

## Spin-gap phase of a quantum spin system on a honeycomb lattice

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We study a quantum spin system on a honeycomb lattice, when it includes frustration and distortion in antiferromagnetic (AF) exchange interactions. We transform the spin system onto a nonlinear  $\sigma$  model (NLSM), preserving the original spin degrees of freedom. Assisted by a renormalization-group argument, the NLSM provides a ground-state phase diagram consisting of an ordered AF phase and a disordered spin-gap phase. The spin-gap phase extends from a strong frustration regime to a strong distortion regime, showing that the disordered ground states are essentially the same in both regimes. In the spin-1/2 case, the spin-gap phase for spin system on a honeycomb lattice is larger than that for the  $J_1$ - $J_2$  model on a square lattice.

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Recently, materials regarded as spin systems on honeycomb lattices have been found. Kataev *et al.*<sup>1</sup> synthesized a spin-1/2 ( $S=\frac{1}{2}$ ) material on a honeycomb lattice  $\text{InCu}_{2/3}\text{V}_{1/3}\text{O}_3$ . They estimated the first-neighbor exchange parameter as 280 K, and found an antiferromagnetic (AF) ordering at 38 K with interlayer interactions. Their results are confirmed in refined samples by Kikuchi *et al.*<sup>2</sup> Miura *et al.*<sup>3</sup> reported other honeycomb-lattice materials  $\text{Na}_3\text{T}_2\text{SbO}_6$  ( $T=\text{Cu, Ni, and Co}$ ). The material with  $T=\text{Cu}$  is an  $S=\frac{1}{2}$  system and shows a spin gap in magnetic susceptibility. The materials with  $T=\text{Ni}$  and  $\text{Co}$  are  $S=1$  and  $S=\frac{3}{2}$  systems, respectively, and both have AF order. So far, the samples are reported to have fairly strong dimerlike distortion<sup>4</sup> ( $T=\text{Cu}$ ) or randomness ( $T=\text{Ni}$  and  $\text{Co}$ ) in exchange interactions on honeycomb lattices. In view of recent experimental activity, materials regarded as various types of spin systems on honeycomb lattices are expected to be synthesized. Hence it is worthwhile to show a theoretical scheme for spin systems on honeycomb lattices.

In two dimensions, there are two kinds of simple bipartite lattices, a square lattice and a honeycomb lattice, if only first-neighbor AF interactions are considered. A spin system on a bipartite lattice has a ground state with AF order in the large-spin-magnitude limit. A question is whether or not a disordered spin-gap phase appears with destroyed AF order, when the spin magnitude becomes small so that quantum fluctuations are large. Quantum fluctuations are further enhanced by frustration among AF interactions and by distortion, or inhomogeneity, in interactions. Hence it would be interesting to study the disordered state under variable strengths of frustration and distortion.

In the square-lattice case, the  $J_1$ - $J_2$  model has been eagerly studied. Theoretically, whether or not a disordered state is formed around  $J_2/J_1=0.5$  has been a controversial problem for a long time. The difficulty originates from the fact that the region of the disordered phase is very narrow in the parameter space even if it exists. Further, a material realizing the  $J_1$ - $J_2$  model in a disordered state has not been found. The difficult synthesis of such a material also seems to come from the narrowness of the disordered phase.

In contrast, a quantum spin system on a honeycomb lattice is a more hopeful candidate for finding a disordered ground state. In fact, the coordination number 3 of the hon-

eycomb lattice is smaller than 4 of the square lattice, and so the AF order for a honeycomb lattice is more fragile. The case without distortion has been treated by numerical diagonalization and linear spin-wave theory,<sup>11</sup> Schwinger-boson mean-field theory,<sup>10</sup> and the nonlinear  $\sigma$  model (NLSM) method.<sup>6</sup> A systematic study of a spin system on a honeycomb lattice with both frustration and distortion is expected.

