spin-disordered phase in the Heisenberg model on the honeycomb lattice with antiferromagnetic interactions up to third neighbors along the line \( J_2 = J_3 \), predicted in1. We use the Schwinger boson technique followed by a mean field decoupling and exact diagonalization for small systems to show the existence of an intermediate phase with a spin gap and short range Néel correlations in the strong quantum limit \((S = \frac{1}{2})\).

PACS numbers:

1. INTRODUCTION

Geometrical frustration in two-dimensional (2D) antiferromagnets is expected to enhance the effect of quantum spin fluctuations and hence suppress magnetic order giving rise to a spin liquid2, and this idea has motivated many researchers to look for its realization2, 3.

One candidate to test these ideas is the honeycomb lattice, which is bipartite and has a classical Néel ground state, but due to the small coordination number \((z = 3)\), quantum fluctuations could be expected to be stronger than those in the square lattice and may destroy the antiferromagnetic long-range order (LRO)2, 12.

The study of frustrated quantum magnets on the honeycomb lattice has also experimental motivations3–7.

The analysis of the hexagonal lattice from a more general point of view has gained lately a lot of interest both coming from graphene-related issues and from the possible spin-liquid phase found in the Hubbard model in such geometry8–14.

Motivated by the previous results, in this paper we show the study of the Heisenberg model on the honeycomb lattice with first \((J_1)\), second \((J_2)\) and third \((J_3)\) neighbors couplings15, along the special line \( J_2 = J_3 \). Using Schwinger boson mean field theory (SBMFT) and exact diagonalization we find strong evidence for the existence of an intermediate disordered region where a spin gap opens and spin-spin correlations decay exponentially. Using exact diagonalization of small clusters we also have calculated the dimer-dimer correlation function that indicates short range dimer-dimer order. Although our results correspond to a specific line, we conjecture that the quantum disordered phase that we have found in the vicinity of the tricritical point extends within a finite region around it. Previous evidence of massive behaviour in the hexagonal lattice Heisenberg model has been found in other regions of the phase space by means of exact diagonalization in ref. 18.

The Heisenberg model on the \( J_1 - J_2 - J_3 \) honeycomb lattice is given by

\[
H = J_1 \sum_{(xy)_1} \hat{S}_x \cdot \hat{S}_y + J_2 \sum_{(xy)_2} \hat{S}_x \cdot \hat{S}_y + J_3 \sum_{(xy)_3} \hat{S}_x \cdot \hat{S}_y, \tag{1}
\]

where \( \hat{S}_x \) is the spin operator on site \( x \) and \( \langle xy \rangle_n \) indicates sum over the \( n \)-th neighbors (see Fig. 1). In the classical limit, \( S \to \infty \), the model displays different zero temperature phases with a tricritical point at \( J_2 = J_3 = \frac{1}{2} J_1 \). At this particular point the classical ground state has a large GS degeneracy18, 19. The Heisenberg model on the honeycomb lattice was studied using SBMFT by Mattsson et al.20 for antiferromagnetic interactions at first and second neighbors. Here we study the Hamiltonian (1) using a rotationally invariant version of this technique, which has proven successful in incorporating quantum fluctuations20, 21.

2. SCHWINGER Bosons MEAN-FIELD THEORY.

In this section we describe in detail the Schwinger boson mean field theory used in the present work. The
SU(2) Heisenberg Hamiltonian on a general lattice can be written as

\[ \hat{H} = \frac{1}{2} \sum_{x,y,\alpha,\beta} J_{\alpha\beta}(x - y) \hat{S}_x \cdot \hat{S}_y, \]  

(2)

where \( x \) and \( y \) are the positions of the unit cells and vectors \( r_\alpha \) correspond to the positions of each atom within the unit cell. \( J_{\alpha\beta}(x - y) \) is the exchange interaction between the spins located in \( x + r_\alpha \in y + r_\beta \).

