Nonlinear σ Model Method for the J_1-J_2 Heisenberg Model: Disordered Ground State with Plaquette Symmetry

Ken'ichi Takano,[1] Yoshiya Kito,[2] Yoshiaki Ōno,[2,*] and Kazuhiro Sano[3]
[1]*Toyota Technological Institute, Tenpaku-ku, Nagoya 468-8511, Japan*
[2]*Department of Physics, Nagoya University, Chikusa-ku, Nagoya 464-8602, Japan*
[3]*Department of Physics Engineering, Mie University, Tsu, Mie 514-8507, Japan*
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A novel nonlinear σ model method is proposed for the two-dimensional J_1-J_2 model, which is extended to include plaquette-type distortion. The nonlinear σ model is properly derived without spoiling the original spin degrees of freedom. The method shows that a single disordered phase continuously extends from a frustrated uniform regime to an unfrustrated distorted regime. By the continuity and Oshikawa's commensurability condition, the disordered ground states for the uniform J_1-J_2 model are plaquette states with fourfold degeneracy.

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The two-dimensional (2D) J_1-J_2 model is a frustrated Heisenberg model with nearest neighbor (NN) and next nearest neighbor (NNN) antiferromagnetic exchange interactions on a square lattice. The model with spin magnitude $S = \frac{1}{2}$ is realized in mother materials of cuprate superconductors, La_2CuO_4, YBa_2CuO_6, and $Sr_2CuO_2Cl_2$ as small-J_2 systems [1,2]. Recently found materials, Li_2VOSiO_4 and Li_2VOGeO_4, are also described by the model in the case of $J_2/J_1 \sim 1$ [3,4]. A particular interest for the J_1-J_2 model is in a gapful disordered state, which may be formed by frustration under strong quantum fluctuations [5]. The subject has been theoretically investigated by various methods [1]: e.g., spin-wave theories [6–8], nonlinear σ model (NLSM) methods [9,10], numerical diagonalizations [11–14], quantum Monte Carlo (QMC) simulations [15–17], series expansions [18–20], and variational methods [21].

For a system only with the NN interactions ($J_2 = 0$), the ground state is believed to have an antiferromagnetic (AF) order. The NNN exchange interactions are expected to induce strong frustration to break the AF order and to form a disordered ground state around $J_2/J_1 = 0.5$. A current leading QMC calculation [15,16] supports the disordered phase with spin gap for $J_2/J_1 \gtrsim 0.4$. Accepting this result, the issue is the character of the ground state in the disordered phase. Candidates examined in recent years are a uniform resonating-valence-bond (RVB) state [21], a plaquette state [16,18], a dimer state [19–21], and a state with both dimer and plaquette structures [17]; their degeneracies are 1, 4, 4, and 8, respectively. Although Oshikawa's commensurability condition [22] is useful to restrict possibilities, it does not completely select one; e.g., it requires that a uniform RVB ground state with spin gap is accompanied with gapless singlet excitations. The character of the ground state is still under debate.

A disordered state is formed also by distortion in the exchange constants, even if there is no frustration ($J_2 = 0$). For a plaquette-type distortion, a disordered state interpreted as a 2D array of plaquette singlets is formed [23]. Here it is a question whether the disordered state by frustration is essentially the same as that by plaquette-type distortion. If it is the same, a disordered phase continuously extends from a regime of strong frustration and weak distortion to a regime of weak frustration and strong distortion in a parameter space. However, if not, there exists a phase boundary between them; then the ground state of the uniform J_1-J_2 model is not plaquettelike. Hereafter we consider the J_1-J_2 model which is extended to include a plaquette-type distortion.

Among various methods to analyze spin systems, an NLSM method is effective to clarify their characters. The first successful example appeared in one dimension. A uniform spin chain with NN interactions is mapped onto an NLSM with an appropriate topological term [24]. Inhomogeneous spin chains with periodicity are treated by refined and extended NLSM methods [25,26]. For 2D systems, an NLSM without topological term is derived for $J_2 = 0$ [27]. For $J_2 \neq 0$, Chakravarty *et al.* [9] analyzed 2D NLSM which represents the uniform J_1-J_2 model. By applying a renormalization group (RG) method to the NLSM, they constructed a standard theory for the quantum phase transition.

