Novel Approach to Description of Spin-Liquid Phases in Low-Dimensional Quantum Antiferromagnets

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We consider quantum spin systems with dimerization, which at strong coupling have singlet ground states. To account for strong correlations, the $S = 1$ elementary excitations are described as a dilute Bose gas with infinite on-site repulsion. This approach is applied to the two-layer Heisenberg antiferromagnet at $T = 0$ with general couplings. Our analytic results for the triplet gap, the excitation spectrum, and the location of the quantum critical point are in excellent agreement with numerical results obtained by dimer series expansions. [S0031-9007(98)06407-2]

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One of the most challenging problems of quantum magnetism is the description of transitions between phases with spontaneously broken symmetry and disordered (spin liquid) phases. The properties of the disordered phases are also of great interest [1].

A variety of quantum spin models has been introduced in connection with the high-T_c cuprates and other recently discovered compounds. Examples include the Heisenberg ladder [2], the two-layer Heisenberg model [3], and 2D square lattice models with dimerization [4]. In all of the above the Hamiltonian favors singlet formation of the spins between the chains (layers) or on neighboring sites. For this class of models the disordered phase is relatively well understood, since the lowest excitation above the singlet is a massive triplet. Another example is the CaV_4O_9 lattice [5], where the spins form a singlet state on a plaquette. There also have been suggestions that dimerization of different kinds may occur in the J_1-J_2 model [6,7].

All of the models mentioned above, except for the ladder, exhibit a quantum phase transition from a disordered dimer phase to a collinear Néel phase with long range order in the ground state as the dimerization decreases. This transition occurs due to competition between singlet formation and antiferromagnetic order. A useful approach to the description of the disordered phase is the bond operator representation for spins, introduced by Chubukov [8] and Sachdev and Bhatt [6]. This representation can be considered as the analog of the usual Holstein-Primakoff transformation for phases with unbroken spin rotational symmetry. Let us consider two $S = 1/2$ spins $\vec{S}_1$, $\vec{S}_2$ and introduce operators for creation of a singlet $s^\dagger|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ and three triplet states $t^\dagger_\alpha, \alpha = x, y, z$ above a fictitious vacuum state $|0\rangle$: $t^\dagger_x|0\rangle = -\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle)$, $t^\dagger_y|0\rangle = \frac{i}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle)$, $t^\dagger_z|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$. Then the following representation is exact [6]:

$$S^\alpha_{1,2} = \frac{1}{2}(\pm s^\dagger t_\alpha \pm t^\dagger_\alpha s - i\epsilon_{\alpha\beta\gamma} t^\dagger_\beta t_\gamma)\,. \qquad (1)$$

The four operators satisfy the usual bosonic commutation relations. In order to ensure that the physical states are either singlets or triplets one has to impose the condition: $s^\dagger s + t^\dagger_\alpha t_\alpha = 1$. For a lattice spin system, the constraint is typically taken into account in a mean-field fashion, i.e., it is not strictly satisfied on every site, but only on average [6]. A slightly different representation can be obtained by choosing the singlet as the ground state. Then Eq. (1) is still valid, but the operator s has the form $s = \sqrt{1 - t^\dagger_a t_a}$, which formally is the resolution of the constraint [3,8]. Again, the form of s ensures that only physical states are present. However, it is very difficult to take the s term into account due to its nonlinear nature. Expansions of the square root to infinite order have been proposed [3]. Unfortunately, there is no small parameter in this expansion and therefore the summation is ambiguous and technically complicated. Alternatively, one can use numerical techniques, based on the Gutzwiller projection method [9].

