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THEORETICAL CALCULATION OF THE MAGNETIC PROPERTIES IN DISORDERED GROUND STATE FOR SPIN-1/2 ANTIFERROMAGNET

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The spin-1/2 quantum antiferromagnet in three-dimensional space can be described by Heisenberg model:two nearest-neighbor spins with strong interaction in z direction consistitute a dimer, every dimer can be considered as a lattice site in the tetragonal lattice. In z direction, the exchange interaction between two nearest-neighbor dimers is $\lambda' J$, and in xy plane it is 2 λJ. In bond-operator representation, within the mean-field decoupling approximation, we have calculated the phase diagram for order-disorder phase transition about λ and λ' at 0 K, obtained the critical value, λ_c=0.292, within which the disordered phase is stable in two-dimensional case(λ'=0), and found that the disordered phase is always stable for $\lambda' < 1$ in one-dimensional case(λ=0). For the disordered phase, the physical quantities, such as energy gap, ground-state energy, two-point correlation functions and correlation lengths in xy plane and in z direction, have been calculated as functions of parameters λ and λ'.

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I. INTRODUCTION

In recent years, spin-1/2 Heisenberg antiferromagnetic model has received considerable attention due to the factors: (a) many new contents contained in this model need to be investigated, and (b) there may exist a possible connection between this model and the mechanism of superconductivity.[1]

In undoped case, the parent compounds of high-temperature superconductors La_2CuO_4 and $YBa_2Cu_3O_{6+x}$ are typical antiferromagnetic insulators, only by doping to some extent, can the antiferromagnetic long range ordering be destroyed, and superconducting phenomenon appears.[2] Thus some authors study their magnetic properties in the insulating phase [3,4], and some others study how the physical mechanism for antiferromagnetism is destroyed, and propose some new physical models, such as the t-J model in which the doped vacancies may disturb antiferromagnetism by their movement[1,5], J_1-J_2 model in which the frustration resulting from the next nearest-neighbor antiferromagnetic interaction between spins will destroy antiferromagnetism in the square lattice.[6]

There is another mechanism that destroys Heisenberg antiferromagnetism without frustration, which may be called dimer mechanism. In this mechanism, two nearest-neighbor spins with opposite spin directions stay in spin singlet due to the stronger exchange interaction between them, the spin triplet with higher energy becomes excited state. When the interaction between spin pairs is small, an energy gap may occur between the triplet and the singlet, thus the excitation from the ground state to excited state is frustrated and the antiferromagnetic long range ordering is difficult to set up, and therefore the system exhibits disordered phase or the so-called dimered phase.

Hida[7] studied the crossover between the Haldane-gap phase and the dimered phase in the spin-1/2 alternating Heisenberg chain by using the exact diagonalization of small size system; Sachdev and Bhatt[8] gave phase diagrams about the dimered phase and the magnetically ordered phase on the square lattice with different spin configurations by bond-operator representation of quantum spins; using the same method, Gopalan *et al.* [9] calculated the properties of the square lattice spin system and spin ladders in dimered phase, such as the excited spectra, energy gap and ground state energy. Connecting with the $YBa_2Cu_3O_{6+x}$, Mills and Monien[10] calculated the critical value of dimered phase in double-layer Heisenberg antiferromagnet, and found the dimered phase stable when the interlayer coupling strength between spins was 4.48 times bigger than the intralayer coupling strength by using the Schwinger boson mean-field theory. This critical value was also calculated by using the other methods, such as the modified spin-wave theory[11], series expansion method[12] and quantum Monte Carlo simulation method.[13] We also studied this critical value by using the bond-operator method of spins, and calculated in detail the magnetic properties of the system in dimered phase.[14]

In this paper, we will extend the double-layer Heisenberg system with spin- 1/2 to three-dimensional(3D) space, so that the system becomes a periodic double-layer structure, and use the bond-operator method of spins to discuss the magnetic properties of this system in disordered phase.