Despite the success, there remains ambiguity in the correspondence of a derived NLSM to the J_1-J_2 model. If one uses a naive mapping in literature, a single spin variable is replaced by the sum of two new variables representing a slowly varying AF motion and a rapid fluctuation. This is not justified because the number of independent variables is abruptly increased. Although the mapping may phenomenologically produce the correct NLSM, there is no way to confirm the correctness within the NLSM method itself. Further the increase of the degrees of freedom leaves ambiguity for the choice of the cutoff. In one dimension, the problem of the degrees of freedom has been overcome in generalized formulations [25,26]. However such a reasonable theory in two dimensions has not been proposed. To construct a qualified 2D NLSM method is a purpose of this Letter. Using

the NLSM method, to determine the character of the ground state for the 2D J_1-J_2 model is the final purpose.

The J_1-J_2 model with plaquette-type distortion is represented by the Hamiltonian:

$$H = \sum_{\langle i,j\rangle} J_{1;ij}\mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle\langle i,k\rangle\rangle} J_{2;ik}\mathbf{S}_i \cdot \mathbf{S}_k, \tag{1}$$

where $\mathbf{S}_i$ is the spin of magnitude S at site i. The first and the second summations are taken over NN and NNN pairs, respectively, in a square lattice. $J_{1;ij}$ takes J_1 or J_1', and $J_{2;ik}$ does J_2, J_2', or J_2'' as shown in Fig. 1(a). The system is reduced to the uniform J_1-J_2 model when $J_1 = J_1'$ and $J_2 = J_2' = J_2''$. In the limit of $J_1' = J_2' = J_2'' = 0$, the lattice is an assembly of isolated plaquettes each of which consists of four spins connected by J_1 and J_2 [Fig. 1(b)]. Also, in the limit of $J_1 = J_2 = J_2'' = 0$, the lattice is an assembly of another kind of isolated plaquettes; each consists of four spins connected by J_1' and J_2' [Fig. 1(c)]. Hamiltonian (1) is invariant under the simultaneous exchanges of J_1 and J_1', and of J_2 and J_2'. The symmetric case of $J_1 = J_1'$ and $J_2 = J_2'$ includes the uniform J_1-J_2 model.

We consider the quantum Hamiltonian (1) in the classical Néel ordered region. The expectation value of $\mathbf{S}_j$ for a spin coherent state at imaginary time τ is given as

$$\langle \mathbf{S}_j \rangle = (-1)^j S\mathbf{n}_j(\tau) \quad \text{with } \mathbf{n}_j^2 = 1, \tag{2}$$

where $(-1)^j$ is a symbol taking + or − depending on the sublattice which the jth site belongs to. The partition function is then written in a path integral formula as

$$Z = \int D[\mathbf{n}_j(\tau)] \prod_j \delta(\mathbf{n}_j^2(\tau) - 1) e^{-A}. \tag{3}$$

The action A at temperature $1/\beta$ is given by

$$A = iS\sum_j (-1)^j w[\mathbf{n}_j] + \int_0^\beta d\tau H(\tau). \tag{4}$$

The first term is the Berry phase term with the solid angle $w[\mathbf{n}_j]$ which the unit vector $\mathbf{n}_j(\tau)$ forms in period β. $H(\tau)$ in the second term is given by

$$H(\tau) = \frac{1}{2}S^2 \sum_{\langle i,j\rangle} J_{1;ij}[\mathbf{n}_i(\tau) - \mathbf{n}_j(\tau)]^2 - \frac{1}{2}S^2 \sum_{\langle\langle i,k\rangle\rangle} J_{2;ik}[\mathbf{n}_i(\tau) - \mathbf{n}_k(\tau)]^2, \tag{5}$$

where the constraint $\mathbf{n}_j^2(\tau) = 1$ in the δ function of Eq. (3) has been used. Hereafter we do not explicitly write the τ dependence of $\mathbf{n}_j(\tau)$.