In what follows we assume that the classical order can be parameterized as

\[ \hat{S}_x = S \sin \varphi_\alpha(x) \]  

(3)

\[ \hat{S}_y = 0 \]  

(4)

\[ \hat{S}_z = S \cos \varphi_\alpha(x), \]  

(5)

with \( \varphi_\alpha(x) = Q \cdot x + \theta_\alpha \), where \( Q \) is the ordering vector and \( \theta_\alpha \) are the relative angles between the classical spins inside each unit cell.

The spin operators \( \hat{S}_x \) on site \( x \) are represented by two bosons \( \hat{b}_{x\sigma} \) \((\sigma = \uparrow, \downarrow)\)

\[ \hat{S}_x = \frac{1}{2} \hat{b}_{x\uparrow} \cdot \hat{b}_{x\downarrow} \]  

(6)

where \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) are the Pauli matrices. This is a faithful representation of the algebra SU(2) if we take into account the following local constraint

\[ 2S = \hat{b}_{x\uparrow} \cdot \hat{b}_{x\downarrow} + \hat{b}_{x\downarrow} \cdot \hat{b}_{x\uparrow}. \]  

(7)

In this representation, the exchange term can be expressed as

\[ \hat{S}_{x+r_\alpha} \cdot \hat{S}_{y+r_\beta} : = \hat{B}_{\alpha\beta}(x, y) \hat{A}_{\alpha\beta}(x, y) : - \hat{A}_{\alpha\beta}^\dagger(x, y) \hat{A}_{\alpha\beta}(x, y) \] 

where \( \hat{A}_{\alpha\beta}(x, y) \) and \( \hat{B}_{\alpha\beta}(x, y) \) are the SU(2) invariants defined as

\[ \hat{A}_{\alpha\beta}(x, y) = \frac{1}{2} \sum_\sigma \sigma \hat{b}_{x,\sigma}^{(\alpha)} \hat{b}_{y,\sigma}^{(\beta)} \]  

(9)

\[ \hat{B}_{\alpha\beta}(x, y) = \frac{1}{2} \sum_\sigma \hat{b}_{x,\sigma}^{(\alpha)} \hat{b}_{y,\sigma}^{(\beta)} \]  

(10)

with \( \sigma = \uparrow, \downarrow \) and double dots \( \langle \cdot \hat{O} \cdot \rangle \) indicate normal ordering of operator \( \hat{O} \). This decoupling is particularly useful to the description of magnetic systems near disordered states, because it allows to treat antiferromagnetism and ferromagnetism in equal footing.

To construct a mean field theory we perform a Hartree-Fock decoupling

\[ \langle \hat{S}_{x+r_\alpha} \cdot \hat{S}_{y+r_\beta} \rangle_{MF} = \langle B_{\alpha\beta}(x - y) \hat{A}_{\alpha\beta}(x, y) - A_{\alpha\beta}^\dagger(x - y) \hat{A}_{\alpha\beta}(x, y) + H.c \rangle \]  

(11)

with

\[ A_{\alpha\beta}^\dagger(x - y) = \langle \hat{A}_{\alpha\beta}^\dagger(x, y) \rangle \]  

(12)

\[ B_{\alpha\beta}^\dagger(x - y) = \langle \hat{B}_{\alpha\beta}^\dagger(x, y) \rangle \]  

(13)

and where \( \langle \cdot \rangle \) denotes the expectation value in the ground state at \( T = 0 \). Because several functions involved in the Hamiltonian depend on the difference \( x - y \) we change variables to \( R = x - y \) and eliminating \( x \) in the sums we obtain

\[ H_{MF} = \frac{1}{2} \sum R_{y,\alpha\beta} J_{\alpha\beta}(R) \left\{ \frac{1}{2} \sum_\sigma \left[ B_{\alpha\beta}(R) \hat{b}_{x,\sigma}^{(\alpha)} \hat{b}_{y,\sigma}^{(\beta)} - \sigma A_{\alpha\beta}(R) \hat{b}_{x,\sigma}^{(\alpha)} \hat{b}_{y,\sigma}^{(\beta)} + H.c. \right] + \langle B_{\alpha\beta}(R) \rangle^2 - \langle A_{\alpha\beta}(R) \rangle^2 \right\}. \]  