II. THEORETICAL METHOD AND CALCULATION OF THE RELATED PHYSICAL QUANTITIES

The Heisenberg model with spatial anisotropy is given by

$$H = J \sum_{\langle ij \rangle z} S_i \cdot S_j + \lambda J \sum_{\langle ij \rangle xy} S_i \cdot S_j + \lambda' J \sum_{\langle ij \rangle z} S_i \cdot S_j \,, \tag{1}$$

where $\langle \cdots \rangle$ represents the sum over the nearest-neighbor lattice sites. The physical meaning of every term can be explained by refering to Fig.1. The first term represents the sum over the stronger exchange interactions between nearest-neighbor spins in z direction, which make the spins dimerized; the second and the third terms are the sums over the exchange interactions between spins in different dimers within the horizontal xy plane and in z direction, respectively. When $\lambda' = 0$,Eq.(1) describes the double-layer antiferromagnet; for $\lambda = 0$, it describes the one-dimensional(1D) antiferromagnet; for $\lambda \gg \lambda' > 0$, the quasi-two-dimensional(quasi-2D) antiferromagnet; for$\lambda' \gg \lambda > 0$, the quasi-1D antiferromagnet; and for $\lambda \sim \lambda'$, the 3D antiferromagnet.

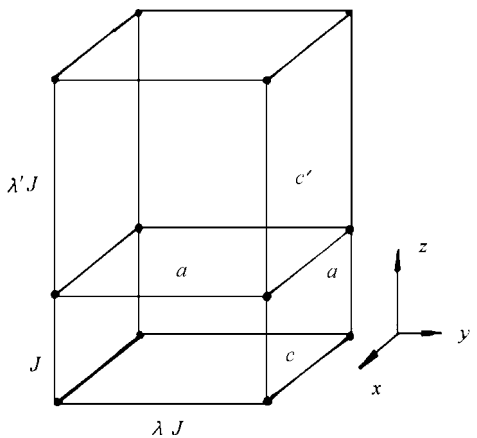

Fig.1. An original cell of periodic double-layer Heisenberg antiferromagnetic model. Every thick line represents the stronger exchange interaction between two spins on the lattice sites, it maybe considered as a dimer.

In the following, we explain briefly the bond-operator method of spins that we will adopt.[8,9] A pair of spins with antiferromagnetic coupling, $JS_i \cdot S_j$, contains four eigenstates, which consist of a singlet with the eigenenergy $-3/4J$ and three triplets, each with an eigenenergy $1/4J$. In the Hilbert sub-space set up with the four eigenstates, four new operators s, t_x, t_y, t_z, corresponding to the annihilations of a singlet and three triplets, respectively, can be formed. According to the completeness of the sub-space or the states created (annihilated) being either singlet or triplets, a constraint for the four new operators

must be satisfied, $s^+s+t_x^+t_x+t_y^+t_y+t_z^+t_z=1$. Finding the relations between spin operator $S_i(S_j)$ and s and $t_\alpha(\alpha=x,y,z)$, and supposing s and t_α are bosonic operators, we can show that all the commutation relations between spin operators are preserved and $S_i^2=S_j^2=3/4$ are also satisfied. In this way, every pair of spins with stronger exchange interaction in the system can be expressed with four new bosonic operators (bond-operators), and thus the Hamiltonian in Eq.(1) becomes

$$H=H_0+\frac{\lambda}{2}(H_1+H_2)+\frac{\lambda'}{2}(H_1'+H_2'), \tag{2}$$

where

$$H_0=J\sum_{li}\left(-\frac{3}{4}s_{li}^+s_{li}+\frac{1}{4}t_{li\alpha}^+t_{li\alpha}\right)-\sum_{li}\mu_{li}(s_{li}^+s_{li}+t_{li\alpha}^+t_{li\alpha}-1), \tag{3}$$

$$H_1=\frac{J}{2}\sum_{l\langle ij\rangle}(t_{li\alpha}^+t_{lj\alpha}s_{lj}^+s_{li}+t_{li\alpha}^+t_{lj\alpha}^+s_{li}s_{lj}+\text{H.c.}), \tag{4}$$