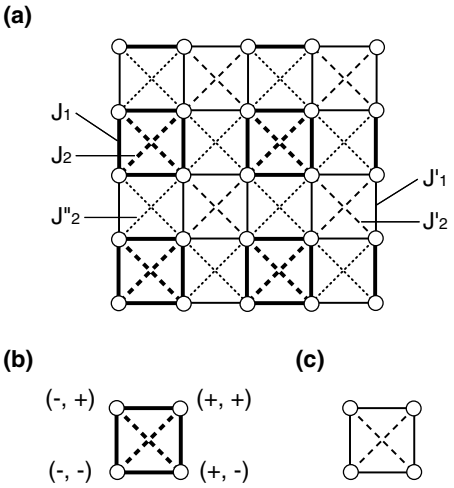

FIG. 1. (a) Lattice of the J_1-J_2 model. Lattice sites are denoted by small circles and exchange constants are by various lines between the circles. (b) A plaquette consisting of four sites connected by J_1 and J_2; this is a block which is a unit in the NLSM formulation. A variable $\mathbf{n}_j$ in the pth block is relabeled as $\mathbf{n}^{\mu\nu}(p)$, where μ and ν take + or −. The value of (μ, ν) at each site is shown. (c) Another kind of plaquette consisting of four sites connected by J_1' and J_2'.

We adopt a plaquette of Fig. 1(b) as a unit of transformation and call it a block; we would choose another kind of plaquette in Fig. 1(c) as a block. We relabel four variables, $\mathbf{n}_j$'s, in the pth block as $\mathbf{n}^{++}(p)$, $\mathbf{n}^{+-}(p)$, $\mathbf{n}^{-+}(p)$, and $\mathbf{n}^{--}(p)$, as shown in Fig. 1(b). By analogy with the one-dimensional case [26], we transform them as

$$\mathbf{n}^{\mu\nu}(p) = \mathbf{m}(p) + a[\mu\nu\mathbf{L}_0(p) + \mu\mathbf{L}_1(p) + \nu\mathbf{L}_2(p)]. \tag{6}$$

Here $\mathbf{L}_0(p)$, $\mathbf{L}_1(p)$, and $\mathbf{L}_2(p)$ describe small fluctuations around $\mathbf{m}(p)$. According to the variable transformation, four original constraints, $[\mathbf{n}^{\mu\nu}(p)]^2 = 1$ ($\mu, \nu = \pm$), are changed to four new constraints, $\mathbf{m}^2(p) = 1$ and $\mathbf{m}(p) \cdot \mathbf{L}_q(p) = 0$ ($q = 0, 1, 2$). Thus we obtained a new set of variables, the number of which is the same as that of the original variables. This plaquette-based transformation is inevitable to keep the original degrees of freedom even in the uniform J_1-J_2 model.

In the continuum limit, the first term of the action (4) is written as $iS\sum_p \sum_{\mu,\nu} \mu\nu w[\mathbf{n}^{\mu\nu}(p)] = i(S/a) \times \int d\tau d^2\mathbf{r}\mathbf{L}_0(\mathbf{m} \times \partial_\tau \mathbf{m})$ with lattice spacing a. For the second term of Eq. (4), we substitute Eq. (6) into Eq. (5) and take the continuum limit. Thus, to the leading order of derivatives and fluctuations, we have the field-theoretic action

$$A = S^2 \int d\tau d^2\mathbf{r}\Big\{\frac{i}{Sa}\mathbf{L}_0 \cdot (\mathbf{m} \times \partial_\tau \mathbf{m}) + J_0'[(\partial_x \mathbf{m})^2 + (\partial_y \mathbf{m})^2 - 2\partial_x \mathbf{m} \cdot \mathbf{L}_1 - 2\partial_y \mathbf{m} \cdot \mathbf{L}_2] + 2(J_1 + J_1')\mathbf{L}_0^2 + (J_0 + J_0')(\mathbf{L}_1^2 + \mathbf{L}_2^2)\Big\} \tag{7}$$

with $J_0 \equiv J_1 - J_2 - J_2''$ and $J_0' \equiv J_1' - J_2' - J_2''$. This action includes all the low-energy excitations surviving the continuum approximation, since the original degrees of freedom are not spoiled in the variable transformation (6). In Eq. (7), $\mathbf{L}_0$, $\mathbf{L}_1$, and $\mathbf{L}_2$ are massive fields [28], so that they are irrelevant to a symmetry change of the ground state.