Among various methods to analyze a two-dimensional spin system, mapping it onto a NLSM and applying a renormalization-group (RG) analysis is effective to study a quantum phase transition. In the case of a square lattice, a spin system with only first-neighbor exchange interactions is shown to be mapped onto a NLSM without a topological term.<sup>5</sup> For the  $J_1$ - $J_2$  model, NLSM methods have been developed and a spin-gap phase studied.<sup>6,7</sup> After that, a refined mapping onto a NLSM was proposed for a  $J_1$ - $J_2$  model on a square lattice with plaquette distortion.<sup>8</sup> In the derivation, the original spin degrees of freedom are properly treated without doubling by a variable transformation, in which a slowly varying AF variable appears for a block of four spins. The derived NLSM has no topological term even if a second-neighbor interaction and a plaquette distortion exist. The RG analysis<sup>7</sup> was successfully applied to the obtained NLSM and provides a phase diagram in the space of parameters describing frustration and plaquette distortion.<sup>8</sup>

In this Rapid Communication, we propose a NLSM method for a spin system on a honeycomb lattice in the case with frustration by second-neighbor AF interactions and dimerlike distortion in first-neighbor AF interactions. The original spin Hamiltonian is mapped onto a field theory in such a manner that six close spins are transformed into one slow modulation of the AF spin configuration and five rapid motions. The degrees of freedom of the original spin model are properly considered through the derivation, and the choice of the cutoff becomes unique.<sup>9</sup> The derived field theory is a NLSM except for an additional term. After dealing with the additional term, we apply the RG analysis<sup>7</sup> to the NLSM to obtain the ground-state phase diagram in the parameter space, and show a disordered region in which there is a spin gap.

The Hamiltonian of the present spin system on a honeycomb lattice is

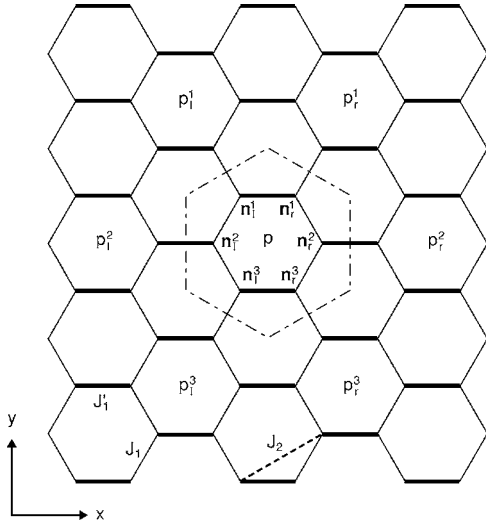


FIG. 1. Spin system on a honeycomb lattice, where a spin with magnitude  $S$  is on a lattice point. The first-neighbor exchange interaction with coupling constant  $J_1$  ( $J_1'$ ) is represented by the thin (bold) solid line. The second-neighbor exchange interaction with coupling constant  $J_2$  is represented by the dotted line; dotted lines should be between any second-neighbor spins, although not drawn to avoid complication. The  $p$ th block is shown by the dash-dotted line; the adjacent blocks are labeled as  $p_l^j$  ( $j=1,2,3$ ;  $u=l,r$ ). Variables  $\mathbf{n}_u^j$  ( $j=1,2,3$ ;  $u=l,r$ ) represent spins in the path-integral formula with spin coherent states.

$$H = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where  $\mathbf{S}_i$  is the quantum spin at site  $i$  and the spin magnitude  $S$  is uniform and arbitrary. The exchange constant  $J_{ij}$  takes the value  $J_1$ ,  $J_1'$ , or  $J_2$  depending on the bond between sites  $i$  and  $j$  as shown in Fig. 1. The distance between first-neighbor spins will be written as  $a$ . We consider the quantum Hamiltonian in the classical Néel ordered region.

Quantum fluctuation is increased by distortion and frustration in exchange constants. To measure the strengths, we introduce distortion parameter  $\delta$  and frustration parameter  $\alpha$  as follows:

$$J_1 = \bar{J}_1(1 - \delta), \quad J_1' = \bar{J}_1(1 + 2\delta), \quad \alpha = J_2/\bar{J}_1. \quad (2)$$

Then  $\bar{J}_1$  is an average of all first-neighbor exchange constants:  $\bar{J}_1 = \frac{1}{3}(2J_1 + J_1')$ .

We construct a path-integral formula by means of spin coherent states. For a spin coherent state labeled by the unit vector  $\mathbf{n}_j(\tau)$  at imaginary time  $\tau$ , the expectation value of  $\mathbf{S}_j$  is represented as  $\langle \mathbf{S}_j \rangle = \zeta_j S \mathbf{n}_j(\tau)$ , where  $\zeta_j$  is + or - depending on the sublattice to which the  $j$ th site belongs. Then the general formula of the partition function at temperature  $1/\beta$  is written as

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

with action

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

The first term in  $A$  is the Berry phase term with the solid angle  $w[\mathbf{n}_j]$  that the unit vector  $\mathbf{n}_j(\tau)$  forms in period  $\beta$ . The Hamiltonian  $H(\tau)$  is written as

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

using the constraint  $[\mathbf{n}_j(\tau)]^2 = 1$ . Hereafter we do not explicitly denote the  $\tau$  dependence of  $\mathbf{n}_j(\tau)$ .