$$H_2=\frac{J}{4}\sum_{l\langle ij\rangle}(1-\delta_{\alpha\beta})(t_{li\alpha}^+t_{lj\alpha}^+t_{li\beta}t_{lj\beta}-t_{li\alpha}^+t_{lj\beta}^+t_{lj\alpha}t_{li\beta}+\text{H.c.}), \tag{5}$$

$$H_1'=-\frac{J}{4}\sum_{\langle ll'\rangle i}(t_{li\alpha t}^+t_{l'i\alpha}s_{l'i}^+s_{li}+t_{li\alpha}^+t_{l'i\alpha}^+s_{li}s_{l'i}+\text{H.c.}), \tag{6}$$

$$\begin{aligned}H_2'=&-\frac{J}{4}\sum_{\langle ll'\rangle i}\left[\frac{1}{2}(1-\delta_{\alpha\beta})(t_{li\alpha}^+t_{l'i\alpha}^+t_{li\beta}t_{l'i\beta}-t_{li\alpha}^+t_{l'i\beta}^+t_{li\beta}t_{l'i\alpha})\right.\\&\left.+\mathrm{i}\varepsilon_{\alpha\beta\gamma}(s_{li}^+t_{l'i\alpha}^+t_{li\beta}t_{l'i\gamma}+s_{l'i}^+t_{li\alpha}^+t_{li\beta}t_{l'i\gamma})+\text{H.c.}\right].\end{aligned} \tag{7}$$

Here l and l' label the dimer layers, i and j label the dimers in a dimer layer, $\alpha(\beta\,\text{or}\,\gamma)$ can take x,y and z, and repeated indices of them indicate a summation; ε is the totally antisymmetric tensor. In Eq.(3) μ_{li} is the chemical potential, by which the constraint $s_{li}^+s_{li}+t_{li\alpha}^+t_{li\alpha}$=1 is introduced.

Because of the$-3J/4$ s$^+$s term in H_0, single s bosons will condense, $<s>=<s^+>=\overline{s}$. Moreover, the translation invariance of the system implies that we may assume $\mu_{li}=\mu$, i.e., site independent. Within the mean-field approximation, by omitting the $H_2(H_2')$ term, the Hamiltonian (2) becomes

$$\begin{aligned}\overline{H}=&-N\left(\frac{3}{4}J+\mu\right)\overline{s}^2+N\mu+\left(\frac{J}{4}-\mu\right)\sum_k t_{k\alpha}^+t_{k\alpha}\\&+\frac{J\overline{s}^2}{4}\sum_k\left[\left(\lambda Z_\parallel\gamma_\parallel(k)-\frac{\lambda'}{2}Z_\perp\gamma_\perp(k)\right)(2t_{k\alpha}^+t_{k\alpha}+t_{k\alpha}t_{-k\alpha}+t_{-k\alpha}^+t_{k\alpha}^+)\right],\end{aligned} \tag{8}$$

where N represents the total number of dimers, with $Z_{\parallel}=4, Z_{\perp}=2, \gamma_{\parallel}(k)$=[cos($k_x a$)+ cos($k_y a$)]/2, $\gamma_{\perp}(k)$=cos($k_z c'$).

Using Bogoliubov transformation to make the mean-field Hamiltonian diagonalized[15], we obtain the excited spectrum from the condensed singlet to the excited triplets

$$\omega_k = \sqrt{\alpha_k^2 - (2\beta_k)^2}\,, \tag{9}$$

where

$$\alpha_k = \frac{J}{4} - \mu + \frac{J\overline{s}^2}{2}\left(\lambda Z_{\parallel}\gamma_{\parallel}(k) - \frac{\lambda'}{2}Z_{\perp}\gamma_{\perp}(k)\right), \tag{10a}$$

$$\beta_k = \frac{J\overline{s}^2}{4}\left(\lambda Z_{\parallel}\gamma_{\parallel}(k) - \frac{\lambda'}{2}Z_{\perp}\gamma_{\perp}(k)\right), \tag{10b}$$

and the ground state energy of the system is

$$E_{\rm g} = -N\left(\frac{3}{4}J + \mu\right)\overline{s}^2 + N\mu + \frac{1}{2}\sum_k(\omega_k - \alpha_k). \tag{11}$$