Now we integrate out the partition function for the action (7) with respect to massive fields $\mathbf{L}_0$, $\mathbf{L}_1$, and $\mathbf{L}_2$. The resultant partition function contains the NLSM action:

$$A_{\text{eff}} = \int d\tau d^2\mathbf{r}\Big\{\frac{1}{8a^2(J_1 + J_1')}(\partial_\tau \mathbf{m})^2 + S^2\Big(\frac{1}{J_0} + \frac{1}{J_0'}\Big)^{-1}[(\partial_x \mathbf{m})^2 + (\partial_y \mathbf{m})^2]\Big\}. \tag{8}$$

There appears no topological term even if the NNN interactions exist. The bare spin-wave velocity is read as $v = 2\sqrt{2}Sa(J_1 + J_1')^{1/2}(1/J_0 + 1/J_0')^{-1/2}$. Action A_{eff} keeps the original invariance against the simultaneous exchanges of J_1 and J_1', and of J_2 and J_2', meaning that the same action is obtained if we use a plaquette in Fig. 1(c), instead of Fig. 1(b), as a block. This result reflects the fact that the variable transformation (6) does not restrict the spin motion to form a singlet on the plaquette of Fig. 1(b).

We apply the RG analysis by Chakravarty *et al.* [9] to the present NLSM. We first introduce rescaled dimensionless coordinates, $x_0 = \Lambda v\tau$, $x_1 = \Lambda x$, and $x_2 = \Lambda y$, with a momentum cutoff Λ of order a^{-1}. The NLSM action (8) is then rewritten as

$$A_{\text{eff}} = \frac{1}{2g_0}\int d^3x\Big(\frac{\partial \mathbf{m}}{\partial x_\mu}\Big)^2 \tag{9}$$

with coupling constant $g_0 = \sqrt{2}\Lambda a S^{-1}(J_1 + J_1')^{1/2} \times (1/J_0 + 1/J_0')^{1/2}$. By RG equations up to one-loop approximation, the quantum phase transition from the AF ordered (Néel) state to a disordered state takes place at $g_0 = 4\pi$. Rewriting this, the phase boundary in the space of the exchange parameters is given by

$$(J_1 + J_1')\Big(\frac{1}{J_0} + \frac{1}{J_0'}\Big) = \frac{2}{\lambda} \quad \text{with } \lambda \equiv \Big(\frac{\Lambda a}{2\pi S}\Big)^2. \tag{10}$$

Parameter λ represents the strength of quantum effect; $\lambda = 0$ in the classical spin limit.

To make the NLSM method complete, we determine the cutoff Λ by considering the number of degrees of freedom for the square lattice. The variable $\mathbf{m}$ is originally defined for each block of size $2a \times 2a$ [Fig. 1(b) and Eq. (6)] and is taken a continuum limit. Hence the correspondence of the momentum spaces is expressed as $(\pi/a)^2 = \pi\Lambda^2$, or the cutoff is given by $\Lambda = \sqrt{\pi}/a$. Thus Eq. (10) unambiguously determines the phase boundary between the ordered and the disordered phases.

In the uniform limit ($J_1 = J_1'$, $J_2 = J_2' = J_2''$), the system depends only on frustration parameter $\alpha \equiv J_2/J_1$ and Eq. (10) is reduced to $\alpha = \frac{1}{2} - \lambda$. Hence, for $S = \frac{1}{2}$ with $\Lambda = \sqrt{\pi}/a$, the critical value for α is given as $\alpha_c \simeq 0.18$. Thus the NLSM method succeeds in producing a critical value satisfying $0 < \alpha_c < \frac{1}{2}$ without any additional assumption or interpretation. The value is smaller than $\sim$0.4 estimated by the QMC simulation [15,16]. The deviation reflects the difference between the dispersions for spin-wave excitations in the lattice and the continuum models and may be reduced by adjusting the cutoff. Since we aim at inspecting the continuity of a phase, we do not need such a phenomenological adjustment.

In the limit of no frustration ($J_2 = J_2' = J_2'' = 0$), the plaquette distortion may cause an order-disorder transition. We denote the strength of the distortion by distortion parameter γ defined as $J_1' = (1 - \gamma)J_1$. Then Eq. (10) produces the critical value $\gamma_c = 2 - \lambda^{-1} + \sqrt{\lambda^{-2} - 2\lambda^{-1}}$. This value decreases from 1 to 0 as λ increases from 0 to $\frac{1}{2}$.