By finding an appropriate variable transformation, we will separate out the slow mode describing AF fluctuations for the honeycomb lattice. We adopt the region enclosed by a dash-dotted line in Fig. 1 as the unit of transformation, and call it a *block*; the first-neighbor blocks to the  $p$ th block are labeled by  $p_l^j$  ( $j=1,2,3$  and  $u=r,l$ ). Accordingly we relabel the six variables  $\mathbf{n}_j$  in the  $p$ th block as  $\mathbf{n}_r^l(p)$  and  $\mathbf{n}_l^r(p)$  ( $j=1,2,3$ ), as shown in Fig. 1. Then we perform the following variable transformation for each block:

$$\begin{aligned} \mathbf{n}_r^1(p) &= \mathbf{m}(p) + 3a[-\mathbf{L}(p) + \mathbf{X}_-(p) + \mathbf{Y}_-(p)], \\ \mathbf{n}_r^2(p) &= \mathbf{m}(p) + 3a[\mathbf{L}(p) + \mathbf{X}_+(p) + \mathbf{Y}_+(p)], \\ \mathbf{n}_r^3(p) &= \mathbf{m}(p) + 3a[-\mathbf{L}(p) - \mathbf{X}_-(p) - \mathbf{Y}_-(p)], \\ \mathbf{n}_l^1(p) &= \mathbf{m}(p) + 3a[\mathbf{L}(p) + \mathbf{X}_-(p) - \mathbf{Y}_-(p)], \\ \mathbf{n}_l^2(p) &= \mathbf{m}(p) + 3a[-\mathbf{L}(p) - \mathbf{X}_+(p) + \mathbf{Y}_+(p)], \\ \mathbf{n}_l^3(p) &= \mathbf{m}(p) + 3a[\mathbf{L}(p) - \mathbf{X}_-(p) + \mathbf{Y}_-(p)]. \end{aligned} \quad (6)$$

Here  $\mathbf{L}(p)$ ,  $\mathbf{X}_\pm(p)$ , and  $\mathbf{Y}_\pm(p)$  describe fluctuation modes around the AF variable  $\mathbf{m}(p)$ . The six original constraints  $[\mathbf{n}_r^j(p)]^2 = 1$  ( $j=1,2,3$  and  $t=r,l$ ) are changed to six new constraints  $[\mathbf{m}(p)]^2 = 1$  and  $\mathbf{m}(p) \cdot \mathbf{L}(p) = 0$ ,  $\mathbf{m}(p) \cdot \mathbf{X}_\pm(p) = 0$ , and  $\mathbf{m}(p) \cdot \mathbf{Y}_\pm(p) = 0$ .<sup>12</sup>

Taking the continuum limit, the first term of action (4) is written as

$$\begin{aligned} iS \sum_p \sum_{j=1}^3 (-1)^j \{w[\mathbf{n}_r^j(p)] - w[\mathbf{n}_l^j(p)]\} \\ \rightarrow -i \frac{2S}{3a} \int d\tau d^2\mathbf{r} (3\mathbf{L} + \mathbf{X}_+) \cdot (\mathbf{m} \times \partial_\tau \mathbf{m}). \end{aligned} \quad (7)$$

In each term of Eq. (5), a calculation like the following is carried out;

$$\begin{aligned} \mathbf{n}_r^2(p) - \mathbf{n}_l^2(p) &= \mathbf{m}(p) - \mathbf{m}(p) + 6a(\mathbf{L} + \mathbf{X}_+) \\ &\rightarrow 3a[\partial_x \mathbf{m} + 2(\mathbf{L} + \mathbf{X}_+)] \end{aligned} \quad (8)$$

up to the lowest order of derivatives and fluctuation variables. Thus the lattice action (4) becomes the field-theoretic action