The chemical potential μ and the mean-field parameter $\overline{s}$ may be determined from the minimization of the ground state energy, thus two equations about μ and $\overline{s}$ are obtained

$$\frac{3}{2} - \overline{s}^2 = \frac{1}{N}\sum_k \frac{\alpha_k}{2\omega_k}\,, \tag{12}$$

$$\frac{3}{2}J + 2\mu = \frac{J}{N}\sum_k\left[\left(\lambda Z_{\parallel}\gamma_{\parallel}(k) - \frac{\lambda'}{2}Z_{\perp}\gamma_{\perp}(k)\right)\frac{\alpha_k - 2\beta_k}{2\omega_k}\right]. \tag{13}$$

Solving for μ and $\overline{s}$ from the above equations, and substituting them into Eqs.(11) and (9), we can obtain the ground state energy $E_{\rm g}$ and the energy gap Δ of the system. When Δ becomes zero, the boundary of the disordered phase (or the phase diagram) may be determined.

Because the system is spatially anisotropic, the two-point correlation functions and the correlation lengths in xy plane and in z direction are not the same. In xy plane, the two-point correlation function is given by

$$< S_{li}\cdot S_{lj} >_{xy} = \frac{\overline{s}^2}{4}\frac{1}{N}\sum_k {\rm e}^{{\rm i}k_{xy}\cdot(R_{li}^{xy} - R_{lj}^{xy})}\frac{\alpha_k - 2\beta_k}{2\omega_k}\,, \tag{14}$$

and in z direction,the two-point correlation function is given by

$$< S_{li}\cdot S_{lj} >_{z} = -\frac{\overline{s}^2}{4}\frac{1}{N}\sum_k {\rm e}^{{\rm i}k_z\cdot(R_{li}^{z} - R_{l'i}^{z})}\frac{\alpha_k - 2\beta_k}{2\omega_k}\,. \tag{15}$$

When the distance between the two points becomes large, the correlation functions in xy plane and in z direction may be expressed, respectively, as follows.

$$< S_{li} \cdot S_{lj} >_{xy} = \begin{cases} 0 \quad , & (\lambda = 0) \\ \dfrac{\overline{s}^2}{8\pi}\sqrt{\dfrac{1}{\lambda d}}\dfrac{a}{r_{xy}} \mathrm{e}^{-r_{xy}/\xi_{xy}} \, , & (\lambda \neq 0, \lambda' = 0) \\ \dfrac{\overline{s}^2}{8\pi^2}\sqrt{\dfrac{2}{\lambda' d}}\left(\dfrac{a}{r_{xy}}\right)^2 , & (\lambda \neq 0, \lambda' \neq 0, \Delta = 0) \\ \dfrac{\overline{s}^2}{8\pi^2}\sqrt{\dfrac{\pi}{\lambda' d}}\left(\dfrac{a}{\xi_{xy}}\right)^2 \left(\dfrac{r_{xy}}{\xi_{xy}}\right)^{-3/2} \mathrm{e}^{r_{xy}/\xi_{xy}} \, , & (\lambda \neq 0, \lambda' \neq 0, \Delta \neq 0) \end{cases} \tag{16}$$

$$< S_{li} \cdot S_{l'i} >_{z} = \begin{cases} 0 \quad , & (\lambda' = 0) \\ -\dfrac{\overline{s}^2}{4}\sqrt{\dfrac{1}{\pi\lambda' d}}\sqrt{\dfrac{\xi_z}{r_z}} \, \mathrm{e}^{-r_z/\xi_z} \, , & (\lambda = 0, \lambda' \neq 0) \\ -\dfrac{\overline{s}^2}{32\pi\lambda}\sqrt{\dfrac{\lambda'}{d}}\left(\dfrac{c'}{r_z}\right)^2 , & (\lambda \neq 0, \lambda' \neq 0, \Delta = 0) \\ -\dfrac{\overline{s}^2}{16\pi^2\lambda}\sqrt{\dfrac{\pi\lambda'}{d}}\left(\dfrac{c'}{\xi_z}\right)^{5/2}\left(\dfrac{r_z}{\xi_z}\right)^{-3/2} \mathrm{e}^{-r_z/\xi_z} \, , & (\lambda \neq 0, \lambda' \neq 0, \Delta \neq 0) \end{cases} \tag{17}$$

