Magnetic order and finite-temperature properties of the two-dimensional frustrated Heisenberg model

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Ground-state and finite-temperature properties of the square-lattice $S=1/2$ Heisenberg model with antiferromagnetic nearest- and next-nearest-neighbor interactions (J_1-J_2 model) are investigated by a spin-rotation-invariant Green's-function theory, where a reasonable agreement with numerical diagonalization data is found. The quantum phase transitions from the Néel and collinear phases into a spin-liquid phase are obtained at $(J_2/J_1)_{c_1}=0.24$ and $(J_2/J_1)_{c_2}=0.83$, respectively, which considerably improves the results by a previous similar approach. The low-temperature magnetic susceptibility at $J_2/J_1 \lesssim 0.5$, the temperature of the susceptibility maximum, and the antiferromagnetic correlation length are found to decrease with increasing frustration. For high-T_c cuprates the relationship between the t-J model and an effective J_1-J_2 model is analyzed.

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I. INTRODUCTION

The study of frustration effects in low-dimensional quantum spin systems is of growing interest, which is mainly due to the availability of new materials. A substantial degree of magnetic frustration exists, for example, in the quasi-one-dimensional (1D) edge-sharing cuprates[1] (e.g., Li_2CuO_2 and $CuGeO_3$) and vanadates[2] (e.g., $MgVO_3$) and in the magnetic salt $(VO)_2P_2O_7$.[3] Frustration plays a role as well in high-T_c cuprates, where its degree, e.g., in La_2CuO_4 is about 5–8 % (Ref. 4) and hole doping may create an effective frustration.[5]

As a generic model with frustration, the square-lattice $S=1/2$ Heisenberg model with antiferromagnetic (AFM) couplings J_1 and J_2 between nearest neighbors (NN) and next-nearest neighbors (NNN), respectively, often referred to as the J_1-J_2 model,

$$H=J_1\left[\sum_{\langle ij\rangle}\vec{S}_i\vec{S}_j+R\sum_{[ij]}\vec{S}_i\vec{S}_j\right], \tag{1}$$

was widely investigated. Here, $R=J_2/J_1$ and $\langle ij\rangle$ ($[ij]$) denote NN (NNN) bonds.

At zero temperature there occur two phases with magnetic long-range order (LRO), the Néel phase for $R<R_{c_1}$ and the collinear phase for $R>R_{c_2}$, and a spin-liquid phase with only short-range order (SRO) in an intermediate region, $R_{c_1}<R<R_{c_2}$, in the vicinity of $R=0.5$. Much work was devoted to the determination of the quantum critical points $R_{c_{1,2}}$ and to the nature of the spin-liquid state, which is not yet clarified.[6–9] Recent diagrammatic[6] and series-expansion approaches[6,10] yield $R_{c_1}\simeq 0.4$ and $R_{c_2}\simeq 0.6$ being in accord with the results of previous theories,[11–13] where R_{c_1} nearly agrees with the value obtained by the variational approach[11] and linear spin-wave theory[12] and R_{c_2} nearly agrees with the critical point found by a modified second-order spin-wave theory[13] (for references and discussions of most of the early work, see Refs. 14 and 15). Those analytical results are corroborated by exact-diagonalization (ED) studies on lattices up to 36 sites supplemented by finite-size scaling,[16] which yield $R_{c_1}=0.34$ and $R_{c_2}=0.68$.

At finite temperatures, most of the previous spin-wave theories are valid only at sufficiently low temperatures, since the temperature-dependent AFM SRO is not adequately taken into account. First steps towards an improved description of SRO in the model (1) at arbitrary temperatures were made by a spin-rotation-invariant Green's-function theory[15] based on an approach first proposed by Shimahara and Takada[17] and a modified spin-wave-theory extended by a mean-field decoupling of the spin-wave interaction.[18] However, in Ref. 15 the window for the spin-liquid phase was found to be too broad, and in Ref. 18 the $T=0$ limit was not considered in detail.