$$\begin{aligned}
A = & S^2 \int \int \int d\tau d^2\mathbf{r} \left( \frac{3}{4}(\bar{J}'_1 - 4J_2)(\partial_x \mathbf{m})^2 \right. \\
& + \frac{3}{4}(J_1 - 4J_2)(\partial_y \mathbf{m})^2 + 18\bar{J}_1 \mathbf{L}^2 + 6\mathbf{L} \cdot (2\bar{J}_1 \mathbf{X}_+ + \mathbf{B}_-) \\
& + 2(J_1 + J'_1 - 3J_2) \mathbf{X}_+^2 - 2\mathbf{X}_+ \cdot (\mathbf{B}_+ - 3J_2 \partial_x \mathbf{m}) \\
& + 2(J_1 - 3J_2)(3\mathbf{X}_-^2 - \sqrt{3}\mathbf{X}_- \cdot \partial_y \mathbf{m} + \mathbf{Y}_+^2) \\
& \left. + 6(\bar{J}'_1 - 3J_2) \mathbf{Y}_-^2 \right) \quad (9)
\end{aligned}$$

with  $\mathbf{B}_+ = \frac{1}{3}[3J'_1 \partial_x \mathbf{m} + i(aS)^{-1} \mathbf{m} \times \partial_\tau \mathbf{m}]$ ,  $\mathbf{B}_- = \frac{1}{3}[(J_1 - J'_1) \partial_x \mathbf{m} - i(aS)^{-1} \mathbf{m} \times \partial_\tau \mathbf{m}]$ ,  $\bar{J}_1 = \frac{1}{3}(2J_1 + J'_1)$ , and  $\bar{J}'_1 = \frac{1}{3}(J_1 + 2J'_1)$ . 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).

Integrating out the partition function with the action (9) with respect to the massive fields  $\mathbf{L}$ ,  $\mathbf{X}_\pm$ , and  $\mathbf{Y}_\pm$ , we obtain the following action:

$$\begin{aligned}
A' = & \int \int \int d\tau d^2\mathbf{r} \left( -i \frac{S\delta}{3a} \mathbf{m} \cdot (\partial_\tau \mathbf{m} \times \partial_x \mathbf{m}) \right. \\
& \left. + \frac{1}{18a^2 \bar{J}_1} (\partial_\tau \mathbf{m})^2 + \frac{1}{4} S^2 \bar{J}_1 [c_+ (\partial_x \mathbf{m})^2 + c_- (\partial_y \mathbf{m})^2] \right) \quad (10)
\end{aligned}$$

with  $c_+ = 1 + \delta - 2\delta^2 - 6\alpha$  and  $c_- = 1 - \delta - 6\alpha$ . Except for the first term this is a NLSM action. The first term is rewritten as

$$-iA_Q = -i \frac{S\delta}{3a} \int dy Q(y) \quad (11)$$

with

$$Q(y) = \int \int d\tau dx \mathbf{m} \cdot (\partial_\tau \mathbf{m} \times \partial_x \mathbf{m}). \quad (12)$$

The quantity  $Q(y)$  is not a topological charge in the three-dimensional Euclidean space. For a fixed value of  $y$ , however,  $Q(y)$  is the topological charge of the field  $\mathbf{m}$  in the Euclidean  $\tau$ - $x$  plane, and is an integer. Further, for continuity,  $Q(y)$  must be the same integer for all values of  $y$ . This means that there is at least a vortex line for  $Q \neq 0$ . Hence, a configuration with  $Q \neq 0$  gives positive contributions of the system size to the second and third terms in the action (10). Thus we remove the first term from the action (10), assuming that configurations with  $Q \neq 0$  are negligible.

The momentum cutoff  $\Lambda$  is determined by keeping the number of degrees of freedom for  $\mathbf{m}$ . The variable  $\mathbf{m}$  is originally defined for each hexagonal block with sides of length  $\sqrt{3}a$  (dash-dotted line in Fig. 1) and with area  $\omega = (9\sqrt{3}/2)a^2$ . Then, in the continuum limit, the momentum cutoff  $\Lambda$  is given as  $a\Lambda = (2/3)(2\pi)^{1/2} 3^{-1/4}$  from  $(2\pi)^2/\omega = \pi\Lambda^2$ .