In this Letter we present an effective analytical method to deal with the hard core constraint. This approach can be applied to any model, for which the excitations in the disordered phase are triplets above a strong coupling singlet ground state. For definiteness we consider the model

$$H = J \sum_{\langle i,j\rangle} \vec{S}_{1i} \cdot \vec{S}_{1j} + \lambda J \sum_{\langle i,j\rangle} \vec{S}_{2i} \cdot \vec{S}_{2j} + J_\perp \sum_i \vec{S}_{1i} \cdot \vec{S}_{2i}\,. \qquad (2)$$

All the spins are $1/2$ and the couplings are antiferromagnetic ($J, J_\perp \geq 0$). The spins $\vec{S}_{1i}, \vec{S}_{2i}$ represent two planes of Heisenberg spins, coupled through the third term in (2). The summation in each plane is over nearest neighbors on a square lattice. In the present Letter we consider two cases: $\lambda = 1$, which corresponds to the two-layer Heisenberg model, and $\lambda = 0$, describing free spins in one of the planes. The latter model is interesting because of its connection to the Kondo lattice model (at half filling) with an additional repulsive Hubbard interaction between the conduction electrons. In the limit when the repulsion is strong, the charge degrees of freedom are frozen, while

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the spin part is described by the Heisenberg Hamiltonian, leading to (2) (at $\lambda = 0$). A simplified version of this model was introduced by Doniach [10] to study the competition between local singlet formation (the Kondo effect) and the induced magnetic (RKKY) interaction between the free spins. The early mean-field treatment of Doniach predicted a critical $J_\perp$, below which the free spins order antiferromagnetically. Recently this model was also studied numerically by Matsushita, Gelfand, and Ishii [11] who also found a finite transition point.

For $J_\perp \gg J$ interplane singlets are favored and the wave function is a product of on-site dimers. The excitations above this strong coupling ground state are triplets. In order to obtain the effective Hamiltonian for the triplets we pair the spins into interplane singlets by using (1). Alternatively, instead of applying the transformation (1), one could use perturbation theory in the "hopping" $J_\perp$ and calculate matrix elements of the type $\langle t_{\alpha i}, s_j | \vec{S}_{1i} \cdot \vec{S}_{1j} | s_i, t_{\alpha j} \rangle = \langle t_{\alpha i}, t_{\alpha j} | \vec{S}_{1i} \cdot \vec{S}_{1j} | s_i, s_j \rangle = 1/4$, $\langle t_{\alpha i}, t_{\beta j} | \vec{S}_{1i} \cdot \vec{S}_{1j} | t_{\gamma i}, t_{\delta j} \rangle = 1/4(\delta_{\alpha\delta}\delta_{\gamma\beta} - \delta_{\alpha\beta} \times \delta_{\gamma\delta})$, etc. The latter method is more useful when additional degrees of freedom are present in the problem, e.g., holes. For a start we neglect the constraint completely [i.e., formally set $s = 1$ in (1)] and obtain the effective Hamiltonian

$$H = H_2 + H_3 + H_4, \tag{3}$$

$$H_2 = \sum_{\mathbf{k},\alpha} A_{\mathbf{k}} t^\dagger_{\mathbf{k}\alpha} t_{\mathbf{k}\alpha} + \frac{B_{\mathbf{k}}}{2} (t^\dagger_{\mathbf{k}\alpha} t^\dagger_{-\mathbf{k}\alpha} + \text{H.c.}), \tag{4}$$

$$H_3 = \frac{(\lambda - 1)J}{4} \sum_{\langle i,j\rangle,\alpha\beta\gamma} \{[i\epsilon_{\alpha\beta\gamma} t^\dagger_{\alpha i} t^\dagger_{\beta j} t_{\gamma j} + \text{H.c.}] + [i \leftrightarrow j]\}, \tag{5}$$

$$H_4 = \frac{(1 + \lambda)J}{4} \sum_{\langle i,j\rangle,\alpha\beta} \{t^\dagger_{\alpha i} t^\dagger_{\beta j} t_{\beta i} t_{\alpha j} - t^\dagger_{\alpha i} t^\dagger_{\alpha j} t_{\beta i} t_{\beta j}\}. \tag{6}$$