In this paper we improve the rotationally symmetric Green's-function theory (formulated in terms of the projection method), as compared with Ref. 15, by another choice of the free-vertex parameter using more input information. We focus on the quantum phase transitions and the temperature dependence of the uniform static spin susceptibility. Moreover, we discuss in detail a possible equivalence of frustration and doping, as conjectured in Ref. 5, and examine the influence of frustration on the AFM correlation length. Our improved approach outlined in Sec. II results, as compared with Ref. 15, in considerable differences with respect to the position of the quantum critical points (see Sec. III), and the frustration and temperature dependence of the magnetic sus-

ceptibility (Sec. IV). On the other hand, our results are in much better agreement with Ref. 16 and with the finite-temperature ED data of Ref. 19.

II. THEORY OF SPIN SUSCEPTIBILITY

The spin susceptibility $\chi_{\vec{q}}^{+-}(\omega)=-\langle\langle S_{\vec{q}}^{+};S_{-\vec{q}}^{-}\rangle\rangle_{\omega}$ ($\langle\langle\ldots;\ldots\rangle\rangle_{\omega}$ denotes the two-time retarded commutator Green's function), determined by the projection method in a generalized-mean-field approximation,[20,21] is given by

$$\chi_{\vec{q}}^{+-}(\omega)=-\frac{M_{\vec{q}}^{(1)}}{\omega^2-\omega_{\vec{q}}^2} \tag{2}$$

with

$$M_{\vec{q}}^{(1)}=-8J_1C_{1,0}(1-\gamma_{\vec{q}})-8J_2C_{1,1}(1-\gamma'_{\vec{q}}), \tag{3}$$

$C_{n,m}\equiv C_{\vec{r}}=\langle S_0^+S_{\vec{r}}^-\rangle$, $\vec{r}=n\vec{e}_x+m\vec{e}_y$, $\gamma_{\vec{q}}=(\cos q_x+\cos q_y)/2$, and $\gamma'_{\vec{q}}=\cos q_x\cos q_y$. The spin-correlation functions are calculated from

$$C_{\vec{r}}=\frac{1}{N}\sum_{\vec{q}}\frac{M_{\vec{q}}^{(1)}}{2\omega_{\vec{q}}}[1+2p(\omega_{\vec{q}})]e^{i\vec{q}\vec{r}}, \tag{4}$$

where $p(\omega_{\vec{q}})=(e^{\omega_{\vec{q}}/T}-1)^{-1}$. The NN correlators are related to the internal energy per site $\varepsilon=3(J_1C_{1,0}+J_2C_{1,1})$.

The spectrum $\omega_{\vec{q}}$ is obtained in the approximation $-\ddot{S}_{\vec{q}}^{+}=\omega_{\vec{q}}^2S_{\vec{q}}^{+}$, where products of three spin operators on different sites occurring in $-\ddot{S}_i^+$ are decoupled introducing vertex parameters $\alpha_i(i=1,2,3)$ as in Refs. 17 and 15. That is, α_1 is attached to $C_{1,0}$; α_2 to $C_{2,0}$, $C_{2,1}$, and $C_{2,2}$; and α_3 is associated with $C_{1,1}$. For $J_2=0$ or $J_1=0$ we take $\alpha_3=\alpha_2$ or $\alpha_3=\alpha_1$, respectively, because for $J_1=0$ we have two noninteracting mutually penetrating Néel sublattices (rotated by $\pi/4$) being equivalent to the complete lattice for $J_2=0$. We obtain