(14)

The mean field Hamiltonian is quadratic in the boson operators and can be diagonalized in real space. However, as we look for translational invariant solutions, it is convenient to transform the operators to momentum space

\[ \hat{b}_{x,\sigma}^{(\alpha)} = \frac{1}{\sqrt{N_c}} \sum_k \hat{b}_{x,\sigma}^{(\alpha)} e^{-ik \cdot (x + r_\alpha)}. \]  

(15)

After some algebra and using the symmetry properties:

\[ J_{\alpha\beta}(R) = J_{\beta\alpha}(-R) \]

\[ A_{\alpha\beta}(R) = -A_{\beta\alpha}(-R) \]  

(16)

\[ B_{\alpha\beta}(R) = B_{\beta\alpha}^*(-R), \]

we obtain the following form for the Hamiltonian
\[ H_{MF} = \frac{1}{2} \sum_{k_{\alpha\beta}} \left\{ \gamma_{\alpha\beta}^B(k) \hat{b}^\dagger_{k_{\alpha\beta}} \hat{b}^\dagger_{k_{-\alpha\beta}} + \gamma_{\alpha\beta}^B(-k) \hat{b}^\dagger_{-k_{-\alpha\beta}} \hat{b}^\dagger_{-k_{\alpha\beta}} - \sigma \gamma_{\alpha\beta}^A(k) \hat{b}^\dagger_{k_{\alpha\beta}} \hat{b}^\dagger_{-k_{\alpha\beta}} - \sigma \gamma_{\alpha\beta}^A(-k) \hat{b}^\dagger_{-k_{\alpha\beta}} \hat{b}^\dagger_{k_{-\alpha\beta}} \right\} \\
- \frac{N_c}{2} \sum_{R_{\alpha\beta}} [B_{\alpha\beta}(R)]^2 - |A_{\alpha\beta}(R)|^2 , \]  
where 
\[ \gamma_{\alpha\beta}^B(k) = \frac{1}{2} \sum_R J_{\alpha\beta}(R) B_{\alpha\beta}(R) e^{-ik(R + r_\sigma - r_\beta)} \]  
\[ \gamma_{\alpha\beta}^A(k) = \frac{1}{2} \sum_R J_{\alpha\beta}(R) A_{\alpha\beta}(R) e^{-ik(R + r_\sigma - r_\beta)} \]  
\[ \gamma_{\alpha\beta}^A(-k) = \frac{1}{2} \sum_R J_{\alpha\beta}(R) A_{\alpha\beta}(R) e^{-ik(R - r_\sigma + r_\beta)} \] 

Now, we impose the constraint on average over each sublattice \( \alpha \) by means of Lagrange multipliers \( \lambda^{(\alpha)} \)

\[ H_{MF} \to H_{MF} + \hat{H}_\lambda \]  
(21)

It is convenient to define a vector operator \( \hat{b}^\dagger(k) = (\hat{b}^\dagger_{k_{\alpha\beta}}, \hat{b}^\dagger_{-k_{\alpha\beta}}) \) where 
\[ \hat{b}^\dagger_{k_{\alpha\beta}} = (\hat{b}^\dagger_{k_{\alpha\beta}}, \hat{b}^\dagger_{k_{-\alpha\beta}}, \cdots, \hat{b}^\dagger_{k_{n_{\alpha\beta}}}) \]  
\[ \hat{b}^\dagger_{-k_{\alpha\beta}} = (\hat{b}^\dagger_{-k_{\alpha\beta}}, \hat{b}^\dagger_{-k_{-\alpha\beta}}, \cdots, \hat{b}^\dagger_{-k_{n_{\alpha\beta}}}) \] 
and \( n_c \) is the number of atoms in the unit cell. Now, we can write the Hamiltonian as