The coefficients in (4) are $A_{\mathbf{k}} = J_\perp + (1 + \lambda)J\xi_{\mathbf{k}}$, $B_{\mathbf{k}} = (1 + \lambda)J\xi_{\mathbf{k}}$, where $\xi_{\mathbf{k}} = [\cos(k_x) + \cos(k_y)]/2$. By using the Bogoliubov transformation $t_{\mathbf{k}\alpha} = u_{\mathbf{k}} \tilde{t}_{\mathbf{k}\alpha} + v_{\mathbf{k}} \tilde{t}^\dagger_{-\mathbf{k}\alpha}$ we obtain for the excitation spectrum at the quadratic level (H_2 only) $\omega^2_{\mathbf{k}} = A^2_{\mathbf{k}} - B^2_{\mathbf{k}}$. The gap $\Delta = \omega_{\pi,\pi}$ is nonzero for $J_\perp > (J_\perp)_c = 2(1 + \lambda)J$ and vanishes at $(J_\perp)_c$, signaling a transition to a Néel ordered phase. The location of the critical point at this level of approximation $(J_\perp)_c = 2J$ ($\lambda = 0$), $4J$ ($\lambda = 1$) differs significantly from the recent numerical results 1.39 ([11], this Letter) and 2.54 [12], respectively. Let us mention that spin wave theory in the ordered phase works rather poorly for this problem, predicting for $\lambda = 1$, $(J_\perp)_c \approx 4.3J$ [3].

We find, in agreement with previous work [2,6], that the effect of the terms H_3 and H_4 on the spectrum is quite small and therefore cannot explain the numerical results. We treat these terms later perturbatively.

The dominant contribution to the renormalization of the spectrum comes from the constraint that only one of the triplet states can be excited on every site: $t^\dagger_{\alpha i} t^\dagger_{\beta i} = 0$. This hard-core condition can be taken into account by introducing an infinite on-site repulsion between the bosons:

$$H_U = U \sum_{i,\alpha\beta} t^\dagger_{\alpha i} t^\dagger_{\beta i} t_{\beta i} t_{\alpha i}, \qquad U \to \infty. \tag{7}$$

Since the interaction is infinite, one has to find the exact scattering amplitude for the triplets. Our treatment is similar to the one used for Fermi gas with hard core, which appears in the theory of nuclear matter and ^{3}He. The approach was initiated by Brueckner [13]. The scattering vertex $\Gamma_{\alpha\beta,\gamma\delta}(\mathbf{K})$, $\mathbf{K} \equiv (\mathbf{k}, \omega)$ in the ladder approximation satisfies the Bethe-Salpeter equation, shown in Fig. 1a. It depends on the total energy and momentum of the incoming particles $\mathbf{K} = \mathbf{K}_1 + \mathbf{K}_2$ and has the structure $\Gamma_{\alpha\beta,\gamma\delta} = \Gamma \delta_{\alpha\gamma}\delta_{\beta\delta}$. Since the interaction is local and nonretarded, the equation for Γ can be readily solved with the result

$$\Gamma(\mathbf{K}) = i\left(\int \frac{d^3Q}{(2\pi)^3} G(\mathbf{Q}) G(\mathbf{K} - \mathbf{Q}) \right)^{-1} = -\left(\frac{1}{N} \sum_{\mathbf{q}} \frac{u^2_{\mathbf{q}} u^2_{\mathbf{k}-\mathbf{q}}}{\omega - \omega_{\mathbf{q}} - \omega_{\mathbf{k}-\mathbf{q}}} + \left\{ \begin{array}{c} u \to v \\ \omega \to -\omega \end{array} \right\} \right)^{-1}. \tag{8}$$

Here $G(\mathbf{Q})$ is the normal triplet Green's function, i.e., $G(\mathbf{k}, t) = -i\langle T(t_{\mathbf{k}\alpha}(t) t^\dagger_{\mathbf{k}\alpha}(0))\rangle$ and the Bogoliubov coefficients $u^2_{\mathbf{k}}, v^2_{\mathbf{k}} = \pm 1/2 + A_{\mathbf{k}}/2\omega_{\mathbf{k}}$. The imaginary part of Γ is determined by the rule $\omega \to \omega + i\delta$.