$$\begin{aligned}\omega_{\vec{q}}^2=&2J_1^2(1-\gamma_{\vec{q}})(1-2\alpha_1C_{1,0}+2\alpha_2C_{2,0}+4\alpha_3C_{1,1}\\&-8\alpha_1C_{1,0}\gamma_{\vec{q}})+2J_2^2(1-\gamma'_{\vec{q}})[1-2\alpha_3C_{1,1}\\&+2\alpha_2(C_{2,2}+2C_{2,0})-8\alpha_3C_{1,1}\gamma'_{\vec{q}}]\\&+8J_1J_2\{(1-\gamma_{\vec{q}})(\alpha_2C_{1,2}-\alpha_1C_{1,0})+(1-\gamma'_{\vec{q}})\\&\times[3\alpha_1C_{1,0}+\alpha_2C_{1,2}-2(\alpha_1C_{1,0}+\alpha_3C_{1,1})\gamma_{\vec{q}}]\},\end{aligned} \tag{5}$$

which agrees with the result of the Green's-function decoupling scheme of Ref. 15. In the limiting cases, by the choice of α_3 we have $(\omega_{\vec{q}'}/J_2)^2(J_1=0)=(\omega_{\vec{q}}/J_1)^2(J_2=0)$ with $\vec{q}'$ lying in the rotated frame $[q'_{x,y}=\frac{1}{2}(\pm q_x+q_y);\ \gamma'_{\vec{q}'}=\gamma_{\vec{q}}]$, since $C_{1,1}(J_1=0)=C_{1,0}(J_2=0)$, $C_{2,2}(J_1=0)=C_{2,0}(J_2=0)$, and $C_{2,0}(J_1=0)=C_{1,1}(J_2=0)$. Note that the theory preserves the rotational symmetry in spin space, that is $\chi_{\vec{q}}^{zz}(\omega)\equiv\chi_{\vec{q}}(\omega)=\frac{1}{2}\chi_{\vec{q}}^{+-}(\omega)$.[20,21]

The magnetic LRO phases in model (1) are reflected by the closure of the spectrum gap at $\vec{Q}_0$ as $T\to0$, so that $\lim_{T\to0}\chi_{\vec{Q}_0}^{-1}$, where $\vec{Q}_0=(\pi,\pi)$ in the Néel phase (I) for $R<R_{c_1}$, and $\vec{Q}_0=\vec{Q}_i(i=1,2)$ with $\vec{Q}_1=(\pi,0)$ and $\vec{Q}_2=(0,\pi)$ in the collinear phase (II) for $R>R_{c_2}$. In phase I, the condensation part $C_{\rm I}e^{i\vec{Q}_0\vec{r}}$ is separated from $C_{\vec{r}}$, and the magnetization m is calculated as in Refs. 20 and 21, where $m^2=\frac{3}{2}C_{\rm I}$. In phase II the correlators are calculated as $C_{\vec{r}}=(C_{\vec{r},1}+C_{\vec{r},2})/2$ preserving the square symmetry (e.g., $C_{1,0}=C_{0,1}$), where

$$C_{\vec{r},i}=\frac{1}{N}\sum_{\vec{q}(\neq\vec{Q}_i)}\frac{M_{\vec{q}}^{(1)}}{2\omega_{\vec{q}}}e^{i\vec{q}\vec{r}}+C_{\rm II}e^{i\vec{Q}_i\vec{r}}. \tag{6}$$

Then the magnetization m is obtained from

$$m^2=\frac{3}{2N}\sum_{\vec{r}}C_{\vec{r},i}e^{-i\vec{Q}_i\vec{r}}=\frac{3}{2}C_{\rm II}. \tag{7}$$

Note that in the limiting cases we have $m(J_1=0)=m(J_2=0)$.[15,16]