\[ H_{MF} = \sum_k \hat{b}^\dagger(k) \cdot D(k) \cdot \hat{b}(k) \]  
(25)

where the 2\( n_c \) x 2\( n_c \) dynamical matrix \( D(k) \) is given by

\[ D(k) = \begin{pmatrix} \gamma_{\alpha\beta}^B(k) + \lambda^{(\alpha)} \delta_{\alpha\beta} & -\gamma_{\alpha\beta}^B(k) + \lambda^{(\alpha)} \delta_{\alpha\beta} \\ \gamma_{\alpha\beta}^B(k) + \lambda^{(\alpha)} \delta_{\alpha\beta} & \gamma_{\alpha\beta}^B(k) + \lambda^{(\alpha)} \delta_{\alpha\beta} \end{pmatrix} . \]

To diagonalize the Hamiltonian we need to perform a para-unitary transformation of the matrix \( D(k) \) that preserves the bosonic commutation relations. We can diagonalize the Hamiltonian by defining the new operators \( \hat{a} = F \cdot \hat{b} \), where the matrix \( F \) satisfy

\[ (F^\dagger)^{-1} \cdot \tau_3 \cdot (F)^{-1} = \tau_3, \quad \tau_3 = \begin{pmatrix} I_{2\times2} & 0 \\ 0 & -I_{2\times2} \end{pmatrix} . \]

With this transformation, the Hamiltonian reads

\[ H_{MF} = \sum_k \hat{a}^\dagger(k) \cdot E(k) \cdot \hat{a}(k) - 2(S + 1)N_c \sum_\alpha \lambda^{(\alpha)} - \langle H_{MF} \rangle , \]  
(27)
Spin correlation function (SSCF) vs distance for several values of the frustration parameter. We numerically solve for finite but very large lattices and finally we extrapolate the results of this self-consistent procedure until the energy and the Lagrange multipliers we compute the energy and the Lagrange multipliers. We perform the calculations for finite systems, then to detect LRO we calculate the gap in the bosonic spectrum as a function of the correlations and the excitation gap. In order to support the analytical results of the MF approach, we have also performed exact diagonalization on finite systems with 18, 24 and 32 spins with periodic boundary conditions for $S = 1/2$ using Spinpack.

3. RESULTS

In Fig. 2(a) we show the ground state energy per unit cell as a function of the frustration for a system of 32 sites calculated by means of SBMFT and ED, showing an excellent agreement between both approaches. The advantage of the SBMFT is that it allows to study much larger systems: we have studied different system sizes up to 3200 sites and extrapolated to the thermodynamic limit.

For the present model we only find commensurate collinear phases and for these phases, the wave vector $Q_0 = Q/2$ where the dispersion relation has a minimum remains pinned at a commensurate point in the Brillouin zone, independently of the value of the frustration $J_2/J_1$. In the thermodynamic limit, a state with LRO is characterized in the Schwinger boson approach by a condensation of bosons at the wave vector $Q_0$. This implies that the dispersion of the bosons in a state with LRO is gapless. As we discussed earlier, we solve the mean field equations for finite systems, then to detect LRO we calculate the gap in the bosonic spectrum as a function of $J_2/J_1$ for different system sizes and perform a finite size scaling finding a finite region where the system remains gapped.

The structure of the different phases can be understood calculating the spin-spin correlation function (SSCF). For $J_2/J_1 < 0.41$ the SSCP is antiferromagnetic in all directions and is long-ranged while for $0.6 < J_2/J_1$. 

FIG. 2: (a): GS energy per unit cell for $S = 1/2$ as a function of $J_2/J_1$ for a lattice of 32 sites. The circles are exact results (ED) and the squares are the SBMFT results. (b): Spin-Spin correlation function (SSCF) vs distance $X$ in the zig-zag direction obtained within SBMFT for a $32 \times 32$ system in $\mathbb{Z}^2$. For $0 < J_2/J_1 < 0.41$, the SSCP correspond to the Néel phase with long-range-order (LRO), for $0.41 < J_2/J_1 < 0.6$ the correlations are short ranged indicating a gap zone with short-range-order (SRO), and for $0.6 < J_2/J_1$ the correlations correspond to the collinear phase (ferromagnetic correlations in the zig-zag direction). The inset in Fig. (b) shows the finite size results for the SSCP obtained by ED and SBMFT for 32 sites. Lines are guides to the eye and different colors are used for clarity.