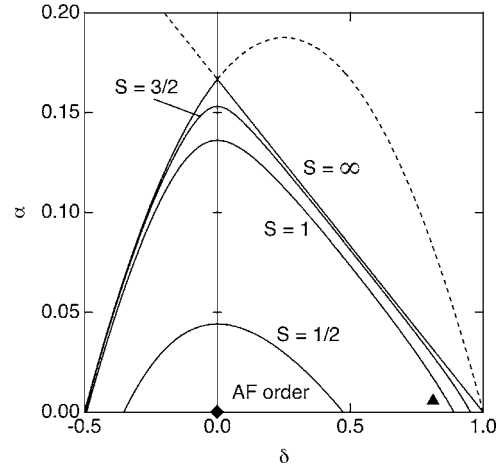


FIG. 2. Phase diagram in the space of the distortion parameter  $\delta$  and the frustration parameter  $\alpha (= J_2/\bar{J}_1)$ . Phase boundaries for  $S = \frac{1}{2}$ , 1, and  $\frac{3}{2}$  and also the classical limit of the AF phase ( $S = \infty$ ) are shown. For each  $S$ , the region below the boundary is an AF ordered phase, while that above is a disordered spin-gap phase. Possible positions for  $\text{InCu}_{2/3}\text{V}_{1/3}\text{O}_3$  and  $\text{Na}_3\text{Cu}_2\text{SbO}_6$  are shown by diamond and triangle symbols, respectively.

We introduce rescaled dimensionless coordinates  $x_0 = \Lambda v_0 (c_+ c_-)^{1/4} \tau$ ,  $x_1 = \Lambda (c_- / c_+)^{1/4} x$  and  $x_2 = \Lambda (c_+ / c_-)^{1/4} y$  with  $v_0 = 3Sa\bar{J}_1/\sqrt{2}$ . The action (10), neglecting the  $A_Q$  term, is then rewritten in a standard NLSM form:

$$A_{\text{eff}} = \frac{1}{2g_0} \int d^3x \left( \frac{\partial \mathbf{m}}{\partial x_\mu} \right)^2 \quad (13)$$

with coupling constant  $g_0 = 3\sqrt{2}a\Lambda S^{-1}(c_+ c_-)^{-1/4}$ .

Applying the one-loop RG analysis by Chakravarty *et al.*<sup>7</sup> to Eq. (13), a quantum phase transition from the Néel phase with AF order to a disordered phase takes place at  $g_0 = 4\pi$ . Rewriting this, the critical value  $\alpha_c$  for the phase boundary in the  $\delta$ - $\alpha$  space is given by

$$\alpha_c = \frac{1}{6} [1 - \delta^2 - \sqrt{\delta^2(1 - \delta)^2 + \lambda_S^2}] \quad (14)$$

with  $\lambda_S = (\sqrt{3}\pi S^2)^{-1}$ . The parameter  $\lambda_S$  is a measure of quantum character of the spin system. In the classical limit of  $\lambda_S = 0$  ( $S = \infty$ ), Eq. (14) reduces to  $\alpha_c = \frac{1}{6}(1 - \delta)$  for  $\delta > 0$  and  $\alpha_c = \frac{1}{6}(1 + \delta - 2\delta^2)$  for  $\delta < 0$ ; the present theory is applicable below the classical line. The phase boundaries (14) for  $S = \frac{1}{2}$ , 1, and  $\frac{3}{2}$  are shown in Fig. 2. For each  $S$ , the region above the solid line represented by the equation is a disordered phase with a spin gap and the region below is an AF ordered phase. Possible positions for the materials  $\text{InCu}_{2/3}\text{V}_{1/3}\text{O}_3$  and  $\text{Na}_3\text{T}_2\text{SbO}_6$  are also shown.

In the case of no distortion ( $\delta = 0$ ), Eq. (14) becomes simply  $\alpha_c = \frac{1}{6}(1 - \lambda_S)$ ; e.g.,  $\alpha_c = 0.044$  ( $S = \frac{1}{2}$ ), 0.136 ( $S = 1$ ), and 0.153 ( $S = \frac{3}{2}$ ). For  $S = \frac{1}{2}$ , the disordered phase has a range of 74% against that of the classical AF order on the  $\alpha$  axis (Fig. 2). The actual portion may be fairly reduced from this, since the NLSM method includes, e.g., a continuum approximation.<sup>13</sup> Whether or not a disordered phase survives

for  $\delta = 0$  will be considered in comparison with the square-lattice case. The portion of the disordered range in the  $J_1$ - $J_2$  model on a square lattice is  $(0.5-0.18)/0.5 \times 100 = 64\%$ .<sup>8</sup> A disordered phase exists more plausibly for a honeycomb lattice than for a square lattice as was physically expected. For  $S \geq 1$ , the disordered phase in Fig. 2 is very narrow, and so its actual existence does not seem to be expected.