Concerning the vertex parameters in our self-consistency scheme, $\alpha_1(T;R)$ is fixed by the sum rule $C_{0,0}=1/2$. To determine the free parameter $\alpha_2(T;R)$, we may adopt different choices. In Ref. 15, $\alpha_2(0,0)$ is fit to $m(T=0;R=0)=0.3$, and the ratio $(\alpha_2-1)/(\alpha_1-1)$ is assumed to be temperature and frustration independent. To obtain more realistic results, in this paper we adjust $\alpha_2(T;R)$ to the ED data for $\varepsilon(T;R)$ available at $T=0$ (Ref. 16) and at finite temperatures and some R values in terms of the specific heat C_v [$\varepsilon(T;R)=\varepsilon(0;R)+\int_0^T dT'C_v(T';R)$].[19] Finally, the parameter α_3 is interpolated between the limiting values at $J_2=0$ and $J_1=0$ (see above). To provide a stable self-consistent numerical solution at given $\varepsilon(T;R)$, we choose $\alpha_3=(1+R)^{-1}e^{-R}\alpha_2+R(1+R)^{-1}\alpha_1$, which yields for most R values nearly the same results as the linear interpolation (e^{-R} replaced by 1) used in Ref. 15.

III. GROUND-STATE PROPERTIES

As seen in Fig. 1, our approach yields second-order quantum phase transitions at $R_{c_1}=0.24$ and $R_{c_2}=0.83$ from the Néel and collinear phases, respectively, into a spin-liquid phase. Let us emphasize that, as compared with the analogous approach of Ref. 15 ($R_{c_1}=0.11$, $R_{c_2}=1.70$), the more realistic choice of the free parameter α_2 (see Sec. II) leads to an appreciable stabilization of LRO. However, as compared with the ED results[16] (cf. Fig. 1) and the recent diagrammatic[6] and series-expansion approaches[6,10] ($R_{c_1}\simeq0.4$, $R_{c_2}\simeq0.6$), our theory overestimates the effects of frustration in destroying the magnetic LRO. Note from Fig. 1 that our calculation taking $\varepsilon(T;R)$ as input underestimates the magnetization m for very small or very large R values ($R\ll1$ or $R\to\infty$) in variation with Ref. 15, where $m(0;0)$ is taken as input.

Let us compare the influence of hole doping and frustration on the Néel order in high-T_c cuprates in relation to the conjecture[5] about the equivalence of doping and frustration. In Ref. 5 an effective AFM J_1-J_2 model was derived from an

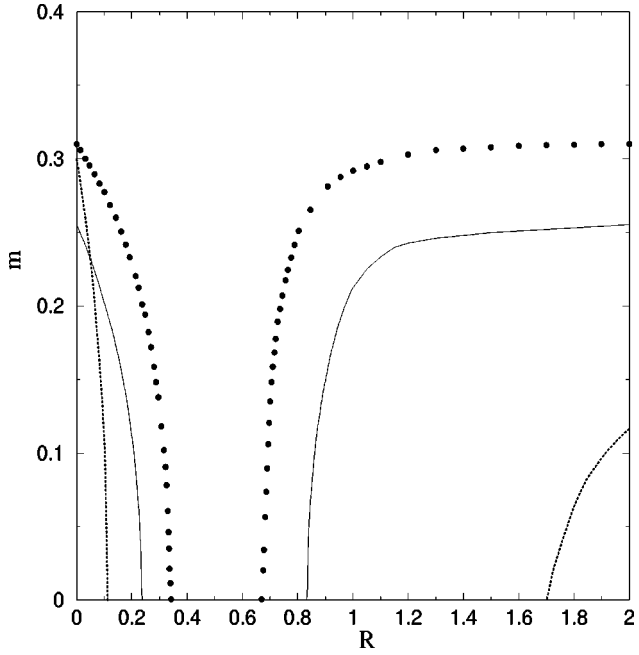

FIG. 1. Magnetization as a function of the frustration parameter $R=J_2/J_1$ at $T=0$ compared with ED data (●, Ref. 16) and the results of Ref. 15 (dotted line).

extended t-J model, where $R=(\delta/2)[1-(\{U/t\}+5)\delta]^{-1}$ (δ is the hole concentration, U and t are the Hubbard-model parameters). Taking $R_c \equiv R_{c_1}=0.4$ (Refs. 6 and 10) and the realistic values $U/t=8$ and $U/t=10$, we get the critical doping for the destruction of Néel order $\delta_c=0.07$ and $\delta_c=0.06$, respectively. This is consistent with experiments and in agreement with $\delta_c=0.06$ in the t-J model (see Ref. 22; $J/t=0.4$). Note that for our value of $R_c=0.24$ nearly the same critical dopings are obtained. That is, concerning the stabilization of Néel order the doping-frustration analogy may be invoked.