FIG. 3: Gap in the boson dispersion as a function of $J_2/J_1$ for $S = 1/2$ from Ref. (i) $J_2/J_1 = 0.5$ ($\gamma = 0.6451$), (ii): Circles correspond to $J_2/J_1 = 0.55$ ($\gamma = 0.911$) and squares correspond to $J_2/J_1 = 0.35$ ($\gamma = 0.758$).
indicates the value of $J$ shows Néel short range order. A plot of the SSCF for $J_2/J_1 = 0$, 0.2, 0.5 and 0.8 obtained within SBMFT is presented in Fig. 2. In Fig. 3 we show the ground state phase diagram as a function of $1/S_c$. The classical phase diagram reduces to that shown in the line $1/S_c = 0$ of Fig. 4 where two collinear phases meet at the classical critical point $J_2/J_1 = 0.5$. On the one hand, for $1/S$ smaller than a critical value $1/S_c(J_2/J_1)$, the correlation functions have LRO, characterized by a condensation of bosons at the wave vector $Q_0$. On the other hand, when $1/S$ is greater than $1/S_c(J_2/J_1)$, the correlation functions have SRO indicating quantum disorder.

In the intermediate region the results found with SBMFT predict a quantum disordered region $0.41 < J_2/J_1 < 0.6$. In this region a gap opens in the bosonic dispersion and the spin-spin correlation function shows Néel short range order followed by the LRO CAF phase for $J_2/J_1 > 0.6$. In Fig. 4 we show the extrapolation of the boson gap as a function of the frustration. The inset shows an example of the finite size scaling for different values of the frustration.

Previous results show that for $0.41 < J_2/J_1 < 0.6$ the ground state has no magnetic order. The main question now is: Is this non magnetic quantum phase a quantum disordered one? Or does it exhibit any other kind of non-magnetic order? To answer this question the knowledge of the spin-spin correlation function is not enough and one has to check for other types of (non-magnetic) ordering patterns.

One kind of non magnetic order that can set in is the dimer long-range order. The dimer operator on a pair of sites $(i,j)$ is defined as $\hat{d}_{i,j} = 1/4 - \mathbf{S}_i \cdot \mathbf{S}_j$, and one usually defines the dimer-dimer correlation between bonds $(i,j)$ and $(k,l)$ as $D_{(i,j),(k,l)} = <\hat{d}_{i,j}\hat{d}_{k,l}> - <\hat{d}_{i,j}> <\hat{d}_{k,l}>$. In order to understand the nature of the ground state in the intermediate region, we have calculated dimer-dimer correlation function defined above by means of exact diagonalization on a 24 sites cluster with periodic boundary conditions for $S = 1/2$. The correlation pattern for dimers on first neighbor bonds is displayed in Fig. 5. We can see that the exact dimer-dimer correlations show a rather fast decay suggesting that there is no dimer order in the ground state, though due to the small size of the cluster studied, this is not conclusive and we cannot discard other ordering patterns.

In summary, the results presented here further support the existence of a region in the intermediate frustration regime where the system does not show quantum magnetic order for $S = 1/2$.

**Note added:** When this manuscript was completed we became aware of two independent works providing an analysis of the model using a combination of exact diagonalizations and an effective quantum dimer model, as well as a self-consistent cluster mean-field theory. Several similar findings show a good correspondence of both approaches.