where $d = \overline{s}/(1/4 - \mu/J)$, $r_{xy} = \mid R^{xy}_{li} - R^{xy}_{lj} \mid$, and $r_z = \mid R^{z}_{li} - R^{z}_{l'i} \mid$; ξ_{xy} and ξ_z are correlation lengths in the xy plane and in the z direction, respectively. They are defined as

$$\xi_{xy} = \sqrt{\frac{\lambda d}{1 - (4\lambda + \lambda')d}} \, a \, , \tag{18}$$

$$\xi_z = \sqrt{\frac{\lambda' d}{2[1 - (4\lambda + \lambda')d]}} \, c' \, . \tag{19}$$

III. CALCULATED RESULTS AND DISCUSSION

A. Order-disorder phase diagram

Form Eqs.(9) and (10), we find that the energy gap in the triplet excited spectrum appears at the position of $(-\pi, -\pi, \pi)$ in Brillouin zone. Because of the interaction between dimers($\lambda \neq 0$ or $\lambda' \neq 0$), the triplets form an energy band, the bottom of which is at the position of $(-\pi, -\pi, \pi)$, and the energy difference between the bottom and the condensed singlets is the energy gap Δ. When Δ becomes zero, the ordering of spins may appear at 0 K, therefore we can draw the diagram of order-disorder phase transition, as shown in Fig.2.

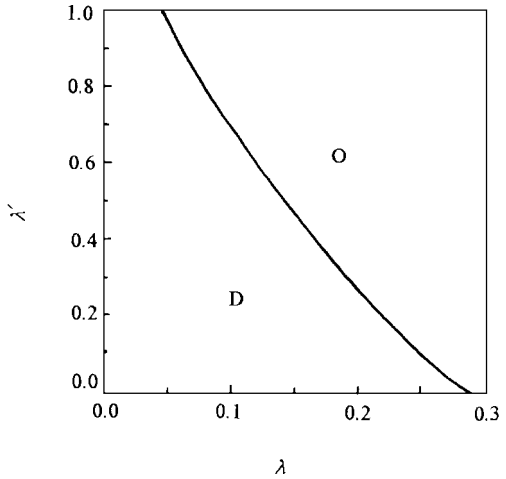

Fig.2. Diagram for order-disorder phase transition. D: disordered phase; O: ordered phase.

As for a system composed of double layers ($\lambda' = 0$), in order to guarantee the disordered phase to be stable the interaction between dimers must be smaller than a critical value, λ_c=0.292. By other methods, such as the Schwinger boson mean-field theory, λ_c= 0.223 was obtained[10]; in modified spin-wave theory, λ_c =0.236 was obtained[11]; by series expansion method, λ_c =0.391 was obtained[12]; and by quantum Monte Carlo simulation, λ_c=0.398 was obtained.[13]

As for the 1D dimer system ($\lambda = 0$), we find that the system is always in disordered phase, and there is not a critical value for λ'. This result is also found in spin ladder.[9] When λ'=1, the system becomes a homogeneous 1D spin chain, which remains in a spin-liquid state without an energy gap. Since we characterise the disordered phase with non-zero energy gap in the present method, the finite energy gap we obtained for λ'=1 does not violate the known result. What is worth mentioning is that the concept of dimer has lost its original meaning when λ'=1. However, this does not prevent the present method from being applied in this limit case, only the approximation assumption should be modified. In fact, the bond-operator method of spins is equivelent to the Schwinger boson theory.

Now we take four special cases to illustrate the calculations of the energy gap, ground state energy, correlation length of the system in the disordered phase: (1) λ'=0 indicates a 2D dimer system with λ_c= 0.292; (2)$\lambda' = 0.1\lambda$, a quasi-2D dimer system with λ_c = 0.272; (3)$\lambda' = \lambda$, a 3D dimer system with λ_c = 0.213; (4)$\lambda' = 10\lambda$, a quasi-1D dimer system with λ_c=0.079.