In Fig. 2 the spin-correlation functions up to the fifth-nearest neighbors are compared with available ED data,[23] where a reasonable agreement is found. For $R\lesssim 0.4$ the magnitudes of all correlators decrease with increasing frustration and are smaller than the corresponding ED results, which is in accord with the reduced values for m as compared with the ED data of Ref. 16 (cf. Fig. 1). As can be seen, the values of C_r^- at $R=0$ and $R=2$ are compatible with the exact relations $C_{1,1}(J_1=0)=C_{1,0}(J_2=0)$, etc. [see below Eq. (5)]. Comparing the sign changes and nonmonotonous R dependences of some correlators with series-expansion results,[10] we get $C_{1,1}=0$ at $R\simeq 0.5$ and a minimum of $C_{2,0}>0$ at $R\simeq 0.6$ contrary to Ref. 10, where $C_{1,1}$ and $C_{2,0}$ are found to vanish in the spin-liquid phase $0.4\lesssim R\lesssim 0.6$.

To check a possible equivalence of frustration and doping effects on the AFM SRO, let us compare the decrease of $|C_{1,0}|$ and $C_{1,1}$ with increasing R and δ in the effective J_1-J_2 model and the t-J model,[22] respectively. In both models $|C_{1,0}|$ decreases from $R=0(\delta=0)$ to $R=0.4$ corresponding to $\delta=0.06$ (see above) by about 20%. For the decrease of $C_{1,1}$ in the J_1-J_2 model at $R=0.4$ we obtain about 70% (the ED data[23] yields 35%; cf. Fig. 2), whereas in the t-J model a