**Acknowledgements:** This work was partially supported by the ESF grant INSTANS, PICT ANPCYT (Grant No 2008-1426) and PIP CONICET (Grant No 1691).
1. D.C. Cabra, C.A. Lamas, H.D. Rosales, Phys. Rev. B 83, 094506 (2011).
2. P. W. Anderson, Science 235, 1196 (1987).
3. M. Metlitski, S. Sachdev, Phys. Rev. B 77, 054411 (2008).
4. R. K. Kaul, M. A. Metlitski, S. Sachdev, C. Xu, Phys. Rev. B 78, 045110 (2008).
5. L. Wang, A. W. Sandvik, Phys. Rev. B 81, 054417 (2010).
6. R. Moessner, S.L. Sondhi, P. Chandra, Phys. Rev. B 64, 144416 (2001).
7. A. Ralko, M. Mambrini, D. Poilblanc, Phys. Rev. B 80, 184427 (2009).
8. A. Mattsson, P. Frojd, T. Einarsson, Phys. Rev. B 49, 3997 (1994).
9. K. Takano Phys. Rev. B 74, 140402 (2006); M. Hermele, Phys. Rev. B 76, 035125 (2007); R. Kumar, D. Kumar, B. Kumar Phys. Rev. B 80, 214428 (2009).
10. S. Okubo et al, J. Phys.: Conf. Series 200, 022042 (2010).
11. Magnetic Properties of Layered Transition Metal Compounds, Ed. L. J. De Jongh, Kluwer, Dordrecht (1990).
12. A. Moller et al, Phys. Rev. B 78, 024420 (2008).
13. A.A. Tsirlin, O. Janson, H. Rosner, Phys. Rev. B 82, 144416 (2010).
14. M. Matsuda, M. Azuma, M. Tokunaga, Y. Shimakawa, N. Kumada Phys. Rev. Lett. 105, 187201 (2010).
15. Z.Y. Meng, T.C. Lang, S. Wessel, F.F. Assaad, A. Muramatsu, Nature 464, 847 (2010).
16. S. Okumura, H. Kawamura, T. Okubo, Y. Motome, J. Phys. Soc. Jpn. 79, 114705 (2010); F. Wang, Phys. Rev. B 82, 024419 (2010); A. Mulder, R. Ganesh, L. Capriotti, A. Paramekanti, Phys. Rev. B 81, 214419 (2010); Y.-M. Lu, Y. Ran, Preprint arXiv:1007.3266.
17. R. Ganesh, D.N. Sheng, Y.-J. Kim, A. Paramekanti, preprint arXiv:1012.0316; M.-T. Tran, K.-S. Kim, preprint arXiv:1011.1700; H.C. Kandpal, J. van den Brink, preprint arXiv:1102.3330; A. Banerjee, K. Damle, A. Paramekanti, preprint arXiv:1012.4546; B.K. Clark, D.A. Abanin, S.L. Sondhi preprint arXiv:1010.3011; H. Mosadeq, F. Shahbazi, S.A. Jafari, preprint arXiv:1007.0127; H. Wadati et al, preprint arXiv:1101.2847.
18. J. B. Fouet, P. Sindzingre, C. Lhuilli, Eur. Phys. J. B 20, 241 (2001).
19. E. Rastelli, A. Tassi, L. Reatto, Physica 97B, 1 (1979).
20. H. A. Ceccato, C. J. Gazza, A. E. Trumper, Phys. Rev. B 47, 12329 (1993).
21. R. Flint, P. Coleman, Phys. Rev. B 79, 014424 (2009).
22. A. E. Trumper, L. O. Manuel, C. J. Gazza, H. A. Ceccato, Phys. Rev. Lett. 78, 2216 (1997).
23. J. Schulenburg, program package SPINPACK, http://www-e.uni-magdeburg.de/jschulen/spin/.
24. A.F. Albuquerque, D. Schwandt, B. Hetényi, S. Capponi, M. Mambrini, A.M. Lauchli, arXiv:1102.5325 (2011)
25. D. J. J. Furnell, R. F. Bishop, P. H. Y. Li, J. Richter, C. E. Campbell, arXiv:1103.3856 (2011)