The basic approximation made in the derivation of $\Gamma(\mathbf{K})$ is that we neglect all anomalous scattering vertices, which are present in the theory due to the existence of anomalous Green's functions, $G_a(\mathbf{k}, t) = -i\langle T(t^\dagger_{-\mathbf{k}\alpha}(t) t^\dagger_{\mathbf{k}\alpha}(0))\rangle$. We have also derived the complete set of equations by taking all vertices into account. However, our key observation is that all anomalous contributions are suppressed by an additional small parameter, present in the theory—the density of triplet excitations $n_i = \sum_\alpha \langle t^\dagger_{\alpha i} t_{\alpha i}\rangle = 3N^{-1} \sum_{\mathbf{q}} v^2_{\mathbf{q}} \approx 0.1$ at $J_\perp/J \approx 2.5$. We find that n_i is quite small throughout the disordered phase, even close to the transition point. Thus the triplet excitations behave as a dilute, strongly interacting Bose gas. Consequently, since an insertion of an anomalous Green's function into the intermediate states of the ladder in Fig. 1a brings powers of $v_{\mathbf{q}}$ into the equation for the amplitude, its contribution is small. Therefore Eq. (8) can be considered as the first term in an expansion in powers of the gas parameter n_i. To be consistent, we also neglect the second term in (8), since it contains $v_{\mathbf{q}}$.

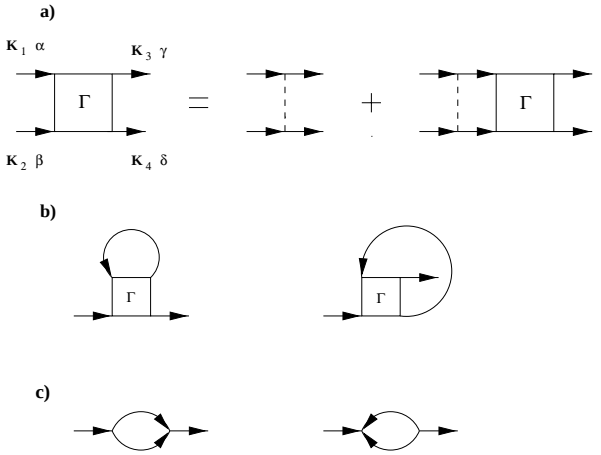

FIG. 1. (a) Equation for the scattering amplitude Γ. (b) Diagrams for the self-energy, corresponding to Γ. (c) One-loop diagrams, arising from the three-point interaction.

The self-energy, corresponding to the scattering amplitude Γ is found as a sum of the diagrams shown in Fig. 1b:

$$\Sigma(\mathbf{k},\omega) = \frac{4}{N}\sum_{\mathbf{q}} v_{\mathbf{q}}^2 \Gamma(\mathbf{k}+\mathbf{q}, \omega-\omega_{\mathbf{q}}). \qquad (9)$$

Let us stress again that at our level of approximation (dilute gas), there is only a normal self-energy. Next, in order to find the renormalized spectrum, one has to solve the coupled Dyson equations for the normal and anomalous Green's functions. Since the procedure is well known from the theory of a Bose gas, we write only the final result for the normal Green's function [14]:

$$G(\mathbf{k},\omega) = \frac{\omega + A_{\mathbf{k}} + \Sigma(\mathbf{k},-\omega)}{[\omega + A_{\mathbf{k}} + \Sigma(\mathbf{k},-\omega)][\omega - A_{\mathbf{k}} - \Sigma(\mathbf{k},\omega)] + B_{\mathbf{k}}^2}. \qquad (10)$$

After separating this equation into a quasiparticle contribution and incoherent background, we find

$$G(\mathbf{k},\omega) = \frac{Z_{\mathbf{k}} U_{\mathbf{k}}^2}{\omega - \Omega_{\mathbf{k}} + i\delta} - \frac{Z_{\mathbf{k}} V_{\mathbf{k}}^2}{\omega + \Omega_{\mathbf{k}} - i\delta} + G_{\mathrm{inc}}. \qquad (11)$$

The renormalized triplet spectrum and the renomalization constant are

$$\Omega_{\mathbf{k}} = Z_{\mathbf{k}}\sqrt{[A_{\mathbf{k}} + \Sigma(\mathbf{k},0)]^2 - B_{\mathbf{k}}^2}, \qquad (12)$$

$$Z_{\mathbf{k}}^{-1} = 1 - \left(\frac{\partial\Sigma}{\partial\omega}\right)_{\omega=0}. \qquad (13)$$