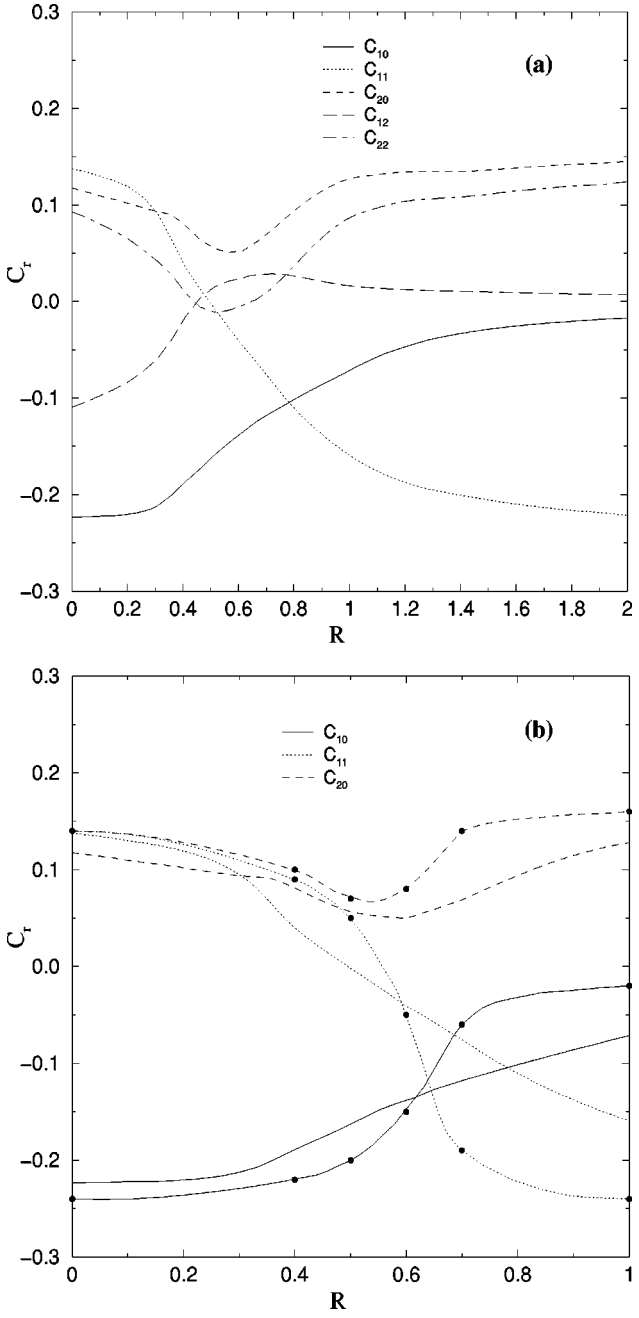

FIG. 2. Spin-correlation functions vs frustration R at $T=0$ (a) compared with the ED results (●) of Ref. 23 (b) joined by the corresponding line style.

decrease of about 40% was obtained. Note, however, that the curvatures of the functions $|C_r^-|$ ($R<0.4$) and $|C_r^-|(\delta)$ are different (convex in the J_1-J_2 model and concave in the t-J model). Altogether, the reduction of SRO by small doping in the t-J model may be roughly simulated by frustration in the J_1-J_2 model. Let us point out that for large-doping levels such an analogy does not hold anymore, since the effective J_1-J_2 model of Ref. 5 is only valid for $\delta\lesssim 0.08$ ($J_1>0$ for $U/t=8$).

Concerning the nature of the intermediate phase ($0.4\lesssim R\lesssim 0.6$) with spontaneously broken translation symmetry[6–9] (spin-dimer order, such as the plaquette-resonating valence-

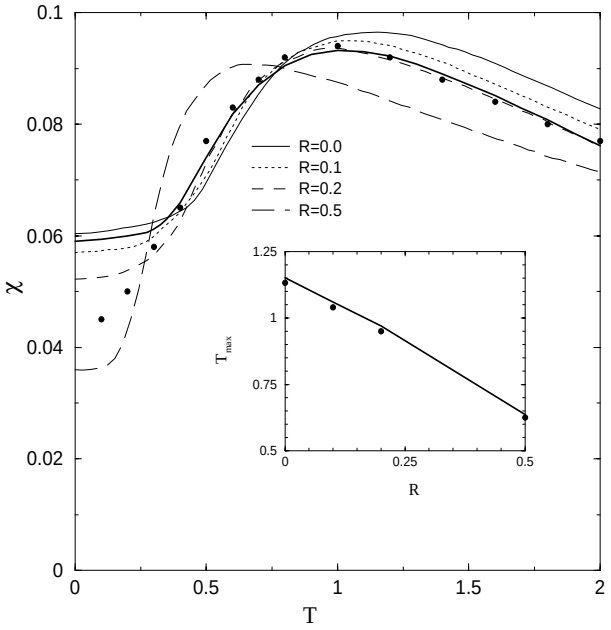

FIG. 3. Uniform static spin susceptibility vs T for different frustration parameters ($J_1=1$). The result for $R=0$ is compared with that obtained by taking the Monte Carlo (MC) data for the internal energy (Ref. 24) as input (thick solid line) and with MC results (●, Ref. 24). The inset shows the position $T_{\max}$ of the maximum in χ vs R together with the ED data (●) of Ref. 19.