We now examine the continuity of the ground state between both the limits above. To be concrete, we parametrize the exchange constants as $J_1' = (1 - \gamma)J_1$, $J_2' = (1 - \gamma)^2 J_2$, and $J_2'' = (1 - \gamma)J_2$ for $0 \le \gamma < 1$. Equation (10) for the phase boundary is reduced to a simple form as $\alpha = (2 - \gamma)^{-1} - \frac{1}{2}\lambda(2 - \gamma)(1 - \gamma)^{-1}$. The phase diagram in the γ-α parameter space is shown in Fig. 2. The bold line with $S = \infty$ is the classical phase boundary between the Néel and the collinear phases [28]. The phase boundary of $S = \frac{1}{2}$ between the gapful and the gapless phases for variable $\mathbf{m}$ is the thin solid line; the state above is gapful, while that below is gapless corresponding to the Néel (AF) ordered state. Boundaries for other spin magnitudes S are also shown by dashed lines.

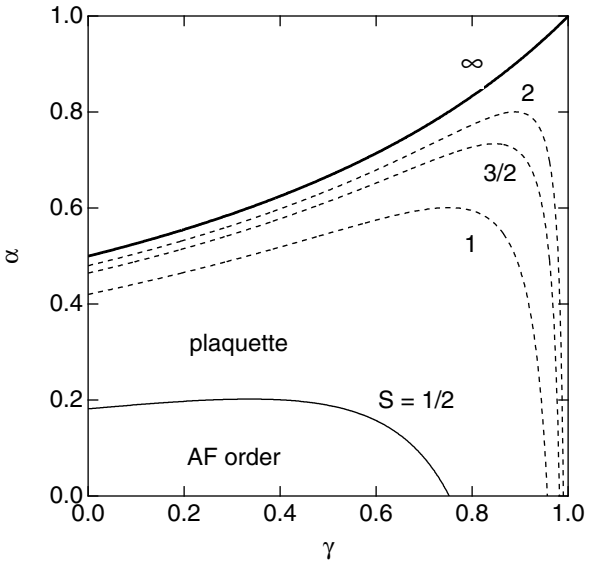

FIG. 2. Phase diagram in the space of distortion parameter γ and frustration parameter α ($= J_2/J_1$). The bold solid line for $S = \infty$ separates the classical Néel and the classical collinear phases. The region between the bold solid and the thin solid lines is the gapful plaquette phase for $S = \frac{1}{2}$. The phase boundaries for $S = 1, \frac{3}{2}$, and 2 are also shown by dashed lines.

The gapful region of **m** in Fig. 2 extends continuously from the uniform limit on the α axis ($\gamma = 0$) to the limit of no frustration on the γ axis ($\alpha = 0$). Remembering that fields $\mathbf{L}_0$, $\mathbf{L}_1$, and $\mathbf{L}_2$ are gapful, there is no gapless excitation throughout the region whether it is triplet or singlet. Hence, the whole gapful region in Fig. 2 is a single disordered phase. In particular, the phase continues to the point of $(\gamma, \alpha) = (1, 0)$ [29]. Hence a disordered ground state on the α axis finally continues to the ground state of the assembly of isolated plaquettes.

Thus there remain two possibilities for a disordered ground state of the uniform J_1-J_2 model, which is on the α axis in the phase diagram (Fig. 2). First, the translational symmetry may be spontaneously broken; then the ground states are fourfold degenerate and one of them continues to the ground state at $(\gamma, \alpha) = (1, 0)$. Second, the symmetry may not be spontaneously broken; then the ground state is unique and is a uniform RVB state with strong fluctuations of plaquette singlets. However, the second possibility is excluded by Oshikawa's commensurability condition [22]. Applying it to the present case, a uniform ground state with triplet excitation gap must be accompanied with other gapless excitations like singlet ones. Such gapless excitations do not exist as we have already shown. We therefore conclude that the disordered ground states for the uniform J_1-J_2 model are fourfold degenerate plaquette states with spontaneously broken translational invariance.

Finally, we discuss possible experiments to detect the plaquette state for materials with $J_2/J_1 \sim 0.45$ which will hopefully be found in the future. In a realistic layered material, the uniform J_1-J_2 model is accompanied with at least weak three dimensionality. Hence, at a finite temperature, the system will spontaneously break the translational symmetry to fall into a plaquette phase. The appearance of a spin gap at the temperature will be observed. The characteristics of the plaquette state appear in the dispersion relation, which reflects the invariance for the translations of $2a$ in the x and the y directions. They will be observed in neutron scattering experiments. In a material where the spin system weakly interacts with the lattice, the spontaneous symmetry breaking for the spin degrees of freedom may induce a plaquette-type lattice distortion, which will be observed by x-ray diffraction. Such a distorted system may correspond to a point deviated from the α axis in the plaquette phase of Fig. 2.