In the limit of no frustration ( $\alpha=0$ ), Eq. (14) is reduced to  $2\delta^3 - 3\delta^2 + 1 - \lambda_S^2 = 0$ . The critical value of distortion for  $S = \frac{1}{2}$  is  $\delta = 0.47$ . The disordered state on the  $\delta$  axis of Fig. 2 is explained by the tendency to form singlet pairs at strong  $J'_1$  bonds.

Summarizing, we formulated a NLSM method for a spin system on a honeycomb lattice in the case that the exchange interaction includes both frustration by second-neighbor AF interactions and dimerlike distortion in first-neighbor AF interactions. Applying the RG analysis by Chakravarty *et al.*,<sup>7</sup> we have the ground-state phase diagram in the parameter space. A disordered spin-gap phase is continuously extended from a strong frustration regime to a strong distortion regime, showing that the disordered ground states in both the regimes are essentially the same. In the case of  $S = \frac{1}{2}$ , the

spin-gap phase for the spin system on a honeycomb lattice is larger than that for the  $J_1$ - $J_2$  model on a square lattice, suggesting that a disordered state likely exists.

The material  $\text{Na}_3\text{T}_2\text{SbO}_6$  with  $T = \text{Cu}$  ( $S = \frac{1}{2}$ ) has a strong dimerlike distortion and a spin gap, unlike the other similar materials with  $T = \text{Ni}$  ( $S = 1$ ) and  $T = \text{Co}$  ( $S = \frac{3}{2}$ ).<sup>3</sup> The distortion may be induced because it is in the spin-gap phase. In fact, within the spin-gap phase, the energy of spins decreases and the lattice energy increases, as the dimerlike distortion increases; the balance determines an actual distortion. On the other hand, within the AF phase, the dimerlike distortion does not reduce the spin energy, which is consistent with little distortion in materials with  $T = \text{Ni}$  and  $\text{Co}$ . To find a material with small distortion in the spin-gap phase, it is reasonable to try  $S = \frac{1}{2}$  materials with strong elastic constants as well as appropriate second-neighbor AF interactions on honeycomb lattices.

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<sup>2</sup>H. Kikuchi, Y. Kubo, Y. Fujii, M. Chiba, Y. Yamamoto, and H. Hori (unpublished).

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<sup>6</sup>T. Einarsson and H. Johannesson, *Phys. Rev. B* **43**, 5867 (1991).

<sup>7</sup>S. Chakravarty, B. I. Halperin, and D. R. Nelson, *Phys. Rev. Lett.* **60**, 1057 (1988); *Phys. Rev. B* **39**, 2344 (1989).

<sup>8</sup>K. Takano, Y. Kito, Y. Ono, and K. Sano, *Phys. Rev. Lett.* **91**, 197202 (2003).

<sup>9</sup>Reference 6 includes a derivation of a NLSM for a honeycomb lattice without distortion. This pioneering work, however, does

not keep the degrees of freedom of the original spin system in variable transformation; i.e., a spin variable is written as two new variables and the doubling is compensated by a regularization.

<sup>10</sup>A. Mattsson, P. Fröjdh, and T. Einarsson, *Phys. Rev. B* **49**, 3997 (1994).

<sup>11</sup>J. B. Fout, P. Sindzingre, and C. Lhuillier, *Eur. Phys. J. B* **20**, 241 (2001).

<sup>12</sup>Seemingly, a prefactor for  $\mathbf{m}(p)$  in each of Eqs. (6) is needed to satisfy the constraint up to the second order of the fluctuation variables; e.g.,  $\{1 - [-\mathbf{L}(p) + \mathbf{X}_-(p) + \mathbf{Y}_-(p)]^2\}^{1/2}$ . However, the factor gives only fourth-order contributions to Eq. (5) and is neglected.

<sup>13</sup>In the literature,  $S$  in only the coupling constant  $g_0$  of a NLSM is sometimes replaced by  $\sqrt{S(S+1)}$ . Following this,  $\lambda_{1/2} \approx 0.735$  for  $S = \frac{1}{2}$  changes to 0.245 and then the disordered region seems plausibly narrow. However, we do not adopt the replacement, since it has not been justified.