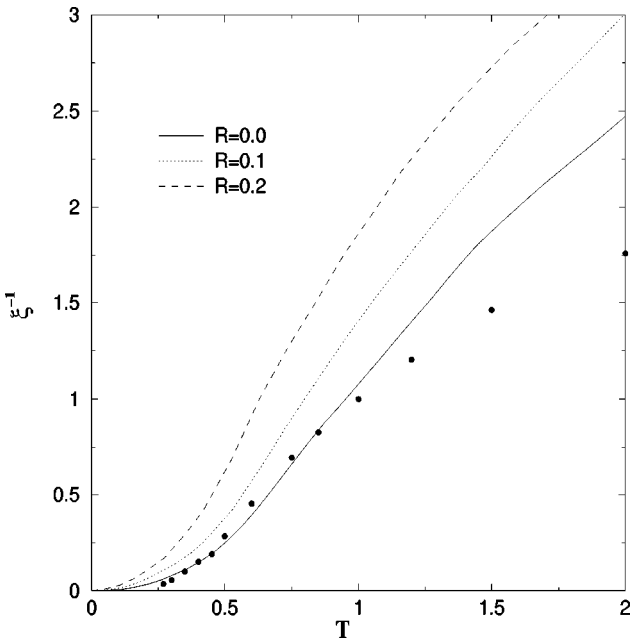

FIG. 4. Inverse antiferromagnetic correlation length vs T for different frustrations ($J_1=1$). The $R=0$ result is compared with the MC data (●) of Ref. 26.

bond state), our present approach cannot probe this spin structure. To this end, the Green's-function theory has to be extended by the introduction of sublattices and of appropriate dimer order parameters, which may be left for further studies.

IV. FINITE-TEMPERATURE RESULTS

At nonzero temperatures ($m=0$) we have solved the self-consistency equations (4), where $\alpha_2(T;R)$ is fit to the ED data for $\varepsilon(T;R)$,[19] to obtain the static-spin susceptibility and the correlation length.

Our results for the uniform static susceptibility $\chi(T;R) \equiv \chi_0(0)$ are depicted in Fig. 3. To judge the quality of our approach for $R=0$ and to illustrate the uncertainty caused by the input of numerical data for ε, we have also calculated $\chi(T;0)$ adjusting α_2 to the Monte Carlo data for $\varepsilon(T;0)$,[24] where the comparison of both theoretical curves with the Monte Carlo results of Ref. 24 yields a reasonable agreement. At $T=0$ and low enough temperatures the susceptibility for $R \lesssim 0.5$ decreases with increasing frustration, which qualitatively agrees with the behavior found by the extended mean-field spin-wave theory of Ref. 18 (there, the $T=0$ values of χ are not given) and ED studies of finite systems[19] [yielding $\chi(T=0)=0$)]. On the other hand, our results are in contradiction to Ref. 15, where the low-temperature susceptibility was found to increase with increasing R, which may be due to the simplified determination of α_2 (see Sec. II). The increase of χ with temperature, the pronounced maximum at $T_{\max}$ near the exchange energy J_1 (see inset), and the crossover to the high-temperature Curie-Weiss behavior are due to the decrease of AFM SRO with increasing temperature. Note that the susceptibility maximum found in Ref. 15 is smoothed out at $R \gtrsim 0.3$. As shown in the inset, $T_{\max}$ decreases with increasing R in very good agreement with the ED results of Ref. 19. In the approach of Ref. 18 the same tendency was found, however, the values of $T_{\max}$ are remarkably higher than the ED data.