B. Energy gap

As shown in Fig.3, when $\lambda = \lambda' = 0$, the dimers are independent of each other, thus the energy difference between the triplet and the singlet is naturally J. When the values of λ and λ' increase, that is, the interaction between dimers increases, the triplets form an en-

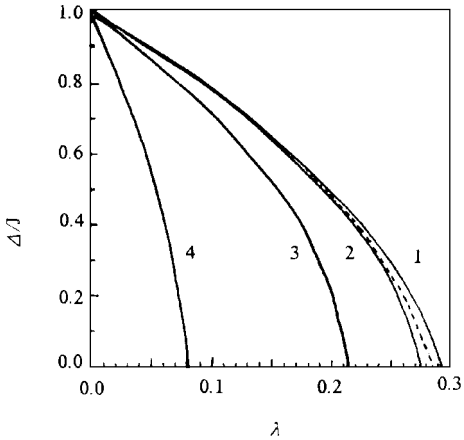

Fig.3. Energy gap as a function of $\lambda(\lambda')$. $1:\ \lambda'=0;\ 2:\lambda'=0.1\lambda; 3:\lambda'=\lambda; 4:\lambda'=10\lambda$. The dashed line is the result by taking into account the H_2 term for the case 1.

ergy band by renormalization, the energy gap between the energy band and the condenced singlets decreases, consequently the stronger the coupling between dimers, the faster the energy gap decreases.

C. Ground state energy

The ground state energy per dimer as a function of $\lambda(\lambda')$ is given in Fig.4. When $\lambda=\lambda'=0$, two spins in a dimer form a singlet with antiparallel spin directions, the energy is $-3J/4$. When the values of λ and λ' increase, the spins in a dimer interact with other spins in the nearest-neighbor dimers, which produces a part of minus energy, so that the ground state energy decreases. From Fig.4 we can also see that the ground state energies for the two cases of $\lambda'=0.1\lambda$ and $\lambda'=0$ are almost the same for a given λ, indicating that the ground state energy is in weak dependence on λ' when λ' is small. From the dimer structure in Fig.1 we may understand this result. In xy plane, every dimer interacts with four nearest-neighbor dimers, the total interacting strength is 8λ; in the z direction, the interacting strength is only $2\lambda'$. Thus when λ' is small, the ground state energy is determined almost by the horizontal coupling strength λ. When $\lambda'=\lambda$, the behavior of the ground state energy is somewhat different from the case $\lambda'=0$, and when λ' further increases ($\lambda'=10\lambda$), the difference between them becomes much bigger.

The ground state energy at the critical point for the 2D dimer system ($\lambda'=0$) is-0.778 J, and it is-0.742 J in modified spin-wave theory.[11]

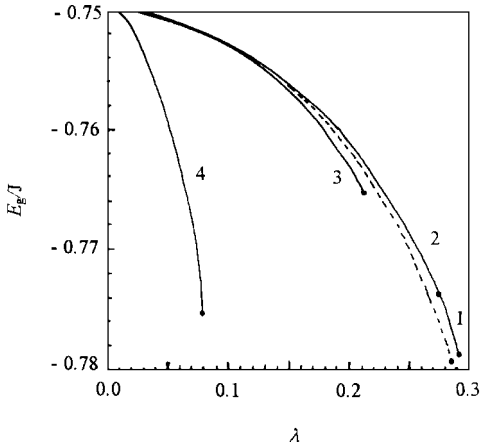

Fig.4. Ground state energy as a function of $\lambda(\lambda')$. The dashed line is the result by taking into account the H_2 term for the case 1.