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*Present address: Department of Physics, Niigata University, Ikarashi, Niigata 950-2181, Japan.

[1] For a review, see E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).
[2] For a review, see M. A. Kastner *et al.*, Rev. Mod. Phys. **70**, 897 (1998).
[3] R. Melzi *et al.*, Phys. Rev. Lett. **85**, 1318 (2000); Phys. Rev. B **64**, 024409 (2001); P. Carretta *et al.*, Phys. Rev. Lett. **88**, 047601 (2002).
[4] H. Rosner *et al.*, Phys. Rev. Lett. **88**, 186405 (2002); Phys. Rev. B **67**, 014416 (2003).
[5] P.W. Anderson, Phys. Rev. **86**, 694 (1952).
[6] P. Chandra and B. Doucot, Phys. Rev. B **38**, 9335 (1988).
[7] F. Mila and T. M. Rice, Physica (Amsterdam) **157C**, 561 (1989); **38C**, 9335 (1988).
[8] H. Nishimori and Y. Saika, J. Phys. Soc. Jpn. **59**, 4454 (1990).
[9] S. Chakravarty *et al.*, Phys. Rev. Lett. **60**, 1057 (1988); Phys. Rev. B **39**, 2344 (1989).
[10] T. Einarsson and H. Johannesson, Phys. Rev. B **43**, 5867 (1991).
[11] E. Dagotto and A. Moreo, Phys. Rev. B **39**, 4744 (1989); Phys. Rev. Lett. **63**, 2148 (1989).
[12] K. Sano *et al.*, J. Phys. Soc. Jpn. **60**, 3807 (1991).
[13] H. J. Schultz and T. A. Ziman, Europhys. Lett. **18**, 355 (1992); H. J. Schultz *et al.*, J. Phys. I (France) **6**, 675 (1996).
[14] L. Capriotti *et al.*, cond-mat/0305099.
[15] S. Sorella, Phys. Rev. Lett. **80**, 4558 (1998).
[16] L. Capriotti and S. Sorella, Phys. Rev. Lett. **84**, 3173 (2000).
[17] M. S. L. du Croo de Jongh *et al.*, Phys. Rev. B **62**, 14 844 (2000).
[18] M. E. Zhitomirsky and K. Ueda, Phys. Rev. B **54**, 9007 (1996).
[19] R. R. P. Singh *et al.*, Phys. Rev. B **60**, 7278 (1999).
[20] V. N. Kotov *et al.*, Phys. Rev. B **60**, 14 613 (1999); O. P. Sushkov *et al.*, Phys. Rev. B **63**, 104420 (2001).
[21] L. Capriotti *et al.*, Phys. Rev. Lett. **87**, 097201 (2001).
[22] M. Oshikawa, Phys. Rev. Lett. **84**, 1535 (2000).
[23] A. Koga *et al.*, J. Phys. Soc. Jpn. **68**, 642 (1999); **68**, 2373 (1999); A. Voigt, Comput. Phys. Commun. **146**, 125 (2002).
[24] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).
[25] I. Affleck, Nucl. Phys. **B257**, 397 (1985); **B265**, 409 (1986).
[26] K. Takano, Phys. Rev. Lett. **82**, 5124 (1999); Phys. Rev. B **61**, 8863 (2000).
[27] T. Dombre and N. Read, Phys. Rev. B **38**, 7181 (1988); E. Fradkin and M. Stone, Phys. Rev. B **38**, 7215 (1988); X.-G. Wen and A. Zee, Phys. Rev. Lett. **61**, 1025 (1988); F. D. M. Haldane, Phys. Rev. Lett. **61**, 1029 (1988).
[28] The present analysis is performed in the classical Néel region, $J_0 + J_0' > 0$.
[29] The NLSM method is not applicable to this point, since the continuum approximation is unsuitable to the isolated plaquettes. However the ground state is not singular at the point. In fact, the ground state is the direct product of the unique singlet ground states for the isolated plaquettes, and the excitation gap is of order J_1. As γ decreases from 1 on the γ axis, the ground state remains nondegenerate with the plaquette symmetry at least for $J_1' \ll J_1$.