Comparing the influence of frustration and doping on the magnetic susceptibility on the basis of the effective J_1-J_2 model with $J_1=J_{1,0}[1-(\{U/t\}+5)\delta]$ (Ref. 5) and the t-J model with $J=J_{1,0}$,[22] we consider the zero-temperature susceptibilities $\tilde{\chi}=J_{1,0}\chi$. Then the decrease of $J_1\chi$ with increasing R (see Fig. 3) corresponds to an increase of $\tilde{\chi}$, where $J_1/J_{1,0}$ is expressed in terms of R (cf. Sec. III), Equally, $\tilde{\chi}$ in the t-J model increases with δ. For the relative change of $\tilde{\chi}(R)$ in the J_1-J_2 model between $R=0$ and $R=0.4$($\delta=0.06$), we obtain $[\tilde{\chi}(0.4)-\tilde{\chi}(0)]/\tilde{\chi}(0)=4.7$, which exceeds the corresponding change in the t-J model calculated by the theory of Ref. 22 by a factor of about 30. Moreover, the maximum of $\tilde{\chi}$ in the t-J model at $\delta_{\max} \simeq 0.3$ cannot be described by a pure spin model. Our results on the spin susceptibility show that there is no general equivalence of doping and frustration. This is in accord with the conclusions of Ref. 25, where some dynamic properties are shown to behave quite differently in the t-J and J_1-J_2 models.

Considering the AFM correlation length $\xi(T;R)$ for $R < R_{c_1}$, the expansion of $\chi_{\vec{q}}(0)$ around $\vec{Q}_0=(\pi,\pi)$, $\chi_{\vec{q}}(0) = \chi_{\vec{Q}_0}(0)(1+\xi^2 k^2)^{-1}$ with $\vec{k}=\vec{q}-\vec{Q}_0$, yields

$$\xi^2=\frac{C_{1,0}-2RC_{1,1}}{8C_{1,0}}-\left(\frac{J_1}{\omega_{\vec{Q}_0}}\right)^2\left\{\frac{1}{2}+11\alpha_1C_{1,0}+\alpha_2C_{2,0}+2\alpha_3C_{1,1}-2R(11\alpha_1C_{1,0}+\alpha_2C_{1,2}+4\alpha_3C_{1,1})-R^2[1-10\alpha_3C_{1,1}+2\alpha_2(C_{2,2}+2C_{2,0})]\right\}. \quad (8)$$

In Fig. 4 the influence of frustration on the temperature dependence of ξ^{-1} is shown. For $R=0$ and $T\lesssim 1$ we obtain a good agreement with the Monte Carlo data of Ref. 26. For $R>0$ there are, to our knowledge, no previous results to compare with. The decrease of ξ with increasing frustration, i.e., the stronger spatial decay of the spin correlators, corresponds to our physical expectation.

V. SUMMARY

To summarize, the strengths of our rotationally symmetric Green's-function theory for the 2D J_1-J_2 model including an improved determination of the free-vertex parameter consist in the possibility to calculate all static magnetic properties at arbitrary temperatures, as compared with most of the previous spin-wave theories being valid only at sufficiently low temperatures. Thereby, the temperature dependence of the magnetic short-range order is well described, as can be seen from the magnetic susceptibility. The frustration effects on its temperature dependence (cf. Fig. 3) are characterized by a decrease of the low-temperature susceptibility at $R\lesssim 0.5$ and a lowering of the maximum temperature $T_{\max}$ with increasing frustration, where the values of $T_{\max}$ are in excellent agreement with the ED results of Ref. 19.

The weaknesses of our approach are visible in the ground-state properties. The approximate time evolution of spin operators, $-\ddot{S}_{\vec{q}}^{+}=\omega_{\vec{q}}^2 S_{\vec{q}}^{+}$, resulting from a decoupling of three-spin-operator products yields the critical points $R_{c_1}=0.24$ and $R_{c_2}=0.83$, which differ from the generally accepted values ($R_{c_1}\simeq 0.3-0.4$, $R_{c_2}\simeq 0.6-0.7$). On the other hand, the region of the nonmagnetic phase is considerably narrowed as compared with Ref. 15 (cf. Fig. 1). The study of the spin structure in this phase is beyond the scope of our present approach.

Analyzing the influence of hole doping in the t-J model for high-T_c cuprates as compared with that of frustration in an effective J_1-J_2 model, it is shown that in the small-doping region a doping-frustration analogy referring to the Néel order and static spin-correlation functions only may be invoked. We conclude that our approach is promising to be applied to other frustrated spin models.

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