The renormalized Bogoliubov coefficients in (11) are

$$U_{\mathbf{k}}^2, V_{\mathbf{k}}^2 = \pm\frac{1}{2} + \frac{Z_{\mathbf{k}}[A_{\mathbf{k}} + \Sigma(\mathbf{k},0)]}{2\Omega_{\mathbf{k}}}. \qquad (14)$$

Equations (8),(9),(12)–(14) have to be solved selfconsistently for $\Sigma(\mathbf{k},0)$ and $Z_{\mathbf{k}}$. From Eq. (11) it is also clear that one has to replace $u_{\mathbf{k}} \to \sqrt{Z_{\mathbf{k}}}\,U_{\mathbf{k}}$, $v_{\mathbf{k}} \to \sqrt{Z_{\mathbf{k}}}\,V_{\mathbf{k}}$ in (8) and (9) [and also in (15) and (16); see below].

We have found that the effect of H_3 and H_4 on the quasiparticle spectrum is small, compared to the renormalization due to H_U. However, these two terms have to be included for the precise determination of the critical point. We treat H_4 in mean-field theory, by splitting the quartic operator products into all possible pairs. This is equivalent to taking only one-loop diagrams (first order in J) into account. These diagrams renormalize the two coefficients

$$A_{\mathbf{k}} \to A_{\mathbf{k}} + 2(1+\lambda)J\xi_{\mathbf{k}}\frac{1}{N}\sum_{\mathbf{q}}\xi_{\mathbf{q}} v_{\mathbf{q}}^2, \qquad (15)$$

$$B_{\mathbf{k}} \to B_{\mathbf{k}} - 2(1+\lambda)J\xi_{\mathbf{k}}\frac{1}{N}\sum_{\mathbf{q}}\xi_{\mathbf{q}} u_{\mathbf{q}} v_{\mathbf{q}}. \qquad (16)$$

This concludes the solution of the two-layer problem ($\lambda = 1$).

To solve the case $\lambda = 0$ we also have to take into account H_3. It is convenient to rewrite H_3 in terms of the Bogoliubov transformed operators $\tilde{t}_{\mathbf{k}\alpha}, \tilde{t}_{\mathbf{k}\alpha}^{\dagger}$, since in this way only the normal Green's functions remain. To one-loop order (J^2) the renormalization of the spectrum is determined by the sum of the two diagrams in Fig. 1c. The formula for the interaction vertex in Fig. 1c is quite lengthy and we do not present it here. Once the vertex is known, the self-energy of Fig. 1c can be easily computed, leading to renormalization of $A_{\mathbf{k}}$ and $B_{\mathbf{k}}$.

The results of the self-consistent numerical solution are summarized in Figs. 2 and 3. Figure 2 shows the

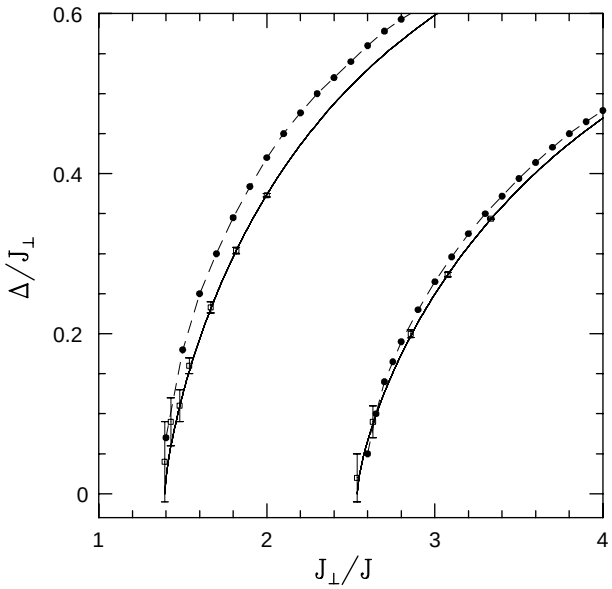

FIG. 2. Triplet gap as a function of interlayer coupling for $\lambda = 0$ (left curves) and $\lambda = 1$ (right curves). The dashed lines with the solid circles are the results of the self-consistent solution. Open squares (with error bars) are from direct Padé approximants to the dimer series while solid lines are from approximants which assume $\nu = 0.71$.