D. Correlation length

We give the correlation lengths in xy plane and in z direction as functions of λ in Figs.5 and 6, respectively. When $\lambda = \lambda'$=0, dimers are independent of each other, the correlation lengths are naturally zero. When the values of λ and λ' increase, the correlation lengths increase too, and they become infinite at the critical points, indicating that the magnetically ordered phase begins to appear in the system. By modified spin-wave theory, Hida gave a result that the correlation length was zero in disordered phase for the 2D dimer system, so he conjectured that the disordered phase was a spin-gas phase.[11] But by series expansion

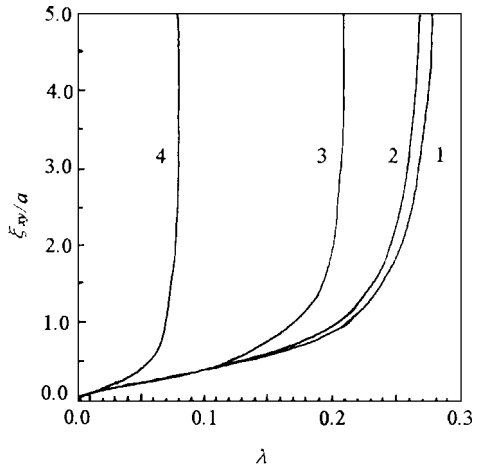

Fig.5. Correlation length in the xy horizontal plane as a function of $\lambda(\lambda')$.

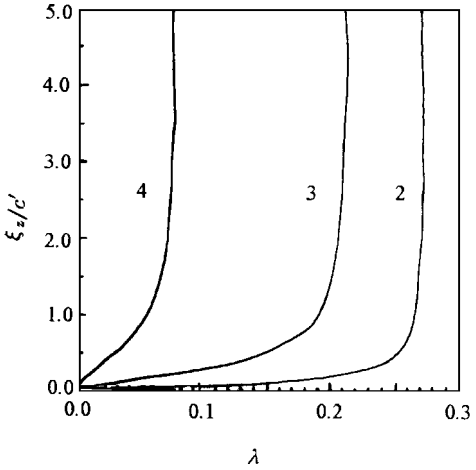

Fig.6. Correlation length in the z direction as a function of $\lambda(\lambda')$.

method, Hila also found the correlation length was finite in the disordered phase[12], which

is in agreement with our results, so the disordered phase is a spin-liquid phase rather than a spin-gas phase.

E. Effect of the H_2 term

In the above calculation, we have omitted the $H_2(H_2')$term in the Hamiltonian (2). In the calculation of the energy gap in spin ladder, Gopalan *et al.*[9] inspected the effect of H_2 term, and found that the influence of it on the energy gap is small. Through investigating two limit cases, the 1D($\lambda = 0$) and 2D($\lambda' = 0$) dimer systems, we also want to inspect the effect of H_2 term on the present system.

When H_2 term is considered, in the same approximation, we can obtain a set of new equations.[9] After solving the equations consistently, we can obtain the energy gap and the ground state energy as functions of λ and λ'. For the 2D case, we give these results in Figs.3 and 4 with a dashed line, respectively. We find that in the 2D case, the critical value λ_c changes from 0.292 to 0.286 after considering H_2 term, and the ground state energy at the critical points changes from-0.778 to-0.779 J. For the 1D case, there is still no critical value for λ' after considering H_2 term, when λ'=0.5, the energy gap changes from 0.728 to 0.726 J, and the ground state energy changes from-0.758 to-0.759 J; whenλ'=1, the energy gap changes from 0.404 to 0.392 J, and the ground state energy changes from-0.788 to-0.792 J.

Therefore, we may conclude that when λ and λ' are not too large($\lambda < \lambda_c, \lambda' < 1$), the H_2 term will influence quantitatively, the physical quantities in the disordered phase, and this influence is small.

IV. CONCLUSION

Using he bond-operator method of spins, we have studied the 3D Heisenberg antiferromagnetic dimer system, and given the diagram of order-disorder phase transition. In 2D, quasi-2D, 3D and quasi-1D dimer structural cases, we have calculated the energy gap, ground state energy, two-point correlation functions and correlation lengths in horizontal plane and in perpendicular direction as functions of the coupling strengths between dimers in the disordered phase, and discussed the variations of these physical quantities, and come to the conclusion that the disordered phase is spin-liquid phase. Finally we have also discussed the approximation adopted in this paper.

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