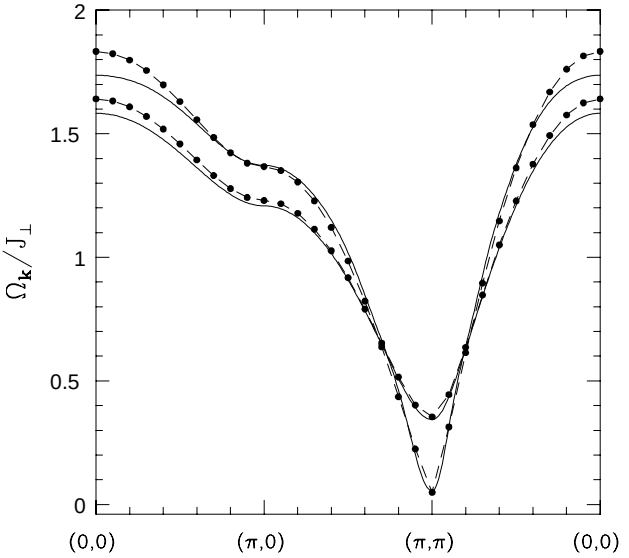

FIG. 3. Triplet excitation spectrum for $\lambda = 1$ along high symmetry directions in the Brillouin zone. The dashed line with solid circles is the self-consistent solution while the solid lines are from direct summation of the dimer series. The upper (at $\mathbf{k} = 0$) dimer series curve corresponds to the critical (within the error bar) spectrum ($J_\perp/J = 2.54$), while the upper analytical curve is computed at $J_\perp/J = 2.6$, in order to have the same gap $\Delta/J_\perp = 0.05$. The lower (at $\mathbf{k} = 0$) curves correspond to $J_\perp/J = 3.33$.

triplet gap $\Delta = \Omega_{\pi,\pi}$ as a function of the interlayer coupling. The transition into the Néel ordered phase occurs at $(J_\perp/J)_c = 2.57$ ($\lambda = 1$), 1.37 ($\lambda = 0$). We have also calculated the gap by using dimer series expansions [12] up to order 11 (10) for $\lambda = 1$ ($\lambda = 0$). The critical points are found at $(J_\perp/J)_c = 2.52(2)$ ($\lambda = 1$), 1.39(4) ($\lambda = 0$), or 2.537(5) ($\lambda = 1$), 1.393(8) ($\lambda = 0$) by fixing the critical exponent $\nu = 0.71$ [12]. The agreement between the analytic method and the dimer series results is excellent. Such a good agreement is better than might have been expected. Our analytic method involves approximations and an error of a few percent is always expected. The gap critical exponent ν, defined as $\Delta \sim (J - J_c)^\nu$, is $\nu = 0.5$ in our analytical calculation, while the dimer series gives $\nu \approx 0.7$, in agreement with the $O(3)$ nonlinear sigma model prediction. Recall that the mean-field approximation gives $\nu = 1$. Our diagrammatic approach is not valid very close to the critical point since the neglected terms in $\Gamma(\mathbf{K})$ are of the form $\sum_{\mathbf{q}} v_{\mathbf{q}}^2/\omega_{\mathbf{q}}$ and thus logarithmically diverge at criticality. However this happens only very near to the critical point.

The comparison of the excitation spectra, presented in Fig. 3, shows that the agreement is very good over almost the whole Brillouin zone. The disagreement between the two curves is largest at $\mathbf{k} = 0$, where it is about 5%.

In conclusion, we have presented an effective analytical approach to take into account the hard-core constraint which appears in the bond operator description of the dimer phase. The triplet excitations are described as a dilute Bose gas with infinite on-site repulsion. We find that the spectrum is renormalized mostly due to the hard core, while the additional three- and four-point interactions are comparatively weak and can be treated perturbatively. The advantages of our formulation are that it is simple and captures the essential physics, being in agreement within a few percent with results obtained by dimer series expansions. Obvious other applications of the method include the 2D Heisenberg model with dimerization, the Heisenberg ladder, and the Kondo lattice model [15]. The method can also be easily generalized to describe phases with spontaneously broken symmetries and nonzero temperature.

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